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Enhancing expected food intake behaviour, hedonics and sensory characteristics of oil-in-water emulsion systems through microstructural properties, oil droplet size and flavour


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Abstract

Food reformulation, either to reduce nutrient content or to enhance satiety, can negatively impact upon sensory characteristics and hedonic appeal, whilst altering satiety expectations. Within numerous food systems, perception of certain sensory attributes, known as satiety-relevant sensory cues, have been shown to play a role in food intake behaviour. Emulsions are a common food structure; their very nature encourages reformulation through structural design approaches. Manipulation of emulsion design has been shown to change perceptions of certain sensory attributes and hedonic appeal, but the role of emulsions in food intake behaviour is less clear. With previous research yet to identify emulsion designs which promote attributes that act as satiety-relevant sensory cues within emulsion based foods, this paper investigates the effect of oil droplet size (d_{4,3}: 0.2 - 50 µm) and flavour type (Vanilla, Cream and No flavour) on sensory perception, hedonics and expected food intake behaviour. By identifying these attributes, this approach will allow the use of emulsion design approaches to promote the sensory characteristics that act as satiety-relevant sensory cues and/or are related to hedonic appeal. Male participants (n =24) assessed the emulsions. Oil droplet size resulted in significant differences (P <0.05) in ratings of Vanilla and Cream flavour intensity, Thickness, Smoothness, Creamy Mouthfeel, Creaminess, Liking, Expected Filling and Expected Hunger in 1 hour’s time. Flavour type resulted in significant differences (P <0.05) in ratings of Vanilla and Cream flavour intensity, Sweetness and Liking. The most substantial finding was that by decreasing oil droplet size, Creaminess perception significantly increased. This significantly increases hedonic appeal, in addition to increasing ratings of Expected Filling and decreased Expected Hunger in 1 hour’s time, independently of energy content. If this finding is related to actual eating behaviour, a key target attribute will have been identified which can be manipulated through an emulsions droplet size, allowing the design of hedonically appropriate satiating foods.

Keywords: Emulsions, Microstructure, Sensory perception, Flavour, Expected satiety, Creaminess

1. Introduction

With the increasing prevalence of global obesity and its related non-communicable diseases, new strategies to promote weight loss and reduce the risk of weight gain are urgently needed. The food industry is increasingly being encouraged to contribute to the alleviation of the obesity burden through product reformulation and the development of the next generation of foods (Norton, Moore and Fryer, 2007). One approach involves increasing the satiating power of foods and
beverages, reducing consumption quantity, and thus energy intake (Blundell, 2010; van Kleef et al., 2012).

Prandial experienced sensory characteristics have been shown to impact upon consumption (de Graaf, 2012). Even subtle differences in sensory characteristics have an impact on eating behaviour (de Graaf and Kok, 2010; McCrickerd et al., 2012; Yeomans and Chambers, 2011; Zijlstra et al., 2009a; Zijlstra et al., 2009b). This indicates that certain sensory characteristics, such as Thickness (Hogenkamp et al., 2011; Mattes and Rothacker, 2001; McCrickerd et al., 2012; Zijlstra et al., 2009b), and the degree to which these are perceived during the prandial experience, act as satietyrelevant sensory cues, changing the food or beverages capacity to generate satiety expectations. Identifying satiety-relevant sensory cues and designing foods with these sensory attributes should increase their satiating power.

The mechanism by which satiety-relevant sensory cues appear to work suggests that people learn to associate sensory characteristics with the subsequent experience of satiety post-consumption (Brunstrom, Shakeshaft and Scott-Samuel, 2008; Yeomans et al., 2014). As such, when presented with similar stimuli, expectations are made about how satiating the food or drink will be. Therefore, an indication of how a food may impact on actual food intake behaviour can be acquired by simply presenting a sensory stimulus and measuring the resulting expectations on food intake behaviour.

Nonetheless, disadvantages of using satiety-relevant sensory cues as a reformulation or design approach have been highlighted: 1) as learned sensory cues are associated with a given caloric value and satiety expectation, producing low-energy dense foods with sensory characteristics (such as Thickness and Creaminess) indicative of a greater energy content, which is not delivered by the food, typically results in compensatory intake (Yeomans and Chambers, 2011); and 2) palatability has been shown to be inversely correlated to satiating power (Drewnowski, 1998; de Graaf, de Jong and Lambers, 1999; Holt et al., 1995), a commercial disadvantage when we consider that hedonic appeal is a driver in consumer purchasing habits (Dhar and Wertenbroch, 2000).

If hedonic properties can be maintained, or even enhanced, an effective formulation or design approach would be to increase the satiating power of foods independently of energy content. Typically, energy dense foods associated with nutrients such as fat have a strong hedonic appeal (Prentice and Jebb, 2003). Within food systems, fat is often structured in the form of an emulsion. An emulsion is comprised of two immiscible liquids, the most common food emulsion being oil dispersed in water (e.g. mayonnaise, milk, dressings, creams), known as an oil-in-water emulsion.

Microstructural reformulation approaches have been shown to alter sensory characteristics and hedonics in model and applied emulsion food systems (Akhtar et al., 2005; Akhtar, Murray and Dickinson, 2006; de Wijk and Prinz, 2005; Kilcast and Clegg, 2002; Lett et al., in press; Mela, Langley and Martin, 1994; Moore et al., 1998; van Aken, Vingerhoeds and de Wijk, 2011; Vingerhoeds et al., 2008). Subsequently, through the manipulation of microstructural properties, the capability to change the capacity to which satiety expectations are generated could be realised, through altering perception intensity of sensory characteristics that act as satiety-relevant cues.

We report: 1) how microstructural differences in emulsion based food systems change perceptions of sensory attributes; 2) sensory attributes that promote hedonic appeal; and 3) sensory attributes that act as satiety-relevant sensory cues, within emulsion systems. Taking a multidisciplinary
approach, combining understanding of food engineering, sensory science, nutrition and food psychology, the work identifies the microstructural properties of emulsion food systems that promote individual sensory attributes and expected food intake behaviours. Most importantly, we aim to identify emulsion designs which may be used to maintain or enhance sensory and hedonic properties, but increase the satiating power of emulsion based foods.

2. Materials and Methods

2.1 Design and participants

The present study investigated the effect of oil droplet size and flavour type within model oil-in-water emulsions on the perception of sensory attributes, hedonics and expected food intake behaviours.

Male participants were recruited via advertisement and screened for food allergies, smoking habits, body mass index (BMI), current medical status and dietary habits (restricted eating) via Dutch eating behaviour questionnaire (DEBQ) (van Strien, et al., 1986). Females were excluded as they typically practice significantly higher levels of restricted eating and other eating behaviours than males (Wardle, 1987). The restricted eating DEBQ consisted of 10 questions having a five-option response format: never (1), seldom (2), sometimes (3), often (4), and very often (5). A restraint score was obtained by summing the scores for the 10 items and dividing by 10. A higher score indicates greater dietary restraint. Potential participants were prevented from participating if they indicated any food allergies, history of smoking, had a BMI above 24.9 Kg/M² or below 18.5 Kg/M², were taking medication known to interfere with sensory perception or food intake or had a DEBQ restricted eating score of >2.4 indicative of the participant occasionally or more often exercising restricted eating behaviour. 24 respondents met the study criteria and were included in the study. Participants were aged 18 - 26, with a mean BMI of 22.8 ± 1.7 Kg/m² and DEBQ restricted eating score of 1.8 ± 0.2. All participants gave written informed consent prior to participation. To guard against expectancy effects, the study was described as an investigation into the sensory analysis of emulsions. Ethical approval for the study was obtained from the University of Birmingham ethics committee.

2.2 Test samples

Samples consisted of an oil-in-distilled water emulsion (1 wt.% sodium caseinate (Excellion EM7, DMV International, The Netherlands)), 2 wt.% sucrose (Silverspoon granulated, British Sugar Plc, UK)) and 15 wt.% sunflower oil (Tesco Plc, UK)) with one of three flavours dependent on flavour condition: 1 wt.% vanilla extract (Nielsen-Massey Vanillas International LLC, The Netherlands), 0.05 wt.% cream flavouring (Frontier Natural Products Co-op, USA) and No flavour.

Emulsions were produced using two different methods dependent upon the required mean droplet size of the emulsion being produced: a high shear mixer (Silverson LSM, Silverson machines Ltd, UK) or a high-pressure homogeniser (GEA Niro Soavi Panda Plus 2000, GEA Niro Soavi, Italy). In a 600ml beaker, 15 wt.% sunflower oil was added to 85 wt.% aqueous phase (1.1 wt.% NaCas, 2.2 wt.% sucrose, 96.6 wt.% distilled water solution). The whole sample was then emulsified for 5 minutes using the high shear mixer. Dependent on oil droplet size being produced the sample was subjected
to a different rotational speed (rpm) and emulsor screen (fine (0.8 mm pores) or medium (1.6 mm pores)) (\(50 \mu m: 2500 \text{ rpm medium screen}\), \(40 \mu m: 3500 \text{ rpm medium screen}\), \(20 \mu m: 5000 \text{ rpm fine screen}\) and \(11 \mu m: 9000 \text{ rpm fine screen}\)). For emulsions produced using the high-pressure homogeniser, first a pre-emulsion was produced using the high shear mixer at 9000 rpm with a fine emulsor screen for 5 minutes using the high shear mixer. The pre-emulsion was then subjected to homogenisation, differing in pressure and number of passes (\(6 \mu m: 20 \text{ Bar} 3 \text{ passes}\), \(2 \mu m: 100 \text{ Bar} 2 \text{ passes}\) and \(0.2 \mu m: 1250 \text{ Bar} 4 \text{ passes}\)). All samples were produced in 400 g batches, under clean and hygienic conditions on the day of evaluation and stored under refrigerated conditions at 2-5 °C.

2.3 Measurement of sensory perception and expected food intake behaviours

Test sessions were scheduled between 10 am and 12 am or 2 pm and 4 pm, Monday to Friday, with sessions lasting 1 hour to 1 hour 30 minutes. Participants were instructed to arrive on one occasion having refrained from consuming any food or beverages other than water 2 hours before their arrival. Participants were seated in individual sensory booths and were presented with 21 40 ml samples in 60 ml twist closure lid pots coded with random 3 digit codes. All samples were served between 5-7 °C and were visually identical. To minimise volatile loss, all samples were served with the lids closed; participants were instructed only to remove the lid of the relevant sample during its analysis and then replace the lid once sample analysis was complete. Sample order was randomised differently for all assessors. Inter-sample duration was at the participant discretion and ranged from approximately 1-3 minutes. A spittoon was provided and subjects were instructed to spit out the sample after their assessment had been made. A bottle containing 400 g of water with 4 wt.% blue food colouring (Silverspoon blue food colouring liquid, British Sugar Plc, UK) was provided to act as a visual portion size reference for food intake expectation questions, which requires the participant to imagine they were to consume a bottle of the specific sample presented. 1 250 ml bottle of still water and 3 dry crackers were provided, and participants were instructed to use these to refresh their palate between samples. In addition to the randomised presentation of samples for each participant, to further minimise the impact of consuming the water and crackers on predicted food intake ratings, participants were instructed to rinse and spit with the water and ensure crackers were completely consumed by the end of the study (this worked out to be 1-2 bites of cracker after each sample).

Measurements of perceived intensity of sensory attributes, hedonics and expected food intake behaviours were made using visual analogue scales (VAS). Fifteen 100 mm randomised VAS’s acquiring information about the intensity of the sensory perception or level of expected intensity of the specific food behaviour e.g. “How <attribute> is sample <code>?”, or “Imagine you consumed an entire bottle of sample <code> right now, how strong would your <intake behaviour> be in <time period>?”, were presented. These questions were anchored with opposing statements left-to-right e.g. “Not at all <target attribute>” (scored as zero) and “Extremely [target attribute]” (scored as 100) (see Table 1). Questions differed slightly in order to be grammatically correct. Pre- and post-test participants rated their mood and appetite via VAS’s comprised of a series of questions in the form “How <word> do you feel?”. The evaluations rated were Full, Hungry, Desire to Eat, Prospective Consumption, Clearheaded, Calm, Happy, Anxious, Tired and Alert, in random order. Before testing, all sensory attributes were discussed individually with participants in accordance to the description shown in Table 1. Participants were also given the opportunity to ask any questions about the study and its protocol to clarify issues, queries or definitions before the test began.
2.4 Data analysis

Data and statistical analysis were carried out using IBM SPSS Statistic (SPSS Statistics 21, SPSS Inc., Chicago, US). The effect of emulsion design (oil droplet size and flavour condition) on sensory perceptions, hedonics and expected food intake behaviour were analysed via general linear model repeated-measures ANOVA. Test-within subject’s sphericity assumed significance was taken at 95% confidence interval and degrees of freedom and P values are presented. If a P value was considered significant, a pairwise comparison post-hoc Bonferroni test was performed to reveal the nature of the differences. Pre- and post-test mood ratings were compared via paired t-test with significance being taken at 95% confidence interval. To assess the direction and variability of relationships between microstructural components and attributes, or attributes and attributes, Pearson’s correlation (r) and coefficient of determination (R^2) were performed. Correlations are linear unless stated. Means and standard error of the mean (SEM) are presented throughout.

3. Results

3.1 Emulsion droplet size

Seven different emulsions varying in droplet size were produced. The volume weighted mean droplet sizes ($d_{4,3}$, µm) were 0.19 ± 0.02, 1.6 ± 0.17, 5.9 ± 0.65, 11.2 ± 0.38, 20.2 ± 0.83, 37.1 ± 0.94 and 48.1 ± 3.3. All samples displayed a unimodal oil droplet size distribution (data not shown). In all subsequent sections droplet sizes will be referred to as 0.2, 2, 6, 11, 20, 40 and 50 µm for simplicity.

3.2 Evaluations of emulsions

The mean sensory and expected food intake ratings are presented in table 2.

3.2.1 Flavour evaluations

The intensity of rated Vanilla flavour was dependent on both oil droplet size ($F(1, 6) = 3.18$) and flavour condition ($F(1, 2) = 18.53, P <0.001$), with no significant interaction ($F(1, 12) = 0.63, P >0.05$). Vanilla flavoured emulsions were perceived as having significantly greater Vanilla flavour compared to Cream ($P = 0.047$) and No flavour ($P <0.000$) emulsions. Additionally, Cream flavoured emulsions were perceived as having significantly greater Vanilla flavour than No flavour emulsions ($P <0.000$).

However, perception of Vanilla flavour also decreased significantly with increasing droplet size ($R^2 = 0.73, P = 0.006$), with a significant difference between droplets of 50 µm and 11 µm ($P = 0.013$).

The intensity of rated Cream flavour was dependent on both oil droplet size ($F(1, 6) = 8.14$) and flavour condition ($F(1, 2) = 7.87, P = 0.001$), with no significant interaction ($F(1, 12) = 0.54, P >0.05$). Cream flavour emulsions were perceived as having significantly greater Cream flavour compared to No flavour emulsions ($P = 0.004$), however not the Vanilla flavoured emulsions ($P >0.05$). Additionally, Vanilla flavoured emulsions were perceived as having a significantly greater Cream flavour than No flavour emulsions ($P = 0.03$). However, the perception of Cream flavour also decreased significantly with increasing droplet size ($R^2 = 0.73, P 0.006$) with 50 µm droplets being rated as less creamy than 0.2, 2, 6 or 11 µm emulsions ($P <0.000, P = 0.006, P = 0.003, P = 0.001$, respectively).
Sweetness intensity was dependent on flavour condition \(F(1, 2) = 8.27, P < 0.000\), but not droplet size \(F(1, 6) = 2.01, P > 0.05\), with no significant interaction \(F(1, 12) = 0.47, P > 0.05\). Vanilla and Cream flavoured emulsions were perceived as significantly sweeter \((P = 0.001, P = 0.02, \text{respectively})\) than the No flavour emulsions.

### 3.2.2 Mouthfeel and texture evaluations

Thickness perception intensity was dependent on droplet size \(F(1, 6) = 2.6, P = 0.02\), but not flavour condition \(F(1, 2) = 0.8, P > 0.05\), with no significant interaction \(F(1, 12) = 0.71, P > 0.05\). Thickness significantly decreased with increasing droplet size \(r = -0.58, R^2 = 0.34\), with a significant difference between droplets of 50 \(\mu m\) and 40 \(\mu m\) \(P = 0.049\).

Creamy Mouthfeel intensity depended on droplet size \(F(1, 6) = 9.69, P < 0.000\), but not flavour condition \(F(1, 2) = 0.84, P > 0.05\), with no significant interaction \(F(1, 12) = 0.98, P > 0.05\). Creamy Mouthfeel intensity significantly decreased with increasing droplet size \(r = -0.92, R^2 = 0.85\), with emulsions with 50 \(\mu m\) droplets being rated as having a less creamy mouthfeel than those with 0.2, 2, 6 or 11 \(\mu m\) droplets \((P < 0.000, P = 0.004, P = 0.003, P = 0.001, \text{respectively})\) and 0.2 \(\mu m\) having a creamier mouthfeel than 20 \(\mu m\) droplets \(P = 0.029\).

Smoothness perception intensity was dependent on droplet size \(F(1, 6) = 3.69, P = 0.002\), but not flavour condition \(F(1, 2) = 1.4, P > 0.05\), with no significant interaction \(F(1, 12) = 0.69, P > 0.05\). The significant difference at a 94% confidence interval was between droplets of 2 \(\mu m\) and 20 \(\mu m\) \((P = 0.059)\). The trend between droplet size and ratings of Smoothness is interesting; a strong polynomial relationship \(R^2 = 0.76\) between droplet size and smoothness was demonstrated, despite there being a weak linear relationship \(R^2 = 0.29\). The polynomial relationship appears to be a result of the increase in perception intensity at 50 \(\mu m\). A strong linear relationship is observed when 50 \(\mu m\) is removed \(R^2 = 0.73\); however, the order 2 polynomial relationship also increases in strength \(R^2 = 0.82\).

Slipperiness perception did not significantly \((P > 0.05)\) differ as a function of droplet size \(F(1, 6) = 0.55\), flavour condition \(F(1, 2) = 1\) or interaction \(F(1, 12) = 0.72\).

### 3.2.3 Overall sensory evaluations

Creaminess perception intensity was dependent on droplet size \(F(1, 6) = 10.47, P < 0.001\), but not flavour condition \(F(1, 2) = 0.37, P > 0.05\), with no significant interaction \(F(1, 12) = 0.76, P > 0.05\). Creaminess significantly decreased with increasing droplet size \(r = -0.94, R^2 = 0.89\), with emulsions with 50 \(\mu m\) being rated as significantly less creamy than those with 0.2, 2, 6, 11 or 20 \(\mu m\) droplets \((P < 0.000, P = 0.003, P = 0.008, P = 0.006, P = 0.037, \text{respectively})\). Emulsions with 0.2 \(\mu m\) droplets were also significantly creamier than those with 20 or 40 \(\mu m\) droplets \((P = 0.019, P = 0.011, \text{respectively})\).

Oiliness perception intensity did not depend on oil droplet size \(F(1, 6) = 0.07, P > 0.05\) or flavour condition \(F(1, 2) = 0.76, P > 0.05\), but a flavour condition*droplet interaction was observed \(F(1, 12) = 2.803, P = 0.001\). Contrasts revealed significant differences in oiliness between 20 \(\mu m\) No flavour emulsions and 6, 20, 40 and 50 \(\mu m\) Vanilla emulsions \((P = 0.011, P = 0.001, P = 0.01, P = 0.001, \text{respectively})\) and 0.2, 2, 6, 11, 40 and 50 \(\mu m\) Cream emulsions \((P = 0.011, P = 0.008, P = 0.025, P = 0.008, P = 0.003, P = 0.047, \text{respectively})\). 0.2 \(\mu m\) No flavour emulsions and 2 and 11 \(\mu m\) Vanilla emulsions were perceived as significantly oilier \((P = 0.001, P = 0.02, \text{respectively})\) and 0.2 \(\mu m\) having significantly less oiliness than 20 or 40 \(\mu m\) droplets \((P = 0.029\).
flavoured emulsions (P = 0.008, P = 0.018, respectively) and 50 µm Cream flavoured emulsions (P = 0.043), 11 µm No flavour emulsions and 20 and 50 µm Vanilla flavoured emulsions (P = 0.021, P = 0.005, respectively) and 20 µm Vanilla emulsions and 50 µm Cream flavoured emulsions (P = 0.04).

Liking was dependent on both droplet size (F (1, 6) = 5.53, P < 0.000) and flavour condition (F (1, 2) = 8.23, P = 0.001), with no significant interaction (F (1, 12) = 0.99, P > 0.05). Vanilla flavoured emulsions were liked significantly more than Cream (P = 0.046) and No flavour (P = 0.008) emulsions, but liking of Cream and No Flavour emulsions was similar (P > 0.05). However, Liking significantly decreased with increasing droplet size (r = - 0.89, R² = 0.79), with 0.2 µm droplet emulsions being liked more than 20, 40 and 50 µm emulsions (P = 0.012, P = 0.011, P = 0.01, respectively) and 11 µm emulsions being more liked than 20 and 50 µm emulsions (P = 0.006, P = 0.045, respectively).

3.2.4 Expected food intake evaluations

Expected Filling was dependent on droplet size (F (1, 6) = 3.08, P = 0.007), but not flavour condition (F (1, 2) = 0.67, P > 0.05), with no significant interaction (F (1, 12) = 0.8, P > 0.05). Expected Filling significantly decreased with increasing droplet size (r = - 0.9, R² = 0.8), the significant difference being between emulsions with droplets of 0.2 µm and 50 µm (P = 0.025).

Expected Hunger in 1 hour was dependent on droplet size (F (1, 6) = 5.8, P < 0.000), but not flavour condition (F (1, 2) = 2, P > 0.05), with no significant interaction (F (1, 12) = 1.1, P > 0.05). Expected Hunger in 1 hour significantly increased with increasing droplet size (r = 0.76, R² = 0.57). The significant difference being between emulsions with droplets of 50 µm and those with 2, 6 and 20 µm droplets (P = 0.017, P = 0.008, P = 0.008, respectively).

Expected Desire to Eat in 1 hour was unaffected by oil droplet size (F (1, 6) = 2.18, P > 0.05) or flavour condition (F (1, 2) = 0.1, P > 0.05), but there was a significant flavour condition*droplet interaction (F (1, 12) = 2.33, P = 0.007). Contrasts revealed significant differences in Expected Desire to Eat in 1 hour for 0.2 µm Vanilla flavoured emulsions and 20 and 50 µm Cream flavoured emulsions (P = 0.034, P = 0.013, respectively) and 11 µm No flavour emulsions (P = 0.048), 2 µm Cream flavoured emulsions and 11, 20 and 50 µm No flavour emulsions (P = 0.026, P = 0.048, P = 0.028, respectively), 6 µm Cream flavoured emulsions and 2 µm No flavour emulsions (P = 0.048), 20 µm Cream flavoured emulsions and 11 and 50 µm No flavour emulsions (P = 0.011, P = 0.009 respectively) and 40 µm Vanilla flavour emulsions (P = 0.025), 40 µm Cream flavoured emulsions and 2 µm No flavour emulsions (P = 0.037) and 50 µm Cream flavoured and 0.2, 2, 6 and 40 µm No flavour emulsions (P = 0.042, P = 0.005, P = 0.015, P = 0.048, respectively).

Ratings of Prospective Consumption and Desire to Eat immediately did not significantly differ as a function of droplet size (F (1, 6) = 1.08, P > 0.05; F (1, 6) = 1.94, P > 0.05, respectively), flavour condition (F (1, 2) = 2.26, P > 0.05; F (1, 2) = 1.94, P > 0.05, respectively), or an interaction (F (1, 12) = 1.08, P > 0.05; F (1, 12) = 1.15, P > 0.05, respectively).

3.3 Sensory attribute – expected food intake behaviour correlations

Attribute-Attribute correlations (see Table 3) highlight the relationship between sensory attributes and prandial outcome expectations.

3.4 Mood ratings
Participants’ mood rating scores were not significantly different pre- and post-test ($P >0.05$). Therefore, differences in sensory ratings were as a result of sample differences and not participants’ mood.

### 4. Discussion

The results of this study indicate that participants, who were untrained, were able to perceive significant differences in flavour, mouthfeel, texture, hedonics and expectations of food intake behaviour as a result of differences in emulsion design: flavour type and oil droplet size.

The microstructural property that had the predominant effect on perceived sensory characteristics, food intake expectations and sample hedonics was oil droplet size. Thus, our findings suggest that greater consideration should be given to this structural component during reformulation of emulsion-based food products. In comparison to previous studies investigating oil droplet size (Akhtar et al., 2005; de Wijk and Prinz, 2005; Vingerhoeds et al., 2008), in this work a larger range of droplet sizes was considered. Our results demonstrate that when a larger oil droplet size range is investigated, many findings emerge that were not evident with narrower range of droplet sizes.

Flavour intensity (Vanilla and Cream) significantly decreased with increasing droplet size, an observation which may relate to the greater surface area with smaller droplets. Thus, the increased contact between the sample and the surface of the mouth could have enhanced flavour intensity, in line with previous findings in other contexts (Malone, Appelqvist & Norton, 2003). However, the observed relationship was mainly due to decreased perception of these properties with 50 $\mu$m droplets, a finding which highlights a future opportunity to decrease flavour intensity. An interesting observation is that a greater number of oil droplet sizes were significantly different to the sample with 50 $\mu$m droplets in the Cream flavoured emulsions, which contained an oil-soluble flavour, than the Vanilla flavoured emulsions that contained a water-soluble flavour. This highlights a potential difference in flavour intensity dependent on the phase location of the flavour within an emulsion system and a surface area effect of droplet size on oil-soluble flavour perception. This would be an interesting area for further investigation.

The main sensory attribute types in which significant differences in perception were generated as a function of oil droplet size were related to mouthfeel and textural sensations. Studies considering Thickness perception and oil droplet size often report increasing Thickness perception with decreasing droplet size. Commonly this is shown to be a result of increasing viscosity with decreasing droplet size, since a strong correlation between viscosity and Thickness perception has been shown previously (Cutler, Morris and Taylor, 1983; Kokini, Kadane and Cussler, 1977; Shama and Sherman, 1973a; Shama and Sherman, 1973b; Wood, 1968). Our observations highlighted a weak linear relationship, with Thickness perception decreasing as droplet size increased, although this was only significant between two oil droplets of adjacent sizes, and so should be interpreted with some caution. This could be a result of the sensory protocol and/or the systems themselves, as suggested by Lett et al., (in preparation), since only subtle viscosity differences in emulsions of these droplet sizes exist, identifying a perceivable difference in Thickness may be challenging to untrained participants.
Our observations do suggest that droplet size effects Smoothness perception, which agrees with previous observations (de Wijk and Prinz, 2005). Our results using a droplet size range of 0.2 - 50 µm highlight significant differences, but only at a 94% confidence interval. This suggests that although statistical significance is shown, oil droplet size may have a lesser influence on Smoothness than the other attributes. However, the trend between oil droplet size and Smoothness was complicated. At the full droplet size range investigated a polynomial trend was shown; on omitting 50 µm droplets (whose data seemed not to fit the trend for other emulsions) a linear increase in smoothness was shown, however a polynomial trend remained and strengthened. Given the known strength of the correlation between friction coefficient and Smoothness perception (de Wijk and Prinz, 2005; Kokini et al., 1984), the polynomial second order trend with friction coefficient with emulsions of these droplet sizes (Lett et al., in preparation) and our current observations that the significant difference in perception occurs between a small and median size droplets, suggests that with such a large droplet size range the relationship between Smoothness and droplet size is polynomial, but why this is so remains unclear.

Creaminess perceptions of the emulsions were not significantly influenced by flavour type, a relationship also demonstrated by Kilcast and Clegg (2002). Instead our observations show that Creamy Mouthfeel and overall Creaminess increases significantly with decreasing droplet size. Given the strength of correlation between Creaminess and Creamy Mouthfeel (r: 0.99, R²: 0.98), this strongly suggests overall Creaminess and Creamy Mouthfeel were assessed as the same attribute. This could be attributed to the synthetic manner in which ordinary consumers, as represented the untrained participants, perceived food, assessing the totality of an attribute, instead of assessing attributes analytically when requested (Frost and Janhoj, 2007). Nevertheless, this observation highlights that Creaminess was predominantly influenced by textural/mouthfeel attributes, a conclusion also reached by Frost and Janhoj (2007) in liquid systems. This further suggests that the mechanism through which oil droplet size modified Creaminess was through altered mouthfeel. When hedonics and expected food intake behaviours is also considered, this observation provides an extremely interesting finding which can be related to a modifiable emulsion design property (Table 2a).

As previously observed in liquid dairy products (Richardson-Harman et al., 2000) and semi-solids (Daget, Joerg and Bourne, 1987; Elmore et al., 1999) and shown here in liquid emulsions, Creaminess is strongly and significantly positively correlated with the sample’s hedonic appeal. When we regard expected food intake behaviours, our results in relation to Creaminess demonstrate a novel and substantial finding.

Expected Filling significantly increased with decreasing droplet size and Expected Hunger significantly decreased with decreasing droplet size. In regards to a predominant sensory characteristic that would be driving these differences, the attribute Thickness (Hogenkamp et al., 2011; Mattes and Rothacker, 2001; McCrickerd et al., 2012; Zijlstra et al., 2009b) displays a strong significant correlation with Expected Filling and hunger in 1 hours time (see Table 3), despite potential erroneous data due to subtleties in viscosity. However, Thickness does not show the strongest correlation (see Table 3). Additionally, Smoothness, Slipperiness and oiliness were not shown to be directly involved in hedonics or any expected food intake behaviours (see Table 3). Instead, the strongest significant correlation for both Expected Filling and hunger was with Creaminess (see Table 3). This suggests with increasing Creaminess we see an increase in Expected...
Filling and a decrease in Expected Hunger. Therefore, Creaminess, as well as being a predominant influence in hedonics (see Table 3), can also generate greater expectations of filling and decreased hunger. If this observation translated to actual eating behaviour, this would highlight Creaminess as a key target attribute, which would allow foods to be engineered via droplet size manipulations to modify eating behaviour, but also maintain hedonic properties (see Table 2a). Clearly, future work should determine if expected ratings translate to real behaviour.

Given our earlier discussion regarding participants considering Creaminess as a textural/mouthfeel attribute, this difference in expected food intake behaviour mediated by Creaminess is suggested to be related to textural/mouthfeel sensations. This could be because texture is one sensory characteristic that reliably predicts nutrient content (Drewnowski, 1990) especially for attributes such as Creaminess which are typically associated with fat content (de Wijk, Rasing and Wilkinson, 2003; Frost and Janhoj, 2007). Thus, for energy-dense foods containing structures such as the oil-water emulsions used here, modifying droplet size could lead to enhanced satiety expectations that could enhance the degree to which participants subsequently respond to the ingested fat, in line with evidence that increased satiety expectations increase satiety generated by other macronutrients (Bertenshaw, Lluch and Yeomans, 2013; McCrickerd, Chambers, & Yeomans, 2014; Yeomans and Chambers, 2011). However, if the increase in expected satiety generated by manipulated droplet size was not matched by adequate nutrient ingestion, data suggests there might be a risk of rebound hunger (Yeomans and Chambers, 2011), and so the use of modified droplet size to generate satiety expectations in the context of low-energy products should be approached with caution. Nevertheless, the observation that droplet size affects expected satiety is important in relation to actual short-term eating behaviour when we consider the effect of expectations on eating behaviour mediators such as ghrelin response, which has been demonstrated to be significantly lower if the preload is assumed to be caloric (Crum et al., 2011). Furthermore, our results still highlight an interesting finding that Creaminess may also provide a functional benefit in relation to actual eating behaviour.

With regards to flavour type, the flavour manipulations were included primarily as a positive control to ensure that the ratings used were significantly sensitive to detect effects, guarding against the possibility than droplet manipulations may have had no effects (although in practice droplet size had very clear effects). As expected, a significant increase in ratings of Vanilla and Cream flavour intensity were observed with the addition of the respective flavour. Interestingly, just the presence of a flavour significantly increased Sweetness and Vanilla and Cream flavour intensities. It is generally considered Sweetness intensity is enhanced by odour, when sweet congruent odours are added to sugar solutions (Cliff and Noel, 1990; Frank and Byran, 1988; Frank, Ducheny and Mize, 1989; Valentin, Chrea and Nguyen, 2006). Odorants like Vanilla and Cream flavours are themselves rated as “sweet” tasting (even though they contain no specific sweet tastants). This enhancement of Sweetness through the presence of odorants has been demonstrated in protocols where samples are swallowed and spat out by participants, as seen within our protocol (Frank, Ducheny and Mize, 1989).

Additionally, our findings highlight a significant increase in Liking was achieved with the addition of Vanilla flavour, compared to No flavour or Cream flavour. Independent of flavour related questions, flavour type did not independently significantly effect the perception of mouthfeel or texture, and did not effect overall or expected food intake behaviour. However, flavour type significantly
influenced expected Desire to Eat in 1 hours’ time and oiliness in an interaction with droplet size. An unexpected result given that an oil droplet*flavour interaction is not shown in any other expected appetite or satiety attributes. However, findings regarding oiliness are more in line with other findings. Lett et al., (in preparation) found that frictional properties form a part of Oiliness perception; however, other influences such as flavour could be involved within the formation of the multi-influenced attribute Oiliness. Our findings support this conclusion, with oiliness perception being a result of an oil droplet*flavour interaction, independent of just flavour or oil droplet size alone. Additionally, as results indicate that flavour only significantly affected perceived flavour intensities, and oil droplet independently affected mouthfeel and textural perceptions. An interaction between the two variables would be expected for a significant difference in perception of an attribute which is comprised of textural and flavour perceptions. Given our observations, using flavour type as a reformulation technique, should only be considered in emulsion based food products when looking to produce a specific flavour or to manipulate Oiliness intensity.

5. Conclusion

The present study has shown that changing oil droplet size significantly altered flavour intensity, Thickness, Smoothness, Creamy Mouthfeel, Creaminess, Liking, Expected Filling and Expected Hunger in 1 hours’ time. Altering the flavour of these emulsions using odour-based flavourants only significantly changed flavour intensity, Sweetness and Liking. The most important observation highlighted in this study is that by altering the emulsion design through decreasing oil droplet size, perceived Creaminess can be significantly enhanced which as a result significantly increases Hedonic appeal as well as increasing Expected Filling and reducing Expected Hunger, independent of energy content. If shown to relate to actual eating behaviour, this would provide a key target attribute which can be manipulated through emulsion design, to produce hedonically appropriate satiating foods.

Acknowledgements

The authors gratefully acknowledge Keri McCrickerd (University of Sussex) for useful discussions.

References


Table 1: Assessment attributes used during measurements of sensory perception, hedonics and expected food intake behaviour analysis, with description.

<table>
<thead>
<tr>
<th>Attribute category</th>
<th>Sensory attribute</th>
<th>Description reference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Flavour</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Vanilla flavour intensity</td>
<td>Degree of perceived vanilla flavour</td>
</tr>
<tr>
<td></td>
<td>Cream flavour intensity</td>
<td>Degree of perceived cream flavour</td>
</tr>
<tr>
<td></td>
<td>Sweetness</td>
<td>Degree of sweet taste associated with table sugar</td>
</tr>
<tr>
<td><strong>Mouthfeel</strong></td>
<td>Smoothness</td>
<td>Degree of absence of any particles, lumps, bumps etc within the sample</td>
</tr>
<tr>
<td></td>
<td>Thickness</td>
<td>Viscous consistency within the mouth; <em>Water to yoghurt</em></td>
</tr>
<tr>
<td></td>
<td>Slipperiness</td>
<td>Degree to which the product slides over the tongue</td>
</tr>
<tr>
<td></td>
<td>Creamy</td>
<td>Soft, smooth with flowing consistency; <em>Water to full fat cream</em></td>
</tr>
<tr>
<td><strong>Overall</strong></td>
<td>Creaminess</td>
<td>Assessment of overall creaminess of the sample</td>
</tr>
<tr>
<td></td>
<td>Oiliness</td>
<td>Assessment of overall oiliness of the sample</td>
</tr>
<tr>
<td></td>
<td>Liking</td>
<td>Overall liking of the sample</td>
</tr>
<tr>
<td><strong>Expected food intake behaviour</strong></td>
<td>Filling</td>
<td>Measure of expected satiation if to consume 400g, referenced to 400g water portion</td>
</tr>
<tr>
<td></td>
<td>Hunger in 1 hours time</td>
<td>Measure of expected satiety if to consume 400g, referenced to 400g water portion</td>
</tr>
<tr>
<td></td>
<td>Prospective Consumption in 1 hours time</td>
<td>Measure of expected quantity consumed, if to consume 400g now of the sample and 400g again in 1 hours time, referenced to 400g water portion</td>
</tr>
<tr>
<td></td>
<td>Desire to Eat immediately</td>
<td>Measure of expected appetite if to consume 400g, referenced to 400g water portion</td>
</tr>
<tr>
<td></td>
<td>Desire to Eat in 1 hours time</td>
<td>Measure of expected appetite in 1 hours time if to consume 400g, referenced to 400g water portion</td>
</tr>
</tbody>
</table>
Table 2 Mean (± SEM) sensory and expected food intake ratings of samples for droplet size (a) and flavour (b) as variables.

(a) Emulsion sample (Droplet size μm)

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<tr>
<th></th>
<th>0.2</th>
<th>2</th>
<th>6</th>
<th>11</th>
<th>20</th>
<th>40</th>
<th>50</th>
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<tr>
<td>Vanilla Flavour</td>
<td>50.7 ± 3.8</td>
<td>48.1 ± 3.5</td>
<td>48.4 ± 3</td>
<td>50.8 ± 3.2</td>
<td>46.9 ± 3</td>
<td>46.3 ± 3.2</td>
<td>39.4 ± 3.7</td>
</tr>
<tr>
<td>Cream Flavour</td>
<td>62.3 ± 3</td>
<td>56.4 ± 3.1</td>
<td>57.6 ± 2.4</td>
<td>56.4 ± 2.7</td>
<td>50.8 ± 3.4</td>
<td>49.7 ± 3.9</td>
<td>45.9 ± 3.4</td>
</tr>
<tr>
<td>Sweetness</td>
<td>51.5 ± 3.6</td>
<td>48.9 ± 3.4</td>
<td>47.9 ± 3.3</td>
<td>52.2 ± 3.6</td>
<td>44.7 ± 3.6</td>
<td>47.3 ± 3.5</td>
<td>46.5 ± 3.8</td>
</tr>
<tr>
<td>Smoothness</td>
<td>61.8 ± 3</td>
<td>63.4 ± 2.9</td>
<td>62.3 ± 3</td>
<td>60.4 ± 3.2</td>
<td>53.3 ± 3.4</td>
<td>54.7 ± 3.7</td>
<td>60.1 ± 3.8</td>
</tr>
<tr>
<td>Thickness</td>
<td>43.3 ± 3.8</td>
<td>40.2 ± 3.4</td>
<td>41.5 ± 3</td>
<td>39.8 ± 3.1</td>
<td>38.5 ± 3</td>
<td>43.4 ± 3.3</td>
<td>32.8 ± 2.9</td>
</tr>
<tr>
<td>Slipperiness</td>
<td>59.3 ± 3.7</td>
<td>58 ± 3.7</td>
<td>56.7 ± 3.5</td>
<td>56 ± 3.2</td>
<td>54.1 ± 2.9</td>
<td>56.6 ± 3.2</td>
<td>58.1 ± 3.9</td>
</tr>
<tr>
<td>Creamy Mouthfeel</td>
<td>63.3 ± 3.4</td>
<td>58.7 ± 3.7</td>
<td>59.8 ± 3.3</td>
<td>58.7 ± 2.9</td>
<td>51.8 ± 3.4</td>
<td>53.3 ± 4</td>
<td>44.6 ± 3.6</td>
</tr>
<tr>
<td>Creaminess</td>
<td>65.5 ± 3.7</td>
<td>59.2 ± 4.1</td>
<td>61.7 ± 3.6</td>
<td>60.3 ± 4</td>
<td>51.1 ± 3.6</td>
<td>50.2 ± 4.1</td>
<td>49 ± 3.5</td>
</tr>
<tr>
<td>Oiliness</td>
<td>45.4 ± 3.9</td>
<td>43.6 ± 4.3</td>
<td>43.6 ± 3.4</td>
<td>40.6 ± 3.6</td>
<td>40.8 ± 3.8</td>
<td>44.7 ± 4.2</td>
<td>44.6 ± 3.7</td>
</tr>
<tr>
<td>Liking</td>
<td>53.8 ± 2.9</td>
<td>47.8 ± 3.4</td>
<td>48 ± 3</td>
<td>50.1 ± 2.6</td>
<td>43.7 ± 2.5</td>
<td>41.9 ± 3.8</td>
<td>40.4 ± 3.7</td>
</tr>
<tr>
<td>Filling</td>
<td>63.2 ± 3.2</td>
<td>61.1 ± 3.7</td>
<td>60 ± 3.7</td>
<td>58.1 ± 2.9</td>
<td>56.4 ± 3.3</td>
<td>57.7 ± 4.3</td>
<td>50.8 ± 4</td>
</tr>
<tr>
<td>Hunger in 1 hours</td>
<td>44.2 ± 5.4</td>
<td>44.9 ± 5.1</td>
<td>45 ± 4.6</td>
<td>49 ± 4.4</td>
<td>45 ± 4.6</td>
<td>46.3 ± 4.8</td>
<td>57.4 ± 4</td>
</tr>
<tr>
<td>Prospective...</td>
<td>57.4 ± 5.3</td>
<td>54.2 ± 5.2</td>
<td>59.3 ± 4.3</td>
<td>59.4 ± 5.3</td>
<td>58.3 ± 4.6</td>
<td>59.9 ± 5</td>
<td>59.3 ± 4.5</td>
</tr>
<tr>
<td>Desire to Eat in 1 h</td>
<td>42.4 ± 4.4</td>
<td>41.8 ± 4.7</td>
<td>42.2 ± 4.3</td>
<td>42 ± 4.6</td>
<td>41.9 ± 4.3</td>
<td>44 ± 4.6</td>
<td>48.3 ± 4.2</td>
</tr>
<tr>
<td>Desire to Eat...</td>
<td>48.8 ± 4.4</td>
<td>46.8 ± 4.6</td>
<td>49.6 ± 4.4</td>
<td>51.9 ± 4.1</td>
<td>49.4 ± 4.5</td>
<td>51.1 ± 4.2</td>
<td>54.7 ± 4.1</td>
</tr>
</tbody>
</table>

(b) Emulsion sample (Flavour)

<table>
<thead>
<tr>
<th></th>
<th>Vanilla</th>
<th>Cream</th>
<th>No flavour</th>
</tr>
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<tbody>
<tr>
<td>Vanilla Flavour</td>
<td>57.7 ± 2.7</td>
<td>46.9 ± 3.9</td>
<td>37 ± 3.3</td>
</tr>
<tr>
<td>Cream Flavour</td>
<td>56.3 ± 3.2</td>
<td>57 ± 2.9</td>
<td>49.2 ± 2.5</td>
</tr>
<tr>
<td>Sweetness</td>
<td>53 ± 3.2</td>
<td>50.8 ± 4.2</td>
<td>41.5 ± 3.2</td>
</tr>
<tr>
<td>Smoothness</td>
<td>61.1 ± 3</td>
<td>58.3 ± 2.9</td>
<td>58 ± 2.7</td>
</tr>
<tr>
<td>Thickness</td>
<td>41.1 ± 2.6</td>
<td>40 ± 3</td>
<td>38.7 ± 2.4</td>
</tr>
<tr>
<td>Slipperiness</td>
<td>57.9 ± 2.6</td>
<td>56.8 ± 3.1</td>
<td>56.3 ± 3.5</td>
</tr>
<tr>
<td>Creamy Mouthfeel</td>
<td>54.6 ± 3.4</td>
<td>57.5 ± 3.1</td>
<td>55.1 ± 3.3</td>
</tr>
<tr>
<td>Creaminess</td>
<td>56.2 ± 3.4</td>
<td>56.7 ± 3.4</td>
<td>54.6 ± 3.4</td>
</tr>
<tr>
<td>Oiliness</td>
<td>43.4 ± 3.6</td>
<td>43.5 ± 3.7</td>
<td>43 ± 3</td>
</tr>
<tr>
<td>Liking</td>
<td>52.3 ± 3</td>
<td>46 ± 3.3</td>
<td>41.2 ± 2.7</td>
</tr>
<tr>
<td>Filling</td>
<td>57.7 ± 3.1</td>
<td>59.8 ± 3.5</td>
<td>57.1 ± 3.2</td>
</tr>
<tr>
<td>Hunger in 1 hours</td>
<td>49.2 ± 4.5</td>
<td>46.3 ± 4.7</td>
<td>46.7 ± 4.1</td>
</tr>
<tr>
<td>Prospective...</td>
<td>61.1 ± 4.6</td>
<td>58.1 ± 5.2</td>
<td>55.6 ± 4.6</td>
</tr>
<tr>
<td>Desire to Eat...</td>
<td>42.6 ± 4.6</td>
<td>43.4 ± 4.5</td>
<td>43.6 ± 4</td>
</tr>
<tr>
<td>Desire to Eat...</td>
<td>50.5 ± 4.5</td>
<td>49.8 ± 4.5</td>
<td>50.7 ± 3.7</td>
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</table>
Table 3: Pearsons correlation (r) Coefficient of determination (Linear R²) of mean sensory attribute, hedonic and expected food intake ratings as a function of one another

<table>
<thead>
<tr>
<th>r</th>
<th>Thickness</th>
<th>Smoothness</th>
<th>Slipperiness</th>
<th>Creamy Mouthfeel</th>
<th>Creaminess</th>
<th>Oiliness</th>
<th>Liking</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filling</td>
<td>0.85*</td>
<td>0.28</td>
<td>0.40</td>
<td>0.96*</td>
<td>0.92*</td>
<td>0.14</td>
<td>0.84*</td>
</tr>
<tr>
<td>Hunger in 1 hours time</td>
<td>-0.85*</td>
<td>0.16</td>
<td>0.03</td>
<td>-0.77*</td>
<td>-0.70</td>
<td>0.09</td>
<td>-0.55</td>
</tr>
<tr>
<td>Prospective Consumption in 1 hours time</td>
<td>-0.09</td>
<td>-0.36</td>
<td>-0.46</td>
<td>-0.36</td>
<td>-0.36</td>
<td>-0.10</td>
<td>-0.35</td>
</tr>
<tr>
<td>Desire to Eat immediately</td>
<td>-0.71</td>
<td>0.31</td>
<td>0.07</td>
<td>0.80*</td>
<td>-0.78*</td>
<td>0.44</td>
<td>-0.66</td>
</tr>
<tr>
<td>Desire to Eat in 1 hours time</td>
<td>-0.65</td>
<td>-0.05</td>
<td>-0.22</td>
<td>-0.71</td>
<td>-0.66</td>
<td>0.01</td>
<td>-0.54</td>
</tr>
<tr>
<td>Liking</td>
<td>0.56</td>
<td>0.36</td>
<td>0.58</td>
<td>0.92*</td>
<td>0.96*</td>
<td>-0.02</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>R²</th>
<th>Thickness</th>
<th>Smoothness</th>
<th>Slipperiness</th>
<th>Creamy Mouthfeel</th>
<th>Creaminess</th>
<th>Oiliness</th>
<th>Liking</th>
</tr>
</thead>
<tbody>
<tr>
<td>Filling</td>
<td>0.73</td>
<td>0.16</td>
<td>0.08</td>
<td>0.92</td>
<td>0.85</td>
<td>0.02</td>
<td>0.70</td>
</tr>
<tr>
<td>Hunger in 1 hours time</td>
<td>0.73</td>
<td>0.00</td>
<td>0.03</td>
<td>0.59</td>
<td>0.50</td>
<td>0.01</td>
<td>0.30</td>
</tr>
<tr>
<td>Prospective Consumption in 1 hours time</td>
<td>0.01</td>
<td>0.21</td>
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<td>0.13</td>
<td>0.13</td>
<td>0.10</td>
<td>0.12</td>
</tr>
<tr>
<td>Desire to Eat immediately</td>
<td>0.50</td>
<td>0.01</td>
<td>0.09</td>
<td>0.65</td>
<td>0.61</td>
<td>0.19</td>
<td>0.44</td>
</tr>
<tr>
<td>Desire to Eat in 1 hours time</td>
<td>0.40</td>
<td>0.05</td>
<td>0.00</td>
<td>0.50</td>
<td>0.44</td>
<td>0.00</td>
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<tr>
<td>Liking</td>
<td>0.32</td>
<td>0.34</td>
<td>0.13</td>
<td>0.85</td>
<td>0.92</td>
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</tbody>
</table>

*correlation coefficient is significant at P < 0.05.
Highlights

- Emulsion oil droplet size ($d_{4,3} \text{ 0.2 - 50 } \mu\text{m}$) and flavour were investigated.
- Sensory perception, hedonics and expected food intake behaviour were explored.
- Sensory ratings, Liking and expected satiety/satiation significantly differed.
- ↓ Oil droplet size = ↑ Creaminess = ↑ Liking, expected satiation and satiety.
- Promising hedonically appropriate satiating emulsion designs were identified.