

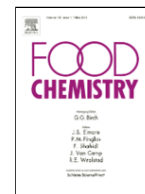


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Impact of drying techniques, seasonal variation and organic growing on flavor compounds profiles in two Italian tomato varieties

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 3-Methyl butanal (PubChem CID: 11552)
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ABSTRACT

The industrial transformation of tomato (*Lycopersicon esculentum* Mill.) produces processed foods, such as dried tomatoes. In this study two varieties (SaAb and PerBruzzo), grown in three cropping systems (one conventional and two organic ones), were processed by two types of small-scale drying (oven or sun drying), over two years of production. The dried samples were analyzed for their non-volatile and volatile composition, relating the results with sensory analysis. The multivariate analysis performed on collected data allowed a detailed comparison of the effects of processing, year-to-year variation and cropping systems. Results indicated that drying methods mainly influenced the composition and flavor profile, also affected by the production year. The cropping system significantly influenced some quality indices, such as the acid and sugar amounts, and the aldehydes, respectively higher and lower in organic samples. The comprehensive PCA analysis allowed discrimination of drying methods and, to a lesser extent, cropping systems.

1. Introduction

The tomato is the edible fruit of the plant *Lycopersicon esculentum* Mill., a species belonging to the *Solanaceae* family. According to the Food and Agriculture Organization of the United Nations (FAOSTAT, 2017), tomato production exceeded 182 million tons in 2017, being widespread all over the world. The popularity of tomatoes depends on the fact that they can be consumed fresh or in

a variety of processed forms, including tomato preserves, tomato-based foods and dried tomatoes.

The flavor of tomato depends on two groups of compounds, the non-volatile molecules and the volatile ones that constitute the aroma; the aroma profile of raw tomatoes has been extensively studied in the past (Carbonell-Barrachina, Agustí, & Ruiz, 2006; Selli, Kelebek, Ayseli, & Tokbas, 2014), as well as its modifications induced by processing (Heredia, Peinado, Rosa, Andrés, & Escriche, 2012; Petro-Turza, 1986), indicating the high complexity of tomato aroma profile.

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Over the years, breeding efforts have mostly been aimed at improving yield, resistance to biotic and abiotic stresses, firmness and external appearance, leading to a decline in consumption of modern cultivars, perceived as less tasty when compared to heirloom varieties, often produced only locally and on a small scale (Bucheli et al., 1999). In order to address the breeding activity to targeting the demand of the consumers, mostly related to nutritional value, sensory quality and acceptance, a different approach has been followed in recent years, based on multivariate data analysis of the relationships between the composition of tomatoes and consumer acceptance. An innovative model highlighted crucial contributions of fructose and citric acid to the flavor of tomatoes and of several volatiles (2-butyl acetate, *cis*-3-hexen-1-ol, 3-methyl-1-butanol, 2-methylbutanal, 1-octen-3-one and *trans,trans*-2,4-decadienal) to the aroma (Tieman et al., 2012). Other studies (Piombino et al., 2013) indicated both positive drivers for liking (soluble solids content, sour taste, total acidity, citrate, herbaceous aroma and 2-isobutylthiazole) and negative ones (diacetyl-like odor).

Dry matter in tomatoes consists mainly of sugars and organic acids, making concentration levels of these molecules crucial in the assessment of fruit palatability, as they act as the main compounds responsible for the sweet and sour/acid tastes of these fruits, respectively. In particular, the importance of the sugar/acid ratio in the overall acceptance of tomatoes was highlighted (Agius, von Tucher, Poppenberger, & Rozhon, 2018; Bennet, 2012). The most abundant sugars in tomatoes are the reducing sugars, glucose and fructose, and only traces of sucrose are found in immature fruits (Davies, Hobson, & McGlasson, 1981; Leyva et al., 2014). The major organic acids in tomatoes are citric, malic and succinic, with citric acid predominating (Agius et al., 2018; Marconi, Floridi, & Montanari, 2007). Other organic acids may appear as a result of technological processing of tomatoes: among these molecules, low levels of aconitic acids deriving from the dehydration of citric acid (Marconi et al., 2007) and the glutamine derivative pyroglutamic acid, albeit already present in fresh fruit, is known to be a marker of thermal treatment (Davies et al., 1981; Marconi et al., 2007; Qiu, Vuist, Boom, & Schutysier, 2018).

In addition to this, it is known that palatability of tomatoes and of their processed derivatives also depends on the umami sensation typical of this fruit (Chew et al., 2017), a feature that can be largely imparted by the high levels of free glutamic acid that increase during ripening (Sorrequieta, Ferraro, Boggio, & Valle, 2010) and as a result of processing (Porretta, Birzi, Ghizzoni, & Vicini, 1995).

As previously stated, behind a deep characterization of tomato flavor there is the relationship of sugars and organic acids with the fundamental contribution of aroma volatiles, mainly composed of alcohols (*cis*-3-hexen-1-ol), aldehydes (*trans*-2-hexenal), as well as ketones and terpenes (6-methyl-5-hepten-2-one and geranyl acetone) from carotenoids breakdown (Lewinsohn et al., 2005).

In the context of an analysis that aims at comparing the flavor of local varieties subjected to small-scale processing, it is also crucial to consider also growing methods for production. In particular, attention should be focused on the increasing consumer interest in organic foods; until now, special attention has been given to comparative studies on the quality of raw organic and conventional products (Rembiałkowska, 2007), though these comparative studies did not show clearly that organic farming increased palatability (Talavera-Bianchi, Chambers, Carey, & Chambers, 2010). However, organic products are often processed, in order to prolong their shelf-life, thus making research efforts on the comparison of organic and conventional processed foods even more relevant in modern food science. Thermal processing, including drying, is a common way to prolong the shelf life of tomatoes, and previous

studies have compared nutritional parameters between raw and processed products (Kerkhofs, Lister, & Savage, 2005; Rodriguez et al., 2015). The effects of drying are positive on the resulting taste, as it is known that drying improves palatability of tomatoes, owing to the formation of Amadori compounds and resulting Maillard reaction products. At the same time, this processing could be detrimental to health-related quality, mainly due to the exposure to heat and light that reduces bioactive phytochemicals and generates off-flavors (Hellwig & Henle, 2014).

The aim of this study was to evaluate the combined effect of cultivation methods (conventional and two types of organic farming) and two different small-scale drying techniques on two tomato varieties over two years of production. The concentration of flavor compounds of tomatoes was assessed by the dosage of non-volatile and volatile compounds and by the correspondent sensory evaluation. This wide pattern of variability was studied, in order to highlight the major variation factors in tomato quality.

2. Materials and methods

2.1. Fruit material

Fruits of two different tomato varieties were studied: variety “SaAb Cra” (selected by CREA) and commercial HF1 “PerBruzzo” (Four Sementi seed company). Samples were collected in the 2015 and 2016 seasons at the experimental fields of CREA-OF Monsampolo del Tronto (Ascoli Piceno, Italy), located in the Tronto Valley (Marche Region) (42°53'N; 13°47'E, 158 m a.s.l.). Three different crop managements were conducted: integrated (CONV), organic with artificial mulching (ORG-PA) and an experimental no tillage organic management with natural mulching (ORG-PN), as described in a previous work (Canali et al., 2013). Field trials were conducted in a randomized strip block experimental design with three replicates for each genotype. Harvests were conducted in the first half of August, on fully ripe fruits, visually assessing the complete red color; they were harvested all at once on the same day, from the first five vines.

Once harvested, samples (around 10 kg for each replicate) were divided into three aliquots, and subjected to three different drying treatments until constant weight, used to calculate the dry matter (dw) content of the products, expressed in g/100 g dw.

- freeze-drying (FREEZE): samples were quickly frozen at -50°C in an air-blast tunnel and successively freeze dried and used as control afterward;
- oven-drying (OVEN): samples were dried for 48 h in an forced-air oven (Thermo-Lab, Codogno, Italy) at 55°C , with internal relative humidity of the air flux around 15–17%;
- solar drying (SUN): samples were dried in a solar-drier apparatus purchased from Termotend S.p.A. (Carpi, Modena, Italy), where inner average temperature and relative humidity had a day–night range (45°C to 17°C and 25% to 50%, respectively); the duration of this process ranged from 7 to 14 days, depending on weather conditions.

The dried materials were reduced to powder (5–10 mesh) in a blender at $4-6^{\circ}\text{C}$ and stored in dark bottles at -20°C until analyses. Every assay was performed at least in duplicate.

2.2. Analysis of the non-volatile fraction

All solvents used were of analytical grade, with a minimum purity of 99.7%, purchased from Sigma-Aldrich and Fluka. Water was purified by ion exchange column (Permax, Treviglio, Italy), while

ultra-pure water was obtained with a Milli-Q apparatus. Solvents were dried by standard methods prior to use. Chromatographic systems were purchased from Jasco (Jasco Europe S.R.L., Cremella, Lecco, Italy), and the software for data analysis was Clarity ver. 2.5.517 (DataApex, Prague, Czech Republic).

2.2.1. Soluble solids content (SSC), titratable acidity (TA) and simple sugars analysis

SSC and TA were measured in duplicate from aqueous extracts of the samples (2 g to 20 mL), treated with ultrasound for 7 min and then centrifuged. SSC was measured by refractometry as °Bx on dry weight (dw), whereas TA was determined by automatic titration with 0.1 M NaOH until an equivalence point established at pH = 8.2. Simple sugars (fructose, glucose and sucrose) were measured by HPLC (Selli et al., 2014) at 85 °C on a Bio-Rad Aminex HPX-87C column (Hercules, CA) with H₂O as mobile phase (0.6 mL/min) and a refractometer as a detector (Jasco RI 1930). The instrument was calibrated with solutions of commercial standards at known concentration and the results were given as g/100 g dw.

2.2.2. Organic acids analysis

The organic acids of tomatoes were evaluated using HPLC (Selli et al., 2014). Samples were extracted with 0.1 M HCl solution (500 mg to 10 mL). The determination was made by using HPLC at 50 °C on a Repromer H⁺ 300 × 8 mm column with H₂SO₄ 3 mM as mobile phase (0.6 mL/min) and UV detection at 214 nm. The instrument was calibrated with solutions of commercial standards at known concentration and the results were given as mg/100 g dw.

2.2.3. Amino acids analysis

Free amino acids in tomato extracts were measured using HILIC-HPLC without pre-column derivatization, similarly to previously described (Bhandare, Madhavan, Rao, & Someswar Rao, 2010). Samples were extracted with 0.1 M HCl solution (500 mg to 10 mL). Separation was achieved at 45 °C using a Lichrosorb Si-60 (10 μm) (Hibar) column at 0.6 mL/min, with an eluant of 85% acetonitrile (99%)/15% aqueous solution containing 0.02 M H₂PO₄ and 3.6 mM KH₂PO₄, with UV detection at 210 nm. The instrument was calibrated with solutions of commercial standards at known concentration and the results were given as mg/100 g dw.

2.3. Tomato extracts fractionation

Samples were extracted in succession with solvents of increasing polarities, namely *n*-hexane, ethyl acetate, methanol and water. Tomato samples (200 mg) were placed in a 10 mL sealed, fritted tube and extracted with 4 mL of the selected solvent for 2.5 h at room temperature (rt) by means of an orbital shaker. The extraction solvent was collected using a Flash VacBiotage apparatus (Biotage Sweden AB, Uppsala, Sweden) and the extraction procedure was reiterated two more times for each solvent. Each step of fractionation was monitored by TLC analysis using silica gel F₂₅₄ aluminum sheets (Merck, Darmstadt, Germany) and *n*-propanol/water 7:3 as the eluent. TLC plates were stained with 0.5% ninhydrin solution in ethanol or with cerium sulfate/ammonium hexamolybdate solution and heating at 150 °C. The three aliquots of solvent were combined and, with the exception of the hexane extracts, evaporated to dryness under reduced pressure (ethyl acetate and MeOH) or freeze-dried (water). Weight data are reported in Supplementary Table 1. Extracts were stored at -20 °C.

2.4. Spectroscopic methods

¹H and ¹³C NMR spectra were acquired at 400.13 and 100.61 MHz, respectively, using a Bruker Avance 400 spectrometer (Bruker, Karlsruhe, Germany) interfaced with a workstation running Windows Operating System (Microsoft, Redmond, WA) equipped with TopSpin software. Chemical shifts are given in ppm (δ) and are referenced to the signals of the solvent (CD₃OD, δ_H = 3.31; δ_C = 49.0; D₂O, δ_H = 4.71) or to 3-(trimethylsilyl)propionic acid-2,2,3,3-d₄ as external standard (δ_C = 0.00). Signal multiplicities in the ¹³C spectra were assigned on the basis of APT (attached proton test) experiments. Signal attribution was made on the basis of COSY (correlated spectroscopy), HSQC (heteronuclear single quantum correlation) and HMBC (heteronuclear multiple bond coherence) experiments. Spectra analyses were carried out using inmr software (www.inmr.net). Mass spectra were acquired with a ThermoFinnigan LCQ Advantage mass spectrometer (GenTech Scientific, Arvade, NY).

2.5. Synthesis of *N*-(1-deoxy-*D*-fructos-1-yl)-*L*-glutamic acid (2)

The procedure of Beksan et al. (2003), was followed with some modifications. To a stirred suspension of *L*-glutamic acid (2.20 g, 15 mmol) in methanol (40 mL) containing 3 Å molecular sieves (500 mg), potassium *tert*-butoxide (3.37 g, 30 mmol) was added portionwise over 30 min under a stream of dry nitrogen. After complete dissolution of the suspended glutamic acid, glucose (1.80 g, 10 mmol) was added in one portion, followed by additional methanol (10 mL). The resulting suspension was stirred at rt for 30 min and then refluxed for 1 h. The reaction mixture was cooled to rt and molecular sieves and unreacted material were removed by filtration. The clear yellow solution was concentrated under reduced pressure to half of the initial volume. Upon addition of dry acetone (ca. 30 mL) at -40 °C, a white solid precipitated. The solid was collected by filtration, washed with dry, cold acetone and dried under reduced pressure over KOH to give *N*-(*D*-glucos-1yl)-*L*-glutamic acid dipotassium salt (1) as a white, amorphous, highly hygroscopic solid (1.83 g, 48%).

¹H NMR (400 MHz, CD₃OD): δ 3.85 (d, *J* = 8.7 Hz, 1H, H-1'); 3.81 (dd, *J* = 12.0, 2.3 Hz, 1H, H-6'a); 3.65 (dd, *J* = 12.0, 5.4 Hz, 1H, H-6'b); 3.48 (dd, *J* = 7.0, 5.7 Hz, 1H, H-2); 3.38–3.35 (m, 1H, H-3'); 3.30–3.26 (m, 1H, H-4'); 3.20–3.16 (m, 1H, H-5'); 3.15 (dd, *J*_a = *J*_b = 8.7 Hz, H-2'); 2.31–2.19 (m, 2H, H-4); 2.10–2.04 (m, 1H, H-3a); 1.94–1.83 (m, 1H, H-3b). ¹³C NMR (101 MHz, CD₃OD): δ 182.9 (COO), 182.1 (COO), 90.8 (C1'), 79.12 (C3'), 79.05 (C5'), 74.9 (C2'), 71.6 (C4'), 62.7 (C6'), 61.6 (C2), 35.9 (C4), 31.7 (C3).

Dry acetic acid was added (0.66 mL) to a stirred solution of 1 (650 mg) in dry methanol (20 mL) and the reaction mixture was refluxed for 30 min under nitrogen. The resulting suspension was cooled to rt and the formed precipitate was filtered off. The obtained clear solution was cooled to -40 °C and upon addition of dry acetone (20 mL) a white solid precipitated. The solid was collected by filtration, washed with dry, cold acetone and dried under reduced pressure over KOH to obtain 2 as a white amorphous solid (350 mg, 53.8%).

¹H NMR (400 MHz, D₂O): δ 3.97–3.94 (m, 1H, H-6'a); 3.93–3.91 (m, 1H, H-4'); 3.83–3.80 (m, 1H, H-5'); 3.72–3.68 (m, 1H, H-6'b); 3.70–3.68 (m, 1H, H-3'); 3.64–3.61 (m, 1H, H-2); 3.22 (d, *J* = 15.0 Hz, H-1'a); 3.18 (d, *J* = 15.0 Hz, H-1'b); 2.43–2.39 (m, 2H, H-4); 2.11–2.03 (m, 2H, H-3). ¹³C NMR (101 MHz, D₂O): δ 179.69 (COO), 174.29 (COO), 95.33 (C2'), 69.86 (C-3'); 69.34 (C-5'); 68.90 (C-4'); 63.84 (C-6'); 63.19 (C-2); 52.59 (C-1'); 33.32 (C-4); 25.42 (C-3). ESI-MS (positive ionization): *m/z* 376.46 (M)

$-2\text{H}^+ + 3\text{Na}^+$). ESI-MS (negative ionization): m/z 308.50 ($\text{M}-\text{H}^+$); 290.50 ($\text{M}-\text{H}_2\text{O}-\text{H}^+$).

Compound 2 was used as standard for the HPLC analysis of the acidic tomato extracts containing the amino acid fraction.

2.6. Analysis of the volatile fraction

2.6.1. Samples preparation and volatiles extraction

For each variety, cropping system, drying method and year, a pool was composed from three biological replicates. Samples were prepared by adding to 2 g of each pool 8 mL of a 20% NaCl solution in a 20-mL vial closed with an aluminum cap and a silicone-PTFE septum. Four technical replicates were prepared for each pool, two for electronic nose (E-nose) measurements and two for gas chromatography-olfactometry (GC-O) analysis. Gas chromatography-mass spectrometry (GC-MS) of selected samples was conducted on the same vials used for E-nose measurements, in order to confirm the sample composition and to identify the compounds not detected by GC-FID and detected by GC-O. The extraction of headspace volatiles for GC was performed by headspace solid-phase microextraction (HS-SPME) using a DVB/CAR/PDMS fiber (absorption step: 45 °C for 30 min; desorption step in the injector port: 250 °C for 5 min in splitless mode).

2.6.2. E-nose

The sensoristic analysis was performed using a PEN3 portable electronic nose (Win Muster Airsense Analytics Inc., Schwerin, Germany) consisting of a sampling section, a detector unit containing the array of sensors and pattern recognition software (Win Muster v. 3.0) for data recording and elaboration. The sensor array is composed of 10 metal oxide semiconductor (MOS) type chemical sensors: W1C (aromatic), W5S (broad range), W3C (aromatic), W6S (hydrogen), W5C (aromatic aliphatics), W1S (broad methane), W1W (sulfur organic), W2S (broad alcohol), W2W (sulfur chlorine), and W3S (methane aliphatics). The sensor response is given by the ratio of the conductivity response of the sensors to the sample gas (G) relative to the carrier gas (G_0) over time (G/G_0). The e-nose analyses were performed after 1 h of conditioning at room temperature. The headspace gas was pumped over the sensor surfaces for 60 s (injection time) at a flow rate of 45 mL min⁻¹, and during this time the sensor signals were recorded. After sample analysis, the system was purged for 120 s with filtered air prior to the next sample injection, to allow re-establishment of the instrument baseline. Each sample was evaluated three times. For each E-nose run, the conductivity G/G_0 of the 10 sensors at the time corresponding to the normalized maximum of all signals was taken as the vector of sensors signal. The average of the runs of each replicate was used for statistical analysis.

2.6.3. GC-MS

The system used for GC-MS was composed of an Agilent 6890 GC connected to an Agilent 5973 MSD detector. Separation was performed on a DB-1 column (60 mm × 0.25 mm I.D., film thickness 0.25 μm) using helium as carrier gas (flow 0.9 mL/min). The column temperature program was: 40 °C for 5 min, 2 °C/min to 160 °C, 4.5 °C/min to 220 °C, 220 °C for 3 min. Injector and detector temperatures were 200 and 240 °C, respectively; interface temperature, 250 °C. The MS settings were as follows: filament voltage, 70 eV; scan range, m/z 45–800; scan speed, 1.4 scan/s. Identification was performed by comparing mass spectra with those of two databases (NIST 08 and Wiley 7 libraries), and comparing their Kovats indices, calculated using *n*-alkanes reference hydrocarbons, with tabulated Kovats indices.

2.6.4. GC-O-FID

In GC-O the volatile compounds are simultaneously detected by FID and human nose after splitting 1:1 of the eluate at the column outlet. The system used was composed of an Agilent 6890 GC unit (Agilent Technologies Italia SpA, Cernusco sul Naviglio, Italy) equipped with an FID and a DB-1 capillary column (injector and detector temperature, 250 °C; carrier gas, He, flow 1.3 mL/min, column length 60 mm × 0.25 mm I.D., film thickness 0.25 μm; temperature program: 50 °C for 5 min, 2.5 °C/min until 160 °C for 3 min, 5 °C/min until 220 °C for 3 min; total duration, 64 min) and connected to an olfactometric system composed of an Olfactory Detector Port ODP2 Gerstel (Gerstel GmbH & Co, KG, Mülheim an der Ruhr, Germany) equipped with the ODPneumatics module to control humidification and make-up gas flows. The GC-O analyses were performed by using a direct intensity method, in which assessors are required to rate both the odor intensity and its duration while the compound is eluting. The olfactometric data (intensity on a 5-point intensity scale where 0 = no odor and 4 = very intense odor, duration and area of each odor event, OE) were collected through a potentiometer with the ODP recorder integrated with the GC software Chemstation Rev A 10.02. The area of each OE is calculated by the software from the intensity and duration values and successively standardized for each assessor, given 100 as maximum value (Mahattanatawee & Rouseff, 2011). FID quantification was performed using solutions of commercial standards at known concentration, when available. The olfactometric analysis was carried out by a panel composed of 7 panelists, aged between 27 and 50 years. Before the analysis of the samples all panelists attended two training sessions to identify the main odor categories. The following standards were used: *n*-hexanal (herbaceous), *trans*-2-hexenal (herbaceous, bug), 2-isobutylthiazole (green tomato), 3-methylbutanoic acid (cheesy, sweaty), geranyl acetone (sweet, floral), citral (citrus, floral), benzaldehyde (almond). Each chromatographic run was conducted in duplicate and divided in two fractions of 25 min, alternately sniffed by the same two panelists.

2.7. Sensory analysis

2.7.1. Samples and sample preparation

The SUN tomato samples were divided into smaller pieces with scissors, in order to make the samples more representative. OVEN samples were divided into smaller pieces using a coffee mill. For both SUN and OVEN samples, 1.0 g was weighed into small plastic beakers with lids (ABENA A/S Aabenraa, Denmark). All samples were coded with 3-digit numbers and were evaluated in three replicates.

2.7.2. Sensory evaluation

A trained sensory panel evaluated the tomato samples described above using sensory descriptive analysis both in 2015 and 2016. In 2015, the sensory panel consisted of 8 assessors, 7 females and 1 male, aged 26 to 61 years; in 2016, the sensory panel consisted of 9 assessors, 7 females and 2 males, aged 26 to 61 years.

The assessors were tested and trained in accordance to international standards (ISO 8586-1, 1993). The sensory evaluation was carried out in a laboratory fulfilling the requirements provided by international standardization (ASTM, 1986). The sensory panel developed the vocabulary based on three representative tomato samples. Prior to the sensory analysis, the assessors attended a two hours training session, where the assessors were introduced to four samples expected to span the perceivable differences for the relevant attributes. After each training session, the assessors received feedback on their

performance using PanelCheck (PanelCheck v.

1.4.0226; www.PanelCheck.com), in order to improve and standardize the panel's discriminating power.

The 2015 and 2016 samples were evaluated differently: 2015 samples were analyzed in a single session ("Cumulative 2015 Test"), while in 2016 three different sensory tests were conducted, as follows: "Test 1 – 2016" = Comparison between SUN and OVEN samples. The following samples were evaluated: SUN-SaAb (CONV), SUN-PerBruzzo (ORG-PA), SUN-PerBruzzo (ORG-PN), OVEN-SaAb (CONV), OVEN-PerBruzzo (ORG-PA) and OVEN-PerBruzzo (ORG-PN); "Test 2 – 2016" = Sensory evaluation of SUN samples. The following samples were evaluated: SUN-SaAb (CONV), SUN-SaAb (ORG-PA), SUN-SaAb (ORG-PN), SUN-PerBruzzo (CONV), SUN-PerBruzzo (ORG-PA) and SUN-PerBruzzo (ORG-PN); "Test 3 – 2016" = Sensory evaluation of the OVEN samples. The following samples were evaluated: OVEN-SaAb (CONV), OVEN-SaAb (ORG-PA), OVEN-SaAb (ORG-PN), OVEN-PerBruzzo (CONV), OVEN-PerBruzzo (ORG-PA) and OVEN-PerBruzzo (ORG-PN). In all tests, samples were served in random order to avoid bias, in three blocks with small breaks in between. White tea, cucumber, water and crackers were served for the assessors to rinse their mouths. A sensory profile for each test was developed for the combined tomato samples, the SUN and OVEN samples, respectively, by the assessors before the evaluation. The descriptors of each profile as well as the used reference samples are shown in Supplementary Table 2. During training and evaluation, the descriptors were evaluated on a 15-cm, non-structured, continuous scale and the ratings were registered using laptops (Compusense, West Guelph, Ontario, Canada). The left side of the scale (= 0) corresponded to the lowest intensity and the right side of the scale (= 15) corresponded to the highest intensity.

2.8. Data analysis

Statgraphics software ver. 5.1 (Manugistics, Rockville, MD) was used to perform the multifactor ANOVA (Tukey HSD test, $p \leq 0.05$) on non-volatile compounds concentrations, e-nose signals, volatile compounds concentrations and GC-OE areas on separate data for each year. Moreover, a multifactor ANOVA was performed on all composition data, in order to assess the effects of the four factors (year, drying, variety, cropping systems) and their interactions. The sensory data were analyzed using a mixed model three-way ANOVA, considering the effects of sample, replicates and assessor, with the assessor and interaction effects as random. Bonferroni LSD values were used to assess the differences between the samples. In order to highlight the main contributors to the differences among samples resulting from sensory data, the sensory descriptors were at first correlated by simple regression with their respective chemical parameters. The reciprocal relationships among significantly correlated variables ($p \leq 0.05$) were further investigated using PCA analysis, performed with PAST software ver. 3.1 (<https://folk.uio.no/ohammer/past/>) on the correlation matrix.

3. Results and discussion

3.1. Non-volatiles

Mean values for non-volatile compounds concentrations are shown in Table 1. The analysis revealed how these indices were strongly influenced by seasonal variation and processing methods, with significance in almost all measured parameters. On the other hand, the differences that arose from the two assayed genotypes and from the cropping systems were limited. Sugar levels were also

significantly affected by the interaction of such factors (Supplementary Table 3).

Non-volatile data are presented in this section as mean values of the three cropping systems, in order to highlight the importance of other factors (Figs. 1–3).

3.1.1. Dry matter, SSC and soluble sugars

The dry matter value (Table 1) for all samples was around 6 g/100 g dw, with no relevant changes between assayed varieties and growing methods. However, a higher variability was found in SUN samples of 2015 compared to SUN from 2016. In 2015, a longer time was needed to complete drying in the solar-drier apparatus, due to unstable weather conditions in 2015. This evidently induced some chemical changes that reflected the variability in dry matter content. The SSC (Table 1, Fig. 1A) depended on the variety, and it showed a significant decrease in SUN and OVEN samples compared to FREEZE. Glucose content (Table 1, Fig. 1B) was on average 21.4 g/100 g dw in FREEZE 2015 samples, not significantly different from FREEZE 2016, where the mean value was 18.9 g/100 g dw. For both years, SUN samples had lower glucose and fructose levels than FREEZE ($p < 0.05$), while no significant differences existed between OVEN and FREEZE. Fructose content (Table 1, Fig. 1C) in FREEZE controls averaged 22.5 g/100 g dw in 2015 and was lower in 2016 at 18.9 g/100 g dw. The reduction in sugar content following thermal treatment was also due to the conversion of soluble sugars into different derivatives, such as Maillard compounds (see below).

The different sugar levels reported in FREEZE in 2015 and 2016 could be an indication that the two sampling year reflected a different ripening stage of the fruit, with 2016 samples being less ripe. This conclusion was also supported by the presence of sucrose, a marker for early ripening stages, in 2016 samples (albeit in very low levels, data not shown), which was absent in 2015.

The CONV and ORG samples gave significantly different sugar contents, especially fructose and total sugars (Table 1), generally lower in ORG tomatoes. There was a strong correspondence with the acidity parameters, especially the citric acid. The higher sugars content found in CONV tomatoes is in full accordance with the data of Györe-Kis, Deak, Lugasi, Csuri-Varga, and Helyes (2013).

3.1.2. TA and organic acids

The first approach to the analysis of the acidic fraction was the measurement of titratable acidity (TA) (Table 1, Fig. 1D). No differences were detected among varieties. Mean TA values for FREEZE 2015 samples were lower than FREEZE 2016 ones (5.0 and 6.5 g/100 g dw citric acid eq., respectively), fully confirming, together with the evidence derived from sugar levels, the differences in ripening stages. In both years, both drying methods reduced TA compared to FREEZE controls; this was particularly evident in SUN 2015 samples. Quantification of citric acid content (Table 1, Fig. 1E), the prevalent organic acid in tomato, revealed that its levels were affected by all factors. In this case, a clear effect of the cropping system was observed, with a higher concentration in ORG samples (Table 1), in accordance with the data reviewed by Araujo and Telhado (2015). The possible cause of this phenomenon could be in the differentiated pattern of ripening in ORG tomatoes with respect to CONV ones, especially in ORG-PN, which are probably less ripe (higher acidity and lower sugar content), due to the stress induced by the root penetration in untilled soil and to the different macronutrients (especially nitrogen) availability caused by the ORG growing system (Lammerts van Bueren et al., 2011).

As for TA, the level of citric acid was higher in FREEZE and lower in dried samples with a reduction of 25% for 2015 and of

Table 1

Concentrations of main non-volatile compounds in dried tomatoes.

			dm (g/ 100 g dw)	SSC (°Bx)	glucose (g /100 g dw)	fructose (g/ 100 g dw)	total sugars(g /100 g dw)	TA (g /100 g dw)	citric acid (mg/100 g dw)	Pyroglutamic acid(mg/100 g dw)	glutamic acid(mg/100 g dw)	Glutamine (mg/100 g dw)	2 (mg/ 100 g dw)	aspartic acid(mg / 100 g dw)	
Year Drying SUN	2015 Variety SaAb	Cropping CONV	7.2 Aa	63.6 Ba	16.1 Ba	15.3 Ba	31.4 Ba	2.9 Ca	2344 Bb	551 Ba	2257 Aa	708 Ba	2912 Ba	153 Ca	
		ORG-PA	7.7 Aa	64.5 Ba	16.3 Ba	16.7 Ba	33.0 Ba	3.1 Ca	4316 Bab	472 Ba	1894 Aa	728 Ba	2716 Ba	131 Ca	
		ORG-PN	7.9 Aa	62.4 Ba	14.3 Ba	15.0 Ba	29.3 Ba	2.9 Ca	4255 Ba	494 Ba	1743 Aa	653 Ba	3442 Ba	190 Ca	
	PerBruzzo	CONV	6.7 Aa	49.2 Ba	10.1 Ba	10.8 Ba	20.9 Ba	2.5 Ca	3735 Bb	517 Ba	1805 Aa	765 Ba	3776 Ba	138 Ca	
		ORG-PA	7.0 Aa	50.9 Ba	11.1 Ba	12.4 Ba	23.5 Ba	2.4 Ca	4343 Bab	545 Ba	1903 Aa	887 Ba	3284 Ba	106 Ca	
		ORG-PN	7.7 Aa	51.8 Ba	9.9 Ba	11.2 Ba	21.1 Ba	2.4 Ca	4655 Ba	512 Ba	2100 Aa	882 Ba	2940 Ba	119 Ca	
	OVEN	SaAb	CONV	5.4 Aa	73.0 Ba	10.8 Ca	20.1 Ba	30.9 Ca	5.2 Ba	4776 Ab	997 Aa	103 Ba	nd	49,503 Aa	1137 Aa
			ORG-PA	5.5 Aa	76.3 Ba	12.2 Ca	20.7 Ba	32.9 Ca	5.3 Ba	4947 Aab	923 Aa	209 Ba	nd	52,931 Aa	1307 Aa
			ORG-PN	6.5 Aa	43.9 Ba	6.7 Ca	11.5 Ba	18.2 Ca	2.9 Ba	5660 Aa	942 Aa	161 Ba	nd	50,769 Aa	1387 Aa
PerBruzzo		CONV	6.0 Aa	55.4 Ba	8.3 Ca	11.8 Ba	20.1 Ca	4.1 Ba	5664 Ab	906 Aa	136 Ba	nd	31,219 Aa	954 Aa	
		ORG-PA	5.9 Aa	45.0 Ba	6.1 Ca	10.0 Ba	16.2 Ca	3.2 Ba	5161 Aab	901 Aa	68 Ba	nd	43,851 Aa	1200 Aa	
		ORG-PN	5.8 Aa	53.2 Ba	8.2 Ca	10.4 Ba	18.6 Ca	3.9 Ba	5625 Aa	799 Aa	131 Ba	nd	38,636 Aa	1170 Aa	
FREEZE	SaAb	CONV	5.6 Aa	74.1 Aa	20.5 Aa	22.5 Aa	42.9 Aa	4.8 Aa	5080 Ab	184 Ca	2323 Aa	1593 Aa	1245 Ba	528 Ba	
		ORG-PA	5.8 Aa	75.5 Aa	23.6 Aa	23.9 Aa	47.5 Aa	5.0 Aa	5875 Aab	216 Ca	2056 Aa	1421 Aa	1549 Ba	488 Ba	
		ORG-PN	6.1 Aa	74.1 Aa	21.8 Aa	22.4 Aa	44.2 Aa	5.4 Aa	6078 Aa	209 Ca	2401 Aa	1218 Aa	1336 Ba	502 Ba	
	PerBruzzo	CONV	5.8 Aa	72.5 Aa	21.5 Aa	23.0 Aa	44.5 Aa	5.1 Aa	5841 Ab	194 Ca	2569 Aa	2236 Aa	1440 Ba	581 Ba	
		ORG-PA	5.7 Aa	72.6 Aa	19.2 Aa	21.3 Aa	40.5 Aa	5.0 Aa	6498 Aab	221 Ca	1619 Aa	1726 Aa	1224 Ba	399 Ba	
		ORG-PN	6.1 Aa	71.4 Aa	21.7 Aa	21.9 Aa	43.6 Aa	5.0 Aa	6715 Aa	167 Ca	1850 Aa	1432 Aa	1408 Ba	425 Ba	
<i>p</i> sign.	Drying	ns	***	***	***	***	***	***	***	***	***	***	***	***	
	Cropping	ns	ns	ns	ns	ns	ns	ns	*	ns	ns	ns	ns	ns	
	Interaction	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns	
Year Drying SUN	2016 Variety SaAb	Cropping CONV	5.8 Aa	51.7 Ba	9.0 Ba	16.5 Ca	25.5 Ca	3.8 Cab	3941 Cb	600 Bb	567 Aa	999 Ab	4600 Ba	276 Ba	
		ORG-PA	5.2 Aa	61.3 Ba	3.1 Bb	6.8 Cb	9.9 Cb	3.6 Cb	3807 Cab	798 Ba	696 Aa	1204 Aa	3529 Ba	341 Ba	
		ORG-PN	6.0 Aa	53.2 Ba	3.4 Bb	6.4 Cb	9.8 Cb	4.2 Ca	4461 Ca	787 Ba	850 Aa	1204 Aab	5371 Ba	380 Ba	
		PerBruzzo	CONV	6.5 Aa	52.8 Ba	9.7 Ba	13.2 Ca	22.9 Ca	3.7 Cab	3369 Cb	476 Bb	634 Aa	889 Ab	2806 Ba	201 Ba
			ORG-PA	5.7 Aa	50.9 Ba	5.8 Bb	10.0 Cb	15.8 Cb	3.6 Cb	4427 Cab	658 Ba	705 Aa	1454 Aa	3716 Ba	249 Ba
			ORG-PN	5.2 Aa	48.5 Ba	4.9 Bb	8.9 Cb	13.7 Cb	4.0 Ca	4510 Ca	789 Ba	540 Aa	937 Aab	3487 Ba	218 Ba

Table 1 (Continued)

			dm (g/ 100 g dw)	SSC (°Bx)	glucose (g /100 g dw)	fructose (g/ 100 g dw)	total sugars(g /100 g dw)	TA (g /100 g dw)	citric acid (mg/100 g dw)	Pyroglutamic acid(mg/100 g dw)	glutamic acid(mg/100 g dw)	Glutamine (mg/100 g dw)	2 (mg/ 100 g dw)	aspartic acid(mg / 100 g dw)
OVEN	SaAb	CONV	6.0 Aa	75.2 Aa	6.3 Ba	16.4 Ba	22.7 Ba	5.0 Bab	4781 Bb	1045 Ab	724 Aa	nd	19,059 Aa	519 Aa
		ORG-PA	5.9 Aa	72.4 Aa	6.0 Bb	14.8 Bb	20.8 Bb	5.1 Bb	5093 Bab	1613 Aa	551 Aa	nd	25,439 Aa	912 Aa
		ORG-PN	5.8 Aa	71.7 Aa	7.6 Bb	14.9 Bb	22.4 Bb	5.1 Ba	5943 Ba	1487 Aa	463 Aa	nd	27,002 Aa	727 Aa
	PerBruzzo	CONV	6.2 Aa	67.5 Aa	4.6 Ba	13.4 Ba	18.1 Ba	5.2 Bab	4630 Bb	907 Ab	668 Aa	nd	18,431 Aa	661 Aa
		ORG-PA	5.5 Aa	70.4 Aa	4.9 Bb	15.2 Bb	20.0 Bb	5.6 Bb	6224 Bab	1364 Aa	678 Aa	nd	23,524 Aa	906 Aa
		ORG-PN	5.6 Aa	66.7 Aa	6.7 Bb	15.0 Bb	21.7 Bb	5.8 Ba	6032 Ba	1122 Aa	591 Aa	nd	25,390 Aa	1087 Aa
FREEZE	SaAb	CONV	6.0 Aa	78.9 Aa	16.0 Aa	20.4 Aa	36.3 Aa	6.5 Aab	6320 Ab	551 Cb	809 Aa	435 Bb	762 Ca	936 Aa
		ORG-PA	5.7 Aa	73.1 Aa	13.6 Ab	17.6 Ab	31.3 Ab	5.9 Ab	6779 Aab	571 Ca	737 Aa	462 Ba	765 Ca	993 Aa
		ORG-PN	5.9 Aa	73.9 Aa	13.8 Ab	19.0 Ab	32.8 Ab	6.7 Aa	6815 Aa	529 Ca	704 Aa	441 Bab	754 Ca	949 Aa
	PerBruzzo	CONV	6.1 Aa	77.1 Aa	15.9 Aa	19.7 Aa	35.5 Aa	6.6 Aab	6703 Ab	456 Cb	448 Aa	281 Bb	902 Ca	603 Aa
		ORG-PA	6.2 Aa	71.3 Aa	13.9 Ab	18.7 Ab	32.6 Ab	6.1 Ab	6758 Aab	491 Ca	466 Aa	292 Ba	786 Ca	628 Aa
		ORG-PN	5.6 Aa	75.7 Aa	13.6 Ab	18.2 Ab	31.8 Ab	7.0 Aa	7951 Aa	451 Ca	536 Aa	336 Bab	681 Ca	722 Aa
<i>p</i> sign.	Drying	ns	***	***	***	***	***	***	***	***	ns	***	***	***
	Cropping	ns	ns	**	*	**	**	**	***	**	ns	ns	*	ns
	Interaction	ns	ns	**	*	**	**	ns	ns	ns	ns	ns	*	ns

Different letters in a column indicate for the same year and for both varieties significant differences (Tukey's test, $p \leq 0.05$), with capital letters corresponding to differences between the two drying methods and lowercase letters corresponding to differences between the three cropping systems. Factors significance: ns = not significant; * = $p < 0.05$; ** = $p < 0.01$; *** = $p < 0.001$; nd = not detected.

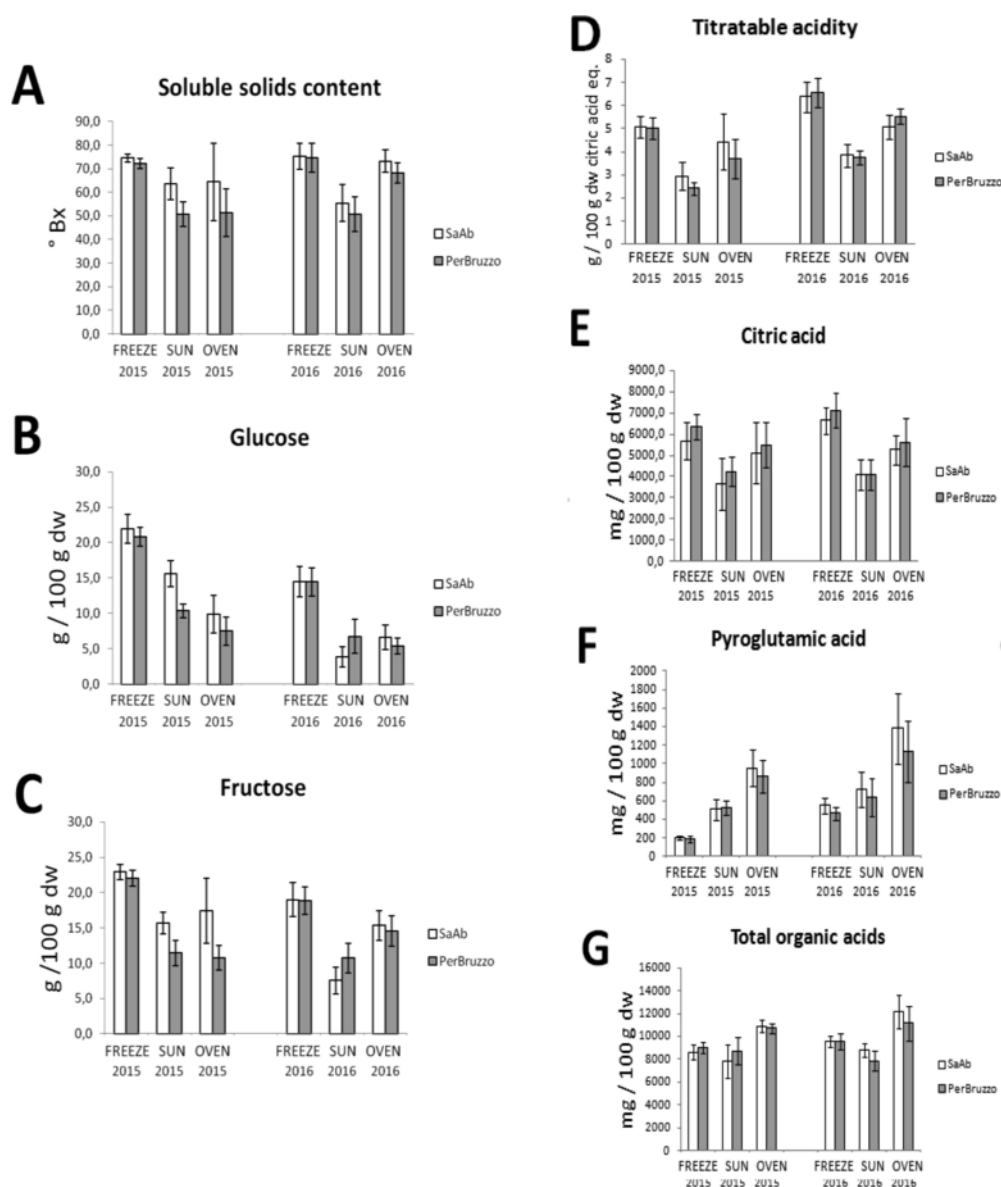


Fig. 1. (A) Soluble solids content; (B) glucose; (C) fructose; (D) titratable acidity; (E) citric acid; (F) pyroglutamic acid; (G) total organic acids, in two different tomato varieties, three types of drying and two sampling years. Each bar is the mean value (\pm SD) of the three growing systems (see Materials and Methods).

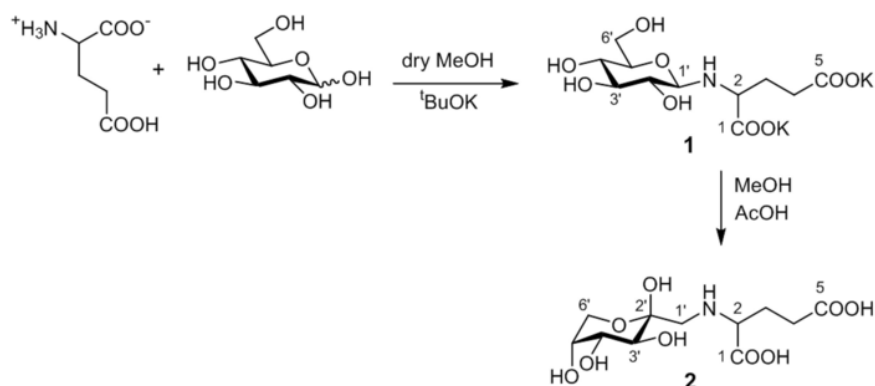
30% for 2016, with no significant differences between SUN and OVEN samples. Pyroglutamic acid derives from glutamine and it is considered a marker for thermal treatment (Schneider, Butz, Ludwig, & Tauscher, 2003). Coherently, pyroglutamic level increased in dried tomatoes (Table 1, Fig. 1F). The highest levels of pyroglutamic acid were detectable in OVEN samples (mean values 0.9g/100g dw in 2015 and 1.3g/100g dw in 2016), although this molecule was present in SUN tomatoes, albeit with lower levels (only 50% compared to OVEN in both years), maybe as a result of the lower working temperatures of the solar drier compared to the hot forced air flux oven; moreover a clear influence of cropping system was detected in ORG dried samples in 2016 season, with a significant higher amount (Table 1). In accordance with previous studies that reported pyroglutamic acid in fresh tomatoes (Mounet et al., 2007), it was also detected in FREEZE samples, with significantly higher levels in 2015 than in 2016 (0.2 and 0.5g/100 g dw, respectively and $p < 0.01$). The data of total organic acid content (Fig. 1G), derived from the sum of citric, pyroglutamic, malic, suc-

cinic, fumaric and *trans*-aconitic, averaged at around 9g/100g dw, with slightly lower contents in SUN samples. Interestingly, the OVEN samples, for the contribution of newly formed organic acids, resulted in a generally higher amount (10–12g/100 g dw) than FREEZE ones. No relevant differences were found in the two sampling years, while some difference was revealed in ORG samples, with a general higher content than the CONV counterparts.

3.1.3. Umami molecules: amino acids and their derivatives

As the umami taste is of great importance in determining tomato flavor and it plays a crucial role in tomato acceptability, quantitative determination of amino acids considered glutamate, the prototypical umami substance, and aspartate, having about 7% of the taste activity of glutamate (Beksan et al., 2003). Moreover, glutamine was also determined. Glutamic acid levels were very different over the two years, with FREEZE 2015 samples significantly richer than FREEZE 2016 ones (mean values 2.1 and 0.6g/100 g dw), for both varieties. This was a further indication that the ripening of

A



B

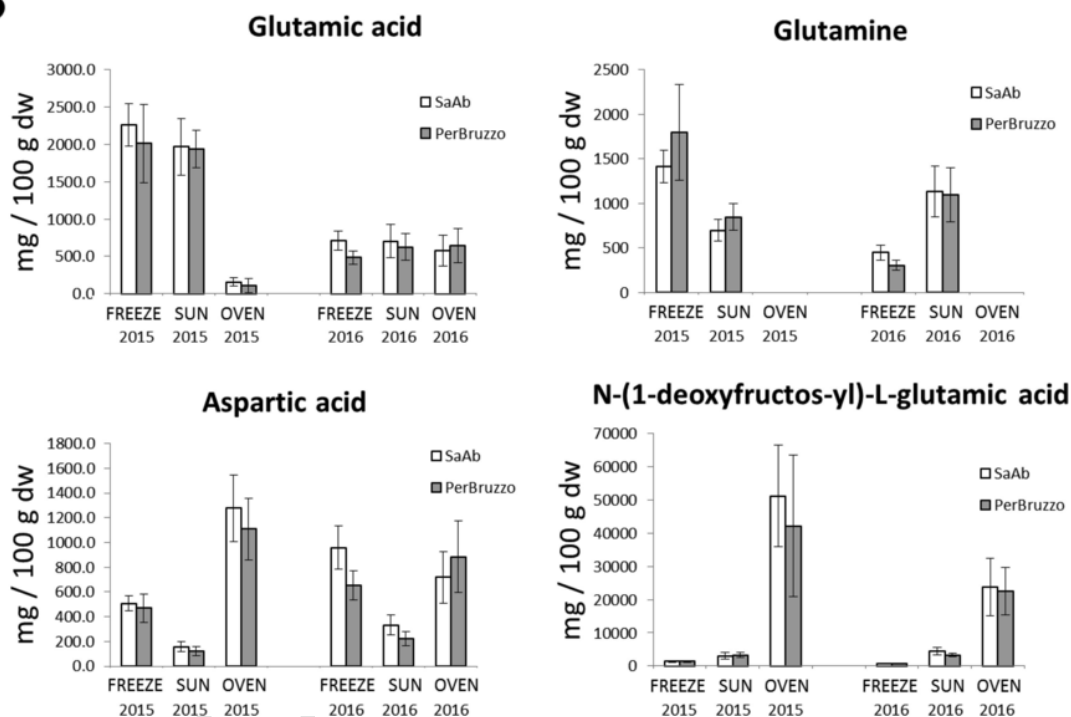


Fig. 2. (A) Chemical synthesis of *N*-(1-deoxy-D-fructos-1-yl)-L-glutamic acid (2); (B) Concentration of the main umami molecules assayed, Each bar is the average (\pm SD) of three growing systems (see Materials and Methods).

these samples was different between the two seasons. In 2015 samples, there was a decrease in the levels of this amino acid due to OVEN drying (-94%), while, unexpectedly, no similar effect was observed in 2016 (Table 1, Fig. 2B). Concentration levels of aspartic acid were significantly lower in SUN than OVEN samples, for both years ($p < 0.01$). The ratio glutamic/glutamine was very different in the two years, with a prevalence of glutamic acid in 2015, that could be an indication of a more advanced ripening (Bortolotti, Boggio, Delgado, Orellano, & Valle, 2003). SUN had considerable levels of glutamine, in accordance with the opposite changes of pyroglutamic acid, previously discussed. Interestingly, in OVEN samples, the glutamine was not detected, because it was completely converted to pyroglutamic acid during the processing.

It is known that the Maillard reaction is chiefly responsible for the development of unique aromas and flavors during thermal processing of foods. It is also known that Amadori compounds, 1-amino-1-deoxyketoses, formed as first stable intermediates of the reaction of reducing sugars with amino acids, are both important aroma precursors and flavor-active compounds (Hellwig & Henle, 2014). *N*-(1-Deoxy-D-fructos-1-yl)-L-glutamic acid (2), arising from the reaction of glutamic acid and glucose, was shown to exhibit a pronounced umami-like flavor quality (Behrens, Meyerhof, Hellfritsch, & Hofmann, 2011; Beksan et al., 2003). Since 2 was identified in dried tomatoes (Behrens et al., 2011), we prepared it by a two-step chemical synthesis following a published procedure (Beksan et al., 2003) with some modifications (Fig. 2A) and used it as reference compound in the analysis of the acidic extracts of tomato sam-

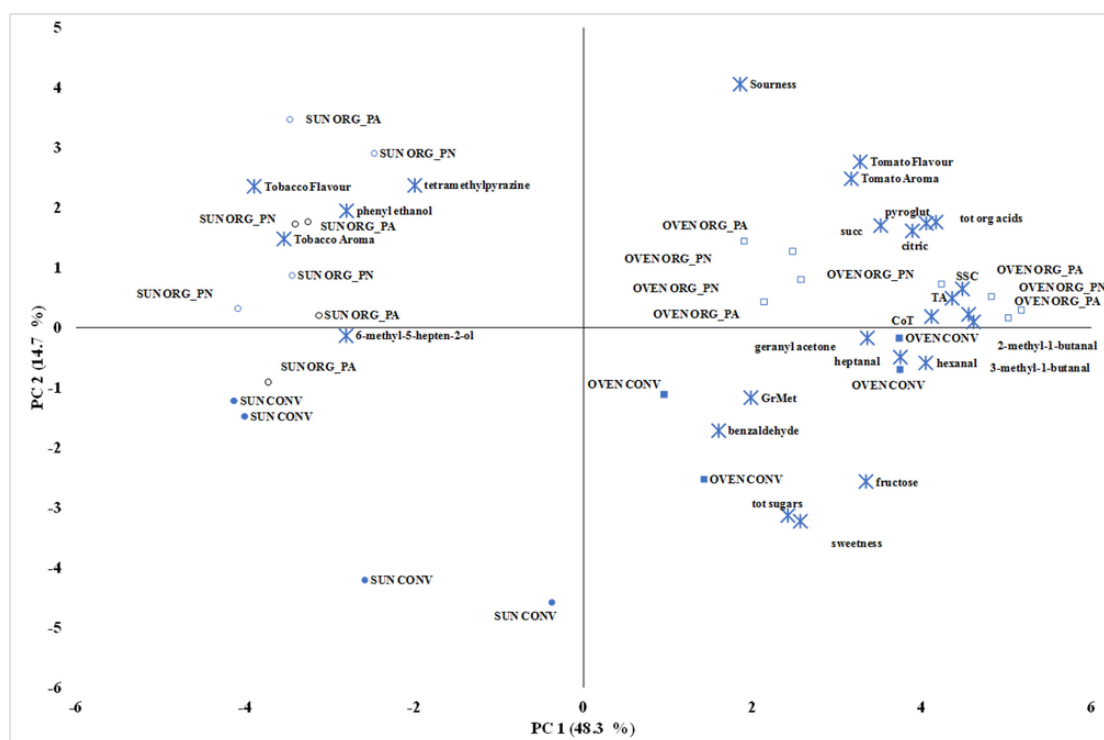


Fig. 3. Biplot of first two PCs of analytical parameters derived from selected non-volatile and volatile compounds, sensory descriptors and odor events. Each point represents a replicate. Symbols significance: Loadings = crosses; OVEN CONV scores = filled squares; OVEN ORG scores = empty squares; SUN CONV scores = filled circles; SUN ORG scores = empty circles. The abbreviations indicating the scores and loadings are explained in the text.

ples. Compound 2 was not detectable in FREEZE tomatoes but was found to be present in both SUN and OVEN samples, thus suggesting that the thermal treatment is responsible for its formation. The average concentration of 2 in OVEN samples is much higher than in the SUN dried ones (about 35 g/100 g dw and 5 g/100 g dw, respectively, with a greater amount in 2015 samples with respect to the 2016 ones). Such values are of the same order of magnitude as those previously found (Behrens et al., 2011).

The cropping generally induced low changes in umami molecules with scarce differences between CONV and ORG tomatoes.

3.2. Volatiles

3.2.1. E-nose profiles

The data analysis indicated that the drying method was the main factor affecting the sensoristic profiles. In 2015, all sensors could distinguish between the two drying methods (Supplementary Fig. 1A), while the cropping system influenced the response of W1C, W3C and W5C (Supplementary Fig. 1B). The E-nose profiles in 2016 samples were more similar, the effect of cropping system not being significant for all sensors, while only 7 out of 10 sensors showed significant G/G_0 differences between the OVEN and SUN samples. Interestingly, in 2016, no differences were observed in the signals of the sulfur-compound-related W1W and W2W sensors (Supplementary Fig. 1A); this in accordance with the data from GC-O analysis (see below).

3.2.2. Volatile compounds concentrations

The main identified (GC-MS) and quantified (GC-FID) compounds (Table 2) were aldehydes (3-methylbutanal, 2-methylbutanal, *n*-hexanal), alcohols (3-methyl-1-butanol, 6-methyl-5-hepten-2-ol), ketones (6-methyl-5-hepten-2-one), acids (3-methylbutanoic acid) and terpenoids (neral, geranial, geranyl acetone, β -ionone). The prevalent compounds were 3-methylbutanoic acid

(ranging from 400 to 3000 ng/g dw) and 6-methyl-5-hepten-2-one (ranging from 100 to 1500 ng/g dw), previously found in tomato volatile extracts (Petro-Turza, 1986; Selli et al., 2014).

The overall influence of selected variables and their interactions are shown in Table 2 and Supplementary Table 4. In this case, the main factors of variation were the year, the drying and the cropping, with the most variability for 6-methyl-5-hepten-2-one and tetramethylpyrazine.

The levels of 6-methyl-5-hepten-2-one, the major lycopene catabolite, were clearly higher in 2015 than in 2016, definitely confirming the different ripening of the two years of sampling. Minor amounts of other apocarotenoids were found, such as β -ionone, deriving from β -carotene cleavage. In the SUN samples tetramethylpyrazine was found, a marker of microbial activity (Zhu, Xu, & Fan, 2010). As for e-nose signals, the drying process was the main source of variation among samples, especially in 2015. In both years, the concentration of the most identified volatiles was higher in OVEN than SUN samples, suggesting that considerable amounts of volatiles were produced by oxidation during the thermal treatment and, on the other hand, that the SUN drying process caused a decrease in volatile content, due both to the lower temperature of the process and to the longer exposure of the samples to the drier air flow. Regarding the other factors, the cropping system overall affected the production of 3-methylbutanal, significantly lower in ORG samples of 2016, and 3-methylbutanoic acid, while significant differences were observed between varieties for 3-methylbutanal, 3-methylbutanoic acid, benzaldehyde, phenyl ethanol, geranyl acetone and β -ionone. Generally, more volatile compounds were found in SaAB than in PerBruzzo, with some difference between the two sampling years (Table 2). Similar volatile profiles were determined in ORG greenhouse tomatoes by Thybo, Edelenbos, Christensen, Sørensen, and Thorup-Kristensen (2006), where differences were found for the sampling years, in accordance with the present data. Other authors (Cuevas, Moreno-Rojas, Arroyo, Daza,

Table 2
Concentrations (ng/g dw) of main volatile compounds in dried tomatoes headspace.

			3-methylbutanal	2-methylbutanal	3-methyl-1-butanol	n-hexanal	3-methylbutanoic acid	n-heptanal	trans-2-heptenal	benzaldehyde	6-methyl-5-hepten-2-one	6-methyl-5-hepten-2-ol	tetram
Year	2015		ng/g dw										
Drying	Variety	Cropping	248Bab	531 Ba	145 Aa	49.9 Ba	1798 Ba	3.5 Ba	10.3 Aab	25.1 Ba	1584 Aa	2210 Aa	22.2 A
SUN	SaAb	CONV											
		ORG-PA	204 Bb	185 Bb	41.3Aa	62.3 Ba	358 Ba	2.7 Ba	3.2 Ab	35.6 Ba	153 Aa	407 Aa	0.6 Aa
		ORG-PN	243 Ba	128 Ba	78.0 Aa	23.5 Ba	1847 Ba	1.1 Ba	21.3 Aa	19.0 Ba	142 Aa	563 Aa	0.5 Aa
	PerBruzzo	CONV	136 Bab	108 Ba	35.1 Aa	38.2 Ba	475 Ba	0.1 Ba	2.2 Aab	12.6 Ba	92 Aa	348 Aa	0.2 Aa
		ORG-PA	343 Bb	114 Bb	52.5 Aa	52.4 Ba	581 Ba	0.2 Ba	0.0 Ab	30.5 Ba	141 Aa	373 Aa	0.2 Aa
		ORG-PN	319 Ba	48 Ba	47.9 Aa	52.4 Ba	473 Ba	0.1 Ba	9.6 Aa	24.1Ba	105 Aa	316 Aa	0.1 Aa
OVEN	SaAb	CONV	3547 Aab	3955 Aa	54.6 Aa	237.7 Aa	3536 Aa	12.9 Aa	9.5 Aab	40.1 Aa	261 Aa	61 Ba	0.0 Aa
		ORG-PA	2194 Ab	2362 Ab	29.8 Aa	264.9 Aa	1045 Aa	16.1 Aa	1.5 Ab	36.9 Aa	200 Aa	38 Ba	0.0 Aa
		ORG-PN	4061 Aa	4082A a	83.8 Aa	373.3 Aa	2874 Aa	19.2 Aa	7.7 Aa	39.9 Aa	375 Aa	81 Ba	0.0 Aa
	PerBruzzo	CONV	3731 Aab	4242 Aa	35.1 Aa	300.7 Aa	879 Aa	8.3 Aa	9.2 Aab	41.0 Aa	253 Aa	52 Ba	0.0 Aa
		ORG-PA	3035 Ab	2878 Ab	52.8 Aa	327.1 Aa	1216 Aa	15.3 Aa	4.9 Ab	41.5 Aa	319 Aa	80 Ba	0.0 Aa
		ORG-PN	4546 Aa	5061 Aa	59.1 Aa	253.4 Aa	1529 Aa	13.1 Aa	4.1 Aa	33.0 Aa	269 Aa	59 Ba	0.0 Aa
	Drying	Interaction	***	***	ns	***	*	***	ns	***	ns	*	ns
<i>p</i> sign.	Cropping	Interaction	***	***	ns	ns	ns	ns	**	ns	ns	ns	ns
	Interaction	Interaction	***	***	ns	ns	ns	ns	*	ns	ns	ns	ns
Year	2016												
Drying	Variety	Cropping	459 Ba	173 Ba	152.8 Aa	147.2 Aa	3311 Bb	11.0 Aa	0.0 Ba	92.5 Aa	467 Ba	88 Ab	37.0 A
SUN	SaAb	CONV											
		ORG-PA	239 Ba	236 Ba	129.6 Aa	44.1 Ab	1192 Ba	1.0 Aa	0.0 Ba	47.6 Aa	367 Bb	286 Aa	304.0
		ORG-PN	298 Ba	137 Ba	230.2 Aa	69.3 Ab	1045 Ba	2.4 Aa	0.0 Ba	36.7 Aa	371 Bab	546 Aa	93.8 A
	PerBruzzo	CONV	356 Ba	150 Ba	283.0 Aa	91.3 Aa	899 Bb	1.3 Aa	0.0 Ba	26.9 Aa	315 Ba	261 Ab	17.3 A
		ORG-PA	290 Ba	58 Ba	299.8 Aa	31.9 Ab	2371 Ba	1.7 Aa	0.0 Ba	20.2 Aa	262 Bb	621 Aa	79.2 A
		ORG-PN	212 Ba	56 Ba	209.2 Aa	25.6 Ab	2874 a	2.1 Aa	0.0Ba	9.8 Aa	511 Bab	354 Aa	201.6
OVEN	SaAb	CONV	2487 Aa	2623 Aa	60.9 Ba	90.3 Aa	3377 Ab	0.8 Aa	9.4 Aa	32.3 Aa	765 Aa	49 Bb	0.0 Ba
		ORG-PA	2451 Aa	3265 Aa	75.9 Ba	72.8 Ab	879 Aa	1.8 Aa	25.3 Aa	31.1 Aa	661 Ab	49 Ba	0.0 Ba
		ORG-PN	2075 Aa	2335 Aa	60.8 Ba	61.4 Ab	879 Aa	0.1 Aa	18.3 Aa	30.2 Aa	535 Aab	47 Ba	0.0 Ba
	PerBruzzo	CONV	3376 Aa	3333 Aa	19.6 Ba	100.8 Aa	878 Ab	6.4 Aa	19.1 Aa	37.2 Aa	794 Aa	53 Bb	0.0 Ba
		ORG-PA	2852 Aa	3156 Aa	68.4 Ba	68.8 Ab	1079 Aa	0.1 Aa	19.6 Aa	30.4 Aa	569 Ab	58 Ba	12.5 B
		ORG-PN	3144 Aa	3243 Aa	90.6 Ba	72.0 Ab	1216 Aa	7.4 Aa	20.7 Aa	33.0 Aa	685 Aab	48 Ba	0.0 Ba
	Drying	Interaction	***	***	***	ns	**	ns	***	ns	***	***	***

<i>p</i> sign.	Cropping	ns	ns	ns	***	**	ns	ns	ns	*	*	ns	**
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Table 2 (Continued)

	3-methylbutanal	2-methylbutanal	3-methyl-1-butanol	<i>n</i> -hexanal	3-methylbutanoic acid	<i>n</i> -heptanal	<i>trans</i> -2-heptenal	benzaldehyde	6-methyl-5-hepten-2-one	6-methyl-5-hepten-2-ol	tetramethylpyrazine
Interaction	ns	ns	ns	ns	ns	ns	ns	ns	ns	*	ns

Different letters in a column indicate for the same year and for both varieties significant differences (Tukey's test, $p \leq 0.05$), with capital letters corresponding to differences between the two drying methods and lowercase letters corresponding to differences between the three cropping systems. Factors significance: ns = not significant; * = $p < 0.05$; ** = $p < 0.01$; *** = $p < 0.001$; nd = not detected.

& Ruiz-Moreno, 2016) reported a significant influence of cropping system on lower aldehydes content of ORG plums, in accordance with the present data on 3-methylbutanal and *n*-hexanal in 2016 (Table 2); moreover, in the latter work, the data variability over the sampling years was fully confirmed.

3.2.3. Gas chromatography–olfactometry (GC–O)

The GC–O results confirmed that the processing method mainly affected the odor quality. The main OE were related to the descriptors “cooked, tomato (CoT)” and “green, metallic (GrMet)” (Supplementary Fig. 2A). The CoT is related to the compound methional, a degradation product of methionine identified by GC–MS and not detectable by FID. The CoT odor was significantly higher in OVEN than SUN tomatoes. 6-Methyl-5-hepten-2-one and 6-methyl-5-hepten-2-ol are responsible for the GrMet; they were affected at $p < 0.05$ by the drying method in 2015 and not affected in 2016 (Supplementary Fig. 2C). The cropping system significantly affected CoT in 2015 ($p < 0.01$).

The minor responses are shown in Supplementary Fig. 2B. Among these, OE “cabbage, sulfurous (CS)”, not detected in SUN tomatoes and significantly influenced by drying ($p < 0.001$), was given by the presence of dimethyl sulfide, identified by GC–MS and not detectable by FID. The OE ChFerm, probably due to 3-methylbutanoic acid, was influenced by variety and cropping system in 2015 ($p < 0.05$), in full accordance with compositional data (Table 2)

3.3. Sensory evaluation

In the cumulative test performed in 2015, six out of nine sensory descriptors differed for OVEN and SUN samples (Table 3). In both 2015 and 2016 data, SUN drying seemed to increase the tobacco aroma and flavor compared to the OVEN samples, while the intensity of tomato flavor was higher in OVEN samples. As for other descriptors, both data from 2015 and 2016 resulted in significant differences for dried tomato aroma, sweetness and sourness, with higher scores in OVEN than SUN samples. In 2015, no variation was found for umami taste among assayed samples, while in 2016, umami taste was higher in the SUN compared to OVEN samples even though the content of glutamic acid was similar for the samples in 2016, so confirming its limited correlation with chemical data (Fig. 2B). In 2015, SUN samples clearly resulted in more bitterness than the OVEN ones, while in 2016 bitterness was to a higher degree affected by the variety, since SaAb was more bitter than PerBruzzo, especially in SUN samples. When SUN samples were tested in 2016, the sensory evaluation showed significant differences between the ORG and the CONV samples for several descriptors. Tobacco aroma, sweetness, sourness, umami, bitterness, tomato flavor, dried fruit flavor, tobacco flavor seemed to vary between the CONV and ORG samples for both varieties (Table 3). The CONV samples of both varieties had a lower intensity of tobacco aroma, sourness, umami, bitterness, tomato flavor and tobacco flavor compared to the ORG samples. In contrast, these samples had higher intensity of dried fruit flavor as well as sweetness. This clear picture of 2016 samples was not observed when the OVEN samples were tested. Fewer descriptors differed between the OVEN compared to SUN samples and only tobacco flavor was lower in CONV compared to ORG samples (Table 3).

3.4. Correlation between chemical composition and sensory descriptors

An explanation of the interactions between sensory descriptors and the respective chemical parameters was performed using sim-

ple regression analysis on a linear model. As expected, the most important determinants for sweetness and sourness were the fructose and the citric acid, the main simple sugar and organic acids of tomato, respectively. Other significant correlations were found for SSC, total sugars, succinic and pyroglutamic acids, TA and total organic acids (Supplementary Table 5). Unexpectedly, umami descriptors scores were not significantly correlated with amino acid concentrations. An in-depth analysis of all the umami molecules would be of great interest and should also include other compounds; including purine 5'-ribonucleotides such as guanosine 5'-monophosphate (GMP), inosine 5'-monophosphate (IMP) and adenosine 5'-monophosphate (AMP), as these molecules were found to induce an umami taste sensation, enhancing synergistically the umami taste of glutamic acid and its corresponding monosodium salt (Behrens et al., 2011).

As for the volatiles (Supplementary Table 6), some positive correlations were found for the compounds influencing the tomato aroma and flavor (Strecker aldehydes, *n*-hexanal, *n*-heptanal, geranyl acetone); the same compounds were negatively correlated with the tobacco aroma and flavor. The opposite situation was found for 6-methyl-5-hepten-2-ol. Moreover, benzaldehyde showed negative correlations with tobacco aroma and flavor, while tetramethylpyrazine and phenyl ethanol were positively correlated with tobacco flavor. Regarding the GC–O data, the CoT odor showed positive correlation with tomato aroma and flavor and negative correlation with tobacco descriptors.

A comprehensive insight into the main factors influencing the differences among samples was achieved using principal component analysis (PCA), which extracted 6 components, together accounting for 88.0% of the data. The first two extracted PC components, plotted in Fig. 3, explained 48.3 and 14.7% of the total variance. As shown in Fig. 3, the expected result regarding the difference between SUN and OVEN samples was clear, with separation on PC1, with negative and positive values for SUN and OVEN samples, respectively. Moreover, the cultivation effect was highlighted by PC2, which divided most of the ORG samples, on the positive axis, from CONV ones, with negative values.

4. Conclusion

It has been proven that in dried tomatoes, the main factor regarding the variability of the measured quality indices is surely the type of processing. This was unexpectedly followed by the sampling year, suggesting that the inner quality indices of tomato may strongly change between two sampling years. According to the meteorological data (Kurze, Lo Scalzo, Campanelli, & Schwab, 2018), the rainfall was significantly higher in 2016, with average daily temperatures from May to August lower in 2016 (21.6 °C) compared to 2015 (22.9 °C). Climatic changes between the two sampling years could have played a significant role for the variation of some previously discussed parameters. The varietal difference did not strongly affect the sample composition, as well as the three different growing conditions, with clear differences between CONV or ORG cropping systems in a limited number of analyzed traits. The relationship between sensory data and chemical indices was significant for fructose and citric acid, the non-volatile compounds responsible for sweetness and sourness, respectively. As for tomato flavor and tomato aroma some volatiles were found correlated, mainly aldehydes. Interestingly, the final approach, using the PCA analysis to discriminate the significantly correlated data, was able to discriminate both the OVEN vs SUN samples, as expected, but also the CONV vs ORG tomatoes, suggesting a multivariate approach with different measured parameters to estimate the impact of the type of cultivation on the quality of tomato. In summary, this

Table 3

Scores of sensory descriptors from assays performed in 2015 ("Cumulative 2015 Test") and 2016 ("Tests 1, 2, 3") sessions (see Materials and Methods). Data are differentiated by type of drying (SUN, OVEN), cultivar (SaAb, PerBruzzo), growing method (CONV, ORG-PA, ORG-PN).

Test	Sample	Tomato flavor	Dried tomato aroma	Sweetness	Sourness	Umami	Bitter	Tobacco aroma	Tobacco flavor	Dried fruit flavor	
Cumulative 2015 Test	SUN-SaAb-CONV	1.5 b	2.7 ab	4.2 abc	6.6	7.8	13.1 a	11.4 a	10.4 b	8.1	
	SUN-SaAb-ORG-PA	3.8 b	1.7 bc	7.6 abc	9.2	6.0	8.0 bc	10.7 a	7.5 b	8.1	
	SUN-SaAb-ORG-PN	4.7b	2.4 bc	6.7 abc	9.4	7.9	7.9 bc	11.5 a	8.0b	9.6	
	SUN-PerBruzzo-CONV	2.4 b	2.5 abc	3.7 bc	8.2	6.9	13.8 a	10.6 a	9.8 b	3.7	
	SUN-PerBruzzo-ORG-PA	3.4 b	2.3 abc	5.4 abc	6.3	7.0	10.6 ab	11.1 a	8.2 b	5.8	
	SUN-PerBruzzo-ORG-PN	3.9 b	2.6 abc	2.7 c	7.7	8.7	12.5 ab	12.8 a	10.6 b	7.8	
	OVEN-SaAb-CONV	11.7 a	8.1 abc	6.9 abc	10.7	8.0	4.7 cd	5.1b	1.7 a	5.1	
	OVEN-SaAb-ORG-PA	12.6 a	8.2 abc	6.8 abc	10.1	6.3	3.9 cd	4.9 b	2.2 a	5.6	
	OVEN-SaAb-ORG-PN	11.9 a	7.9 abc	9.1 a	9.6	7.3	2.6 d	5.3 b	3.0 a	7.9	
	OVEN-PerBruzzo-CONV	12.9 a	9.4 a	9.0 ab	9.5	7.8	3.8 cd	4.1 b	1.2 a	6.9	
	OVEN-PerBruzzo-ORG-PA	12.4 a	8.6 ab	8.9 ab	10.3	6.5	3.6 cd	4.0 b	1.2 a	7.1	
	OVEN-PerBruzzo-ORG-PN	12.7 a	8.9 ab	7.9 abc	11.2	7.6	3.5 cd	4.5 b	1.6 a	6.9	
	<i>p</i> -value	***	***	***	ns	ns	***	***	***	***	ns
	Bonferroni LSD	4.37	6.97	5.46	6.38	7.67	4.86	6.80	5.24	7.92	7.92
Test 1–2016	OVEN-PerBruzzo-ORG-PA	11.7 a	4.2 ab	8.6 ab	8.2 ab	8.6 bc	6.8 ab	6.0 bc	4.7	4.1 bc	
	OVEN-PerBruzzo-ORG-PN	11.3 a	5.0 ab	6.4 abc	11.1 a	9.2 abc	8.3 ab	4.2 c	4.5	4.3 bc	
	OVEN-SaAb-CONV	8.3 ab	7.0 ab	9.8 ab	8.3 ab	6.6 c	5.9 b	5.6 bc	4.6	7.8 ab	
	SUN-PerBruzzo-ORG-PA	8.0 ab	3.5 b	3.0 c	7.8 ab	12.7 a	12.2 a	10.9 a	10.4	3.6 c	
	SUN-PerBruzzo-ORG-PN	7.3 b	4.2 ab	5.2 bc	8.5 ab	10.7 ab	9.9 ab	9.7 ab	10.8	5.4 bc	
	SUN-SaAb-CONV	5.2 b	7.8 a	11.1 a	4.6 b	7.1 bc	4.0 b	7.8 abc	4.7	11.3 a	
	<i>p</i> -value	***	*	***	*	***	**	***	***	***	
	Bonferroni LSD	4.00	4.49	4.91	5.32	4.01	6.20	4.66	5.05	4.04	
	OVEN-SaAb-CONV	6.0b	7.6	11.9 a	4.3b	4.2b	3.9	6.6	3.9b	9.3 a	
	OVEN-SaAb-ORG-PA	11.2 a	4.0	6.8 b	9.3 a	9.3 ab	8.3	8.0	9.4 a	3.6 b	
OVEN-SaAb-ORG-PN	10.2 ab	6.5	6.8 b	9.6 a	10.0 ab	8.3	8.5	6.8 ab	4.5 ab		
OVEN-PerBruzzo-CONV	7.3 ab	5.3	8.7 ab	7.6 ab	6.8 ab	6.1	6.5	4.5 b	7.2 ab		
OVEN-PerBruzzo-ORG-PA	8.6 ab	5.0	8.1 ab	7.8 ab	7.5 ab	6.7	7.6	6.0 ab	4.9 ab		
OVEN-PerBruzzo-ORG-PN	10.8 ab	5.1	6.8 b	11.3 a	9.5 a	9.2	5.6	6.4 ab	4.7 ab		
<i>p</i> -value	*	ns	**	**	*	ns	ns	***	*		
Bonferroni LSD	5.30	4.79	4.45	4.96	5.29	6.53	5.18	3.54	5.10		
Test 3–2016	SUN-SaAb-CONV	6.3 b	9.8 a	12.0 a	4.2 b	3.8 b	3.7 c	3.6 b	2.7 b	11.8 a	
	SUN-SaAb-ORG-PA	11.4 a	5.4 ab	3.9 b	10.2 a	10.3 a	11.7 a	9.1 a	10.3 a	3.6 b	
	SUN-SaAb-ORG-PN	12.4 a	3.5 b	3.7 b	10.7 a	11.0 a	11.8 a	8.8 a	11.5 a	2.4 b	
	SUN-PerBruzzo-CONV	4.5 b	7.8 ab	11.5 a	3.2 b	4.0 b	2.2 c	3.8 b	2.8 b	12.0 a	
	SUN-PerBruzzo-ORG-PA	12.2 a	4.2 b	5.7 ab	8.4 a	9.7 a	9.5 ab	9.7 a	11.1 a	4.4 b	
	SUN-PerBruzzo-ORG-PN	11.1 a	6.9 ab	6.1 ab	9.2 a	8.4 a	8.6 b	5.2 ab	9.0 a	5.3 b	
	<i>p</i> -value	***	**	**	***	***	***	***	**	***	
	Bonferroni LSD	4.26	4.64	6.30	3.93	3.88	3.21	4.80	6.08	5.09	

Different letters indicate statistically significant difference within each set of data, factors significance: ns = not significant, * = $p < 0.05$; ** = $p < 0.01$; *** = $p < 0.001$.

quality evaluation approach was reliable and potentially useful, since, to the best of our knowledge, no comprehensive characterization of flavoring compounds has been performed on organic tomatoes.

Declaration of Competing Interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.foodchem.2019.125062>.

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