

Air–Liquid Partition Coefficients of Aroma Volatiles in Frozen Sugar Solutions

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Static headspace gas chromatography was used to measure air–solution partition coefficients of homologous series of methyl ketones and ethyl esters in aqueous sugar solutions. The measurements were performed in a temperature range from 25 to –25 °C. A rather unexpected temperature dependence of the partition coefficients was observed at subzero temperatures; that is, partition coefficients were found to increase in the temperature interval from 0 to –10 °C. A simple model, based on the freeze concentration effect, is proposed to explain the observed temperature dependence.

KEYWORDS: Air–solution partition coefficient; methyl ketones; ethyl esters; sucrose; fructose; freeze concentration

INTRODUCTION

Despite an extensive literature on ice cream flavor, relatively little information is available on the physicochemical properties of aroma molecules in frozen matrices (1, 2). It is not fully clear, for example, how aroma molecules partition among the different phases of an ice cream matrix (viz., water, fat, and air) and how partition processes are influenced by ice crystal formation or fat crystallization. It is also not clear yet to what extent aroma molecules interact or “bind” to milk proteins or other ingredients in an ice cream matrix (such as polysaccharides or small-molecule surfactants) and how this influences delivery of flavor during consumption. Many studies deal with the effects of flavor–matrix interactions and low molecular weight sugars on aroma release, but experiments are usually performed at positive temperatures and results do not necessarily hold at subzero temperatures (3–6). The purpose of the present study is to improve our understanding of the properties of aroma molecules in frozen matrices. Results will be presented for relatively simple model matrices, namely, aqueous sugar solutions containing fructose or sucrose at various concentrations. Results obtained for more complex systems, more closely related to real ice cream matrices, will be reported separately. The aroma components added to the sugar solutions are model “flavors” selected from a homologous series of methyl ketones and ethyl esters. The compounds were selected on the basis of their physicochemical properties and cover a wide range of volatility and polarity. Static headspace gas chromatography (GC) was used to measure the equilibrium volatile concentration in the headspace above the (partially frozen) sugar solutions in a temperature range from 25 to –25 °C. From the results air–

solution partition coefficients were calculated for the various model flavors used. A simple model, based on the well-known freeze concentration effect, will be proposed to explain the temperature dependence of the partition coefficients.

MATERIALS AND METHODS

Materials and Sample Preparation. Homologous series of methyl ketones (acetone, 2-butanone, ..., 2-nonanone) and ethyl esters (ethyl acetate, ethyl butanoate, ethyl pentanoate) were used in the study. The materials were obtained from Sigma-Aldrich or Acros Organics and were used without further purification. Sucrose and fructose were obtained from Fisher Scientific Ltd. Samples were prepared by adding a mixture of the different methyl ketones and ethyl esters to an aqueous sucrose or fructose solution. Sucrose and fructose levels were 4.9, 9.6, 14.2, and 18.5 wt %. The concentration of each of the volatile compounds in solution was ~50 mg/L, which is well below the aqueous solubility limit at 25 °C. Small portions of solution (3 mL) were transferred to 10 mL glass vials and sealed. The samples were stored at –18 °C in a freezer until further analysis.

Headspace Analysis Using Gas Chromatography. The concentration of volatile compounds in the headspace above the aqueous sugar solutions was measured in the temperature range between –25 and 25 °C. Sample vials were placed in a thermostatic bath and allowed to equilibrate until the concentration of aroma volatiles in the headspace had reached a constant value (~2 days). After equilibration, the temperature in the vials was within 0.5 °C of the desired temperature. Headspace samples (200 µL) were taken from the vials using a gastight syringe and injected into the injector port of a gas chromatograph. The chromatograph was equipped with a Quadrex BTR-CW column (i.d. = 0.53 mm, 30 m, film thickness = 1.0 µm) and a flame ionization detector. Temperatures of the injector and detector were 150 and 250 °C, respectively. The temperature programming for gas chromatographic analysis was 40 °C (5 min), raised at 7 °C/min to 180 °C (0 min). The volatile concentration in the headspace (in milligrams per liter) was determined by comparing the GC peak area with the peak area of standard solutions of methyl ketones and ethyl

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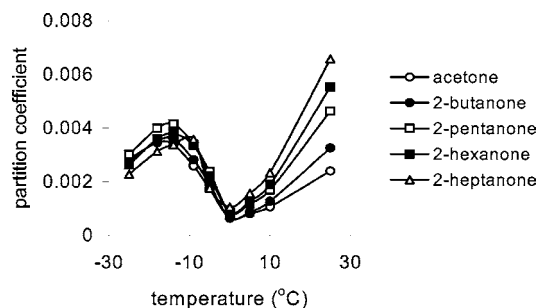


Figure 1. Air–solution partition coefficients of various methyl ketones in a 14.2 wt % sucrose solution as a function of temperature.

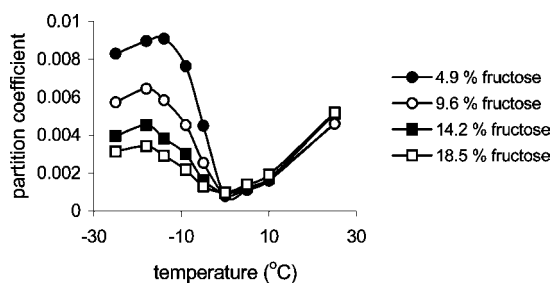


Figure 2. Air–solution partition coefficients of 2-pentanone in aqueous fructose solutions.

esters at 15, 30, 60, 120, and 250 mg/L. All headspace analyses were made in triplicate; relative standard deviation was 5%. Partition coefficients were calculated as $K_i = C_{i,A}/C_{i,L}$, where $C_{i,A}$ and $C_{i,L}$ represent the concentration (in grams per liter) of a volatile component i in air and liquid, respectively.

RESULTS

Air–Solution Partition Coefficients of Methyl Ketones and Ethyl Esters as a Function of Temperature. As an example, **Figure 1** shows results for a series of methyl ketones in a 14.2 wt % sucrose solution. The figure shows a strong dependence of partition coefficient on temperature for all methyl ketones studied. In the temperature range from 25 to 0 °C the partition coefficients decrease with decreasing temperature, indicating a gradual reduction of the concentration of methyl ketones in the headspace above the sugar solution. The behavior becomes more complex at subzero temperatures, where the sugar solutions are (partially) frozen. At these temperatures, partition coefficients first increase with decreasing temperature until a maximum is reached at ca. –10 °C. The partition coefficients then decrease again at still lower temperatures. Similar results were obtained for the series of ethyl esters in a 14.2 wt % sucrose solution (data not shown).

As expected, partition coefficients vary with the number of carbon atoms in the aroma molecule. At a constant positive temperature, partition coefficients of methyl ketones increase in the order acetone, 2-butanone, ..., 2-heptanone. Partition coefficients for the ethyl ester series increase in the order ethyl acetate, ethyl butanoate/ethyl pentanoate. This order is no longer observed at negative temperatures.

Air–Solution Partition Coefficients of Methyl Ketones and Ethyl Esters as a Function of Sugar Concentration. The effect of sugar concentration on the partition coefficients of methyl ketones and ethyl esters was investigated for temperatures in the range between –25 and 25 °C. As an example, **Figure 2** shows results for 2-pentanone in 4.9, 9.6, 14.2, and 18.5 wt % fructose solutions. A small increase of partition coefficient with fructose concentration is observed for temperatures between 0 and 25 °C. The effect of fructose is much more pronounced at

negative temperatures. At these temperatures a significant reduction of partition coefficient is observed upon the addition of fructose. Similar effects have been observed for other methyl ketones and ethyl esters investigated (in both fructose and sucrose solutions).

MODELLING

In this section we aim to develop a simple quantitative model to explain the observed effects of temperature and sugar on aroma partition. The model will be developed in three steps: we first consider the effect of temperature on aroma partition in the absence of ice formation; we then discuss the possible effects of sugar on aroma partition and, finally, the effects of ice formation and freeze concentration are considered. To facilitate the discussion, we start with a brief review of some basic concepts relevant for aroma partition (for a more detailed description the reader is referred to refs 7–10).

Basic Concepts and Definitions. The partitioning of a volatile component i between an aqueous phase and a gas phase (headspace) can be expressed in several ways, one of which is

$$K_i = C_{i,A}/C_{i,L} \quad (1)$$

where K_i is the air–liquid partition coefficient and $C_{i,A}$ and $C_{i,L}$ are the concentrations (in g/L) of the volatile in air and liquid, respectively. Alternatively, equilibrium can be expressed as

$$H_i = P_i/x_i \quad (2)$$

where P_i is the partial pressure of the volatile in the gas phase and x_i is the mole fraction of the volatile in the aqueous phase, all at equilibrium. If the volatile is highly diluted, K_i and H_i are independent of the concentration of the volatile in the aqueous phase and can be regarded as constants; under these conditions H_i can be related to fundamental thermodynamic parameters

$$H_i = \gamma_i^\infty P_i^{\text{sat}} \quad (3)$$

where γ_i^∞ is the activity coefficient (at infinite dilution) of the volatile i in water and P_i^{sat} is the vapor pressure of the pure compound i . The activity coefficient γ_i^∞ is a measure of the “degree of affinity” of i for the aqueous phase. Equations 2 and 3 can be combined to

$$P_i = x_i \gamma_i^\infty P_i^{\text{sat}} \quad (4)$$

Partition coefficient and partial pressure are related by the expression

$$K_i = P_i \times \frac{M_i}{\phi_i P \rho_L V_g} \quad (5)$$

where M_i is the molecular mass of the volatile, ϕ_i is the mass fraction of the volatile in solution, P is the (total) pressure in the system, ρ_L is the density of the aqueous phase, and V_g is the molar volume of the gas.

Effect of Temperature on Aroma Partition in the Absence of Ice Formation. We first consider the situation of a pure (aroma) component i . The vapor pressure of a pure component is related to temperature by the well-known Clausius–Clapeyron equation (11)

$$P_i^0 = P_{\text{ref}}^0 \exp \left[-\frac{|\Delta H_{\text{vap}}|}{R} \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}} \right) \right] \quad (6)$$

where ΔH_{vap} is the enthalpy of vaporization (J/mol), R is the gas constant ($= 8.31 \text{ J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$), and P_{ref}^0 is the vapor pressure at a reference temperature T_{ref} (in K). Equation 6 implicitly assumes that the enthalpy of vaporization is independent of temperature in the temperature range of interest. We now consider the situation of a volatile component i in water and assume that a similar expression holds for the partial pressure of the volatile in the headspace, $P_i(T)$

$$P_i(T) = P_{i,\text{ref}} \exp\left[-\frac{|\Delta H_{\text{hydr}}|}{R}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right] \quad (7)$$

where $P_{i,\text{ref}}$ is the volatile partial pressure at a reference temperature T_{ref} and ΔH_{hydr} represents the enthalpy of hydration (i.e., the amount of energy released when 1 mol of volatiles dissolves from the gas phase into a large amount of water forming an infinite dilute solution). In the derivation of eq 7, ΔH_{hydr} is assumed to be independent of temperature in the temperature range of interest, which is clearly an approximation considering the relatively large temperature variation in our experiments. Equation 7 can be inserted into eq 5 to yield an approximate expression for the temperature dependence of the air–solution partition coefficient

$$K_i(T) \cong K_{i,\text{ref}} \exp\left[-\frac{|\Delta H_{\text{hydr}}|}{R}\left(\frac{1}{T} - \frac{1}{T_{\text{ref}}}\right)\right] \quad (8)$$

where $K_{i,\text{ref}}$ is the partition coefficient at T_{ref} . In the derivation of eq 8 several simplifying assumptions were made; that is, the total pressure in the system, the density of the aqueous phase, and the molar volume of the gas phase were all assumed to be independent of temperature. The latter simplifications are probably not strictly necessary, but eq 8 is considered to be sufficiently accurate for our purposes. When $K_i/K_{i,\text{ref}}$ is plotted versus $1/T - 1/T_{\text{ref}}$ on a semilogarithmic scale, a straight line is predicted with a slope proportional to ΔH_{hydr} .

Effect of Sugar on Aroma Partition. Soluble, nonvolatile components such as sugars can affect the partition of volatiles between an aqueous solution and the headspace. Headspace concentrations may either increase or decrease upon the addition of nonvolatiles. To explain the effect, two mechanisms have to be considered. First, nonvolatile components can affect the activity coefficient γ_i^∞ of the volatile molecules in solution, that is, the affinity of the volatile for the solution. A second mechanism to be considered is simply a consequence of the increase of the mole fraction volatile, x_i , when a nonvolatile is added to the solution (assuming that the mass concentration of the volatile is kept constant). Briefly, imagine a volatile present in pure water and in a sucrose solution at the same mass concentration (mass of volatile divided by mass of solution). Although mass concentrations are equal in both cases, the mole fraction (moles of volatile divided by moles of solvent) will be larger in the sugar solution than in pure water. Provided that γ_i^∞ remains constant in the presence of sugar, eq 4 then predicts a larger amount of volatiles in the headspace above the sugar solution. Voilley et al. (12) used this effect to explain the increase of acetone and 1-octanol concentration in the headspace of aqueous sucrose solutions.

The change of mole fraction volatile in solution, x_i , due to addition of sugar (or any other nonvolatile) is easily calculated. Consider an aqueous solution containing m_i grams of volatile, m_s grams of sugar, and m_w grams of water (total mass $m_t = m_i + m_s + m_w$). We assume that the mass fraction sugar $\phi_s = m_s/m_t$ is varied, whereas the mass fraction volatile $\phi_i = m_i/m_t$ is

kept constant. For the mole fraction volatile, x_i , we may then write

$$x_i = \frac{m_i/M_i}{m_i/M_i + m_s/M_s + m_w/M_w} \quad (9)$$

where M_i , M_s , and M_w represent the molecular weight of the volatile, sugar, and water, respectively. We use the definitions of ϕ_s and ϕ_i and some straightforward calculus to rewrite eq 9 as

$$x_i = x_{i,0} \frac{M_s[M_i + \phi_i(M_w - M_i)]}{M_i M_s + \phi_i(M_s M_w - M_i M_s) + \phi_s(M_i M_w - M_i M_s)} \quad (10)$$

where $x_{i,0}$ is the mole fraction of the volatile in the absence of sugar. When the mass concentration of the volatile is relatively small ($\phi_i \ll 1$), eq 10 simplifies to

$$x_i \cong x_{i,0} \frac{M_s}{M_s + \phi_s(M_w - M_s)} \quad (11)$$

Substitution of eq 11 into eq 4 then yields an expression for the partial pressure of the volatile in the headspace, P_i , in the presence of sugar

$$P_i \cong P_{i,0} \frac{M_s}{M_s + \phi_s(M_w - M_s)} \quad (12)$$

where $P_{i,0}$ is the partial pressure of the volatile in the absence of sugar. It is mentioned that eq 12 is valid, provided that the sugar does not affect the activity coefficient of the volatile. Equation 12 may in turn be substituted into eq 5 to yield an expression for the air–solution partition coefficient in the presence of sugar

$$K_i \cong K_{i,0} \frac{\rho_0 M_s}{\rho_s [M_s + \phi_s(M_w - M_s)]} \quad (13)$$

where $K_{i,0}$ is the partition coefficient in the absence of sugar and ρ_0 and ρ_s represent the density of pure water and the sugar solution, respectively. The density of a fructose or sucrose solution is approximated by the following equation (13):

$$\rho_s \text{ (in kg/L)} \cong 0.2\phi_s^2 + 0.37\phi_s + 1.0 \quad (14)$$

Equation 14 describes the density of a sugar solution at $T = 20^\circ\text{C}$; in our analysis we assume the equation also applies at the other temperatures of interest. It is emphasized that eq 13 only holds when sugar does not affect the activity coefficient of the volatile. The validity of this assumption, obviously, depends on the nature of the volatile. Voilley et al. (12) showed that high concentrations (up to 50%) of sucrose and (other sugars) did not significantly affect the activity coefficients of acetone and 1-octanol in solution; Chandrasekaran and King (14), on the other hand, reported a substantial increase of the activity coefficients of ethyl acetate and several other volatiles in the presence of high levels of fructose.

Influence of Ice Formation and Freeze Concentration. We now consider the problem of aroma partition between an aqueous sugar solution and a headspace at subzero temperatures. At the temperature range of interest ($-25^\circ\text{C} < T < 0^\circ\text{C}$) the aqueous solution is partially frozen, depending on the amount and type of sugar present. Sugar and aroma molecules are excluded from the ice crystals and concentrated in the residual,

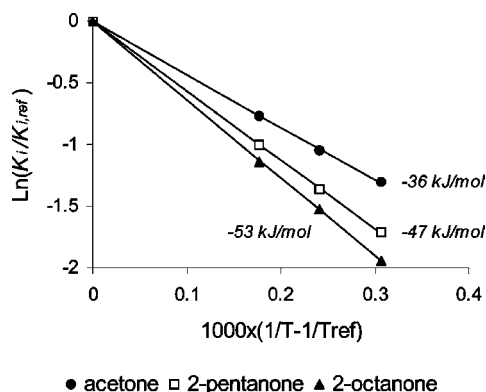


Figure 5. Logarithm of air–water partition coefficient as a function of inverse temperature for acetone, 2-pentanone, and 2-octanone in pure water. Data are plotted relative to the air–water partition coefficient at a reference temperature $T_{ref} = 25\text{ }^{\circ}\text{C}$.

Table 1. Enthalpy of Hydration of Methyl Ketones and Ethyl Esters in Pure Water

compound	ΔH_{hydr} (kJ/mol), this work	ΔH_{hydr} (kJ/mol), literature ^a
acetone	-35.6	-41.5 ± 0.3
2-butanone	-44.4	-45.5 ± 0.5
2-pentanone	-46.5	-49.0 ± 0.5
2-hexanone	-51.7	-52.6 ± 1.0
2-heptanone	-46.5	-56.4 ± 0.6
2-octanone	-52.5	
2-nonanone	-43.5	-62.7 ± 2.0
ethyl acetate	-41.3	
ethyl butanoate	-44.0	
ethyl pentanoate	-48.8	

^a Average values reported by A. V. Plyasunov et al. (17).

of $K_i(T)$. At lower temperatures, the increase in terms I and II levels off and $K_i(T)$ is dominated by the exponentially decreasing term III.

DISCUSSION

We first consider the temperature dependence of the partition coefficients of methyl ketones and ethyl esters at positive temperatures. Under these conditions the variation of the partition coefficient with temperature is described by eq 8. According to this equation the logarithm of the partition coefficient K_i is proportional to inverse temperature. A plot of $\text{Ln } K_i$ versus $1/T$ is thus expected to yield a straight line with a slope proportional to ΔH_{hydr} . In **Figure 5**, we have plotted the logarithm of the partition coefficients of acetone, 2-pentanone, and 2-octanone in (pure) water as a function of inverse temperature. A linear dependence of $\text{Ln } K_i$ on inverse temperature is indeed observed in the temperature range of interest (i.e., from 25 to 0 $^{\circ}\text{C}$). The enthalpy of hydration is determined from the slope of the plots to be $\Delta H_{hydr} = -36\text{ kJ/mol}$ for acetone, -47 kJ/mol for 2-pentanone, and -53 kJ/mol for 2-octanone. Plots of $\text{Ln } K_i$ versus $1/T$ were also made for the other methyl ketones and ethyl esters used in this study. In all cases a linear behavior was observed as predicted by eq 8. **Table 1** summarizes the hydration enthalpies for the various methyl ketones and ethyl esters in pure water. For comparison, the table also shows literature data (17); the latter are seen to be in fair agreement with the results reported here (except for 2-nonanone). Plots of $\text{Ln } K_i$ versus $1/T$ were also made for methyl ketones and ethyl esters dissolved in sucrose and fructose solutions. Just as for the systems without sugar, linear plots were obtained in

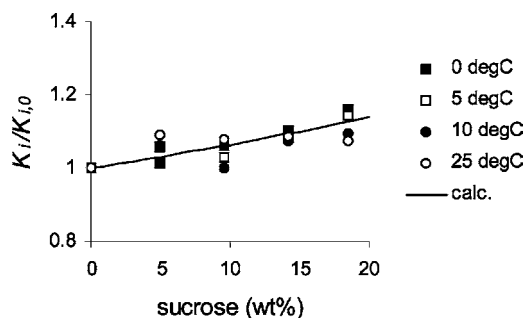


Figure 6. Air–solution partition coefficient of 2-pentanone as a function of sucrose concentration in solution. Data are plotted relative to the partition coefficient of 2-pentanone in pure water (at the relevant temperature). Drawn line is the result of a calculation based on eq 13.

all cases. Hydration enthalpies were again calculated from the slopes of the plots. Sugar was not found to have a significant influence on the hydration enthalpy of methyl ketones and ethyl esters at the sugar concentrations used. Only small variations ($<10\%$) in hydration enthalpy were observed when the sugar concentration was varied between 0 and 18.5 wt %.

Figure 6 shows partition coefficients of 2-pentanone measured at different sucrose concentrations and different temperatures relative to the partition coefficient in pure water. The drawn line in **Figure 6** is the result of a calculation based on eq 13. This equation predicts the variation of the air–solution partition coefficient with sugar concentration, assuming a constant value for the activity coefficient of the volatile. The latter assumption does not seem unreasonable in view of the fact that ΔH_{hydr} is not significantly influenced by the presence of sucrose. The results in **Figure 6** show a fair agreement between the experimental and calculated curves.

We now consider the temperature dependence of partition coefficients at subzero temperature. **Figures 7** and **8** show results of a calculation based on eq 15 together with experimental data for ethyl butanoate and 2-heptanone in different sucrose solutions. The variables used in the calculation are $\Delta H_{hydr} = -44.0\text{ kJ/mol}$ and $K_{i,ref} = 0.012$ for ethyl butanoate and $\Delta H_{hydr} = -46.5\text{ kJ/mol}$ and $K_{i,ref} = 0.006$ for 2-heptanone [ΔH_{hydr} is the heat of hydration in pure water and $K_{i,ref}$ is the air–(pure) water partition coefficient at a reference temperature $T_{ref} = 25\text{ }^{\circ}\text{C}$]. The ice content in the sucrose solution (ϕ_{ice} in eq 15) was taken from the ice–temperature curves in **Figure 3**. The agreement between the calculated and experimental results is satisfactory for the entire temperature range of interest. Equation 15 correctly describes the increase of the experimental air–solution partition coefficient at negative temperature and the occurrence of a maximum at $T \cong -10\text{ }^{\circ}\text{C}$. Note that the increase in partition coefficient at subzero temperature is most pronounced for samples with a low sucrose concentration; this feature is nicely reproduced in the calculations (the pronounced increase is explained from the relatively large freeze concentration effect in low concentration sugar solutions). In view of the large number of assumptions made in the derivation of eq 15, it cannot be excluded that the good agreement between experimental and calculated results is to some extent fortuitous. Indeed, larger deviations between calculated and experimental partition coefficients were observed for some of the other methyl ketones and ethyl esters investigated, in particular for the more hydrophilic molecules such as acetone and 2-butanone. In this case the calculations underestimate the increase of the partition coefficients at negative temperature (experimental partition coefficients are 30–50% higher than calculated values). The discrepancy might be due to the assumption in the calculations

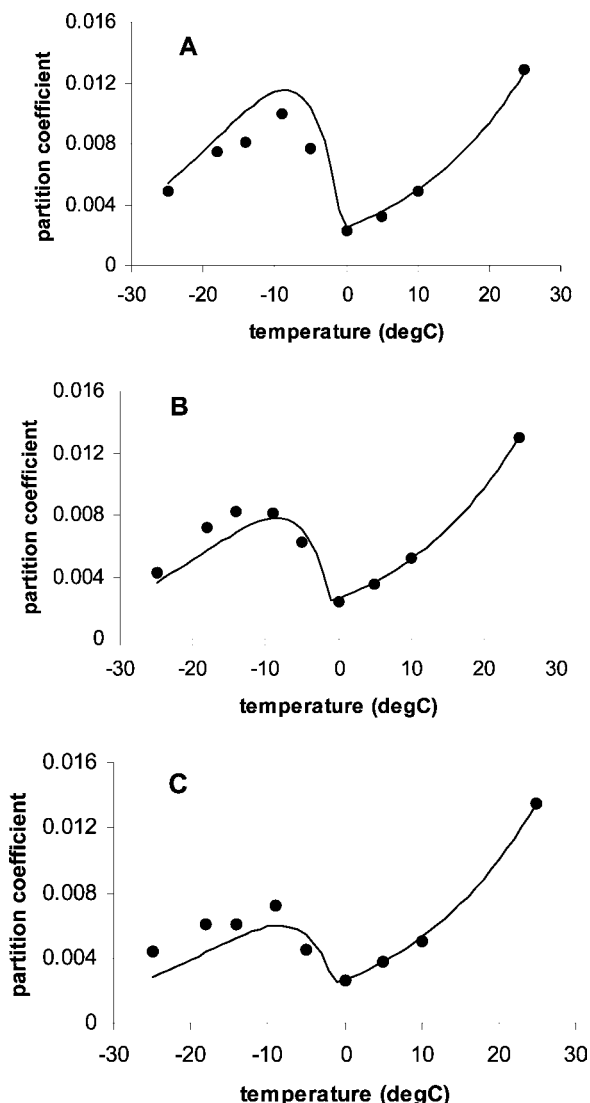


Figure 7. Partition coefficient of ethyl butanoate in aqueous sucrose solutions as a function of temperature (A, 9.6, B, 14.2, and C, 18.5 wt % sucrose). Drawn line is the result of a calculation based on eq 15 (see text for explanation). Variables used in the calculation: $\Delta H_{\text{hydr}} = -44.0$ kJ/mol and $K_{i,\text{ref}} = 0.012$ (at $T_{\text{ref}} = 25$ °C).

that the activity coefficient of aroma molecules is independent of the sugar concentration in solution. This assumption may be reasonable at relatively low sugar concentrations, but becomes questionable at high concentrations (i.e., at the sugar concentrations found in freeze-concentrated solutions). If the activity coefficient in freeze-concentrated sugar solutions is higher than in pure water solutions, this could explain why larger partition coefficients are observed at negative temperatures for some of the aroma compounds. Significant deviations between calculated and experimental results were also observed for 2-nonanone. In this case, calculated partition coefficients (at subzero temperatures) are larger than experimental values. It is speculated that this discrepancy is due to the low solubility of 2-nonanone in aqueous solutions [~ 370 mg/L at 25 °C in pure water (18)]. At low temperature, when freeze concentration takes place, the solubility limit of 2-nonanone may be exceeded. This might then explain why the experimental air–solution partition coefficients are lower than the calculated values.

In summary, air–solution partition coefficients of methyl ketones and ethyl esters in sugar solutions showed a rather unexpected temperature dependence at subzero temperature. The

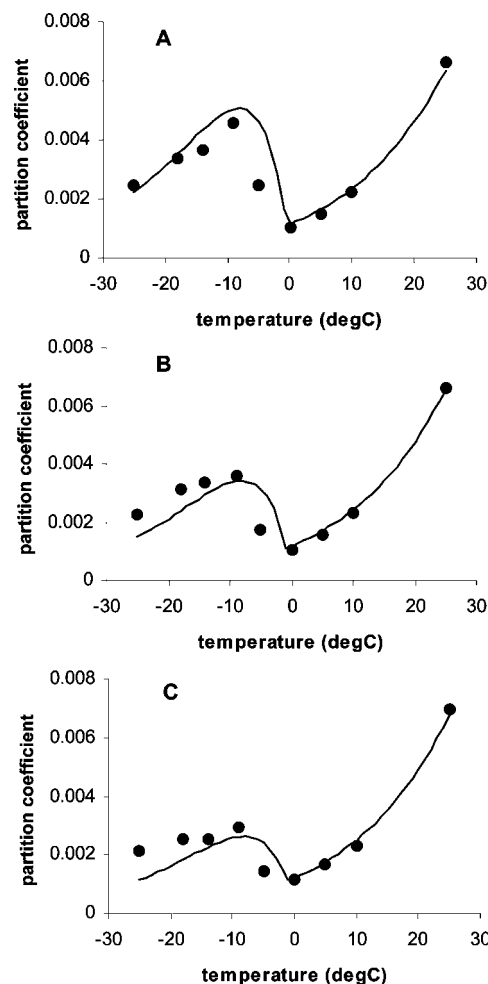


Figure 8. Partition coefficient of 2-heptanone in aqueous sucrose solutions as a function of temperature (A, 9.6, B, 14.2, and C, 18.5 wt % sucrose). Drawn line is the result of a calculation based on eq 15. Variables used in the calculation: $\Delta H_{\text{hydr}} = -46.5$ kJ/mol and $K_{i,\text{ref}} = 0.006$ (at $T_{\text{ref}} = 25$ °C).

temperature dependence was explained in a satisfactory way using a simple model based on the well-known freeze concentration effect. Although quantitative differences between experimental and calculated results were observed in some cases, the main feature (i.e., the increase of the partition coefficient at negative temperature) was reproduced well by the model.

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