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AN OPERATIONAL METHOD FOR ASSESSING TRAFFIC-RELATED AIR POLLUTION IN URBAN STREETS

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Abstract

Urban air quality has been a topic of major public concern and scientific research in recent years. Several theoretical and experimental studies have focused on the assessment of air quality within street canyons and other microenvironments (intersections, motorways, parking spaces, etc.), where population exposure to traffic-related pollutants is relatively high.

The aim of this study was to develop a practical methodology for assessing traffic-related air pollution in urban streets, after testing available monitoring and modelling techniques. To meet this objective, a large amount of original air quality, meteorological and traffic data were collected during four intensive short-term and one long-term monitoring campaigns carried out in the region of Paris from December 1998 to December 2001. These campaigns covered three representative street canyon sites (Bd. Voltaire, Rue de Rennes, Av. Leclerc – Pl. Basch) as well as a motorway service station (RN10 petrol station).

Passive and active monitoring techniques were used to sample a wide range of inorganic (CO, NO_x and O_3) and organic gases (benzene, toluene, xylene, ethylbenzene, formaldehyde, acetaldehyde, etc.) at different heights and distances from the kerb. Indicative background measurements were also taken during the same sampling periods. Furthermore, relevant meteorological (synoptic and local) and traffic information was obtained on each site.

The analysis of the data gave insights into the dispersion and transformation processes taking place within the streets. Channelling effects induced by parallel to the road axis winds gave rise to relatively high kerbside pollution levels. On the other hand, perpendicular synoptic winds generated air vortices within the canyons, which resulted in steep crossroad concentration gradients. In that case, higher pollution levels were observed on the leeward than on the windward side of the streets. A significant reduction of concentrations with height above the ground was also observed within two of the street canyons (Bd. Voltaire and Av. Leclerc). In all cases, roadside concentrations were several times higher than the corresponding urban background values.

This spatial variability indicates a strong transport effect on the pollutant distribution within urban canyons, caused by the synoptic wind and influenced by the geometry of the street. That may have serious implications in terms of population exposure and compliance with air quality legislation. In this context, the siting of permanent monitoring equipment becomes crucial.

A relationship between CO and benzene as well as an exponential expression linking pollutant concentrations at different heights within the canyons were empirically deduced. Five dispersion models of different levels of complexity (STREET-SRI, OSPM, AEOLIUS, CAR-International, and CALINE4) were used to calculate CO and benzene concentrations at the campaign sites. The Computational Fluid Dynamic code PHOENICS was also tested for one location.

The comparison between observed and predicted values revealed the advantages and drawbacks of each model in association with the configuration of the street and the meteorological conditions. Furthermore, a sensitivity and uncertainty analysis involving three of the available models (STREET-SRI, OSPM and AEOLIUS) was carried out. OSPM was slightly modified in order to allow user access to certain internally coded parameters.

An operational method combining multi-site sampling and dispersion modelling was finally proposed for assessing air quality in urban streets, taking into account the pronounced spatial and temporal variability of traffic-related air pollution, the modelling uncertainty, the practical constraints related to measurements and models, and the needs of decision makers. This methodology may find wider application in air quality management, urban and transport planning, and population exposure studies.

Sommaire

Cette thèse a été réalisée dans le cadre d'une coopération entre l'INERIS et l'Université de Greenwich. Elle a été financée par le projet "Etudes des Microenvironnements" du Ministère Français de l'Environnement.

L'objectif principal de cette recherche est l'amélioration des connaissances sur les processus physico-chimiques qui gouvernent la pollution dans le milieu urbain, ainsi que le développement de méthodes de mesure et d'évaluation de la pollution dans ce milieu.

Quatre campagnes intensives de courte durée et une de longue durée ont été réalisées pour prélever des polluants atmosphériques dans la région parisienne pendant la période entre décembre 1998 et décembre 2001. Ces campagnes ont eu lieu dans trois rues "canyons" de Paris (Bd. Voltaire, Rue de Rennes et Av. Leclerc – Pl. Basch) et à une station service sur la Route Nationale 10 (Rambouillet).

Les polluants mesurés étaient: CO, NO_x, O_3, COV (benzène, toluène, xylène, cyclohexane, éthylbenzène, 1,3,5 TMB, 1,2,4 TMB, formaldéhyde, acétaldéhyde, etc.). Les mesures ont été réalisées par prélèvement actif, passif ou encore par monitorage continu, à différentes hauteurs et distances du trottoir. La pollution de fond a également été mesurée pendant les mêmes périodes. Une remorque laboratoire sur les sites a permis d'installer les appareils de mesure et de saisie des données météorologiques locales. D'autres données météorologiques ont été fournies par Météo France, et les données du trafic par la Mairie de Paris et vérifiées par comptage sur place.

Les niveaux de concentration de CO observés pendant les campagnes étaient faibles et souvent inférieurs à 2 ppm. Cependant, l'intérêt du monitorage de ce composé réside dans le fait qu'il est un excellent traceur pour d'autres substances. En ce qui concerne les concentrations des polluants réglementés, comme le benzène, il est intéressant de remarquer que les niveaux de fond sont toujours inférieurs à la valeur limite européenne de 5 $\mu g/m^3$, alors que dans les rues canyons, cette limite est souvent dépassée. En outre, une forte variabilité spatiale et temporelle des concentrations de COV a été observée dans les rues.

Cinq modèles de dispersion ont été choisis pour effectuer les simulations numériques: STREET-SRI, OSPM, AEOLIUS, CAR-International et CALINE4. Un modèle numérique (PHOENICS) a également été testé. Ces modèles prennent en compte les mécanismes physiques de dispersion des polluants à proximité des sources et les réactions chimiques rapides. Les données d'entrée liées aux sources d'émissions et aux conditions météorologiques ont été obtenues en utilisant différentes méthodes de calcul proposées dans la littérature. Les résultats des simulations ont été traités à l'aide de logiciels statistiques appropriés. Pour chaque modèle, les paramètres d'entrée les plus importants ont été identifiés. De plus, une étude d'incertitude impliquant trois des modèles disponibles a été réalisée. Enfin, OSPM a été sensiblement modifié afin de permettre l'accès à certains paramètres internes du modèle.

Enfin, une méthode opérationnelle combinant différentes techniques de monitorage, et l'utilisation de modèles mathématiques a été présentée pour l'évaluation de la qualité de l'air dans les rues urbaines. Cette méthode prend en compte la variabilité spatiale et temporelle de la pollution atmosphérique liée à la circulation automobile, les contraintes pratiques liées aux mesures et à la modélisation, et les besoins des organismes responsables de la qualité de l'air. Les applications immédiates peuvent servir à l'évaluation de l'exposition des populations séjournant dans des zones urbaines, y compris à plusieurs mètres d'altitude.

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Preface

This study formed part of a research collaboration project between the Institut National de l'Environnement Industriel et des Risques (INERIS) and the University of Greenwich, which was mainly funded by the French Ministry for the Environment.

Research supervision was provided by Professor B. Fisher from the School of Earth and Environmental Sciences (and later the Environment Agency) and Professor K. Pericleous from the School of Computing and Mathematical Sciences of the University of Greenwich, and Dr N. Gonzalez-Flesca from INERIS.

The author of the thesis designed and carried out a series of air quality monitoring campaigns in the region of Paris (France). Furthermore, he analysed and interpreted the monitoring data using a variety of mathematical models. This combination of monitoring and modelling techniques enabled the author to develop an operational methodology for assessing traffic-related air pollution in urban streets. The conclusions of this study may help local authorities, government agencies and environmental professionals in their tasks related to roadside air quality by optimising the use of available techniques.

A substantial part of the research results have been published by the author in peer-reviewed scientific journals and presented in international conferences during his PhD. His academic supervisors co-authored these articles and presentations. Technical personnel from INERIS were acknowledged for carrying out the chemical analysis of the VOC samples and calibrating the monitoring instruments.

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$h\nu$	photon of light
M	molecule of N_2 or O_2
ε	turbulent kinetic energy dissipation rate
k	turbulent kinetic energy
H	height of the canyon
W	width of the canyon
L	length of the canyon (road distance between two major intersections)
C	ambient pollutant concentration
$K_{axial/radial}$	empirical constants for deviation from ideal behaviour (diffusive sampling)
D	diffusion coefficient of a gas in ambient air / traffic density in OSPM
m	mass of the pollutant sampled
t	time of exposure
C_i	pollutant concentration at a given location "i"
C_b	background concentration of the pollutant
C_p	pollutant concentration due to emissions released within the petrol station
C_m	pollutant contribution from vehicle traffic in the proximity of the petrol station.
α	slope of the linear relationship relating CO and benzene / proportionality constant in OSPM
β	intercept in the linear relationship relating CO and benzene
C_s	pollutant concentration due to emissions released within the canyon
$C_S^{\ L}$	leeward receptor concentration due to emissions released within the canyon
$C_S^{\ W}$	windward receptor concentration due to emissions released within the canyon
K	empirical parameter of STREET-SRI model
Q	rate of release of emissions in the street
x	horizontal distance between the receptor and the centre of the nearest traffic lane
z	height of the receptor
h_o	height of initial pollutant dispersion (effective release height)
u	street-level wind speed
U	roof-level wind speed
U_S	constant accounting for the vehicle induced turbulence in STREET-SRI model
C_d	pollutant contribution from the direct flow from the source to the receptor
C_r	pollutant contribution due to the recirculation of pollutants within the vortex
F	factor depending on the synoptic wind in OSPM

σ_w	vertical velocity fluctuation due to mechanical turbulence in OSPM
σ_{wo}	traffic-induced turbulence in OSPM
b	aerodynamic drag coefficient
N	number of vehicles using the street per time unit
V	average vehicle speed
S^2	road surface occupied by a single vehicle
L_r	width of the recirculation zone in OSPM
L_t, L_{S1}, L_{S2}	dimensions of the recirculation zone in OSPM
σ_{wt}	ventilation velocity of the canyon in OSPM
λ	proportionality constant in OSPM
F_{roof}	proportionality constant in OSPM
F_{vortex}	proportionality constant in OSPM
r	wind speed dependent factor reflecting the strength of the vortex in OSPM
ϕ	angle of the roof-level wind with respect to the street
$U_{critical}$	critical velocity for vortex formation within the canyon
z_o	surface roughness length
F_{wind}	empirical constant in OSPM
U_a	wind speed corresponding to a meteorological mast
F_{mast}	empirical parameter in OSPM
H_α	height of anemometer
C(z)	pollutant concentration at height z
A, B, q	regression coefficients in exponential expression of vertical benzene reduction
C_r	pollutant concentration a reference height z_r
C_p	predicted pollutant concentrations
C_o	observed pollutant concentrations

Glossary

ABLWT	Atmospheric Boundary Layer Wind Tunnel
ADEME	Agence de l'Environnement et de la Maîtrise de l'Energie (France)
ADMS-Urban	Second generation urban-scale dispersion model
ADREA-HF	CFD code for simulating vapour cloud dispersion in complex terrain
AEOLIUS Full/Screen	Urban canyon model (Met Office, U.K.)
AEOLIUSQ	Emission model (Met Office, U.K.)
AirGIS	A decision-support GIS tool (based on OSPM)
AIRPARIF	Air quality monitoring network for the region of Paris (France)
APPS	Three-dimensional numerical model
APRAC	Air Pollution Research Advisory Committee (U.S.A.)
AQMA	Air Quality Management Area
AUTO-OIL II	European research programme
BLASIUS	Atmospheric boundary layer wind tunnel (Meteorological Institute of Hamburg University)
BTX	Benzene Toluene Xylene
CALGRID	Urban scale photochemical model
CALINE4	California Line Source Dispersion Model
CAR International	Calculation of Air pollution from Road traffic model (dispersion model)
CAR-FMI	Gaussian line source model for calculating pollution from road networks
CARMEN	Urban canyon model
CARSMOG	Extension of CAR model
CFD	Computational Fluid Dynamics
CFX-5	Numerical CFD code
CFX-TASCflow	Numerical CFD code
C_6H_6	Benzene
CHENSI	CFD model
CITY	Wind sub-model (see SCAM)
CO	Carbon monoxide
COPERT	Computer Program to Calculate Emissions from Road Traffic
COPREM	Constrained Physical Receptor Model
CORINAIR	Co-ordination of Information on Air Emissions
CPBM	Canyon Plume Box Model (dispersion model)
DAPPLE	Dispersion of Air Pollution & Penetration into the Local Environment project
DIAL	Differential Absorption Lidar (line measurements)
DOAS	Differential Optical Absorption Spectroscopy (line measurements)
DEFRA	UK Department for Environment, Food and Rural Affairs (formerly DETR)
DETR	UK Department of the Environment, Transport and the Regions
DMRB	Design Manual for Roads and Bridges (screening dispersion model)
EC	Commission of the European Communities
EMFAC	Emission model (Air Resources Board – California, USA)
EnFlo	Meteorological wind tunnel (University of Surrey, U.K.)
EPA	Environmental Protection Agency (U.S.A.)
EU	European Union
FAC2	Fraction of predictions within a factor of two (statistical evaluation)
FB	Fractional Bias (statistical model evaluation)
FID	Flame Ionisation Detector
FloVENT	Nunerical CFD code
FLUENT	General purpose CFD model
Fluidyn-PANACHE	Commercial CFD model

GC	Gag Chromatography
GEM	Micro-scale Lagrangian particle model
GIS	Geographic Information System
HC	Hydrocarbons
HEATX	Nunerical CFD code
HIWAY-2	Highway air pollution model (U.S.EPA)
HPLC	High Performance Liquid Chromatography
H/W	Canyon aspect ratio (height-to-width)
IARC	International Agency for Research on Cancer
IMPACT	Road traffic emission model (ADEME)
INDIC AIRVIRO	Regional-scale dispersion model
INERIS	Institut National de l'Environnement Industriel et des Risques (France)
JEA	Japan Environmental Agency
KSS	Kinetic Sequential Sampling system (for particulate matter)
L-2B	Wind tunnel (von Karman Institute, Belgium)
LDA	Laser Doppler Anemometry (wind tunnel modelling)
LES	Large Eddy Simulation (CFD numerical principles)
LIFE	European research project
MAPS	Dispersion model (modified version of STREET-SRI)
MEMO	MEsoscale MOdel
MERCURE	Atmospheric dispersion model
METRAS	Mesoscale dispersion model
MG	Geometric Mean Bias (statistical model evaluation)
MICRO-CALGRID	Microscale dispersion model
MIMO	MIcroscale MOdel (coupled with MEMO).
MISKAM	Numerical CFD model
MITRAS	Microscale dispersion model (coupled with METRAS)
MOBILE	Mobile source emission factor model (U.S.EPA)
MOBILEV	German emission model
NERI	National Environmental Research Institute (Denmark)
NMSE	Normalised Mean Square Error (statistical model evaluation)
NO_x	Nitrogen oxides (NO and NO_2)
O_3	Ozone
OMG volume-source	Osaka Municipal Government volume-source model
OSPM	Operational Street Pollution Model (NERI, Denmark)
PAH	Polycyclic Aromatic Hydrocarbons
Paramics	Traffic simulation package
PART5	Emission model for particulate matter
PHOENICS	General purpose CFD model
PIV	Particle Image Velocimetry
$PM_{2.5}/PM_{10}$	Particulate Matter
PROKAS-V	Gaussian urban scale model
PROKAS-B	Street canyon module of PROKAS-V
PUFFER	Gaussian puff model
QA/QC	Quality Assurance / Quality Control
RANS	Reynolds Averaged Navier-Stokes flow equations (CFD principles)
RPE	Radiello Perkin Elmer tube
RSM	Advanced turbulence model
SCAM	Numerical CFD model (consisted of the wind sub-model CITY and the dispersion sub-model SCALAR)
SCALAR	Dispersion sub-model (see SCAM)
SCV	Spatial Coefficient of Variation

SF_6	Hexafluorosulfide (tracer gas)
SIMPLE	Semi-Implicit Method for Pressure Linked Equations (CFD solver)
SIRANE	Dispersion model for street networks
SLAQ	Street Level Air Quality model (for particulate matter)
SO_2	Sulphur dioxide
SPM	Suspended Particulate Matter
SRI	Stanford Research Institute
STAR-CD	General purpose CFD model
STREET (SRI)	Urban canyon model (Stanford Research Institute)
STREET (Targeting)	Urban canyon model (based on MISKAM)
STREET BOX	Dispersion model
STREETBOX	Air quality monitoring instrument
TEACH-2E	Numerical CFD code
TEOM	Tapered Element Oscillating Microbalance (automatic PM_{10} monitor)
TNO-Traffic	Gaussian plume-type dispersion model
TOKYO	Line source dispersion model (Tokyo Metropolitan Government, Japan)
TRAPOS	Optimisation of Modelling Methods for Traffic Pollution in Streets
TSP	Total Suspended Particles
URBCAP	Research project (France)
USIAM	Integrated assessment model
VG	Geometric Mean Variance (statistical model evaluation)
VOC	Volatile Organic Compounds
WHO	World Health Organisation
WMO	World Meteorological Organisation

Chapter 1
Introduction

1.1. Background

The increasing awareness of scientist and public about the acute and chronic health effects of several traffic-related pollutants (NO_2, CO, hydrocarbons, etc.) has led in recent years to a significant number of relevant epidemiological studies mainly concerning urban populations (Burnett et al., 1998; Hoek et al., 2000). Although the mechanisms are not fully explained from a medical point of view, epidemiological evidence suggests that ambient air pollution is a contributing cause of morbidity and mortality (Bates, 1992).

For assessing health risks related to air pollution, it is necessary to quantify the exposure of the population to the various hazardous substances released in the atmosphere. The key assumption in previous research on the topic has been that ambient concentrations of air pollutants can be used as an indicator of population exposure, despite the fact that people in European cities typically spend the majority of their time (up to 90%) indoors (Gonzalez-Flesca et al., 2000; Hertel et al., 2001). Baek and Perry (1997) demonstrated experimentally the importance of ambient air quality in determining the quality of indoor air in two major Korean cities. Another field experiment conducted by Kingham et al. (2000) in the area of Huddersfield (England) suggested that outdoor pollution may give a useful measure of exposure to traffic-related pollutants as part of epidemiological studies.

In most cases so far, the population exposure to air pollution has been assessed through crude assumptions. It has been assumed, for example, that concentrations observed at a single or a few permanent monitoring stations within a city are representative of the exposure of the entire urban population (Fenger, 1999). This is in line with current European legislation relevant to health protection. The Council Directive related to limit values for benzene and carbon monoxide in ambient air (European Commission, 2000) specifies that only one fixed sampling point is enough for assessing compliance with limit values for the protection of human health in urban agglomerations with less than 250,000 of population. This practice is in contradiction with findings from current research, which show a significant small-scale spatial variability of traffic-related pollution in urban areas (Hewitt, 1991; Croxford et al., 1996; Monn et al., 1997; Croxford and Penn, 1998; Monn, 2001).

Nowadays, most large European cities are covered to some extent by air quality monitoring networks, which provide continuous measurements of key pollutants (e.g. NO_x, SO_2, CO). Nevertheless, a more detailed spatial profile of ambient concentrations is often needed for population exposure studies than is usually available

(WHO, 1999). This need is more pronounced in areas with high population density, strong emission sources, and limited natural ventilation (e.g. urban streets and avenues). For this reason, alternative sampling techniques not entailing the high cost and practical constraints (e.g. bulk of equipment, power supply requirements) of continuous air quality monitoring need to be tested. In addition, dispersion models should be used to provide concentration estimates in areas that are not sufficiently covered by measurements or to explore future emission and traffic scenarios.

1.1.1. Traffic-related air pollution

Since the industrial revolution and for most of the 20th century, urban air pollution was considered as a local problem mainly associated with domestic heating and industrial emissions, which are now controllable to a great extent. Despite significant improvements in fuel and engine technology, present day urban environments are mainly dominated by traffic emissions (Fenger, 1999; Colvile et al., 2001). It is now generally recognised that many of the substances directly emitted by vehicles in the ambient air or indirectly produced through photochemical reactions represent a serious hazard for human health (Hoek et al., 2000; Nyberg et al., 2000; Dab et al., 2001).

The main traffic-related pollutants are CO, NO_x, hydrocarbons, and particles. CO is an imperfect fuel combustion product. Combustion also produces a mixture of NO_2 and NO, of which more than 90% is in the form of NO. A wide range of unburned and chemically transformed hydrocarbons (e.g. benzene, toluene, ethane, ethylene, pentane, etc.) is emitted by motor vehicles through a number of different processes (e.g. evaporation, fuel tank displacement, oil seep, etc.). Finally, particles of condensed carbonaceous material are emitted mainly by diesel and poorly maintained petrol vehicles.

Atmospheric pollutants are responsible for both acute and chronic effects on human health (WHO, 2000). CO is an asphyxiating pollutant that reduces the ability of blood to carry oxygen to the different organs (Burnett et al., 1998). Therefore, short-term exposure to high CO concentrations might cause an acute health impact. On the other hand, pollutants like benzene have a cumulative effect on human health. Long-term exposure to high benzene levels increases the risk for an individual to suffer from cancer (Cicolella, 1997). Furthermore, there are gases like NO_2 that are responsible for both short- and long-term health effects. Depending on the effects related to each substance, atmospheric pollutants are regulated with respect to different exposure times. For example, in the European air quality guidelines, standards are set for benzene as one year averages, for CO as eight hour averages, and for NO_2 as both one hour and one year averages (European Commission, 1999; 2000).

Particulate matter with aerodynamic diameter below 10 μm (PM_{10}) and especially the finer fraction with aerodynamic diameter below 2.5 μm ($PM_{2.5}$) was found to associate with increased daily mortality and asthma (Dockery and Pope, 1994; Anderson at al., 1992; Harrison and Yin, 2000). Furthermore, ultrafine particles (i.e. aerodynamic diameter < 100 nm) are likely to represent a major health risk (Seaton et al., 1995). Nevertheless, current European legislation addresses only total PM_{10} as 24-hour and one year averages, while U.S. legislation regulates both PM_{10} and $PM_{2.5}$ as three year averages (EPA, 1996). Although roadside concentrations differ significantly from background levels, all outdoor environments are subject to the same regulatory standards for ambient air quality.

In urban environments and especially in those areas where population and traffic density are relatively high, human exposure to hazardous substances is expected to be significantly increased. This is often the case near busy traffic axes in city centres, where urban topography and microclimate may contribute to the creation of poor air dispersion conditions giving rise to contamination hotspots. High pollution levels have been observed in street canyons, which is a term frequently used for urban streets flanked by buildings on both sides. Within these streets, pedestrians, cyclists, drivers and residents are likely to be exposed to pollutant concentrations exceeding current air quality standards.

1.1.2. Air quality monitoring and modelling

The impact of air pollution on urban environments has led to numerous modelling studies related to the influence of buildings and other urban structures on pollutant accumulation/dissipation patterns (Georgii, 1969; Oke, 1988; Bitan, 1992). The main features of pollutant dispersion within urban canyons are well understood through the pioneering work of Johnson et al. (1973), Dabberdt et al. (1973), Hotchkiss and Harlow (1973), Nicholson (1975) and others.

Nowadays, automated monitoring networks operate in many European cities providing detailed air quality information on a regular basis. There are several techniques available for monitoring gaseous pollutants (e.g. continuous monitoring using standard gas analysers, diffusive and pumped sampling using tubes filled with an appropriate adsorbent, grab sampling using canisters) and particulate matter (e.g. filtration and impaction). Each one of them can be associated with a number of advantages and disadvantages that make it suitable or not for a specific application.

Dispersion models are also widely used for assessing roadside air quality by providing predictions of present and future air pollution levels as well as temporal and spatial variations (Sharma and Khare, 2001). When used in a knowledgeable way, they can be very useful in giving insights into the physical and chemical processes that govern the dispersion and transformation of atmospheric pollutants.

1.2. Motivation and aims

This study was motivated by the widely expressed need for evaluating and improving existing monitoring and modelling methodologies for assessing air quality in roadside microenvironments. In recent years, a plethora of sampling devices and mathematical models have been developed and made commercially available. However, the main users – local authorities, regulatory bodies, consultants, etc. – have been generally given little strategic guidance about how to best employ these techniques as well as about their applicability in particular situations (Cooper, 1987). Furthermore, only a limited number of model inter-comparison and harmonisation exercises have been conducted (Lohmeyer et al., 2002).

In the UK, local authorities expressed concern about the lack of necessary expertise to undertake effectively their new responsibilities following the publication of the National Air Quality Strategy (NAQS), which put more emphasis on local action (Beattie et al., 2002).

Given the fact that the total number of receptors is limited by practical constraints, local authorities, public health agencies, etc., have to rely to a certain extent on mathematical models for assessing air quality. Taking into account the great variety of urban environments, different models might apply for example to street canyons, wide avenues, motorways, intersections and urban background locations. Furthermore, there is a need for original data sets containing detailed air quality, meteorological and traffic information for validating these models.

Street canyons raise great concern in terms of air quality due to the relatively high pollution concentrations and population density occurring in these locations compared to the background areas (Skov et al., 2001). In big urban agglomerations like Paris and London, a large number of people live, work, commute or walk in busy streets flanked by relatively tall buildings. A question that needs to be answered is how representative a permanent monitoring station supported by modelling can be of the actual population exposure in such environments (Fisher, 2001).

The aim of this research is to create a comprehensive air quality database for model validation purposes, address the issue of data representativeness, test different monitoring/modelling approaches, and finally propose a sound methodology for assessing roadside air quality.

1.3. List of research objectives

- Create an original air quality database for microscale (i.e. street canyon) model validation.

- Assess ambient air quality in a variety of roadside microenvironments in Paris with respect to the national and international standards.

- Identify implications for population exposure studies.

- Test, evaluate and possibly improve available dispersion models that may be used by local authorities in street canyon applications.

- Evaluate passive sampling as an alternative technique for dispersion model validation.

- Propose a practical methodology for estimating model uncertainty.

- Assess the effectiveness of the "Stage 2" vapour recovery system in reducing air pollution in the vicinity of petrol stations.

- Develop an operational methodology for assessing traffic-related air pollution in urban streets.

1.4. Tested hypothesis and methodology

For optimising the standard air quality assessment procedures, it is important to identify the best air pollution indicators as well as the minimum amount of monitoring/sampling data needed to establish air pollution levels in areas where regulatory standards are likely to be exceeded. These data should include information on spatial and temporal variation patterns.

Air quality monitoring should be complemented with dispersion modelling in order to optimise resources. The main questions to be answered are the following: Which is the most appropriate mathematical model for a specific application? Which are the most relevant model input data (e.g. meteorological data, emission factors, etc.)? In which way modelling results should be interpreted and how much decision-makers should rely on them?

In this research, different air quality monitoring/sampling methodologies were tested in a variety of roadside locations during short a long time periods. Three intensive short-term monitoring campaigns and one long-term sampling campaign were carried out in central Paris. An additional monitoring campaign was carried out in a motorway service station within the region of Paris. A wide range of models of different level of complexity (screening, semi-empirical, CFD) were tested using the available field data. Input meteorological and traffic data were locally collected and/or obtained from remote sources. Finally, emphasis was put on practical issues (cost-effectiveness, user-friendliness, etc.) that may be important to common users.

The present study built on the experience gained from two previous sampling campaigns and relevant model simulations carried in London and Paris by the same research team (Jones at al., 1998; 2000).

The scope of this research is original and involves a number of overlapping scientific disciplines like mathematical modelling, environmental chemistry and engineering, urban meteorology, atmospheric physics and air quality management.

1.5. Outline of the thesis

This thesis is structured in six chapters. Chapter 1 is the introduction, which includes the aim and objectives of the study. Chapter 2 is a review of monitoring and modelling methodologies that have been used in the past for assessing roadside air quality. A great number of relevant research studies are also summarised in this chapter.

Chapter 3 presents the monitoring/sampling techniques applied and the results obtained during three intensive short-term monitoring campaigns in urban street canyons (Boulevard Voltaire, Rue de Rennes and Avenue Leclerc), one long-term sampling campaign in a complex urban intersection (Place Basch), and one short-term sampling campaign in a motorway service station (Route Nationale 10). The sites of the campaigns, the sampling protocol and monitoring/sampling results are separately presented for each individual "case study". An overall discussion on the results follows at the end of this chapter.

Chapter 4 is dedicated to mathematical modelling. It starts with a description of the three models that are more extensively used in this study (STREET-SRI, OSPM and AEOLIUS), highlighting their empirical parameters and assumptions. Furthermore, a sensitivity analysis for certain internal parameters of OSPM is presented as well as a number of modifications and extensions to this model developed in this study. The methodologies followed for creating model inputs and evaluating model results are described in detail. The calculated pollutant concentrations are presented separately for each one of the six models involved in this study

(STREET-SRI, OSPM, AEOLIUS, CAR International, PHOENICS, and CALINE4) and compared with the observed values. The model uncertainty is estimated using two original methodologies. Finally, at the end of this chapter there is an overall discussion on the performance and suitability of the models.

Chapter 5 integrates the main findings of the previous chapters into a practical methodology for assessing traffic-related air pollution in urban streets. In addition, it tackles the problem of comparing model predictions with regulatory standards, discusses implications for population exposure studies, and briefly presents some air pollution mitigation measures.

A summary of the main findings and conclusions is given in Chapter 6, together with a list of research achievements, a discussion on the limitations of this study and some recommendations for further research.

Chapter 2

Review of street canyon monitoring and modelling

2.1. Street canyon characteristics

2.1.1. Canyon geometry

The term *street canyon* ideally refers to a relatively narrow street with buildings lined up continuously along both sides (Nicholson, 1975). However, the same term has been used to refer to larger streets, also called *avenue canyons*. In the real world, a broader definition of the term has been applied, including urban streets that are not necessarily flanked by buildings continuously on both sides, allowing thus for some openings on the walls of the canyon.

The dimensions of a street canyon are usually expressed by its aspect ratio, which is the height (H) of the canyon divided by the width (W). A canyon might be called *regular*, if it has an aspect ratio of approximately equal to 1 and no major openings on the walls. An avenue canyon may have an aspect ratio below 0.5, while a value of 2 may be representative of a deep canyon. Finally, the length (L) of the canyon usually expresses the road distance between two major intersections, subdividing street canyons into short ($L/H \approx 3$), medium ($L/H \approx 5$), and long canyons ($L/H \approx 7$). Urban streets might be also classified in *symmetric* (or *even*) canyons, if the buildings flanking the street have approximately the same height, or *asymmetric*, if there are significant differences in building height.

2.1.2. Wind flow

The climate of street canyons is primarily controlled by the micro-meteorological effects of urban geometry rather than the mesoscale forces controlling the climate of the boundary layer (Hunter et al., 1992). A clear distinction should be made between the synoptic above roof-top wind conditions and the local wind flow within the cavity of the canyon (Fig. 2.1). Depending on the synoptic wind (or free-stream velocity), three main dispersion conditions can be identified: (i) low wind conditions, for synoptic winds lower than 1.5 m/s, (ii) perpendicular or near-perpendicular flow for synoptic winds over 1.5 m/s blowing at an angle of more than 30° to the major canyon axis, and (iii) parallel or near-parallel flow for winds over 1.5 m/s blowing from all other directions. In the case of perpendicular flow, the upwind side of the canyon is usually called *leeward*, and the downwind *windward*.

The emphasis has often been on the two-dimensional nature of the flow, studying vertical cross-sections at mid-canyon level. When the above roof flow is perpendicular to the canyon and the wind speed is greater than 1.5 to 2 m/s, flow may be described in terms of three regimes, depending on the dimensions of the street (Oke, 1988) (Fig. 2.2): (a) *isolated roughness* flow, (b) *wake interference* flow, and (c) *skimming* flow. For wide canyons (H/W<0.3), the buildings are well spaced and act essentially as isolated roughness elements, since the air travels a sufficient distance downwind of the first building before encountering the next obstacle. As buildings become more closely spaced (H/W $\approx$ 0.5), the disturbed air flow has insufficient distance to readjust before encountering the downwind building, resulting in wake interference flow. In the case of regular canyons (H/W $\approx$ 1), the bulk of the synoptic flow skims over the canyon producing the skimming flow, which is characterised by the formation of a single vortex within the canyon (Hunter et al., 1992).

From a three-dimensional point of view, a reflection of the wind off the windward wall of the canyon should be ideally observed in the case of skimming flow (Nakamura and Oke, 1988; Johnson and Hunter, 1999). For oblique roof-level winds, this reflection may induce a spiral wind flow through the canyon. Other complex channelling effects might be produced for winds parallel to the street axis. Additional low pressure areas and wind circulation is created near intersections, resulting in horizontal corner vortices. In relatively short canyons, corner vortices might be strong enough to inhibit a stable vortex perpendicular to the street in the mid-section. This ventilation effect fades with increasing street length (Theurer, 1999).

The strength of the wind vortices inside the canyon mainly depends on wind speed at roof-top level. However, the local wind flow is also affected by the mechanical turbulence induced by moving vehicles (Eskridge and Rao, 1986) or by urban roughness elements within the street (e.g. trees, kiosks, balconies, slanted building roofs, etc.) (Hoydysh and Dabberdt, 1994; Theurer, 1999). Furthermore, the shape and strength of the wind vortices might be also affected by the atmospheric stability and other thermal effects induced by the differential heating of the walls and/or the bottom of the canyon (Sini et al., 1996; Kim and Baik, 2001).

In relatively deep canyons (H/W>1.3), the main wind vortex is usually displaced towards the upper part of the cavity, with almost stagnant air below (DePaul and Shieh, 1986). As the aspect ratio increases (H/W $\approx$ 2), a weak counter-rotating secondary vortex may be observed at street level (Pavageau et al., 1996).

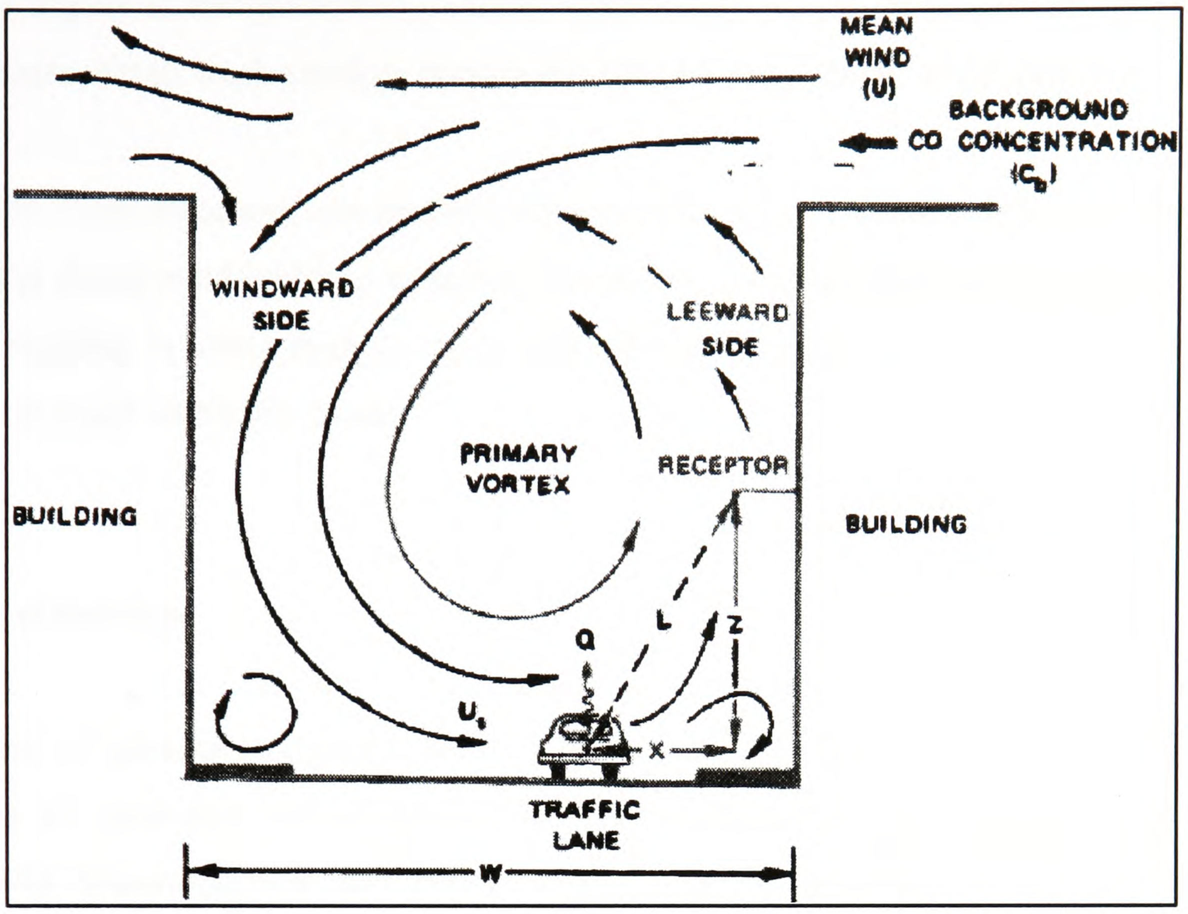


Fig. 2.1: Pollutant dispersion in a regular street canyon (Dabberdt et al., 1973).

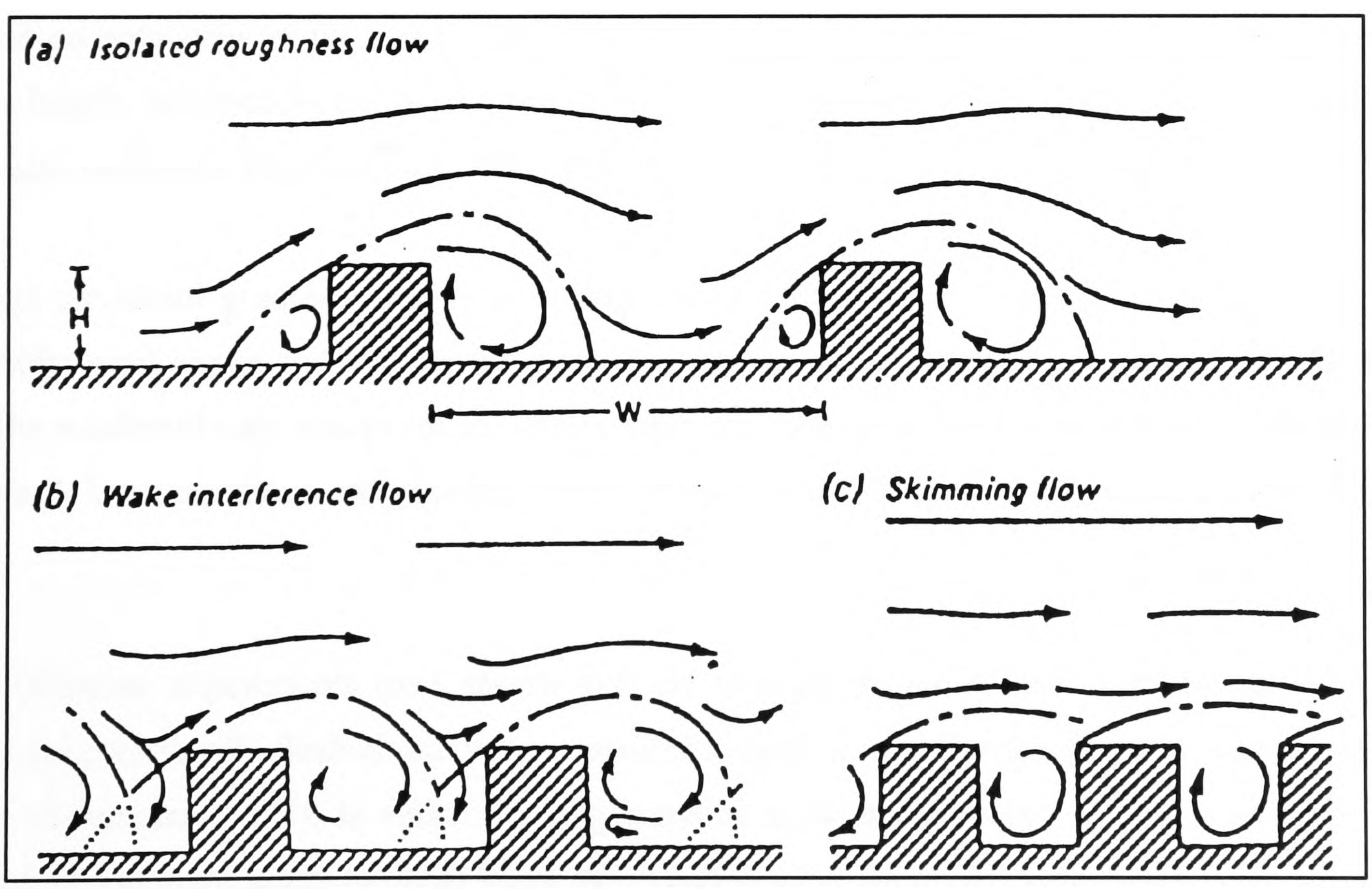


Fig. 2.2: Perpendicular flow regimes in urban canyons for different aspect ratios (Oke, 1988).

For even higher aspect ratios (H/W $\approx$ 3), a third weak vortex might be also formed (Jeong and Andrews, 2002). In most cases, small week vortices occupy the bottom side corners of the canyon.

Depending on the wind direction, asymmetric canyons may be sub-divided into two categories: (i) *step-up* canyons, when the down-wind building is higher than the up-wind building, and (ii) *step-down* canyons, when the down-wind building is lower than the up-wind building. In these cases, mid-section wind vortices might be displaced or reversed within the cavity.

2.1.3. Pollutant dispersion

The concentration of gaseous pollutants within a street canyon depends generally on the rate at which the street exchanges air vertically with the above-roof level atmosphere and laterally with connecting streets (Riain et al., 1998). Skimming flow, a feature of regular canyons, provides minimal ventilation of the canyon and is relatively ineffective in removing pollutants (Hunter et al., 1992).

Field measurements (DePaul and Sheih, 1985; Qin and Kot, 1993) show increased concentrations of traffic-related pollutants on the leeward side of the canyon, and decreasing concentrations along with height above the ground on both sides of the street. The increased leeward concentrations are due to the accumulation of pollutants locally advected by the large wind vortex that covers most of the canyon. Minor pollution hotspots might be also created in small cavities where additional recirculation phenomena can take place.

Street-level crossroad gradients observed in wind tunnel experiments (Hoydysh and Dabberdt, 1988) for perpendicular wind conditions indicate that concentrations are generally a factor of two greater for the leeward than for the windward side, except for step-down canyons where windward concentrations are slightly greater than leeward. Concentrations are generally lower in the step-up canyons relative to the even and step-down notches.

Flow visualisation experiments have shown that the strength of the canyon vortices varies. As a result, pollutants are periodically flushed out of the canyon (Pavageau et al., 1996), a phenomenon known as *canyon breathing* (Scaperdas, 2000). In relatively long canyons without connecting streets, maximum street-level concentrations are more likely to occur when the synoptic wind is parallel to the street axis. In that case, the accumulation of emissions along the line source outweighs the ventilation induced by the parallel winds (Soulhac et al., 1999; Dabberdt and Hoydysh, 1991).

Low synoptic winds create a well-known meteorological situation that favours air pollution built-up in urban areas (Qin and Kot, 1993; Vignati et al., 1996; Jones et al., 2000). There is evidence that when the synoptic wind speed is below about 1.5 m/s, the wind vortex within the canyon tends to disappear and the air stagnates in the street (DePaul and Sheih, 1986). In that case, the mechanical turbulence induced by moving vehicle as well as the atmospheric (i.e. thermal) stability conditions might play a significant role in the dispersion of traffic-generated pollutants.

Fine and especially ultrafine particles are expected to disperse in the air like gases. The larger-sized particles, however, are greatly affected by gravity and thus have a shorter residence time in the air (Chan and Kwok, 2000). For this reason, the coarse fraction of the total suspended particles (TSP) exhibits larger vertical concentration gradients than those usually observed for gases or fine particles.

2.1.4. Pollutant transformation

Due to the very short distances between sources and receptors, only very fast chemical reactions have a significant influence on the measured concentrations within street canyons (Berkowicz et al., 1997). For this reason, most traffic-related pollutants (e.g. CO and hydrocarbons) can be considered as practically inert species within these distances. This is not the case either for NO_2, which dissociates extremely fast in the presence of light, or for NO, which also reacts very fast with O_3 (Palmgren et al., 1996). Hence, the reactions of practical interest in street canyon studies are the following:

$$NO + O_3 \longrightarrow NO_2 + O_2 \qquad (2.1)$$

$$NO_2 + h\nu \longrightarrow NO + O \qquad (2.2)$$

$$O + O_2 + M \longrightarrow O_3 + M \qquad (2.3)$$

where $h\nu$ represents a photon of light, and M a molecule (usually N_2 or O_2) that carries away some of the energy released in the reaction (de Nevers, 1995). These three reactions represent a cyclic pathway driven by photons (i.e. photochemical cycle). The time scales of these photochemical reactions are of the order of tens of seconds, thus comparable with residence times of the pollutants in a street canyon.

It is expected that the relationship between relatively stable chemical species emitted by vehicles would not vary significantly within urban streets. This is very helpful for epidemiological studies, because a single or only few indicators can be identified for assessing population exposure to roadside air pollution (Kingham et al., 2000). Sillman (1999), and Jenkin and Clemitshaw (2000) have produced reviews on the formation of photochemical pollutants in larger urban and rural areas.

2.1.5. Population exposure

From a population exposure point of view, air quality in street canyons is of a major importance, since the highest pollution levels and the larger targets of impact are often concentrated in this kind of streets (Hertel et al., 2001). The so-called *canyon effect* (i.e. the reduced natural ventilation in urban streets) results in greater health impacts (e.g. indicated by an increased number of respiratory hospital admissions) and damage costs for the exposed population (Spadaro and Rabl, 2001).

Personal exposure can be calculated as the product of the pollutant concentration and time spent in a specific *microenvironment*, which is defined as a confined space (e.g. bedroom, office, car, parking, pavement, etc.) where pollutant concentrations are assumed to be uniform (Colls and Micallef, 1997). The total personal exposure will be then the sum of all such products. However, the assumption of spatial uniformity of air pollution might be erroneous for certain microenvironments like street canyons, where strong spatial concentration gradients are often observed. In these cases, exposure calculations should be refined by subdividing microenvironments into sub-microenvironments, taking into account pollution hot spots and refined human breathing zones (e.g. for residents, pedestrians, cyclists, drivers, etc.).

Relatively few examples of this approach can be found in the literature. In a study attempting to quantify residential exposure to exhaust gases in Oslo (Larssen et al., 1993), a correction coefficient was introduced to account for changes in ambient concentrations with height over street level. Furthermore, Croxford and Penn (1998) suggested that a side of the street factor should be introduced, if the prevailing wind direction is perpendicular or near-perpendicular to the street axis. Finally, other authors (Ashmore et al., 2000; Adams et al., 2001) examined the personal exposure levels in transport microenvironments and the effects of traffic management on them.

2.2. Air quality monitoring

2.2.1. Monitoring techniques

Air quality monitoring methods can be broadly divided into two different categories: (a) *line measurements*, when measurements are performed along an optical path, and (b) *point measurements*, when measurements are carried out by taking samples at one spot.

The most commonly used line measurement method is the Differential Optical Absorption Spectroscopy (DOAS), which is an open path optical monitoring technique based on the differential absorption of ultraviolet or visible light (Platt and Perner, 1983). Another line system is the Differential Absorption Lidar (DIAL), which directs pulses from a tuneable dye laser into the air, and measures the back-scattered signals with a detector. These methods are useful for monitoring background pollution at certain height above the ground.

Point measurement techniques, which are adequate for both roadside and background air quality monitoring, can be classified into different categories according to the type of pollutant (i.e. gaseous or particulate) and the physical principle of detection. In the present study, only point measurement techniques were used.

Gaseous pollutants

For gaseous pollutants, available point measurement methods can be subdivided into three categories: (I) *continuous monitoring*, (II) *pre-concentration* and (III) *grab sampling*.

(I) Continuous monitoring is a technique implying the use of a pump for drawing continuously air samples and delivering them to a gas analyser. CO infrared analysers, NO_x chemiluminescence analysers, and O_3 ultraviolet analysers are commonly used.

(II) Pre-concentration techniques capture the pollutant (or a chemical derived from it) from the sampled air for later quantitative analysis in the laboratory by standard methods (Colls, 1997). There are (a) passive and (b) active pre-concentration techniques.

(a) Passive (or *diffusive*) sampling relies on the diffusion of gas molecules down a concentration gradient without pumping. The gas molecules are eventually captured on an adsorbent (e.g. activated charcoal). This method is commonly used for measuring NO_2 and volatile organic compounds (VOC). After removal from the passive device (usually a tube) with thermal or solvent desorption, the samples can be analysed using gas (GC) or high performance liquid chromatography (HPLC) together with an adequate detector (e.g. a flame ionisation detector or "FID").

(b) Active (or *pumped*) sampling is a pre-concentration technique that uses a pump to suck air samples through an adsorbing material, which can then be analysed using the same methods as for passive sampling.

(III) Grab (or *whole air*) sampling techniques capture a sample of the air itself using an appropriate device (e.g. canister, syringe, bag, etc.) and take it to the laboratory for analysis. Canisters (i.e. evacuated stainless steel bottles that are opened in the ambient air and filled with the sample) are widely used for hydrocarbon measurements. They are especially useful for measuring light hydrocarbons (e.g. butadiene).

Particulate pollutants

The main purpose of particulate sampling is to obtain mass concentration and chemical composition data, preferably as a function of particle diameter. The principal methods for extracting particles from an air stream are filtration and impaction (Boubel et al., 1994).

Mass (i.e. gravimetric) measurements are usually made by pre- and post-weighing adequate filters or impaction surfaces. The size distribution may be determined by classifying atmospheric aerosols[1] by aerodynamic diameter (e.g. using cascade impactors), electrical mobility (e.g. using differential mobility analysers) or light scattering properties (e.g. using optical particle counters). Furthermore, the Tapered Element Oscillating Microbalance (TEOM) system is often used for real time PM_{10} monitoring.

Finally, the chemical composition of aerosols, which is useful in determining their sources and fate in the atmosphere, can be determined using direct elemental analysis techniques (e.g. x-ray fluorescence spectroscopy or neutron activation analysis), atomic absorption spectroscopy for heavy metals, and ion chromatography (McMurry, 2000; Winegar and Keith, 1993).

[1] An aerosol is defined as a suspension of liquid or solid particles in gas (McMurry, 2000)

2.2.2. Relationship between pollutants

It is very helpful for epidemiological studies when a single indicator (or *tracer*) can be identified for air pollution, because this can then be used to indicate general levels of population exposure in urban areas (Kingham et al., 2000).

Given the practical advantages and constraints of different air quality monitoring techniques, it may be more convenient to identify a set of possible pollution indicators, each one of them meeting a particular need. The chosen compounds should come from the same sources (e.g. road traffic) and have the same fate with the group of pollutants they are intended to represent. This can be checked by estimating the strength of correlation between any possible indicator with a number of other pollutants sampled in a variety of locations.

Two commonly used indicators for traffic-related pollution are the CO and the benzene (Mukherjee and Viswanathan, 2001). This is because they are mainly of vehicular origin and are practically inert within urban streets.

2.2.3. Spatial variability of air pollution

Diffusive NO_2 sampling has been often used to establish the spatial variability of air pollution in urban areas (Laxen and Noordally, 1987; Hewitt, 1991; Monn et al., 1997). A criticism of this might be that this compound, although easily monitored using passive tubes, is not the best indicator for traffic pollution. This is because NO_2 only represents a small fraction (less than 10%) of the total NO_x directly emitted from motor vehicles. In addition to that, it is highly reactive within very short transport distances and therefore it is not expected to correlate strongly with other more conservative traffic-related pollutants.

For the reasons explained in the previous paragraph, CO and benzene are more suitable for revealing the spatial gradients of urban air pollution. CO has been used as a tracer to indicate large differences in air pollution levels between neighbouring streets in central London (Croxford et al., 1996). Finally, diffusive benzene and aldehyde sampling has been used to identify strong concentration gradients within short distances in two medium size French cities (Gonzalez-Flesca et al., 1999; 2000).

2.2.4. Temporal variability of air pollution

While diffusing sampling may be seen as an efficient technique for describing the spatial variability of urban air pollution, continuous gas analysers (e.g. for CO, NO_x, and O_3) can provide reliable short-term (e.g. hourly) average concentrations at a limited number of monitoring locations within a city.

Adequately located gas analysers can capture the diurnal fluctuation pattern of air pollution within a street. This fluctuation that might be due to variable vehicle traffic and atmospheric dispersion conditions cannot be reflected on diffusive sampling measurements, which only provide longer term (e.g. daily or weekly) averages.

Besides the traditional gas analysers (briefly described in Section 3.1.2), there are alternative air quality monitoring instruments available, like the recently commercialised STREETBOX (Croxford and Penn, 1998). This system appears to combine the advantages of passive samplers (e.g. small size, portability) and continuous analysers (e.g. short averaging times). Nevertheless, it needs to be further validated before being widely used.

2.2.5. Response time

The response time, which is the time over which the sample is taken, is one of the major factors that will determine the suitability of a sampling method. Standard gas analysers are sufficiently sensitive and fast to give real time (i.e. typical response time: 1-2 min) measurements of CO, NO_x and O_3 concentrations. The results can be then averaged over a short time period (e.g. 1-8 hours) and be compared to the regulatory standards.

Diffusive samplers have a relatively long response time (i.e. typically from one/two days to four weeks), which makes them less suitable for observing atmospheric pollutants responsible for short-term health effects. On the other hand, long response times might be preferable when sampling substances like benzene, whose health effects are due to cumulative exposure. In these cases, peak concentrations are of minor concern and therefore diffusive samplers appear to be the ideal choice (Brown et al., 1999; Cocheo et al., 2000; Skov et al., 2001).

Furthermore, diffusive samplers are portable devices and do not need electrical power supply, which makes them very suitable for spatial distribution measurements (including vertical distributions within canyons), air quality mapping, human exposure studies, and detection of long-term pollution trends.

2.2.6. Siting considerations

The total number of air quality monitoring stations or sampling locations within a city is limited by practical constraints. Since pollutant concentrations might vary with a factor of 5 from a street canyon to an urban background area (Palmgren and Kemp, 1999), the selection of monitoring/sampling locations becomes fundamental.

Permanent air quality stations within a city may be classified into two broad categories: (I) the traffic-oriented (*roadside* or *kerbside*) and (II) the urban background stations. Roadside stations are usually located on the pavement of busy streets, avenues or intersections, within few meters distance from the roadway and with their sampling head at 1.5 - 3 m height above ground. On the other hand, background stations are placed in parks or other urban locations away from road traffic.

Monitoring stations and/or sampling points directed at the protection of human health should be located near places of expected air pollution hotspots, but also must be reasonable with respect to population exposure over the averaging times associated with the regulatory values. Sampling locations should be adequately selected so as to avoid measuring very small microenvironments. As a guideline, a sampling point should be representative of air quality levels in a surrounding area of no less than 200 m^2 at traffic-oriented sites and of several square km^2 at urban background sites (European Commission, 2000). Furthermore, sampling sites should be representative of similar locations not in their immediate vicinity.

As far as the microscale siting of the samplers is concerned, operators need to make sure that there are no physical obstructions (buildings, balconies, trees, etc.) affecting the airflow around the sampling inlet. Furthermore, the sampling should not be carried out in the immediate vicinity of sources in order to avoid direct intake of undiluted exhaust emissions. Other factors that need to be taken into account are the access and security of equipment, the safety of public and operators, the co-location of sampling points for different pollutants, the planning requirements, etc.

Finally, one should be cautious when comparing monitoring data (i.e. absolute values) from different cities. The data are often based on one or few monitoring stations placed at critical sites and thus represent microenvironments rather than large urban areas (Fenger, 1999).

2.3. Air Quality Modelling

2.3.1. Classification of air quality models

There is a plethora of air quality models developed to meet the needs of a variety of end-users. Dispersion models are widely used for assessing roadside air quality by providing predictions of present and future air pollution levels as well as temporal and spatial variations (Sharma and Khare, 2001). More specifically, they find application in air quality and traffic management, urban planning, interpretation of monitoring data, pollution forecasting, population exposure studies, etc.

Although there are no clear-cut distinctions between the different types of air quality models, several authors (Zannetti, 1990; Moussiopoulos et al., 1996; Scaperdas, 2000) have attempted to classify them according to the spatial scale (i.e. from local to global), the physical and mathematical principles (i.e. statistical, box, Gaussian, CFD, etc.), the level of complexity (i.e. empirical, semi-empirical, numerical, etc.), and the scope (i.e. policy, research, etc.). Brief definitions of the most commonly used types of models are given below:

- **Parametric (or *operational* models)**: These are mathematical models that express pollutant concentration as a function of a set of variable parameters, conditions and empirically derived constants.

- **Empirical models**: Mathematical models mainly derived from statistical analysis of field monitoring or laboratory data.

- **Statistical models**: They are based on statistical techniques (e.g. regression, frequency distribution, etc.) for analysing trends and relationships between air quality and meteorological data in order to forecast pollution episodes. They are intrinsically limited since they do not establish cause-effect relationships. They can be useful in short-term forecasting.

- **Receptor models**: They consider the observed concentrations at a receptor point and attempt to apportion the contributions from various sources.

- **Semi-empirical models**: This category consists of several types of mathematical models (e.g. Gaussian plume, box models, etc.) based on a combination of theoretical analysis and empirical parameterisation. There is no clear distinction between empirical and semi-empirical models.

- **Gaussian models**: Their main assumption is that the concentration of the plume follows a Gaussian distribution in both the horizontal and vertical directions. These are the most common air pollution models.

- **Box models**: They assume mass conservation of the pollutant and uniform mixing throughout the volume of a three-dimensional Eulerian box, which might represent a whole city or just a street canyon. This simple modelling approach can be useful for a first approximation.

- **Screening models**: Simple (i.e. empirical or semi-empirical) models enabling a quick *screening* of likely air pollutant concentrations. They require a small amount of input information and usually assume average meteorological conditions.

- **Numerical (or *computational*) models**: These are advanced mathematical models that solve the governing flow and dispersion equations numerically for given boundary conditions, using either Eulerian or Lagrangian approaches.

- **Eulerian models**: They solve numerically (or analytically under special, simplifying assumptions) the atmospheric diffusion equation using a fixed reference system. The computational domain is divided in a number of *boxes*.

- **Lagrangian models**: As an alternative to Eulerian approach, these models describe fluid elements (called *puffs*, *parcels* or *particles*) that follow the instantaneous flow. Particle motion can be simulated using both deterministic and statistical velocities. The Lagrangian reference system follows the average atmospheric motion.

- **Computational Fluid Dynamic (CFD) models**: Advanced Eulerian models able to deal with complex boundary conditions using fine-scale grids.

- **Reduced-scale models**: They are also called *physical* models, in contrast with all other *mathematical* models. They are based on the principle that by reducing the geometrical scale of a given flow domain and adjusting the reference parameters (e.g. flow velocity), the original full-scale conditions can be reproduced experimentally. The most commonly used technique is wind tunnel modelling.

Some of these (often overlapping) categories and corresponding models are presented in Table 2.1. It should be remembered that these are only basic model types. Models belonging to one (or more) of the above categories can also include chemical transformation, plume rise, dry and wet deposition or other sub-models.

Finally, air pollution models can be also classified with respect to their spatial scale in one of the following broad categories: macroscale models (length scale exceeding 1000 km), mesoscale models (length scale between 1 and 1000 km), microscale models (length scale below 1 km). The following sections will present microscale models applicable to pollutant dispersion within street canyons.

2.3.2. Street canyon models

Gaussian plume models

These are sets of equations describing the three-dimensional concentration field generated usually by a point source. They assume that the concentrations from a continuously emitting source are proportional to the emission rate, inversely proportional to the wind speed, and that the time averaged pollutant concentrations horizontally and vertically are well described by Gaussian (i.e. bell-shaped) distributions (Boubel et al., 1994). In its simplest form, the Gaussian plume model assumes that there are no chemical or removal processes taking place and that pollutant material reaching the ground or the top of the mixing layer as the plume grows is reflected back towards the plume centreline.

Gaussian plume models rely on the appropriate selection of the plume spread sigma functions (in both the horizontal and vertical sense), which are generally expressed in terms of Pasquill atmospheric stability classes or Monin-Obukhov similarity theory parameters (Zannetti, 1990). Models using the latter approach dispose of height dependent sigma functions and are known as second generation Gaussian plume models (Carruthers et al., 1994).

Apart from industrial applications (i.e. point sources), specially designed Gaussian plume models can be used to calculate pollutant concentrations over urban agglomerations (i.e. area sources) and in the vicinity of highways (i.e. line sources). Gaussian models are not directly applicable to small-scale dispersion within the urban canopy, since they treat buildings and other obstacles only via a surface roughness parameterisation (Scaperdas, 2000). Nevertheless, in some cases, they include specialised modules for street canyons. This is the case of ADMS-Urban (Owen et al., 1999), a second generation urban-scale dispersion model that includes a street canyon module nested within the core Gaussian code.

Table 2.1: Classification of commonly used dispersion models.

Parametric						Numerical		
Empirical		Semi-empirical				Eulerian		Lagrangian
Statistical	Receptor	Screening	Box	Street canyon	Gaussian	Microscale (CFD)	Urban scale	Stochastic particle
Derwent and	COPREM	CAR International	STREET-SRI	CPBM	ADMS-Urban	PHOENICS	MEMO	GEM
Middleton (1996)	Karim and	AEOLUS Screen	MAPS	OSPM	INDIC AIRVIRO	FLUENT	METRAS	Addison et al. (2000)
Stedman et al. (2001)	Ohno (2000)	AEOLIUSQ	STREET BOX	AEOLIUS	TNO-Traffic	STAR-CD	CALGRID	Jicha et al. (2000)
		STREET	Nicholson (1975)	SLAQ	CAR-FMI	CFX-TASCflow		Xia and Leung
		UK DMRB		OMG	PROKAS-V	PANACHE		(2001a,b)
				Hotchkiss and	HIWAY-2	MERCURE		
				Harlow (1973)	CALINE4	CHENSI		
					APRAC	MISKAM		
					PUFFER	MIMO		
						MITRAS		
						ADREA-HF		
						FloVENT		

CALINE4

CALINE4, the latest version of the CALINE series of pollutant dispersion models, is one of the most validated models available for assessing the impact of vehicle traffic on roadside air quality (Benson, 1984). It has been widely used in scientific and engineering applications mainly concerning highway development and management (Jones et al, 2000).

The model uses Gaussian plume theory to simulate the dispersion of pollutants emitted from a line source. This is divided in a series of elements, which are modelled as equivalent finite line sources located normal to the wind direction. The region directly over the road, called the *mixing zone*, is treated as a zone of uniform emission and turbulence. Within the mixing zone, vehicle induced turbulence (both mechanical and thermal) is taken into account (Benson, 1992).

CALINE4 includes street canyon, bridge, intersection, and parking lot modules. Under the *depressed section* mode, the model is able to calculate pollutant concentrations in urban canyons. The street canyon algorithm devised by Turner (1970) computes the effect of single or multiple horizontal reflections of the plume on the walls of the canyon. In this case, the road axis and the wind velocity are assumed to be parallel to the horizontal topographic boundary (i.e. the walls of the canyon), resulting in equal concentrations on both sides of the street.

TNO and CAR

TNO-Traffic is a Gaussian plume-type model that describes the dispersion of traffic exhausts (Eerens et al., 1993). It is based on an extensive programme of wind tunnel experiments which covered a great number of different street configurations, including urban canyons and intersections (van den Hout et al., 1994). In this model, the traffic is represented by line sources divided into series of small point sources.

CAR (or CAR International) is a simplified version of the same model, in which only the most representative street configurations were categorised (den Boeft et al., 1996). For each street type (e.g. highway, urban road, etc.) a source-receptor relationship is specified as a function of the distance between the receptor and the street axis. CAR uses annual average wind speeds and assumes that there is no prevailing wind direction. Thus, the user obtains the same yearly averages and percentiles on either side of the street. In all cases, the effect of trees and moving vehicles on street-level wind velocity is taken into account. A recent extension of CAR is the CARSMOG model, which calculates hourly roadside concentrations of traffic-related pollutants. CAR model versions should not be confused with CAR-FMI (Härkönen et al., 1995), which is a Gaussian line source model for calculating pollution from road networks.

STREET-SRI

Johnson et al. (1973) used a single box model, together with some simplified assumptions concerning initial dispersion and car induced turbulence, to derive a street canyon sub-model usually called STREET or SRI (i.e. Stanford Research Institute), which formed part of a multipurpose urban diffusion model for inert pollutants (APRAC). It is based on the assumption that concentrations of the pollutant occurring on the roadside consist of two components, the urban background concentration and the concentration component due to vehicle emissions generated within the specific street. Then, it calculates pollutant concentrations on both sides of the street, taking into account the height and distance of the simulated receptor from the kerb.

On the leeward side of the canyon, concentrations are assumed to be inversely proportional to the distance between the line source and the receptor point. On the windward side, the vertical decrease of concentrations due to entrainment of fresh air through the top of the canyon is taken into account (see mathematical description in Section 4.11). For parallel or near-parallel synoptic winds, the average of the leeward and windward values may give the pollutant concentration on both sides of the street, although the model is not specifically designed for this situation. STREET-SRI was parameterised using data from a regular street canyon and for this reason it might need re-calibration before being applied to other canyon geometries.

CPBM

The Canyon Plume Box Model (Yamartino and Wiegand, 1986) combines a Gaussian plume model for the direct impact of pollutants emitted in the street, with a box model that accounts for the additional impact of pollutants trapped within the wind vortex formed inside the canyon. The wind flow in the canyon is reproduced using the methodology proposed by Hotchkiss and Harlow (1973) for the two transverse components of the wind velocity and a logarithmic expression for the longitudinal component. An empirical model that takes into account wind generated turbulence as well as thermal effects induced by solar radiation and moving vehicles is used to calculate the turbulent sigma parameters representing the standard deviation of flow velocities about the mean flow.

The plume generated inside the canyon is divided into three segments, which are assumed to follow straight line trajectories and disperse according to Gaussian plume formulae. The impact resulting from the recirculation component is calculated from the consideration of the mass budget inside the canyon. On the leeward side of the street, the total impact is calculated by adding the direct plume to the recirculated fraction. On the windward side, where the only contribution arises from the recirculation component, the dilution of the concentrations due to the entrainment of fresh air is also taken into account. For winds parallel to the street axis or for very low wind speeds, a simpler plume model is used.

OSPM and AEOLIUS

AEOLIUS (Buckland, 1998) is based on concepts and techniques previously used for the development of the Operational Street Pollution Model (Hertel and Berkowicz, 1989a), which was evolved from the CPBM. AEOLIUS and OSPM are semi-empirical models that calculate concentrations of exhaust gases on both sides of a canyon assuming three different contributions: (a) the contribution from the direct flow of pollutants from the source to the receptor, (b) the recirculation component due to the flow of pollutants around the vortex generated within the recirculation zone of the canyon, and (c) the urban background contribution. A Gaussian plume algorithm is used for the calculation of the direct contribution and a simple box model for deriving the recirculation component (see mathematical description in Section 4.11).

The vortex is formed inside the canyon, if the synoptic wind is not parallel to the street axis. The length of the vortex (along the wind direction) is 2 times the upwind building height. For synoptic winds below 2 m/s, the length of the vortex decreases with the wind speed (Berkowicz, 2000a). The width of the recirculation zone cannot exceed the width of the canyon in any case. The relation between street- and roof-level winds in the canyon is given by a logarithmic relationship that takes into account the surface roughness length, the height of initial dispersion of car exhausts and the synoptic wind direction. Finally, the mechanical turbulence in the street due to the wind and vehicle traffic is empirically derived.

AEOLIUS (the Full version) is based on the same formulation as OSPM. Nevertheless, some discrepancies between predictions from the two models cannot be excluded, due to differences in coding, parameterisation and data pre-processing techniques. There are also two screening versions of AEOLIUS, namely AEOLIUS Screen and AEOLIUSQ Emission, made available by the U.K Met Office.

2.3.3. Receptor models

The models described in the previous sections may be also defined as *source-oriented* models. Such models rely on the use of best available emission estimates and meteorological data to predict pollutant concentrations at various roadside locations. An alternative approach is the *receptor-oriented* modelling, which is based on the detailed analysis of the pollutant collected at one or more monitoring sites. This analysis, also called *source apportionment* or *chemical mass balance* (Gordon et al., 1984), attempts to determine which sources contributed to the concentration measured at the receptor point. If the pollutant of interest is chemically inert (e.g. CO), there is no way to distinguish between different sources. But if the pollutant consists of a variety of chemical species (e.g. particulate matter), then from its chemical composition one can make inferences about the sources.

Receptor-oriented models, such as the Constrained Physical Receptor Model (CPRM) (Wåhlin et al., 2001), are mostly used to test the predictions made by source-oriented models as well as the accuracy of the emission estimates that are used in them (Karim and Ohno, 2000).

2.3.4. Computational Fluid Dynamic models

Computational Fluid Dynamics (CFD) modelling is a general term used to describe the analysis of systems involving fluid flow, heat transfer and associated phenomena (e.g. chemical reactions) by means of computer-based numerical methods that solve the fundamental equations of fluid motion. It is a powerful modelling technique spanning a wide range of industrial and more recently environmental and biomedical applications (Gosman, 1999).

What distinguishes CFD from other Eulerian models is their capability to deal with complex shaped walls and other boundary conditions (e.g. in aircraft and automobile design) using flexible fine-scale grids. Furthermore, they usually include advanced turbulence treatment schemes, which makes them suitable for small-scale pollutant dispersion applications.

CFD codes are structured around numerical algorithms that can tackle fluid flow problems. In order to provide easy user access, most commercial CFD packages include user-friendly input and output interfaces. Hence, they contain three main elements: (I) The pre-processor, which serves to input problem parameters, generate the grid of the computational domain, select the physical and chemical phenomena that need to be treated, define the fluid properties, and finally specify the appropriate boundary conditions. (II) The solver, which first approximates numerically the unknown flow variables, then discretises the governing flow equations using these approximations, and finally solves the resulting system of algebraic equations. (III) The post-processor, which displays the grid and geometry of the domain, plots vectors (e.g. wind velocity) and contours (e.g. pollutant concentration) and may even provide animation facilities for dynamic result display.

Physical principles

CFD modelling is based on the numerical solution of the governing fluid flow and dispersion equations, which are derived from basic conservation and transport principles: (a) the mass conservation (or *continuity*) equation, (b) the three momentum conservation (or *Navier-Stokes*) equations in x, y, z, and (c) the transport equation for pollutant concentration. The equations of state (obtained through the thermodynamic equilibrium assumption) and the Newtonian model of viscous stresses are also enlisted to close the system numerically. The initial and boundary conditions have to be specified by the user.

Furthermore, atmospheric turbulent processes need to be modelled. Existing turbulence models can be classified in two broad categories: (I) The classical models based on Reynolds Averaged Navier-Stokes (RANS) flow equations (e.g. the k-ε model, which is by far the most used and validated); (II) the Large Eddy Simulation (LES) models, which are computationally very demanding and therefore mainly used in research applications (Versteeg and Malalasekera, 1995).

Numerical principles

There are three different streams of numerical solution techniques: (a) finite difference, (b) fine element, and (c) spectral methods. The main differences between them are associated with the way in which the flow variables are approximated and with the discretisation processes. The finite volume method, which was originally developed as a special finite difference formulation, is now the most well established and thoroughly validated method (it is central to most popular CFD codes: PHOENICS, FLUENT, STAR-CD).

According to this method, the flow domain is divided into individual finite control volumes (or *computational cells*). The differential flow equations are then integrated over each cell in order to transform them into a set of approximated algebraic difference equations between all nodal points of the grid. An advantage of the finite volume method is that mass and momentum conservation is imposed at cell level, which ensures that the discretised form of the flow equations integrated over the entire domain is also conservative.

An iterative approach is required for solving the system of algebraic difference equations resulting from the discretisation method. The most popular solution procedures are the TDMA, a line-by-line solver of the algebraic equations, and the SIMPLE algorithm. The Semi-Implicit Method for Pressure Linked Equations (SIMPLE), originally proposed by Patankar and Spalding (1972), is a predictor-corrector method. That means that velocities are predicted by solving the momentum conservation equations using the most recent estimate of the pressure field, and then the pressure field is corrected by using the imbalances in the mass conservation equations. The other conservation equations are then solved, and the procedure is iterated until reaching convergence (i.e. when the imbalance in all conservation equations reaches a sufficient low value).

Applications and codes

When pollutant dispersion is examined within a street canyon, the computational domain should be sufficiently extended to stabilise the air inflow and outflow through the geometrical boundaries of the area.

The relief of the buildings (e.g. due to the presence of balconies) or the street (e.g. due to the presence of vegetation, parked cars, etc.) can be taken into account by introducing a roughness coefficient for each surface of the domain. Alternatively, a volume resistance (i.e. in the form of a porous medium) can be assigned to represent tree foliage.

The space discretisation is usually not uniform, since a higher resolution is required near the canyon walls and the roadway. Finally, fields of pollutant concentrations, wind velocity and other physical quantities (e.g. turbulent kinetic energy and eddy diffusivity) may be reproduced.

The commercially available general-purpose CFD codes PHOENICS, FLUENT, STAR-CD, CFX-TASCflow and Fluidyn-PANACHE have been used in a number of street canyon applications. Other numerical models like MERCURE (Carissimo et al., 1995), CHENSI (Levi Alvares and Sini, 1992) and MISKAM (Eichhorn, 1995) were specially designed to simulate pollutant dispersion at local scale.

MISKAM was used to create a database of numerical three-dimensional simulations that was integrated in a screening model called STREET (Petit et al., 2000). Furthermore, the street canyon module PROKAS-B, which forms part of the Gaussian urban scale model PROKAS-V, was also based on dimensionless concentrations calculated using a version of MISKAM. Finally, the microscale models MIMO and MITRAS were also specially designed for street canyon applications and nested within the mesoscale MEMO and METRAS, respectively (Ehrhard et al., 2000).

2.3.5. Reduced-scale models

The reduced-scale (or *physical*) models are based on the principles of similarity, which means that by reducing the geometrical scale of a given flow domain and adjusting the reference parameters (e.g. flow velocity), the original full-scale conditions can be reproduced. Reduced-scale modelling can be carried out in a wind tunnel or a water tank facility. Although wind tunnels have been more widely used for simulating pollutant dispersion than water tanks, the same principles and considerations apply to both the methods.

Similarity is usually expressed in the form of non-dimensional quantities with a physical meaning, such as the Reynolds number and the Froude number. The Reynolds number represents the ratio of inertia/viscous forces in the fluid and is responsible for turbulence similarity, while the Froude number is responsible for buoyant convection similarity. Other quantities representing species diffusion may also be important. It should be remembered that it is not generally possible to satisfy all these numbers when scaling down from a full size street to a wind tunnel model.

Three monitoring techniques are usually involved in wind tunnel experiments: (a) flow visualisation, which helps to explore the range of possible flow and dispersion patterns obtained for different building arrangements, (b) tracer dispersion, which is used to quantify concentrations at receptor locations within the canyon, and (c) Laser Doppler Anemometry (LDA), which is used to study in more detail the patterns observed during flow visualisation experiments. Finally, Particle Image Velocimetry (PIV) is a valuable new experimental technique being used in flow measurements (Vinçont et al., 2000).

Despite the scaling difficulties, wind tunnel modelling can efficiently approximate real atmospheric conditions in urban streets. Furthermore, it allows isolating and studying separately each one of the phenomena involved in microscale pollutant dispersion. Reduced-scale modelling has often been used as a complementary tool to numerical modelling and been proved especially useful in model development and validation (Baker and Hargreaves, 2001). Nevertheless, differences between wind tunnel and full-scale experimental data should be carefully considered when validating numerical models (Schatzmann et al., 1999).

2.3.6. Model features and requirements

Dispersion model predictions are in most cases a function of meteorology, street geometry, receptor location, traffic volumes and emission factors. The acquisition and pre-processing of these data is an important part of any modelling study, since the performance of a model greatly depends on the quality of the inputs.

Traffic data

Detailed traffic information, including traffic volumes, fleet composition (e.g. ratio of light/heavy duty vehicles) and average vehicle speeds, is normally required for running street canyon models. Part of this information (e.g. traffic volumes and average vehicle speeds) might be obtained from automatic detectors permanently or temporarily operating in the street of interest. The vehicle fleet composition, however, is rarely available for a specific location and time period and for this reason has often to be estimated from on site spot measurements. At least few manual traffic counts should be always taken to assure the quality of data obtained from automatic traffic networks.

Emissions

All street canyon models require vehicle emission factors (e.g. g/km per single vehicle) or emission rates (e.g. g/km per hour) as input, although some operational models (e.g. CAR International) might include default national emission factors for certain countries. In certain models (e.g. AEOLIUS), separate emission factors for small and large vehicles need to be specified. The emission rates in a street can be derived from the traffic volumes and the composite emission factors of the pollutants. A number of methodologies and models may be applied to determine the appropriate fleet-average emission factors.

The CORINAIR working group (sponsored by the European Commission) developed a methodology for calculating emissions, including appropriate emission factors, from road traffic (Eggleston et al., 1993). The methodology was transformed into the computer program COPERT (Ntziachristos and Samaras, 1997; 2000). The same methodology was adopted by ADEME (1998) to develop IMPACT, a road traffic emission model which quantifies fuel consumption and atmospheric releases of a specified vehicle fleet in a given year in France. Emissions are calculated for two vehicle operating modes: hot and cold start. The required input parameters are traffic composition, average vehicle speed, length and slope of the road segment of interest. In addition, the month of the year is used to estimate average ambient temperatures, which are further used for calculating evaporative and cold running emissions. The model provides default values for the average travelling distance and the fraction of this distance run with a cold engine in France.

The U.S. EPA has developed and regularly revised MOBILE, which is also a mobile source emission factor model. MOBILE distinguishes moving vehicles into three operating modes: cold start, hot stabilised and hot start. The model inputs include the vehicle miles travelled by each specified type of vehicle, ambient temperature, terrain altitude, calendar year, average vehicle speed, etc. Default values applying to the U.S. vehicle fleet are provided within the model. A related EPA model, PART5, can be used to calculate emission factors for particulate matter. In California, the Air Resources Board's EMFAC model is used in place of MOBILE.

An important aspect that differentiates MOBILE from COPERT is that the latter bases its assumptions on fuel sale statistics, while MOBILE assesses annual mileage accumulation rates by using data from traffic surveys. Other significant differences lie in the way the two models account for the effect of low ambient temperatures and cold start emissions (Zachariadis and Samaras, 1999). A detailed comparative analysis of MOBILE5a and COPERT was presented by Samaras and Zachariadis (1994).

The German MOBILEV emission model can also be used to calculate yearly or hourly average emissions for a single street or a street network using available information on emission factors, traffic mode, street characteristics and vehicle fleet composition. Congested traffic and cold start emissions are also taken into account.

Casella Stanger in association with AEA Technology developed a spreadsheet model for NO_x and PM_{10} emission factors on behalf of the U.K. Department of the Environment (DEFRA), to assist local authorities in the air quality review and assessment process. Furthermore, the protocol used by Buckland and Middleton (1999) can be applied for estimating composite emission factors for most regulated pollutants in the U.K. This methodology is based on predefined emission factors specific to each vehicle category. The fleet composition is then used to derive a composite emission factor for the road segment of interest. Finally, inverse modelling can be applied to estimate actual fleet emissions from roadside measurements using an operational street canyon model (Palmgren et al., 1999).

Although not exhaustive, the above discussion gives an idea of the existing emission calculation methodologies and available models. It should be stressed that emission factors have to be regularly updated to reflect changes in fuel standards, vehicle fleet composition and engine technology (Stedman et al., 2001). It is generally recognised that emission factors represent one of the most important sources of uncertainty in modelling traffic pollution (Kühlwein and Friedrich, 2000).

Meteorological data

The amount of required meteorological information for air quality modelling is proportional to the complexity of the selected model. Simple models for screening applications (e.g. CAR and AEOLIUS Screen) only require the average wind speed over a period of time, assuming that there is no prevailing wind direction. Relatively more complex street canyon models (e.g. OSPM and AEOLIUS Full) require time series of wind speed and direction for the dispersion calculations, ambient temperature and global radiation for the photochemistry algorithm, and (in some cases) atmospheric pressure for unit conversion. In addition to this information, CFD codes (e.g. PHOENICS) require certain specifications concerning atmospheric turbulence and wind profiles. Finally, the atmospheric stability and mixing height need to be specified in order to run Gaussian plume models (e.g. CALINE4).

It should be remembered that meteorological data obtained simultaneously at different weather stations located within few kilometre distances from each other might differ significantly, especially for short averaging periods. Recent research studies have shown that model simulations carried out using airport winds generally produce lower and less accurate air quality predictions compared to those produced using local wind data (Manning et al., 2000).

Street geometry

Again, more advanced models require a larger amount of input information. Simple models may only need the height and the width of the canyon as input (e.g. AEOLIUS Screen), or just the type of the street and the distance between simulated receptor and road axis (e.g. CAR). Semi-empirical models (e.g. OSPM) may additionally require the length and the orientation of the canyon and allow for some gaps between the buildings. A surface roughness coefficient might be also provided by the user. Although there are both experimental and theoretical methods for estimating the roughness length of an urban surface (Pal Arya S., 1988), arbitrary values (≈ 0.6 m) are often used.

Relatively simple mathematical models are generally not able to capture the details of the urban canopy (e.g. trees, slanted building roofs, balconies, parked cars, etc.), which might have a significant influence in small-scale pollutant dispersion within street canyons (Gayev and Savory, 1999; Rafailidis, 2000). By contrast, CFD models are able to closely reproduce the details of the urban canopy, if the necessary input information is available. Four main types of boundary conditions imposed to the physical limits of the simulated area have to be specified: (a) the walls, (b) the inlets, (c) the outlets, and (d) the planes of symmetry (i.e normal velocities are set to zero at a symmetry boundary and the values of all other properties just outside the solution domain are equal to their values just inside the domain). Most commercial CFD codes provide the necessary graphical and numerical tools for treating complex street configurations, including fixed and moving obstacles (Theurer, 1999; Venetsanos et al., 2001).

Background concentrations

Even the simplest urban canyon models require a background pollution value as input, to account for the fraction of the pollutant that is not emitted within the simulated street. The urban background concentration can be defined in several ways. Ott and Eliassen (1973) suggested the existence of an urban CO background as a relatively constant concentration that would be observed at a number of locations throughout the city, providing that the observer was at least 200 feet from the nearest street. Other authors have suggested the use of roof-top measurements as an estimate of the urban background levels. An alternative approach is to simulate the entire urban area using a larger scale model in order to determine background levels contributed by non-localised sources. The main disadvantage of this method is that it requires additional input data, which are subject to uncertainties (Cooper, 1987).

Berkowicz (2000b) developed a simple model for urban background pollution that can be used in combination with OSPM. ApSimon et al. (2001) adopted ADMS-Urban within the integrated assessment model USIAM to define contributions from different sources at different background receptor locations. CAR includes a simple algorithm for deriving urban background using the regional background (i.e. background due to distant sources) and the diameter of the built-up city area. Nevertheless, the commonest (and more reliable) modelling practice is to use background concentrations obtained from measurements at urban locations that are not directly affected by local sources.

Deposition and resuspension

The mass of TSP and PM_{10}, usually measured in fixed monitoring stations, is dominated by the coarser fraction of airborne particulate matter. The PM_{10} fraction may be transported over long distances (Vignati et al., 1999). It is unlikely for an all-purpose street canyon model to be able to reproduce atmospheric aerosol concentrations measured on the kerbside, unless long range transport, local and regional non-traffic sources, relative humidity, deposition and resuspension processes are adequately taken into consideration.

The number of measured particles is dominated by ultrafine particles (i.e. the smaller fraction of $PM_{2.5}$). Significant correlation at street level was observed between traffic-related gases (NO_x, CO) and ultrafine particle numbers detected in a street canyon in Copenhagen (Wåhlin et al, 2001), indicating that traffic was their major source in the urban air. Ultrafine particles are generally expected to behave like inert gases within short distances from their sources. Therefore, their concentration may be successfully calculated using urban canyon models (e.g. OSPM) originally developed for gaseous pollutants (Le Bihan et al., 2001).

Chemistry sub-models

Simple street canyon models like STREET-SRI can only calculate concentrations of passive compounds (e.g. CO). On the other hand, models that are used in regulatory applications need to take fast photochemical reactions into account in order to calculate NO_2 concentrations. OSPM uses a simplified chemistry algorithm to account for the transformation of reactive species (i.e. NO_x and O_3) inside a street canyon. AEOLIUS includes a subroutine that calculates statistically NO_2 from NO_x concentrations by means of an empirical curve-fit formula derived from measurements in London (Derwent and Middleton, 1996). In that case, a maximum ratio is set to prevent the NO_2 exceeding 25% of the NO_x at high concentrations. CAR uses an empirical relationship derived from the more elaborate TNO model to calculate street-level NO_2 concentrations, depending on O_3 levels, the fraction of total NO_x directly emitted as NO_2, and the type of the street. Finally, CALINE4 includes the Discrete Parcel Model for NO_x chemistry.

General-purpose CFD models are only able to provide concentrations of inert pollutants, since they do not usually take photochemistry into account. However, specially designed microscale models may combine CFD codes with simplified chemistry algorithms. For example, WinMISKAM adds to MISKAM a simple NO-NO_2 conversion model. A simple photochemistry algorithm linked with MISKAM is also implemented in the street and neighbourhood scale MICRO-CALGRID model, as an alternative to the full chemistry scheme implemented in the urban scale CALGRID model (Stern and Yamartino, 2001).

2.4. Street canyon studies

Several modelling and experimental field studies aiming at establishing pollutant dispersion and transformation patterns within street canyons have been carried out in the past. Depending on their objectives, different modelling and monitoring techniques have been adopted. Some of these studies were purely experimental, which means that they were exclusively based on full- and/or reduced-scale measurements. At the other end of the spectrum, some purely theoretical studies mainly focusing on the investigation of different wind flow and pollutant dispersion regimes using mathematical models can be also found in the literature.

Most commonly, street canyon studies combine both mathematical modelling and experimental work. They may follow two different research approaches. The first one is based on the use of relatively simple parametric models and data obtained from field and/or wind tunnel experiments. Usually, the objective of this kind of studies is to determine the spatial and temporal variability of roadside air pollution, validate operational models, estimate population exposure, etc. The second approach is based on the use of advanced CFD models and experimental data from wind tunnel and/or field measurements. The objective of these studies is usually to obtain a detailed description of the wind and concentration fields within the urban canopy under well-defined dispersion conditions.

Recently, the European research network TRAPOS (Optimisation of Modelling Methods for Traffic Pollution in Streets) gave new insights in a number of street canyon related issues: (a) the influence of moving vehicles on pollutant dispersion and turbulence in urban streets (Kastner-Klein et al., 2000 and 2001; Vachon et al., 2001); (b) the thermal effects on flow and dispersion within street canyons especially under low wind conditions (Kovar-Panskus et al., 2001a; Louka et al., 2001); (c) the sensitivity of flow and turbulence characteristics to the geometry of the street and its surroundings (Kovar-Panskus et al., 2001b; Kastner-Klein and Rotach, 2001; Leitl et al., 2001; Chauvet et al., 2001); (d) the dispersion and transformation of traffic-related particles (Le Bihan et al., 2001; Wåhlin et al., 2001). TRAPOS included field and wind tunnel measurements, as well as mathematical simulations carried out with advanced numerical (MISKAM, CHENSI, MIMO, CFX-TASCflow) and a simpler parametric model (OSPM). A significant part of the work within the network was devoted to the inter-comparison and evaluation of these models (Louka et al., 2000; Sahm et al., 2001; Ketzel et al., 2001) (Fig. 2.3 and 2.4).

In another recent comparison exercise, 24 modellers from 21 different institutions used a range of models to predict pollutant concentrations in the same street canyon in Hannover (Germany). Large discrepancies were identified in the emission and dispersion modelling results obtained from different participants. The fact that individual modellers obtained different results even when they used the same model revealed the influence of the human factor on the quality of the simulations as well as the need for establishing standard operational procedures (Baechlin et al., 2001; Lohmeyer et al., 2002). In the following sections, representative studies covering all aspects of street canyon research are briefly discussed.

2.4.1. Full-scale experiments

DePaul and Sheih (1985; 1986) carried out a tracer gas (SF_6) experiment in an urban street canyon in Chicago (U.S.A.) in order to obtain measurements of pollutant retention times and resulting concentrations within the canyon. The mean wind velocities were determined by analysing trajectories of air balloons that were released in the street. Nakamura and Oke (1988) studied the climate of urban canyons using field observations of wind and temperature from a street canyon in Kyoto (Japan). These observations were used to derive simple algorithms relating the above roof-level to the within-canyon meteorological conditions.

Pfeffer et al. (1995) presented measurements of NO_2, CO, benzene, soot and other atmospheric pollutants carried out in two busy street canyons in Dusseldorf and Essen (Germany), as a part of a pilot study preparing the implementation of new regulations included in the German Federal Clean Air Act. The correlation between different pollutants and the influence of the wind conditions on measured concentrations were investigated.

Namdeo et al. (1999) presented results from a monitoring study on traffic-related particulate pollution in urban areas. Field measurements of airborne fine and coarse particulate matter were taken in an urban street canyon in Nottingham (U.K.) and the correlation of the observed concentrations with traffic was studied. Venegas and Mazzeo (2000) reported CO concentrations measured in a deep street canyon in Buenos Aires (Argentina).

Vertical concentration gradients of CO were observed by Zoumakis (1995) in a busy street canyon in Athens (Greece). The monitoring results were used to derive an empirical expression relating pollutant concentration and height above the ground. Gaseous pollutants (CO, NO_x, O_3) and aerosol particle concentrations were measured at two different heights within an urban street canyon in Lahti (Finland) by Väkevä et al. (1999). The main objective of this study was to investigate the factors leading to the formation of vertical concentration profiles within the canyon.

TSP, PM_{10} and $PM_{2.5}$ concentrations were measured in two open streets and two canyon sites in Hong Kong by Chan and Kwok (2000). These measurements showed that the dispersion of particulate matter was affected by the prevailing wind direction and the aspect ratio of the street. An exponential reduction of TSP and PM_{10} with height was observed.

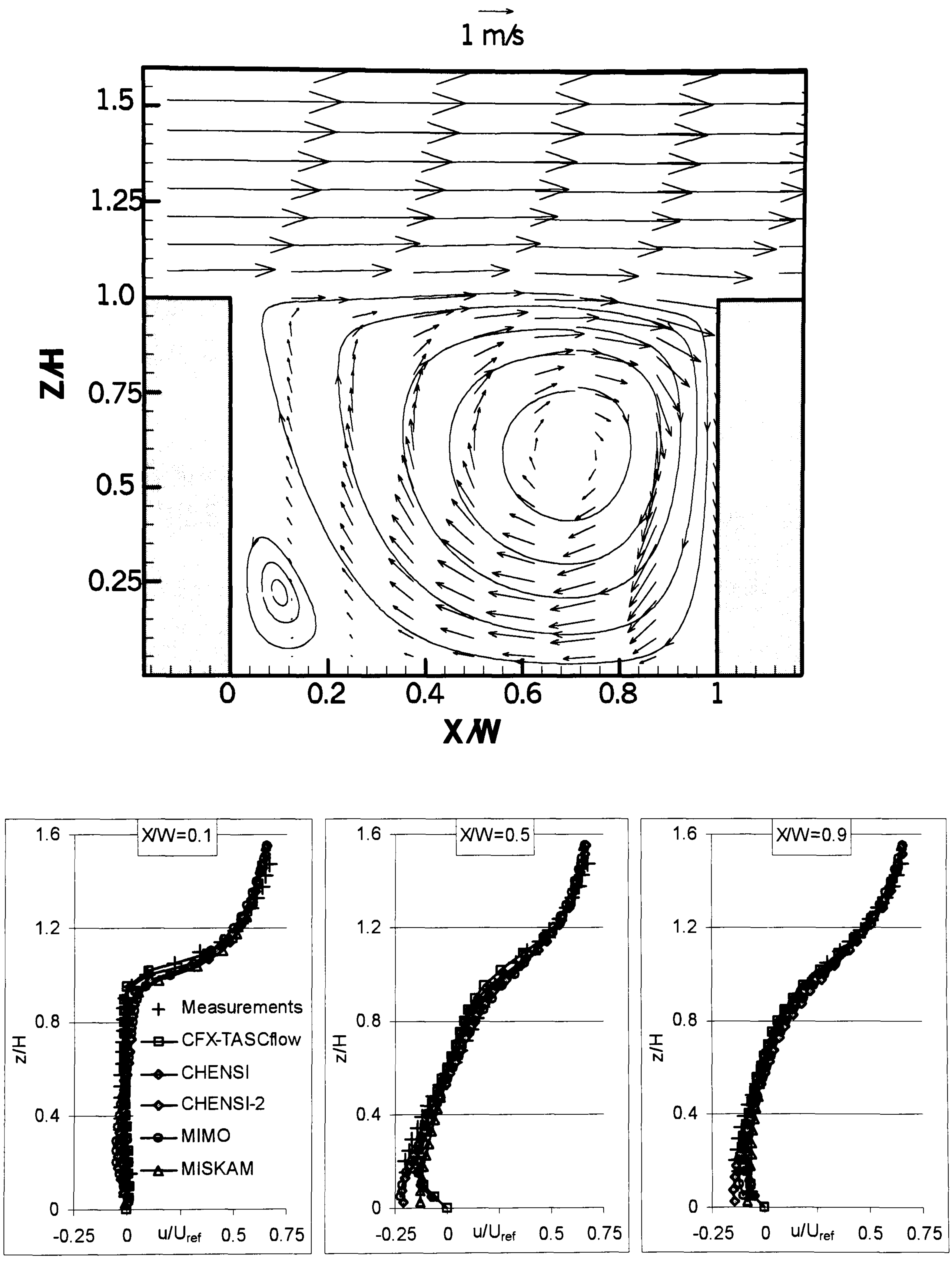
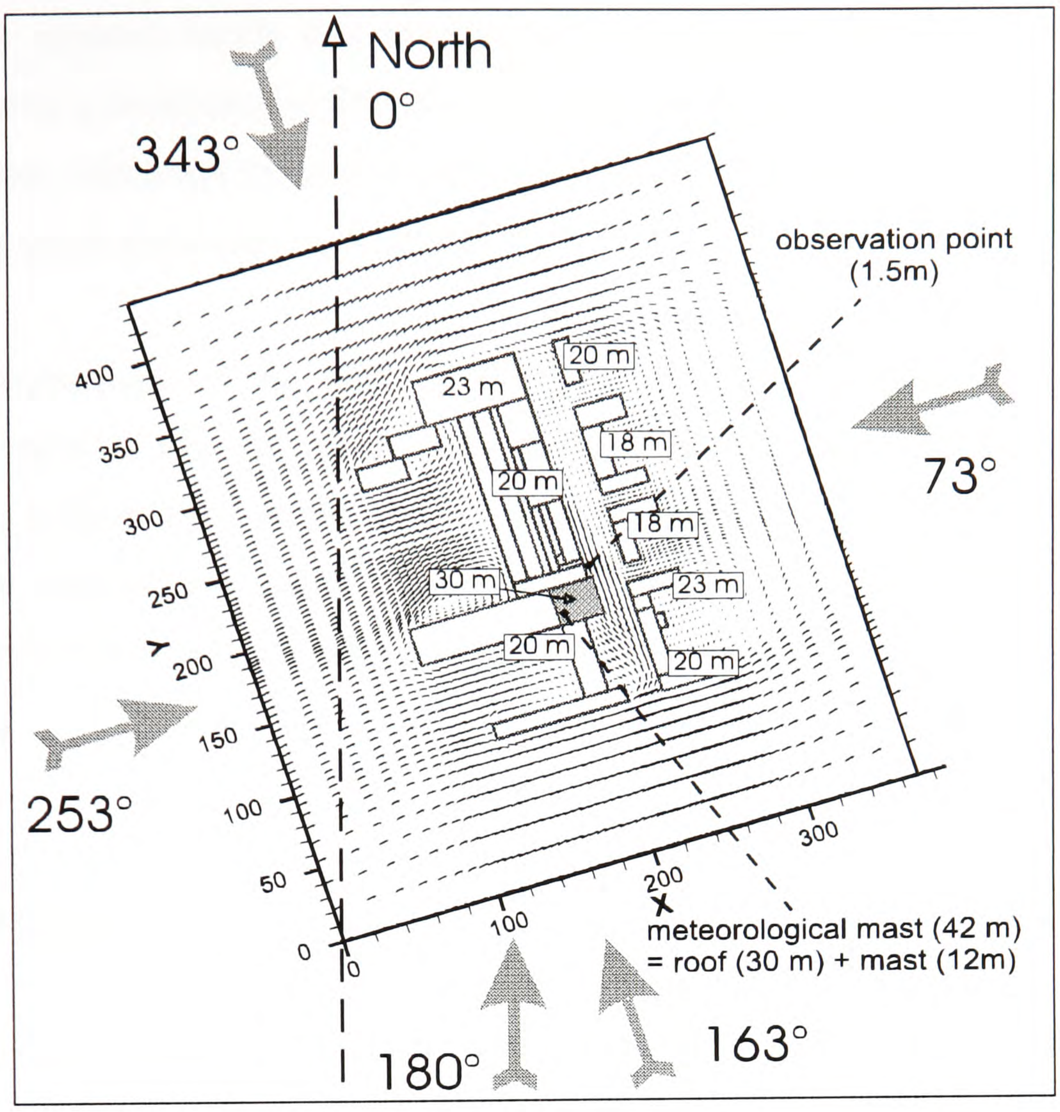


Fig. 2.3: Flow field within and above a wide street canyon as it was reproduced using CHENSI (top), and vertical profiles of the normalised horizontal wind component (u/U_{ref}) measured in a wind tunnel (BLASIUS) and predicted using five different CFD models (bottom). H and W are the height and the width of the canyon, and z and x the height and the distance of the receptor from the canyon wall, respectively (Sahm et al., 2001).



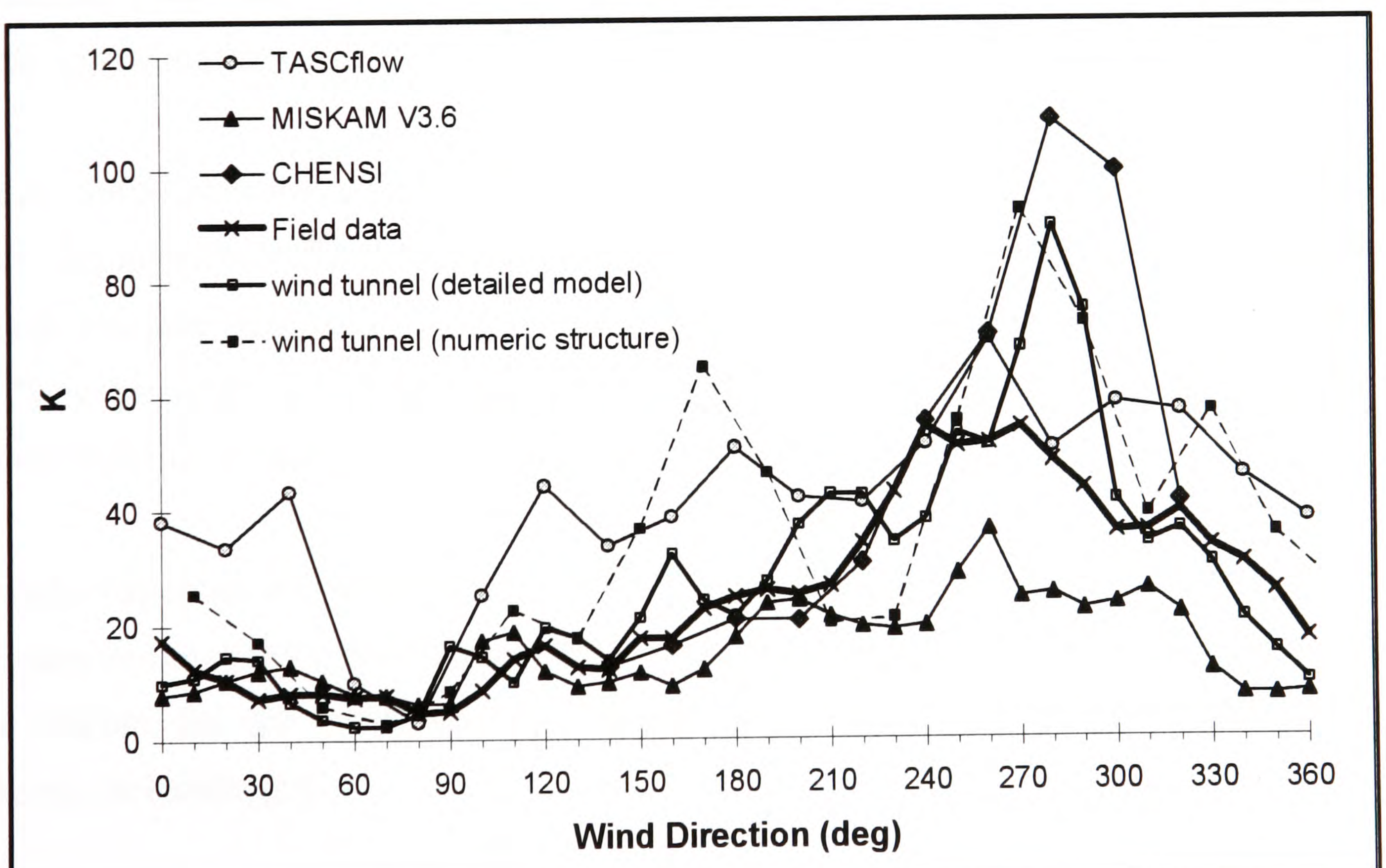


Fig. 2.4: (a) Flow field in a complex street canyon in Hannover reproduced using MISKAM (wind direction: 250°) (top); (b) field data, normalised calculated concentrations (K), and wind tunnel measurements (EnFlo) using the exact shape of the real buildings (detailed model) or building shapes adapted to the resolution of the grid used in the CFD simulations (numerical structure) (bottom) (Louka et al., 2000).

Vachon et al. (2000) reported results (i.e. concentration, temperature and wind fields) from a full-scale experiment carried out in a street canyon (Rue de Strasbourg) in Nantes, France. This was the first campaign of the URBCAP project, which has the aim of assessing pollutant transformation processes within the urban canopy and validating small-scale dispersion models.

Finally, within the framework of the LIFE RESOLUTION project (Wright, 2001), benzene and NO_2 measurements were taken in four European cities (Dublin, Madrid, Paris and Rome) in order to assess pollution levels with reference to established air quality standards, optimise the design of monitoring networks, and provide experimental data to support the validation of urban dispersion models. The sampling, carried out with diffusive tubes, covered a wide range of urban and suburban locations, including a number of street canyons.

2.4.2. Reduced-scale experiments

Leisen and Sobottka (1980) made a comparison between field observations from two street canyons in Cologne (Germany) and wind tunnel measurements in order to investigate pollutant dispersion within urban streets and develop simulation models.

Meroney et al. (1996) presented a wind tunnel study of car exhaust dispersion from street canyons in an urban environment. The main objective of this study was to investigate how pollution dispersion is affected by street geometry and particular emphasis was put on the design of a line source to realistically represent traffic emissions. The experiments were performed in the atmospheric boundary layer wind tunnel (BLASIUS) of the Meteorological Institute of Hamburg University (Germany).

In a later study, Pavageau and Schatzmann (1999) investigated the concentration fluctuations in a reduced-scale urban canyon simulated within BLASIUS. Experimental data sets of wind tunnel measurements carried out in this facility for the validation of microscale dispersion models are available on the Internet (http://www.mi.uni-hamburg.de).

The differences between reduced- and full-scale experiments were illustrated by Liedtke et al. (1999), who compared field measurements obtained in a street canyon in Hannover with wind tunnel results. The generic effect of using a simplified model of a street canyon in the wind tunnel was studied by taking measurements using different scaled models that included various levels of detail of the real canyon geometry. Significant differences were found in the results. In a later study, Schatzmann et al. (2000) showed how wind tunnel data can be used to supplement and enhance the value of field measurements for model validation purposes.

Rafailidis (1999) investigated the influence of atmospheric thermal stratification on urban street canyon ventilation in the EnFlo wind tunnel of the University of Surrey (U.K.). The measurements indicated that stable stratification conditions result in trapping the pollutants within the canyon. In a number of other wind tunnel studies (Rafailidis, 1997; 2000), the influence of building area density and roof shape on the wind field above and inside the urban canopy were highlighted.

Uehara et al. (2000) also investigated the effects of thermal stratification on the wind flow in and above urban street canyons using the atmospheric diffusion wind tunnel at the Japanese National Institute for Environmental Studies (Ogawa et al., 1981). The results showed that the wind vortex within the canyon becomes weaker when the atmosphere is stable.

Kastner-Klein and Plate (1999) presented results from tracer dispersion experiments performed in a neutrally stratified atmospheric boundary layer wind tunnel in the Institute of Hydrology and Water Resources of the University of Karlsruhe (Germany). The influence of systematic parameter variation (i.e. building configuration, roof shape, wind direction) on the concentration field within a street canyon was studied.

Gerdes and Olivari (1999) studied the wind and concentration fields generated within even and asymmetric street canyons under perpendicular winds using the L-2B wind tunnel of the von Karman Institute (Belgium). A strong influence of the surrounding landscape on pollutant dispersion was observed. The ratio of the height of the walls flanking the street was found to have a significant effect on the concentration patterns, while the width of the canyon was proved to be of less importance.

2.4.3. Parametric modelling using field and/or wind tunnel measurements

Johnson et al. (1973) developed STREET-SRI using data from the San Jose Street Canyon Experiment in California. Sobottka and Leisen (1980a; 1980b) created a modified version, called MAPS, which is quite similar in form and performance with the original model.

Nicholson (1975) developed a simple box model that yields street-level average CO concentrations in urban canyons under perpendicular and parallel wind conditions. Model results were proved to be in reasonable agreement with field data obtained in Frankfurt (Germany), Madison and Chicago (U.S.A.).

Yamartino and Wiegand (1986) developed CPBM using data from an extensive field monitoring programme in Bonner Strasse (Cologne). Part of the experimental data (CO and NO_x) was used to evaluate the model. In the same study, the performance of CPBM (only for CO predictions) was compared with the performance of STREET-SRI and its modified version MAPS.

Hoydysh and Dabberdt (1988; 1994), and Dabberdt and Hoydysh (1991) carried out flow visualisation and tracer concentration measurements in the atmospheric boundary layer wind tunnel (ABLWT) of the Environmental Science and Services Corporation (U.S.A.). Pollutant dispersion was simulated using reduced-scale models for street canyons (both even and asymmetric) and intersections. Wind tunnel results were compared with concentrations calculated using STREET-SRI and the analytical model developed by Hotchkiss and Harlow (1973). Finally, a simple exponential law describing vertical concentration profiles was established.

Hertel and Berkowicz (1989a) developed OSPM using measurements obtained in Jagtvej Street in Copenhagen (Denmark). An intensive monitoring site was established in connection with a permanent air quality station operating in this street. A selection of the obtained wind and turbulence data was analysed by Nielsen (2000). OSPM has been applied to several street canyons in Copenhagen, Utrecht, Oslo, Helsinki, Beijing and other major cities (Berkowicz et al., 1996; Hertel and Berkowicz, 1989b; 1989c; Kukkonen et al., 2001; Fu et al., 2000).

Based on the same principles, Buckland (1998) formulated AEOLIUS, which has been mainly used in the UK (Manning et al., 2000). Sacré et al. (1995) presented a slightly modified version of OSPM. Finally, Jensen et al. (2001) developed a decision-support tool (AirGIS) based on OSPM, which applies a geographic information system (GIS) for mapping traffic emissions, air quality and human exposure levels at residential/professional addresses and in streets.

Kono and Ito (1990a) presented the OMG VOLUME-SOURCE model, a microscale dispersion model that estimates concentrations of traffic-related pollutants in an urban area within 200 m from the roadside. The model parameters were determined using experimental data from five locations in Osaka (SF_6 was released as a tracer gas). Model results were compared by the same authors (Kono and Ito, 1990b) with concentrations calculated using three line source dispersion models, namely the JEA model, the TOKYO model, and the HIWAY-2 model (Peterson, 1980).

Qin and Kot (1993) took measurements of CO and NO_x at different heights and distances from the kerb within three asymmetric street canyons in Guangzhou City (China). STREET-SRI and a Gaussian plume model were used in this study to obtain CO and NO_x estimates, which were found to be in reasonable agreement with the observed values.

Lanzani and Tamponi (1995) presented the microscale Lagrangian particle model GEM for the atmospheric dispersion of primary pollutants in the urban canopy. In the same study, GEM was validated against field measurements and compared with CPBM and STREET-SRI. The STREET-SRI model was also used in two independent studies in Argentina to calculate CO concentrations in street canyons in Cordoba (Stein and Toselli, 1996) and Buenos Aires (Bogo et al., 2001). A reasonably good agreement was found between measurements and predictions.

Hargreaves and Baker (1997) developed a Gaussian puff model, called PUFFER, to simulate the dispersion of vehicular pollutants in urban street canyons. This model, which explicitly takes into account vehicle induced turbulence, enables the user to investigate realistic transient situations such as traffic congestion and non-steady above canyon wind fields. A short sensitivity analysis and a comparison with STREET-SRI were also included in the same study.

Gualtieri and Tartaglia (1998) developed a comprehensive air quality model, including traffic, emission and dispersion sub-models, for assessing pollutant concentrations in urban areas. A semi-empirical street canyon algorithm based on STREET-SRI and field measurements from Firenze (Italy) (Tartaglia et al., 1995; Gualtieri and Tartaglia, 1997) was included in this model, which was finally integrated in a GIS.

Hassan and Crowther (1998a) developed a single box model for calculating first estimates of pollutant concentrations within urban canyons. The model parameters were derived using field CO measurements taken in Hope Street, Glasgow (U.K.). Furthermore, Hassan and Crowther (1998b) used PHOENICS to simulate wind flow and pollutant dispersion within the same canyon. The accuracy of the two-dimensional steady state numerical simulations was examined by comparing the predicted results with the field measurements.

Micallef and Colls (1999) developed a semi-empirical emission-dispersion model for predicting the temporal and spatial distribution of airborne particulate matter in street canyons. This model called SLAQ includes emission modules, meteorological pre-processors, modules for within-canyon processes, dispersion modules, and modules accounting for external influences. The dispersion module is mainly based on CPBM. Model features include a correction for the heat island effect, dry and wet deposition, particle settling, etc. SLAQ was evaluated against measurements obtained in a street in Loughborough (U.K.) using the automated near real time Kinetic Sequential Sampling (KSS) system (Colls and Micallef, 1999).

Coppalle et al. (1999, 2001) measured NO_x and CO concentrations at different background and kerbside locations, including a street canyon (Rue Crevier), in a medium size French city (Rouen) during four weeks in winter. Using the obtained experimental data for parameterisation, Coppalle (1999) developed a simple mathematical model that calculates the vertical pollutant distribution inside a street canyon under low wind conditions.

In the framework of AUTO-OIL II programme (Skouloudis, 2000), a large number of air quality modelling simulations were carried out in order to assess the compliance with the new EU air quality standards for NO_2, O_3, CO, benzene, and PM_{10}. An advanced methodology was established, incorporating models of different spatial scales, to provide air quality simulations down to street level. Two urban canyon models, namely OSPM and MICRO-CALGRID, were evaluated using measurements from Viale Murrilo (Milan) and Schildhorn Strasse (Berlin).

Mensink and Lewyckyj (2001) developed the simple mathematical model STREET BOX, which assumes a uniform concentration distribution within a street canyon and is based on the concept of a turbulent intermittent shear flow shed from the roof of the upwind building. Model results were compared with benzene concentrations measured in ten streets in Antwerp (Belgium). Furthermore, benzene, CO and NO_x concentrations calculated with STREET BOX were compared with values predicted using OSPM for a street canyon in Hannover. A discrepancy of 30% between predictions from the two models was reported.

Mukherjee and Viswanathan (2001) used the street canyon and Gaussian line source modules of the regional-scale dispersion model INDIC AIRVIRO to simulate ambient CO concentrations on two major roads in Singapore. The street canyon module based on STREET-SRI gave predictions comparable to the measured values at both the sites, despite the significant differences in street geometry.

Finally, Addison et al. (2000) presented an integrated method for predicting the spatial pollutant distribution within a street canyon. This method was based on a Lagrangian stochastic particle model superimposed on a known velocity and turbulence field. A traffic simulation package (Paramics) was used to model the flow of vehicles in realistic traffic conditions. This model is expected to be calibrated in the future using roadside measurements.

2.4.4. CFD modelling using field and/or wind tunnel measurements

Okamoto et al. (1994; 1996) developed a two-dimensional numerical air quality model that can be applied to street canyon cross-sections under perpendicular wind conditions. It contains a wind field and a diffusion sub-model; the latter based on a Monte Carlo particle scheme. The model was evaluated using databases from field measurements carried out in three typical roadways surrounded by tall buildings in Tokyo (Japan).

Furthermore, the predictive performance of the model was compared with the performance of STREET-SRI and of the APPS three-dimensional numerical model.

Leitl and Meroney (1997) used FLUENT to simulate numerically wind tunnel experiments conducted by Rafailidis et al. (1995) in the BLASIUS facility of the University of Hamburg. Several simplified two- and three-dimensional simulations were carried out to study the effect of emission rate and source design on flow structures and pollutant dispersion within the canyon. The advantages of using numerical CFD codes for optimising wind tunnel experiments were highlighted.

In a later study, Meroney et al. (1999) compared numerical simulations carried out with FLUENT against other wind tunnel data from BLASIUS corresponding to several building shapes including a street canyon. Johnson and Hunter (1998) carried out a preliminary comparison between wind tunnel data from BLASIUS and simulations of wind flow and pollutant dispersion within street canyons using SCAM. This is a numerical code that consists of the wind model CITY and the dispersion model SCALAR (Johnson and Hunter, 1995).

Yoshikawa and Kunimi (1998) reported the development of an air quality simulation system, which calculates traffic volumes, evaluates the effects of building structures on pollutant dispersion along roadways, and takes into account photochemical reactions. The dispersion model, which serves as the platform of the overall simulation system, is a standard CFD code slightly modified to take into account vehicle induced turbulence inside street canyons. The model was validated against field data from an earlier tracer gas (SF_6) diffusion experiment carried out in Tokyo.

Riain et al. (1998) measured CO concentrations at different heights within an asymmetric canyon in central London, U.K. The FloVENT code was used in that case to simulate the concentration and wind fields created in the street. Soulhac et al. (1999) studied pollutant dispersion within street canyons (both even and asymmetric) and intersections using wind tunnel and numerical simulations carried out with MERCURE and CHENSI. The results were compared with two simple models: CARMEN for flow in a single street canyon and SIRANE for flow in a street network (Soulhac and Perkins, 1998; Soulhac et al., 2001).

Both CFD simulations using STAR-CD and wind tunnel measurements in EnFlo were carried out on a model arrangement of two intersecting street canyons, allowing the accuracy of predictions to be assessed (Scaperdas, 2000). It was found that even small changes in building alignment had a significant effect in the dispersion of pollutant in the street. Monitoring data from a permanent air quality station in central London were also used in this study (Scaperdas and Colvile, 1999).

Ketzel et al. (2000) carried out mathematical simulations of pollutant dispersion within street canyons using the relatively simple OSPM and the more complex MISKAM code. The results were compared with wind tunnel simulations and field measurements from two permanent monitoring stations in Copenhagen and Hannover.

Huang et al. (2000) developed a two-dimensional numerical code, which was evaluated using data sets from tracer gas dispersion experiments carried out in an asymmetric street canyon in Tokyo. Chan et al. (2002) used FLUENT to simulate the wind flow and pollutant dispersion within an isolated street canyon. The validation of the numerical model was carried out using an extensive experimental database obtained from BLASIUS. Different turbulence models and street canyon configurations were studied. It was found that wider streets and lower buildings are favourable to pollutant dilution within canyons.

Garcia Sagrado et al. (2002) studied the two-dimensional wind flow and pollutant dispersion within urban canyons by means of wind tunnel measurements (L-2B wind tunnel, von Karman Institute) and numerical simulations carried out with FLUENT. It was observed that pollutant concentrations decreased with increasing the height of the downwind canyon wall. The influence of a third building situated upwind was also investigated.

2.4.5. Theoretical CFD modelling

Hunter et al. (1992) carried out a numerical investigation of typical three-dimensional flows within urban canyons in order to identify the key parameters that determine the transition between the different flow regimes for synoptic winds perpendicular to the street axis (see Section 2.2). Lee and Park (1994) developed a parameterisation scheme whereby the pollutant concentrations in an urban street canyon can be estimated from the source term, the meteorological conditions, and the street geometry using a two-dimensional time-dependant flow model.

Sini et al. (1996) used CHENSI to study the influence of the geometrical aspect ratio of a street, which led to a refinement of Oke's (1988) classification into three flow regimes. In addition, it was shown that the differential heating of street surfaces (e.g. building facades) can influence the dispersion conditions within the canyon. Assimakopoulos et al. (1999) used MIMO to assess the influence of the numerical treatment of the wall boundary on the wind field and concentration patterns within two different two-dimensional street canyons.

Jicha et al. (2000) adopted a three-dimensional Eulerian-Lagrangian approach to study pollutant dispersion in an idealised street canyon taking traffic-induced turbulence into account. The Eulerian approach was based on the CFD code STAR-CD into which a Lagrangian model was integrated. Craig et al. (2001) also used STAR-CD coupled with a mathematical optimisation algorithm to identify the configuration of an idealised urban geometry that minimises pollution peaks. This methodology may be used to optimise traffic patterns or to modify street geometry for air pollution control.

Xia and Leung (2001a; 2001b) used a Lagrangian particle together with a two-dimensional wind field model to simulate flow patterns for different building configurations within the urban canopy. The flow was visualised numerically by discharging a large number of particles into the computational domain. It was found that the higher concentrations did not always appear on the leeward side of the canyon and that the flow pattern was highly dependent on the configuration of the buildings and surrounding urban canopy.

Theodoridis and Moussiopoulos (2000) investigated the influence of building density and roof shape on the wind and dispersion characteristics in an urban area using CFX-TASCflow. In a later study, Theodoridis et al. (2001) applied the same model for simulating wind fields and pollutant dispersion in a complex urban area. In that case, two advanced turbulence models (namely the k-ε and the RSM) were adopted and two grid refinement levels were tested. Venetsanos et al. (2001) carried out flow and dispersion calculations using ADREA-HF, a CFD code for simulating vapour cloud dispersion in complex terrain (Bartzis, 1991), to study the effects of moving vehicles on air pollution patterns within street canyons. The calculations were performed in a moving co-ordinate system with the car and site geometry fully resolved.

Chan et al. (2001) carried out a number of three-dimensional numerical simulations using CFX-5 in order to study flow regimes and corresponding pollutant dispersion characteristics for various types (i.e. aspect ratios) of urban street canyons. Some guidelines related to the geometry of the canyon were established for efficient pollutant dispersion. Jeong and Andrews (2002) used two numerical codes (TEACH-2E and HEATX) to study the two-dimensional flow structure of skimming flow fields in a street canyon at high Reynolds number. The critical aspect ratios of the transition between different vortex regimes were identified.

Finally, Walton et al. (2002) and Walton and Cheng (2002) performed Large Eddy Simulations (LES) to study the turbulent structure and physical dispersion mechanisms of pollutants within street canyons. The LES were implemented by incorporating a dynamic sub-grid scale model into the commercially available CFX code. Comparisons with the k-ε model showed that LES predicted more accurately the turbulence statistics of the flow.

2.5. Discussion

Considerable effort has been made in recent years to improve the scientific understanding of dispersion and transformation phenomena governing urban air quality. A large number of research studies have focused on street canyons, where the highest levels of air pollution often occur and the larger targets of impact are concentrated.

The natural ventilation of urban streets is reduced mainly due to the presence of buildings. Within the urban canopy, wind vortices, low-pressure areas and channelling effects may be created under certain meteorological conditions, giving rise in some cases to air pollution hotspots. For example, high concentration levels have been often observed on the leeward side of regular canyons under perpendicular wind conditions. Furthermore, photochemical activity especially during sunny days may induce high street-level concentrations of secondary pollutants.

Most authors have adopted different combinations of monitoring and modelling techniques for assessing air quality in urban street. There are several methods for monitoring roadside particulate and gaseous pollutants, each one of them having a number of advantages and drawbacks, which make them suitable or not for a specific application.

There are several factors that need to be taken into account when choosing how and where a particular pollutant is to be measured. Some of them are: (1) The response time; (2) the specificity of the method, which means whether the method measures only the pollutant of interest or it has a response to some other pollutants; (3) the sensitivity of the device, which refers to its detection limits; (4) the stability/reliability of the instrument, which is relevant to its calibration/maintenance requirements; (5) the uncertainty associated with the method; (6) the accessibility of the selected monitoring site; (7) and finally the cost.

Passive sampling can be used to obtain air quality data of high spatial resolution (both vertically and horizontally). On the other hand, active sampling can provide high temporal resolution. Hence, a combination of passive and active methods may be ideally used to capture short-term air pollution episodes and hotspots within a canyon.

Mathematical and physical models are needed to optimise air quality monitoring, provide estimates for regulatory purposes, study different street geometries, and finally test future emission and traffic scenarios. Depending on their mathematical/physical principles, they may be more or less suitable for a number of applications.

Gaussian plume models are popular because of their relative simplicity and the possibility of easily including special features like deposition, source buoyancy, etc. Although they are mainly designed to simulate point or line sources in open terrain, they may also include complex terrain, street canyon and intersection modules (e.g. CALINE4). They can produce time series of pollutant concentrations and are used in a wide range of engineering and scientific applications. The main disadvantages of this method are the restricted number of different canyon configurations allowed and the relatively large amount of input information required.

Semi-empirical parametric models (e.g. STREET-SRI, OSPM, AEOLIUS, etc.) are the most commonly used tools in regulatory street canyon applications. They are specially designed to produce time series of pollutant concentrations within near-regular canyons, and they require a relatively small amount of input information and computational resources. On the other hand, they are based on a number of empirical assumptions and parameters that might not be applicable to all urban environments. For this reason, they should be re-calibrated against a small (at least) number of field measurements, if they are to be applied to new locations.

CFD is a numerical modelling technique that can be applied to many different fields of engineering and scientific research. As far as roadside air quality is concerned, the main advantage of the method is that it can reproduce the entire flow and concentration fields within urban canyons of any configuration, if the necessary input data are available. Furthermore, the details of urban canopy can be efficiently taken into account, thanks to the fine grid generation capabilities of modern CFD models.

Traditionally, CFD has been seen as a modelling technique requiring long computational times and expensive hardware/software resources. However, recent computer hardware developments have contributed to the spread of CFD modelling, since the speed and memory capacities of PCs are now sufficient for relatively small applications. Furthermore, CFD codes have become much easier to use due to improvements in interface facilities (although they still require a reasonable level of knowledge of flow physics). It should be kept in mind that the main objective of an environmental CFD exercise is to improve the understanding of the behaviour of a system, rather than obtaining results readily comparable with regulatory standards.

Physical (reduced-scale) modelling in wind tunnels has proved very useful in investigating specific characteristics of pollutant dispersion within the urban canopy (e.g. effects of roof shape, moving and fixed obstacles, etc.). Although wind tunnel experiments have the advantage of providing controlled dispersion conditions (e.g. wind velocity, stability, etc.), they might be seen as relatively expensive and difficult to set up. Wind tunnel measurements are often used in the development and validation of mathematical models.

For the selection of the appropriate dispersion model, one should be aware of the capabilities, underlying assumptions and limitations of the available software. Although they vary greatly in terms of complexity, simple and advanced models can be both useful in different air quality applications (Berkowicz, 1997).

Chapter 3

Field campaigns

3.1. Introduction

A series of air quality monitoring campaigns was carried out at different roadside locations within the region of Paris from December 1998 to December 2001 (Fig. 3.1). The experiments were designed to cover a variety of weather conditions and street configurations, mainly focusing on busy street canyons. The objectives of these field campaigns were the followings:

- Assess the traffic-related air pollution in environments where ambient concentrations and population exposure are expected to be relatively high.
- Cover a variety of street configurations including regular and asymmetric street canyons, an urban intersection and a motorway.
- Cover different weather conditions during winter and summer.
- Optimise the time duration of air quality sampling/monitoring.
- Evaluate the performance of different air quality monitoring techniques and establish a cost-efficient sampling strategy.
- Identify air pollution indicators and possibly devise empirical relationships between key traffic-related pollutants.
- Create high quality experimental data sets for calibrating/validating microscale dispersion models.
- Establish the contribution of petrol stations (with and without vapour recovery systems) to the local air pollution levels under different weather conditions.

In order to meet these objectives, the sites, timing and sampling protocol of the campaigns were carefully selected. Intensive monitoring campaigns were carried out in two regular street canyons (Bd. Voltaire in winter and Rue de Rennes in summer), one asymmetric canyon (Av. Leclerc), and one motorway service station (Route Nationale 10). In addition, long-term measurements were taken in the vicinity of a complex urban intersection (Pl. Basch). Results from these monitoring campaigns have been reported by Vardoulakis et al. (2000a; 2002a) and Gonzalez-Flesca et al. (2001; 2002).

In Sections 3.1.1 – 3.1.4, the experimental methods common to all campaigns (i.e. sampling strategy, monitoring equipment, analytical methods, etc.) are presented. Furthermore, in Sections 3.2 – 3.6, the sites of the campaigns, the detailed sampling protocol and the experimental results for each individual case study are separately presented and interpreted. An overall discussion on the monitoring results, including comparisons with regulatory standards and relevant values found in the literature closes Chapter 3.

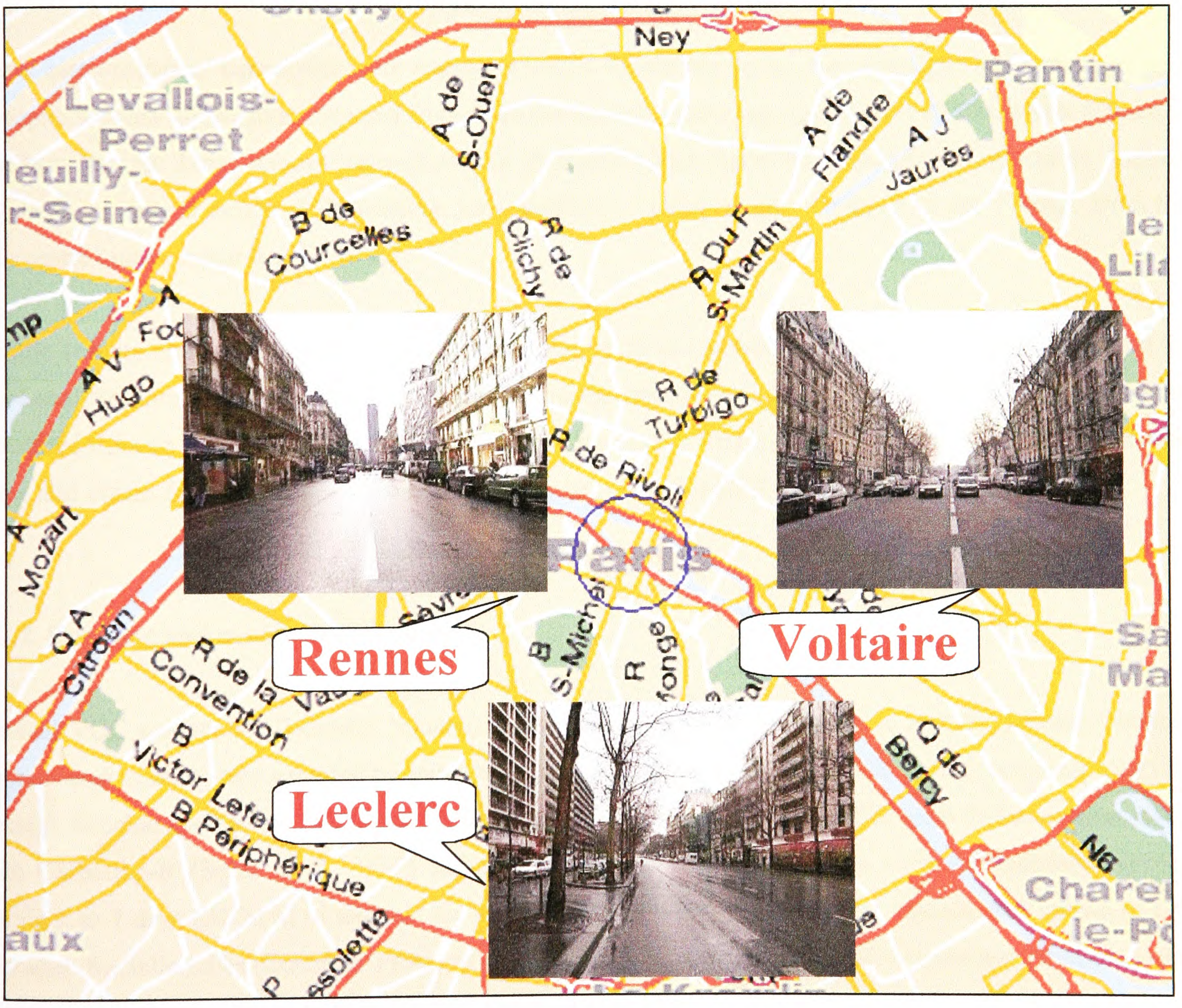


Fig. 3.1: The sites of the three intensive air quality monitoring campaigns in central Paris: Bd. Voltaire (December, 1998); Rue de Rennes (July, 1999); Av. Leclerc (July, 2001).

3.1.1. Sampling strategy

In order to reveal the small-scale spatial and temporal variability of traffic-related pollutants in the selected streets, a combination of different air quality monitoring techniques was tested. Both active and passive means were used to sample a wide range of gases at different roadside and background locations.

Real time CO, NO_x and O_3 monitoring was carried out using a main sampling line established on the kerbside. A weatherproof mobile cabin (trailer) was used to shelter the monitoring equipment and data logger. Using the same sampling line, active (i.e. pumped) sampling of volatile organic compounds (VOC) was conducted by drawing ambient air through a tube filled with the appropriate adsorbent. In addition, passive (e.g. diffusive) VOC samplers were located at different heights and distances from the kerb.

Local meteorological parameters were measured within the streets during the campaigns and compared with synoptic weather information obtained from three permanent monitoring stations operated by Météo France in the Paris Region. These stations were situated in Montsouris Park (anemometer height: 26 m) and St. Jacques Tower (anemometer height: 56 m) within Paris, and Orly Airport at approximately 12 km distance from the city centre (anemometer height: 10 m).

Traffic volume and average vehicle speed were obtained from automatic counters permanently operated by the Local Authority of Paris (Mairie de Paris) within the selected streets. The vehicle fleet composition was estimated from on site spot measurements during the campaigns. Finally, roadside CO and NO_x concentrations were also obtain from the air quality monitoring network of Paris (AIRPARIF).

3.1.2. Sampling and monitoring equipment

Several VOC compounds (benzene, toluene, m+p-xylene, o-xylene, ethylbenzene, styrene, pentane, hexane, heptane and octane) were sampled by pumping roadside air at a constant flow for several one-hour intervals through Supelco glass tubes filled with Carbotrap-B. Radiello Perkin Elmer (RPE) axial diffusive tubes filled with Carbotrap-B (Bates et al., 1997) and sheltered within aluminium boxes were continuously exposed during 2 to 7 days (24/24 h) to ambient air in order to obtain long-term benzene, toluene and xylene (BTX) averages. Radiello tubes (Gonzalez-Flesca et al., 1999) and Sep Pak DNPH-SILICA cartridges (Waters, 1994) were respectively used for passive and active aldehyde sampling.

A carbon monoxide infrared analyser (UNOR 610), a nitrogen oxides chemiluminescence analyser (Mégatec 42-C), and an ozone ultraviolet analyser (Environnement S.A., 03 41M) were used to obtain roadside measurements every second. These devices, sheltered inside the mobile unit, were interfaced with a STADUP data logger, through which data could be observed in real time and recorded as 4 min moving averages, before being further averaged for an hour.

A three-dimensional ultrasonic anemometer (WindMaster, Gill Instruments) and a weather mini-station (AANDERAA) were used to measure street-level wind speed and direction, temperature, relative humidity and global radiation. These instruments were attached to a mast (at 4 to 5 m height above ground) located on the kerbside near the trailer.

3.1.3. Analytical methods

After removal from the tubes with thermal desorption, VOC samples were analysed in the laboratory using gas chromatography (column type: CP-SIL 5CB, 50 m × 0.32 mm, 1.2 μm) + FID. Aldehyde samples were solvent-desorbed and analysed in the laboratory using high performance liquid chromatography (column type: KROMASIL C18, 150 mm × 3 mm, 3.5 μm) + UV detector.

Fick's First Law of diffusion applied to passive samplers with axial (e.g. RPE) and radial (e.g. Radiello) geometry can be used to calculate the ambient concentrations of gases (C) according to the following expressions:

$$C_{axial} = K_{axial}\left(\frac{L}{A}\right)\frac{1}{D}\frac{m}{t} \qquad (3.1)$$

$$C_{radial} = K_{radial}\left[\frac{\ln(r/r_a)}{2\pi h}\right]\frac{1}{D}\frac{m}{t} \qquad (3.2)$$

where $K_{axial/radial}$ are empirical constants accounting for deviations from ideal behaviour, (L/A) and $[\ln(r/r_a)/2\pi h]$ are constants depending on the dimensions of the axial and radial samplers respectively (Appendix I), D is the diffusion coefficient of the gas in ambient air, m is the mass of the pollutant sampled, and t the time of exposure (Bates et al., 1997).

3.1.4. QA/QC programme

A quality assurance / quality control (QA/QC) programme including sampling duplicates, field and laboratory blanks, and instrument calibration with standard gases before, during and after the campaigns was followed. The QA/QC programme of the chemical laboratory (INERIS) included the validation of equations (3.1) and (3.2). That was achieved by exposing diffusive tubes to dynamically generated contaminated atmospheres.

In the field, an additional mechanical microvane and three-cup anemometer was used to assure the quality of the wind measurements. That was placed at street level next to the ultrasonic anemometer and interfaced with the STADUP data logger. Finally, manual vehicle counts were taken during part of the campaigns and compared for consistency with the information obtained from the automatic traffic monitoring network.

3.1.5. Measurement uncertainty

The uncertainty associated with air quality measurements may be reduced by using accurate monitoring instruments and implementing sound QA/QC methodologies, but it cannot be totally eliminated.

Scientific equipment manufacturers usually quote overall uncertainty ranges within which measurement errors are expected to be found. For example, an uncertainty of approximately 5% in the CO concentrations detected with a standard infrared analyser should be expected. On the other hand, the uncertainty attributed to benzene measurements carried out with diffusive tubes may approximately be 15%.

Finally, it should be remembered that the choice of the sampling site might introduce a much larger uncertainty component in the obtained concentrations than the analytical error.

3.2. Boulevard Voltaire

Case study 1: Intensive monitoring campaign in a regular street canyon during winter (Dec. 1998)

3.2.1. Description of the site

The first monitoring campaign was carried out during winter 1998 (14-18 December) in Boulevard Voltaire, between Rue des Boulets and Rue de Montreuil junctions. This site is a regular street canyon, typical example of the urban topography of Paris. Uniform six-storey buildings line up continuously on both sides of a busy four-lane traffic axis with large pavements and leafless trees (in winter). The aspect ratio (H/W) of the canyon is approximately equal to 0.8 and the street axis bearing from the north 140° (Fig. 3.2).

Measurements were taken within a straight road segment of approximately 300 m. Traffic lights were operating at both ends of the canyon, and there was a pedestrian crossing at a distance of 34 m from the main sampling point. The average traffic volume in Bd. Voltaire was 30,000 veh/day during sampling. Urban background measurements were also carried out in an adjacent park location (Impasse des Jardiniers), at a distance of approximately 100 m from the canyon.

3.2.2. Sampling protocol

Diffusive RPE samplers were located at two different heights (1st and 5th floor) near the walls of the canyon, and at one background site. The exact locations of all passive samplers as well as the dimensions of the canyon are indicated in Fig.3.3. The tubes remained exposed to ambient concentrations for five days (24/24h).

The mobile monitoring unit was parked on the east side of the canyon and a main sampling line was established at the kerb with its inlet at 3.7 m height above the ground and 8.0 m distance from the canyon wall. Ambient air was pumped through this line and analysed continuously. CO, NO_x and O_3 measurements were recorded during daytime (12/24 h). During one day of the campaign (15 December), active VOC sampling was conducted through the main sampling line using Supelco tubes.

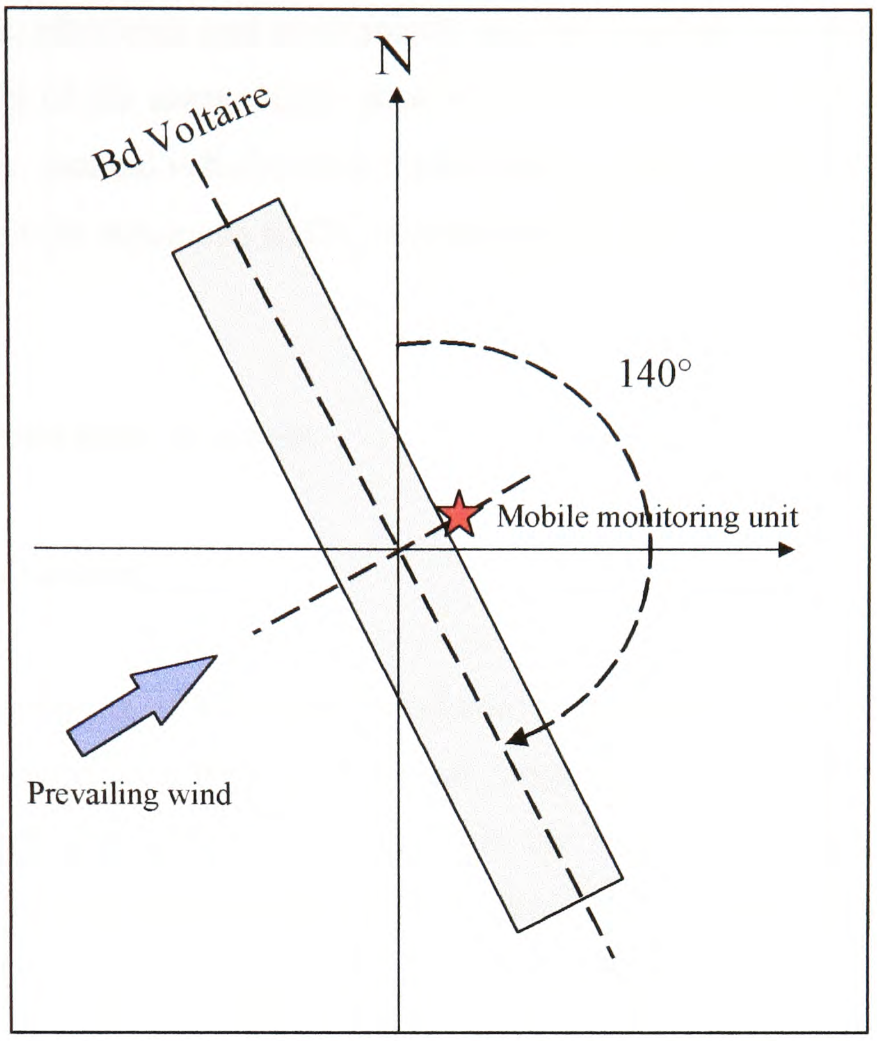


Fig. 3.2: Layout, orientation and prevailing wind direction in Bd. Voltaire during measurements.

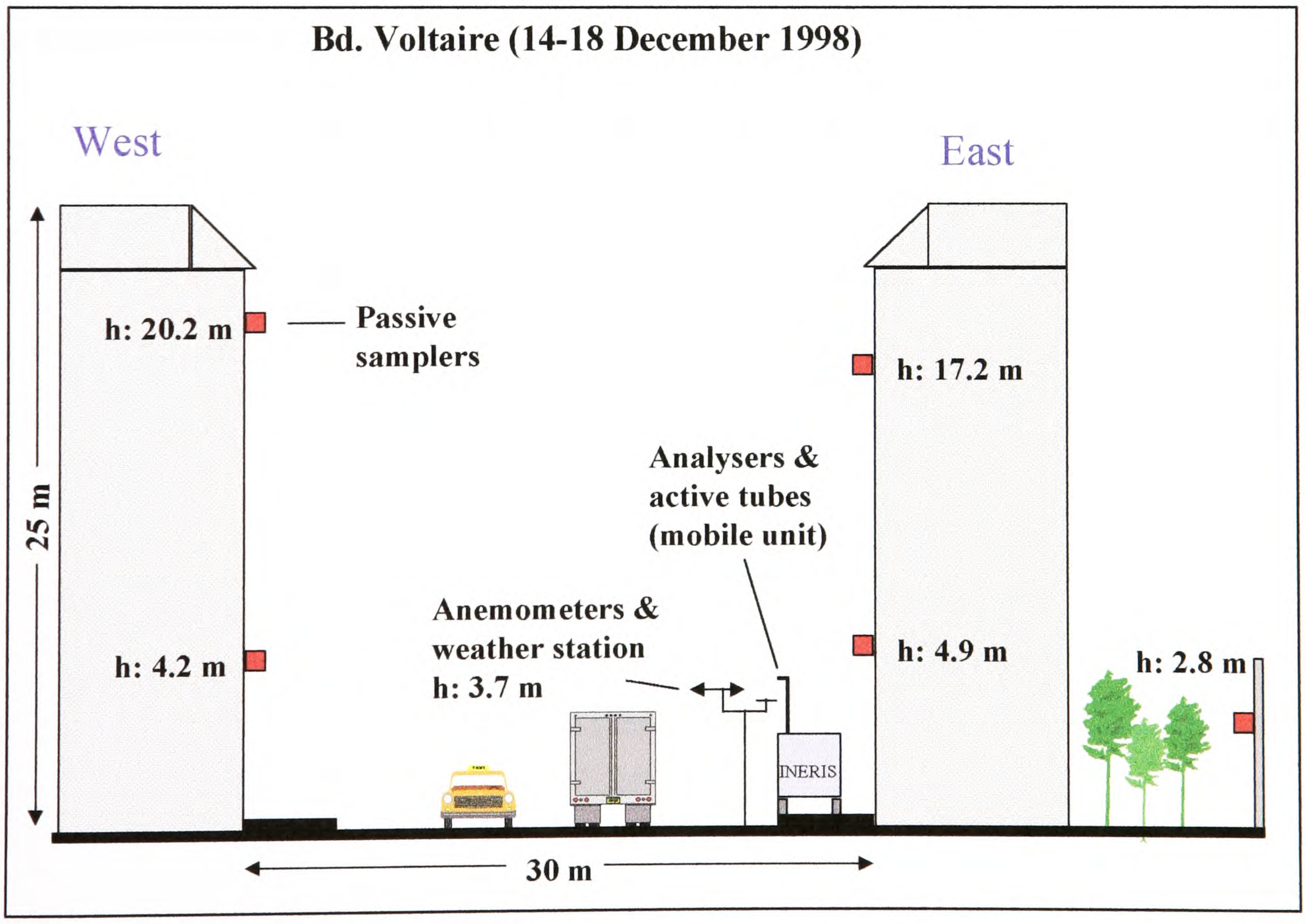


Fig. 3.3: Canyon dimensions and location of monitoring equipment in Bd. Voltaire.

The two anemometers (i.e. ultrasonic and mechanical) and the weather mini-station were located at the kerb near the trailer. The height of the anemometer mast was 3.7 m above the ground and the distance from the canyon wall 8.5 m. Finally, manual vehicle counts were taken during 10 hours of the campaign and compared with the data obtained from the automatic traffic monitoring network.

3.2.3. Analysis and interpretation of results

Relationship between pollutants

The simultaneous active sampling of VOC and continuous monitoring of CO during one day of the campaign allowed to calculate the correlation between different compounds of interest (Table 3.1), and to establish empirical relationships between their concentrations on the kerbside. A very strong correlation between benzene, toluene, m+p-xylenes, heptane and CO was identified. A quite strong correlation between these compounds and other hydrocarbons (pentane, hexane, octane, ethylbenzene, o-xylene) was also observed.

The measurements carried out with diffusive tubes at different sampling locations within Bd. Voltaire also showed a very strong correlation between benzene and toluene concentrations (Fig. 3.4). The experimental toluene to benzene ratio (by volume) in that case was 2.9.

Table 3.1: Correlation coefficients of air pollutants measured with active sampling in Bd. Voltaire.

	Benzene	Toluene	m+p-Xylenes	Pentane	Hexane	Heptane	Octane	Ethylbenzene	o-Xylene
CO	0.93	0.90	0.83	0.71	0.75	0.91	0.79	0.77	0.72
Benzene		0.99	0.88	0.85	0.91	0.96	0.88	0.85	0.86
Toluene			0.90	0.87	0.94	0.97	0.88	0.88	0.88
m+p-Xylenes				0.78	0.90	0.87	0.87	0.99	0.92
Pentane					0.83	0.85	0.68	0.75	0.72
Hexane						0.87	0.82	0.90	0.92
Heptane							0.87	0.84	0.84
Octane								0.89	0.89
Ethylbenzene									0.93

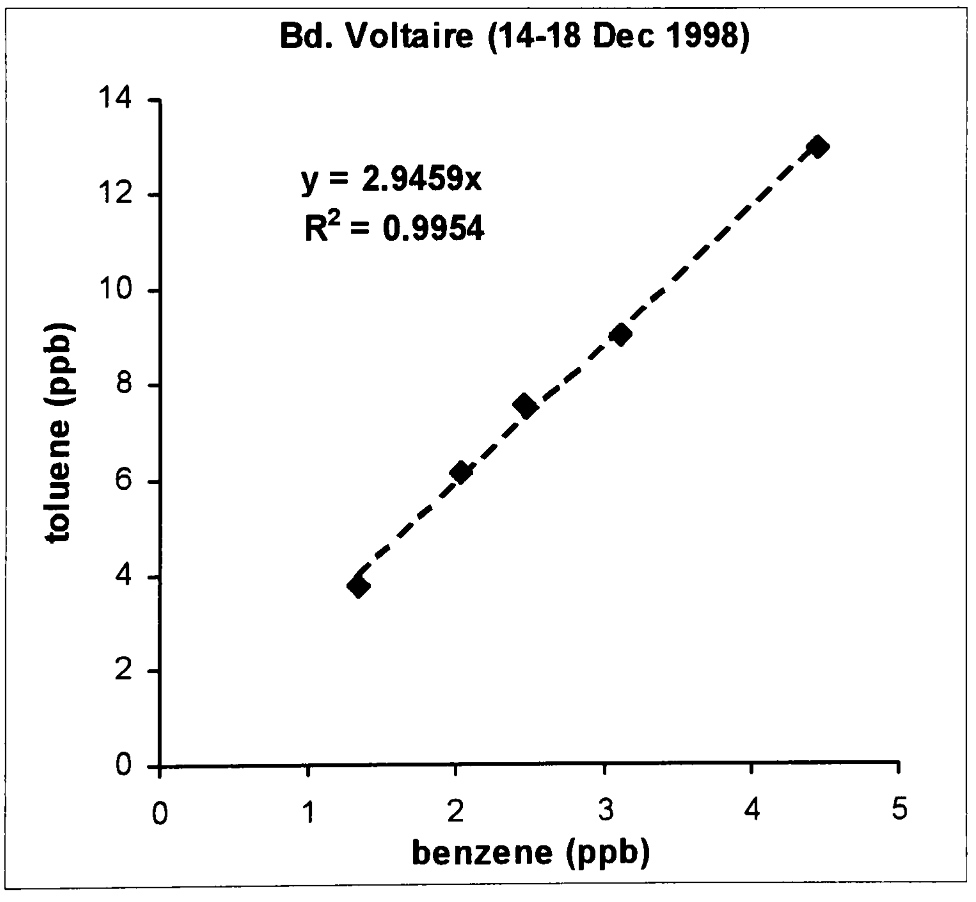


Fig. 3.4: Average toluene (ppb) vs. benzene (ppb) concentrations measured with passive sampling in Bd. Voltaire.

Wind flow and dispersion conditions

During part of the measurements in Bd. Voltaire, there was evidence of a wind vortex regime being established within the canyon. That was identified using the local wind information obtained at street level. For perpendicular or near-perpendicular synoptic winds (200°-260°), a downward airflow was observed on the windward side of the street (Fig. 3.5).

In addition, an elastic-type reflection of the wind off the windward wall of the canyon was detected for synoptic winds greater than 2 m/s. Using synoptic wind data from Montsouris station, street level wind directions were calculated applying a simple relationship deduced from the elastic reflection assumption. The calculated values were then compared with the wind directions actually measured in the street using the 3D ultrasonic anemometer. A very good agreement ($r = 0.78$) between measured and calculated wind directions was finally observed for winds above 2 m/s (Fig. 3.6).

Furthermore, the influence of the synoptic wind speed and direction on the dispersion of pollutants at street level is illustrated in Fig. 3.7. It can be seen that the lower CO concentrations (normalised with respect to the traffic volume) occurred for synoptic wind directions between 200° and 260° (thus perpendicular or near-perpendicular to the street axis) and wind speeds higher than 2 m/s. For these dispersion conditions, the natural ventilation of the canyon was optimised.

The influence of the synoptic wind direction on pollutant dispersion within the canyon was also illustrated on the pollution roses plotted for CO and NO (Fig. 3.8). Hourly mean CO and NO concentrations, normalised with respect to the wind speed and traffic volume, were assigned to the corresponding synoptic wind directions. Then, the arithmetic mean of the concentrations was calculated for each of 36 equal wind direction sectors. Both CO and NO roses demonstrated a clear dependence of pollution levels on the synoptic wind direction. It can be seen that, keeping the other factors constant, winds parallel or near-parallel to the street axis (i.e. from SE directions) favoured pollution built-up on the kerbside, while perpendicular winds (i.e. from SW directions) provided better dispersion conditions.

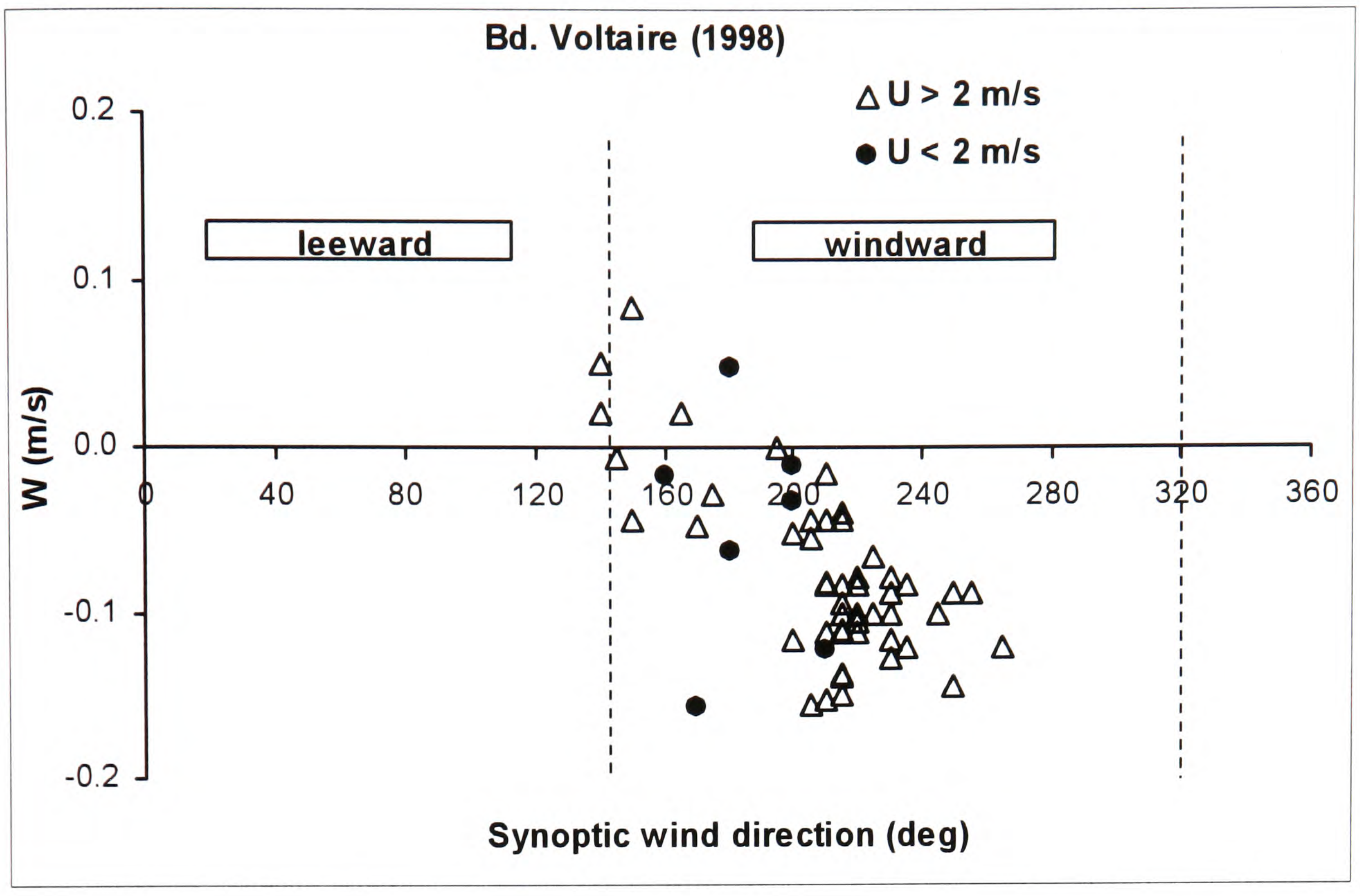


Fig. 3.5: Vertical street level wind speed (W) vs. synoptic wind direction in Bd. Voltaire (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

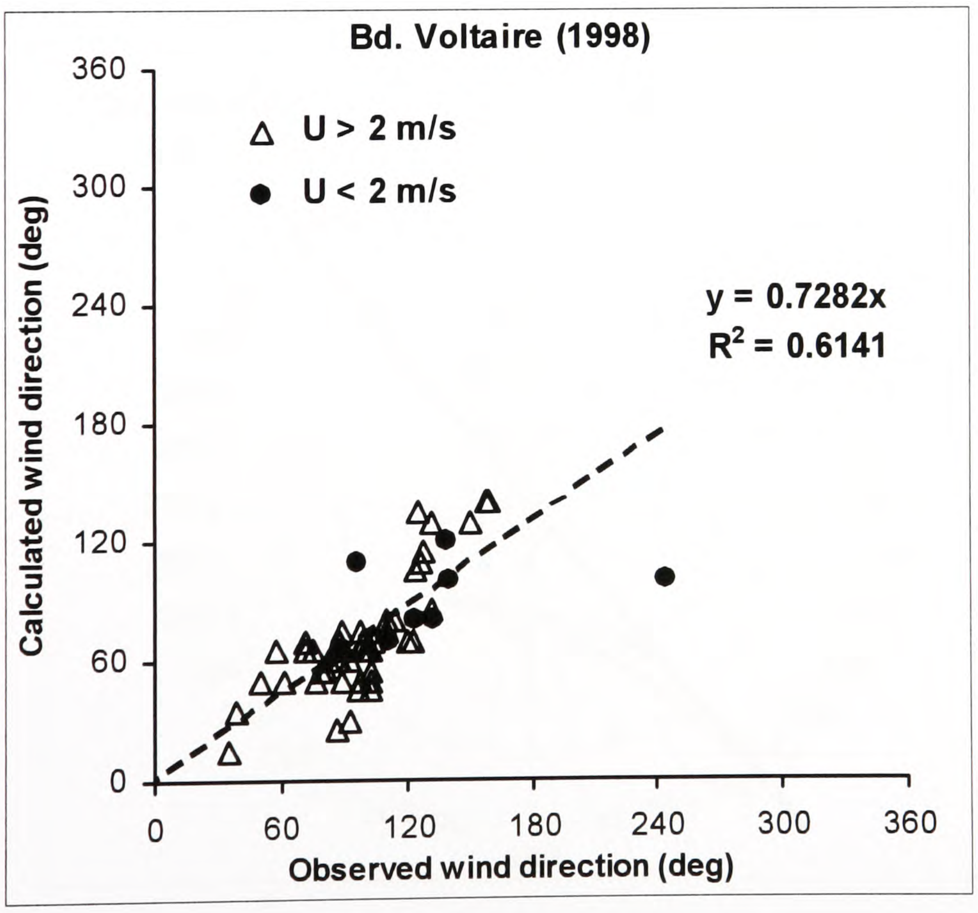


Fig. 3.6: Calculated wind directions: $[Dir]_{local} = 2\,[\text{Street bearing}] - [Dir]_{synoptic}$ vs. observed street-level wind directions in Bd. Voltaire (sorted for synoptic wind speed above and below 2 m/s).

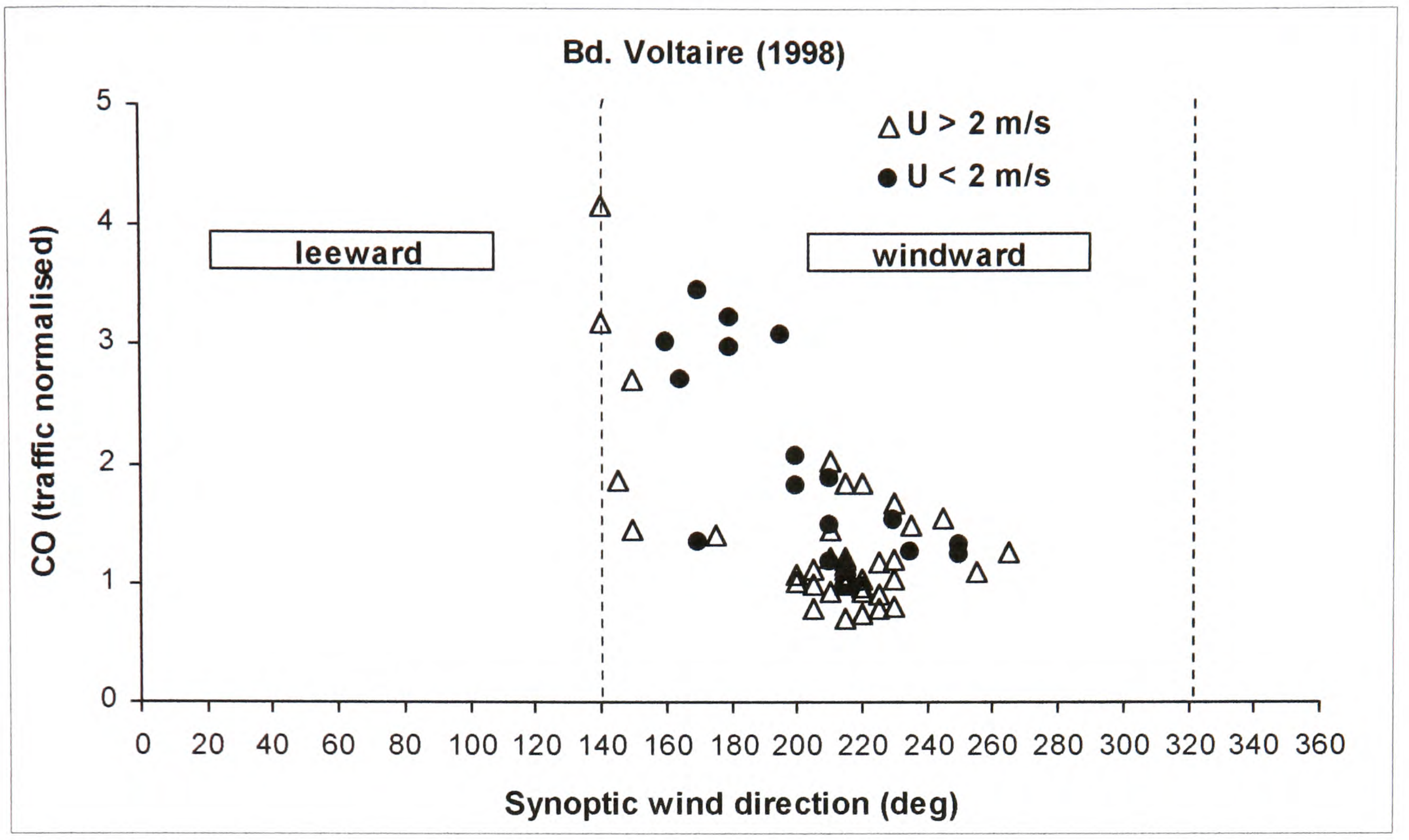


Fig. 3.7: Traffic normalised CO concentrations vs. synoptic wind direction in Bd. Voltaire (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

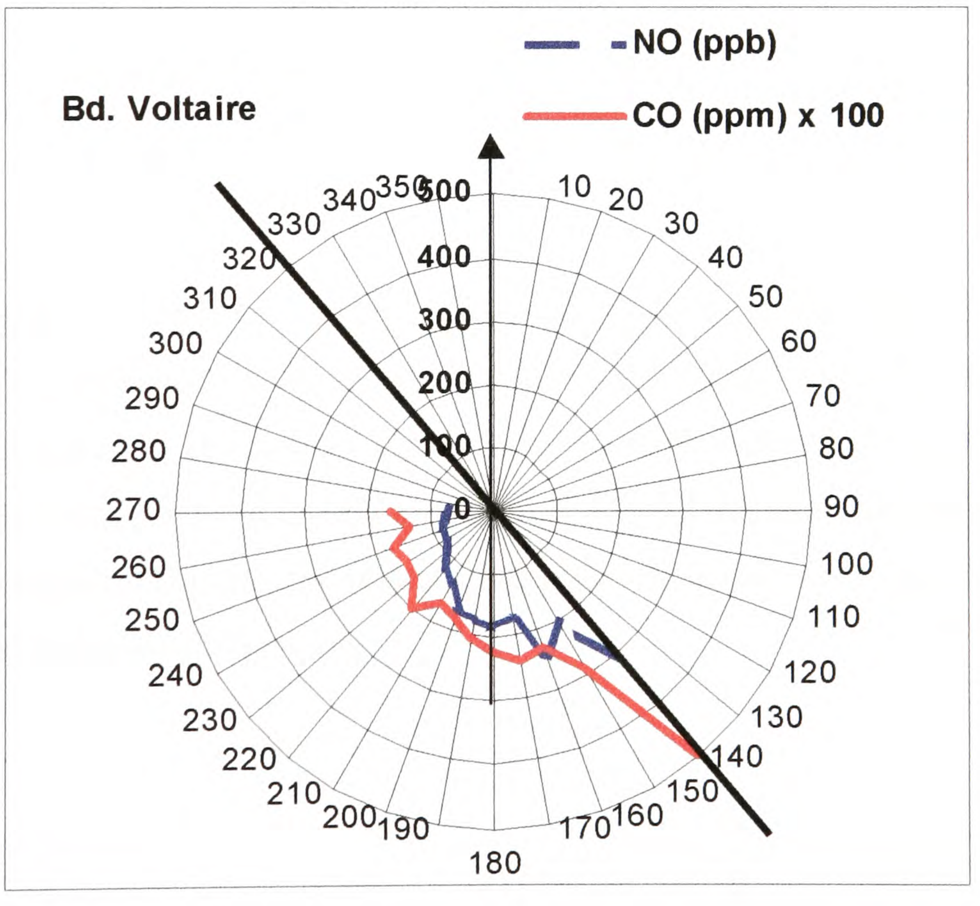


Fig. 3.8: Traffic and wind speed normalised CO and NO concentration roses in Bd. Voltaire. (The heavy straight line indicates the direction of the street.)

Spatial variability

Diffusive VOC samplers were deployed to reveal the spatial variability of traffic pollution within Bd. Voltaire and in relation with urban background levels. Using BTX as indicators, strong concentration gradients were identified in the horizontal and vertical sense within the canyon. The highest benzene concentrations were detected at street level, on the side of the canyon that was up-wind (i.e. leeward) most of the time (Fig. 3.9). At the leeward sampling locations, weekly benzene averages were from 55% (5^{th} floor) to 80% (1^{st} floor) higher than the values measured at approximately the same height on the windward side of the canyon (i.e. down-wind).

A substantial reduction in ambient benzene concentrations along with the height above the street was also observed. The weekly benzene averages measured on 5^{th} floor balconies were from 20% (on the windward side) to 30% (on the leeward side) lower than at 1^{st} floor level.

Finally, benzene concentrations measured on the kerbside were 2 to 3.5 times higher than those detected in the selected background location. Even the benzene values detected at 17-20 m height near the walls of the buildings facing the street were significantly higher than the background value. The same trends were also observed for the other VOC sampled with diffusive tubes during the campaign.

Temporal variability

Two peaks of CO were observed in Bd. Voltaire during morning hours on the 16^{th} and 17^{th} of December (Fig. 3.10). The first one can be explained by the presence of low wind conditions (≤ 2.5 m/s) in the region during the third day of the campaign (16^{th} December). On the other hand, the second CO peak may be attributed both to the relative low wind speed (2.5-3.0 m/s) and to the parallel wind direction during part of the fourth day of measurements (17^{th} December). During this winter campaign, O_3 levels were very low at all times, due to negligible photochemical activity in the region.

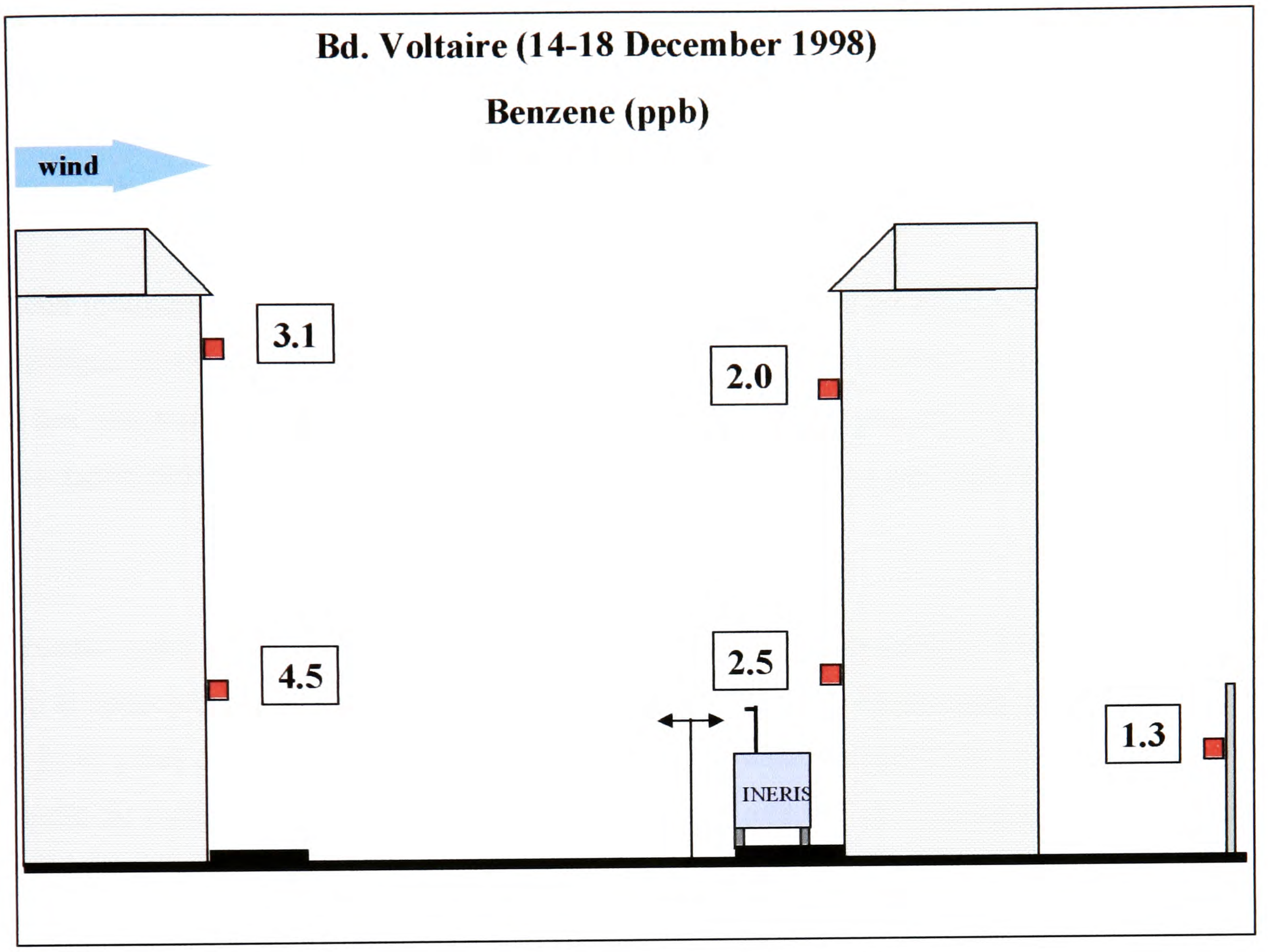


Fig. 3.9: Benzene (ppb) concentrations measured with passive sampling in Bd. Voltaire (14-18 Dec. 1998)

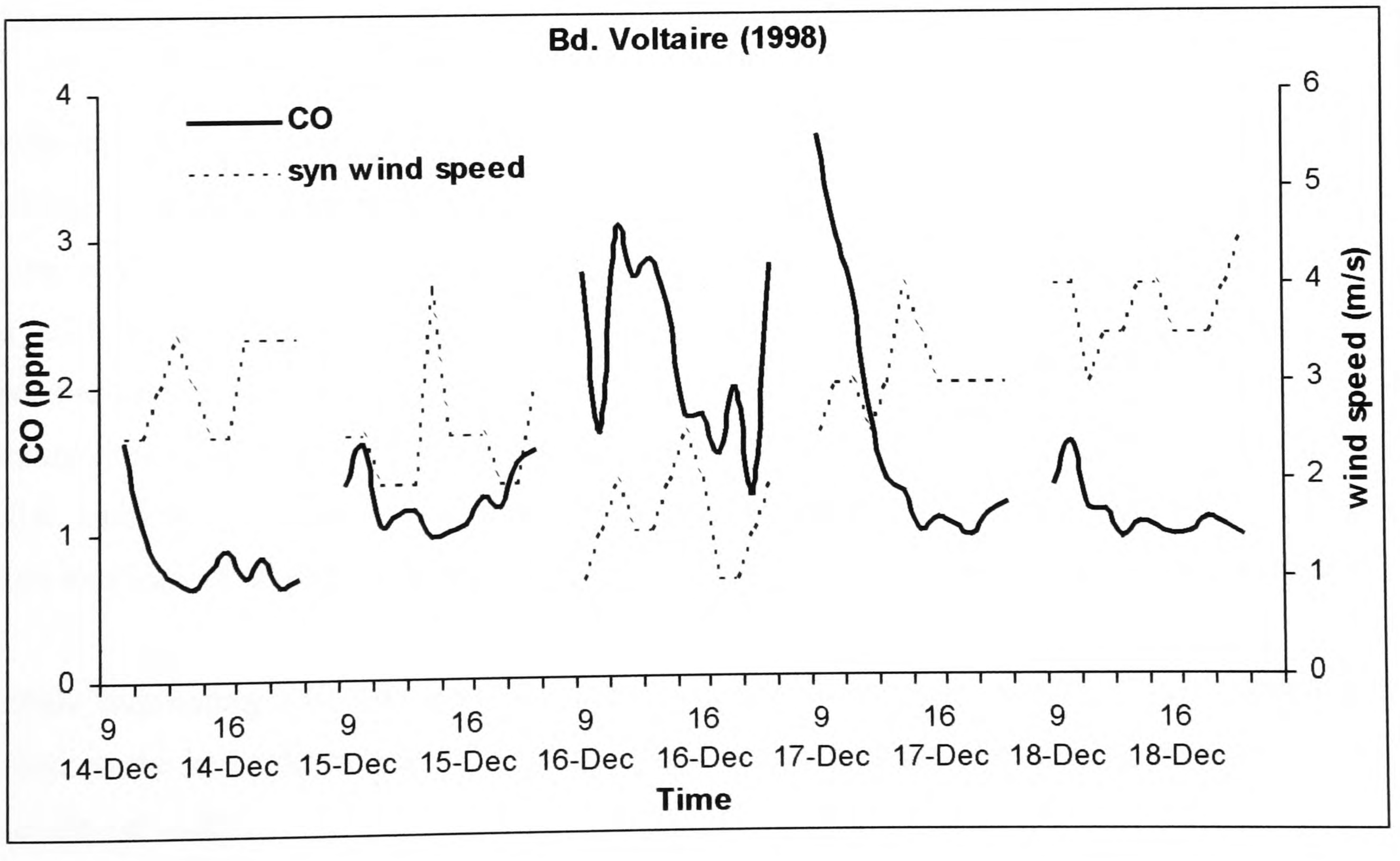


Fig. 3.10: Hourly mean CO (ppm) concentrations and synoptic wind speeds in Bd. Voltaire.

3.3. Rue de Rennes

Case study 2: Intensive monitoring campaign in a regular street canyon during summer (July 1999)

3.3.1. Description of the site

The second monitoring campaign was conducted in July 1999 in Rue de Rennes, between Rue d'Assas and Rue Coëtlogon junctions. This is a regular street canyon flanked by uniform six-storey buildings on both sides. It has four traffic lanes, cars parked on both sides of the street and relatively large pavements without trees. The aspect ratio (H/W) is approximately equal to 1.1 and the street axis bearing from the north 32° (Fig. 3.11).

Measurements were taken within a straight road segment of approximately 150 m. Traffic lights were operating at both ends of the canyon at a distance of at least 50 m from the monitoring unit, which was located at 37 m from a bus stop. The average traffic volume during measurements was 23,000 veh/day. Two green areas (Jardin du Luxembourg and Sq. Recamier) not directly affected by road traffic were selected for urban background measurements. These locations were at a distance of approximately 300 m from the canyon in opposite directions.

3.3.2. Sampling protocol

Two different sets of diffusive RPE samplers were used to examine separately VOC levels during weekend and working weekdays. A more detailed spatial resolution of VOC concentrations was obtained with respect to the previous campaign in Bd. Voltaire by increasing the number of passive sampling locations. In Rue de Rennes, apart from the measurements at two different heights (1st and 5th floor) near the walls of the canyon, samplers were also placed on the kerbside within the human breathing zone (h = 1.5 m), and at two different background sites. Diffusive aldehyde sampling was also carried out during weekdays. Radiello samplers were exposed to ambient air within the pedestrian breathing zone and near the walls of the canyon at 1st floor level. The exact locations of all passive samplers as well as the dimensions of the canyon are indicated in Fig. 3.12.

The mobile monitoring unit was parked on the east side of the canyon and a main sampling line was established on the kerbside with its inlet finally placed at 2.9 m height above the ground and 3.3 m distance from the canyon wall.

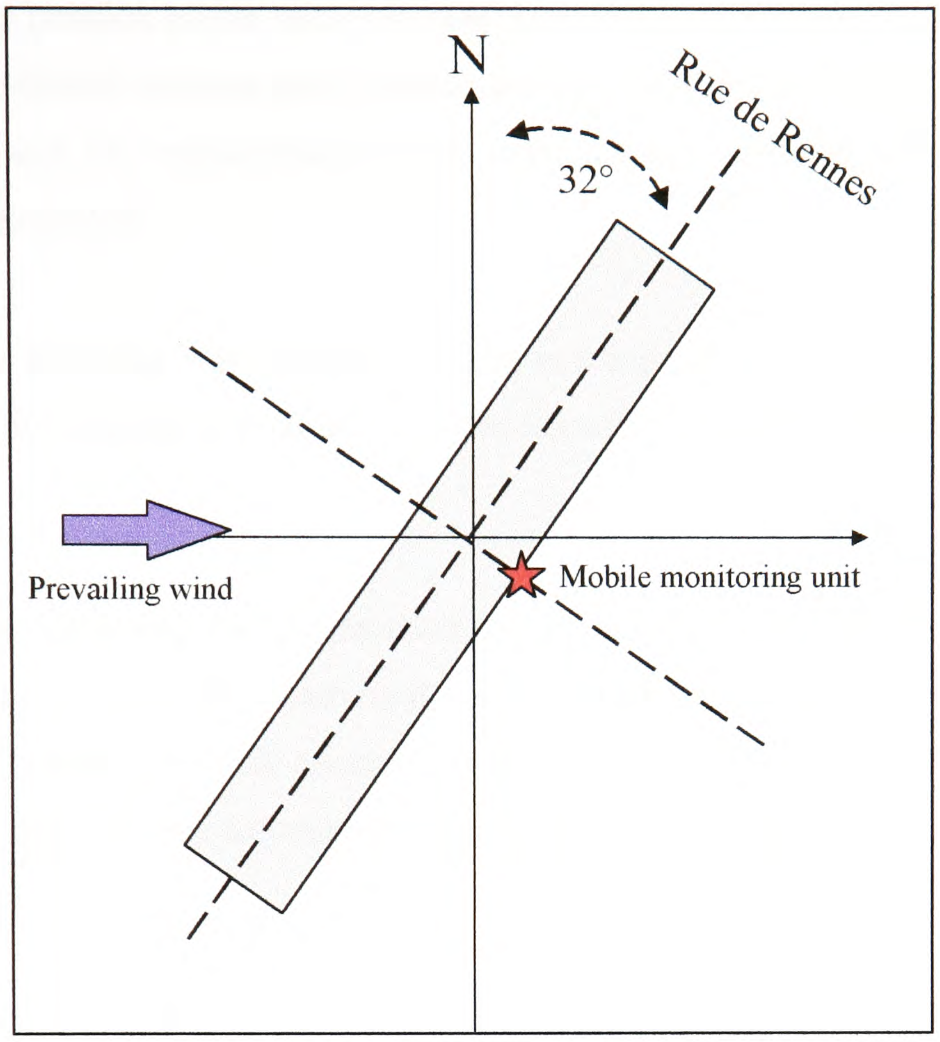


Fig. 3.11: Layout, orientation and prevailing wind direction in Rue de Rennes during measurements.

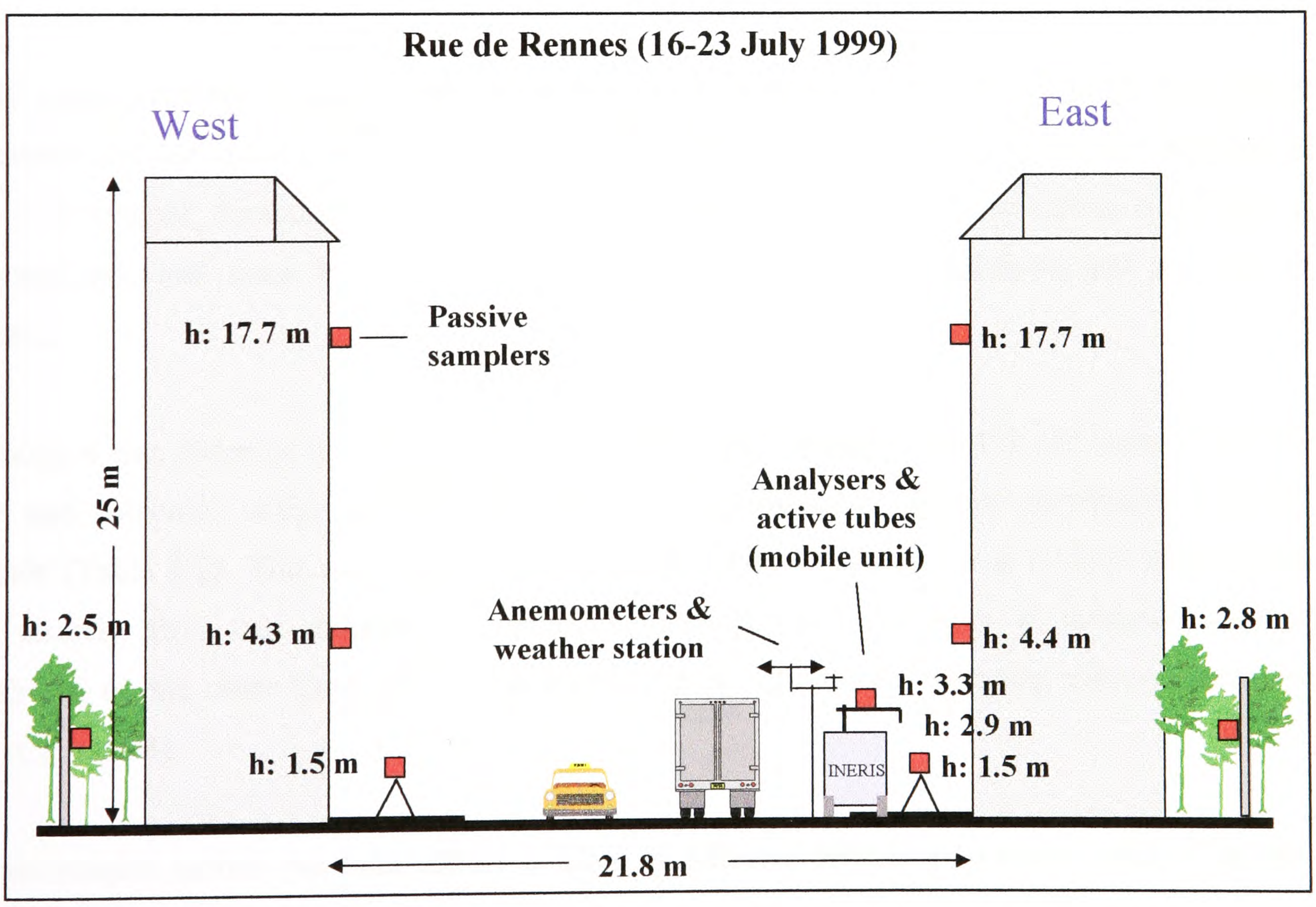


Fig. 3.12: Canyon dimensions and location of monitoring equipment in Rue de Rennes.

It should be noted that the position of the inlet was readjusted soon after its initial installation at the kerb in order to avoid sampling undiluted exhaust gases coming directly from idling or slowly moving cars. After this readjustment, CO, NO_x and O_3 measurements were continuously recorded during day and night-time throughout the campaign (24/24 h).

Active VOC and aldehyde sampling was conducted during two days of the campaign (20-21 July) by drawing ambient air at a constant flow during several one and two hour intervals through Supelco and Sep Pak tubes, respectively.

The two anemometers (i.e. ultrasonic and mechanical) and the weather mini-station were located at the kerb near the trailer. The height of the anemometer mast was 3.9 m above the ground and the distance from the canyon wall 5.0 m. Finally, manual vehicle counts were taken during 25 hours of the campaign and compared with the information obtained from the automatic traffic monitoring network.

3.3.3. Analysis and interpretation of results

Relationship between pollutants

The VOC concentrations obtained with active tubes were statistically compared with the simultaneous CO measurements carried out through the same sampling line. A very strong correlation between benzene, toluene, m+p-xylenes, formaldehyde and CO was identified (Table 3.2). A less strong correlation between these compounds and other hydrocarbons (ethylbenzene, o-xylene, acetaldehyde and styrene) was also established.

Furthermore, it was observed that formaldehyde correlated very strongly with CO and some of the VOC (e.g. benzene and toluene), while acetaldehyde showed a considerably weaker correlation with the same compounds (Table 3.2). This may lead to the conclusion that formaldehyde is mainly of vehicular origin, coming directly from car exhausts or indirectly through the oxidation of unburned hydrocarbons. Acetaldehyde, on the other hand, may come from a variety of sources, including photochemical processes (Ferrari et al., 1998).

The measurements carried out with diffusive tubes at different sampling locations within Rue de Rennes during weekdays and a weekend revealed a very strong correlation between benzene and toluene concentrations (Fig. 3.13). The experimental toluene to benzene ratio (by volume) during the whole campaign was 3.4.

Table 3.2: Correlation coefficients of air pollutants measured with active sampling in Rue de Rennes.

	Benzene	Toluene	Ethylbenzene	m+p-Xylene	Styrene	o-Xylene	Formaldehyde	Acetaldehyde
CO	0.94	0.89	0.61	0.97	0.49	0.67	0.93	0.69
Benzene		0.91	0.40	0.98	0.22	0.49	0.95	0.70
Toluene			0.62	0.98	0.37	0.72	0.92	0.58
Ethylbenzene				0.98	0.92	0.98	0.81	0.46
m+p-Xylene					0.82	0.99	0.65	0.21
Styrene						0.87	0.75	0.44
o-Xylene							0.81	0.46
Formaldehyde								0.83

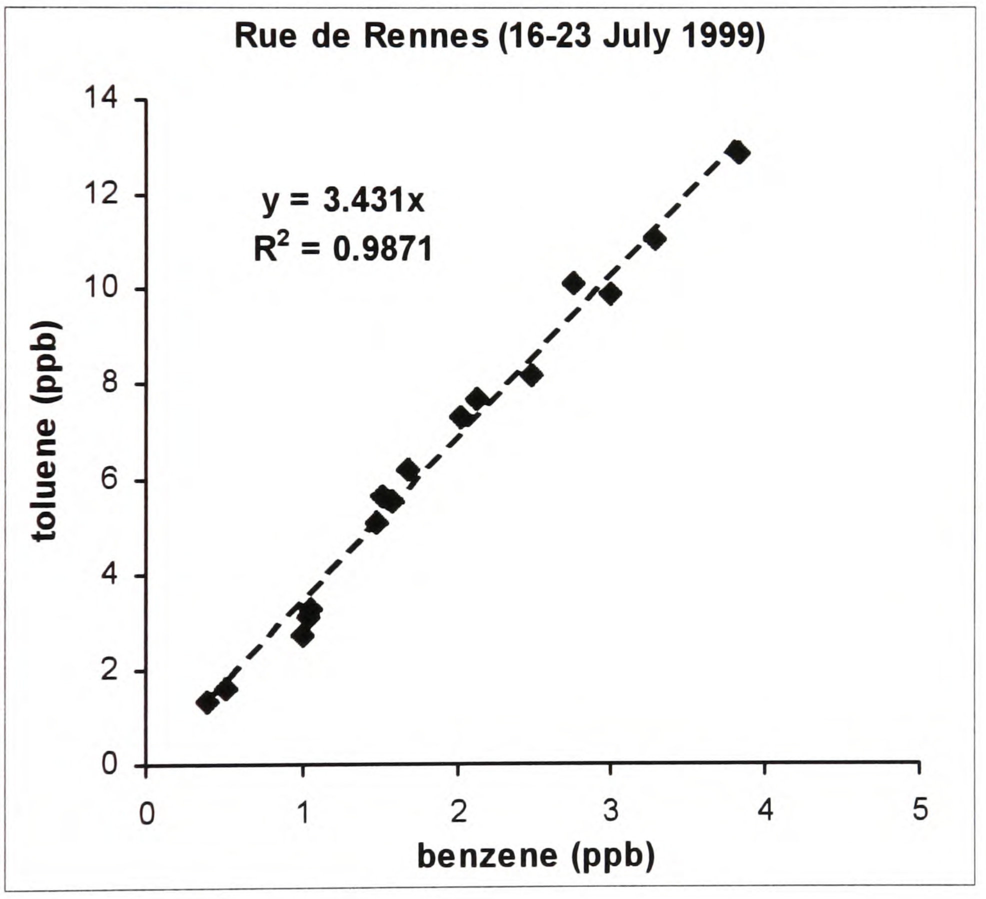


Fig. 3.13: Average toluene (ppb) vs. benzene (ppb) concentrations measured with passive sampling in Rue de Rennes.

Wind flow and dispersion conditions

Due to the vortex formation inside the canyon during perpendicular or near-perpendicular wind conditions (90°-150° and 270°-330°), a downward airflow was established on the windward side of the street (especially for synoptic winds above 2 m/s) and an upward flow on the leeward side (Fig. 3.14).

In addition, an elastic-type reflection of the wind off the windward wall of the canyon was detected for synoptic winds greater than 2 m/s. Using roof-level wind data from Montsouris station, a set of street level wind directions was calculated according to the elastic reflection assumption. A good agreement ($r = 0.70$) between measured and calculated wind directions was finally observed for winds above 2 m/s (Fig. 3.15).

The influence of the synoptic wind speed and direction on the dispersion of pollutants at street level is illustrated in Fig. 3.16. It can be seen that the lower traffic normalised CO concentrations occurred for synoptic wind directions from 270° to 360°, thus for windward flow. No clear dependence of the normalised concentrations on the wind speed was observed in this graph.

The influence of the synoptic wind direction on pollutant dispersion within the canyon was also illustrated on the pollution roses plotted for CO and NO_x (Fig. 3.17). Again, hourly mean CO and NO_x concentrations, normalised with respect to the wind speed and traffic volume, were assigned to the corresponding synoptic wind directions and mean concentrations were calculated for each wind direction sector. Both roses demonstrated a clear dependence of pollution levels on the synoptic wind direction. Fig. 3.17 shows that, keeping the other factors constant, winds parallel or near-parallel to the street axis (i.e. from NE and SW directions) favoured pollution build on the kerbside, while perpendicular winds (i.e. from NW and SE directions) provided better dispersion conditions. Furthermore, it can be observed that normalised concentrations were higher at least by a factor of 2 for winds blowing from SE (i.e. leeward flow) than for winds coming from NW (i.e. windward flow).

Spatial variability

Using BTX as indicators, strong concentration gradients were identified in the horizontal and vertical sense within the canyon during weekdays and a weekend (Fig. 3.18). The highest average benzene concentrations were detected within the pedestrian breathing zone (height = 1.5 m), on the side of the street which was

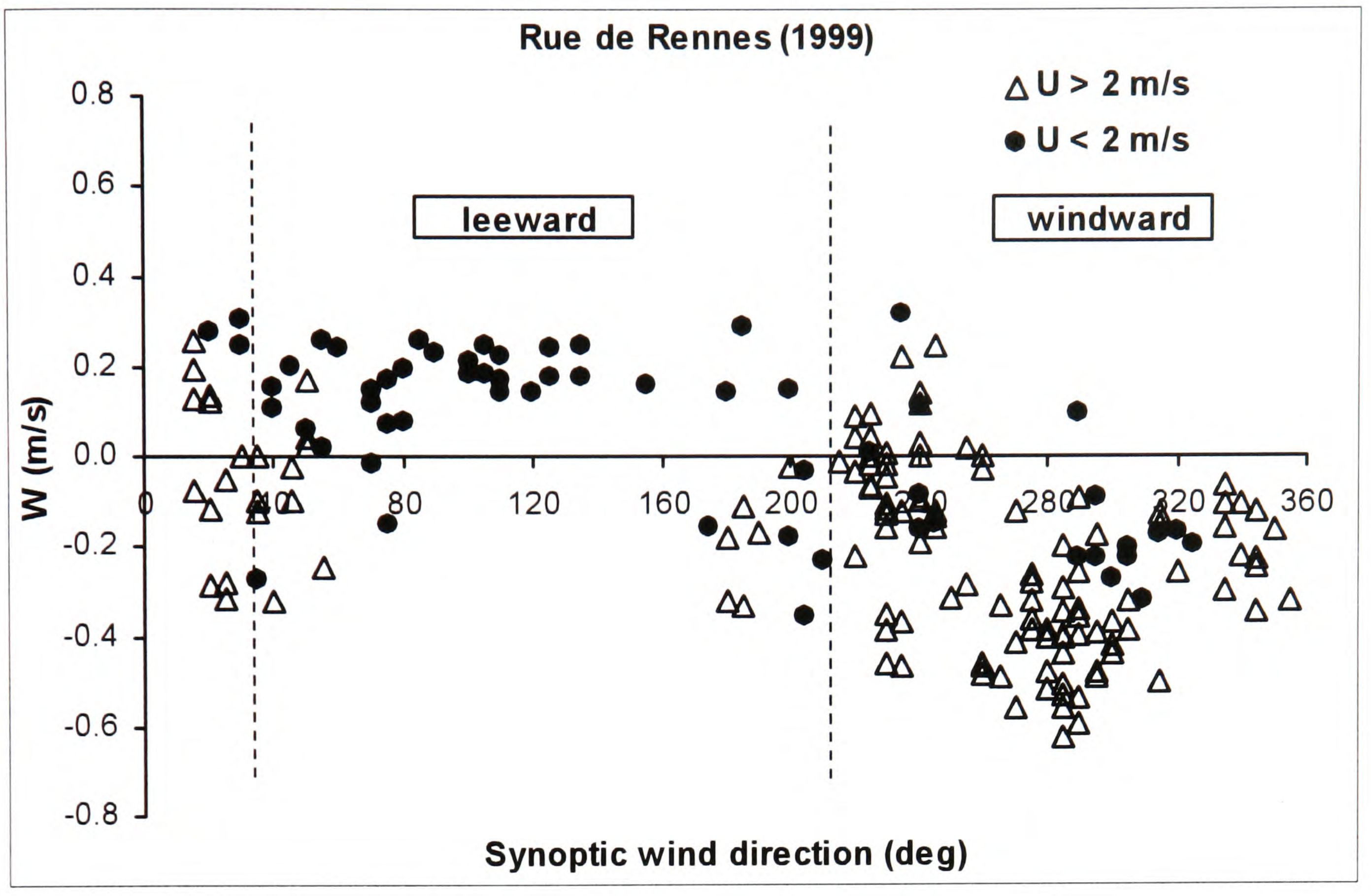


Fig. 3.14: Vertical street level wind speed (W) vs. synoptic wind direction in Rue de Rennes (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

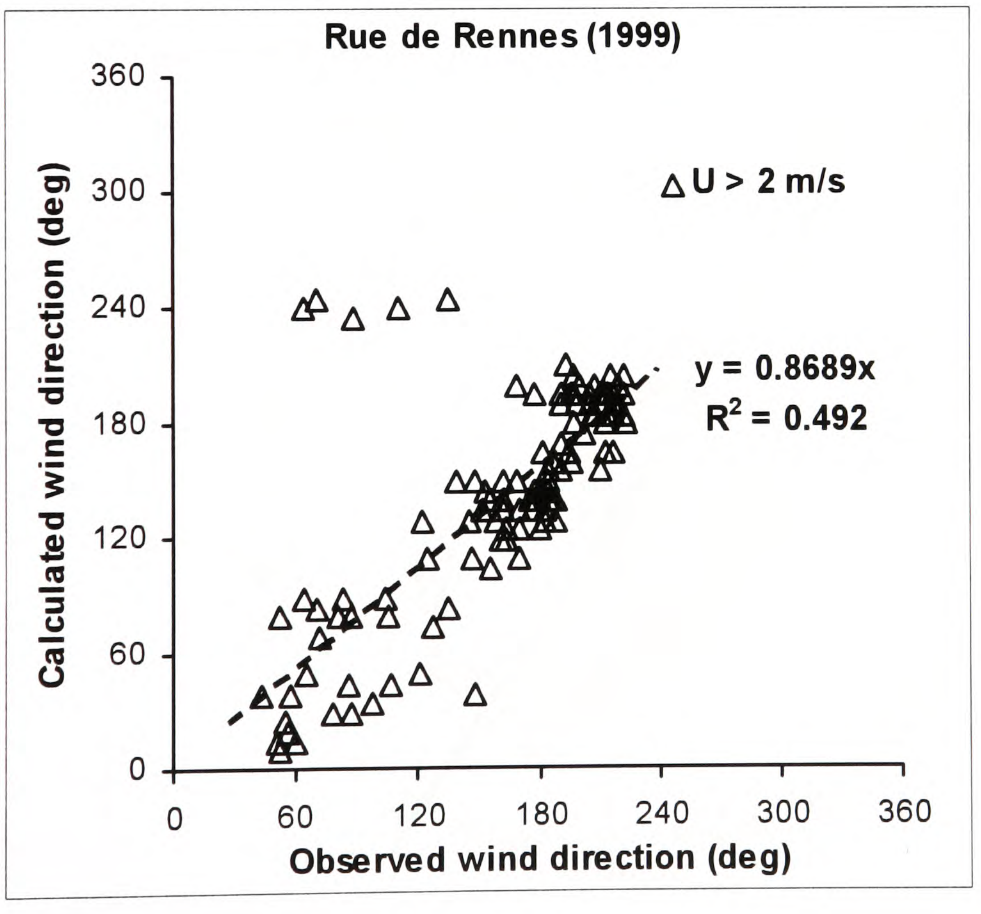


Fig. 3.15: Calculated wind directions: $[Dir]_{local} = 2\,[\text{Street bearing}] - [Dir]_{synoptic}$ vs. observed street level wind directions in Rue de Rennes.

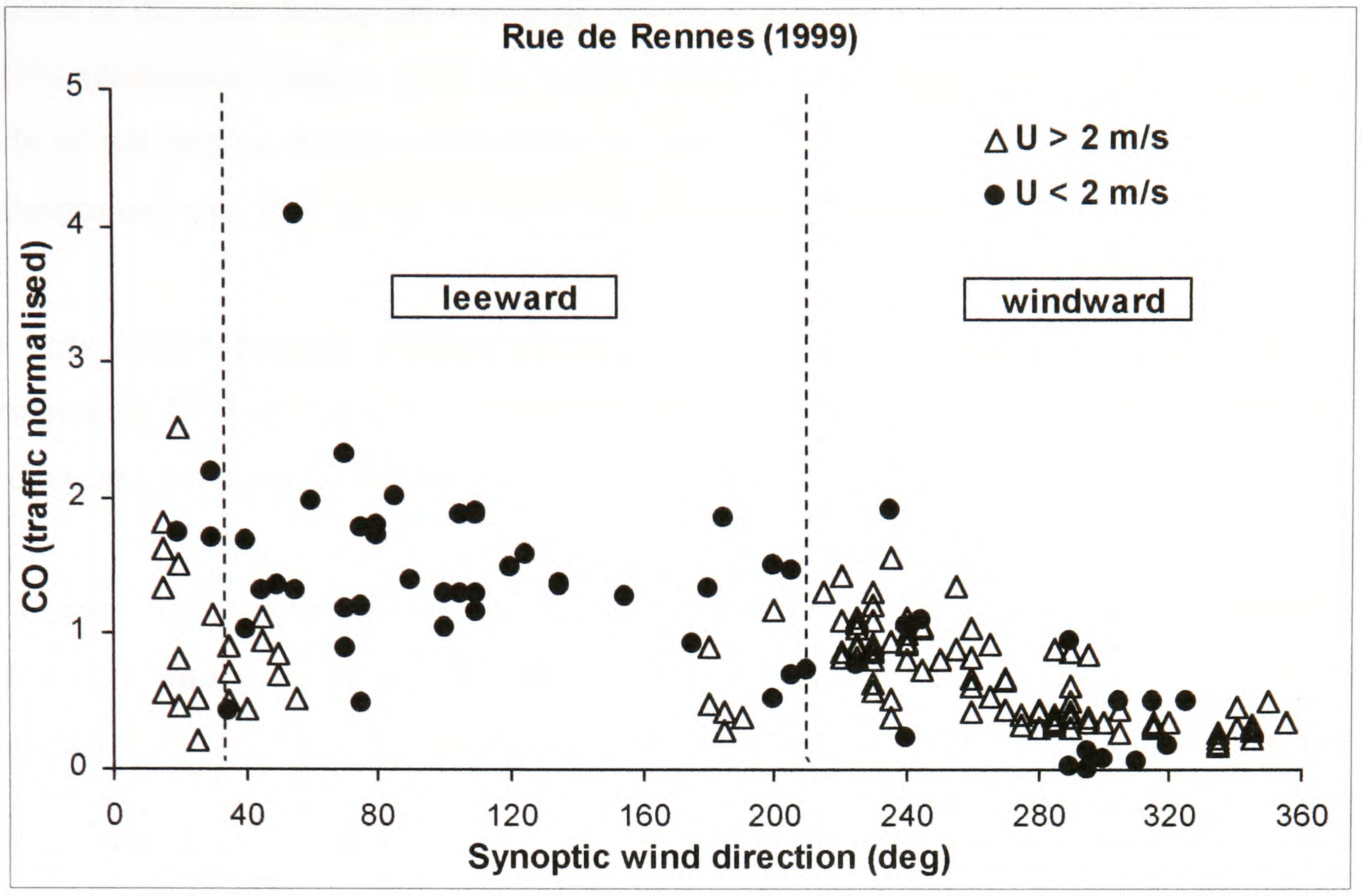


Fig. 3.16: Traffic normalised CO concentrations vs. synoptic wind direction in Rue de Rennes (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

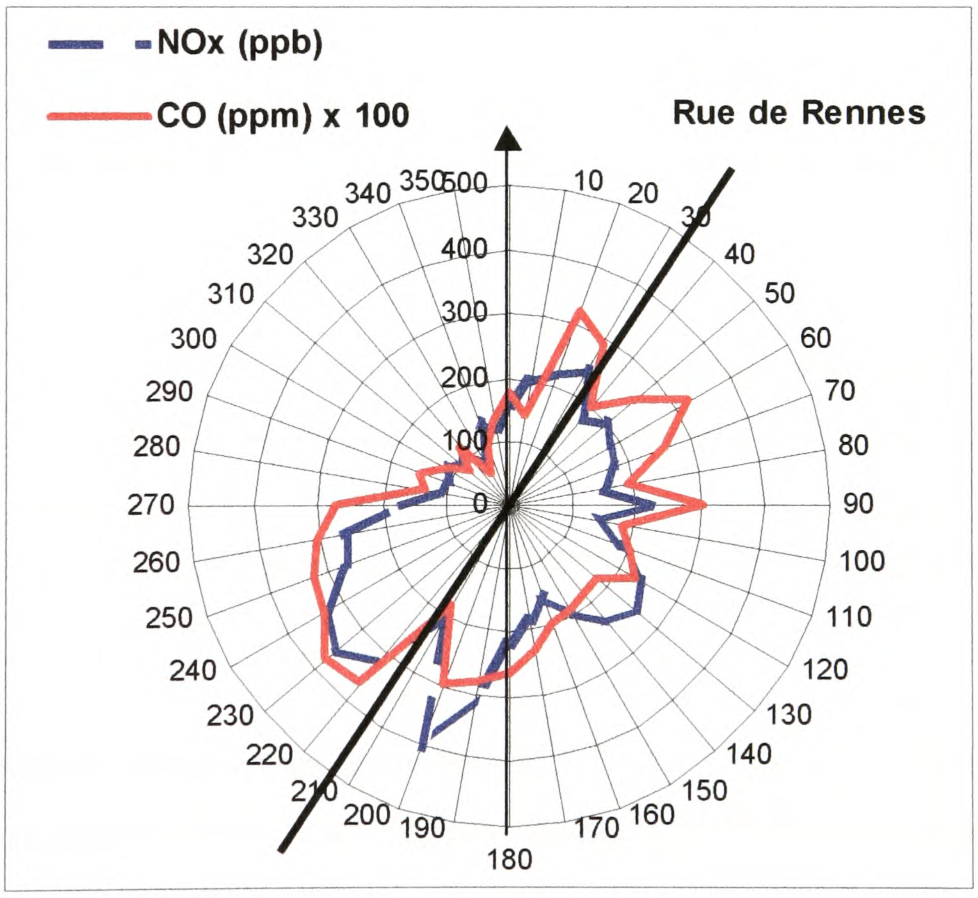


Fig. 3.17: Traffic and wind speed normalised CO and NO_x concentration roses in Rue de Rennes. (The heavy straight line indicates the direction of the street.)

leeward for most of the time during the sampling period. The leeward values were 34% (1st floor), 40% (5th floor) and 65% (pavement) higher than the values measured at approximately the same heights on the windward side of the canyon during the weekdays. During the weekend, the crossroad increase in ambient benzene concentrations was 50% on the 1st floor and 55% on the pavement.

A significant reduction in benzene concentrations along with the height was observed. The weekly benzene averages measured on 5th floor balconies were approximately 30% lower than the values observed on 1st floor level during the same sampling period, on both the sides of the canyon.

Finally, the kerbside concentrations were 3 to 6 times higher than those detected in the adjacent urban background locations during the working week and 2 to 4 higher during the weekend. The same trends were also identified for the rest of the VOC compounds sampled with passive tubes during the campaign.

Temporal variability

Interestingly, the highest CO concentrations in Rue de Rennes were detected on Saturday 17th of July (Fig. 3.19), on a day when the total traffic volume was slightly lower than during the weekdays. These relatively high values can be mainly attributed to the low winds (1-2 m/s) blowing over Paris on that day. During the following weekdays (19-23 July) winds became stronger (2.5-5.5 m/s) and as a result ambient concentrations were reduced. The recorded CO levels were consistent with the passive VOC measurements, which were also higher during the weekend than during the rest of this campaign.

Moderate photochemical activity was observed in Rue de Rennes. NO concentrations peaked during the morning rush hours (Fig. 3.20). The NO_2 peak levels were delayed a few hours, as expected due to the time required to oxidise NO. Ozone gradually increased during the day, producing higher concentrations in the afternoon. This is because most of the NO must be converted in NO_2 before ozone builds up in the atmosphere (Boubel et al., 1994).

The relatively low winds and the strong insolation on Sunday 18th, in combination with low NO emissions due to reduced road traffic, gave rise to a minor ozone episode on that day with values almost reaching 50 ppb in early afternoon. On another occasion (20-21 July), relatively high ozone levels were observed during late evening and night. This might be explained by long-distance transport of pollutants. It is not unusual for large quantities of ozone formed in rural areas during daytime to be advected over long distances, reaching urban centres during late afternoon and evening.

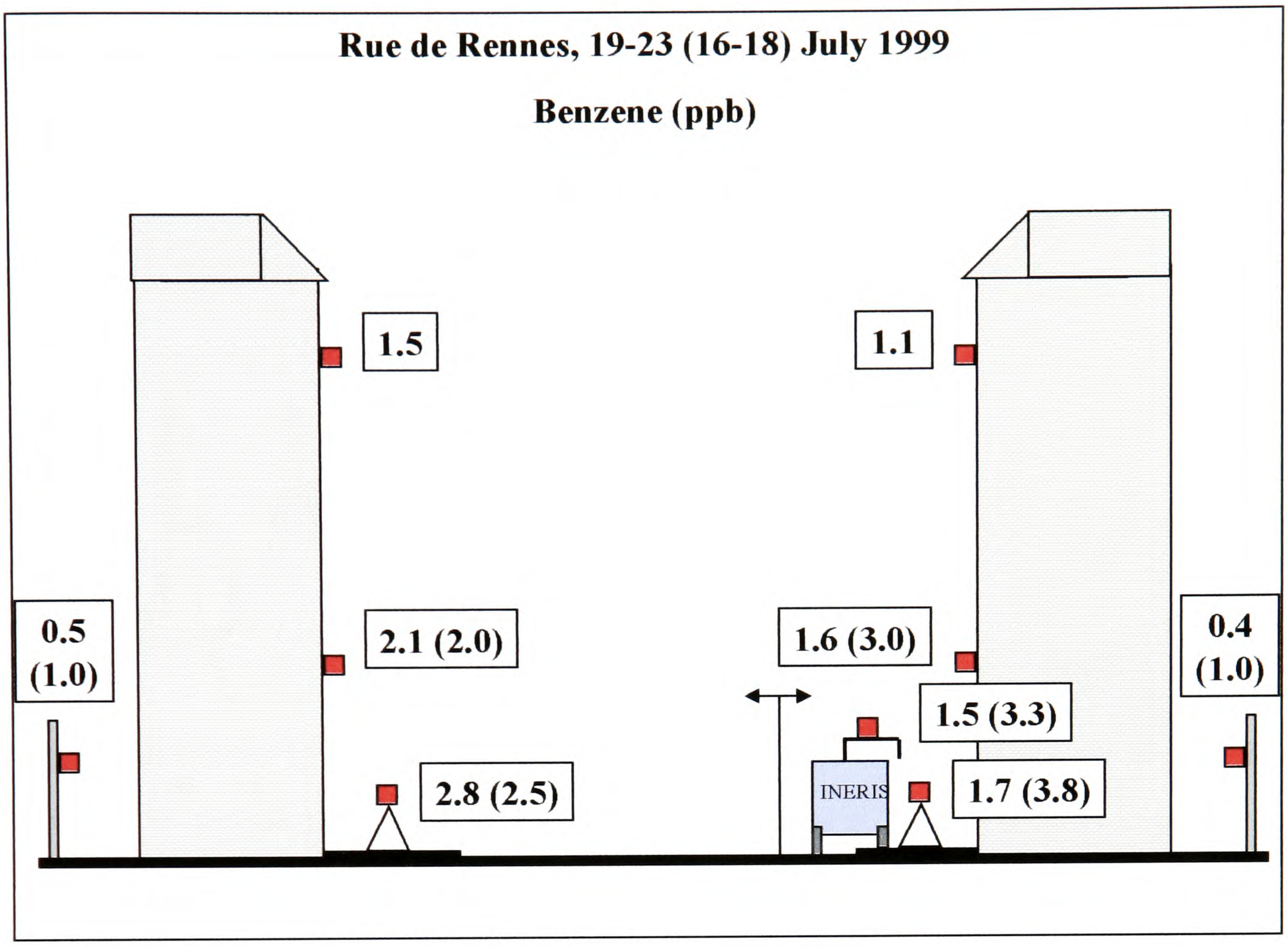


Fig. 3.18: Benzene (ppb) concentrations measured with passive sampling in Rue de Rennes during weekdays (19-23 July); and a weekend (16-18 July, values in parenthesis).

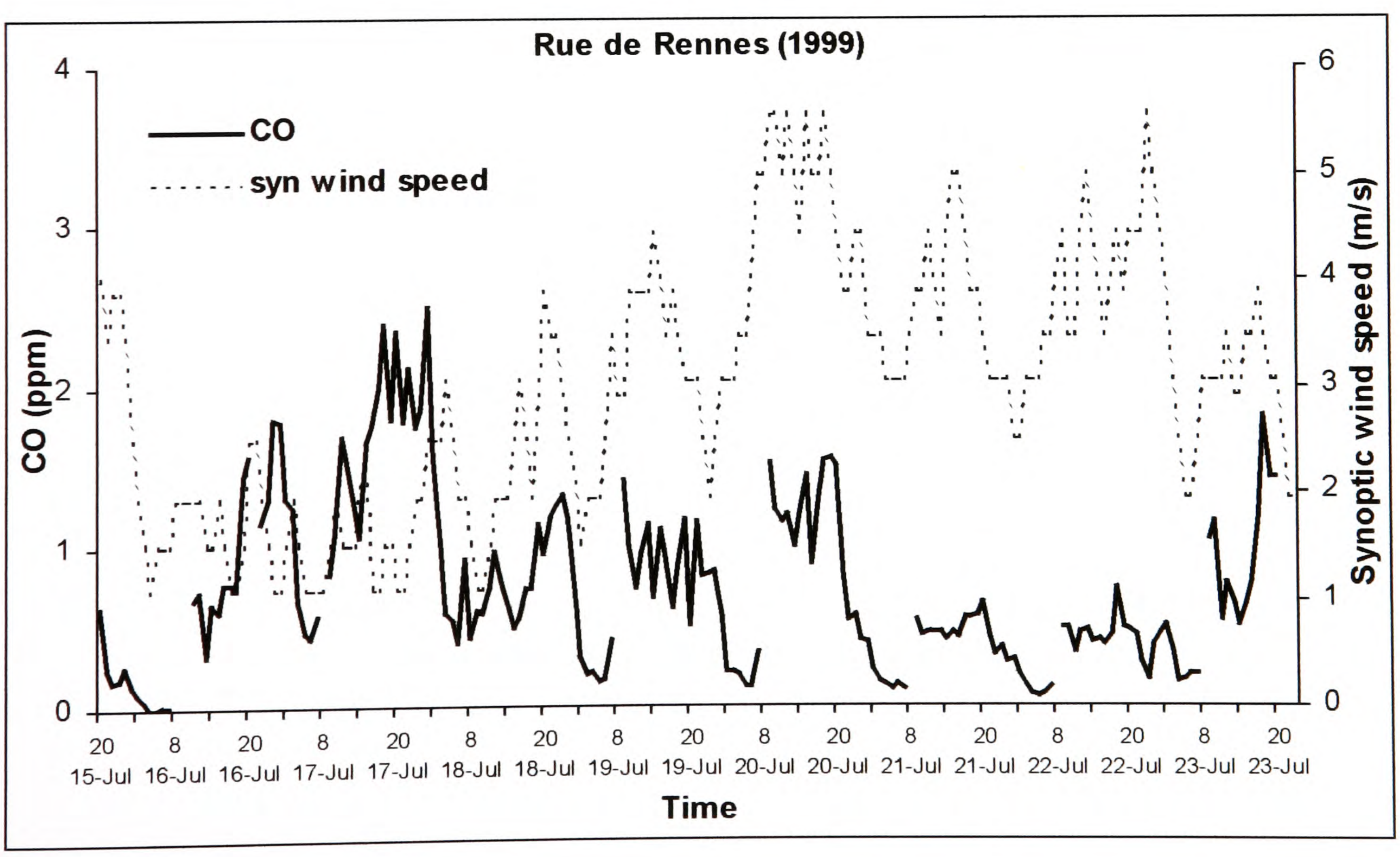


Fig. 3.19: Hourly mean CO (ppm) concentrations and synoptic wind speeds in Rue de Rennes.

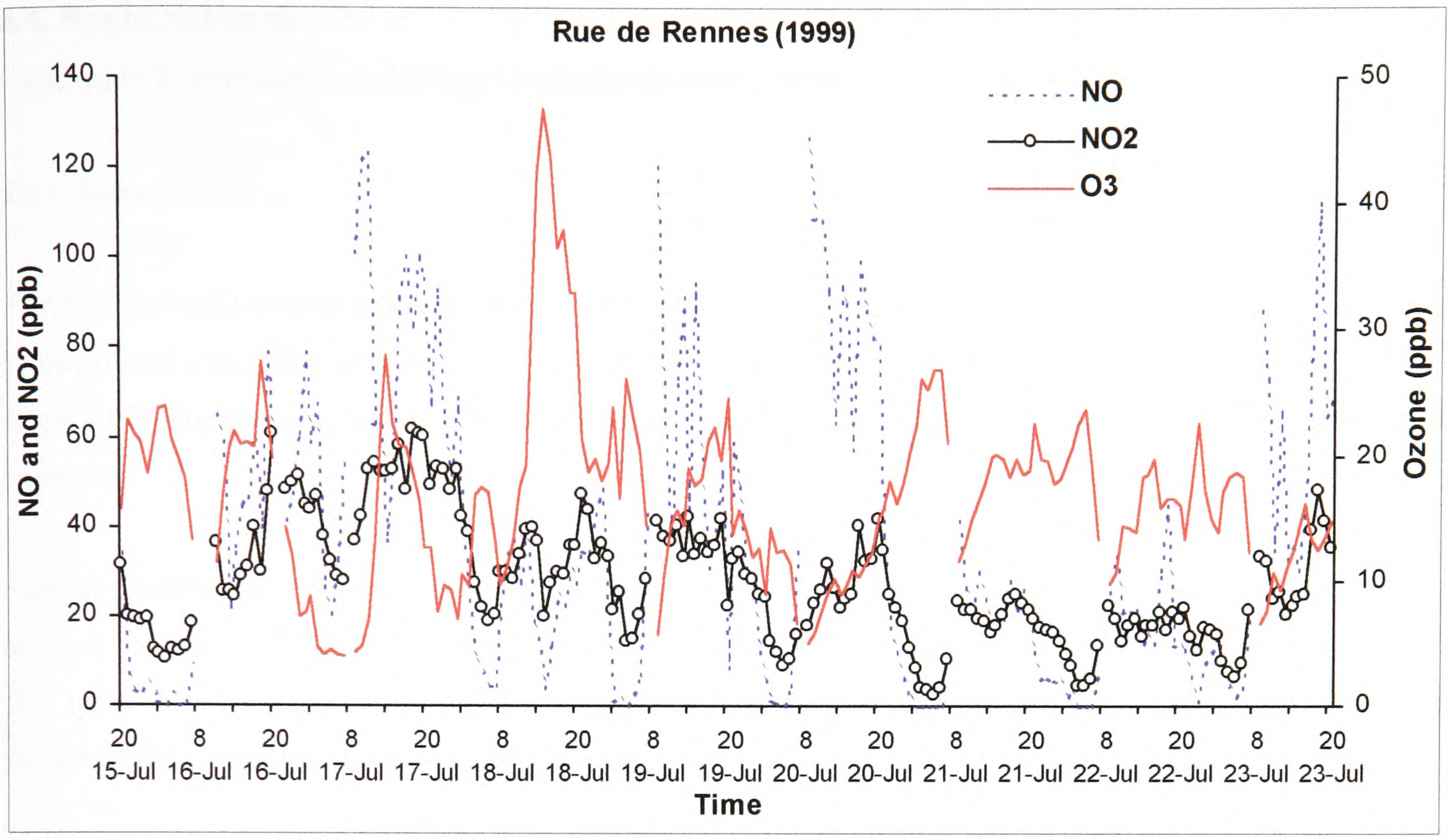


Fig. 3.20: Hourly mean NO, NO_2 and O_3 (ppb) concentrations observed in Rue de Rennes

3.4. Route Nationale 10

Case study 3: Intensive monitoring campaign in a motorway petrol station (November 1999)

3.4.1. Introduction

Petrol is a complex petroleum product mainly consisting of paraffins, olefins, naphthenes and aromatic hydrocarbons containing from 3 to 11 carbon atoms. The exact composition of the fuel varies according to its origin. BTX compounds, mainly occurring in unleaded petrol, can be treated as tracers for traffic-generated pollution.

From a regulatory point of view, the European Commission (2000) has established a limit value of 5 $\mu g/m^3$ for benzene in ambient air. In France, the "Loi sur l'Air" (MATE, 1998) determines an air quality objective value of 2 $\mu g/m^3$ for the same compound. Fuel storage and delivery activities in service stations have also been subject of EU regulation due to the VOC releases involved (European Commission, 1994).

Tanks containing petrol can emit VOC vapours due to filling and emptying activities (i.e. displacement losses), as well as to due changes in temperature and atmospheric pressure (i.e. breathing losses). Every time an empty tank is filled, the corresponding air volume saturated with petrol vapour is displaced into the atmosphere. Displacement losses can increase both occupational exposure to VOC in the immediate proximity of the pumps (for people working in the station, car drivers and passengers), and population exposure in the surroundings of the station (Periajo et al., 1997; Kearney and Dunham, 1986).

For the reduction of these emissions, vapour recovery devices can be put in place. These systems return the VOC saturated volume of air that has been displaced from the tank being filled to the tank being emptied during the delivery of the fuel. The set of equipment used for vapour recovery during the loading of a storage tank is called *Stage 1* control, while the system used for the same purpose during the refuelling of a vehicle tank is called *Stage 2* control.

3.4.2. Description of the site

An intensive monitoring campaign was carried out in a modern petrol station by the RN 10 motorway, near the small town of Prunay in the south of Rambouillet (Yvelines, France) in November 1999.

The specific petrol station was selected for this campaign because of the availability of modern refuelling facilities and vapour recovery systems (which could be turned on and off), and because of the relatively constant quantities of petrol sold.

The monitoring site was on a flat, well ventilated terrain, where the only sources of BTX were the service station and the RN10 motorway (a typical linear source). The service station was located on the west side of the N – S oriented RN10 motorway. The average traffic volume during measurements was 1,400 veh/hour and the total volume of fuel (diesel + petrol) sold 11,000 l/day.

3.4.3. Sampling strategy

The concentration of a pollutant at a given location and time equals the summation of the contributions from different emission sources. The major factor that determines the dispersion of gaseous pollutants in the atmosphere is the wind velocity and its related turbulent effects.

Considering the case of the petrol station, the concentration C_i of a pollutant at a given location can be expressed in the following way:

$$C_i = C_b + C_p + C_m \qquad (3.3)$$

Where C_b is the background concentration of the pollutant, C_p is the concentration due to emissions released within the station, and C_m is the contribution from vehicle traffic in the proximity of the station. C_m and C_p are expected to vary as a function of time and distance from the source. The total concentration C_i of the pollutant, as well as the background contribution C_b, can be measured using adequate equipment (e.g. passive samplers). By contrast, the other two independent contributions C_p and C_m need to be calculated.

In order to study the efficiency of the vapour recovery system, one should be able first to quantify the variation of VOC releases within the station, and then attribute this variation to different factors (vapour recovery, traffic volume, accidental spillage, weather conditions, composition of the fuel, etc.). In addition, the variation of C_p should be significant compared to C_b and C_m values.

Diffusive Perkin Elmer (PE) tubes were used to detect BTX concentrations at 20 sampling locations, which can be classified in three different "levels" of proximity to the source: (I) next to the fuel pumps, (II) within their surrounding environment, and (III) in the background. The samplers placed nearer the source (level I) were located in pairs at two different heights above the ground ($h_1 = 0.2$ m and $h_2 = 2.0$ m).

Furthermore, continuous CO and wind measurements (24/24 h) were taken at 3.5 m height above the ground using the mobile air quality monitoring unit and the two anemometers (i.e. ultrasonic and mechanical). The exact location of the monitoring equipment is indicated in Fig. 3.21.

Manual field notes of traffic volume and average vehicle speed were taken during the campaign. Finally, the quantity of petrol sold in the station was automatically recorded by fuel pump meters.

The duration of the campaign was two weeks. During the first week (2-9 November 1999), the vapour recovery system of the station was operating, while during the second week (15-22 November 1999) it was disconnected.

3.4.4. Analysis and interpretation

Mapping of monitoring results

Weekly mean BTX concentrations for all sampling locations at 2 m height are presented in Fig. 3.22. The concentrations detected near the ground (at 0.2 m height) at proximity level I were significantly higher than the rest of the measurements. For this reason, they were treated separately.

The results shown in Fig. 3.22 were used to plot a number of iso-concentration maps (Fig. 3.21 and Appendix II). Concentration mapping was carried out using the kriging method with a linear variogram model.

The cartography of the pollutants (i.e. iso-concentration lines on a site map) is an efficient means of visualising sampling results and interpolated values. Amongst the different interpolation methods applicable to this case, kriging was considered as the most appropriate one because of the irregular distribution of the sampling points and the possibility of using a variogram model (Journel, 1989).

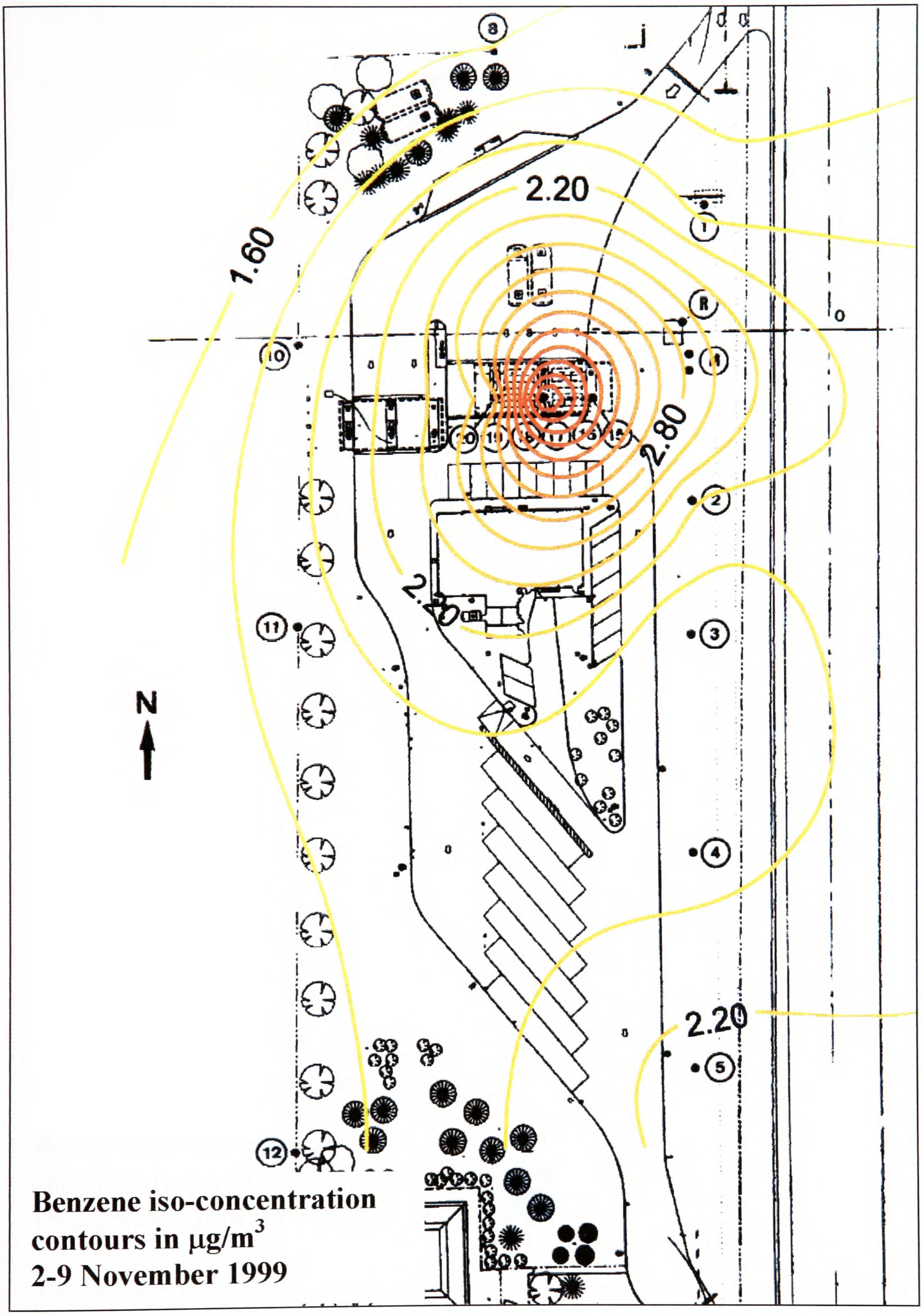


Fig. 3.21a: Benzene iso-concentration contours (µg/m^3) in the RN10 petrol station while Stage 2 vapour control was operating (2-9 November 1999) - R: mobile monitoring unit, M: meteorological monitoring equipment, 1-20: passive sampling locations (plan view).

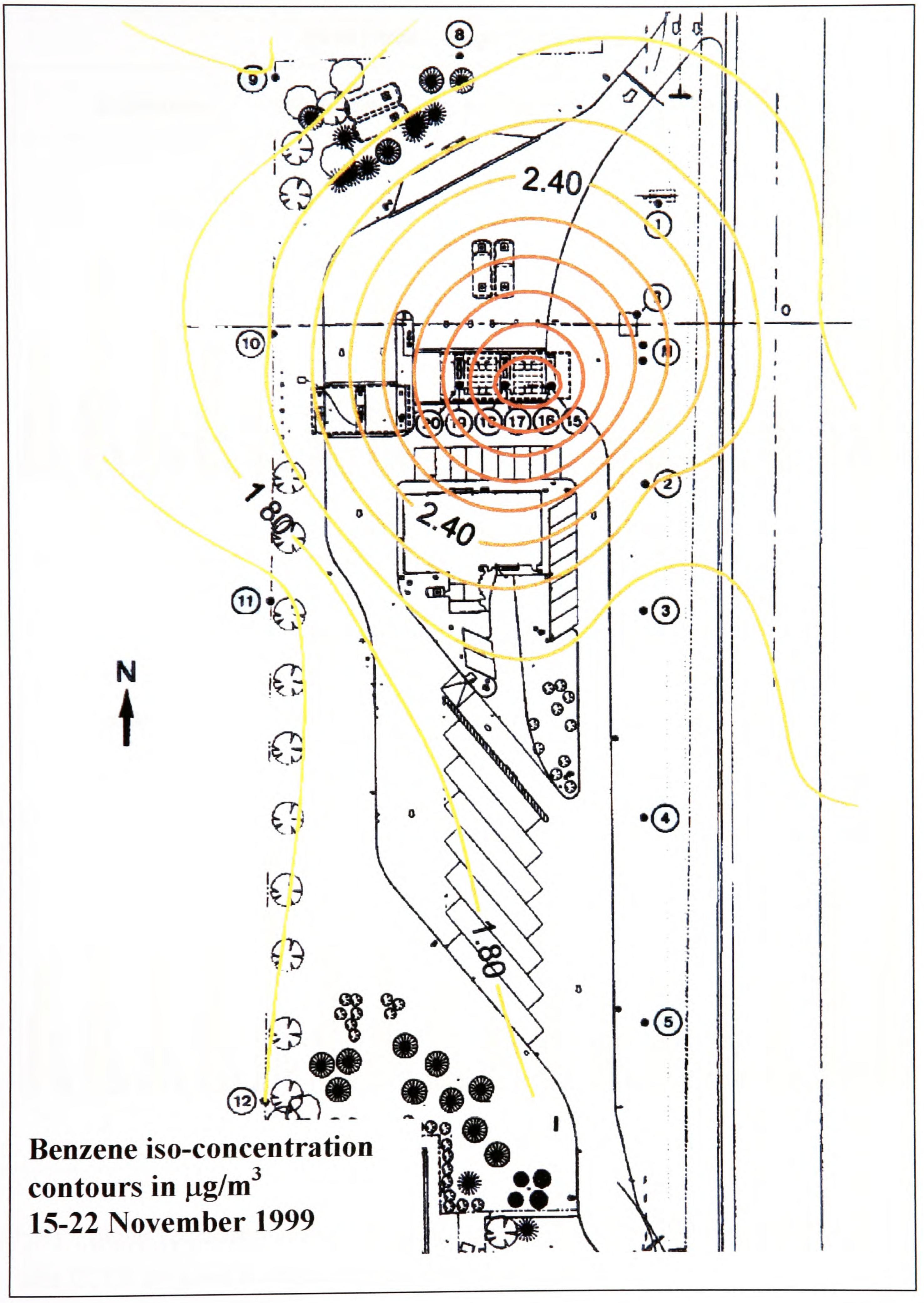
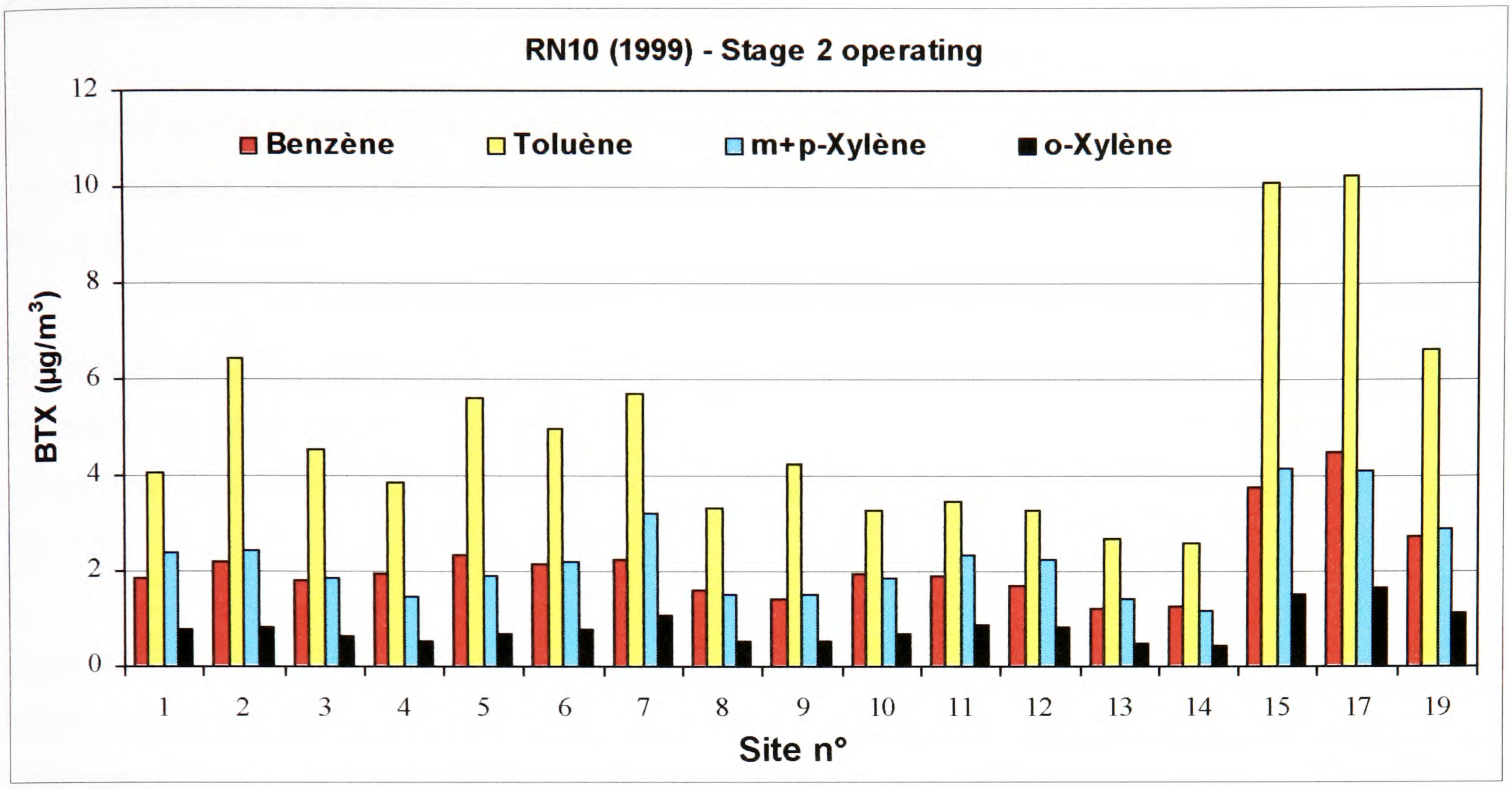


Fig. 3.21b: Benzene iso-concentration contours (µg/m^3) in the RN10 petrol station while Stage 2 vapour control was disconnected (15-22 November 1999) - R: mobile monitoring unit, M: meteorological monitoring equipment, 1-20: passive sampling locations (plan view).



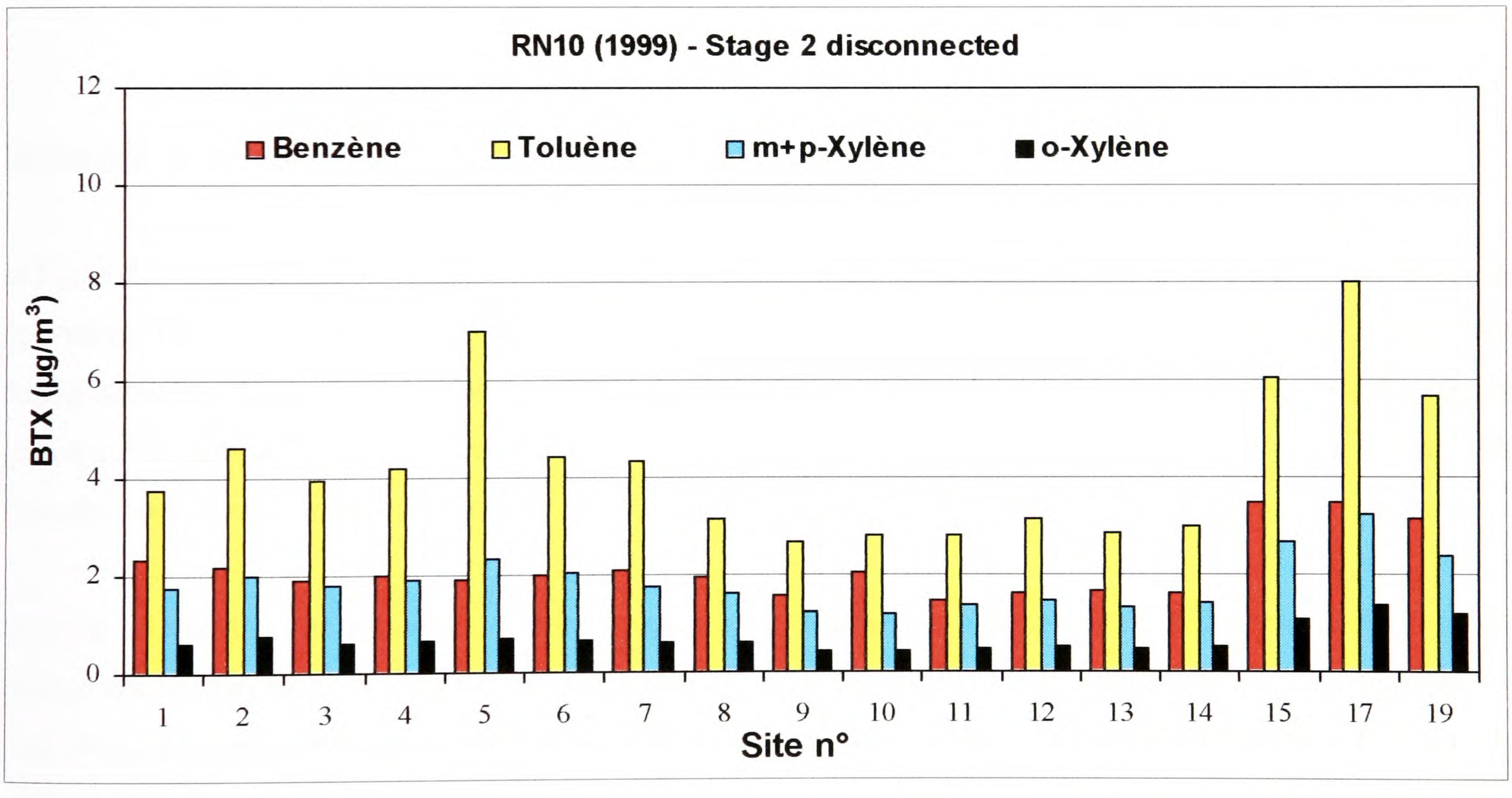


Fig. 3.22: BTX ($\mu g/m^3$) concentrations at 19 passive sampling sites in the RN10 petrol station.

Level I: sites 15, 17, 19; Level II: sites 1-12; Level III: sites 13, 14.

(a) Stage 2 control operating (top), (b) Stage 2 control disconnected (bottom).

Relationship between pollutants and spatial variability

The regular profile of the BTX concentrations measured in all sampling locations (with and without Stage 2 control operating) revealed the common origin (i.e. petrol combustion and evaporation) of VOC emissions (Fig. 3.22).

The background BTX values, which were (as expected) much lower than those measured near the station or the motorway, were approximately the same for both level III locations (i.e. sites 13 and 14). Near the fuel pumps (level I), concentrations were up to a factor of 4 higher than the background values, as it can be seen in Fig. 3.22.

Large differences in BTX concentrations were also detected between the sampling locations of different height near the pumps (level I). The concentrations observed close to the ground (h_1 = 0.2 m) were significantly higher than those measured at h_2 = 2.0 m. This could be explained by the short distance between (h_1) samplers and car exhausts, as well as by the occurrence of accidental spillage losses of fuel on the ground.

Dispersion conditions

In Fig. 3.22, it can be seen that BTX concentrations were in fact higher while the vapour recovery system was operating. This rather unexpected result can be explained by the differences in meteorological conditions during sampling. In particular, mean wind speed was higher (increasing from 2.5 to 3.4 m/s) during the second period of the campaign, when the vapour recovery system was disconnected. The prevailing wind was coming from the west during both the periods of the campaign.

In order to establish the influence of wind speed on the ambient pollution levels, BTX concentrations were plotted against the reciprocal of the wind speed (Fig. 3.23) for the two different periods of the campaign (a: with Stage 2 control, b: without Stage 2 control). Only the measurements of higher temporal resolution are presented in this graph. These correspond to the sampling points near the pumps (level I), where passive tubes were replaced every 48 hours. The concentration variations observed in Fig. 3.23 could be mainly attributed to the changes in wind speed, since the station was all the time upwind with respect to the RN 10 motorway and the quantity of fuel sold remained almost constant.

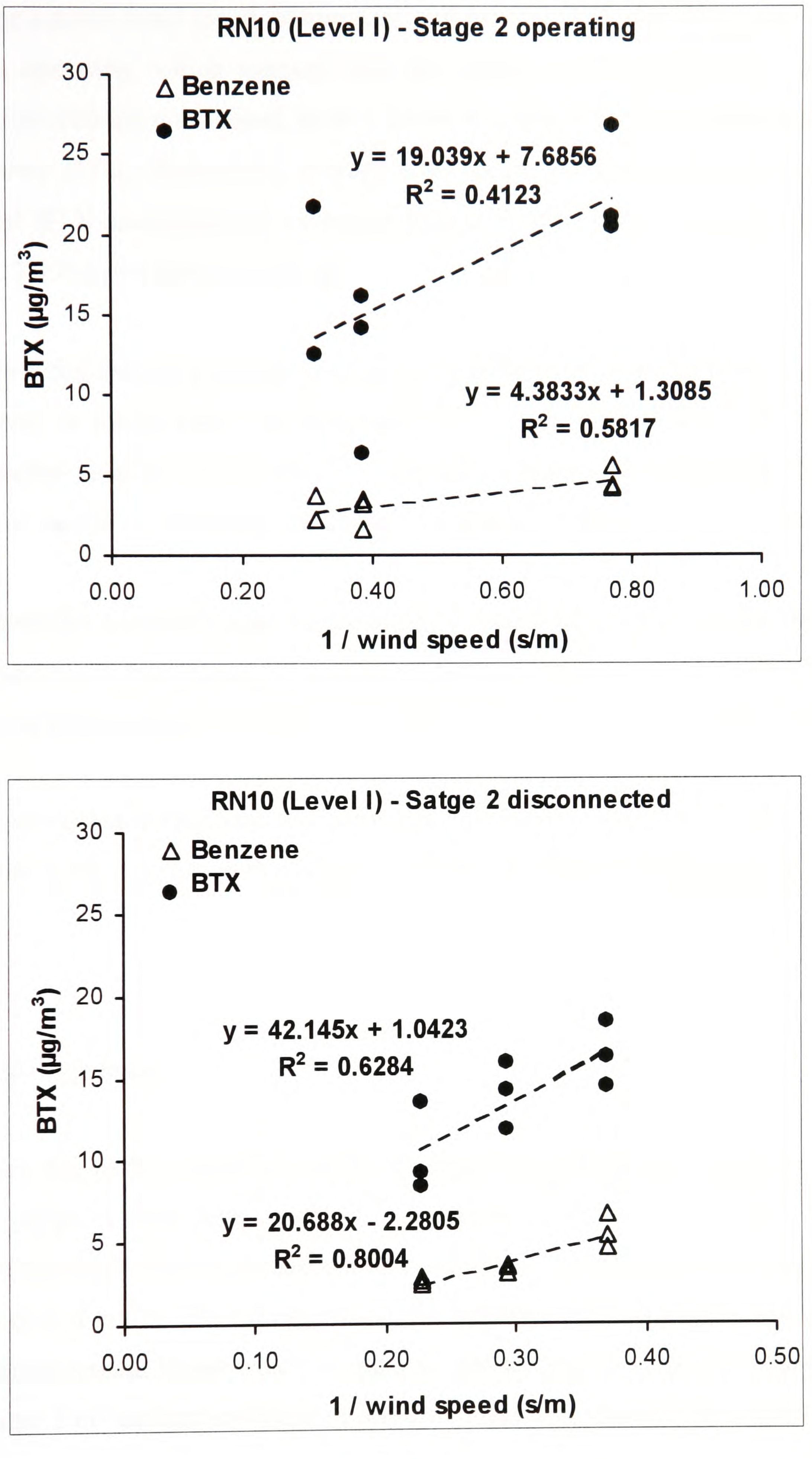


Fig. 3.23: BTX and benzene ($\mu g/m^3$) concentrations vs. 1 / wind speed in the RN10 petrol station (a) Stage 2 control operating (top), (b) Stage 2 control disconnected (bottom).

It can be seen that for a given wind speed, benzene as well as total BTX concentrations were lower when the Stage 2 control was operating, which suggests that the system worked efficiently. The efficiency of the system, however, varies with the wind speed, as it is shown in Table 3.3. During the sampling periods without Stage 2 vapour recovery (15-22 November), average wind speeds were always above 2 m/s. For this reason, the benzene and total BTX concentrations corresponding to 2 m/s (Table 3.3) were extrapolated from the values shown in Fig. 3.23b using linear regression.

Furthermore, it appears that there is a critical wind speed value above which the system is no longer effective. While vapour recovery is taking place, benzene reduction is generally higher than total BTX reduction, possibly due to the higher volatility of benzene. Nevertheless, it should be remembered that measurements are not only affected by evaporative emissions, but also by combustion releases from vehicles using the station.

The benzene concentrations measured in the surrounding environment (level II) during both time periods were normalised with respect to the wind speed to reveal the impact of Stage 2 control. It should be noted that there is no need to normalise further, since the station was all the time upwind with respect to the RN 10 motorway (thus traffic volume did not significantly affect measurements) and the quantity of fuel sold remained almost constant. Fig. 3.24 shows that normalised concentrations were lower when the vapour recovery system was operating, which leads to the conclusion that Stage 2 control also reduces pollution in the surroundings of the station (level II).

Displacement and spillage losses

Evaporative emissions due to the loss of few drops of petrol while filling the tank of a vehicle can be easily calculated using a simple closed box model. For example, if 1 ml (≈ 5 drops) of petrol containing approximately 1% of benzene were evaporated within an isolated 2 m^3 envelop of air, that would give rise to a benzene concentration of 4 mg/m^3. On the other hand, the loading of 40 l of petrol from a storage to a vehicle tank would induce displacement losses which would give rise to benzene concentrations of approximately 85 mg/m^3 within the same 2 m^3 isolated envelope of air. This value was deduced by applying Raoult's Law and assuming there was no vapour recovery and that the partial pressure of benzene in the tank was approximately 1 mm Hg. From this simple calculation, it can be concluded that displacement losses during petrol delivery without Stage 2 control are much more significant (about 20 times higher in this example) than small spillage losses.

Table 3.3: Impact of Stage 2 control on BTX concentrations for different wind conditions.

Wind speed (m/s)	Concentrations ($\mu g/m^3$) Stage 2 control OFF		Stage 2 control ON		Reduction (%) = (1-Co/C) x 100	
	Benzene	BTX	Benzene	BTX	Benzene	BTX
3.5	3.6	13	2.56	13.1	29	0
3.3	4	13.8	2.6	13.4	35	3
2	8.06 *	22.1 *	3.5	17.2	56	22

* Extrapolated value

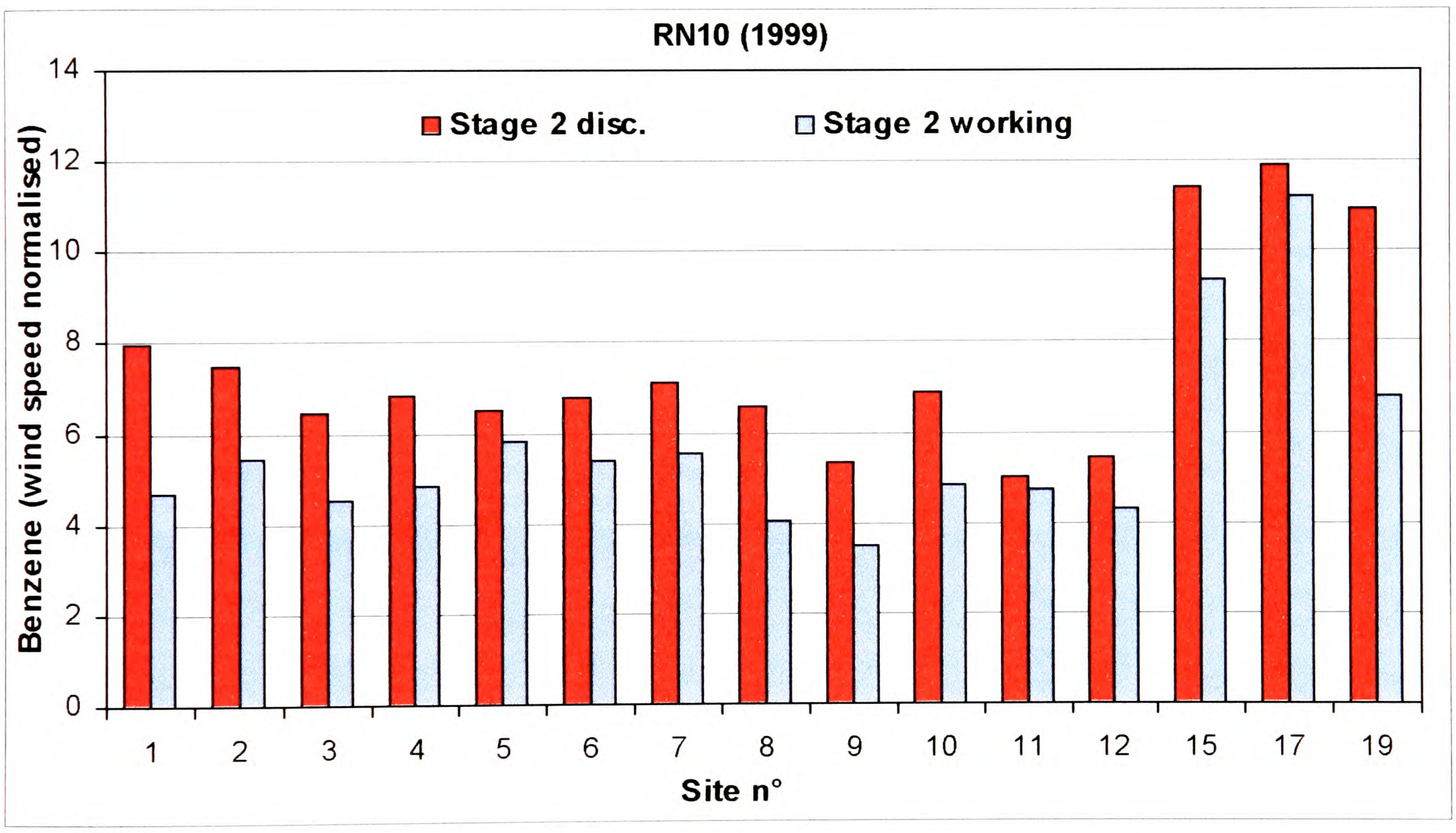


Fig. 3.24: Wind speed normalised benzene concentrations in RN10 petrol station for Stage 2 control operating or disconnected.

3.5. Place Basch

Case study 4: Long-term monitoring campaign in an asymmetric street canyon leading to a major urban intersection (June – December 2001).

3.5.1. Description of the site

A long-term monitoring campaign (June-December 2001) was carried in a complex urban site in Paris, comprising the busy intersection of Pl. Basch and the large asymmetric canyon of Av. Leclerc.

Av. Leclerc is a busy road axis linking Bd. Périphérique (i.e. the major ring motorway of Paris) in the south with the city centre in the north. Roadside measurements were taken within the straight road segment between Rue Sarrette (south) and Pl. Basch (north), and around Pl. Basch where four avenues are intersecting (Av. Leclerc, Rue d'Alésia, Av. Maine, and Av. Moulin). In addition, urban background measurements were carried out in Montsouris Park, at approximately 800 m distance in the SE of the roadside monitoring site.

Av. Leclerc has four traffic lanes in the direction towards Pl. Basch and two lanes towards Bd. Périphérique. In the road segment between Rue Sarrette and Rue Daudet, there is a parking lane on the east side, separated by a narrow traffic island from the rest of the street (Fig. 3.25). Parking is not allowed at any other location in Av. Leclerc. Finally, there are large pavements (10 m wide on the east side and 8 m wide on the west side) and rows of big trees on both sides of the street and on the traffic island.

The height and shape of the urban canopy surrounding Av. Leclerc is not uniform, since there is a mixture of traditional (usually) six-storey buildings, modern tower blocks, and few detached houses. The large road segment between Rue Sarrette and Rue Daudet can be considered as an asymmetric street canyon, with the buildings on the east side being approximately 12 m taller than those on the west side.

3.5.2. Sampling protocol

Diffusive RPE samplers were exposed to ambient air during 28 consecutive seven day periods from the 3rd of June to the 17th of December 2001. The samplers were placed at the ten different roadside locations indicated in Fig. 3.25 and one urban background site (Montsouris Park). At the locations A, B, D, E, F, G, and I, samples were taken at 1.3 m distance from the kerb.

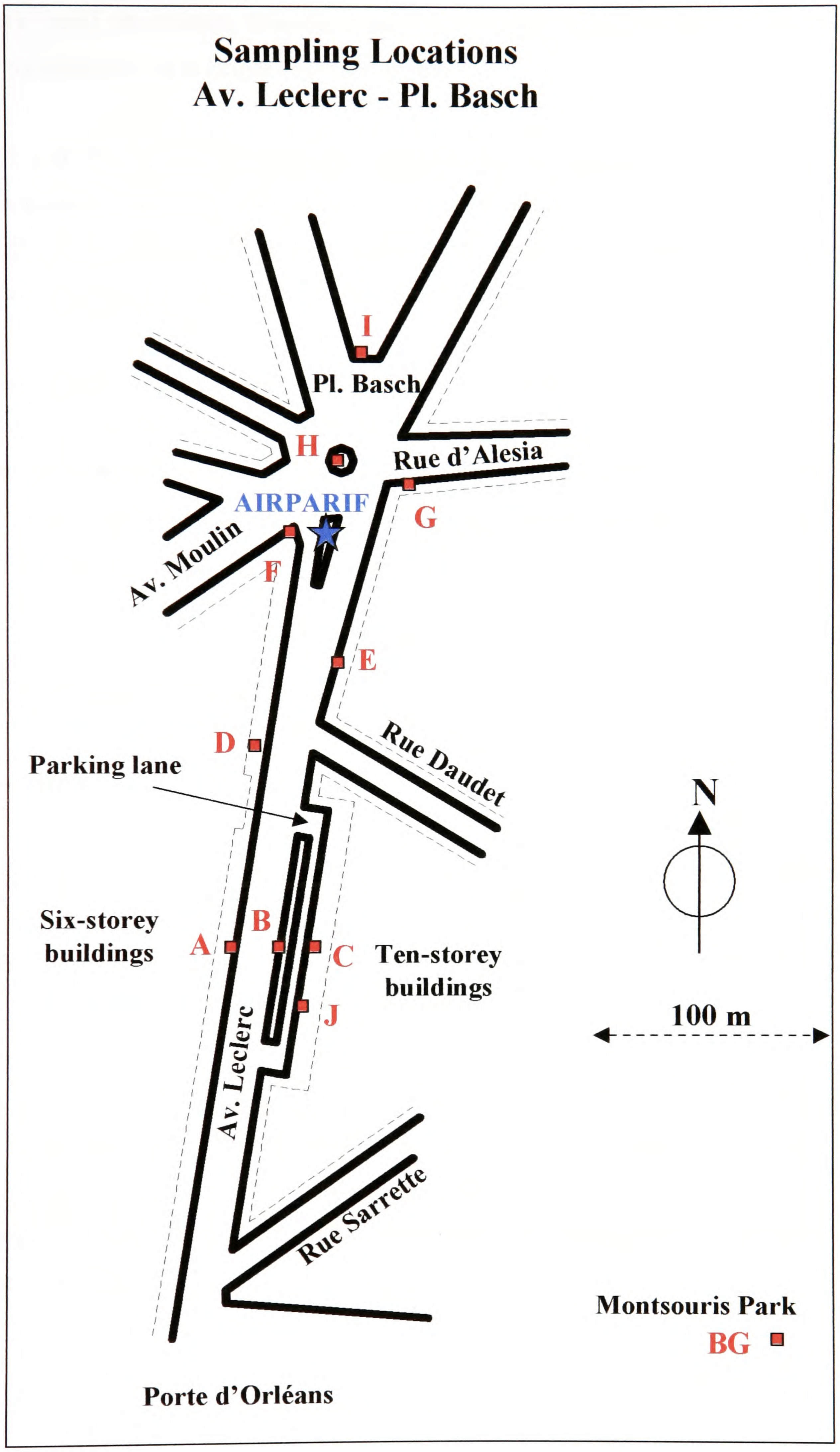


Fig. 3.25: Layout of the AIRPARIF monitoring site and passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park (plan view - Montsouris Park not in scale).

At the locations C and J, the distance from the nearest kerb was 2 m, and for location H (roundabout) 4.5 m. All samplers were placed at 2.6 m height above the ground.

Continuous CO and NO_x measurements were obtained from a roadside air quality monitoring station (AIRPARIF) permanently operating in Pl. Basch. As indicated in Fig. 3.25, the exact location of this station is on a narrow traffic island in the middle of a pedestrian crossing in Av. Leclerc, very near Pl. Basch. Its sampling inlet was at approximately 2 m height above the ground.

Synoptic meteorological data (i.e. wind speed and direction, atmospheric pressure and temperature) were obtained from the Montsouris and Orly weather stations. Local street-level wind measurements were only available during an intensive monitoring campaign carried out on the same site (see Section 3.6). Finally, manual vehicle counts were taken during 37 hours and compared with the data obtained from the automatic traffic monitoring network.

3.5.3. Analysis and interpretation of results

Relationship between pollutants

The passive sampling measurements at different locations within Av. Leclerc and Pl. Basch during seven months revealed a very strong correlation between benzene and toluene concentrations (Fig. 3.26). The experimental toluene to benzene ratio (by volume) during the whole campaign was approximately equal to 4.0.

Furthermore, a strong correlation between weekly benzene, toluene, ethylbenzene, m+p-xylenes and o-xylene concentrations observed at location H (roundabout) and the average CO values obtained from the AIRPARIF station during the same time periods was established (Table 3.4). It should be noted that from all diffusive benzene measurements, those taken at location H showed the best correlation (r = 0.85) with the continuous CO values from AIRPARIF.

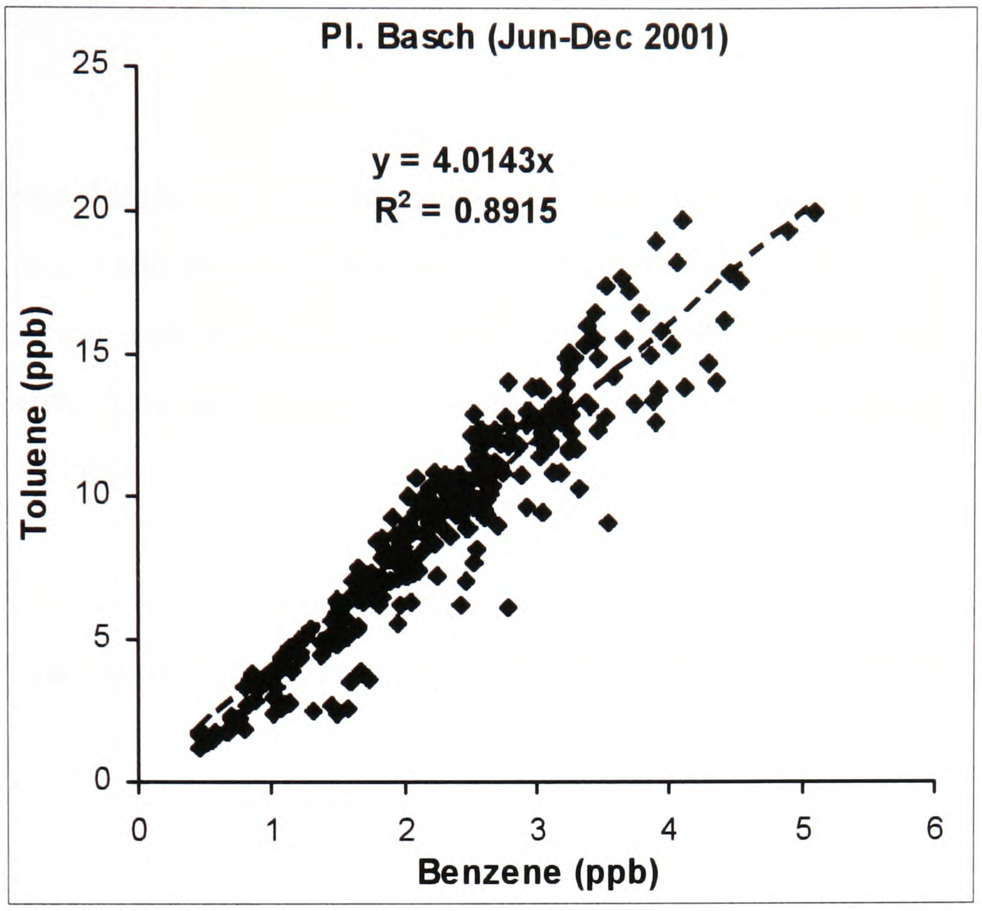


Fig. 3.26: Average toluene (ppb) vs. benzene (ppb) concentrations measured with passive sampling in Pl. Basch (Jun-Dec 2001).

Table 3.4: Correlation coefficients of BTX concentrations measured with passive sampling and CO values from AIRPARIF in Pl. Basch.

	Benzene	Toluene	Ethylbenzene	m+p-Xylene	o-Xylene
CO	0.85	0.86	0.81	0.86	0.86
Benzene		0.92	0.83	0.89	0.92
Toluene			0.94	0.96	0.96
Ethylbenzene				0.98	0.97
m+p-Xylene					0.99

Wind conditions

Using data obtained from Montsouris weather station (3-hour mean values), wind roses were plotted for the whole year 2001 (Fig. 3.27) and separately for each month of this year (Fig. 3.28). It can be seen that the wind was predominantly blowing from SW directions in the region of Paris in the year 2001 and especially for the months of January, March, April and October. By contrast, the prevailing wind was from NE directions during May, November and December.

Higher wind speeds were observed in Montsouris station during the winter season, although the monthly averages did not vary substantially throughout the year (Fig. 3.29). In most cases, winds remained within the range of 2 - 4 m/s.

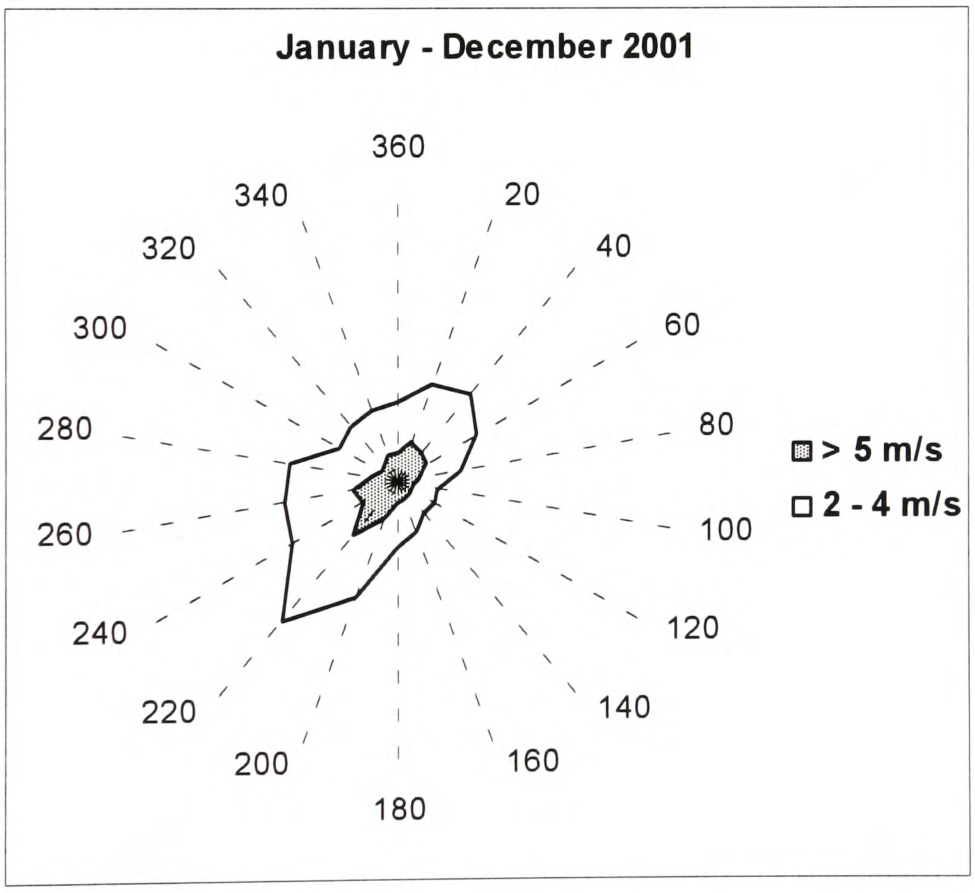


Fig. 3.27: Wind rose for the year 2001 in Paris (Montsouris station).

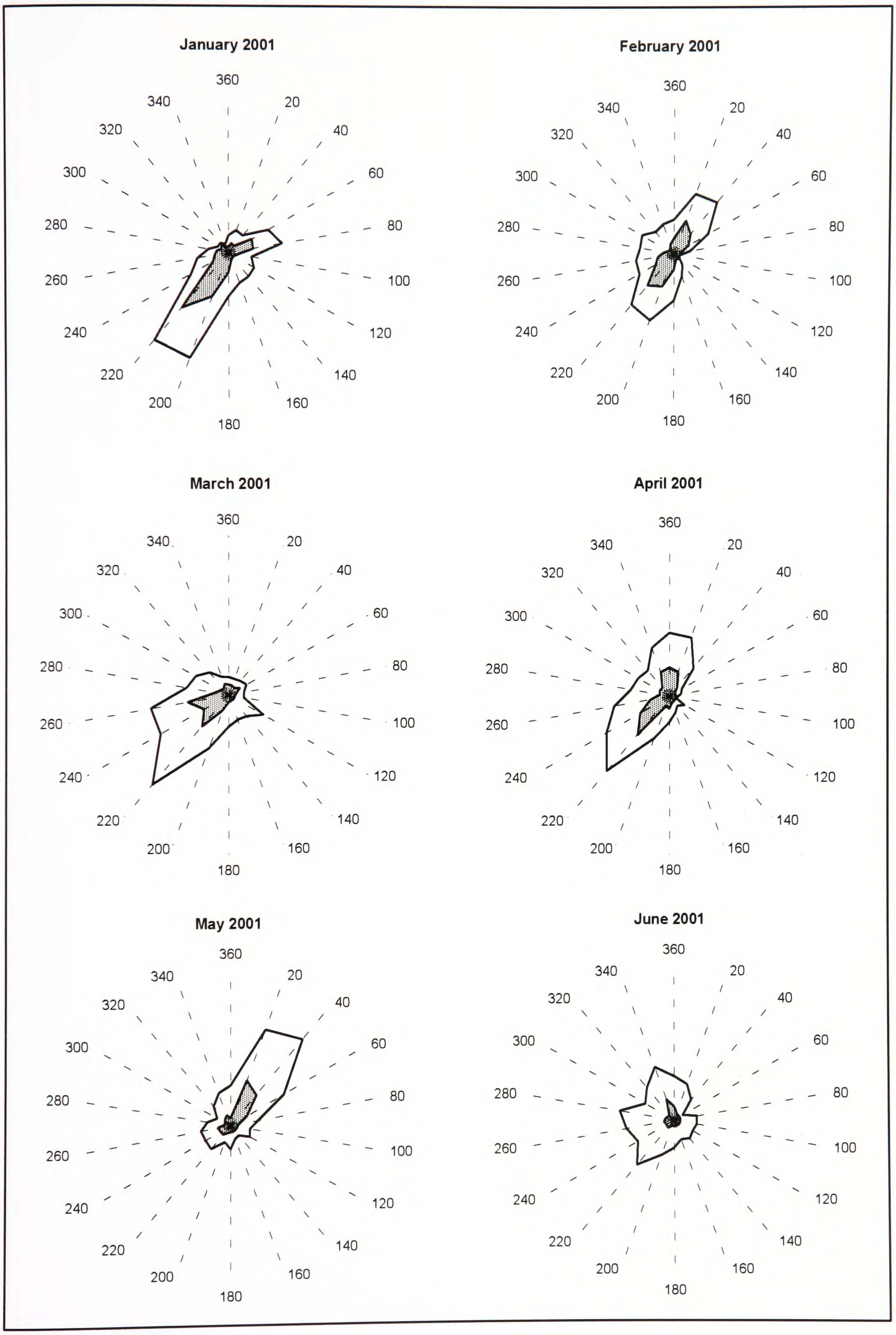


Fig. 3.28a: Monthly wind roses for the year 2001 in Paris (Montsouris station)

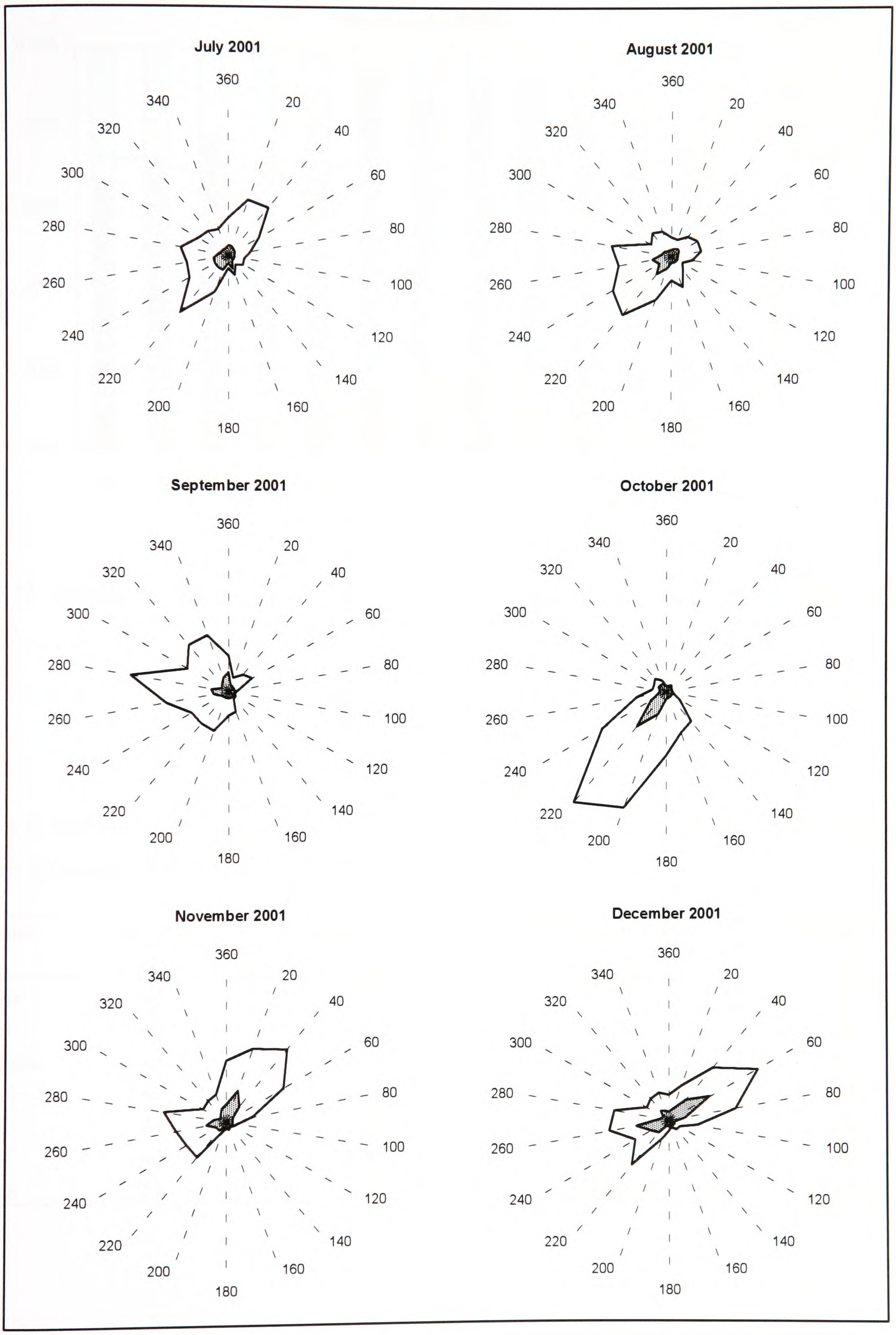


Fig. 3.28b: Monthly wind roses for the year 2001 in Paris (Montsouris station)

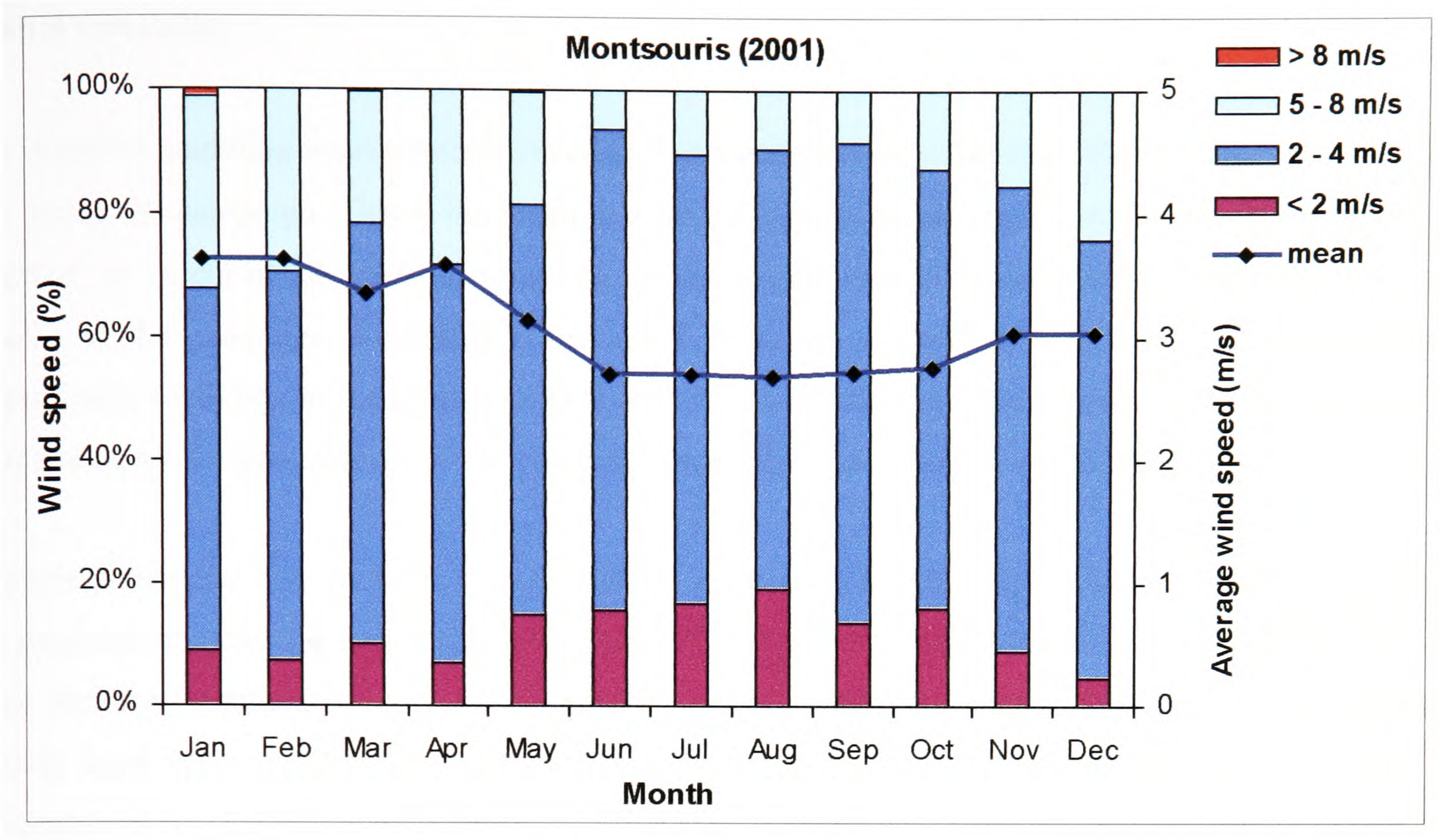


Fig. 3.29: Monthly average wind speed and wind speed categories for the year 2001 in Paris (Montsouris station).

Table 3.5: Monthly variation of the spatial coefficient of variation (SCV) in Pl. Basch and Av. Leclerc during the year 2001 calculated using diffusive benzene measurements.

Month	Samples N°	Benzene average ppb	Standard deviation ppb	Spatial CV %
June	40	2.10	0.57	27
July	43	2.30	0.57	26
August	46	2.24	0.48	24
Sept	44	2.48	0.74	34
Oct	44	3.14	0.77	25
Nov	40	2.45	0.71	26
Dec	22	2.51	0.69	27

Spatial variability

The diffusive sampling measurements revealed that location H (roundabout) was the most polluted sampling site during the campaign. This is the location were the highest weekly mean concentrations of benzene were detected for 16 out of 28 weeks of sampling, as well as the highest benzene value averaged over the seven months of the campaign. Locations D, A and F were the second, third and fourth most polluted sites respectively, according to the same criteria (Fig. 3.30). It should be noted that these three locations are on the west site of Av. Leclerc, which was more often leeward than the opposite side of the canyon.

The lowest benzene concentrations were observed at locations C and J, on the mostly windward pavement of the asymmetric canyon of Av. Leclerc (i.e. east side). Apart from the prevailing wind direction, the relative large distance (approximately 12 m) of receptors C and J from the main traffic lanes of Av. Leclerc may explain these relatively low concentrations. Furthermore, it should be remembered that the width of the pavements in Av. Leclerc is not uniform and that vehicle emissions within the parking lane were much lower than in the main traffic lanes.

The spatial coefficient of variation (SCV), which is a statistical measure of the spatial variability of air pollution, was then calculated by dividing the standard deviation of benzene concentrations observed at different roadside locations by the mean value calculated for the same time period. Coefficients of variation were initially calculated for each seven-day sampling period and then averaged over a month.

As it can be observed in Table 3.5, the monthly SCV did not vary significantly during the measurements, except for the month of September. The spatial variation of benzene during this month was 30% higher than the average value for the rest of the campaign. This might be explained by the westerly winds (thus perpendicular to Av. Leclerc) that were prevailing over Paris during that period, inducing steep crossroad gradients within the asymmetric canyon.

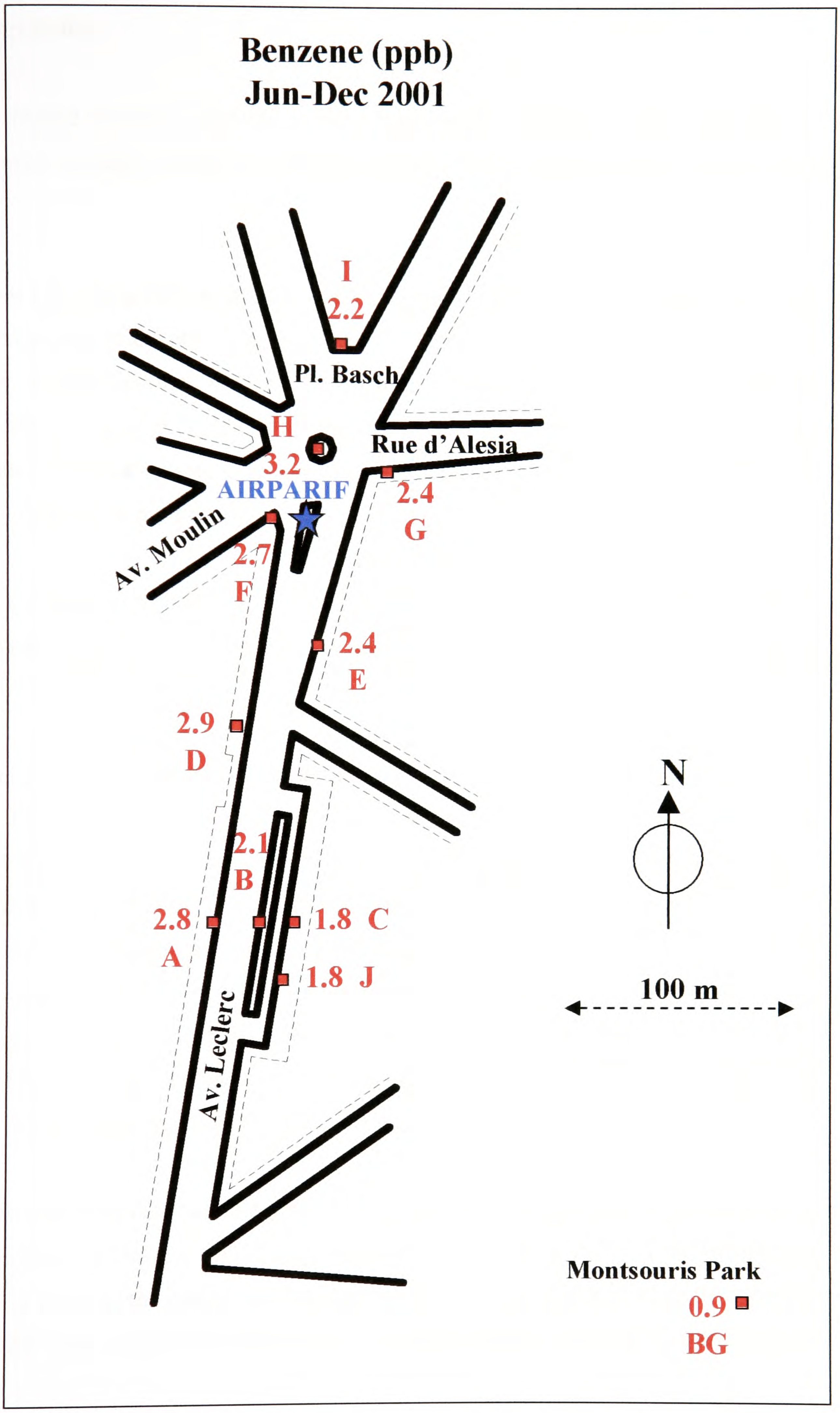


Fig. 3.30: Average benzene concentrations (ppb) during seven months (Jun-Dec 2001) at 11 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park (plan view).

Temporal variability

There is no marked seasonal variation in the observed air pollution levels in Pl. Basch and Av. Leclerc, although in most roadside sampling locations slightly lower benzene levels occurred during the summer months.

At all locations (including the background site), the highest benzene concentrations (seven-day averages) were observed in the month of October (Fig. 3.31), due to the relatively low winds coming from the S and SW, thus parallel to Av. Leclerc (average wind speed = 2.8 m/s). These wind conditions probably induced substantial wind flow along Av. Leclerc, transporting polluted air masses from Bd. Périphérique towards Pl. Basch. The lowest benzene concentrations were at most roadside locations observed in August, mainly due to the reduced traffic density during this month.

The benzene averages corresponding to the roadside sampling locations in Pl. Basch and Av. Leclerc followed roughly the same monthly variation pattern as the CO concentrations recorded at the AIRPARIF station in Pl. Basch (Fig. 3.32).

The crossroad gradients at locations A – B and D – E gave evidence of a canyon effect taking place within Av. Leclerc. From June to November, monthly averages at the locations A and D on the west side were respectively higher than those measured at the locations B and E at approximately the same level on the east side of the street. That was probably because A and D locations were most of the time on the leeward side of the canyon under the W and SW winds prevailing during this period. Only in December, the crossroad concentration gradients were reversed within Av. Leclerc. During this month, benzene averages were higher at receptor B than at receptor A and at receptor E than at receptor D, probably due to the prevailing NE winds. This was confirmed by the fact that concentrations at receptors C and J on the east side of the street took also their highest values during the month of December.

During all (but one) seven-day sampling periods, background benzene concentrations at Montsouris Park were significantly lower than those detected at all roadside locations (Fig. 3.31). Monthly averages were within the range of 0.5 – 1.0 ppb at the background, except for the month of October when the benzene average value in Montsouris Park was 1.5 ppb. On the roadside, monthly benzene averages ranged between 1.3 and 4.0 ppb (Fig. 3.32).

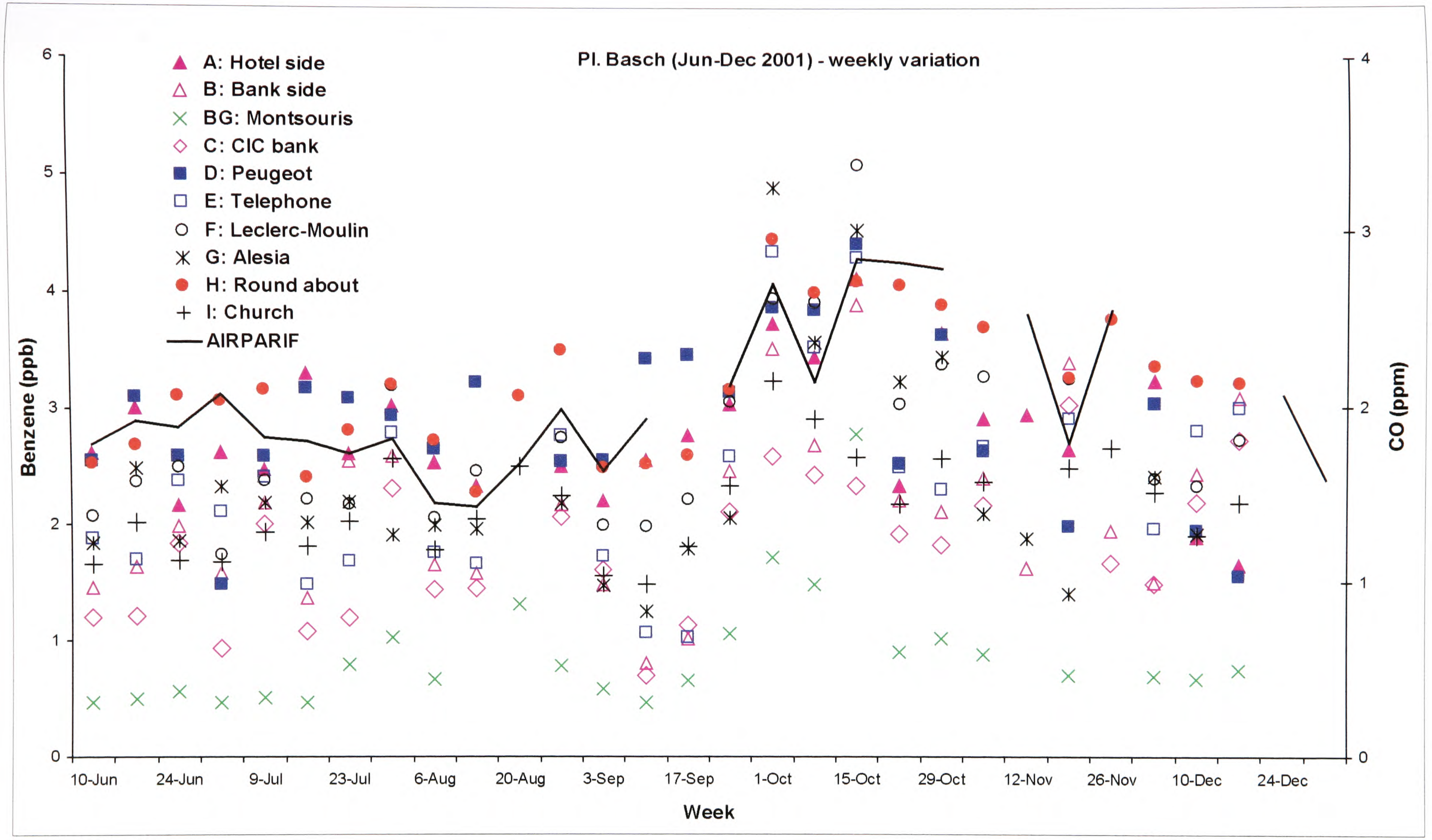


Fig. 3.31: Weekly benzene (ppb) variation in 10 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park; CO (ppm) variation in the AIRPARIF monitoring station in Pl. Basch (Jun-Dec 2001).

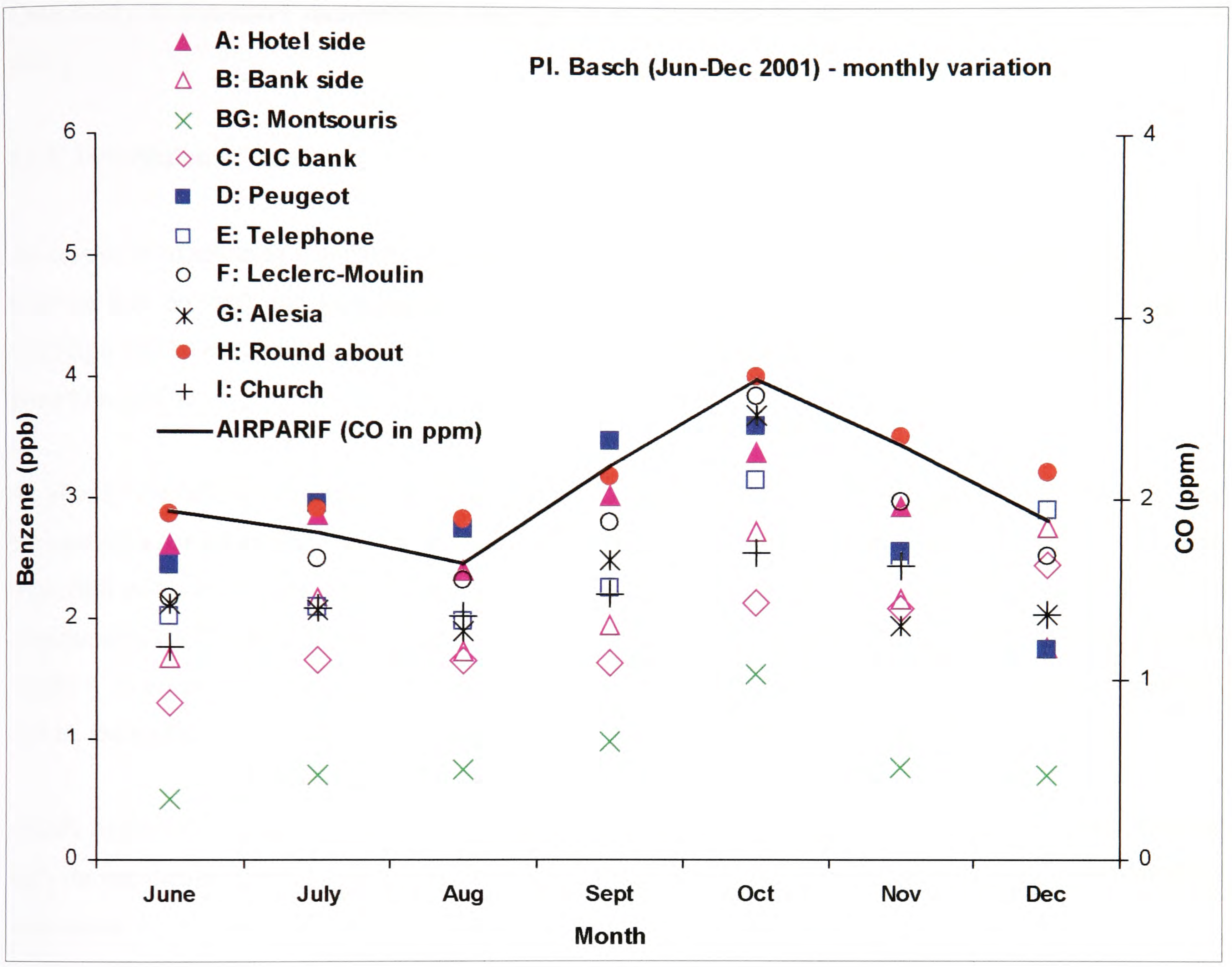


Fig. 3.32: Monthly benzene (ppb) variation in 10 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park; CO (ppm) variation in the AIRPARIF monitoring station in Pl. Basch (Jun-Dec 2001).

3.6. Avenue Leclerc

Case study 5: Intensive monitoring campaign in an asymmetric street canyon during summer (July 2001)

3.6.1. Description of the site

An intensive monitoring campaign was carried out in Av. Leclerc in July 2001, within the road segment between Rue Sarrette and Rue Daudet, where the separate parking lane is located. The results from this campaign will be examined together with the passive sampling measurements obtained in Pl. Basch during the same time period.

As already explained in Section 3.5.1, this part of Av. Leclerc is an asymmetric avenue canyon, with large pavements, a narrow traffic island separating the parking lane from the rest of the street, and three rows of big trees (full of leaves during summer time). Three modern ten-storey building blocks (height = 36 m) line up continuously on the east side of the canyon and a mixture of lower traditional and modern buildings (average height = 24 m) are situated on the west side (Fig. 3.33). The length of the straight road segment of interest is 116 m, the width 45 m (including pavements) and the street axis bearing from the north 18°.

Traffic lights were operating at all junction indicated in Fig. 3.25. The traffic volume in Av. Leclerc was very high during measurements (approximately 66,000 veh/day) and congested during the morning and afternoon rush hours.

3.6.2. Sampling protocol

In addition to the 11 sampling locations of the long-term campaign, diffusive RPE samplers were exposed at two different heights (2nd and 5th floor) near the walls of the asymmetric canyon in Av. Leclerc during the week of 9-16 July. During the following seven days (16-23 July), RPE as well as Radiello samplers were exposed at a total number of 19 different locations in Av. Leclerc and Pl. Basch, covering two heights at street level (1.5 and 2.6 m) and two or three heights (2nd, 5th and 10th floor) near the walls of the asymmetric canyon (Fig. 3.33).

During the second week of the campaign (16-23 July), the mobile monitoring unit was parked on the east side of the canyon (inside the separate parking lane) and a main sampling line was established at 2.9 m height above the ground and 9.2 m distance from the canyon wall. Real time CO, NO_x and O_3 measurements were recorded during day and night-time throughout the campaign (24/24 h). Continuous CO and NO_x

measurements were also obtained from the AIRPARIF monitoring station in Pl. Basch. Finally, active VOC sampling was conducted during one day (20 July) by drawing ambient air during several one hour intervals at a constant flow through Supelco tubes.

The two anemometers (i.e. ultrasonic and mechanical) and the weather mini-station were located at the kerb, near the trailer, in a relatively open space between the trees. The height of the anemometer mast was 4.8 m above the ground and the distance from the canyon wall 10.3 m. Finally, manual vehicle counts were taken during 10 hours of the campaign and compared with the data obtained from the automatic traffic monitoring network.

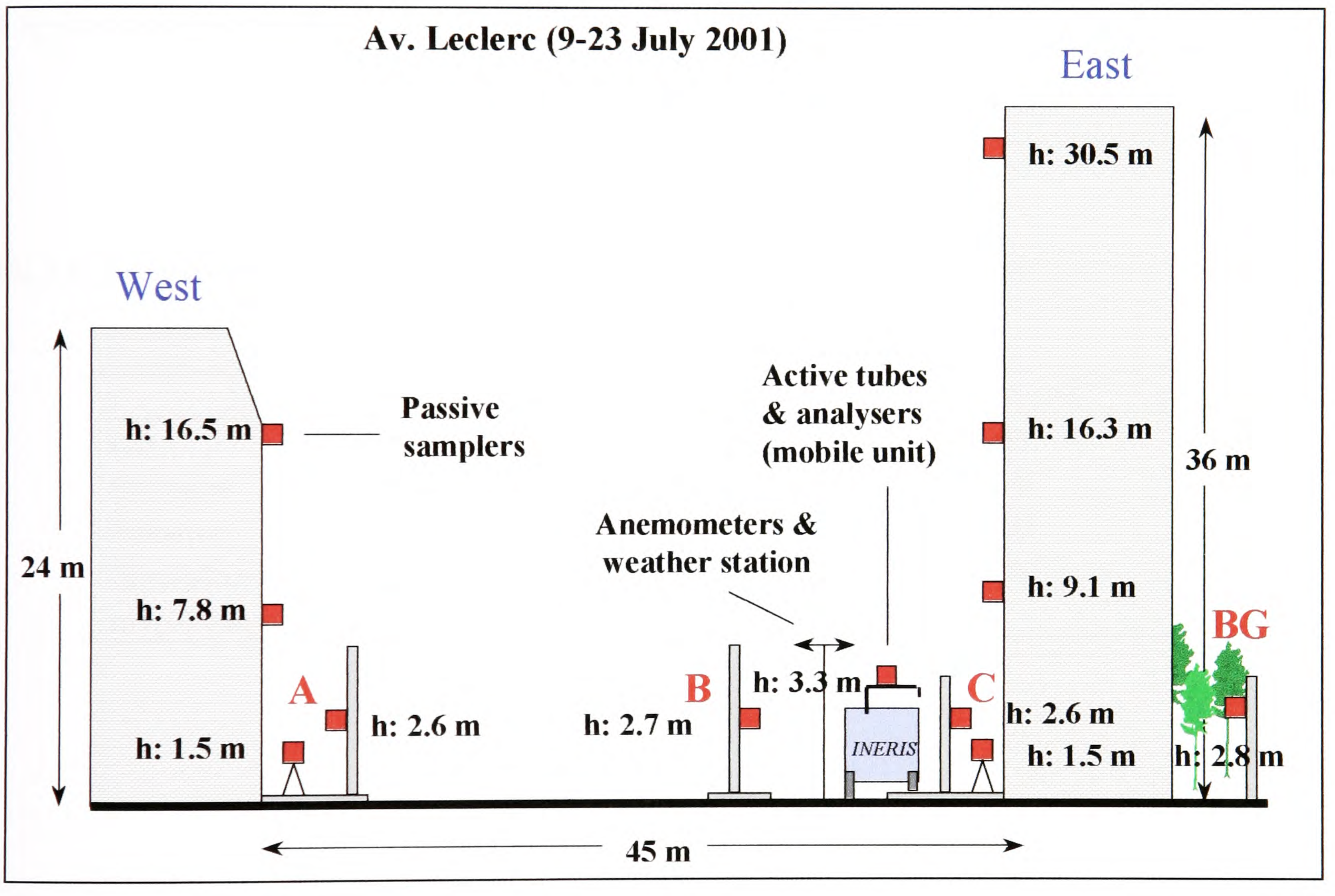


Fig. 3.33: Canyon dimensions and location of monitoring equipment in Av. Leclerc.

3.6.3. Analysis and interpretation of results

Relationship between pollutants

The active VOC sampling carried out through the same manifold as the continuous CO measurements on the 20^{th} of July revealed a strong correlation between benzene, toluene, m+p-xylenes, o-xylene, ethylbenzene and CO in Av. Leclerc (Table 3.6).

Furthermore, the measurements carried out with diffusive tubes at different sampling locations within Av. Leclerc during two consecutive weeks (9-16 and 16-23 July) revealed a very strong correlation between benzene and toluene concentrations. The experimental toluene to benzene ratio (by volume) during this period was approximately equal to 4.0 (Fig. 3.34).

Table 3.6: Correlation coefficients of air pollutants measured with active sampling in Av. Leclerc.

	Benzene	Toluene	Ethylbenzene	m+p-Xylene	o-Xylene
CO	0.88	0.97	0.81	0.80	0.84
Benzene		0.93	0.71	0.75	0.81
Toluene			0.75	0.80	0.82
Ethylbenzene				0.94	0.93
m+p-Xylene					0.91

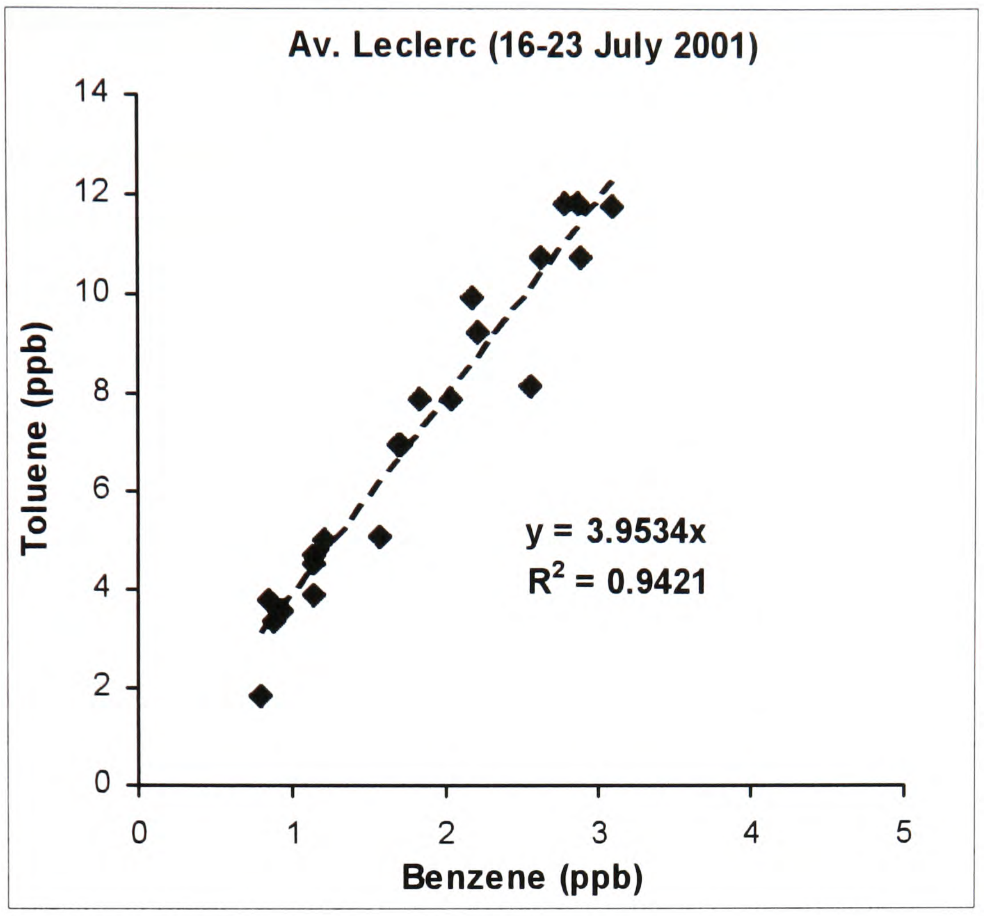


Fig. 3.34: Average toluene (ppb) vs. benzene (ppb) concentrations measured with passive sampling in Av. Leclerc (16-23 July 2001).

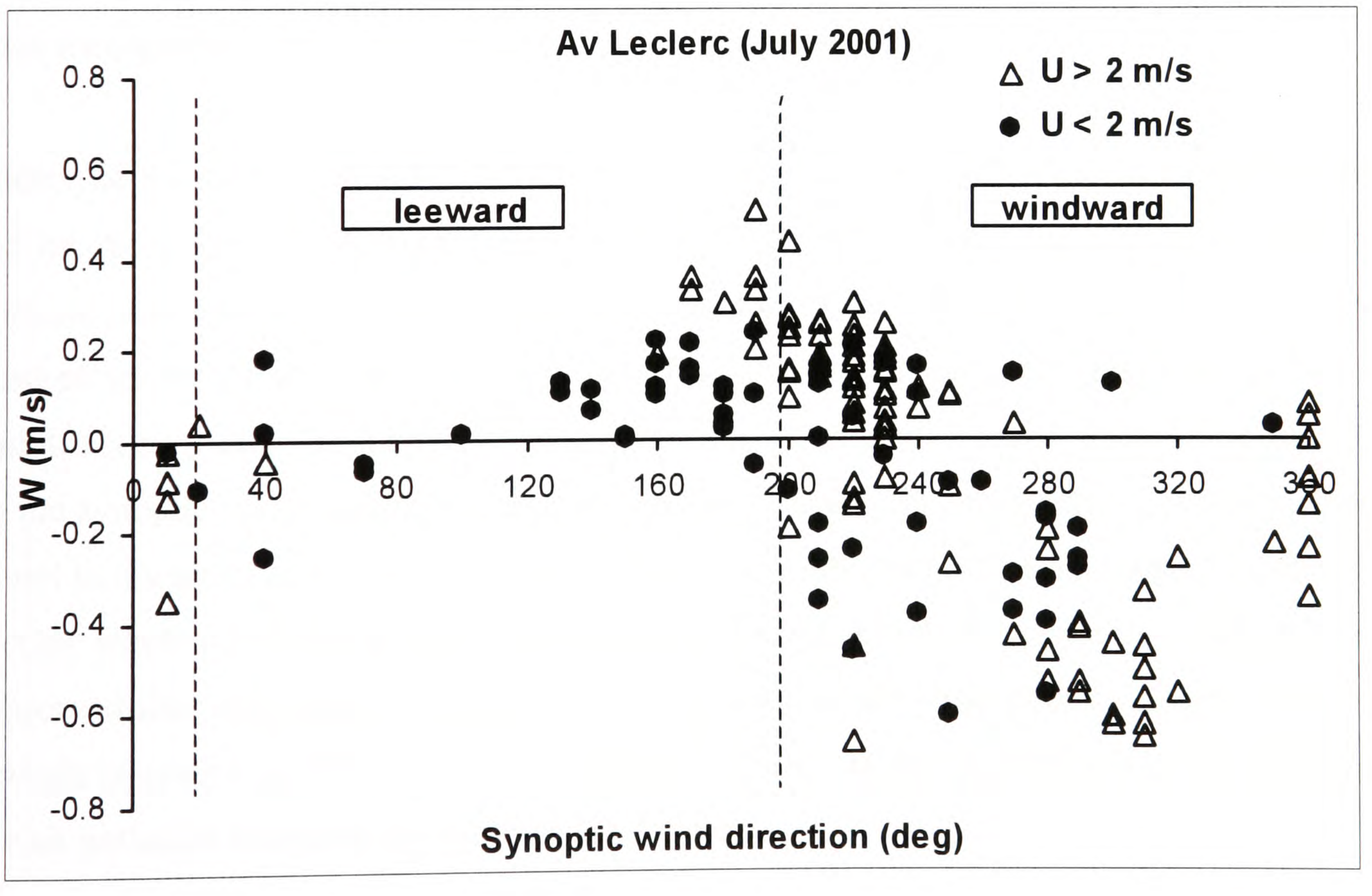


Fig. 3.35: Vertical street level wind speed (W) vs. synoptic wind direction in Av. Leclerc (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

Wind flow and dispersion conditions

During part of the intensive campaign in Av. Leclerc (16-23 July), there was evidence of a wind vortex being formed within the asymmetric canyon. When the synoptic wind was blowing from 260° to 320° (thus perpendicular to the street axis), a downward airflow was detected on the windward side of the street, especially for synoptic winds above 2 m/s (Fig. 3.35).

Furthermore, there was certain evidence of an elastic-type reflection of the wind off the windward wall of the canyon for synoptic winds greater than 2 m/s. Using the synoptic wind data from Montsouris station, a set of street level wind directions was calculated according to the elastic reflection assumption. A reasonable agreement ($r = 0.65$) between measured and calculated wind directions was finally observed for winds above 2 m/s (Fig. 3.36). However, this assumption was probably too idealistic in the case of Av. Leclerc, which is an asymmetric canyon with irregular walls and big trees on both sides.

The influence of the synoptic wind speed and direction on the dispersion of pollutants at street level is illustrated in Fig. 3.37. It can be seen that the lower traffic normalised CO concentrations in Av. Leclerc occurred for relatively high synoptic winds ($U > 2$ m/s) blowing from directions between 230° and 320°, thus for windward flow. Furthermore, it can be observed that low wind conditions ($U < 2$ m/s) as well as near-parallel and leeward flow favoured pollution built-up in the street.

The influence of the synoptic wind direction on pollutant dispersion within the asymmetric canyon was also illustrated on the pollution roses plotted for CO and NO_x (Fig. 3.38). Again, hourly mean CO and NO_x concentrations observed in Av. Leclerc and normalised with respect to the wind speed and traffic volume, were assigned to the corresponding synoptic wind directions and mean concentrations were calculated for each wind direction sector. Both CO and NO_x roses (Fig. 3.38) demonstrated a clear dependence of pollution levels on the synoptic wind direction. They showed that, keeping the other factors constant, winds parallel or near-parallel to the street axis (i.e. from S and N directions) favoured pollution built-up on the kerbside, while perpendicular winds (i.e. from W and NW directions) provided better dispersion conditions. Furthermore, it can be observed that normalised CO and NO_x concentrations were significantly higher for southerly winds than for winds coming from the north. This might be explained by the contribution of Bd. Périphérique, which is a major air pollution source in the south of Av. Leclerc.

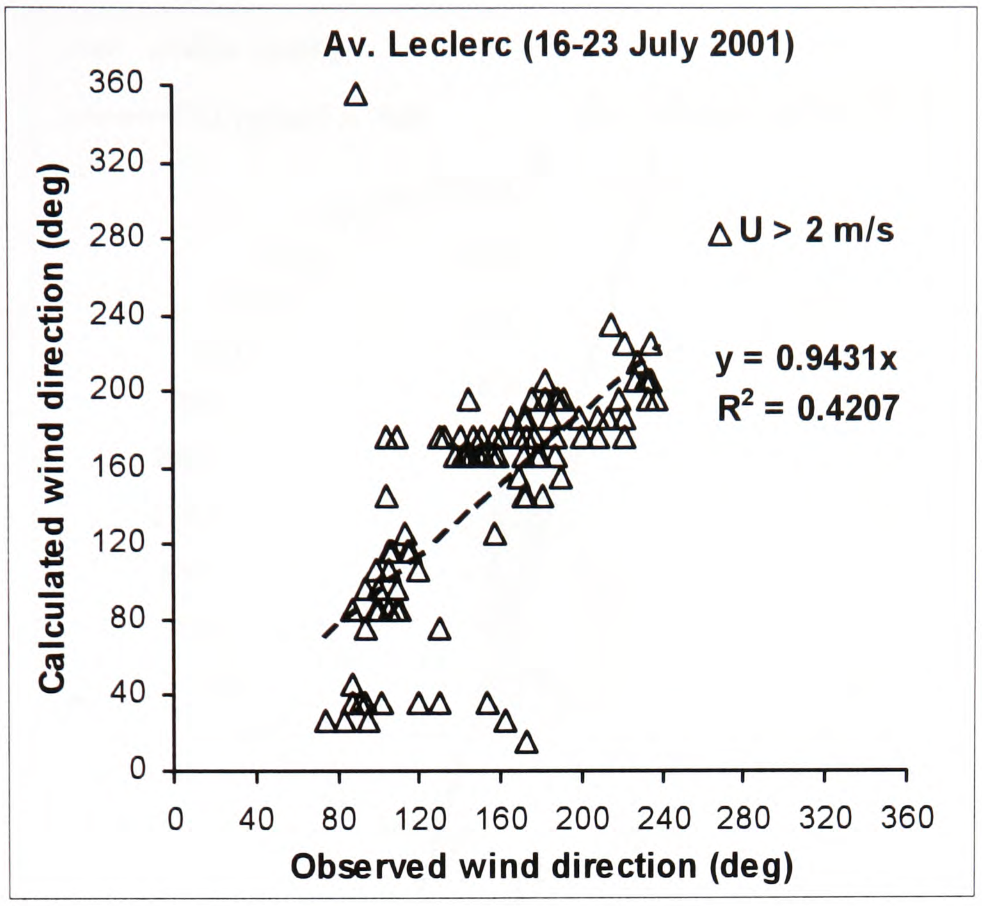


Fig. 3.36: Calculated wind directions: $[Dir]_{local} = 2$ [Street bearing] - $[Dir]_{synoptic}$ vs. observed street level wind directions in Av. Leclerc.

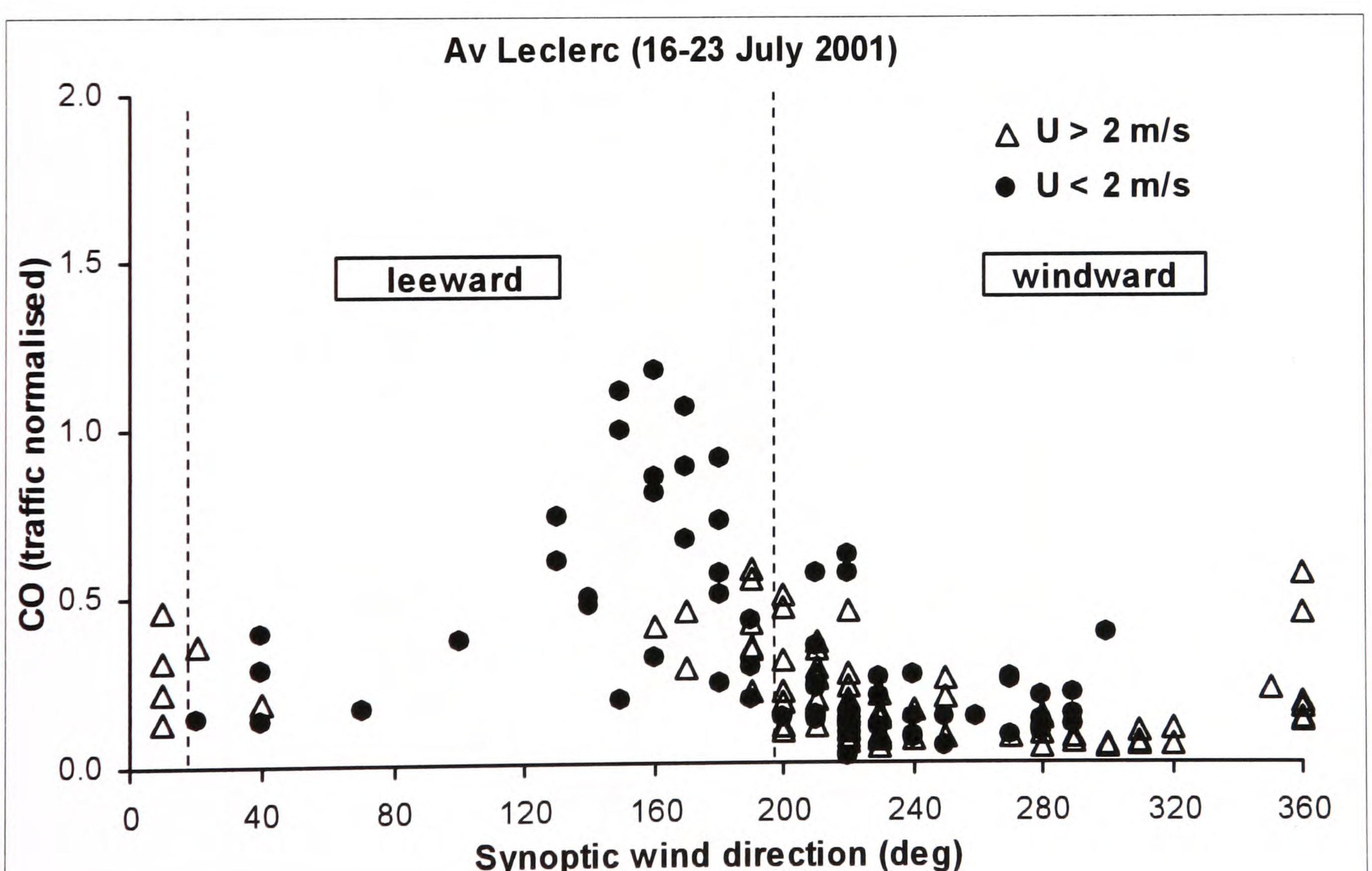


Fig. 3.37: Traffic normalised CO concentrations vs. synoptic wind direction in Av. Leclerc (sorted for synoptic wind speed above and below 2 m/s). The dotted lines indicate the orientation of the street.

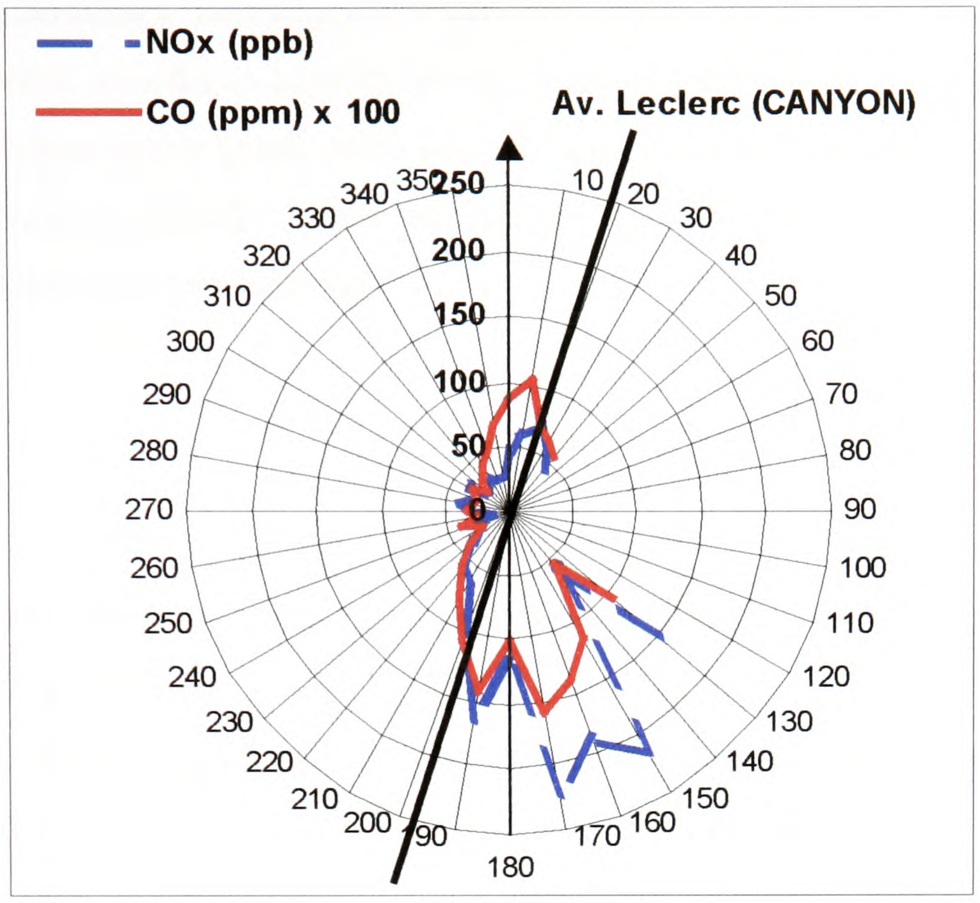


Fig. 3.38: Traffic and wind speed normalised CO and NO_x concentration roses in Av. Leclerc. (The heavy straight line indicates the direction of the street.)

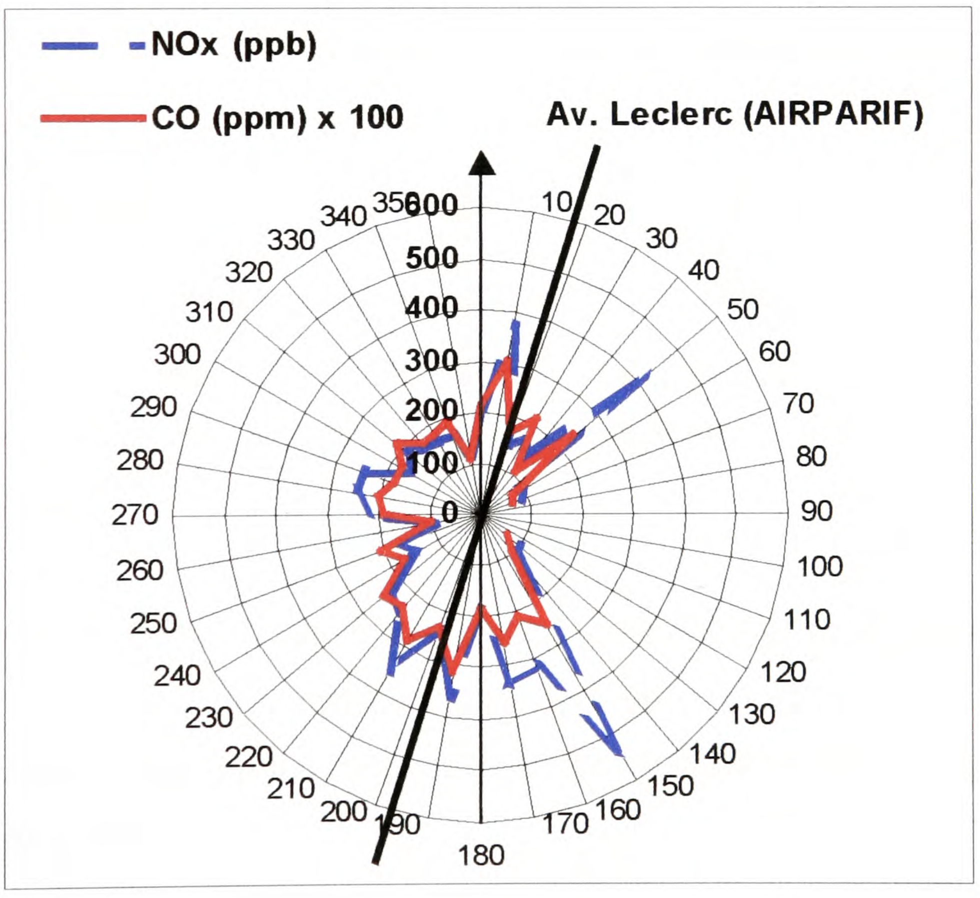


Fig. 3.39: Traffic and wind speed normalised CO and NO_x concentration roses in Pl. Basch – AIRPARIF monitoring station. (The heavy straight line indicates the direction of Av. Leclerc.)

Following the same methodology, normalised CO and NO_x concentrations from the AIRPARIF station were used to plot pollution roses, in order to identify the influence of wind direction on pollutant dispersion in Pl. Basch (Fig. 3.39). According to this graph, there was no clear dependence of the observed pollution levels on the synoptic wind direction, although higher normalised CO and NO_x concentrations were more often associated with parallel and near-parallel winds.

Spatial variability

Using BTX as indicators, strong concentration gradients were identified in the horizontal sense within Av. Leclerc during the two consecutive weeks of the intensive campaign (Fig. 3.40 and 3.41). Within the asymmetric canyon, the highest average benzene concentrations were detected near the kerb, on the side of the street that was leeward for most of the sampling period.

During the week of 9-16 July, moderate winds (average wind speed = 2.7 m/s) from W and NW directions were prevailing in the region, inducing perpendicular flow within Av. Leclerc. That resulted in significantly higher street-level benzene concentrations at location A on the west side (i.e. leeward) of the canyon compared to those measured at B, C and J on the east side (i.e. windward) (Fig. 3.40).

During the following week (16-23 July), slightly stronger winds (average wind speed = 3 m/s) from S and SW directions were prevailing in the site, giving rise to a combination of perpendicular and parallel wind conditions in Av. Leclerc. This meteorological situation resulted in lower crossroad gradients (East-West) and higher benzene concentrations at the locations H and I (Fig. 3.41), which were mainly affected by the accumulation of vehicle emissions along the heavily trafficked segment of Av. Leclerc linking Bd. Périphérique with Pl. Basch.

Kerbside concentrations were 2.3 to 7.1 times higher than those detected in Montsouris Park during the first week (9-16 July) and 1.4 to 3.6 times higher during the second week (16-23 July). It can thus be concluded that the impact of Bd. Périphérique on the benzene levels in Montsouris Park was greater during the second week due to the southerly winds.

The vertical benzene gradients near the walls of the canyon were relatively weak during both sampling periods. The reduction of ambient concentrations from the 2nd to the 5th floor was less than 10% and 20% on the leeward and windward side, respectively, during the first week (Fig. 3.42).

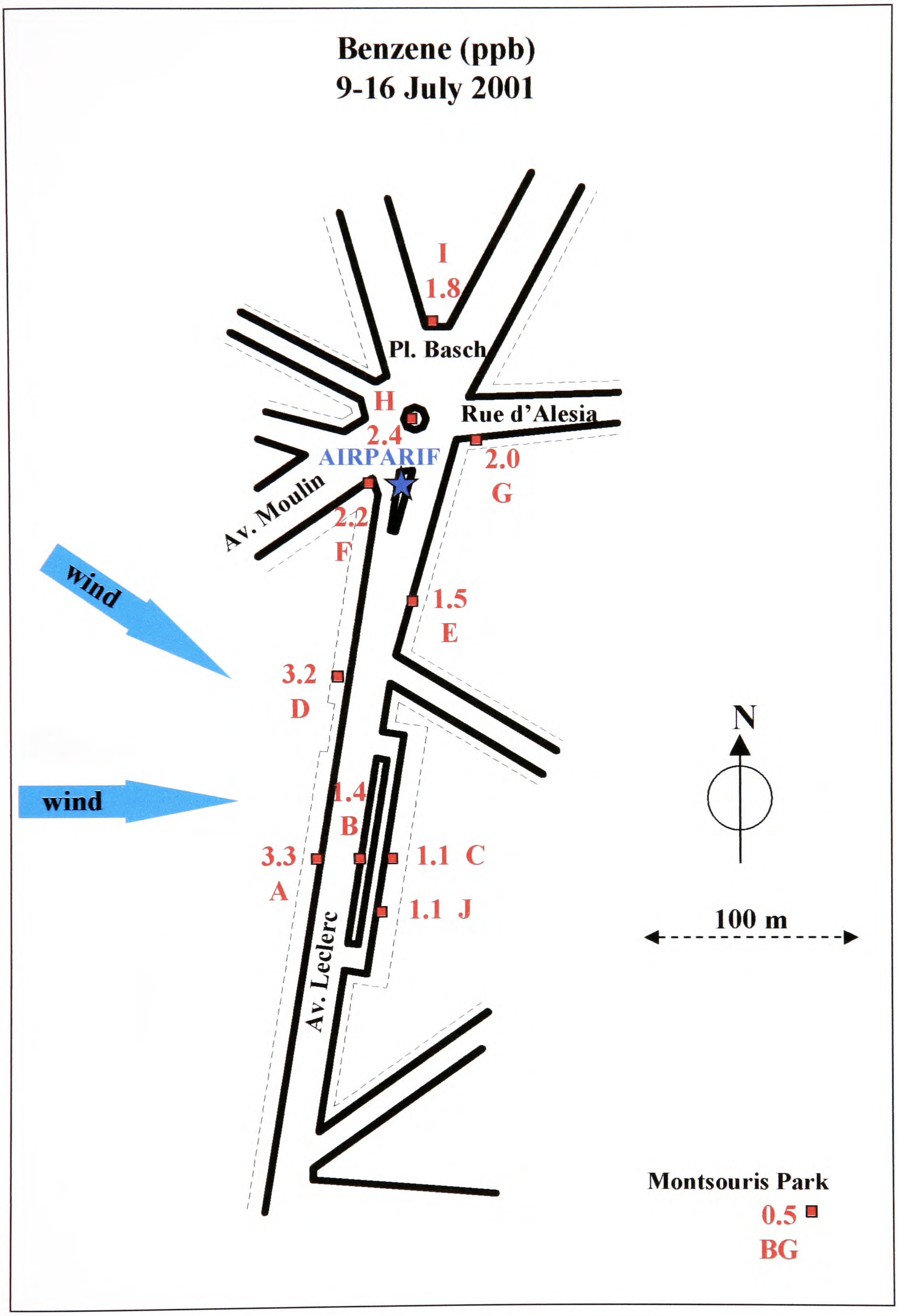


Fig. 3.40: Average benzene concentrations (ppb) during one week (9-16 July 2001) at 11 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park (plan view).

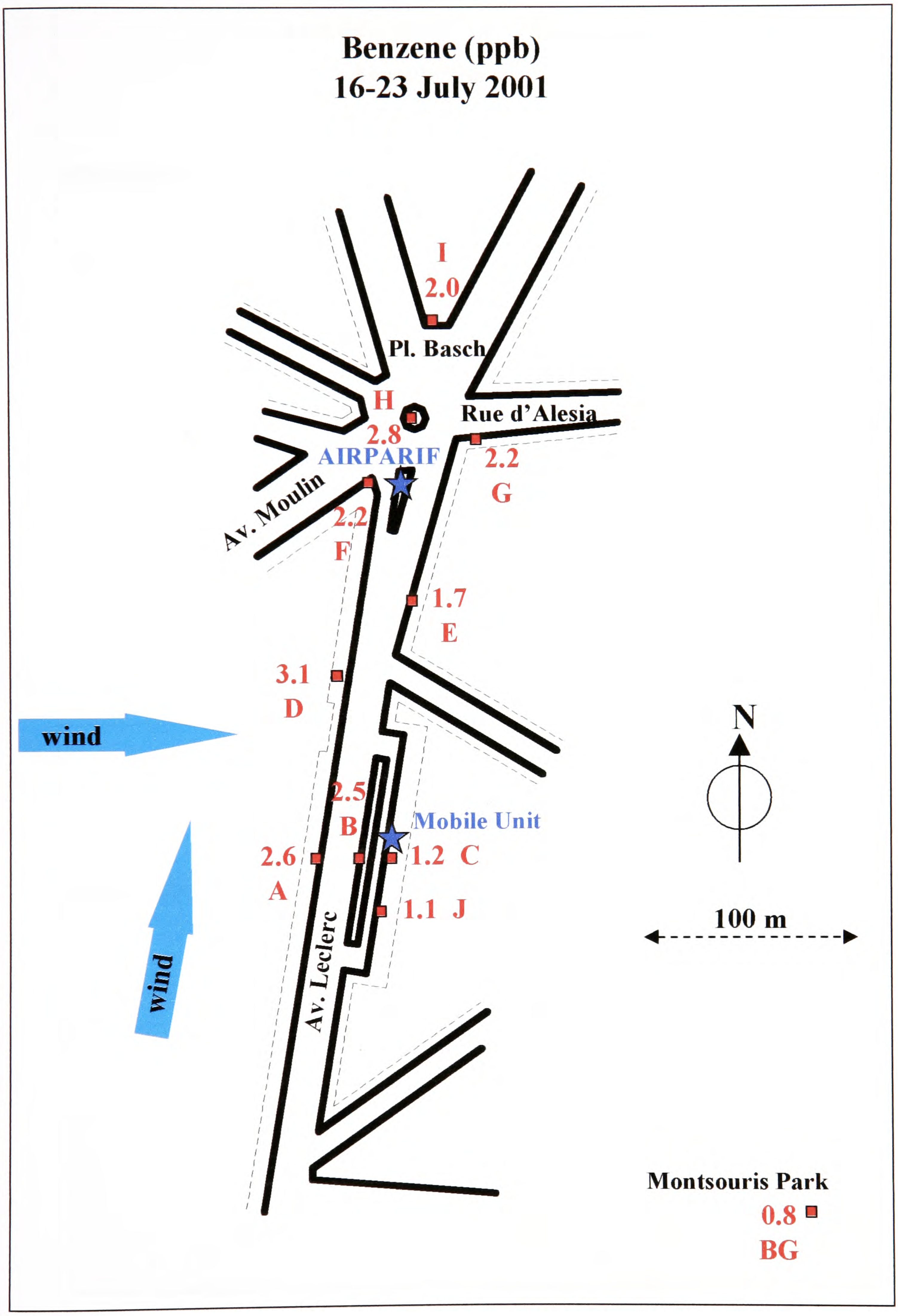


Fig. 3.41: Average benzene concentrations (ppb) during one week (16-23 July 2001) at 11 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park (plan view).

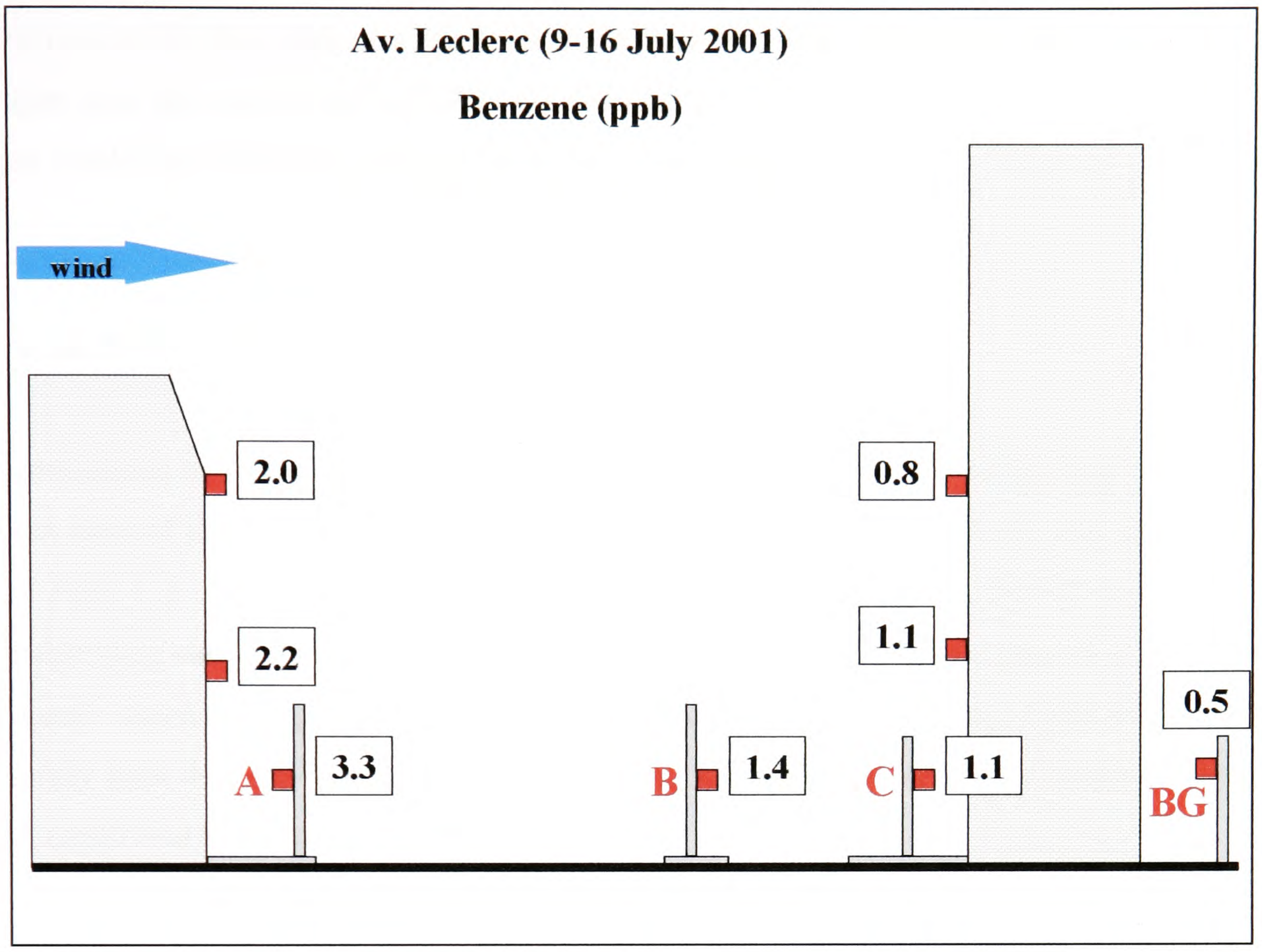


Fig. 3.42: Average benzene concentrations (ppb) during one week (9-16 July 2001) in Av. Leclerc.

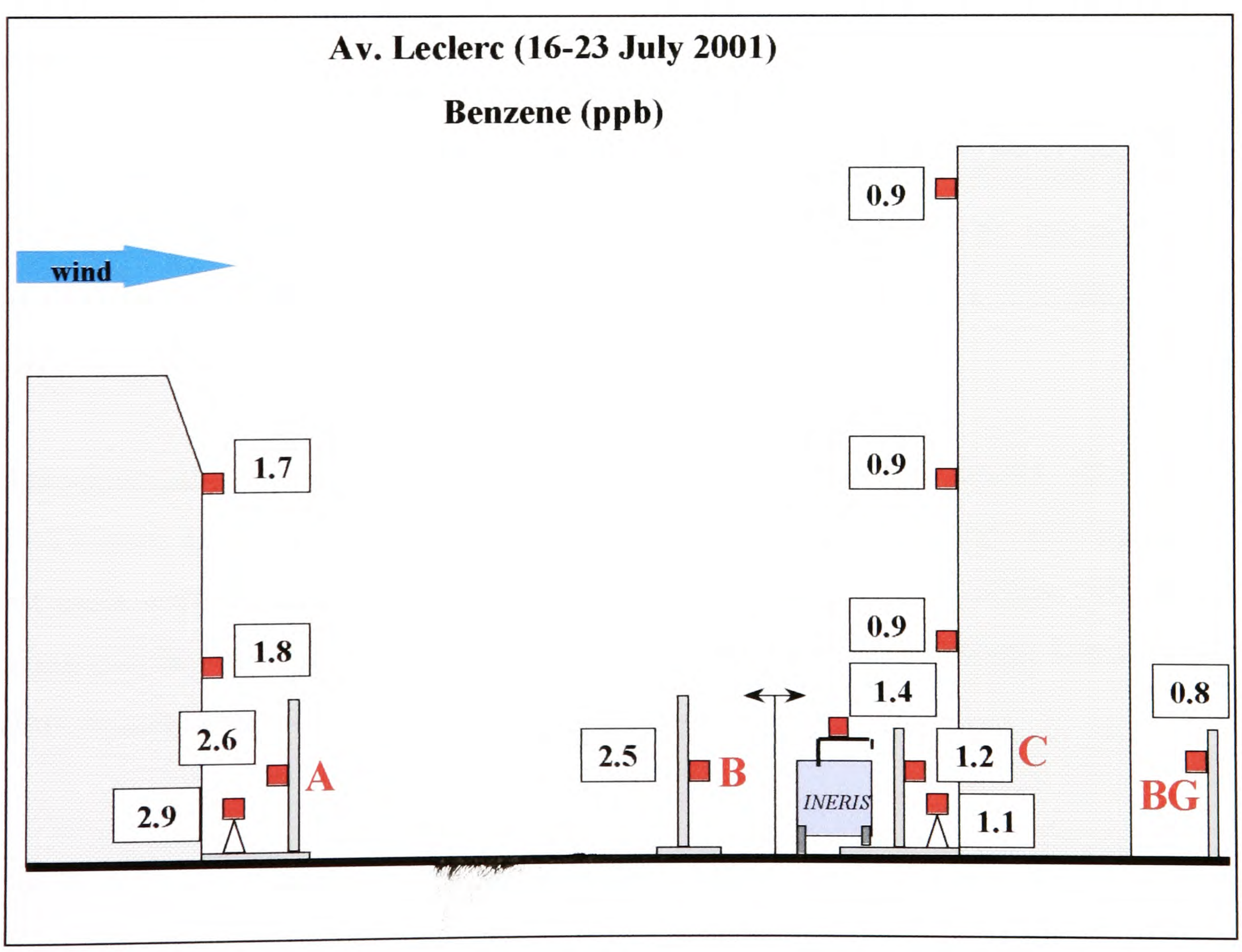


Fig. 3.43: Average benzene concentrations (ppb) during one week (16-23 July 2001) in Av. Leclerc.

During the second week, there was no significant difference between the benzene concentrations measured at different heights near the canyon walls (Fig. 3.43). Finally, the same trends were confirmed by the other VOC concentrations (including aldehydes) measured during this campaign.

Temporal variability

The CO concentrations observed in Av. Leclerc (16-23 July) using the mobile monitoring unit were very low. They remained most of the time below 1 ppm and only for few hours during the second day of the intensive campaign (17 July) CO values exceeded 2 ppm (Fig. 3.44). On the other hand, the CO levels detected at the permanent monitoring site in Pl. Basch (AIRPARIF) were several times higher than in Av. Leclerc (trailer) during the same monitoring period. Furthermore, it was observed that the AIRPARIF measurements correlated closely with the daily traffic pattern in this area. The NO_x concentrations measured in Av. Leclerc and Pl. Basch confirmed these trends (Fig. 3.45).

The very low CO and NO_x concentrations observed between the 18th and 21st of July in Av. Leclerc can be attributed to the relatively strong synoptic winds (2-6 m/s) recorded in Montsouris Park (Fig. 3.46) and the mainly windward position of the monitoring unit within the canyon (Fig. 3.47). The relatively high CO and NO_x concentrations observed on the 17th of July in Av. Leclerc correspond to a period of strong synoptic winds (3-6 m/s in Montsouris Park) coming from the south (wind directions: 160°-220°). Under these meteorological conditions, the impact of Bd. Périphérique on the concentrations measured in Av. Leclerc was expected to be high. By contrast, the CO and NO_x concentrations measured in Pl. Basch did not reflect the variability of the synoptic meteorological conditions, probably due to the very short distance between the receptor (AIRPARIF station) and the sources (i.e. vehicles).

Moderate photochemical activity was observed in Av. Leclerc during the second week of the intensive campaign (16-23 July). Generally, NO concentrations peaked during the morning rush hours. The NO_2 peak levels in most cases were delayed a few hours, because of the time needed to oxidise NO. Ozone gradually increased during the day, reaching higher levels during the afternoon and evening hours. However, ozone remained below 40 ppb in all cases (Fig. 3.48).

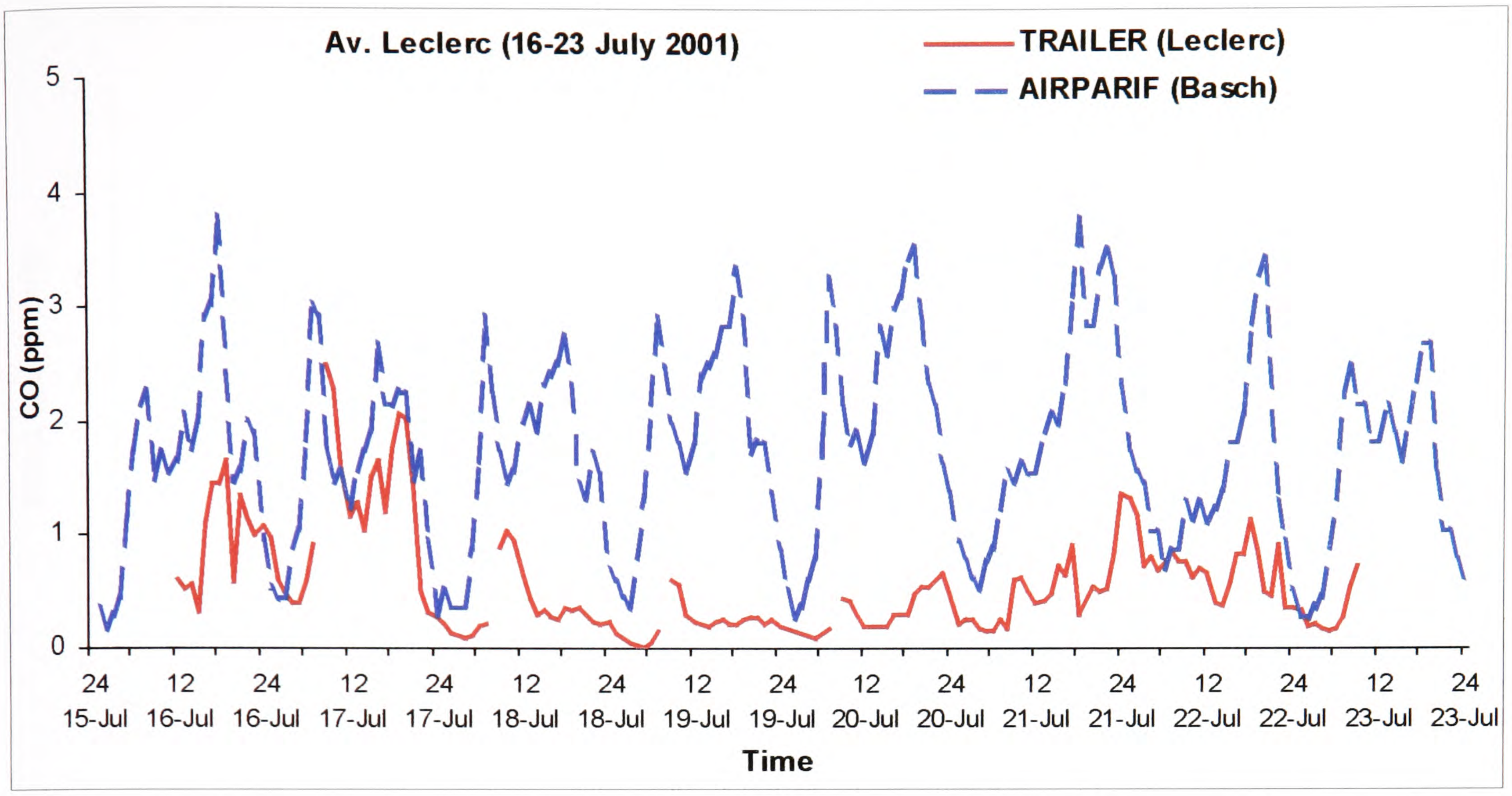


Fig. 3.44: Hourly mean CO (ppm) concentrations measured in Av. Leclerc (trailer) and in Pl. Basch (AIRPARIF).

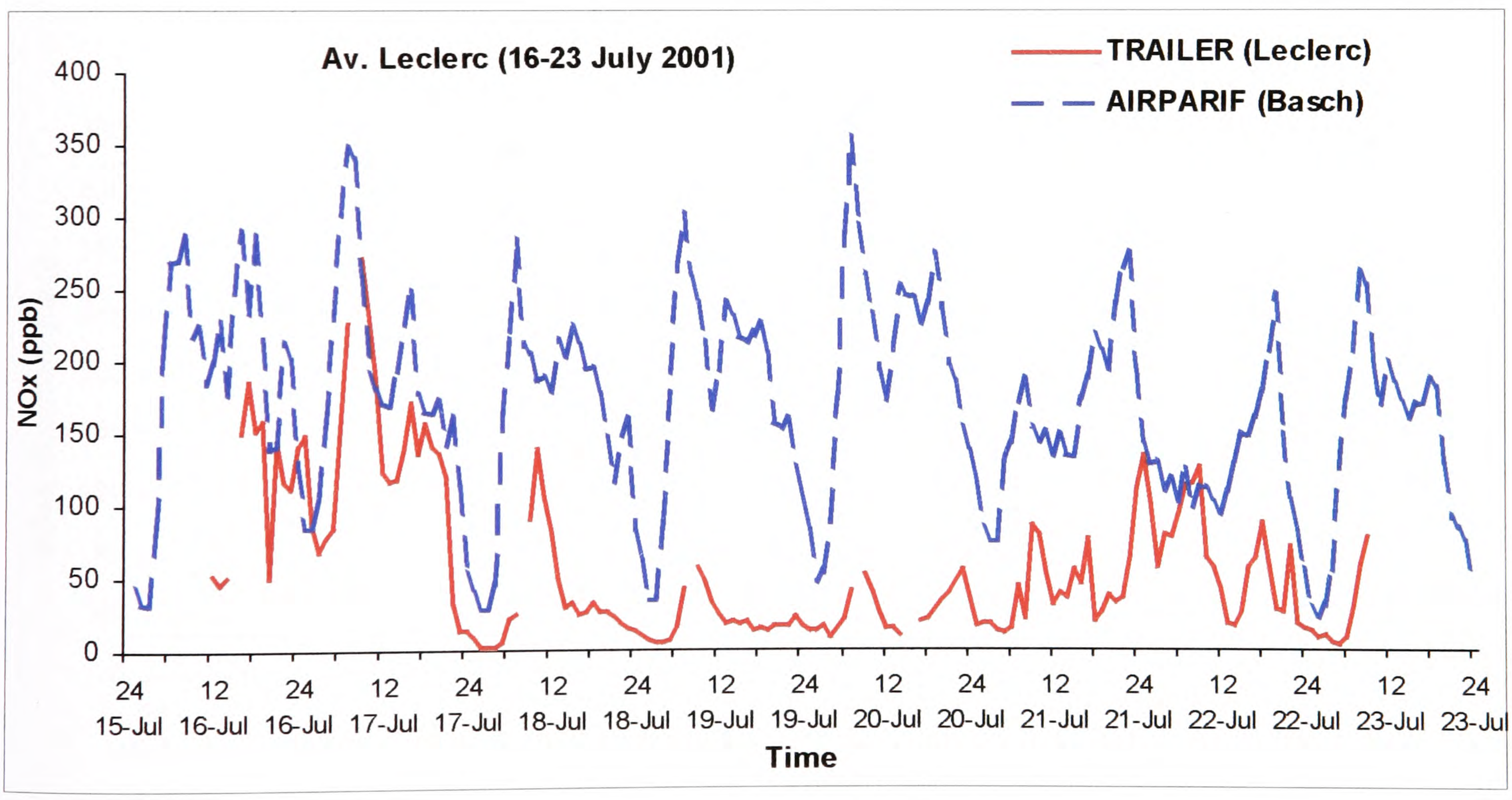


Fig. 3.45: Hourly mean NO_x (ppb) concentrations measured in Av. Leclerc (trailer) and in Pl. Basch (AIRPARIF).

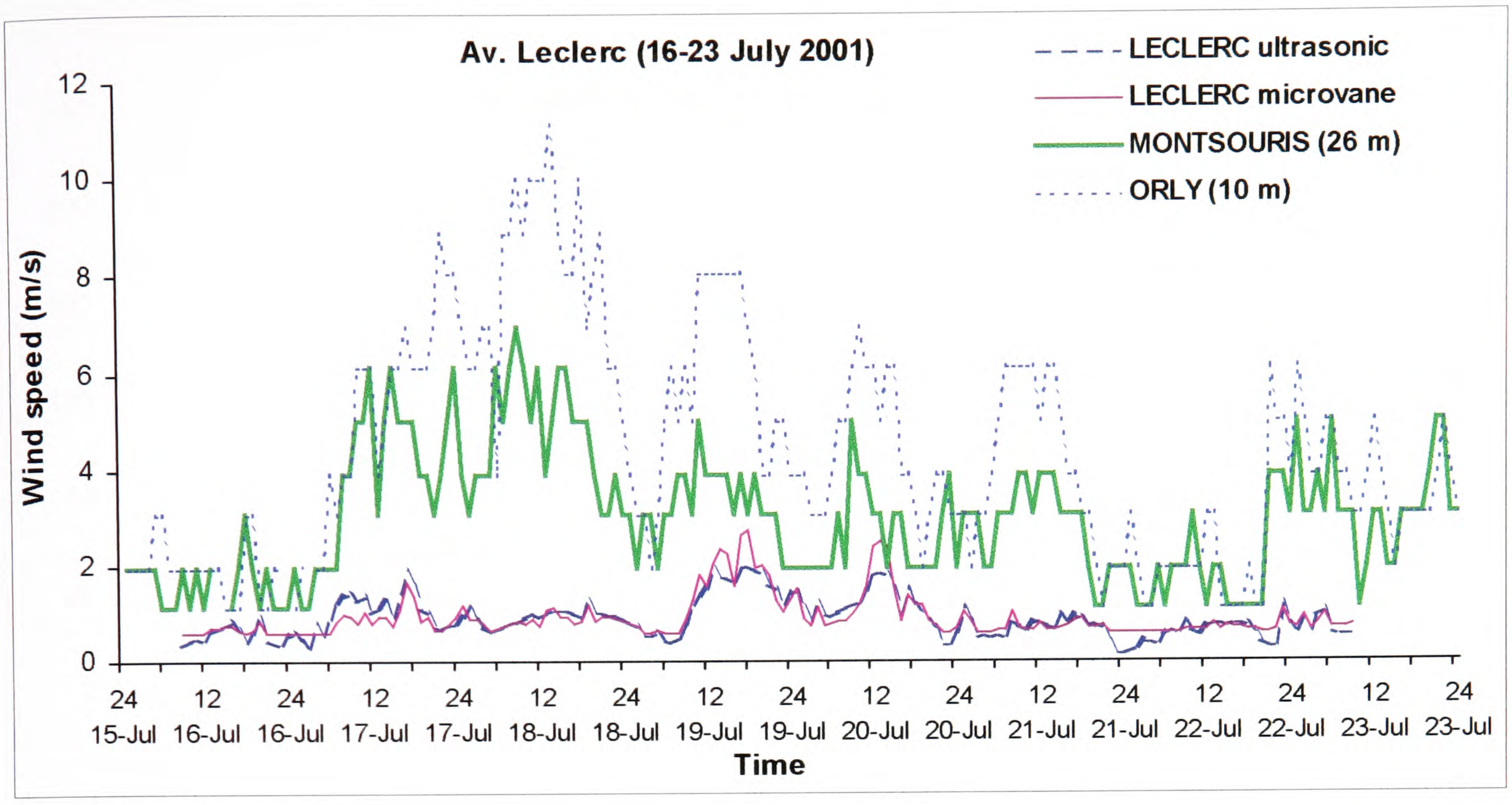


Fig. 3.46: Synoptic wind speed measured in Montsouris Park and Orly Airport, and local wind speed measured in Av. Leclerc with a 3D ultrasonic and a mechanical microvane anemometer.

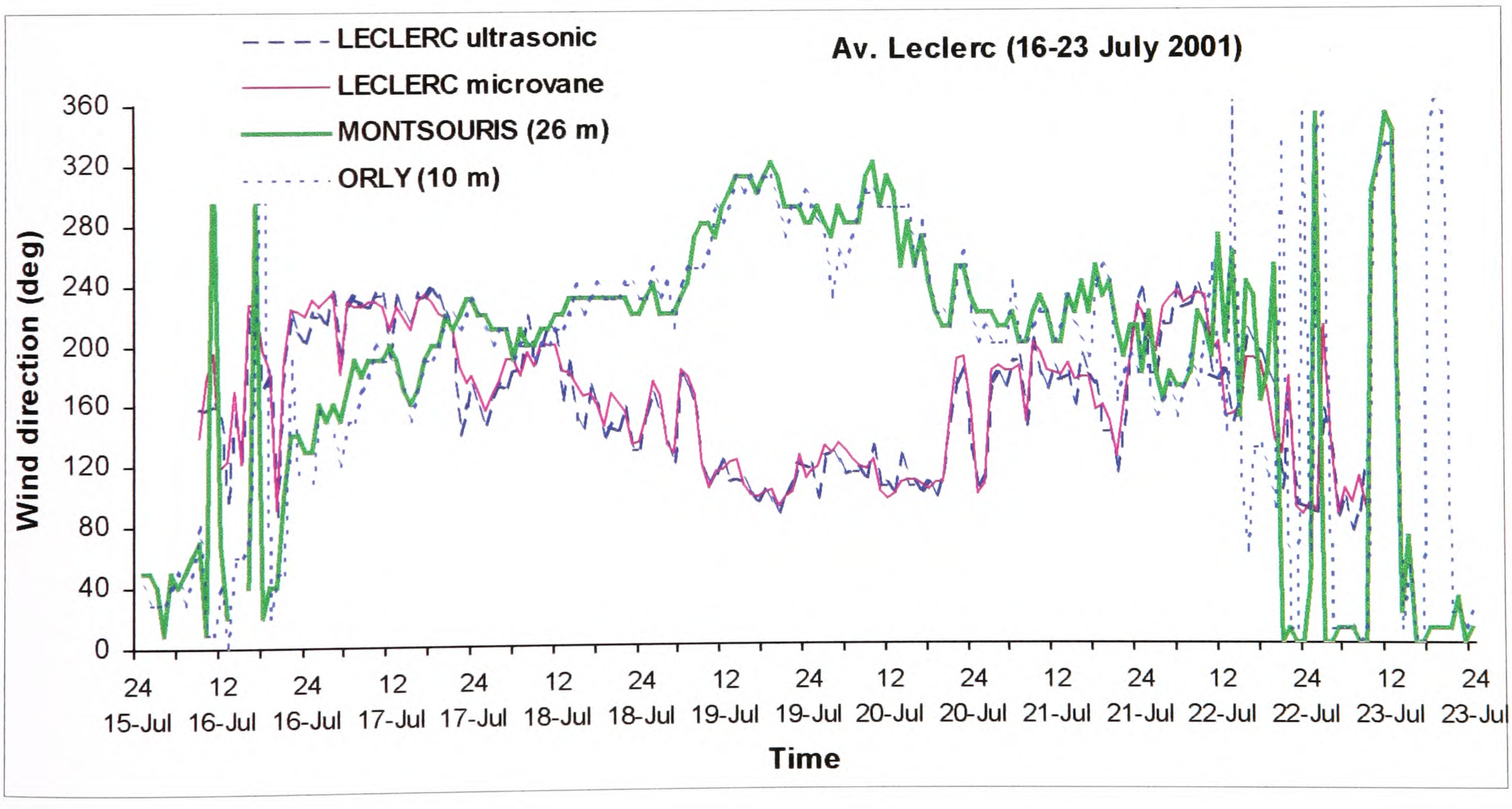


Fig. 3.47: Synoptic wind direction measured in Montsouris Park and Orly Airport, and local wind direction measured in Av. Leclerc with a 3D ultrasonic and a mechanical microvane anemometer.

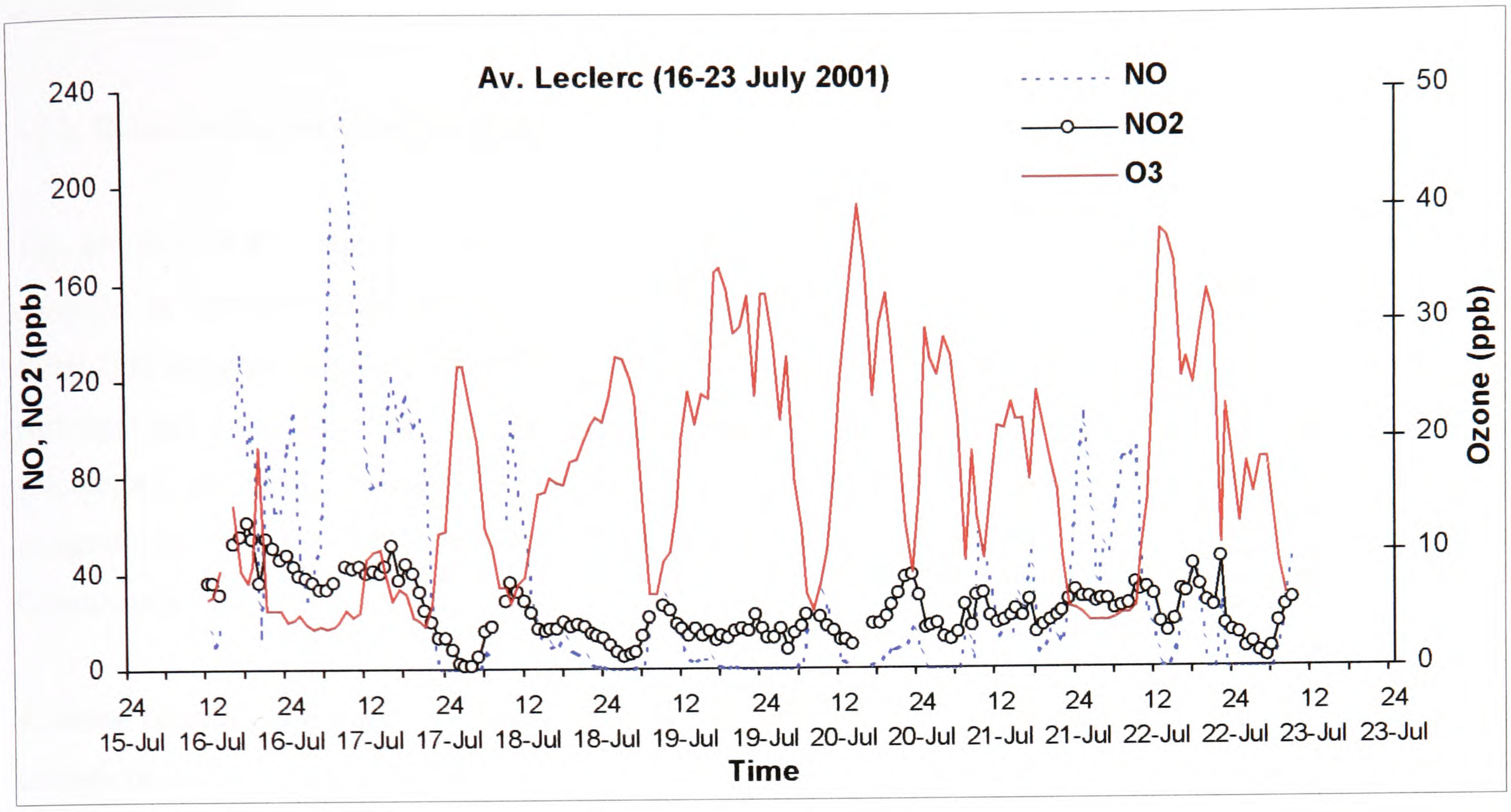


Fig. 3.48: Hourly mean NO, NO_2 and O_3 (ppb) concentrations observed in Av. Leclerc (mobile unit).

3.7. Discussion

3.7.1. Relationship between pollutants

The diffusive VOC samplers revealed a very strong correlation between benzene and toluene concentrations observed at different locations within Bd. Voltaire, Rue de Rennes, Pl. Basch and Av. Leclerc. This correlation suggests that these compounds, which come from the same source (i.e. petrol vehicles), do not take part significantly in chemical reactions within the canyons. The experimental toluene to benzene ratio (by volume) was 2.9 in Bd. Voltaire, 3.4 in Rue de Rennes, and 4.0 in Pl. Basch and Av. Leclerc. These values are in agreement with the ratio (approximately 3.2) observed by Palmgren et al. (1999) in a street canyon in Copenhagen.

A strong correlation between several VOC (especially BTX) and CO was established during the monitoring campaigns in Paris using simultaneously active sampling (i.e. pumped tubes) and continuous monitoring (i.e. a standard gas analyser). The observed correlation was in agreement with findings from previous studies (Hansen and Palmgren, 1996; Giugliano et al., 2000) that have shown that CO, although a pure combustion product, correlates highly with several aromatic VOC, which are not only emitted through combustion but also through direct fuel evaporation.

The negligible chemical reactivity corresponding to the short diffusion times of CO and benzene in canyon streets as well as their strong correlation suggest that they can both be used as traffic pollution indicators. Due to their common origin and fate in urban environments, it is expected that a simple linear relationship can be established between them:

$$\text{Benzene (ppb)} \approx \alpha \text{ CO (ppm)} + \beta \qquad (3.4)$$

where α and β are regression coefficients. These coefficients obtained for Bd. Voltaire (Fig. 3.49), Rue de Rennes (Fig. 3.50), and Av. Leclerc (Fig. 3.51) agree with results reported by Jones et al. (2000) after a field experiment in Paris, and Pfeffer et al. (1995) after measurements in two German cities. In addition, they are within the range of values observed by Palmgren et al. (1999). All coefficients are summarised in Table 3.7.

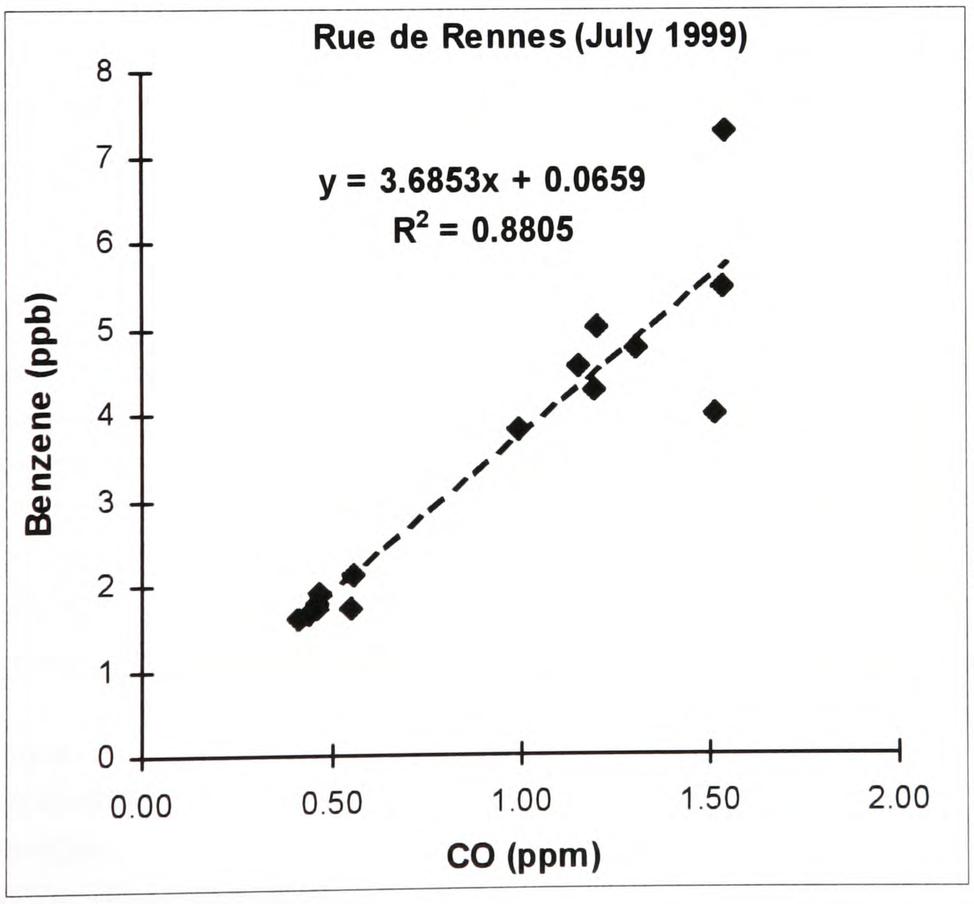


Fig. 3.49: Hourly benzene (ppb) vs. CO (ppm) concentrations measured with active sampling in Bd. Voltaire (Dec.1998).

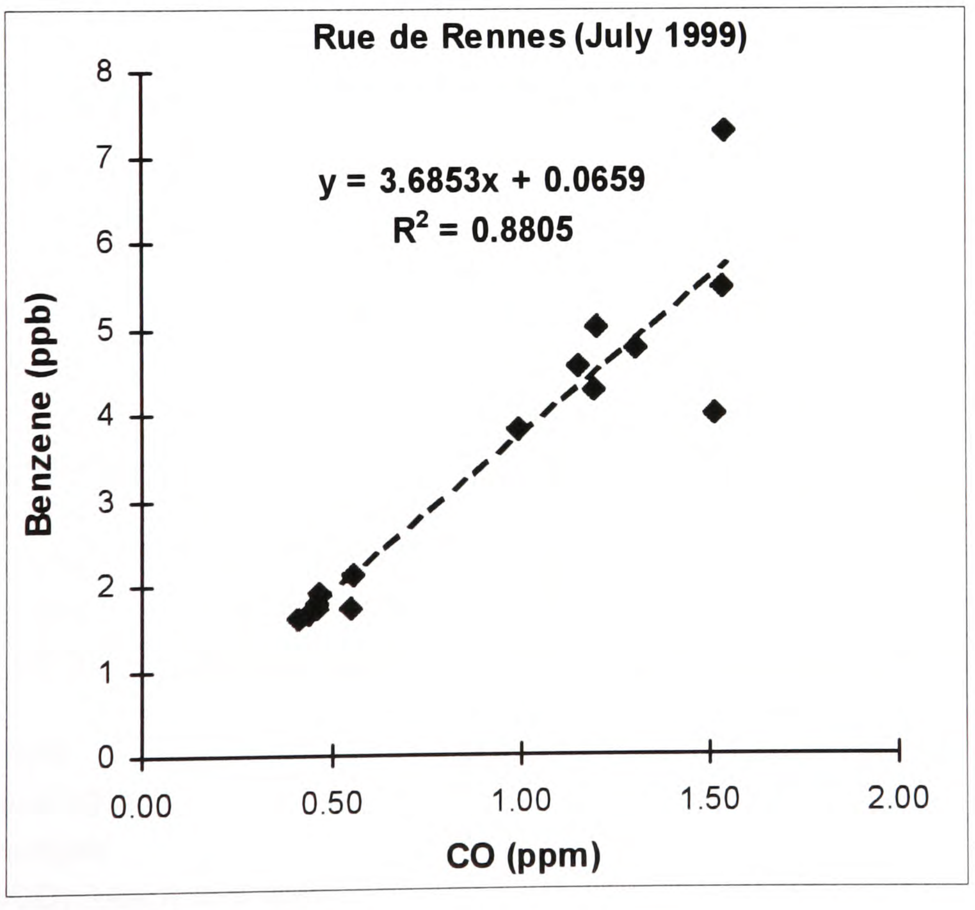

Rue de Rennes (July 1999)
y = 3.6853x + 0.0659
R^2 = 0.8805
Benzene (ppb)
CO (ppm)
0
1
2
3
4
5
6
7
8
0.00
0.50
1.00
1.50
2.00

Fig. 3.50: Hourly benzene (ppb) vs. CO (ppm) concentrations measured with active sampling in Rue de Rennes (July 1999).

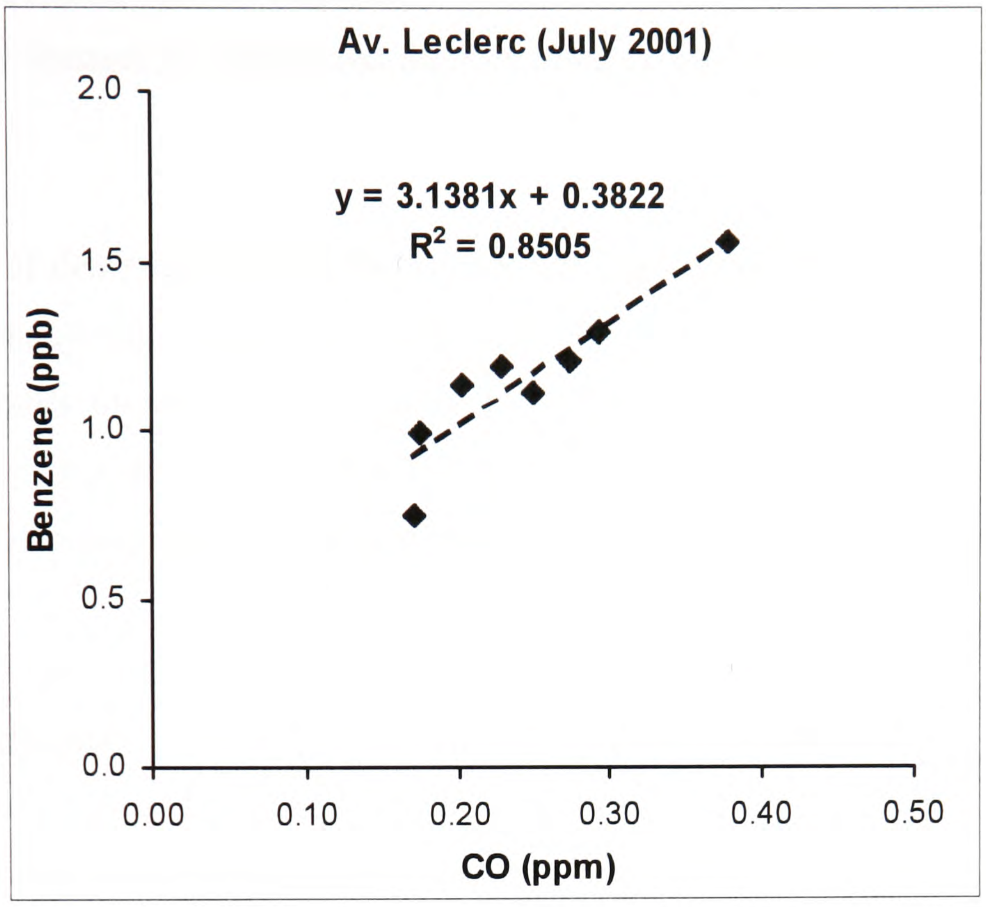


Fig. 3.51: Hourly benzene (ppb) vs. CO (ppm) concentrations measured with active sampling in Av. Leclerc (July 2001).

Table 3.7: Linear regression coefficients of the relationship between benzene and CO, and toluene/benzene ratios derived from measurements in Paris and other European cities.

Authors	City	Location	Year	Benzene / CO			Toluene / Benzene	
				alfa	beta	r	ratio	r
Pfeffer et al.	Dusseldorf	Corneliusstreet	1993	3.91	0.81	0.97	-	-
Pfeffer et al.	Essen	Hindenburgstreet	1993	3.27	1.66	0.94	-	-
Palmgren et al.	Copenhagen	Jagtvej street	1996	2.36	0.08	0.96	3.23	0.98
Jones et al.	Paris	Bd. Peripherique	1997	2.23	1.03	0.98	3.05	0.99
Vardoulakis et al.	Paris	Bd. Voltaire	1998	3.97	0.15	0.93	2.95	0.99
Vardoulakis et al.	Paris	Rue de Rennes	1999	3.69	0.07	0.94	3.43	0.99
Vardoulakis et al.	Paris	Avenue Leclerc	2001	3.14	0.38	0.92	3.95	0.97

Pfeffer et al.: Daily averages
Palmgren et al.: Hourly averages
Jones et al.: Daytime averages
Vardoulakis et al. (Benz/CO): Hourly averages
Vardoulakis et al. (Tolu/Benz): Weekly averages

In general, the CO to benzene ratio is expected to remain roughly the same in urban environments as far as there are no significant changes in vehicle and fuel technology, fleet composition, traffic patterns, or ambient temperature.

However, a reduction of this ratio (α) with the year of the monitoring campaign in Paris can be observed in Table 3.7. This is probably due to the relative reduction in the benzene content of fuels sold in France in recent years. The parallel increase of the toluene to benzene ratio observed during the same campaigns confirms this hypothesis. For this reason, relationship (3.4) should be regularly updated, if it is to be routinely used for estimating CO concentrations using benzene measurements in urban areas and vice versa.

3.7.2. Dispersion conditions

The formation of vertical wind vortices inside the three street canyons of this study (Bd. Voltaire, Rue de Rennes and Av. Leclerc) under perpendicular wind conditions was investigated by examining the direction (vertical and horizontal) and strength of the local street-level wind. In all three cases, there was evidence of a wind vortex being created within the canyon for synoptic winds above 2 m/s. Even in the case of Av. Leclerc, in which the assumption of elastic wind reflection was initially considered over-idealistic, there was further evidence of vortex formation in Fig. 3.47. It can be clearly seen on this graph that the synoptic wind (i.e. Montsouris) and the local wind (i.e. Leclerc ultrasonic) come from almost opposite directions during two days of near-perpendicular wind conditions (19-20 July).

The wind vortices gave rise to relatively high CO concentrations on the leeward side of each street. Nevertheless, the pollution roses plotted for Bd. Voltaire, Rue de Rennes and Av. Leclerc (Fig. 3.8, 3.17 and 3.38) showed that winds parallel or near-parallel to the street axis induced the highest roadside concentrations, while perpendicular winds generally reduced pollution levels (especially on the windward side of the streets). That confirms previous studies showing that in relatively long canyons without connecting streets, the accumulation of emissions along the line source outweighs the ventilation induced by the parallel winds (Soulhac et al., 1999; Dabberdt and Hoydysh, 1991).

3.7.3. Spatial variability

Diffusive BTX sampling was proved an efficient technique for revealing the spatial variability of air pollution in roadside microenvironments. The benzene hot spots detected on the predominantly leeward side of Bd. Voltaire, Rue de Rennes and Av. Leclerc were an indirect manifestation of the wind vortex formation within street canyons. This is in agreement with field observations made by Qin and Kot (1993) in three urban canyons in Guangzhou City (China).

A substantial reduction in ambient benzene concentrations along with the height above the ground was also observed in the two symmetric street canyons of Bd. Voltaire and Rue de Rennes. Nevertheless, this variation was smaller than the vertical CO gradients observed in a busy street canyon in Athens, as reported by Zoumakis (1995). In the asymmetric canyon of Av. Leclerc, no clear trend was identified probably due to the presence of big trees, which increased mechanical turbulence and hence vertical mixing within the street.

3.7.4. Temporal variability

Real time CO and NO_x monitoring was used to detect the temporal variability of air pollution in the selected streets. The mobile monitoring unit (i.e. trailer) was located on the east side of Bd. Voltaire, Rue de Rennes and Av. Leclerc, which was most of the time windward during the intensive campaigns. As expected, the time profiles of roadside CO and NO_x concentrations were mainly affected by the variation of the synoptic wind and the traffic flow pattern. The daily maximum concentrations corresponded roughly to the morning and evening rush hours as well as to the lowest recorded roof-level winds. Finally, the canyon effect (i.e. higher leeward concentrations under perpendicular winds) was reflected on the continuous CO and NO_x measurements.

The CO and NO_x values recorded at the AIRPARIF monitoring site in Pl. Basch were much higher than the values observed within the asymmetric canyon sector of Av. Leclerc during the same time period. That mainly reflected the influence of the high traffic density and the short distance between the AIRPARIF station and the sources (i.e. car exhausts). Furthermore, there was evidence that during southerly wind conditions, polluted air masses from Bd. Périphérique were transported toward Pl. Basch, thus contributing to the relatively high CO and NO_x readings.

3.7.5. Compliance with regulations

Comparing the obtained measurements with EU limit values for ambient air (Table 3.8), it was observed that the NO_2 averages over the intensive campaigns exceeded the annual threshold of 40 μg/m^3 (21 ppb). Although the averaging times were different, these exceedences indicate that the air quality objective for NO_2 might not be met in the long run.

By contrast, the hourly mean NO_2 concentrations remained always below the short-term limit value of 200 μg/m^3 (104.6 ppb). Finally, the ambient CO and O_3 levels detected during these campaigns were much lower than the limit values of 10 mg/m^3 (8.5 ppm) and 110 μg/m^3 (55 ppb) respectively.

Benzene concentrations measured with diffusive samplers on the kerbside of Bd. Voltaire, Rue de Rennes, Av. Leclerc and Pl. Basch were several times higher than those detected in the respective background locations. Attention should be drawn to the fact that observed roadside levels for benzene (averaged over 2 to 7 days) exceeded the yearly EU limit value of 5 μg/m^3 (1.6 ppb) for ambient air, although background concentrations remained in almost all cases below the same threshold. In Pl. Basch in particular, long-term (i.e. seven-month) benzene averages at all eleven roadside locations were clearly above the EU limit, while the average value in Montsouris Park was below this threshold for the same time period.

3.7.6. Representativeness of measurements

The strong spatial and temporal variability of traffic-related air pollution detected during the field campaigns in Paris raised the question of how representative the site and time period of air quality measurements actually can be.

Berkowicz et al. (1996) argued that roadside observations are site-dependent and not representative for a larger urban area, after comparing monitoring data from two streets in Copenhagen (Jagtvej and Bredgade). It was demonstrated that measured concentrations could be very different at these two sites, mainly due to the different position of the monitoring stations within the streets. In a later study, Scaperdas and Colvile (1999) showed that air quality measurements made at the intersection of two street canyons in Central London (Marylebone Road) were strongly dependent on the interaction of the local wind flow with the geometry of the streets and buildings surrounding the receptor.

Table 3.8: Measured pollutant concentrations against regulatory limit values. The statistical quantities for O_3, CO and NO_2 were calculated using real time kerbside measurements. The min and max benzene concentrations correspond to diffusive measurements at different sampling locations. The campaign benzene averages were calculated over the available street-level observations.

Pollutant	O_3			CO			NO_2			Benzene		
Unit	ppb			ppm			ppb			ppb		
Averaging time	8 hour		campaign	8 hour		campaign	1 hour		campaign	campaign		
	median	max	average	median	max	average	median	98 percentile	average	min	average ***	max
Bd. Voltaire	5.49	6.91	5.51	1.14	2.39	1.42	27.89	48.37	30.94	1.34	3.50	4.46
Rue de Rennes	16.68	28.56	16.74	0.67	2.00	0.74	25.06	58.17	27.63	0.40	2.43	3.80
Av. Leclerc ****	18.24	31.12	16.19	0.54	1.62	0.56	21.17	54.06	23.84	0.80	2.07	3.09
EU limit value	55 (110 µg/m^3) *			8.5 (10 mg/m^3)			104.6 (200 µg/m^3)		21 (40 µg/m^3) **	1.6 (5 µg/m^3) **		

* French limit value for the protection of human health ** Annual mean *** Roadside average **** 16 - 23 July 2001

In general, monitors should be located near places of expected pollution hotspots but also must be reasonable as related to population exposure over the averaging times associated with ambient air quality regulations. It has been suggested in the past that receptors should be placed at the edge of pavement, at the corner of two intersecting streets. Although such locations may satisfy the criterion of maximum ambient concentrations, it can be argued that pedestrians do not usually spend a lot of time at street corners (Cooper, 1987).

Further to the general siting considerations presented in Section 2.2.6, specific EU guidelines require that sampling points be located between 1.5 m (breathing zone) and 4 m above the ground, at least 25 m away from the edge of any major junctions and at least 4 m from the centre of the nearest traffic lane. For CO, the sampling inlet should be no more than 5 m from the kerbside and for benzene it should be located near the building line (but at least 0.5 m away from the nearest wall) (European Commission, 2000).

The permanent AIRPARIF monitoring station in Pl. Basch does not fulfil the above described siting criteria, since the sampling inlet is placed in the middle of a very busy avenue and very close to a major intersection. At this position, the direct intake of car emissions unmixed with ambient air cannot be avoided. Not surprisingly, the AIRPARIF station in Pl. Basch has recorded the highest CO and NO_x concentrations during recent years in the region of Paris (AIRPARIF, 1999). Furthermore, the CO and NO_x values recorded in this station were much higher than the concentrations observed within the asymmetric canyon section of Av. Leclerc during the intensive monitoring campaign of July 2001. Even though people have access to the narrow traffic island where the AIRPARIF station is permanently located, they do not stay there for long. Therefore, it can be argued that this is not a reasonable monitoring site for estimating population exposure in Paris, although it may be still useful for observing traffic emission trends.

The monitoring campaigns in Bd. Voltaire, Rue de Rennes, Av. Leclerc and Pl. Basch demonstrated that diffusive sampling may efficiently complement continuous monitoring by quantifying the small-scale spatial variability of air pollution in urban streets. In the present study, that was achieved by deploying a limited number of passive samplers at roadside and urban background locations during representative time periods. The sampling sites were carefully selected so as to represent microenvironments where population exposure to traffic-related pollution was expected to be high. Furthermore, an effort was made to cover different seasons and weather conditions. The same sampling strategy was successfully adapted to the specific needs of the motorway service station campaign (RN10).

Continuous monitoring in Pl. Basch (AIRPARIF) showed that the CO average for the year 2001 was very close to the mean CO value over the June to December sampling period (Fig. 3.52). On the other hand, the monthly benzene averages obtained using diffusive samplers were not always representative for longer time periods. For example, in October 2001 benzene concentrations were significantly higher than the campaign (seven-month) averages obtained at the same sites. Weekly benzene averages were also significantly deviating in some cases from the long-term mean values. Therefore, it might be concluded that at least two months of passive sampling (preferably covering two different seasons) are needed in order to obtain representative roadside benzene concentrations, readily comparable to the annual EU threshold.

Finally, field data from few monitoring/sampling sites can be further interpreted and generalised to a wider variety of urban streets using mathematical modelling, as it will be shown in Chapter 4.

3.7.7. Vapour recovery systems

Multi-site BTX sampling in a motorway petrol station (RN10) allowed for a first evaluation of the benefits of Stage 2 vapour control. The adopted methodology gave reliable results without requiring excessive measurements or calculations. Nevertheless, mathematical modelling will be needed in order to examine pollutant dispersion under unfavourable weather conditions (see Section 4.6.6).

It was demonstrated that Stage 2 vapour recovery reduced BTX and especially benzene levels near the fuel pumps and in the surroundings of the selected petrol station. Consequently, population exposure to these substances was mitigated during the operation of the system. Although vapour recovery generally reduced VOC emissions due to displacement losses, the effectiveness of the system was proved to be inversely proportional to the local wind speeds recorded during sampling.

The emissions released in this motorway petrol station gave rise to benzene concentrations that appeared to be lower than those generally occurring in busy urban streets (Hartle, 1993; Vardoulakis et al., 2002a). Therefore, the operation of Stage 2 vapour control in petrol station located in already polluted urban environments is only expected to bring marginal reductions in ambient VOC levels, although the total mass of releases into the atmosphere will be reduced (Gonzalez-Flesca et al., 2002).

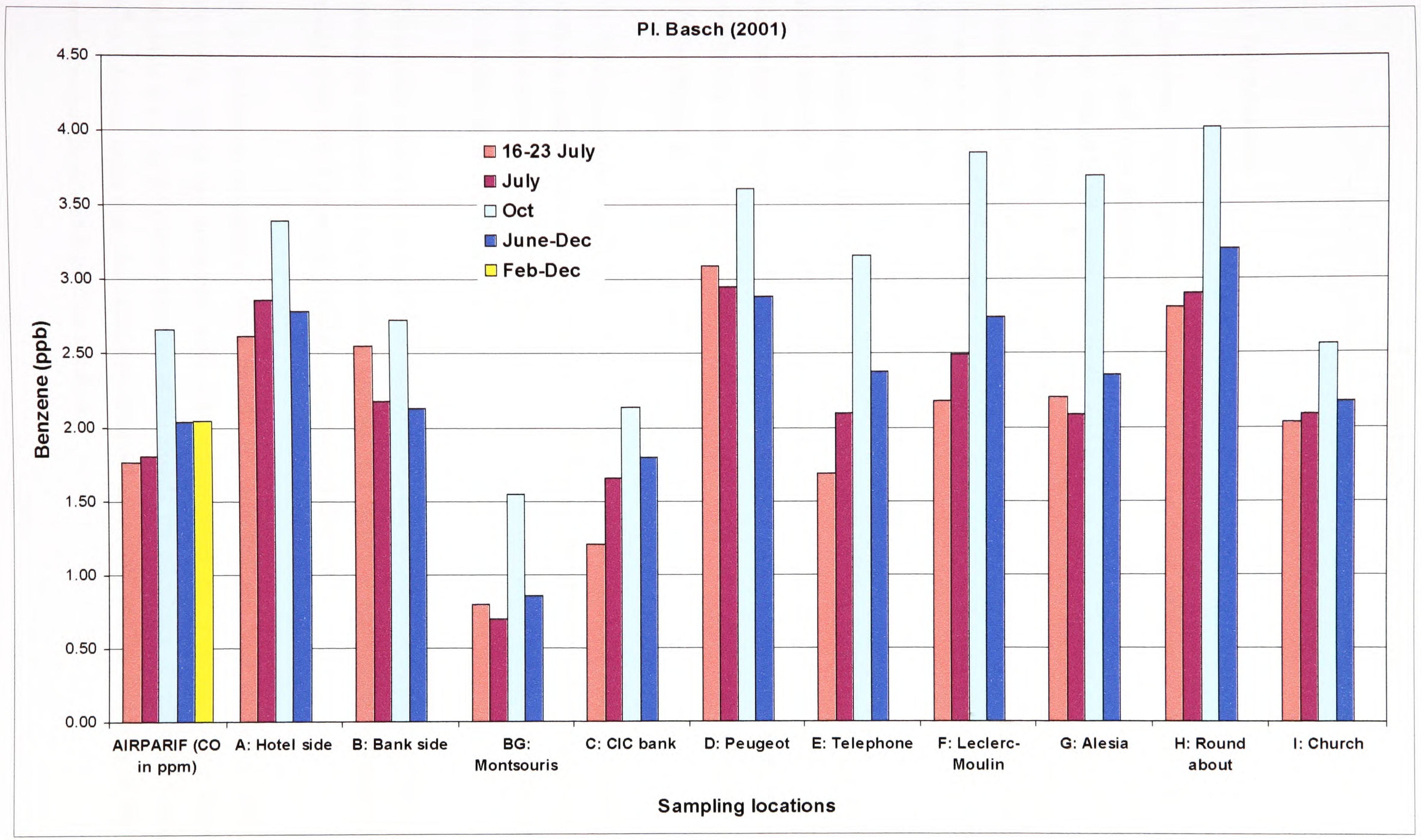


Fig. 3.52: Benzene (ppb) concentrations at 10 passive sampling locations in Pl. Basch, Av. Leclerc and Montsouris Park, and CO (ppm) concentrations at the AIRPARIF monitoring station in Pl. Basch for different averaging periods during the year 2001.

Chapter 4

Dispersion modelling

4.1. Introduction

Mathematical modelling has been widely used for assessing ambient air quality. Government departments, agencies, and local authorities increasingly (but not exclusively) rely on air pollution models for making decisions related to air quality and traffic management, urban planning, and public health. As a result, the model users' community has become larger and more diverse. A recent questionnaire survey targeted at environmental health officers in the UK showed that three-quarters of the responding officers had already used an air pollution model and many of them were proposing to increase the use of this tool in the near future (Beattie et al., 2001).

In the present study, three popular semi-empirical models (STREET-SRI, OSPM and AEOLIUS) were mainly used to calculate CO, NO_x and benzene concentrations within the street canyons of Bd. Voltaire, Rue de Rennes and Av. Leclerc (Vardoulakis et al., 2000a; Vardoulakis et al., 2002a; 2002b). In addition, preliminary calculations with a screening model (CAR International) and a CFD code (PHOENICS) were carried out (Vardoulakis et al., 1999).

The Gaussian plume model CALINE4 was used to estimate the contribution of RN10 to the concentrations occurring near the motorway service station of Rambouillet (RN10) under unfavourable weather conditions (Gonzalez-Flesca et al., 2002). Finally, CALINE4 was also tested for the street canyon of Rue de Rennes (Vardoulakis et al., 2000b).

The selected mathematical models (or variations of them) are likely to be used by local authorities, air quality monitoring networks and government agencies in a variety of applications including air quality and traffic management, urban planning, population exposure studies, etc.

In the following paragraphs, these models are described and their empirical assumptions highlighted. Modelling results are compared with field measurements and evaluated using statistical tools. Finally, emphasis is put on the parameterisation of the models, the calculation of road traffic emissions, the quality of meteorological input data, the estimation of model uncertainty and the suitability of the models for different street configurations and dispersion conditions.

4.1.1. Description of models

STREET-SRI

This empirical model calculates series of hourly concentrations at different receptor locations. The concentrations (C) of the pollutant occurring on the roadside consist of two components, the urban background concentration (C_b) and the concentration component (C_s) due to vehicle emissions generated within the street:

$$C = C_s + C_b \qquad (4.1)$$

The C_s component was derived from a simple box model (Johnson et al., 1973). On the leeward side of the street, pollutant concentrations are proportional to the rate of release of emissions Q (mg/m s) in the street, and inversely proportional to the slant distance $(x^2 + z^2)^{1/2}$ between the receptor and the nearest traffic lane (Fig. 2.1), and the roof-level wind speed U (m/s). This is expressed by the relationship:

$$C_S^L = K \frac{Q}{\left(\sqrt{(x^2 + z^2} + h_o\right)\ (U + U_S)} \qquad (4.2)$$

where K is an empirical constant parameter (usually given the value of 7), x is the horizontal distance between the receptor and the centre of the nearest traffic lane, z is the height of the receptor, h_o is a constant that accounts for the height of initial pollutant dispersion (empirical value: 2 m), and U_S is a constant that accounts for the additional air movement induced by vehicle traffic (empirical value: 0.5 m/s).

On the windward side, the original expression given by Johnson et al. (1973) was revised by Dabberdt et al. (1973) to account for vertical decrease of concentrations due to entrainment of fresh air through the top of the canyon. On that side of the street, pollutant concentrations are proportional to the rate of release of emissions Q, and inversely proportional to the width of the canyon W and the roof-level wind speed U. The entrainment through the top of the canyon is assumed to vary linearly with height z. The final expression is:

$$C_S^W = K \frac{Q}{W(U + U_S)} \frac{H - z}{H} \qquad (4.3)$$

where H is the height of the canyon. For parallel or near-parallel synoptic winds, the average of (4.2) and (4.3) should be calculated.

OSPM and AEOLIUS

AEOLIUS (the Full version) and OSPM are semi-empirical dispersion models based on the same mathematical formulation. They combine Gaussian plume theory with empirical box model techniques to calculate concentrations of exhaust gases in a street canyon assuming three different contributions: The contribution (C_d) from the direct flow of pollutants from the source to the receptor, the recirculation component (C_r) due to the flow of pollutants around the main vortex generated within the recirculation zone of the canyon, and the urban background contribution (C_b):

$$C = C_d + C_r + C_b \tag{4.4}$$

A Gaussian plume model is used to calculate the direct contribution at a receptor located at a down-wind distance x from the line source:

$$dC_d = \sqrt{\frac{2}{\pi}} \frac{Q \cdot dx}{W \cdot u \cdot \sigma_z(x)} \tag{4.5}$$

where u is the street-level wind speed, and $\sigma_z(x)$ the vertical dispersion coefficient given by the expression:

$$\sigma_z(x) = \sigma_w \frac{x}{u} + h_o \tag{4.6}$$

where σ_w is the vertical velocity fluctuation due to mechanical turbulence, and h_o the effective release height of car exhausts due to initial dispersion. Equation (4.5) is integrated along the street-level paths illustrated in Fig. 4.1, giving the expression:

$$C_d = \sqrt{\frac{2}{\pi}} \frac{Q}{W \sigma_w} F \tag{4.7}$$

where F is a factor depending on the synoptic wind. If the roof-top wind is perpendicular to the street axis and the recirculation zone covers the whole canyon, then $F = ln\ \{[\ h_o + (\sigma_w /u)\ W\ ]\ /\ h_o\}$. The vertical velocity fluctuation σ_w due to mechanical turbulence generated by the wind and the moving vehicles in the street is described by the relationship:

$$\sigma_w = \sqrt{(\alpha u)^2 + \sigma_{wo}^2} \qquad (4.8)$$

where α is a proportionality constant given empirically the value of 0.1, which corresponds to typical levels of mechanically induced turbulence, and σ_{wo} is the traffic-induced turbulence. This turbulence is calculated using a simple approach that considers vehicles in the street as moving distortion elements creating additional mechanical turbulence (Hertel and Berkowicz, 1989c):

$$\sigma_{wo}^2 = b^2 V^2 D \qquad (4.9)$$

where b is an aerodynamic drag coefficient (given empirically the value of 0.3), V is the average vehicle speed, and D the density of moving elements given by the relative area of the street occupied by vehicles:

$$D = \frac{N \cdot S^2}{V \cdot W} \qquad (4.10)$$

where N is the number of vehicles using the street per time unit, S^2 the road surface occupied by a single vehicle, and W the width of the canyon. The combination of equations (4.9) and (4.10) gives the following expression:

$$\sigma_{wo} = b\left(\frac{N \cdot V \cdot S^2}{W}\right)^{1/2} \qquad (4.11)$$

according to which, the vehicle induced turbulence is proportional to the traffic flow ($N{\cdot}V$) and inversely proportional to the width of the canyon.

The contribution from the recirculation zone is calculated using a simple box model, which assumes that the recirculation zone has the shape of a trapezium (Fig. 4.2). The ventilation of the recirculation zone takes place through the edges L_t, L_{S1} and L_{S2} of the trapezium. The pollutant inflow rate (per unit length) is $Q{\cdot}L_r /W$, where

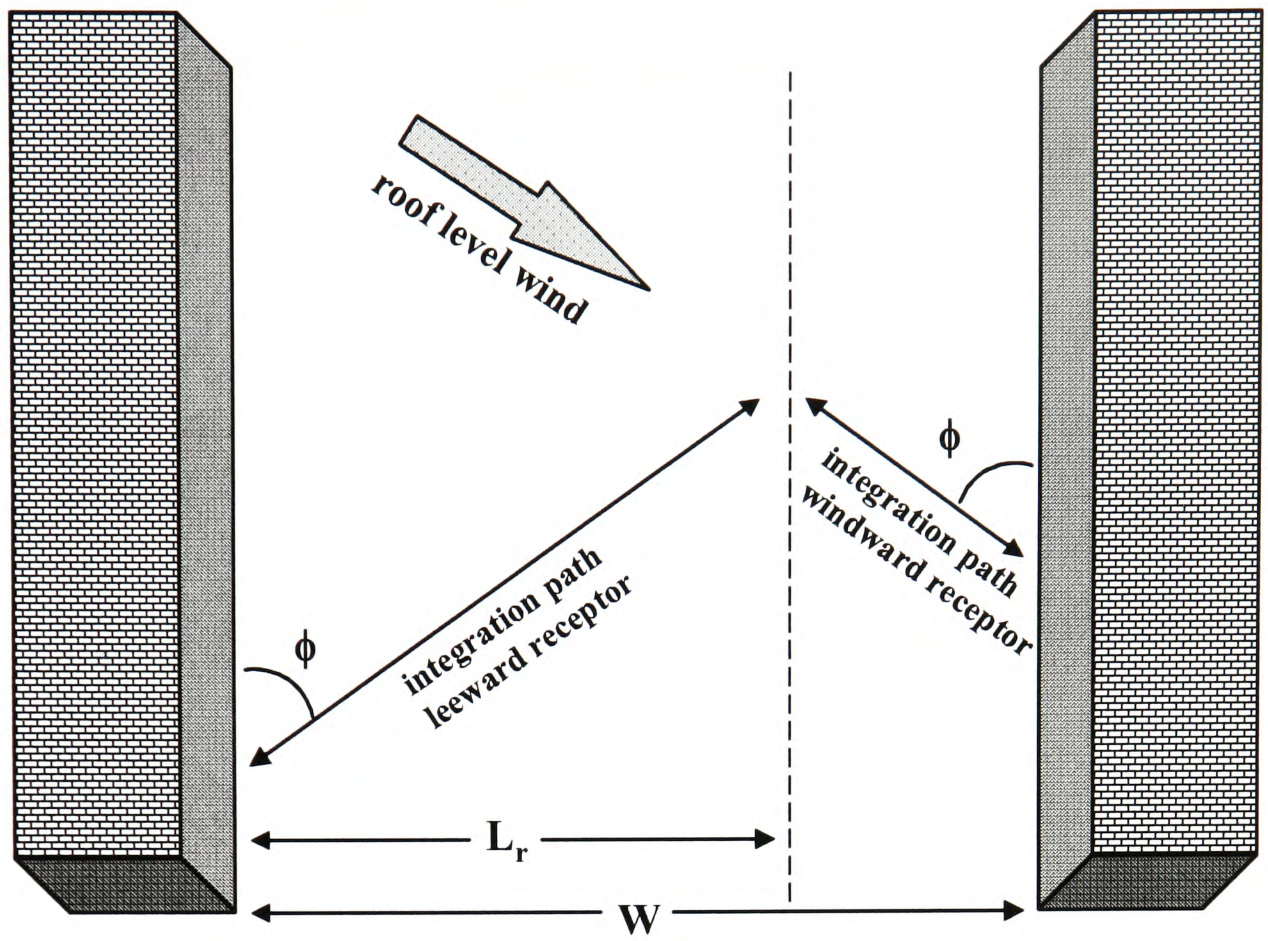


Fig. 4.1: OSPM and AEOLIUS: Formation of the recirculation zone within a street canyon (plan view).

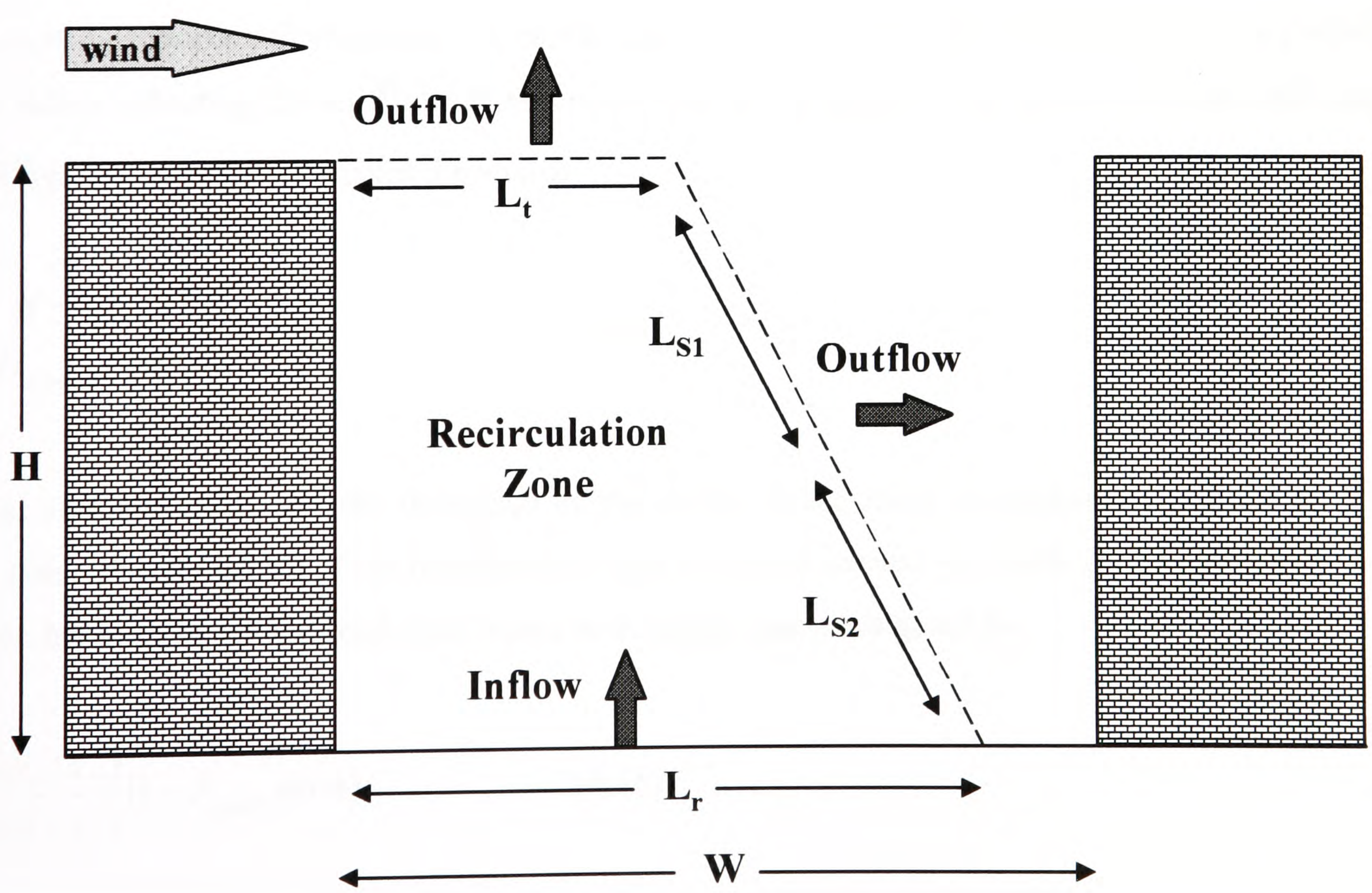


Fig. 4.2: OSPM and AEOLIUS: Geometry of the recirculation zone (cross section).

L_r is the width of the recirculation zone, and the outflow rate is $C_r \cdot (\sigma_{wt} L_t + UL_{S1} + uL_{S2})$. Assuming that the pollutants are well mixed inside the trapezium, the recirculation contribution is then given by the relationship:

$$C_r = \frac{Q}{W} \frac{L_r}{\sigma_{wt} L_t + UL_{S1} + uL_{S2}} \qquad (4.12)$$

where U is the roof-level wind speed, and σ_{wt} the ventilation velocity through the top of the canyon, expressed as:

$$\sigma_{wt} = \sqrt{(\lambda U)^2 + F_{roof} \sigma_{wo}^2} \qquad (4.13)$$

where λ and F_{roof} are proportionality constants given the value of 0.1 and 0.4, respectively. Although λ is currently given the same constant value as α, it might be sensitive to atmospheric stability variations. The extension L_r of the recirculation zone is defined as:

$$L_r = F_{vortex} \cdot H \cdot r \sin \phi \qquad (4.14)$$

where F_{vortex} is a proportionality constant given the value of 2, H is the height of the canyon, r is a wind speed dependent factor reflecting the strength of the vortex, and ϕ the angle of the roof-level wind with respect to the street (Fig. 4.1). The factor r takes the values:

$$\begin{Bmatrix} r = 1 & if & U > U_{critical} \\ r = U / U_{critical} & otherwise & \end{Bmatrix} \qquad (4.15)$$

The critical velocity $U_{critical}$ for the formation of the vortex in the street is empirically defined as 2 m/s. It should be noted that the width of the recirculation zone L_r cannot exceed the width of the canyon in any case. The relation between street and roof-level winds in a regular canyon is given by:

$$u = U \frac{\ln(h_o / z_o)}{\ln(H / z_o)} (1 - F_{wind} \sin \phi) \qquad (4.16)$$

where z_o is the surface roughness length of the area under consideration, h_o the effective release height of car exhausts, and F_{wind} an empirical constant given the value of 0.2. The wind speed at roof-level (U) is calculated from the input wind (U_a), which corresponds to a meteorological mast of generally different height, using the simple relationship:

$$U = F_{mast} \cdot U_a \qquad (4.17)$$

The empirical parameter F_{mast} is given the value of 0.82, which is derived from a logarithmic law similar to expression (4.16).

On the leeward side of the street, concentrations are calculated as the sum of the direct and recirculation contributions, while on the windward side only the direct contribution of emissions generated outside the recirculation zone are taken into account. If the recirculation zone extends throughout the whole canyon, then the windward concentrations are calculated from only the recirculation component. For near-parallel flow, emissions from outside the recirculation zone may contribute to the leeward concentrations. When the wind speed is near zero or parallel to the street axis, the concentrations on both sides of the canyon become equal. In all cases, the background contribution should be added to obtain the final result.

4.2. Model sensitivity

The above described models contain a number of constant parameters that have been empirically defined using experimental data. Although they might have a significant influence on model predictions and hence on the interpretation of the results, these constants have drawn relatively little attention so far. Comparing model results with experimental data, inappropriate values of such model constants might be falsely interpreted as unsatisfactory emission factors or meteorological input data (Buckland and Middleton, 1999).

STREET-SRI model includes three empirical parameters K, h_o and U_S (eqs. 4.2 and 4.3), which have been adjusted to observed results. Johnson et al. (1973) derived initially the values of $K = 7$, $h_o = 2$ m and $U_S = 0.5$ m/s, using data from the San Jose Street Canyon Experiment. The value of $K = 7$ is presumably valid for canyons having $H/W \cong 1$, which is comparable to the aspect ratio of the street canyons in San Jose. A subsequent evaluation by Dabberdt et al. (1973) did not suggest dramatic variation in K for two narrower canyons with H/W of 1.5 and 2 in St. Luis. Yamartino and Wiegand (1986) kept the original values for h_o and U_S, but allowed K to rise to an optimal value of 10.2 in their evaluation of STREET-SRI model using measurements from Bonner Strasse ($H/W \cong 1$), Cologne. In a research study in China, K was given the value of 6 to simulate concentrations observed in an asymmetric street canyon in Guangzhou City (Qin and Kot, 1993). Finally, in a recent experiment in a street canyon in Buenos Aires ($H/W \cong 1$), best fit was obtained for

K = 8 (Bogo et al., 2001). Obviously, the value of K weighs heavily on calculations, since it is directly proportional to the model output.

Buckland and Middleton (1999) and Manning et al. (2000) carried out sensitivity studies by varying a number of input variables and internal empirical parameters within AEOLIUS and assessing their impacts on the calculated NO_x and CO concentrations, respectively. It was found that predicted values were almost linearly proportional to the emission factors and traffic volume. Canyon geometry did not significantly affect perpendicular concentrations for a given traffic flow and wind speed. However, that was not the case for parallel concentrations, which increased with canyon height due to the longer integration path before the plume escaped from the canyon, and decreased with canyon width. Increasing surface roughness was shown to enhance calculated concentration. Concentrations decreased at higher traffic speeds as the turbulent mixing increased, although this parameter did not appear to have a dramatic effect on the results. A change in the model coding of the extent of the vortex across the street produced only a minor change in leeward concentrations. Buckland and Middleton (1999) altered the model to let the user specify whether the wind speed was measured at 10 m height (as it is usually the case in airports) or at roof-level. That had only a small effect on the results. A constant value (0.1 m/s) was tested for replacing equation (4.11), which calculates vehicle-induced turbulence. This generated much larger concentrations. Finally, it was shown that the assumed magnitudes of arbitrary model constants such as b or λ had a marked effect on calculated concentrations.

4.2.1. Internal model parameters

In the present study, a sensitivity analysis was performed to determine the importance of various internal model parameters on the output of OSPM. Minor modifications were made in the code of the model, so as to enable the user to define externally the values of the following empirical constants: α, b, λ, z_o, h_o, F_{roof}, F_{vortex}, F_{mast}, F_{wind}, and $U_{critical}$. The values of these parameters were then perturbed to observe the effects on the predicted CO concentrations for perpendicular (i.e. leeward and windward) and parallel wind conditions. For the sensitivity runs, the characteristics of the regular canyon of Rue de Rennes were used, while the input wind speed was maintained constant (3.5 m/s). A large number of diagrams were produced (Fig. 4.3 - 4.12).

As expected, most of the conclusions from the previous sensitivity analysis for AEOLIUS were also valid for OSPM. An increase in surface roughness (z_o) enhanced parallel and leeward concentrations, but had no effect on windward concentrations (Fig. 4.3). Larger z_o values would be expected to reduce the street-level wind, but increase the turbulence within the canyon (Manning et al., 2000). However, OSPM simply reduced the street-level wind, which in turn lowered the mechanical turbulence (eq. 4.8) and increased concentrations.

The effective release height (h_o) had almost the opposite effect on the calculated results (Fig. 4.4). This parameter, which is user defined in AEOLIUS but originally coded inside OSPM (h_o = 2 m), also represents the height of the simulated roadside receptor. In this analysis, parallel and leeward concentrations decreased with increasing h_o, because of the consequent increase in street-level wind speed (eq. 4.16). The values on the windward side remained constant, because in this case the recirculation zone covered the whole canyon.

OSPM perpendicular concentrations were proved to be quite sensitive to the value of λ (Fig. 4.5), which sets the rate at which material is dispersed out of the top of the canyon, but there was no effect on the parallel concentrations. That was expected because λ only affects the recirculation component C_r (eqs. 4.12 and 4.13), which disappears when the synoptic wind becomes parallel to the street axis ($\sin\phi$ = 0) (eq. 4.14). It should be also noted that λ might be sensitive to seasonal weather variations, since atmospheric stability can play a role in the ventilation of street canyons.

Parameter α, which corresponds to the mechanical turbulence created by the wind at street level, had no marked effect on OSPM results for the values tested (Fig. 4.6). That is because the traffic-induced turbulence (σ_{wo}) dominates over the wind generated turbulence in equation (4.8), due to the relatively high traffic flow and the low street-level wind speed introduced in the sensitivity simulations.

As it can be observed in Fig. 4.7, the aerodynamic drag coefficient b had certain influence on the parallel and leeward concentrations. This proportionality parameter weighs heavily in the calculation of the traffic-induced turbulence (eq. 4.11), which is the dominant term in equation (4.8). On the other hand, it only plays a small role in the calculation of the recirculation contribution (eq. 4.12), which determines windward concentrations. However, when assigning empirical values to b, it should be remembered that the aerodynamic drag of cars is likely to diminish in the future due to improvements in the design of new vehicles.

The influence of the coefficient F_{mast} (eq. 4.17) was found to be of minor importance (Fig. 4.8). Nevertheless, OSPM code was here slightly modified by replacing relationship (4.17) with the following:

$$U = U_a \frac{\ln\left(\dfrac{H}{z_o}\right)}{\ln\left(\dfrac{H_a}{z_o}\right)} \qquad (4.18)$$

which allows the user to specify the height of the anemometer (H_a); this information may be easily and accurately obtained from weather station operators.

By altering the value of F_{vortex} (eq. 4.14), a significant effect on leeward and windward concentrations was produced due to changes in the dimensions of the recirculation zone. For example, a change from the standard value of 2 to the value of 1 results in a 31% and 85% decrease in leeward and windward CO concentrations, respectively (Fig. 4.9). When the recirculation zone becomes very small ($F_{vortex} < 0.4$), the windward concentrations become higher than the leeward concentrations, due to the increasing direct contribution of emissions generated outside the recirculation zone. The model is insensitive to values $F_{vortex} \geq 2$, for which the wind vortex covers the whole canyon. No effect was expected on the parallel concentrations, since the recirculation zone disappears in that case.

The F_{wind} coefficient (eq. 4.16) had a significant influence only on leeward concentrations (Fig. 4.10), since it mainly affects the dispersion of pollutants coming directly from the source. An increase from the standard value of 0.2 to the value of 0.4 produced 11% higher leeward concentrations.

Fig. 4.11 shows that when the synoptic wind (3.5 m/s) was below the critical wind speed $U_{critical}$ for vortex formation (eq. 4.15), the calculated windward and leeward concentrations were significantly lower than when the vortex was formed inside the canyon. For winds parallel to the street axis, the vortex always disappears.

Finally, the coefficient F_{roof} (eq. 4.13) had no marked effect on the model results for the range of tested values (Fig. 4.12). This is because the traffic-induced turbulence (σ_{wo}) generally plays a very small role in the dispersion of pollutants through the top of a regular canyon.

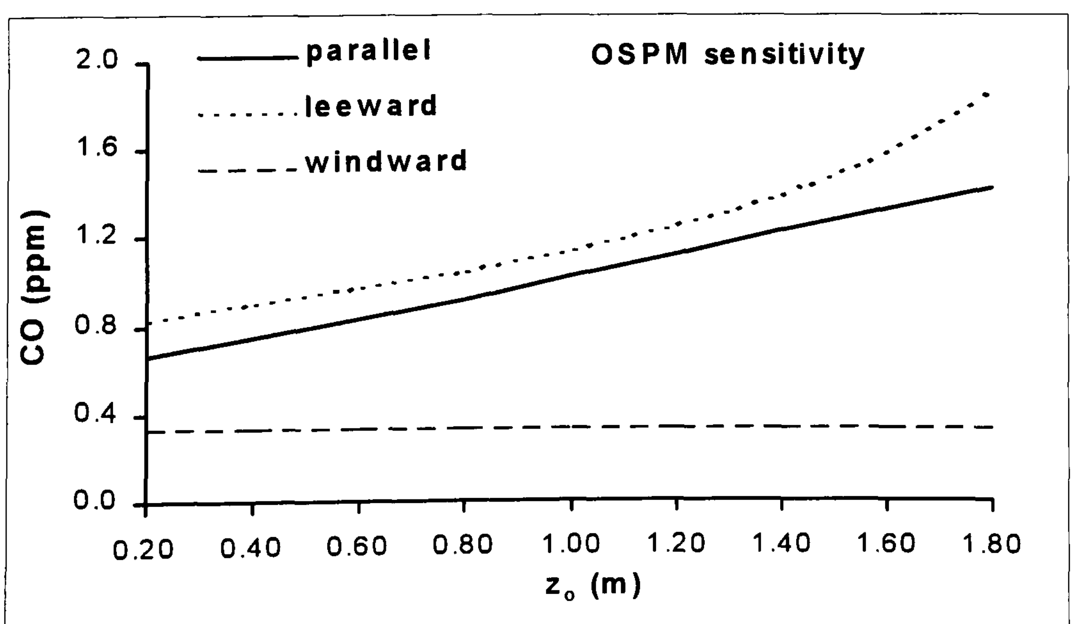


Fig. 4.3: OSPM sensitivity to the surface roughness length (z_o) for three different wind regimes.

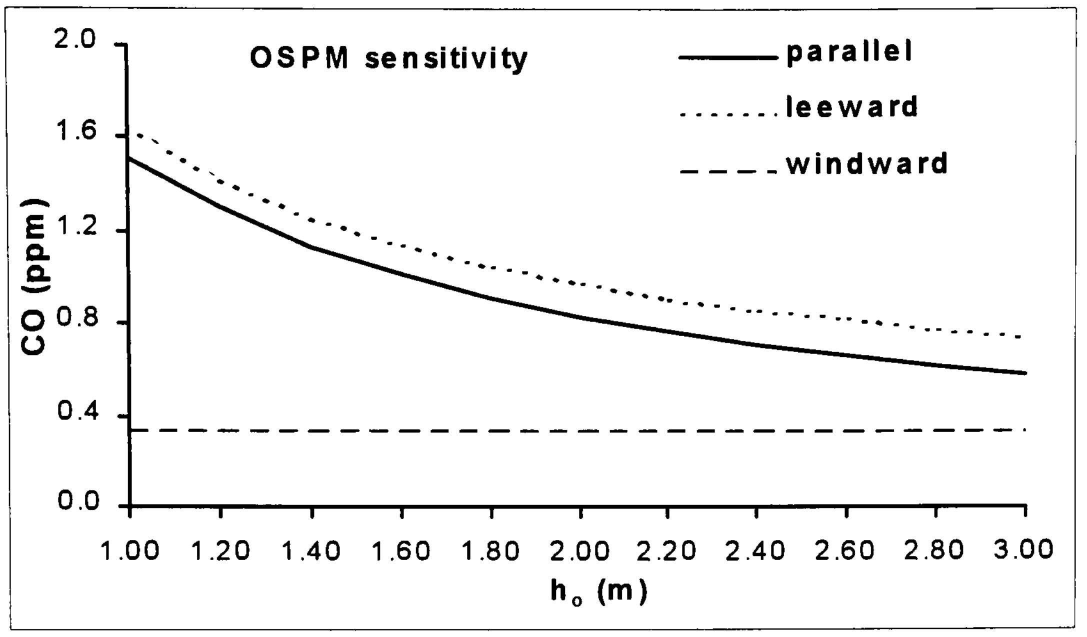


Fig. 4.4: OSPM sensitivity to the effective release height (h_o) for three different wind regimes.

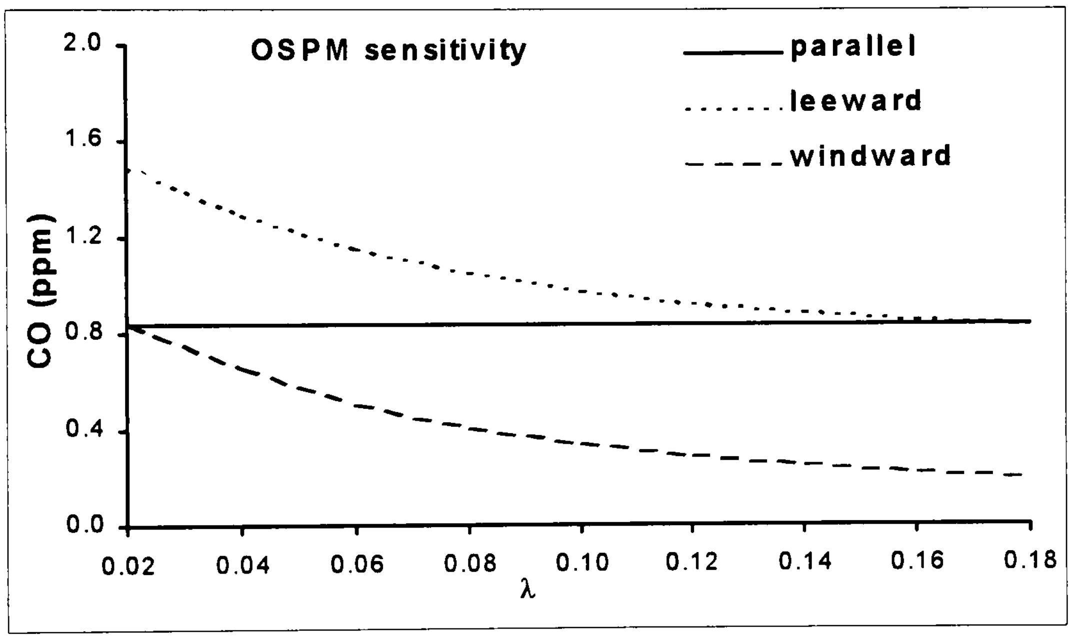


Fig. 4.5: OSPM sensitivity to the canyon ventilation coefficient (λ) for three different wind regimes.

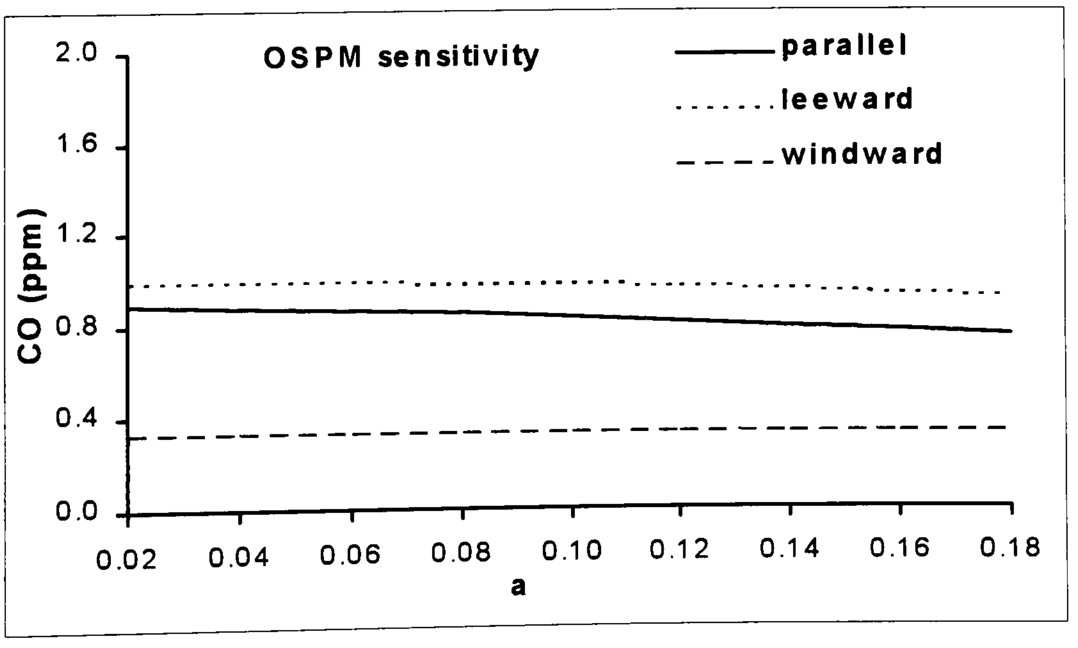


Fig. 4.6: OSPM sensitivity to the mechanical turbulence coefficient (α) for three different wind regimes.

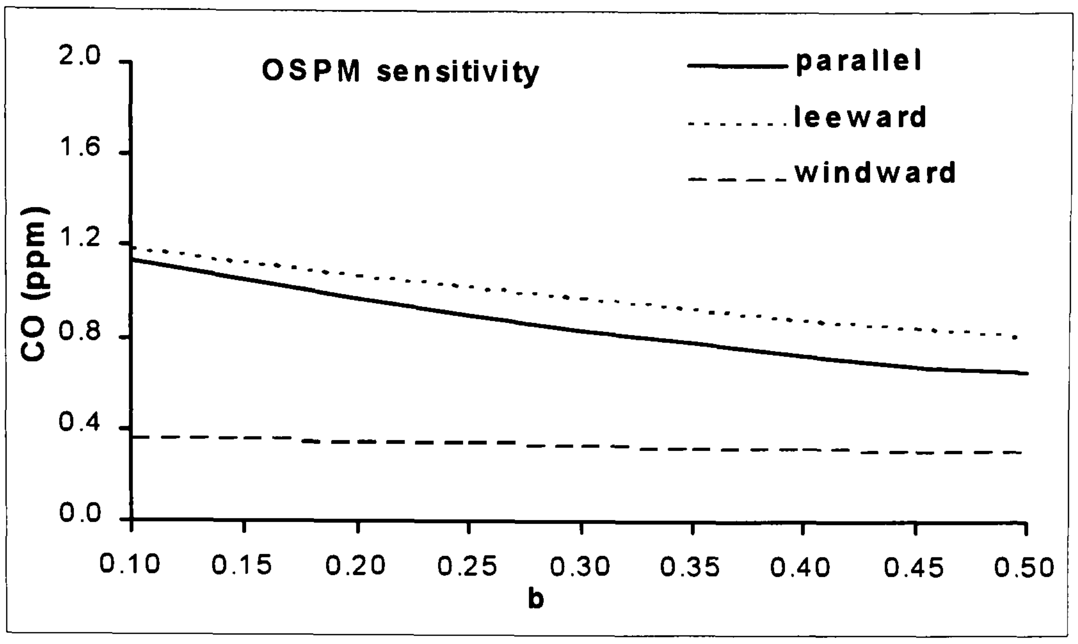


Fig. 4.7: OSPM sensitivity to the aerodynamic drag coefficient (b) for three different wind regimes.

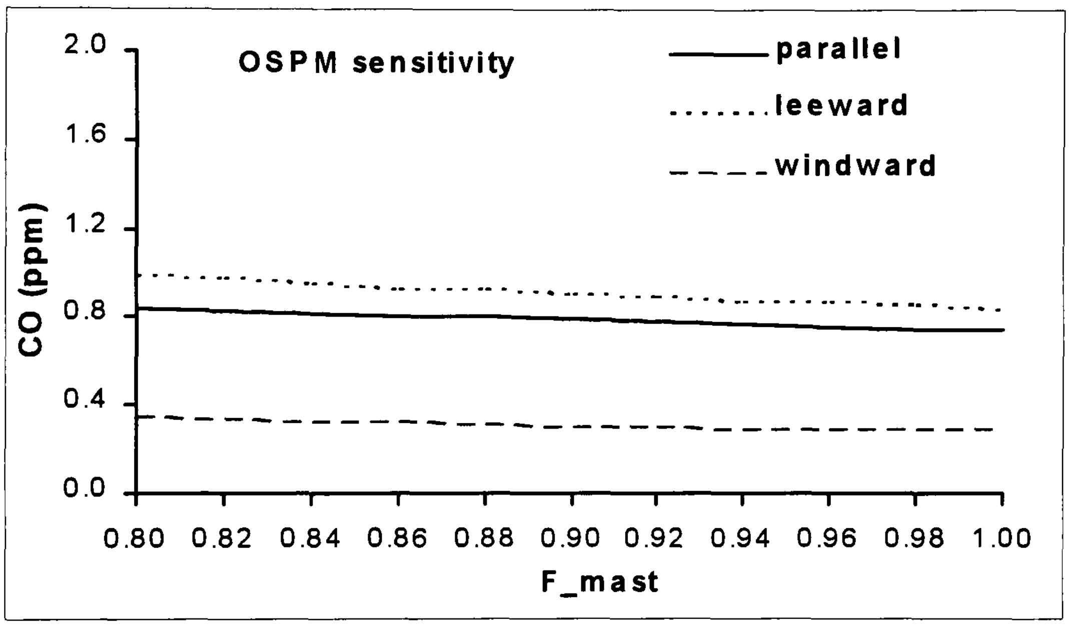


Fig. 4.8: OSPM sensitivity to the anemometer height coefficient (F_{mast}) for three different wind regimes.

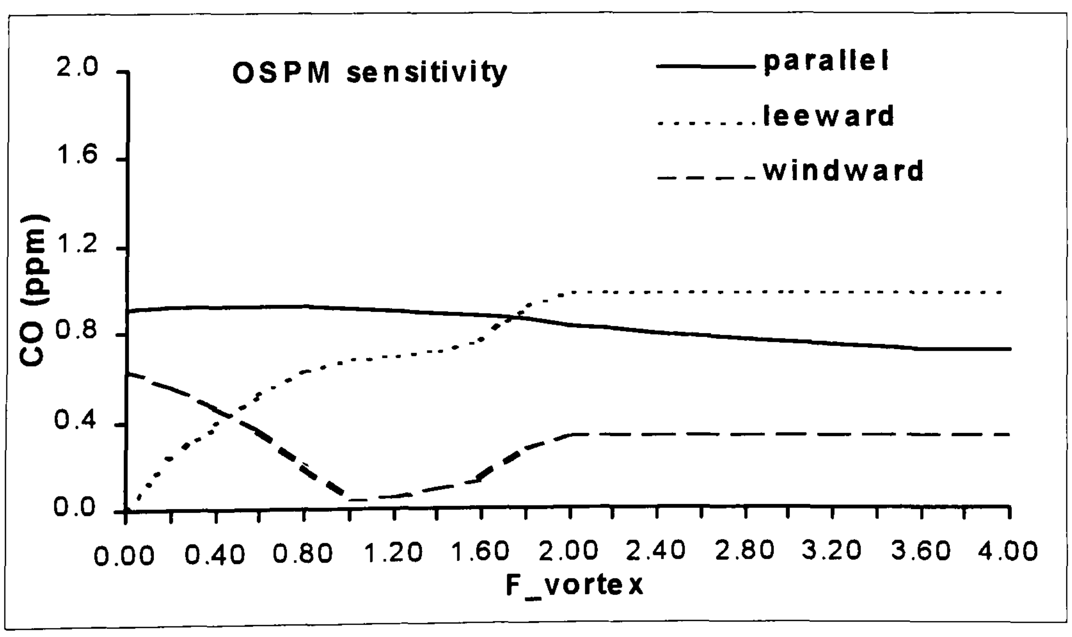


Fig. 4.9: OSPM sensitivity to the vortex length coefficient (F_{vortex}) for three different wind regimes.

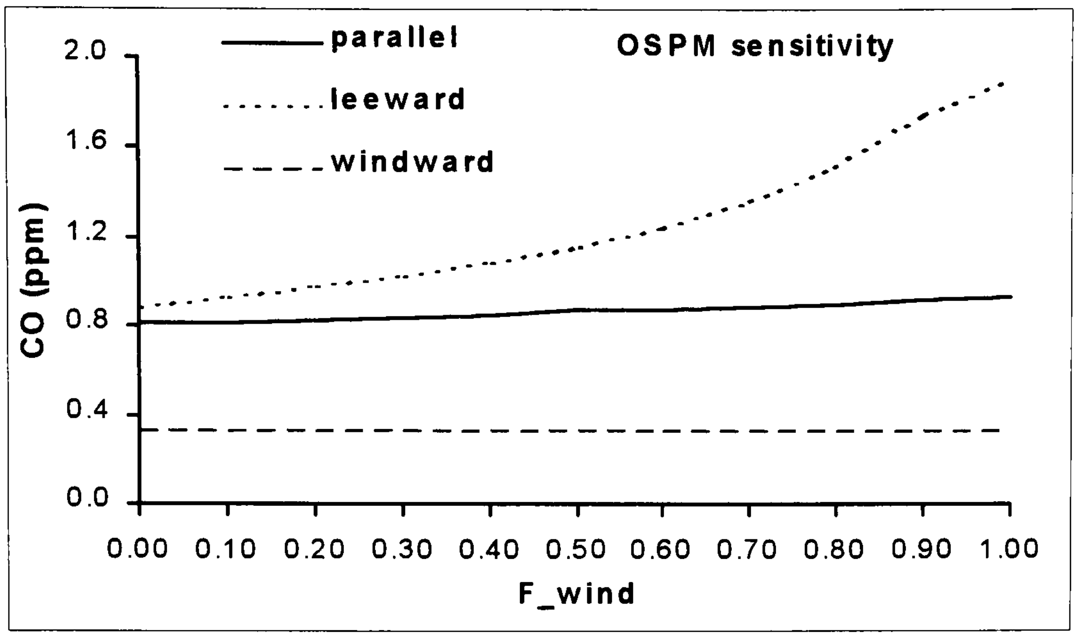


Fig. 4.10: OSPM sensitivity to the street-level wind coefficient (F_{wind}) for three different wind regimes.

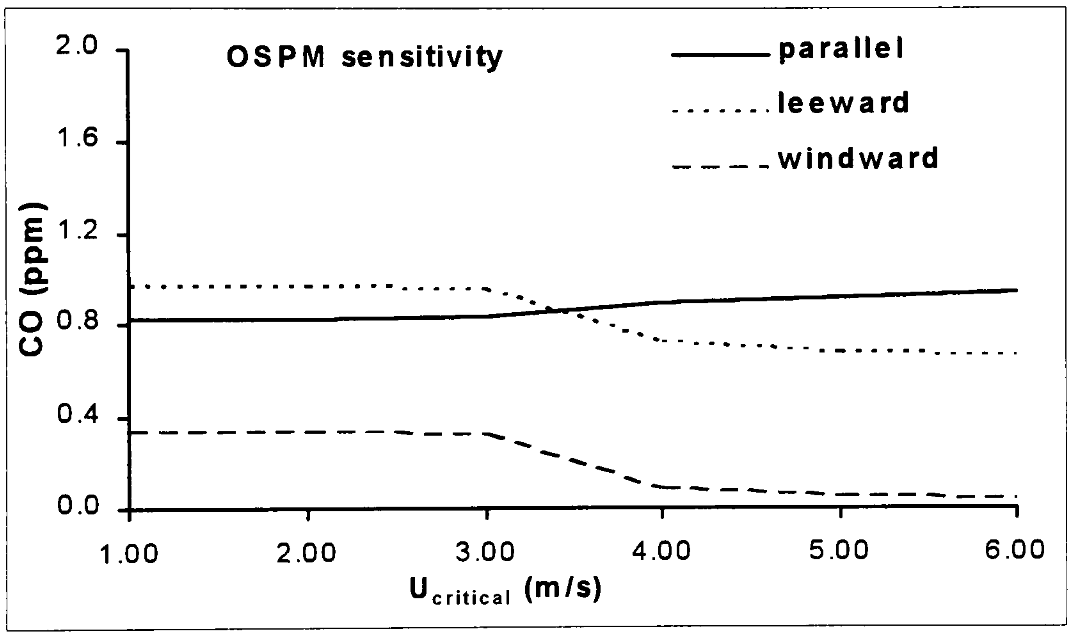


Fig. 4.11: OSPM sensitivity to the critical wind speed for vortex formation ($U_{critical}$) for three different wind regimes.

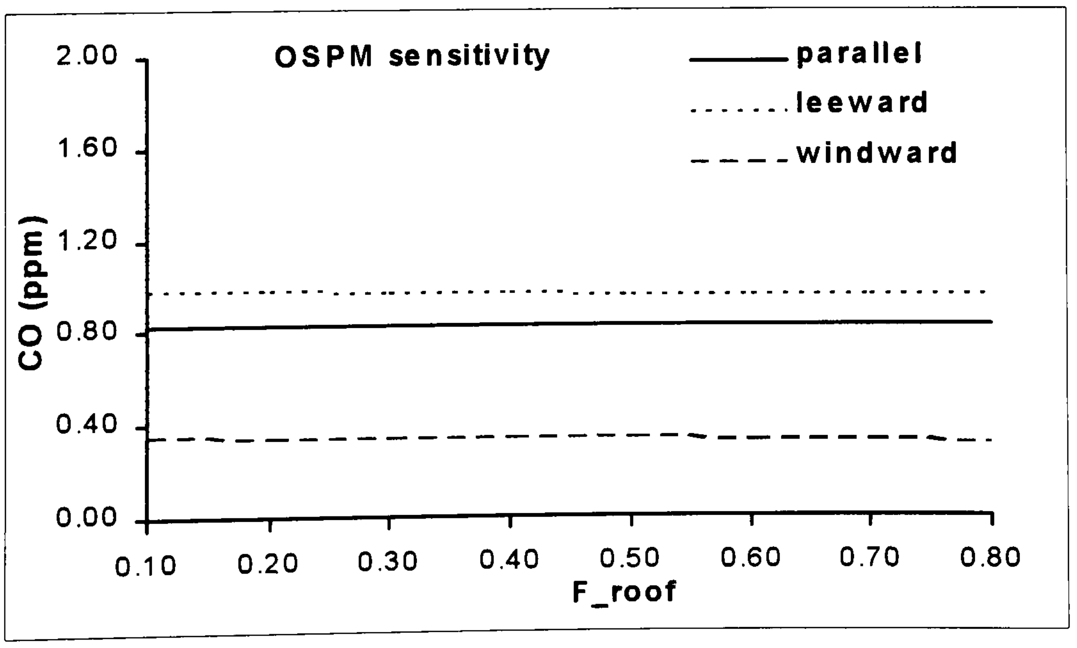


Fig. 4.12: OSPM sensitivity to the traffic turbulence coefficient (F_{roof}) for three different wind regimes.

4.3. Vertical concentration profiles

Relatively simple street canyon models (e.g. OSPM, AEOLIUS, CAR, etc.) only calculate street-level pollutant concentrations, without giving the user the possibility of choosing the height of the simulated receptors.

OSPM, for example, calculates pollutant concentrations only at the effective height of exhaust gas release (≈ 2 m) on both sides of the street. However, pollution levels at different vertical distances from the road surface may be important from a population exposure point of view. For this reason, an external algorithm that enables the user to establish vertical pollution profiles should be introduced.

It has been suggested by several authors in the past (Capannelli et al., 1977; Huang, 1979; Dabberdt and Hoydysh, 1991; Zoumakis, 1995) that the vertical concentration profiles C(z) in a street canyon generally satisfy a law of exponential reduction with height (z), although more complex patterns depending on the side of the street, the distance from the walls, and the small-scale features of the buildings may be also observed (Kastner-Klein and Plate, 1999; Jicha et al., 2000). According to the exponential reduction law, the vertical concentration profile in the street is given by the expression:

$$C(z) \approx A \exp\left[-B\left(\frac{z}{H}\right)^q\right] \qquad (4.19)$$

where A, B, and q are regression coefficients. Although empirically defined, the coefficients A, B and q are generally dependent on the wind direction, atmospheric stability and the aerodynamic characteristics of the canyon (Zoumakis, 1995). According to Georgii (1969), and Hoydysh and Dabberdt (1988), the vertical profiles may be reasonably well approximated by the simple exponential function, where q = 1. Zoumakis (1995) proposed for the q values ranging from 1 to 4.5, and for B from 1.1 to 2.9. Moreover, he identified a significant dependence of q on B. Sacré et al. (1995) suggested that the coefficient B varies in a way that can be empirically described by the relationship:

$$B = 1.6 \cos\phi + 0.4 \qquad (4.20)$$

where ϕ is the angle between roof-top wind and street axis. From equation (4.19), a general expression relating pollutant concentrations at two different heights in the street can be deduced:

$$C(z) \approx C_r \exp\left(-B \frac{z^q - z_r^q}{H^q} \right) \qquad (4.21)$$

where C_r is the concentration of the pollutant at a reference height z_r on either side of the canyon.

4.4. Input data

The required input data for the dispersion model simulations of this study were collected at the sites of the monitoring campaigns during the corresponding sampling periods or obtained from competent authorities.

For the street canyon simulations, synoptic wind data obtained from three different Météo France weather stations were used (see Section 3.1.1), while locally measured winds were input into CALINE4 in the case of the RN10 motorway.

Traffic volumes and average vehicle speeds provided by the automatic traffic monitoring network of Paris were used in the calculations, except for the case of RN10 where traffic density had to be estimated from manual counts. The site-specific vehicle fleet composition was in all cases estimated from field observations.

The dimensions of the streets and the exact receptor locations were manually measured during the campaigns. Finally, the model background input for CO was estimated from relationship (3.4), using the relative street and background contributions derived from diffusive benzene measurements.

4.4.1. Traffic emissions

The rate of release of emissions in the street was derived from the traffic volumes and the composite emission factor of the pollutant. Two different methods were applied for calculating CO emission factors: (a) The protocol used by Buckland and Middleton (1999), which was based on values specific to UK vehicles, and (b) the IMPACT road traffic emission model commercialised by ADEME (see Section 2.3.6).

The values estimated using both the methods were compared for consistency with CO emission factors specific to the French vehicle fleet reported in other recent studies (Touaty and Bonsang, 2000; Jones et al., 2000). All values are summarised in Table 4.1.

Table 4.1: CO emission factors for road transport in the region of Paris.

Author	CO emission factors (g/km veh)			
	Bd. Voltaire 1998	Rue de Rennes 1999	Av. Leclerc 2001	Paris 1996-7
Buckland and Middleton	7.23	8.73	7.99	
IMPACT (ADEME)	20.29	10.91	10.08	
Touaty and Bonsang				8.11
Jones et al.				8.82

4.5. Statistical methods

Six statistical evaluation methods are commonly used to quantify differences between predicted (C_p) and observed (C_o) concentrations (Cox and Tikvart, 1990; Yadav and Sharan, 1996):

1. The fractional bias (FB), which provides information on the tendency of the model to overestimate or underestimate the observed concentrations (overbars denote mean values):

$$FB = \frac{\left(\overline{C}_o - \overline{C}_p\right)}{0.5\left(\overline{C}_o + \overline{C}_p\right)} \qquad (4.22)$$

2. The normalised mean square error (NMSE), which provides information on the overall deviations between predicted and observed concentrations:

$$NMSE = \frac{\overline{\left(C_o - C_p\right)^2}}{\overline{C_o} \cdot \overline{C_p}} \qquad (4.23)$$

3. The correlation coefficient, which describes the degree of association between variables:

$$r = \frac{\sum[(C_o - \overline{C}_o)(C_p - \overline{C}_p)]}{\sqrt{\sum(C_o - \overline{C}_o)^2 \sum(C_p - \overline{C}_p)^2}} \qquad (4.24)$$

4. The fraction of predictions within a factor of two (FAC2):

$$0.5 \leq \left(C_p / C_o\right) \leq 2 \qquad (4.25)$$

5. The geometric mean variance (VG), which is a substitute for the normalised mean square error (NMSE):

$$VG = \exp\left[\overline{\left(\ln C_o - \ln C_p\right)^2}\right] \qquad (4.26)$$

6. The geometric mean bias (MG), which is a substitute for the fractional bias (FB):

$$MG = \exp\left(\overline{\ln C_o} - \overline{\ln C_p}\right) \qquad (4.27)$$

The geometric mean variance (VG) and the geometric mean bias (MG) find relevance when there is a wide range of C_p and C_o values in the data set (Hanna, 1993).

4.6. Modelling results

4.6.1. STREET-SRI

Equations (4.2) and (4.3) were used to simulate CO concentrations in Bd. Voltaire, Rue de Rennes and Av. Leclerc. The rate of release of emissions Q was derived from the traffic volume and the composite emission factor for the specific street calculated according to Buckland and Middleton's (1999) methodology. Input wind data were obtained from Montsouris station. Then, the calculated hourly averages were compared with the concentrations measured on the kerbside during the campaigns.

Although the model reproduced reasonably well the diurnal variation pattern of CO in the three canyons, it appeared to underestimate the observed concentrations mainly in Bd. Voltaire (Fig. 4.13), but also in Rue de Rennes (Fig. 4.14). In the case of Av. Leclerc, the model significantly over-predicted most of the observed CO values (Fig. 4.15).

In Fig. 4.16, the calculated concentrations for Rue de Rennes were stratified into three different wind regimes, where it can be reasonably argued that the physical processes affecting dispersion were similar. This analysis revealed a good linear agreement ($r = 0.79$) between observations and predictions on the leeward side of the canyon when the wind was perpendicular to the street axis. On the other hand, the performance of the model was not satisfactory for the windward side of the street when the wind was perpendicular, and for both sides during parallel wind conditions. In these cases, the model mostly under-predicted the ambient CO concentrations.

Using the empirical relationship (3.4), average benzene values were calculated for different locations in the streets over the passive sampling periods and added to the observed background concentrations. The comparison of the total calculated values with the relevant field measurements showed a very good linear correlation for Bd. Voltaire, although the model seemed to significantly under-predict the observed concentrations (Fig. 4.17). In the case of Rue de Rennes, a very good general agreement was observed, despite some small under-predictions related to the lower concentrations observed near the top of the canyon (Fig. 4.18). Finally, in the case of Av. Leclerc, the model clearly over-predicted all the observed benzene values, although a good correlation between measurements and predictions was again identified (Fig. 4.19).

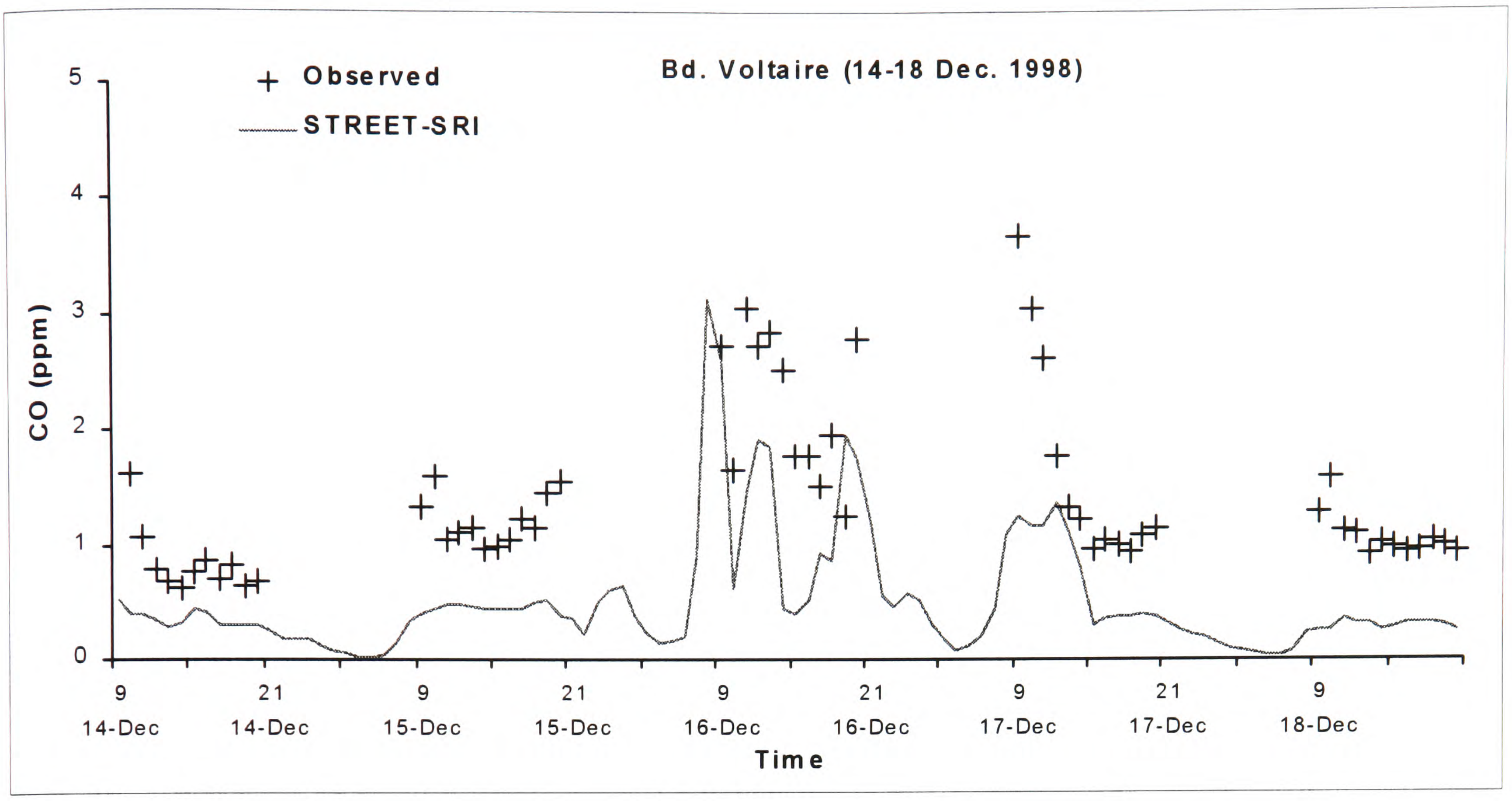


Fig. 4.13: Hourly mean CO (ppm) concentrations observed in Bd. Voltaire and STREET-SRI predictions.

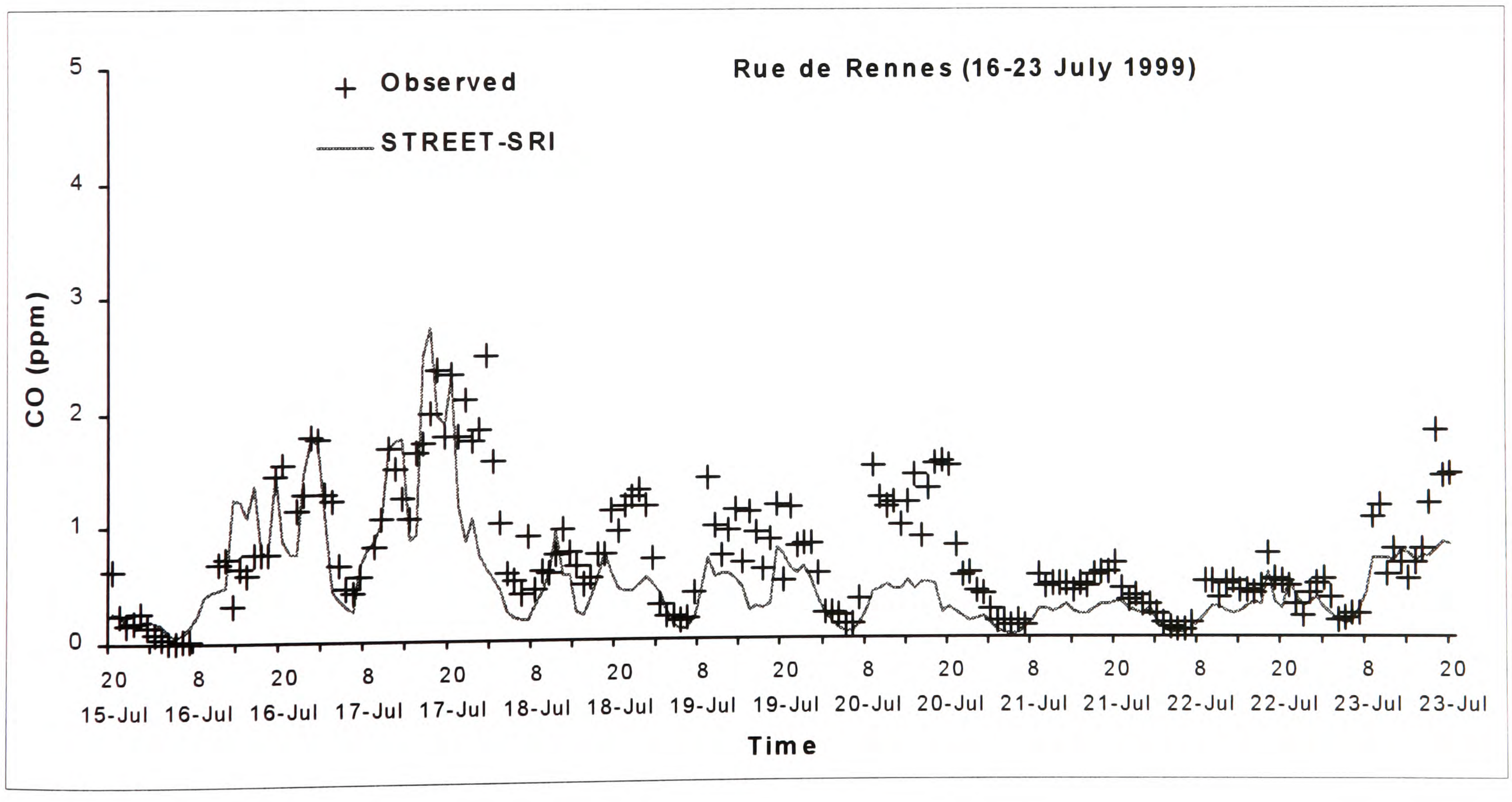


Fig. 4.14: Hourly mean CO (ppm) concentrations observed in Rue de Rennes and STREET-SRI predictions.

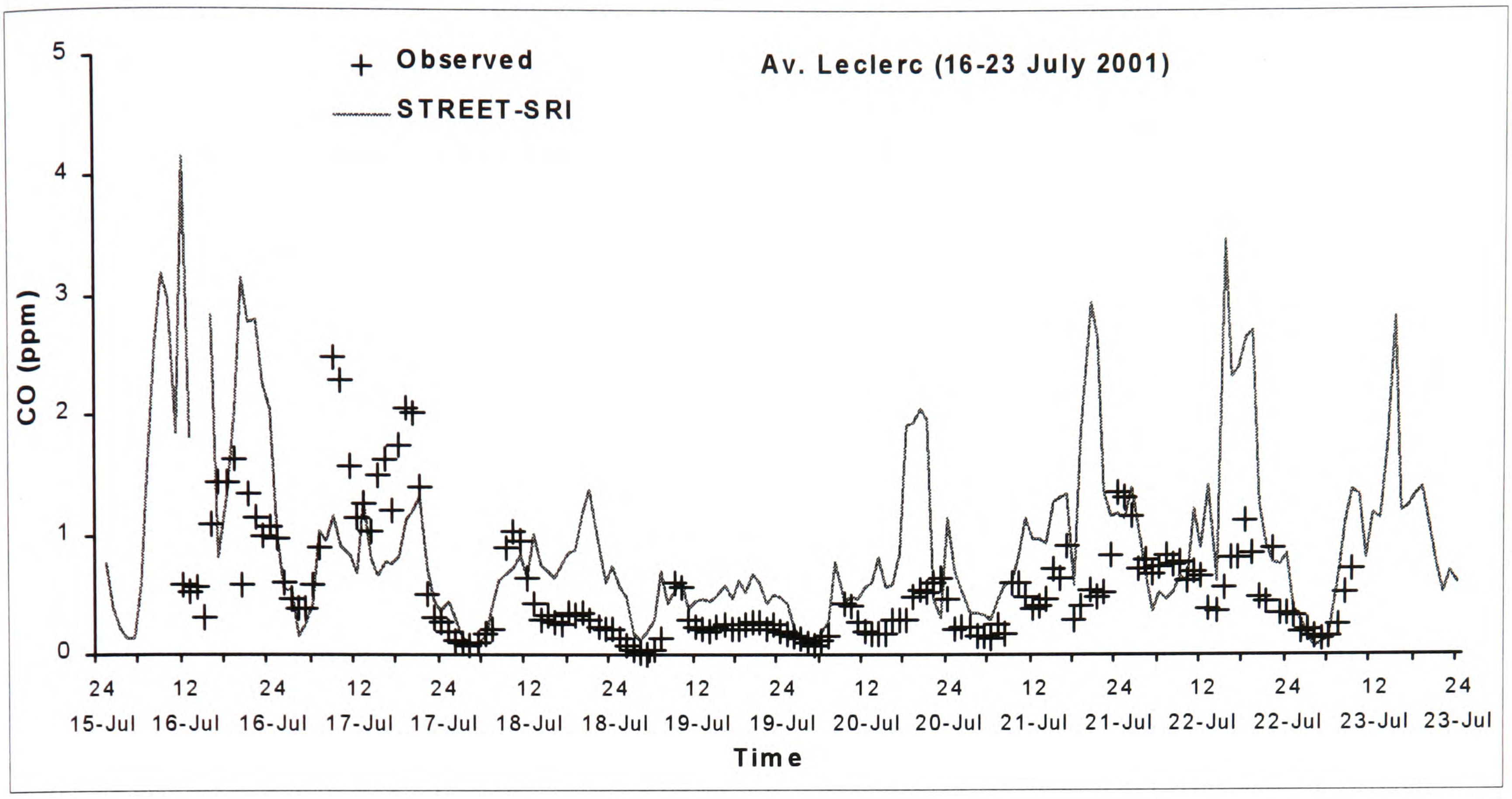


Fig. 4.15: Hourly mean CO (ppm) concentrations observed in Av. Leclerc (mobile unit) and STREET-SRI predictions.

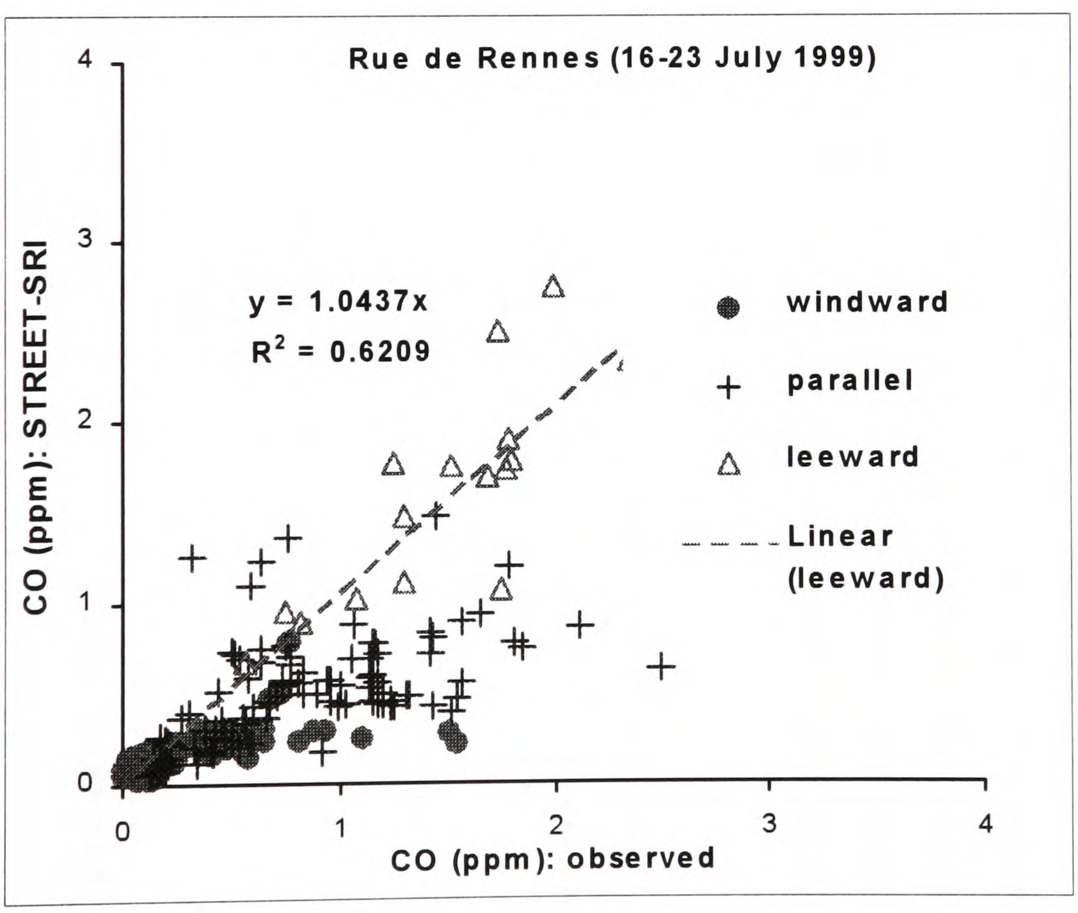
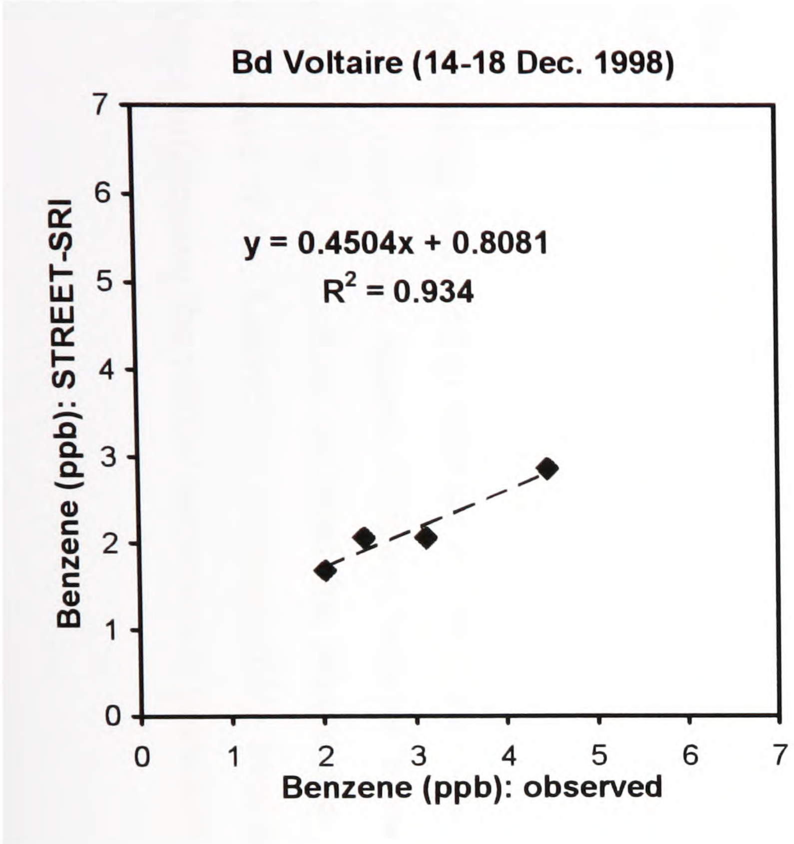
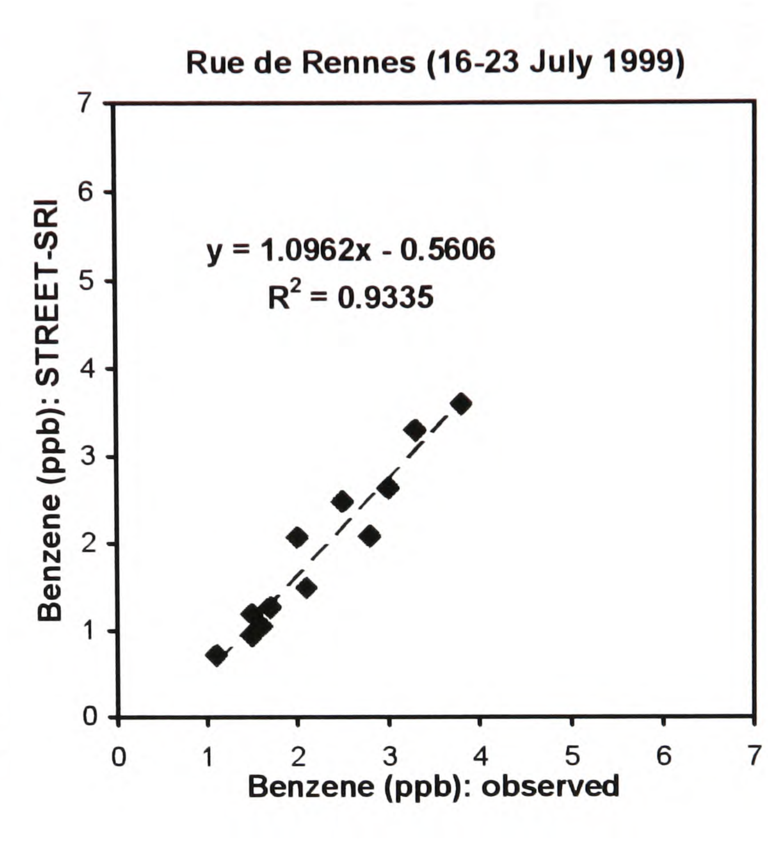


Fig. 4.16: STREET-SRI predictions vs. hourly mean CO (ppm) concentrations observed in Rue de Rennes for three different wind regimes.





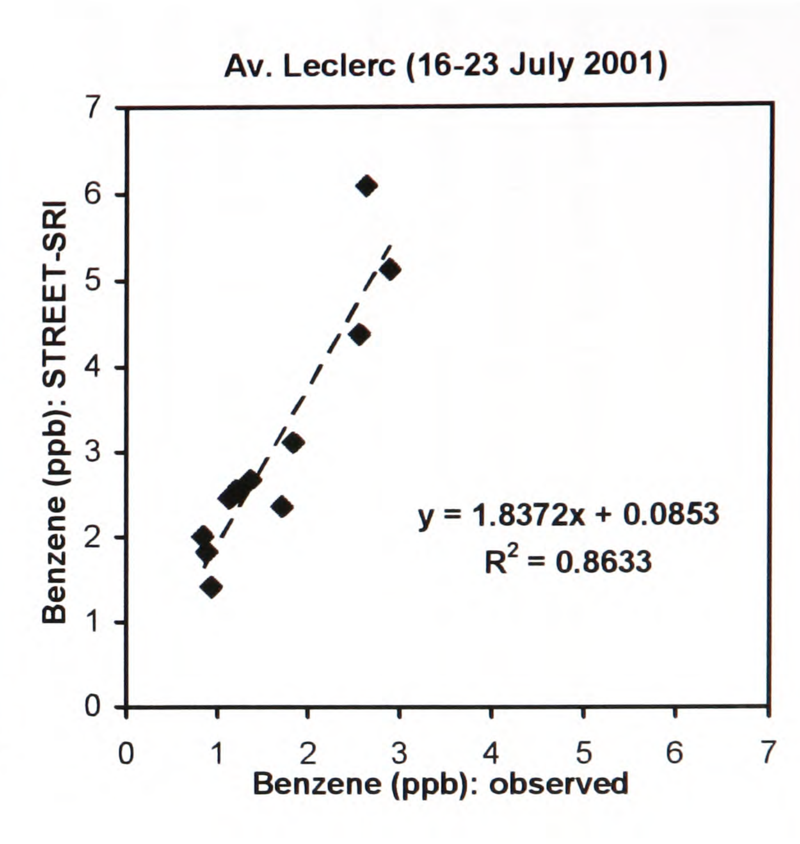


Fig. 4.17: STREET-SRI predictions vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Bd. Voltaire (left).

Fig. 4.18: STREET-SRI predictions vs. 2-5 days mean benzene (ppb) concentrations obtained with passive sampling in Rue de Rennes (centre).

Fig. 4.19: STREET-SRI predictions vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Av. Leclerc (within the asymmetric canyon segment) (right).

4.6.2. OSPM

Using the same input data sets, OSPM reproduced quite successfully the hourly CO pattern at street level in Bd. Voltaire (Fig. 4.20) and Rue de Rennes (Fig. 4.21), despite some significant under-predictions in the case of Bd. Voltaire. Again, large over-predictions were identified when model results were compared to the CO values measured in Av. Leclerc (Fig. 4.22).

Comparing separately the model output for different wind regimes (Fig. 4.23), a good linear agreement ($r = 0.79$) was established between predicted and measured values on the leeward side of the road. Large scatter and under-predictions were observed for parallel and windward flow, respectively.

Expression (4.21) was used to calculate CO concentrations at receptor heights corresponding to the different passive sampling locations in Bd. Voltaire and Rue de Rennes, using the street level output of OSPM as reference value. Applying relationship (3.4), average benzene values were obtained and added to the observed background concentrations. The comparison of the total calculated values with the concentrations observed in the field showed that the simple exponential expression ($q = 1$ and $B = 1$) gave the best agreement between predictions and measurements. The application of equation (4.20) for the calculation of B as a function of ϕ did not significantly improve predictions. Thus, the following simple exponential expression is proposed for assessing vertical concentration profiles in street canyons:

$$C(z) \approx C_r \exp\left(-\frac{z - z_r}{H}\right) \qquad (4.28)$$

Despite some under-predictions in the case of Bd. Voltaire, expression (4.28) reproduced successfully the benzene profiles detected in the two regular canyons of Bd. Voltaire (Fig. 4.24) and Rue de Rennes (Fig. 4.25).

The same relationship was used to estimate vertical concentration gradients within the asymmetric canyon of Av. Leclerc. The comparison against diffusive benzene measurements showed relatively large scatter and over-predictions (Fig. 4.26), which indicated that relationship (4.28) might not be applicable to non-regular canyons. In the case of Av. Leclerc, the presence of big trees on both sides of the canyon may have been an additional factor influencing the vertical dispersion of pollutants in the street.

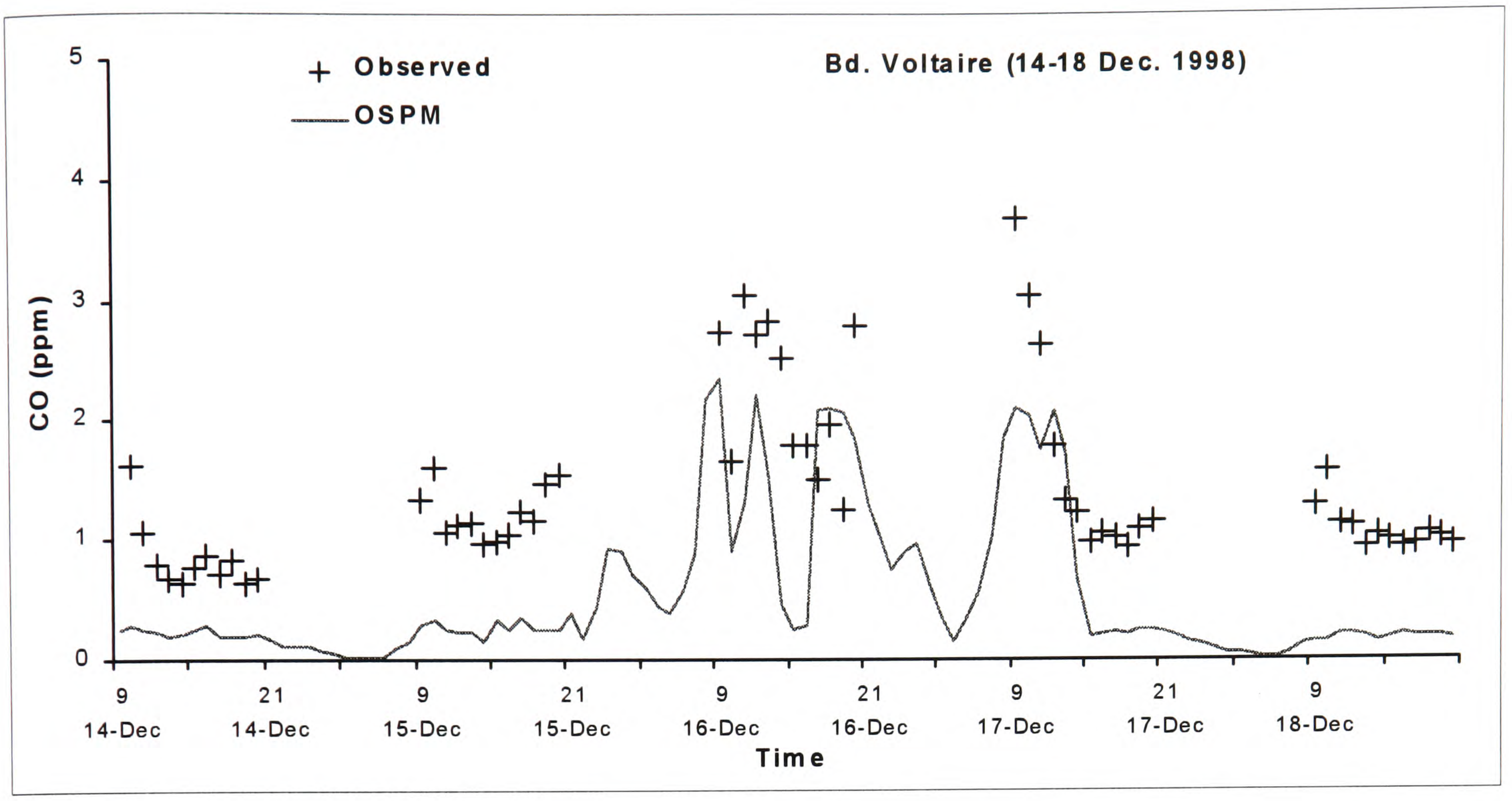


Fig. 4.20: Hourly mean CO (ppm) concentrations observed in Bd. Voltaire and OSPM predictions.

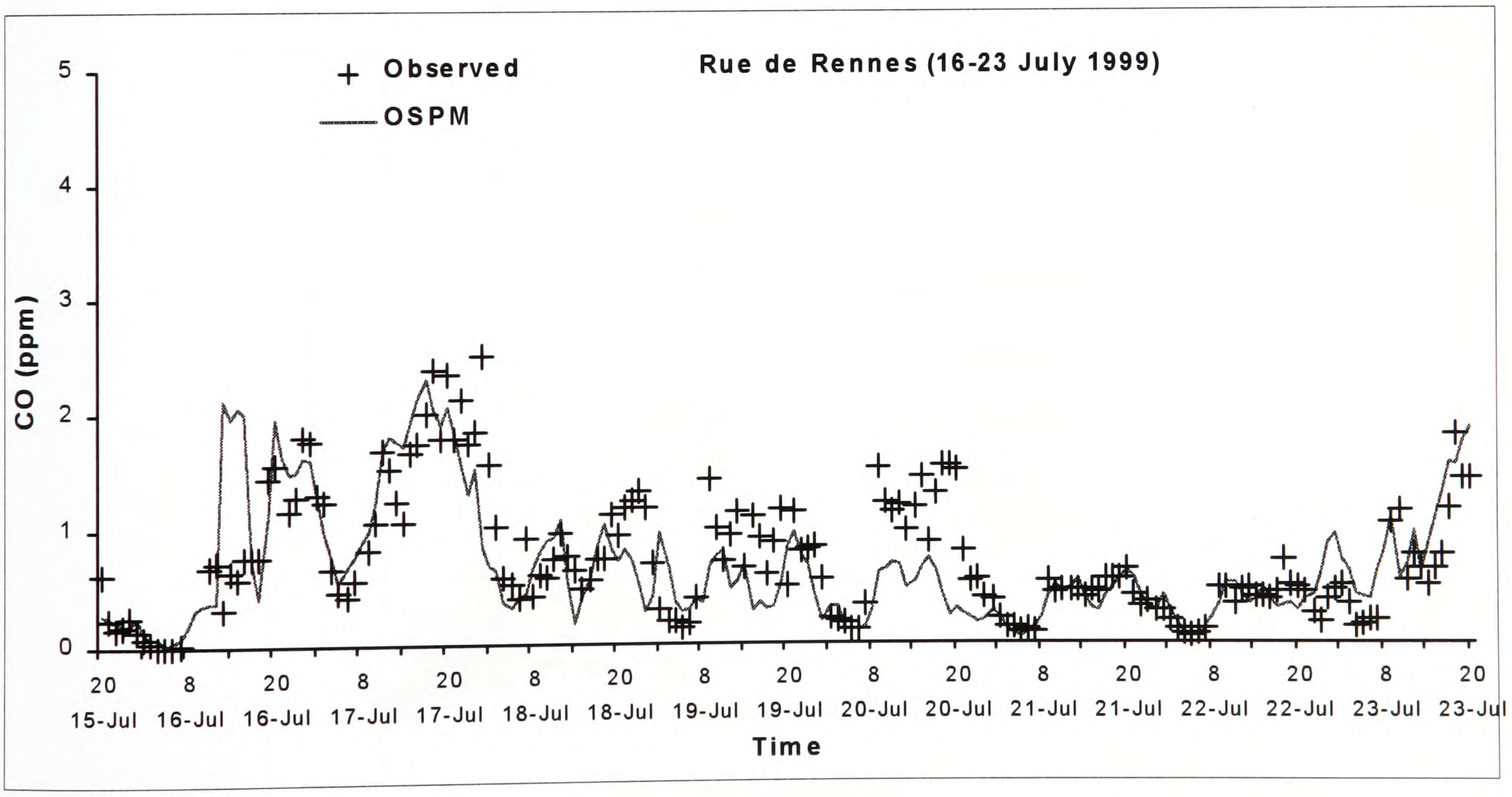


Fig. 4.21: Hourly mean CO (ppm) concentrations observed in Rue de Rennes and OSPM predictions.

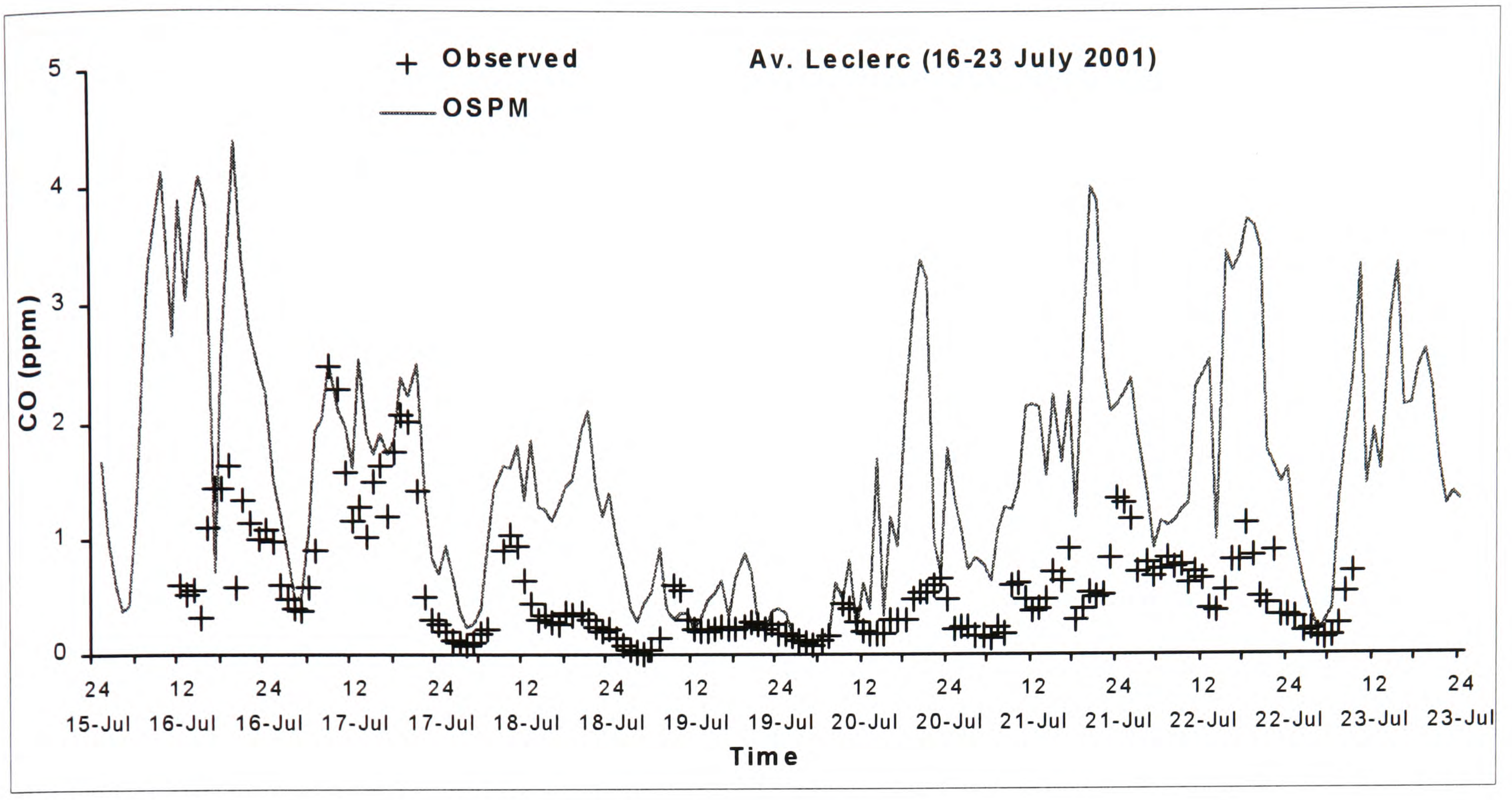


Fig. 4.22: Hourly mean CO (ppm) concentrations observed in Av. Leclerc (mobile unit) and OSPM predictions.

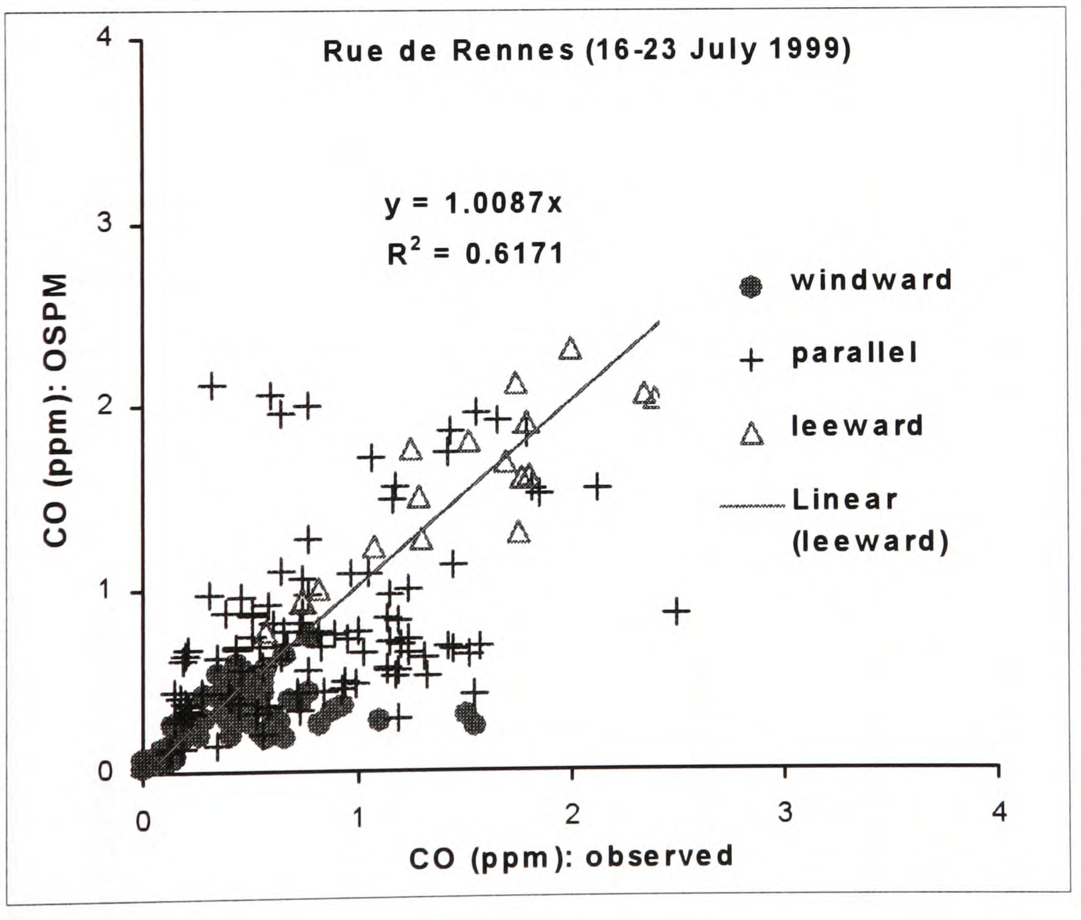


Fig. 4.23: OSPM predictions vs. hourly mean CO (ppm) concentrations observed in Rue de Rennes for three different wind regimes.

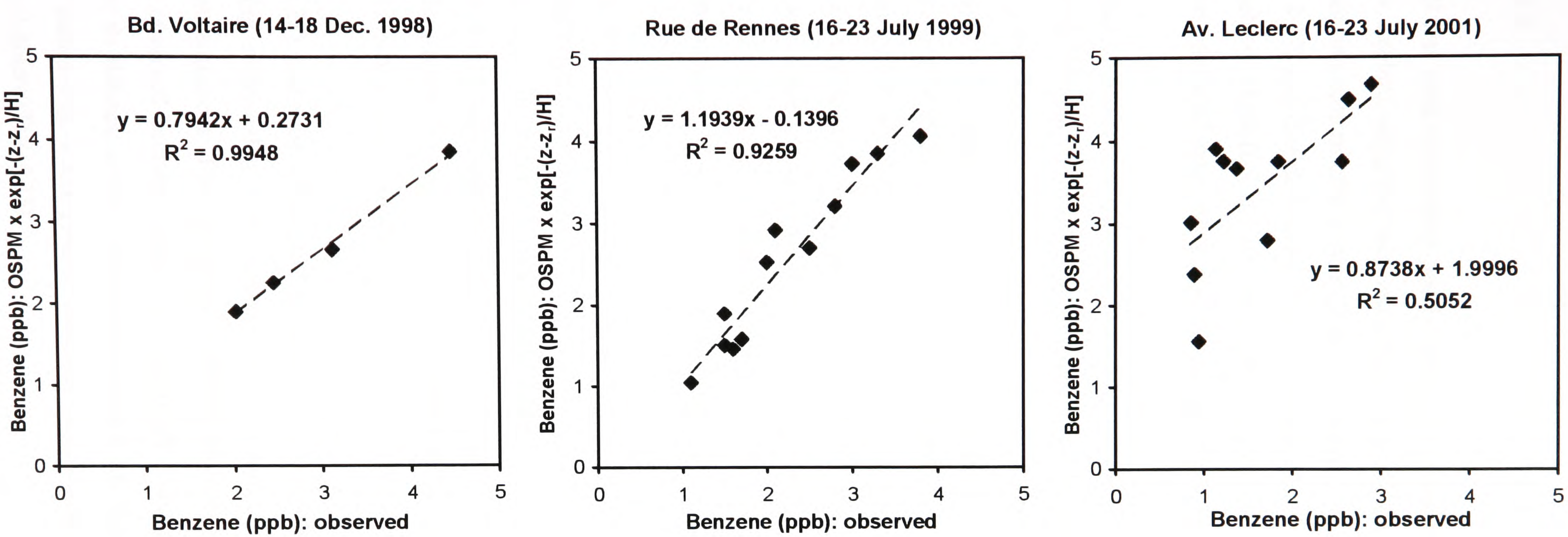


Fig. 4.24: OSPM predictions (in association with relationship 4.28) vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Bd. Voltaire (left).

Fig. 4.25: OSPM predictions (in association with relationship 4.28) vs. 2-5 days mean benzene (ppb) concentrations obtained with passive sampling in Rue de Rennes (centre).

Fig. 4.26: OSPM predictions (in association with relationship 4.28) vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Av. Leclerc (within the asymmetric canyon segment) (right).

4.6.3. AEOLIUS

AEOLIUS Full was used to calculate hourly mean CO concentrations at street level in Bd. Voltaire, Rue de Rennes and Av. Leclerc, using the same input data sets as for the previous simulations. It can be observed that the diurnal variation pattern of CO was clearly reproduced by the model throughout the campaigns in Bd. Voltaire (Fig. 4.27) and Rue de Rennes (Fig. 4.28), and roughly reproduced in the case of Av. Leclerc (Fig. 4.29). Nevertheless, AEOLIUS significantly under-predicted the CO concentrations observed in Bd. Voltaire and greatly over-predicted those measured in Av. Leclerc (especially the higher daytime CO values).

The performance of AEOLIUS was evaluated under different dispersion conditions by sorting the available data from Rue de Rennes into different wind speed and direction classes. Hourly measured versus calculated concentrations were separately plotted for the following situations (Fig. 4.30): (a) All wind directions sorted into two different wind speed classes (synoptic wind speed above or below 2.5 m/s); (b) southerly winds parallel or near-parallel to the street, sorted into two different wind speed classes (synoptic wind speed above or below 2.5 m/s); (c) northerly winds parallel or near-parallel to the street, sorted into two different wind speed classes (synoptic wind speed above or below 2.5 m/s); (d) westerly winds perpendicular or near-perpendicular to the street, sorted into two different wind speed classes (synoptic wind speed above or below 2.5 m/s); (e) easterly winds perpendicular or near-perpendicular to the street, sorted into two different wind speed classes (synoptic wind speed above or below 2.5 m/s).

Comparing the correlation coefficients and regression lines obtained for the different regimes (Fig. 4.30), it can be concluded that the performance of the model was enhanced for perpendicular winds. The best model predictions corresponded to easterly winds (Fig. 4.30e), i.e. when the receptor was on the leeward side of the canyon. For westerly perpendicular winds (Fig. 4.30d), i.e. when the receptor was windward, the model seemed to under-predict the observed concentrations. Finally, the variability in correlation, which was clearly wind direction dependent, did not appear sensitive to changes in wind speed.

Expression (4.28) was used to estimate vertical CO concentration gradients within the three canyons of the study. Applying relationship (3.4), average benzene values were obtained and added to the observed background concentrations. The comparison against measurements revealed a good linear correlation between predicted and observed benzene values, but significant under-predictions in the case of Bd. Voltaire (Fig. 4.31). For Rue de Rennes, a very good general agreement was established between benzene measurements and predicted values (Fig. 4.32). Finally, relatively large scatter and over-predictions were again observed in the case of Av. Leclerc (Fig. 4.33).

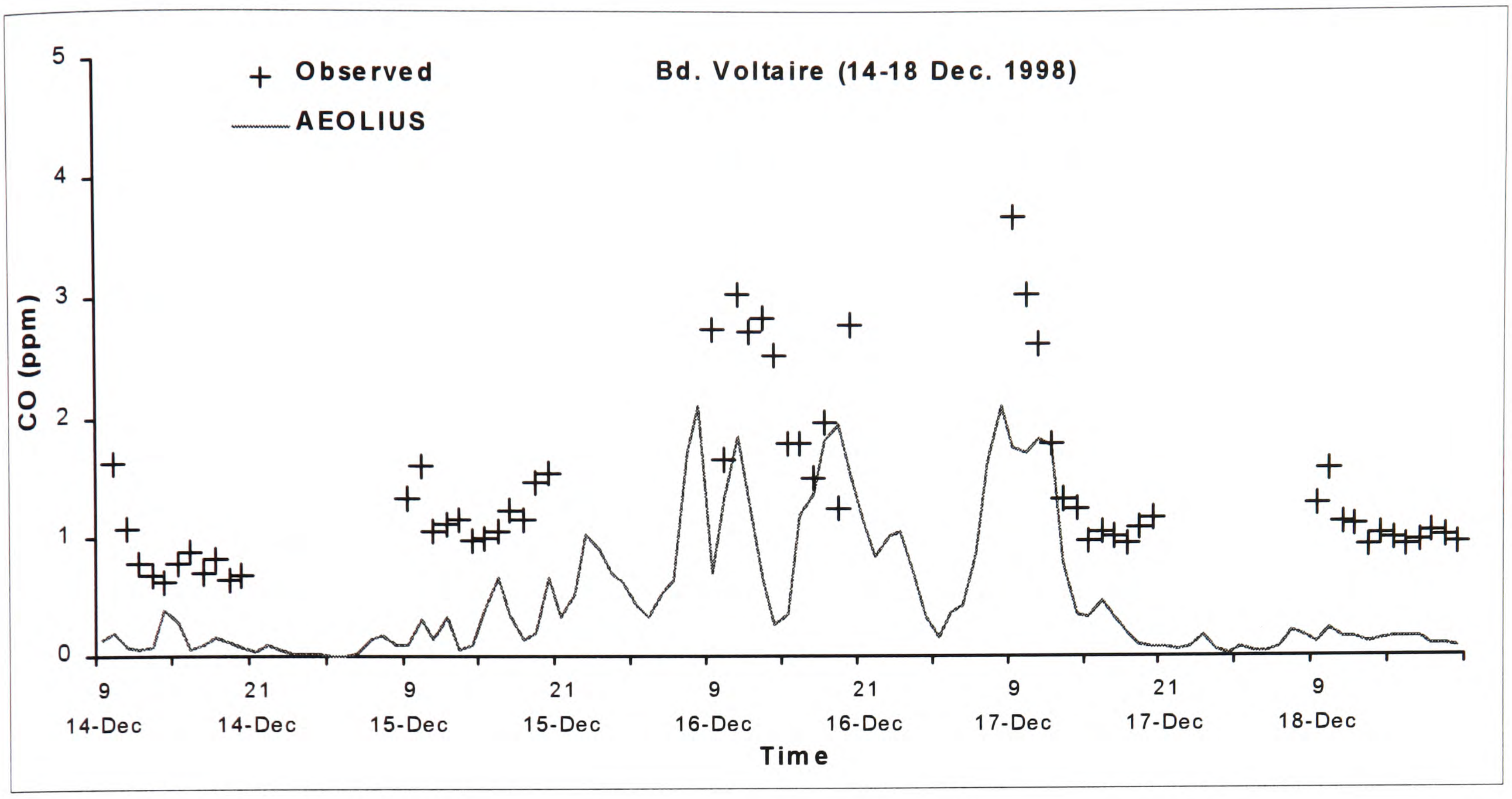


Fig. 4.27: Hourly mean CO (ppm) concentrations observed in Bd. Voltaire and AEOLIUS (Full) predictions.

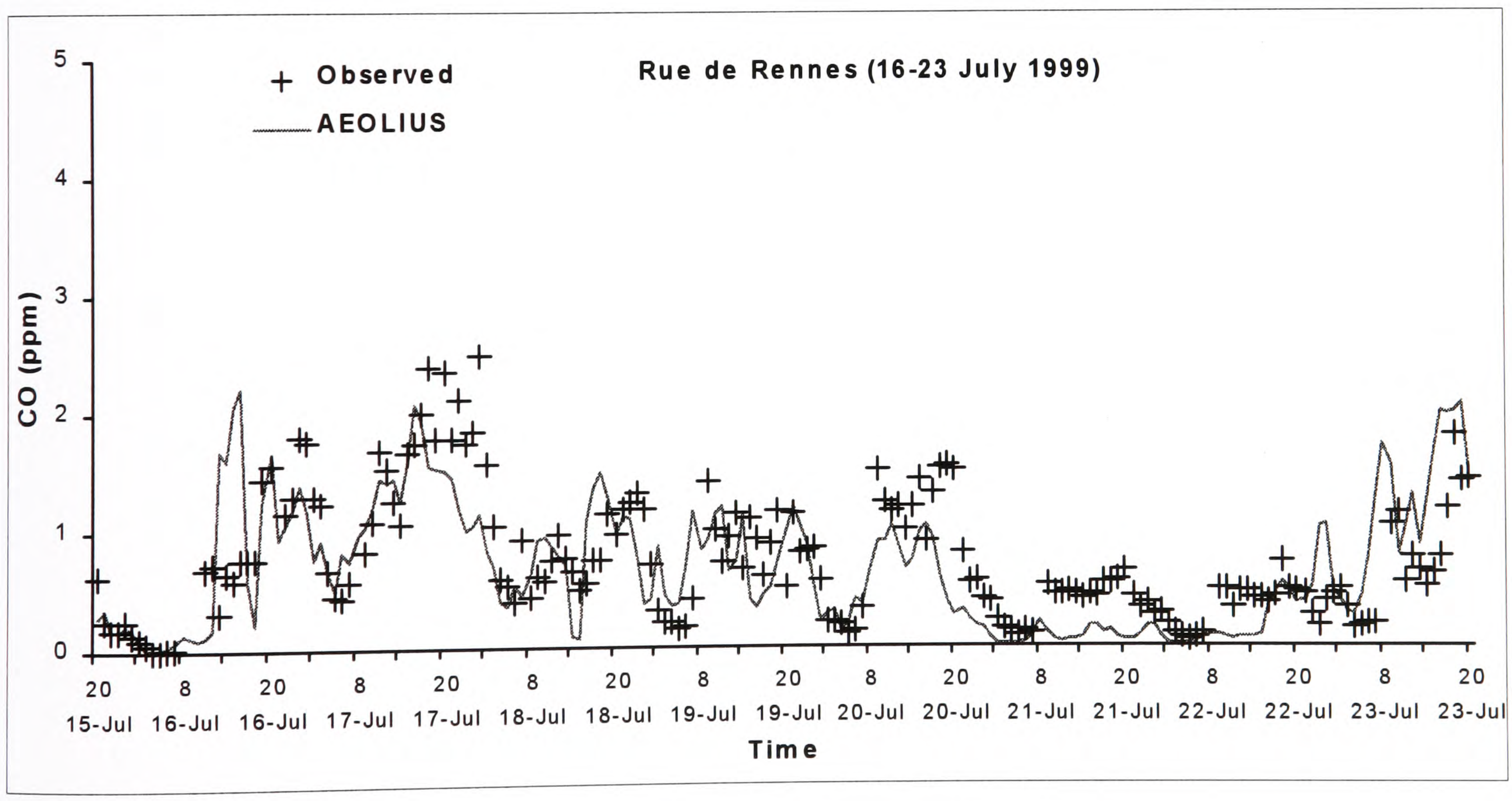


Fig. 4.28: Hourly mean CO (ppm) concentrations observed in Rue de Rennes and AEOLIUS (Full) predictions.

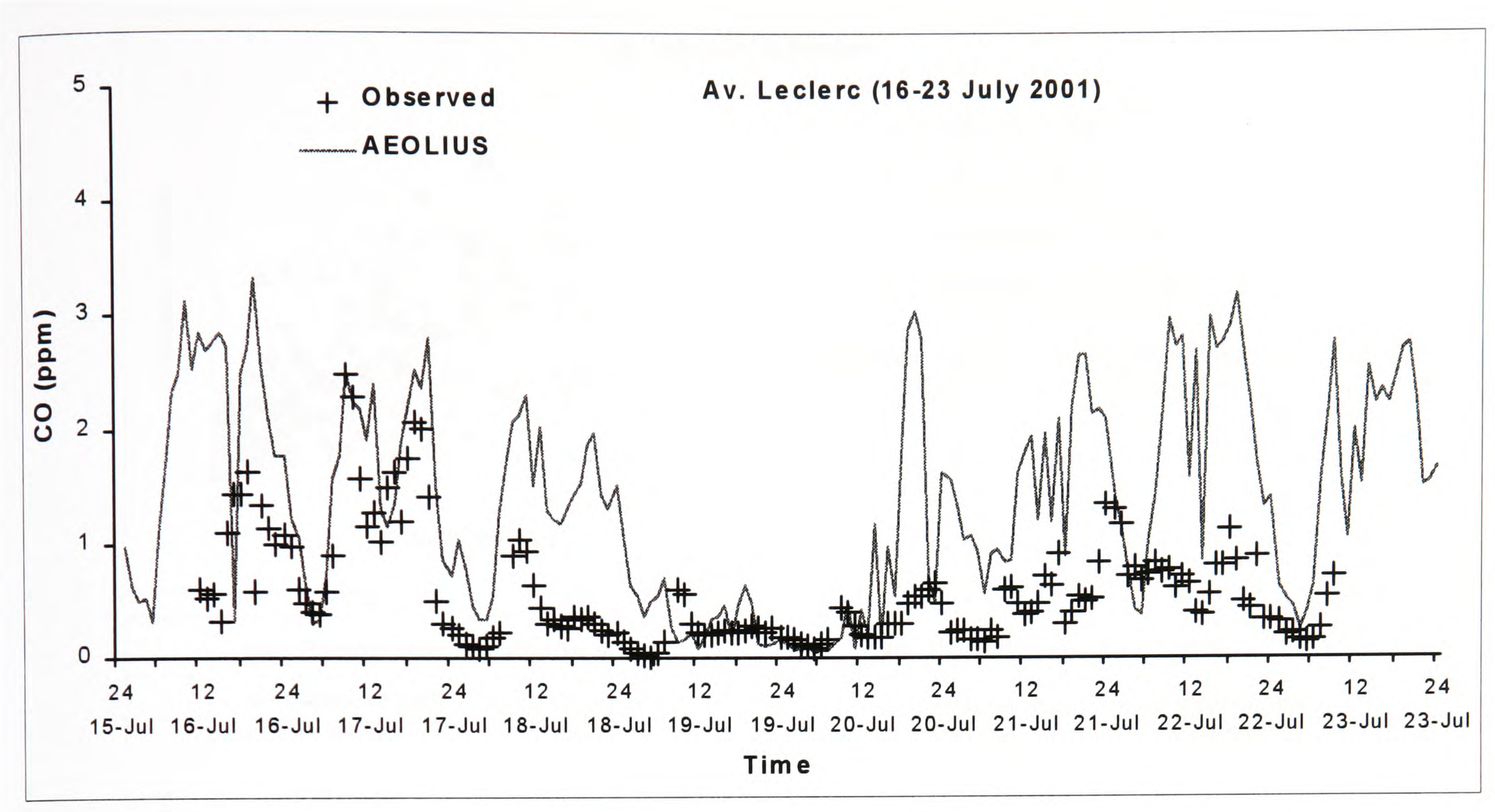


Fig. 4.29: Hourly mean CO (ppm) concentrations observed in Av. Leclerc and AEOLIUS (Full) predictions.

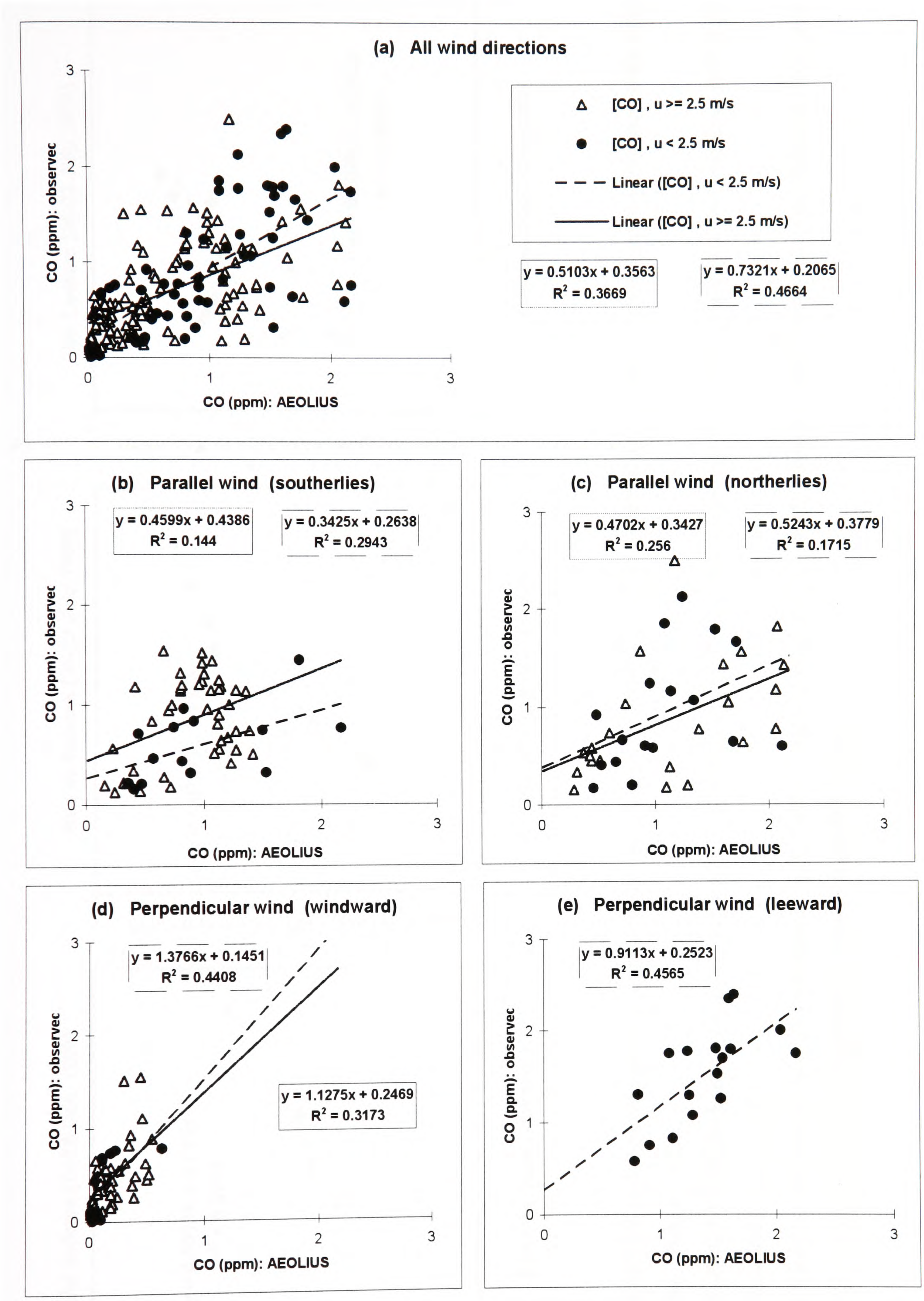
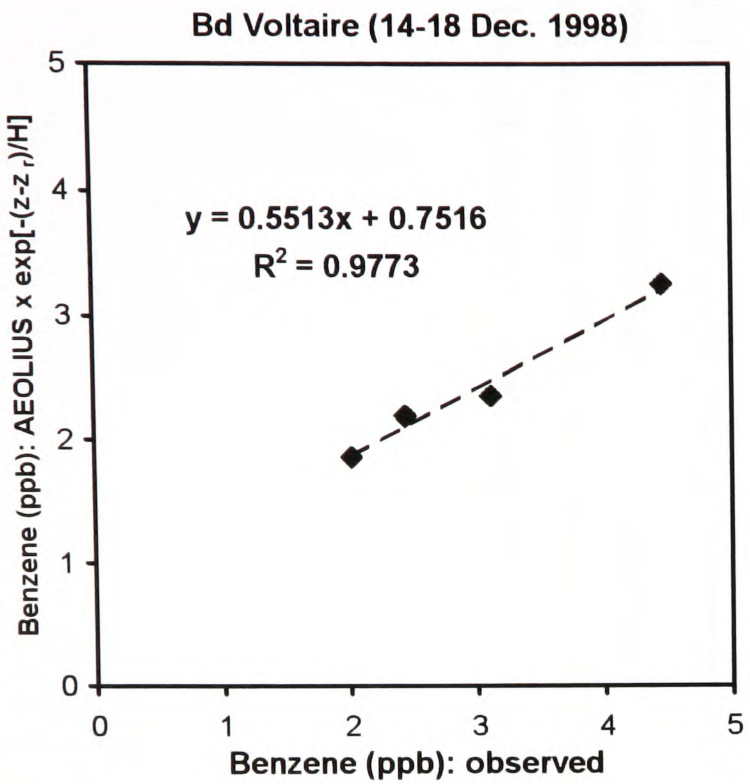
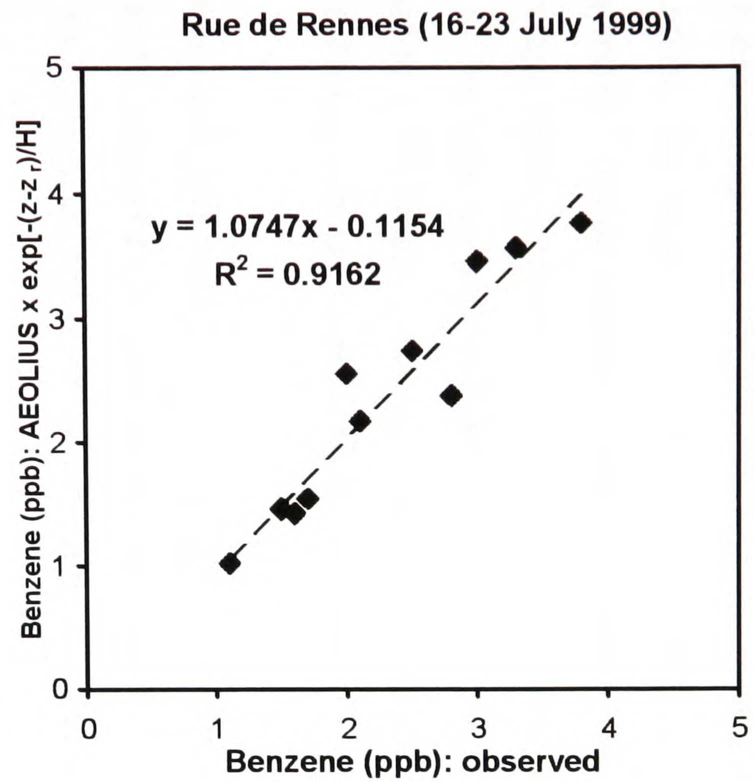


Fig. 4.30: AEOLIUS (Full) performance for different wind regimes using CO (ppm) concentrations from Rue de Rennes.





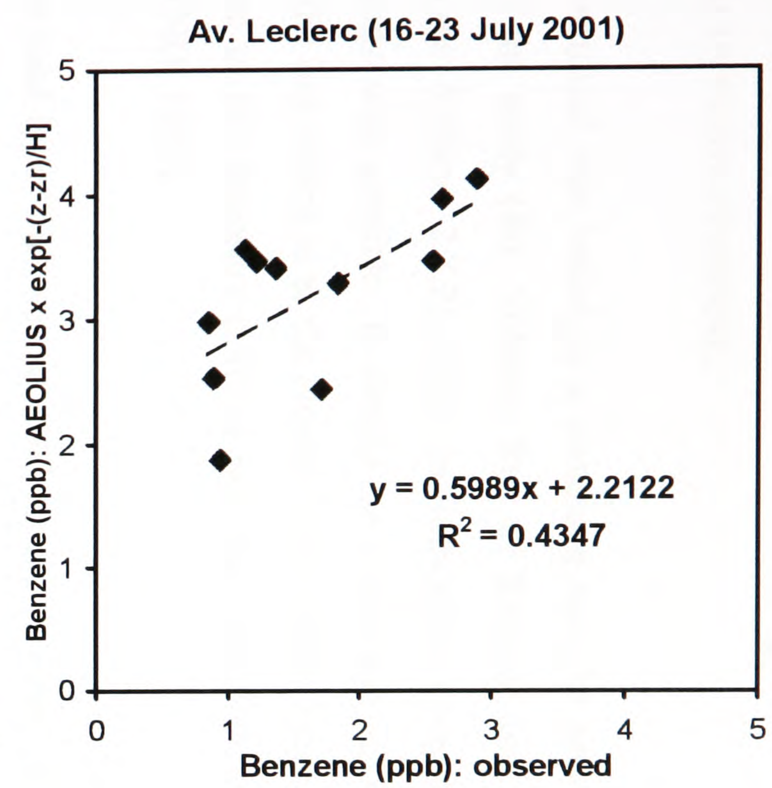


Fig. 4.31: AEOLIUS (Full) predictions (in association with relationship 4.28) vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Bd. Voltaire (left).

Fig. 4.32: AEOLIUS (Full) predictions (in association with relationship 4.28) vs. 2-5 days mean benzene (ppb) concentrations obtained with passive sampling in Rue de Rennes (right).

Fig. 4.33: AEOLIUS (Full) predictions (in association with relationship 4.28) vs. weekly mean benzene (ppb) concentrations obtained with passive sampling in Av. Leclerc (within the asymmetric canyon segment) (right).

4.6.4. CAR INTERNATIONAL

CAR International was tested as a screening tool for assessing air pollution levels within the three street canyons of the study (Bd. Voltaire, Rue de Rennes, and Av. Leclerc). Since this model provided yearly averages (see Section 2.3.2), only an indicative comparison with the available shorter-term kerbside measurements was possible. It should be noted that in CAR International, the kerbside receptor is by definition the one which is at the closest to the traffic location usually occupied by pedestrians. Therefore, if the distance to the exposure point is not equal for both sides of the road, the shorter distance should be selected (TNO, 1995).

The canyon road type "3b" was selected for Bd. Voltaire and Rue de Rennes, which means that the buildings flanking the streets had to be at a distance from the road axis of less than 1.5 times the height of the canyons. For Av. Leclerc, the larger street canyon type "3a" was selected. With respect to the vehicle speed, the "stagnating traffic" option (average speed: 13 km/h) was chosen for Av. Leclerc, while the "normal city traffic" (average speed: 19 km/h) was consider to be more representative for Bd. Voltaire and Rue de Rennes. The effect of the trees on pollutant dispersion within Av. Leclerc was taken into account by assigning "tree factor" equal to 1.25. Finally, ten-meter yearly average wind speeds from Orly Airport were used in all cases in order to calculate average roadside CO concentrations.

Yearly benzene averages were then calculated from the corresponding CO predictions using relationship (3.4). Since the model predicted pollutant concentrations at 1.5 m height above the ground (den Boeft et al., 1996), expression (4.28) was applied to obtain estimates at higher sampling locations.

CAR International seemed to over-predict benzene concentrations in all selected street (Table 4.2), although a straightforward comparison might not be applicable due to the pronounced small-scale spatial variability of traffic-related air pollution within the canyons. This model was unable to reproduce the strong crossroad gradients observed, because it did not take into account the prevailing regional wind direction.

Table 4.2: Benzene (ppb) predictions using CAR International in association with expression (3.4) for Bd. Voltaire, Rue de Rennes and Av. Leclerc, and comparison with roadside measurements (on the west and east side of these streets).

Year	Street	Vehicle volume	Distance receptor m	Height receptor m	Road type	Speed type	Tree factor	Wind speed m/s	CO emission factor g/km veh	CAR_Int.* benzene ppb	Obs. West benzene ** ppb	Obs. East benzene ** ppb
1998	Voltaire	30390	14	4.5	3b	c	1	4.08	7.23	3.1	4.5	2.5
1998	Voltaire	30390	14	4.5	3b	c	1	4.08	20.29	6.4	4.5	2.5
1999	Rennes	22371	8	1.5	3b	c	1	4.16	8.73	3.5	2.8	1.7
1999	Rennes	22371	10	4.4	3b	c	1	4.16	8.73	2.6	2.1	1.6
1999	Rennes	22371	8	1.5	3b	c	1	4.16	10.91	4.3	2.8	1.7
1999	Rennes	22371	10	4.4	3b	c	1	4.16	10.91	3.2	2.1	1.6
2001	Leclerc	66343	10	2.6	3a	d	1.25	4.24	12.64	7.9	2.8	
2001	Leclerc	66343	20	2.6	3a	d	1.25	4.24	12.64	4.4		1.8

* In association with expression (3.4)

** Bd. Voltaire: five-day averages

Rue de Rennes: five-day averages

Av. Leclerc: six-month averages

4.6.5. PHOENICS

PHOENICS is a general purpose CFD code able to simulate complex air flows and concentration fields within cavities, around obstacles, etc. (see Section 2.3.4). In this study, only a small number of model runs were carried out in order to test the applicability of a CFD code in pollutant dispersion within street canyons, and identify possible advantages and drawbacks. The regular canyon of Bd. Voltaire was selected for this exercise.

The dispersion calculations were restricted to the area between the buildings flanking the street. In that case, the total width of the canyon (30 m) was divided into three parts in order to separate the roadway from the pavements. Bd. Voltaire was simulated as a 50 m long linear source, with the buildings lining up uniformly on both sides. The discretisation of the model domain (50 m $\times$ 50 m $\times$ 40 m) was not uniform. Higher grid density applied to the areas near the ground and the canyon walls. Furthermore, the relief of the buildings and the street surface was taken into account by assigning a roughness coefficient for each physical limit of the domain.

Local street-level wind measurements and synoptic wind data from Montsouris station were used to calibrate and then run the model. Since there was no information about the shape of the wind field above the urban canopy available, it was assumed that the vertical profile of the wind entering into the model domain was uniform. Finally, the heat exchange between the air masses and the physical limits of the domain was neglected, because of the limited insolation during the campaign period (December 1998).

From the detailed analysis of the synoptic wind data, four representative meteorological situations were identified: (a) Moderate SW winds (2 - 3.5 m/s), corresponding mainly to the first two days of the campaign, (b) weak southerly winds (< 2 m/s), corresponding to the third day of the campaign, (c) relatively strong SW winds (> 3.5 m/s), prevailing during the fifth day of the campaign, and (d) moderate SE winds (2 - 3.5 m/s), thus parallel to the street axis, during the morning hours of the fourth day of the campaign.

CO emissions were calculated according to Buckland and Middleton's (1999) methodology and were then uniformly distributed throughout the roadway. In order to take into account the fast initial dispersion of exhaust gases due to the mechanical and thermal turbulence induced by moving vehicles, an effective release height of 1.6 m was introduced in the calculations.

The urban background concentration was estimated from diffusive benzene measurements carried out in a green space in the vicinity of Bd. Voltaire. The background CO contribution (calculated using expression 3.4) was then injected into the model domain at roof-top level.

For the simulation of the turbulent flow, a traditional k-ε model was applied. In order to reduce the computational time, a simplifying two-dimensional approximation was made, according to which the synoptic wind was modelled to be normal to the street axis. All calculations were steady state.

Indicative wind and concentration fields were produced for synoptic wind speeds of 2.5 m/s, 4 m/s and 2 m/s (Fig. 4.34 and Appendix III). Selected CO values were compared with relevant field measurements from Bd. Voltaire. As it can be seen in Table 4.3, the orders of magnitude were respected in all cases.

Table 4.3: CO (ppm) predictions using PHOENICS compared to roadside observations in Bd. Voltaire for three different synoptic wind speeds.

Case	Wind speed m/s	PHOENICS CO ppm	Observed CO ppm
1st	2.5	2.05	1.60
2nd	4.0	1.10	0.97
3rd	2.0	2.60	1.46

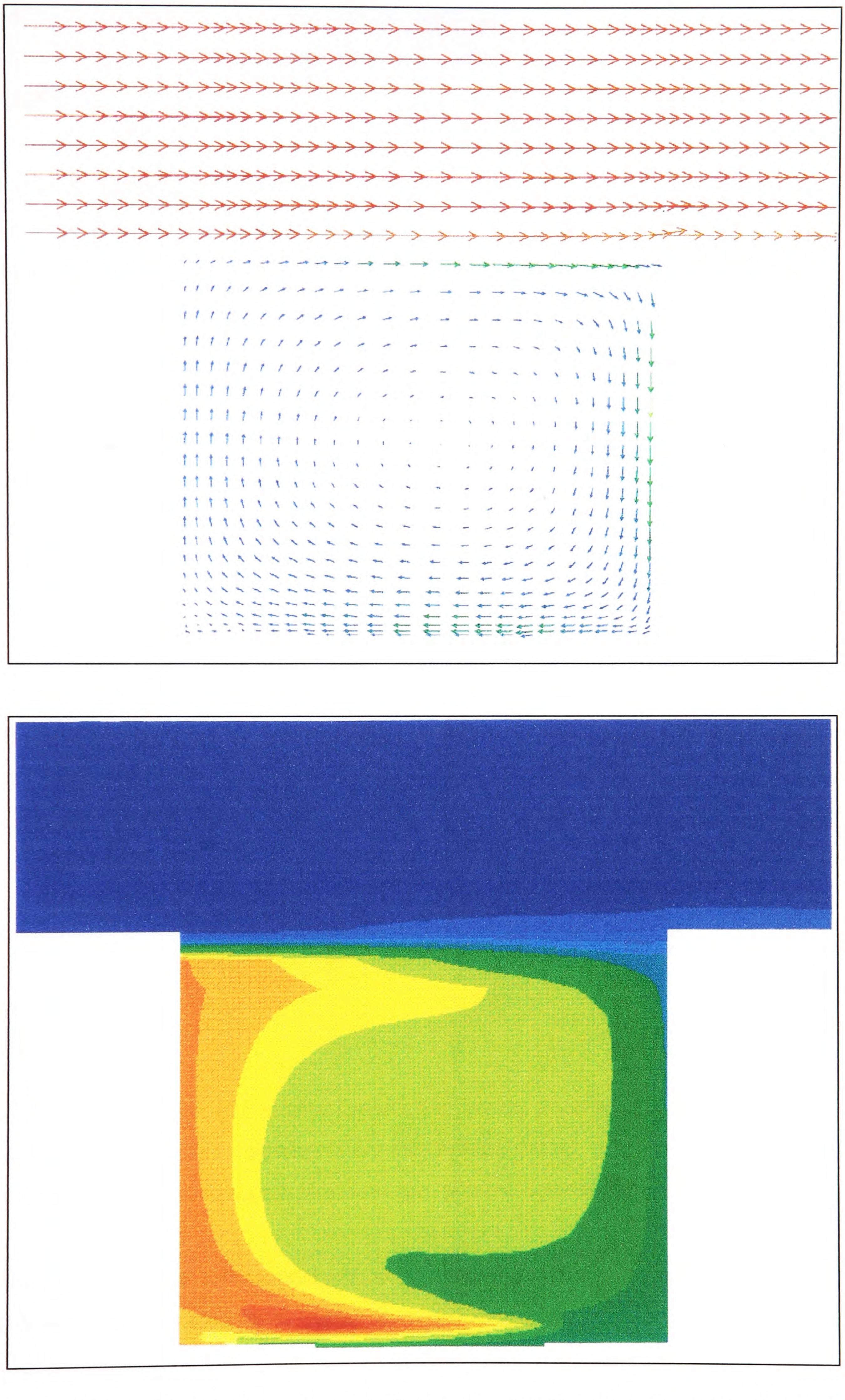


Fig. 4.34: Wind field (top) and CO concentration field (bottom) in Bd. Voltaire calculated with PHOENICS (synoptic wind speed entering the model domain: 2 m/s).

4.6.6. CALINE4

Motorway (RN10)

CALINE4 was used to estimate the likely contribution from RN10 motorway to the BTX concentrations occurring at the level II sampling locations of the Stage 2 implemented petrol station in the south of Rambouillet (see Section 3.4).

On a flat terrain, CALINE4 is able to calculate pollutant concentrations for multiple receptors at distances up to 500 m from the source, if the necessary input information is available (i.e. emission factors, site topography, meteorological and traffic data). In the case of RN10, the local wind speed average (2.5 m/s) and ambient temperature (7°C) were used, while the most unfavourable wind direction was selected for each receptor (i.e. worst case mode). The input CO emission factors were calculated using the site-specific vehicle fleet composition (Buckland and Middleton, 1999). Applying empirical relationship (3.4), benzene concentrations at 12 receptor locations surrounding the petrol station were finally predicted (Table 4.4).

As expected, N-NE and S-SE wind directions represented the worst meteorological scenarios in terms of pollutant dispersion at the selected receptor locations. Under these conditions, winds blowing almost parallel to the motorway accumulated the pollutants emitted along the upwind segments of this linear source. Comparing the benzene values of Table 4.4 and Fig. 3.22, it might be concluded that the motorway contribution is stronger than the petrol station contribution to the total benzene levels occurring at the level II receptor locations under worst case wind conditions.

Street canyon (Rue de Rennes)

Although CALINE4 is applicable to street canyons and intersections, it has been used in relatively few studies assessing pollutant dispersion within confined urban microenvironments. For this reason, it was decided to test this model using one of the available street canyon data sets (i.e. Rue de Rennes).

In such a case, the main limitation of the model is that its canyon ("depressed section") mode can only be used for synoptic winds parallel to the street axis. During the monitoring campaign in Rue de Rennes, a continuous fifty-two hour time period of parallel or near-parallel winds (i.e. SW) was identified. For this period, an input data set including CO emission factors (Buckland and Middleton, 1999), canyon dimensions, traffic data, meteorological information including atmospheric stability class and mixing height (Benson, 1984), and urban background contributions was created.

Hourly mean CO concentrations were calculated at a roadside receptor corresponding to the continuous monitoring location on the east side of Rue de Rennes and compared with the measurements from the mobile unit. As it can be observed in Fig. 4.35, the diurnal variation pattern of CO was well reproduced by CALINE4 for this time period of prevailing parallel winds.

Table 4.4: Road traffic contribution to the CO and benzene levels in the surroundings of the RN10 petrol station (under worst case wind conditions) calculated using CALINE4 and expression (3.4).

Site N°	Wind direction * deg	CALINE4 CO ppm	CALINE4 ** Benzene µg/m³	CALINE4 ** Benzene ppb
1	169	0.4	4.8	1.48
2	12	0.4	4.8	1.48
3	12	0.4	4.8	1.48
4	11	0.4	4.8	1.48
5	11	0.4	4.8	1.48
9	19	0.2	2.4	0.74
10	161	0.2	2.4	0.74
11	161	0.2	2.4	0.74
12	161	0.2	2.4	0.74

* Worst case ** In association with expression (3.4)

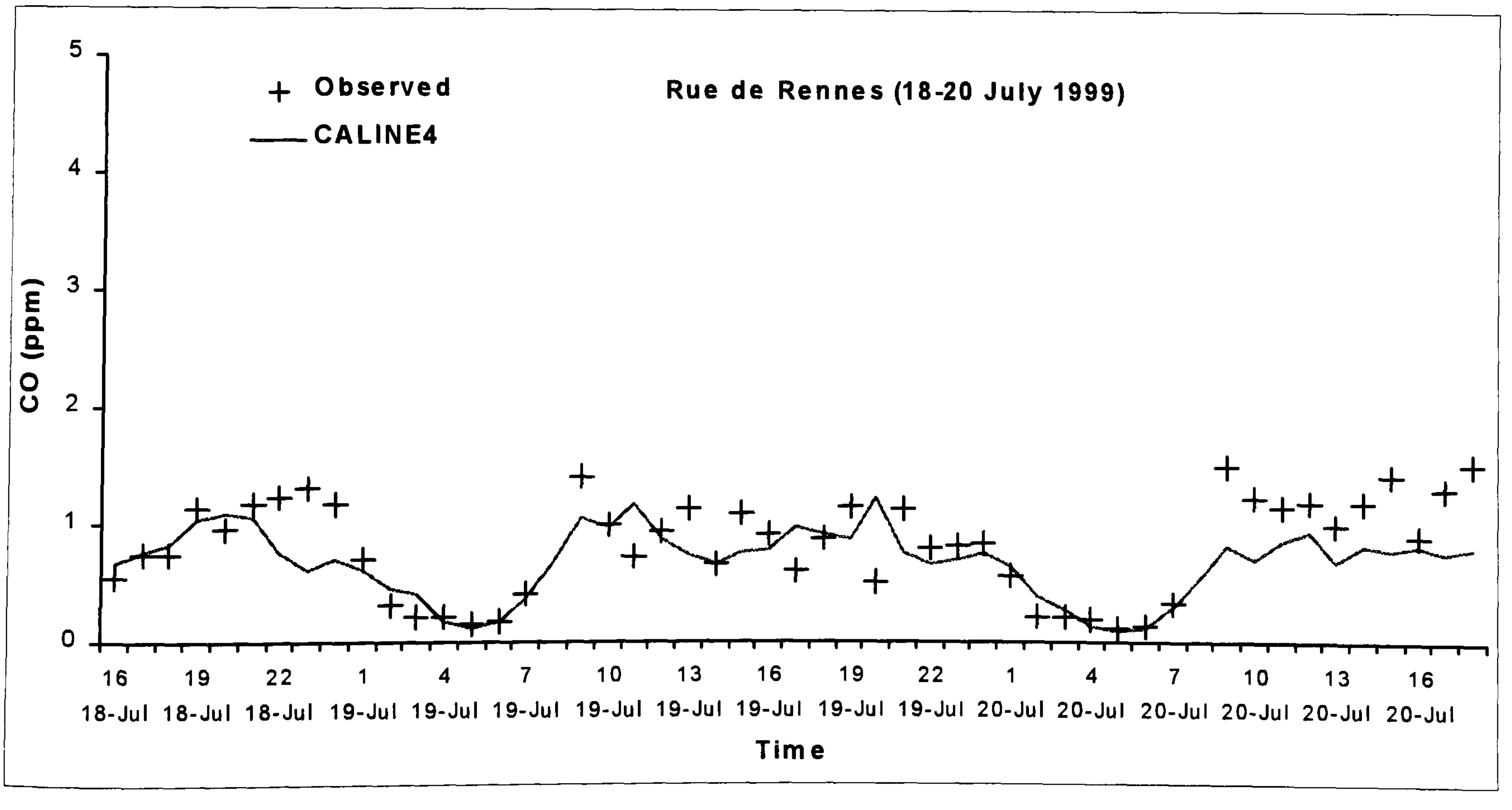


Fig. 4.35: Hourly mean CO (ppm) measurements and CALINE4 predictions during a two-day period of parallel and near-parallel winds in Rue de Rennes.

4.7. Model uncertainty

Most of the air quality modelling work has been so far based on the "deterministic" approach of using only one dispersion model for a specific application. The selected model provides estimates of average concentrations using a specific meteorological and emission data set. A serious weakness of this method lies on the fact that many uncertainties, not only related to the calculations and input variables, but also to the very nature of atmospheric processes, are ignored. That might have serious implications for exposure studies, since the area and number of people exposed to a predicted pollution level may be very sensitive to the uncertainties associated with this prediction (Fisher and Ireland, 2001).

The total uncertainty involved in modelling simulations can be considered as the sum of three components (Hanna, 1988): (a) The uncertainty due to errors in the model physics, (b) the uncertainty due to input data errors, (c) the uncertainty due to stochastic processes (e.g. turbulence) in the atmosphere. It may be possible to reduce the first component of model uncertainty by introducing more physically realistic and computationally efficient algorithms. It may also be possible to eliminate some of the effects of input data errors once more accurate monitoring instruments can be set up at representative locations. However, the stochastic fluctuations are a natural characteristic of the atmosphere that cannot be eliminated. Therefore, practical methodologies need to be developed to quantify total model uncertainty and present results in a meaningful way. If uncertainties (however large) are explicitly reflected on model results, policy makers will still make decisions, but an inappropriate level of reliance on the results will be avoided (Pielke, 1998; Dabberdt and Miller, 2000).

Uncertainty related to model physics is presumed to be much larger than the uncertainty due to input errors (Freeman et al., 1986). For this reason, research efforts have been mainly focused on devising models that will be more consistent with reality and thereby minimise model uncertainty. Less work has been done so far to incorporate input data uncertainty into model results.

The Monte-Carlo analysis is one of the commonly used methods for propagating input data uncertainties through air quality models. It has been applied to models of different levels of complexity, from Gaussian plume (Irwin et al., 1987) to complex photochemical codes (Hanna et al., 1998; 2001). This method enables an evaluation of the output of the model for many sets of combinations of the input parameters. These data sets are obtained by random sampling from the distribution assigned to each one of the uncertain input variables. Two important advantages of this statistical method are that it can be applied to a complete set of about 100 or more input parameters, and that it is widely used in the analysis of other environmental problems. On the other hand, it has the limitation that the estimates of uncertainty in the inputs are often based on informal processes (e.g. the professional judgement of the modeller), and that the cost of the method in terms of computer simulation time might be quite high, since a relatively large number (>100) of model runs is preferable (Irwin et al., 1987; Hanna et al., 1998).

Freeman et al. (1986) developed a theoretical formula for propagating input data uncertainties through dispersion models. Results obtained using this formula were in good agreement with Monte-Carlo uncertainty estimates for unstable atmospheric conditions, but large inconsistencies were observed for neutral and stable conditions. These results also showed that even small uncertainties in the inputs might cause large uncertainties in the predictions.

Dabberdt and Miller (2000) used a probabilistic method for quantifying the uncertainty related to model predictions in an accidental gas release application. An ensemble set of 162 simulations was created by specifying a best estimate together with two additional values that bounded the likely range of uncertainty in estimating four input parameters (i.e. wind speed, wind direction, source strength, and plume rise). A best estimate together with a "second choice" was specified for atmospheric stability. Finally, contour concentration patterns (both deterministic and probabilistic) and histograms of the probability of occurrence of concentrations at specific receptor locations were produced to illustrate the uncertainty in the predictions.

Another way to estimate uncertainty in model predictions is by determining the input parameters to which the model in use is most sensitive. A sensitivity analysis indicates how much of the overall uncertainty in the model predictions is associated with the individual uncertainty in each model input (McRae and Seinfeld, 1983). Sensitivity studies are not, strictly speaking, uncertainty analyses, since they do not combine the uncertainties of the model inputs to provide a realistic estimate of the uncertainty in the model output. Nevertheless, knowledge of the model's sensitivity to different variables is necessary in order to decide where emphasis should be placed in estimating total uncertainty (Hanna, 1988).

One of the objectives of the present study was to examine practical methodologies for quantifying the uncertainty in urban air quality model predictions. Two methodologies, involving the use of three semi-empirical street canyon models (STREET-SRI, OSPM and AEOLIUS), were developed and then applied to the street canyons of Bd. Voltaire, Rue de Rennes, and Av. Leclerc.

4.7.1. Statistical evaluation of models

STREET-SRI, OSPM and AEOLIUS were initially used in a traditional manner to create time series of CO best estimates. Instead of defining a priori uncertainty ranges in different input variables, three independent meteorological data sets and three different emission factors (as described in Section 4.4 and 4.4.1) were used to create ensemble sets of 27 model realisations for each time step.

In the same manner, ensemble sets of 27 model realisations were also created for benzene. In that case, instead of time series, weekly averages corresponding to different receptor locations in the streets were calculated using expressions (3.4) and (4.28).

The six statistical evaluation measures described in Section 4.5 were then applied to quantify the differences between predicted and observed concentrations. The results of this analysis for 1- and 8-hour CO predictions in Bd. Voltaire, Rue de Rennes and Av. Leclerc are presented in Tables 4.5, 4.6 and 4.7, respectively. It can be seen that for longer averaging times (i.e. 8 hours), the performance of all three models was enhanced. That was expected, since the uncertainty attributed to the turbulent atmospheric processes generally decreases as averaging times increase.

Although there were no dramatic differences in the statistics for the different trials, it might be concluded that OSPM performed slightly better than the other two models in the case of Rue de Rennes. For OSPM, the correlation coefficient ranged from 0.68 to 0.77 for 1-hour averages and from 0.81 to 0.89 for 8-hour averages. For the same model, the FAC2 values ranged from 0.66 to 0.78 for 1-hour averages and from 0.80 to 0.92 for 8-hour averages. The optimum FB values (i.e. near zero) were observed for simulations carried out with emission factors calculated according to Buckland and Middleton (1999) and wind data obtained from Montsouris station.

When the same statistical measures were applied to the Bd. Voltaire data, STREET-SRI seemed to give slightly better predictions than the other two models. For STREET-SRI, the FAC2 reached values of 0.93 and 1.00 for 1-hour and daytime averages, respectively (night measurements were not available in Bd. Voltaire). In that case, it was revealed that emission factors calculated using IMPACT gave the best agreement (FB between -0.22 and -0.51 for 1-hour averages), while the other two emission factors produced large under-predictions (FB between 0.39 and 0.77 for 1-hour averages). A reason for this was probably the fact that IMPACT accounted also for the very significant cold start/running emissions in Bd. Voltaire, an urban environment with many parking spaces, during winter. The use of urban meteorological data was again found to improve the model results.

In the case of Av. Leclerc, the statistical evaluation of the simulations revealed significant model over-predictions (i.e. negative FB values). STREET-SRI gave slightly better predictions than the other two models, with the FAC2 values ranging between 0.48 and 0.68 for 1-hour averages and between 0.43 and 0.76 for 8-hour averages. Nevertheless, the correlation between CO measurements and the values calculated with STREET-SRI was relatively weak (correlation coefficients 0.35 - 0.39 for 1-hour averages and 0.45 - 0.46 for 8-hour averages). The emission factor according to Buckland and Middleton (1999) and the wind data from Orly Airport appeared to slightly enhance the predictions. That was probably due to the fact that the wind speeds measured at Orly Airport were simply stronger than the winds measured in Montsouris Park, which made model over-predictions less pronounced. During this campaign, wind data from St-Jacques Tower were not available.

Finally, some significant discrepancies between OSPM and AEOLIUS predictions were identified (e.g. NMSE and FAC2 values in Table 4.6), despite the fact that both the models are based on the same formulation. This was probably due to certain differences in coding, parameterisation and data pre-processing techniques between the two models.

Table 4.5: Statistical evaluation of 27 model simulations for predicting 1- and 8-hour CO concentrations in **Bd. Voltaire**, using three models (OSPM, STREET-SRI, and AEOLIUS), three wind data sets (from M: Montouris Park, S: St-Jacques Tower, and O: Orly Airport), and three emission factors (according to 1: Buckland and Middleton, 2: IMPACT-ADEME, 3: Touaty and Bonsang).

Statistical evaluation	Correlation coefficient		Fractional bias		NMSE		MG		VG		FAC2	
Ideal value	1.00		0.00		0.00		1.00		1.00		1.00	
Averaging time	1 hour	12 hour	1 hour	12 hour	1 hour	12 hour	1 hour	12 hour	1 hour	12 hour	1 hour	12 hour
OSPM (M1)	0.75	0.97	0.77	0.77	0.98	0.71	3.33	3.00	4.24	3.34	0.24	0.40
OSPM (M2)	0.75	0.97	-0.22	-0.22	1.02	0.42	1.18	1.06	1.03	1.00	0.75	1.00
OSPM (M3)	0.75	0.97	0.67	0.67	0.80	0.53	2.98	2.68	3.29	2.65	0.24	0.40
OSPM (S1)	0.81	0.97	0.62	0.62	0.64	0.50	3.07	2.93	3.51	3.17	0.31	0.40
OSPM (S2)	0.82	0.97	-0.39	-0.39	1.24	0.98	1.09	1.04	1.01	1.00	0.51	0.60
OSPM (S3)	0.82	0.97	0.51	0.51	0.52	0.39	2.72	2.59	2.72	2.48	0.31	0.40
OSPM (O1)	0.75	0.97	0.67	0.67	0.88	0.60	3.94	3.55	6.54	4.97	0.24	0.40
OSPM (O2)	0.75	0.97	-0.33	-0.33	1.79	1.06	1.40	1.27	1.12	1.06	0.32	0.20
OSPM (O3)	0.75	0.97	0.56	0.57	0.79	0.49	3.54	3.18	4.93	3.81	0.24	0.40
STREET (M1)	0.74	0.97	0.76	0.76	0.93	0.71	2.52	2.45	2.35	2.23	0.19	0.20
STREET (M2)	0.74	0.97	-0.23	-0.23	0.49	0.18	0.90	0.87	1.01	1.02	0.92	1.00
STREET (M3)	0.74	0.97	0.66	0.66	0.72	0.51	2.25	2.18	1.93	1.84	0.31	0.40
STREET (S1)	0.75	0.96	0.77	0.77	0.93	0.71	2.59	2.62	2.48	2.53	0.27	0.20
STREET (S2)	0.75	0.96	-0.22	-0.22	0.39	0.29	0.92	0.93	1.01	1.00	0.93	1.00
STREET (S3)	0.75	0.96	0.67	0.67	0.72	0.52	2.31	2.34	2.02	2.06	0.31	0.20
STREET (O1)	0.49	0.91	0.50	0.50	1.62	0.57	2.97	2.75	3.28	2.79	0.15	0.20
STREET (O2)	0.49	0.91	-0.51	-0.50	4.58	1.95	1.06	0.98	1.00	1.00	0.73	0.80
STREET (O3)	0.49	0.91	0.39	0.40	1.68	0.54	2.65	2.45	2.59	2.24	0.15	0.20
AEOLIUS (M1)	0.72	0.98	0.92	0.93	1.41	1.11	4.43	3.71	9.17	5.58	0.22	0.20
AEOLIUS (M2)	0.71	0.98	0.00	0.01	0.72	0.22	1.64	1.37	1.28	1.11	0.42	0.60
AEOLIUS (M3)	0.72	0.98	0.83	0.83	1.13	0.84	3.94	3.29	6.56	4.14	0.25	0.40
AEOLIUS (S1)	0.79	0.97	0.68	0.69	0.74	0.58	3.27	2.98	4.07	3.29	0.36	0.40
AEOLIUS (S2)	0.79	0.97	-0.28	-0.27	0.87	0.70	1.21	1.10	1.04	1.01	0.56	0.80
AEOLIUS (S3)	0.80	0.97	0.58	0.58	0.58	0.44	2.91	2.65	3.13	2.59	0.37	0.40
AEOLIUS (O1)	0.69	0.96	0.88	0.89	1.41	1.04	6.66	5.37	36.50	16.85	0.20	0.20
AEOLIUS (O2)	0.69	0.96	-0.05	-0.04	1.39	0.77	2.47	2.00	2.26	1.62	0.27	0.20
AEOLIUS (O3)	0.70	0.96	0.79	0.79	1.19	0.83	6.04	4.84	25.43	12.00	0.22	0.20

Table 4.6: Statistical evaluation of 27 model simulations for predicting 1- and 8-hour CO concentrations in **Rue de Rennes**, using three models (OSPM, STREET-SRI, and AEOLIUS), three wind data sets (from M: Montouris Park, S: St-Jacques Tower, and O: Orly Airport), and three emission factors (according to 1: Buckland and Middleton, 2: IMPACT-ADEME, 3: Touaty and Bonsang).

Statistical evaluation	Correlation coefficient		Fractional bias		NMSE		MG		VG		FAC2	
Ideal value	1.00		0.00		0.00		1.00		1.00		1.00	
Averaging time	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour
OSPM (M1)	0.68	0.82	0.10	0.00	0.38	0.15	1.08	1.04	1.01	1.00	0.76	0.88
OSPM (M2)	0.68	0.82	-0.12	-0.15	0.42	0.21	0.87	0.84	1.02	1.03	0.78	0.84
OSPM (M3)	0.68	0.82	0.17	0.14	0.40	0.18	1.17	1.12	1.03	1.01	0.73	0.88
OSPM (S1)	0.77	0.89	0.13	0.11	0.31	0.12	1.18	1.12	1.03	1.01	0.78	0.88
OSPM (S2)	0.77	0.89	-0.09	-0.11	0.34	0.15	0.94	0.90	1.00	1.01	0.76	0.88
OSPM (S3)	0.77	0.89	0.21	0.19	0.33	0.14	1.27	1.21	1.06	1.04	0.76	0.92
OSPM (O1)	0.71	0.81	0.19	0.16	0.41	0.23	1.26	1.22	1.06	1.04	0.71	0.80
OSPM (O2)	0.71	0.81	-0.03	-0.06	0.41	0.25	1.01	0.98	1.00	1.00	0.78	0.84
OSPM (O3)	0.71	0.81	0.27	0.24	0.45	0.26	1.36	1.31	1.10	1.08	0.66	0.80
STREET (M1)	0.72	0.83	0.40	0.38	0.55	0.34	1.56	1.48	1.22	1.17	0.65	0.72
STREET (M2)	0.72	0.83	0.18	0.17	0.42	0.20	1.25	1.18	1.05	1.03	0.78	0.84
STREET (M3)	0.72	0.83	0.47	0.45	0.64	0.37	1.68	1.61	1.31	1.07	0.57	0.64
STREET (S1)	0.76	0.89	0.54	0.52	0.72	0.45	1.90	1.80	1.51	1.41	0.49	0.44
STREET (S2)	0.76	0.89	0.33	0.31	0.46	0.21	1.52	1.44	1.19	1.14	0.65	0.76
STREET (S3)	0.76	0.89	0.61	0.59	0.84	0.56	2.05	1.93	1.67	1.54	0.43	0.40
STREET (O1)	0.64	0.74	0.58	0.56	0.95	0.67	1.98	1.86	1.59	1.47	0.44	0.48
STREET (O2)	0.64	0.74	0.37	0.35	0.67	0.41	1.58	1.49	1.23	1.17	0.57	0.60
STREET (O3)	0.64	0.74	0.65	0.63	1.08	0.79	2.13	2.00	1.77	1.62	0.38	0.44
AEOLIUS (M1)	0.64	0.80	0.11	0.07	0.45	0.19	1.40	1.24	1.12	1.05	0.59	0.68
AEOLIUS (M2)	0.66	0.81	-0.11	-0.14	0.47	0.23	1.13	1.00	1.02	1.00	0.56	0.64
AEOLIUS (M3)	0.66	0.80	0.19	0.15	0.46	0.20	1.51	1.35	1.19	1.09	0.59	0.72
AEOLIUS (S1)	0.71	0.86	0.16	0.13	0.43	0.17	1.69	1.56	1.29	1.22	0.49	0.64
AEOLIUS (S2)	0.71	0.87	-0.06	-0.09	0.47	0.20	1.45	1.26	1.15	1.06	0.46	0.60
AEOLIUS (S3)	0.71	0.87	0.24	0.21	0.45	0.18	1.82	1.69	1.40	1.32	0.49	0.64
AEOLIUS (O1)	0.67	0.80	0.40	0.36	0.64	0.36	2.00	1.90	1.57	1.51	0.51	0.56
AEOLIUS (O2)	0.67	0.80	0.19	0.15	0.48	0.24	1.61	1.54	1.24	1.20	0.57	0.68
AEOLIUS (O3)	0.67	0.80	0.47	0.44	0.72	0.43	2.16	2.06	1.75	1.69	0.47	0.56

Table 4.7: Statistical evaluation of 18 model simulations for predicting 1- and 8-hour CO concentrations in **Av. Leclerc**, using three models (OSPM, STREET-SRI and AEOLIUS), two wind data sets (from M: Montouris Park and O: Orly Airport), and three emission factors (1: Buckland and Middleton, 2: IMPACT-ADEME, 3: Touaty and Bonsang). Wind data from S: St-Jacques Tower were not available during this campaign.

Statistical evaluation	Correlation coefficient		Fractional bias		NMSE		MG		VG		FAC2	
Ideal value	1.00		0.00		0.00		1.00		1.00		1.00	
Averaging time	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour	1 hour	8 hour
OSPM (M1)	0.51	0.60	-0.83	-0.85	1.57	1.37	0.38	0.36	2.60	2.83	0.35	0.24
OSPM (M2)	0.51	0.60	-1.01	-1.03	2.32	2.08	0.30	0.29	4.31	4.79	0.25	0.19
OSPM (M3)	0.51	0.60	-0.84	-0.86	1.61	1.40	0.37	0.36	2.68	2.92	0.35	0.24
OSPM (S1)												
OSPM (S2)												
OSPM (S3)												
OSPM (O1)	0.55	0.62	-0.67	-0.70	1.38	1.15	0.52	0.49	1.53	1.67	0.49	0.43
OSPM (O2)	0.55	0.62	-0.87	-0.89	2.05	1.79	0.41	0.39	2.20	2.46	0.43	0.38
OSPM (O3)	0.55	0.62	-0.69	-0.71	1.41	1.19	0.51	0.48	1.57	1.71	0.49	0.43
STREET (M1)	0.39	0.45	-0.25	-0.31	0.93	0.66	0.74	0.68	1.10	1.17	0.63	0.76
STREET (M2)	0.39	0.45	-0.48	-0.54	1.22	0.95	0.58	0.54	1.34	1.48	0.48	0.43
STREET (M3)	0.39	0.45	-0.27	-0.33	0.95	0.67	0.72	0.67	1.11	1.18	0.62	0.76
STREET (S1)												
STREET (S2)												
STREET (S3)												
STREET (O1)	0.35	0.46	-0.13	-0.17	1.30	0.71	0.95	0.88	1.00	1.02	0.68	0.71
STREET (O2)	0.35	0.46	-0.35	-0.40	1.62	0.97	0.76	0.70	1.09	1.14	0.68	0.67
STREET (O3)	0.35	0.46	-0.14	-0.18	1.31	0.72	0.94	0.86	1.00	1.02	0.68	0.71
AEOLIUS (M1)	0.53	0.61	-0.82	-0.84	1.63	1.36	0.43	0.41	2.03	2.24	0.36	0.43
AEOLIUS (M2)	0.53	0.61	-1.01	-1.02	2.40	2.08	0.34	0.32	3.16	3.58	0.27	0.33
AEOLIUS (M3)	0.53	0.61	-0.83	-0.85	1.67	1.40	0.42	0.40	2.08	2.30	0.35	0.43
AEOLIUS (S1)												
AEOLIUS (S2)												
AEOLIUS (S3)												
AEOLIUS (O1)	0.55	0.62	-0.70	-0.72	1.46	1.17	0.56	0.50	1.40	1.63	0.40	0.43
AEOLIUS (O2)	0.55	0.62	-0.89	-0.91	2.17	1.82	0.44	0.39	1.93	2.39	0.31	0.38
AEOLIUS (O3)	0.55	0.62	-0.71	-0.73	1.49	1.21	0.55	0.49	1.43	1.67	0.40	0.43

4.7.2. Concentration ranges using three models

A comparison of statistical performance measures helps to determine if one model or input data set is better than another for a specific application. In certain cases, however, model results may deviate quite significantly (especially for short averaging times), without this being reflected on the overall statistics.

Medians together with maximum and minimum concentrations were calculated for each ensemble set of 27 model realisations corresponding to a specific time and location within Bd. Voltaire and Rue de Rennes. In the case of Av. Leclerc, only 18 model realisations were available. The extreme concentrations were thought to give a rough estimate of model uncertainty in the predictions (Vardoulakis et al., 2001a). As it can be seen in Fig. 4.36 and 4.37, approximately 92% and 95% of CO observations in Bd. Voltaire and Rue de Rennes, respectively, lie within the predicted concentration ranges. For Av. Leclerc, only 74% of CO measurements are within the predicted ranges (Fig. 4.38). In the same manner, error bounds were also attached to 8-hour mean CO concentrations in Rue de Rennes and Av. Leclerc, since CO standards are written as 8-hour averages. It can be seen that approximately 96% of the 8-hour mean measurements lie within the predicted bounds for Rue de Rennes (Fig. 4.39). For Av. Leclerc the respective value was only 77% (Fig. 4.40).

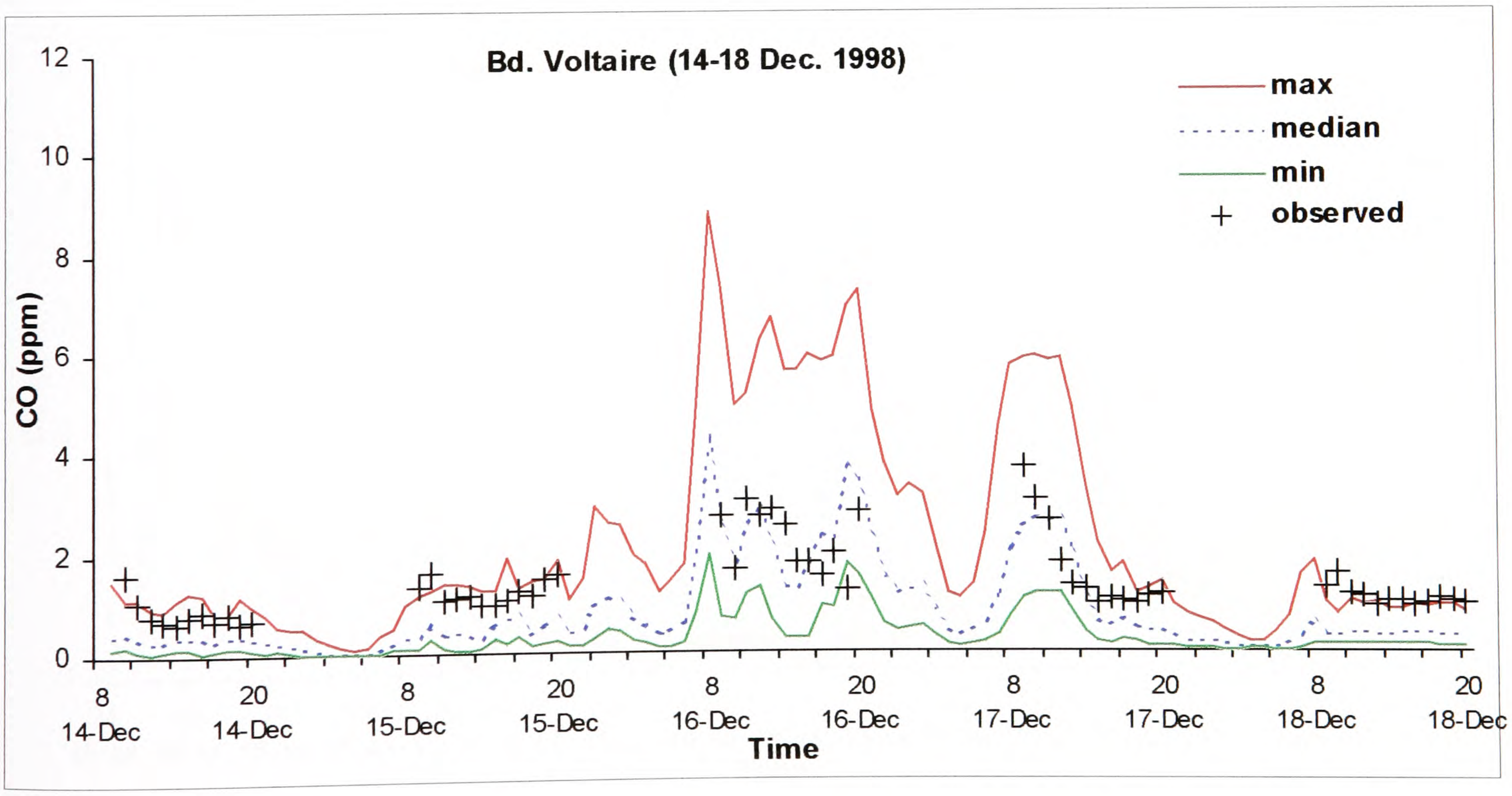


Fig. 4.36: Time series of best 1-hour CO estimates (median), error bounds (max, min), and observed concentrations in Bd. Voltaire produced using three models.

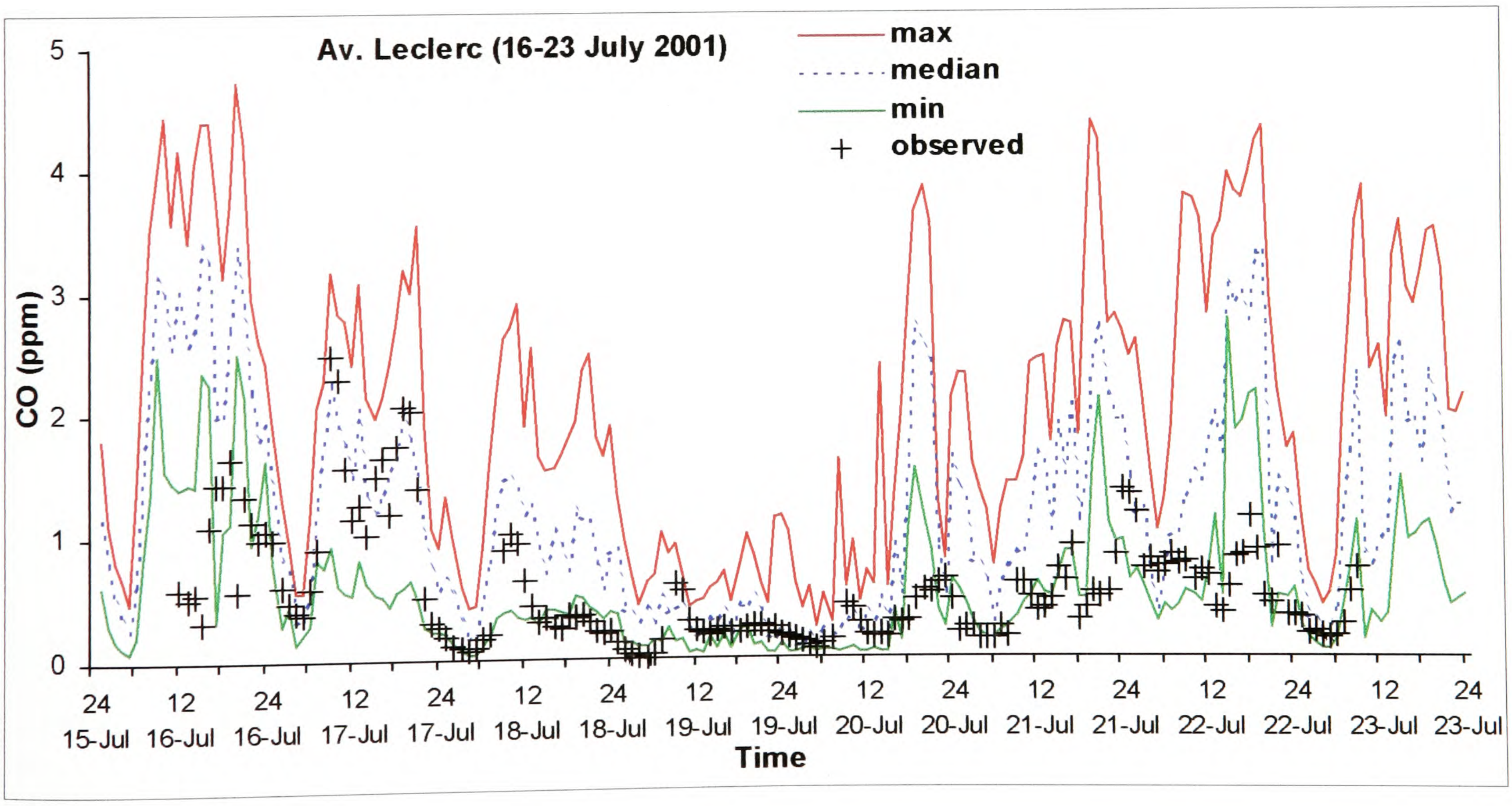


Fig. 4.37: Time series of best 1-hour CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using three models.

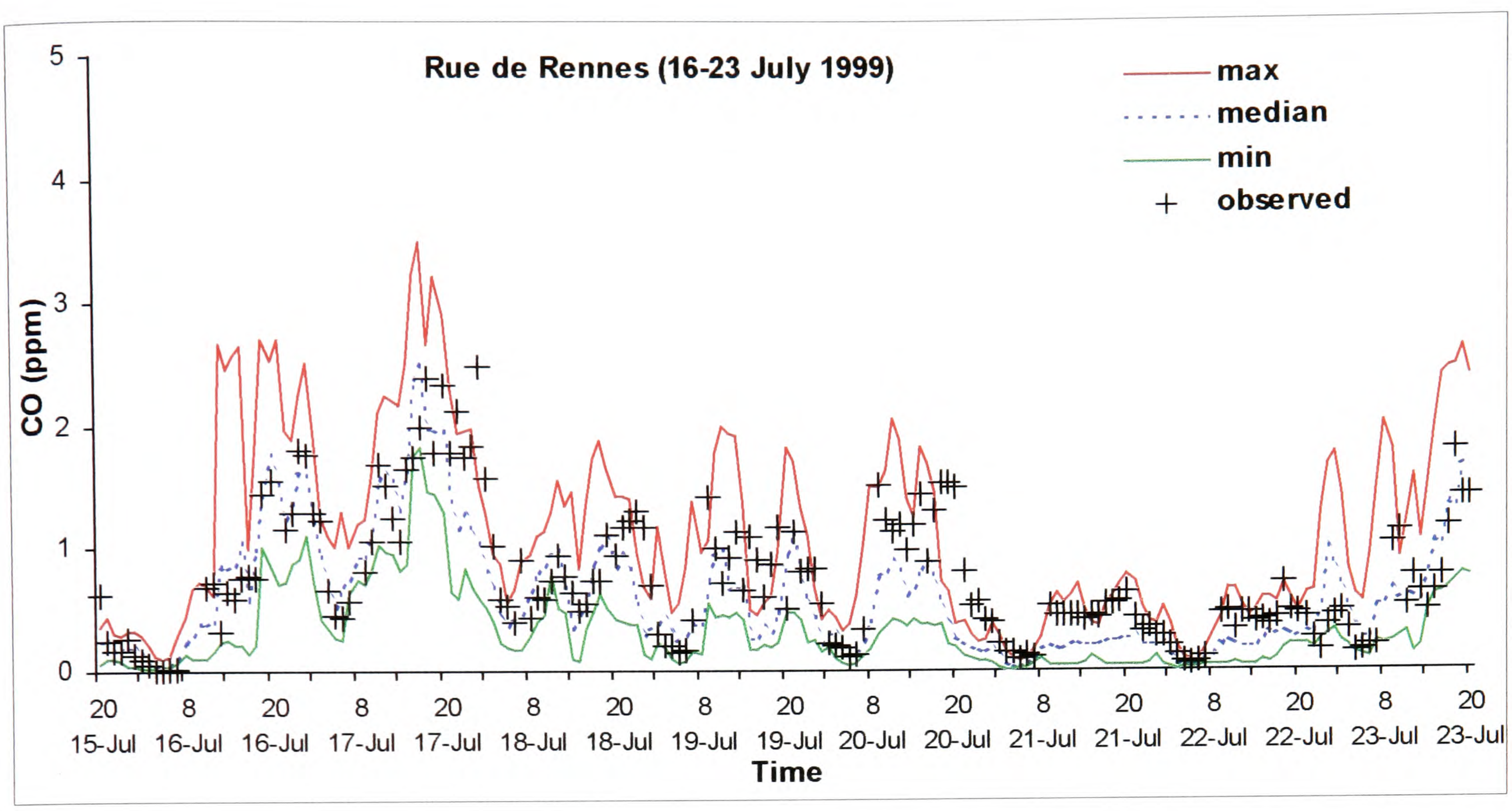


Fig. 4.38: Time series of best 1-hour CO estimates (median), error bounds (max, min), and observed concentrations in Av. Leclerc produced using three models.

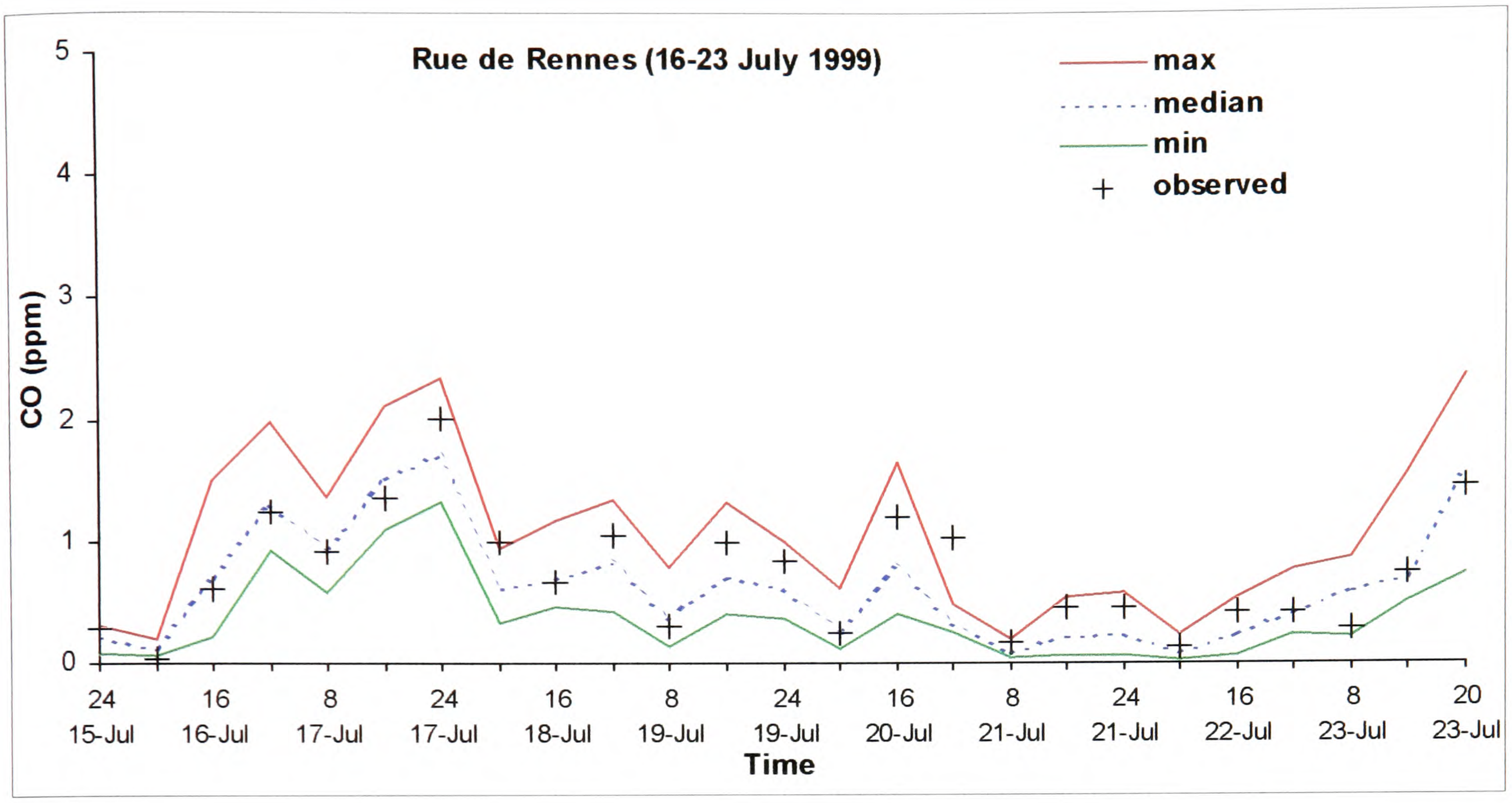


Fig. 4.39: Time series of best 8-hour CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using three models.

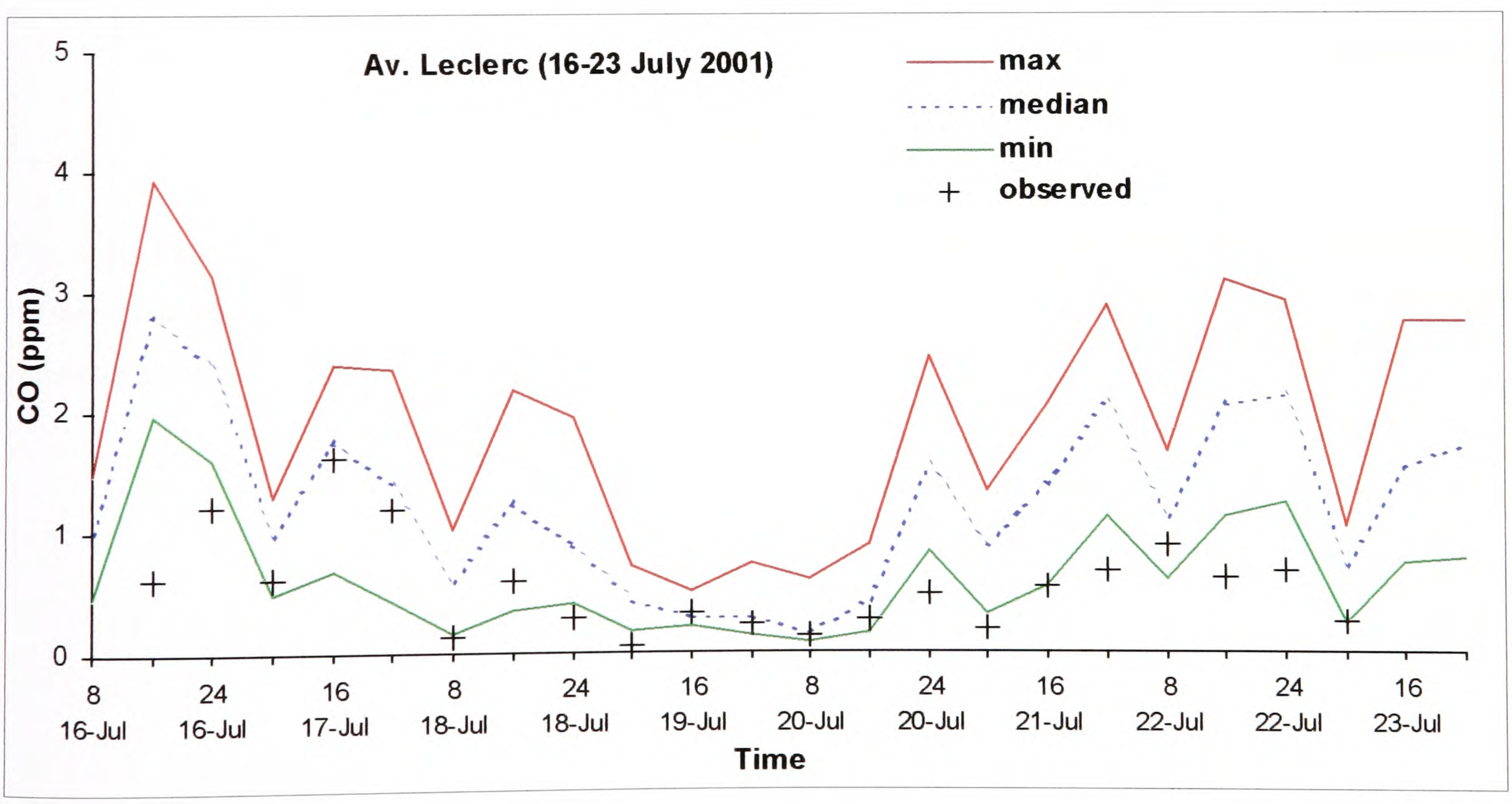


Fig. 4.40: Time series of best 8-hour CO estimates (median), error bounds (max, min), and observed concentrations in Av. Leclerc produced using three models.

The same method was used to obtain a rough estimate of model uncertainty in benzene predictions corresponding to different sampling locations within the three canyons of this study. As it can be seen in Fig. 4.41 and 4.42, all observed benzene concentrations (weekly averages) lie within the estimated error bounds in the two regular canyons of Bd. Voltaire and Rue de Rennes. By contrast, that was the case only for one benzene value out of 11 in the asymmetric canyon of Av. Leclerc (Fig. 4.43).

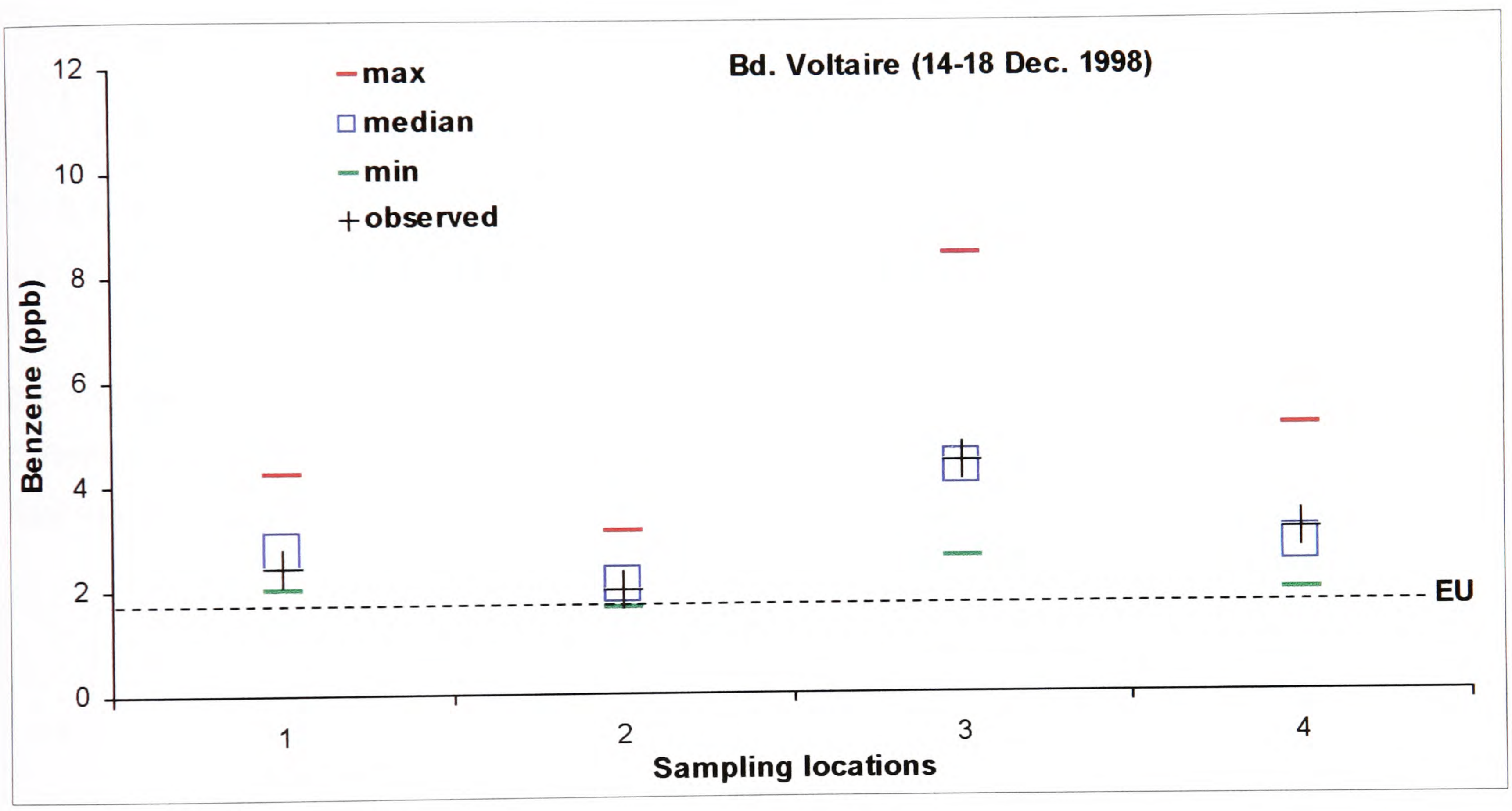


Fig. 4.41: Best weekly benzene estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Bd. Voltaire, produced using three models. The dashed line shows the EU limit value for benzene (1.6 ppb).

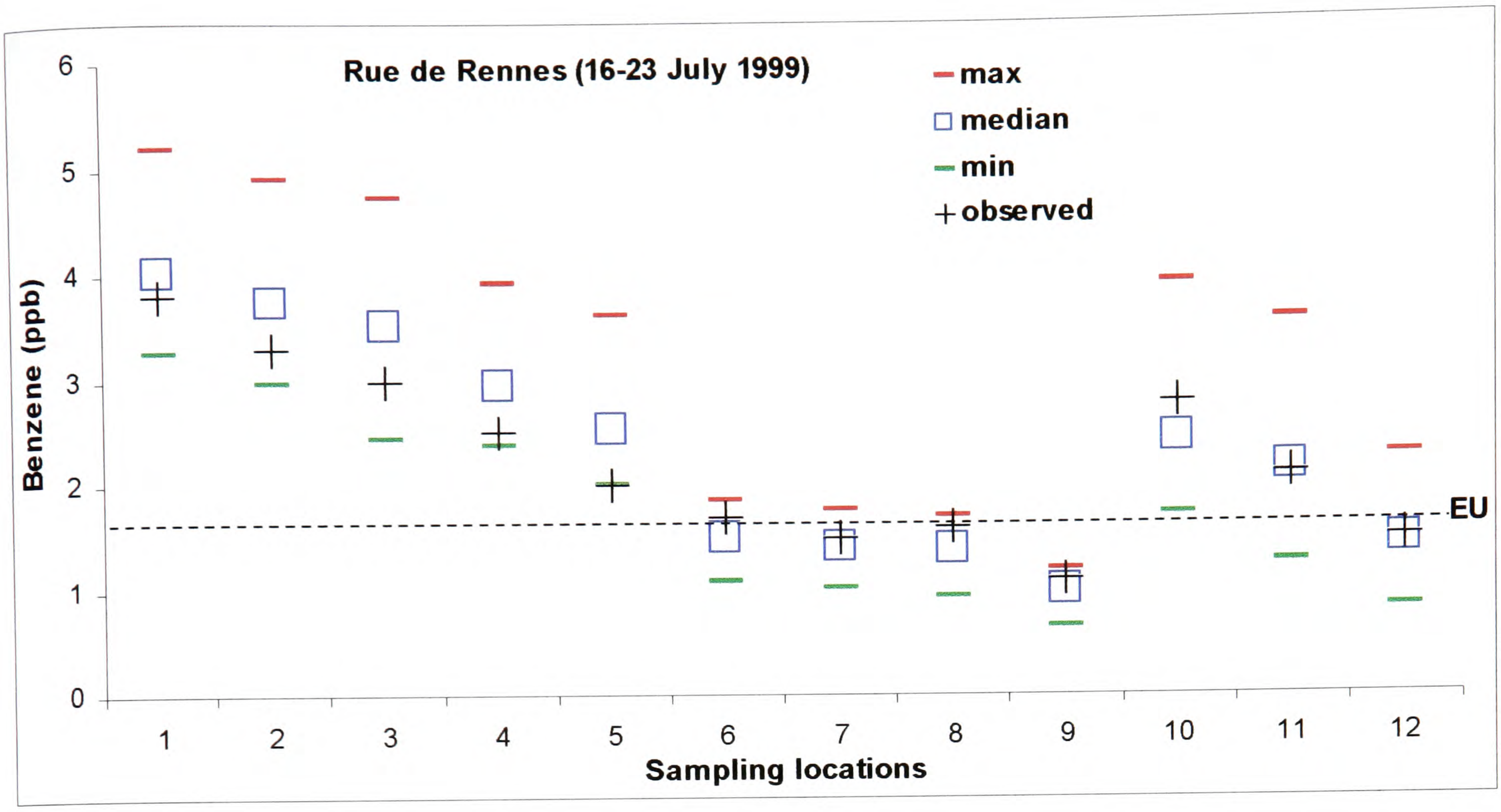


Fig. 4.42: Best weekly benzene estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Rue de Rennes produced using three models. The dashed line shows the EU limit value for benzene (1.6 ppb).

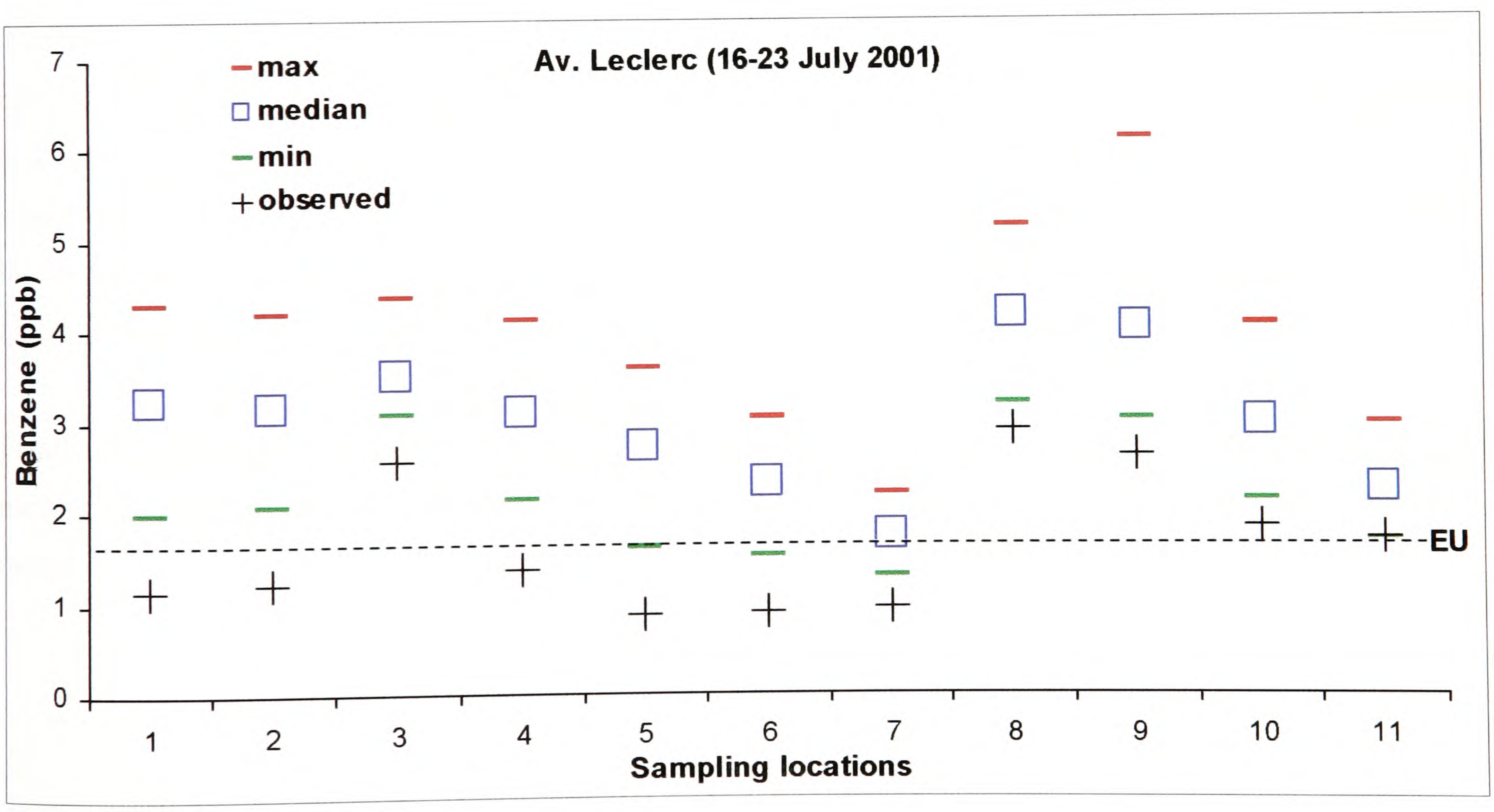


Fig. 4.43: Best weekly benzene estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Av. Leclerc, produced using three models. The dashed line shows the EU limit value for benzene (1.6 ppb).

4.7.3. Concentration ranges using one model

Assuming that there is only one model and one input data set available, it is still possible to have a rough estimate of uncertainty in model predictions. This can be achieved by assigning a best estimate together with two additional values that may bound the likely range of uncertainty related to certain internal model parameters. In the present study, λ, b, and z_o were selected because they were found to have a significant effect on OSPM results (see Section 4.2.1).

Using the meteorological data from Montsouris station and the emission factors calculated according to Buckland and Middleton (1999), an ensemble set of 27 OSPM simulations was again created by only varying the values of b and z_o by 33% and the value of λ by 50%, as shown in Table 4.8. The "max" and "min" values assigned to the three selected parameters were in a reasonable agreement with values previously tested by Buckland and Middleton (1999), and Manning et al. (2000).

Although the number of simulations were here the same as in the examples of Section 4.7.2, the estimated concentration ranges were significantly narrower (Fig. 4.44). As a result, more than 20% of the observed CO concentrations in Rue de Rennes fell outside the estimated concentration ranges.

A more rigorous approach would require that probability functions be developed for each "sensitive" input or internal model parameter, and that these be randomly sampled to obtain improved ensemble sets (Dabberdt and Miller, 2000). On the other hand, this approach would require a much larger number of model runs, which would increase the time and consequently the cost of simulations. Furthermore, it is more important to specify the width rather than the shape of any probability functions describing the uncertainty of the variables (Alcamo and Bartnicki, 1987).

Finally, it should be stressed that this single model approach can be applied only if the user has access to a number of empirical model parameters, which is usually not the case since they are often coded inside the model.

Table 4.8: Values of three internal OSPM parameters (b: aerodynamic drag coefficient, z_o: surface roughness length, and λ : canyon ventilation coefficient) used to create an ensemble set of 27 model simulations.

Coefficients		Estimated values		
		max	standard	min
Aerodynamic drag	(b)	0.40	0.30	0.20
Surface roughness	(z_0)	0.80	0.60	0.40
Canyon ventilation	(λ)	0.15	0.10	0.05

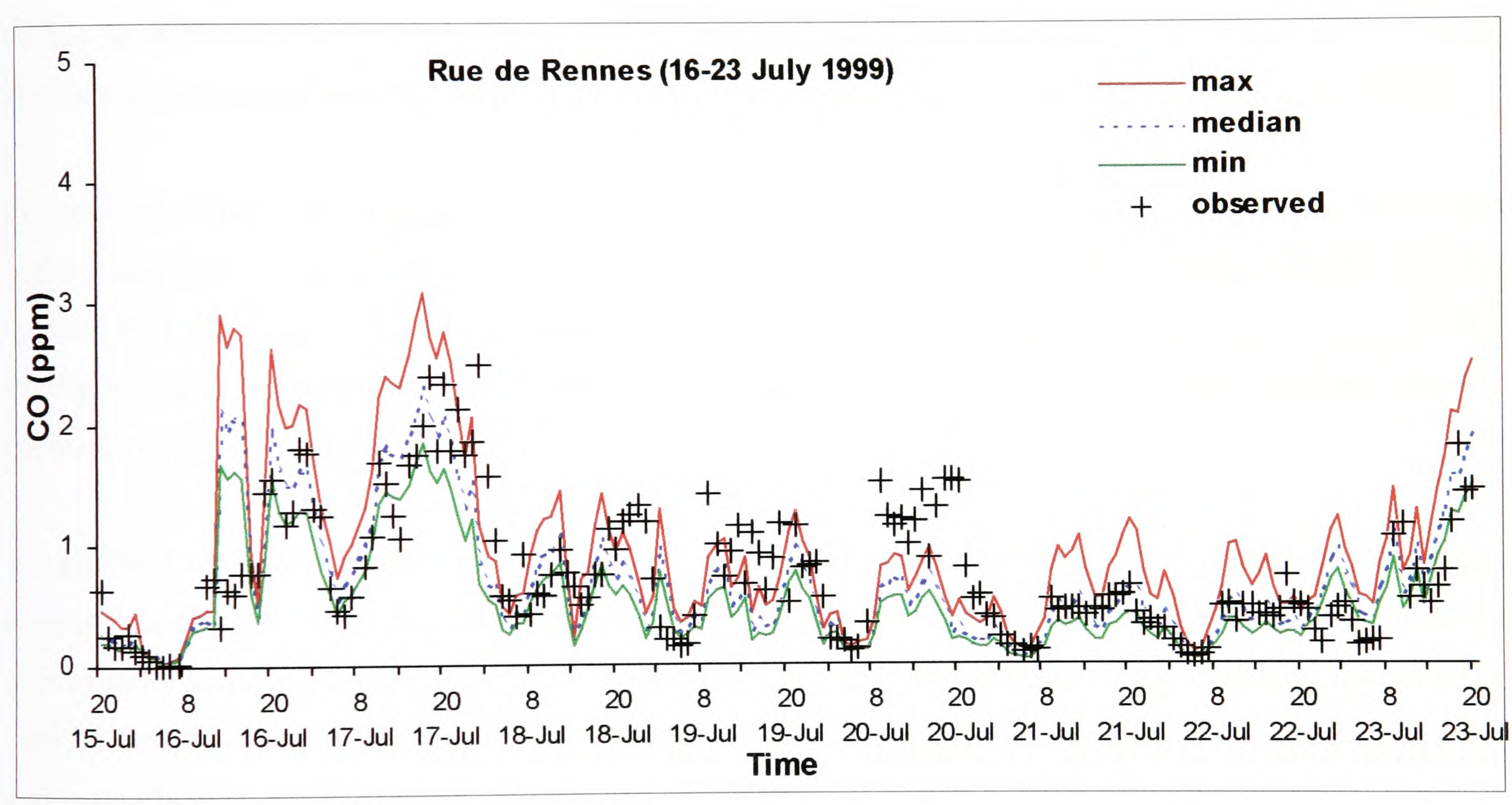


Fig. 4.44: Time series of best 1-hour CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using one model (OSPM).

4.8. Discussion

4.8.1. Emission factors

The CO emission factor calculated using IMPACT was 25% higher than the value calculated according to Buckland and Middleton (1999) for Rue de Rennes during the summer campaign. For Bd. Voltaire, the IMPACT model estimate was 180% higher than the value calculated using Buckland and Middleton's (1999) methodology (Table 4.1). This large discrepancy between the two methods may be justified by the fact that IMPACT also added the very significant cold start/running emissions (during winter) to the hot running values. Cold start/running emissions are indeed expected to be important for urban driving conditions, because of the relative large number of short trips carried out with cold engines, especially in winter. In an on-road experiment in Belgium (De Vlieger, 1997), it was found that the average CO vehicle emissions measured during the cold phase were 4 to 40 times higher than emissions with a hot start. Other experiments showed significant seasonal differences in cold starts due to ambient temperature variations (Mensink et al., 2000).

Furthermore, it has been reported that emissions obtained from aggressive driving can be up to four times higher than those obtained from normal driving (De Vlieger, 1997), and that emission factors typically increase by a factor up to ten during congestion compared to smooth driving conditions (Sjodin et al., 1998). Finally, it should be remembered that the vast majority of fleet emissions come from a small number of poorly maintained vehicles (Singh and Huber, 2000).

The selection of the appropriate emission model or methodology is crucial, since model predictions are almost linearly proportional to the estimated emission factors. The methodology implemented in IMPACT may be seen as more suitable for the present study (especially for the winter campaign in Bd. Voltaire) than the one used by Buckland and Middleton (1999), because it takes into account the average vehicle speed and the seasonal influences in vehicle cold start/running emissions.

Finally, it should be noted that all model simulations were carried out using CO emission factors. This approach was adopted in order to avoid the use of benzene emission factors, which would have introduced a higher uncertainty component in the calculations (due to evaporative losses, etc.). In all cases, benzene predictions were derived from CO values using relationship (3.4), which was thought to be more reliable.

4.8.2. Meteorological data

Meteorological data obtained simultaneously at different weather stations located within few kilometre distances from each other might differ significantly, especially for short averaging periods. When the first street canyon models were developed a few decades ago, it was assumed that the local roof-level wind information needed as an input would not be generally available, and airport wind data would have to be used. For this reason, empirical expressions relating airport and local roof-level winds were derived (Johnson et al., 1973).

Exploring the sensitivity of AEOLIUS to wind data from different sources, Manning et al. (2000) observed that model concentrations were significantly lower when airport rather than local winds were used. That was in agreement with the present study, which showed that simulations carried out using wind data from Orly Airport generally produced lower and less accurate predictions compared to those produced using urban wind data.

Nowadays, there is at least one weather station permanently operating within every European capital. It is, therefore, suggested that wind information from airports be avoided, when suitable urban meteorological measurements (obtained under the same quality criteria) are available for running street canyon models.

4.8.3. Model performance and suitability

In this study, three operational models specially designed for street canyon applications were extensively used (STREET-SRI, OSPM and AEOLIUS). In addition, a screening air quality model (CAR International), a Gaussian plume model (CALINE4), and a CFD code (PHOENICS) were tested using part of the available data.

Statistical methods were used for the evaluation and inter-comparison of the models. Nevertheless, it should be pointed out that the review and evaluation of the scientific components of a model are often of greater importance than their strictly statistical evaluation (Hanna, 1988).

Regular street canyons

STREET-SRI, OSPM and AEOLIUS simulated reasonably well the diurnal variation pattern of roadside CO concentrations within the regular street canyons of Bd. Voltaire and Rue de Rennes, although in some cases they seemed to under-predict the observed values. That was partly due to the large uncertainties associated to the CO emission factors (especially in the case of Bd. Voltaire) as well as to the meteorological input data. Furthermore, these under-predictions might have been also related to the unsatisfactory representation of the concentration field under parallel and windward flow regimes.

That was consistent with the findings reported by Manning et al. (2000) after simulating field measurements carried out in a busy street canyon in Leek, Staffordshire (UK). It was observed that for windward flow, AEOLIUS predictions were less accurate, because there was no direct contribution of pollutants to the monitor. Better predictions were produced when the monitor was leeward, due to the fact that the pollution emitted in the canyon in that case was by far the dominant quantity measured.

In another application, Buckland (1998) observed that AEOLIUS consistently under-estimated the measured mixing ratios when the monitor was on the windward side of a busy street canyon in London (Cromwell Road), which is also in agreement with the present study.

The observed crossroad and vertical benzene gradients in Rue de Rennes and Bd. Voltaire were closely reproduced by OSPM and reasonably well reproduced by AEOLIUS and STREET-SRI. In the case of OSPM and AEOLIUS, the vertical concentration profiles were obtained by coupling the models with an empirically derived exponential function (4.28).

However, it should be emphasised that this relationship is not applicable to traffic-related substances with very short chemical lifetime (e.g. NO, NO_2). It has been experimentally demonstrated that concentrations of reactive species may even increase with height within a street canyon, when the weather conditions favour photochemical activity (Väkevä et al., 1999).

It can be generally concluded that STREET-SRI, OSPM and AEOLIUS are useful tools for assessing the spatial and temporal variability of traffic-related pollutants within regular street canyons. Although these models are already relatively easy to set-up, they can be further improved by establishing user-friendly input/output interfaces.

CAR International is a user-friendly model able to provide annual concentration averages at a height of 1.5 m above the pavement, for a distance of 5 – 30 m between the receptor and the road axis. This model, coupled with expression (4.28) for height correction, showed a tendency to over-predict the benzene concentrations observed in Bd. Voltaire and Rue de Rennes. It should be, however, noted that the observed benzene values were weekly averages and for this reason not directly comparable with the yearly estimates of the model.

An analysis of air quality measurements carried out in Dutch streets revealed that long-term (yearly) concentration averages are much less affected by the presence of buildings than short-term (hourly) values (Eerens et al., 1993). On the other hand, the measurements of the present study provided evidence of strong crossroad gradients within urban streets due to the surrounding buildings. These gradients are expected to persist throughout the year within street canyons that are perpendicular or near-perpendicular to the prevailing wind direction. In these cases, CAR International is unlikely to provide reliable estimates, since it does not take into account the direction of the wind.

CALINE4 predictions were in good agreement with CO measurements carried out in Rue de Rennes during a two-day period of prevailing parallel winds. As already discussed in Sections 2.3.2 and 4.6.6, the street canyon mode of CALINE4 is not compatible with perpendicular wind conditions, which is the main drawback of the model in this kind of applications.

Finally, PHOENICS was successfully tested using data from Bd. Voltaire. This numerical model requires a significantly larger amount of computational resources compared to the semi-empirical models used in this study (i.e. STREET-SRI, OSPM and AEOLIUS). Therefore, it might not be appropriate for routine calculations of air quality levels within regular street canyons.

Asymmetric street canyon

STREET-SRI, OSPM and AEOLIUS were used to calculate hourly mean CO concentrations at street level within the asymmetric canyon of Av. Leclerc. Furthermore, the empirical relationships (3.4) and (4.28) were applied to obtain weekly benzene estimates directly comparable with the passive sampling measurements in the same street.

The comparison between observations and calculated values revealed the tendency of all three models to significantly over-predict the CO and benzene concentrations within the street. In addition, the diurnal variation pattern of CO was only roughly reproduced by these models (Fig. 4.15, 4.22 and 4.29). That was also reflected on Fig. 4.19, 4.26 and 4.33, which showed larger scatter between benzene measurements and predictions in Av. Leclerc than in the regular canyons of Bd. Voltaire (Fig. 4.17, 4.24 and 4.31) and Rue de Rennes (Fig. 4.18, 4.25 and 4.32) using the same models.

These discrepancies may be explained by the complex geometry of Av. Leclerc, a large asymmetric canyon that differs significantly from San Jose and Jagtvej Street, where the original data for the initial parameterisation of STREET-SRI and OSPM, respectively, were collected. In addition, the exponential relationship (4.28) was derived using data from two regular street canyons (Bd. Voltaire and Rue de Rennes) and might not be applicable to asymmetric or wide canyons.

The three rows of big trees inside Av. Leclerc complicated the situation further (at least during summer) by creating additional mechanical turbulence and reducing wind speed at street level. It should be noted that OSPM, AEOLIUS and STREET-SRI do not taken into account the influence of the trees, which might have a significant impact on the measured concentrations.

Finally, the fact that the observed values within the step-up canyon of Av. Leclerc were lower than the model predictions is consistent with a recent study showing that the height increase of the downstream building decreases the pollutant concentrations in the street (Garcia Sagrado et al., 2002). In an earlier study, Hoydysh and Dabberdt (1988) had also demonstrated that concentrations were generally a factor of two lower in the step-up notch relative to the even and step-down notches of their experimental set-up. Although differential wall height was used as model input in the case of Av. Leclerc, model results (i.e. STREET-SRI, OSPM and AEOLIUS) did not appear sensitive to the asymmetry of the canyon. This is probably a reason for which these models greatly over-predicted the observed pollutant concentrations on the kerbside of that street.

CAR International was tested against the monitoring data from the asymmetric canyon of Av. Leclerc. Not surprisingly, the yearly benzene predictions were again much higher than the seven-month average concentrations observed at street level. That was probably because CAR International was developed using experimental data representative of Dutch roadside environments. Furthermore, the model only provides a limited number of road type and average vehicle speed options (see Section 2.3.2), which are not representative of the complex geometry and traffic pattern of Av. Leclerc.

The effect of the trees on pollutant dispersion was taken into account in CAR International by using a standard multiplying factor that reflected the reduction in street-level wind speed due to the presence of trees. However, this might be seen as an over-simplifying approach, since it disregarded the increase in mechanical turbulence induced by trees and other fixed obstacles in Av. Leclerc. If this additional turbulence had been taken into account, it would have reduced to a certain extent the predicted roadside concentrations.

Given the inherent limitations of parametric models, numerical CFD codes (e.g. PHOENICS) may be used to predict pollution levels within asymmetric canyons and other complex urban environments (e.g. intersections, parking spaces, etc). These models, although more demanding in terms of input information and computational resources, can provide a physically realistic representation of the wind turbulence within asymmetric canyons, taking into account the differential height of the buildings as well as the presence of other roughness elements (e.g. trees, parked cars, kiosks, etc.).

Motorway

CALINE4 is a valuable tool for simulating the dispersion of traffic-generated pollutants in open terrain. The advantage of this model is its ability of predicting concentrations at many user-defined receptor locations, at different heights and distances from the kerb. In the case study of RN10 motorway, CALINE4 calculated efficiently pollutant concentrations at 12 receptor locations corresponding to the passive sampling sites near the Stage 2 implemented petrol station.

4.8.4. Modelling uncertainty

Two methodologies were developed to derive best CO and benzene estimates and related error bounds. The first methodology that involved three models (STREET-SRI, OSPM and AEOLIUS) and different input data sets appeared to be more successful. In that case, the vast majority of pollutant concentrations observed in the regular canyons of Bd. Voltaire and Rue de Rennes fell within the estimated error (or *confidence*) bounds. On the other hand, a large number of measurements from Av. Leclerc were below the minimum predicted values, indicating that the selected models were probably inappropriate for this application.

It might be argued that the error limits produced were in some cases so large that they hardly provided any useful information. The fact is that, although large uncertainties do exist, dispersion models are usually applied in a traditional "deterministic" way, often returning results with several significant digits. It is, therefore, preferable to include error bounds (however large they may be) in the predictions in order to avoid inappropriate reliance on modelling results.

An alternative method using fuzzy numbers to treat predictions from more than one model has been proposed by Fisher and Ireland (2001). This method, which provides probability weightings on model predictions, may be applied at a later stage of an air quality assessment, if a more advanced interpretation of the modelling results is needed.

As far as the choice of models is concerned, the intention was to define the appropriate degree of complexity for the specific applications, so as to minimise uncertainties. It might be incorrectly assumed that the total uncertainty in predictions always decreases as the complexity of a model increases. This is only true for the uncertainty attributed to errors in the physical description of the model domain (e.g. incorrect assumptions, oversimplifications, etc.). On the other hand, advanced models require a larger amount of input information, which inevitably introduces a larger data uncertainty component in their calculations.

Chapter 5

Urban air quality management

5.1. Introduction

In most European countries, municipal authorities have an important role in ensuring that air quality objectives are achieved. Particularly in the UK, the National Air Quality Strategy requires local authorities to conduct periodic reviews and assessments of ambient air quality. In the cases where the objectives are not likely to be met by the end of the year specified in the Air Quality Regulations, Air Quality Management Areas (AQMA) have to be designated. In such cases, the local authorities have to carry out a detailed assessment of present and future air quality levels in the area concerned, and prepare an action plan in order to reach the prescribed objectives (DETR, 2000).

The compliance with regulatory standards is only one of the tasks related to air quality that local authorities have to undertake. In addition to that, traffic management and transport planning studies need to be carried out on a regular basis. To manage these tasks, local authorities need efficient air quality monitoring and modelling tools and methodologies. As far as actions need to be taken at local level, these tools should be user-friendly, well documented, and produce reliable results at a relatively low acquisition and operational cost.

5.2. Comparing model predictions with regulatory standards

It has already been discussed that the traditional method of applying air quality models disregards uncertainty (Section 4.7). There is therefore a risk of ending up with a misleading classification of urban environments in only two, "yes" or "no" polluted categories. What is really needed is a probabilistic comparison of predicted values against regulatory limits that takes into account best model estimates as well as their related error bounds.

Having successfully calculated the concentration ranges in Bd. Voltaire and Rue de Rennes (Section 4.7.2), a probabilistic method (Ramsey and Argyraki, 1997) was adopted for assessing the compliance of different kerbside locations with an ambient air quality standard. According to this method, the predicted benzene concentrations (Fig. 4.41 and 4.42) were classified in four different categories with respect to the EU limit value of 5 $\mu g/m^3$ (i.e. 1.6 ppb): (a) "exceeding the limit" if the predicted minimum value for one location was above the threshold, (b) "probably exceeding" if the predicted median was above the limit while the minimum was below, (c) "possibly exceeding" if the predicted median was below the limit but the maximum above, and finally (d) "not exceeding" if the predicted maximum lay below the threshold. According to this classification, locations 1, 3, and 4 in Fig. 4.41 were found to exceed the EU threshold, while location 2 was *probably* exceeding the same limit value. In Fig. 4.42, sampling locations 1 – 5, and 10 were exceeding the threshold, location 11 was *probably* exceeding, locations 6 – 8, and 12 were *possibly* exceeding, and finally only location 9 was found not to exceed the limit.

Although the averaging times in Fig. 4.41 and 4.42 do not directly correspond with the EU standard value for benzene (i.e. annual average), these examples show a precautionary way of applying a discrete air quality criterion. It should be remembered that it is possible to relate short-term averages to annual standards (e.g. for benzene) by means of an appropriate surrogate (e.g. CO) measured throughout the year. An alternative approach would allow for a certain degree of tolerance to be associated with the criterion itself, instead of attaching error bounds to the predictions.

5.3. Implications for population exposure

Two main elements which should be considered when exposure information is required for health impact assessment are the representativeness of the available environmental data for the population at risk, and the averaging time appropriate for creating a link with human health (Krzyzanowski, 1997). Traffic generated pollutants are responsible for both acute and chronic effects on human health. For this reason, they are regulated with respect to different exposure times depending on their properties (see Section 1.1.1).

From a toxicological point of view, benzene is one of the most notorious traffic-related compounds. It has been classified in the "group 1" of carcinogenic substances by the International Agency for Research on Cancer (IARC, 1987) and there is no threshold value below which benzene is not dangerous for human health according to World Health Organisation (WHO, 2000). WHO proposes a *unit risk excess* of 6×10^{-6} per $\mu g/m^3$ for leukaemia, based on a linear extrapolation model without threshold. This is to say that if one million people are exposed to 1 $\mu g/m^3$ of benzene for a lifetime, 6 members of this population are expected to suffer from leukaemia at some stage of their lives. In the UK, like in most EU countries, benzene emissions to ambient air are predominantly derived from road transport and mainly from petrol fuelled vehicles (IEH, 1999).

Although the spatial inhomogeneity of air pollution in urban streets has been quite early raised as an issue (Capannelli et al., 1977), it has been seldom taken into consideration. Given the fact that a large number of residents (e.g. approximately half a million people in Paris) live in the close proximity of urban streets, the strong crossroad and vertical concentration gradients observed may have serious implications in terms of total population exposure.

A further complication is that the actual exposure of individuals to poor air quality is largely a function of indoor air quality, which depends on a variety of factors, including indoor sources and ventilation rates. However, indoor air quality is partly determined by outdoor levels and it is reasonable to assume that outdoor air pollution is a surrogate for personal exposure (Fisher, 2001).

In a study attempting to quantify residential exposure to vehicle exhaust gases in Oslo (Larssen et al., 1993), a correction coefficient was introduced to account for changes in ambient CO concentrations with height over street level. In that case, the coefficient was arbitrarily given the value of 1 for the basement, the ground and the 1st floor of the building facing the street, 0.5 for the second and 3rd floor, and 0.25 for any level above 3rd floor. Using instead equation (4.28), a correction factor of 0.7 was estimated for the 2nd floor, 0.6 for the 3rd floor and so on, giving thus a more physically realistic representation of the vertical pollution profile on the facade of the building.

The present study added evidence to the hypothesis that people living on the leeward side of urban canyons which are perpendicular or near-perpendicular to the prevailing wind direction may be exposed to higher air pollution levels than those living on the windward side. As already suggested by other authors (Croxford and Penn, 1998), the side of the street factor should be taken into account in studies trying to link traffic-related air pollution with public health impact.

Alternative approaches based on traffic counts for assessing the impact of traffic-related air pollution have been proved less reliable than field measurements and/or dispersion modelling, since they do not take into account several important factors like street geometry, wind conditions and urban background levels (Raaschou-Nielsen et al., 2000).

Finally, concerning the benzene concentrations observed near the RN10 petrol station, it may be concluded that pedestrians are likely to be exposed to higher benzene levels in busy urban canyons (e.g. Bd. Voltaire, Rue de Rennes and Av. Leclerc) than in the vicinity of a Stage 1 & 2 implemented motorway service station. Furthermore, under unfavourable wind conditions, the contribution from vehicle traffic in adjacent streets to the pollution levels near the station can be very significant and even higher than the contribution of the station itself.

5.4. An operational method for assessing roadside air quality

In considering their options for achieving air quality objectives and implementing potential mitigation measures, competent authorities need to assess present and future air pollution levels in a variety of urban, suburban and rural sites. The monitoring campaigns and model simulations conducted within the framework of the present study gave insights into certain issues that may play an important role in air quality assessments. The following paragraphs attempt to integrate the lessons learned in the previous chapters into a practical methodology for assessing traffic-related air pollution mainly in urban streets.

Since the total number of permanent air quality monitoring stations in a city is limited due to practical constraints (cost and bulk of equipment, power supply, etc.), alternative measurement and modelling techniques are also needed in order to assess urban air quality with respect to population exposure and compliance with regulations. Furthermore, the strong spatial and temporal variability of traffic-related air pollution detected within the selected streets in Paris raised the question of how representative the site and time period of air quality measurements actually can be.

The present study demonstrated that, in addition to the continuous roadside and background monitoring, a limited number of intensive short-term field campaigns has to be carried out in a variety of urban locations in order to establish the small-scale spatial variability of air pollution, identify representative sampling locations and potential hotspots. These campaigns require a significant amount of resources including diffusive and pumped samplers, meteorological monitoring instruments, continuous gas analysers (sheltered inside a mobile unit), and technical staff (see Sections 3.1.1 and 3.1.2).

Although benzene and CO levels should be mainly observed to establish the spatial and temporal variability of gaseous air pollution respectively, their correlation with other traffic-related pollutants (NO_x, O_3, toluene, xylenes, aldehydes, etc.) may be also examined. During these campaigns a relatively large number of diffusive tubes should be deployed within the selected streets, at different heights and distances from the kerb on both sides (see Sections 2.2.6 and 3.7.6).

Short-term monitoring campaigns may have duration of one to two weeks and be repeated twice a year to cover both summer and winter periods. The repeated measurements enable to regularly update the empirical CO-benzene relationship (3.4) so as to keep up with changes in vehicle fleet composition, fuel quality and/or engine technology. Furthermore, these short-term measurements provide useful information on the diurnal variation pattern of air pollution and the local dispersion conditions (e.g. wind vortex formation).

It should be borne in mind that intensive monitoring campaigns, like those carried out in Bd. Voltaire, Rue de Rennes and Av. Leclerc, require a relative large amount of resources and for this reason they cannot have long duration or be repeated many times throughout the year.

Long-term air quality measurements are usually needed to examine compliance with regulatory standards. Permanent monitoring networks can provide long-term air quality data (e.g. CO and NO_x) of high temporal resolution (e.g. hour by hour). However, they might fail to convincingly represent the spatial distribution of key pollutants like benzene, since they only comprise a limited number of stations within a city (see Section 1.1). For this reason, it is recommended to couple continuous monitoring with long-term passive sampling at a limited number of well-selected roadside locations within the area surrounding the monitoring station.

The most representative long-term sampling locations can be identified during intensive short-term campaigns. At the selected long-term sites, diffusive samplers should be replaced weekly or fortnightly during several months covering summer and winter periods. Relevant meteorological and traffic data can be obtained from the nearest permanent weather stations and traffic counters operating during the same time periods. Finally, a QA/QC programme should be followed throughout sampling and analysis (see Section 3.1.4).

Obviously, it is not possible to take measurements in every single street within a city where population exposure is likely to be high, due to the practical constraints related to air quality monitoring. Therefore, dispersion modelling should be used to generalise monitoring results so as to cover a great number of similar urban locations. Furthermore, models can be used to test different traffic and meteorological situations.

For example, Mukherjee and Viswanathan (2001) carried out scenario simulations aiming to answer the question of "what happens if the average vehicle speed and/or the road traffic volume increases?" on an expressway in Singapore. Using a semi-empirical modelling approach (see Section 2.4.3), they demonstrated that an increase in traffic flow should be associated with an increase in the average vehicle speed up to 85 km/h, as an optimum planning strategy for the future.

For regulatory purposes, it is recommended to use a simple screening model initially, before adopting a more sophisticated approach that will include simulations with a more complex code. For quick air quality surveys, simulations with a simple model like CAR International might suffice. Semi-empirical models like STREET-SRI, OSPM and AEOLIUS coupled with a number of empirical relationships (i.e. expressions 3.4 and 4.28) should be used to assess air quality in regular street canyons.

On the other hand, numerical CFD codes (e.g. PHOENICS) are required for simulating pollutant dispersion in more complex urban sites. These locations may include asymmetric canyons, urban intersections, streets surrounded by detached buildings of different size, urban streets with rows of big trees and/or other fixed obstacles. Finally, Gaussian plume models like CALINE4 can be used to provide concentration estimates near motorways in open terrain or within wide streets surrounded by relatively low buildings (e.g. two storey houses).

Air quality monitoring network design may require both parametric modelling for an initial selection of the streets to be implemented and then CFD simulations in association with diffusive BTX measurements, in order to identify representative locations within these streets.

It should be remembered that the accuracy of model predictions is bounded by the accuracy of input data such as emission factors, traffic and meteorological data, street geometry, etc. Therefore, it is recommended that more than one models and input data sets be used to calculate concentration ranges that reflect the uncertainty related to the predictions (see Section 4.7.2). In regulatory applications, a probabilistic classification of the simulated sites into four categories (i.e. *unpolluted, possibly polluted, probably polluted, polluted*) with respect to an air quality objective should be adopted in order to account for the uncertainty in modelling (see Section 5.2).

Available monitoring data need to be used to validate and possibly calibrate any models applied to new locations. That should include model validation against continuous CO values obtained from an adequate roadside monitoring station, as well as multi-site validation against passive BTX measurements from different locations within the same street (Vardoulakis et al., 2001b).

It should be stressed that the application of dispersion models is optimised when a small (at least) number of relevant field measurements is available. If this is not the case, decision-makers should use modelling results very cautiously.

The combination of monitoring and modelling techniques described in the above paragraphs may be seen as a practical and cost-effective approach to urban air quality management that avoids costly monitoring as well as excessive reliance to models. Furthermore, artificial intelligence tools (i.e. knowledge based reasoning) may be used to facilitate the selection of the most appropriate monitoring/modelling techniques for a specific urban street application with reference to stored experience.

5.5. Mitigation of traffic-related air pollution

Although the detailed investigation of air pollution mitigation measures is beyond the scope of the present study, some potential control measures are briefly discussed in the following paragraphs.

The mitigation of traffic-related air pollution and its related impacts at a local scale generally rely on: (a) technical solutions for improving fuel quality and vehicle engine efficiency, (b) transport planning solutions for reducing road traffic emissions, (c) urban planning measures for reducing population exposure, and (d) individual life-style choices for reducing both traffic emissions and personal exposure.

Technical solution may include better vehicle and engine design, fuel modification (in terms of volatility, hydrocarbon types or additive content) as well as the use of alternative fuels, and more efficient catalytic converters.

Transport planning can include traffic restrictions, road pricing, changes in traffic signal timing, road widening and creation of bus lanes, replacing of at-grade intersections with grade-separated interchanges in order to reduce traffic congestion, etc. (Cooper, 1987; Petit, 1998/9).

Site layout can also be adequately modified to relocate pedestrian occupancy areas away from busy intersections, parking garage entrances, and other highly polluted locations. The development of new motorways bypassing urban agglomerations is a rather controversial solution for reducing population exposure, since it might increase the length of vehicle journeys and encourage the use of private cars.

Other control measures include encouraging public transport, car sharing and parking schemes, employer programs to encourage van pooling, bicycling, walking, staggered work hours, etc. (Oduyemi and Davidson, 1998).

The involvement of central and local government is crucial for informing the public and giving incentives for making environmentally friendly choices (e.g. reducing car dependence). Furthermore, the challenge for local authorities is to integrate air pollution mitigation measures into more general policy packages like transport, energy and land use, economic and sustainable development plans (Beattie et al, 2002). In the future, legislation is expected to be less important than persuasion in changing public perception and everyday practices for improving air quality (Fisher, 2001).

Chapter 6

Conclusions

6.1. Summary of main findings and conclusions

As highlighted in the literature review (Chapter 2), many street canyon studies have been carried out in the past revealing the main features of wind flow and pollutant dispersion in such environments. Most of these studies relied to a certain extent on field measurements and mathematical modelling, without however exploring the full range of available techniques. This is where the present research study finds its scientific relevance.

Most of the individual monitoring and modelling techniques involved in this project had already been used before, but the way they were here combined and optimised was original. Rather than devising new tools, this study tested existing ones and focused on the development of practical methodologies that will help users to take full advantage of available sampling devices and models avoiding costly practices.

To achieve this, an original air quality database was created from field measurements obtained during short- and long-term monitoring campaigns conducted in three street canyons and one urban intersection in central Paris. An additional monitoring campaign was carried out in a motorway service station within the region of Paris. The advantage of these campaigns was that they revealed the small-scale spatial and temporal variability of traffic-related air pollution in urban locations that are representative of high population exposure and likely exceedences of regulatory standards.

Using BTX as indicators, strong spatial concentration gradients were identified in all urban sampling sites. The receptors on the leeward side of the buildings were exposed to substantially higher concentrations than those on the windward side, due to the helical circulation of the air within the three street canyons under perpendicular or near-perpendicular synoptic winds.

A substantial reduction of pollution levels along with height above ground was also observed within the two regular canyons of Bd. Voltaire and Rue de Rennes. For these cases, an exponential relationship (4.28) that reproduces the vertical concentration gradients on both sides of the streets was empirically derived.

Given the strong spatial pollution gradients observed in urban canyons, the placement of air quality sampling equipment becomes crucial both in scientific experiments and routine measurements. Furthermore, there is evidence that human exposure to traffic pollution should be assessed not only as a function of residence distance from the road, but also as a function of side of the street and height above ground.

It was demonstrated that the very high CO and NO_x levels recorded in the permanent AIRPARIF monitoring station of Pl. Basch were not representative of the ambient concentrations to which pedestrians were exposed. Certain misunderstanding may be created with respect to air pollution levels occurring in the vicinity of this junction, if the small-scale spatial variability of pollutant concentrations is not taken into account.

Roadside CO was used as indicator of the short-term temporal variability of traffic-related air pollution in the selected streets. The highest observed concentrations were associated with low wind conditions and/or synoptic winds parallel to the street axis.

BTX compounds and CO were highly correlated during the experiments, confirming that vehicle traffic can be considered as their dominant source near busy roads. The toluene to benzene ratio remained almost constant at all sampling locations of each campaign, which is in agreement with the chemical lifetime and the residence time of these substances in urban streets. Nevertheless, this ratio increased with the year of the measurements, probably due to the reduction in the benzene content of fuels sold in France.

A simple proportionality relationship was empirically established between CO and benzene. This is particularly helpful because it allows to calculate roadside benzene concentrations from CO measurements or predictions, avoiding thus the use of uncertain benzene emission factors.

Three semi-empirical mathematical models (STREET-SRI, OSPM and AEOLIUS) were validated against continuous CO and multi-site diffusive benzene measurements obtained in the regular street canyons of Bd. Voltaire and Rue de Rennes. All three models gave very satisfactory weekly benzene estimates for Rue de Rennes, but they appeared to under-predict (especially STREET-SRI and AEOLIUS) the concentrations measured in Bd. Voltaire. The hourly CO predictions were generally less successful, revealing poor model performance under certain wind regimes (i.e. parallel and windward flow).

In the case of Av. Leclerc, STREET-SRI, OSPM and AEOLIUS failed to closely predict the observed roadside concentrations. Two key parameters that were not treated properly within these models were the differential height of the canyon walls and the urban vegetation. Therefore, a more sophisticated modelling approach (e.g. CFD) may be more appropriate for simulating pollutant dispersion within asymmetric canyons and other complex urban locations.

STREET-SRI, OSPM and AEOLIUS, had already been validated in the past against continuous measurements from roadside air quality monitoring stations. In the present study, the diffusive benzene measurements enabled to test the models for different receptor locations within the same street. Most importantly, that was achieved without investing a great amount of resources (e.g. expensive instrumentation, power supply, etc.). For this reason, it is believed that diffusive sampling will be increasingly used in the future as an alternative technique for creating data sets for model validation purposes.

CAR International was tested against the same data sets showing clearly the advantages (e.g. relative simplicity and user-friendliness) and limitations (e.g. oversimplification with respect to the wind conditions, traffic patterns and street configurations) of a screening model. The Gaussian plume model CALINE4 was proved very useful in a motorway application, but less practical for street canyon simulations.

The RN10 case study demonstrated that Stage 2 vapour recovery can mitigate to a certain extent local air pollution by reducing ambient BTX levels, although it might only bring marginal reductions in the vicinity of petrol stations operating within heavily polluted areas.

The sensitivity of STREET-SRI, OSPM and AEOLIUS to certain input variables (i.e. emission factors and meteorological data) and internal parameters was studied in detail. Emphasis was put on the sensitivity of OSPM to the full set of empirical constants coded inside the model. A "flexible" version of OSPM that allows user access to the internal model parameters was produced.

Practical methodologies for obtaining first estimates of model uncertainty were developed and tested using air quality data from the field experiments in Bd. Voltaire, Rue de Rennes, and Av. Leclerc. To achieve this, two different approaches were adopted to create ensemble sets of 27 model realisations, which were then used to derive best pollutant (i.e. CO and benzene) concentration estimates and related error bounds. Statistical techniques were applied to evaluate the performance of each simulation. It was shown that the use of wind information obtained from urban monitoring stations optimised the application of models. Large uncertainties related to vehicle emission factors were identified.

A probabilistic method for assessing air pollution in urban streets was developed. Although advanced statistical and error propagation techniques were avoided, the proposed methodology certainly increased to some extent the complexity of the simulations. It is, however, believed that it can contribute to reducing the risk of misinterpreting modelling results and making erroneous management decisions.

The present study finally proposed the use of appropriate parametric models in association with a limited number of short- and long-term diffusive BTX measurements for establishing pollution levels and concentration gradients within urban streets. The necessary long-term meteorological, traffic and real time CO, NO_x, and O_3 data can be mainly obtained from permanent monitoring stations. After further validation, this operational method may find application in local air quality reviews and assessments, and population exposure studies.

6.2. List of research achievements

- Air pollution levels and compliance with regulations were assessed in four representative roadside locations in Paris (Bd. Voltaire, Rue de Rennes, Av. Leclerc and Pl. Basch).

- Original data sets (including air quality, meteorological and traffic information) for street canyon model validation were created.

- The small-scale spatial variability of air pollution in urban streets was revealed and its implications for population exposure studies were highlighted.

- The representativeness of real time air quality measurements recorded in a permanent monitoring station (i.e. AIRPARIF station in Pl. Basch) was evaluated.

- Two principal traffic pollution indicators were identified (i.e. CO and benzene) and an empirical relationship between them was established.

- Sensitive internal parameters of three commonly used operational models (STREET-SRI, OSPM and AEOLIUS) were identified.

- OSPM was modified in order to allow user access to the internal model parameters.

- An empirical relationship reproducing vertical concentration gradients within regular canyons was derived.

- It was confirmed that meteorological data from urban weather stations are generally more reliable as street canyon model inputs than data obtained from remote airport stations.

- It was concluded that cold start/running emissions and average vehicle speed should be taken into account when deriving vehicle CO emission factors.

- A practical methodology for estimating the uncertainty associated to street canyon model predictions was developed.

- A probabilistic methodology for assessing compliance with air quality standards taking into account model uncertainty was proposed.

- It was demonstrated that diffusive sampling is a useful technique for multi-site model validation.

- The suitability and performance of different mathematical models (screening, semi-empirical, numerical CFD) in microscale pollutant dispersion applications was assessed.

- The efficiency of the "Stage 2" vapour recovery system in reducing BTX levels in the vicinity of implemented petrol stations was evaluated.

- An operational method for assessing traffic-related air pollution in urban streets was finally proposed.

6.3. Limitations of the study

Due to practical constraints, it was not possible to cover with measurements all types of urban locations affected by road traffic in Paris. Monitoring mainly focused on heavily trafficked street canyons, which were thought to represent reasonably well the urban topography of Paris. Consequently, other urban microenvironments that may also be important in terms of population exposure and compliance with regulations (e.g. intersections, narrow streets, parking spaces, etc.) were neglected.

The pollutants treated in this study were the major traffic-related gases CO, NO_x, O_3 and VOC (mainly BTX). Relevant particulate matter data were not available. Although discussed, the photochemical activity taking place at the monitoring sites during the field campaigns was not modelled, mainly due to the non-availability of relevant background ozone concentrations.

The thermal effects (e.g. differential wall heating) and traffic queuing patterns were neglected in the dispersion model simulations. Furthermore, the influence of surrounding buildings and adjacent streets was only taken into account through a number of empirical assumptions (i.e. height of the canyon walls, average surface roughness, urban background contribution, etc.).

It should be stressed that further validation against experimental data will be required before the empirical expression (4.28) relating pollutant concentrations at different heights within regular canyons may be of general use. In addition, the empirical relationship between CO and benzene (3.4) needs to be regularly updated to reflect changes in engine technology, fuel quality and vehicle fleet composition.

The thorough validation and use of CFD codes in specific street canyon applications was thought to fall beyond the scope of the present study, which focused on the optimisation of operational dispersion models.

It should be finally noted that uncertainties related to air quality measurements (e.g. sampling and analytical errors) were not taken into consideration. These uncertainties, though generally smaller than model uncertainties, may have some implications in decision making, if the predicted concentrations are close to an air quality standard.

6.4. Recommendations for further research

There is scope for further modelling of pollutant dispersion within asymmetric canyons and urban intersections. Detailed CFD simulations should be carried out for a variety of complex urban sites and empirical models might be derived. Furthermore, artificial intelligence in the form of *case base reasoning* may be applied in order to create a database storage and recovery system containing a great amount of street canyon data already available in the literature.

The recently launched DAPPLE project (Dispersion of Air Pollution and Penetration into the Local Environment) is expected to provide further insight into the formation of localised pollution hotspots and the implications for personal exposure in urban microenvironments.

Mitigation measures based on scenario simulations should be developed for polluted urban microenvironments. Dispersion models can be used to test the effectiveness of potential traffic management measures (e.g. creation of bus lanes), street geometry modifications (e.g. road widening) and flow control measures (e.g. flow deflectors/screens, building design, tree planting, etc.) prior to their implementation.

Future research on urban air quality is expected to focus on topics related to low wind conditions, thermal effects due to solar radiation, and microscale dispersion around fixed and moving obstacles. Fine and ultrafine particles, polycyclic aromatic hydrocarbons (PAH), 1,3-butadiene, formaldehyde and some other VOC are also expected to draw more attention in the future.

Finally, despite the large number of existing codes, there is still a need for scientifically sound, user-friendly and well-documented air quality models, as well as for high quality experimental data sets.

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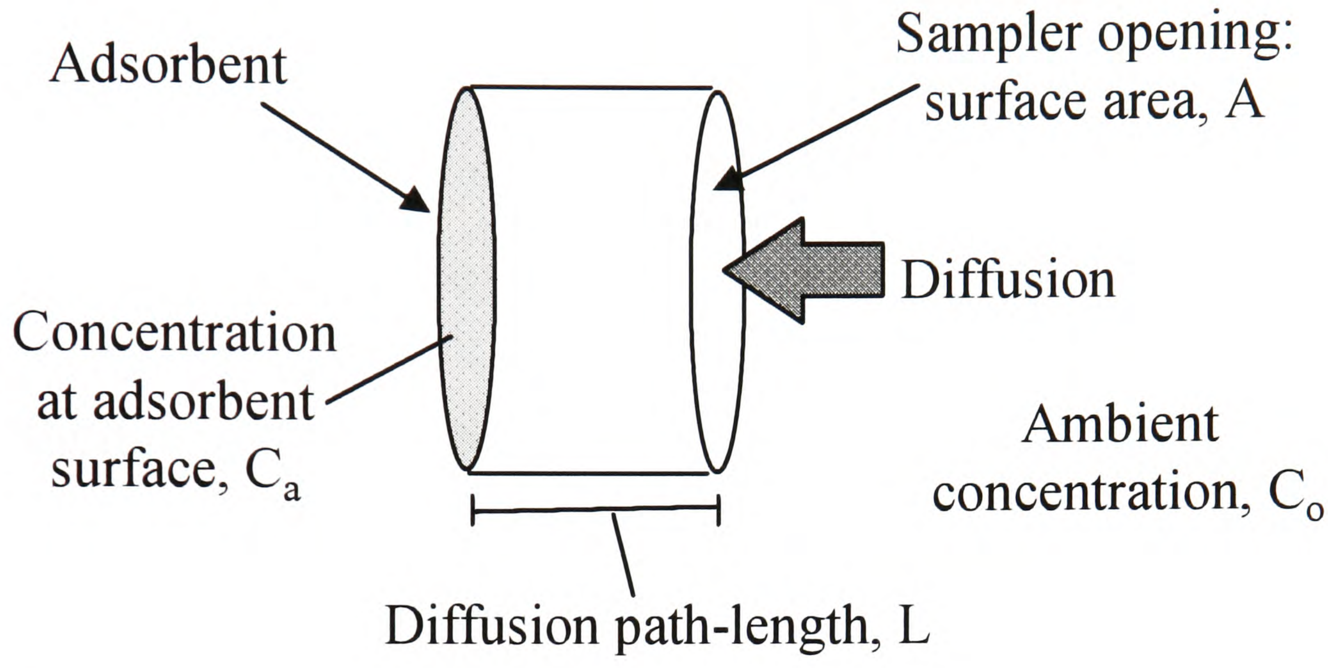
Appendix I

Diffusive sampler geometry and sampling shelter

(A) Axial sampler



(B) Radial sampler

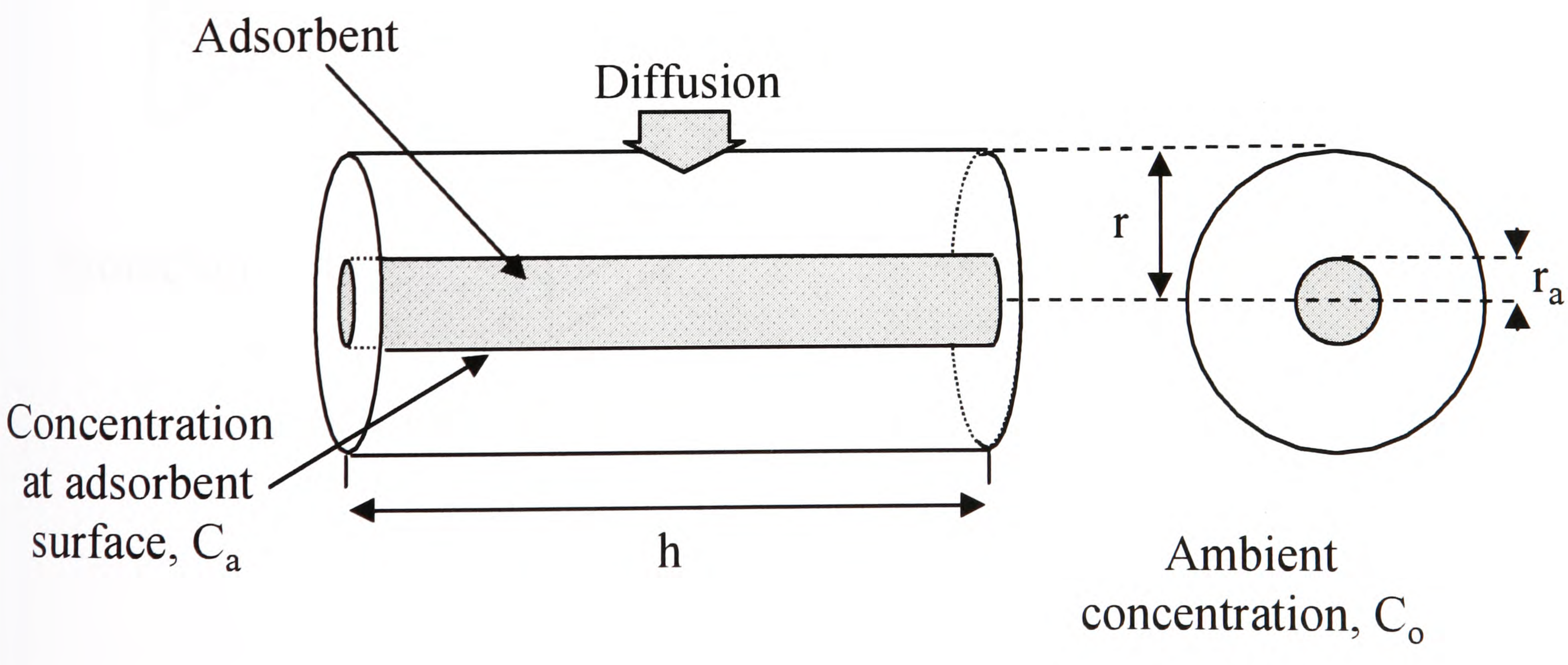


Fig. AI-1: Diffusive sampler geometry

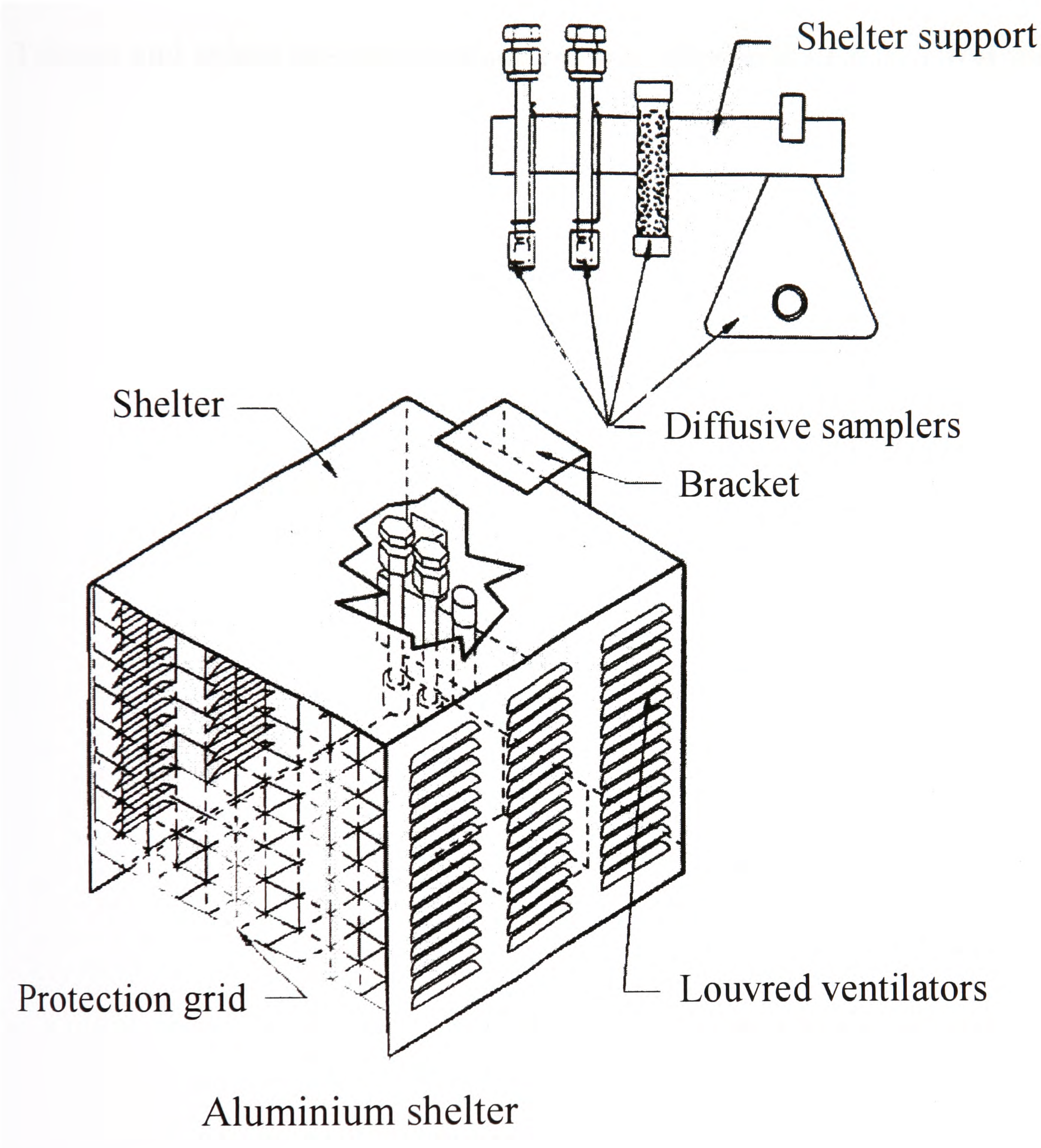


Fig. AI-2: Shelter for passive sampling

Appendix II

Toluene and xylene iso-concentration contour maps in the RN10 petrol station

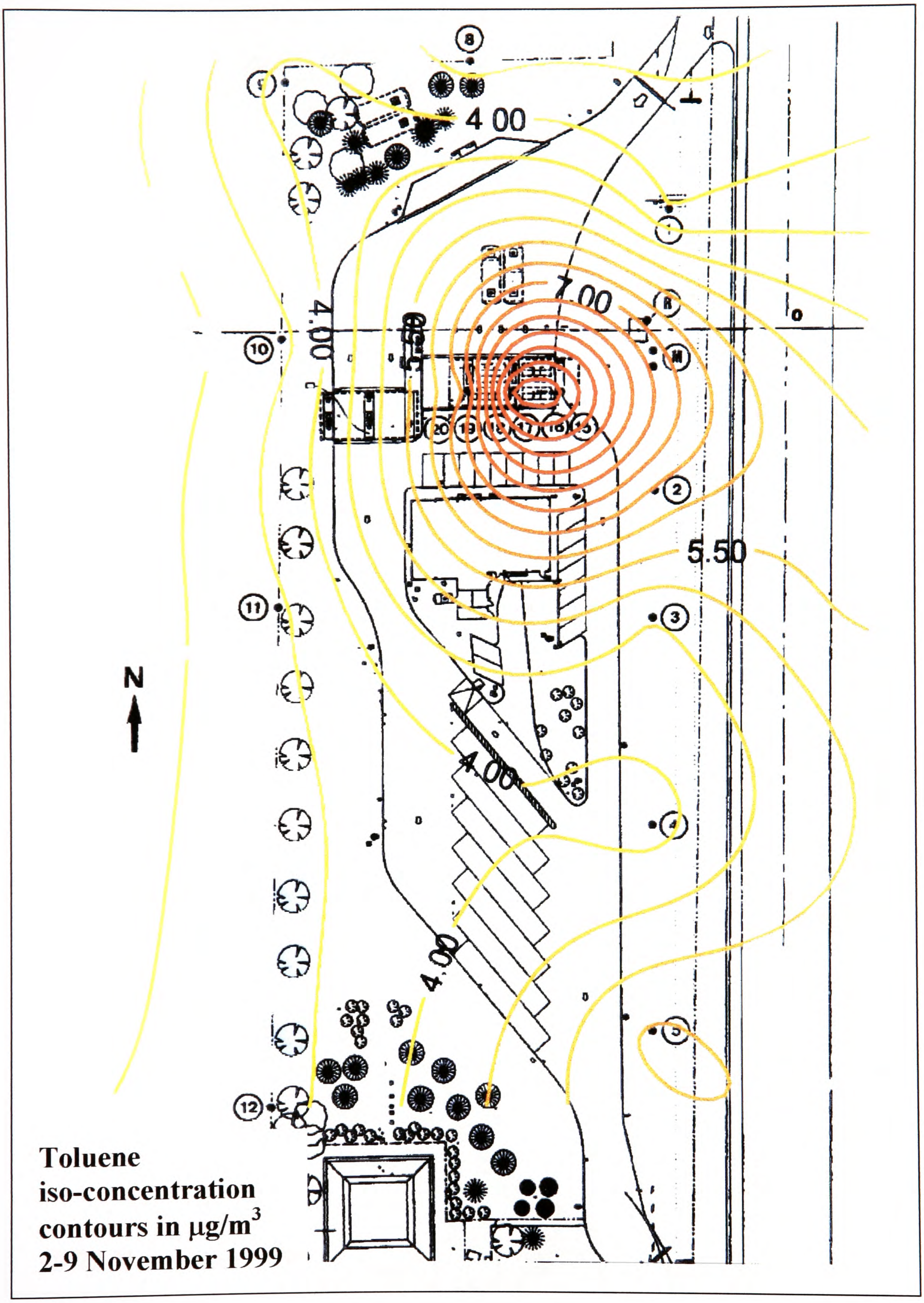


Fig. AII-1a: Toluene iso-concentration contours ($\mu g/m^3$) in the RN10 petrol station while Stage 2 vapour control was operating (2-9 November 1999).

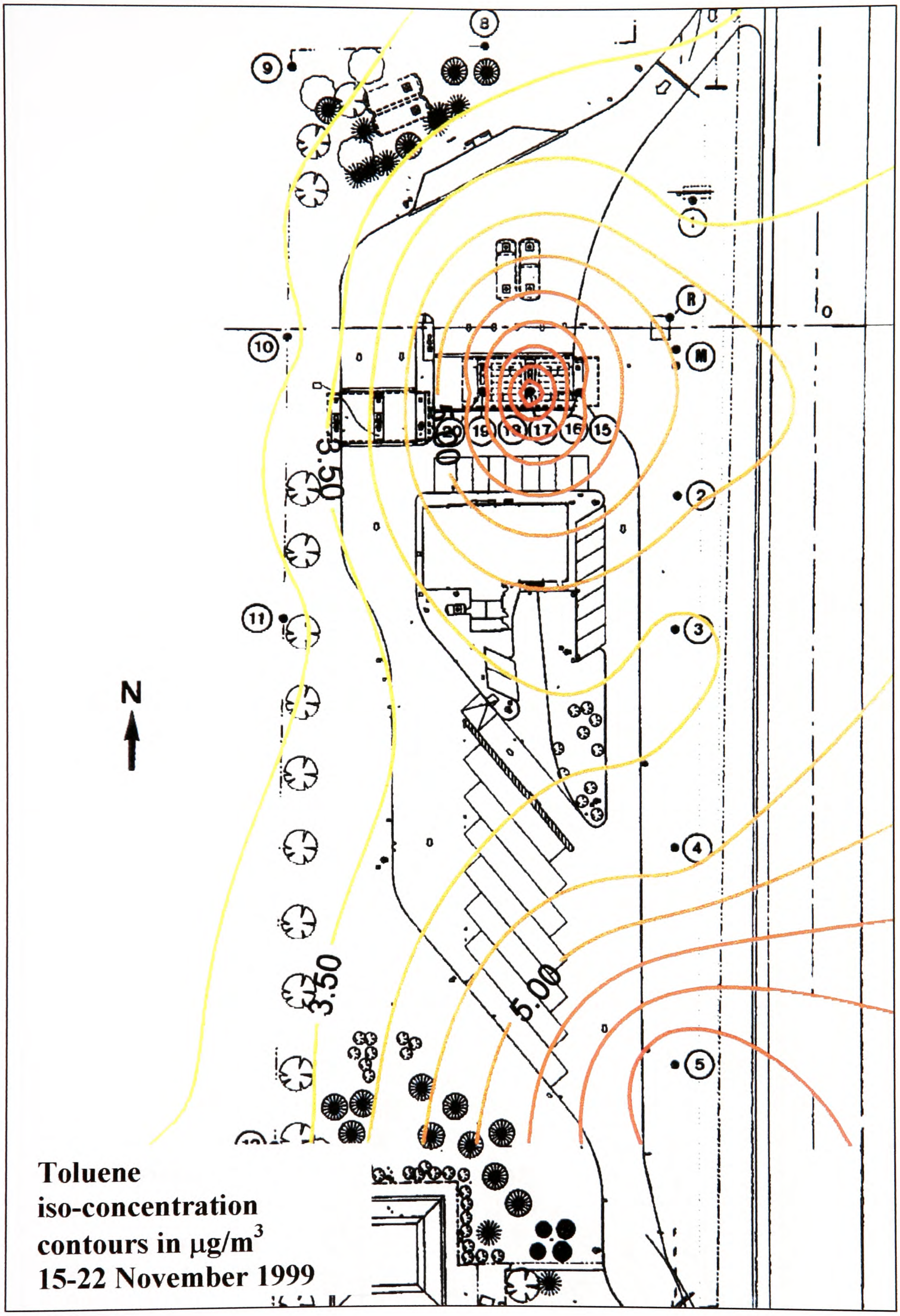


Fig. AII-1b: Toluene iso-concentration contours (µg/m^3) in the RN10 petrol station while Stage 2 vapour control was disconnected (15-22 November 1999).

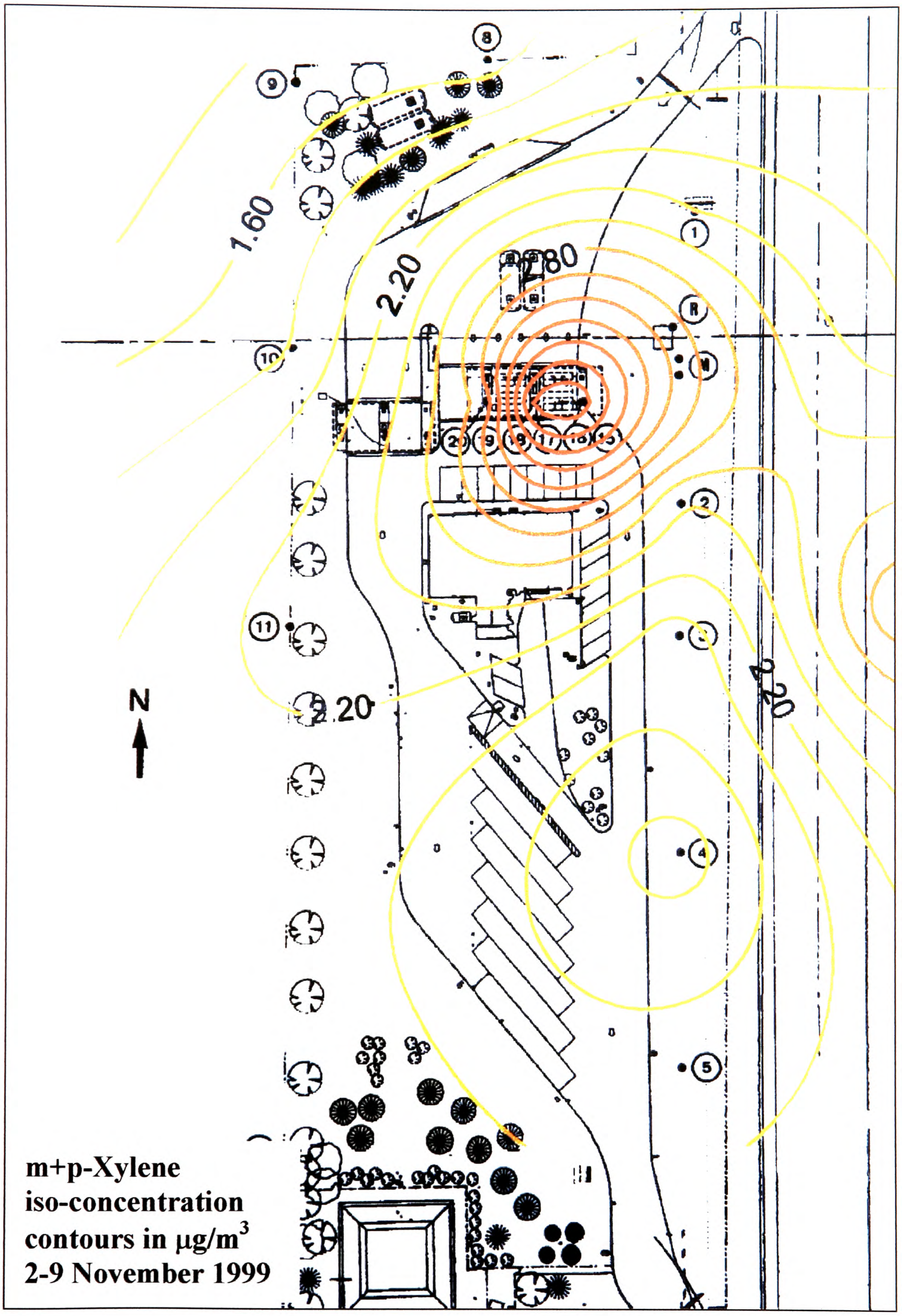


Fig. AII-2a: m+p-Xylene iso-concentration contours (µg/m³) in the RN10 petrol station while Stage 2 vapour control was operating (2-9 November 1999).

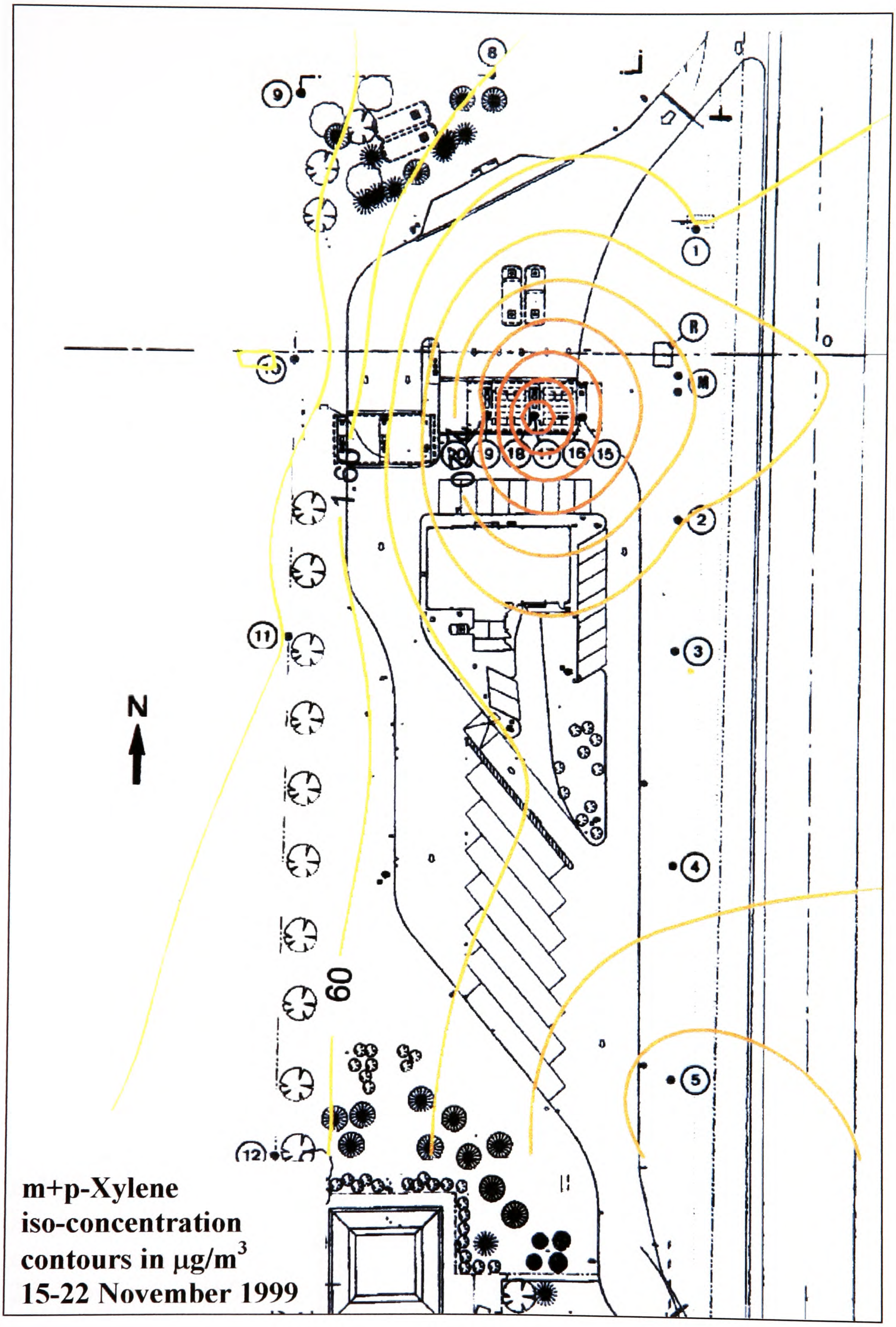


Fig. AII-2b: m+p-Xylene iso-concentration contours (μg/m³) in the RN10 petrol station while Stage 2 vapour control was disconnected (15-22 November 1999).

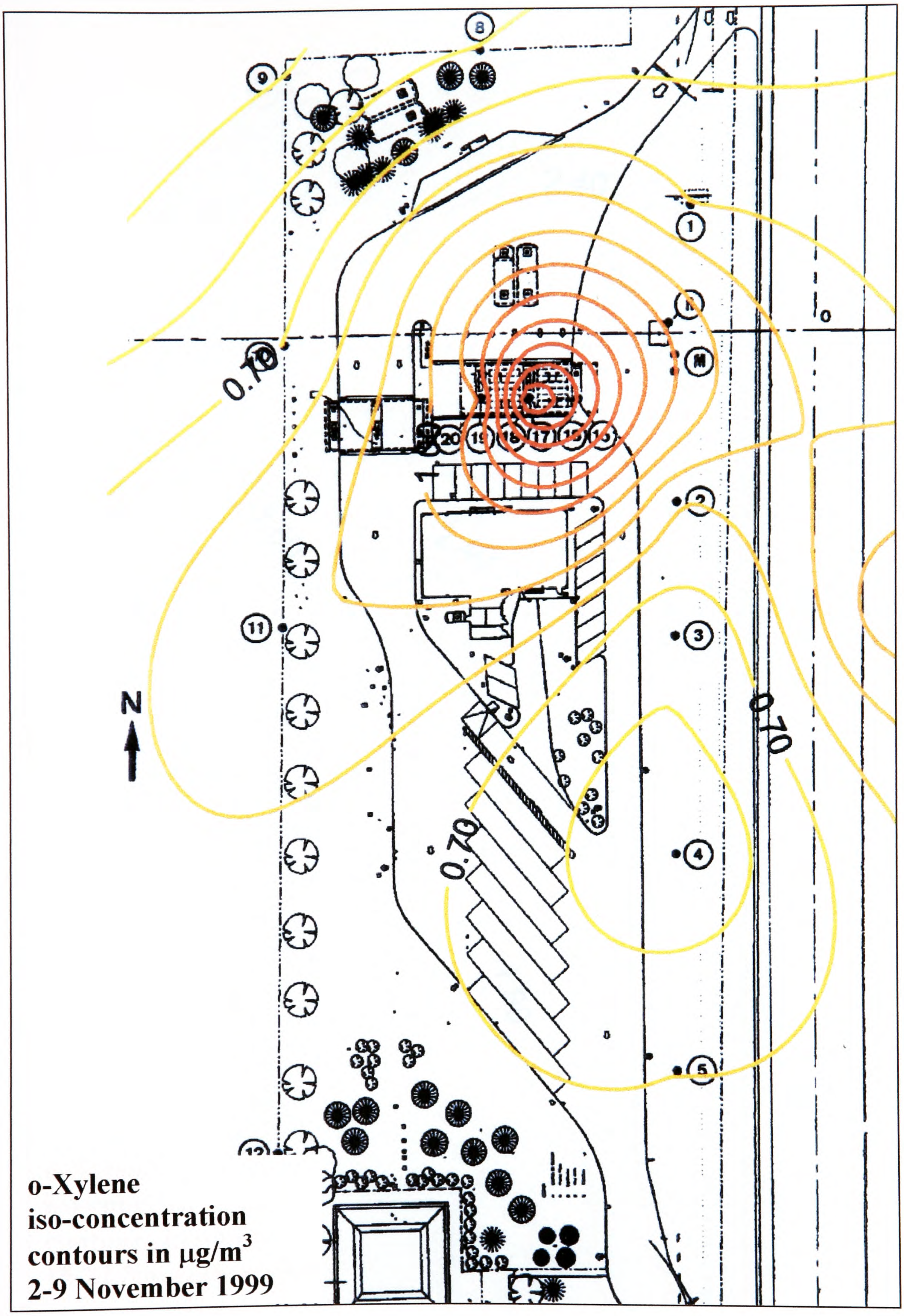


Fig. AII-3a: o-Xylene iso-concentration contours ($\mu g/m^3$) in the RN10 petrol station while Stage 2 vapour control was operating (2-9 November 1999).

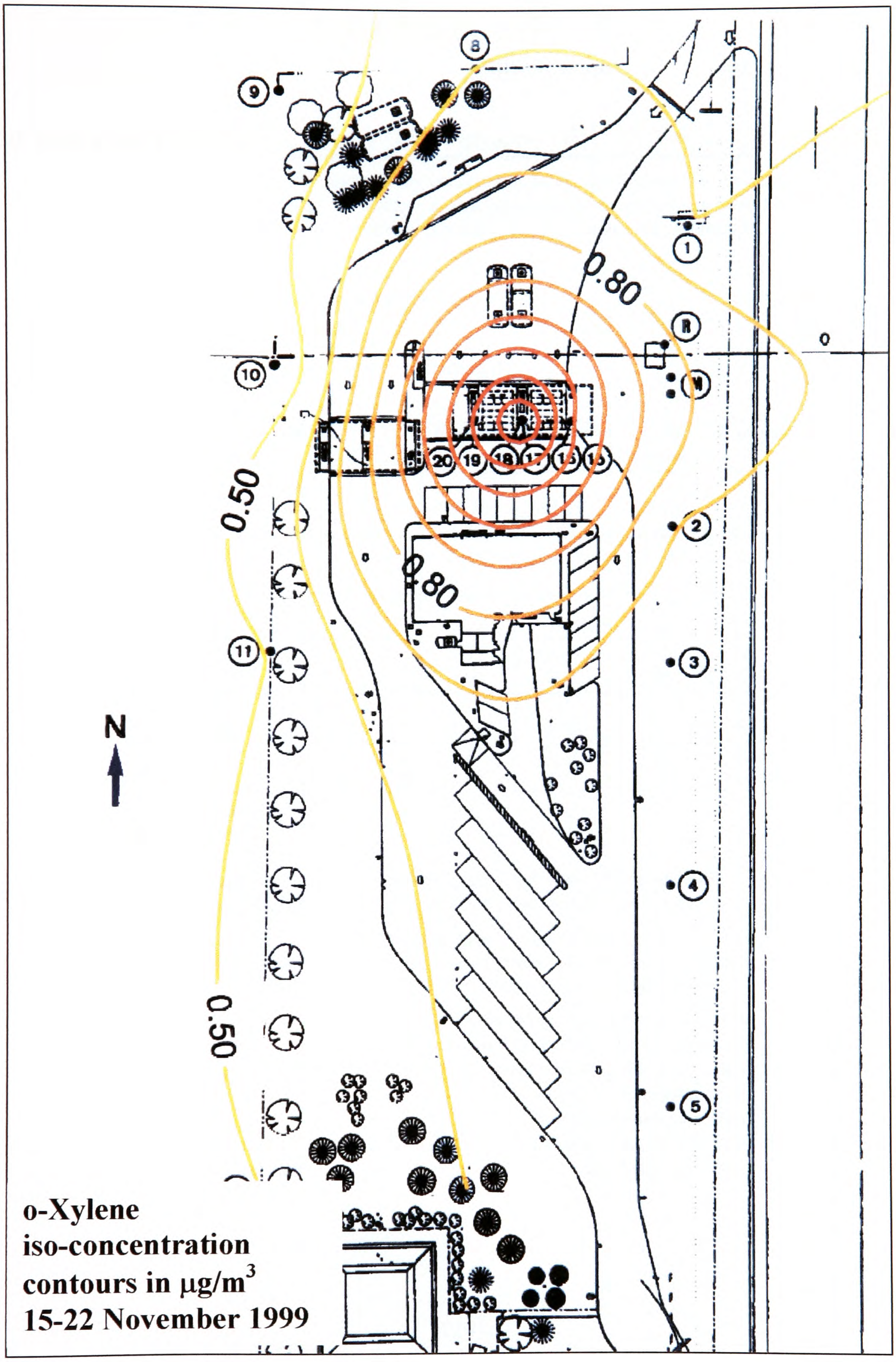


Fig. AII-3b: o-Xylene iso-concentration contours (μg/m³) in the RN10 petrol station while Stage 2 vapour control was disconnected (15-22 November 1999).

Appendix III

Wind and concentration field in Bd. Voltaire (PHOENICS simulations)

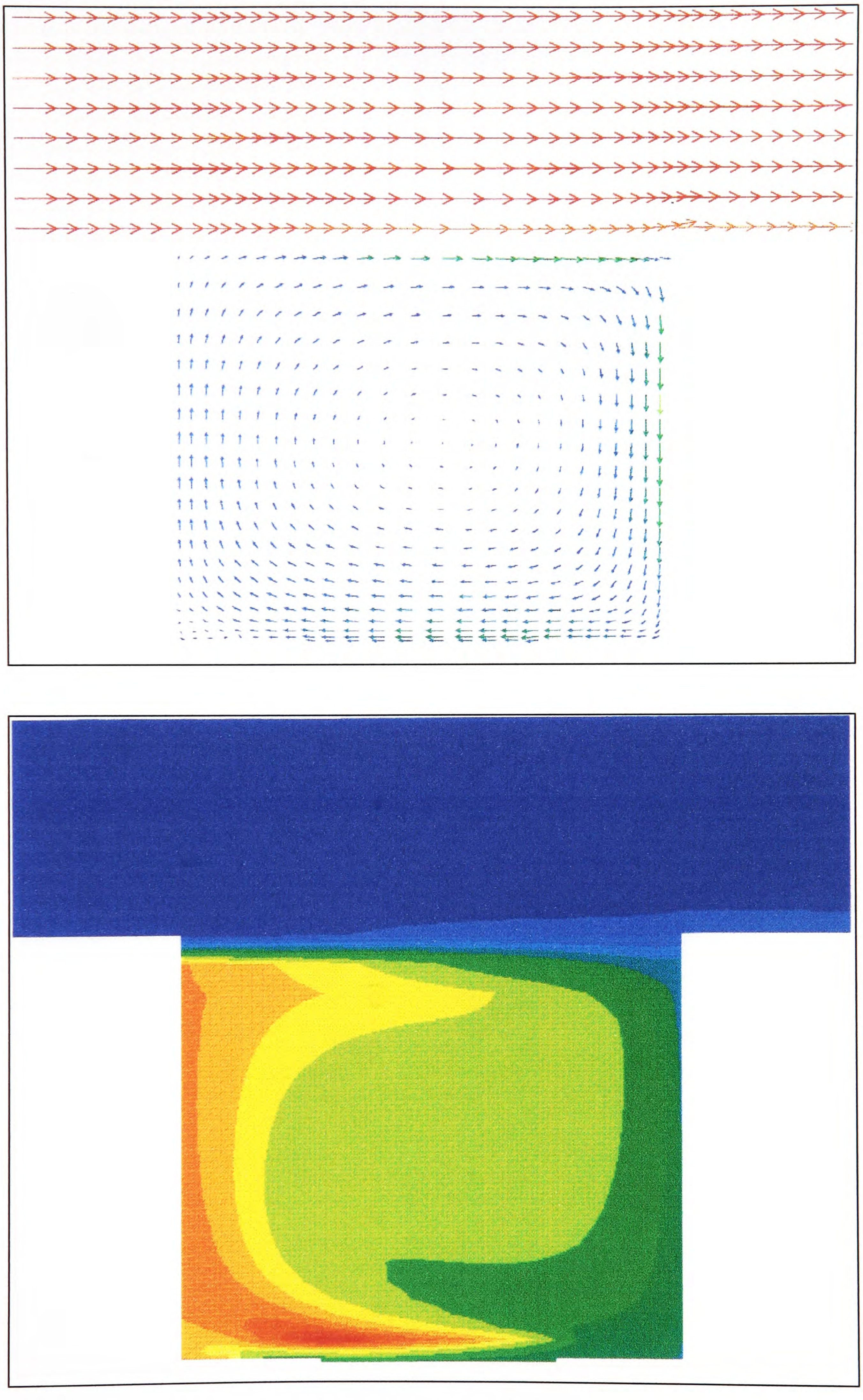


Fig. AIII-1: Wind field (top) and CO concentration field (bottom) in Bd. Voltaire calculated with PHOENICS (synoptic wind speed entering the model domain: 2.5 m/s).

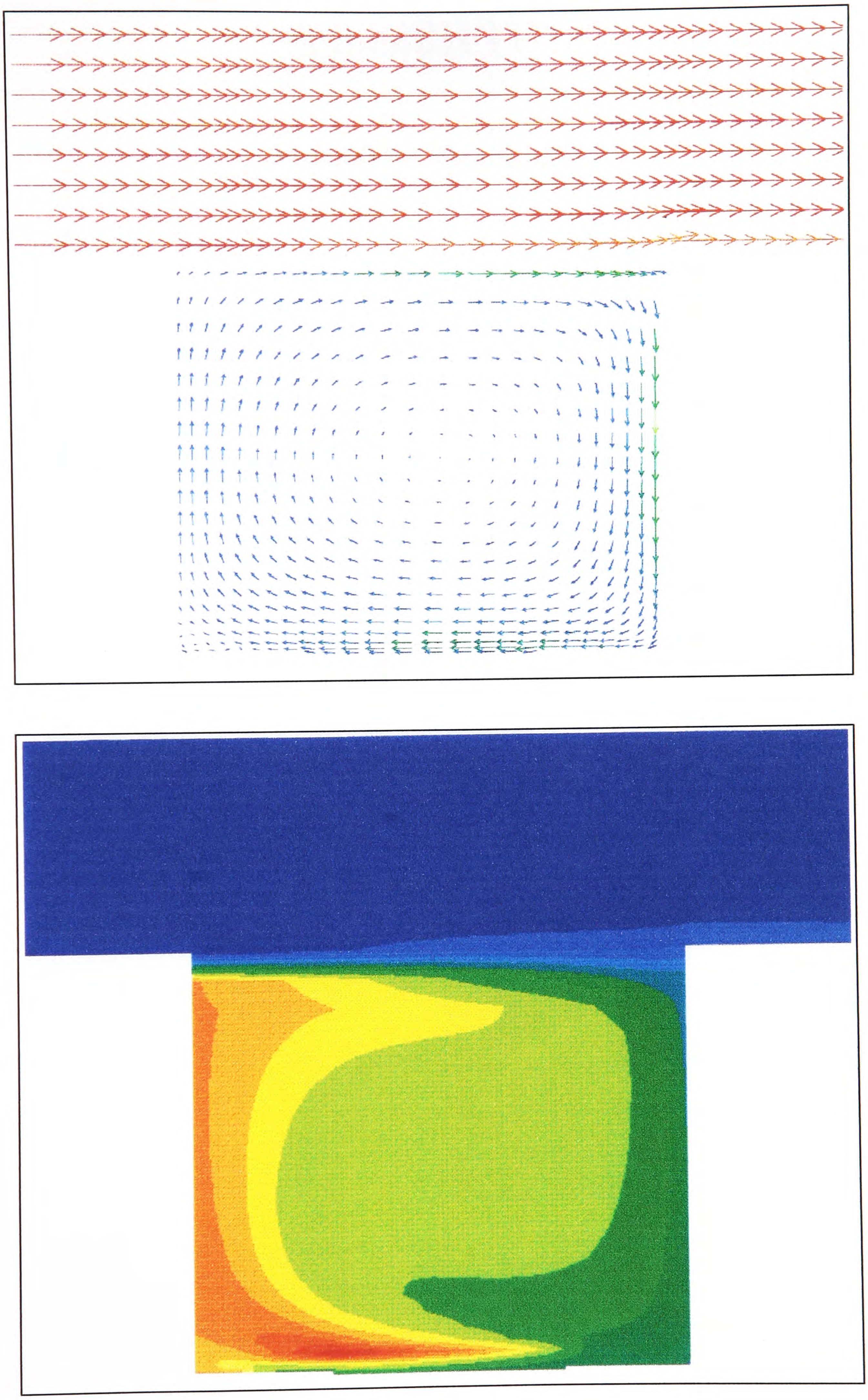


Fig. AIII-2: Wind field (top) and CO concentration field (bottom) in Bd. Voltaire calculated with PHOENICS (synoptic wind speed entering the model domain: 4 m/s).

Appendix IV

Journal Publications

PERGAMON

Atmospheric Environment 36 (2002) 1025–1039

ATMOSPHERIC ENVIRONMENT

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Assessment of traffic-related air pollution in two street canyons in Paris: implications for exposure studies

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Abstract

The small-scale spatial variability of air pollution observed in urban areas has created concern about the representativeness of measurements used in exposure studies. It is suspected that limit values for traffic-related pollutants may be exceeded near busy streets, although respected at urban background sites. In order to assess spatial concentration gradients and identify weather conditions that might induce air pollution episodes in urban areas, different sampling and modelling techniques were studied.

Two intensive monitoring campaigns were carried out in typical street canyons in Paris during winter and summer. Steep cross-road and vertical concentration gradients were observed within the canyons, in addition to large differences between roadside and background levels. Low winds and winds parallel to the street axis were identified as the worst dispersion conditions. The correlation between the measured compounds gave an insight into their sources and fate. An empirical relationship between CO and benzene was established. Two relatively simple mathematical models and an algorithm describing vertical pollutant dispersion were used. The combination of monitoring and modelling techniques proposed in this study can be seen as a reliable and cost-effective method for assessing air quality in urban micro-environments. These findings may have important implications in designing monitoring studies to support investigation on the health effects of traffic-related air pollution.

Keywords: Air quality; Urban canyon; Roadside monitoring; Modelling; Spatial variability; Dispersion

1. Introduction

The increasing awareness of scientists and public about the acute and chronic health effects of several traffic-related pollutants (NO_2, CO, hydrocarbons, etc.) has led in recent years to a significant number of relevant epidemiological studies mainly concerning urban populations (Burnett et al., 1998; Hoek et al., 2000). Although the mechanisms are not fully explained from a medical point of view, epidemiological evidence suggests that ambient air pollution is a contributing cause of morbidity and mortality (Bates, 1992).

For assessing health risks related to air pollution, it is necessary to quantify the exposure of the population to the various hazardous substances released in the atmosphere. The key assumption in previous research on the topic has been that ambient concentrations of air pollutants can be used as an indicator of population exposure, despite the fact that people in European cities typically spend the majority of their time (up to 90%) indoors (Gonzalez-Flesca et al., 2000). Baek et al. (1997)

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PII: S1352-2310(01)00288-6

demonstrated experimentally the importance of ambient air quality in determining the quality of indoor air in two major Korean cities. Another field experiment conducted by Kingham et al. (2000) in the area of Huddersfield (England) suggested that outdoor pollution may give a useful measure of exposure to traffic-related pollutants as a part of epidemiological studies.

In most cases so far, the population exposure to air pollution has been assessed through crude assumptions. It has been assumed, for example, that concentrations observed at a single or a few permanent monitoring stations within a city are representative of the exposure of the entire urban population (Fenger, 1999). This is in line with current European legislation relevant to health protection. The proposed Council Directive related to limit values for benzene and carbon monoxide in ambient air (European Commission, 1998) specifies that only one fixed sampling point is enough for assessing compliance with limit values for the protection of human health in urban agglomerations with less than 250,000 of population. This practice is in contradiction with findings from current research, which show a significant small-scale spatial variability of traffic-related pollution in urban areas.

Nowadays, most large European cities are covered to a certain extent by air quality monitoring networks, which provide continuous measurements of key pollutants (e.g. NO_x, SO_2, CO). Nevertheless, a more detailed spatial profile of ambient concentrations is often needed for population exposure studies than that is usually available (WHO, 1999). This need is more pronounced in areas with high population density, strong emission sources, and limited natural ventilation (e.g. urban streets and avenues). For this reason, alternative sampling techniques, not entailing high cost and practical constraints (e.g. bulk of equipment, power supply requirements) of continuous air quality monitoring, should be tested. In addition, dispersion models can be used to provide concentration estimates in areas that are not sufficiently covered by measurements or to explore future emission and traffic scenarios.

The objective of this study is to propose a sound methodology for assessing air quality in urban micro-environments. For this purpose, different monitoring and modelling techniques were tested during a series of field experiments carried out in Paris and London (Jones et al. 1998, 2000; Vardoulakis et al., 2000). Two of these experiments were conducted at representative street canyon sites in Paris during winter (Bd. Voltaire, December 1998) and summer (Rue de Rennes, July 1999). The observed pollution levels were compared with values calculated using two different dispersion models.

2. Experimental methods

2.1. Description of the sites

Two street canyons, typical examples of the urban topography of Paris (i.e. busy four lane streets with large pavements and uniform buildings lining up continuously on both sides) were selected for the field measurements. In both the locations, the population exposure to traffic-related pollution was expected to be high.

The winter campaign was carried out in Bd. Voltaire, between Rue des Boulets and Rue de Montreuil junctions. The height-to-width (H/W) ratio for Bd. Voltaire was approximately equal to 0.8. Measurements were taken within a straight road segment of approximately 300 m. Traffic lights were operating at both ends of the canyon, and there was a pedestrian crossing at a distance of 34 m from the main sampling point. The street axis bearing from the north was 140°. The average traffic volume in Bd. Voltaire was 30,000 veh/d. Background measurements were also taken in an adjacent park location, at a distance of 100 m from the canyon.

The summer campaign was conducted in Rue de Rennes, between Rue d'Assas and Rue Coëtlogon junctions. The H/W ratio was approximately equal to 1.1. Traffic lights were operating at both ends of the selected road segment at a distance of at least 50 m from the monitoring unit, which was located at 37 m from a bus stop. The street axis bearing from the north was 32°, and the average traffic volume during measurements 23,000 veh/d. Two green areas located at a distance of approximately 300 m from the canyon in opposite directions were selected for background pollution measurements.

2.2. Sampling protocol

Parallel techniques, both active and passive, were used to sample a wide range of traffic-related atmospheric pollutants at different heights and distances from the kerb. Real time CO, NO_x, and O_3 monitoring was carried out on the eastern side of both the canyons throughout the respective campaigns. A main sampling line was established at the kerb with its inlet at 3.7 (Bd. Voltaire) and 2.9 m (Rue de Rennes) heights above the ground. Active (i.e. pumped) sampling of volatile organic compounds (VOC) and aldehydes was conducted through the same sampling line during part of the campaigns.

In Bd. Voltaire, passive (i.e. diffusive) VOC samplers were located at two different heights (first and fifth floors) near the walls of the canyon, and at one background site. The devices remained exposed to ambient concentrations for five days. In Rue de Rennes, two different sets of passive samplers were used to examine separately the VOC levels during the weekend and working weekdays. A more detailed spatial resolution of VOC concentrations was obtained by increasing the number of sampling locations. In this case, apart from measurements near the walls of the canyon, samples were also taken on the kerbside within the human breathing zone ($h = 1.5$ m), and at two different background sites. Passive aldehyde sampling was also carried out in Rue de Rennes during weekdays. Aldehyde samplers were placed within the pedestrian breathing zone and near the walls of the canyon at the first floor level. The exact locations of all passive samplers are indicated in Fig. 1.

Local meteorological parameters were measured at street level and compared with synoptic weather information obtained from a permanent monitoring station located in park Montsouris, within a few km distance from the experimental sites. Hourly traffic volume and average vehicle speed were obtained from automatic counters permanently operating in Bd. Voltaire and Rue de Rennes. The vehicle fleet composition was estimated from on-site spot measurements during the campaigns.

2.3. Sampling and analytical equipment

Several VOC compounds were sampled using active and passive means. Roadside air was pumped at a constant flow for several one hour intervals through Supelco glass tubes filled with Carbotrap-B. Radiello Perkin Elmer axial diffusive tubes filled with Carbotrap-B and sheltered in aluminium boxes were continuously exposed for two and five days. After removal from the

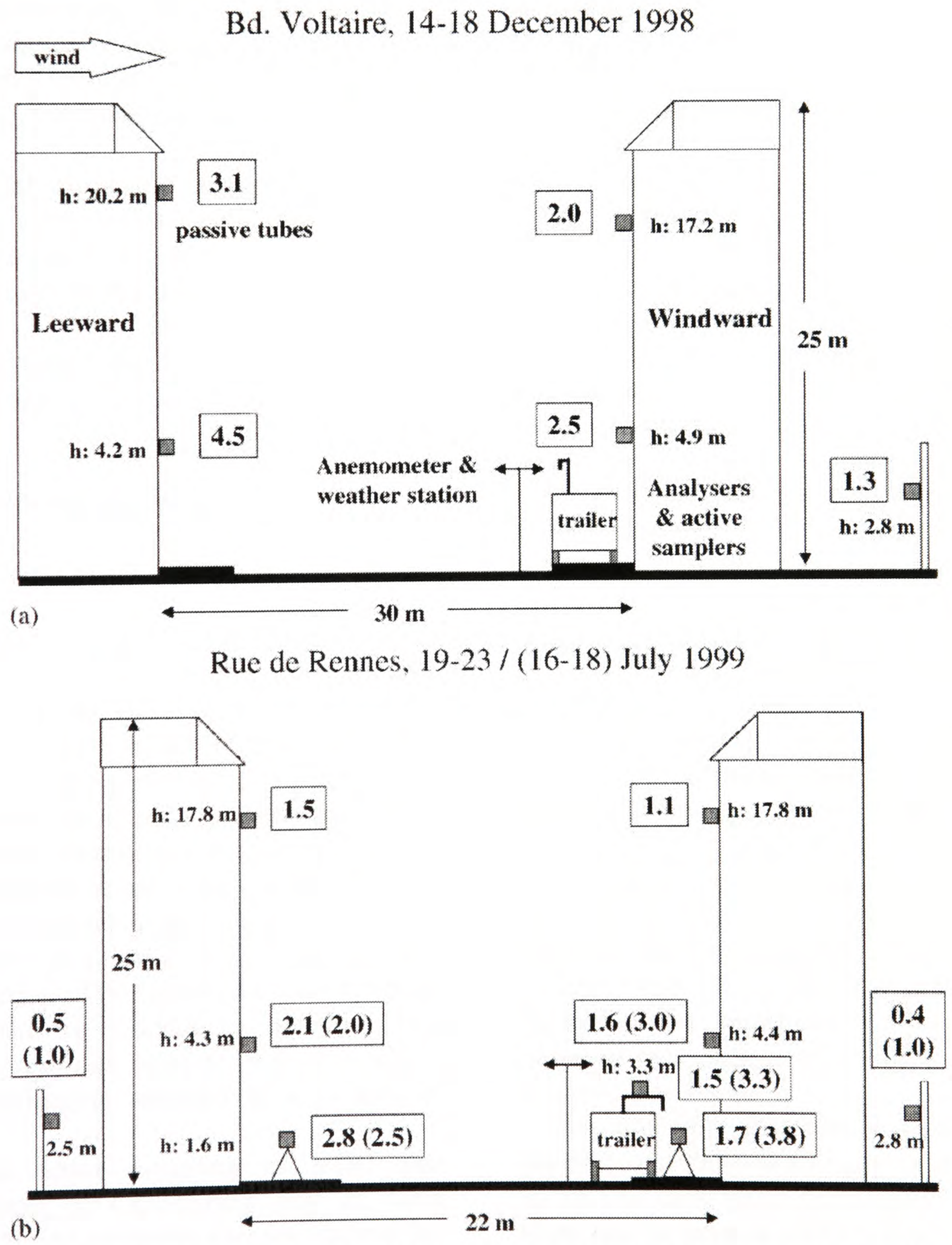

Fig. 1. Spatial variation of benzene (ppb) measured with passive sampling in: (a) Bd. Voltaire, and (b) Rue de Rennes during weekdays (19–23 July), and a weekend (16–18 July, values in parenthesis).

tubes with thermal desorption, VOC were analysed in the laboratory using gas chromatography (column type: CP-SIL 5CB, 50 m×0.32 mm, 1.2 μm) + FID.

Radiello samplers and Sep Pak DNPH-SILICA cartridges were, respectively, used for passive and active aldehyde measurements. The samples were solvent-desorbed and analysed in the laboratory using high performance liquid chromatography (column type: KROMASIL C18, 150 mm×3 mm, 3.5 μm) + UV detector.

A carbon monoxide infrared analyser (UNOR 610), a nitrogen oxides chemiluminescence analyser (Mégatec 42-C), and an ozone ultra-violet analyser (Environnement S.A., 03 41 M) were used to obtain roadside measurements every second. These devices, sheltered in an air-conditioned portable cabin (trailer), were interfaced with a STADUP data logger, through which data could be observed in real time and recorded as 4 min moving averages, before being further averaged for an hour. A quality assurance programme, including sampling duplicates, blanks and instrument calibration with standard gases was followed during sampling and analysis.

A three-dimensional ultrasonic anemometer (WindMaster, Gill Instruments) and a weather mini-station (AANDERAA) were used for measuring street-level wind speed and direction, temperature, relative humidity and global radiation. These instruments were attached to a mast (at approximately 4 m above ground) located on the kerbside near the trailer.

3. Experimental results and discussion

3.1. Relationship between pollutants

It is very helpful for epidemiological studies when a single indicator can be identified for air pollution, because this can then be used to indicate general levels of population exposure in urban areas (Kingham et al., 2000). Given the practical advantages and constraints of different air quality monitoring techniques, it may be more convenient to identify a set of possible pollution indicators, each one of them meeting a particular need. The chosen compounds should come from the same sources (e.g. road traffic) and have the same fate with the group of pollutants they are intended to represent. This can be checked by estimating the strength of correlation of any possible indicator with a number of other pollutants sampled in a variety of locations.

The simultaneous active sampling of VOC and continuous monitoring of CO during part of both the campaigns enabled to calculate the correlation between different compounds of interest (Table 1), and to establish empirical relationships between their concentrations on the kerbside. A very strong correlation between benzene, toluene, xylenes (BTX) and CO was established during both campaigns. A quite strong correlation between these compounds and other hydrocarbons (pentane, hexane, heptane, octane, ethylbenzene) was also observed. This is in agreement with findings from previous studies (Hansen and Palmgren, 1996; Giugliano et al., 2000) which have shown that CO, although a pure combustion product, correlates highly with several aromatic VOC, which are not only emitted through combustion but also through direct fuel evaporation.

Another observation that can be made is that formaldehyde correlates very strongly with CO and some of the VOC (e.g. benzene and toluene), while acetaldehyde shows a considerably weaker correlation with the same compounds (Table 1b). This may lead to the conclusion that formaldehyde is mainly of vehicular origin, coming directly from car exhausts or indirectly through the oxidation of unburned hydrocarbons. Acetaldehyde, on the other hand, comes from a variety of sources, including photochemical processes (Ferrari et al., 1998).

It is expected that the relationship between relatively stable chemical species coming from the same source would not vary significantly within urban environments. As far as street canyons are concerned, due to the very short distances between sources and receptors, only very fast chemical reactions have a significant influence on the measured concentrations (Berkowicz et al., 1997). For this reason, not only very stable gases like CO, but also some relatively more reactive ones like benzene and other VOC can be considered as practically inert within these distances. This is not the case either for NO_2, which dissociates extremely fast in the presence of light, or for NO, which also reacts very fast with O_3. The time scales of these chemical reactions are of the order of tens of seconds, thus comparable with residence times of pollutants in the street.

The very strong correlation between benzene and toluene concentrations measured with diffusive tubes at different sampling locations in Bd. Voltaire and Rue de Rennes (Fig. 2) suggests that these compounds, which come from the same source (i.e. petrol vehicles), do not take part significantly in chemical reactions within the canyons. The experimental toluene to benzene ratio (by volume) was 2.9 in Bd. Voltaire and 3.4 in Rue de Rennes. These values are in agreement with the ratio observed by Palmgren et al. (1999) in a street canyon in Copenhagen.

The negligible chemical reactivity corresponding to the diffusion times of CO and benzene in canyon streets as well as their good correlation suggest that both of them can be used as traffic pollution indicators. Due to their common origin and fate in urban environments, it is expected that a simple proportionality relationship

Table 1
Correlation coefficients of air pollutants measured with active sampling in: (a) Bd. Voltaire, and (b) Rue de Rennes

	Benzene	Toluene	*m*+*p*-Xylenes	Pentane	Hexane	Heptane	Octane	Ethylbenzene	*o*-Xylene
(a) Bd. Voltaire									
CO	0.93	0.90	0.83	0.71	0.75	0.91	0.79	0.77	0.72
Benzene		0.99	0.88	0.85	0.91	0.96	0.88	0.85	0.86
Toluene			0.90	0.87	0.94	0.97	0.88	0.88	0.88
m+*p*-Xylenes				0.78	0.90	0.87	0.87	0.99	0.92
Pentane					0.83	0.85	0.68	0.75	0.72
Hexane						0.87	0.82	0.90	0.92
Heptane							0.87	0.84	0.84
Octane								0.89	0.89
Ethylbenzene									0.93

(b) Rue de Rennes

	Benzene	Toluene	Ethylbenzene	*m*+*p*-Xylene	Styrene	*o*-Xylene	Formaldehyde	Acetaldehyde
CO	0.94	0.89	0.61	0.97	0.49	0.67	0.93	0.69
Benzene		0.91	0.40	0.98	0.22	0.49	0.95	0.70
Toluene			0.62	0.98	0.37	0.72	0.92	0.58
Ethylbenzene				0.98	0.92	0.98	0.81	0.46
m+*p*-Xylene					0.82	0.99	0.65	0.21
Styrene						0.87	0.75	0.44
o-Xylene							0.81	0.46
Formaldehyde								0.83

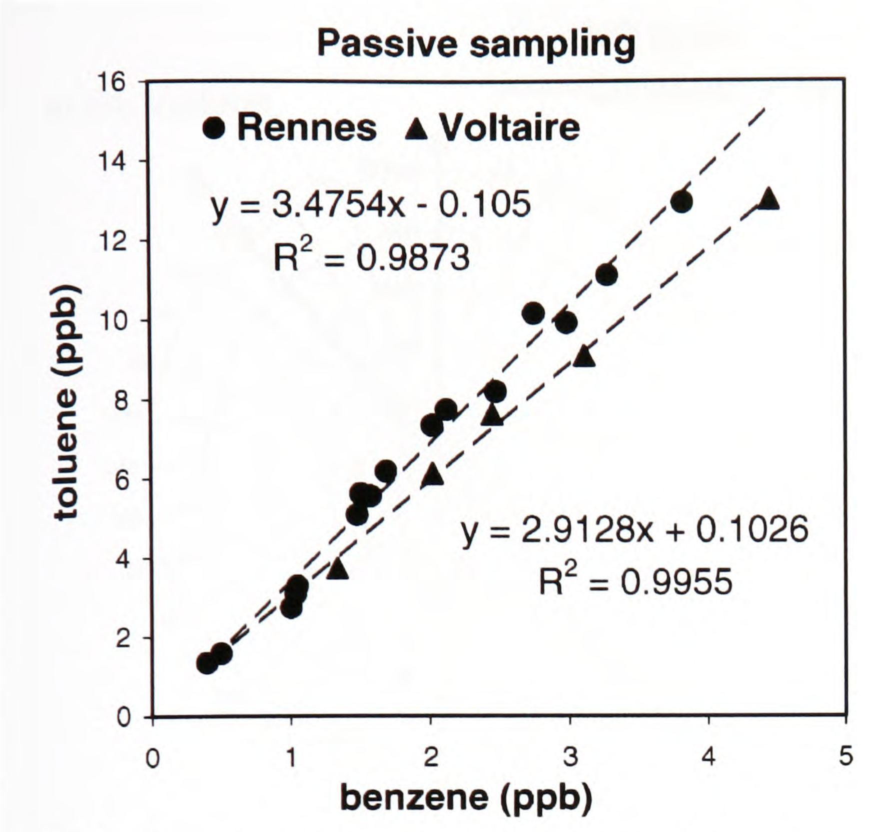

Fig. 2. Average toluene (ppb) vs. benzene (ppb) concentrations measured with passive sampling in Bd. Voltaire and Rue de Rennes.

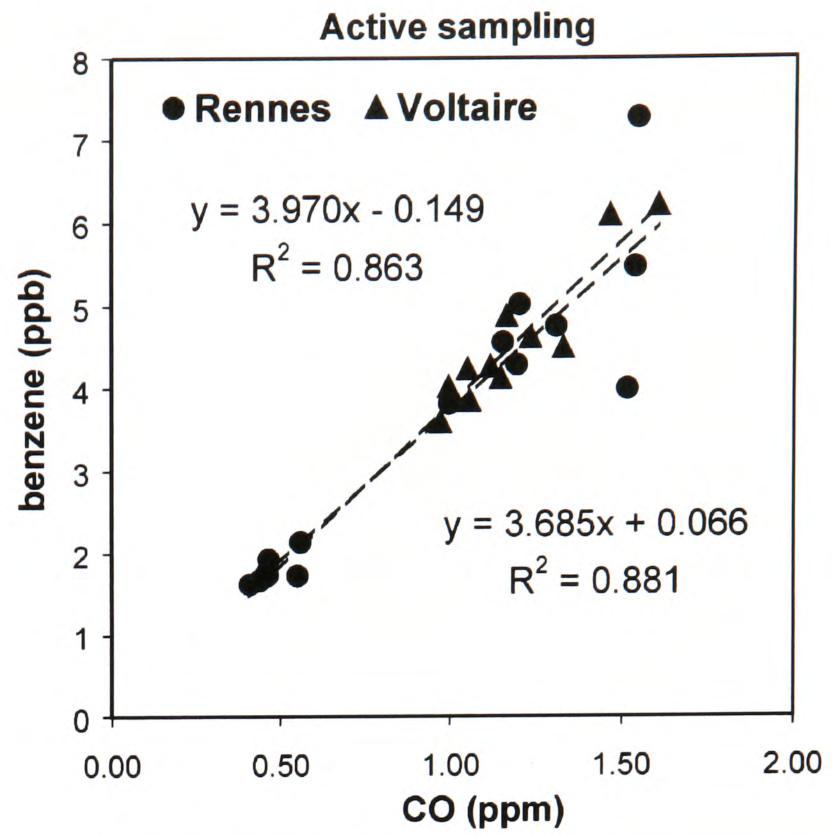

Fig. 3. Average benzene (ppb) vs. CO (ppm) concentrations measured with active sampling and real time monitoring, respectively, in Bd. Voltaire and Rue de Rennes.

can be established between them:

$$\text{Benzene(ppb)} \approx \alpha\, \text{CO(ppm)}, \qquad (1)$$

where $\alpha = 3.8$ for Bd. Voltaire and 3.7 for Rue de Rennes (Fig. 3). These values agree with results reported by Jones et al. (2000) after a field experiment in Paris, and Pfeffer et al. (1995) after measurements in two German cities. In addition, they are within the range of values observed by Palmgren et al. (1999).

The CO to benzene ratio is expected to remain roughly the same in urban environments as far as there are no significant changes in vehicle and fuel technology, fleet composition, traffic patterns, or ambient temperature. Therefore, the relationship (1) may be generally applied to estimate CO concentrations using benzene measurements in urban areas and vice versa.

3.2. Spatial variability

In several field experiments (Laxen and Noordally, 1987; Hewitt, 1991; Monn et al., 1997), diffusive NO_2 sampling has been used for establishing the spatial variability of air pollution in urban areas. A criticism of this might be that this compound, although easily monitored using passive tubes, is not the best indicator for traffic generated pollution. This is because NO_2 only represents a small fraction (less than 10%) of the total NO_x directly emitted from traffic. In addition to this, it is highly reactive within very short transport distances and therefore is not expected to correlate strongly with other more conservative pollutants like CO and benzene.

Croxford et al. (1996) used CO as a tracer to indicate large differences in air pollution levels between neighbouring streets in central London. Gonzalez-Flesca et al. (1999) used diffusive benzene and aldehyde sampling to identify strong concentration gradients within short distances in a medium size French town.

In the two field experiments in Paris, diffusive VOC samplers were deployed to reveal the spatial variability of traffic pollution within the streets and in relation with urban background levels. Using BTX as indicators, strong concentration gradients were identified in the horizontal and vertical sense within the canyons. The highest benzene concentrations were detected at street level, on the side of the canyon that was upwind (i.e. leeward) most of the time (Fig. 1). At the leeward sampling locations, weekly benzene averages were from 30% to 80% higher than the values measured on the side of the canyon that was most of the time facing the wind (i.e. windward).

The formation of pollution hot spots on the leeward side of the street, which is in agreement with previous field observations (Qin and Kot, 1993), may be explained by the creation of an air vortex within the canyon. This single vortex appears in a regular canyon of H/W ratio approximately equal to one, when synoptic (i.e. above roof-level) winds greater than 2 m/s blow at an angle of more than 30° to the street axes (Oke, 1988). During part of the measurements in Bd. Voltaire and Rue de Rennes, a vortex regime was established within the canyon. This was identified using the wind information obtained at street level. For perpendicular

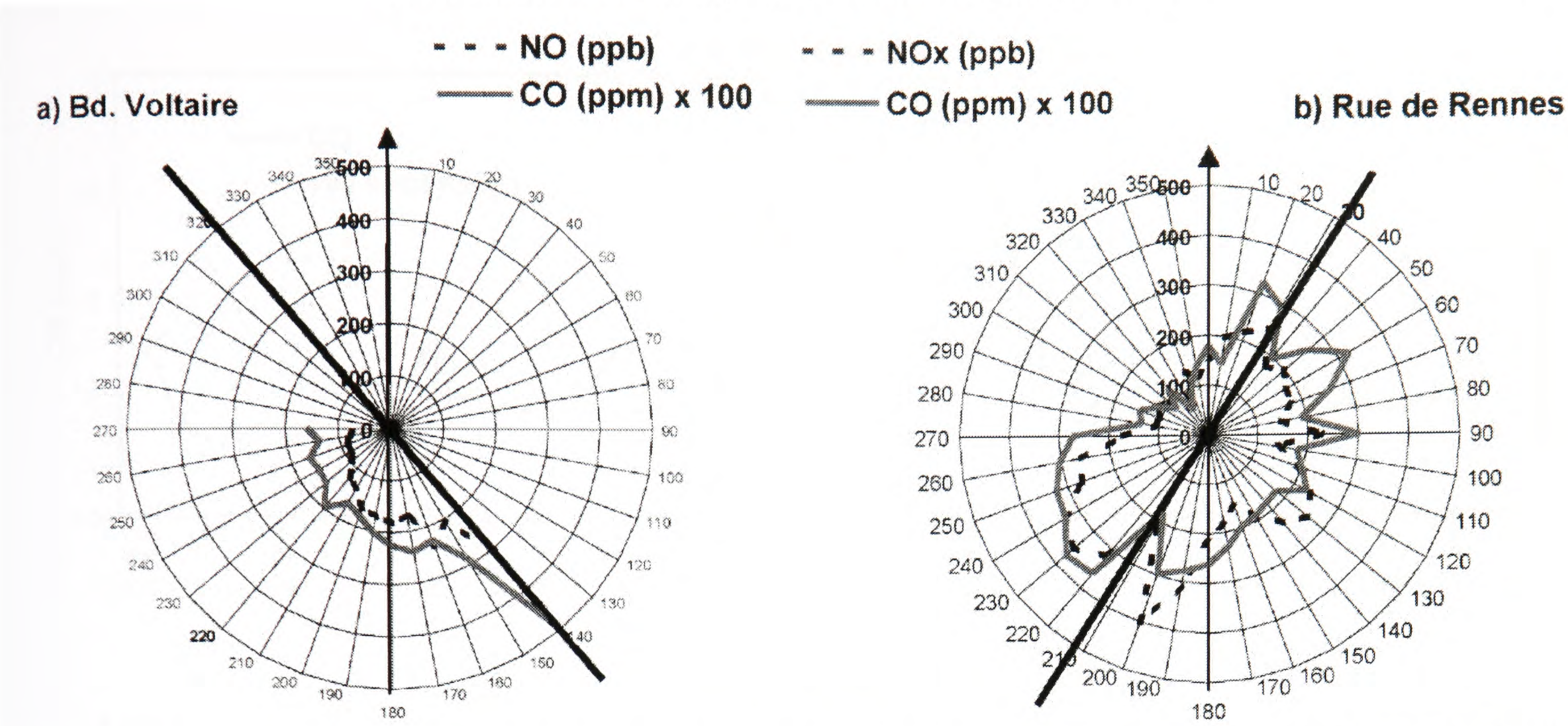

Fig. 4. Pollution roses: (a) Normalised CO and NO concentrations in Bd. Voltaire, (b) Normalised CO and NO_x concentrations in Rue de Rennes. (The heavy straight line indicates the direction of the street).

or near-perpendicular synoptic winds, a downward airflow was observed on the windward side of the street and an upward flow on the leeward side. In addition, an elastic-type reflection of the wind off the windward wall of the canyon was detected for synoptic winds greater than 2 m/s.

The influence of the synoptic wind direction on the dispersion of pollutants in the two canyons is illustrated on the pollution roses plotted for CO and NO_x (Fig. 4). Hourly mean CO and NO_x concentrations, normalised with respect to the wind speed and traffic volume, were assigned to the corresponding synoptic wind directions. Then, the arithmetic mean of the concentrations was calculated for each of the 36 equal wind direction sectors. Both roses demonstrated a clear dependence of pollution levels on the synoptic wind direction. It can be seen that, keeping the other factors constant, winds parallel or near-parallel to the street axis favour pollution built-up on the kerbside, while perpendicular winds provide better dispersion conditions. This is because for relatively long canyons without connecting streets, the accumulation of emissions along the line source outweighs the ventilation induced by the parallel winds (Soulhac et al., 1999; Dabberdt and Hoydysh, 1991). Furthermore, in Fig. 4b it can be observed that the normalised concentrations in Rue de Rennes were higher at least by a factor of 2 for winds blowing from SE (i.e. leeward flow) than for winds coming from NW (i.e. windward flow), confirming thus the vortex formation hypothesis for the perpendicular wind regime.

A substantial reduction in ambient benzene concentrations along with the height above the street was also observed. The weekly benzene averages measured on fifth floor balconies were from 20% to 30% lower than at the first floor level. Nevertheless, this variation was smaller than the vertical CO gradients observed in a busy street canyon in Athens, as reported by Zoumakis (1995).

Finally, benzene concentrations measured on the kerbside were two to six times higher than those detected in the selected background locations. Even the values detected at 17–20 m height near the walls of buildings facing the street were significantly higher than the background levels. Attention should be drawn to the fact that the observed roadside levels for benzene (averaged over two to five days) exceeded the proposed EU limit value of 5 $\mu g/m^3$ (1.6 ppb) for ambient air, although background concentrations remained in all cases below the same threshold.

3.3. Temporal variability

While benzene measurements described the spatial variability of pollution, CO was used as an indicator of the temporal variability of inert compounds. As expected, the time profile of roadside CO was affected by the variation of the synoptic wind speed and the traffic flow in the street. In Fig. 5, it can be seen that daily maximum concentrations corresponded roughly to the morning and evening rush hours as well as to the lowest recorded winds.

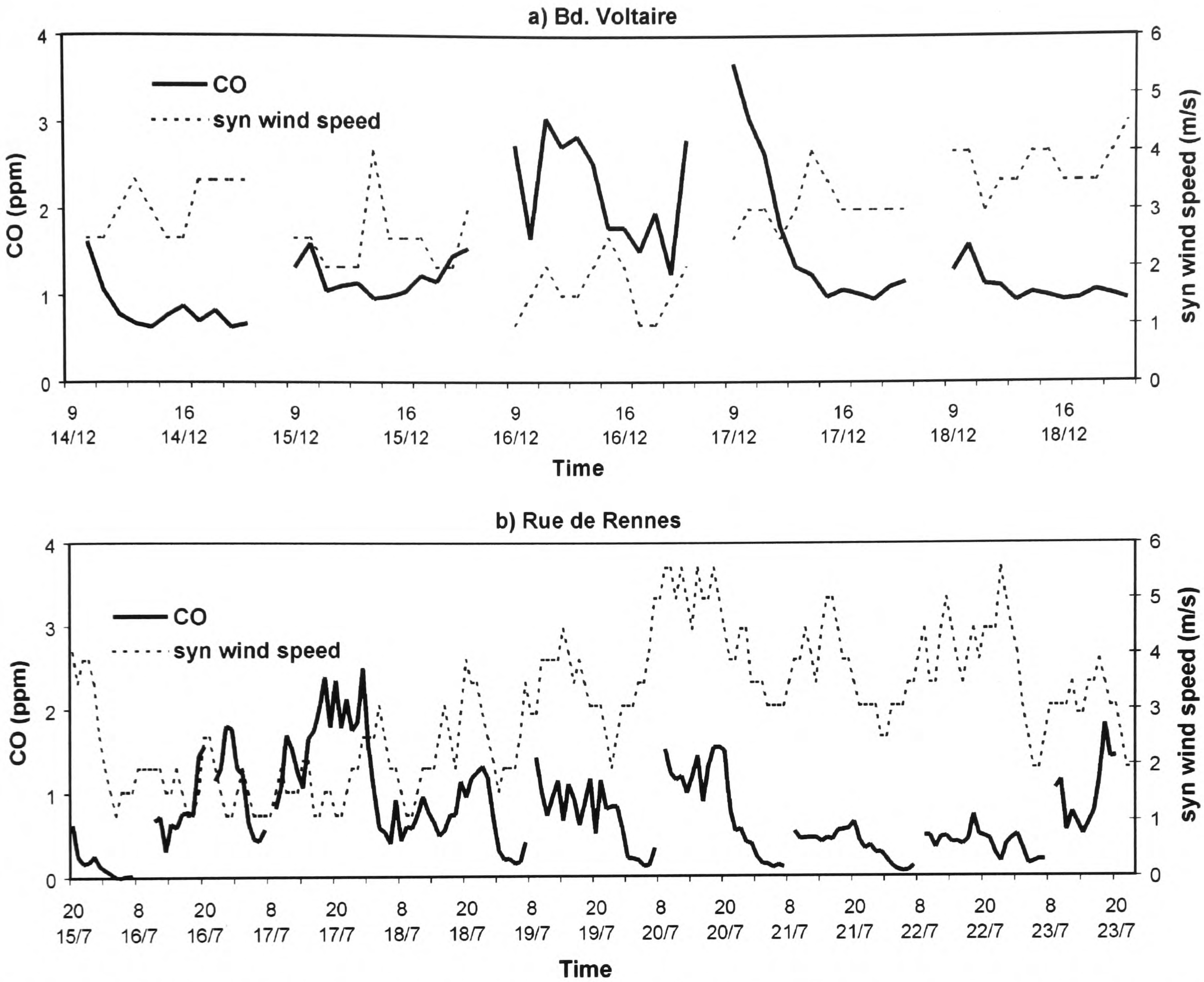

Fig. 5. Hourly mean CO (ppm) concentrations and synoptic wind speeds during measurements in: (a) Bd. Voltaire, and (b) Rue de Rennes.

Two peaks of CO were observed in Bd. Voltaire during morning hours (16th and 17th of December; Fig. 5a). The first one can be explained by the presence of low wind conditions ($\leqslant 2.5\,m/s$) in the region. The second CO peak may be attributed both to the relative low wind speed (2.5–3.0 m/s) and to the parallel wind direction.

Interestingly, the highest CO concentrations in Rue de Rennes were detected on a Saturday (17 July; Fig. 5b), when traffic volume was slightly smaller than that during weekdays. Again, this episode can be mainly attributed to the low winds (1–2 m/s) blowing over Paris on that day. During the following week (19–23 July) winds became stronger (2.5–5.5 m/s) and as a result, concentrations were reduced. CO levels were consistent with passive VOC measurements, which were also higher during the weekend than those during the rest of this campaign.

Moderate photochemical activity was observed in Rue de Rennes. NO concentrations peaked during the morning rush hours (Fig. 6). The NO_2 peak levels were delayed a few hours, as expected due to the time required to oxidise NO. Ozone gradually increased during the day, producing higher concentrations in the afternoon. The relatively low winds and the strong insolation on Sunday (18th), in combination with low NO emissions due to reduced road traffic, gave rise to a minor ozone episode on that day with values reaching 50 ppb in early afternoon. On another occasion (20–21 July), relatively high ozone levels were observed during late evening and night. This might be explained by long-distance transport of pollutants. It is not unusual for large quantities of ozone formed in rural areas during daytime to be advected over long distances, reaching urban centres during late afternoon and evening. During the winter campaign in Bd. Voltaire, O_3 levels were very

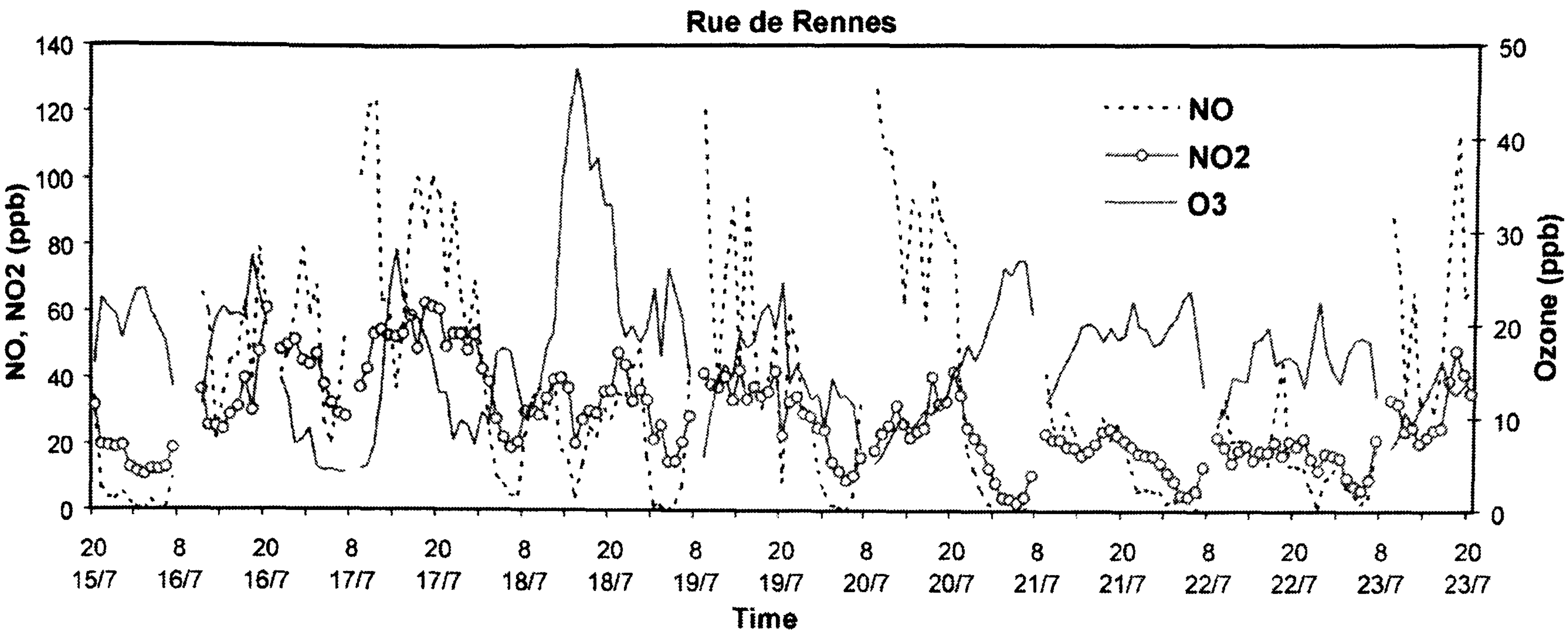

Fig. 6. Hourly mean NO, NO_2 and O_3 (ppb) concentrations observed in Rue de Rennes.

low at all times, due to negligible photochemical activity in the region.

Comparing measurements with proposed EU limit values for ambient air (Table 2), it was observed that the NO_2 averages over the campaigns exceeded the annual threshold of $40\,\mu g/m^3$ (21 ppb). Although the averaging times were different, these exceedances indicate that the air quality objective for NO_2 might not be met in the long run. By contrast, the hourly mean NO_2 concentrations remained always below the short-term limit value of $200\,\mu g/m^3$ (104.6 ppb). Finally, the ambient CO and O_3 levels detected during both campaigns were much lower than the limit values of $10\,mg/m^3$(8.5 ppm) and $110\,\mu g/m^3$ (55 ppm), respectively.

4. Modelling methods

4.1. Model formulation and requirements

In this study, two relatively simple models were proposed for the simulation of pollutant dispersion within canyons. Computational fluid dynamic codes were avoided, due to the great amount of input information required and the practical constraints related to their use (computational time, software/hardware requirements, etc.). It should be stressed that the accuracy of air quality models is bounded by the accuracy of input data such as vehicle emission factors, local traffic and meteorological data, the geometry of the site, etc., which are rarely available in great detail for routine applications. This is why a semi-empirical modelling approach was adopted here.

An empirical model (STREET) developed by Johnson et al. (1973) was used to calculate series of hourly CO concentrations at several receptor locations in Bd. Voltaire and Rue de Rennes. This model is based on the assumption that the concentrations (C) of the pollutant occurring on the kerbside consist of two components, namely the urban background concentration (C_b) and the concentration component (C_s) due to vehicle emissions generated within the street:

$$C = C_s + C_b. \tag{2}$$

The C_s component has been derived from a simple box model. The concentrations on the leeward side of the street are given by the following expression:

$$C_s = K\frac{Q}{\left(\sqrt{x^2+z^2}+2\right)(U+0.5)}, \tag{3}$$

where K is an empirical constant parameter given the value 7, Q the rate of release of emissions in the street (expressed in mg/m s), U the roof-level wind speed (m/s), z the height (m) of the receptor and x the horizontal distance (m) between the receptor and the centre of the nearest traffic lane. On the windward side, the original expression given by Johnson et al. (1973) was revised by Dabberdt et al. (1973) to account for vertical decrease of concentrations due to entrainment of fresh air through the top of the canyon. The final equation was:

$$C_s = K\frac{Q}{W(U+0.5)}\frac{H-z}{H}, \tag{4}$$

where H is the height (m) and W the width (m) of the canyon. For parallel or near-parallel synoptic winds, the average of Eqs. (3) and (4) was calculated. This model has been widely used for scientific and engineering applications (Qin and Kot, 1993; Hoydysh and Dabberdt,1988).

The operational street pollution model (OSPM), developed by Hertel and Berkowicz, was also used in

Table 2
Measured pollutant concentrations against regulatory limit values. The statistical quantities for O_3, CO and NO_2 were calculated using real time kerbside measurements. The min and max benzene concentrations correspond to passive sampling measurements at different locations. The campaign benzene averages were calculated over the available street-level observations

	O_3			CO			NO_2			Benzene		
Unit	ppb			ppm			ppb			ppb		
Averaging time	8 h		Campaign average	8 h		Campaign average	1 h		Campaign average		Campaign average	
	Median	Max		Median	Max		Median	98th Percentile		Min		Max
Bd. Voltaire	5.49	6.91	5.51	1.14	2.39	1.42	27.89	48.37	30.94	1.34	3.50	4.46
Rue de Rennes	16.68	28.56	16.74	0.67	2.00	0.74	25.06	58.17	27.63	0.40	2.43	3.80
EU limit value	55 (110 μg/m^3)[a]			8.5 (10 mg/m^3)			104.6 (200 μg/m^3)		21 (40 μg/m^3)[b]		1.6 (5 μg/m^3)[b]	

[a] French limit value for the protection of human health.
[b] Annual mean.

this study. This semi-empirical model calculates concentrations of exhaust gases assuming three different contributions to the kerbside levels: the contribution (C_d) from the direct flow of pollutants from the source to the receptor, the recirculation component (C_r) due to the flow of pollutants around an horizontal wind vortex generated within the so-called recirculation zone of the canyon, and the urban background contribution (C_b):

$$C = C_d + C_r + C_b. \quad (5)$$

The direct component is calculated by applying Gaussian plume dispersion theory, while the recirculation component is given by a box model algorithm. On the leeward side of the street, concentrations are calculated as the sum of the direct and recirculation contributions, while on the windward side, only the direct contribution of emissions generated outside the recirculation zone are taken into account. If the recirculation zone extends throughout the whole canyon, then the windward concentrations are calculated from only the recirculation component. When the wind speed is near zero or parallel to the street axis, the concentrations on both sides of the canyon become equal. In all the cases, the background contribution should be added to obtain the final results. A detailed description of this model is given elsewhere (Berkowicz et al., 1997; Berkowicz, 2000). OSPM has been successfully applied to several street canyons in Copenhagen, Utrecht, Oslo, Helsinki, and to a wide street canyon in Beijing (Berkowicz et al., 1996; Kukkonen et al., 2001; Fu et al., 2000).

4.2. Modelling results

Eqs. (3) and (4) were used to simulate CO concentrations in Bd. Voltaire and Rue de Rennes. The rate of release of emissions Q in the street was calculated from the traffic volume and the composite emission factors for each pollutant. Emission factors were derived from the vehicle fleet composition specific to the site of interest, following a methodology described by Buckland and Middleton (1999). The estimated values were compared for consistency with emission factors specific to the French vehicle fleet reported in other recent studies (Touaty and Bonsang, 2000; Jones et al., 2000). The model background input for CO was estimated using relationship (1). The relative contributions from the street and the background were derived from benzene measurements.

The hourly averages derived from the model (STREET) were compared with concentrations measured on the kerbside. Although the model reproduces, reasonably well, the diurnal variation pattern of CO, in some cases, it appeared to underestimate the observed concentrations. In Fig. 7, the calculated concentrations for Rue de Rennes were stratified into three different wind regimes, where it can be reasonably argued that the physical processes affecting dispersion were similar. This

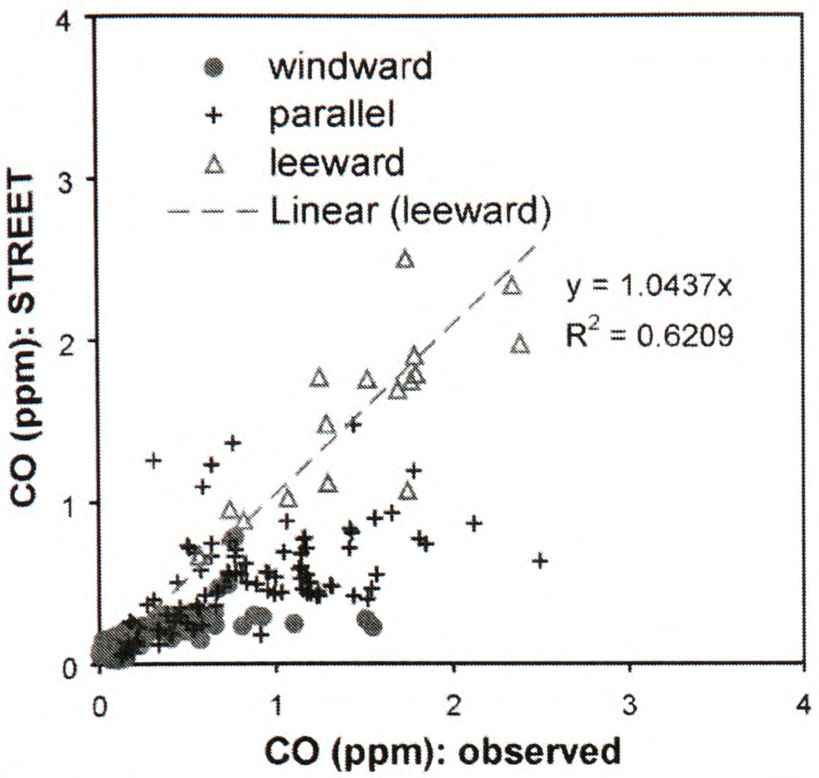

Fig. 7. STREET predictions vs. hourly mean CO (ppm) concentrations measured at street level in Rue de Rennes for three different wind regimes.

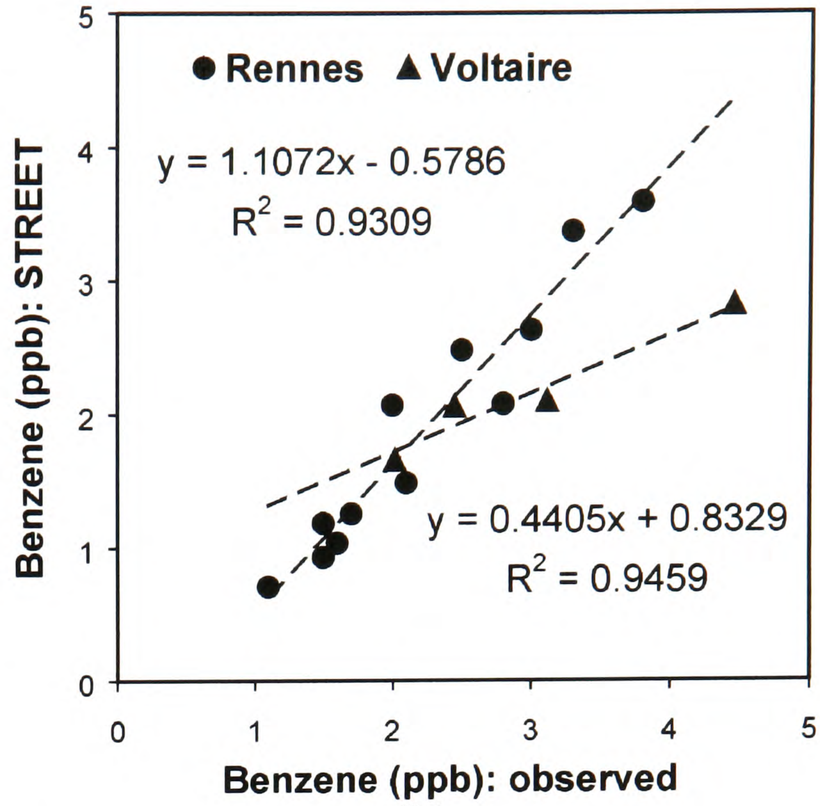

Fig. 8. STREET predictions vs. 2–5 days mean benzene (ppb) concentrations obtained with passive sampling in Bd. Voltaire and Rue de Rennes.

analysis revealed a good linear agreement ($r = 0.79$) between observations and predictions on the leeward side of the canyon when the wind blows perpendicular to the street axis. On the other hand, the performance of the model was not satisfactory for the windward side of the street when the wind is perpendicular, and for both sides when the wind is parallel. In these cases, the model under-predicted the ambient concentrations.

Using the empirical relationship (1), average benzene values were calculated for different locations in the streets over the passive sampling periods (two to five day averages) and added to the observed background concentrations. The comparison of the total calculated values with the street measurements showed (Fig. 8) a very good general agreement for Rue de Rennes, although the model seemed to slightly under-predict the low concentrations observed near the top of the canyon. For Bd. Voltaire, the model under-predicted all measured values.

Using the same input data set, OSPM reproduced, quite successfully, the hourly mean CO concentrations at street level. Comparing separately the model output for different wind regimes (Fig. 9), a good linear agreement ($r = 0.79$) was again established between predicted and measured values on the leeward side of the road. Large scatter and under-predictions were observed for parallel and windward flow, respectively.

4.3. Vertical profiles

OSPM calculates pollutant concentrations on both sides of the canyon, at the effective height of release of gases in the street (≈ 2 m), without giving the user the possibility of choosing the height of the receptors. However, pollution levels at different vertical distances from the road may be important from a population exposure point of view. For this reason, an external algorithm that enables the user to establish vertical pollution profiles should be introduced.

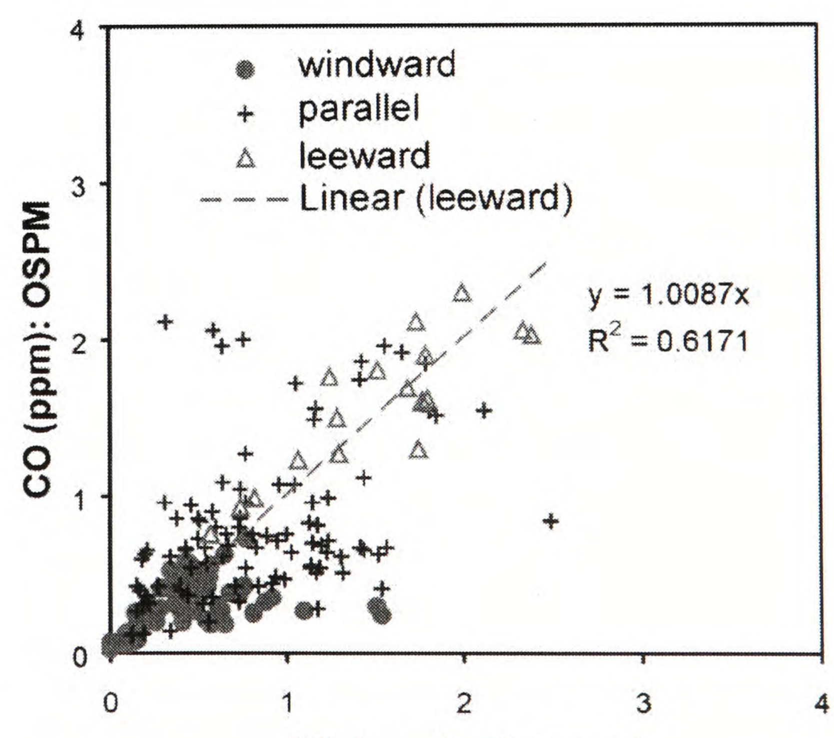

Fig. 9. OSPM predictions vs. hourly mean CO (ppm) concentrations measured at street level in Rue de Rennes for three different wind regimes.

It has been suggested by several authors in the past (Capannelli et al., 1977; Huang, 1979; Dabberdt and Hoydysh, 1991; Zoumakis, 1995) that the vertical concentration profile $C(z)$ in the street satisfies a law of exponential reduction with height (z):

$$C(z) \approx A\exp\left[-B\left(\frac{z}{H}\right)^q\right], \tag{6}$$

where A, B, and q are regression coefficients. Although empirically defined, the coefficients A, B and q are generally dependent on the wind direction, atmospheric stability and the aerodynamic characteristics of the canyon (Zoumakis, 2000). According to Georgii (1969), and Hoydysh and Dabberdt (1988), the vertical profiles may be reasonably well approximated by the simple exponential function, where $q = 1$. Zoumakis (2000) proposed for the q values ranging from 1 to 4.5, and for the B values from 1.1 to 2.9. Moreover, he identified a significant dependence of q on B. Sacré et al. (1995) suggested that the coefficient B varies in a way that can be empirically described by the relationship:

$$B = 1.6\cos\Phi + 0.4, \tag{7}$$

where Φ is the angle between roof-top wind and street axis.

From Eq. (6), a general expression relating pollutant concentrations at two different heights in the street can be deduced:

$$C(z) \approx C_r\exp\left(-B\frac{z^q - z_r^q}{H^q}\right), \tag{8}$$

where C_r is the concentration of the pollutant at a reference height z_r on either side of the canyon.

This function was used to calculate CO concentrations at receptor heights corresponding to the passive sampling locations in Bd. Voltaire and Rue de Rennes, using the street level output of OSPM as reference value. Applying relationship (1), average benzene values were obtained and added to the observed background concentrations. The comparison of the total calculated values with the concentrations observed in the street showed that the simple exponential expression ($q = 1$ and $B = 1$) gave the best agreement between predictions and measurements. The application of Eq. (7) for the calculation of B as a function of Φ did not significantly improve predictions. Thus, the following simple exponential expression is proposed for assessing vertical concentration profiles in street canyons:

$$C(z) \approx C_r\exp\left(-\frac{z - z_r}{H}\right). \tag{9}$$

Despite some under-predictions in the case of Bd. Voltaire, expression (9) reproduced satisfactory the benzene profiles detected in the two canyons (Fig. 10). However, it should be emphasised that this relationship is not applicable to traffic-related substances with very short chemical lifetime (e.g. NO, NO_2). It has been experimentally demonstrated (Väkevä et al., 1999) that NO_2 concentrations may even increase with height within a street canyon, when the weather conditions favour photochemical activity. Finally, it should be stressed that further validation against experimental data will be required before expression (9) may be of practical use.

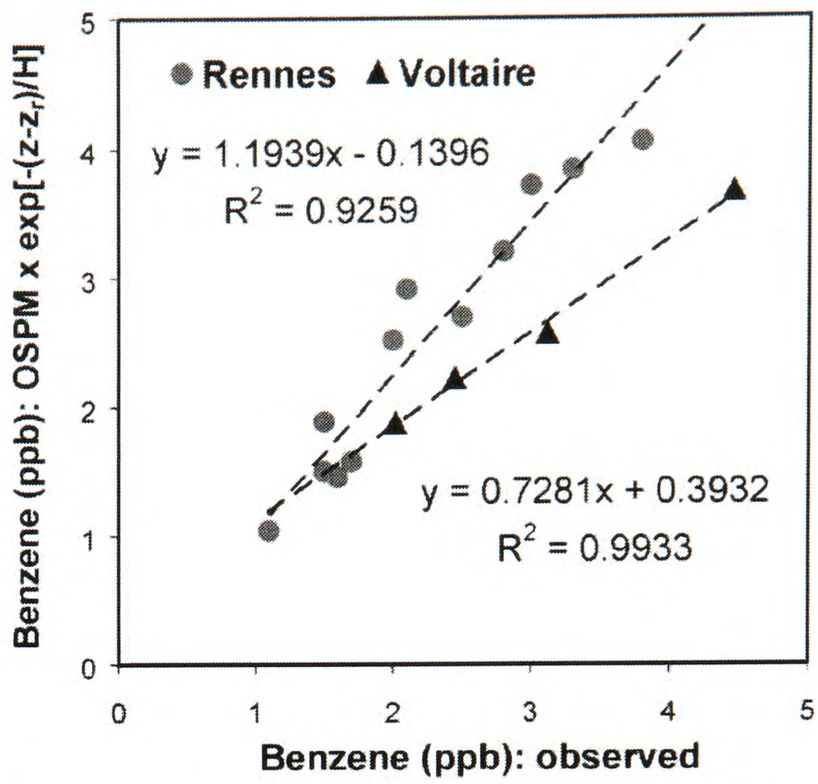

Fig. 10. OSPM predictions $\times \exp[-(z-z_r)/H]$ vs. 2–5 days mean benzene (ppb) concentrations obtained with passive sampling in Bd. Voltaire and Rue de Rennes.

5. Implications for population exposure

Two main elements which should be considered when exposure information is required for health impact assessment are the representativeness of the available environmental data for the population at risk, and the averaging time appropriate for creating a link with human health (Krzyzanowski, 1997). Traffic-generated pollutants are responsible for both acute and chronic effects on human health. For this reason, they are regulated with respect to different exposure times depending on their properties. In national and EU air quality guidelines, limit values are set for benzene as one year averages, for CO as eight hour averages, and for NO_2 as both one hour and one year averages (Table 2). Air quality monitoring and modelling methods should correspond to these averaging times.

Both the models used to simulate concentrations in Bd. Voltaire and Rue de Rennes gave acceptable 2–5 day average predictions. Nevertheless, the same method might not always give satisfactory short-term results. As an example, 8-h mean CO estimates were produced using OSPM and STREET and compared with

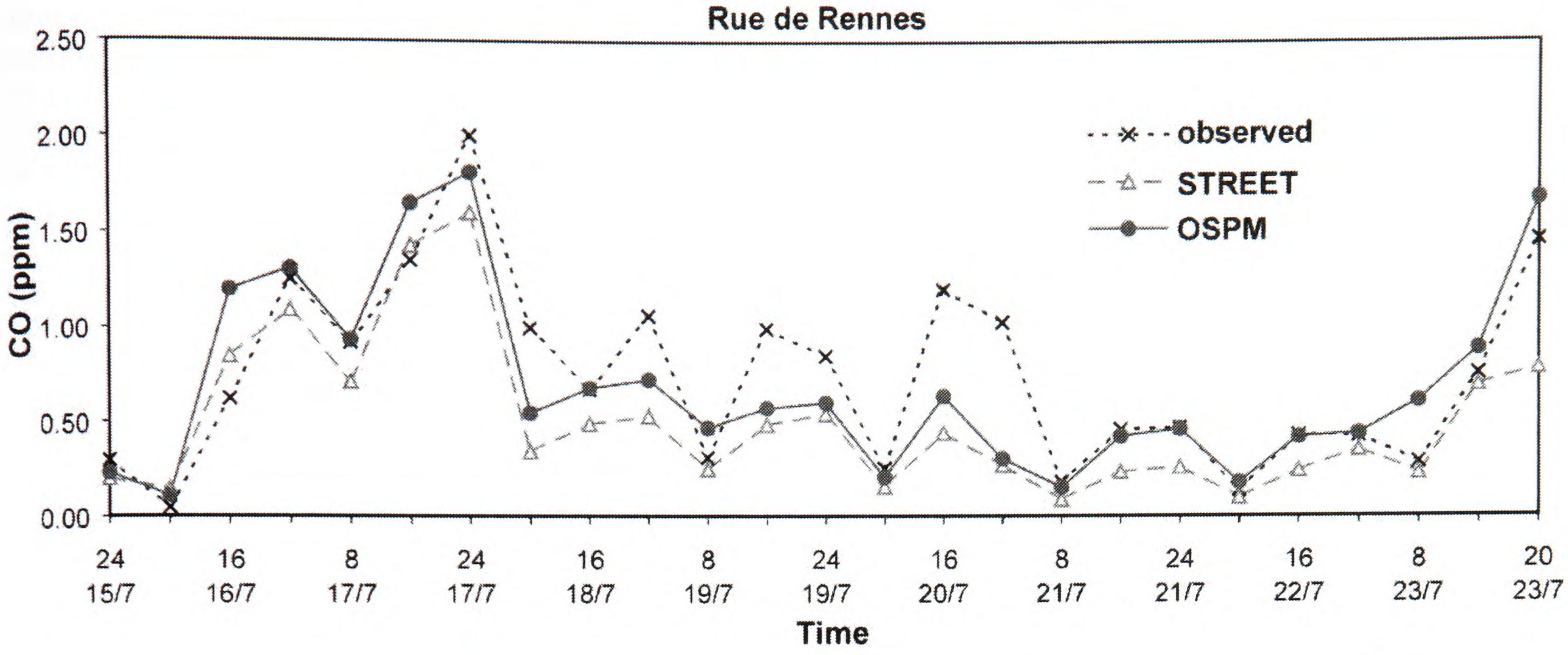

Fig. 11. STREET and OSPM predictions, and 8-h CO (ppm) averages observed at street level in Rue de Rennes.

concentrations measured at street level in Rue de Rennes (Fig. 11). It can be seen that although both the models reproduced the general variation pattern of pollution reasonably well, they led to certain number of under-predictions. These might be related to the unsatisfactory representation of the concentration field under parallel and windward flow regimes. For this reason, modelling results concerning current pollution levels should be validated with at least a small number of on-site measurements (e.g. passive sampling).

Although the spatial inhomogeneity of air pollution in urban streets has been quite early raised as an issue (Capannelli et al., 1977), it has been seldom taken into consideration. Given the fact that a large number of residents (approximately half a million people in Paris) live in the close proximity of urban streets, the strong cross-road and vertical concentration gradients observed may have serious implications in terms of total population exposure.

In a study attempting to quantify residential exposure to vehicle exhaust gases in Oslo (Larssen et al., 1993), a correction coefficient was introduced to account for changes in ambient CO concentrations with height over street level. In that case, the coefficient was arbitrarily given the value 1 for the basement, the ground and the first floor of the building facing the street, 0.5 for the second and third floors, and 0.25 for any level above the third floor. Using instead Eq. (9), a correction factor of 0.7 was estimated for the second floor, 0.6 for the third floor and so on, giving thus a more physically realistic representation of the vertical pollution profile on the facade of the building.

The present study added evidence to the hypothesis that people living on the leeward side of urban canyons which are perpendicular or near-perpendicular to the prevailing wind direction may be exposed to higher air pollution levels than those living on the windward side. As already suggested by other authors (Croxford and Penn, 1998), the side of the street factor should be taken into account in studies trying to link traffic-related air pollution with public health impact.

6. Conclusions

Two comprehensive air quality data sets were created after intensive monitoring in two street canyons in Paris. Using BTX as indicators, strong spatial concentration gradients were identified in both the sampling sites. The receptors on the leeward side of the buildings were exposed to substantially higher concentrations than those on the windward side, due to the helical circulation of the air within the canyon under perpendicular or near-perpendicular synoptic winds. A substantial reduction of pollution levels along with heights above the ground was also observed.

Given the strong spatial variability of pollution levels, the placement of roadside air quality monitors becomes crucial both in scientific experiments and routine measurements. Furthermore, there is evidence that human exposure to traffic pollution should be assessed not only as a function of residence distance from the road, but also as a function of side of the street and height above ground.

Roadside CO was used as an indicator of the short-term temporal variability of traffic-related pollution. BTX compounds and CO were highly correlated during both experiments, confirming that road traffic can be considered as their dominant source near streets. The benzene to toluene ratio remained almost constant at all sampling locations, which is in agreement with the chemical lifetime and the residence time of these substances in the street. A simple proportionality relationship was empirically established between CO and benzene. This is particularly helpful because it allows to calculate benzene concentrations in the street from CO measurements or predictions, avoiding thus the use of uncertain benzene emission factors.

Weekly benzene averages were closely reproduced by OSPM and reasonably well reproduced by STREET at different receptor locations. A simple exponential function was proposed for assessing vertical concentration profiles in street canyons. The performance of both the models was less satisfactory when predicting short term (1–8 h) CO averages under windward and parallel wind regimes. Given the limitations and uncertainties associated with dispersion modelling, this should be very cautiously used in decision making, especially when field measurements are not available.

The present study proposes the use of appropriate parameterised models in association with a limited number of passive BTX measurements for establishing pollution levels and concentration gradients within urban streets. The necessary meteorological and real time CO, NO_x, and O_3 data can be obtained from a limited number of monitoring stations. After further validation, this method may be adopted to provide information required for population exposure studies or to identify possible exceedences of regulated pollutants in urban areas.

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Model sensitivity and uncertainty analysis using roadside air quality measurements

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Abstract

Most of the air quality modelling work has been so far oriented towards deterministic simulations of ambient pollutant concentrations. This traditional approach, which is based on the use of one selected model and one data set of discrete input values, does not reflect the uncertainties due to errors in model formulation and input data. Given the complexities of urban environments and the inherent limitations of mathematical modelling, it is unlikely that a single model based on routinely available meteorological and emission data will give satisfactory short-term predictions.

In this study, different methods involving the use of more than one dispersion model, in association with different emission simulation methodologies and meteorological data sets, were explored for predicting best CO and benzene estimates, and related confidence bounds. The different approaches were tested using experimental data obtained during intensive monitoring campaigns in busy street canyons in Paris, France. Three relative simple dispersion models (STREET, OSPM and AEOLIUS) that are likely to be used for regulatory purposes were selected for this application. A sensitivity analysis was conducted in order to identify internal model parameters that might significantly affect results. Finally, a probabilistic methodology for assessing urban air quality was proposed.

Keywords: Air pollution; Model sensitivity; Uncertainty; Street canyon; Traffic emissions; Meteorological data

1. Introduction

Mathematical modelling has been widely used for assessing ambient air quality. Government departments, agencies, and local authorities increasingly (but not exclusively) rely on air pollution models for making decisions related to air quality and traffic management, urban planning, and public health. As a result, the model users' community is becoming larger and more diverse.

Most of the air quality modelling work has been so far based on the "deterministic" approach of using only one dispersion model for a specific application. The selected model provides estimates of average concentrations using a specific meteorological and emission data set. A serious weakness of this method lies in the fact that many uncertainties, not only related to the calculations and input variables, but also to the very nature of atmospheric processes, are ignored. That it might have

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serious implications for exposure studies, since the area and number of people exposed to a predicted pollution level may be very sensitive to the uncertainties associated with this prediction (Fisher and Ireland, 2001).

The total uncertainty involved in modelling simulations can be considered as the sum of three components (Hanna, 1988): (a) the uncertainty due to errors in the model physics, (b) the uncertainty due to input data errors, (c) the uncertainty due to stochastic processes (e.g. turbulence) in the atmosphere. It may be possible to reduce the first component of model uncertainty by introducing more physically realistic and computationally efficient algorithms. It may also be possible to eliminate some of the effects of input data errors once more accurate monitoring instruments are set up at representative locations. However, the stochastic fluctuations are a natural characteristic of the atmosphere that cannot be eliminated. Therefore, practical methodologies need to be developed to assess total model uncertainty and present results in a meaningful way. If uncertainties (however large) are explicitly reflected on model results, policy makers will still make decisions, but an inappropriate level of reliance on the results will be avoided (Pielke, 1998; Dabberdt and Miller, 2000).

The Monte-Carlo analysis is one of the commonly used methods for propagating input data errors through air quality models. It has been applied to models of different levels of sophistication, from Gaussian plume (Irwin et al., 1987) to complex photochemical codes (Hanna et al., 1998, 2001). This method enables an evaluation of the output of the model for many sets of combinations of the input parameters. These data sets are obtained by random sampling from the distribution assigned to each one of the uncertain input variables. Two important advantages of this statistical method are that it can be applied to a complete set of about 100 or more input parameters, and that it is widely used in the analysis of other environmental problems. On the other hand, it has the limitation that the estimates of uncertainty in the inputs are often based on informal processes (e.g. the professional judgement of the modeller), and that the cost of the method in terms of computer simulation time might be quite high, since a relatively large number (>100) of model runs is preferable (Irwin et al., 1987; Hanna et al., 1998).

Freeman et al. (1986) developed a theoretical formula for propagating input data errors through dispersion models. Results obtained using this formula were in good agreement with Monte-Carlo uncertainty estimates for unstable atmospheric conditions, but large inconsistencies were observed for neutral and stable conditions. These results also showed that even small uncertainties in the inputs might cause large uncertainties in the predictions.

Dabberdt and Miller (2000) used a probabilistic method for quantifying the uncertainty related to model predictions in an accidental release application. An ensemble set of 162 simulations was created by specifying a best estimate together with two additional values that bounded the likely range of uncertainty in estimating four input parameters (i.e. wind speed, wind direction, source strength, and plume rise). A best estimate together with a "second choice" was specified for atmospheric stability. Finally, contour concentration patterns (both deterministic and probabilistic) and histograms of the probability of occurrence of concentrations at specific receptor locations were produced to illustrate the uncertainty in the predictions.

Another way to estimate uncertainty in model predictions is by determining the input parameters to which the model in use is more sensitive. A so-called sensitivity analysis indicates how much of the overall uncertainty in the model predictions is associated with the individual uncertainty in each model input (McRae and Seinfeld, 1983). Sensitivity studies are not, strictly speaking, uncertainty analysis, since they do not combine the uncertainties of the model inputs to provide a realistic estimate of the uncertainty in the model output. Nevertheless, knowledge of the model's sensitivity to different variables is necessary in order to decide where emphasis should be placed in estimating total uncertainty (Hanna, 1988).

In this study, the sensitivity of three street canyon models (STREET, OSPM and AEOLIUS) to certain input variables and internal parameters was examined. Furthermore, practical methodologies for assessing model uncertainty were tested using air quality measurements from two street canyons in Paris.

2. Experimental

2.1. Field measurements

The data used in this study were collected during two field experiments carried out in Paris during winter (Bd. Voltaire, December 1998) and summer (Rue de Rennes, July 1999). The two sites were busy four lane streets with large pavements and uniform buildings lining up continuously on both sides. The aspect ratios (i.e. height/width) for Bd. Voltaire and Rue de Rennes were approximately equal to 0.8 and 1.1, respectively.

A number of traffic-related air pollutants were sampled during these campaigns. Hourly CO concentrations were measured at two roadside locations using an infrared analyser, and weekly benzene averages were obtained at different heights and distances from the kerb using diffusive samplers in both the canyons. The ensemble of these measurements revealed a significant spatial and temporal variability of air pollution in urban streets.

The background contribution to the roadside concentrations was derived from benzene measurements (in green spaces adjacent to the street monitoring sites) and an empirical relationship relating benzene to CO concentrations. Benzene background values ranged from 0.4 ppb in Rue de Rennes to 1.3 ppb in Bd. Voltaire (weekly averages), while roadside values were from 2 to 6 times higher depending on the sampling location within the street (e.g. pavement, balcony, etc.). The experimental set-up and monitoring results from these two campaigns were presented and fully discussed elsewhere (Vardoulakis et al., 2002).

Synoptic meteorological information was obtained from three permanent weather stations operated by Meteo France. Two of them were located within Paris: in Montsouris park (anemometer height: 26 m) and St. Jacques tower (anemometer height: 56 m). The third station was located in Orly airport, at approximately 12 km distance from central Paris (anemometer height: 10 m).

Hourly traffic data were obtained from automatic counters permanently operating in both the streets. The average traffic volumes in Bd. Voltaire and Rue de Rennes during measurements were approximately 30,000 and 23,000 veh/day, respectively. The vehicle fleet composition was estimated from on site spot measurements.

2.2. Traffic emissions

The rate of release of emissions in the street was derived from the traffic volumes and the composite emission factor of the pollutant. Two different methods were applied for calculating CO emission factors: (a) the protocol used by Buckland and Middleton (1999), which was based on values specific to UK vehicles, and (b) the IMPACT road traffic emission model commercialised by ADEME (1998). This model uses COPERT II methodology (Ntziachristos and Samaras, 1997) to quantify fuel consumption and atmospheric releases of a specified vehicle fleet in a given year in France. The required input parameters are traffic composition, average vehicle speed, length and slope of the road segment of interest. In addition, the month of the year is used to estimate average ambient temperatures, which are further used for calculating evaporative and cold start/running emissions. The model provides default values for the average travelling distance and the fraction of this distance run with a cold engine in France.

The values estimated using both the methods were compared for consistency with CO emission factors specific to the French vehicle fleet reported in other recent studies (Touaty and Bonsang, 2000; Jones et al., 2000). All values were summarised in Table 1.

Table 1
CO emission factors from road transport in the region of Paris

Author	CO emission factors (g/km veh)		
	Bd. Voltaire	Rue de Rennes	Paris
Buckland and Middleton	7.23	8.73	
IMPACT ADEME	20.29	10.91	
Touaty and Bonsang			8.11
Jones et al.			8.82

3. Street canyon models

Three parametric models (STREET, OSPM, and AEOLIUS) were used to simulate pollutant dispersion within the canyons. These relatively simple models (or variations of them) are likely to be used by local authorities or air quality monitoring networks in a variety of applications including air quality and traffic management, urban planning, population exposure studies, etc. In the following paragraphs, these models are briefly described and some of their empirical assumptions highlighted. References are provided for further in-depth reading.

3.1. STREET

This empirical model calculates series of hourly concentrations at different receptor locations within a street canyon. The concentrations (C) of the pollutant occurring on the roadside consist of two components, the urban background concentration (C_b) and the concentration component (C_s) due to vehicle emissions generated within the street

$$C = C_s + C_b. \tag{1}$$

The C_s component was derived from a simple box model (Johnson et al., 1973). On the leeward side of the street, pollutant concentrations are given by the expression

$$C_S^L = K\frac{Q}{(\sqrt{(x^2+z^2} + h_0)(U + U_S)}, \tag{2}$$

where K is an empirical constant parameter (usually given the value of 7), Q is the rate of release of emissions in the street, x is the horizontal distance between the receptor and the centre of the nearest traffic lane, z is the height of the receptor, h_0 is a constant that accounts for the height of initial pollutant dispersion (empirical value: 2 m), U is the roof-level wind speed, and U_S is a constant that accounts for the additional air movement induced by vehicle traffic (empirical value: 0.5 m/s).

On the windward side, the original expression given by Johnson et al. (1973) was revised by Dabberdt et al. (1973) to account for vertical decrease of concentrations due to entrainment of fresh air through the top of the canyon. The final equation was

$$C_S^W = K\frac{Q}{W(U+U_S)}\frac{H-z}{H}, \tag{3}$$

where H is the height and W the width of the canyon. For parallel or near-parallel synoptic winds, the average of Eqs. (2) and (3) should be calculated. Variations of this model have been widely used in scientific and engineering applications (Qin and Kot, 1993; Hoydysh and Dabberdt, 1988).

3.2. OSPM and AEOLIUS

AEOLIUS (the Full version) and OSPM are semi-empirical dispersion models based on the same mathematical formulation. They combine Gaussian plume theory with empirical box model techniques to calculate concentrations of exhaust gases in a street canyon assuming three different contributions: the contribution (C_d) from the direct flow of pollutants from the source to the receptor, the recirculation component (C_r) due to the flow of pollutants around the horizontal vortex generated within the recirculation zone of the canyon, and the urban background contribution (C_b)

$$C = C_d + C_r + C_b. \tag{4}$$

A Gaussian plume model is used for the calculation of the direct contribution

$$C_d = \sqrt{\frac{2}{\pi}}\frac{Q}{W\sigma_w}F, \tag{5}$$

where F is a factor depending on the synoptic wind, and σ_w is the vertical velocity fluctuation due to mechanical turbulence generated by wind and vehicle traffic in the street. This is described by the relationship

$$\sigma_w = \sqrt{(\alpha u)^2 + \sigma_{wo}^2}, \tag{6}$$

where u is the street-level wind speed, α is a proportionality constant (given empirically the value of 0.1), and σ_{wo} is the traffic-induced turbulence, defined as

$$\sigma_{wo} = b\left(\frac{NVS^2}{W}\right)^{1/2}, \tag{7}$$

where b is an aerodynamic drag coefficient (given empirically the value of 0.3), N the number of vehicles using the street per time unit, V the average vehicle speed, S^2 the road surface occupied by a single vehicle, and W the width of the canyon.

The contribution from the recirculation zone is calculated using a simple box model, which assumes that the pollutants are well mixed inside the box. This is expressed by the relationship

$$C_r = \frac{Q}{W}\frac{L_r}{\sigma_{wt}L_t + U_t L_{S1} + uL_{S2}}, \tag{8}$$

where L_r, L_t, L_{S1} and L_{S2} are dimensions of the recirculation zone, which has the shape of a trapezium; σ_{wt} is the ventilation velocity of the canyon expressed as

$$\sigma_{wt} = \sqrt{(\lambda U)^2 + F_{roof}\sigma_{wo}^2}, \tag{9}$$

where U is the roof-level wind speed, and λ and F_{roof} are proportionality constants given the value of 0.1 and 0.4, respectively. The extension L_r of the recirculation zone is defined as

$$L_r = F_{vortex}Hr\sin\phi, \tag{10}$$

where F_{vortex} is a proportionality constant given the value of 2, H is the height of the canyon, r is a wind speed dependent factor reflecting the strength of the vortex, and ϕ the angle of the roof-level wind with respect to the street. The factor r takes the values

$$r = \begin{cases} 1 & \text{if} \quad U > U_{critical}, \\ U/U_{critical} & \text{otherwise.} \end{cases} \tag{11}$$

The critical velocity $U_{critical}$ for the formation of the vortex in the street is empirically defined as 2 m/s. It should be noted that the width of the recirculation zone L_r cannot exceed the width of the canyon in any case. The relation between street and roof-level winds in a regular canyon is given by

$$u = U\frac{\ln(h_0/z_0)}{\ln(H/z_0)}(1 - F_{wind}\sin\phi), \tag{12}$$

where z_0 is the surface roughness length of the area under consideration, h_0 the effective release height of car exhausts due to initial dispersion, and F_{wind} an empirical constant given the value of 0.2. The wind speed at roof-level (U) is calculated from the input wind (U_a), which corresponds to a meteorological mast of generally different height, using the simple relationship

$$U = F_{mast}U_a. \tag{13}$$

The empirical parameter F_{mast} is given the value of 0.82, which is derived from a logarithmic law similar to expression (12).

On the leeward side of the street, concentrations are calculated as the sum of the direct and recirculation contributions, while on the windward side only the direct contribution of emissions generated outside the recirculation zone are taken into account. If the recirculation zone extends throughout the whole canyon, then the windward concentrations are calculated from only the recirculation component. For near-parallel flow, emissions from outside the recirculation zone may contribute to the leeward concentrations. When the wind speed is near zero or parallel to the street axis, the concentrations on both sides of the canyon become equal. In all cases, the background contribution should be added to obtain the final result. A more detailed description of OSPM is given elsewhere (Berkowicz et al., 1997; Berkowicz, 2000).

OSPM has been applied to several street canyons in European and Asian cities (Berkowicz et al., 1996; Kukkonen et al., 2001; Fu et al., 2000), while AEOLIUS has been mainly used in the UK (Buckland, 1998).

4. Sensitivity analysis

The above described models contain a number of constant parameters that have been empirically defined using experimental data. Although they might have a significant influence on model predictions and hence on the interpretation of the results, these constants have drawn relatively little attention so far. Comparing model results with experimental data, inappropriate values of such model constants might be falsely interpreted as unsatisfactory emission factors or meteorological input data (Buckland and Middleton, 1999).

STREET model includes three empirical parameters K, h_0 and U_S (Eqs. (2) and (3)), which have been adjusted to observed results. Johnson et al. (1973) derived initially the values of $K = 7$, $h_0 = 2\,m$ and $U_S = 0.5\,m/s$, using data from the San Jose Street Canyon Experiment. The value of $K = 7$ is presumably valid for canyons having $H/W \cong 1$, which is comparable to the aspect ratio of San Jose Street. A subsequent evaluation by Dabberdt et al. (1973) did not suggest dramatic variation in K for two narrower canyons with H/W of 1.5 and 2 in St. Luis. Yamartino and Wiegand (1986) kept the original values for h_0 and U_S, but allowed K to rise to an optimal value of 10.2 in their evaluation of STREET model using measurements from Bonner Strasse ($H/W \cong 1$), Cologne. In a research study in China, K was given the value of 6 to simulate concentrations observed in an asymmetric street canyon in Guangzhou City (Qin and Kot, 1993). Finally, in a recent experiment in a street canyon in Buenos Aires ($H/W \cong 1$), best fit was obtained for $K = 8$ (Bogo et al., 2001). Obviously, the value of K weighs heavily on calculations, since it is directly proportional to the model output.

Buckland and Middleton (1999) and Manning et al. (2000) carried out sensitivity studies by varying a number of input variables and internal empirical parameters within AEOLIUS and assessing their impacts on the calculated NO_x and CO concentrations, respectively. It was found that predicted values were almost linearly proportional to the emission factors and traffic flow. Canyon geometry did not significantly affect perpendicular concentrations for a given traffic flow and wind speed. However, that was not the case for parallel concentrations, which increased with canyon height due to the longer integration path before the plume escaped from the canyon, and decreased with canyon width. Increasing surface roughness was shown to enhance calculated concentration. Concentrations decreased at higher traffic speeds as the turbulent mixing increased, although this parameter did not appear to have a dramatic effect on the results. A change in the model coding of the extent of the vortex across the street produced only a minor change in leeward concentrations. Buckland and Middleton (1999) altered the model to let the user specify whether the wind speed was measured at 10 m height (as it is usually the case in airports) or at roof level. That had only a small effect on the results. A constant value (0.1 m/s) was tested for replacing Eq. (7), which calculates vehicle-induced turbulence. This generated much larger concentrations. Finally, it was shown that the assumed magnitudes of arbitrary model constants such as b or λ had a marked effect on calculated concentrations.

4.1. Internal parameters

In the present study, a sensitivity analysis was performed to determine the importance of various internal model parameters on the output of OSPM. Minor modifications were made in the code of the model, so as to enable the user to define externally the values of the following empirical constants: α, b, λ, z_0, h_0, F_{roof}, F_{vortex}, F_{mast}, F_{wind}, and $U_{critical}$. The values of these parameters were then perturbed to observe the effects on the predicted CO concentrations for perpendicular (i.e. leeward and windward) and parallel wind conditions. For the sensitivity runs, the characteristics of the regular canyon of Rue de Rennes were used, while the input wind speed was maintained constant (3.5 m/s). A large number of diagrams similar to Figs. 1–3 were produced (not shown).

As expected, most of the conclusions from the previous sensitivity analysis for AEOLIUS were also valid for OSPM. An increase in surface roughness (z_0) enhanced parallel and leeward concentrations, but had no effect on windward concentrations (Fig. 1). Larger z_0 values would be expected to reduce the street-level wind, but increase the turbulence within the canyon (Manning et al., 2000). However, OSPM simply reduced the street-level wind, which in turn lowered the mechanical turbulence (Eq. (6)) and increased concentrations.

The effective release height (h_0) had the opposite effect on the calculated results. This parameter, which is user defined in AEOLIUS but originally coded inside OSPM ($h_0 = 2$), also represents the height of the simulated roadside receptor. In this analysis, parallel and leeward concentrations decreased with h_0, while the values on the windward side remained constant.

OSPM perpendicular concentrations were proved to be quite sensitive to the value of λ (Fig. 2), which sets the

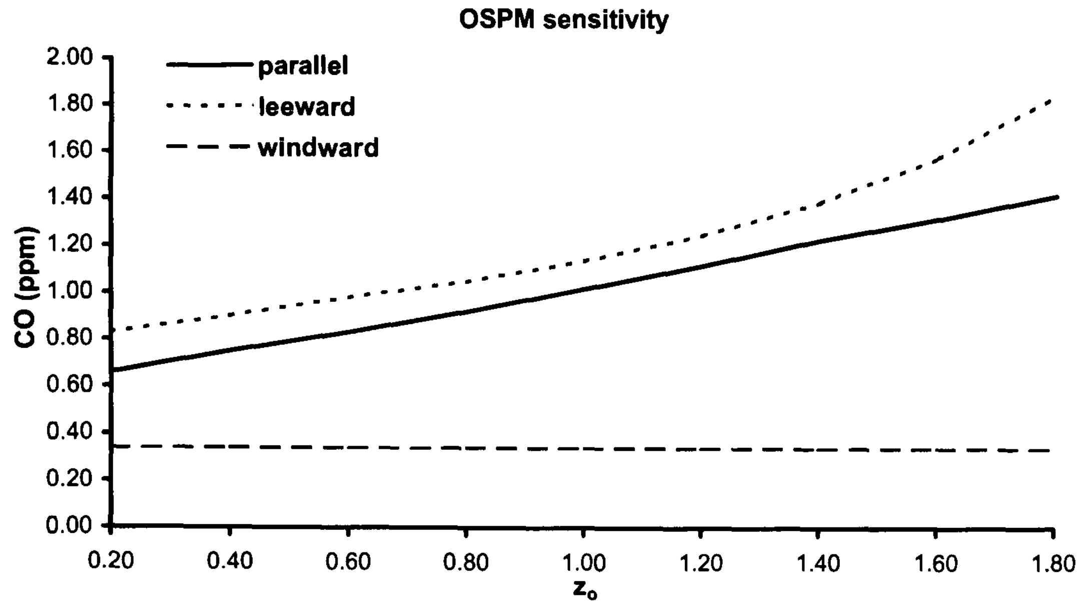

Fig. 1. OSPM sensitivity to the surface roughness length (z_0) for three different wind regimes.

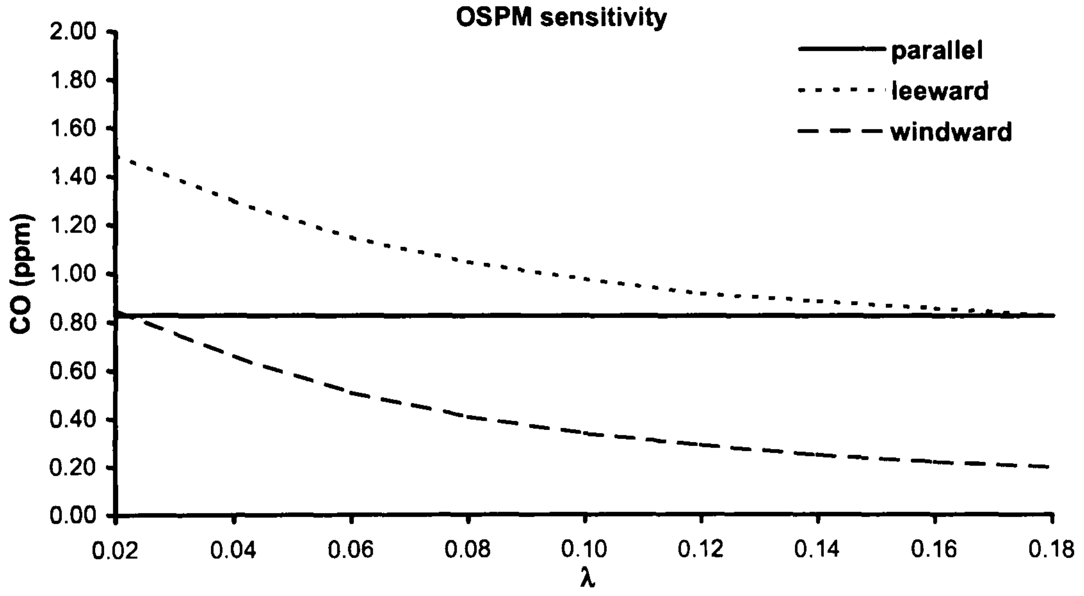

Fig. 2. OSPM sensitivity to the canyon ventilation coefficient (λ) for three different wind regimes.

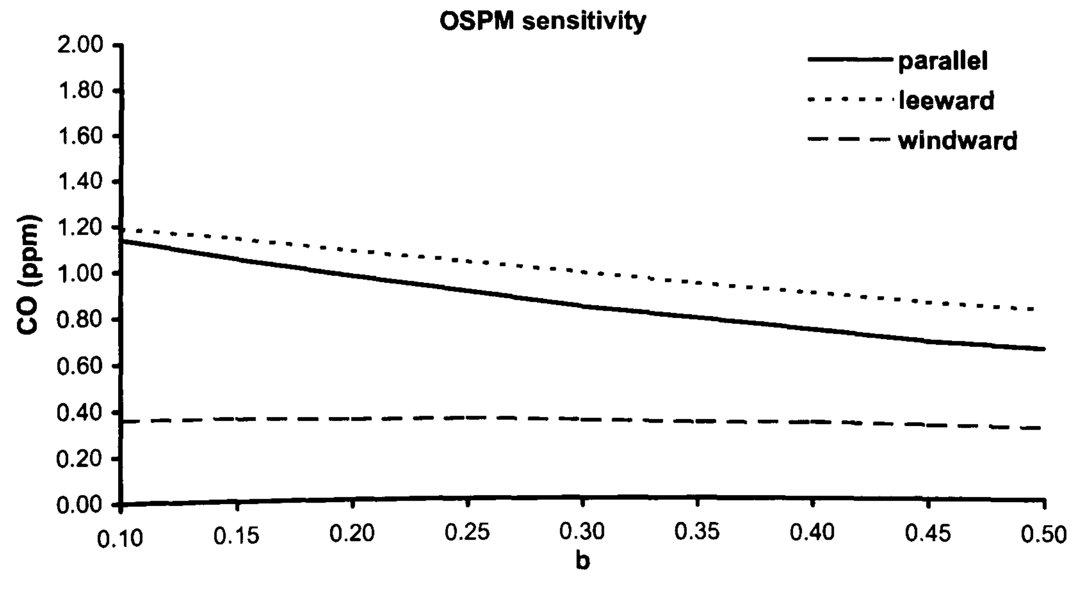

Fig. 3. OSPM sensitivity to the aerodynamic drag coefficient (b) for three different wind regimes.

rate at which material is dispersed out of the top of the canyon, but there was no effect on the parallel concentrations. It should be noted that λ might be sensitive to seasonal weather variations, since atmospheric stability can play a role in the ventilation of street canyons. Parameter α had no marked effect on OSPM results.

As it can be observed in Fig. 3, the aerodynamic parameter b had certain influence on the parallel and leeward concentrations, due to the role of cruising vehicles in the calculation of the mechanical turbulence in the street. For the range of values tested, b had no significant effect on the windward concentrations. When assigning empirical values to b, it should be remembered that the aerodynamic drag of cars is likely to diminish in the future due to improvements in the design of new vehicles.

The influence of the coefficient F_{mast} (Eq. (13)) was found to be of minor importance. Nevertheless, OSPM code was slightly modified here by replacing relationship (13) with the following:

$$U = U_{\mathrm{a}} \frac{\ln(H/z_0)}{\ln(H_{\mathrm{a}}/z_0)}, \tag{14}$$

which allows the user to specify the height of the anemometer (H_α); this information may be easily and accurately obtained from weather station operators.

Altering the value of F_{vortex} (Eq. (10)), a significant effect on leeward concentrations was produced due to changes in the dimensions of the vortex. For example, a change from the standard value of 2 to the value of 1 resulted in a 31% decrease in leeward CO concentrations. No marked effect was observed on the windward and parallel concentrations for such a change.

The F_{wind} coefficient (Eq. (12)) had a small influence only on leeward concentrations, since it mainly affects the dispersion of pollutants coming directly from the source. An increase from the standard value of 0.2 to the value of 0.4 produced 11% higher leeward concentrations. Finally, the coefficient F_{roof} (Eq. (9)) and the critical wind speed U_{critical} for vortex formation (Eq. (11)) had no marked effect on the model results for the range of tested values.

4.2. Emission factors

The CO emission factor calculated using IMPACT was 25% higher than the value calculated according to Buckland and Middleton (1999) for Rue de Rennes during the summer campaign. For Bd. Voltaire, the IMPACT model estimate was 180% higher than the value calculated using Buckland and Middleton's (1999) methodology (Table 1). This large discrepancy between the two methods may be justified by the fact that IMPACT also added the very significant cold start/running emissions (during winter) to the hot running values. Cold start/running emissions are indeed expected to be important for urban driving conditions, because of the relative large number of short trips carried out with cold engines, especially in winter. In an on-road experiment in Belgium (De Vlieger, 1997), it was found that the average CO vehicle emissions measured during the cold phase were 4 to 40 times higher than emissions with a hot start. Other experiments showed significant seasonal differences in cold starts due to ambient temperature variations (Mensink et al., 2000).

Furthermore, it has been reported that emissions obtained from aggressive driving can be up to 4 times higher than those obtained from normal driving (De Vlieger, 1997), and that emission factors typically increase by a factor up to 10 during congestion compared to smooth driving conditions (Sjodin et al., 1998). Finally, it should be remembered that the vast majority of fleet emissions come from a small number of poorly maintained vehicles (Singh and Huber, 2000).

The selection of the appropriate emission model or methodology is crucial, since model predictions are almost linearly proportional to the estimated emission factors. The methodology implemented in IMPACT may be seen as more suitable for the present study (especially for the winter campaign) than the one used by Buckland and Middleton (1999), because it takes into account the average vehicle speed and the seasonal influences in vehicle start/running emissions.

4.3. Meteorological data

Meteorological data obtained simultaneously at different weather stations located within few kilometre distances from each other might differ significantly, especially for short averaging periods. When the first street canyon models were developed few decades ago, it was assumed that the local roof-level wind information needed as an input would not be generally available, and airport wind data would have to be used. For this reason, empirical expressions relating airport and local roof-level winds were derived (Johnson et al., 1973).

Exploring the sensitivity of AEOLIUS to wind data from different sources, Manning et al. (2000) observed that model concentrations were significantly lower when airport rather than local winds were used. This was in agreement with the present study, which showed that simulations carried out using wind data from Orly airport generally produced lower and less accurate predictions compared to those produced using urban wind data.

Nowadays, there is at least one weather station permanently operating within every European capital. It is, therefore, suggested that wind information from airports be avoided, when suitable urban meteorological measurements (obtained under the same quality criteria) are available for running street canyon models.

5. Uncertainty analysis

5.1. Statistical evaluation

STREET, OSPM and AEOLIUS were initially used in a traditional manner to create time series of CO best estimates in Bd. Voltaire and Rue de Rennes. Instead of defining a priori uncertainty ranges in different input variables, three independent meteorological data sets and three different emission factors (as described in Section 2) were used to create ensemble sets of 27 model realisations for each time step.

In the same manner, ensemble sets of 27 model realisations were also created for benzene. In that case, instead of time series, weekly averages corresponding to different receptor locations in the streets were calculated following a practical methodology described by Vardoulakis et al. (2002).

Four statistical evaluation measures were then applied to quantify the differences between predicted and observed concentrations (Yadav and Sharan, 1996): (1) the fractional bias (FB), which provides information on the tendency of the model to over-estimate or under-estimate the observed concentrations (negative values show overpredictions), (2) the normalised mean square error (NMSE), which provides information on the overall deviations between predicted and observed concentrations, (3) the correlation coefficient, which describes the degree of association between variables, and (4) the fraction of predictions within a factor of two (FAC2).

The results of this analysis for 1- and 8-h CO predictions in Rue de Rennes are presented in Table 2. It can be seen that for longer averaging times (i.e. 8 h), the performance of all three models was enhanced. That was expected, since the uncertainty attributed to the turbulent atmospheric processes generally decreases as averaging times increase.

Although there were no dramatic differences in the statistics for the different trials, it might be concluded that OSPM performed slightly better than the other two models in the case of Rue de Rennes. For OSPM, the correlation coefficient ranged from 0.68 to 0.77 for 1-h averages and from 0.81 to 0.89 for 8-h averages. For the same model, the FAC2 values ranged from 0.66 to 0.78 for 1-h averages and from 0.80 to 0.92 for 8-h averages. The optimum FB values (i.e. near zero) were observed for simulations carried out with emission factors calculated according to Buckland and Middleton (1999) and wind data obtained from Montsouris station.

When the same statistical measures were applied to the Bd. Voltaire data, STREET seemed to give slightly better predictions than the other two models. For STREET, the correlation coefficients ranged from 0.74 to 0.75 for 1-h averages and from 0.96 to 0.97 for day averages (night measurements were not available in Bd. Voltaire). In that case, it was revealed that emission factors calculated using IMPACT gave the best agreement ($FB \cong 0$ for 1-h averages), while the other two emission factors produced large underpredictions ($FB \cong 1$ for 1-h averages). A reason for this was probably the fact that IMPACT accounted also for the very significant cold start/running emissions in Bd. Voltaire, an urban environment with many parking spaces, during winter. The use of urban meteorological data was again found to improve the model results.

Finally, some significant discrepancies between OSPM and AEOLIUS predictions were identified (e.g. NMSE and FAC2 values in Table 2), despite the fact that both the models are based on the same formulation. This is probably due to certain differences in coding, parameterisation and data pre-processing techniques between the two models.

5.2. Concentration ranges using three models

A comparison of statistical performance measures helps to determine if one model or input data set is better than another for a specific application. In certain cases, however, model results may deviate quite significantly (especially for short averaging times), without this being reflected on the overall statistics.

Medians together with maximum and minimum concentrations were calculated for each ensemble set of 27 model realisations corresponding to a specific time and location within the two streets in Paris. The extreme concentrations were thought to give a rough estimate of model uncertainty in the predictions (Vardoulakis et al., 2001). As it can be seen in Figs. 4 and 5, approximately 92% and 95% of CO observations in Bd. Voltaire and Rue de Rennes, respectively, lie within the predicted concentration ranges. In the same manner, error bounds were also attached to 8-h mean CO concentrations (Fig. 6), since CO standards are written as 8-h averages.

The same method was used to obtain a rough estimate of model uncertainty in benzene predictions corresponding to different sampling locations within the two canyons. As it can be seen in Figs. 7 and 8, all observed benzene concentrations (weekly averages) lie within the estimated error bounds.

An alternative method using fuzzy numbers to treat predictions from more than one model has been recently proposed (Fisher and Ireland, 2001). This method, which provides probability weightings on model predictions, may be applied at a later stage of an air quality assessment, if a more advanced interpretation of the modelling results is needed.

As far as the choice of models is concerned, the intention was to define the appropriate degree of sophistication for the specific applications, so as to minimise uncertainties. It might be incorrectly assumed that the total uncertainty in predictions always decreases

Table 2
Statistical evaluation of 27 model simulations for predicting 1- and 8-h CO concentrations in Rue de Rennes, using three models (OSPM, STREET, and AEOLIUS), three wind data sets (from M: Montouris park, S: St-Jacques tower, and O: Orly airport), and three emission factors (according to (1): Buckland and Middleton, (2): IMPACT-ADEME, (3): Touaty and Bonsang)

Statistical evaluation	Correlation coefficient		Fractional bias		NMSE		FAC2	
Ideal value	1.00		0.00		0.00		1.00	
Averaging time	1 h	8 h	1 h	8 h	1 h	8 h	1 h	8 h
OSPM (M1)	0.68	0.82	0.10	0.00	0.38	0.15	0.76	0.88
OSPM (M2)	0.68	0.82	−0.12	−0.15	0.42	0.21	0.78	0.84
OSPM (M3)	0.68	0.82	0.17	0.14	0.40	0.18	0.73	0.88
OSPM (S1)	0.77	0.89	0.13	0.11	0.31	0.12	0.78	0.88
OSPM (S2)	0.77	0.89	−0.09	−0.11	0.34	0.15	0.76	0.88
OSPM (S3)	0.77	0.89	0.21	0.19	0.33	0.14	0.76	0.92
OSPM (O1)	0.71	0.81	0.19	0.16	0.41	0.23	0.71	0.80
OSPM (O2)	0.71	0.81	−0.03	−0.06	0.41	0.25	0.78	0.84
OSPM (O3)	0.71	0.81	0.27	0.24	0.45	0.26	0.66	0.80
STREET (M1)	0.72	0.83	0.40	0.38	0.55	0.34	0.65	0.72
STREET (M2)	0.72	0.83	0.18	0.17	0.42	0.20	0.78	0.84
STREET (M3)	0.72	0.83	0.47	0.45	0.64	0.37	0.57	0.64
STREET (S1)	0.76	0.89	0.54	0.52	0.72	0.45	0.49	0.44
STREET (S2)	0.76	0.89	0.33	0.31	0.46	0.21	0.65	0.76
STREET (S3)	0.76	0.89	0.61	0.59	0.84	0.56	0.43	0.40
STREET (O1)	0.64	0.74	0.58	0.56	0.95	0.67	0.44	0.48
STREET (O2)	0.64	0.74	0.37	0.35	0.67	0.41	0.57	0.60
STREET (O3)	0.64	0.74	0.65	0.63	1.08	0.79	0.38	0.44
AEOLIUS (M1)	0.64	0.80	0.11	0.07	0.45	0.19	0.59	0.68
AEOLIUS (M2)	0.66	0.81	−0.11	−0.14	0.47	0.23	0.56	0.64
AEOLIUS (M3)	0.66	0.80	0.19	0.15	0.46	0.20	0.59	0.72
AEOLIUS (S1)	0.71	0.86	0.16	0.13	0.43	0.17	0.49	0.64
AEOLIUS (S2)	0.71	0.87	−0.06	−0.09	0.47	0.20	0.46	0.60
AEOLIUS (S3)	0.71	0.87	0.24	0.21	0.45	0.18	0.49	0.64
AEOLIUS (O1)	0.67	0.80	0.40	0.36	0.64	0.36	0.51	0.56
AEOLIUS (O2)	0.67	0.80	0.19	0.15	0.48	0.24	0.57	0.68
AEOLIUS (O3)	0.67	0.80	0.47	0.44	0.72	0.43	0.47	0.56

as the complexity of a model increases. This is only true for the uncertainty attributed to errors in the physical description of the model domain (e.g. incorrect assumptions, oversimplifications, etc.). On the other hand, sophisticated models require a larger amount of input information, which inevitably introduces a larger data uncertainty component in their calculations.

5.3. Concentration ranges using one model

Assuming that there is only one model and one input data set available, it is still possible to have a rough estimate of uncertainty in model predictions. This can be achieved by assigning a best estimate together with two additional values that may bound the likely range of uncertainty related to certain internal model parameters. In this case, λ, b, and z_0 were selected because they were found to have a significant effect on OSPM results (see Section 4.1).

Using the meteorological data from Montsouris station and the emission factors calculated according to Buckland and Middleton (1999), an ensemble set of 27 OSPM simulations was again created by only varying the values of b and z_0 by 33% and the value of λ by 50%, as shown in Table 3. The "max" and "min" values assigned to the three selected parameters were in a reasonable agreement with values previously tested by Buckland and Middleton (1999), and Manning et al. (2000).

Although the number of simulations here was the same as in the examples of Section 5.2, the estimated concentration ranges were significantly narrower (Fig. 9). As a result, more than 20% of the observed CO concentrations fell outside the estimated ranges.

A more rigorous approach would require that probability functions be developed for each "sensitive" input or internal model parameter, and that these be randomly sampled to obtain improved ensemble sets

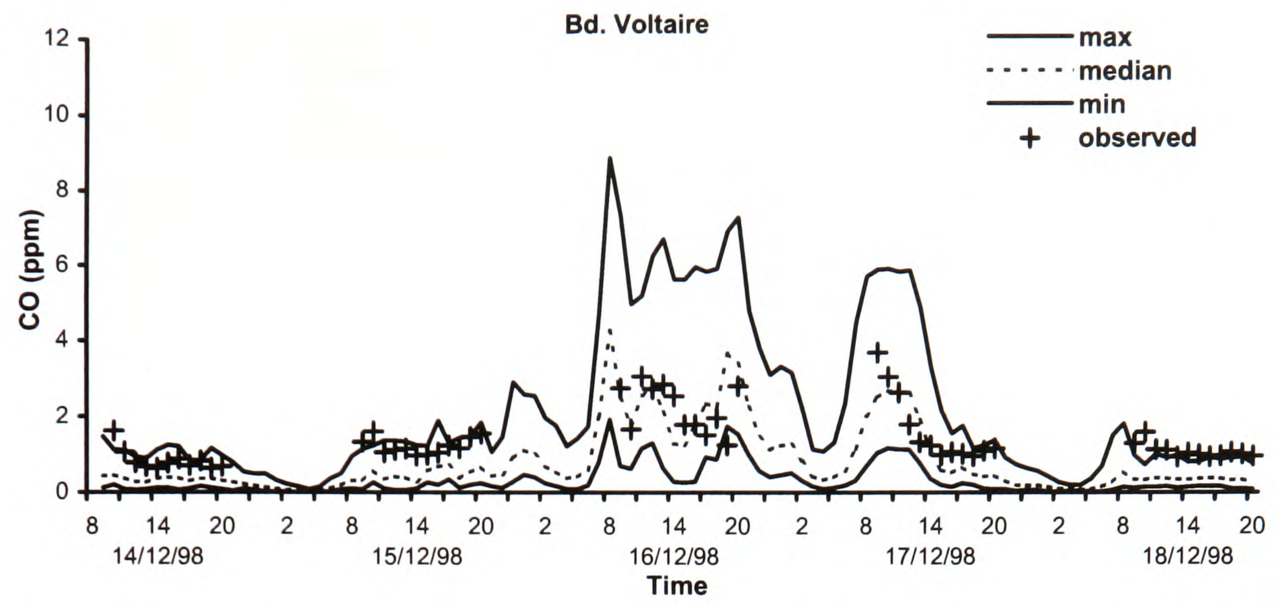

Fig. 4. Time series of best 1-h CO estimates (median), error bounds (max, min), and observed concentrations in Bd. Voltaire produced using three models.

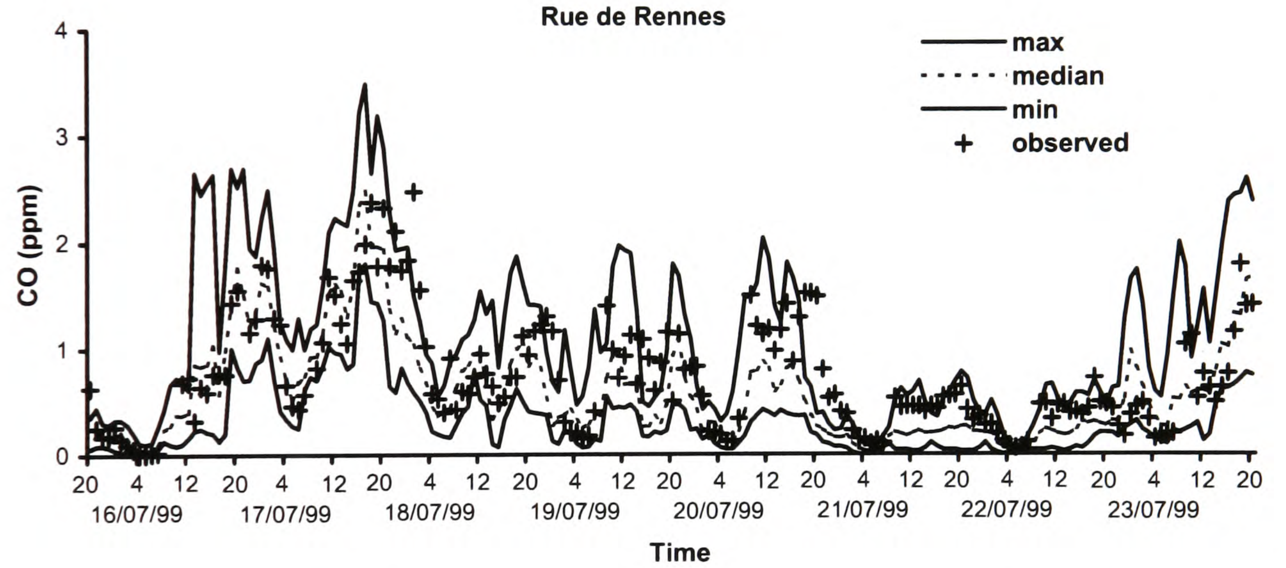

Fig. 5. Time series of best 1-h CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using three models.

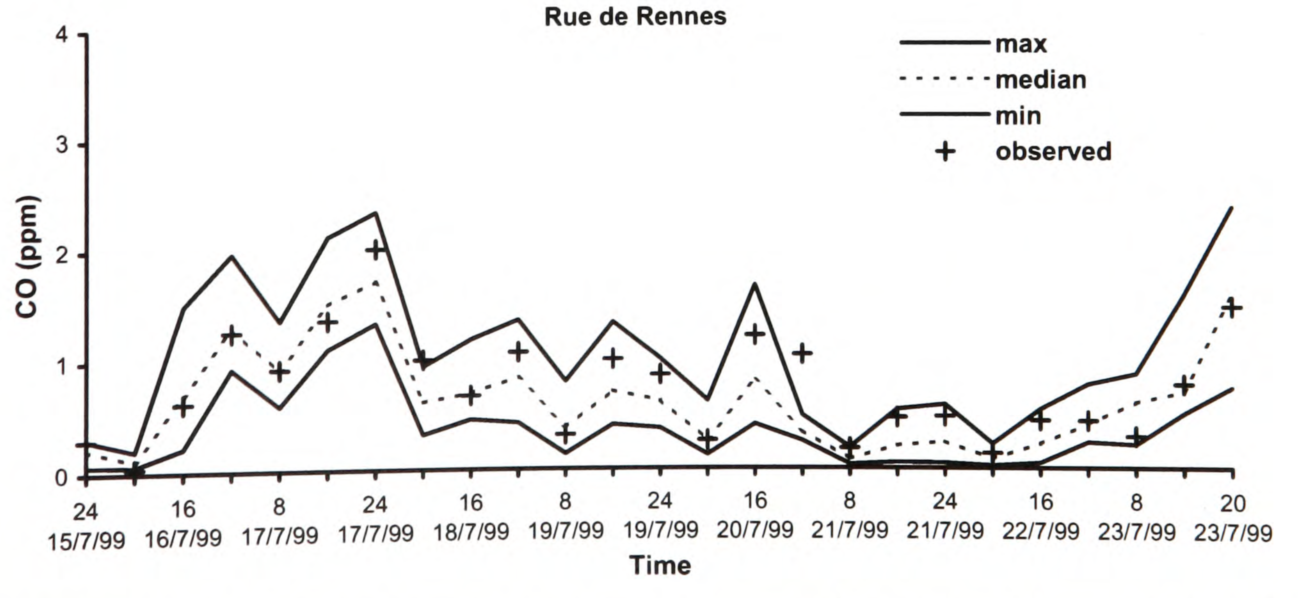

Fig. 6. Time series of best 8-h CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using three models.

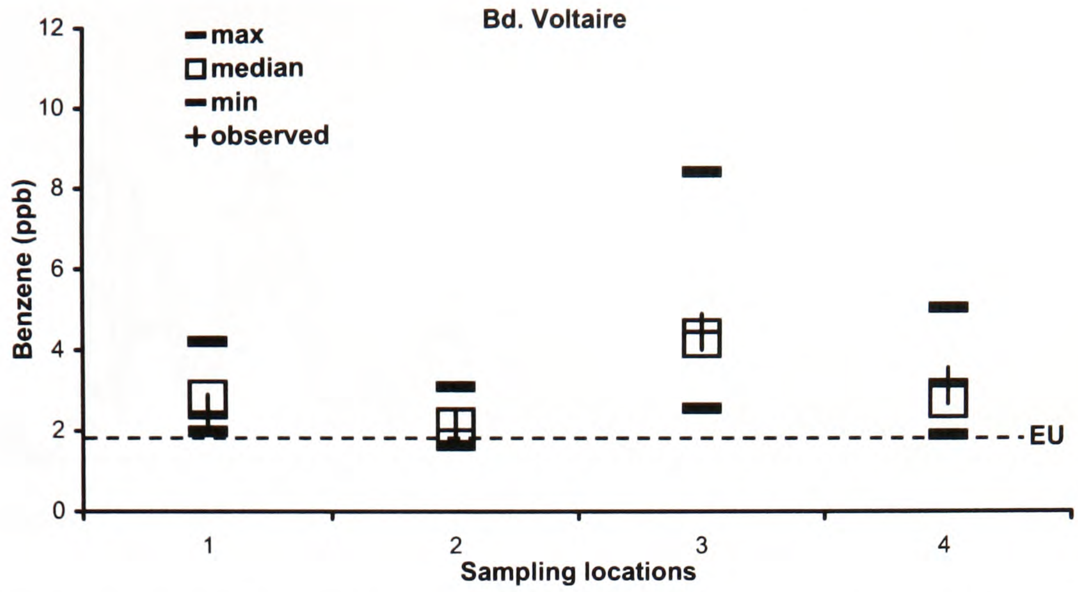

Fig. 7. Best weekly benzene estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Bd. Voltaire produced using three models. The dashed line shows the EU proposed limit value for benzene (1.66 ppb).

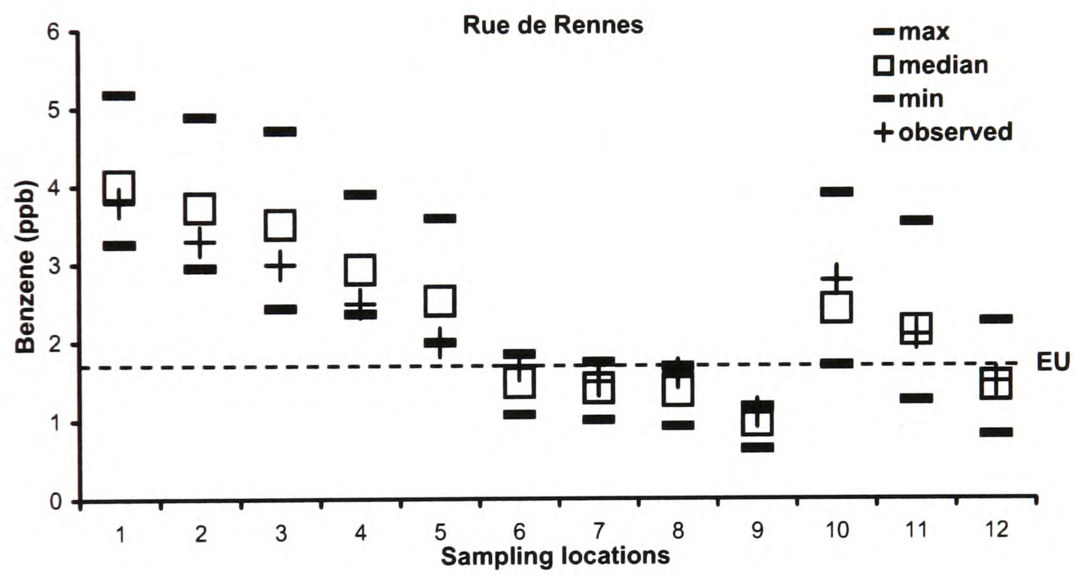

Fig. 8. Best weekly benzene estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Rue de Rennes produced using three models. The dashed line shows the EU proposed limit value for benzene (1.66 ppb).

(Dabberdt and Miller, 2000). On the other hand, this approach would require a much larger number of model runs, which would increase the time and consequently the cost of simulations. Furthermore, it is more important to specify the width rather than the shape of any probability functions describing the uncertainty of the variables (Alcamo and Bartnicki, 1987).

Finally, it should be stressed that this single model approach can be applied only if the user has access to a number of empirical model parameters, which is usually not the case since they are often coded inside the model.

Table 3
Values of three internal OSPM parameters (b: aerodynamic drag coefficient, z_0: surface roughness length, and λ: canyon ventilation coefficient) used for creating an ensemble set of 27 model simulations

Coefficients	Estimated values		
	Max	Standard	Min
Aerodynamic drag (b)	0.40	0.30	0.20
Surface roughness (z_0)	0.80	0.60	0.40
Canyon ventilation (λ)	0.15	0.10	0.05

5.4. Comparison with regulatory standards

It has already been discussed that the traditional method of applying air quality models disregards uncertainty. There is therefore a risk of ending up with a misleading classification of urban environments in only two, "yes" or "no" polluted categories. What is really needed is a probabilistic comparison of predicted values against regulatory limits that takes into account best model estimates as well as their related error bounds.

Having calculated the concentration ranges (Section 5.2), a probabilistic method (Ramsey and Argyraki, 1997) was adopted for assessing the compliance of

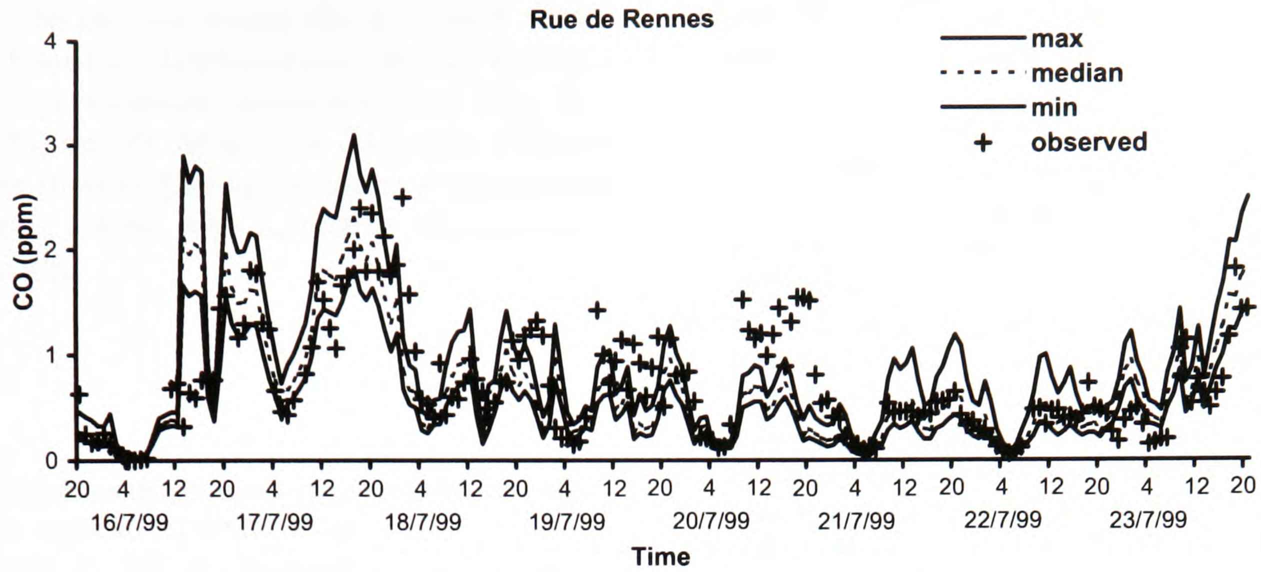

Fig. 9. Time series of best 1-h CO estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes produced using one model (OSPM).

different kerbside locations with an ambient air quality standard. According to this method, the predicted benzene concentrations (Figs. 7 and 8) were classified in four different categories with respect to the proposed EU limit value of 5 μg/m^3 (i.e. 1.66 ppb): (a) "exceeding the limit" if the predicted minimum value for one location was above the threshold, (b) "probably exceeding" if the predicted median was above the limit while the minimum was below, (c) "possibly exceeding" if the predicted median was below the limit but the maximum above, and finally (d) "not exceeding" if the predicted maximum lay below the threshold. According to this classification, locations 1, 3, and 4 in Fig. 7 were found to exceed the EU threshold, while location 2 was *probably* exceeding the same limit value. In Fig. 8, sampling locations 1–5, and 10 were exceeding the threshold, location 11 was *probably* exceeding, locations 6–8, and 12 were *possibly* exceeding, and finally only location 9 was found not to exceed the limit.

Although the averaging times in Figs. 7 and 8 do not directly correspond with the EU standard value for benzene (i.e. annual average), these examples show a precautionary way of applying a discrete air quality criterion. It should be remembered that it is possible to relate short-term averages to annual standards (e.g. for benzene) by means of an appropriate surrogate (e.g. CO) measured throughout the year. An alternative approach would allow for a certain degree of tolerance to be associated with the criterion itself, instead of attaching error bounds to the predictions.

6. Conclusions

The sensitivity of regulatory street canyon models to certain input variables (i.e. emission factors and meteorological data) and internal parameters was studied. Emphasis was put on the sensitivity of OSPM to the full set of empirical constants coded inside the model.

Practical methodologies for obtaining first estimates of model uncertainty were tested using air quality data from two field experiments in Paris. Two different approaches were used to create ensemble sets of 27 realisations, which were then used to derive best pollutant (i.e. CO and benzene) concentration estimates and related error bounds. Statistical techniques were applied to evaluate the simulations. It was shown that the use of wind information obtained from urban monitoring stations optimised the application of models. Large uncertainties in vehicle emission factors were identified.

A probabilistic method for assessing air pollution in urban streets was proposed. Although sophisticated statistical and error propagation techniques were avoided, the above methodologies certainly increased to some extent the complexity and amount of modelling work. It is, however, believed that they can contribute to reducing the risk of misinterpreting modelling results and making erroneous management decisions.

It should be finally noted that uncertainties related to air quality measurements (e.g. sampling and analytical errors) were not taken into consideration. These uncertainties, though generally smaller than model uncertainties, may have some implications in decision making, if the predicted concentrations are close to an air quality standard.

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Research Articles

BTX Concentrations Near a Stage II Implemented Petrol Station

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Abstract. A combined monitoring and dispersion modelling methodology was applied for assessing air quality at three different levels of proximity to the selected service station: (I) next to the fuel pumps, (II) in the surrounding environment, and (III) in the background. Continuous monitoring and passive sampling were used for achieving high temporal and spatial resolution, respectively. A Gaussian dispersion model (CALINE4) was used for assessing the road traffic contribution to the local concentrations under different meteorological conditions.
It was established that Stage 2 vapour recovery reduces BTX concentrations not only near the pumps, but also in their surrounding environment. However, there is evidence that the efficiency of the system is wind speed dependent. The modelling simulation of the worst case wind scenario revealed the significance of local traffic emissions. It was shown that the traffic contribution even from a single road in the vicinity of the station can, under certain conditions, be higher than the contribution of the station itself to the local BTX levels. Finally, after comparison with previous studies, the concentrations measured near the service station (which was situated in a rural environment) appear to be lower than those observed in busy street canyons in city centres.
It can be concluded, although Stage 2 recovery system effectively reduces working VOC losses in service stations, that it will only have a limited positive impact on local air quality if the service station is located in a heavily polluted area.

Keywords: Air quality; aromatic hydrocarbons; benzene; BTX concentrations; displacement loss; dispersion modelling; naphthenes; olefins; paraffins; petrol; population exposure; service station; spillage loss; research articles; Stage 1 and 2 control; vapour recovery; VOCs; volatile organic compounds (VOCs); wind conditions

Introduction

Petrol is a complex petroleum product mainly consisting of paraffins, olefins, naphthenes and aromatic hydrocarbons containing from 3 to 11 carbon atoms. The exact composition of the fuel varies according to its origin. BTX compounds, mainly occurring in unleaded petrol, can be treated as tracers for motor generated pollution.

From a toxicological point of view, benzene is the most notorious of these compounds. As a matter of fact, it has been classified in group 1 of the IARC [1]. Furthermore, according to the World Health Organisation (WHO) [2], there is no single threshold value for benzene below which there is no danger for human health. WHO proposes a unit risk excess of 6×10^{-6} per µg/m^3 for leukaemia, based on a linear extrapolation model without threshold. That is to say, if one million people are exposed to 1 µg/m^3 of benzene for a lifetime, 6 members of this population are expected to suffer from leukaemia at some stage of their lives.

From a regulatory point of view, the limit value of 5 µg/m^3 has been proposed for benzene by the European Commission (EC) [3]. For France, the Loi sur l'Air [4] determines an air quality objective value of 2 µg/m^3 for the same compound.

Fuel storage and delivery activities in service stations have also been a subject of EC regulation due to the VOC releases involved [5]. Tanks containing petrol can emit VOC vapours due to filling and emptying activities (displacement losses), as well as changes in temperature and atmospheric pressure (breathing losses). Every time an empty tank is filled, the corresponding air volume saturated with petrol vapour is displaced into the atmosphere. Displacement losses can increase both occupational exposure to VOC (for people working in the station, car drivers and passengers) in the immediate proximity of the pumps, and population exposure in the surroundings of the station [6-7]. For the reduction of these emissions, vapour recovery devices can be put in place. These systems return the VOC saturated volume of air which has been displaced from the tank being filled to the tank being emptied during the delivery of the fuel. The set of equipment used for vapour recovery during the loading of a storage tank is called Stage 1 control, while the system used for the same purpose during the refuelling of a vehicle tank is called Stage 2 control.

The question raised is whether Stage 2 control is really efficient. Does this system make a significant difference in terms of ambient VOC levels and correspondent human exposure near service stations? Before giving an answer, one should establish the contribution of service stations (with and without vapour recovery taking place) and road traffic to the local air pollution under different weather conditions.

A research project, commissioned by the French Ministry of Environment, was carried out by INERIS to address this question. A two week monitoring campaign was carried out in a modern service station by the RN 10 motorway, near the small town of Prunay in the south of Rambouillet (Yvelines, France) in November 1999. During the first week, the vapour recovery system of the station was operating, while during the second one it was disconnected. The adopted methodology, presented below in detail, was based on real measurements and model simulations.

1 Methods

1.1 Site description

The specific service station was selected for the campaign because of the availability of modern refuelling facilities and vapour recovery systems (which could be turned on and off), the flat topography of the site (which increases confidence over modelling results), the good natural ventilation of the area (which makes simple Gaussian models applicable), the existence of only one traffic axis (RN 10) (a good linear source), the easily measured background contribution, and finally the relatively constant quantities of petrol sold in the station.

1.2 Sampling strategy

The concentration of a pollutant at a given location and time equals the summation of the contributions from different emission sources. The major factor which determines the dispersion of gaseous pollutants in the atmosphere is the wind (speed and direction) and its related turbulent effects.

Considering the case of the service station, the concentration C_i of a pollutant at a given location can be expressed in the following way:

$$C_i = C_o + C_s + C_r \tag{1}$$

Where C_o is the background concentration of the pollutant, C_s is the concentration due to emissions released within the station, and C_r is the contribution from vehicle traffic in the proximity of the station. C_r and C_s is expected to vary as a function of time and distance from the source. The total concentration C_i of the pollutant, as well as the background contribution C_o, can be measured using adequate equipment. By contrast, the other two independent contributions C_s and C_r need to be calculated.

In order to study the efficiency of the vapour recovery system, one should be able first to quantify the variation of VOC releases within the station, and then attribute this variation to different factors (vapour recovery, traffic volume, accidental spillage, weather conditions, composition of the fuel, etc.) In addition, the variations of C_s should be significant compared to C_o and C_r values.

A mobile monitoring unit (trailer-lab) and diffusive samplers were used for taking measurements at 20 sampling locations (**Fig. 1**, Appendix), which can be classified in three different 'levels' of proximity to the source: (I) next to the fuel pumps, (II) within their surrounding environment, and (III) in the background. The samplers placed nearer the source (level I) were located in pairs at two different heights (h_1 = 0.2 m, h_2 = 2.0 m).

1.3 Measurement techniques

Two anemometers, one 3-D ultrasonic (WindMaster, Gill Instruments, Hampshire, UK) and one mechanical (microvane and three-cup), were used for monitoring local wind speed and direction (24 hours per day). Air temperature (AANDERAA, 3455) and relative humidity (AANDERAA, 3445) recorders were also used.

A carbon monoxide infra-red analyser (UNOR 610) interfaced with a data logger (STADUP) was continuously monitoring ambient air (24 hours per day). This equipment was sheltered in the weather-proof trailer-lab parked next to the service station. Perkin Elmer (PE) diffusive samplers (adsorbent: Carbotrap B) were used for the multisite BTX measurements. The PE tubes were sheltered in specially designed aluminium boxes, and regularly replaced.

Field notes were taken for traffic volume and vehicle speed. The quantity of petrol sold in the station was controlled by the fuel pump meters.

1.4 Analytical methods

BTX samples were analysed using thermal desorption and gas chromatography + FID (WCOT ID = 0.32 mm, 50 m CP 5/2 5 CB, 1.2 µm). A quality assurance (QA/QC) programme, including sampling duplicates, blanks and instrument calibration with standard gases was followed during the sampling and analytical work. Ambient BTX concentrations were calculated from the relationship:

$$C = \left(\frac{L}{S}\right)\frac{1}{D}\frac{m}{t} \tag{2}$$

Where C is the ambient concentration of the gas, L/S is a constant depending on the dimensions of the sampling tube, D is the diffusion coefficient of the gas in ambient air, m is the mass of the pollutant sampled, and t the time of exposure [8]. The QA/QC programme included the validation of the equation 2 by exposing the PE tubes to dynamically generated contaminated atmospheres.

1.5 Cartography and modelling

The cartography of the pollutants (i.e. the plotting of iso-concentration lines on a site map) is an efficient means of visualising sampling results and interpolated values. Amongst the different interpolation methods applicable to this case, kriging was considered to be the most appropriate one because of the irregular distribution of the sampling points and the possibility of using variogram models [9].

Carbon monoxide and benzene concentrations were modelled using CALINE4, the last version of the Gaussian dispersion model developed by the California Department of Transportation (USA). This model uses Gaussian plume theory as well as the concept of a mixing zone to simulate the dispersion of pollutants emitted from a line source [10]. Model input requirements include emission factors, description of the site topography, meteorological and traffic data. CALINE4 calculates the pollutant concentrations for multiple receptors at distances up to 500 m from the source.

2 Results and Discussion

Measurements were taken for two different time periods: During the first one, Stage 2 recovery system was operating, and it was disconnected during the second one. General meteorological, traffic and fuel data averaged during different periods of time are presented in **Table 1**.

Table 1: General information averaged over different time periods

Averaging period	2-9/11/99	15-22/11/99
Wind speed (m/s)	2.5	3.4
Wind direction (deg)	242	243
Temperature (°C)	6.9	0.5
Volume of fuel (diesel+petrol) sold	11000 l/day	
Diesel / Petrol partition	3 / 1	
Traffic volume (veh/h)	1400	
Fleet composition	74% passenger cars 7% LGV 18.5% HGV 0.2% buses 0.3% motos	

LGV: Light-Good Vehicles
HGV: Heavy-Good Vehicles

Weekly mean BTX concentrations for all locations at 2 m height are presented in **Fig. 2a** (without vapour recovery) and **Fig. 2b** (with vapour recovery). The concentrations detected nearer the ground (at 0.2 m height) at proximity level I were significantly higher than the rest of the measurements and, for this reason, they are treated separately. The results shown in Fig. 2a and b (Appendix) were used for the plotting of the iso-concentration maps, one of which is presented in Fig. 1. Concentration mapping was carried out using the kriging method with a linear variogram model.

2.1 Displacement and spillage losses

The regular profile of the BTX concentrations measured in all sampling locations (with and without Stage 2 control) reveals the common origin (i.e. petrol combustion and evaporation) of VOC emissions. The background BTX concentrations, which were (as expected) much lower than those measured near the station or the motorway, were approximately the same for all level III locations. Near the fuel pumps (level I), concentrations are up to a factor of 4 higher than the background values, as can be seen on Fig. 1.

Large differences in BTX concentrations were detected between the sampling locations of different height near the pumps (level I). The concentrations observed close to the ground were significantly higher than those measured at 2 m height. This could be explained by the short distance between samplers and car exhausts, as well as by the occurrence of accidental spillage losses of fuel on the ground. Evaporative emissions due to the loss of a few drops of petrol while filling the tank of a vehicle can be easily calculated using a simple closed box model. For example, if 1 ml (i.e. 5 drops) of petrol containing approximately 1% of benzene were evaporated within an isolated 2 m^3 envelop of air, that would give rise to a concentration of 4 mg/m^3 for benzene. On the other hand, the loading of 40 l of petrol from a storage to a vehicle tank would induce displacement losses which would give rise to benzene concentrations of approximately 85 mg/m^3 within the same 2 m^3 isolated envelope of air (assuming there is no vapour recovery control, and the partial pressure of benzene in the tank is approximately 1 mm Hg). From this simple calculation, it can be concluded that displacement losses during petrol delivery without Stage 2 control are much more significant (about 20 times higher in this example) than small spillage losses.

2.2 Dispersion conditions

In Fig. 2a and 2b, it can be seen that BTX concentrations were in fact higher while the vapour recovery system was operating. This rather unexpected result can be explained by the differences in meteorological conditions during sampling. In particular, mean wind speed was higher (increasing from 2.5 to 3.4 m/s) during the second period of the campaign, when the vapour recovery system was disconnected. In order to establish the influence of wind speed on the ambient pollution levels, BTX concentrations were plotted against the reciprocal of the wind speed in **Fig. 3** (Appendix) for the two different periods of the campaign (a: without Stage 2 control, b: with Stage 2 control). Only the measurements of higher temporal resolution are presented in Fig. 3. Those correspond to the sampling points near the pumps (level I), where passive tubes were replaced every 48 hours, covering thus a wider range of meteorological conditions. The observed concentration variations could be mainly attributed to the changes in wind speed, since the station was always upwind with respect to the RN 10, and the quantity of fuel sold remained almost constant. It can be seen for a given wind speed, that benzene as well as total BTX concentrations were lower when the Stage 2 control was operating, which suggests that the system works efficiently. The efficiency of the system, however, varies with the wind speed, as it is shown in **Table 2**. Furthermore, it appears that there is a critical wind speed value above which the system is no longer effective.

Table 2: Impact of Stage 2 control on BTX concentrations for different wind conditions

Wind speed (m/s)	Concentrations ($\mu g/m^3$)				Reduction (%) = (1-Co/C) x 100	
	Stage 2 control OFF		Stage 2 control ON			
	Benzene	BTX	Benzene	BTX	Benzene	BTX
3.5	3.6	13	2.56	13.1	29	0
3.3	4	13.8	2.6	13.4	35	3
2	8.06 *	22.1 *	3.5	17.2	56	22

* extrapolated value

While vapour recovery is taking place, benzene reduction is generally higher than total BTX reduction, possibly due to the higher volatility of benzene. Nevertheless, it should be remembered that measurements are not only affected by evaporative emissions, but also by combustion releases from vehicles using the station.

The benzene concentrations measured in the surrounding environment (level II) during both time periods were normalised with respect to the wind speed to reveal the impact of Stage 2 control. It should be noted that there is no need to normalise further, since the station was always upwind with respect to the RN 10 (thus traffic volume did not affect measurements, $C_i = C_o + C_s$), and the quantity of fuel sold remained almost constant. **Fig. 4** (Appendix) shows that normalised concentrations are lower when the vapour recovery system is operating, which leads to the conclusion that Stage 2 control also reduces pollution in the surroundings of the station (level II).

2.3 Road traffic contribution

CALINE4 was used to estimate the possible contribution from road traffic (RN 10) to the BTX levels occurring at the level II sampling locations. The meteorological input parameters were: wind speed = 2.5 m/s, and ambient temperature = 7°C. The model was run for the most unfavourable wind conditions (i.e. worst case mode). CO emission factors were taken from literature [11] and were adapted to the site-specific fleet composition before being introduced into the model. The outcome of the simulation was used to calculate benzene concentrations by applying an empirically established CO/benzene relationship [14]. This approach was adopted in order to avoid the use of benzene emission factors, which would have introduced a higher uncertainty component in the calculations. The results are presented in **Table 3**.

Table 3: Road traffic contribution to the CO and benzene levels in the surroundings of the service station (under worst case wind conditions)

Site n°	Wind Dir (deg)	CO (ppm)	Benz. (µg/m³)
1	169	0.4	4.8
2	12	0.4	4.8
3	12	0.4	4.8
4	11	0.4	4.8
5	11	0.4	4.8
9	19	0.2	2.4
10	161	0.2	2.4
11	161	0.2	2.4
12	161	0.2	2.4

As expected, NNE and SSE wind directions represent the worst case meteorological scenario. Under these conditions, winds blowing almost parallel to the road axis accumulate the pollutants emitted along the upwind segments of what can be considered as a linear source (i.e. RN 10). As far as the receptors of level II are concerned, under worst case wind conditions the emissions from RN 10 contribute more strongly than the petrol station itself to the total benzene levels.

Finally, comparing the benzene concentrations observed during this study with levels usually occurring in urban environments, it can be concluded that the average benzene concentrations to which pedestrians may be exposed in busy street canyons are generally higher than those occurring in the surroundings (level II) of a Stage 1and 2 implemented service station located in a rural area [14-15].

3 Conclusions

The adopted methodology of using multisite sampling combined with dispersion modelling allowed for a first evaluation of the benefits of Stage 2 control in a motorway service station. This deterministic approach gave reliable results without requiring excessive measurements or calculations.

It has been demonstrated that Stage 2 vapour recovery mitigates BTX and especially benzene levels near the fuel pumps and in the surrounding environment of the service station. Consequently, population exposure to these substances is expected to reduce during the operation of the system. Although vapour recovery reduces VOC emissions due to displacement losses, the effectiveness of the control device is proved to be inversely proportional to the local wind speed.

Fuel spillage losses in a service station should be taken into consideration when evaporative emissions are calculated in personal exposure studies. Under unfavourable wind conditions, the contribution from vehicle traffic in adjacent streets to the pollution levels near the station can be very significant and even higher than the contribution of the station itself.

The emissions released from a Stage 2 implemented service station give rise to benzene concentrations which appear to be lower than those generally occurring in busy urban streets. For this reason, the operation of the system in a service station located in an already polluted urban environment is only expected to bring marginal reductions in VOC levels, although the total mass of releases into the atmosphere will be reduced.

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Appendix

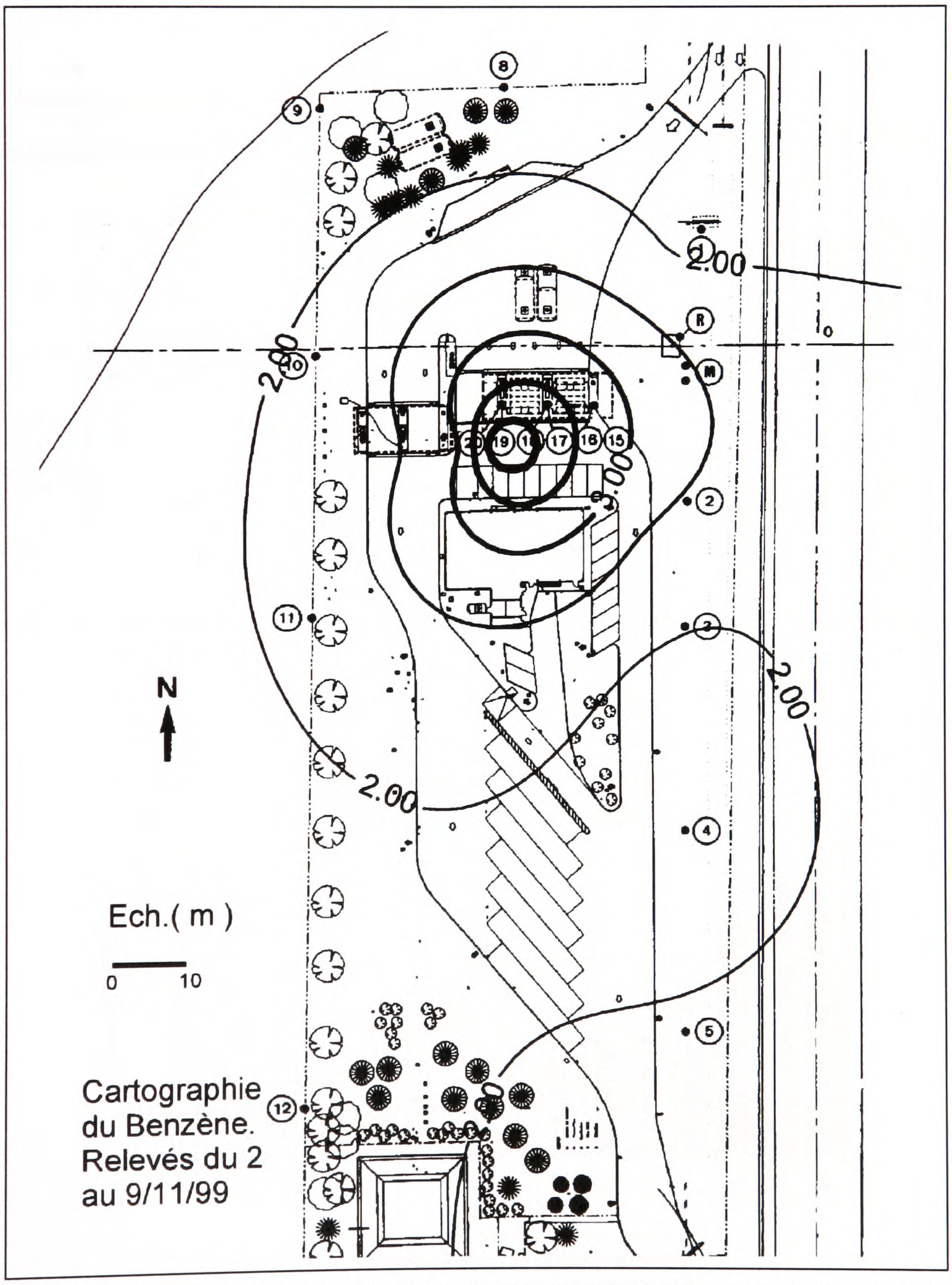

Fig. 1: Benzene iso-concentration map (Stage 2 control working). R: trailer-lab; M: meteorological monitoring equipment. Sampling sites: 1,2,3. Concentrations expressed in µg/m³.

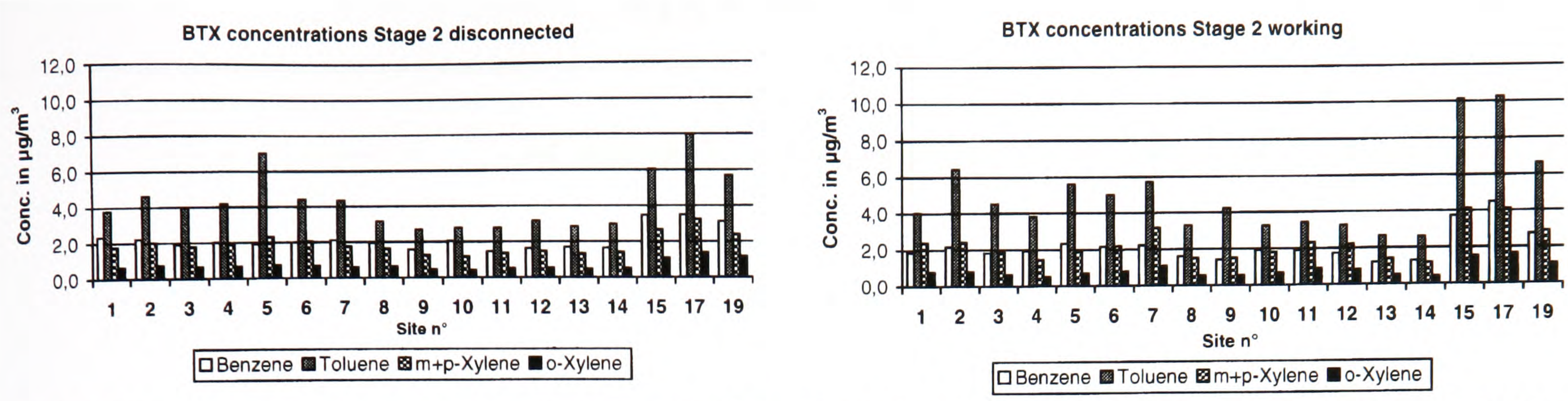

Fig. 2a,b: BTX concentrations. Level I: sites 15, 17, 19. Level II: sites 1-12. Level III: sites 13, 14 (a: Stage 2 control disconnected, b: Stage 2 control working).

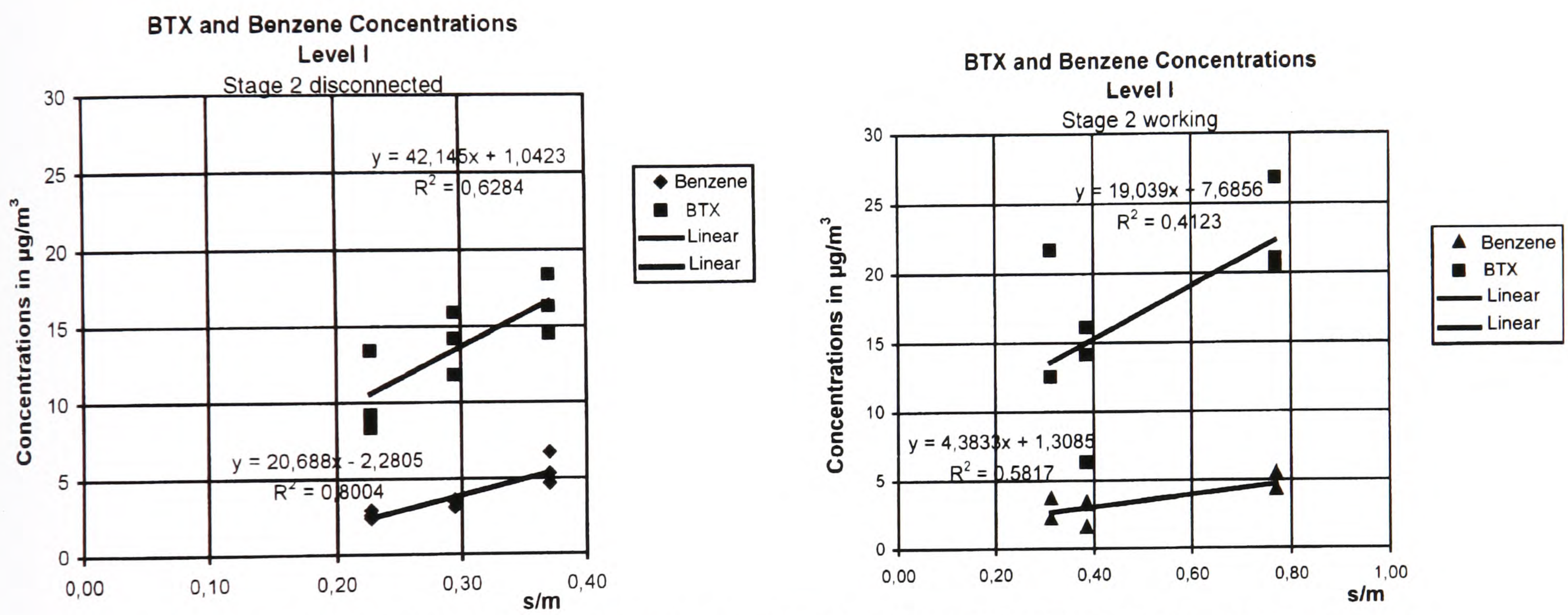

Fig. 3a,b: BTX and benzene concentrations vs. 1/wind speed (a: Stage 2 control disconnected, b: Stage 2 control working).

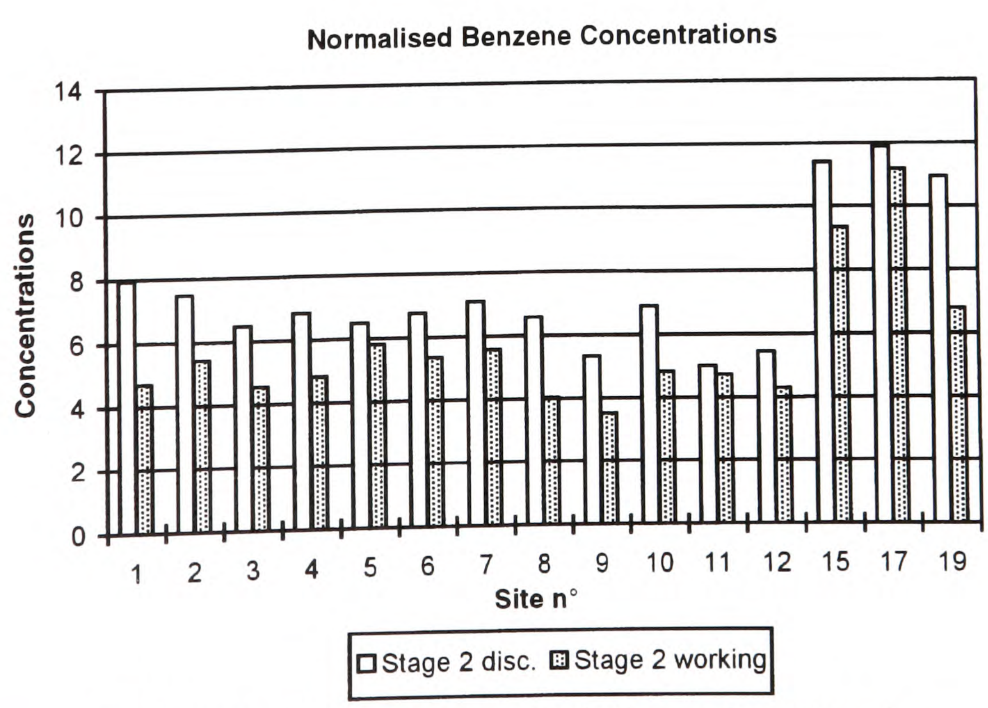

Fig. 4: Benzene concentrations normalised with respect to wind speed.

Appendix V

Conference Proceedings

Eighth International Conference on Air Pollution
Cambridge, UK, 24-26 July 2000, WIT Press, pp. 505-513

Air quality monitoring and modelling techniques for street canyons: the Paris experience

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Abstract

A better understanding of the dispersion and transformation of atmospheric pollutants in urban micro-environments is required to address the increasing public concern about human exposure in such areas. A joint research program has been established between INERIS (France) and University of Greenwich (UK) with the aim of developing efficient air quality monitoring and modelling methodologies to cover the needs of public health and road traffic managers in Europe.

An intensive monitoring campaign was conducted at a representative canyon street in Paris in winter 1998. This experiment was designed to establish the spatial and temporal variation of pollution within the canyon, and test readily available dispersion models. Active and passive techniques were used to sample a wide range of traffic generated pollutants (VOC and inorganic gases) at different heights and distances from the kerb. Local meteorological and traffic information was also obtained. The observed CO and NO concentrations were compared with predicted values, calculated using AEOLIUS, the street canyon model developed by the UK Meteorological Office.

The results demonstrate strong spatial pollution gradients within the canyon, large differences between roadside and background pollution levels, and pronounced temporal variability.

1 Introduction

In recent years, the increasing public concern about the adverse health effects of atmospheric pollution has led to the revision of air quality legislation at national and European level. Consequently, more stringent regulations have been proposed by the European Commission for several toxic gases mainly induced by road traffic (CO, NO_2, benzene, ozone, etc.)

In most European countries, municipal authorities have an important role in ensuring that the air quality objectives are achieved. Particularly in the UK, the Local Air Quality Management legislation (Part IV of the Environment Act 1995) requires local authorities to conduct periodic reviews and assessments of air quality. In the cases where the objectives specified in the Air Quality Regulations 1997 are not likely to be met by the end of 2005, an Air Quality Management Area (AQMA) has to be declared. In such a case, the local authority has to carry out an assessment of present and future air quality in the area concerned, and prepare an action plan in order to reach the objectives [1].

The compliance with regulatory standards is one of the tasks related to air quality that local administration has to undertake. In addition to that, health risk assessments, traffic management and transport planning studies need to be carried out on a regular basis. To manage these tasks, competent authorities need efficient monitoring and modelling tools and methodologies to determine pollutant concentrations in ambient air. As far as actions need to be taken at local level, these tools should produce reliable results at a low acquisition and operational cost. Furthermore, they should be user-friendly and well documented.

Within this context, particular attention should be given to those environments where population exposure to atmospheric pollutants is likely to be significantly higher than the urban average (e.g. busy streets and intersections, highways, petrol stations or industrial plants). A series of air quality monitoring campaigns is being conducted in Paris by the *Institut National de l'Environnement Industriel et des Risques (INERIS - France)* with the collaboration of the *University of Greenwich (UK)*, aiming to develop a standard method for the characterisation of such environments.

The experimental approach adopted in this study comprises: (a) the use of mobile air quality and meteorological monitoring equipment for the spatial and temporal characterisation of urban micro-environments; (b) the application of simple dispersion models for assessing their potential as predictive tools.

2 Field experiment

2.1 Site description

A field campaign was carried out in December 1998 in Bd. Voltaire, a typical four lane avenue of Paris with wide pavements and car parkings on both sides of the street. The part of Bd. Voltaire which was selected for this experiment constitutes a nearly perfect canyon with uniform buildings lining up continuously on both sides, height-to-width ratio of 0.8, and no major intersections along a straight road segment of approximately 300 m. Traffic lights were operating at both ends of the canyon, and there was a pedestrian crossing at a distance of 34 m from the main sampling point. The street axis bearing from the north was 140° and therefore nearly perpendicular to the westerly winds prevailing in the region during the measurements.

2.2 Measurement techniques

Parallel techniques, both active and passive, were used during one week to sample a wide range of traffic related atmospheric pollutants (CO, NOx, O_3, VOC). For the same period of time, local meteorological and traffic data were also collected.

Continuous CO, NOx, and O_3 analysers based on radiation absorption principles were used for the description of the temporal variation of pollution levels in the street. These analytical instruments were sheltered in a trailer, which was parked on the east side of the road.

Diffusive VOC samplers were located at different heights on both sides of the canyon in order to reveal the spatial variability of the compounds. For the estimation of the urban background contribution to the concentrations measured in the street, passive samplers were also placed in an adjacent park location.

Active sampling for VOC was carried out during one day of the campaign. Ambient air was pumped for one hour periods at a constant flow rate through a tube filled with the appropriate adsorbent. The hourly VOC concentrations obtained using pumped tubes were directly comparable with the observed CO levels, since both measurements were carried out through the same sampling line.

Carbotrap-B Supelco [2] was used as adsorbent for passive and active VOC sampling. After removal from the tubes with thermal desorption, the VOC samples were analysed in the laboratory using Gas Chromatography (column type: CP-SIL 5CB, 50 m x 0.32 mm, 1.2 μm). A quality assurance programme, including sampling duplicates, blanks and instrument calibration with standard gases was followed during the sampling and analytical work.

A 3-D ultrasonic anemometer and a weather mini-station were used for meteorological monitoring. These instruments were situated on the top of a mast (of 3.7 m height) next to the trailer, at a distance of 8.5 m from the wall of the nearest building. The location of the sampling and monitoring equipment is shown in Figure 3. Traffic data were continuously recorded by an automated counter located at the end of the canyon.

3 Results

3.1 Correlation between CO and benzene

Most of the atmospheric pollutants generated by traffic in urban environments (e.g. VOC, CO) can be considered as inert compounds due to the very short distances between sources and receptors. For this reason, the proportionality between them is expected to be nearly constant in a specific area and for a period of time with no significant changes in vehicle fleet composition and traffic pattern. If a simple mathematical relationship expressing such a proportionality between CO and benzene is established, then it will be possible to estimate CO concentrations using benzene measurements and vice-versa on the basis of this relationship [3].

In order to establish such a relationship, active VOC sampling was performed in Bd. Voltaire during one day of the campaign, simultaneously with CO monitoring and through the same sampling line. Figure 1 shows the hourly variation of the detected benzene, toluene, m,p-xylenes, and CO concentrations. Figure 2 illustrates the linearity between CO and benzene measurements (correlation coefficient = 0.93, slope = 3.97×10^{-3}, and intercept = 0.15×10^{-3}). The empirical formula obtained from this regression, benzene (ppb) = 4 $\times$ CO (ppm), can be consequently used for the calculation of CO levels at all locations where benzene measurements are available, or for the estimation of benzene variation with time. This simple methodology makes use of the practical advantages of passive-active VOC sampling (low cost and autonomy-portability of samplers) and continuous CO monitoring (accuracy and high time resolution) to provide a detailed temporal and spatial description of the pollution levels in an urban canyon.

3.2 Spatial and temporal variability

Using benzene as an indicator, strong pollution gradients were identified in the horizontal and vertical sense within the canyon. Differences of more than 2 ppb of benzene were observed at street level between the two opposite sides of the canyon, and of more than 1 ppb between roadside and urban background (Figure 3). A hot-spot of benzene (weekly average: 4.5 ppb) was detected on the leeward (up-wind) side of the street at 4.2 m height. In addition to the horizontal pollution gradients, a significant reduction in benzene concentrations along with the height was also observed. Very similar trends were identified for the rest of the VOC compounds sampled with passive tubes during the campaign.

While benzene measurements described the spatial variability of pollution in the street, CO continuous monitoring provided detailed information on the temporal variability of inert pollutants. Two peaks of CO were observed during the campaign. The first one (16th December) can be explained by the presence of low wind conditions (wind speed $\leq$ 2.5 m/s) in the region, and the second one (17 December) by the presence of relative low winds (2.5 - 3.0 m/s) blowing from directions parallel to the street axis (Figure 4).

4 Model simulations

The observed CO and NO concentrations were compared with predicted values, calculated using AEOLIUS, the street canyon model developed by the UK Meteorological Office [4]. This model is designed to calculate a series of hourly concentrations of a pollutant at a single receptor location on either side of the street. It requires synoptic meteorological data, traffic information, emission factors, and description of the topography [5].

AEOLIUS is based on the OSP Model [6], which calculates the concentration of pollutants in the street as the sum of three components: the contribution from the direct flow of pollutants from the source to the receptor, the recirculation component due to the flow of pollutants around the horizontal wind vortex generated within the canyon, and the urban background contribution. The direct component is calculated by the model using a simple plume dispersion algorithm, while the recirculation component is calculated using a box model formulation. Finally, the background contribution is an additive term introduced by the user.

In this application, the diurnal variation of measured CO and NO concentrations was clearly reproduced by the model (Figures 5 and 6). Nevertheless, the model predictions were quantitatively satisfactory only after introducing a constant fitting parameter. Given the strong pollution gradients in the street, this adjustment may be justified by the fact that the model was not configured to calculate the concentration of pollutants at the exact location where the measurements were taken. In addition to that, it is expected that the predicted concentrations would be closer to the observed values if locally measured (above-roof) wind data had been used for running the model. Finally, AEOLIUS appears not to be able to reproduce the measured peaks of pollution on an hourly basis, which is expected as models are rarely able to capture extreme pollution events [7].

5 Conclusions

A combination of air quality monitoring and modelling techniques has been proposed for assessing air quality in urban canyons. During the presented campaign, the simultaneous use of passive and active sampling systems provided a detailed description of the temporal and spatial variation of atmospheric pollution in the street and its vicinity. The strong horizontal and vertical concentration gradients may raise questions about the representativeness of routinely obtained air quality data from fixed monitoring stations.

The empirical CO-benzene relationship established for the specific environment can be used for achieving a better time and space resolution of roadside concentrations for both compounds. AEOLIUS can give reliable CO and NO predictions when it is carefully calibrated by measurements.

Following these air quality monitoring-modelling techniques, an optimum use of resources can be achieved. The relatively low cost of monitoring devices (e.g. passive and active tubes), the public domain software (AEOLIUS), and the limited amount of computational time and user expertise involved, make this method a cost-effective tool for the determination of air quality levels in urban canyons.

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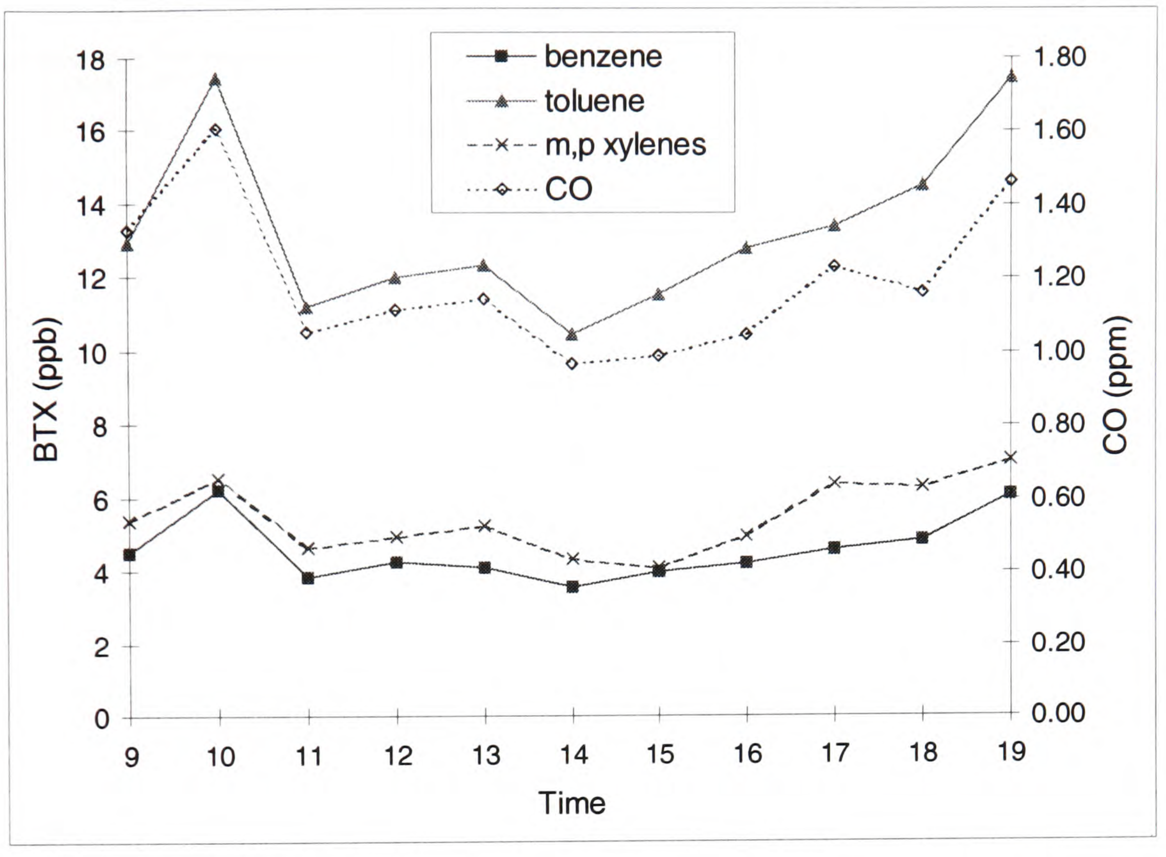

Figure 1: Daily variation of benzene, toluene, m,p-xylenes, and CO

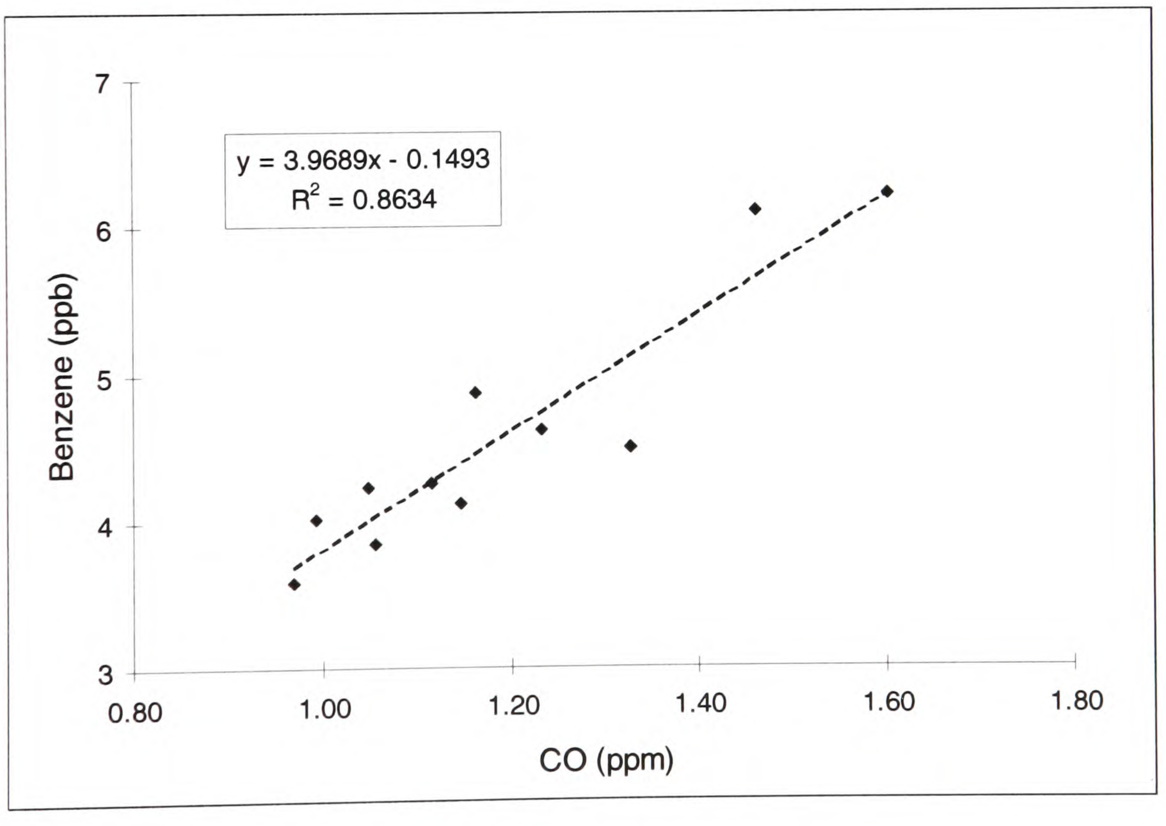

Figure 2: Benzene against CO

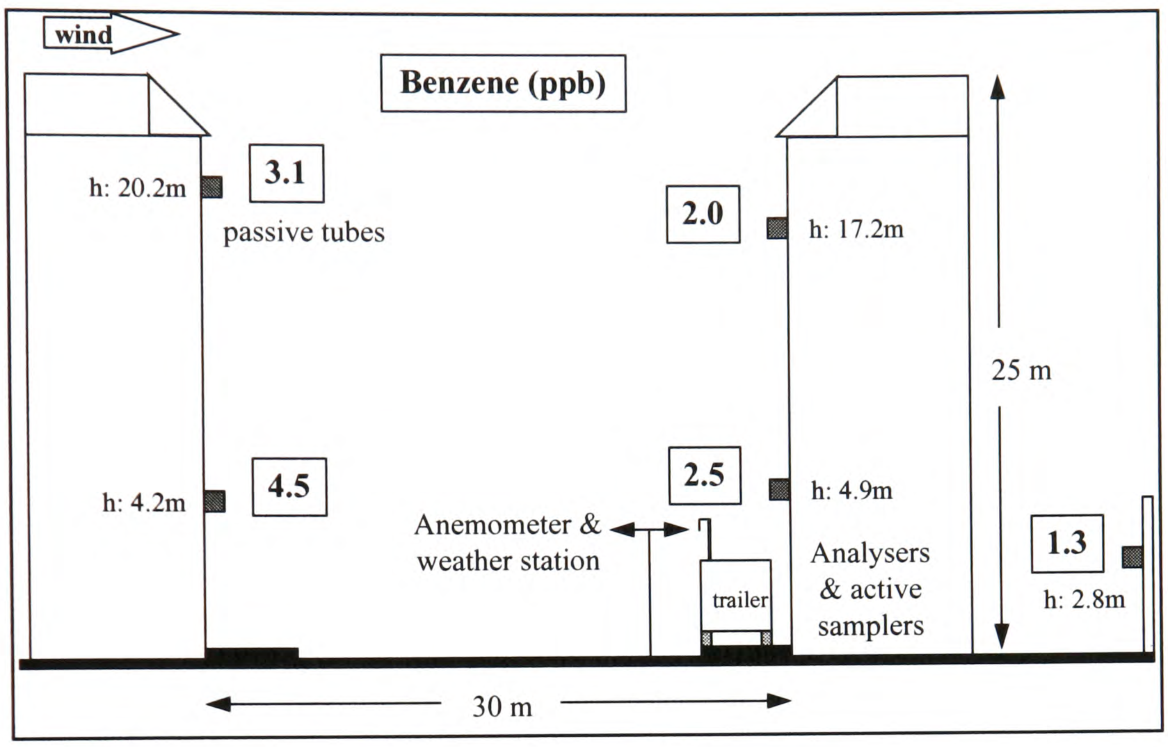

Figure 3: Spatial variability of benzene in Bd. Voltaire (Paris)

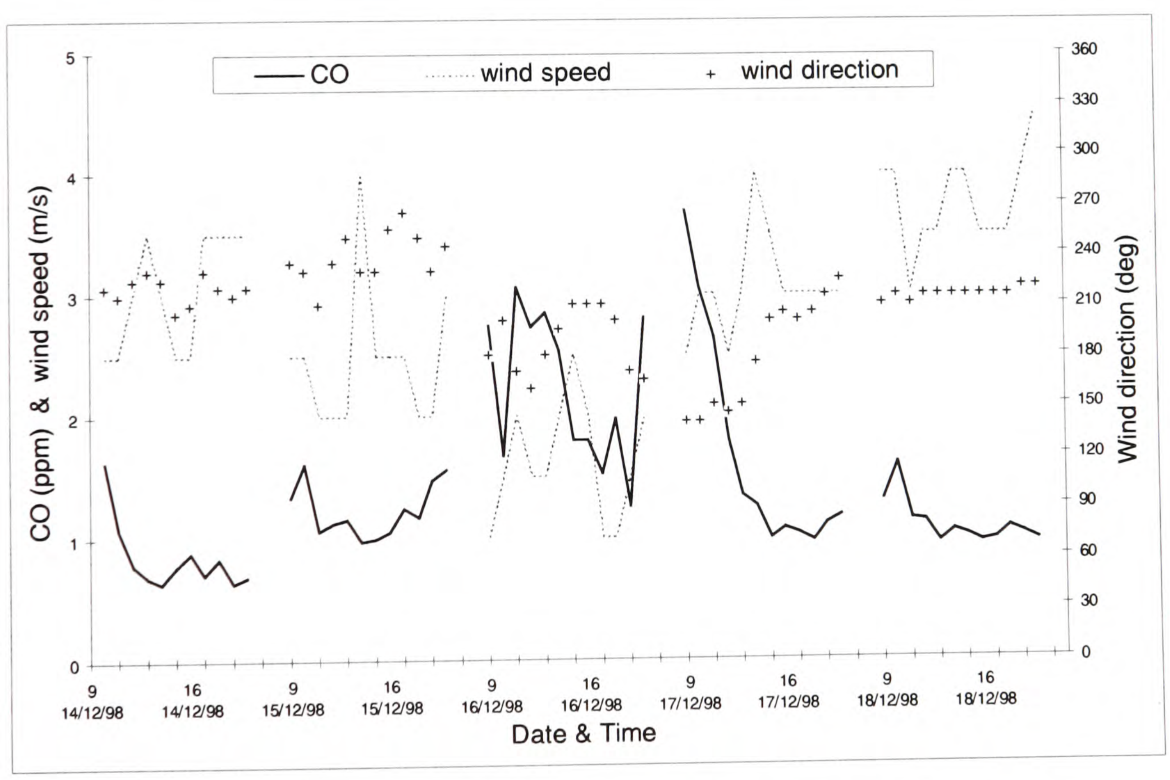

Figure 4: Time series of CO, with above-roof wind and direction

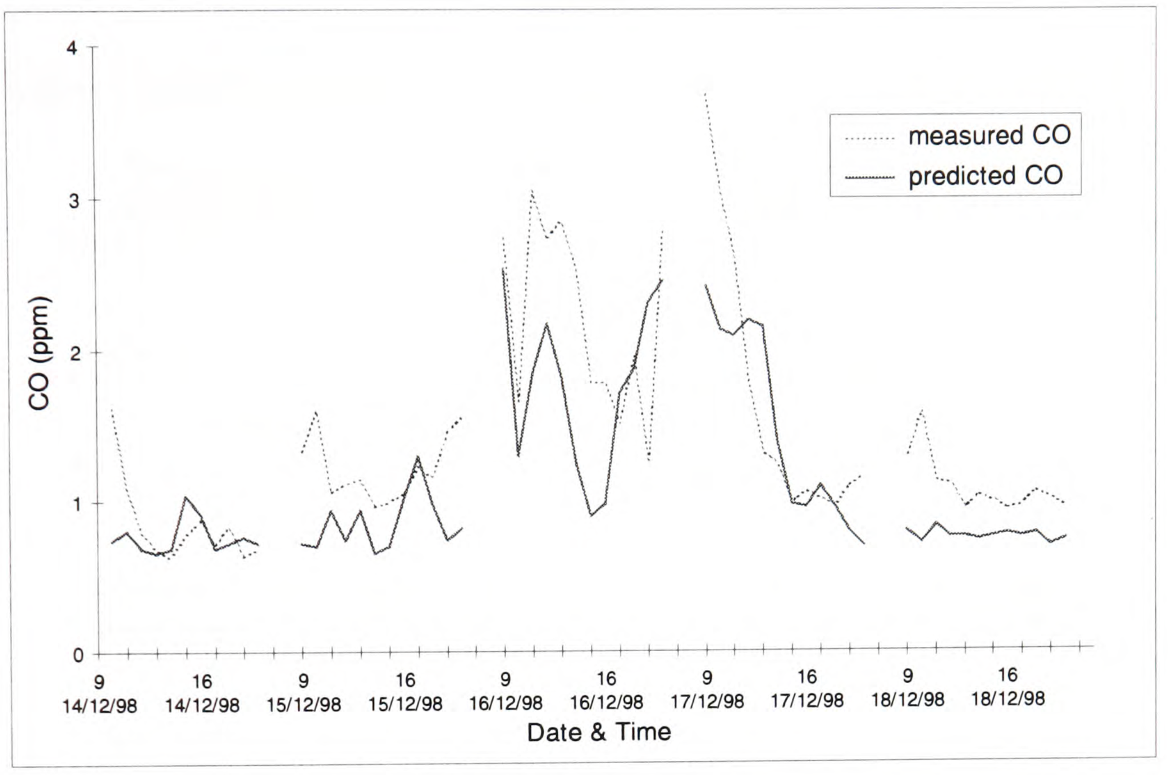

Figure 5: Time series of measured and predicted CO

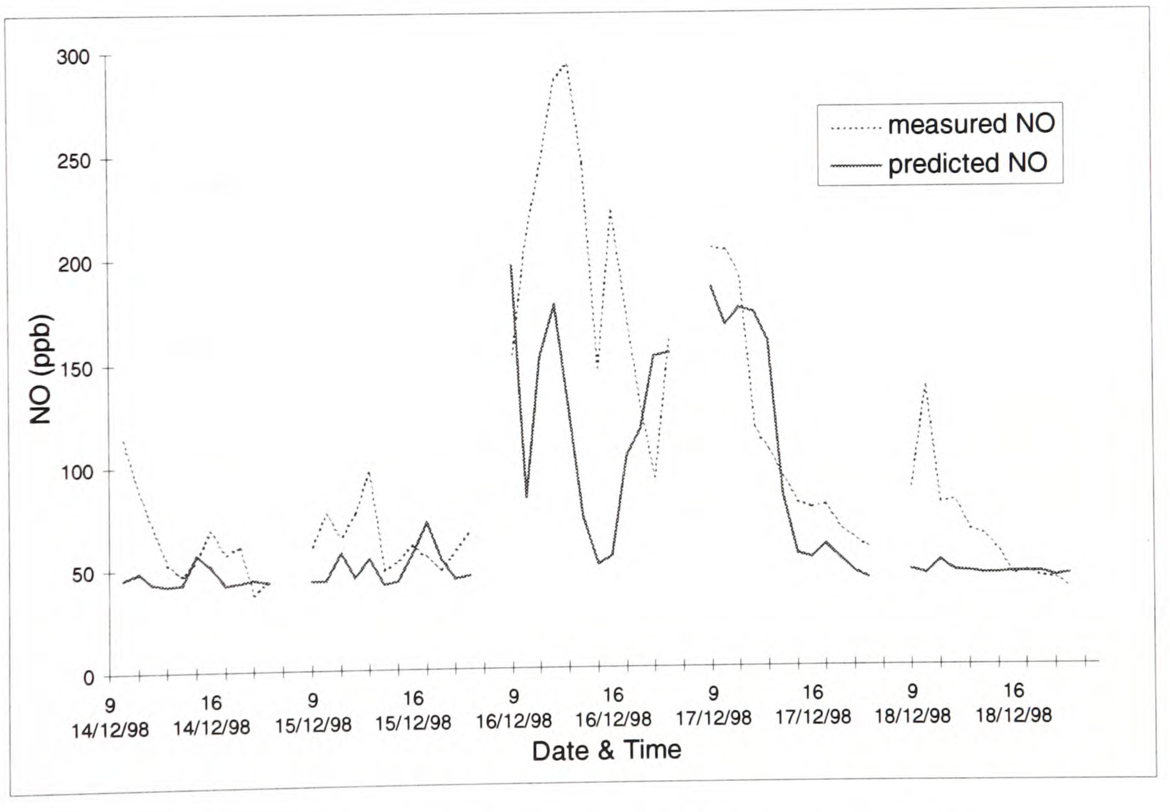

Figure 6: Time series of measured and predicted NO

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Diffusive sampling for the validation of urban dispersion models

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Abstract: Although widely used in workplace, indoor and ambient air quality assessments, diffusive sampling has not yet been established as a common tool for dispersion model validation. In this study, three mathematical models (STREET, OSPM and AEOLIUS) that are likely to be used for regulatory purposes were validated against experimental data obtained in two street canyons in Paris. Passive tubes were used to sample a wide range of traffic-related organic compounds at different heights and distances from the kerb. Model input information (site geometry, meteorological and traffic data) was obtained from the competent authorities and compared with on site observations. An algorithm describing vertical pollutant dispersion and an empirical relationship between CO and benzene were used. Diffusive sampling might be seen as a practical and cost-effective method for creating data sets for dispersion model validation.

Keywords: Model validation; Diffusive sampling; Street canyon; Benzene

1. Introduction

Nowadays, several street canyon models of different levels of sophistication are commonly used by local authorities, air quality networks, and research institutions in Europe. Most of these models have been parameterised and validated against real time measurements obtained from roadside monitoring stations.

However, these one-site continuous measurements do not reflect the strong spatial variability of traffic pollution revealed by a number of recent studies [1,2]. This variability, which might have serious implications in terms of population exposure, can be efficiently monitored using diffusive sampling. Furthermore, it may be reproduced by adequately validated dispersion models.

Diffusive sampling has become a popular method for assessing air quality due to a number of practical advantages (e.g. no need for power supply, portability of samplers, etc.) Although widely used in workplace, indoor and ambient air quality studies [3,4], it has not yet been established as a common tool for the validation of dispersion models.

The objective of this paper is to present a model validation method involving multisite diffusive sampling. This method was applied to three street canyon models (STREET, OSPM and AEOLIUS) that were validated against experimental data obtained at two different urban sites in Paris, France.

2. Experimental

2.1. Monitoring sites and equipment

Two air quality monitoring campaigns were conducted in street canyons in Paris during winter (Bd. Voltaire, December 1998) and summer (Rue de Rennes, July 1999). The two sites were busy four lane streets with large pavements and uniform buildings lining up continuously on both sides. The height-to-width (H/W) ratios for Bd. Voltaire and Rue de Rennes were approximately equal to 0.8 and 1.1, and the average traffic volumes during measurements were 30,000 and 23,000 veh/day, respectively.

Active (i.e. pumped) and passive (i.e. diffusive) tubes were used to sample benzene, toluene, xylenes (BTX) and other volatile organic compounds (VOC) in both the canyons. CO, NO_x and O_3 were continuously monitored using infrared, chemiluminescence and ultra-violet analysers, respectively. Local meteorological parameters were measured at street level and compared with synoptic weather information obtained from a permanent monitoring station located in Montsouris park, within few km distance from the experimental sites. Hourly traffic volumes and average vehicle speeds were obtained from automatic counters operating in both the streets. Finally, the vehicle fleet composition was estimated from on site spot measurements.

2.2. Diffusive sampling and analysis

While active sampling was conducted at only one kerbside location in each canyon (height of inlet: 3.7 and 2.9 m in Bd. Voltaire and Rue de Rennes respectively), diffusive samplers were placed at several roadside and background locations, at different heights and distances from the kerb.

In Bd. Voltaire, passive tubes were located at two different heights (1st and 5th floor) near the walls of the canyon, and at one background site (Fig. 1). The devices remained exposed to ambient concentrations for five days. In Rue de Rennes, two different sets of passive tubes were used to examine separately BTX levels during weekend and working weekdays. A more detailed spatial resolution was obtained by increasing the number of sampling locations. In this case, apart from measurements near the walls of the canyon, samples were also taken on the kerbside (h=1.5 m), and at two different background sites (Fig. 2).

The diffusive sampling was carried out using Radiello Perkin Elmer axial tubes filled with Carbotrap-B and sheltered in aluminium boxes [5]. After removal from the tubes with thermal desorption, VOC were analysed in the laboratory using Gas Chromatography (column type: CP-SIL 5CB, 50 m$\times$0.32 mm, 1.2 μm) + FID. A quality assurance programme, including sampling duplicates and blanks was followed during sampling and analysis.

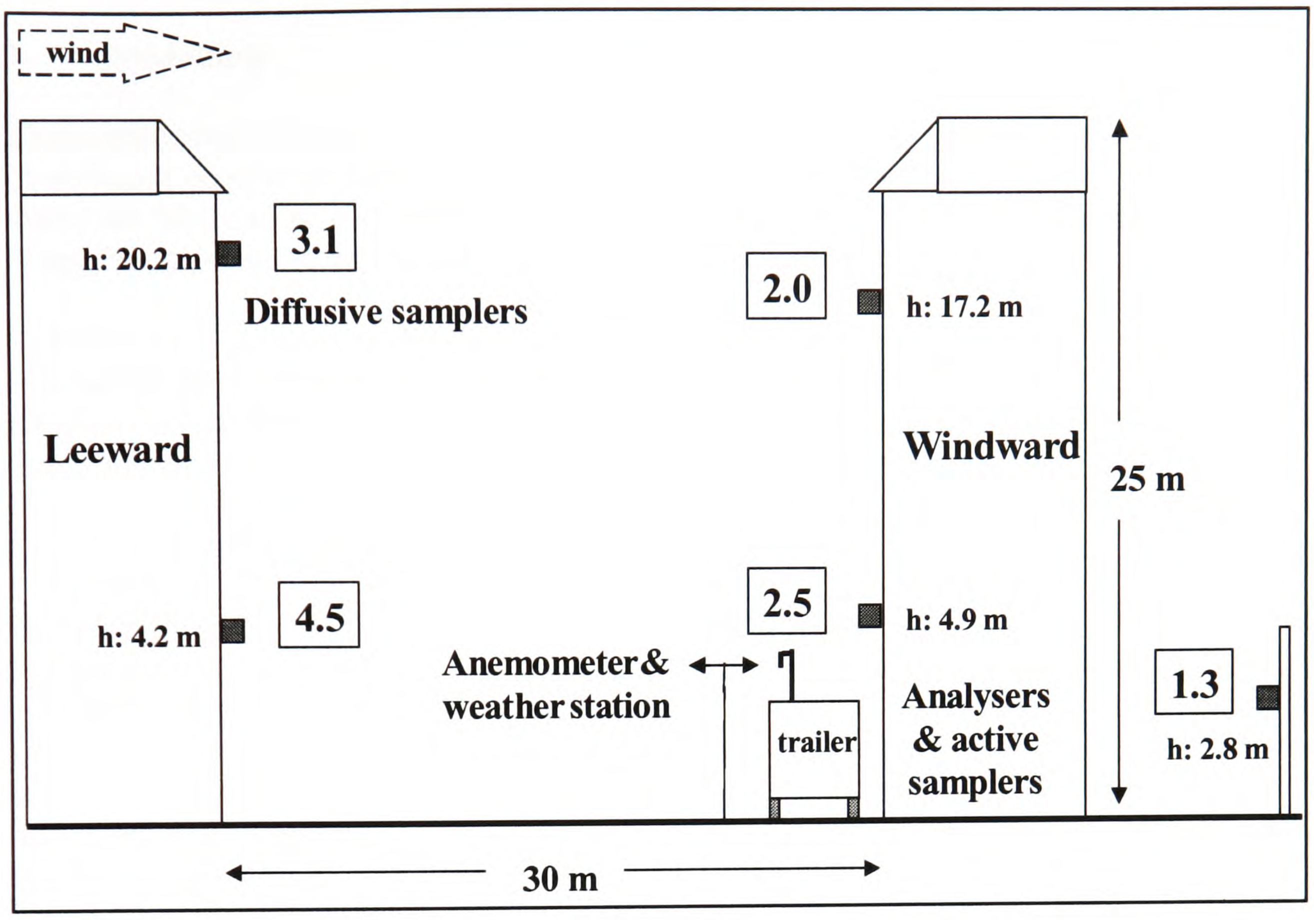

Fig. 1: Diffusive sampling for benzene (ppb) in Bd. Voltaire (14-18 December1998)

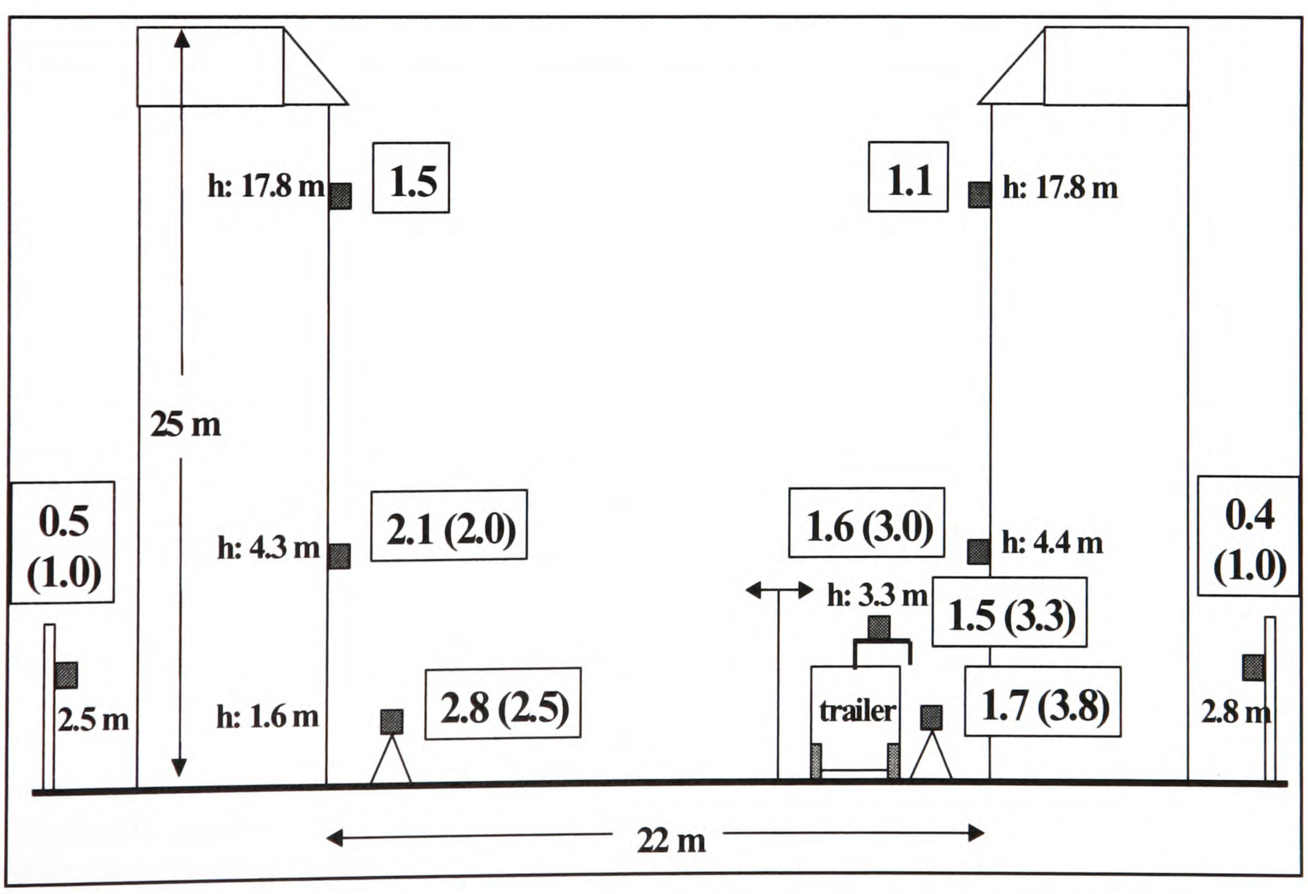

Fig. 2: Diffusive sampling for benzene (ppb) in Rue de Rennes during weekdays (19-23 July) and a weekend (16-18 July 1999, values in parenthesis).

3. Modelling

Three mathematical models, STREET, OSPM, and AEOLIUS, were used for the simulation of pollutant dispersion within the canyons. These relatively simple codes (or variations of them) are likely to be involved in a variety of applications including air quality and traffic management, urban planning, population exposure studies, etc.

STREET [6,7] is a box model that uses two different empirical algorithms to reproduce CO concentrations on either side (i.e. leeward and windward) of a street canyon. When the wind direction is parallel or near-parallel to the axis of the canyon, concentrations on the two opposite sides of the street become equal and they are calculated by averaging the results from the two algorithms. The final CO values are obtained by adding the urban background contribution to the kerbside concentrations.

OSPM [8] is a semi-empirical code, which was evolved from the CPBM model [9]. It is designed to produce series of hourly pollutant concentrations at a single receptor location on either side of a street canyon. It assumes three different contributions to the kerbside levels: (a) the contribution from the direct flow of pollutants from the source to the receptor, (b) the recirculation contribution due to the flow of pollutants around an horizontal wind vortex generated within the so called recirculation zone of the canyon, and (c) the urban background contribution. The direct component is calculated applying Gaussian dispersion theory, while a box model algorithm gives the recirculation component. On the leeward side of the street, concentrations are calculated as the sum of the direct and recirculation contributions, while on the windward side, only the direct contribution of emissions generated outside the recirculation zone are taken into account. If the recirculation zone extends throughout the whole canyon, then the windward concentrations are calculated from only the recirculation component. When the wind speed is near zero or parallel to the street axis, the concentrations on both sides of the canyon become equal.

These two models, STREET and OSPM, have been used in many scientific and engineering applications [10,11]. AEOLIUS is a more recent model based on the same formulation as OSPM and mainly used in the U.K. [12].

In STREET, the user externally defines the height (z) of the receptor and its distance from the kerb. By contrast, OSPM and AEOLIUS produce pollutant concentrations only at street level ($\approx$ 2 m), without giving the user the possibility of choosing the height of the receptors. This limitation was overcome by introducing an algorithm that enables the user to establish vertical pollution profiles in the street [13]:

$$C(z) = C_r \exp\left(-\frac{z - z_r}{H}\right) \qquad (1)$$

where C_r is the concentration of the pollutant at a reference height z_r on either side of the canyon (H: height of the canyon). Furthermore, an empirical relationship was introduced, so as to allow the calculation of benzene concentrations from CO predictions:

$$\text{Benzene (ppb)} = \alpha \cdot \text{CO (ppm)} \qquad (2)$$

The proportionality constant α was experimentally derived from simultaneous BTX and CO measurements in both the canyons (3.8 and 3.7 in Bd. Voltaire and Rue de Rennes, respectively).

As inputs, all three models required synoptic wind information, traffic and emission data, as well as the dimensions of the canyons. The rate of release of CO in the street was calculated from hourly traffic volumes and emission factors, which were derived from the site-specific vehicle fleet composition [14]. The relative pollutant contributions from the street and the background were derived from diffusive benzene measurements.

4. Results and discussion

STREET, OSPM and AEOLIUS were initially used to simulate hourly CO averages in Bd. Voltaire and Rue de Rennes. The results showed a good agreement with the continuous CO measurements in both the streets [13,15]. In the present study, the three models were further validated against diffusive benzene measurements.

Applying the empirical relationship (2) to the CO concentrations calculated with STREET, average benzene values were produced for different receptor locations in the street over the passive sampling periods (i.e. 2 to 5 day averages) and added to the observed background concentrations. The comparison of the total calculated values with the observations showed a very good general agreement for Rue de Rennes (Fig. 3a), although the model seemed to slightly under-predict the low concentrations observed near the top of the canyon. For Bd. Voltaire, the model under-predicted all measured values (Fig. 3b).

Furthermore, relationship (1) was used to calculate CO concentrations at receptor heights corresponding to the passive sampling locations in Bd. Voltaire and Rue de Rennes, using the street level OSPM and AEOLIUS outputs as reference values. Applying relationship (2), average benzene values were obtained and added to the observed background concentrations. Despite some slight under-predictions in the case of Bd. Voltaire, OSPM[1] reproduced successfully the concentration profiles detected in the two canyons (Fig. 3c and 3d). AEOLIUS[1] gave also very good predictions for Rue de Rennes (Fig. 3e), but under-predicted the values observed in Bd. Voltaire (Fig. 3f).

The tendency of all three models to under-predict pollutant concentrations in the case of Bd. Voltaire (i.e. the models under-predicted the CO concentrations from which the benzene averages were calculated) might be attributed to under-estimated CO emissions in this street. More diffusive benzene measurements from other urban canyons are needed for further validating the models as well as the empirical relationship (1). It should be also emphasised that this expression is not applicable to traffic-related pollutants with very short chemical lifetime, like NO_2, which can be also sampled with diffusive tubes. It has been experimentally demonstrated [16] that NO_2 concentrations may even increase along with height within a street canyon, when the weather conditions favour photochemical activity.

The present study showed how diffusing sampling can be used to test the performance of urban dispersion models at different receptor locations within a street. While high spatial resolution might be achieved using passive tubes, the temporal resolution obtained from these measurements is relatively low (e.g. weekly averages). Therefore, the "spatial" validation of the models against diffusive sampling results should be coupled with

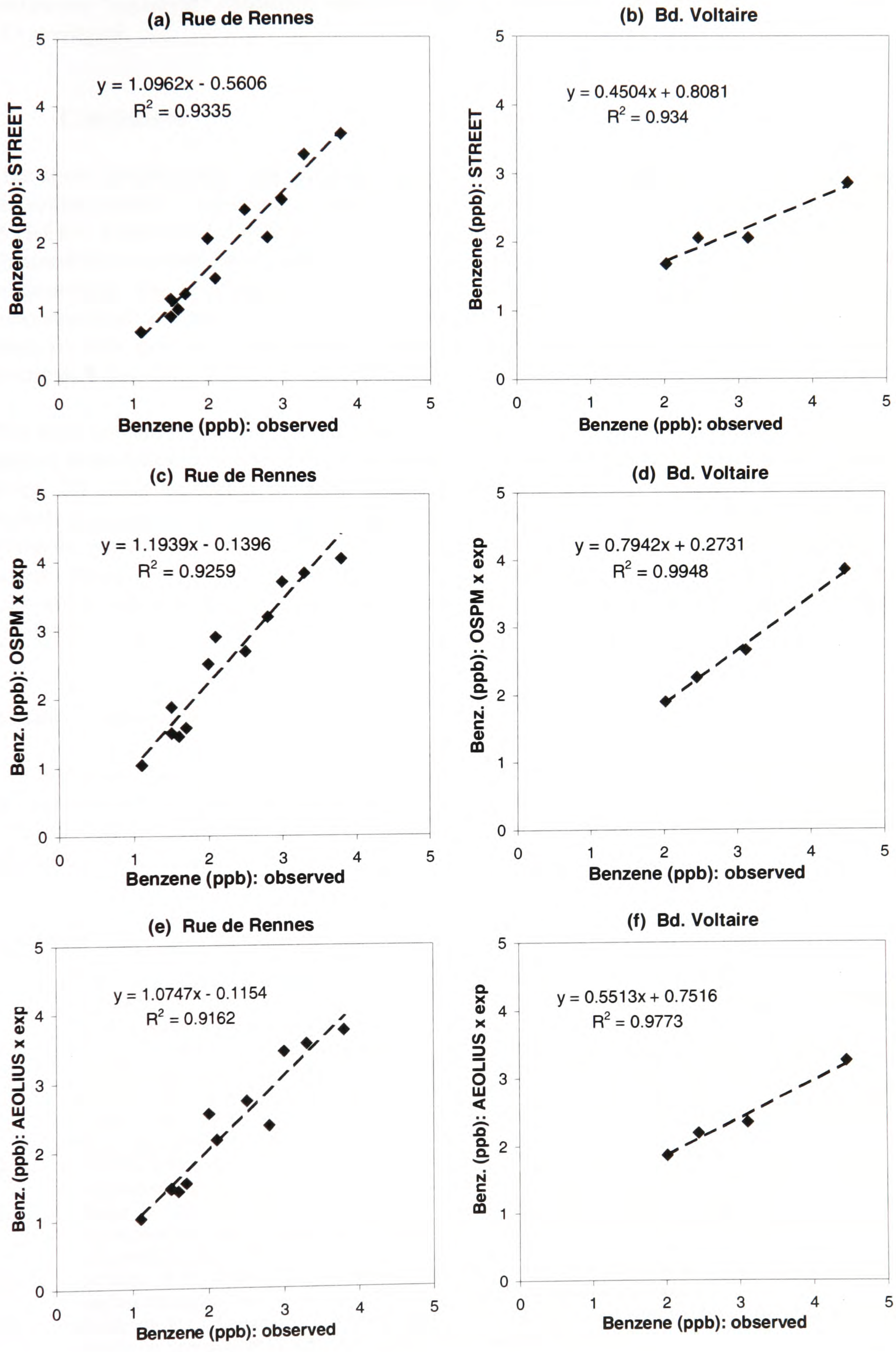

Fig. 3: STREET, OSPM and AEOLIUS predictions vs. observed benzene concentrations obtained with diffusive sampling in Rue de Rennes and Bd. Voltaire.

traditional "temporal" validation methods against continuous monitoring data (e.g. hourly CO averages).

5. Conclusions

This study demonstrated that diffusive sampling can be a useful tool for validating urban dispersion models. Following a relatively simple methodology, three mathematical models (STREET, OSPM and AEOLIUS) were validated against multisite benzene measurements obtained in two street canyons in Paris. This methodology involved the use of two empirical relationships: the first one for reproducing vertical pollution profiles using street level concentrations, and the second one for calculating benzene values from CO predictions. All three models gave very satisfactory benzene estimates for Rue de Rennes, but under-predicted (especially STREET and AEOLIUS) the concentrations measured in Bd. Voltaire.

The three models, STREET, OSPM and AEOLIUS, had already been validated in the past against continuous measurements from urban air quality monitoring stations. In the present study, the main advantage of using diffusive sampling was that it enabled us to test the models for different receptor locations within the same street. Most importantly, that was achieved without investing a great amount of resources (e.g. sophisticated instrumentation, power supply, etc.) For this reason, it is believed that diffusive sampling will be increasingly used in the future as an alternative technique for creating data sets for model validation.

Acknowledgements

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Louvain-la-Neuve, Belgium, 15-19 October 2001

ESTIMATES OF UNCERTAINTY IN URBAN AIR QUALITY MODEL PREDICTIONS

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1. INTRODUCTION

The deterministic approach of running a single dispersion model using a specific input data set of fixed values has been traditionally adopted in air quality management. In some cases more than one model has been involved in inter-comparison studies and different emissions scenarios have been taken into account in order to assess future air quality trends. However, most of the modelling work has been so far oriented towards deterministic simulations of ambient air pollution. This approach does not reflect the uncertainties attached to the input data, model formulation and stochastic variability of the atmosphere. Given the complexities of urban environments and the inherent limitations of mathematical modelling, it is unlikely that a single model based on routinely available meteorological and emission data will give satisfactory short-term predictions. Moreover, deterministic air quality modelling might lead in same cases to erroneous decisions with serious financial and social implications when used for regulatory and planning purposes.

In this study, a method involving the use of more than one urban dispersion model, in association with different emission simulation methodologies and meteorological input data from different sources, is proposed for predicting best CO and benzene estimates, and related confidence bounds. This method was tested using experimental data obtained during intensive monitoring campaigns in busy street canyons in Paris, France. Three relatively simple dispersion models (OSPM, AEOLIUS and STREET), which are likely to be used for regulatory purposes, were selected for this application. The comparison between simulated and observed concentrations demonstrated the advantages and limitations of this approach. Without resorting to sophisticated statistical techniques, this

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study explores practical methods for quantifying and presenting model uncertainties in a way that can be easily understood by local authority officers and general public.

2. INPUT DATA

Two comprehensive data sets were created from field experiments conducted at representative street canyon sites in Paris during winter (Bd. Voltaire, December 1998) and summer (Rue de Rennes, July 1999). The two sites are busy four lane streets with large pavements and uniform buildings lining up continuously on both sides. The height-to-width (H/W) ratios for Bd. Voltaire and Rue de Rennes were approximately equal to 0.8 and 1.1, respectively.

Traffic-related atmospheric pollutants were sampled during five and eight days in Bd. Voltaire and Rue de Rennes, respectively. Continuous CO monitoring and diffusive benzene sampling was carried out in both the canyons throughout the respective campaigns. The relative street and background contributions to the pollution levels observed on the roadside were derived from benzene measurements. The experimental lay out and monitoring results from these campaigns were presented elsewhere (Vardoulakis et al., 2001).

Synoptic wind data and other meteorological information were obtained from three permanent monitoring stations operated by Meteo France. Two of them were located in urban settings within Paris: (a) park Montsouris, and (b) St. Jacques tower. The third station was located at (c) Orly airport, in approximately 12 km distance from central Paris.

The average traffic volumes in Bd. Voltaire and Rue de Rennes during measurements were 30,000 and 23,000 veh/day, respectively. Hourly traffic data were obtained from automatic counters permanently operating in both the streets. The vehicle fleet composition was estimated from on site spot measurements during the campaigns.

The rate of release of emissions in the street was derived from traffic volumes and composite emission factors. Two different methods were applied for calculating CO emission factors: (a) The protocol used by Buckland and Middleton (1999), and (b) the IMPACT road traffic emission model commercialised by ADEME (1998). This model uses COPERT II methodology (Ntziachristos and Samaras, 1997) to quantify fuel consumption and atmospheric releases of a specified vehicle fleet in a given year in France. The required input information includes traffic volume and composition, average vehicle speed, and length and slope of the road segment of interest. In addition, the month of the year is used to estimate average ambient temperatures, which are further used for calculating evaporative and cold running emissions. The model provides default values for the average travelling distance and the fraction of this distance run with a cold engine in France.

The estimated values from both the methods were compared for consistency with CO emission factors specific to the French vehicle fleet reported in other recent studies (Touaty and Bonsang, 2000; Jones et al., 2000). All values are summarised in Table 1.

Table 1: CO emission factors from road transport in the region of Paris

Author	CO emission factors (g/km veh)		
	Bd. Voltaire	Rue de Rennes	Paris
IMPACT (ADEME, 1998)	20.29	10.91	
Buckland and Midletton (1999)	7.23	8.73	
Touaty and Bonsang (2000)			8.11
Jones et al. (2000)			8.82

3. STREET CANYON MODELS

In this study, three parameterised models (STREET, OSPM, and AEOLIUS) were used to simulate pollutant dispersion within canyons. These relatively simple codes (or variations of them) have been widely used by local authorities and air quality networks in Europe. They are likely to be involved in a variety of applications including air quality and traffic management, urban planning, population exposure studies, etc.

Like most empirical street canyons models, they are based on the assumption that a wind recirculation zone is formed within the canyon when the roof-top wind blows perpendicularly or near-perpendicularly to the street axis (Berkowicz et al., 1997). This air vortex causes a downward flow of relatively clean air on the windward side of the canyon and an upward flow of air mixed with exhaust gases on the leeward side. This flow gives rise to high cross-road and vertical pollution gradients in the street.

Empirical models have been proved to describe quite efficiently concentration gradients in regular canyons, especially for the perpendicular wind situation. Nevertheless, more sophisticated modelling techniques might be needed to simulate dispersion under low wind conditions or in more complex urban environments (e.g. asymmetric and deep canyons, intersections, etc.). In these cases, the single vortex assumption does not appear realistic.

STREET and OSPM have been parameterised according to different field experiments and used successfully in many scientific and engineering applications (Johnson et al., 1973; Qin and Kot, 1993; Kukkonen et al., 2001). AEOLIUS is a more recent model based on the same formulation as OSPM and mainly used in the U.K. (Buckland, 1998).

4. QUANTIFYING UNCERTAINTY

The total uncertainty involved in modelling simulations can be considered as the sum of three components (Hanna, 1988): (a) The uncertainty due to errors in the model physics, (b) the uncertainty due to input data errors, (c) the uncertainty due to stochastic processes (e.g. turbulence) in the atmosphere.

STREET, OSPM and AEOLIUS were initially used in a traditional manner to produce estimates of roadside CO concentrations in Bd. Voltaire and Rue de Rennes. Three independent meteorological data sets and three different emission factors (as described in paragraphs 2) were used for each canyon, so as to create an ensemble set of 27 simulations for each case. It was considered that the use of different models enabled us to account, to a certain extent, for the uncertainty in model formulation. Furthermore, the use of independent wind and emission data sets was thought to represent the uncertainties due to errors in model inputs. This approach might be more realistic than defining a priori uncertainty ranges in different input variables.

The same methodology was followed for creating ensemble sets of weekly benzene averages corresponding to different receptor locations in both the canyons.

For quantifying the total uncertainty in model predictions, medians together with maximum and minimum concentrations were calculated for each set of 27 model outputs corresponding to a specific receptor location and time. The extreme concentrations were thought to bound the likely ranges of total uncertainty in the predictions. As it can be seen in Fig. 1 and 2, approximately 95% and 92% of CO observations in Rue de Rennes and Bd. Voltaire, respectively, fell within the predicted concentration ranges. In the case of benzene, all predicted values were within the estimated error bounds (Fig. 3 and 4).

5. DISCUSSION

In the previous examples, the observed concentrations fell in most cases within the estimated error (or "confidence") bounds. Nevertheless, it might be argued that these error limits were so large that they did not really provide any useful information. The fact is that, although large uncertainties do exist, dispersion models are still used in a traditional "deterministic" way, often returning results with several significant digits. It is, therefore, preferable to include error bounds (however large they may be) in the predictions in order avoid inappropriate reliance on modelling results.

A more rigorous approach would require that probability functions be developed for input and internal model parameter, and these be randomly sampled to obtain improved ensemble sets (Dabberdt and Miller, 2000). This, however, would risk to geometrically increase the time and consequently the cost of the simulations. It is believed that ensembles of less than 30 simulations, as shown in this study, can provide satisfactory air quality predictions and error bounds.

Model users some times assume that the total uncertainty in predictions decreases as the complexity of the model increases. This is true only for the uncertainty attributed to errors in the physical description of the model domain (e.g. incorrect assumptions, oversimplifications, etc.). On the other hand, sophisticated models require a larger amount of input information, which inevitably introduces a larger data uncertainty component in their calculations. It is, therefore, very important to define the appropriate degree of sophistication for a specific application, so as to achieve the lowest possible level of uncertainty in modelling results.

5.1. Uncertainty in emissions

It has been suggested by other authors (Kühlwein and Friedrich, 2000) that emission factors represent one of the most important source of uncertainty in modelling traffic pollution. That was confirmed in the present study. CO emission factors calculated with IMPACT were 25% and 180% higher than those calculated following Buckland and Middleton's methodology for Rue de Rennes and Bd. Voltaire, respectively (Table 1).

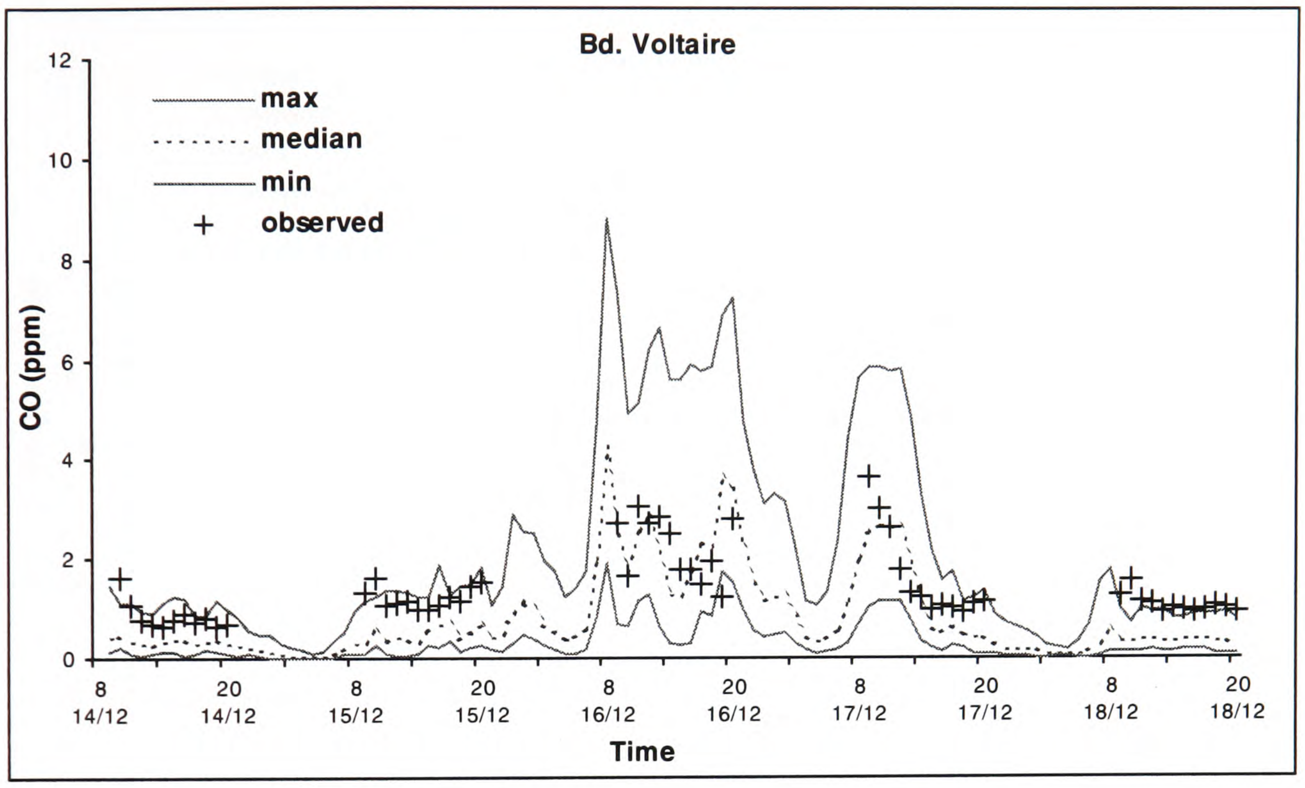

Fig. 1: Time series of CO best estimates (median), error bounds (max, min), and observed concentrations in Bd. Voltaire, Paris.

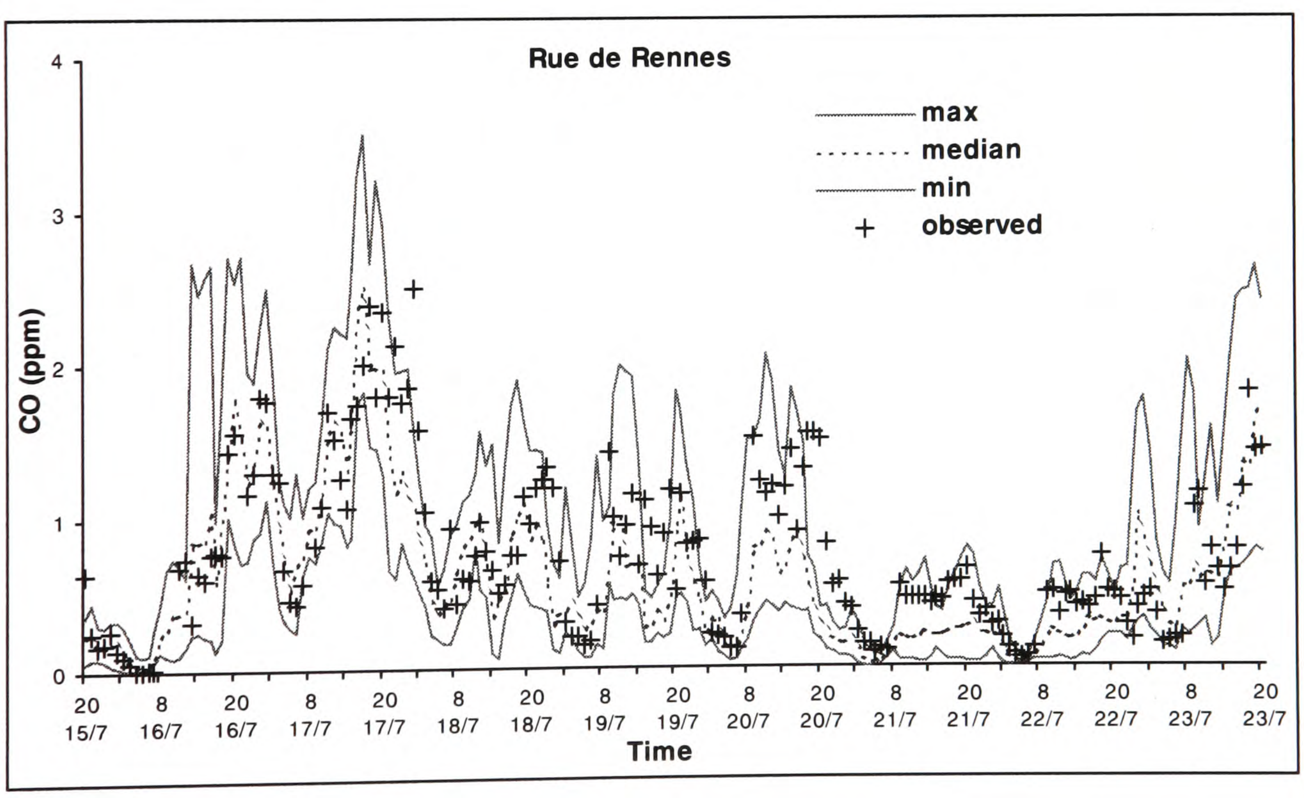

Fig. 2: Time series of CO best estimates (median), error bounds (max, min), and observed concentrations in Rue de Rennes, Paris.

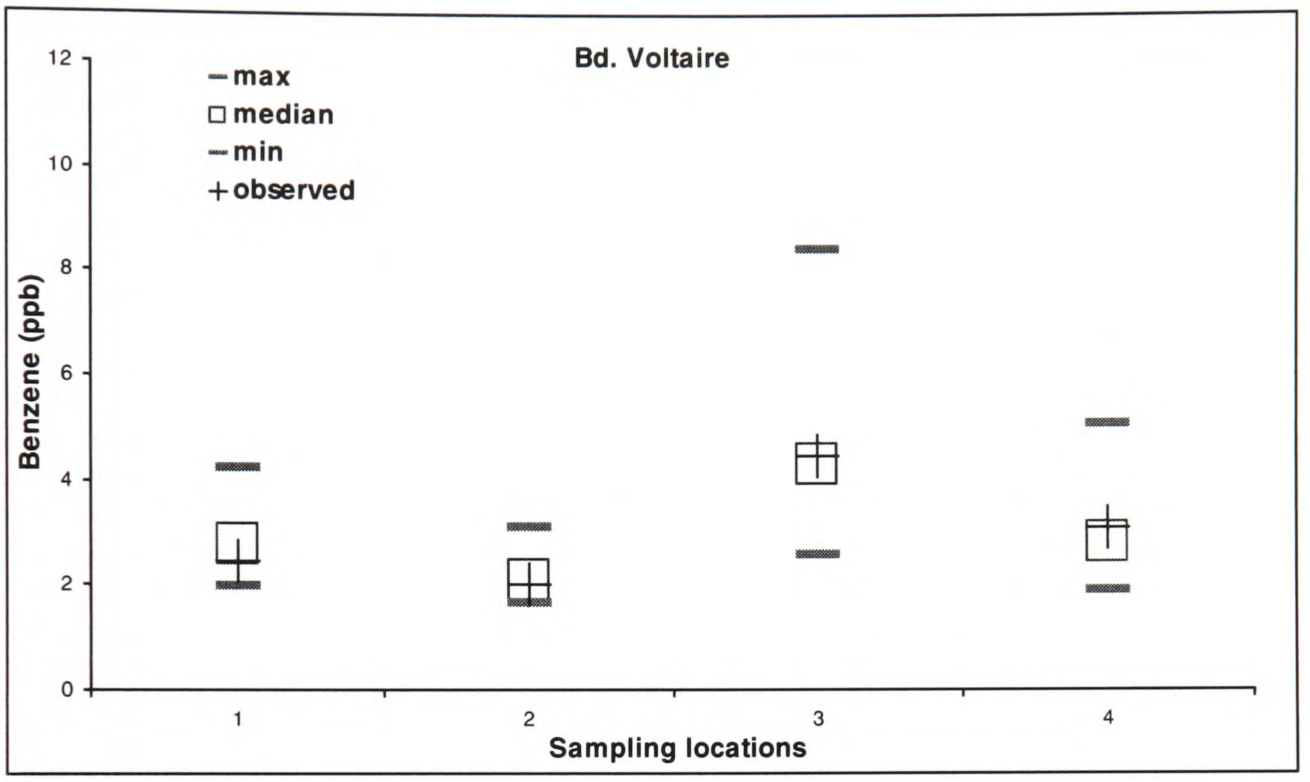

Fig. 3: Benzene best estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Bd. Voltaire, Paris.

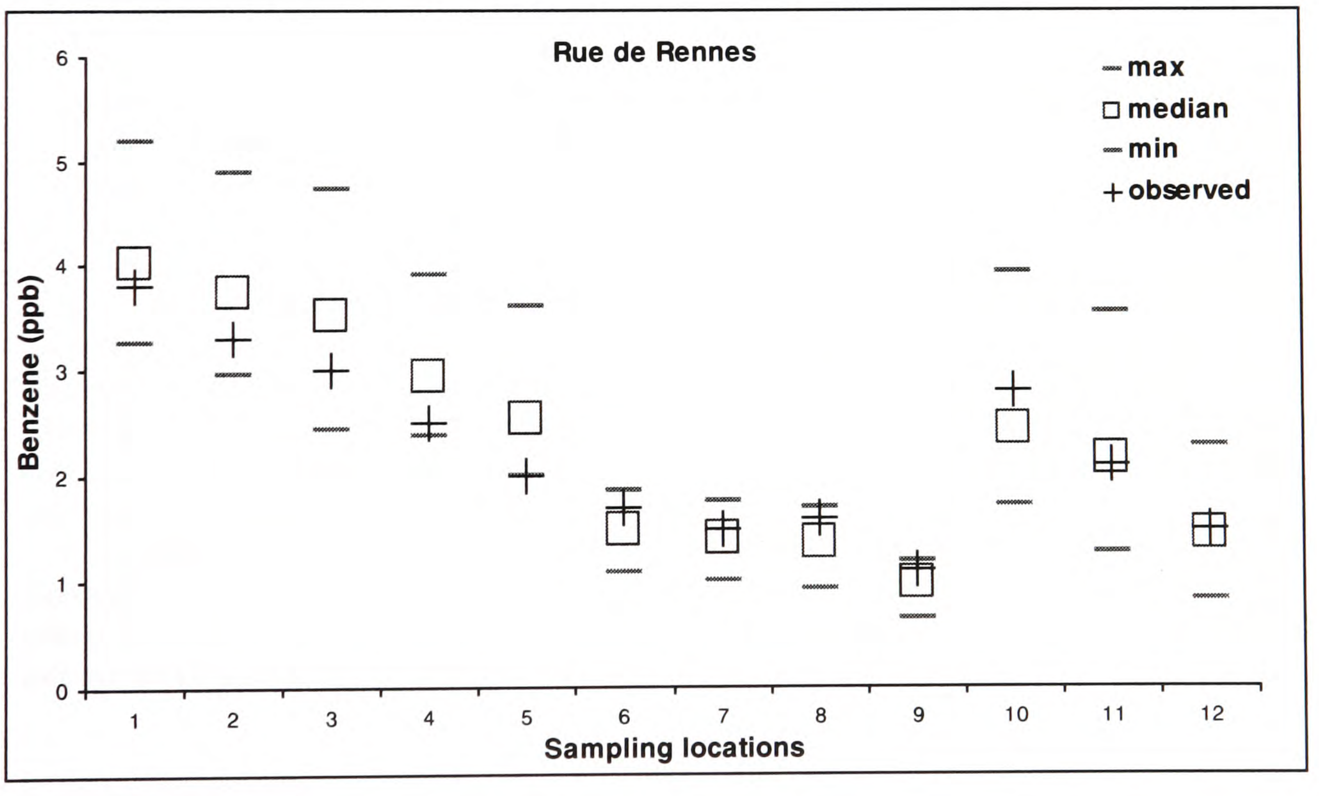

Fig. 4: Benzene best estimates (median), error bounds (max, min), and observed concentrations at different receptor locations in Rue de Rennes, Paris.

The large discrepancies, especially in the case of Bd. Voltaire, were probably due to the very significant cold start/running emissions calculated by IMPACT and added to the hot running values.

Cold start emissions are indeed expected to be important for urban driving conditions, because of the relative large number of short trips carried out with cold engines, especially in winter. In an on-road experiment in Belgium (De Vlieger, 1997), it was found that the average CO vehicle emissions measured during the cold phase were 4 to 40 times higher than emissions with a hot start (i.e. a hot engine + a warmed-up three way catalyst). Other experimental results showed significant seasonal differences in cold starts due to ambient temperature variations (Mensink et al., 2000).

Further uncertainties in vehicle emissions may be related to the driving behaviour. It has been reported that emissions obtained from aggressive driving can be up to four times higher than those obtained from normal driving (De Vlieger, 1997). In addition to this, emission factors typically increase by a factor up to ten during congestion compared to smooth driving conditions (Sjodin et al., 1998). Finally, it should be remembered that the vast majority of fleet emissions come from a small number of poorly maintained vehicles (Singh and Huber, 2000). For all these reasons, it is practically impossible to determine vehicle emission factors with great accuracy.

5.2. Uncertainty in meteorology

Although meteorological data from different urban monitoring stations might differ significantly for short time periods (i.e. few hours), it is mainly between urban and airport wind data where larger discrepancies are usually encounter.

When the first street canyon models were developed few decades ago, it was assumed that the local roof-top wind information needed as an input would not be generally available, and that airport wind data would have to be used. For this reason, empirical expressions relating airport and roof-top winds were derived (Johnson et al., 1973).

Exploring the sensitivity of AEOLIUS to wind data from different sources, Manning et al. (2000) observed that model concentrations were significantly lower when airport wind speeds were used rather than local roof-top winds. That was in agreement with findings of the present study, which showed that simulations carried out using airport winds produced lower concentrations than those produced using wind data from the other two urban sites. For this reason, predictions made using Orly airport data were eventually excluded from the calculation of the error bounds in Fig. 1, 2, 3, and 4.

Nowadays, there is usually at least one weather station permanently operating in big European cities like Paris. It is, therefore, suggested that only wind information from urban monitoring sites be used, when available, for street canyon simulations. The use of airport wind data is expected to increase uncertainty in predictions.

6. CONCLUSIONS

A practical methodology for quantifying uncertainties in air quality model predictions was developed and tested on experimental data obtained in two street canyons in Paris.

This method was based on the use of three different models, independent meteorological data sets and different emission factors, for creating ensemble sets of street canyon simulations. That enabled us to calculate best estimates of CO and benzene concentrations, and related error bounds.

It was not the intention of the authors to simulate all possible sources of uncertainty in urban dispersion modelling. Uncertainties due to stochastic atmospheric processes or due to errors in some of the input parameters (e.g. dimensions of the street, traffic volumes, etc.) were not here taken into consideration. However, it is believed that the above presented methodology strikes a reasonable balance between simplicity and reliability on urban dispersion models.

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