

# Aroma Volatility from Aqueous Sucrose Solutions at Low and Subzero Temperatures

Marco Covarrubias-Cervantes, Dominique Champion, Frédéric Debeaufort,<sup>†</sup> and Andrée Voilley\*

Laboratoire d'Ingénierie Moléculaire et Sensorielle de l'Aliment et des Produits de Santé, ENSBANA, Université de Bourgogne, 1 Esplanade Erasme, 21000 Dijon, France

The gas-liquid partition coefficients of ethyl acetate and ethyl hexanoate have been measured in water and aqueous sucrose solutions from 25 to -10 °C by dynamic headspace. Experiments were carried out on sucrose solutions at temperatures where no ice formation was possible. Results showed that when sucrose concentration increased, aroma volatility increased except for ethyl hexanoate and in the highest sucrose concentration solution (57.5%). A quasi-linear temperature decrease on aroma volatility was observed in sucrose solutions from 25 to around 4 and 0 °C. Then, from 0 to -10 °C, aroma volatility did not decrease: ethyl acetate volatility remained constant but that of ethyl hexanoate increased. Enthalpy of vaporization and activity coefficients of the aroma compounds were calculated.

## KEYWORDS: Aroma; low and subzero temperatures; aqueous sucrose solutions; partition coefficients

## INTRODUCTION

In a general way, flavor release from foods depends on many parameters. For instance, as the serving temperature was increased from -14 to -7.8 °C, vanilla flavor was more marked (1). The same authors observed ice cream also became sweeter as the serving temperature was raised. Sherbets and ice creams of high sugar concentrations or pronounced flavor were liked better when served at temperatures below -12 °C.

Various sensory evaluations on ice creams or frozen desserts have been carried out. Several parameters such as ice cream eating temperature or ice cream ingredients, such as fat and sucrose, have been the object of sensory evaluations.

Moreover, the flavorings used in those sensory studies on ice cream were flavor extracts, which are complex mixtures of aroma compounds. Each of those aroma compounds may interact with the nonvolatile constituents of the matrix. These physicochemical interactions depend on the nature of both the aroma compounds and the nonvolatile components of the matrix. In addition, structural and rheological properties of the matrix influence the retention or release of volatile compounds. Therefore, the study of physicochemical interactions of flavor compounds in simple media definitely could give deeper information on the specific flavor—matrix interactions.

Various observations and works dealing with aroma retention in frozen and freeze-dried food have been carried out. Most of the studies dealt with fruits, vegetables, seafood, meat, and some processed food products. For instance, the evolution of strawberry flavor compounds during frozen storage is the subject of several studies (2-4). The storage of strawberry after 1 month of storage at -18 °C resulted in different trends of behavior depending on the aroma compound nature. On the one hand, a sharp decrease in esters concentration (C < 9 such as methyl 2-methylbutanoate, ethyl butanoate, and butyl acetate) but an increase in nerolidol concentration was observed. On the other hand, frozen storage did not affect key flavor compounds such as mesifurane and Furaneol (2).

Aroma compound behavior in frozen food (in fact, in all foodstuffs) depends on their chemical nature. Their volatility had been reported in terms of flavor compound polarity: the more polar molecules showed an enhancement, whereas the nonpolar ones showed a volatility depression in sucrose solutions (5). Several studies at 25 °C showed that volatility of various polar aroma compounds increased as sucrose concentration increased in the media (6-9). Although some other authors showed that nonpolar compounds such as *d*-limonene, *n*-butylbenzene (10), ionone, naphthalene (11), 2-heptanone, 2-heptanal (6), methyl butanoate, and hexanal (12) decreased in volatility, sucrose concentration increased (up to 60%) in the aqueous solution.

Regardless of the above observations, no explanations of the aroma behavior as a function of temperature have been proposed yet. Indeed, various physical, chemical, and physicochemical phenomena are responsible for the aroma behavior in food at low and subzero temperatures: physicochemical interactions with other food constituents and molecular mobility (mass transfer and diffusion).

The actual state of knowledge on aroma property variations in food at low and subzero temperatures appears in both natural

<sup>\*</sup> Corresponding author (telephone +33 3 80 39 66 59; fax +33 3 80 39 66 11; e-mail voilley@u-bourgogne.fr).

<sup>&</sup>lt;sup>†</sup> Current address: IVT-Dijon, BP 17867, 21078 Dijon Cedex, France

Table 1. Physicochemical Characteristics of Aroma Corr	ipounds
--	---------

	purity	molar	melting	log P <sup>b</sup> at	saturated vapor pressure <sup>c</sup> (Pa) at		
chemical compound	(%)	wt (g/mol)	point <sup>a</sup> (°C)	25 °C	25 °C	−5 °C	−10 °C
ethyl acetate, $C_4H_8O_2$ ethyl hexanoate, $C_8H_{16}O_2$	99.5 99	88.11 144.22	-83.40 -67.50	0.67 2.79	12510 215	2374 20	1723 13

<sup>a</sup> Fenaroli (31). <sup>b</sup> Log P: molecule hydrophobicity expressed by the logarithm of the liquid–liquid partition coefficient between water and 1-octanol. From Rekker (32). <sup>c</sup> Covarrubias-Cervantes et al. (25).

and processed food products. However, to better understand these variations, data on physicochemical interactions and mobility of aroma in food at those temperatures are needed. Moreover, it seems that the structure and physical state of food strongly influence aroma retention and release. Indeed, physicochemical characteristics data at low temperatures complement the aroma release study, especially when physical changes take place in food.

Therefore, the objective of this work was to measure the gasliquid partition coefficients of aroma compounds as a function of temperature (low and subzero) in simple media, as a first approach. Water and aqueous sucrose solutions were chosen to study aroma volatility from 25 to -10 °C.

#### MATERIALS AND METHODS

**Material.** Aroma Compounds. To better understand aroma volatility from aqueous solutions at low and subzero temperatures, two aroma compounds from the same chemical family [ethyl acetate (EA) and ethyl hexanoate (EH)] were selected (**Table 1**). Both aroma compounds were supplied by Aldrich-Sigma Co. with a minimum purity of 98%. Because this work dealt with low and subzero temperatures, the flavor compounds were selected to avoid aroma crystallization at the studied temperatures. Neither of the aroma compounds freezes at -10 °C, which is the lowest temperature of analysis (**Table 1**).

Preparation of Aqueous Solutions. The solutions of analysis were distilled water and aqueous sucrose (Prolabo) solutions at two concentrations: 30 and 57.5% (sucrose weight/solution weight, w/w). Pure sucrose (99%) was dissolved in distilled water at the given concentration and stirred at room temperature using a magnetic stirrer. Sucrose concentrations were chosen to study sucrose concentration effects on aroma volatility at both positive and negative temperatures, without ice crystals. The freezing point of each sucrose solution was previously determined from the water–sucrose state diagram (13). The calculated freezing points of 30 and 57.5% (w/w) sucrose solutions were -3 and -10 °C, respectively. Prior to aroma release determination, a 50 ppm (microliters of aroma/liter of solution) concentration of each aroma compound was added at room temperature in the studied solutions.

**Methods.** Measurement of Gas-Liquid Partition Coefficients of Aroma Compounds. The exponential dilution headspace technique was used to measure the air-liquid partition equilibrium of aroma compounds in both water and aqueous sucrose solutions. This technique was chosen because it reaches aroma equilibrium in less time than a static headspace technique. That was convenient for our study because thermodynamic equilibria at low and subzero temperatures may take long periods to reach the equilibrium state. Another advantage of this technique is that aroma quantification in the liquid phase is not necessary (14, 15).

A given volume of the solution ( $\sim 10 \text{ mL}$ ) was poured into a headspace glass tube to determine the flavor compound volatility. The headspace tube was put into a thermostated bath where temperature (*T*) was controlled ( $T \pm 0.1$  °C) by a Julabo cryobath FH-45 (Seelbach, Germany). Experiments were carried out at both low and subzero temperatures: 25, 10, 4, 1, 0, -5, and -10 °C.

Once the solution achieved the temperature of analysis, a stripping gas (nitrogen) was bubbled through the solution at a constant flow rate of 30 mL min<sup>-1</sup>; 1 mL of the headspace was injected at regular time intervals into a gas chromatograph (GC) until aroma exhaustion. A

Chrompack CP9000 GC with a flame ionization detector (FID) was used in all of the experiments. A Shimadzu C-R6A Chromatopac integrator was coupled to the GC. The column was a packed column of 3 m length and 2.2 mm internal diameter, 10% carbowax 20M, and 100–120 mesh. Analysis conditions were as follows: oven temperature, 100 and 120 °C isotherm; detector temperature, FID, 200 °C; injector temperature, 190 °C; nitrogen (carrier gas) flow rate, 30 mL min<sup>-1</sup>; hydrogen flow rate, 23 mL min<sup>-1</sup>; air flow rate, 232 mL min<sup>-1</sup>; headspace nitrogen (stripping gas) flow rate, 30 mL min<sup>-1</sup>.

The gas-liquid partition coefficients of aroma compounds were calculated by the exponential dilution method (14, 15). Equation 1 allowed us to calculate the molar air-liquid partition coefficient of aroma compound *i* at infinite dilution ( $K_i^{\infty}$ ):

$$K_i^{\infty} = \frac{RTN}{P_{\rm T}D_{\rm N_2}} \frac{1}{t} \times \ln \frac{S_0}{S} \tag{1}$$

*t* is time (s), *R* is the ideal gas constant (8.3142 J mol<sup>-1</sup> K<sup>-1</sup>), *T* is temperature (K),  $D_{N_2}$  is stripping gas flow (m<sup>3</sup> s<sup>-1</sup>), *N* is aroma moles number in the liquid phase,  $P_T$  is total pressure of the system (Pa),  $S_{t_0}$  and  $S_t$  are peak surfaces of the compound *i* in the air phase at time  $t_0$  and *t*, respectively; and  $K_i^{\infty}$  is related to the mass partition coefficient  $K_{\text{mass}}$  by the equation

$$K_i^{\infty} = K_{\text{mass}} \times \frac{M_{\text{air}}}{M_{\text{solvent}}}$$
 (2)

where  $M_{\text{solvent}}$  is the molar mass of the solvent or liquid phase (g mol<sup>-1</sup>) and  $M_{\text{air}}$  is the molar mass of air with moisture (g mol<sup>-1</sup>) obtained from the air moisture diagram (25). From both Dalton's and Henry's laws, the activity coefficient ( $\gamma_i^{\infty}$ ) of a volatile compound *i* at infinite dilution is related to the air-liquid partition coefficient, expressed as the molar fraction, by the equation

$$\gamma_i^{\infty} = K_i^{\infty} \times \frac{P_{\rm T}}{P_i^{s}} \tag{3}$$

where  $P_i^{s}$  is the saturated vapor pressure of compound *i*.

To be sure that no experimental artifacts were the cause of the observed behavior, experiments were repeated from at least three up to five times.

*Statistical Analysis.* Comparison of square means and Duncan's groups of results was carried out by using the statistics software SAS 8.2 (17) with a p < 0.01.

## RESULTS

Aroma Volatility from Water and Sucrose Solutions at 25 °C. Volatilities of EA and EH in water and aqueous sucrose solutions at 30 and 57.5% were measured at 25 °C (Figure 1). The higher the sucrose concentration is, the higher the mass partition coefficient ( $K_{mass}$ ) of EA is. The EH volatility in water is ~3 times higher than that of EA. However, in the 30% sucrose solution, the EH volatility is only twice higher than EA volatility. Finally, in the 57.5% sucrose solution, EA and EH volatilities are not significantly different, 11.60 and 13.63, respectively. It seems that as sucrose concentration increases in the solution, the difference between aroma volatilities is reduced to similar



Figure 1. Volatility of aroma compounds from aqueous sucrose solutions at 25  $^{\circ}$ C.

values. In contrast, EH volatility in the 57.5% sucrose solutions is lower than in the 30% sucrose solutions.

Aroma Volatility from Water and Sucrose Solutions as a Function of Sucrose Concentration. Aroma volatility as a function of sucrose concentration at 4 and 0 °C is shown in Figure 2. When sucrose concentration was increased to 57.5% at 4 and 0 °C, aroma volatility was higher, being more important for EH than for EA. In the 57.5% sucrose solution at 25 °C (Figure 1), the EH partition coefficient drops to a value similar to that of EA. The latter presents no significant difference compared to EH in the 57.5% sucrose solution at 4 and 0 °C.

Aroma Volatility from Water and Aqueous Sucrose Solutions as a function of Temperature. The gas-liquid partition coefficients ( $K_{mass}$ ) of EA and EH were measured in water and 30 and 57.5% sucrose solutions as a function of temperature (Figure 2). All of the gas-liquid partition coefficients were measured in aqueous sucrose solutions without ice crystals. Aroma volatility from water was measured down to 1 °C. The 30% sucrose solution freezing point being -3 °C, experiments were carried out down to 0 °C. In 57.5% sucrose solution, aroma volatility was determined down to -10 °C because the freezing point of this solution is -10 °C.

It was observed that the volatility from water and 30% sucrose solution of both EA and EH decreases with temperature down to 1 and 0 °C, respectively. Values are significantly different (with a p < 0.01) between 25 and 4, 1, or 0 °C, but not between 4, 1, and 0 °C. Although differences between partition coefficients of the two compounds decrease as sucrose concentration increases, the later is true up to values of the same order of magnitude of volatility between aroma compounds, which is achieved in the 57.5% sucrose solution.

The temperature effect on partition coefficients of the two ethyl esters in a 57.5% sucrose solution displays different behaviors depending on the aroma compound: EA shows a decrease in partition coefficient between 25, 10, 4, 0, and -5°C; significant differences (p < 0.01) between values at 25, 10, 4, and 0 °C were obtained. Values at 0 and -5 °C are not significantly different; however, an increase in volatility is observed at -10 °C, with a partition coefficient as high as that obtained at 4 °C.

In the case of EH, partition coefficients also decrease from 25 to 4 °C; then an increase in volatility is observed from 4 to -10 °C. No significant difference (p < 0.01) is obtained for values at 10 and -10 °C.

Activation energies ( $E_a$ ) of the aroma molar partition coefficients in water and 30 and 57.5% sucrose solutions were calculated as a function of temperature (from 25 to 0 °C). An Arrhenius plot [ln  $K_i^{\infty}$  vs the temperature reciprocal (K<sup>-1</sup>)] allows  $E_a$  calculations. In a general way, when the sucrose concentration increases in the aqueous solution, the  $E_a$  increases,

too. In water, an activation energy of  $-52 \text{ kJ mol}^{-1}$  was obtained for EA, which is in agreement with literature data (-47.3 kJ mol<sup>-1</sup>) (*18*). For EH, the  $E_a$  is  $-63 \text{ kJ mol}^{-1}$ .

When sucrose concentration increases in the solution (30% sucrose), the  $E_a$  for both compounds is the same,  $E_a = -46$  kJ mol<sup>-1</sup>. It is observed that the  $E_a$  of each aroma compound increases between 2 and 3 times in the 57.5% sucrose solution with respect to water in the range from 25 to 0 °C, with  $E_a = -28$  and -19 kJ mol<sup>-1</sup> for EA and EH, respectively.

### DISCUSSION

Effect of Sucrose Concentration on Aroma Volatility. The sucrose concentration effect on aroma volatility is shown in Figures 1 and 2. The EA partition coefficient increases with sucrose concentration up to 57.5% whatever the temperature of analysis. This is in agreement with  $E_a$  values observed in the literature (12, 19–21).

In the case of EH, volatility increases with sucrose concentration except at 25 °C, where the volatility was lower in the 57.5% sucrose solution than in the 30% sucrose solution. It is also observed in **Figure 2** that as the sucrose concentration increases, aroma partition coefficient values tend to be of the same order of magnitude as observed for EA. This is true in the range between 1 and 25 °C.

The chemical nature of the aroma compound strongly determines the aroma-solvent interactions (12, 19). Composition and nonvolatile solute concentration in solutions may also influence aroma volatility in different manners. Aroma compound solubility or activity coefficients are physicochemical parameters that give information about interactions between aroma and solvent molecules and/or aroma and substrate molecules (22). Aroma profile modification in sucrose solutions was observed by Voilley and Sauvageot (23). They reported that the odor intensity was lower in a 50% sucrose solution than in water.

The different nature of physicochemical interactions involved in the ternary systems (water, flavor compounds, and sucrose) could explain the different aroma behavior in sucrose solutions.

It is well-known that these interactions could modify the resistance to mass transfer of volatiles, especially if headspace measurements were done under dynamic conditions. This means then that the release is determined not only by the partition coefficient but also by the resistance to mass transfer. The resistance to mass transfer increases with viscosity of the medium and with the volatility of the flavor compounds in the medium. Therefore, it is possible that the resistance to mass transfer has influenced the dynamic headspace of ethyl hexanoate (most volatile compound) over the 57.5% sucrose solution (most viscous medium) more drastically than the other headspace concentrations. This may explain why lower concentrations of EH are released from the 57.5% sucrose solution than from 30% solution. The equal release of EA and EH from the 57.5% sucrose solution at 25 °C suggests that, at this temperature, the release of both compounds is diffusion controlled (24). At lower temperatures, the release of the flavor compounds will be less diffusion controlled because of the lower volatility at lower temperatures.

Effect of Temperature on Aroma Volatility. In Table 1, the volatility of the pure aroma compounds (saturated vapor pressure) is given at 25, -5, and -10 °C. Volatility increases exponentially as temperature increases and as the aliphatic chain carbon number decreases (25). However, the behavior is modified because of the influence of cosolutes or solvent. In the case of both water and the 30% sucrose solution, the



Ethyl hexanoate



Figure 2. Volatility of ethyl acetate and ethyl hexanoate from aqueous sucrose solutions at 0, 30, and 57.5% as a function of temperature.



Figure 3. Volatility,  $K_i^{\circ}$  or  $K_{mol}$  ( $\Box$ ), and solubility ( $\diamond$ ) of ethyl acetate and ethyl hexanoate in a 57.5% aqueous sucrose solution as a function of temperature (solubility values from ref 27).



Figure 4. Activity coefficients ( $\gamma_i^{\circ\circ}$ ) of ethyl acetate and ethyl hexanoate in aqueous sucrose solutions at 0 ( $\triangle$ ), 30 ( $\diamondsuit$ ), and 57.5% ( $\Box$ ) as a function of temperature.

volatility of EA and EH followed the same decrease as temperature decreases to 1 and 0 °C, respectively, near their freezing points.

The aroma volatility increase from water and sucrose solutions with temperature obtained in this work is consistent with results observed by Jouquand et al. (26). They observed an increase of aroma volatility (hexanal, 2-octanone, ethyl butanoate, 2-heptanone, 2-hexanone, 2-hexenal, 2-butanone, and 1-hexanol) from water when the temperature increased from 60 to 80 °C.

Aroma volatility is inversely related to solubility; this is expressed by the well-known Henry's law. Hence, an increase in aroma solubility in the sucrose solutions as a function of temperature would be expected. Therefore, the molar volatility

and solubility (27) of EA and EH in the 57.5% sucrose solution as a function of temperature are compared (**Figure 3**).

In the temperature range from 25 to 4 °C, for both aroma compounds, solubility and volatility decrease with temperature. This behavior was expected and can be described by the Clausius-Clapeyron equation. However, from 4 to -5 and -10°C, different behaviors for each equilibrium are observed. Indeed, for EA a maximum solubility is observed at a temperature near that where volatility is the lowest (near -5 °C). The same trend is observed for EH, although there is a slight shift of the temperature range where the maximum solubility and the lowest volatility are observed. This means that the physicochemical interactions (expressed by the activity coefficient,  $\gamma_i^{\infty}$ ) between the aroma compounds and the solution at this temperature range (around -5 °C) could be very important. To verify this hypothesis, **Figure 4** gives the activity coefficients  $(\gamma_i^{\infty})$  of EA and EH in the aqueous solutions (0, 30, and 57.5%), calculated according to the temperature (eq 3). For aroma compounds in water,  $\gamma_i^{\infty}$  decreases as temperature decreases to 1 °C, which can be interpreted as interactions of the same nature between flavor molecules and water (hydrogen bonds). As the sucrose concentration increases to 30 and 57.5% in the solution,  $\gamma_i^{\infty}$  increased 2 and 5 orders of magnitude with respect to water, respectively. Also, as the temperature decreased, the aroma activity coefficients increased. This shows that aroma compounds establish strong and/or great numbers of interactions not only when the sucrose concentration increases but also when the temperature decreases to -10 °C. This occurs at this temperature and for a 57.5% sucrose solution, where the highest  $\gamma_i^{\infty}$  was observed. The previous results could partially explain the decrease and then the increase of aroma volatility in the sucrose solutions as a function of temperature.

Some authors have suggested (28, 29) or considered (30) the influence of the solution's density to better predict thermodynamic equilibria such as solubility, partition coefficients, and Henry's constants of volatile compounds in aqueous binary systems as a function of temperature. The density of aqueous sucrose solutions increases when temperature decreases. If so, a salting-out effect on aroma would be observed. The latter effect may be opposite (an increase with the temperature decrease) to that predicted by the Clausius–Clapeyron equation. To relate the behavior of a thermodynamic equilibrium, such as partition coefficient, of aroma in water or in an aqueous solution as a function of temperature, density should be taken into account. Further application of such an equation will be adapted and/or applied on aroma volatility from water and sucrose solutions.

Results obtained in this work could be useful to explain sensory observations obtained in frozen food such as ice cream with the aim to better understand the increase of flavor perception at subzero temperatures. To better understand if there is a temperature or density effect on aroma volatility, experiments with other aqueous solutions, such as polyols, should be carried out. The effect of ice or not on aroma volatility should be also studied in the future in order to clarify the effects of density and ice content on aroma release from frozen food.

## ACKNOWLEDGMENT

We are thankful to Rob Vreeker, Mark Malone, and Jeroen Klooster for very helpful discussions.

## LITERATURE CITED

 Guinard, J.-X.; Zoumas-Morse, C.; Mori, L.; Panyam, D.; Kilara, A. Effect of sugar and fat on the acceptability of vanilla ice cream. J. Dairy Sci. 1997, 79, 1922–1927.

- (2) Douillard, C.; Guichard, E. Comparison by multidimensionnal analysis of concentrations of volatile compouds in fourteen frozen strawberry varieties. *Sci. Aliment* **1989**, *9*, 53–76.
- (3) Hakala, M. A.; Lapveteläinen, A. T.; Kallio, H. P. Volatile compounds of selected strawberry varieties analyzed by purgeand-trap headspace GC-MS. J. Agric. Food Chem. 2002, 50, 1133–1142.
- (4) Schieberle, P.; Hofman, T. Evaluation of the character impact odorants in fresh strawberry juice by quantitative measurements and sensory studies on model mixtures. *J. Agric. Food Chem.* **1997**, 45, 227–232.
- (5) Roberts, D. D.; Elmore, J. S.; Langley, K. R.; Bakker, J. Effects of sucrose, guar gum, and carboxymethylcellulose on the release of volatile flavor compounds under dynamic conditions. *J. Agric. Food Chem.* **1996**, *44*, 1321–1326.
- (6) Nawar, W. W. Some variables affecting composition of headspace aroma. J. Agric. Food Chem. 1971, 19, 1057–1059.
- (7) Voilley, A.; Simatos, D.; Loncin, M. Gas-phase concentration of volatiles in equilibrium with a liquid aqueous phase. *Lebensm-Wiss. Technol.* **1977**, *10*, 45–49.
- (8) Voilley, A.; Bosset, J. Nouvelle technique de détermination rapide de la volatilité de composants d'arômes dans des milieux à forte viscosité (New technique of fast determination of the volatility of flavour components in high viscosity media). *Sci. Aliment* **1986**, *5*, 169–174.
- (9) Bredie, W. L. P.; Mottram, D. S.; Birch, G. G. Aroma binding in maltodextrin solutions. In *Trends in Flavour Research*; Maarse, H., Van der Heij, D. G., Eds.; Elsevier Science: Amsterdam, The Netherlands, 1994; pp 139–143.
- (10) Massaldi, H. A.; King, C. J. Simple technique to determine solubilities of sparingly soluble organics: solubility and activity coefficients of *d*-limonene, *n*-butylbenzene, and *n*-hexyl acetate in water and sucrose solutions. *J. Chem. Eng. Data* **1973**, *18*, 393–397.
- (11) De Roos, K. B.; Wolswinkel, K. Non-equilibrium partition model for predicting flavour release in the mouth. In *Trends in Flavour Research*; Maarse, H., Van der Heij, D. G., Eds.; Elsvier Science: Amsterdam, The Netherlands, 1994; pp 15–32.
- (12) Nahon, D. F.; Harrison, M.; Roozen, J. P. Modeling flavor release from aqueous sucrose solutions, using mass transfer and partition coefficients. *J. Agric. Food Chem.* **2000**, *48*, 1278– 1284.
- (13) Blond, G.; Simatos, D.; Catté, M.; Dussap, C. G.; Gros, J. B. Modelling of the water-sucrose state diagram below 0 °C. *Carbohydr. Res.* **1997**, 298, 139–145.
- (14) Richon, D.; Sorrentino, F.; Voilley, A. Infinite dilution activity coefficients by inert gas stripping method: extension to the study of viscous and foaming mixtures. *Ind. Eng. Chem. Process Des. Dev.* **1985**, *24*, 1160–1165.
- (15) Sorrentino, F.; Voilley, A.; Richon, D. Mesure de la volatilité de substances d'arôme à l'aide de deux techniques. *Sci. Aliment* **1984**, *4*, 105–110.
- (16) Bimbenet, J.; Loncin, M. Bases du Génie des Procédés Alimentaires; Masson: Paris, France, 1995.
- (17) SAS Institute. SAS Statistics software, release 8.2; Cary, NC.
- (18) Sanders, R. Compilation of Henry's law constants for inorganic and organic species of potential importance in environmental chemistry (version 3); personal website: http://www. mpch-mainz.mpg.de/~sander/res/henry.html.
- (19) Lubbers, S. G., E. The effects of sugars and pectin on flavour release from a fruit pastille model system. *Food Chem.* 2003, *81*, 269–273.
- (20) Godshall, M. A. Rôle du saccharose dans la rétention des arômes et dans le renforcement de la flaveur des aliments; Mathlouthi, M. R., Ed.; Chapman and Hall: Paris, France, 1995; pp 263– 280.
- (21) Friel, E. N.; Linforth, R. S. T.; Taylor, A. J. An empirical model to predict the headspace concentration of volatile compounds above solutions containing sucrose. *Food Chem.* **2000**, *71*, 309– 317.

- (22) Buttery, R. G.; Bomben, J. L.; Guadagni, D. G.; Ling, L. C. Some considerations of the volatilities of organic flavor compounds in foods. J. Agric. Food Chem. 1971, 19, 1045–1048.
- (23) Voilley, A.; Sauvageot, F. Effet du saccharose sur l'intensité de l'odeur de deux substances d'arôme en solution aqueuse. *Sci. Aliment* **1987**, 7, 59–64.
- (24) De Roos, K. B. Physicochemical models of flavour release from foods. In *Flavour Release*; Roberts, D. D., Taykor, A. J., Eds.; Washington, DC, 2000; pp 126–141.
- (25) Covarrubias-Cervantes, M.; Mokbel, I.; Champion, D.; Jose, J.; Voilley, A. Saturated vapour pressure of aroma compounds at various temperatures. *Food Chem.* **2004**, *85*, 221–229.
- (26) Jouquand, C.; Ducruet, V.; Giampaoli, P. Partition coefficients of aroma compounds in polysaccharide solutions by the phase ratio variation method. *Food Chem.* **2004**, *85*, 467–474.
- (27) Covarrubias-Cervantes, M.; Bongard, S.; Champion, D.; Voilley, A. Temperature effect on solubility of aroma compounds in various aqueous solutions. *Lebensm-Wiss. Technol.* 2004, in press.

- (28) Hu, Y.; Azevedo, E.; Ludecke, D.; Prausnitz, J. Thermodynamics of associated solutions: Henry's constants for nonpolar solutes in water. *Fluid Phase Equilibr.* **1984**, *17*, 303–321.
- (29) Hu, Y.; Ying-Nian, X.; Prausnitz, J. Molecular thermodynamics of gas solubility. *Fluid Phase Equilibr*. **1985**, *23*, 15–40.
- (30) Miklos, G.; Dewulf, J.; Van Langenhove, H. Temperature dependence of Henry's law constant in an extended temperature range. *Chemosphere* **2002**, *48*, 757–762.
- (31) Fenaroli, G. Fenaroli's Handbook of Flavor Ingredients, 2nd ed.; Furia, T. E., Bellanca, N., Eds.; CRC Press: Cleveland, OH, 1975.
- (32) Rekker, R. F.; de Kort, H. M. The hydrophobic fragmental constant; an extension to a 1000 data point set. *Eur. J. Med. Chem.*—*Chim. Ther.* **1979**, *14*, 479–488.

Received for review April 6, 2004. Revised manuscript received August 13, 2004. Accepted August 15, 2004. M.C.-C. is grateful to CONACyT-Mexico for financial support.

JF040171D