Effects of Inhomogeneous Spatial Distribution of Aroma Compounds on Perceived Aroma Intensity and Human Eating Behavior for Neutral pH Gels

Satomi NAKAO, Sayaka ISHIHARA, Makoto NAKAUMA and Takahiro FUNAMI*

San-Ei Gen F.F.I., Inc., 1-1-11 Sanwa-cho, Toyonaka, Osaka 561-8588, Japan

Received February 12, 2013; Accepted April 7, 2013

Effects of inhomogeneous spatial distribution of aroma compounds were investigated on the perceived aroma intensity and human eating behavior using polysaccharide gels as a food model. Gels tested were structured using gel-in-gel configuration to create different degrees of inhomogeneous spatial aroma distribution, and their pH values were set at neutral (6.7 – 6.8). There were no differences in mechanical properties between the structured gels. A greater degree of inhomogeneous spatial aroma distribution increased the perceived aroma intensity, increased the duration of oral processing and suprahyoid musculature activity during oral processing, decreased the particle size of the bolus, and increased the saliva content in the bolus. Effects of pH were compared with our previous report using the same experimental design but different pH (approximately 4.2), confirming a similar trend in the results. A strategy of food structure design for enhanced perceived aroma intensity and human eating behavior was suggested regardless of gel pH.

Keywords: inhomogeneous spatial distribution, aroma, gels, perceived intensity, muscle activity, saliva, gel pH.

Introduction

Flavor, composed of aroma and taste, is one of the most important elements for food palatability, and methods to increase flavor intensity depend on a strategy of food structure design. It has been reported that the perceived intensity of sweetness is increased by inhomogeneous spatial distribution of sugar in layered structure gels mainly of gelatin (Holm *et al.*, 2009; Mosca *et al.*, 2010, 2013.). Also, increase in the perceived intensity of saltiness by inhomogeneous spatial distribution of salt has been reported for bread (Noort *et al.*, 2010, 2012.) and sausages (Mosca *et al.*, 2013.) using either a layer or encapsulation technique. These can be explained by the prevention of adaptation or of gradual decrease in the receptor response, either of which was caused by a continuous and prolonged exposure to taste stimuli, particularly at high doses (Meiselman, 1972). In relation to these, pulsatile stimulation increases perceived intensity of sweetness (Bursseg *et al.*, 2010a, b) and saltiness (Busch *et al.*, 2009, Meiselman and Halpern, 1973) in the case of liquids. As can be seen, most of the previous studies have focused on tastants

rather than aroma. Since perceived flavor intensity is known to be modulated by a cross-modal interaction between aroma and taste (Tournier *et al.*, 2007), similar effects of aroma on flavor perception should be investigated further in relation to food structure.

The release profile of aroma from foods depends on human eating behavior, particularly during chewing for size reduction, although large individual differences can be observed (Hodgson *et al.*, 2003; Mestres *et al.*, 2005; Pionnier *et al.*, 2004; Tarrega *et al.*, 2011). Chewing affects the nasal airflow, and the pulses of air pumped from the mouth with each chew affect aroma release through the retronasal pathway (Hodgson *et al.*, 2003) because the air serves as a carrier for the transport of aroma to the olfactory epithelium (Pionnier *et al.*, 2004). The aroma release profile is also associated with the perceived aroma intensity (Baek *et al.*, 1999). This suggests that human eating behavior should affect perceived aroma intensity through changes in the release profile and vice versa.

With this background, we previously reported (Nakao *et al.*, in press), using food gels, how inhomogeneous spatial aroma distribution affects the perceived aroma intensity and human eating behavior, represented, for example, by muscle

*To whom correspondence should be addressed.

E-mail: tfunami@saneigenffi.co.jp

activity during oral processing. In our previous study, gels tested were from one polysaccharide mixture and were structured using gel-in-gel configuration, in which small gel cubes were dispersed in the matrix gels at a pH of approximately 4.2. Different degrees of inhomogeneous spatial aroma distribution, without changing mechanical properties, were achieved among the gel samples by balancing the aroma concentration in the dispersed gels and that in the matrix gels. Results showed that a greater degree of inhomogeneous spatial aroma distribution increased the perceived aroma intensity and also increased the duration of oral processing and suprahyoid musculature activity during oral processing. On the other hand, it decreased the particle size of the expectorated bolus just before swallowing and increased the saliva content in the bolus.

For gels of low pH or acidified gels, the sour taste contributes to the enhanced perceived aroma intensity (Tournier *et al.*, 2007) and promotes saliva secretion during eating (Liem *et al.*, 2004). Results from our previous study may involve synergistic multi-modal effects between aroma and taste, and the effects of aroma itself cannot be determined exclusively. Also, food products distributed in the market have a range of pH, depending on flavor, and for wide industrial usage of this food structure design, investigations under different pH conditions are necessary to confirm the validity and robustness of results from our previous study. In this

study, therefore, gels of the same experimental design as our previous study, but without added acidulant, were used as test samples. The purpose of this study is to investigate the effects of inhomogeneous spatial aroma distribution on the perceived aroma intensity and human eating behavior using neutral pH gels.

Materials and Methods

Materials As texturizing agents, KELCOGEL® (deacetylated gellan gum) and CARRAGEENAN CS-599 (iota-carrageenan) were used for gelation, and SAN ARTIST® PG (microbial fibrous cellulose) was used for dispersion, as in our previous study (Nakao *et al.*, in press). As a food color, SAN-RED® 3743-EM (a red cabbage extract) was used to dye gels for visualization when particle size of the expectorated bolus was measured. As aroma compounds, Apple Base 3087FA was used because of the absence of variation in preference among subjects. These food ingredients were all commercially available (San-Ei Gen F.F.I., Inc., Osaka, Japan). Other ingredients, including granulated sugar and calcium lactate pentahydrate (for gelling promotion), were of food grade with > 99% purity. Concentration represents w/w% unless otherwise specified.

Gel sample preparation Formulations of gel samples are described in Table 1. As in our previous study (Nakao *et al.*, in press), six gel samples having different structures and/

Table 1. Formulation of gel samples

(a) Structured gels								
	Concentration (w/w%)							
	Dispersed gels				Matrix gels			
	S _{NO}	S _{LOW}	S _{HIGH} Fr1	S _{HIGH} Fr2	S _{NO}	S _{LOW}	S _{HIGH}	
Granulated sugar	5.00	5.00	5.00	5.00	5.00	5.00	5.00	
SAN ARTIST® PG	0.45	0.45	0.45	0.45	0.45	0.45	0.45	
KELCOGEL®	0.15	0.15	0.15	0.15	0.15	0.15	0.15	
CARRAGEENAN CS-599	0.10	0.10	0.10	0.10	0.10	0.10	0.10	
Calcium lactate pentahydrate	0.10	0.10	0.10	0.10	0.10	0.10	0.10	
Aroma compounds	0.30	0.60	1.50	NA	0.30	NA	NA	
De-ionized water	93.90	93.60	92.70	94.20	93.90	94.20	94.20	
(b) Non-structured gels								
	Concentration (w/w%)							
	N ₀	N _{0.1}	N _{0.3}					
Granulated sugar	5.00	5.00	5.00					
SAN ARTIST® PG	0.45	0.45	0.45					
KELCOGEL®	0.15	0.15	0.15					
CARRAGEENAN CS-599	0.10	0.10	0.10					
Calcium lactate pentahydrate	0.10	0.10	0.10					
Aroma compounds	NA	0.10	0.30					
De-ionized water	94.20	94.10	93.90					

NA: not applicable

or different aroma distributions were prepared; three samples from structured gels, termed S, and three samples from non-structured gels, termed N. Structured gels contained small cube gels (6 mm $\times$ 3), dispersed at 50% weight ratio in the matrix gels (cylindrical shape of 25 mm in diameter and 15 mm in height). The composition of texturizing agents was the same between the dispersed and matrix gels. For structured gels, aroma concentration in either the dispersed gels or the matrix gels was arranged in three steps of concentration gradient, although the whole aroma concentration in one gel sample was constant at 0.3%. These were S_{NO} : no concentration-gradient of aroma, in which both the dispersed gels and the matrix gels contained 0.3% aroma compounds, S_{LOW} : the smaller concentration-gradient of aroma, in which all dispersed gels contained 0.6% aroma compounds, and S_{HIGH} : the larger concentration-gradient of aroma, in which 40% of the dispersed gels contained 1.5% aroma compounds (S_{HIGH} Fr1) and the remaining 60% of the dispersed gels did not contain aroma compounds (S_{HIGH} Fr2) (Fig. 1).

Preparation of gel samples was in accordance with our previous study (Nakao *et al.*, in press) except that acidulant was not added in the present study. Measured pH values were 6.8 for S_{NO} , 6.7 for S_{LOW} , and 6.7 for S_{HIGH} .

Differences in the aroma concentration between the dispersed gels (only those containing aroma) and the matrix gels and its constancy were confirmed instrumentally for each gel sample using hexyl acetate as a benchmark, because hexyl acetate is one of the dominant chemicals in the aroma compounds used. The differences were 12.0 (as a relative peak area to that for the internal standard, 3-heptanol) for S_{NO} , 110.7 for S_{LOW} , and 470.9 for S_{HIGH} , where the extraction using dichloromethane as a solvent from those two gel constituents was subjected to GC-MS analyses. For comparison, non-structured gels (i.e. bulk gels) of 0%, 0.1%, and 0.3% aroma compounds, termed N_0 , $N_{0.1}$, and $N_{0.3}$, were prepared

without a concentration gradient. Measured pH values were 7.0 for N_0 , 6.9 for $N_{0.1}$, and 6.8 for $N_{0.3}$.

Mechanical properties Measurement procedure was in accordance with our previous study (Nakao *et al.*, in press), and determined the fracture load and the fracture strain of each gel sample by compressing the entire surface area using an aluminum flat plunger, 100 mm in diameter, at a cross-head speed of 1mm/s. All measurements were carried out at 20°C. Data are presented as means $\pm$ SD of triplicate determinations.

Sensory evaluation Three females and three males, ranging from 26 to 40 years of age (average age 32.0 years), participated as subjects upon their informed consent, and their ability to recognize differences in aroma intensity was confirmed preliminarily. All subjects were healthy and free from chewing and swallowing problems. These subjects were the same as in our previous study (Nakao *et al.*, in press) and had already experienced an identical session. The evaluation procedure was in accordance with our previous study (Nakao *et al.*, in press), and required subjects to process each gel sample by tongue-palate compression for size reduction (i.e. palatal reduction). Subjects evaluated perceived aroma intensity using a visual analogue scale (VAS) method by marking a score of each gel sample on a 100 mm length scale. The length on the scale corresponds directly to the score. The larger the length, the greater the perceived intensity. Data are presented as means $\pm$ SD of the six subjects in duplicate.

Electromyography (EMG) The same subjects as in the sensory evaluation participated in this test. As mentioned, these subjects were the same as in our previous study (Nakao *et al.*, in press) and had already experienced an identical session. The evaluation procedure was in accordance with our previous study (Nakao *et al.*, in press), and recorded the EMG activities from either the left or right masseter muscle and from the suprahyoid musculature. The EMG signal from

Fig. 1. Schematic drawings of structured gel samples.

S_{NO} : No concentration-gradient of aroma, in which both the dispersed gels and the matrix gels contained 0.3% aroma compounds; S_{LOW} : the smaller concentration-gradient of aroma, in which all dispersed gels contained 0.6% aroma compounds (0.3% in total); S_{HIGH} : the larger concentration-gradient of aroma, in which 40% of the dispersed gels contained 1.5% aroma compounds (S_{HIGH} Fr1) and the remaining 60% of the dispersed gels did not contain aroma compounds (S_{HIGH} Fr2) (0.3% in total).

the masseter muscle was not apparent, while the EMG signal from the suprahyoid musculature was detected continuously during oral processing (data not shown). Therefore, the duration of oral processing and the activity of the suprahyoid musculature during oral processing were determined. Each EMG variable was standardized with the corresponding datum for non-structured gel of 0% aroma (N_0) for minimization of differences in individual oral physiology and in electrode placement among subjects. Data are presented as means $\pm$ SD of the six subjects in duplicate.

Particle size in the expectorated bolus before swallowing The same subjects as in the sensory evaluation participated in this test. As mentioned, these subjects were the same as in our previous study (Nakao *et al.*, in press) and had already experienced an identical session. The evaluation procedure was in accordance with our previous study (Nakao *et al.*, in press), and required subjects to expectorate a bolus just before swallowing, after palatal reduction using structured gels only. The number of gel particles and area of each gel particle (presented by the two-dimensional area on a filter paper) were identified. Also, coefficients of skewness and kurtosis of the particle size distribution were determined. Skewness and kurtosis represent the degree of asymmetry and sharpness of the distribution, respectively. Data are presented as means $\pm$ SD of the six subjects in duplicate.

Saliva content in the expectorated bolus The same subjects as in the sensory evaluation participated in this test. As mentioned, these subjects were the same as in our previous study (Nakao *et al.*, in press) and had already experienced an identical session. The evaluation procedure was in accordance with our previous study (Nakao *et al.*, in press), and determined the sugar content in the expectorated bolus to reveal the weight of gel particles in the bolus (according to sugar content in the formulation of each gel sample). Saliva content was thus determined by the difference in the weights between the bolus and gel particles. Data are presented as means $\pm$ SD of the six subjects in duplicate.

Statistics Data were examined by one-way analysis of variance (ANOVA), followed by the Bonferroni test. Significance was defined at $p < 0.05$ to confirm the difference in the mean values between gel samples using JSTAT software.

Results

Mechanical properties For structured gels (S_{NO} , S_{LOW} , and S_{HIGH}), no differences were found between gel samples in the fracture load, ranging from 1.51 to 1.55 N, or in the fracture strain, ranging from 16.7% to 17.0% (Table 2). For non-structured gels (N_0 , $N_{0.1}$, and $N_{0.3}$), no differences were found between gel samples in the fracture load, ranging from 4.72 to 4.98 N, or in the fracture strain, ranging from 23.1% to

Table 2. Mechanical properties of gel samples by compression tests

Sample		Fracture load (N)	Fracture strain (%)
Structured gels	S_{NO}	1.53 ± 0.29	16.8 ± 1.26
	S_{LOW}	1.55 ± 0.29	16.7 ± 1.22
	S_{HIGH}	1.51 ± 0.28	17.0 ± 0.63
Non-structured gels	N_0	4.85 ± 0.25	23.1 ± 0.42
	$N_{0.1}$	4.98 ± 0.24	23.3 ± 0.86
	$N_{0.3}$	4.72 ± 0.22	23.3 ± 0.91

Data are represented as means $\pm$ SD of triplicate determinations. The entire surface area of gels was compressed uniaxially using an aluminum flat plunger, 100 mm in diameter, at a crosshead speed of 1 mm/s. Gels were molded into a cylindrical shape of 25 mm in diameter and 15 mm in height.

23.3% (Table 2). The fracture load and the fracture strain for structured gels were both lower than the corresponding data for non-structured gels, even if the same texturizing agents were used at the same concentration. It was difficult to prepare structured gels of equivalent fracture load and fracture strain to non-structured gels using the same gelling agent (without changing the composition).

Sensory evaluation For structured gels (S_{NO} , S_{LOW} , and S_{HIGH}), differences were found between gel samples in the VAS score of the perceived aroma intensity, increasing from 50.3 to 78.1 with increasing degree of inhomogeneous spatial distribution of aroma compounds, although the overall aroma concentration was identical (Fig. 2a). For non-structured gels (N_0 , $N_{0.1}$, and $N_{0.3}$), differences were found between gel samples in the same VAS score, increasing from 6.8 to 46.6 with increasing overall aroma concentration (Fig. 2b). Scores were comparable between S_{NO} and $N_{0.3}$.

EMG For structured gels (S_{NO} , S_{LOW} , and S_{HIGH}), each EMG variable tended to increase with increasing degree of inhomogeneous spatial distribution of aroma compounds, and differences were found between S_{HIGH} and S_{NO} or S_{LOW} for the duration of oral processing and between S_{NO} and S_{HIGH} for suprahyoid EMG activity (Fig. 3a and c). For non-structured gels (N_0 , $N_{0.1}$, and $N_{0.3}$), no differences were found between gel samples in each EMG variable (Fig. 3b and d).

Particle size in the expectorated bolus before swallowing For structured gels (S_{NO} , S_{LOW} , and S_{HIGH}), mean particle size tended to decrease with increasing degree of inhomogeneous spatial distribution of aroma compounds, and differences were found between S_{HIGH} and S_{NO} or S_{LOW} (Fig. 4a). Also, the number of gel particles in the expectorated bolus tended to increase with increasing degree of inhomogeneous spatial distribution of aroma compounds, and differences were found between S_{HIGH} and S_{NO} or S_{LOW} (Fig. 4b). On the other hand, no differences were found between gel samples in co-

Fig. 2. Sensory scores for perceived aroma intensity.

(a) Structured gels; (b) non-structured gels. Six subjects with an average age of 32.0 years participated. Data are presented as means $\pm$ SD of the six subjects in duplicate. Data with an asterisk are significantly different ($p < 0.05$).

Fig. 3. EMG variables during oral processing.

(a) and (b): Duration of oral processing; (c) and (d): suprahyoid musculature activity. (a) and (c): structured gels; (b) and (d): non-structured gels. The same subjects as in the sensory evaluation participated. Each EMG variable was standardized with the corresponding datum for non-structured gels at 0% aroma N₀. Data are presented as means $\pm$ SD of the six subjects in duplicate. Data with an asterisk are significantly different ($p < 0.05$).

Fig. 4. Geometric properties of the expectorated bolus before swallowing.

(a) Mean particle size ; (b) the number of particles; (c) coefficient of skewness; (d) coefficient of kurtosis. The same subjects as in the sensory evaluation participated. Skewness and kurtosis represent the degree of asymmetry and sharpness of the distribution, respectively. Data are presented as means $\pm$ SD of the six subjects in duplicate. Data with an asterisk are significantly different ($p < 0.05$).

efficient of skewness or coefficient of kurtosis with respect to particle size distribution (Fig. 4c and d).

Saliva content in the expectorated bolus For structured gels (S_{NO}, S_{LOW}, and S_{HIGH}), saliva content in the expectorated bolus tended to increase with increasing degree of inhomogeneous spatial distribution of aroma compounds, and a difference was found between S_{NO} and S_{HIGH} (Fig. 5).

Discussion

Mechanical properties Lower fracture load and strain for structured gels in comparison with the corresponding data for non-structured gels can be explained by the function of the dispersed gels in lowering cohesiveness or internal binding of the matrix gels, giving rise to a stress concentration upon compression and making the matrix gels easier to collapse.

Sensory evaluation For structured gels, the degree of inhomogeneous spatial distribution of aroma compounds

Fig. 5. Saliva content in the expectorated bolus before swallowing.

The same subjects as in the sensory evaluation participated. Data are presented as means $\pm$ SD of the six subjects in duplicate. Data with an asterisk are significantly different ($p < 0.05$).

should dominate the increase in the perceived aroma intensity since the factor of textural difference is excluded from mechanical measurements. From a physiological point of view, this result can be attributed to the prevention of adaptation or of the gradual decrease in receptor response. Under continuous stimulation, adaptation will cause the response threshold of the cell to rise to the level of the stimulus, thus decreasing the firing rate of the receptor cells (Moore, 1994). This is applicable to the results of the present study. Continuous stimulation from homogeneous spatial aroma distribution increases the response threshold of receptor cells, thus lowering the perceived aroma intensity as a result of adaptation. In contrast, discontinuous stimulation from inhomogeneous spatial aroma distribution causes a stimulation cycle, where the response threshold of receptor cells can return to baseline or can become even lower than baseline during oral processing. This is due to the concentration gradient of aroma compounds in the structured gels. A greater degree of inhomogeneous spatial aroma distribution gives rise to a larger stimulus gap, occurring in a discontinuous manner during oral processing, and this should result in higher perceived aroma intensity. No difference in the perceived aroma intensity between S_{NO} and $N_{0.3}$ in spite of different fracture characteristics suggests that the impact of aroma distribution should be much greater than the mechanical properties of gels on the perceived aroma intensity in the present study.

Results from the present study are similar to those from our previous study (Nakao *et al.*, in press), and indicated that the effects of inhomogeneous spatial aroma distribution on the perceived aroma intensity do not depend on the pH condition of structured gels. Perceived aroma intensity is greater at the lower pH when compared at the same degree of inhomogeneous spatial aroma distribution. This is the most apparent for S_{HIGH} (i.e. at the largest degree of inhomogeneous spatial aroma distribution), where VAS scores from the previous study (i.e. at the lower pH) and from the present study are 88.0 and 78.1, respectively. This can be explained by the cross-modal interaction between taste (sourness) and aroma (Tournier *et al.*, 2007).

EMG As seen in non-structured gels, no changes in the duration of oral processing or in suprahyoid activity during oral processing upon increase in aroma concentration is in accordance with a previous report, which described that human eating behavior is not affected by aroma concentration (Sprunt *et al.*, 2002), suggesting that non-structured gels lacking a concentration gradient of aroma compounds should not alter human eating behavior. In the case of structured gels, inhomogeneous spatial distribution of aroma compounds alters human eating behavior even though the overall aroma concentration is identical, which is quite different

from non-structured gels. The chewing process should result in the formation of a homogeneous bolus (Mosca *et al.*, 2010), and this is true of palatal reduction. Both mechanical and chemical aspects are involved in bolus homogeneity. For structured gels, aroma can be released in a pulsative manner from the dispersed gels upon compression. It is thus speculated that the subjects should try more palatal reduction to form a chemically homogeneous bolus in terms of aroma concentration. This can be a dominant reason for the longer duration of oral processing caused by the greater degree of inhomogeneous spatial aroma distribution. On the other hand, a longer duration of oral processing does not represent greater effort for the formation of a mechanically homogeneous bolus, because mechanical properties are not different between structured gels.

Results from the present study are similar to those from our previous study (Nakao *et al.*, in press), and indicated that the effects of inhomogeneous spatial aroma distribution on human eating behavior do not depend on the pH condition of structured gels. The duration of oral processing and suprahyoid musculature activity during oral processing are smaller at the lower pH when compared at the same degree of inhomogeneous spatial aroma distribution. This is the most apparent for S_{HIGH} , where durations of oral processing from the previous study (i.e. at the lower pH) and from the present study are 12.2 s and 14.3 s, respectively. Also, suprahyoid musculature activities during oral processing from the previous study (i.e. at the lower pH) and from the present study are $0.27 \text{ mV} \times \text{s}$ and $0.36 \text{ mV} \times \text{s}$, respectively. These can be explained by the enhancement of initial swallowing by acid or sourness (Logemann *et al.*, 1995), shortening the duration and lowering coincident muscle activity or eating effort.

As mentioned, human eating behavior relates to the release profile of aroma through the retronasal pathway (Hodgson *et al.*, 2003; Mestres *et al.*, 2005; Pionnier *et al.*, 2004; Tarrega *et al.*, 2011), which is applicable not only to chewing but also to palatal reduction as an oral strategy for size reduction. For structured gels, the total amount of aroma release through the retronasal pathway increases and/or the opportunity to perceive the dynamic change in aroma intensity during oral processing increases by inhomogeneous spatial aroma distribution due to the prolonged duration of oral processing. It is hypothesized that the increase in the perceived aroma intensity can be attributed to the discontinuous stimuli and higher release rate of aroma compounds, and this should be confirmed in the future. A previous report (Bylaite *et al.*, 2004) indicates that the non-equilibrium release rate of volatile aroma compounds has a greater influence on human perceived intensity than an equilibrium saturated concentration, which supports the hypothesis.

Also, increases in the duration of oral processing and in the muscle activity during oral processing may increase the feeling of satiety, playing a role in obesity prevention from both a physiological and a psychological point of view, as indicated by previous reports (Hendrik *et al.*, 2011; Maruyama *et al.*, 2008; Otsuka *et al.*, 2006). Intentional increases in chewing frequency during eating and in eating time should be a measure for satiety enhancement, thus leading to the prevention of obesity (Hendrik *et al.*, 2011; Maruyama *et al.*, 2008; Otsuka *et al.*, 2006). These previous reports may support the hypothesis of health benefits of structured gels with inhomogeneous spatial aroma distribution, even though the gels are processed by palatal reduction.

Particle size of the expectorated bolus before swallowing

For structured gels, no changes in the particle size distribution, represented by coefficients of skewness and kurtosis, suggest that the mode of palatal reduction should not be changed substantially by the degree of inhomogeneous spatial aroma distribution. A greater degree of size reduction relates to longer duration of oral processing and larger suprahyoid activity during oral processing on the EMG recording, presenting greater efforts by subjects to average the aroma concentration in the bolus, as previously mentioned. Increase in the perceived aroma intensity with increasing degree of inhomogeneous spatial aroma distribution can also be associated with smaller particle size of the bolus after palatal reduction. This is supported by a previous report (Arvisenet *et al.*, 2006) on aroma release from reduced apples of different average particle sizes, using a model mouth system, showing that the smallest particles allow the release of the highest amount of volatile compounds. Aroma release occurs at the surface of the sample, and the larger surface area available for diffusion (or mass transfer) due to a smaller particle size results in greater aroma release (Harrison *et al.*, 1998).

Results from the present study are similar to those from our previous study (Nakao *et al.*, in press), and indicated that the effects of inhomogeneous spatial aroma distribution on the bolus particles do not depend on the pH condition of structured gels. Particle size, represented by its mean area, is larger and the number of particles is smaller at the lower pH when compared at the same degree of inhomogeneous spatial aroma distribution. This is the most apparent for S_{HIGH} , where the particle sizes from the previous study (i.e. at the lower pH) and from the present study are approximately 1.5 mm² and 1.2 mm², respectively. Also, particle numbers from the previous study and the present study are approximately 2400 and 3300, respectively. These can be explained by the difference in the duration of oral processing.

Saliva content in the expectorated bolus Since saliva secretion is increased by human mastication (Humphrey and

Williamson, 2001), prolonged duration of oral processing is responsible for increased saliva content. Inhomogeneous spatial aroma distribution should be one of the strategies to enhance saliva secretion.

Results from the present study are similar to those from our previous study (Nakao *et al.*, in press), and indicated that the effects of inhomogeneous spatial aroma distribution on saliva secretion do not depend on the pH condition of structured gels. Saliva content in the bolus is larger at the lower pH when compared at the same degree of inhomogeneous spatial aroma distribution. This is the most apparent for S_{HIGH} , where the saliva contents from the previous study (i.e. at the lower pH) and from the present study are approximately 2.8 g and 2.4 g, respectively. This can be explained by the enhancement of saliva secretion by acid or sourness (Liem *et al.*, 2004), overcoming the effects on shortening of the duration of oral processing.

Conclusion

Inhomogeneous spatial aroma distribution enhances the perceived aroma intensity and human eating behavior for gels processed by palatal reduction. These findings are similar to those from our previous study at a lower pH, elucidating that the same strategy of food structure design is effective for the wide pH range encountered in food production. Results from the present study can be utilized for product development, particularly in foods for the elderly or for nursing-care purposes, where soft gels are frequently used as matrixes, as in jellies or puddings.

References

- Arvisenet, G., Billy, L., Royer, G. and Prost, C. (2006). Role of mastication on the release of apple volatile compounds in a model mouth system. *Dev. Food Sci.*, **43**, 465-468.
- Baek, I., Linforth, R.S.T., Blake, A. and Taylor, A.J. (1999). Sensory perception is related to the rate of change of volatile concentration in-nose during eating of model gels. *Chem. Senses.*, **24**, 155-160.
- Burse, K.M.M., Brattinga, C., de Kok, P.M.T. and Bult, J.H.F. (2010a). Sweet taste enhancement through pulsatile stimulation depends on pulsation period not on conscious pulse perception. *Physiol. Behav.*, **100**, 327-331.
- Burse, K.M.M., Camacho, S., Knoop, J. and Bult, J.H.F. (2010b). Sweet taste intensity is enhanced by temporal fluctuation of aroma and taste, and depends on phase shift. *Physiol. Behav.*, **101**, 726-730.
- Busch, J.L.H.C., Tournier, C., Knoop, J.E., Kooyman, G. and Smit, G. (2009). Temporal contrast of salt delivery in mouth increases salt perception. *Chem. Senses*, **34**, 341-348.
- Bylaite, E., Ilgunaite, Z., Meyer, A.S. and Adler-Nissen, J. (2004).

- Influence of λ -carrageenan on the release of systematic series of volatile flavor compounds from viscous food model systems. *J. Agric. Food Chem.*, **52**, 3542-3549.
- Harrison, M., Campbell, S. and Hills, B.P. (1998). Computer simulation of flavor release from solid foods in the mouth. *J. Agric. Food Chem.*, **46**, 2736-2743.
- Hendrik J.S., Kemsley E.K., Henri S.T. and Henry C.J.K. (2011). Does prolonged chewing reduce food intake? Fletcherism revisited. *Appetite*, **57**, 295-298.
- Hodgson, M., Linforth, R.S.T. and Taylor, A.J. (2003). Simultaneous real-time measurements of mastication, swallowing, nasal airflow, and aroma release. *J. Agric. Food Chem.*, **51**, 5052-5057.
- Holm, K., Wendin, K. and Hermansson, A.M. (2009). Sweetness and texture perceptions in structured gelatin gels with embedded sugar rich domains. *Food Hydrocolloids*, **23**, 2388-2393.
- Humphrey, S.P. and Williamson, R.T. (2001). A review of saliva: Normal composition, flow, and function. *J. Prosthet. Dent.*, **85**, 162-169.
- Liem, D.G., Westerbeek, A., Wolterink, S., Kok, F.J. and de Graaf, C. (2004). Sour taste preferences of children relate to preference for novel and intense stimuli. *Chem. Senses*, **29**, 713-720.
- Logemann, J.A., Pauloski, B.R., Colangelo, L., Lazarus, C., Fujii, M. and Kahrilas, P.J. (1995). Effects of a sour bolus on oropharyngeal swallowing measures in patients with neurogenic dysphagia. *J. Speech Hear. Res.*, **38**, 556-563.
- Maruyama, K., Sato, S., Ohira, T., Maeda, K., Noda, H., Kubota, Y., Nishimura, S., Kitamura, A., Kiyama, M., Okada, T., Imano, H., Nakamura, M., Ishikawa, Y., Kurokawa, M., Sasaki, S. and Iso, H. (2008). The joint impact on being overweight of self reported behaviours of eating quickly and eating until full: cross sectional survey. *Br. J. Med. Psychol.*, **337**, 1091-1093.
- Meiselman, H.L. (1972). Human taste perception. *CRC Crit. Rev. Food Technol.*, **3**, 89-119.
- Meiselman, H.L. and Halpern, B.P. (1973). Enhancement of taste intensity through pulsatile stimulation. *Physiol. Behav.*, **11**, 713-716.
- Mestres, M., Moran, N., Jordan, A. and Buettner, A. (2005). Aroma release and retronasal perception during and after consumption of flavored whey protein gels with different textures. 1. In vivo release analysis. *J. Agric. Food Chem.*, **53**, 403-409.
- Mosca, A.C., van de Velde, F., Bult, J.H.F., van Boekel, M.A.J.S. and Stieger, M. (2010). Enhancement of sweetness intensity in gels by inhomogeneous distribution of sucrose. *Food Qual. Prefer.*, **21**, 837-842.
- Mosca, A.C., Bult, J.H.F. and Stieger, M. (2013). Effect of spatial distribution of tastants on taste intensity, taste intensity fluctuation and consumer preference of (semi-)solid food products. *Food Qual. Prefer.*, **28**, 182-187.
- Moore, P.A. (1994). A model of the role of adaptation and disadaptation in olfactory receptor neurons: implications for the coding of temporal and intensity patterns in odor signals. *Chem. Senses*, **19**, 71-86.
- Nakao, S., Ishihara, S., Nakauma, M. and Funami, T. (2013). Inhomogeneous spatial distribution of aroma compounds in food gels for enhancement of perceived aroma intensity and muscle activity during oral processing. *J. Texture Stud.*, DOI:10.1111/jtxs.12023.
- Noort, M.W.J., Bult, J.H.F., Stieger, M. and Hamer, R.J. (2010). Saltiness enhancement in bread by inhomogeneous spatial distribution of sodium chloride. *J. Cereal Sci.*, **52**, 378-386.
- Noort, M.W.J., Bult, J.H.F. and Stieger, M. (2012). Saltiness enhancement by taste contrast in bread prepared with encapsulated salt. *J. Cereal Sci.*, **55**, 218-225.
- Otsuka, R., Tamakoshi, K., Yatsuya, H., Murata, C., Sekiya, A., Wada, K., Zhang, H.M., Matsushita, K., Sugiura, K., Takefuji, S., Ouyang, P., Nagasawa, N., Kondo, T., Sasaki, S. and Toyoshima, H. (2006). Eating fast leads to obesity: findings based on self-administered questionnaires among middle-aged Japanese men and women. *J. Epidemiol.*, **16**, 117-124.
- Pionnier, E., Chabanet, C., Mioche, L., Quere, J.L.L. and Salles, C. (2004). In vivo aroma release during eating of a model cheese: relationships with oral parameters. *J. Agric. Food Chem.*, **52**, 557-564.
- Sprunt, J.C., Raithatha, C.E. and Smith, A.C. (2002). Swallow indicator methodology as an enhancement to combined time-intensity measurement of flavour release and electromyography for monitoring mastication. *Food Qual. Prefer.*, **13**, 47-55.
- Tarrega, A., Yven, C., Semon, E. and Salles, C. (2011). In-mouth aroma compound release during cheese consumption: relationship with food bolus formation. *Int. Dairy J.*, **21**, 358-364.
- Tournier, C., Sulmont-Rosse, C. and Guichard, E. (2007). Flavour perception: aroma, taste and texture interactions. *Food*, **1**, 246-257.