

Effect of temperature, pH, and food additives on tomato product volatile levels

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Abstract

Temperature, NaCl, pH, pectin, protein, sucrose, and oil were varied in tomato juice, as a model for flavored tomato sauces, to determine the effect on volatile levels. The headspace concentrations of different tomato juice samples were measured by selected ion flow tube-mass spectrometry (SIFT-MS). Temperature produced the greatest increase, followed by the addition of NaCl. pH and pectin produced no significant difference, while protein, sucrose, and oil decreased volatile levels. Sensory testing showed that NaCl, control, and sucrose had the highest aroma intensity and consumer preference followed by pectin and milk protein and finally oil. In general, the higher the volatile concentration, the stronger the preference, without an indication that any change in volatile ratios produced a detectible off-odor.

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Introduction

Tomatoes are an important agricultural commodity, which possess a characteristic taste and aroma. The production of tomato products rose from 6 to 14 million metric tons from 1980 to 2012 (United States Department of Agriculture, 2012a). Processed tomato products account for 80% of total tomato consumption. Americans consume three-fourths of their tomatoes in processed form. In the late 1980s, the increasing popularity of pizza, pasta, and salsa led to an increase in U.S. consumption of processed tomatoes (United States Department of Agriculture, 2012b). The largest use of processed tomatoes is for sauces which account for 35%, followed by tomato paste, 18%; canned whole tomato products, 17%; and catsup and juice, 15% (United States Department of Agriculture, 2012b).

Many ingredient variations in tomato products are due to a desire to improve the flavor. Consequently, salt, sugar, and oil contents are varied among different brands, and proteins and pectin have been widely used as a fat replacer, thickener, and forming agent. Altering these additives, however, results in different levels of aroma volatiles being released. Salt increases flavor release due to the salting-out phenomenon (Bakierowska and Trzeszczynski, 2004; Ventanas *et al.*, 2010a; Martínez *et al.*, 2012). NaCl decreases the availability of water in food, thus increasing volatility of aroma compounds (Rabe *et al.*, 2003). Sucrose, however, decreases aroma

compounds due to sucrose-sucrose interaction and sucrose-water interaction (Richardson *et al.*, 1987; Roberts *et al.*, 1996; Starzak *et al.*, 2000).

Oil and protein are frequently added to tomato products and also have been reported to have a large effect on flavor volatile levels. Higher contents of either oil (Rosett *et al.*, 1997; Chung *et al.*, 2003; Ventanas *et al.*, 2010b) or protein (Macleod *et al.*, 1988; Thanh *et al.*, 1992; Kuhn *et al.*, 2006) reduce the volatility of some aroma compounds due to their hydrophobic characteristics. The resistance to mass transfer of volatile compounds in fat and oil is higher than in water, thus the release of fat-soluble flavor compounds is delayed because flavor compounds must be released from the lipid phase to the aqueous phase and then released from the aqueous phase to the headspace; thus the volatility of fat-soluble volatile compounds in high-fat-content foods is lower than in low-fat foods (Jo and Ahn, 1999; Guinard *et al.*, 2002). Protein has hydrophobic interactions with volatile compounds depending on the polarity of the protein structure, temperature, and degree of denaturation (Macleod *et al.*, 1988).

Pectin and pH may be adjusted intentionally to change flavor or texture. pH can either increase or decrease flavor perception and volatility (Chen *et al.*, 1984; Hansson *et al.*, 2001a; Kim *et al.*, 2009); pectin can physically entrap flavor molecules within the food matrix, interact with aroma compounds, or form hydrophobic micelles, which capture hydrophobic compounds. These interactions reduce the volatility

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of some aroma compounds (Chinachoti, 1995; Guichard, 1996; Boland *et al.*, 2006).

Tomato products are consumed at different temperatures and the temperature of food is an important factor for food volatile levels (Cardello and Maller, 1982; Zellner *et al.*, 1988; Ventanas *et al.*, 2010b). Flavor release, which is a function of temperature according to Henry's law, is increased by temperature (Bakierowska and Trzeszczynski, 2004; Klooster *et al.*, 2005). The goal of this research was to determine how temperature, pH, and food additives affect the headspace volatile concentration and consumer acceptability of the aroma of flavored tomato products, using tomato juice as a model system.

Materials and Methods

Temperature and pH

To adjust temperature to simulate different serving temperatures, samples were placed into a 25°C or 50°C water bath or refrigerated at 5°C for 1 h to fully equilibrate. Tomato juices (pH 4.26) were adjusted to pH 2.5 and 8.5 with 2 M citric acid solution (Archer Daniels Midland, Decatur, Illinois, U.S.A.) and 1 M sodium hydroxide (Sigma, St. Louis, MO, U.S.A.) using a Model 10 pH Meter (Fisher Scientific, Waltham, Mass., U.S.A.). For 350 mL juice, 40 mL citric acid solution was used to adjust to pH 2.5 and 30 mL NaOH was used to adjust to pH 8.5.

Sodium chloride, pectin, protein, sucrose and oil

To 350 mL tomato juice was added one of the following additives: 1% (w/v) whey protein isolate (Industrial Food Ingredients, Minneapolis, MN, USA), milk protein isolate (Protient Inc., Norfolk, NE, U.S.A.), or collagen (Arnhem Group, Cranford, NJ, USA); 5% or 10% olive oil (Kroger CO., Cincinnati, OH, USA) plus 1% mono and diglycerides (Continental Custom Ingredient, Inc., West Chicago, IL, USA); 5% or 10% (w/v) NaCl (Sigma-Aldrich, Co., St. Louis, MO, USA); 5% or 10% (w/v) sucrose (Domino Food, Inc., Yonkers, NY, USA) or 1% (w/v) low-methoxyl pectin (TIC Gums, Inc., Belcamp, MD, USA). To test volatile levels in the pectin and proteins themselves, 1% pectin, 1% milk protein isolate, 1% whey protein isolate or 1% collagen was added to deionized water.

Measurement of volatile compound concentrations of different samples

Treatments used 50 mL tomato juice filled in 500-mL Pyrex bottles and closed with a silicon septum

cap. Hot break tomato juice (5.72 ± 0.05 %TS and 4.76 ± 0.11 %SS) was produced from fresh Roma tomatoes, which were washed, chopped, pumped through a 88°C hot break system and extracted into juice. The samples were equilibrated in a 45°C water bath for 1 h before measurement unless specified otherwise.

The concentrations of volatile compounds of samples were analyzed by selected ion-flow tube mass spectrometry (SIFT-MS) (SYFT Voice 200, Syft LTD, Christchurch, New Zealand). During the measurement, 2 needles were used. The short passivated needle (5.5 cm), which is the inlet to the apparatus, was placed in the middle of the septum. The long needle (27 cm), which allowed air inside to maintain atmospheric pressure in the bottle, was placed next to the short needle. Water (45°C) was used as a blank between each sample and all samples were measured for 3 min. The flow tube pressure during the machine run was 0.060 ± 0.002 torr. The temperature of the capillary and arm was automatically maintained at 120°C. Each sample was measured using 5 replicates.

The method used a selected ion mode with H_3O^+ , O_2^+ , and NO^+ as precursor ions, developed based on volatile compounds which contribute to tomato flavor and derived from Xu and Barringer (2010) and Azcarate and Barringer (2010). Each volatile compound concentration was calculated by using known kinetic parameters. The concentration [M] of selected volatiles was calculated using the product count rate [Ip], reaction rate constant [k], precursor ions count rate [I], and reaction time [t] as shown in the equation: $[M] = Ip / Ikt$ (Spanel and Smith, 1999). Some compounds produce the same mass for a given precursor ion, in which case a mixture of the interfering compounds were reported. The following pairs are mixtures: isobutanal and 2-methylpropanal, 1-butanol and isobutyl alcohol, benzaldehyde and methyl benzoate, and, (E,E)-2,4-decadienal and citral. Cyclic terpenes are a mixture of alpha-terpinene, terpinolene, β -pinene R-limonene, α -pinene, S-limonene, (+)-aromadendrene, and (E)-cayophyllene. Compounds with irresolvable conflicts or low concentrations are not reported.

Sensory evaluation

The experiment was divided into 2 parts; aroma intensity and preference evaluation. Fifty untrained participants were asked to sniff 20 mL tomato samples with 10% NaCl, 10% sucrose, 1% pectin, 10% oil, 1% milk protein or the control at room temperature and rank the aroma intensities and their preference for each sample from 1 to 6. To avoid bias, three-

digit random numbers were used to represent each treatment and the positions of the samples on the tray were randomized. The samples were filled in an opaque container and closed with a small 6 cm x 6 cm aluminum cap for sniffing.

Statistical analysis and odor active value calculation

Odor active values (OAV) of individual compound were calculated as the ratio of the headspace concentrations obtained and their odor thresholds in water. One-way analysis of variance (ANOVA) was performed on volatile and OAV data by Minitab (Minitab Inc., State college, PA, USA), and Tukey's test was carried out to determine significant differences among mean values of volatile concentrations, OAV, and sensory data. The independent, two-sample T-test was performed on the pectin result. The Friedman test was carried out for statistical difference among groups for sensory data. A significance level of 0.05 was applied throughout the study.

Results and Discussion

The various treatments had different effects on the volatile levels that appear in the headspace, and thus the samples may be perceived differently by the consumer during consumption. Increasing temperature and NaCl increased volatile levels, pH and pectin had no significant effect, and protein, sucrose and oil decreased volatile levels.

Temperature

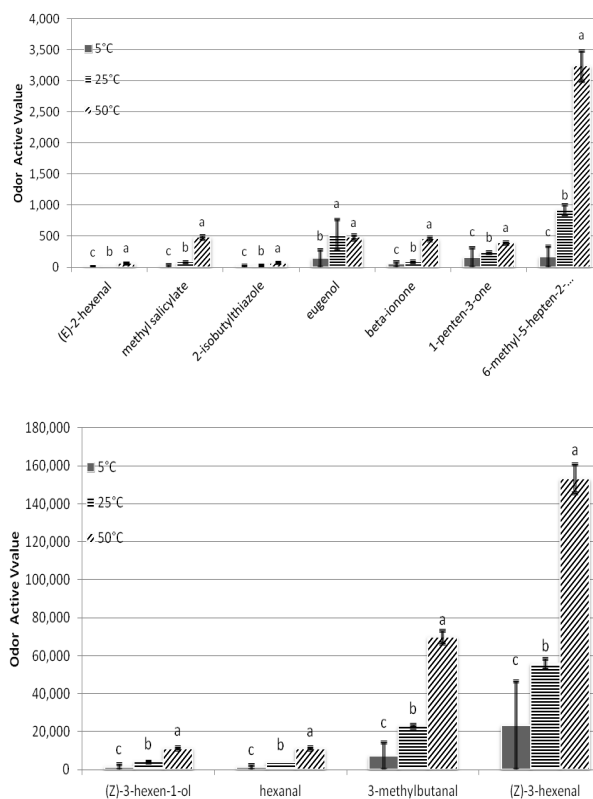
Different temperatures were used to simulate different consumer consumption conditions for different tomato products. Comparing the effect of temperature to the other additives, it is apparent that temperature had a much larger effect on the volatile levels. When the temperature increased, the volatile levels all increased, but by different amounts (Table 1). From 5 to 25°C, the increase varied from 45 to 530% (average 188%), depending on the volatile. There was a 65-518% increase (average 209%) when the temperature increased from 25 to 50°C. The air/water partition coefficient of volatile compounds in the headspace increases as temperature increases, thus the concentration of volatile compounds in the headspace increases and the flavor intensity is stronger (Jouquand *et al.*, 2004).

The volatiles considered important to create tomato aroma are (Z)-3-hexanal, (Z)-3-hexanol, hexanal, 1-penten-3-one, 3-methylbutanal, (E)-2-hexenal, 6-methyl-5-hepten-2-one, methyl salicylate, 2-isobutythiazole, methional, eugenol, and β -ionone

(Buttery, 1993). The odor activity value (OAV) of methional, which was described as "boiled potato like" (Guen *et al.*, 2000), was less than 1 for all treatments, and so should not be detectible to the consumer. All of the other compounds are at detectible levels (Figure 1). (Z)-3-hexanal, (Z)-3-hexanol, (E)-2-hexenal, and hexanal are associated with "fresh green" aroma in tomato juice (Kazeniak and Hall, 1970; Goodman *et al.*, 2002; Xu and Barringer, 2010). 3-methylbutanal was described as "malty" (Fickert *et al.*, 1998) and 2-isobutythiazole has a spoiled wine-like, slightly horseradish-type flavor, becoming rancid, medicine like and metallic if it is used at levels above 50 ppm in tomato juice (Kazeniak and Hall, 1970). The use of 2-isobutythiazole has been patented for use in tomato products as a means of enhancing tomato flavor (Christiansen *et al.*, 2011). "Fruit-like" aroma and "cooked-stewed tomato", at concentrations around 0.75 ppm, was used to describe 6-methyl-5-hepten-2-one (Kazeniak and Hall, 1970). Eugenol was described as "clove" aroma (Laing *et al.*, 1983; De Wijk and Cain, 1994) and β -ionone was described as floral and raspberry (Perez-Cacho and Rouseff, 2008). Methyl salicylate was described as "maraschino-cherry" or "almond" aroma (De Wijk and Cain, 1994) and gave a "canned tomato" note (Kazeniak and Hall, 1970). 1-penten-3-one was described as "fresh/sweet" (Tandon *et al.*, 2000) or "green" (Krumbien and Auerswald, 1998).

(Z)-3-hexanal, 3-methylbutanal, hexanal and (Z)-3-hexen-1-ol had the highest OAV in all treatments, and so should have the greatest aroma impact (Figure 1 and 2). The other volatiles varied slightly in order depending on the temperature or additive used. This might change aroma perceived by consumer; however, the ratios between the volatiles did not change greatly.

The overall effect of temperature on volatile levels is explained by the partition coefficient and Henry's law equations (Bakierowska and Trzeczynski, 2004; Klooster *et al.*, 2005). The partition coefficient $K = C_G/C_L$ (Bakierowska and Trzeczynski, 2004) and the Henry's law constant ($\text{atm} \cdot \text{m}^3 \cdot \text{mol}^{-1}$) is often described as $H_p = RT/K$; where K is the water/gas partition coefficient value, T is the system temperature (K), and R is the gas constant equaling $0.08206 \times 10^{-3} (\text{m}^3 \cdot \text{atm} \cdot \text{mol}^{-1} \cdot \text{K}^{-1})$. An increase in temperature increases the Henry's law constant of volatile compounds. The temperature dependent aroma partition coefficient can be described with Clausius-Clapeyron equations (Covarrubias-Cervantes *et al.*, 2004; Klooster *et al.*, 2005). According to these equations, the logarithm of the partition coefficient is proportional to the inverse



*Odor activity value for each volatile that does not share a letter is significantly different

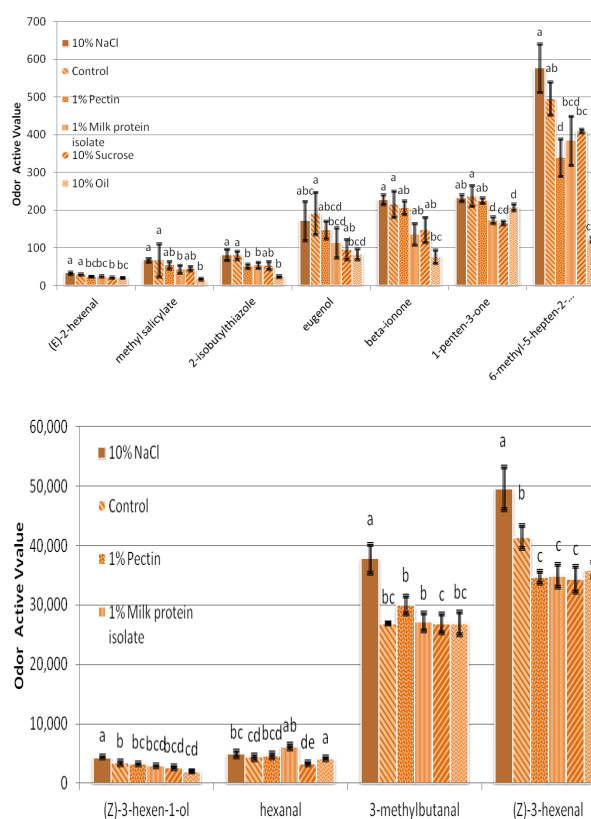
Figure 1. The effect of temperature on odor activity value of volatiles important to tomato aroma

of temperature.

Because of the change in concentration of volatiles and ratio of volatiles in the headspace, temperature plays an important role in the flavor perception of tomato products. Previous studies of odor and flavor perception in a flavored model system have shown that increasing temperature increased perceived flavor intensity (Ventanas *et al.*, 2010a). The odor intensity of cheese soup was stronger at 63°C than at 33°C, and the odor and flavor intensity of carrot, meat patty, and mashed potato increased as serving temperature increased from 25 to 65°C (Kähkönen *et al.*, 1995; Ryyanen *et al.*, 2001).

NaCl

NaCl is frequently added to foods to improve the flavor. NaCl and temperature were the only 2 treatments in this study that increased volatile levels (Tables 1 and 2). As NaCl increased from 0 to 10%, half of the volatile levels significantly increased while the other half of the volatiles showed no significant difference. The increase was 0-60% (average 10%) depending on the volatile. The addition of 10% NaCl produced the highest % increase for alcohols (average 29%) followed by ketones (average 22%), aldehydes (average 19%), and esters (average 16%) (Table 2). In apple juice aroma analysis, an increase



*Odor active value for each volatile that does not share a letter is significantly different

Figure 2. The effect of NaCl, pectin, milk protein isolate (MPI), sucrose, and oil on odor activity value of volatiles important to tomato aroma

from 0-37.5% salt increased the off-aroma because of higher alcohol levels in the headspace (Poll and Flink, 1984). Alcohol showed the highest increase with the addition of salt followed by aldehydes and esters producing an increase in both aroma intensity and off-aroma (Poll and Flink, 1984).

Among the volatiles considered to be most important, only the headspace concentration of (Z)-3-hexenal, (Z)-3-hexen-1-ol, hexanal, and 3-methylbutanal significantly increased at 10% NaCl. These volatiles also had the highest OAV values among important tomato volatiles.

Others have also found an increase in flavor perception and intensity by NaCl addition. In meat broth, the addition of 0.5% NaCl increases the intensity of overall flavor and broth-like flavor (Ventanas *et al.*, 2010a). The mushroom flavor intensity of cooked bologna sausage was shown to be increased by an increase from 1.4 to 2.0% salt (Ventanas *et al.*, 2010b). A reduction from 0.37 to 0.18% salt has previously been shown to decrease total volatile compound levels by 50% in vegetable soup (Mitchell *et al.*, 2011). The partition coefficient of volatiles in a model system increases as salt concentration increases, therefore producing a higher concentration in the headspace (Bakierowska and

Table 1. The effect of temperature (5, 25, and 50°C) on headspace concentration of tomato juice volatiles (ppb)

Volatile compounds (ppb)/ treatments	5°C	25°C	50°C
(E)-2-heptenal	1.71 ^c	3.64 ^b	7.41 ^a
(E)-2-hexenal	0.87 ^c	1.87 ^b	7.32 ^a
(E)-2-nonenal	0.53 ^c	1.22 ^b	3.46 ^a
(E)-2-octenal	0.91 ^c	1.84 ^b	4.94 ^a
(E)-2-pentenal	1.20 ^c	3.48 ^b	12.67 ^a
(E,Z)-2,6-nonadienal	1.51 ^b	2.35 ^b	5.59 ^a
(Z)-3-hexen-1-ol	6.02 ^c	16.36 ^b	44.19 ^a
(Z)-3-hexenal	2.80 ^c	6.71 ^b	18.38 ^a
1-hexanol	3.85 ^c	7.76 ^b	22.66 ^a
1-octen-3-ol	4.68 ^c	7.71 ^b	15.66 ^a
1-penten-3-one	3.65 ^c	5.48 ^b	9.09 ^a
1-propanol	3.33 ^c	16.70 ^b	51.19 ^a
2,3-butanediol	4.09 ^c	12.55 ^b	47.68 ^a
2,3-butanedione	5.87 ^c	11.45 ^b	30.71 ^a
2-isobutylthiazole	0.51 ^c	0.89 ^b	2.04 ^a
2-pentanol	3.59 ^c	10.36 ^b	33.93 ^a
2-pentanone	4.08 ^c	7.47 ^b	16.62 ^a
2-pentylfuran	2.25 ^c	4.38 ^b	11.50 ^a
3-methylbutanal	7.88 ^c	25.23 ^b	76.87 ^a
6-methyl-5-hepten-2-one	11.48 ^c	62.80 ^b	220.16 ^a
Acetaldehyde	27.73 ^c	57.83 ^b	117.67 ^a
Acetone	91.87 ^c	357.85 ^b	1,099.10 ^a
Benzaldehyde	2.51 ^c	9.92 ^b	37.69 ^a
benzene ethanol	0.66 ^c	1.60 ^b	5.41 ^a
benzyl alcohol	0.60 ^c	3.13 ^b	18.22 ^a
beta-ionone	0.40 ^c	0.72 ^b	3.83 ^a
cyclic terpenes	7.56 ^b	28.91 ^b	134.34 ^a
Decanal	0.94 ^c	2.62 ^b	7.04 ^a
dimethyl disulfide	1.61 ^c	3.70 ^b	12.32 ^a
dimethyl sulfide	33.60 ^c	118.16 ^b	541.33 ^a
dodecanal	0.46 ^c	0.73 ^b	1.75 ^a
Ethanol	136.07 ^c	628.35 ^b	2071.74 ^a
ethyl acetate	18.2 ^c	54.97 ^b	119.68 ^a
Eugenol	0.84 ^b	3.16 ^a	2.88 ^a
Furfural	2.01 ^c	4.71 ^b	13.64 ^a
Guaiacol	10.18 ^c	14.84 ^b	30.89 ^a
Hexanal	6.81 ^c	19.62 ^b	55.86 ^a
hexanoic acid	29.11 ^c	113.08 ^b	298.20 ^a
hexyl acetate	19.05 ^c	73.65 ^b	297.57 ^a
Isobutanal	11.89 ^c	30.71 ^b	65.47 ^a
Methanol	1,947.90 ^c	6,388.76 ^b	19,514.43 ^a
Methional	1.51 ^c	3.80 ^b	13.01 ^a
methyl hexanoate	20.67 ^c	80.26 ^b	235.26 ^a
methyl salicylate	0.85 ^c	3.40 ^b	19.43 ^a
methylbutanoic acid	26.16 ^c	77.56 ^b	174.76 ^a
Nonanal	2.97 ^c	5.28 ^b	11.54 ^a
Octanal	5.85 ^c	13.94 ^b	29.00 ^a
Phenylacetaldehyde	4.15 ^c	6.96 ^b	17.00 ^a
Propanal	9.87 ^c	21.73 ^b	49.52 ^a
propanoic acid	8.91 ^c	16.72 ^b	33.44 ^a

*Means in the same row that do not share a letter are significantly different

Trzeszczynski, 2004).

Na⁺ and Cl⁻ bind water molecules with a strong dipole interaction and form hydration shells thus depleting the availability of water molecules for the solubilization of flavor compounds (Rabe *et al.*, 2003; Ventanas *et al.*, 2010a; Mitchell *et al.*, 2011). NaCl decreases the free water of solubilization available for polar compounds as well as creating the iceberg effect around hydrophobic molecules (Shinoda, 1977; Blokzijl and Engberts, 1993; Rabe *et al.*, 2003). This results in an increased concentration of the volatile compounds in the remaining free water. The iceberg effect occurs when numerous layers of water molecules orient around the organic solute, inducing solubility of a nonpolar organic molecule in water. However, the iceberg effect depends on both solvent properties and intrinsic thermodynamic properties of a compound. Therefore, this effect does not occur for all compounds.

pH

The pH of tomato products may be adjusted to

keep it out of the low acid food category, but is not typically adjust for flavor. Changing the pH from 2.5 to 8.5 produced anywhere from 155% increase to 55% decrease, depending on the volatile (Table 3). However, the majority of volatile showed no significant effect of pH. There was also no correlation between pH (2-9) and volatile concentrations in the headspace of a soft drink model system (Hansson *et al.*, 2001a). They found that volatile levels in the headspace can either increase or decrease due to the interaction between the dissociated form of citric acid and volatiles but not due to the change in pH, which may explain why the headspace concentration of some volatiles were significantly higher with the addition of citric acid to fruit pulp (Marsh *et al.*, 2006).

Pectin

Pectin and other thickeners may be added to increase the viscosity of processed tomato products. The addition of pectin produced a significant decrease in only 18% of the volatile compounds with the rest

Table 2. The effect of NaCl (5% and 10%), sucrose (5% and 10%), oil (5% and 10%), and 1% protein (whey protein isolate (wpi), milk protein isolate (mpi), and collagen) on headspace concentration of tomato juice volatiles (ppb)

Volatiles/Treatments	Control	5%NaCl	10%NaCl	5% Sucrose	10% Sucrose	5%Oil	10%Oil	WPI	MPI	Collagen
(E)-2-heptenal	2.66 ^a	2.34 ^{abc}	2.59 ^{ab}	2.33 ^{abc}	2.07 ^c	2.08 ^c	2.10 ^c	2.16 ^{bc}	2.15 ^c	2.29 ^{abc}
(E)-2-hexenal	3.32 ^{ab}	3.02 ^{bc}	3.69 ^a	2.87 ^{cd}	2.48 ^{def}	2.27 ^f	2.33 ^{ef}	2.73 ^{cdef}	2.76 ^{cde}	2.86 ^{cd}
(E)-2-nonenal	1.02 ^a	0.80 ^{abc}	0.88 ^{ab}	0.73 ^{bc}	0.71 ^{bc}	0.57 ^{bc}	0.61 ^c	0.75 ^{abc}	0.80 ^{abc}	0.85 ^{abc}
(E)-2-octenal	5.35 ^a	4.35 ^b	5.55 ^a	3.50 ^f	3.31 ^f	1.90 ^d	1.65 ^d	3.50 ^c	3.59 ^c	3.81 ^{bc}
(E)-2-pentenal	8.97 ^b	8.11 ^c	9.84 ^a	7.36 ^e	7.00 ^{ed}	5.71 ^e	5.43 ^e	7.05 ^d	7.12 ^d	7.19 ^d
(E,Z)-2,6-nonadienal	2.21 ^a	1.52 ^{bc}	1.91 ^{ab}	1.18 ^c	1.36 ^{bc}	1.38 ^{bc}	1.58 ^{bc}	1.52 ^{bc}	1.51 ^{bc}	1.84 ^{ab}
(Z)-3-hexen-1-ol	13.71 ^b	13.71 ^b	17.41 ^a	10.89 ^{cd}	10.52 ^d	8.67 ^e	8.18 ^e	12.29 ^{bc}	11.65 ^{cd}	12.28 ^{bc}
(Z)-3-hexenal	4.98 ^b	4.71 ^{bc}	5.97 ^a	4.28 ^{cd}	4.13 ^{cd}	3.71 ^d	4.32 ^{cd}	4.12 ^{cd}	4.20 ^{cd}	3.96 ^d
1-hexanol	4.85 ^b	4.85 ^{bc}	6.00 ^a	3.97 ^d	3.76 ^{da}	3.17 ^e	3.22 ^e	3.89 ^d	3.96 ^d	4.22 ^{cd}
1-octen-3-ol	6.76 ^a	5.62 ^{abc}	6.63 ^{ab}	4.91 ^{cd}	4.75 ^{cd}	3.83 ^d	4.5 ^{cd}	4.95 ^{cd}	4.94 ^{cd}	5.52 ^{bc}
1-penten-3-one	5.47 ^a	4.57 ^{cd}	5.35 ^{ab}	4.07 ^{def}	3.82 ^f	4.82 ^{abc}	4.78 ^{bcd}	4.00 ^{ef}	3.98 ^{ef}	4.40 ^{def}
1-propanol	15.49 ^c	18.57 ^b	24.88 ^a	14.8 ^{cd}	14.13 ^d	14.81 ^{cd}	14.56 ^{cd}	13.92 ^d	14.53 ^{cd}	14.35 ^{cd}
2,3-butanediol	14.35 ^b	14.12 ^{bc}	18.02 ^a	12.65 ^{cde}	11.55 ^e	12.38 ^{de}	12.85 ^{bde}	13.71 ^{bcd}	13.3 ^{bcd}	13.34 ^{bcd}
2,3-butanedione	13.99 ^{bc}	14.47 ^b	17.78 ^a	14.86 ^b	12.1 ^{de}	14.27 ^b	13.59 ^{bcd}	14.37 ^b	12.63 ^{cde}	11.89 ^e
2-isobutylthiazole	2.34 ^a	1.98 ^{ab}	2.37 ^a	1.56 ^b	1.56 ^b	0.67 ^c	0.71 ^c	1.62 ^b	1.58 ^b	1.85 ^{ab}
2-pentanol	8.02 ^c	9.63 ^b	12.66 ^a	7.89 ^{cd}	7.21 ^{de}	7.07 ^{de}	6.62 ^e	7.56 ^{cd}	7.36 ^{cde}	7.82 ^{cd}
2-pentanone	5.66 ^a	4.79 ^b	5.97 ^a	4.61 ^b	4.24 ^b	4.25 ^b	4.56 ^b	4.47 ^b	4.51 ^b	4.82 ^b
2-pentylfuran	4.68 ^a	3.85 ^{bc}	4.35 ^{ab}	3.57 ^c	3.36 ^c	2.16 ^d	2.26 ^d	3.61 ^{bc}	3.32 ^c	3.79 ^{bc}
3-methylbutanal	29.66 ^{de}	34.72 ^{bc}	41.67 ^a	37.68 ^b	29.62 ^{de}	32.93 ^{cd}	29.68 ^{de}	32.84 ^{cd}	29.94 ^{de}	26.91 ^e
6-methyl-5-hepten-2-one	33.69 ^{ab}	33.51 ^{ab}	39.20 ^a	29.60 ^{bc}	27.87 ^{bc}	10.74 ^d	8.35 ^d	27.91 ^{bc}	26.12 ^c	31.16 ^{bc}
acetaldehyde	58.59 ^{bcd}	60.72 ^{bc}	70.55 ^a	59.45 ^{bc}	49.45 ^e	59.2 ^{cd}	55.56 ^{cd}	61.41 ^b	58.71 ^{bcd}	53.49 ^{de}
acetone	537.63 ^{bc}	564.51 ^b	713.19 ^a	501.8 ^{de}	467.91 ^e	486.73 ^{de}	493.41 ^{de}	512.76 ^{cd}	511.73 ^{cd}	500.06 ^{de}
benzaldehyde	11.52 ^{bc}	12.50 ^b	17.64 ^a	10.19 ^{cd}	9.19 ^{de}	8.54 ^e	8.16 ^e	11.53 ^{bc}	12.40 ^b	10.98 ^{bc}
benzene ethanol	2.29 ^{ab}	2.07 ^{abc}	2.69 ^a	1.83 ^{bc}	1.77 ^{bc}	1.35 ^{bc}	1.67 ^c	1.82 ^{bc}	1.71 ^{bc}	1.80 ^{bc}
benzyl alcohol	4.42 ^{ab}	3.73 ^{bc}	4.90 ^a	3.73 ^{bc}	3.16 ^c	3.27 ^c	4.05 ^{abc}	4.15 ^{abc}	3.75 ^{bc}	3.92 ^{abc}
beta-ionone	1.82 ^a	1.56 ^{ab}	1.92 ^a	1.39 ^{ab}	1.24 ^{bc}	0.54 ^d	0.65 ^{cd}	1.32 ^{ab}	1.15 ^{bcd}	1.64 ^{ab}
cyclic terpenes	3.61 ^a	3.05 ^{ab}	3.65 ^a	2.94 ^{ab}	2.58 ^b	0.91 ^c	0.92 ^c	2.79 ^b	2.51 ^b	3.14 ^{ab}
decanal	1.21 ^c	0.80 ^d	0.93 ^{cd}	0.70 ^d	0.64 ^d	2.34 ^b	3.11 ^a	0.77 ^d	0.88 ^{cd}	0.93 ^{cd}
dimethyl disulfide	6.89 ^b	7.16 ^b	9.64 ^a	5.97 ^b	5.69 ^{bc}	3.84 ^c	3.88 ^c	9.56 ^a	7.21 ^b	5.41 ^{bc}
dimethyl sulfide	5,191.22 ^a	4,758.39 ^b	5,339.91 ^a	4,252.64 ^c	4,214.55 ^c	2,960.12 ^d	2,651.15 ^e	4,300.67 ^c	4,323.73 ^c	4,242.86 ^c
dodecanal	0.63 ^a	0.40 ^b	0.53 ^a	0.38 ^b	0.34 ^b	0.40 ^b	0.47 ^{ab}	0.41 ^b	0.49 ^{ab}	0.50 ^{ab}
ethanol	579.41 ^c	544.51 ^b	638.80 ^a	481.19 ^d	464.09 ^d	367.28 ^e	344.33 ^e	474.81 ^d	478.88 ^d	481.89 ^d
ethyl acetate	14.95 ^a	13.18 ^b	16.26 ^a	11.16 ^c	10.68 ^c	10.63 ^c	10.66 ^c	11.22 ^c	11.79 ^{bc}	12.85 ^b
eugenol	1.15 ^a	0.83 ^{abc}	1.03 ^{ab}	0.66 ^{bc}	0.57 ^c	0.45 ^c	0.50 ^c	0.69 ^{bc}	0.68 ^{bc}	1.10 ^{ab}
furfural	19.53 ^a	16.36 ^b	18.66 ^a	14.89 ^{bc}	14.46 ^c	10.22 ^d	9.53 ^d	14.72 ^{bc}	14.38 ^c	15.44 ^{bc}
guaiaicol	16.91 ^a	12.72 ^b	15.77 ^a	9.79 ^{cd}	9.2 ^{cd}	7.89 ^d	8.5 ^d	12.08 ^{bc}	11.96 ^{bc}	12.55 ^b
hexanal	22.34 ^{cd}	20.72 ^{de}	25.16 ^{bc}	18.89 ^{de}	16.90 ^f	21.83 ^{cd}	20.88 ^d	27.11 ^b	31.19 ^a	19.76 ^{de}
hexanoic acid	6.99 ^a	4.92 ^{bcd}	5.90 ^b	4.38 ^{cd}	3.97 ^d	4.19 ^{cd}	4.52 ^{cd}	4.17 ^{cd}	4.55 ^{cd}	5.03 ^{bc}
hexyl acetate	6.57 ^a	4.65 ^{bc}	5.38 ^b	3.63 ^a	3.64 ^a	3.52 ^a	3.75 ^{de}	3.98 ^{cde}	4.34 ^{de}	4.83 ^{bc}
isobutanol	23.62 ^{cd}	21.90 ^{de}	25.77 ^{ab}	19.30 ^{ef}	17.79 ^f	18.91 ^{ef}	17.68 ^f	27.21 ^a	24.65 ^b	20.74 ^{ef}
methanol	10,556.66 ^{bcd}	10,945.79 ^{bc}	13,086.63 ^a	10,223.26 ^{de}	9,699.44 ^e	10,504.15 ^{bcd}	11,147.12 ^b	10,149.26 ^{de}	10,364.96 ^{de}	10,107.51 ^{de}
methional	3.50 ^{ab}	3.19 ^{abc}	3.86 ^a	2.58 ^{de}	2.48 ^{de}	2.18 ^e	2.43 ^{de}	3.00 ^{bcd}	3.03 ^{bd}	2.92 ^{bcd}
methyl hexanoate	6.52 ^a	4.38 ^{bcd}	5.27 ^b	3.75 ^d	3.64 ^d	3.61 ^d	3.92 ^{cd}	3.78 ^d	4.00 ^{cd}	4.89 ^{bc}
methyl salicylate	2.69 ^a	2.23 ^{abc}	2.70 ^a	1.99 ^{bc}	1.82 ^{bc}	0.69 ^d	0.71 ^d	1.85 ^{bc}	1.77 ^c	2.33 ^{ab}
methylbutanoic acid	12.74 ^a	11.02 ^b	13.44 ^a	9.80 ^{bcd}	9.05 ^d	9.09 ^d	9.24 ^d	9.58 ^{cd}	9.69 ^{cd}	10.74 ^{bc}
nonanal	3.71 ^a	2.15 ^{bcd}	2.68 ^{abcd}	1.78 ^{cd}	1.58 ^d	2.36 ^{abcd}	2.88 ^{abcd}	3.61 ^{ab}	2.47 ^{abcd}	3.16 ^{abc}
octanal	9.43 ^a	6.25 ^{bcd}	7.59 ^{ab}	5.27 ^{de}	4.89 ^{de}	3.84 ^e	4.00 ^e	6.19 ^{bcd}	5.79 ^{bcd}	7.10 ^{bc}
phenylacetaldehyde	19.08 ^a	16.64 ^{bcd}	18.48 ^{ab}	15.13 ^{def}	14.71 ^f	14.01 ^f	14.94 ^{def}	16.37 ^{cde}	16.81 ^{bd}	17.07 ^{bc}
propanal	15.54 ^d	15.09 ^d	17.95 ^c	12.89 ^{ef}	11.56 ^f	19.75 ^b	21.38 ^a	14.37 ^{de}	13.56 ^{ef}	13.62 ^{ef}
propanoic acid	10.15 ^a	8.73 ^b	10.40 ^a	7.64 ^b	7.45 ^b	7.70 ^b	8.31 ^b	7.80 ^b	8.04 ^b	8.33 ^b

*Means in the same row that do not share a letter are significantly different

showing no difference (Table 3). Previous studies have found a decrease in volatile compound levels with the addition of 1-2.5% pectin (Boland *et al.*, 2006; Hansson *et al.*, 2001b; Lubbers and Guichard, 2003). Pectin was expected to decrease volatile levels because the overall intensity and typical flavor note in jam and some volatile levels decreased when 0.5% pectin was added (Guichard, 1996). The partition coefficient of compounds in a gel system and a fruit pastille was decreased by 1-2.5% pectin because of flavor entrapment (Lubbers and Guichard, 2003; Boland *et al.*, 2006). In this study, the addition of 1% pectin also produced a sharp increase for 1-propanol due to the initial 1-propanol levels in the pectin itself.

Hydrocolloids may delay flavor release due

to 3 mechanisms (Boland *et al.*, 2004). One is the physical entrapment of flavor molecules within the food matrix. The presence of an entangled polymer network in thickened systems inhibits the transport of small molecules, such as flavor volatiles from within the gel system to the surface. The macromolecule network seems to play a vital role in flavor release, and entanglement of the polygalacturonic chains may be responsible for the decrease of flavor mobility in a gel, thus reducing the release of volatiles into the vapor phase. Pectin significantly increased viscosity of tomato juice, from 0.07 to 0.30 Pa-s. Second, the stretching of pectin molecules in water aligns them with other molecules and they form micelles, which are more hydrophobic because bound water

Table 3. The effect of pH (2.5, 4.26, AND 8.5) AND 1% pectin on headspace concentration of tomato juice volatiles (ppb)

Volatile compounds (ppb)/ treatments	pH 2.5	pH 4.26	pH 8.5	Control	1% Pectin	Pectin in water
(E)-2-heptenal	2.95 ^a	2.79 ^a	2.96 ^a	3.15 ^a	2.65 ^a	2.15
(E)-2-hexenal	2.31 ^b	2.36 ^b	6.03 ^a	2.81 ^a	2.71 ^a	0.81
(E)-2-nonenal	1.16 ^a	1.10 ^a	0.99 ^a	0.96 ^a	1.15 ^a	1.04
(E)-2-octenal	3.20 ^a	2.64 ^b	3.02 ^{ab}	3.98 ^a	2.38 ^a	1.06
(E)-2-pentenal	6.73 ^a	5.66 ^b	6.72 ^a	7.59 ^a	5.78 ^a	0.64
(E,Z)-2,6-nonadienal	2.14 ^a	1.81 ^a	1.96 ^a	2.18 ^a	1.69 ^a	1.90
(Z)-3-hexen-1-ol	13.16 ^a	12.23 ^a	12.78 ^a	12.77 ^a	13.25 ^a	5.54
(Z)-3-hexenal	3.98 ^a	4.11 ^a	4.00 ^a	4.83 ^a	4.17 ^a	2.00
1-hexanol	4.44 ^a	4.22 ^a	4.51 ^a	5.23 ^a	5.07 ^a	2.97
1-octen-3-ol	6.63 ^a	6.06 ^b	5.97 ^b	7.54 ^a	6.50 ^b	4.90
1-penten-3-one	4.92 ^a	4.74 ^a	4.84 ^a	5.84 ^a	5.19 ^a	4.08
1-propanol	47.29 ^a	38.52 ^b	18.38 ^c	14.68 ^b	554.92 ^a	466.37
2,3-butane diol	11.61 ^a	10.40 ^c	11.07 ^b	13.26 ^a	13.27 ^a	3.41
2,3-butanedione	11.04 ^a	11.44 ^a	11.73 ^a	14.66 ^a	15.85 ^a	7.08
2-isobutylthiazole	0.74 ^b	1.63 ^a	1.66 ^a	1.78 ^a	1.50 ^a	0.57
2-pentanol	7.20 ^a	6.47 ^b	7.32 ^a	7.32 ^b	8.19 ^a	2.39
2-pentanone	4.86 ^b	5.42 ^a	5.61 ^a	5.56 ^a	5.29 ^b	3.89
2-pentylfuran	4.71 ^a	4.30 ^a	3.71 ^b	4.44 ^a	3.89 ^a	2.33
3-methylbutanal	22.43 ^a	23.91 ^a	21.09 ^b	26.61 ^a	33.06 ^a	3.97
6-methyl-5-hepten-2-one	59.20 ^a	62.11 ^a	63.40 ^a	26.48 ^a	23.08 ^a	1.96
Acetaldehyde	49.99 ^{ab}	51.84 ^a	46.22 ^b	56.35 ^a	100.33 ^a	53.94
Acetone	424.11 ^a	363.11 ^b	403.20 ^a	465.49 ^a	488.48 ^a	11.5
Benzaldehyde	9.87 ^a	8.37 ^b	9.62 ^a	10.94 ^a	12.41 ^a	1.89
benzene ethanol	2.84 ^a	1.63 ^b	1.56 ^b	2.11 ^a	2.12 ^a	0.74
benzyl alcohol	2.11 ^{ab}	1.71 ^b	2.35 ^a	4.32 ^a	3.98 ^a	2.36
beta-ionone	1.40 ^a	1.10 ^a	1.23 ^a	2.07 ^a	1.74 ^a	0.81
cyclic terpenes	3.31 ^b	3.78 ^b	5.44 ^a	3.38 ^a	5.27 ^a	5.41
Decanal	1.87 ^a	1.56 ^b	1.52 ^b	1.10 ^a	1.15 ^a	1.32
dimethyl disulfide	5.99 ^a	4.50 ^b	5.80 ^a	8.28 ^a	5.99 ^a	0.75
dimethyl sulfide	3,184.00 ^a	2,413.29 ^b	3,178.46 ^a	4485.41 ^a	2883.59 ^a	1.24
dodecanal	0.67 ^a	0.59 ^a	0.57 ^a	0.70 ^a	0.50 ^a	0.64
Ethanol	376.53 ^a	310.95 ^b	369.96 ^a	490.39 ^a	383.87 ^a	22.38
ethyl acetate	12.01 ^a	13.78 ^b	12.31 ^a	14.16 ^a	14.90 ^a	12.02
Eugenol	1.70 ^a	0.90 ^b	0.84 ^b	1.26 ^a	0.88 ^b	1.02
Furfural	12.29 ^a	9.26 ^b	11.04 ^b	16.88 ^a	11.95 ^b	1.56
Guaiacol	15.71 ^a	13.53 ^a	13.86 ^a	18.05 ^a	14.78 ^b	12.00
Hexanal	27.69 ^a	30.14 ^a	24.08 ^a	18.10 ^a	23.95 ^a	11.60
hexanoic acid	4.77 ^b	5.81 ^a	4.51 ^b	6.63 ^a	5.96 ^a	6.81
hexyl acetate	6.21 ^a	6.42 ^a	5.84 ^a	8.23 ^a	6.28 ^a	7.20
Isobutanal	18.52 ^a	17.65 ^a	15.92 ^b	21.20 ^a	19.83 ^b	6.47
Methanol	7,993.39 ^a	6,853.69 ^b	8,149.25 ^a	11,647.21 ^a	12,650.70 ^a	80.06
Methional	3.04 ^a	2.62 ^b	3.03 ^a	3.95 ^a	3.75 ^a	2.05
methyl hexanoate	4.57 ^a	4.94 ^a	3.97 ^b	6.56 ^a	5.63 ^b	6.75
methyl salicylate	3.49 ^a	3.23 ^{ab}	2.75 ^b	2.52 ^a	2.18 ^a	0.81
methylbutanoic acid	11.94 ^a	12.01 ^a	10.39 ^b	14.25 ^a	13.62 ^a	13.61
Nonanal	6.58 ^a	5.11 ^a	4.96 ^a	3.76 ^a	3.16 ^a	3.82
Octanal	11.66 ^a	10.49 ^{ab}	9.42 ^b	9.75 ^a	7.27 ^b	5.94
Phenylacetaldehyde	7.13 ^a	7.65 ^a	8.30 ^a	18.04 ^a	15.96 ^a	15.89
Propanal	14.13 ^a	14.23 ^a	12.38 ^b	13.97 ^a	15.69 ^a	9.53
propanoic acid	7.68 ^b	7.69 ^b	10.69 ^a	10.56 ^a	9.70 ^b	6.39

*Means in the same row for pH or pectin that do not share a letter are significantly different.

Statistical calculations were done separately for pH and pectin treatments

is replaced by intermolecular hydrogen bonds (Chinachoti, 1995). The more nonpolar compound may be captured in the hydrophobic parts of the pectin solution, while the other less hydrophilic compounds are not affected (Hansson *et al.*, 2001b). The third mechanism may involve interactions between the flavor molecules and the gel components (Boland *et al.*, 2004).

Protein

Meat, milk, and cheese are commonly added to tomato sauce therefore the addition of collagen, milk protein isolate, and whey protein isolate were tested. The volatile concentrations in tomato juice significantly decreased with the addition of 1% protein (Table 2). Milk protein produced 4-41% decrease (average 24%). Whey protein isolate

produced a 2-42% decrease (average 9%). Collagen produced a 2-29% decrease (average 15%). Others have also found protein-flavor binding interaction by proteins such as whey protein isolate and milk protein isolate (Macleod *et al.*, 1988; Mottram *et al.*, 1996; Kuhn *et al.*, 2006). Different proteins have different binding sites, but for most volatiles in this study there were no significant differences between the 3 types of proteins tested. The strongest affinity to soy protein is aldehydes, followed by ketones and alcohols, with no binding affinity to acid (Macleod *et al.*, 1988). In this study, aldehydes and ketones were also decreased by the addition of protein more than alcohols were (Table 2). Therefore, the decrease in volatile levels by proteins is likely due to the hydrophobicity of individual compounds (Macleod *et al.*, 1988; Landy *et al.*, 1995; Kuhn *et al.*, 2006).

In sulfide volatiles, sulfhydryl groups form disulfide bridges with cysteine and cysteine amino acid units in the protein (Mottram *et al.*, 1996). In this current study, sulfides were affected by collagen more than the other proteins. This may be due to the fact that collagen contains more -SH containing amino groups than milk proteins (Gordon and Ziegler, 1955; Steven and Jackson, 1967; Husdan *et al.*, 1977)

Although most of the volatiles decreased with the addition of protein, hexanal and acetaldehyde increased in concentration. The increase was probably caused by the initial volatile levels in the proteins themselves. Most volatile levels were low in 1% whey protein isolate, milk protein isolate, and collagen in deionized water but hexanal was 10-29 ppm and acetaldehyde was 27-31 ppm.

Sucrose

The volatile levels of most aroma compounds significantly decreased as the amount of sucrose increased from 5% to 10% (Table 2). Sucrose at 10% produced a 14-47% decrease (average 32%). For the important tomato compounds, only 3-methylbutanal and 6-methyl-5-hepten-2-one showed no significant decrease from control. Interestingly, there was a strong negative correlation ($R= 0.90$) between the effect of NaCl and sucrose. The volatiles that had the greatest percent increase with NaCl showed the least change with sucrose, and those that had the lowest increase with NaCl had the greatest decrease with sucrose. Both may be due to the effect of the solute on water mobility. In previous studies, contradictory results have been found, where sucrose has been shown to either increase volatiles due to the salting-out effect or to decrease volatile levels due to sucrose-water interactions (Roberts *et al.*, 1996; Covarrubias-Cervantes *et al.*, 2004; Piccone *et al.*, 2012).

Oil

Oil is normally added to tomato sauce. Oil at 10% produced a 10-83% decrease (average 33%). Most of the volatile levels significantly decreased as the amount of oil increased from 0% to 10% and terpenes showed the greatest decrease followed by esters, aldehydes, alcohols, and ketones (Table 2). Oil at 10% changed the order of the OAV values for the important volatiles compared to the rest of treatments. The order of β -ionone, eugenol, 1-penten-3-ol, and 6-methyl-5-hepten-2-one was different from the other treatments. Thus, there might be a slight change in aroma perceived by the consumer. Previous studies have also shown a decrease in volatiles with an increase in oil content (Kaneko *et al.*, 1994; Chung *et al.*, 2003; Ventanas *et*

al., 2010b). The higher fat content in cooked bologna decreased the volatility of some volatiles because fat acts as a solvent for hydrophobic compounds, thus diminishing their release (Ventanas *et al.*, 2010b). Fat also reduced the release of hydrophobic compounds in ice cream (Chung *et al.*, 2003). An increase of fat in salad dressing reduces garlic and pepper flavor intensity due to its viscosity (Guinard *et al.*, 2002). The resistance to mass transfer of volatile compounds in fat and oil is higher than in water, thus the release of fat-soluble flavor compounds is delayed because flavor compounds must be released from the lipid phase to the aqueous phase and then released from the aqueous phase to the headspace; thus the volatility of fat-soluble volatile compounds in high-fat-content foods is lower than in low-fat foods (Jo and Ahn, 1999; Guinard *et al.*, 2002).

Sensory

Sensory evaluation was conducted to determine the effect of the additives on aroma intensity and preference of tomato juice. NaCl (10%), control, and sucrose (10%) produced the highest aroma intensity followed by pectin (1%) and milk protein (1%) while oil (10%) had the lowest aroma intensity. The average rank scores were 4.68a, 4.08a, 3.82ab, 3.18b, 3.10b, 2.14c respectively (means that do not share a letter are significantly different). The order of the aroma intensity was similar to the volatile results from SIFT-MS, with NaCl producing the highest volatile levels (Table 2) and perceived aroma intensity (4.68), and oil producing the lowest (2.14). For aroma preference, the average rank scores for 10% NaCl, control, 10% sucrose, 1% pectin, 1% milk protein, and 10% oil were 4.68a, 4.16a, 4.30a, 3.18b, 2.78b, 1.82c respectively (means that do not share a letter are significantly different). Consumer preference was very similar to aroma intensity, with the highest aroma intensity (10% NaCl) being the most preferred (4.68). Samples that were significantly different from the control were pectin, milk protein, and oil, which were significantly less preferred.

Conclusions

Temperature and food additives affect tomato juice volatile levels. Temperature produced the greatest increase in volatiles among all treatments. The addition of NaCl also increased volatile levels, due to the salting-out effect. This suggests that NaCl should increase the aroma intensity of tomato products, though sensory aroma results were not significantly different from the control. Pectin and pH produced little significant difference in volatile

levels. Proteins decreased volatile levels and aroma intensity due to protein-flavor binding. Thus, the addition of protein or the applications of protein-based fat replacers decreased the volatility of flavor compounds, changing the flavor profile of low-fat tomato products. Sucrose also decreased volatile levels. Oil produced the greatest decrease in volatile levels and perceived aroma among treatments due to the emulsion system affecting mass transfer of volatile compounds.

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