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Modeling the Partition of Volatile Aroma Compounds from a Cloud Emulsion

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Parameters determining the partitioning behavior of volatile compounds between a cloud emulsion and the gas phase were measured under static equilibrium headspace conditions, using volatiles (e.g., ethyl hexanoate, cymene, and octanol) representing different volatilities and different degrees of hydrophobicity. The significant factors were the molecular characteristics of the volatile and the concentration of the oil phase. The nature of the lipid (C8 and C12 triglycerides), particle size, and emulsifier type (modified starch and gum arabic) did not significantly alter volatile partitioning. An empirical model based on the partition behavior and physicochemical parameters of 67 volatile compounds was produced. This predicted the partition of volatiles ($R^2 = 0.83$) in cloud emulsions as a function of lipid content. The significant terms ($P < 0.05$) in the empirical model were Log P, Log solubility, the dipole vector, and the oil fraction.

KEYWORDS: **Lipid; emulsifier; modeling; QSPR**

INTRODUCTION

Cloud emulsions are widely used in beverages (e.g., citrus drinks) to give the products an opaque appearance, which is more appealing to the consumer. Although the emulsions have a low oil concentration (2−5 g/L), the addition of a cloud emulsion changes the properties of the beverage phase, thus altering volatile compound partition. As a result, the aroma profile above the product changes and this may affect the overall perceived flavor. Flavorists use a combination of experience and science to adjust flavor formulations to compensate for the type of cloud emulsion. However, it should be possible to predict volatile behavior through scientific principles and the literature contains several relevant papers describing the situation in a range of oil in water emulsions (*1*−*4*).

McNulty and Karel (*5*) predicted the behavior of volatile compounds in oil−water systems on the basis of oil−water−air partitioning and then validated some of the predictions using a homologous series of alcohols. They concluded that the oil fraction (i.e., the relative amount of oil to water) was the key factor influencing flavor release. The viscosity of the oil phase had some role, but large changes in viscosity were required to change release rate by an order of magnitude. Similarly, the solid fat index had some effect on release. The effect of surfactants was limited to a change in oil−water partition and not to any interfacial effect. Further experimental studies on oil−water systems were carried out by Landy (*6*) who determined the effects of emulsions on volatiles both under static equilibrium and under dynamic dilution conditions. Similar experiments have also been reported from our laboratory (*7*).

The model proposed by Hills and Harrison (*2*) was based on mass transfer theory. Experimental evidence using heptan-2-one validated their model in that resistance to mass transfer at the interface was the rate-limiting step. They too predicted that oil fraction was an important factor but also predicted that oil droplet size would affect the mass transfer coefficient and therefore the rate of release.

The models described above were validated with a limited number (and range) of volatile compounds. With the advent of rapid methods for following flavor release (*8*), it is now possible to obtain more experimental data about flavor release. To obtain a robust model, it is particularly important to carry out validation experiments with a wide range of volatile compounds. These should cover the full range of chemicals found in food aroma (pyrazines, esters, aldehydes, and ketones) as well as the full range of physicochemical parameters (volatility, solubility, and polarity).

The initial aim of this paper was to test the emulsion hypotheses in the literature using an emulsion with a low oil fraction. This system restricts manipulation of the oil fraction but allows the effect of emulsifier type and amount to be observed. The effect of the emulsifier:lipid ratio can also be tested at these oil fractions. For instance, a 0.2% lipid emulsion can easily carry 0.4% emulsifier, whereas at higher levels, the solubility of the emulsifier becomes the problem. Landy et al. (*6*) pointed out that if a compound had a high affinity for the oil phase, its concentration in the aqueous phase would be so

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low that any interfacial binding would be insignificant. In the experimental protocol, it is therefore useful to have a range of volatile concentrations and oil:emulsifier ratios to overcome this potential problem. The effects of different lipid types and the interfacial area were also tested. The published models in the literature require the calculation of some fundamental parameters such as mass transfer at the interface or release rates. An alternative way of modeling the process was the second aim of this paper. Quantitative structure property relationships (QSPR) are an empirical way of describing observed behavior through physicochemical properties of the molecules involved (*9−11*). The technique has proved successful in other flavor applications (*12*, *13*).

MATERIALS AND METHODS

Chemicals. Tricaprylin (C8 triglyceride), ethyl butyrate, ethyl hexanoate, ethyl octanoate, decanal, decanol, anethole, ethyl decanoate, isoamylbutyrate, terpinolene, alpha damascenone, ethyl undecanoate, nonanone, menthofuran, octanal, menthone, cymene, octanone, ethyl methyl furan, carvone, linalool, hexyl acetate, octanol, isoamyl acetate, ethyl pentanoate, cyclohexanone, ethyl-2-methylbutyrate, benzaldehyde, dimethylpyrazine, butanone, (*E*)-2-hexenal, hexanol, pyrazine, 2,3-diethylpyrazine, 3-ethyl- 2-methylpyrazine, methyl furan, 2-methylbutanol, and ethyl lactate were obtained from Sigma-Aldrich (Poole, U.K.). Limonene was obtained from ACROS Organics (Loughborough, U.K.). Gum arabic, citric acid, and potassium sorbate were obtained from Fischer Scientific (Loughborough, U.K.). Modified starch was obtained from Procter and Gamble (Cincinnati, U.S.A.). Trilaurin (C12 triglyceride) was obtained from Larodan Fine Chemicals, AB (Malmo, Sweden). A cloud emulsion consisting of hydrogenated coconut oil and gum arabic was obtained from Firmenich SA (Geneva, Switzerland).

Sample Preparation. Solutions of volatiles were individually prepared at various volatile concentrations according to their solubility limit. The volatiles were dispersed in 1 L of distilled water and shaken using an SF1 flask shaker (Stuart Scientific, Redhill, U.K.) for 1h. Cloud emulsions were then added at various emulsion concentrations to 100 mL aliquots of the volatile solutions at 0−5 g/kg of lipid, dependent upon the experimental requirements. The final volatile concentrations were in the range of 5−15 ppm. All samples were shaken on the flask shaker for 10 min and left to equilibrate overnight (room temperature).

Emulsion Preparation. The emulsifying agent, either gum arabic (100 g) or modified starch (50, 100, or 200 g/kg), citric acid (10 g/kg), and potassium sorbate (25 ppm) were dispersed in water using a high shear blender (Silverson Machines Ltd., Chesham, U.K.) for 10 min before the addition of the oil phase, either hydrogenated coconut oil or synthetic triglycerides (C8 and C12; 100 g/kg). The mixture was blended for a further 25 min. An aliquot of this crude emulsion was passed through a homogenizer (APV, Crawley, U.K.) three times at 4500 psi. All emulsions were stored at 4 °C until required.

Droplet Size Distribution. The droplet size distribution of the model emulsions was determined using the Malvern Mastersizer S system (Malvern Instruments, U.K.). A 300RF lens was used to cover a particle size range of 0.05−880 μm, using a presentation grid code 3NAD (which relates to the predicted scattering pattern). The presentation grid code corresponded to the type of Mastersizer used, the relative particle refractive index (real), the relative refractive index (imaginary), and the dispersant refractive index. A polydispersed analysis was used on the emulsion sample. The obscuration value, i.e., the amount of laser light lost due to the sample, was set to around 15−20%, which lies within the ideal range, and produced a volume concentration of about 0.0020%. Three separate samples of each emulsion were analyzed in duplicate. The interfacial surface area distribution was estimated from the average particle size radius using the equation defining the surface area of a sphere (eq 1), the assumption being that the emulsion droplets were wholly spherical in their shape.

$$\text{surface area of a sphere} = 4\pi r^2 \tag{1}$$

where r is the radius of the emulsion droplet.

The mean interfacial surface area for the emulsions following high shear mixing was 5355 cm^2/g. This was increased to 30 000 cm^2/g after five passes through the homogenizer.

Equilibrium Headspace Measurements Using Atmospheric Pressure Ionization−Gas Phase Analyzer (API−GPA). The emulsion sample (100 mL) was placed in a glass bottle (total volume 123 mL), sealed, and then equilibrated at room temperature overnight. After the sample was equilibrated, the bottle was connected to the API sampling tube. The API−GPA (*8*) was operated in a positive ion mode with a source temperature of 75 °C and a corona pin voltage of 4 kV. Compounds were measured as their $[M + H]^+$ ion, with ion intensities monitored in single ion mode, and at a cone voltage of 18 V. With a headspace of 25 mL and a sampling rate of 5−6 mL/min, dilution of the headspace was minimized. The API−MS ion trace (for all volatile compounds tested) increased rapidly to a plateau value, which remained constant during the sampling period and was taken as the equilibrium headspace concentration. Data were expressed as the relative headspace intensity (RHI; %) calculated as

$$\frac{\text{volatile headspace intensity above the emulsion sample} \times 100}{\text{volatile headspace intensity above the aqueous control}} \tag{2}$$

Model Development. A database of physicochemical parameters relating to the 39 volatiles was produced using the chemical modeling program CAChe (CAChe 3.1, Oxford Molecular, Oxford, U.K.). The terms and values derived by CAChe were correlated with the experimental data to find the most important factors influencing volatile−emulsion interactions. This was performed using partial least squares regression analysis in Guideline+ (Camo ASA, Oslo, Norway). The significant terms identified by Guideline+ were transferred into Design Expert (Design Expert 5, Minneapolis, U.S.A.) and modeled using a multilinear regression (MLR) method.

RESULTS AND DISCUSSION

Data Acquisition and Analysis. There are several experimental problems to consider when measuring the headspace concentration of volatile compounds (*14*). The major problem is avoiding dilution of the headspace by incoming air when sampling headspace into the analyzer. The experimental procedure used direct sampling into the API−GPA source to overcome these problems. The results obtained (an almost instantaneous increase to a plateau value) suggested that the approach was successful. Because the purpose of this study was to determine the effects of different emulsion compositions on the partition of volatile compounds between the liquid and the gas phases, all data were expressed relative to water using eq 2 to calculate a RHI value. To aid interpretation, a value of 100% indicated the same behavior as in water, values <100 indicated that volatile compounds were retained by the emulsion, and values >100 indicated that there was some “salting out” effect from the aqueous phase to the gas phase.

Effect of Lipid Concentration. The static equilibrium headspace above three volatiles (ethyl butyrate, octanone, and ethyl octanoate) in cloud emulsions at various lipid concentrations (0−2 g/kg) showed differences in volatile partitioning. The partition of the aroma compounds was reduced to different extents by the addition of the emulsion as shown by the RHI values (**Figure 1**). The ethyl butyrate partition was little changed by the addition of emulsion (RHI ~100%) whereas the other compounds showed decreased RHI values. The volatile headspace concentration above emulsions containing octanone did not decrease noticeably until the lipid concentration in the emulsion was greater than 0.5 g/kg. After this, octanone steadily decreased in RHI value to 25 and then 50% at 1 and 2 g/kg of lipid, respectively. The RHI value for ethyl octanoate, however, decreased to approximately 80% in the emulsions containing 0.25 g/kg of lipid, the lowest lipid concentration analyzed.

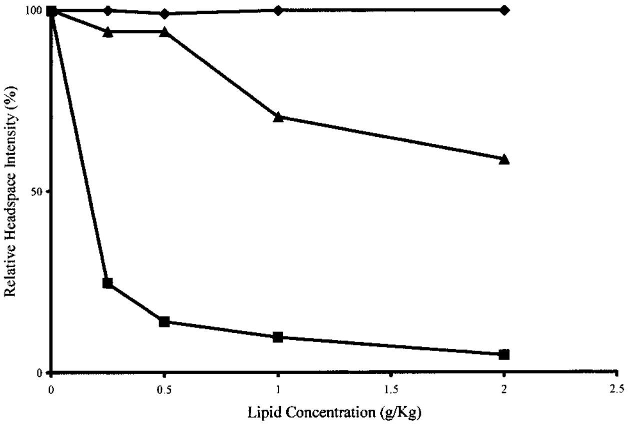

Figure 1. Effect of lipid concentration on the RHI of ethyl butyrate (◆), octanone (▲), and ethyl octanoate (■) in a hydrogenated coconut oil oil–water emulsion, emulsified with gum arabic. Each value is based on two replicates.

Further increases in lipid concentration to 2 g/kg reduced the RHI of ethyl octanoate by a further 10%. Hydrophobicity is an obvious parameter to explain the behavior of the volatiles in the presence of emulsions (*15*). The log *P* values for the three compounds were calculated using CAChe software (ethyl butyrate 1.2, octanone 2.6, and ethyl octanoate 2.8). Although octanone and ethyl octanoate have similar log *P* values, their behavior in the presence of cloud emulsion is very different (**Figure 1**). This suggests that log *P* alone provides a crude estimate of partition behavior in the presence of emulsions.

The effect of the emulsion oil fraction on volatile behavior has been studied previously (*16*−*18*). All studies showed greater retention of hydrophobic compounds with increasing lipid content, which agrees with our findings. Most studies on emulsions compared the effect of lipid concentration on volatile behavior at very different oil fractions and/or relatively high lipid concentrations. From our data, it is clear that small changes in the quantity of lipid at very low lipid concentrations can significantly affect the partition of aroma compounds.

Effect of Emulsifier Type. It is possible that emulsifiers interact with volatile compounds either through a binding effect (many aroma compounds are amphoteric) or by changing the mass transport properties of the liquid interfacial boundary layer. Binding can be measured with the static equilibrium headspace analysis whereas the mass transport effect will only be seen in dynamic release experiments. Two common emulsifiers with different functionalities were used on the premise that they might alter the binding capacity of the cloud emulsions, which would result in a changed volatile headspace concentration above the emulsions.

Three volatiles with different affinities for the lipid phase were chosen. Cymene is a highly nonpolar, lipophilic volatile, which might be expected to preferentially inhabit the oil phase. Ethyl hexanoate and octanol are moderately lipophilic compounds containing polar and nonpolar regions, giving them an amphiphilic nature. As such, they may interact with the lipid phase and the oil−water interface, possibly acting as cosurfactants (*19*).

The results in **Figure 2** showed that there was no significant difference ($P < 0.05$) in volatile partitioning between cloud emulsions emulsified with gum arabic and cloud emulsions emulsified with modified starch, at either lipid concentration. The reduction in volatile headspace, caused by the emulsion, ranged from 30 (octanol dissolved in 2 g/kg of emulsion) to 96% (cymene dissolved in 5 g/kg of emulsion). These differences in volatile partitioning produced a range of volatile concentrations in the aqueous phase, which according to the work of Landy et al. (*6*), should have allowed us to determine any significant emulsifier−volatile interactions.

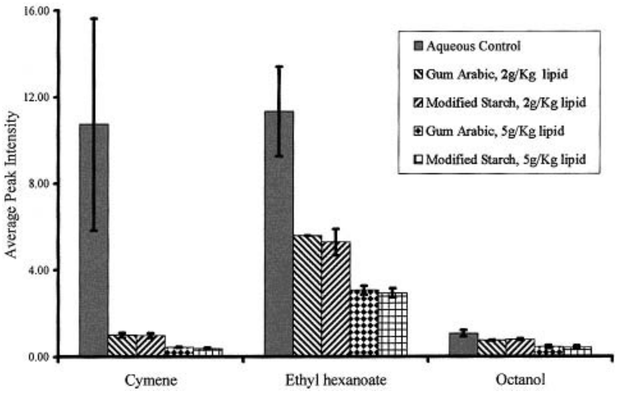

Figure 2. Effect of emulsifier type on the RHI of three volatiles. Each value is based on three replicates (± standard deviation).

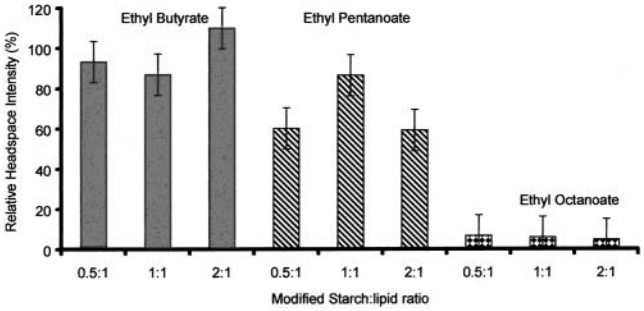

Figure 3. Effect of the concentration of modified starch in a cloud emulsion on the RHI of three esters. Each value is based on three replicates (± standard deviation).

Effect of Emulsifier Concentration. To investigate the effect of emulsifier concentration, volatile behavior was analyzed above emulsions with various amounts of emulsifier present (2.5, 5, and 10 g/kg), while keeping the lipid concentration constant (5 g/kg). Three esters (ethyl butyrate, ethyl pentanoate, and ethyl octanoate) were used as volatile probes in this experiment as the increase in chain length would increase compound hydrophobicity and produce emulsions with different amounts of volatile in the aqueous phase, making it easier to observe any emulsifier−volatile interactions. However, the results showed that there were no significant differences in volatile behavior ($P < 0.05$) at any emulsifier:lipid ratio (**Figure 3**).

The results suggest that regardless of the nature of the volatile and its concentration in the dispersed and continuous phase, neither the emulsifier type nor the concentration of emulsifier at the oil−water interface (**Figure 3**) significantly affected equilibrium volatile partitioning in cloud emulsions. Landy and co-workers (*6*) obtained different results but using a dynamic headspace dilution technique in which both partition and mass transport are important factors. They also used different emulsifiers (sodium caseinate and sucrose stearate). Direct comparisons of the two sets of data are therefore not valid.

Effect of Lipid Type. Lipid type may also affect the partition of volatile compounds between an emulsion and the air phase (*5*, *20*). To investigate this hypothesis, the behavior of three volatiles (octanol, cymene, and ethyl hexanoate) was studied in cloud emulsions containing either tricaprylin (C8:0 triglyceride) or trilaurin (C12:0 triglyceride), emulsified with gum arabic. The triglycerides were chosen due to the differences in

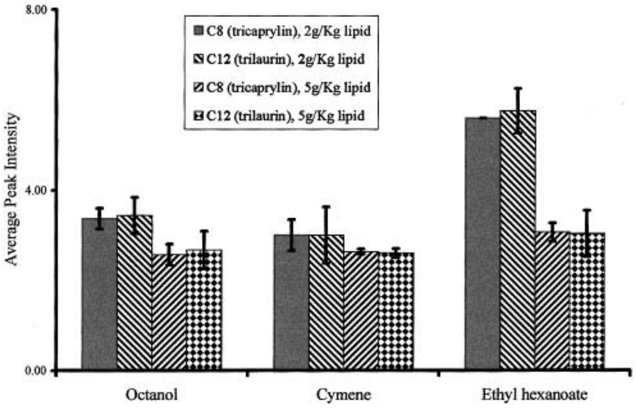

Figure 4. Effect of lipid type on the RHI of three volatiles. Each value is based on three replicates (± standard deviation).

their chain length and physical state at ambient temperatures; tricaprylin is an oil at room temperature (mp 4 °C), whereas trilaurin is a solid (mp 47 °C). It was hypothesized that emulsions made with pure tricaprylin would produce amorphous, liquid emulsion droplets, whereas those made with pure trilaurin might contain solid droplets. The solid regions have the potential to crystallize and may not absorb volatiles as readily (*21*), thereby altering the partitioning of a volatile into the emulsion phase. This should increase the volatile headspace concentration above such an emulsion relative to an emulsion containing a purely amorphous oil phase. However, the results did not support this idea, as the data showed no significant effect ($P < 0.05$) on volatile behavior when changing the nature of the lipid phase (**Figure 4**). Therefore, using liquid or solid triglyceride as the oil phase in the production of emulsions may not significantly affect volatile partitioning.

Effect of Emulsion Droplet Size. The effect of particle size in a cloud emulsion was studied to observe the effect of interfacial surface area on volatile partitioning with the hypothesis that the greater the surface area, the greater the potential binding to the oil–water interface. Emulsions with very different droplet size profiles were prepared using different emulsification processes. Following high shear mixing, the mean interfacial surface area of the emulsion droplets was 5355 cm^2/g. Further homogenization increased this to an average interfacial surface area of 30 000 cm^2/g.

The data showed that there was no significant effect ($P < 0.05$) of particle size on equilibrium volatile partitioning (**Figure 5**). This may be because the volatiles did not interact with the interfacial region and hence were not affected by changes in it. These results are consistent with the work of Landy et al. (*6*) who also found no effect of the interfacial surface area on the volatility of ethyl esters in lipid-containing systems. They suggested that the low quantities of protein at the interface for the sodium caseinate samples meant that not enough volatile could bind to the interface to show a significant effect of altering interfacial surface area. However, it could also be argued that reducing the amount of emulsifier present in the system would reduce the amount of free emulsifier, which could potentially interact with aroma compounds. Producing a lipid-containing system with a low quantity of emulsifier bound at the oil–water interface would therefore provide a clearer insight into the effect of interfacial surface area on volatile binding. As our samples contained very low amounts of emulsifier (0.5–2 g/kg), it can be suggested that volatiles, independent of functionality, do not significantly interact with the oil–water interface.

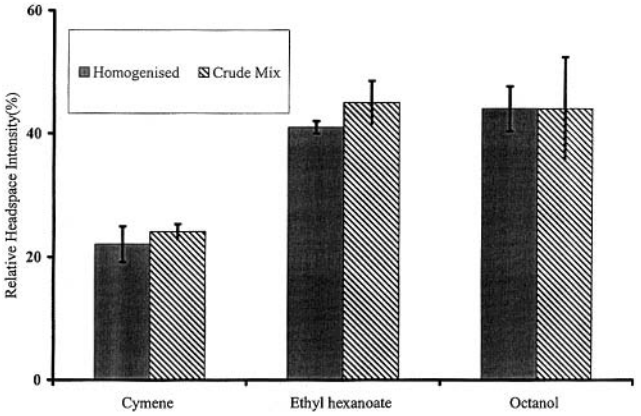

Figure 5. Effect of droplet size distribution on the RHI of three volatiles (0.5 g/kg of lipid and emulsifier in the cymene sample, 2 g/kg of lipid and emulsifier in the remaining volatile samples). Each value is based on three replicates (± standard deviations).

Modeling Volatile Behavior in a Cloud Emulsion. From the results above, the only significant factors affecting equilibrium partitioning in this cloud emulsion were the nature of the volatile and the lipid concentration. This type of behavior is ideal for modeling using the QSPR approach, which relates partition coefficient to the physical and chemical properties of the volatile compounds. Partition behavior was measured for 39 compounds in 0, 1, and 2 g/kg cloud emulsions, and data were expressed as RHI (eq 2) values. For each volatile compound, 72 different physicochemical terms were calculated using CAChe, a computer package that estimates physicochemical and topological parameters using the structure and geometry of each molecule based on quantum mechanics (*9*). From the 72 terms, those that were significant in describing volatile partition in the model system were identified using partial least squares regression. These significant descriptors were then built into a model to describe the effect of lipid content on partitioning, using a MLR algorithm to yield eq 3.

$$\begin{gathered}\text{lipid effect} = \\ +107 \\ -6.3 \times (\text{Log } P)^2 \\ -3.2 \times \text{Log solubility} \\ +0.28 \times (\text{dipole vector})^2 \\ +10 \times \text{lipid concentration (g/kg)} \\ +0.39 \times (\text{Log } P)^4 \\ -2 \times (\text{Log } P)^2 \times \text{lipid concentration (g/kg)} \\ +7.6 \times \text{Log solubility} \times \text{lipid concentration (g/kg)} \\ -0.93 \times (\text{dipole vector})^2 \times \text{lipid concentration (g/kg)} \quad (3)\end{gathered}$$

A quadratic model provided the best fit to the data using analysis of variance analysis. The statistically significant descriptors ($P < 0.05$) were $(\text{Log } P)^2$, Log solubility, $(\text{dipole vector})^2$, and the lipid concentration. The correlation between the experimental data set and the results estimated by the model was good with a correlation coefficient (R^2) of 0.83. The value of the predictive R^2 was 0.8, indicating that the model had good predictive capabilities. The fact that the predictive R^2 did not deviate substantially from the correlation coefficient R^2 showed

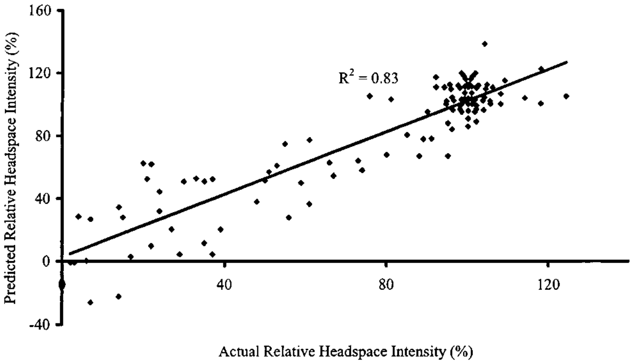

Figure 6. Experimental vs predicted model values for the behavior of 39 different volatiles in cloud emulsions containing 0, 1, and 2 g/kg of lipid.

that the terms used in the model were valid and capable of describing the current data set.

The predictive capacity of the model was evaluated by predicting the behavior of a further 28 volatiles, whose values were not included in the preliminary model, under the same experimental conditions. The volatiles used in the test set were chosen to reflect the distribution of relative headspace intensities shown in the initial model data set. The model predicted the behavior of the 28 volatiles with an R^2 of 0.83, which confirmed the robustness and predictive capability of the model. **Figure 6** shows how well the model predicted the effect of lipid in cloud emulsions on the RHI of volatiles.

Although the descriptors in the model were used together to predict the behavior of volatiles in a cloud emulsion, they were not of equal importance. The lipid concentration and the terms associated with Log *P* were the most influential factors describing volatile partitioning in a cloud emulsion. Log solubility and the dipole vector were not as influential in the model equation but were still significant terms as corrective descriptors in the model. Log solubility is based on terms such as solvent accessibility surface area and is an estimate of a solute's ability to dissolve in water (*22*). The model revealed that a volatile, which possessed a low Log solubility value, such as ethyl undecanoate, would partition into the lipid phase to a greater extent than those with a higher Log solubility value, such as pyrazine. The dipole vector correlates with a volatile's polarity and allows the determination of a volatile's suitability for solvation and how it orients itself within a solvent. The dipole vector is also related to polarizability, which is a dispersive force related to vapor pressure (*23*). Volatiles with a high (dipole vector)^2 value, such as decanal, showed greater interaction with the emulsion than those with a low value, such as dimethyl pyrazine.

In the literature, few attempts have been made to correlate physicochemical parameters to volatile behavior in emulsions. However, recent efforts have been made to correlate Log *P*, and other hydrophobic estimators, with volatile behavior in lipid-containing systems with various degrees of success (*24*, *25*). Pirapez and co-workers showed that the increased retention of aroma compounds in fresh cheese samples with higher triolein concentrations was strongly influenced by the structure of the volatile. They found that a capacity factor (pertaining to reversed-phase high-pressure liquid chromatography) and Log *P* successfully described the relationship between volatile structure and retention in triolein samples. Roberts et al. (*25*) studied the behavior of volatiles in oil and water matrixes using a retronasal simulator to mimic aroma behavior during consumption. This dynamic technique showed a general trend between Log *P* and flavor compound volatility in oil and water matrixes. Compounds with a high Log *P* were more volatile in water than oil. However, this term did not predict adequately the relative volatility of flavor compounds within oil and water solutions under dynamic conditions. This may be due to the fact that the predictive capacity of log *P* was based on a data set of eight compounds. The limited variation in molecular functionalities may have been inadequate to fully assess this term's capabilities, as compared to the 39 different compounds used to produce our predictive model. Furthermore, their correlation of Log *P* with volatility rate constants was based on linear regression analysis. Our model shows that the interaction between Log *P* and the oil fraction was not linear (**Figure 7**), due to the quadratic Log *P* term in eq 2. Volatile compounds showed a greater response to changes in lipid concentration as (Log *P*)^2 increased (i.e., compounds become less hydrophilic and more hydrophobic) up until a certain point, whereupon this trend was reversed. This might be explained by steric hindrances as volatiles that possess a higher (Log *P*)^2 value are typically larger and more bulky molecules. However, although various steric parameters were included in the original 72 physicochemical descriptors, none of them was significant in the model describing partition behavior, suggesting that was not the origin of the effect.

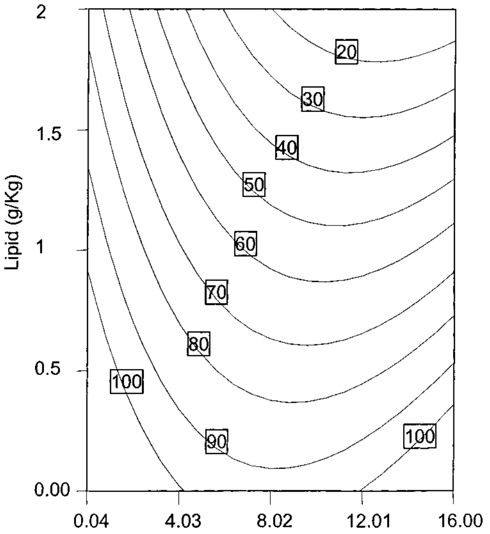

Figure 7. Contour plot showing the RHI values at various lipid concentrations against (Log *P*)^2. Log solubility and the (dipole vector)^2 were constant values of –3.19 and 7.88, respectively.

The work of Agatonovic-Kustrin et al. (*26*) provided a model describing the transfer of drug compounds into human milk using QSAR, and it correlates very well with our model. One of the physicochemical parameters found to be highly significant in their model was Log *P*, which is in strong agreement with our findings. The solvent accessibility surface area, related to Log solubility, was also important, as were functionality terms, such as amino, hydroxy, nitro, and methyl counts, which would exert an influence on the dipole vector term shown in our model. Their model also included numerous other descriptors, including a steric energy term and a structural flexibility term, which may be required to adequately describe the behavior of large molecules with high (Log *P*)^2 values.

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