



A systematic analysis on tomato powder quality prepared by four conductive drying technologies

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ABSTRACT

Four pilot-scale conductive dryers, namely a vacuum drum dryer (VDD), a drum dryer (DD), an agitated thin film dryer (ATFD) and a refractance window dryer (RWD), were used to dry tomato puree. Drying induced colour differences between the reconstituted puree and the original puree and strongly affected the volatile and non-volatile profiles of the powders. Principal component analysis (PCA) identified four separated groups corresponding to the different drying methods, indicating that the drying methods caused significant variance in compound profiles. Subsequently, pairwise comparison of different dried powders was performed by partial least square discriminant analysis (PLS-DA). This resulted in a selection of discriminative volatile and non-volatile markers. RWD and VDD produced powders with high volatile markers that may be related to aroma retention. Conversely, DD dried products contained more non-volatile markers that can be related to taste perception. ATFD processed powders had a lower level of discriminant compounds.

Industrial relevance: Tomato products are frequently thermally processed and dehydrated. However, processing negatively affects the sensory quality of tomato products. In this study, four conductive drying processes, i.e. vacuum drum drying (VDD), drum drying (DD), agitated thin film drying (ATFD) and refractance window drying (RWD) were studied for being energy-efficient drying methods, while suitable for mild (e.g. due to the reduced pressure) drying of pastes and slurries, such as tomato puree. The pilot-scale drying experiments and subsequent statistical analyses of results on quality markers contributed to unravel the impact of the different conductive drying technologies on tomato powder quality. This study may be considered a starting point for selection of conductive drying technologies for the efficient production of high quality tomato powders and other vegetable powders.

1. Introduction

Tomato (*Lycopersicum esculentum*) is a healthy source of nutrients such as fibres, proteins, vitamins, lycopene and other antioxidants (Gahler, Otto, & Böhm, 2003; Qiu, Vuist, Boom, & Schutyser, 2018). Drying is one of the commonly applied methods to preserve tomatoes and process tomatoes into powders that can be incorporated in for example soups and sauces.

Conductive drying is a good option to dry tomato puree into powders, as it is energy efficient and especially suitable for pastes, slurries and concentrated solutions (Devahastin & Mujumdar, 2006; Sahni & Chaudhuri, 2012). Throughout the years different types of conductive dryers have been developed, such as drum dryers (DD), agitated thin film dryers (ATFD) and refractance window dryers (RWD) (Fudym, Carrère-Gée, Lecomte, & Ladevie, 2003; Nindo & Tang, 2007; Pawar,

Patil, Mujumdar, & Thorat, 2011).

Drum drying is successfully applied with viscous paste or pureed foods, such as pre-gelatinized starches, mashed potatoes, caseinate and fruit purees (Kalogianni, Xynogalos, Karapantsios, & Kostoglou, 2002; Rodriguez, Vasseur, & Courtois, 1996; Trystram & Vasseur, 1992). The puree is applied to the outer surface of the rotating drum, where it is rapidly dried by heat supplied through the steel wall by steam condensing inside the drum (Daud, 2006). The dried product film is then scraped off from the drum surface, which allows reuse of the hot drum surface. Drum drying is one of the more energy-efficient drying techniques as it consumes on average 40% less energy than spray drying (Nindo & Tang, 2007), but the product is exposed to the boiling temperature and eventually to the wall temperature, which can result in severe quality loss. The thermal damage can be alleviated by applying reduced pressure to lower the boiling temperature, although the capital

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costs of vacuum drum dryers (VDD) remain a concern (Qiu, Boom, & Schutyser, 2018).

Agitated thin film drying (ATFD) is a continuous drying process carried out under reduced pressure. The ATFD mainly consists of a cylindrical drying chamber with a heating jacket and an internal rotor with fixed blades. The blades agitate and spread the liquid feed as a thin film across the heated surface. The liquid flows down by gravity and progressively passes through different phases, from liquid, to a paste and finally into a solid. The dried solid product is fractured by the rotor blades into smaller particles. The entire process can be easily operated under reduced pressure and is therefore suitable for heat and oxygen sensitive products (Pawar et al., 2011). Nevertheless, successful drying with an ATFD is strongly dependent on the material properties, such as its stickiness and viscoelasticity. A higher tip velocity of the blades provides stronger shearing, which can make the process more robust to the product properties, but it may also lead to local overheating due to friction and thermal damage to the product (Qiu, Boom, & Schutyser, 2018).

Refractance window drying (RWD) is a relatively new gentle drying technique, which has been applied on drying pureed or sliced fruits and vegetables under atmospheric conditions (Abonyi et al., 2002; Zotarelli, Carciofi, & Laurindo, 2015). During RWD, a thin film of the food product is dried on a transparent polyethylene conveyor belt moving over circulating hot water. At the end of the drying process, the dried film moves over a cooling water bath, where the products cool down below its glass transition temperature to avoid stickiness and facilitate scraping-off (Azizi, Jafari, Mirzaei, & Dehnad, 2017). RWD is aimed at heat sensitive products with a low drying temperature (Moses, Norton, Alagusundaram, & Tiwari, 2014). However, RWD has limitations with respect to capacity, throughput and scale-up, as very thin films need to be casted to accommodate reasonable drying rates (Shende & Datta, 2018).

Several studies evaluated the effects of different drying technologies on the perceived freshness and nutritional properties of various food products, such as asparagus, carrots, strawberry, mango, etc. (Abonyi et al., 2002; Caparino et al., 2012; Jafari, Azizi, Mirzaei, & Dehnad, 2016; Nindo, Sun, Wang, Tang, & Powers, 2003; Rostami, Dehnad, Jafari, & Tavakoli, 2018). However, no studies have yet been conducted to compare the effect of different conductive drying methods on tomato powders quality, especially in terms of the retention of taste and aroma specifically. The changes in perceived fresh-tomato flavour during the drying process can be related to loss and/or formation of specific volatile (e.g. alcohols, aldehydes) and non-volatile compounds (e.g. sugars, amino acids), which have been described in previous studies (Kazeniak & Hall, 1970; Malmendal et al., 2011; Petro-Turza, 1986; Rambla et al., 2015; Yilmaz, 2001). These changes may lead to the perception of lower quality by consumers (Qiu et al., 2015). Assessment of the volatile and non-volatile profiles in processed fruits and vegetable products, combined with multivariate data analysis, has been applied to investigate the process impact on flavour quality (Aganovic et al., 2014; Koutidou, Grauwet, Van Loey, & Acharya, 2017; Vervoort et al., 2012). Nevertheless, to the best of our knowledge, no research has been performed to evaluate the effect of drying methods on the flavour quality of tomato powder by this approach.

The objective of this study is thus to create a better understanding of the relationship between organoleptic quality of tomato powders (colour and flavour) and conductive drying methods. The tomato powders were prepared with four conductive drying technologies (DD and VDD, ATFD and RWD) using different drying conditions (temperature and initial solid content). The flavour quality was evaluated by quantitatively determining the concentrations of key volatile and non-volatile compounds, which contribute to tomato flavour.

2. Materials and methods

2.1. Preparation of tomato puree

The hot-break tomato puree used in this study was purchased from AGRAZ, S.A.U. (Badajoz, Spain). The tomato puree was sterilized and aseptically packed in multilayer polyethylene bags. The bags with puree were stored under freezing conditions in a galvanized steel drum, while transported to ILVO (Melle, Belgium), Bodec (Helmond, the Netherlands), ANDRITZ Gouda (Waddinxveen, the Netherlands), and Unilever R&D (Vlaardingen, the Netherlands). The puree was kept frozen until it was ready for drying. The tomato puree was maintained at 20–22 °Brix and pH 4–4.4 with a moisture content of ~0.75 kg/kg wet basis before drying.

2.2. Drying experiments

The frozen tomato puree was thawed just before it was used for drying. To vary the moisture content, a specific amount of puree was thoroughly mixed with added tap water. Thereafter, different batches of diluted tomato puree were obtained with moisture contents of 0.80, 0.81 and 0.82 kg/kg wet basis. The range of the moisture content is small because the initial moisture content of the puree was restricted by the consistencies that could be used with the RWD. For the ATFD, more diluted puree (0.86 and 0.93 kg/kg wet basis) was needed. The prepared puree was dried to below 0.04 kg/kg wet basis by regular and vacuum drum drying, agitated thin film drying or refractance window drying. The drying conditions of each experiment are shown in the Supplementary Data I and were selected after several trials and with input from local experts of each drying technology.

2.2.1. Drum drying

Two types of pilot scale double drum dryers, i.e. a regular drum dryer (DD) and a vacuum drum dryer (VDD), were utilized for drying tomato puree in this study. The DD (Tummers Machinebouw B.V., Hoogerheide, the Netherlands) had two hollow steel drums with 0.30 m external diameter and 0.30 m length. The drums were internally heated by steam with a pressure varying from 1.0 to 3.0 bars, which provided a hot wall temperature ranging from 99.6 to 133.5 °C. The rotation speed of the drums was set at ~1.6 RPM. The VDD (ANDRITZ Gouda, Waddinxveen, the Netherlands) consisted of two steel drums with 0.20 m external diameter and 0.48 m length, was enclosed in a vacuum chamber and operated under reduced pressure (60 mbar). The drums rotated at a fixed speed of ~2.6 RPM and a heating temperature ranging from 81.3 to 133.5 °C, by using steam of different pressures (0.5 to 3.0 bar). The vacuum chamber itself was maintained at 80 °C to prevent the condensation of vapour.

For both drum drying experiments, the clearance between the two drums was fixed at 0.2 mm. The drum temperature was first stabilized before feeding the puree to the dryer. The prepared puree with different moisture contents (0.80 to 0.82 kg/kg wet basis) was poured manually and evenly over the hot feeding pool and passed through the gap of the drums forming a thin layer attaching on the drum surface. After traveling approximately three fourths of the drum circumference, the dried sample was scraped from the drum surface by doctor blades. The dried product collected from the same drying conditions was mixed together for further analysis.

2.2.2. Agitated thin film drying (ATFD)

A pilot scale ATFD was used to dry tomato puree (Bodec, Helmond, the Netherlands). The ATFD consisted of a cylindrical drying chamber with a heating jacket and a rotor with three fixed blades. The drying chamber had an internal diameter of 0.20 m and an effective length of 0.40 m, and the diameter of the blades was 0.198 m. The clearance between the blade tip and the hot surface was 1.0 mm. Before drying, the drying chamber was pre-heated by steam via the wall to the

required temperature (60, 75 or 80 °C). The prepared puree (0.86 and 0.93 kg/kg wet basis) was then pumped into the system with a flow rate of 6 kg/h. The rotation speed of the blades was varied from 500 RPM to 700 RPM, depending on the possibility of scraping the dried product from the drying surface. The dried product was collected at the bottom of the dryer for further analysis. The whole system was operated under a reduced pressure of 50 mbar.

2.2.3. Refractance window drying (RWD)

Refractance window drying was performed with a custom made system (ILVO, Melle, Belgium). The total effective length of the dryer was 3 m in the direction of the belt motion, including a heating part of 2 m and a cooling part of 1 m. The conveyor belt was made of polyethylene terephthalate. During drying, the prepared puree (0.80 to 0.82 kg/kg wet basis) was continuously applied to the plastic conveyor belt with a film thickness of ~1.5 mm. It is emphasized that it was difficult to apply thinner films of tomato puree (i.e. between 0.5 and 0.7 mm) with the available applicator, because of tissue particles present in the puree and its high viscosity. After multiple trials, it was found that 1.5 mm was the minimum thickness for preparing homogeneous tomato puree films. The film of puree was gradually dried while conveyed across the surface of recirculating hot water (85 and 95 °C), and then was cooled down by recirculating cold water (30 °C). The water removal from the film was facilitated by an air suction hood (~20 °C) over the puree at an average air velocity of 1.5 m/s. The belt was set in motion with a speed of 0.1 m/min. At the end of the belt, the dried product was scraped off the belt and collected for further analysis.

2.3. Colour measurements of reconstituted tomato puree

For colour measurements ~5 g of dried tomato powder was reconstituted by adding hot distilled water to attain the water content similar to that of the original tomato puree. The reconstituted puree was mixed with a spatula until a homogeneous puree was obtained. Subsequently, it was stored at room temperature for 24 h for full hydration. The prepared sample was transferred in a 5 cm diameter transparent PE petri dish. Their colour parameters (L^* , a^* , and b^*) were measured with a colorimeter (LABSCAN XE, Hunterlab, Murnau, Germany), with the original tomato puree as a reference. The colorimeter was calibrated using white and black standard reflective calibration plates (Hunterlab, Murnau, Germany). The total colour difference between original and reconstituted puree (ΔE^*) was calculated by (Pathare, Opara, & Al-Said, 2013):

$$\Delta E^* = \sqrt{(L_{sample}^* - L_{ref}^*)^2 + (a_{sample}^* - a_{ref}^*)^2 + (b_{sample}^* - b_{ref}^*)^2} \quad (1)$$

All measurements were conducted in duplicate.

2.4. HS-SPME-GC-MS analysis of volatiles

About 1 g reconstituted puree was homogenised in a 20 mL headspace vial sealed with a screw cap and a PTFE/silicon septum (Supelco, Sigma-Aldrich, Zwijndrecht, Netherlands). The vial was heated at 60 °C for 30 min. Subsequently, the headspace vial was exposed to a PDMS fibre (Agilent Technologies, Santa Clara, USA) to adsorb the volatile compounds. This fibre had been preconditioned at 250 °C for 30 min before use. After extraction, the fibre was transferred to the injection port of a GC-MS, where the adsorbed volatiles were thermally desorbed for 1 min at 250 °C. The GC-MS analysis was conducted on an Agilent 7890A-5975C system (Agilent Technologies, Santa Clara, USA), equipped with an MPS autosampler (Gerstel, Mülheim, Germany). The volatiles were injected in splitless mode, and separated on a DB-Wax column with 20 m × 0.18 mm and 0.3 µm film thickness (Agilent Technologies, Santa Clara, USA). Helium was the carrier gas with a constant rate of 1 mL/min. The column oven started with a temperature of 35 °C for 4 min. It was then heated to 230 °C with a rate of 4.6 °C/min

and maintained at this temperature for 4 min. Mass spectra were obtained by electron ionisation at 1758 eV, with a scanning range of 20–250 *m/z*. MS ion source and quadrupole temperatures were 230 and 150 °C, respectively. All the measurements were conducted in triplicate.

The obtained chromatograms were processed using ChemStation and MassHunter software (Agilent Technologies, Santa Clara, USA). The peak areas were used for multivariate data analysis.

2.5. NMR spectrometry analysis of non-volatiles

2.5.1. Materials

Deuterated oxide for NMR (D₂O, 99.96 atom % D) was purchased from Euriso-top (Saclay, France). Ethylenediaminetetraacetic-d₁₂-acid (EDTA-d₁₂, 98 atom % D) was purchased from Cambridge Isotope Laboratories, Inc. (Tewksbury, U.S.A.). 3-(Trimethylsilyl)propionic-2,2,3,3-d₄ acid, sodium salt (TSP-D₄, 98 atom % D) was purchased from Sigma-Aldrich® (Zwijndrecht, Netherlands). (Difluoro-trimethylsilyl-methyl)phosphonic acid (DFTMP) was purchased from Bridge Organics Co (Vicksburg, U.S.A.).

2.5.2. Sample preparation

Tomato powder (50 mg) was dissolved in 3 mL D₂O. Next, 600 µL of this sample solution were diluted with 100 µL of internal standard solution (0.2 mg/mL of TSP-d₄ and 0.1 mg/mL DFTMP in D₂O), 100 µL of 5 mg/mL EDTA-d₁₂ solution in D₂O and 300 µL of 0.2 M phosphate buffer in D₂O (pD 7.4, containing 0.05% w/w NaN₃ in D₂O). The NMR sample solution was mixed for 3 min using an Eppendorf Thermomixer®C (Hamburg, Germany) at room temperature. Then the solution was centrifuged at 17,000g and room temperature for 5 min. Subsequently, 650 µL of the supernatant was transferred to a 5 mm NMR tube for analysis.

2.5.3. NMR spectrometry analysis

1D ¹H NMR spectra were recorded at 25 °C on a Bruker Avance III 600 NMR spectrometer (Bruker BioSpin GmbH, Rheinstetten, Germany) equipped with a 5 mm cryoprobe. The probe was tuned to detect ¹H resonances at 600.25 MHz. 64 scans were collected in data points of 57 k with a relaxation delay of 10 s, an acquisition time of 4 s and a mixing time of 100 ms. Low power water suppression (16 Hz) was applied for 0.99 s. The resultant data were processed in TopSpin software Version 3.5 pl 1 (Bruker BioSpin GmbH, Rheinstetten, Germany). The data of the free induction decay (FID) were Fourier transformed after multiplying by an exponential window function with a line-broadening factor of 0.15 Hz. Manual phase and baseline correction was applied to all 1D ¹H NMR spectra. The spectra were referenced against the methyl signal of TSP (δ 0.0 ppm). The measurements were carried out in duplicate.

The NMR spectra were imported in Chenomx software (Chenomx NMR Suite Professional v7.63, Edmonton, Alberta, Canada) to calculate the concentrations of the pre-selected target non-volatile compounds. The concentrations of the compounds were expressed as milligram per gram sample (mg/g). A relative error of 10% was taken into account.

2.6. Multivariate data analysis

Experimental data of volatiles and non-volatiles were used for multivariate data analysis using SIMCA 14 (Sartorius Stedim, Malmö, Sweden). As a pre-processing step, all data were mean-centred and then weighed by their standard deviation to give them equal variance. Hereafter, principal component analysis (PCA) was performed to evaluate the variance of each sample and detect outliers without considering the information of the classes (i.e. drying methods). Subsequently, in order to detect the inter-class variance (differences between the samples dried with different conductive drying technologies), partial least square discriminant analysis (PLS-DA) was carried out. For PLS-DA, different volatiles and non-volatiles were considered

as X-variables and the four drying techniques as categorical Y-variables. Considering the complexity of PLS-DA, only pairwise comparison was performed (in total 6 comparisons). The lowest number of latent variables (LVs) resulting in the separation was selected for each comparison. To assess model performance, a permutation test was conducted and r^2 (goodness of fit) and Q^2 (goodness of prediction) values were calculated.

Variable importance in projection (VIP) scores were calculated to identify discriminant compounds, which contributed the most to the explanation of Y-variable variance estimated by PLS-DA. In the present work, X-variables with a VIP score higher than 1 were considered as discriminant markers.

3. Results and discussions

3.1. Performance of the four drying technologies for producing tomato powders

The four dryers required different preconditioning. The original tomato puree (0.75 kg/kg wet basis) could be directly fed to the DD and VDD, while the RWD and ATFD required some dilution.

For DD and VDD, significant amounts of water were evaporated during boiling in the feeding pool. After the tomato passed through the gap between the two drums, it formed a moist, semi-solid film without visible boiling, as bubbles could not nucleate and grow (Kim, 2009). It is assumed that water vapour migrates through pores in the film from the evaporation front inside the film towards the surface of the film (Qiu, Kloosterboer, Guo, Boom, & Schutyser, 2019). The drying processes lasted ~17 and ~28 s for VDD and DD, respectively.

For RWD, dilution of the feed was required due to the applicator design, the minimum belt speed and the maximum drying temperature of the applied RWD: the original puree was too viscous to be formed into a homogeneous thin film, while a puree that was too diluted could not be totally dried. At a drying temperature of 85 °C only the puree with a water content of 0.80 kg/kg could be successfully dried. Due to the low drying temperature (< 95 °C), no boiling took place, and water evaporated due to the difference in relative humidity between the air and the film. Thus, the residence time of RWD (~30 min) was much longer than that of drum drying. The long residence time during RWD is related to the thicker film (1.5 mm) applied. It can be potentially reduced to a few minutes by applying a thinner film (i.e. between 0.5 and 0.7 mm).

For ATFD, also dilution of the original puree had to be carried out (0.86 and 0.93 kg/kg wet basis). The puree with a moisture content of 0.86 kg/kg could only be dried at low temperatures (60 °C). At higher temperatures, the concentrated puree transformed into a sticky mass forming large lumps, which could not be easily dried and scraped from the wall. Unfortunately, it is impossible to visually inspect the drying behaviour in the stainless steel ATFD equipment. The relatively difficult drying of tomato puree in ATFD could be related to its high concentration of glucose and fructose, which contributed to sticky behaviour during drying (Qiu, Boom, & Schutyser, 2018). The rotation speed of the blades had to be well controlled as well: faster rotation facilitates powder formation, but can also lead to local overheating and damage to the product. Therefore the feed rate, feed moisture, drying temperature and blade rotation speed should be well tuned for proper operation of ATFD. To conclude, RWD and ATFD are more challenging in their operation and require more optimization for successful drying of tomato puree.

The different drying methods resulted in different product morphology (Fig. 1). DD and RWD yielded smooth, thin flakes. VDD produced a sausage-like product due to the high chamber temperature (80 °C): after scraping-off, the film remained in the rubbery state and thus curled into a sausage shape along with the rotation of the drums (Kitson & MacGregor, 1982). During ATFD, the product was fractured into small particles by the rotating action of the blades.

3.2. Impact of drying technologies on colour

Fig. 2 shows the luminosity values of the reconstituted tomato purees. All reconstituted samples had lower L^* values than the reference (original tomato puree) indicating a darker colour. This may be attributed to non-enzymatic browning (Maillard reaction) during drying (Caparino et al., 2012). Comparing each drying method, the L^* values of VDD (except the samples dried at 133.5 °C) and DD dried samples were closer to the reference than those from RWD and ATFD. The reconstituted ATFD dried puree was darker, due to local overheating by the blade rotation, in spite of the low drying temperature. Overheating might lead to caramelization of sugars contributing to darkening during drying. The colour degradation was more pronounced at a higher rotation speed. The unexpected darker samples produced by RWD could be attributed to the long drying time, and the extended exposure to oxygen. As the product stayed in a moist state with high water activity for long time, more browning or Maillard reactions might occur, although the drying temperature was much lower than during VDD and DD. The impact of temperature was more pronounced for VDD than the other methods. The possible reason could be that during VDD the product temperature quickly increases to the wall temperature, leading to more pronounced overheating and darkening. The effect of the moisture content on the feed was limited due to the small range of the moisture content considered.

The colour of tomato puree is red, which is mainly due to the presence of lycopene (Shi & Maguer, 2000). It is commonly regarded as a measure of quality and can be represented by a^* values (Fig. 3). Similar to L^* , all reconstituted purees had lower a^* values than the reference, mainly because of the degradation of lycopene during the process (Shi & Maguer, 2000). The decrease of a^* was more pronounced with ATFD and RWD, probably also due to high blade rotation and long drying time, respectively. The impact of the temperature was more pronounced for VDD and DD. The higher temperature led to decrease of a^* in VDD, whereas increase of a^* in DD. On the one hand, a higher temperature may induce more lycopene degradation. On the other hand, an increased temperature leads to faster drying of the tomato matrix, which may prevent penetration of oxygen and limit the oxidation of the lycopene (Goula & Adamopoulos, 2005). For VDD, the effect of temperature is dominant since the vacuum creates a low-oxygen environment. For the DD, the influence of oxidation is probably more pronounced. Again, the impact of moisture content was limited.

Fig. 4 shows the total colour difference between the reconstituted puree. The VDD (81.3 °C) and DD (99.6 °C, 0.75 kg/kg) dried samples had minimal colour changes, indicating that their colour quality was closest to that of the reference. Increasing temperature induced larger colour changes for VDD, while smaller changes were observed for DD. Especially, VDD led to more significant colour changes than DD for drying at 133.5 °C. This could be explained by faster increase in the product temperature during VDD as the samples were dried much faster, which resulted in a longer exposure to a high temperature and thus more thermal damage. Therefore, under applied high wall temperatures, the drums of VDD could be rotated faster to shorten the residence time and to prevent overheating. Another option to reduce thermal damage is to use low-pressure steam to operate at lower drum wall temperature. Amongst the applied conditions, the effect of temperature and moisture content was not obvious for RWD and ATFD.

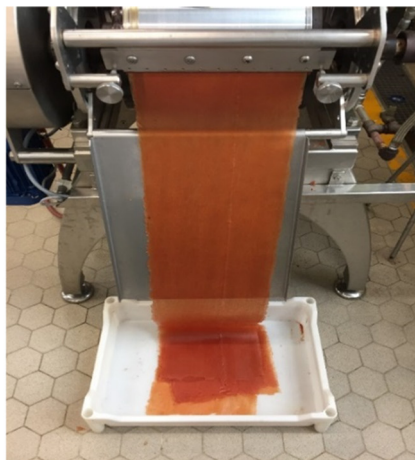
3.3. Impact of drying technologies on volatile and non-volatile profiles

To assess the effect of the conductive drying methods on tomato flavour quality, the concentrations of volatile and non-volatile compounds in the tomato powders were analysed. We selected 14 volatiles and 16 non-volatile compounds that are key compounds for flavour of tomato products (shown in Supplementary Data II) (Krumbein, Peters, & Brückner, 2004; Malmendal et al., 2011; Yilmaz, 2001). Some key volatile compounds, previously reported, e.g. hexanal, cis-3-hexenal,

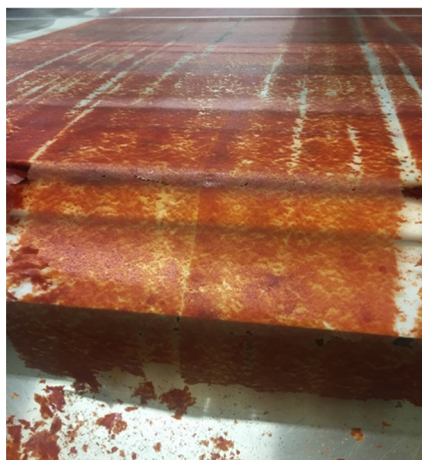
(A) Vacuum drum drying



(B) Drum drying



(C) Refractance window drying



(D) Agitated thin film drying



Fig. 1. Visual images of the tomato products produced from (A) vacuum drum drying; (B) drum drying; (C) refractance window drying and (D) agitated thin film drying.

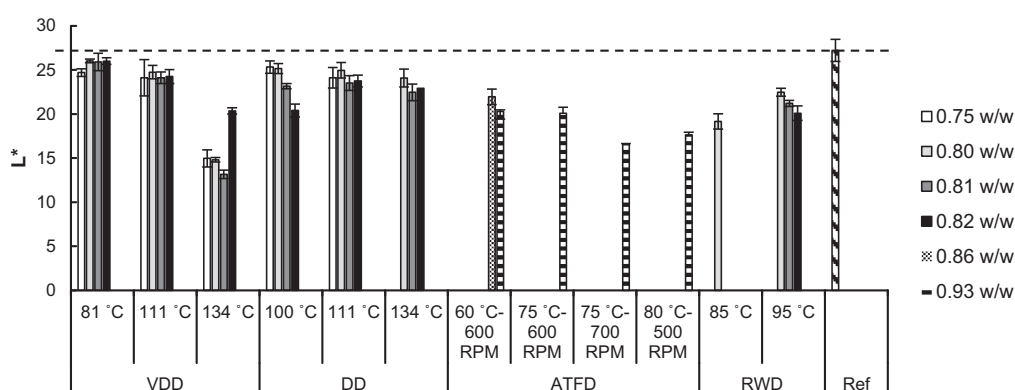


Fig. 2. Luminosity of reconstituted tomato powders dried from puree at different drying conditions. The error bars represent the standard deviation of the experimental data ($n = 2$).

hexanol and cis-3-hexenol (Yilmaz, 2001), were not detected either in the tomato powders or the original tomato puree, possibly due to the hot-break process used for the production of the starting puree. These volatiles, known as C_6 aldehydes, are derived from the oxidation of fatty acids via the lipoxygenase pathway. Inactivation of the enzymes at higher temperatures during hot-break (ranging from 85 to 100 °C) resulted in the reduced generation of these volatiles (Xu & Barringer,

2009). In addition, the hot-break processing may have further reduced their level especially that of cis-3-hexenal, because of their instability to heat and metal surfaces (Buttery, Teranishi, Ling, & Turnbaugh, 1990; Goodman, Fawcett, & Barringer, 2002; Sies & Crouzet, 1977).

3.3.1. Principal component analysis (PCA)

Principal component analysis was first applied to the data of

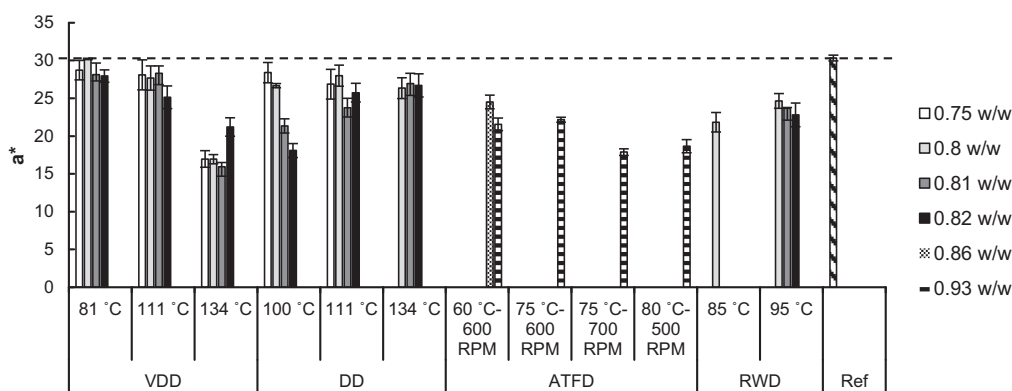


Fig. 3. Redness of reconstituted tomato powders dried from puree at different drying conditions. The error bars represent the standard deviation of the experimental data ($n = 2$).

volatiles and non-volatiles. The first two principal components (PCs) could explain 33.6% and 21.0% of the total variability, respectively. The score plot (Fig. 5) showed the differences in the volatile and non-volatile profiles of the different tomato powders. Four separated groups could be identified, corresponding to the different drying methods, which indicates that the variance of the compound profiles caused by the drying methods was more significant than that caused by the drying conditions within a single drying technology, i.e. temperature and initial moisture content.

3.3.2. Partial least square discriminant analysis (PLS-DA)

In order to further understand the variance, partial least square discriminant analyses (PLS-DA) were carried out to pairwise compare each drying method. All PLS-DA models had Q^2 values > 0.5 and were statistically significant, indicating that the groups could be discriminated beyond chance (Hu et al., 2018). All the PLS-DA models were validated without overfitting according to permutation tests (Supplementary Data III).

A PLS-DA bi-plot using the first two latent variables (LVs) was constructed to visualize the different impact of VDD and DD on tomato powder quality (Fig. 6). The VDD and DD dried samples are well separated and their variance can be explained mainly by the first LV, indicating processing impact. The significance of the individual compounds for the discrimination of differently processed samples increased with their distance from the centre. In Fig. 6, compounds with higher concentrations in the VDD dried samples are projected close to the VDD side (mainly in the left-hand side of the plots). In contrast, compounds located closer to the DD side, have lower concentration in the VDD dried powders. PLS-DA bi-plots of the other comparisons are not shown.

To quantitatively rank the importance of volatiles to the

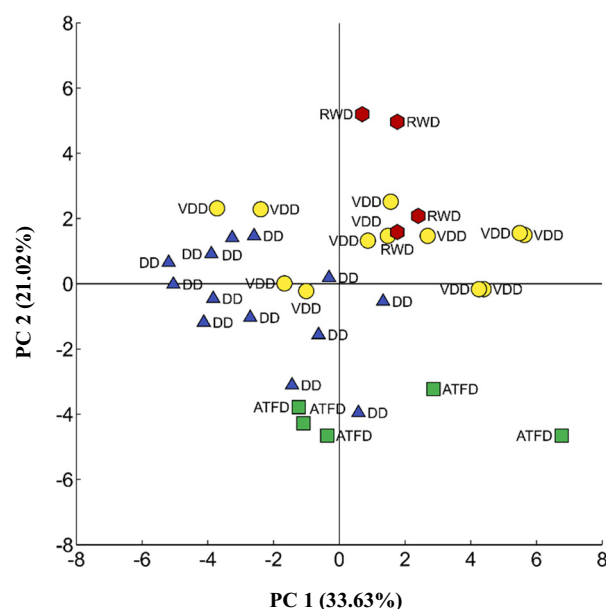


Fig. 5. A score plot of the PCA performed on the volatile and non-volatile data of the tomato powders obtained from: vacuum drum drying (yellow circle), regular drum drying (blue triangle), agitated thin film drying (green square) and refractance window drying (red diamond). The variance explained by each component is indicated on the respective axis.

discrimination between VDD and DD, variable-importance-in-projection (VIP) scores were calculated. In the present work, only compounds with $VIP > 1$ were selected and considered as discriminant markers,

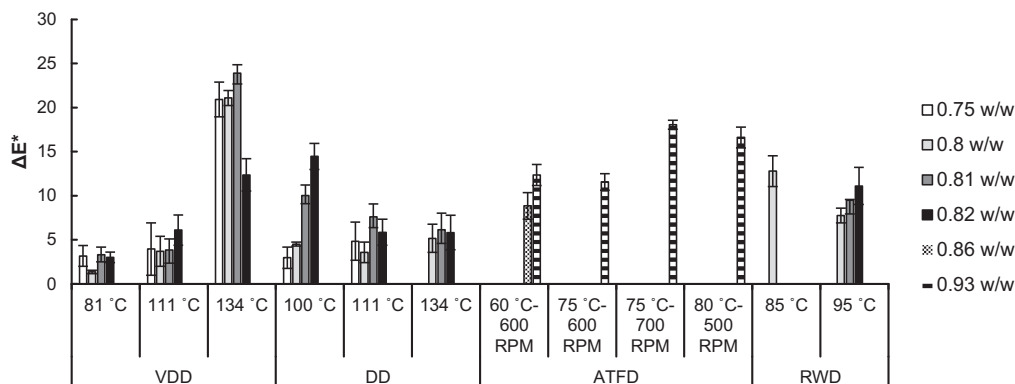


Fig. 4. Total colour difference of reconstituted tomato powders dried from puree at different drying conditions. The error bars represent the standard deviation of the experimental data ($n = 2$).

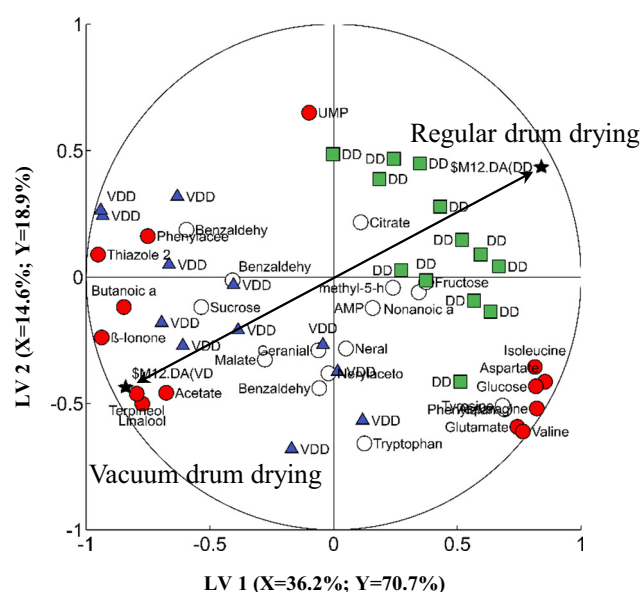


Fig. 6. A PLS-DA bi-plot visualizing impact differences between vacuum drum drying (blue triangle) and regular drum drying (green square). The circles represent the volatile and non-volatile compounds detected, of which the selected discriminant compounds (VIP > 1) are highlighted in red. The X- and Y-variance explained by each latent variable (LV) is indicated on the respective axis.

and were highlighted in red (Fig. 6). Table 1 shows the regression coefficients of the discriminant markers in the indicated class (VDD). Positive regression coefficients indicate higher concentrations in VDD,

and vice versa. Six volatiles and eight non-volatiles markers were selected. Nine markers had positive coefficients, indicating their higher concentration in VDD dried powders. All the discriminative volatiles were detected in a higher abundance in VDD.

The identification of the discriminative compounds provides valuable information about differences amongst drying technologies. For example, the higher concentration of 2-isobutylthiazole in VDD can be understood from the limited presence of oxygen in the vacuum chamber, as 2-isobutylthiazole is sensitive to oxidation (Jeyaprakash, Frank, & Driscoll, 2016). Phenylacetaldehyde was also detected in larger amounts in VDD. This component can be associated with a cooked flavour, originating from Strecker degradation of amino acids (a minor pathway of the Maillard reaction) (Granvogl, Beksan, & Schieberle, 2012). More Strecker and/or Maillard reactions could occur during VDD as the product temperature quickly increased to the wall temperature due to the fast drying and thus the product was exposed to high temperatures for longer time during the process. Conversely, many of the compounds with negative coefficients were amino acids in the VDD samples, which might also be explained by Strecker and/or Maillard reactions considering that amino acids are substrates for these reactions. In addition to the chemical changes, fast drying and solidification of the tomato matrix during VDD may also entrap volatiles in the matrix, especially because of the presence of sugars, thus facilitating the retention of volatiles despite the lower pressure that would in principle favour the release of volatiles (Jeyaprakash et al., 2016).

Comparing the impacts of VDD and ATFD on product quality, eight volatiles and five non-volatiles were selected as markers (Table 1). Most compounds (twelve) had positive regression coefficients, indicating they were more abundant in the VDD class. Similar results were observed when comparing ATFD with DD or RWD. The comparison

Table 1

Discriminant volatile and non-volatile compounds for each comparison, selected in tomato powders based on VIP values, and listed in decreasing order of regression coefficients.

VDD versus DD		VDD versus ATFD		VDD versus RWD	
Compound	Regression coefficient (VDD)	Compound	Regression coefficient (VDD)	Compound	Regression coefficient (VDD)
α -Terpineol	0.17	Geranial	0.10	Tryptophan	0.14
Linalool	0.16	β -Ionone	0.10	Malate	0.11
β -Ionone	0.15	Neral	0.10	Nerylacetone	0.01
3-Methylbutanoic acid	0.12	α -Terpineol	0.09	3-Ethylbenzaldehyde	−0.06
Acetate	0.12	Tryptophan	0.09	6-Methyl-5-hepten-2-one	−0.08
2-Isobutylthiazole	0.08	Linalool	0.08	Geranial	−0.08
Phenylacetaldehyde	0.03	2-Isobutylthiazole	0.08	Neral	−0.08
Valine	0.02	3-Methylbutanoic acid	0.07	α -Terpineol	−0.11
Glutamate	0.01	AMP	0.07	Linalool	−0.12
Asparagine	−0.004	Nerylacetone	0.07	β -Ionone	−0.16
Glucose	−0.01	Citrate	0.07		
Aspartate	−0.04	Malate	0.06		
Isoleucine	−0.06	Nonanoic acid	−0.07		
UMP	−0.11				

DD versus ATFD		DD versus RWD		ATFD versus RWD	
Compound	Regression coefficient (DD)	Compound	Regression coefficient (DD)	Compound	Regression coefficient (ATFD)
AMP	0.16	AMP	0.09	Nonanoic acid	0.07
Citrate	0.15	3-Ethylbenzaldehyde	−0.08	6-Methyl-5-hepten-2-one	−0.06
α -Terpineol	0.13	2-Isobutylthiazole	−0.10	Nerylacetone	−0.07
Geranial	0.08	3-Methylbutanoic acid	−0.11	AMP	−0.07
Neral	0.07	α -Terpineol	−0.13	Neral	−0.08
Fructose	0.05	Linalool	−0.13	Geranial	−0.08
Glucose	0.05	β -Ionone	−0.13	Citrate	−0.09
Tryptophan	0.04			2-Isobutylthiazole	−0.09
Isoleucine	0.03			Linalool	−0.09
Asparagine	0.03			3-Methylbutanoic acid	−0.09
Aspartate	0.03			β -Ionone	−0.10
Valine	0.03			α -Terpineol	−0.10
Glutamate	0.02				
Acetate	−0.15				

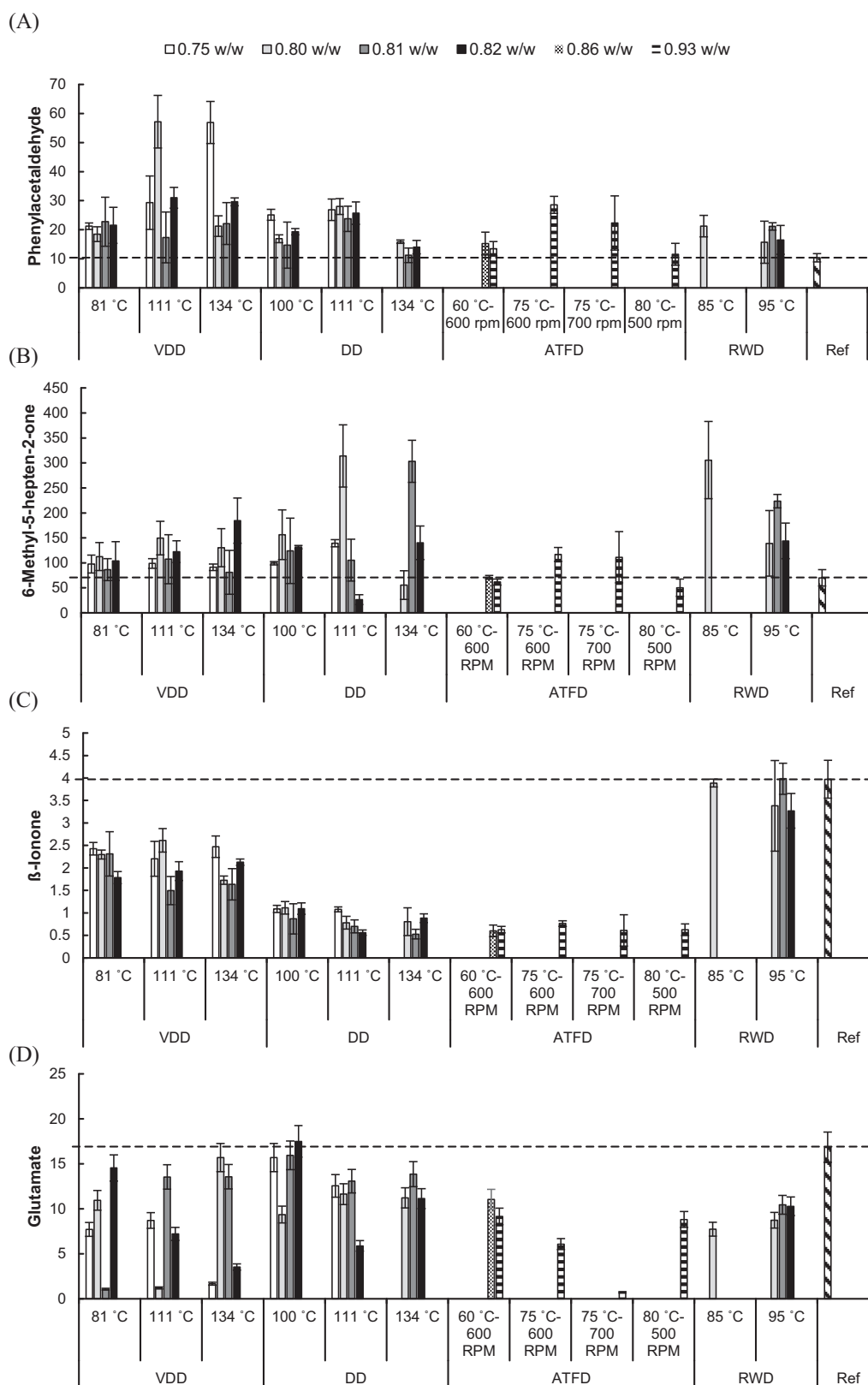


Fig. 7. Discriminative components (A) phenylacetaldehyde, (B) 6-methyl-5-hepten-2-one, (C) β -ionone and (D) glutamate of reconstituted tomato powders dried from puree at different drying conditions. The Y-axis of volatiles indicates the peak area $\times 10^5$. The error bars represent the standard deviation of the experimental data ($n = 3$). The Y-axis of non-volatiles indicates the concentration in mg/g. The error bars represent the standard deviation of the experimental data ($n = 2$).

between DD and ATFD showed that thirteen of the fourteen markers (three volatiles and eleven non-volatiles) were in higher concentration in DD dried powders (Table 1). Regarding ATFD versus RWD, eleven of

the thirteen markers (eleven volatiles and two non-volatiles) were more retained in RWD (Table 1). These findings show that ATFD retained less flavour compounds than the others, under the applied drying

conditions. This may be due to the thermal damage induced by the fast blade rotation. Therefore, more optimization of the processing parameters, i.e. feed moisture content and blade rotation speed, etc., is necessary in order to produce tomato powders of higher quality.

When comparing RWD with VDD, ten markers (eight volatiles and two non-volatiles) were selected, while seven of them had negative regression coefficients in VDD. Similarly, in the comparison of RWD and DD, six of the seven markers had a negative coefficient in DD. In both comparisons, all the discriminative volatiles (except nerylacetone) were detected in high levels in RWD (Table 1). The reason could be attributed to the low operating temperatures ($< 95^{\circ}\text{C}$) applied in RWD, resulting in less release of volatiles from tomato puree. The more abundant volatiles might contribute to higher aroma retention in RWD processed products. Specifically, for VDD compared to RWD, a lower concentration of 6-methyl-5-hepten-2-one in VDD could also be related to the absence of oxygen in VDD. 6-Methyl-5-hepten-2-one is regarded as a marker compound for lycopene degradation as it forms when lycopene is oxidized (Cremer & Eichner, 2000; Thakur, Singh, & Nelson, 1996). This was also consistent with the observation that VDD dried powders had higher a^* values than RWD.

Concluding, RWD processing has the best potential to produce powders with a high level of volatile compounds (i.e. retained aromas), followed by VDD. Although DD processing led to lower levels of volatiles compared to RWD and VDD, it produced powders with more non-volatiles, known to contribute to overall taste of tomato. ATFD processed powders contained lowest amount of (non)-volatiles compared to other conductive drying technologies, making ATFD probably less suitable for drying of tomato products.

3.3.3. Changes of discriminative compounds during drying processes

To further substantiate differences between the drying methods, Fig. 7 shows the individual plots of several discriminative markers. The concentrations of these markers were also compared to the original tomato puree (as a reference) to visualize how they changed after drying. It can be observed that phenylacetaldehyde and 6-methyl-5-hepten-2-one had higher concentration in the tomato powders. Especially, formation of phenylacetaldehyde was more pronounced for VDD at high temperature, probably because more Strecker/Maillard reactions occurred. Formation of 6-methyl-5-hepten-2-one in VDD and ATFD dried powders was less than RWD and DD, which is related to less oxidation of lycopene in a low oxygen environment. Differently, the concentration of β -ionone decreased during drying probably due to its low boiling point (Narain, Galvao, de Santana, & da Silveira Moreira, 2010). RWD dried powders had the highest β -ionone level practically similar to the reference, due to its mild operating conditions. Again, all the dried powders, except those obtained from DD at 100°C and 0.82 w/w, contained less glutamate (known as a marker for umami taste) compared to the reference. One possible reason could be that glutamate converted into pyroglutamic acid during thermal processing, which contributes to a more bitter and undesirable sour taste of the product (Qiu, Vuist, et al., 2018).

4. Conclusions

Tomato puree was dried using four different conductive drying methods and the organoleptic quality (colour and flavour) of the resulting powders were compared. The drying methods strongly affected the final powder morphology of the dried products. All the drying methods resulted in a change in colour when comparing the reconstituted and the original tomato puree. The colour changes were most obvious for ATFD and RWD, due to the high local shear and long drying time, respectively.

In addition to colour, the different technologies also led to different volatile and non-volatile profiles in the powders that might be related to perceived flavour quality. In general, RWD resulted in a higher level of discriminative volatiles, due to the lower operation temperature.

Comparing to DD, discriminative volatiles were more abundant in VDD, while discriminative non-volatiles especially amino acids were less. Under the applied conditions, ATFD dried powders had a lower level of flavour compounds. However, since the ATFD is operated under vacuum and at lower drying temperature, we had expected to prepare tomato powders with more flavour compounds. Possibly, further process optimization, i.e. feed moisture content and blade rotation speed, etc., might improve ATFD processing.

RWD has potential for making powders with high retention of volatiles, with amongst other aromas. However, RWD is a relative slow process for drying of concentrated tomato puree, and thus has limitations with respect to capacity and scale-up. Following RWD, VDD can be an alternative to retain aromas during drying. Nevertheless, also its high capital costs need to be concerned. Although DD dried products contain less volatiles, they have higher concentration of certain non-volatile compounds, i.e. amino acids, which contributes to the taste. ATFD is probably less suitable for sticky materials like tomato, while it can be applied to products with limited sticky behaviour, such as spinach juice (Qiu, Boom, & Schutyser, 2018). The results in the present work may be considered a starting point for selecting conductive drying technologies for the production of tomato powders and even other vegetable powders of desired quality. Sensory analysis of processed powders combined with corresponding volatile and non-volatile profiles is recommended to achieve more detailed information about how powder flavour quality is perceived by consumers.

Choice of technology in practice also depends on the costs involved. In a recent study, a techno-economical assessment was made for VDD, RWD, ATFD compared to spray drying (de Smidt, Wemmers, & Spoelstra, 2017). This assessment was based on water evaporation and not on specific product cases. The results in this paper can be combined with this assessment to guide selection of the most appropriate conductive drying technology for tomato powder.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ifset.2019.03.013>.

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Conflict of interest

There is no conflict of interest.

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