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Sensory and analytical characterization of the "cool-melting" perception of commercial spreads

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Abstract

Butters, margarines and table spreads are water-in-oil emulsions. Melting characteristics of these products are important for flavor release and consumer acceptance. One characteristic that is believed to discriminate butters from margarines is a cooling sensation perceived in-mouth while consuming these products. Here, we investigated different methods to characterize sensorically and analytically the "cool-melting" properties of commercial butter and margarines. Our results show that butter indeed can be distinguished from margarines based on their "cool-melting" properties. Furthermore, changes in enthalpy as measured through DSC and solid fat content are good predictors of the "cool-melting" effect of spreads.

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Practical applications

By understanding the mechanisms of the "cool-melting" perception of spreads, and linking them to analytical measurements, we can create an in-vitro quantification method of "cool-melting." This method can eventually help directing product development to achieve the desire product profile and increase consumer acceptance and liking of margarines and low-fat spread products. In this study we did not assess the impact of "cool-melting" on consumer perception, which would be the next step in understanding the drivers of liking of spread products.

KEYWORDS

butter, cooling, enthalpy, margarine, melting, w/o, water in oil emulsions

1 | INTRODUCTION

The relationship between fat liking and meltability is highly dependent on the type of product. Butter has been described to give a particularly pleasant in-mouth texture with a significant cooling impact on the tongue and a rapid and sharp mouth melt, with no coated or waxy mouthfeel (Lomneth, Blair, Parnell, Park, & Tao, 1983). The cooling sensation has been associated with almost instantaneous absorption of the heat of crystallization due to a steep melting profile (Chrysan, 2005). According to Borwankar (Borwankar, Frye, Blaurock, & Sasevich, 1992), perceived meltability of butters, margarines and table spreads represents a combined perception of cooling sensation, accompanying the melting of fat crystals; and the sensation of flow, relating to the rheological transitions. They explain that the cooling effect is only relevant for butter and highfat margarines, where both components of melting perception occur at body temperature. Conversely, in the case of reduced fat products, the cooling is imperceptible and only the component related to the flowability plays a role (Borwankar et al., 1992). Kodali et al. (2013) suggested that if a fat has a high amount of solids at room temperature and melts quickly at or below body temperature it creates a smooth cooling sensation in the mouth as it absorbs the energy from the mouth cavity.

Typically butter contains 80–84% milk fat which is characterized by a relatively high level of short-chain fatty acids (C4–C10) (Frede, 2011). Although butter has a unique flavor and a natural image, it also has some aspects that could be considered negative, such as its high fat content, high content of saturated fat, and reduced spreadability at
 TABLE 1
 Typical fatty acid composition of milk fat and margarine (Adapted from Gunstone and Harwood, 2007; O'Brien, 2009)

| Fatty acid composition | Milk fat% | Margarine% |
|---------------------------|-----------|------------|
| Saturated fatty acids | 63.3 | 28.0 |
| Butyric C4:0 | 3.2 | - |
| Caproic C6:0 | 2.2 | - |
| Caprylic C8:0 | 1.3 | - |
| Capric C10:0 | 2.5 | 1.0 |
| Lauric C12:0 | 3.3 | 4.0 |
| Myristic C14:0 | 11.8 | 2.0 |
| Palmitic C16:0 | 26.5 | 15.0 |
| Stearic C18:0 | 12.5 | 6.0 |
| Unsaturated fatty acids | 33.1 | 70.0 |
| Palmitoleic C16:1 | 2.3 | 0.5 |
| Oleic C18:1 | 24.5 | 35.0 |
| Linoleic C18:2 (omega 6) | 3.0 | 20.0 |
| Linolenic C18:3 (omega 3) | 0.5 | 14.5 |
| Trans fatty acids | 2.8 | 0 |
| Other fatty acids | 3.6 | 2.0 |

cold temperature (Mortensen, 2011). Margarine on the other hand, is made from vegetable oils like sunflower, soybean and rape seed oil, mixed with other, harder fats (Freeman & Melnikov, 2015). Its principal constituents are unsaturated fatty acids (Arellano, Norton, & Smith, 2015). Variations of fat blend properties in combination with emulsifier mixes and water phase structurants allow a wide range of margarines/ spreads to be produced with various fat contents (low-medium and high fat), textures (hard and soft), desired physical, and nutritional properties (Freeman & Melnikov, 2015) The number and size of crystals in margarine varies with the composition of the fat/oil source(s) and with its processing. Typical levels of fatty acids for milk fat and margarines are shown on Table 1 (Gunstone & Harwood, 2007, O'Brien, 2009). Further information on the properties and composition of different butters and margarines can be found in (Nazir, Moorecroft, & Mishkel, 1976, Fearon, 2003, Vaisey-Genser, 2003).

The composition of the spread blend is designed to deliver good spreadable consistency and mouthfeel, but given the type of oils used, many margarines lack the cooling sensations given by the saturated short chain fatty acids in butter.

Rheology of spread products, water in oil (w/o) emulsions, is mainly governed by the phase volume fraction and droplet size. On the different parameters which could modify the rheological properties and thus sensory perception, the crystal network structure has been the subject of only few studies dealing with spreads. The network structure is characterized by the presence of dispersed aqueous droplets that relate to the three-dimensional crystal network providing a solid character to spreads (Shiota, Isogai, Iwasawa, & Kotera, 2011).The melting characteristics of a spread are essentially those of the fat blend used in its composition, and they are responsible for many of the product's

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sensory characteristics. Desirable mouth melt requires rapid melting at mouth temperature (35–37C) for prompt flavor release and clearance. Rheological properties of margarines and spread products are of critical importance for consumer acceptance and liking.

Sensory evaluation of spreads focuses normally on appearance (e.g., color, gloss), spreadability, mouthfeel (e.g., Firmness, melting), taste (e.g., salty, bitter) and flavor attributes (e.g., buttery, creamy, rancid) (Vaisey-Genser, 2003; Freeman & Melnikov, 2015). Many of these attributes have been correlated to specific analytical measurements such as solid fat content (N-Line), and differential scanning calorimetry DSC for fat melting, Stevens for measuring hardness, brittleness, etc. and salt release as an indirect measurement of breakdown of the spread (Fearon, 2003; Freeman & Melnikov, 2015). Interestingly, the "cool-melting" perception has been mentioned in several review articles and book chapters when describing the properties of margarines and butters (Borwankar et al., 1992, Chrysan, 2005), but so far this attribute has not been taken up as a standard attribute to be evaluated when characterizing spread products.

Most studies describe the "cool-melting" sensation of spreads and potential theories of why it is elicited (Chrysan, 2005, Frede, 2011). Vaisey-Genser mentions that rapid mouth melt of table spreads provide a cooling sensation due to absorption of the heat of crystallization during melting and is accompanied by prompt flavor release (Vaisey-Genser, 2003). The cooling sensation has been estimated by recording the temperature drop in a 35C metal sensing head placed in contact with the product (Lomneth et al., 1983). However in no instance there is mention of a reliable method that could be used to predict this property. Here we focused mainly on characterizing the "cool-melting" property of spreads sensorically and analytically, in an effort to develop tools for *in-vitro* quantification of this phenomenon.

2 | MATERIALS AND METHODS

Ten commercial products were chosen from different European countries (Figure 1) and were classified according to UK legislation. Three butters (unsalted) were selected from different countries to ensure that innate variances within milk composition and processing were covered. Two blends (milk fat + vegetable oil) were included: one high fat blend based on butter and one low fat blend based on margarine that also contained a portion of shea fat. These blends are labelled here as "melanges." Three margarines (80–90% fat) and two low fat spreads (<80% fat). One product, referred as "wrapper" (Block margarine), is meant for cooking and baking and not for spreading on bread. It was included as a negative control as it does not melt at mouth temperature. All products were evaluated before their expiration date.

2.1 Sensory evaluation

The chosen commercial spreadable butters and margarines were given to a trained sensory panel (n = 14) for evaluation of the "cool-melting" property. The panel used is dedicated to spreads evaluation and is familiar with the methodology and terminology used for spreads evaluation. To avoid confusion by the panellists between the new quality "cool-melting" (referring to the specific cooling effect elicited while the

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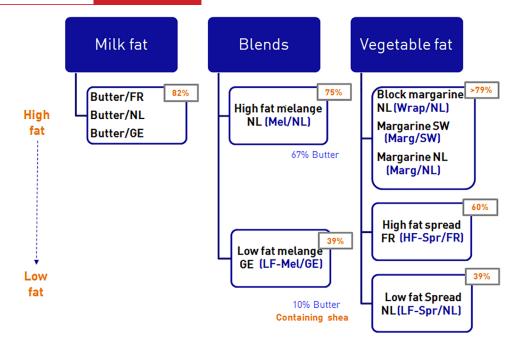


FIGURE 1 Commercial spreads tested for cool melting. Last two letters indicate the country of origin. Classification is based on UK regulation. Blue labels are used to identify the samples further in the study. Numbers in orange indicate the percentage of fat present in the spread

sample is melting inside the mouth) and 'melting' (related to the breakdown + rate of fat melting in mouth), the term "cool-melting" was simplified to the attribute "cooling" for the sensory evaluation, and will be used throughout the rest of the document. Four sessions were necessary for training and evaluation of the spreads.

The evaluation method consisted on a descriptive analysis based on a variant of the Spectrum method (Meilgaard, Civille, & Carr, 2015). The method is known as the UFASM method (Unilever Foods Absolute Scaling Method (SOP 001/01Kooyman, 2005), which similar to the spectrum method uses a 16 point category scale (0-15), however it differs from the spectrum method on that it does not use absolute intensity scales but scales based on the specific product category, in this case panellists are given reference standards that would primarily cover the world of spreads. Having fixed scales enables to compare intensities of attributes relative to each other. The scale intensities are determined based on a wide range of spreads. Panellists also received standard sour solutions and were trained to score the intensity of all attributes according to the sour scale references (water solutions of 0.3, 0.4, 0.6, 0.9, and 1.2 g/L citric acid corresponding to values of 2, 4, 7, 10, and 13 on the scale) (Kooyman, 2005). Panellist rated the attributes "melting" defined as the breakdown + rate of fat melting (slow to fast) and "saltiness" (low to high) for all samples in a sequential monadic test.

A progressive profiling (time intensity) was performed only on the attribute "cooling" defined as the cooling intensity (loss of energy) elicited in mouth during the melting of the spread (from low to high) using the same category scale (0-15) as described before. Ratings were performed at fixed timings from 0 to 80 s (each timing was considered as an individual attribute on the score sheet). At 0 s the panellists

placed the spoon in their mouths, the samples were spit at 60 s and "aftertaste cooling intensity" was rated at 80 s. Panellists rated "cooling" intensity for all samples in duplicate.

A minimum of 2 sessions were necessary for training purposes, where panellists learnt to distinguish the cooling sensation by tasting samples of the baking& cooking spread wrapper/NL which elicits no cooling sensation and samples other spreads and butter as the positive control.

Two dummy samples were given at the start of each session as reminder for low (wrapper NL) and high "cool-melting" (Butter NL). Panellists received 10 spread samples per session each on 2 plastic spoons (2 g per spoon) one spoon to evaluate melting and saltiness and a second spoon to rate cooling. Samples were served at room temperature (~20C, to mimic the temperature of consumption), presented in duplicate, 3-digit coded and in random order. Ratings were recorded using FIZZ® software. Rinsing with lukewarm water and/or jasmingreen tea was performed between sample tasting to normalize mouth temperature, cream crackers were also offered to help cleanse the mouth from any fat residue Panellist were given 5 min break in between samples, and had a break after five samples were tested.

Resulting average curves from the sequential profiling are plotted in Figure 2. Individual curves were further analysed to obtain the area under the curve (AUC), the cooling maximum (Cmax) and the time at which Cmax was achieved (Tmax).

2.2 | In-mouth temperature measurements

In-mouth temperature measurements were executed with an infrared thermometer to determine the actual heating rate in mouth. Samples

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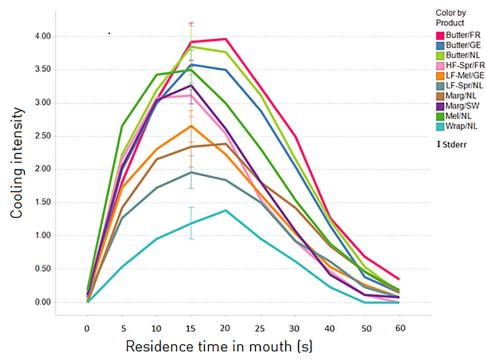


FIGURE 2 Cool melting profile of commercial spreads. Each curve is the average over 26 measurements. For better visualization only one standard error is shown

were tested at fridge temperature making use of a plastic tea spoon and were only taken out of the fridge on the moment of measurement. Sample temperature was initially checked and immediately placed on the tongue, the subject was instructed to make minimal movements with the tongue. In-mouth temperature was measured on the product at fixed points in time (opening the mouth for each measuring point) for 2 minutes, independently of the sample being molten or not.

2.3 | N-lines

N-lines, which quantify solid fat content at different temperatures, were determined for all commercial products using a standard NMR procedure (ISO 8292-1:2008 2008).

N-lines were determined by a minispec mq20 Solid Fat Content Analyser from Bruker, using an automation unit robot by Da Vinci laboratory solutions and an Ecoline RE106 thermostat from Lauda. Samples were molten beforehand at 80C followed by fat separation. The fat phase underwent a stabilization step at 0C, for 1 h. The solid fat content was measured at individual temperatures ranging from 5C to 50C (N-values) with 30 min stabilisation at each temperature before the measurement. Slopes were calculated in relevant temperature regions.

To mimic in-mouth conditions during product consumption, a new method was developed. This new method is referred to as "Direct Nline on product." The products were introduced directly into NMRtubes and measured at three different temperatures without preliminary fat separation or temperature changes. Measurement were done at 5C to simulate refrigeration conditions, 20C to simulate ambient conditions and 35C to resemble in-mouth conditions. Relevant slopes were calculated in different regions and correlated against AUC and Cmax.

2.4 Differential scanning calorimetry (DSC)

DSC is a thermal analysis technique to measure the temperature and heat flows associated with phase transitions in materials as a function of time and temperature (Heussen, 2016). It provides information about the melting enthalpy in J/g. Here we developed an in-mouth simulation method for measuring "cool-melting" properties. Measurements were performed on a Perkin Elmer power compensated DSC8000 equipped with and intracooler III. 20 to 40 mg of samples was measured in a stainless steel sample pan. Measurements were conducted under a Nitrogen atmosphere. The methodology consisted on stabilizing the sample at 5C for 3 min and subsequently heating the sample at a rate of 200C/min from 5C up to 37C, followed by isothermal conditions at 37C for 20 minutes. The heating rate was estimated from the in-mouth temperature measurements described before. Resulting enthalpy curves were plotted against time. Total and partial areas were calculated at different temperatures and slopes were also calculated.

2.5 | Salt release measurements

Salt release was measured based on conductivity in water as a function of temperature using a conductivity module 856 with Tiamo light software from Metrohm, conductivity electrode with PT1000 (Metrohm Y16915130) following a SOP. Two parameters were considered: the 306

TABLE 2 Salt content and % released at 35C per individual spread. Average sensory ratings for saltiness (low-high), melting (slow-fast) and lingering cooling (low-high) measured on a 16 point category scale (n = 26)

| Product | Salt content g/100 g spread declared on label | Salt Release % @ 35C | Saltiness intensity | Melting | Lingering cooling 80 s |
|-----------|---|-------------------------|------------------------|---------|---------------------------|
| Butter/FR | 0.03 | 96.5 | 1.55 | 6.00 | 0.38 |
| Butter/NL | 0.00 | 82.9 | 1.33 | 5.74 | 0.19 |
| Butter/GE | 0.03 | 91.9 | 1.41 | 6.07 | 0.20 |
| Mel/NL | 0.00 | 100 | 2.09 | 10.21 | 0.24 |
| Marg/NL | 0.02 | 0.0 | 1.75 | 5.20 | 0.19 |
| Marg/SW | 0.90 | 1.0 | 6.93 | 9.46 | 0.11 |
| LF-Mel/GE | 0.53 | 100 | 6.66 | 9.73 | 0.12 |
| HF-Spr/FR | 0.32 | 100 | 6.32 | 10.03 | 0.03 |
| LF-Spr/NL | 0.35 | 12.0 | 4.72 | 4.86 | 0.12 |
| Wrap/NL | 1.50 | 0.1 | 1.48 | 2.31 | 0.03 |

temperature at which 100% salt release occurs and percentage of salt release at individual temperatures, such as 35C and 37C.

2.6 | Fame and MDT

Fame is a method to determine the fatty acid composition of edible oils and fats by capillary (high speed) Gas chromatography (Duchateau, van Oosten, & Vasconcellos, 1996). Samples are methylated in-situ with TMSH and subsequently separated over a GC column with medium polar stationary phase. Fatty acids are separated according to chain length and level of unsaturation. Detection is based on FID. The process is performed under a SOP.

MDT is a method to determine the content of free fatty acids, mono, di and triacylglycerides in oils and fats by means of GC. The free OH groups are silylated with BSTFA/Pyridine. Separation takes place on a non-polar stationary phase capillary GC column with FID detection. The method is run under a SOP.

2.7 Data analysis

Repeated measures ANOVA was performed on raw data and on the AUC and Cmax using a Sattherth correction using SAS® 9.4. A posthoc Student Neumann's Keuls (SNK) comparison test was performed on the means using an alpha of 0.05.

3 | RESULTS

3.1 Sensory measurements

Although not a standard quality measured in spreads, the "coolmelting" sensation, later on referred only as "cooling," was well understood by the sensory panel, which was able to capture the differences among spreads. Figure 2 shows the average cooling intensity plotted at individual time points per product obtained from the sequential profiling test (most error bars were left out for better visualisation). At an early stage most products behave similarly, however over the residence

TABLE 3 Comparison of cool melting area under the curve (AUC), Cooling maximum (Cmax) and Time at maximum cooling (Tmax) for commercial spread products. Mean comparison based on SNK posthoc analysis with alpha = 0.05

| Product | N | AUC LSmean | AUC stderr | SNK | Cmax LSmean | Cmax Stderr | SNK | Tmax LSmean | Tmax Stderr | SNK |
|-----------|----|---------------|---------------|-----|----------------|----------------|-----|----------------|----------------|-----|
| Butter/FR | 26 | 135.0 | 3.2 | а | 4.7 | 0.1 | а | 15.9 | 0.4 | а |
| Butter/NL | 26 | 119.5 | 3.2 | ab | 4.6 | 0.1 | а | 15.6 | 0.4 | ab |
| Butter/GE | 26 | 117.7 | 3.2 | b | 4.6 | 0.1 | а | 14.4 | 0.4 | ab |
| Mel/NL | 26 | 113.6 | 3.2 | b | 4.4 | 0.1 | а | 13.8 | 0.4 | ab |
| Marg/NL | 26 | 91.4 | 3.2 | с | 3.9 | 0.1 | b | 14.0 | 0.4 | ab |
| Marg/SW | 26 | 80.7 | 3.2 | cd | 3.8 | 0.1 | b | 13.6 | 0.4 | ab |
| LF-Mel/GE | 26 | 74.8 | 3.2 | de | 3.5 | 0.1 | b | 13.3 | 0.4 | ab |
| HF-Spr/FR | 26 | 74.8 | 3.2 | de | 3.4 | 0.1 | b | 12.5 | 0.4 | ab |
| LF-Spr/NL | 26 | 61.6 | 3.2 | e | 2.4 | 0.1 | с | 12.3 | 0.4 | b |
| Wrap/NL | 26 | 28.2 | 3.2 | f | 1.6 | 0.1 | d | 12.7 | 0.4 | ab |

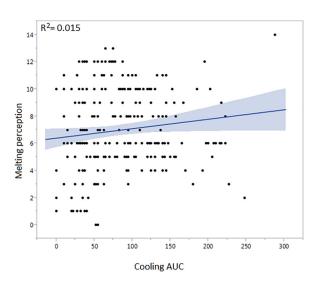


FIGURE 3 Correlation analysis of melting perception versus cooling AUC

time we can see big differences in the cooling perception of the products having clear distinctions among butters, margarines and low fat spreads. The wrapper product (block margarine), which has been designed for cooking and baking and not for spreading, has the lowest cooling effect reason why it was chosen as negative control.

Maximum cooling intensity is reached between 10 s and 20 s, which is also the range where the biggest significant differences are observed among samples. Samples are normally molten between 60 and 80 s. according to the in-mouth temperature measurements (data not shown) and confirmed by the isothermal DSC (Figure 7). By the time the panel spit the sample out (60 s) the majority of the samples are

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already molten. To assess how persistent the cooling perception is, we measured cooling after the sample was spitted out (80 s.). Results show no differences in aftertaste intensity among products (Table 2) and ratings of lingering cooling are very low. The fact that profiles of individual products change and intercross over time (Figure 2), make it difficult to compare products. To correct for these effects, we calculated the area under the curve (AUC), the cooling maximum (Cmax) and the time at which Cmax is reached (Tmax, Table 3), and we used this data to further correlate the sensory perception to the analytical measurements.

Analysis based on AUC or Cmax give a clearer distinction among products (Table 3). Grouping based on statistical differences vary slightly among the 2 parameters but in both cases we can see significant differences between the butters and the margarines tested. Based on cooling maxima, three categories could be established: a high cooling category, an intermediate cooling category, and a low cooling category. These results confirm the original hypothesis that butters differ from margarines in their "coolmelting" perception, having an overall higher cooling. In respect to the third parameter the time to reach maximum cooling, only slight differences were found among the products which Tmax ranged from 12.3 to 15.9.

The melting profile of a spread is a relevant characteristic to predict the "cool-melting," the closer the fat melts to body temperature, the higher the cooling sensation. The "cool-melting" sensation can in a way be defined as the energy spent to melt the fat at 35–37C, the higher the energy spent the higher the cooling perception. As this property is linked to the meltability of the fat, it makes sense that low fat margarines score lower on cooling as water will also serve as a buffer and less energy will be required from our mouth to melt the sample.

Conversely, the sensory attribute referred as "melting," which is an attribute normally described during spreads evaluation, does not

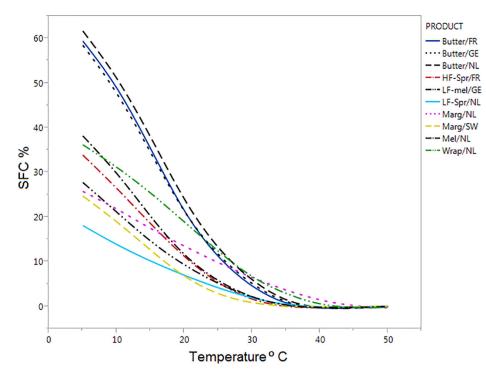


FIGURE 4 N-line curves for commercial spreads- Changes in percentage of solid fat content over a temperature range

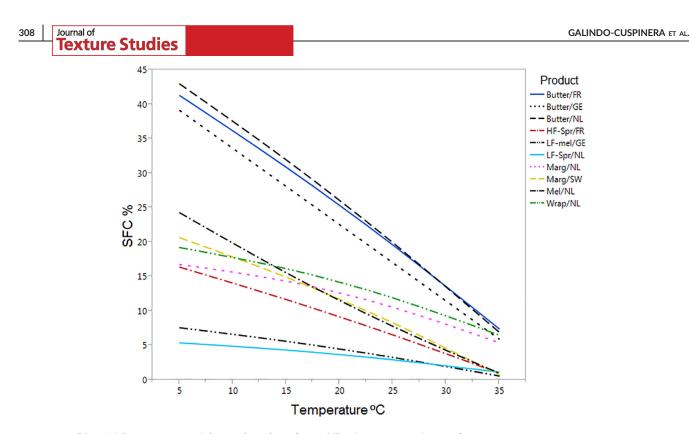


FIGURE 5 Direct N-line on commercial spread products (no stabilization or separation step)

correlate to the attribute cooling as shown in Figure 3. This is due to the way melting is defined by the sensory panel, which actually measures a combination of spread breakdown and rate of melting. The breakdown is related to the inversion of the emulsion from w/o to o/w during in-mouth processing and with the incorporation of saliva, and does not necessarily require extra energy expenditure, which would explain the lack of good correlation with the cooling perception.

Given that commercial samples although labelled as unsalted had still a small amount of salt and the cooking and baking spread as well as the products from Sweden contain a considerable amount of salt we decided to measure the saltiness perception to avoid dumping effects. In parallel, salt release was measured for these samples (Table 2). As expected salt release at 35C correlates well to saltiness perception ($R^2 = 0.936$) however, contrary to what has been shown in other experiments no good correlations were found between salt release and melting perception ($R^2 = 0.275$). We believe this might be due to the low levels of salt present in the majority of samples, and that only few samples are leading the correlation at higher salt levels.

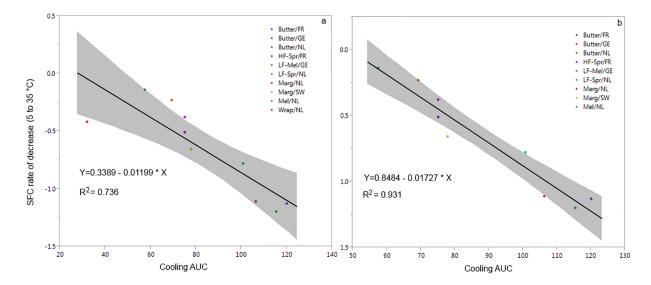


FIGURE 6 Relation between cooling perception (AUC) and the decrease of SFC in commercial spreads. (a) Including the block margarine (wrap/NL) $R^2 = 0.736$, (b) excluding the wrapper from the analysis, $R^2 = 0.931$

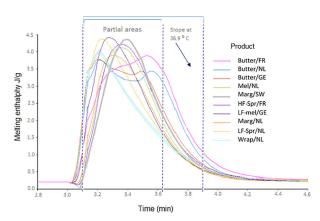


FIGURE 7 Melting enthalpy of commercial spreads using the inmouth simulation method

3.2 | Physical measurements

3.2.1 | N-line

N-line analysis was carried out with the intention of investigating the possible correlation between the rate of decrease in solid fat content around mouth temperature and the cooling perception. The solid fat content (SFC) of a spread at a given temperature is primarily a function of the molecular composition (TAG composition)(Gribnau, 1992) and the polymorph state (dependent on TAG composition and crystallization conditions). Higher solid fat contents relate to higher levels of saturated fatty acids (SAFA) present in the TAG mixture (Bot & Flöter, 2013).

When comparing conventional N-lines to cooling perception (Figure 4), three suggested parameters can be pointed out: (1) the shape of the curve, (2) the amount of solids at 20C, and (3) the amount of solids at body temperature (between 30 and 35C)

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Curve shape: Samples that show higher cooling present an inverted S shaped melting curve. A steep melting curve contrary to a flat one means that the solid fat content decreases more rapidly than the other samples in a minor temperature range giving rise to the cool sensation in mouth.

Solid content: At room temperature (20C), there are significantly more solids present for butters and wrapper NL, than for the other samples. More crystals present in the system means more energy required for the transition from crystal to liquid fat. Low fat product on the other hand contain more water, which act as a heat sink, therefore less energy is spent for melting and hence less cooling. Around body temperature, between 30C and 35C (mouthfeel), steeper slopes suggest more rapid mouth melt (Vaisey-Genser, 2003), which would contribute to cooling perception. The rate of decrease is more accentuated for butters (steeper slope) in this temperature rate.

Weak correlations ($\sim R^2 = 0.24$) were found between the calculated N-line parameter (N-values and all calculated slopes) and AUC or Cmax (not shown). Therefore, the conventional N-line method cannot be taken as a straightforward predictor of "cool-melting," though some visual and calculated parameters might give a good hint. Melting characteristics of fats are history dependent i.e. depending on the way in which the fat has been crystalized. Melting and recrystallization can lead to formation of different crystal forms and consequently different melting behaviour of the mixture compared the original product (Borwankar et al., 1992). This is important to consider when looking at N-line in relation to sensory data as consumers do not ingest reconstituted fat, but a final product. Since the total time-temperature profile of a product determines its actual crystalline state, measurements of N-lines directly on products should provide more relevant data.

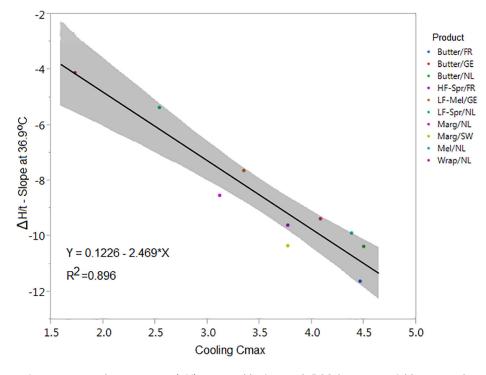


FIGURE 8 Cooling maximum vs. rate of energy spent (ΔH) measured by in-mouth DSC for commercial butters and margarines

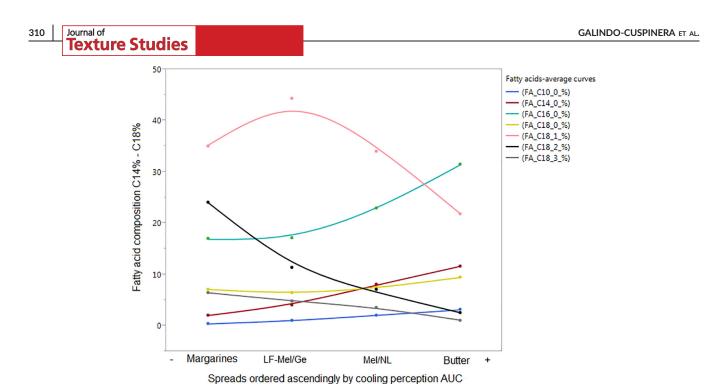


FIGURE 9 Average fatty acid content (%) by spread type (FAME). Cooling intensity increases from left (low) to right (high). The margarine group include all vegetable spreads, LF-Mel/GE and Mel/NL refer to the blends containing low and high butter content

Figure 5 shows the curves obtained from the Direct N-line method. The solid fat percentages are in relation to the total product (water included) and not to the total fat content as in the conventional method. Slopes were determined as the rate of decrease of solid fat content and can be linked to energy expenditure.

In terms of shape, the butter group have clearly the highest SFC from all samples, showing a steeper decrease, with remaining solids at 35C. The best correlation is found between the cooling perception

(AUC) and the slope of the curve between 5 and 35C derived from the Direct N-line method (R^2 = 0.736). The wrapper NL is shown as an outlier in this graph, shifting the curve downwards (Figure 6a). This margarine as mentioned before, is not meant to be spreadable and therefore its structure and composition is very different from the other products. For this experiment, it was only used as a negative cooling control. If left out from the analysis the correlation coefficient would increase to 0.931 (Figure 6b). However the fact that it is showing as an

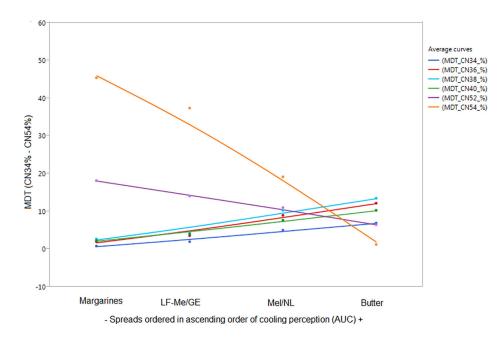


FIGURE 10 Mono, di, and triglyceride content (MDT) vs spread type. The margarine group include all vegetable spreads, LF-Mel/GE and Mel/NL refer to the blends containing low and high butter content

outlier, indicates that this method might not be as robust to predict the "cool-melting" of products that contain higher concentration of fats with melting points above 37C. Further tests would need to be performed to assess predictability for these products.

Advantages of the Direct N-line on product over the conventional method include: (1) The actual product is measured instead of the reconstituted fat phase; (2) The product is measured in its stable polymorph crystal form, which is also the structure that is evaluated by the consumer; (3) It is less time consuming; (4) Shows better correlations with cooling perception, compared to the conventional Nline method.

3.3 | In-mouth DSC simulation method

Curves obtained with the in-mouth DSC method, are plotted in Figure 7. Samples take an average of 15.3 s to reach 35C and 36.6 s to reach the temperature of 36.9C. Butters are positioned on the right side of the plot covering a wide region and showing 2 peaks, the second appearing at around mouth temperature. All the other samples exhibit one single peak. Correlations were calculated using different parameters obtained from the DSC curves including partial areas (5–35C, 20–35C, 5–37C) and slopes. Slopes were calculated when the sample reached 36.9C (indicated in the graph) and were used to compare the energy expenditure among samples.

Data evaluation show good correlations among partial areas and cooling AUC or Cmax ($R^2 = 0.73$ and $R^2 = 0.78$), however, the best correlation is achieved when plotting the Cooling maximum vs. the rate of energy spent to melt the spread (Figure 8; Δ H/t at 36.9C, $R^2 = 0.896$). Slope values are associated with the energy used for melting a specific amount of spread sample in grams, per time interval. The higher the slope the higher the energy that is required for one gram of sample to go through the phase transition from solid to liquid in the same time range, therefore when the rate of change is faster, the cooling sensation is higher. Note that the Wrapper reference is fitting well, while it is normally an outlier in other methods applied. What this method implies is that products that require more energy for melting around body-temperature will have a higher cooling perception in mouth.

3.3 | Fat composition and "cool-melting"

The amount of crystalline fat (SFC) is related to the level of saturated fatty acids (SAFA) in the specific TAG mixture. Products were analysed for FAME & MDT to collect information on their TAG (triacyl-glyceride) mixture composition. FAME delivers information on fatty acid composition while MDT gives information about the composition in terms of mono-, di- and triglycerides based on carbon number count. Given the small number of samples tested in this study, we only describe the trend but no strong conclusions can be made based only on the fat composition. We can clearly see a trend of the type of fatty acids and esterification level between types of spreads and the cooling perception (Figures 9 & 10). In general the content of C18:1 and C18:2 fatty acids decrease when increasing the milk fat content (44.3 to 21.8% for

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C18:1 and 24.1 to 2.6% for C18:2) and is linked to an increase of cooling perception, while C16:0 and C14:0 fatty acid content increase with addition of milk fat (17 to 31.5% for C16:0 and 2.0 to 11.6% for C14:0). Although the full FA profile changes depending on the type of spread these peaks are the most prominent and represent a high percentage of the fatty acids present in the spreads tested. This information suggests that by increasing the proportion of lower chain fatty acids C14-C16 and/or decreasing the proportion of C18, we might increase the "cool-melting" perception in spreads. Further research is needed to corroborate this statement.

In the case of MDT (Figure 10), the analysis show a sharp decrease on CN 54 with the increase in milk fat content (from 45.4% in margarine to 1.23% average in butters). On the other hand CN36 (1.8 to12.2%), CN38 (2.6 to 13.4%) and CN40 (2.2 to 10.3%) increase steadily with the increase in milk fat content. Similar to the fatty acid content, every mix show a different profile, but these peaks are the most prominent. Here again we can see that by modifying the composition of the different acylglycerides we might be able to change the "cool-melting" perception elicited by the spread.

4 | CONCLUSIONS

"Cool-melting" is a property of spreads that can be measured sensorically and can discriminate between butters and margarines. The cooling sensation is not directly related to the total amount of fat in the product but to the type of fats present and to their melting properties, having butters the highest rating on cooling followed by high fat margarines and blends (melanges) and lastly low-fat spreads.

The rate of energy expenditure for melting as measured by the inmouth DSC simulation method is the best measurement to predict the "cool-melting" property of a spread.

The rate of decrease of the solid fat content in the direct product (Nline-direct product method) is another way to predict the "coolmelting" property of a spread, however care should be taken when using it as it might underperform when predicting samples with low cooling perception as seen with the Wrapper/NL, compared to the inmouth DSC simulation method, which did not show outliers.

We have shown that it is possible to create an in-vitro methodology based on the modified DSC or N-line methods to predict the "cool-melting" of spread products, which can help direct future product development. Further studies are required to assess the impact that "cool-melting" has on the general liking of spreads.

ETHICAL STATEMENTS

Conflict of Interest: The authors declare that they do not have any conflict of interest.

Ethical Review: This study was approved by the Unilever-Independent ethical committee (U-IEC).

Informed Consent: Written informed consent was obtained from all study participants.

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