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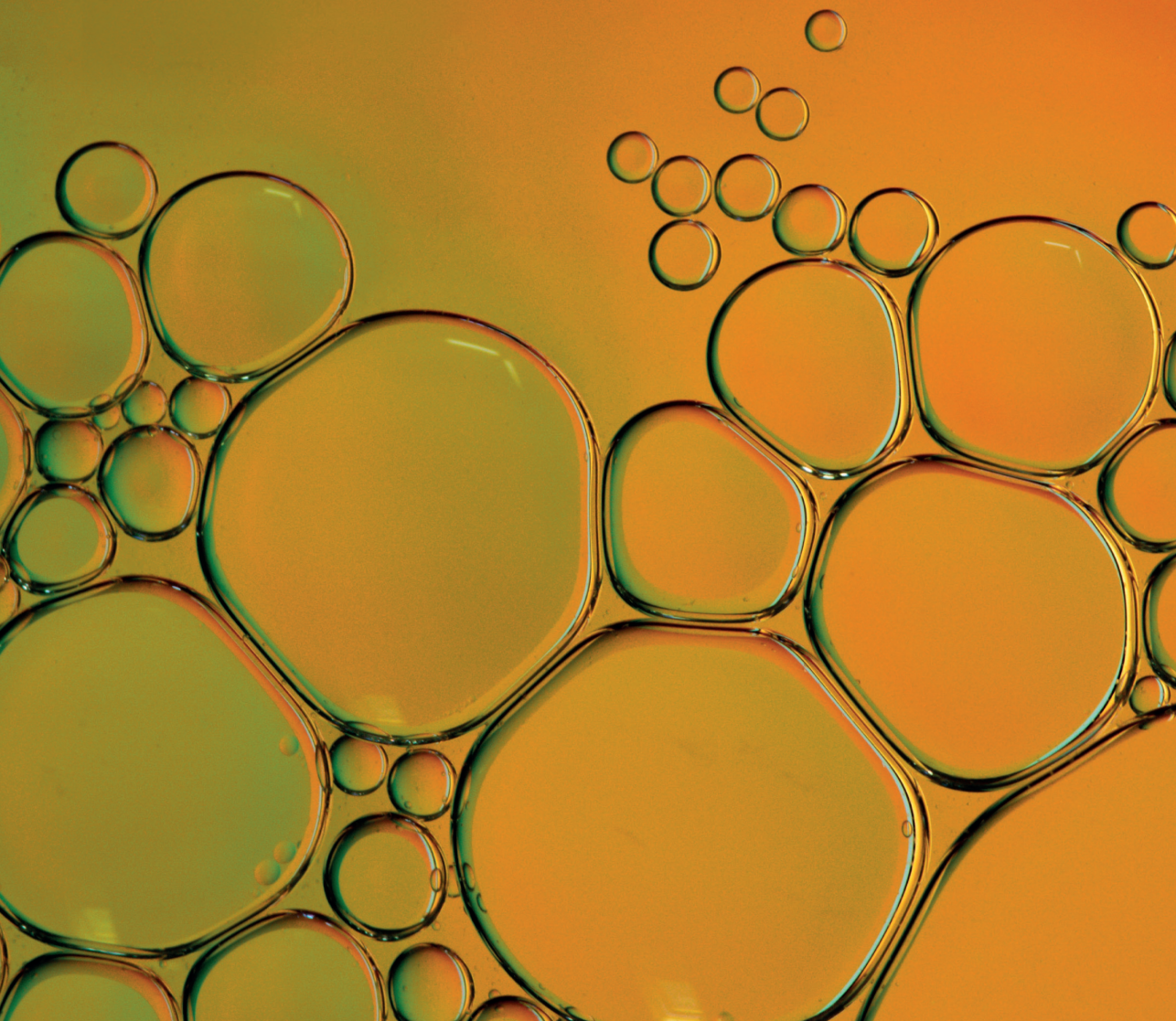
A novel approach to understand taste

Georgios Agorastos



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provincie limburg

The research described in this thesis has been made possible due to a support of the Dutch Province of Limburg. This dissertation was conducted at the facilities of T.A.S.T.E. Foundation, faculty of science and engineering Maastricht University and department of food physics, physical chemistry Wageningen University and Chemelot innovation and learning labs (CHILL).

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DISSERTATION

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Contents

General introduction

General Introduction

Human food consumption has fundamentally changed. For a long time, food consumption was limited to what was available. In the Western world, food is abundantly available which implies a fundamental shift in the driving forces of dietary behavior. Consequently, there is an increasing interest in understanding the drivers behind people's food choices (May-Wilson et al., 2022). Understanding the predominant driving forces of food choice has gained extra relevance since the negative effects of food consumption have become more evident.

These days, most foods are mass produced and mass distributed. The role of the food industry and food retail needs to be considered in influencing food behavior. Liking and deliciousness are drivers of food choice, which implies that taste is a critical determinant of consumers' food choices (Clark, 1998). In developing new products, sensory characteristics are important for food manufacturers and researchers.

Currently, many new food products or reformulations that are launched in the market fail on the market. A recent report from the U.S. indicates that 85 per cent of the new consumer-packaged goods fail within two years (Salnikova et al., 2019). Failure rates could differ extremely among various food product categories, nevertheless, the sensorial match between product characteristics and consumer expectation is believed to be one of the most important parameters for a successful strategy of new product development (Rudder et al., 2001). To start hypothesizing, understanding the taste of food products and the expectations of consumers are vital for the development of a new product formulation and reformulation strategy. Some initial questions could be:

- Does the basic understanding of taste/flavor need re-evaluating?
- Does the basic understanding of tasting food and beverages need re-evaluating?
- Does the above have implications for sensory evaluation methods?

To kick off, it is useful to have a closer look at sensory attributes, oral processing and subsequent definitions.

Taste, odor, mouthfeel and flavor

The complexity of taste and flavor is intriguing, not only in terms of the sensory origins, but also in terms of the definition and expression of the sensory attributes. Therefore, it is important to begin by distinguishing the differences between taste, tasting, and taste perception. Taste is related to the presence of chemical components in food and beverages. Tasting is the process when food compounds interact with saliva and receptors. Taste perception is the interpretation in the brain after transduction.

During the consumption of foods and beverages, a wide variety of chemical components are released into the mouth. Those components can be classified into volatiles and non-volatiles which are responsible for different oral and nasal sensations. The volatiles are transported from the mouth to the nasal cavity and can be described as odorants and irritants. Volatile molecules are released from the food matrix upon consumption and activate the olfactory system (Zarzo,

2007). The non-volatile ones are characterized as tastants, irritants and textural compounds. Tastants are mostly hydrophilic components that activate the taste receptor of the gustatory system (di Lorenzo et al., 2009), while irritant and textural components are registered by the trigeminal system (Gawel et al., 2018; Guinard & Mazzucchelli, 1996; Klosse, 2013). Flavor has also been defined by the ISO as "the complex combination of the olfactory, gustatory and trigeminal sensations perceived during tasting. Flavor may be influenced by tactile, thermal, painful and/or kinesthetic effects" (ISO 5492:2008). Therefore, three neural pathways are involved in flavor registration: the gustatory, olfactory and trigeminal systems (figure 1). Foods and drinks must have these characteristics, otherwise, there would be nothing to register.

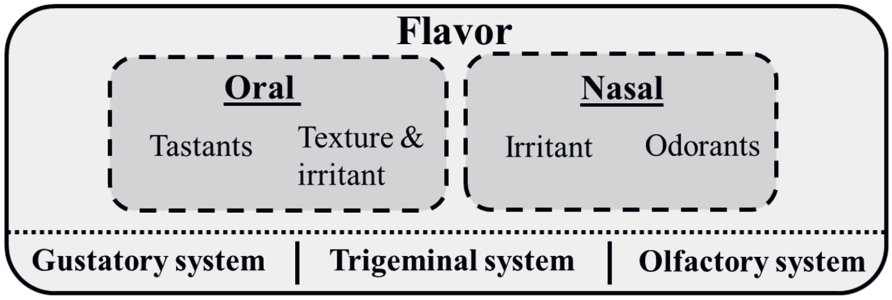

Figure 1. Schematic illustration of flavor interpretation in different neural systems

Mouthfeel is generally described as the sensory term corresponding to textural and irritant sensations. Mouthfeel refers to the tactile, irritant and thermal sensations that can be perceived in the mouth during oral tasting (Gawel et al., 2018). The stimuli of these sensations originates from the activation of chemosensory and somatosensory receptors within the oral cavity (Gawel et al., 2018; Guinard & Mazzucchelli, 1996). In contrast to taste and aroma, mouthfeel is generally considered to be related to signals transmitted by the trigeminal receptors (Hewson et al., 2009; Reynolds, 2010). Thus, mouthfeel as a part of the total flavor sensation is an important sensorial parameter.

Additionally, it has been reported that tastants influence mouthfeel sensations as well (Liu et al., 2017). In the oral cavity, tastants affect mouthfeel sensations by altering rheological behavior. Consequently, mouthfeel sensations are not solely dependent on irritants and textural components, which calls for a broader definition. A narrow interpretation of oral sensations can lead to miscommunication among different disciplines regarding mouthfeel. It is hypothesized that this confusion may account for the high rejection rates of new foods that fail to meet the sensory requirements of consumers.

To integrate the common sensorial aspects of taste and mouthfeel, Klosse (2013) proposed a mouthfeel model with three axes: contracting, coating and drying. These three categories represent all elements of taste, based on irritants, and textural components that initiate mouthfeel sensations. As a result of this model, communication among different disciplines has been proven to be more effective between food designers, consumers, and researchers.

Therefore, the current dissertation was focused on revealing mouthfeel properties such as drying and coating using instrumental and sensorial techniques.

Oral processing and the physical aspects upon consumption

During oral processing, food components are released from the food matrix and interact with saliva. Therefore, saliva is considered the solvent of volatile and non-volatile components, which is subject to biochemical and physiological interactions with food components, such as electrostatic, ionic, or enzymatic interactions. Saliva is a bio-fluid with multiple roles. Salivary lubrication plays a significant role in oral processing. When saliva and food components interact, salivary film integrity is altered, resulting in the activation of the mechanoreceptor (Stokes et al., 2013).

Physiological and physical measurements can provide insight into mouthfeel sensations during eating in order to explain how the state of food is perceived. The viscoelasticity of food and beverages is thought to contribute to most texture characteristics of semisolid foods (Aktar et al., 2019). Therefore, many researchers investigated the association of food rheological behavior with textural attributes, such as mouthfeel. Additionally, the formation of thin layers and their lubricating properties have recently been recognized as important factors contributing to oral sensations (Mosca & Chen, 2017).

As oral processing is a dynamic phenomenon, Hutchings and Lillford, (1988) proposed a breakdown model for solid and semisolid food during mastication (figure 2). This three-dimensional model characterizes a food product based on the degree of structure, degree of lubrication and time. Consequently, this schematic representation illustrates the dynamic nature of oral processing, in which food structures and perception change over time, while the food oral breakdown can be captured in terms of particle size change and saliva secretion and incorporation (Guinard & Mazzucchelli, 1996).

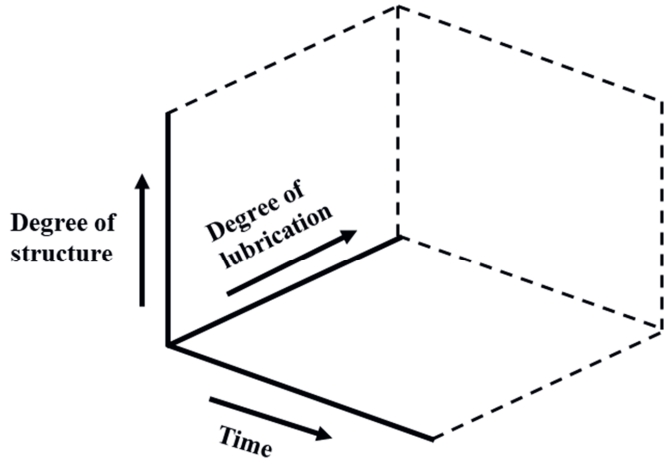

Figure 2. Hutchings and Lillford's philosophy of breakdown path (Hutchings & Lillford, 1988).

Taking all this into account, one can conclude that mouthfeel sensations are derived from the food state and the interactions of food compounds with saliva. It is important to note that oral processing is a dynamic process in which a variety of complicated interactions occur during consumption. In order to gain a deeper understanding of mouthfeel sensations, it is necessary to acquire a deeper understanding of the different molecular characteristics of the food components that are responsible for those interactions. A critical aspect of mouthfeel sensations is the interaction of food and saliva compounds, so it is necessary to investigate both the physical changes in salivary integrity as well as the effects of saliva on the food matrix closely.

Evaluation of organoleptic food properties

Until now, the evaluation of flavor-related characteristics has primarily relied on sensory evaluation. The sensory evaluation consists of a set of well-established methods providing useful information regarding the food perception of humans. The most popular sensory tests are (1) based on their ability to evaluate the overall differences among products (discrimination tests), (2) characterization of the sensorial attributes (descriptive tests) and (3) evaluation of the degree of consumer acceptance and satisfaction (affective or hedonic tests) (Heymann & Lawless, 2013).

Food quality characteristics are evaluated by humans as part of the sensory evaluation process, and therefore subjective (Huang et al., 2004). Methods such as these involve flavor, texture, taste, and odor perception, which are evaluated by experts or panelists of consumers (Poláková et al., 2008). Even though trained panelists possess a high level of experience, their judgment can be affected by psychological and physical factors (Huang et al., 2004). Performing subjective sensory analyses can be time-consuming and subject to a wide variety of sources of error. Furthermore, sensory evaluation is limited by the low repeatability and reproducibility of the results obtained due to several subjective and objective factors (Stone et al., 2012). By

nature, those methods can be biased by individual preferences and may be subjective to day-to-day variations (Huang et al., 2004; Polášková et al., 2008). Another disadvantage of sensory evaluation is the ability of humans to distinguish components of mixtures. Rarely, humans cannot distinguish more than three or four components in odor or taste mixtures (Laing, 1991).

A way of overcoming the uncertainty of sensory evaluation would be to measure taste objectively. Objective sensory methods include instrumental analysis and could be beneficial for various reasons as they are highly repeatable and reproducible since instruments do not suffer from fatigue or adaptation (Ebeler & Thorngate, 2009; Huang et al., 2004; Polášková et al., 2008; Smyth & Cozzolino, 2013). The development of new analytical and statistical methods has improved taste prediction. However, there is a challenge in interpreting instrumental data in terms of multimodal perception, which is mouthfeel. Multivariate analysis has been frequently used to find relationships between sensory and instrumental data. Additionally, the sensory information needs to be comprehensive or provided in a form that reflects the composition of the stimulus as it reaches the receptor (Williams, 1994).

However, to accurately correlate instrumental data to sensory attributes, the sensory information must be as precise and as meaningful as possible. This implies reducing response variation or examining data on an individual basis, as well as defining sensory attributes clearly (Chambers & Koppel, 2013).

Aims and outline of this thesis

Getting new insights on the chemical characteristics regarding mouthfeel sensation in order to be better able to predict taste. This thesis focuses on the current sensory lexicons, chemical characteristics responsible for mouthfeel sensations, and the ability to predict mouthfeel sensations via instrumental and multi-variance analysis. A new approach to evaluating mouthfeel sensations is also shown in this dissertation by integrating mouthfeel sensations through the mouthfeel model and its dimensions.

First, the current status of mouthfeel classification and different mouthfeel lexicons/wheels is described and discussed in **chapter 2**. In this chapter, the first attempts to classify mouthfeel are explained. The confusion of sensory attributes and the necessity of a common mouthfeel vocabulary are highlighted. The current mouthfeel wheels in different food products are reviewed and the applications of the mouthfeel model are described.

The importance of saliva on mouthfeel sensations is summarized in **chapter 3**. The saliva origin and properties are given in this chapter, while the physical and physicochemical properties of saliva are described by emphasizing the properties of salivary proteins. Also, the role of saliva on the part of tactile sensation origin is described in detail. Changes in saliva composition of elderly people or as a result the use of pharmaceuticals are highlighted, which indicates that research in this field merits further elaboration.

The effect of the molecular weight of polyphenols on salivary lubrication properties is explained in **chapters 4 and 5**. Measuring friction by soft-tribology, which mimics the human mouth, this study shows that larger polyphenols increase the friction of the system, while lower

pH values additionally increase friction. The disruption of the salivary film is dependent on the interaction of salivary proteins with polyphenols where aggregate formation occurs.

Chapter 6 elaborates on the effect of mineral salts on the lubrication behavior of human saliva. Different cationic valences were studied where trivalent salts cause loss of lubrication behavior while monovalent salts improve lubrication. Additionally, the interaction between phenols and cationic valences indicates that trivalent salt interacts with phenolic components which provide inhibition of lubrication loss.

The application of the mouthfeel model is presented in **chapter 7.** Commercial beers were analyzed, both chemically and sensorially. Using a chemometric application, sensorial parameters were found to be highly correlated with instrumental techniques regarding the molecular characteristics of beers. The general discussions on the studies described, as well as final conclusions, are formulated in **chapter 8**.

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Review of mouthfeel classification. A new perspective on food perception.

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Abstract

Characteristics of a food product are the backbone of sensory research, and it is essential to describe the food flavor with well-defined and agreed-upon concepts. This paper reviews the current bibliography related to taste/flavor perception, with a particular focus on mouthfeel. A summary of the current mouthfeel vocabularies is given, and research approaches are evaluated. A general mouthfeel model is presented that overarches product categories and has shown its use in practice. The intention is to contribute to an increased understanding of taste and flavor and mouthfeel sensations.

This paper reveals the ambiguity of terms that are regularly used in literature. This is influenced by different focuses in research. Three classes of research related to mouthfeel are identified: (1) product oriented (molecular attributes), (2) product/human oriented (human interface: receptors, saliva, chewing, etc.) and (3) human oriented (after swallowing).

For the future of research in flavor of foods and beverages, it is essential to have consensus on the definitions of relevant concepts and to have a model (classification) based on an approach that is generally accepted. A mouthfeel model is potentially a powerful tool for food producers and researchers alike since it can be used to classify food based on the differences in food composition. Generalist descriptors that can be used to describe mouthfeel in foods and beverages can improve the communication between diverse audiences and contribute to the understanding of taste, flavor and particularly mouthfeel.

Introduction

Currently, the discrimination of taste, smell, texture and flavor is described as following. In theory, taste is suggested to be related to five basic tastes: sweetness, sourness, bitterness, saltiness, and umami, with the gustatory system to mediate the sense of taste via the taste receptors cells (Yarmolinsky et al., 2009). The sense of smell is related to the detection of odorant components (volatile components) via the olfactory system (Zarzo, 2007). Szczesniak in the 60's defined the food texture as "the sensory and functional manifestation of the structural, mechanical and surface properties of food detected through the senses of vision, hearing, touch, and kinesthetics" (Szczesniak, 1963; Szczesniak, 2002). The term flavor finally is used to describe the combination of taste and smell sensations together with other sensations involving the olfactory, gustatory, and trigeminal sensations perceived during the tasting (Hewson et al., 2009; ISO 5492:2008; Reynolds, 2010) (figure 1).

A different aspect of food consumption apart from food texture is mouthfeel. Mouthfeel refers to the tactile aspects of texture perception during consumption and it is defined by Guinard & Mazzucchelli, (1996) that mouthfeel encompasses all the "tactile (feel) properties perceived from the time at which solid, semi-solid or liquid foods or beverages are placed in the mouth until they are swallowed." Recently, Gawel et al. (2018) defined the mouthfeel sensations as "The tactile, irritant and thermal sensations resulting from the activation of chemosensory and somatosensory receptors within the oral cavity by chemical stimuli". Unlike the sensation of

taste and smell, mouthfeel sensations are related to signals transmitted to the brain by the trigeminal system (Hewson et al., 2009; Reynolds, 2010). Scientists suggest mouthfeel is an important factor since, together with gustation and smell, mouthfeel contributes to the food perception and is crucial for the liking and acceptance of consumers (Guinard & Mazzucchelli, 1996; Kemp et al., 2011). In order to communicate about mouthfeel, consumers and food experts use sensory terms. Sensory terms form the language of how consumers describe a food product during tasting. Mouthfeel and texture are multi-parameter sensory attributes since many words are used to describe them (Szczesniak, 2002).

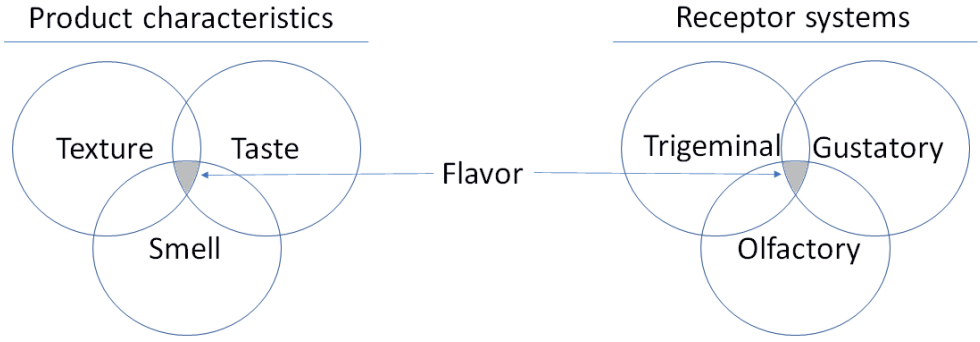

Figure 1. Schematic representation of flavor, taste, smell and texture discrimination.

Sensory lexicons and so-called sensory wheels are often used to categorize sensory terms. These standardized vocabularies are a way to objectively describe the sensory properties of consumer products (Lawless & Civille, 2013). Sensory lexicons can be an important communication tool among diverse audiences, such as sensory panelists, product developments, marketers, and suppliers (de Pelsmaeker et al., 2019; Gawel et al., 2000; Larssen et al., 2018; Pickering & Demiglio, 2008). Also, they may help with clarifying the different interpretations of the same sensory attribute due to differences in perception, background, and culture (Suwonsichon, 2019). In addition, lexicons allow product developers, and other product researchers to understand and study the sensory attributes of food products, which can be applied in product development, product maintenance, quality control, and shelf-life studies (Koppel & Chambers, 2010).

This paper first reviews the publications on descriptors of mouthfeel, sensory wheels and other classification attempts, including the mouthfeel model developed by Klosse (2013). This model deviates from the traditional division between taste and texture and classifies all kinds of mouthfeel sensations into three broad categories. This model provides a new approach that could potentially be useful in communication between consumers and food experts.

Research methods to generate mouthfeel vocabulary

During lexicon development, trained panelists first taste a number of samples within a product category in order to create a comprehensive frame or references for the sensorial space

(Lawless et al., 2012). After describing each sample from a list of selected terms the panelist group narrows the attribute list down through elimination of redundancies and vague terms (ASTM, 1996).

For the creation of a mouthfeel lexicon, it is important that the terms are established with the input of consumers, so that consumers will also be able to use them easily. There are two strategies for classifying mouthfeel terms. The first way to classify mouthfeel terms is by allocating them into logical groups with physical meaning defined well by the researchers. The second approach is that categories are derived from panelists' responses to grouping tasks without reference to their physical meaning. Then the mouthfeel terms are grouped according to a certain method of pairing or sorting comparisons (Hayakawa, 2015). Terms are generated by trained assessors' panels, but the disadvantage is that those terms may not always be understood by consumers (Varela & Ares, 2012). However, terms defined by consumers provide valuable input to the descriptive analysis (Heymann & Lawless, 2013).

For communication, it is imperative that sensory terms are clearly defined and unambiguous. It needs to be clear where one attribute begins and ends. Fuzzy conceptional boundaries need to be avoided. The sensory terms present several challenges to the understanding and interpretation of consumer data. It is difficult to understand synonyms using terminology with fuzzy conceptual boundaries (Ishii & O'Mahony, 1991). It is often unclear where one attribute ends or begins, for example, gritty and grainy or in the deformation characteristics differentiating crispy from crunchy. Ultimately, this should enable a high statistical correlation and conceptual overlap in the use of sensory terms. To avoid this problem, multivariate statistical analyses such as principal components analysis or cluster analysis can be applied. Such techniques can simplify many sensory attributes into fewer dimensions and clarify the data by finding dimensions or factors that are minimally correlated.

A technique to overcome the overlapping of the sensory parameters is sorting by using multidimensional scaling (MDS). Humans have the ability to organize and categorize perceptive features between objects based on similarities and dissimilarities, which allows for the utilization of sorting in sensory studies (Faye et al., 2004). Sorting has been used in sensory studies in order to ascertain the qualitative variation of the products. Sorting requires little training, does not require the use of a quantitative rating system and avoids biasing panelists (Cartier et al., 2006; Faye et al., 2004; Lawless et al., 1995; Saint-Eve et al., 2004). Sorting was introduced to the sensory evaluation in a cheese-based study by Lawless et al. (1995) with references to earlier free sorting of non-food personality impressions by Rosenberg et al. (1968). MDS is used in the analysis of data provided by sorting (Abdi et al., 2007; Lawless et al., 1995). MDS can provide clues for the perceptual features of the products tested by interpretation of the dimensional structure or patterns of clustering in the plots generated. The use of MDS can illustrate the difference among panelists by revealing how they cognitively group a set of stimuli according to some attributes. Stimuli that are located close together are considered similar and stimuli that are further apart, are considered different based on the dimension(s) of interest.

First attempts of mouthfeel terms categorization

The efforts of researchers to introduce some order and classification of mouthfeel and texture terms are introduced. As mentioned above, mouthfeel and texture are multi-parameter sensory attributes (Szczesniak, 2002).

Szczesniak and colleagues made the first effort for classifying and defining texture and mouthfeel terms. Szczesniak (1979) established that the awareness of texture and mouthfeel is as important as that of flavor. In a study for the determination of the sensory dimension of the term mouthfeel, they investigated the mouthfeel properties of 33 beverages. 103 Persons were asked to name as many terms as they could think of describing how beverages felt in the mouth. Based on the results of this study, Szczesniak (1979) concluded that mouthfeel can be reduced into 11 categories (table 1).

Table 1. Classification of sensory mouthfeel properties for beverages based on Szczesniak, (1979).

Category	**Terms**
Viscosity-related terms	Thin, thick, viscous
Feel on soft tissue surfaces	Smooth, pulpy, creamy
Carbonation-related terms	Bubbly, tingly, foamy
Body-related terms	Heavy, watery, light
Chemical effects	Astringent, burning, sharp
Coating of the oral cavity	Mouthcoating, clinging, fatty, oily
Resistance to tongue movement	Slimy, syrupy, pasty, sticky
After-feel-mouthfeel	Clean, drying, lingering, cleansing
After-physiological	Refreshing, warming, thirst-quenching, filling
Temperature-related	Cold, hot
Wetness-related	Wet, dry

Influenced by Szczesniak, Yoshikawa et al. (1970) tried to reduce the terms of texture and mouthfeel by analyzing a texture concept consisting of 40 terms to describe the texture of solid and liquid foods texture profiles were constructed for 79 foods employing the constant-sum method. Using a controlled association test, expressions describing texture were collected in response to 97 food stimulus words. Yoshikawa et al. (1970) used multivariate analysis to analyze the data. The multivariate analysis yielded eight orthogonal factors. These were: hard-soft, cold-warm, oily-juice, elastic-flaky, heavy, viscous, smooth, and gummy. In 1971 Henry et al. (1971) analyzed 15 texture and mouthfeel terms (chewy, springy, stringy, gummy, oily, grainy, breakable, firm, frangible, fluffy, sticky, smooth, fat-like, lumpy and elastic) using

texture profile analyses (TPA) in commercial desserts. Using factor analysis found that four of them (fluffiness, stickiness, firmness, and chewiness) were the most important.

For a study of the perceived textural dimensions of fruit-based beverages, Ingate & Christensen (1981) investigated the degree of correlations between mouthfeel characteristics. During the study untrained panelists had to rate 35 different beverages on 16 different mouthfeel terms. Ingate & Christensen (1981) developed a mouthfeel profile for fruit-based beverages, which includes two major dimensions: density/thickness, and chemical irritant effects. Brown et al. (1978) examined the sensory and instrumental data of many beers, using 13 sensory terms and with stepwise discriminant analysis, identified two major mouthfeel related axes: the body, and viscosity. Kokini et al. (1977) studied 12 textural terms in liquid and semi-solid foods. A liquid system of gums and fluid foods such as honey, corn syrup, etc. was used. Using regression analysis, they concluded that there are three important terms: thick, smooth, and slippery. Then they generated a three-dimensional space where the three key terms formed the base vectors, and the remaining textural attributes were dependent vectors with a specific magnitude and direction in this space. In the following study by Kokini et al. (1984), the analysis was repeated using 27 foods in conjunction with 25 textural and mouthfeel terms. The statistical analysis again resulted in the three already mentioned key terms: thick, smooth, and slippery.

In an attempt to recategorize the mouthfeel attributes of semisolid foods from Szczesniak (1979), Weenen et al. (2003) investigated three categories of semisolids: mayonnaise and dressings, custard desserts and warm sauces. Based on quantitative descriptive sensory analysis and using principal component analysis (PCA), concluded to categorize the mouthfeel of semisolids products, into two categories: texture and non-texture. The texture attributes were visco-elasticity, surface-feel, bulk homogeneity/heterogeneity, adhesion/cohesion, wetness/dryness, and fat sensations and non-texture attributes were temperature and irritation.

Various other studies investigated the terms of mouthfeel in semisolid food (table 2). Wendin et al. (1999) used three texture descriptors for the analysis of mayonnaise and dressings: thickness, fattiness, and toughness. In another semi solid food (mayonnaise) study, Åström (1998) distinguished texture from mouthfeel by using the terms oily and mouthcoating. For an analysis of custard, Kampp (1995) used mouthfeel terms such as firm, rubbery, airy, creamy, and sticky. de Cock & Vanhemelrijck (1995) analyzed starch-based emulsified sauces using the mouthfeel terms firm, slippery, sticky, grainy, mealy, melt rate, thick, creamy, and jumpy.

Table 2. Classification of mouthfeel attributes in semi-solids foods after the recategorization from Weenen et al. (2003).

Category	Class	Terms
Texture	Viscoelasticity	Thick, thin, melting, compact, airy, gummy, heavy, light, firm, spreadable, consistent, tough, dense, jelly-like, work to swallow, syrupy, pasty, viscosity, body, density
	Surface feel	Smooth, rough, astringent, grainy, powdery, gritty, slippery, puckery, chalky
	Bulk homo/heterogeneity	Smooth, heterogeneous, lumpy, slimy

	Co/adhesion	Sticky
	Wetness & dryness	Dry, mealy, moist, wet, watery
	Fat-related	Fatty, coating, creamy, creamy/soft, oily, greasy
Non-texture	Temperature	Cold, hot, warm, cooling
	Irritation	Prickling, mouth-watering, saliva forming, pungent, burning, sharp, chemical irritation

The classification of sensory terms proposed by Szczesniak, and colleagues was generally accepted among food scientists and technicians. Their works provided the base of the texture profile published by the International Standard Organization (ISO, 1994). The limitations of this terminology approach were noted by Nishinari et al. (2008). Nowadays the reliance on consumer judgment has become bigger. A need for developing consumers-oriented mouthfeel vocabularies and well-established clarification of the mouthfeel terminology is essential since profiling panels and consumers do not always speak the same language.

Table 3. Summary of attribute categories based on mouthfeel terms in food products.

Categories	**Products**	**References**
Viscosity-related	Beverages	Szczeniak et al. (1979)
Feel on soft tissue surface		
Carbonation-related		
Body-related		
Chemical affects		
Coating of the oral cavity		
Resistance to tongue movement		
After-feel mouthfeel		
After-physiological		
Temperature-related		
Wetness-related		
Hard-soft	n.g.	Yoshikawa (1970)
Cold-warm		
Oily-juicy		
Elastic-flaky		
Heavy		
Viscous		
Smooth		
Gummy		
Fluffiness	Desserts	Henry et al. (1971)
Stickiness		
Firmness		
Chewiness		

Density-thickness	Fruit-based beverages	Ingate & Christensen (1981)
Chemical irritant		
Body	Beer	Brown & Clapperton, (1978)
Viscosity		
Thick	Liquid and semi-solid food	Kokini et al. (1977)
Smooth		
Slippery		
Visco-elasticity	Semi-solids food	Weenen et al. (2003)
Surface-feel		
Bulk homogeneity/heterogeneity		
Adhesion/cohesion		
Wetness/dryness		
Fat sensations		
Temperature		
Irritation		
Thickness	Mayonnaise and dressings	Wendin et al. (1999)
Fattiness		
Toughness		
Oily	Mayonnaise	Åström (1998)
Mouthcoating		
Firm	Custard	Kampp (1995)
Rubbery		
Airy		
Creamy		
Sticky		
Firm	Starch-based emulsified sauces	Cock & Vanhemelrijck (1995)
Slippery		
Sticky		
Grainy		
Mealy		
Melt rate		
Thick		
Creamy		
Jumply		

Mouthfeel vocabularies and wheels

Sensory lexicons or sensory wheels are useful tools for communication between different disciplines such as food researchers, consumers, marketers, etc. This chapter summarizes the mouthfeel wheels of food products based on the main attribute groups (table 4).

In a study to create a structured vocabulary in order to assist tasters in a better interpretation of ''in mouth'' sensations of red wine, Gawel et al. (2000) asked the opinions of experienced tasters after tasting an extensive range of commercial red wines. A tasting panel consisting of 12 males and two females used different sensory terms to describe the mouthfeel sensations of wines. Based on the results of the sensory terms and definitions, Gawel et al. (2000) formulated a mouth sensory wheel of red wines. The mouthfeel wheel consisted of the following multiple and interacting sensations: acidity, sweetness, bitterness, retro nasal aroma perception (flavor), viscosity, warmth, and astringency. To detect any natural grouping of terms, the above-mentioned authors used an aggregate data matrix in order to identify similarities between the terms using cluster analysis. Gawel et al. (2000) identified three groups of oral sensations: astringency, feel (irritation, heat, texture, and weight) and acidity. Even though the authors recognized inherent difficulties in proposing these terms they represent a vocabulary developed by a relatively small number of wine-tasters. The lexicon of Gawel et al. (2000) suggested that it may not be comprehensive and that the wheel may need revision. Pickering & Demiglio (2008) provided a more detailed explanation and justification for the white wine mouthfeel. The wheel is divided into discrete sensations and integrated percepts. The discrete sensation had the following groups: tingle, pucker, mouth water, mouth dynamics, fullness, surface texture, irritation, mouth coat, overall drying, and length.

Brown et al. (1978) developed a flavor wheel for beer including the following attributes: warming, carbonation, powdery, astringent, metallic, mouthcoating, and alkaline in the mouthfeel sensations. Langstaff et al. (1991) classified three tactile sensations (carbonation, fullness, and after-feel), recommending nine sensory terms to describe these dimensions. The nine sensory terms were: stickiness, astringency, oily mouth coating for the after-feel dimension, viscosity, and density for the fullness dimension and total carbon dioxide, foam volume, bubble size, and sting for the carbonation dimension respectively.

In a study to develop the vocabulary and a sensory wheel for marine oils, Larssen et al. (2018) used twenty-two assessors. The assessors used the free choice profiling technique and the terms that they found were grouped after a discussion with six-panel leaders. During the sensory analysis and language development, the use of reference standards was useful for the panel in order to familiarize themselves with the product and the scaling system. The reference standards were developed or adjusted based on NMKL (2005). For the characterization of the mouthfeel of marine oils, Larssen et al. (2018), found 5 groups: hot, astringent, pungent, thin, and thick.

For a better understanding of mouthfeel attributes Bertino & Lawless (1993) analyzed the data of 35 female and 33 male panelists. The panelists had to sort the attributes of mouthfeel in health oral products, with pairwise similarity ratings of the attributes and ratings of each

attribute on property scales. Bertino & Lawless (1993) used multidimensional scaling in order to find qualitative relationships among mouthfeel attributes. Based on the similarity estimates they suggested three groups of oral sensations: numbing, astringent, and hot.

Using the free choice profiling technique used by de Pelsmaeker et al. (2019) for the sensory description of chocolate. de Pelsmaeker et al. (2019) used a similar approach as Gawel et al. (2000) and Larssen et al. (2018), categorizing the mouthfeel sensations into 5 groups: stickiness, graininess, dryness, creaminess, and coating.

Van der Stelt et al. (2020) developed a hierarchical structure of mouthfeel terminology based on medical nutrition products (MNP). Using taxonomic free sorting developed a hierarchical structure that is represented as a wheel. Using trained panelists analyzed 32 products resulting in 9 umbrella terms and 51 individual attributes. The 9 categories of this wheel were: consistency/body, product texture in the mouth, manipulation in the mouth, mouth effect, mouth-coating, product behavior, mechanics of swallowing, immediate after effect, and throat effect after swallowing.

The creation and organization of lexicon wheels is a qualitative process. The terms are generated by a group of expert panelists after a discussion of the similarities between a group of attributes, categorize the similar attributes in the same categories. Since sensory panels, sensory scientists, product developers, marketing professionals, and suppliers may have different understandings of the same attributes because of differences in perception, background knowledge, and culture, sensory lexicons can play an important role in effective communication among diverse audiences (Koppel & Chambers, 2010; Suwonsichon, 2019). However, some studies tested the effectiveness of mouthfeel lexicons and suggested that the mouthfeel lexicons are too complicated for the panelists (DeMiglio et al., 2002). The effectiveness of the mouthfeel wheel was also studied by King et al. (2003). The results suggested that the mouth wheel was too complex for the training panelists. Similarly, other authors found the definition of the sub-qualities of the mouthfeel wheel very confusing (DeMiglio et al., 2002). The outcome of this study suggests that the mouthfeel wheel needs further modifications and simplification in order to be effective. A simpler mouthfeel classification needs to be used for product developers and panelists to better understand the product characteristics.

Table 4. Summary of main attribute groups of the mouthfeel wheels in food products.

Mouthfeel wheels group attributes	**Products**	**References**
Astringency	Red wine	Gawel, Oberholster & Francis (2000)
Feel		
Acidity		
Tingle	White wine	Pickering & Deminglio (2008)
Pucker		
Mouth water		

Mouth dynamics		
Fullness		
Surface texture		
Irritation		
Mouth coat		
Overall drying		
Length		
Carbonation	Beer	Langstaff et al. (1991)
Fullness		
After-feel		
Warming	Beer	Clapperton (1973)
Carbonation		
Powdery		
Astringency		
Metallic		
Mouthcoating		
Alkaline		
Hot	Marine oils	Monteleone & Hersleth (2018)
Astringency		
Pungent		
Thin		
Thick		
Numbing	Oral health products	Bertino & Lawless (1993)
Astringency		
Hot		
Stickiness	Chocolate	de Palsmeaker et al. (2019)
Graininess		
Dryness		
Creaminess		
Coating		
Consistency/body	Medical nutrition product	van der Stelt et al. (2020)
Product texture in the mouth		
Manipulation in the mouth		
Mouth effect		
Mouth-coating		
Product behavior		
Mechanics of swallowing		
Immediate after effect		
Throat effect after swallowing		

A mouthfeel model developed in practice

Klosse (2013) proposes an overarching empirical mouthfeel model that classifies the mouthfeel sensations of all kinds of products. Following the free choice profiling methodology, panelists were asked to evaluate mouthfeel attributes. The mouthfeel attributes are categorized into three dimensions: contracting, drying, and coating (figure 2). The dimensions are related to a combination of sensations related to many types of tactile experiences, including texture, thermal effects, and chemical influences of different types of fats, proteins, carbohydrates, acids, minerals, metals, and irritants. The mouthfeel model classifies food products into three dimensions based on their component composition and intensity. The mouthfeel model gives the ability for the classification of mouthfeel and taste based on product characteristics. This approach is based on products and aims at developing scales on the three dimensions and assessing the taste of food products more objectively, giving the possibility of characterizing food components related to mouthfeel perception.

In general, contracting is the result of acidity, saltiness, and "irritants" that cause a contraction in the mouth, just as carbon dioxide (CO_2) does in beverages (e.g., sparkling or mineral water, sparkling wines, beer, and soda). Organic acids and minerals are reported to enter the ion-channel of taste cells directly (Gilbertson et al., 2000). Oral irritants, e.g., capsaicin, spices ginger, which are mediated by the nociceptors of the trigeminal system enter directly into ion-channels as well (Byrnes & Hayes, 2013; McCleskey & Gold, 1999; Simons et al., 2019). This common ion-channel mediated mechanism seems to induce a contracting mouthfeel. Multiple receptor types expressed on sensory nerve fibres serve to detect chemesthetic compounds (give the sensation of irritation, pain, temperature, and roughness in the oral cavity) (Roper, 2014) that can contribute to mouthfeel associated with foods and beverages. A common denominator of contracting seems to be that this category is mediated through ion-channels. In general, contracting gives the impression of refreshment or cleansing the mouth.

The coating dimension in the mouthfeel model of Klosse (2013) is illustrated by the residual film which remains in the mouth after swallowing. Fats, like butter, cream, animal fats, oils, and for instance peanut butter can induce a similar sensation. The word coating was chosen because the elements that lead to a coating sensation leave a thin coating layer in the mouth as fats and/or dissolved sugars do (Rolls et al., 1999). Several factors such as type and concentration of fat, polysaccharides, and proteins, the physicochemical properties of food and the morphology of the tongue are hypothesized to influence the oral coating formation (Camacho et al., 2014; Camacho et al., 2015; de Wijk et al., 2009; Dresselhuis et al., 2008; Fleury et al., 2002; Lynch et al., 1993; Madrigal-Galan & Heymann, 2006; Pivk et al., 2008; Ranc et al., 2006; Vingerhoeds et al., 2009). Saliva becomes thicker and more viscous. However different fats and mono- and disaccharides have been demonstrated to induce a similar coating mouthfeel. Physiologically these aspects of mouthfeel are sensed by the mechanoreceptors of the trigeminal system. Their acuity is quite remarkable; even small differences in viscosity can be perceived and particles as small as 5µm can be detected (Guinard & Mazzucchelli, 1996).

The third dimension of the model is drying. Several factors may account for dryness. First is the absorption of saliva, which happens with dry (devoid of moisture) solid food such as toast, biscuits, crackers, puff pastry, crusts, crisps, and chips. In general, dryness refers to the 'bite' of something. Some textures are hard and break easily, and sometimes the break is somewhat soft, but resistant, resulting in a chewy texture with a certain elasticity. Components that absorb fluids (including saliva) are drying. Consider how rice, potatoes, or bread absorb fluids. Drying is neither contracting, nor coating, but it can affect the perception of taste and quality. For instance, in the crust of freshly roasted meat or toast or puff pastry. Some young red wines can be 'drying' or 'puckering' (Gawel et al., 2000). Likewise, some other bitter-tasting substances such as strong coffee or tea can be astringent, bitter, or acid.

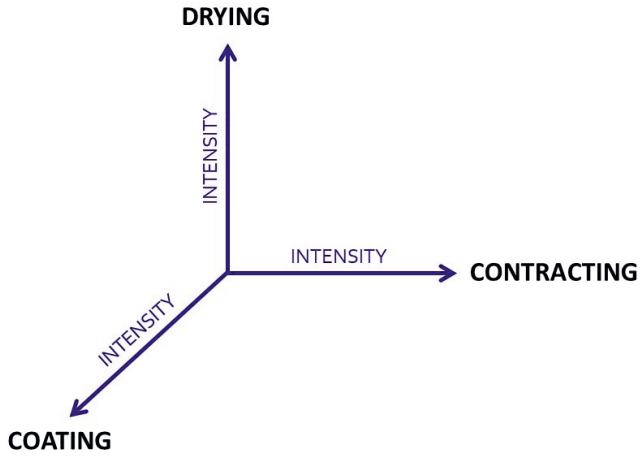

Figure 2. The mouthfeel model represented by the three dimensions (Klosse, 2013).

In general, the terms found in this review were found to fit rather well in the three dimensions of Klosse's mouthfeel model (2013). A further investigation of this mouthfeel model is needed. A mechanistic model which includes the effects of different ingredients in the model will be a useful tool for the food industry, giving the ability to have a more precise tool for food and mouthfeel classification.

Discussion

This review revealed many different terms and lexicons to describe mouthfeel. Many different words have been reported. Many of these attempts are the result of a focus on one specific product category which results in terms that specifically apply to that category. While Klosse's model is an overarching model for mouthfeel classification.

Next, it is important to note that there are different research approaches. Clearly, this influences the formulation of concepts which adds to the ambiguity. Figure 1 in the introduction shows two foci of research. One is oriented toward product characteristics and the other is toward the

perception of foods and beverages. As soon as tasting becomes a part of taste, the objectivity gets lost because of interpersonal differences. This is certainly the case when taste is defined as a human experience with all external influences.

Consequently, three types of food perception research related to mouthfeel classification can be distinguished.

- product focused
- product/human focused
- human focused

The "product" approach focuses on the food components and the physicochemical characteristics of a product before consumption. This approach classifies taste and texture using the compositional factors of food, the molecular composition of a food product, without the involvement of a human. It is the most objective approach to mouthfeel classification. Principal Component Analysis (PCA) is one of the tools used to analyze the data.

The second approach ("product/human") classifies mouthfeel during human consumption. It includes the interaction of different food ingredients with human saliva (chewing, dilution, enzymatic reaction, etc.) and receptors. As saliva bathes the sensory receptors, food components interact first with saliva. It is less objective because of all kinds of human differences in receptors, saliva composition and flow, chewing and swallowing behavior. This suggests an interaction of taste and texture. These would so to say merge into mouthfeel. This approach suggests a new concept of flavor classification (figure 3).

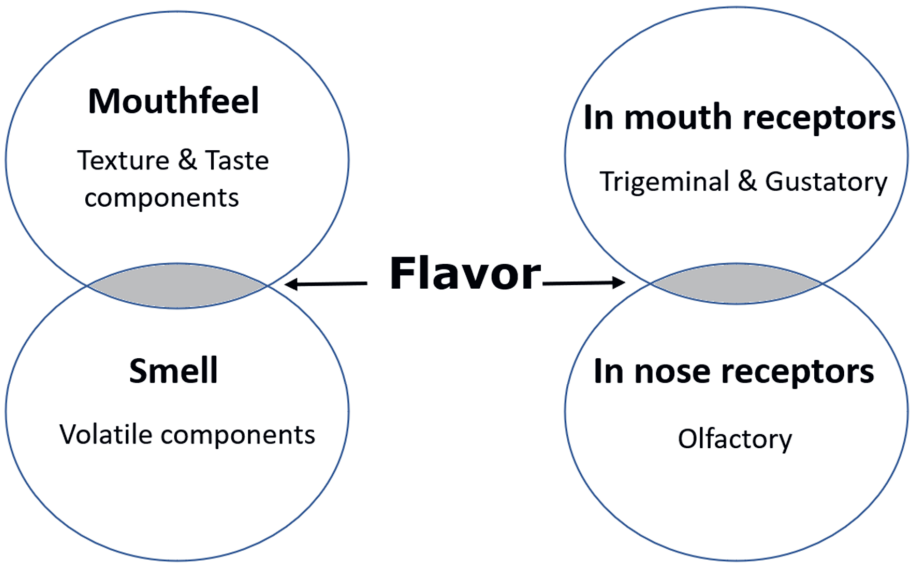

Figure 3. A new perspective for food classification.

The third category of analysis is the “human” approach. This type of research focuses on flavor perception after swallowing and processing in the brain. Spence (2017) introduced ”Gastrophysics” to describe this domain. It is defined as ‘the scientific study of those factors that influence the human multisensory experience while tasting food and drink’. Under this new concept, it was introduced a new definition the “flave”. Flave is the complete integration of all senses during tasting.

The different levels of food perception research are summarized in table 5. Clearly, different research orientations give different results, and this adds to discrepancies in the results of sensory research in general. An effective mouthfeel model for the classification of taste in food products is a useful tool. The complexity of the many sensory terms can lead to errors in the classification of food products. An objective, well-accepted model will improve the communication between product specialists, researchers, sensory panels, etc. A more robust and general classification system will resolve misunderstandings of attribute perception between people from different cultures and backgrounds. The most objective classification will potentially be determined on the product level and be based on real and measurable characteristics.

Table 5. Classification of mouthfeel based on different approaches.

Product	**Product/human**	**Human**
Taste and texture before swallowing	Taste and texture during swallowing: mouthfeel	Perception of all intrinsic and extrinsic factors: flave
Objective	Objective/subjective	Subjective
No human involvement	Human physiology (saliva)	Human interpretation
Food molecules	In mouth interactions of molecules	Brain

Conclusion

A review of different classification approaches of mouthfeel was given in this paper. In summary, this review suggests mouthfeel as a tool for food classification. Already from the ’60s and ’70s, researchers understood that mouthfeel properties are as important as flavors. Many authors studied mouthfeel, defined specific sensory terms and developed mouthfeel

lexicons and wheels. These are useful steps, but often based on specific food categories. It is suggested that mouthfeel wheels are too complex for the panelists and consumers since there are too many attributes. This leads to a systematic error during the product development of food products. There is a need for a more concise and overarching model that is relatively easy to understand. The mouthfeel model suggested by Klosse (2013) summarizes the important sensory parameters related to mouthfeel in three dimensions: drying, coating and contracting. Concluding, mechanistic modeling of the mouthfeel model is needed. Further investigation of the effects of food composition on the mouthfeel dimensions can illustrate a predictive model for taste and flavor classification.

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On the importance of saliva in mouthfeel sensations

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Abstract

Mouthfeel is an important parameter for the total flavor perception of food and beverages. Saliva plays an essential role in oral processing via interaction with food components, which alter the salivary properties. Physiological differences such as age and health influence salivary properties, which lead to different mouthfeel sensations among individuals. Medication affects salivary integrity as well. Despite the importance of saliva in the mouthfeel, researchers largely underestimated the significance of saliva on oral sensations. This study reviews the role, properties and involvement of saliva in the tactile sensations of mouthfeel and the changes due to age, drugs and disease.

Introduction

Mouthfeel refers to the physical-textural sensations in the mouth during and after the consumption of food or beverages. Together with gustation and olfaction, mouthfeel contributes to food perception and is crucial for the liking and acceptance by consumers (Guinard & Mazzucchelli, 1996). Mouthfeel sensations were defined as "the tactile, irritant and thermal sensations resulting from the activation of chemosensory and somatosensory receptors within the oral cavity by chemical stimuli" (Gawel et al., 2018). Important tactile sensations are the drying and coating sensations (Klosse, 2013). A drying sensation refers to the drying feeling in the mouth, which is associated with salivary lubrication loss, while a coating sensation is associated with the presence of oral coatings in the mouth after consumption.

Saliva is naturally present in the mouth and forms a layer in the oral cavity. Therefore, its involvement in mouthfeel sensations is essential. Saliva is a key parameter for mouthfeel sensation as it strongly interacts with food components. Food components are never in direct contact with the oral receptors. Saliva covers the oral cavity and therefore the flavor receptors. It should be noted that trigeminal receptors (mouthfeel receptors) are not in direct contact with saliva since they are embedded in the epithelium of the oral mucosa (Mosca & Chen, 2017). The epithelium of the oral mucosa is covered by salivary proteins. Hence, the access of food components to trigeminal receptors depends on the ability to diffuse through both saliva and epithelium mucosa (Canon et al., 2018).

One of the most important roles of saliva is to lubricate and protect the oral surfaces. It is the natural lubricant of the oral surfaces, with an important role of the salivary proteins (such as mucins, proline-rich proteins, etc.) in the lubrication. These proteins are capable of forming salivary films (pellicles). The mucosal pellicle is a "bio"-film that covers the oral cavities (Odanaka et al., 2020). This film is unique among other biological lubricants, since it protects the oral cavity against wear and provides low friction coefficients (Boyd et al., 2021).

During oral processing, food components are released from the food matrix and interact with saliva. Therefore, saliva can be considered as the solvent of volatile and non-volatile components and undergoes biochemical and physiological interactions with the food components, such as electrostatic, ionic, or enzymatic interactions. These interactions alter the integrity of the salivary film, which results in the activation of the trigeminal receptors (Stokes et al., 2013). Tactile sensations originate from the activation of mechanoreceptors. These

receptors respond to mechanical pressure or distortion of the frictional force in the mouth, which depends on the lubrication of the oral cavity (van Aken, 2010). On the other hand, chemical irritation (chemesthesis) such as numbing, burning and tingling occur due to the presence of irritant compounds which activate sensory receptors that detect pain (nociceptors) and temperature (thermoreceptors) (Simons et al., 2019). Therefore, mouthfeel is described by both tactile and chemesthetic sensations.

This review aims to highlight the important role of saliva in mouthfeel sensations. It focuses specifically on the tactile sensations of mouthfeel. First, saliva characteristics will be discussed, followed by a more detailed look at the role of saliva in tactile sensations and the physiological factors that affect them. Emphasis is given to the effect of ageing, disease and medication.

Saliva characteristics

Saliva origin and composition

Saliva is a complex mixture consisting of water, cellular debris, bacteria, and secretions from the salivary glands in the mouth. Saliva consists of 99.5 % water, 0.3 % protein, and 0.2 % inorganic compounds (Laguna et al., 2017). The protein fraction mainly consists of enzymes, glycoproteins, immunoglobulins, and a wide variety of polypeptides such as statherins, cystatins, histatins, mucins and proline rich proteins (PRPs) (Humphrey & Williamson, 2001). The inorganic fraction consists of electrolytes, present in various concentrations, such as potassium, sodium, bicarbonate, and chloride (Schipper et al., 2007). The pH of saliva ranges between 6.2 and 7.4 (Schipper et al., 2007) and its buffer capacity counteracts changes in pH caused by foods or drinks (Lenander-Lumikari & Loimaranta, 2000). This buffering capacity is mainly dependent on the histidine-rich, low-molecular-weight polypeptides, bicarbonate, and phosphate ions (Humphrey & Williamson, 2001). This complex matrix makes saliva properties difficult to understand completely.

Saliva is produced by major and minor salivary glands. Submandibular, sublingual glands and parotid are the major salivary glands, while the minor glands are spread at the lower lip, tongue, palate, cheeks, and pharynx (Silvers & Som, 1998). Saliva differs depending on whether it is stimulated or unstimulated. Unstimulated saliva is primarily secreted by the submandibular and sublingual glands, whereas stimulated saliva is mainly secreted by the parotid glands. Saliva varies in composition, depending on the salivary glands that produced it. The activation level of each of these glands depends on the type of saliva stimulation. This increased stimulation of the parotid glands is caused through neural reflexes, instigated by chemical stimulation from food components (either via the olfactory or gustatory system) and mechanical stimulation from chewing (Mosca & Chen, 2017). Additionally, the composition of saliva is influenced by diet, emotional stress, time of the day, gender, age, disease, and pharmacological agents (Brandão et al., 2017). The variations in salivary composition are mostly expressed in compositional differences, in buffer capacity, mucin and total protein concentration, and amylase activity (Engelen et al., 2007).

Salivary proteins related to mouthfeel sensations

Salivary proteins are responsible for the viscoelastic properties of saliva. More than 400 proteins are present in saliva, but only some specific proteins proline-rich proteins (PRPs), α-amylase, statherin, cystatins, P-B peptide, and mucins) are important for the mouthfeel sensations (Condelli et al., 2006; de Freitas & Mateus, 2001; Gambuti et al., 2006; Messana et al., 2008; Xie et al., 2005). Salivary proteins have different biological functions, depending on their structure and composition (Brandão et al., 2017; Castagnola et al., 2011). Different types of saliva stimulation affect protein composition. For instance, unstimulated saliva contains higher amounts of proteins compared to stimulated saliva (Schipper et al., 2007). The main salivary proteins and their average distribution in unstimulated saliva are listed in table 1.

Table 1. The molecular weight (MW), isoelectric point (pI) and average distribution of the main salivary proteins in unstimulated saliva.

Salivary proteins	***Mw (kDa)***	***pI***	***Concentration***
Mucins		2-3	>15 %
MUC5B	2000-4000		
MUC7	130-180		
aPRP	28	4	>15 %
bPRP	10-20	>10	5-15 %
gPRP	70	7	5-15 %
a-Amylase	54	4.1	>15 %
Cystatins	13	9.3	1-5 %
Statherin	6	4.4	1-5 %

Data are from: Bennick, (1982); Boze et al., (2010); Delavat et al., (2012); Gibbins et al., (2014); Laterza et al., (2002); Oho et al., (1992); Perez & Proust, (1987); Schenkels et al., (1995); Valente et al., (2018).

The largest class of salivary proteins are the proline-rich proteins (PRPs), representing 60 % of the total weight of proteins (Castagnola et al., 2011). Their name originates from their high proline content, i.e., 25 to 42 % of the amino acids in these proteins (Bennick, 1982). PRPs are classified into three categories: acidic, basic and glycosylated. The differences between these categories are related to charges and the presence or absence of glycosylation in their structure. Basic PRPs have a high iso-electric point of around 10 (Gawel, 1998). Hence, in physiological saliva pH (around 7), these proteins are positively charged (Baliga et al., 2013). Acidic PRPs are very similar in structure to basic PRPs, except for the extension of the N-terminal, which contains a higher number of acidic amino acid residues, resulting in a pI of 4-5 (Gawel, 1998; McArthur et al., 1995). The glycosylated proline rich proteins (gPRPs) make up to 15 % of the total salivary proteins (Boze et al., 2010). The pI of gPRPs is reported to be around 7, and hence, at average saliva pH, they are nearly charged neutral (Oho et al., 1992). They are the most important of the PRPs for the lubrication of the oral cavity (Boze et al., 2010; Sarni-Manchado et al., 2008).

Mucins are large glycoproteins secreted by the submandibular and sublingual glands (Cook et al., 2017) at a concentration of around 1.2 mg/mL in healthy individuals (Kejriwal, 2014). They

are present freely in saliva and as transmembrane mucins attached to epithelial cells through hydrophobic interactions (Cook et al., 2017; Gibbins et al., 2014). In general, all mucins have a similar structure, despite their charge density and oligosaccharide content, and serve the same function of protecting delicate tissues up to a certain degree (Perez-Vilar & Hill, 1999). They are high molecular weight proteins carrying high-density oligosaccharide side chains with a molecular weight varying between 130 to 4000 kDa (Cook et al., 2017). These glycan sidechains are attached to the protein's backbone and constitute 50-90 % of the total molecular weight of the mucins. Because of hydrogen bonding between the sugar units and hydrophobic interactions of their non-polar groups, they tend to self-aggregate (Bansil & Turner, 2006). Mucins found in the oral cavity are divided into two groups, based on their molecular weight: the high molecular-weight MUC5B (2000-4000 kDa), and the low molecular weight MUC7 (130-180 kDa) (Gibbins et al., 2014; Schipper et al., 2007). MUC7 contains less heterogeneous glycosylation, with a smaller backbone and smaller oligosaccharide side chains compared to MUC5B. They are secreted by all salivary glands except for the parotid gland (Thomsson, 2002). The gel-like characteristic of saliva is due to the presence of this type of mucin. Their gelling properties are thought to originate from the hydrophobic interactions between the core protein's cysteine-rich hydrophobic regions (Bromberg & Barr, 2000), the hydrogen bonds between the sugar side chains and crosslinking mediated by calcium (Raynal et al., 2003).

Mucosal pellicle

Salivary proteins are either present freely in the saliva (flowing saliva) or specifically anchored onto mouth surfaces (salivary pellicles) (Ployon et al., 2016). The adsorption of salivary proteins onto oral surfaces is a highly selective process, where mostly mucins and to a lesser extent PRP, α-amylase, statherin, cystatins, lactoferrin, and immunoglobulin A adsorb onto any oral surface. These thin films, also called pellicles, are divided into the enamel pellicle (forming on the teeth) and mucosal pellicle (forming on the epithelia) (Hannig et al., 2017). The mucosal pellicle is a thin film of 30 to 100 nm of salivary proteins consisting of two layers covering the oral mucosa. This pellicle is made of salivary proteins anchored onto oral epithelial by covalent and non-covalent bonds (Gibbins et al., 2014). Changes in the integrity of the mucosal pellicle activate the mechanoreceptors that sense mouthfeel sensations (Nayak & Carpenter, 2008; Soares et al., 2017).

How this pellicle is formed is not yet exactly known, but two mechanisms have been proposed. The first mechanism proposes that mucins (mostly MUC5B) anchored onto the oral mucosa form the first layer of the mucosal pellicle. These mucins facilitate the anchoring of a pellicle of crosslinked PRPs, histadin, and statherin through hydrophobic and hydrogen bonding, forming the second layer (Gawel et al., 2018; Gibbins et al., 2014; Lendenmann et al., 2000; Macakova et al., 2010; Ployon et al., 2016; Svendsen & Lindh, 2009). The second mechanism proposes that the hydrophobically bound mucin layer is stabilized by the smaller proteins through cross-linking. In either case, the result is the formation of a thin lubricative layer, where “molecular brushes” are formed by mucins oligosaccharide's side chains. These brushes face outward and repel each other by steric effects and osmotic pressure (Gawel et al., 2018).

Salivary physical properties

As a biological fluid, saliva possesses unique physical properties. Mouthfeel sensations are mainly affected by the changes in both salivary properties and the integrity of the salivary films. Those salivary properties are dominated by protein composition and functionality.

Saliva has differences in rheological and lubricational properties depending on the stimulated and unstimulated manners, while those properties differ between different salivary glands (Prinz et al., 2007). The stimulated saliva provides less lubrication than the unstimulated. This is related to the higher protein content and lower viscosity of unstimulated saliva (Prinz et al., 2007). Stimulated saliva has a continuous and more compact matrix, while unstimulated saliva has a filamentous network (Vijay et al., 2015). Unstimulated saliva has a low viscosity, high elasticity, stickiness, water holding capacity, and lubrication properties, which is thought to be mainly caused by the presence of mucins (Humphrey & Williamson, 2001; R. G. Schipper et al., 2007). In addition, it shows non-Newtonian, shear-thinning behavior (Bhat et al., 2010). The apparent viscosity is highly variable and depends greatly on the mucin composition (Schipper et al., 2007; Stokes & Davies, 2007). Stimulated saliva has a different composition (Ash et al., 2014; Stokes & Davies, 2007) and consequently different physicochemical properties compared to unstimulated saliva. Stimulated saliva shows Newtonian behavior, with a shear-independent viscosity, and much lower viscosity and lubricating properties than unstimulated saliva (Mosca & Chen, 2017). The latter can be explained as stimulated saliva is 80 % secreted by the parotid gland, containing little to no mucins, which results in much less strong lubricational properties and thus a higher friction coefficient.

Mucins and gPRP are important salivary proteins with lubrication abilities (Pascal et al., 2009; Stokes & Davies, 2007). Around 20 % of the saliva proteins are mucins and 70 to 80 % of mucins contain carbohydrates. The main functions of mucins proteins are related to lubrication, hydration and protection of the oral cavity (Huq et al., 2007; Messana et al., 2008). MUC5B and MUC7 both contain the glycosylated chains important for lubrication. Due to its size and large glycosylated fraction, the MUC5B is considered to have the most impact on oral lubrication (Davies et al., 2014). The glycosylated parts of gPRP extend into the bulk fluid to form the lubricating film with the protein component of the gPRP anchored to the oral surface.

Mucins are known to provide oral lubrication via two mechanisms. Firstly, because of their high water-retaining capacity. Mucins have an isoelectric point between 2 and 3, giving them a negative net charge at physiological saliva pH. Because of this negative net charge, the mucins are surrounded by a hydration shell of water molecules. Under shear, these bound water molecules are exchanged for free water (Ma et al., 2015; Yakubov et al., 2009). The second mechanism works via the continuous de- and reabsorption of mucins onto the oral mucosa, dissipating energy and contributing to low friction (Crouzier et al., 2015). Furthermore, mucins coatings on a surface can also lead to steric repulsion between two surfaces which gives lower friction (Yakubov et al., 2009). Based on these mechanisms mucoproteins can provide lubrication to the oral surfaces.

The role of saliva in tactile sensations

Drying sensation

The sensation of drying/astringency in the mouth is important for beverage acceptance by consumers. This sensation is associated with a drying and puckering feeling in the mouth. Even though the mechanisms of drying are not fully understood, some important parameters that influence this sensation are known. The interaction between phenolic compounds and proteins is the most important parameter for astringency sensation. Salts of multivalent metallic cations, ethanol, mineral, organic acids and polyphenols are responsible for astringent sensations (Bajec & Pickering, 2008).

Currently, two commonly accepted theories describe astringency. These theories are distinguished as either friction-based or receptor-based. The first assumes that astringent components interact with saliva proteins via hydrogen bonding and hydrophobic interactions and then aggregate. The changes in the salivary film are responsible for the activation of the mechanoreceptors, which elicit the astringency sensation (Canon et al., 2018, Gawel, 1998; Gawel et al., 2018; Mosca & Chen, 2017; Soares et al., 2017, Chen, 2009; Gibbins & Carpenter, 2013). The second theory hypothesizes that the astringency sensation is caused by the direct interaction of polyphenols with the salivary proteins adhered to buccal mucosal cells that form the mucosal pellicle (Canon et al., 2021). In this review, the friction-based theory will be discussed in greater detail since it has recently gained quite some attention.

Friction-based astringency

Astringency is described as a tactile sensation since a strong correlation has been found between the oral friction and astringency intensity. Rossetti et al. (2009) found that a decrease in human salivary lubrication caused by epigallocatechin gallate (EGCG) increased the friction coefficient in tribology measurements using smooth polydimethylsiloxane (PDMS) surfaces and increase tactile perceptions of astringency by a sensory panel. Although the exact molecular mechanism is unclear and thought to vary with the nature of the astringent, some proposals have been made and are schematically represented in figure 1.

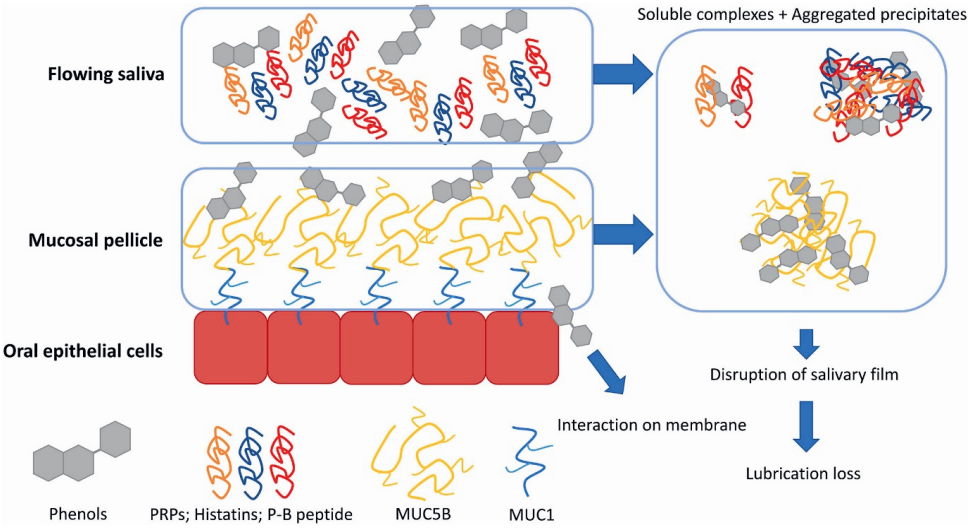

Figure 1. Schematic overview of the proposed multimodal perception of astringency perceived in the oral cavity triggered by the phenolic compounds (after Huang & Xu, 2021).

Astringency is associated with the changes in oral surface properties caused by the astringent compound, changes in lubricating and rheological properties, and further activation of receptors. The underlying mechanisms are proposed to be a decrease in salivary lubrication by (1) protein precipitation and (2) changes in the salivary pellicle, and (3) activation of the mechanoreceptors (Gibbins & Carpenter, 2013). These mechanisms will be further explained below.

The first stage of the friction-based theory is the formation of aggregates between the "flowing" salivary proteins and polyphenols. PRPs have been reported to effectively form insoluble complexes with polyphenols (Brandão et al., 2017; Ployon et al., 2018; Sarni-Manchado et al., 2008). Furthermore, other proteins like histatins and statherin have a strong binding affinity with polyphenols (Brandão et al., 2017; Soares et al., 2011). These proteins are present in the flowing saliva, providing the first defense line against the polyphenol's interaction with salivary proteins. When the concentration of polyphenols exceeds the binding capacity of salivary proteins of the flowing saliva, the remaining polyphenols will interact with the mucins in the mucosal pellicle. The aggregation of mucins by the remaining polyphenol affects the integrity of the salivary film. The astringents disrupt the lubricious mucin coating on the oral mucosa, exposing oral receptors and increasing friction by decreasing the lubricity of the mucosal pellicle upon aggregation of the mucins (Ranc et al., 2006). It is suggested that tannins and positively charged compounds induce mucin aggregation by crosslinking (Davies et al., 2014; Gibbins et al., 2014). This reduces the lubrication ability of the mucins, as aggregated mucin cannot provide lubrication. These variations in the mucosal pellicle change the lubrication behavior of saliva (Laguna et al., 2017). The lost ability of salivary proteins to provide

lubrication causes a friction increase in the oral cavity, which elicits astringency via the activation of the mechanoreceptors (Ma et al., 2016; Nayak & Carpenter, 2008).

The interaction of polyphenols and salivary proteins occurs in three steps (Charlton et al., 2002; Jöbstl et al., 2004). The first step is the interaction between the hydrophobic phase (via π-π interaction) of the aromatic rings of the polyphenols and the hydrophobic sites (pyrrolidine ring of the proline residues) of the salivary proteins (soluble complex). The peptides (carboxyl and $-NH_2$ groups) are cross-linked with polyphenols (hydroxyl group), by cooperating hydrogen bonding. In the second step, the protein-polyphenols complex self-associates via hydrogen bonding to form soluble, large complexes and aggregates. In the last step, the aggregates are large enough to induce phase separation.

Additionally, there are some other important parameters that influence the formation of the aggregates between polyphenols and salivary proteins. These parameters include the ratio of protein types: polyphenols, molecular weight, pH, temperature, ionic strength and the type of polyphenols (Bajec & Pickering, 2008). Lastly, the degree of polymerization, galloylation, interflavanic bond, and the number of hydroxyl groups present in the B-ring of the flavanic nucleus are important additional parameters that influence aggregate formation (García-Estévez et al., 2018) which lead to changes in friction and therefore astringency perception.

Coating sensation

The coating sensation is caused by a residual film (oral coatings and saliva). This film sticks onto the oral surface after swallowing, giving lower friction. Oral coatings are known as the residuals of food and beverages that coat the oral mucosa after food consumption. Oral coatings (fats, proteins, and carbohydrates) have the ability to act as lubricants between the mouth surfaces in the oral mucosa. A higher amount of oral coating decreases the frictional forces between mouth surfaces, giving a higher coating sensation.

Many foods and beverages have a coating character, because of the liking of sugar and fat in modern foods and drinks. Nevertheless, there is limited information available on the chemical and physical properties of oral coatings (Camacho et al., 2014). It has been postulated that changes in the structure of the thin layer of adsorbed proteins (salivary pellicle), that coat oral surfaces, are detected by mechanoreceptors. These changes in the oral surfaces are due to the covalent and non-covalent interactions of saliva or mucosa with food ingredients on oral surfaces (Silletti et al., 2007).

Fats lubricate the movement of the food bolus surface along the oral tissue, which decreases perceived dryness and roughness and increases perceived fattiness and creaminess (de Wijk et al., 2009). Additionally, Dresselhuis et al. (2008) explained that the adhesion and spreading of emulsion droplets on the oral surfaces are a combination of electrostatic, steric and hydrophobic interactions, with hydrophobic interactions being the most important. These interactions act as separation distances between emulsion droplets and the surface.

An example of the coating sensation is given by the changes in the emulsion stability during consumption. Emulsions aggregate immediately in the mouth caused by the interactions between emulsion components with salivary proteins. The interaction of saliva-emulsion increases the viscosity of the coating layer while lowering the friction forces between the oral surfaces. These changes in oral surfaces can be detected by the mechanoreceptors located in the tongue surface suggesting the sensations of soft coating, and velvety coating (van Aken, 2010).

Under normal conditions in the mouth, the tongue surface behaves strongly hydrophilic due to the presence of the mucous layer. Van Aken (2010) suggested that increased sensitivity to coalescence promotes the deposition of oil on the tongue surface, which makes the surface more hydrophobic. This increases the retention of emulsified fat on the tongue. To conclude, the coating layer of the aggregated emulsion droplets is responsible for the feeling of a velvety coating as mentioned above.

Finally, several factors such as the type and concentration of fats, polysaccharides, and proteins, the physicochemical properties of food and the morphology of the tongue are hypothesized to influence the oral coating formation (Camacho et al., 2014, 2015; de Wijk et al., 2009; Dresselhuis et al., 2008; Fleury et al., 2002; Lynch et al., 1993; Madrigal-Galan & Heymann, 2006; Pivk et al., 2008; Vingerhoeds et al., 2009). In literature it has been found that mouthfeel sensations such as creamy, coating after-feel, fatty and slippery increase with higher oil content in emulsions (van Aken, 2010), while the addition of thickener in emulsions can also form a layer similar to oil on oral surfaces or imitate an oil layer, giving the coating sensation (van Aken, 2010; Vingerhoeds et al., 2009).

Saliva characteristics and ageing

It is reported that taste and smell loss is associated with ageing. Texture and mouthfeel perception has been shown to vary due to age as well. Older adults perceive soups as less creamy, sweet, fatty and elastic and dairy products as more astringent compared to younger persons (Kremer et al., 2005; Kremer et al., 2007; Withers et al., 2013). Apparently, changes in oral physiology due to age result in altered oral sensations.

Age is responsible for changes in salivary properties such as saliva flow, calcium and mucin content and ionic concentrations. Those changes over time affect the quantity and quality of saliva (Xu et al., 2019). A significant reduction of salivary flow in older adults generally occurs (Affoo et al., 2015). Elderly individuals have lower resting and stimulated salivary flow compared with younger adults (Vandenberghe-Descamps et al., 2016). The reduced salivary flow is commonly associated with decreased lubrication, protection, oral clearance, mucosal surfaces hydration and coating abilities in the oral cavity (Chaudhury et al., 2015; Lee et al., 2002). Those changes have been reported to negatively impact the nutritional status and alter the sensory perception (Norton et al., 2021). Interestingly, stimulated saliva is influenced less by age (Affoo et al., 2015), this may limit changes from salivary flow following food consumption and therefore sensory perception, but further research is needed. Currently, little is known about how mouthfeel changes with age. Many researchers suggested the importance

of mouthfeel to compensate for taste and smell loss in older adults. The age-dependent loss of both quality and quantity of saliva decreases hydration properties, increases the ionic concentration and viscosity of saliva. The formation of an optimal pellicle will thus be hampered (table 2).

Table 2. Age-, disease- and drug-induced effects on saliva. Drugs are classified as compounds with mild, moderate or severe anticholinergic activity. The changes may hamper the formation of an adequate salivary pellicle.

CHANGES IN SALIVA

Age	**Disease**	**Medication**		
Salivary flow rate ↓	HIV/AIDS	Drugs on the Anticholinergic Burden Scale		
Ionic concentration ↓	Sjőgrens syndrome	Mild	Moderate	Severe
Hydration↓	Sarcoidosis	e.g., Diazepam	e.g., Carbamazepine	e.g., Amitriptyline
Viscosity↑	Rheumatoid arthritis	Furosemide	Meperidine	Atropine
Mucin degradation↑	Diabetes	Metoprolol	Pethidine	Clozapine
Absorption ability↓	Radiation therapy	Ranitidine		Nortriptyline
	Chemotherapy			Prometazine
				Scopolamine

Oral tribology using a smooth in vitro PDMS surface might be a promising method to quantify friction and lubrication of mixtures of food and saliva of elderly individuals. Subsequently, well-designed in vivo sensory studies will enable alignment of these in vitro data with perceived mouthfeel.

Influence of medication

Despite the increased use of medication associated with ageing, several connected features do not receive adequate attention. Ageing for example changes the pharmacology of drugs (Bast & Drent, 2022). Knowledge of this item exists but is not used to personalize and thereby optimize pharmacotherapy. Another well-known phenomenon is the so-called anti-cholinergic accumulation (Bast & Drent, 2022). Many drugs have an anticholinergic component in their action (Dinh et al., 2021; Mate et al., 2015). Obviously, anticholinergic agents display this effect, but also drugs that are not known as anti-cholinergic drugs show this characteristic. Simultaneous use of multiple drugs, so-called polypharmacy, has been suggested to also lead to more anticholinergic activity. This anticholinergic accumulation may lead to signs with are often regarded as typical age-related symptoms like physical and cognitive impairment. Common anticholinergic effects like blurred vision, cardiac effects, and dry mouth are also

well-known in this regard. Changes in saliva production and characteristics due to multiple drug use can occur. In this way, drug-induced changes in taste can occur during ageing (Proesmans et al., 2019). This may impact food intake and thus influence the condition of the patient. To classify drugs with anticholinergic effects the so-called Anticholinergic Burden Scale (ABS) has been developed (Proesmans et al., 2019). The ABS is primarily focused on the prevention of cognitive impairment in elderly receiving multiple drugs (polypharmacy). In the ABS drugs with mild, moderate and severe anticholinergic activity are discerned (table 2). It would be advisable to investigate whether this scale could also be used in relation to mouthfeel. Drug-induced aberration of mouthfeel might be prevented in this way. More knowledge of this subject is imperative.

Xerostomia

Oral health, nutritional status and taste can be negatively influenced by saliva-related diseases such as xerostomia. This syndrome results in an absence of saliva which causes eating difficulties and changes taste perception (Muñoz-González et al., 2018). Researchers indicate that a large proportion of the elderly has diminished salivary secretion (xerostomia), either due to the effects of medication or age-related physiological changes (Nagler & Hershkovich, 2005). Such changes could also alter the viscoelastic properties of saliva in the elderly since the reduced lubricating ability is characteristic of xerostomia.

Salivary differences between individuals showed an age-related reduction in salivary flow rate, accompanied by an increase in salivary viscoelasticity and protein content. The increased salivary viscoelastic behavior is an effect of the reduced water content, which leads to increased salivary protein concentration (Nagler & Hershkovich, 2005). Additionally, a significant difference between the viscoelastic properties of the parotid and submandibular/sublingual saliva has been observed. The viscoelastic differences between the salivary glands suggest a potential difference in the salivary proteins profiles, where mucins and glycoproteins are more present in submandibular/sublingual saliva (Aguirre et al., 1989; Nagler & Hershkovich, 2005).

As mentioned, the rheological and lubrication function of saliva is highly related to the mucin-type and concentration (Inoue et al., 2008). Interestingly, previous researchers observed that despite the increase in protein content of mucins, functionality and degree of glycan/protein proportion decreased among patients with drying mouth (xerostomia) (Chaudhury et al., 2015; Lai et al., 2009; Veeregowda et al., 2012). Chaudhury et al. (2015) proposed that the salivary lubrication loss was correlated to negatively charged sialylated residue loss. This decrease reduces water retention and less hydrated mucins which reduce the lubrication ability via hydration (Chaudhury et al., 2015). Therefore, a change in mucin glycosylation is the responsible mechanism for altered saliva rheological behavior for people suffering from dry mouth syndrome. The alteration of mucin structure in elderly people influences the perception of flavor and should therefore be considered an interesting topic for further investigation. Better insights could help develop food products for people suffering from dry mouth syndrome.

Cancer

Saliva flow and composition can be negatively impacted by cancer treatments in a variety of ways. Radiation therapy to the head and neck region can result in salivary gland hypofunction to varying degrees depending on the cumulative radiation dose to the salivary gland tissue (Jensen et al., 2010). With radiation therapy, it is anticipated that a lethal dose of radiation will be delivered only to the tumor area, causing a minimal amount of radiation exposure to surrounding tissues. It is however impossible to avoid radiation affecting salivary glands, oral mucosa, and jaws since they fall within the blast radius. As a result of this, irreversible damage to the gross architecture is caused by a gradual replacement of ductal remnants and fibrous tissues with lymphocytes and plasma cells, resulting in a loss of salivary fluid secretion (Wong, 2014). This results in saliva becoming "sticky, thick, and viscous". An increase in saliva viscosity could interfere with the transport of taste stimuli to taste receptors, resulting in a reduction in the perception of taste stimuli (Comeau et al., 2001; Wong, 2014). Additionally, the high viscous saliva results in xerostomia, taste dysfunction and dysphagia.

Conclusion

This review highlights the important role of saliva in mouthfeel sensations. Saliva has undoubtedly a great influence on mouthfeel sensations. Salivary properties are associated with the presence of glycosylated proteins like mucins and gPRPs. Changes in the viscoelastic behavior of saliva arise the tactile sensations. The change in the salivary physical properties is covered by the interactions between salivary proteins and food components. More insights into the interactions between salivary proteins and food components are essential for understanding the mechanisms behind mouthfeel sensations. Additionally, physiological parameters influence saliva integrity and properties which lead to different mouthfeel sensations among individuals. The development of instrumental analyses for measuring this impact of food components on saliva properties could potentially be the key to the further development of predicting mouthfeel models. With existing knowledge, simple gastronomical features could already be used for the elderly. Proteins (e.g., milk) in black tea can mask the astringent effect of polyphenols or citric acid with tea might be used to stimulate salivary flow. With the ageing population new gastronomical challenges arise.

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Lubrication behavior of ex-vivo salivary pellicle influenced by tannins, gallic acid and mannoproteins

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Abstract

The objective of this study was to investigate the influence of tannins and gallic acid on the salivary lubrication behavior. Furthermore, the effects of pH and mannoproteins in combination with gallic acid on the lubrication of saliva were studied. The addition of gallic acid and tannins were found to increase friction caused by the removal of the saliva film. Tannins resulted in higher friction compared to gallic acid. Lowering pH increased friction of gallic acid mixtures with saliva, due to stronger interactions between gallic acid and saliva. The increased friction caused by gallic acid was inhibited by the addition of mannoproteins due to the hydrogen bond interactions between gallic acid and mannoproteins, thereby decreasing the complex formation between gallic acid and salivary proteins. A correlation of 0.96 was found between the hydrodynamic diameter of the aggregate and the delta friction suggesting that the formation of aggregates determined the lubrication behavior.

Introduction

Mouthfeel plays a significant role in the sensory experience of alcoholic or nonalcoholic beverages, like wine, beer, or tea (Gawel et al., 2018). In beverages, mouthfeel contributes to the perceived texture and taste perception. This sensation arises partly from the influence of polyphenols on the integrity of the saliva film that coats oral mucosa (Laguna et al., 2017).

Astringent mouthfeel is an important indicator of the sensory quality of a wide range of beverages (Bajec & Pickering, 2008). Astringency is commonly associated with a drying mouthfeel, even though it is a very complex sensation with various definitions and origins (receptor and lubrication based). Astringency perception in beverages has been associated with plant-based polyphenols (Gawel et al., 2018). These polyphenols interact with salivary proteins through non-covalent interactions and result in the depletion of the lubricating salivary film (Rudge et al., 2021). Loss of salivary lubrication has been reported to be related to the astringency sensation (Bajec & Pickering, 2008). When mucins and proline-rich proteins have been suggested as the most important salivary proteins for oral lubrication (Rudge et al., 2021).

Tannins and gallic acid are some of the main phenolic components that interact with salivary proteins. Gallic acid is known to contribute to the astringency of beverages like red wines, while tannins are perceived as more astringent components compared to gallic acid (Frank et al., 2011; Hufnagel & Hofmann, 2008). Since tannins contain larger molecules than gallic acids such as catechins, ellagitannins etc., the molecular weight influences the astringency perception (Ma et al., 2016). Additionally, phenols concentration and the wine matrix (like ethanol, pH, and polysaccharides) are important parameters for the impact on the astringency perception (Gawel et al., 2018).

The mechanisms behind astringent perception are not completely known and are still under debate. At the moment, two mechanisms have been suggested to explain astringency sensation, the receptor and friction-based mechanism (Canon et al., 2021). The most discussed one is the friction-based mechanism, by which astringency is explained by the interaction of tannins with specific salivary proteins (Charlton et al., 2002). This theory suggests that mucins, proline-rich

proteins (PRPs) and histatins readily interact with tannins (Bennick, 2002). The mucus layer in the mouth consists of salivary proteins that are non-covalently bound to oral mucosal cells. Green (1993) suggested that the astringent sensation is mainly caused by the interaction of polyphenols with salivary proteins forming insoluble polyphenols-proteins precipitates (via cross-linking) in the mouth. Those aggregates increase the friction in the oral cavity and reduce the lubrication of the salivary film, giving an astringent/dry sensation. The interaction of polyphenols and salivary proteins is suggested to happen in three stages (Jöbstl et al., 2004). The first stage is the interaction between the aromatic rings of polyphenols and the hydrophobic sites (pyrrolidine ring of the proline residues) of the salivary proteins to form soluble complexes. In the second stage, the protein-polyphenols complexes self-associate via hydrogen bonding to form larger soluble complex aggregates. The peptide groups (carboxyl and -NH2) of the salivary proteins are cross-linked with the addition of extra polyphenol hydroxyl groups via hydrogen bonding. In the last stage, these complex aggregates grow further until they become insoluble, and are large enough to precipitate and induce phase separation.

The formation of the above-mentioned aggregates between polyphenols and salivary proteins depends on a variety of parameters, such as the ratio between proteins and polyphenols, pH, temperature, ionic strength, and the type of polyphenol (Bajec & Pickering, 2008). Even though the formation of aggregates, which leads to loss of lubrication, is considered the main precursor to astringency sensations, researchers have suggested that astringency can also arise without the occurrence of such interactions. This supports that astringency is a more complicated sensation that depends on more than one single physical or chemical mechanism (Rossetti et al., 2009). Therefore, a receptor-based theory suggests the astringency sensation can be origin from direct interaction with the salivary proteins adhered to buccal mucosal cells (Canon et al., 2021).

The beverage matrix is a rich composition of many elements. Depending on the composition, the matrix can induce or reduce astringent perception. For instance, polysaccharides or proteins in wine (like mannoproteins and arabinogalactan) may influence the astringency sensation as they have been shown to disrupt the interaction and aggregation between salivary proteins and polyphenols (Watrelot et al., 2017). The pH is another astringency modulator that may affect the interactions. Previous studies have shown that the astringency sensation increases (Gawel et al., 2018). This increase in astringency sensation may be related to an increase in phenol groups, which may form hydrogen bonds with salivary proteins (Rudge et al., 2021).

An emerging tool that helps understanding oral processing is tribology (Stokes et al., 2013). Tribology is the science of wear, friction and lubrication and includes how interacting surfaces and other tribo-elements behave in relative motion. Soft-tribology can mimic aspects of in-mouth lubrication by applying human saliva and soft surfaces. The use of soft-tribology helps to study mouthfeel characteristics (such as astringency) by simply monitoring the friction change. Previous studies related to the mouthfeel properties of tea, milk, and wine have used tribology to scale the astringency sensations (Laguna et al., 2017). Even though lubrication behavior is important for the understanding of astringency perception, limited studies have focused on the mechanisms behind lubrication losses.

The present work aims to investigate the effect of gallic acid and tannins on the lubrication properties of human saliva at different pH levels. Furthermore, the masking effect of mannoproteins in the lubrication properties was tested. The outcome of this study provides new insights into the effect of different conditions on the lubrication properties of human saliva. The outcome of this research has the potential to help the food industry in influencing the beverage matrix and the astringent sensation.

Material and methods

Model solutions

The model solutions (MS) were made based on astringent components that can be found in wine. Samples were prepared with or without the presence of gallic acid (Sigma-Aldrich Corp, St. Louis, MO, USA), tannins (65 % w/w total polyphenols in gallic acid equivalent, 38 % w/w total catechin equivalent Lamothe-Abiet, France), a 15 % w/v mannoprotein solution produced by grape yeast (Lamothe-Abiet, Bordeaux, France), potassium dihydrogen phosphate, and meta-phosphoric acid (Merck Millipore, Darmstadt, Germany). Demineralized water was used as the solvent for all MS. All samples were formulated on the same day as the analysis in duplicate. The concentrations of gallic acid range between 0.5 and 4 g/L. The mannoproteins had a constant concentration of 400 mg/L for all the gallic acid combinations. All samples were covered with aluminum foil and stored at 4 °C for a maximum of 24 hours to prevent degradation. Gallic acid and tannin concentrations were chosen to represent both the presence of compounds as well as the values of the total amount of polyphenols that can be found in wines (Büyüktuncel et al., 2014; Petković et al., 2015).

To investigate the effect of pH, the components of the MS were diluted in demineralized water or a phosphate buffer at pH 3. A phosphate buffer was used instead of tartaric acid since it has been recognized as an astringent compound (Huang & Xu, 2021). The pH 3 was selected since it represents the most acidic beverages. The buffer solution was prepared by complete dissolution of 0.34 % (w/v) potassium hydrogen phosphate in demineralized water. A phosphoric acid solution (1M) was used to adjust the pH to 3.

Saliva collection

It is suggested that there are no fluids capable of mimicking the properties of real human saliva (Stokes & Davies, 2007). Therefore, for both tribological and aggregate formation measurements, fresh unstimulated human saliva was provided by healthy non-smoking donors, four male subjects (age 22-25, Caucasian) after their consent. This physiological group was selected to minimize the variation in the composition of saliva (Xu et al., 2019). This part of the study has been approved by the Ethical Review Committee at Maastricht University [ethics reference (ERCIC_335_23_03_2022)].
The saliva collection followed by a procedure described by Rudge and co-workers (Rudge et al., 2021). To minimize the variation of the human saliva due to circadian rhythms through the day (Dawes, 1975), the collection took place between 8 and 10 AM. Consumption of food was

avoided for at least 2 hours before collection. The saliva was collected without stimulation of the salivary glands (Brossard et al., 2016). During salivating, the first milliliters of saliva were discarded as possible contaminants could still be present in the mouth. The saliva was collected and stored on ice to prevent degradation by enzymes. After collection, saliva was centrifuged at 10000 rpm (9520 g) at 4 °C for 10 minutes to remove remaining debris. The supernatant was stored in ice and used immediately after centrifugation since the viscoelasticity of saliva decreases during storage (Stokes & Davies, 2007).

Tribological measurements

A dynamic tribological approach was used to measure the changes in the frictional coefficient of saliva upon the addition of the MS. All tribological measurements were performed with an Anton Paar Rheometer MCR302 (Austria). A tribology cell (BC12.7/SS 52837) was used to measure the lubrication properties of the samples in combination with saliva. Polydimethylsiloxane (PDMS) pins were used since PDMS is a prevailing material currently used in tribology (Rudge et al., 2021). The friction was measured using a commercial (glass) ball on a three (PDMS pin setup. The glass ball had a diameter of 12.7 mm and PDMS pins a diameter of 6 mm and a height of 6 mm with a modulus of around 2 MPa. Glass ball and PDMS pins were obtained by the rheometer manufacturer.

The measurements were performed in triplicates. A normal force, Fn, of 1 N was applied (Laguna et al., 2017). The experiments were carried out at a constant rotational speed of 1mm/s to gain boundary regime friction profiles, as this regime is believed to be closely related to the perception of astringency in humans (Prakash et al., 2013). Similarly, to the protocol used by Rudge et al. (2021) the measurements were taken within a period of ten minutes, where the first five minutes were used for the salivary proteins to cover the PDMS pins (ex-vivo salivary pellicle). The salivary layer allows the glass probe to slide against the PDMS pins by the addition of 0.5 mL of saliva. When the five minutes passed and a constant friction coefficient was obtained, the MS were added in a 1:1 (saliva: MS) ratio as has been suggested by other researchers (Laguna et al., 2017; Rudge et al., 2021).

An example of measurement is shown in figure 1. With the use of these graphs, the difference in friction coefficient ($\Delta\mu$) was calculated as $Av.CoF_1$ - $Av.CoF_2$, where $Av.CoF_1$ is the friction coefficient obtained when salivary proteins fully covered the PDMS surface, and $Av.CoF_2$ is the friction coefficient after the addition of the MS. All the friction coefficients represent mean values by taking each average value based on five points.

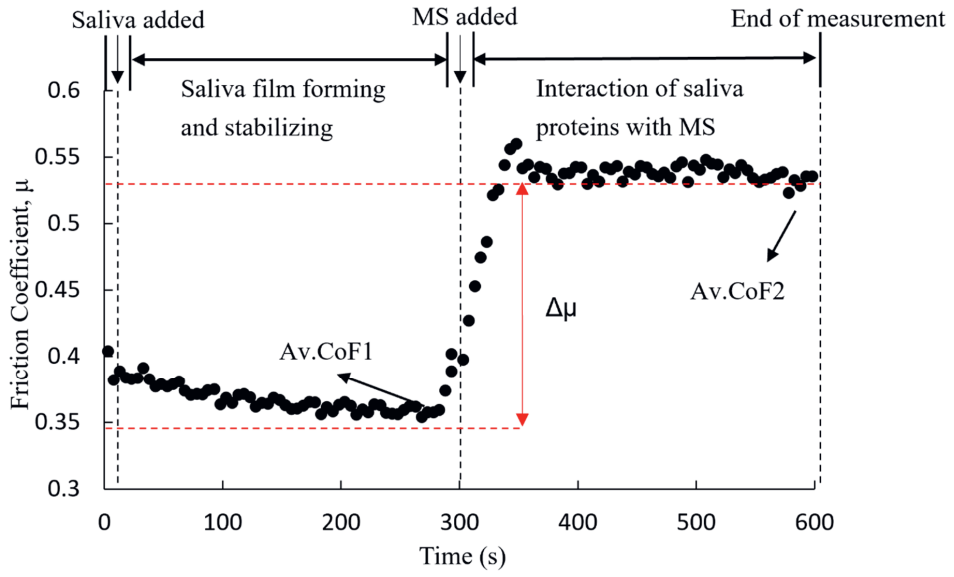

Figure 1. A schematic representation of the friction coefficient, μ, as a function of time obtained by the dynamic protocol. Saliva was added at time 0 of the measurement. After obtaining a constant baseline value as the result of saliva addition ($AvCoF_1$), the model solution (MS) was added after 300 s. The $Av.CoF_2$ was calculated using only the data points after stabilization of the interaction between saliva and MS stabilized.

Zeta-potential and particle size distribution

Zeta-potential and particle size distribution were measured by dynamic light scattering using a Zetasizer Ultra (Malvern Instruments). The particle size distribution was measured in triplicate (173° Backscatter), with a refractive index (RI) of 1.450 for the protein, and an RI of 1.330 for the water. First, pure saliva and astringent compounds solutions were measured. Afterwards, Saliva-MS of 0.9ml (1:1) were mixed in Eppendorf tubes for 5 min at 20 °C. The samples were diluted 100 times prior to measurement. For the zeta potential determination, the samples were tested 3 times at a maximum voltage of 60 mV for a maximum of 100 runs per test. The cuvettes were cleaned between each measurement with demi-water, ethanol, and again demi-water and subsequently dried with pressurized air. The measuring cell was changed every 5 samples because of electrolysis (blackening) of the electrodes.

Statistical analysis

The results were analyzed using a multivariate analysis of variance (MANOVA) and a Pearson correlation. When the values from MANOVA were significantly different ($p < 0.05$), an additional Tukey-Kramer HSD (honestly significant difference) test was used to identify the differences between the parameters. To verify the assumptions of normal distribution and homogeneity of variances, Shapiro-Wilk test and Levene's test were used. All the statistical

analyses were performed by the software R (R core team and foundation for statistical computing), R-studio version 4.0.3 using the R package “agricolae”.

Results and discussion

Differentiation of the influence of tannins and gallic acid on lubrication properties of human saliva

Changes in the lubrication behavior of saliva are associated with interactions between salivary proteins and polyphenols. Gallic acid and tannins are components that are abundantly present in beverages like wine, tea, etc, and are known to provide an astringent sensation. Those components are different in molecular weight, which is expected to influence lubrication. The change in the saliva lubrication behavior upon the addition of gallic acid and tannins was investigated in this study. Different concentrations of gallic acid and tannins were used to identify patterns in the increase of friction. The results are shown in figures 2a and b.

Within 5 minutes, the friction coefficient reached a constant value (baseline) due to the formation of a saliva film on the substrate. The friction coefficient was found to give a constant value of on average 0.37. Such lubrication properties have been found previously, with saliva friction coefficients ranging between 0.25 and 0.35 at low loading forces (Chen, 2009).

The low friction coefficient for saliva is associated with the adsorption of salivary proteins on the PDMS surface. The hydrophobic nature of the PDMS provides good adhesion for the salivary proteins (Carpenter et al., 2019). Upon the addition of the MS, i.e., gallic acid and tannin solutions, the saliva film loses its lubrication properties and friction increases (figures 2a and b). As can be seen, tannins lead to a larger increase in the friction coefficient than gallic acid. The molecular weight of the astringent compounds seems to influence the lubrication properties of saliva. Additionally, for both components, a clear effect of concentration was also observed.

To gain better insights into the effect on the friction coefficients upon the addition of the different astringent agents, the $\Delta\mu$ values after the addition of the MS were determined. These results are given in figure 2c. To examine any water potential effect upon the addition of the MS, demi-water was used as a reference. The addition of demi-water into saliva gave a $\Delta\mu$ value around 0, indicating that water itself did not change the salivary lubrication.

The difference between the two astringent compounds gallic acid and tannins is noticeable (figure 2c). Upon addition of both gallic acid and tannins, $\Delta\mu$ significant increased ($p < 0.001$) with gallic acid and tannins. However, gallic acid solutions gave low values for $\Delta\mu$ of 0.044, 0.078 and 0.141 for concentrations of 1, 2 and 3g/L, respectively, while the $\Delta\mu$ values for tannins were significantly higher at values of 0.349, 0.421 and 0.501 for concentrations of 1, 2 and 3 g/L, respectively. Changes in friction of saliva upon addition of polyphenols were demonstrated by other researchers as well (Watrelot et al., 2017). Additional observation between the tannin and gallic acid solutions was obtained regarding the different rates of the changes in lubrication behavior. Similar to friction changes, the rates were higher in tannin’s presence compared to gallic acid.

The differences in the Δμ values can be explained by the difference in the polyphenol-protein interactions. Those interactions are mainly driven by two types of interactions, i.e., hydrogen bonds and hydrophobic interactions. Hydrophobic stacking interactions occur via the binding sites of the peptides (proline residues) together with the preceding amide bond and amino acid, between the galloyl ring (phenolic compounds) and the pyrrolidine ring face of proline (García-Estévez et al., 2018). Proline residues are the main binding sites of the salivary proteins for the hydrophobic interactions with phenolic compounds. Additionally, the hydrogen bonds form between the hydroxyl groups of the phenolic compounds and the carbonyl and amino groups of the salivary protein groups. The last-mentioned interactions are believed to stabilize the formation of the aggregates between salivary proteins and phenolic compounds (Charlton et al., 2002).

As these interactions are more pronounced for compounds with a larger molecular weight, the difference in Δμ between the gallic acid and tannin solutions can be explained by the molecular weight of the components. As the tannins are larger, they are expected to form more bonds with multiple salivary proteins, i.e., PRPs, mucins, statherin and P-B peptide, which leads to the formation of larger aggregates. Such relation was also suggested by Laguna et al. (2017), who showed that the aggregate size, obtained by interactions between oak tannins and salivary proteins, increased astringency perception. Therefore, the increase in friction observed in this study can be related to an increase in astringency perception upon consumption.

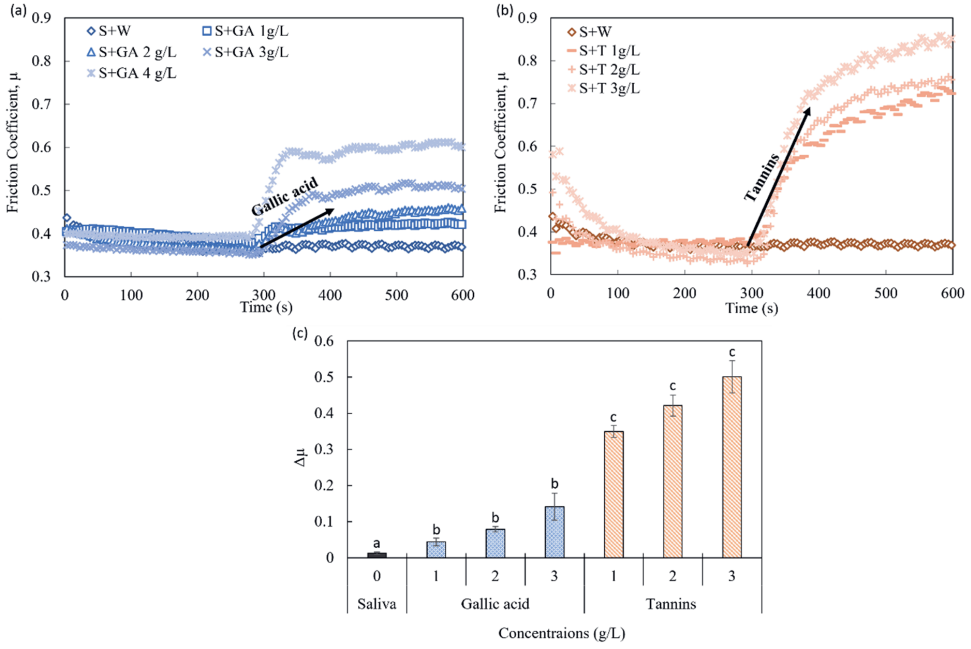

Figure 2. Friction coefficient as a function of time for gallic acid (GA) (a) and tannins (T) (b) in demi-water solution at different concentrations mixed with human saliva in a 1:1 ratio. W refers to water, and S refers to saliva. The friction coefficients represent mean values based on five measurement points. (c) Δμ values as a function of saliva, gallic acid and tannins concentrations. Gallic acid (dotted bars) and tannins (striped bars) solutions were mixed with human saliva in a 1:1 ratio. The values are presented as a mean value ± standard error.

The letters [a-c] in each bar identify that the samples are significantly different according to Tukey's HSD test: $p < 0.05$.

The current findings show that molecular weight affects the absolute change in the friction coefficient, as a result of differences in the interactions between the astringent components and the salivary proteins.

Effect of pH on the lubrication properties of human saliva in combination with a gallic acid series.

Not only is the molecular weight of polyphenols important for astringency perception in beverages, but pH is another parameter that has been found to influence astringency. To investigate the influence of pH on the lubrication behavior, gallic acid was selected, as this astringent compound was found to give more reproducible results than tannins, since the lower solubility of tannins in buffer solution and sample inhomogeneity (data not shown).

The gallic acid solutions were diluted either in demi-water or a buffer solution of pH 3. A significant increase in friction ($p < 0.001$) upon the addition of gallic acid to ex vivo saliva pellicle was noticed for both gallic acid solutions prepared with demi-water and phosphate buffer solutions (figure 3a). In both solutions, the gallic acid caused an increase in the friction coefficient, presented as $\Delta\mu$, in the presence of ex vivo salivary film, as already discussed in the previous section.

The addition of buffer solution alone in ex vivo saliva pellicle was not able to significantly increase the delta friction coefficient, as can be seen in figure 3a. However, in the presence of gallic acid, a clear effect of pH on changes in the friction coefficient can be observed. Addition of gallic acid diluted in a pH 3 buffer gave significant higher $\Delta\mu$ values ($p < 0.001$) than addition of gallic acid diluted in demi-water when added to saliva. Especially for the lowest concentration of GA, the effect of pH was clear. The low concentrations of 0.5 and 1 g/L showed a significant difference between the two solutions. This shows that the effect of pH is especially important for low concentrations of polyphenols. This may be related to differences in the aggregate formation of salivary proteins at these different pH values.

To verify that indeed the aggregate formation was influenced by changes in pH, the size of the aggregates was measured, just as the hydrodynamic diameter, formed by salivary proteins and gallic acid solutions (figure 3b). As can be observed, the hydrodynamic diameter of human salivary proteins was around 127 nm. This value is similar to the findings presented by Laguna et al. (2017). The addition of gallic acid into saliva (1:1) resulted in a significant ($p < 0.001$) increase in the hydrodynamic diameter for all the concentrations in both buffer and demi-water solutions. This increase in size confirms the aggregate formation between the salivary proteins and gallic acid, for which the more pronounced aggregate formation was observed in the buffer solution. This indeed confirms that more aggregation was obtained at lower pH values.

In water, a significant ($p < 0.001$) increase in the aggregate formation was obtained only at or above a concentration of 2 g/L. In buffer solutions, the increase was significant for all concentrations, and the increase in the hydrodynamic diameter seems to be linear with gallic

acid concentrations. At the highest gallic acid concentration of 4 g/L the largest diameter of 809 nm was observed. The aggregate formation of both series was found to correlate with the Δμ values, as shown in figure 3c. The Pearson's correlation between the Δμ and aggregate size for gallic acid was 0.96 ($p < 0.05$) respectively. These results show that higher friction can indeed be associated with aggregate formation. As fewer salivary proteins attached to the PDMS surface are available, no salivary proteins are available to provide lubrication in the oral cavity.

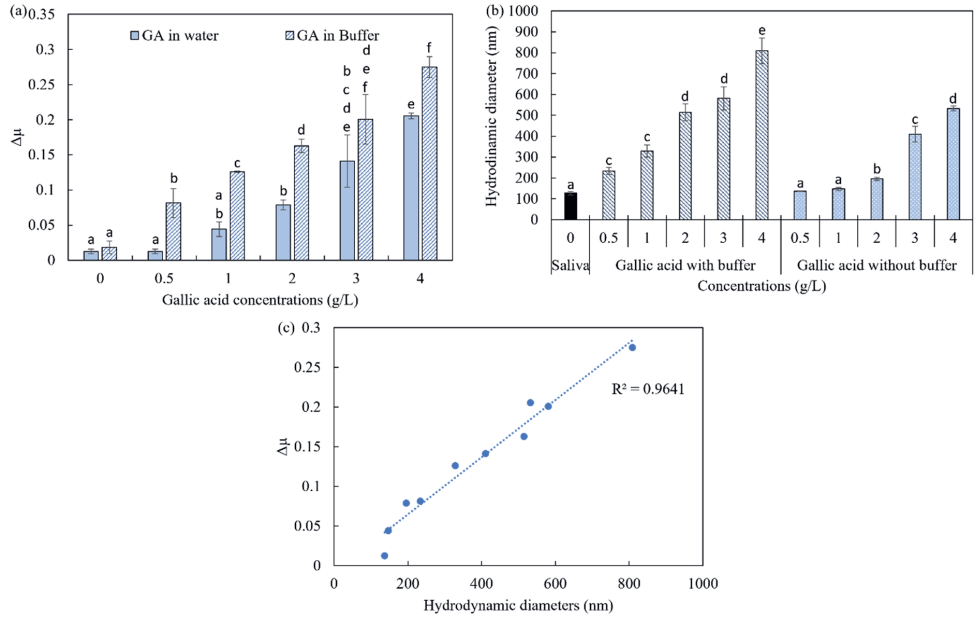

Figure 3. (a) Δμ values as a function of gallic acid concentration for gallic acid (GA) diluted in water (solid bars) and phosphate buffer (pH 3) (striped bars). All the solutions were added in the presence of ex vivo salivary film. (b) The hydrodynamic diameter of saliva–gallic acid aggregates in buffer (striped bar) and without buffer (dotted bars). The values are presented as a mean value ± standard error. The letters $^{a\text{-}e}$ in each bar identify that the samples are significantly different according to Tukey's HSD test: $p < 0.05$. (c) Δμ as a function of hydrodynamic diameter for gallic acid. The line is the best through the data points.

The higher Δμ and larger aggregate size for lower pH values (in buffer) can be explained by the different interactions. Changes in pH will affect the binding affinity by altering the charge and configuration of the protein. Repulsive electrostatic interaction may limit the attractive hydrophobic interactions and hydrogen bond formation. Salivary proteins at physiological pH carry a negative electrical charge (Rykke et al., 1995). Mucins are responsible for this net negatively charge since their isoelectric point is around 2.5 (Veerman et al., 1992). At lower pH values, the electrostatic repulsion between mucins and polyphenol reduces as the charge density of the mucins reduces at pH values close to the iso-electric point. Once the repulsive

interaction decreases, the attractive interactions become more relevant, which induces aggregate formation between proteins and polyphenols.

To investigate the effect of pH on the charge of salivary proteins and gallic acid, the zeta-potential and pH of the resulting protein-gallic acid complexes/aggregates were measured. The pure saliva had a pH of 7.3 and a zeta potential of -15.5 mV. Upon the addition of the buffer solution of pH 3, the pH decreased to a value of 5.8 and the zeta-potential was -15.6 mV (table 1). The phosphate buffer at pH 3 failed to bring the pH to a value of 3 due to the high buffering ability of saliva (Bardow et al., 2000). The addition of gallic acid decreased the pH values in the buffer solution. The pH values of the mixtures (saliva-MS) decreased from 5.8 to 3.9 when the gallic acid concentration was increased to 4 g/L. Because of the pH reduction, the zeta-potential of the mixtures changed from -15.6 to -13.9 mV. In water, the gallic acid solutions did not decrease the pH for low concentrations, but only at high gallic acid concentrations (3 and 4 g/L), the pH change was substantial: the pH decreased from 7.3 without gallic acid to 4.7 with 4 g/L gallic acid. Also, the zeta-potential was only slightly lower for higher concentrations of gallic acid. Based on these differences in the zeta-potential between the MS in water and buffer the conclusion was drawn that the salivary proteins become less charged at lower pH values. Less negatively charged proteins could potentially aggregate easier due to less electrostatic repulsion between gallic acid and proteins. The lower net electrical charge in the buffer solutions confirms the more aggregate formation between salivary proteins and polyphenols. This contributes to the higher loss of salivary lubrication behavior.

Table 1. pH and zeta-potential values of pure saliva or mixtures of saliva with model solutions with and without buffer solutions.

pH 3 buffer	**Mixture**	**pH**	**Zeta-potential (mV)**
Without buffer	Saliva	7.3 ± 0.2	-15.5 ± 0.1
	Saliva GA (0.5 g/L)	6.8 ± 0.1	-15.8 ± 0.6
	Saliva GA (1 g/L)	6.4 ± 0.1	-15.5 ± 0.1
	Saliva GA (2 g/L)	6.0 ± 0.1	-14.6 ± 0.1
	Saliva GA (3 g/L)	4.9 ± 0.1	-13.9 ± 0.7
	Saliva GA (4 g/L)	4.7 ± 0.1	-13.8± 1.2
Buffer	Saliva	5.8 ± 0.1	-15.6 ± 0.1
	Saliva GA (0.5 g/L)	5.4 ± 0.1	-13.9 ± 0.9
	Saliva GA (1 g/L)	4.6 ± 0.1	-14.0 ± 1.4
	Saliva GA (2 g/L)	4.4 ± 0.1	-13.9 ± 0.9
	Saliva GA (3 g/L)	4.1 ± 0.1	-13.6 ± 0.3
	Saliva GA (4 g/L)	3.9 ± 0.1	-13.9 ± 0.3

Remark: The values are presented in mean value ± standard deviation.

Although the overall charge of the complexes/aggregates decreased, the charge of the different proteins in the saliva is not the same, due to differences in their structure. Mucins contribute

up to 30 % of the proteins present in saliva, while PRPs contribute the other 70 % (Carpenter et al., 2019). PRPs can be categorized into basic, acidic (bPRPs), acidic (aPRPs) and glycosylated (gPRPs), of which the basic and glycosylated have the highest proline content. The isoelectric point of mucins is around 2.5 (Veerman et al., 1992), of acidic PRPs around 4.5 (McArthur et al., 1995), of bPRPs around 9.5, and that of gPRPs around 7 (Boze et al., 2010). At physiological pH (around 7), mucins and acidic PRPs are thus responsible for the negative charge of saliva. However, at lower pH values, the acidic PRPs become more neutral, and the charge of mucins also decreases (Veerman et al., 1992). In addition, gallic acid has a pKa value of around 4.4. At lower pH values, gallic acid therefore also becomes less charged, which was also confirmed by the lower zeta-potential of -2.1 mV when present in the buffer solution.

As both gallic acid and mucins became less charged at lower pH, the electrostatic repulsion was minimized, and attractive hydrogen bonds and hydrophobic interactions became more pronounced. These interactions increased aggregate formation and resulted in higher Δμ values in most acidic conditions. In contrast, for higher pH values, a higher charge provided more electrostatic repulsion and thus less aggregate formation, resulting in a lower change in the friction coefficient.

Other studies have also shown the influence of pH on saliva protein-polyphenol interactions (Rinaldi et al., 2012). Those studies focused mostly on the impact of pH on aggregate formation and astringency perception. However, few studies tested the effect of pH on the lubrication properties of saliva (Vardhanabhuti et al., 2011; Wang et al., 2020). In a study by Vardhanabhuti et al. (2011), the effect of β-lactoglobulin on the aggregation behavior at two different pH values (3.5 and 7) was studied. They observed that the changes in pH influence the interaction between β-lactoglobulin and salivary proteins; at pH 3.5, more aggregate formation was observed and higher friction. In the presence of tannins, Wang et al. (2020) showed that a faster collapse of the salivary pellicle was obtained at lower pH. However, Wang et al. (2020) did not find a significant difference in the resulting friction coefficients between different pH values. Those friction-based studies demonstrate the further need for a better understanding of the mechanisms behind the changes in salivary lubrication.

The results of this research provide new insights on the salivary lubrication loss upon the addition of gallic acid influenced by pH. Lower pH in combination with gallic acid leads to more aggregate formation. A linear relationship was found between concentration and Δμ values in low pH. Those changes in friction due to pH are in line with already mentioned studies related to higher astringency perception and aggregate formation due to lower pH. Lastly, it can be concluded that the extent of lubrication loss is not only due to the size of the polyphenols but also to parameters that directly influence the aggregate formation, such as pH.

Masking effect of mannoproteins

Astringent components are known to alter the lubrication of saliva by aggregating salivary proteins. However, components like mannoproteins can provide a “masking” effect by

inhibiting the aggregation, and thus reduce astringent perception (Wang, Wang, et al., 2021). Mannoproteins are known to disrupt the interaction between salivary proteins and polyphenols (Watrelot et al., 2017). However, it is not known how mannoproteins influence the lubrication behavior of saliva. As aggregate formation and lubrication losses were more pronounced in a buffer solution of pH 3, it was investigated whether mannoproteins would be able to reduce this effect under these conditions.

Mannoproteins are proteins located in the outermost layer of the yeast cell wall. These proteins are naturally present in wine and known to mask astringency. Mannoproteins are glycoproteins with a polysaccharide backbone representing about 80 % of the molecule, which is highly abundant in mannose monomer residues (Gonçalves et al., 2002). About 20 % of the molecule consists of protein residues, which are linked to the polysaccharide part via amide bonds at asparagine amino acid residues, or ether bonds at serine or threonine residues (Moreno & Peinado, 2012).

The addition of mannoproteins solution (400 mg/L) itself resulted in a higher friction coefficient (0.45) compared to saliva (0.37). The effect of the addition of mannoproteins to gallic acid systems regarding the Δμ values is shown in figure 4a. The mannoproteins had a constant concentration of 400 mg/L for all the gallic acid combinations. This concentration was chosen to represent the concentration of mannoproteins in wine (Wang, Olarte Mantilla, et al., 2021). The addition of mannoproteins in the gallic acid solutions gave a significant decrease in the delta friction values ($p < 0.001$). It seems that at lower concentrations of gallic acid of 0.5 and 1.0 g/L, the mannoprotein was able to provide stronger masking of friction, i.e., lower values of Δμ, than at higher concentrations of gallic acid. This indicates that there is a specific amount of binding affinity of mannoproteins to gallic acid.

These results indeed indicate the ability of mannoproteins as inhibitors for lubrication loss. However, two main mechanisms have been suggested to explain the reduction of astringency by mannoproteins and polysaccharides. The first mechanism suggests the formation of protein/polyphenol/mannoprotein ternary soluble complexes (Manjón et al., 2020; Ramos-Pineda et al., 2018). The other mechanism states that only interactions between polyphenols and mannoproteins occur (Brandão et al., 2017). To investigate the exact mechanism that occurs in this study, the hydrodynamic diameter (aggregate size) and the zeta-potential of the MS with and without mannoproteins was measured.

Figure 4b presents the hydrodynamic diameter (nm) for pure saliva, different concentrations of gallic acid and the addition of mannoprotein in gallic acid solutions with different concentrations. The diameter showed a similar trend with Δμ (figure 4a), for which the addition of gallic acid increased the diameter, while with the addition of mannoproteins, the diameter was smaller.

The addition of mannoprotein resulted in a significant ($p < 0.001$) decrease in the hydrodynamic diameter. For lower gallic acid concentrations of 0.5 and 1 g/L, this effect was minimal, but the decrease of the aggregated salivary proteins became more noticeable at 2 g/L. At this concentration, the aggregates of gallic acid-salivary proteins had a diameter of 515 nm without mannoprotein, but this size decreased to 292 nm with the addition of mannoprotein. The

decrease of the hydrodynamic diameter was again smaller for a concentration of 4 g/L, as the original size of 809 nm decreased to 719 nm only. Although again a relation could be observed between Δμ and the hydrodynamic diameter, the correlation was less pronounced (0.873).

It was observed that at low gallic acid concentrations, the addition of mannoproteins did not significantly increase the diameter of the aggregates. Contrary, Δμ was significantly reduced at low gallic acid concentrations by the addition of mannoproteins. Additionally, at higher gallic acid concentrations, both the aggregate diameter and Δμ values were slightly smaller upon the addition of mannoproteins. Therefore, the prevention of salivary lubrication loss, induced by mannoproteins, can be explained partly by the aggregate size but also by the interactions between gallic acid and mannoproteins. Those results suggest that smaller aggregate sizes induce less friction in the system. It seems that in this particular case the phenol and the mannoprotein interact with each other before aggregating salivary proteins through hydrogen bond interactions. This can explain the similar aggregate size but lower friction at low gallic acid concentrations with and without the addition of mannoproteins. The current outcome suggests that the second-mentioned masking mechanism (interaction between phenol and mannoproteins) applies.

The masking ability of mannoproteins can be further explained by the affinity of mannoproteins to bind with phenolic components, with or without salivary proteins. Since gallic acid and mannoproteins interact with each other via hydrogen bond, there are less “free” gallic acid molecules that can interact with the salivary proteins. Therefore, when the gallic acid concentration exceeds the limit of the binding ability of mannoproteins, the remaining “free” gallic acid forms aggregates with the salivary proteins, which leads to the decrease of the salivary lubrication properties. For a better understanding of the binding between mannoproteins, gallic acid and/or salivary proteins, the zeta-potential and pH values of the samples were measured.

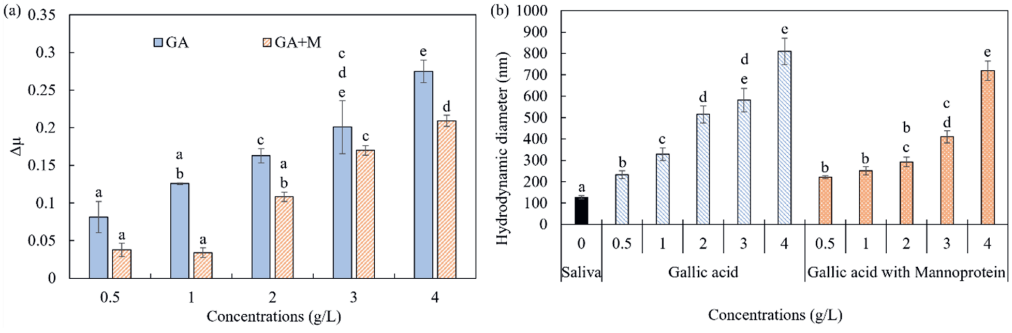

Figure 4. (a) Δμ values as function of gallic acid concentration for gallic acid (GA) (solid bars) and gallic acid plus mannoproteins (M) (striped bars). All the solutions were added in the presence of an ex vivo salivary film. (b) Hydrodynamic diameter of pure saliva (solid bar), saliva - gallic acid combinations (striped bars) against saliva - gallic acid – mannoproteins (dotted bars). All the solutions were diluted in pH 3. The values are presented as a mean value ± standard error. The letters $^{a\text{-}e}$ in each bar identify that the samples are significantly different according to Tukey’s HSD test: $p < 0.05$.

The addition of mannoprotein in gallic acid-saliva solutions gave a slight increase in the pH values of the solutions compared to gallic acid-saliva solutions without mannoproteins (table 2). The pH values for solutions with mannoproteins varied between 6.1 and 4.0 for concentrations of gallic acid of 0.5 and 4 g/L respectively, whereas pH values without mannoproteins varied between 5.4 to 3.9. Additionally, the zeta-potential values were slightly affected by the addition of mannoprotein (table 2). The zeta-potential values in the mannoproteins solutions were lower compared to the values without mannoproteins. This difference can be explained by the negative charge of the mannoproteins since mannoproteins in buffer pH 3 had a zeta-potential of -4.1 mV. This shows that the presence of mannoproteins lowers the zeta potential of the gallic acid solutions, but the effect was minimal.

The small differences in the zeta-potential suggested that the electrostatic interactions are not dominant for the binding inhibition between salivary proteins and gallic acid via mannoprotein. The smaller aggregate formation is therefore not related to electrostatic effects but must be related to other interactions. The main interactions that facilitate the lower binding between gallic acid and salivary proteins are more related to hydrogen bond formation or hydrophobic interactions. It is likely that the mannoproteins can bind to the gallic acid molecules through both types of interactions, which then prevents the gallic acid molecules from binding with the salivary proteins. This would thus explain the decrease in the hydrodynamic radius of the aggregates.

Table 2. Zeta-potential and pH values after mixing with human saliva (1:1) with different concentrations of gallic acid (buffer pH 3) with and without mannoproteins (1:1).

Mannoproteins (g/L)	**Gallic acid (g/L)**	**pH**	**Zeta-potential (mV)**
0	0.5	5.4 ± 0.1	-13.9 ± 0.9
	1	4.6 ± 0.1	-14.1 ± 1.4
	2	4.4 ± 0.1	-13.9 ± 0.9
	3	4.1 ± 0.1	-13.6 ± 0.3
	4	3.9 ± 0.1	-13.9 ± 0.3
0.4	0.5	6.1 ± 0.1	-15.7 ± 0.1
	1	5.4 ± 0.2	-15.1 ± 0.6
	2	4.6 ± 0.1	-13.8 ± 0.1
	3	4.4 ± 0.1	-15.1 ± 1.5
	4	4.0 ± 0.1	-14.4 ± 0.3

Remark: The values are presented in mean value ± standard deviation

The hydrogen bonds form between the hydroxyl-groups of the gallic acid with the oxygen from the sugar linkages of the mannose, which account for 80 % of the mannoproteins (Casassa, 2017). Hydrophobic interactions may occur as well but are expected to be less prominent. As the mannoproteins consist of only 20 % of proteins, of which only approximately 9 % of the

amino acid residues of yeast mannoproteins have an aromatic ring (Liu et al., 2015), the binding affinity through hydrophobic interactions is expected to be limited. This indicates the importance of hydrogen bonds for the aggregate formation between gallic acid and (manno)proteins.

The results of this experiment indicate that mannoproteins can inhibit the binding between gallic acid and salivary proteins, as the addition of mannoproteins resulted in less aggregate formation. The formation of ternary protein/gallic acid/mannoprotein complexes is thus less likely to occur. The inhibition of the aggregate formation allows salivary proteins to lubricate the oral surfaces. This results in lower Δμ values, even though large aggregates are still obtained. Especially at low concentrations of gallic acid, where mannoproteins can bind almost completely to all gallic acid molecules. The masking mechanism is likely to be caused by the complex formation of gallic acid and mannoproteins only, which leaves the salivary proteins available for lubrication. This is in contrast with the different studies that suggest that ternary complexes are formed. This shows that the masking effect of mannoproteins may vary with different phenolic components. This can be due to differences in the molecular structure, as this may influence the affinity of mannoproteins to bind with phenolic components, which may determine whether they only aggregate with the astringent components, or also with saliva together.

These findings give new insights into the masking effect of mannoproteins on gallic acid-induced lubrication loss. The influence of mannoproteins on the aggregate formation between gallic acid and salivary proteins was revealed. The current results are in line with sensory studies, and thus indicate the importance of salivary lubrication behavior in understanding astringency perception. The involvement of other wine components should be further investigated. This would provide a better understanding of the mechanism behind the "masking" effect of friction.

Conclusion

Lubrication properties of saliva, during food consumption, are known to be correlated with astringency sensation. This study investigated the effect of tannins and gallic acid on the lubrication properties of saliva. Tannins and gallic acid were found to reduce salivary lubrication. Tannins lead to a significantly higher increase in friction than gallic acid. pH was shown to have a significant effect, as the charge of the components determined the degree of aggregation. Less electrostatic repulsion between salivary proteins and polyphenols at lower pH values increased the degree of aggregation, which was shown to be linearly related to changes in friction.

Mannoproteins provided a masking effect for the lubrication loss. Mannoproteins showed affinity to bind with astringent components to create gallic acid-mannoprotein complexes. The addition of mannoproteins, therefore, inhibited the aggregation of salivary proteins with gallic acid, thereby providing enough salivary lubrication ability. The changes in salivary lubrication losses could further link to astringency perception.

This study gives insights into the interactions between salivary proteins and polyphenols that occur during food and beverage consumption. The current outcomes show that instrumental lubrication analysis can be a valuable tool for investigating mouthfeel sensations. This is important for the design of new products or product reformulation.

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How phenols affect the mouthfeel of beverages

The effect of cations and epigallocatechin gallate on in vitro salivary lubrication

Submitted

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Instrumental classification of beer based on the mouthfeel

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Abstract

Beer is one of the most consumed alcoholic beverages in the world. Classification of beer helps the consumer to find a preferred beer. Sensory assessments of taste are commonly done by sensory panels and therefore susceptible to subjectivity. Mouthfeel is an important parameter for the total perception of beer flavor. Three dimensions of mouthfeel are distinguished: contracting, coating, and drying. In this study 24 beer samples were evaluated chemically. The data were matched with sensorial data obtained from a trained panel. Different chemical analyses were performed; total acidity (TA), total flavonoids (TF), total polyphenols (TPC), total sugars (TS), color, pH, carbon dioxide content, ethanol, bitterness units (BU) and total iso-α-acids (TIA). The data were analyzed by performing several statistical techniques such as analysis of variance, principal component analysis, agglomerative hierarchical cluster analysis and multiple factor analysis. Sensory data obtained from trained panelists on the different mouthfeel attributes correlated with the data found instrumentally. The drying dimension could be expressed using the TPC, BU, TIA and pH. Contracting compounds correlated positively with TA and negatively with pH. As expected, ethanol was strongly associated with burning sensations and carbon dioxide with carbonation. The results of this experiment indicate that commercial beers can be classified into three mouthfeel attributes: drying, coating and contracting. The Principal Component Analysis (PCA) in this study confirmed the dimensions of the mouthfeel model and identified drying and coating as opposites, contracting forces interact on these dimensions. Moreover, these attributes were shown to be quantifiable by instrumental analysis which suggests that a data-driven approach based on mouthfeel could reduce subjectivity in the analysis of taste.

Introduction

Beer is a widely consumed alcoholic beverage. After water and tea, it is the third most popular drink globally (Piazzon et al., 2010). The flavor of the beer is a crucial parameter for consumer acceptance. There are many options in the brewing process which implies that for the consumer it is hard to get an impression of the flavor of an unfamiliar beer. Classification of beer types is used to give the consumer an impression of the taste that is to be expected. Classification is generally done by subjective methods that are likely to be expensive, time-consuming and less reliable than instrumental methods. A more objective, data-driven approach to assessing and classifying taste requires a good understanding of the origin and different characteristics of compounds that amount to taste (Stone et al., 2012). Furthermore, chemical and/or physical analytical protocols need to be established based on the related characteristics of oral sensations. Lastly, chemometric data need to be interpreted and correlated to taste perception by humans.

The term flavor is defined as the conjoint of odor, aroma, taste, and mouthfeel perceptions by the respiratory and alimentary tract of the food or beverage (Agorastos et al., 2020; Croissant et al., 2011). Volatile compounds in food and beverages stimulate the olfactory system (Croissant et al., 2011; Ryan et al., 2008). Taste perception is the result of non-volatile compounds, which activate receptors in the mouth (primarily in the tongue). Mouthfeel

perception is partly related to the tactile, irritant, and thermal sensations resulting from the activation of chemosensory and somatosensory receptors within the oral cavity by chemical stimuli (Baert et al., 2012; Gawel et al., 2018). Mouthfeel is not limited to the neural trigeminal system as gustatory tastants have been reported to affect mouthfeel as well by altering the rheological behavior of saliva (Liu et al., 2017). Consequently, mouthfeel sensations are not solely dependent on irritants and textural components, which calls for a broader definition.

Mouthfeel is a crucial aspect of the overall drinking experience of beer. According to previous studies, mouthfeel of beer encompasses various sensory attributes such as carbonation, astringency, smoothness, fullness and mouthcoating (Krebs et al., 2019, 2021; Langstaff et al., 1991a, 1991b). Understanding how compounds affect the mouthfeel of beer can help brewers create products that are more appealing to consumers. A valuable tool for comprehending mouthfeel sensations is the classification system proposed by Klosse (2004). A recent review on mouthfeel sensations summarized the benefits of using the mouthfeel model in taste classification (Agorastos et al., 2020). This model classifies mouthfeel sensations into three dimensions, drying, coating and contracting. Each of these dimensions has a scale of intensity, which results in a three-dimensional model. The tactile sensations are summarized by the drying and coating dimensions, while the irritant, thermal, chemesthetic and gustatory elements may affect contracting in mouthfeel (Klosse, 2004, 2013). The mouthfeel model can be used for the prediction of mouthfeel sensations based on the chemical composition and other characteristics of beers. It has been successfully applied in practice. This is the first time to apply the model instrumentally and investigate whether the model can be used for computational methods to predict taste qualities. Interestingly, despite the importance of mouthfeel, there is limited information on the chemical characteristics and responsible subclasses which influence mouthfeel perception of cereal-based beverages (Krebs et al., 2019, 2021).

Beer is a complex mixture of many different components. The general composition of beer consists of water, carbon dioxide (CO_2), alcohol, and extracts (low molar mass compounds and several polymers). Carbohydrates, ethanol, polyphenol, carbon dioxide and glycerol are present in larger quantities in beer. In total there are over 800 other organic components also present in low concentrations (Buiatti, 2008; Eßlinger, 2006). However, the contributions of different beer components to beer mouthfeel sensations are still largely unknown.

Beer contains a variety of polyphenols that are important for chemical stability. These include prenylated flavonoids, phenolic acids, simple phenols, flavanols, hydroxycoumarins flavones (Gerloff et al., 2010), proanthocyanidins, tannins, and amino phenolic compounds (Zhao et al., 2010). Apart from their antioxidant potential, these compounds play an important role in flavor (bitterness, astringency, harshness) and color (Gerloff et al., 2010). Most polyphenol compounds are derived from hops and malt (Montanari et al., 1999). The bitterness from hops is commonly known; the roasting of the malt, which influences the color of the beer is also an indicator of the amounts of polyphenols. The darker the color. the more polyphenols are present (Buiatti, 2008).

The major components that contribute to bitterness in beer are humulone, cohumulone and adhumulone. While the minor components that contribute to bitterness in beer are prehumulone and posthumulone. A-acids are isomerized during the boiling process of wort, forming respectively trans/cis-iso-α-acids (Anderson et al., 2019; Caballero et al., 2012; De Keukeleirc,

2000). The presence of iso-α-acids has been suggested to be the most dominant factor of bitterness in beer. Iso-α-acids are present in beer in the range of 15-100 mg/l (Anderson et al., 2019; Caballero et al., 2012; De Keukeleirc, 2000).

Carbohydrates contribute to mouthfeel, color, carbonization and foam production. Carbohydrates are estimated to range between 1 and 6% of beer. The final product contains mostly non-fermentable carbohydrates, such as dextrins, α -glucans, remnants of enzymatic starch hydrolysis and some polysaccharides components. However, fermentable carbohydrates can be added to the final product (Kunz et al., 2012). This is commonly done to further sweeten the beer (Buiatti, 2008; Floridi et al., 2001).

Finally, organic acids (such as acetic, lactic, malic acid etc.) are the "by-product" of yeast fermentation and are crucial for the taste of beer. Together with carbonic acid, they are responsible for the pH of beer and are present in the ranges of 200-500 mg/l. The pH of beer typically ranges from 4 to 5. Sour beers contain much higher concentrations of organic acids and are specially brewed for their acidic flavor.

The purpose of this study was to investigate the application of the mouthfeel model in beer classification. The relationship between compositional analysis and sensory data was also investigated in order to provide new insights into mouthfeel quantification. Trained panellists evaluated the sensorial properties of 24 beer samples. Nine different mouthfeel attributes, related to beer, were selected. The chemical and sensory data were further investigated using chemometric techniques. This study provides new insights into beer classification through the application of the mouthfeel model and the identification of relationships between sensory and chemical analysis. The outcome of this study can help the industry with a better understanding and predicting of the mouthfeel of beers and ultimately help consumers find the beer of their preference.

Materials and methods

Beer Samples

For the current study, 24 commercial beer samples were selected, based on the different beer styles provided by Scover (Scover, 2022). Scover is an online application which helps consumers with beer selection based on their preferences. To gain a better illustration of beer characteristics each beer style was represented by two different beer samples. The beer categories which are present in Scover and the current study are present in table 1.

Table 1. Beer samples and styles based on Scover.

Beer Sample	Beer Styles (in Dutch)	Beer Styles (in English)
PA	Strak en fruitig	contracting and fruity
KB	Strak en fruitig	
KV	Bitterzoet	bittersweet
GSE	Bitterzoet	
SD	Kruidig en fris	herbal and fresh
ML	Kruidig en fris	

PC	Allemansvriend	everyone's friend
LTB	Allemansvriend	
MM	Vol en fruitig	full and fruity
RL	Vol en fruitig	
CO	Rijk en verwarmend	rich and warming
LTT	Rijk en verwarmend	
HJ	Fris doordrinkbaar	refreshing and easily drinkable
PI	Fris doordrinkbaar	
WTD	Warm en vol	warm and full
LTQ	Warm en vol	
MN	Krachtig	powerful
BOP	Krachtig	
BCI	Toegankelijk karaktervol	accessible with a lot of character
CN	Toegankelijk karaktervol	
WH	Vrolijk en fris	happy and refreshing
DS	Vrolijk en fris	
LTD	Rijk en strak	rich and contracting
WD	Rijk en strak	

Chemical characterization

For each analytical method, except for the measurements of carbon dioxide concentration, the beer samples were degassed in an ultra-sonification bath for 20 minutes. The sonification bath contained ice to avoid high temperatures due to sonification. The chemical analysis for every method was performed in duplicate except for the BU and carbon dioxide measurements.

The pH was determined by a pH meter (FiveEasy^{TM}, FE20, Switzerland). The pH meter was calibrated at pH 4.0 and 7.0 before all measurements. The total (titratable) acidity of the beer samples was determined by potentiometric titration. 10 ml of beer samples was added to 50 ml of distilled water and the sample was titrated against 0.005 M sodium hydroxide. The volume of NaOH required to attain a pH of 8.2 was determined using a pH meter. Total acid was expressed in terms of tartaric acid equivalents (AOAC Method 2000).

The ethanol content was determined by gas chromatography headspace analysis (GC-HS) with a flame ionization detector (FID) (6890, Agilent Technologies, Germany) at 300 ºC. The injection volume of the samples was 0.2 µl and the injection speed was 1.0 µl/sec. The used column was a HP-5MS-UI (30m, 0.25mm, 0.25µm, Agilent Technologies, Germany) at column temperature of 50 ºC.

Measurement of color, total polyphenol and total flavonoids, were performed based on European Brewing Congress (EBC) and American Society of Brewing Chemists (ASBC) methods (Spectroquant® Prove, 2021). The measurements were performed on a Jasco V-650 Spectrophotometer (Thermo Fisher, Germany). For the color measurement samples were centrifuged at 4000 rpm (3095rcf) to reduce sample turbidity. 1 ml supernatant was further filtered over a 0.22 µm PTFE filter and measured at 430 and 700 nm. SRM values were calculated by multiplying 12.7 times the dilution factor and the absorbance at 430 nm. Total polyphenols values were expressed in gallic acid equivalent. 20 µl of degassed beer sample diluted in 150 µl of Milli-Q water. 200 µl of Colin-Ciocalteu reagent (Sigma-Aldrich, Germany), 3 ml of Milli-Q water and 600 µl 20% w/v Na_2CO_3 were added, and the solution was placed in a dark room for an hour. The samples were measured at 760 nm. Total flavonoid measurements were expressed in catechin equivalent. Beer samples were diluted ten times and mixed with a color reagent in a 1:5 v/v ratio. The color reagent was prepared by dissolving 50 mg 4-dimethylaminocinnamaldehyde (Sigma-Aldrich, Germany) in a mix of 12.5 ml HCl (37 %) and 37.5 ml methanol. The samples were measured at 640 nm.

To determine the carbon dioxide content (CO_2) samples were measured at 25 ^{0}C with a Haffmans Impack 2000 CO_2 (Pentair Haffmans, Switzerland). The BU analysis was performed by the Beermaster (Gallery™ Plus Beermaster Discrete Analyzer, Thermo Fisher, Germany).

HPLC (Shimadzu Nexara-I LC-2040C 3D, UK) was used for the quantification of iso-α and α acids. This method has been modified from the original method proposed by Heyerick (2003). The detector was a Photodiode-Array Detector (PDA) at 270 nm for iso-α acids, ρ-iso-α-acids and 314 nm for α acids. A pre-column shim-pack GIST(G) C18 (4.0 x 10 mm, 3.0 µm) (Shimadzu, UK) and a column XSelect HSS T3 (100 x 3.0 mm, 2.5 µm) (WATERS, USA) was used. The eluents were demi water of pH 2.10 and acetonitrile HPLC gradient grade (Sigma-Aldrich, Germany). A flow of 0.8 ml per minute and an injection volume of 10 µl was applied. DCHA ICS-R3, DCHA ICS-I4 and DCHA ICS-A1 standards (Lavor Veritas, Switzerland) were used for the calibration curve. From these standards, a combined stock solution of 200 mg/L was made in 55 % water adjusted to pH 2.10 with orthophosphoric acid (85 %):45 % acetonitrile HPLC grade to match the starting eluent and sample matrix. To remove matrix interference, a liquid-liquid (LLE) extraction was performed. 15 ml glass tube was filled with 6 ml n-heptane, 1 ml methanol, 0,25 ml of orthophosphoric acid (85 %) (Sigma-Aldrich, Germany) and 6 ml of beer sample. The tubes were mixed on a turntable for thirty minutes and thereafter centrifuged at 3000 rpm (1741 rcf) for five minutes. 5 ml of the organic layer was transferred to a new glass tube of 15 ml and evaporated under N_2 flow. The completely evaporated samples were reconstituted with 1 ml of 55 % (v/v) water adjusted to pH 2.10 with orthophosphoric acid (85 %): 45 % (v/v) acetonitrile HPLC grade to match the starting eluent. Samples were filtered over a 0.22 µm PTFE filter and stored in HPLC vials before injection. Samples were prepared in duplicate. The calibration series was made with the same liquid-liquid extraction method from the sample preparation.

The concentration of total fermented sugars (TS) was determined using an HPLC (Shimadzu Nexara-I LC-2040C 3D, UK) method with a RID detector (Shimadzu RID-10A, UK) set to 40 degrees Celsius. A pre-column filter 2 µm SS Frit A-101 (GL Sciences, Germany) and column InertSustain NH_2 (300 mm x 4.6 mm, 5 µm) (GL Sciences, Germany) were used. The separation was done under isocratic elution of 85 % acetonitrile HPLC grade and 15 % (v/v)

MilliQ water. The flow was set to 1.0 ml per minute and the injection volume was 10 µl. Samples were filtered over a 0.22 µm PTFE filter to remove solids and stored in HPLC vials before injection. This method has been modified from the original (Moussa et., 2012).

Sensory analysis

A trained panel assessed the beer's sensory properties. Nineteen experienced beer tasters (beer sommeliers) were selected for the sensory evaluation. Nineteen experienced panelists were selected by using an online screening questionnaire. The panelists were screened based on age, gender, non-smoking and absence of oral disorders.

The panelists were trained on beer-related sensory attributes based on previous studies (Langstaff et al., 1991a, 1991b; Schmelzle, 2009). Three sessions of three hours each were taken for this training. Those sessions consisted of the explanation of the mouthfeel sensations/attributes and the mouthfeel intensities that occur in beer samples.

The beers were classified based on the sensory attributes such as drying, coating, contracting, sweetness, acidity, bitterness, carbonation, burning and taste intensity (Langstaff et al., 1991a, 1991b; Schmelzle, 2009). The intensity of each attribute was rated on a 10 cm unstructured line scale which was displayed on a screen, where low and high intensity was provided in the line scale.

Panelists were given a sample of 50 ml of freshly opened beer bottles or cans. The samples were stored at around 4 ^{o}C prior to the sessions. Samples were presented in amber vials labelled with random three-digit codes throughout the study to minimize expectation error and to reduce bias from appearance. Panelists were given three-minute breaks between each sample to avoid palate fatigue with a 30-minute comfort break at the midpoint of evaluation (after 8 samples). During the break, panelists used water to cleanse the palate and minimize sample carry-over. Data was collected using Qualtrics^{XM} software (Qualtrics International).

Statistical analysis

To analyze the results obtained by the sensory and chemical analysis, several chemometric tools were addressed. Describing complex systems often requires multiple parameters, which implies that a multivariate data analysis needs to be used. All statistical analyses were performed by the software R (R core team and foundation for statistical computing), R-studio version 4.0.3. Intensity scores of the nineteen panelists for each sensory attribute were calculated by applying analysis of variance (ANOVA). The F test was used to investigate the statistical significance of beer samples and assessors and Tukey's test was used to further determine the statistical difference among the beer samples. Statistically significant attributes (p-value < 0.05) were selected for agglomerative hierarchical cluster analysis (AHC) of all samples. Principal component analysis (PCA) was used as a dimension reduction methodology. The object of PCA is to reduce the dimensionality of the dataset while retaining most of the original variability in the data. PCA was performed to identify sensory profiles from different clusters based on the AHC analysis. Finally, loadings for the chemical analysis were analyzed in conjunction with the sensory data through multifactor analysis (MFA).

Results

Sensory analysis and classification.

Results from the ANOVA showed significant differences ($p < 0.05$) among the 24 beer samples and all the sensory attributes. Therefore, a Tukey multiple comparison test was performed for the comparison of the means among the sensory attributes. It is clear from table 2 that the samples with similar letters are statistically not significantly different for a specific sensory attribute.

Panelists were able to discriminate the 24 beer samples among the sensory attributes of coating, drying, sweetness, bitterness, and taste intensity. Table 2 shows the large variation and statistical differences that were observed among those sensations. Contrary, panelists clearly distinguished less efficient beer samples for the sensory attributes of contracting, acidity, carbonation and burning. The variation between the sensory attributes scores contracting, acidity, carbonation, acidity and burning was lower compared to the other sensory attributes.

The beer samples KB, PA and RL scored average intensities above 4 for acidity and contracting sensation. KB, PA and RL presented 5.16, 4.52 and 4.24 for acidity and 5.08, 4.92 and 4.50 for contracting respectively. Contrarily, beer samples KV, PC, PI, BCI and LTT scored lower in the intensity of acidity and contracting attributes.

The highest scores in bitterness were found in beer samples KV, BOP, GSE and PI. The lower bitterness intensities were observed in samples RL, KB and MM samples. Similarly, the drying intensity values, KV, MN and BOP samples were perceived as the most drying, while RL had the lowest intensity mean.

Samples LTD, MM, RL, and KB scored as the sweetest and most coating with values 5.84, 7.05, 6.92 and 6.57 but also 5.84, 5.67, 5.55 and 4.94 respectively. Beers BOP and ML scored the lowest intensities on those sensations.

Regarding the burning and carbonation attributes no large differences were observed. The MM sample showed the lowest score regarding the burning sensation, while WTD had the highest mean value. Furthermore, DS ranked as the beer with the highest carbonation sensation with a value of 6.53, while KV gave the lowest value. Finally, the panelists scored the beer samples based on the total taste intensity. KB, KV and GSE samples were perceived as the most intense with scores of 7.34, 7.29 and 7.08 respectively. With a score of 3.65 BCI ranked lowest in intensity.

Table 2. Sensory characterization of 24 beer samples

Code	Acidity	Bitterness	Burning	Carbonation	Coating	Contracting	Drying	Sweetness	Taste intensity
PA	4.52 ± 0.44^{bc}	3.69 ± 0.50^{bcdfi}	2.00 ± 0.51^{a}	5.47 ± 0.47^{ab}	3.30 ± 0.60^{abd}	4.92 ± 0.34^{ab}	4.13 ± 0.57^{abcde}	4.12 ± 0.65^{abcdf}	5.27 ± 0.37^{abcde}
KB	5.16 ± 0.48^{c}	1.10 ± 0.32^{h}	1.29 ± 0.38^{a}	5.50 ± 0.36^{ab}	4.94 ± 0.67^{abcd}	5.08 ± 0.60^{a}	2.50 ± 0.50^{bef}	6.57 ± 0.53^{ghi}	7.34 ± 0.32^{d}
KV	1.90 ± 0.25^{a}	6.45 ± 0.45^{ae}	2.40 ± 0.64^{a}	4.33 ± 0.32^{b}	3.64 ± 0.73^{abcd}	3.00 ± 0.45^{ab}	6.05 ± 0.25^{a}	3.07 ± 0.38^{abc}	7.29 ± 0.25^{d}
GSE	2.13 ± 0.3^{a}	5.80 ± 0.52^{aefg}	2.70 ± 0.51^{a}	4.97 ± 0.42^{ab}	4.63 ± 0.75^{abcd}	3.52 ± 0.45^{ab}	4.77 ± 0.39^{abcd}	4.01 ± 0.51^{abcdef}	7.08 ± 0.40^{bd}
SD	3.25 ± 0.37^{abc}	4.50 ± 0.50^{cdefg}	2.22 ± 0.56^{a}	6.10 ± 0.47^{ab}	3.19 ± 0.42^{abcd}	3.85 ± 0.50^{ab}	5.29 ± 0.51^{ad}	3.29 ± 0.52^{abcf}	5.56 ± 0.44^{abcde}
ML	3.06 ± 0.60^{abc}	4.82 ± 0.33^{adefg}	2.21 ± 0.59^{a}	5.42 ± 0.41^{ab}	2.52 ± 0.49^{a}	3.64 ± 0.55^{ab}	5.10 ± 0.38^{acd}	2.57 ± 0.27^{b}	4.93 ± 0.43^{ace}
PC	1.92 ± 0.34^{a}	3.50 ± 0.40^{bcdi}	1.98 ± 0.53^{a}	4.86 ± 0.32^{ab}	3.62 ± 0.50^{abcd}	3.06 ± 0.47^{ab}	3.77 ± 0.30^{abcdef}	3.82 ± 0.48^{abcdef}	4.22 ± 0.46^{ae}
LTB	2.59 ± 0.46^{ab}	2.91 ± 0.42^{bcdhi}	2.95 ± 0.59^{a}	5.87 ± 0.34^{ab}	4.75 ± 0.41^{abcd}	3.15 ± 0.42^{ab}	2.48 ± 0.40^{bef}	$4.90 \pm 0.32^{\ cdefgh}$	4.82 ± 0.45^{ace}
MM	3.72 ± 0.49^{abc}	1.94 ± 0.39^{hi}	0.90 ± 0.22^{b}	5.73 ± 0.33^{ab}	5.67 ± 0.54^{cd}	3.92 ± 0.48^{ab}	2.23 ± 0.43^{ef}	7.05 ± 0.38^{i}	5.82 ± 0.40^{abcd}
RL	4.24 ± 0.58^{abc}	1.05 ± 0.21^{h}	1.10 ± 0.44^{ab}	5.43 ± 0.29^{ab}	5.55 ± 0.58^{bcd}	4.50 ± 0.52^{ab}	1.62 ± 0.43^{f}	6.92 ± 0.68^{hi}	4.86 ± 0.49^{ace}
CO	2.87 ± 0.31^{ab}	3.07 ± 0.41^{bcdhi}	2.85 ± 0.72^{a}	5.72 ± 0.48^{ab}	4.44 ± 0.74^{abcd}	3.72 ± 0.49^{ab}	3.66 ± 0.53^{abcdef}	$4.42 \pm 0.59a^{bcdefg}$	5.02 ± 0.43^{abce}
LTT	2.60 ± 0.50^{ab}	3.60 ± 0.53^{bcdi}	2.82 ± 0.68^{a}	6.37 ± 0.41^{ab}	5.17 ± 0.62^{abcd}	2.85 ± 0.28^{ab}	3.35 ± 0.60^{bcdef}	5.66 ± 0.35^{defghi}	5.15 ± 0.46^{abcd}
HJ	2.99 ± 0.46^{abc}	4.34 ± 0.52^{acdefg}	1.83 ± 0.37^{a}	4.75 ± 0.37^{ab}	3.77 ± 0.51^{abcd}	3.18 ± 0.50^{ab}	$4.34 \ \pm 0.55^{abcde}$	3.78 ± 0.46^{abcdf}	4.56 ± 0.45^{ace}
PI	1.81 ± 0.32^{a}	5.57 ± 0.48^{aefg}	1.90 ± 0.48^{a}	4.71 ± 0.41^{ab}	3.24 ± 0.40^{abd}	3.25 ± 0.41^{ab}	3.86 ± 0.59^{abcde}	3.01 ± 0.46^{abc}	4.98 ± 0.42^{abce}
WTD	2.44 ± 0.46^{a}	4.30 ± 0.50^{bcdfg}	3.22 ± 0.66^{a}	5.10 ± 0.41^{ab}	4.30 ± 0.50^{abcd}	3.15 ± 0.45^{ab}	4.29 ± 0.50^{abcde}	$4.32 \pm 0.38^{\ abcdefg}$	5.97 ± 0.43^{abcd}
LTQ	2.21 ± 0.40^{a}	3.72 ± 0.48^{bcdi}	2.89 ± 0.57^{a}	5.62 ± 0.35^{ab}	6.00 ± 0.51^{c}	2.97 ± 0.31^{ab}	3.85 ± 0.41^{abcdef}	6.37 ± 0.28^{eghi}	6.51 ± 0.40^{bcd}
MN	2.41 ± 0.53^{a}	5.73 ± 0.54^{aeg}	2.33 ± 0.34^{a}	5.07 ± 0.44^{ab}	3.18 ± 0.41^{abd}	3.25 ± 0.56^{ab}	5.92 ± 0.68^{a}	3.52 ± 0.30^{abcdf}	6.13 ± 0.42^{abcd}
BOP	2.47 ± 0.43^{ab}	6.52 ± 0.45^{a}	2.40 ± 0.46^{a}	4.49 ± 0.48^{ab}	2.97 ± 0.55^{a}	3.54 ± 0.51^{ab}	5.87 ± 0.38^{a}	2.77 ± 0.50^{ab}	5.93 ± 0.37^{abcd}
BCI	1.90 ± 0.31^{a}	3.37 ± 0.32^{bcdi}	1.49 ± 0.50^{a}	4.94 ± 0.43^{ab}	3.29 ± 0.49^{abcd}	2.71 ± 0.30^{b}	3.02 ± 0.45^{bcdef}	3.37 ± 0.34^{abcf}	3.65 ± 0.25^{e}
CN	2.03 ± 0.26^{a}	4.29 ± 0.48^{bcdefg}	2.10 ± 0.53^{a}	5.33 ± 0.58^{ab}	2.82 ± 0.42^{ab}	3.55 ± 0.45^{ab}	4.50 ± 0.48^{abcde}	3.10 ± 0.44^{abc}	4.73 ± 0.54^{ace}
WH	2.54 ± 0.41^{ab}	2.05 ± 0.48^{bhi}	1.04 ± 0.25^{ab}	5.03 ± 0.47^{ab}	5.45 ± 0.62^{abcd}	3.02 ± 0.39^{ab}	2.27 ± 0.37^{bef}	4.69 ± 0.35^{abcdefg}	5.13 ± 0.40^{ace}
DS	3.45 ± 0.54^{abc}	2.41 ± 0.52^{bchi}	3.34 ± 0.69^{a}	6.53 ± 0.52^{a}	4.32 ± 0.46^{abcd}	4.31 ± 0.67^{ab}	2.82 ± 0.53^{bcdef}	5.33 ± 0.52^{cdefghi}	5.67 ± 0.42^{abcd}
LTD	2.04 ± 0.35^{a}	3.85 ± 0.42^{bcdhi}	2.42 ± 0.45^{a}	5.17 ± 0.47^{ab}	5.68 ± 0.58^{cd}	2.57 ± 0.39^{b}	3.75 ± 0.53^{abcdef}	5.84 ± 0.35^{deghi}	6.10 ± 0.39^{abcd}
WD	2.64 ± 0.36^{ab}	2.74 ± 0.43^{bcdhi}	2.17 ± 0.44^{a}	5.18 ± 0.51^{ab}	4.90 ± 0.57^{abcd}	2.94 ± 0.38^{ab}	2.93 ± 0.40^{bcef}	4.46 ± 0.51^{abcdefg}	5.10 ± 0.53^{ace}

Remark: The values are presented in mean value ±standard error. The letters in each bar identify that the samples are significantly different according to Tukey's HSD test: $p < 0.05$

To visualize the data structure by reducing the data dimensionality while retaining as much as possible the information PCA was used (Berrueta et al., 2007). The PCA plot of the panelist's scores for the 24 beers among nine sensory attributes is shown in figure 1a. The first two principal components using all the variables accounted for 65.2 % of the total variance (Dim 1, 46.8 %; Dim 2, 18.4 %). This indicates that the visualization of the correlation plot accounts for most of the variance within the sensory scores. Overall, the Dim-1 differentiated the beer samples successfully based on the bitterness-drying and sweetness-coating, whereas the dim-2 differentiated effectively the beers based on the contracting and acidity variables.

From the biplot (figure 1a), it can be observed that three different groups among the sensory parameters have been formed that are correlated with each other. First, bitterness, drying and burning attributes had the highest correlation. A similar pattern between bitterness and dry sensations has been noticed by Dietz et al. (2021). Additionally, contracting and acidity were the second-highest correlated group, while sweetness and coating formed the last. The drying/bitterness/burning group was negatively correlated with the sweetness/coating group. The dimensions of the mouthfeel model were clearly observed. Dietz et al. (2021) also observed a similar relationship between drying compounds and sweetness. This finding was not highlighted, however, since the aim of the study was different. In conclusion, there is a clear pattern for the sensory attributes and their correlation. This important finding is crucial for the application of beer classification.

Additionally, a hierarchical clustering analysis (HCA) was performed on the PCA (figure 1b). Cluster 1 (blue) appeared on the left side of the dim-1 and spread across both segments of dim-2 with eight samples in the upper half and four in the lower half. Cluster 2 (yellow) is presented on the right side of dim-1 and the lower half of dim-2. Last, cluster 3 was found on the right side of dim-1 and mostly on the upper half of dim-2, only sample MM was lower.

The clusters defined by HCA analysis could be explained through different sensory profiles (figure1a and b). Cluster 1 was characterized by drying, bitterness and burning sensations. Cluster 2 on the other hand, was generally characterized by sweetness and coating. Finally, cluster 3 was associated with higher acidity and contracting, whereas drying, bitterness, sweetness and coating were not associated with cluster 3. In a previous consumer study, Ramsey et al. (2018) showed as well that three main clusters were able to capture the liking characteristics of beer samples. Their three clusters were characterized by C1: tingly and fullness/body sensations, C2: malty and sweet sensations and C3 sour, bitter and tingly sensations.

These outcomes suggest that the beer samples could potentially be classified based only on the three different dimensions of the mouthfeel model. Furthermore, for coating and drying dimensions, it can be suggested that these are the opposite sides of the tactile dimension while contracting can be categorized as a separate dimension based on irritant sensations.

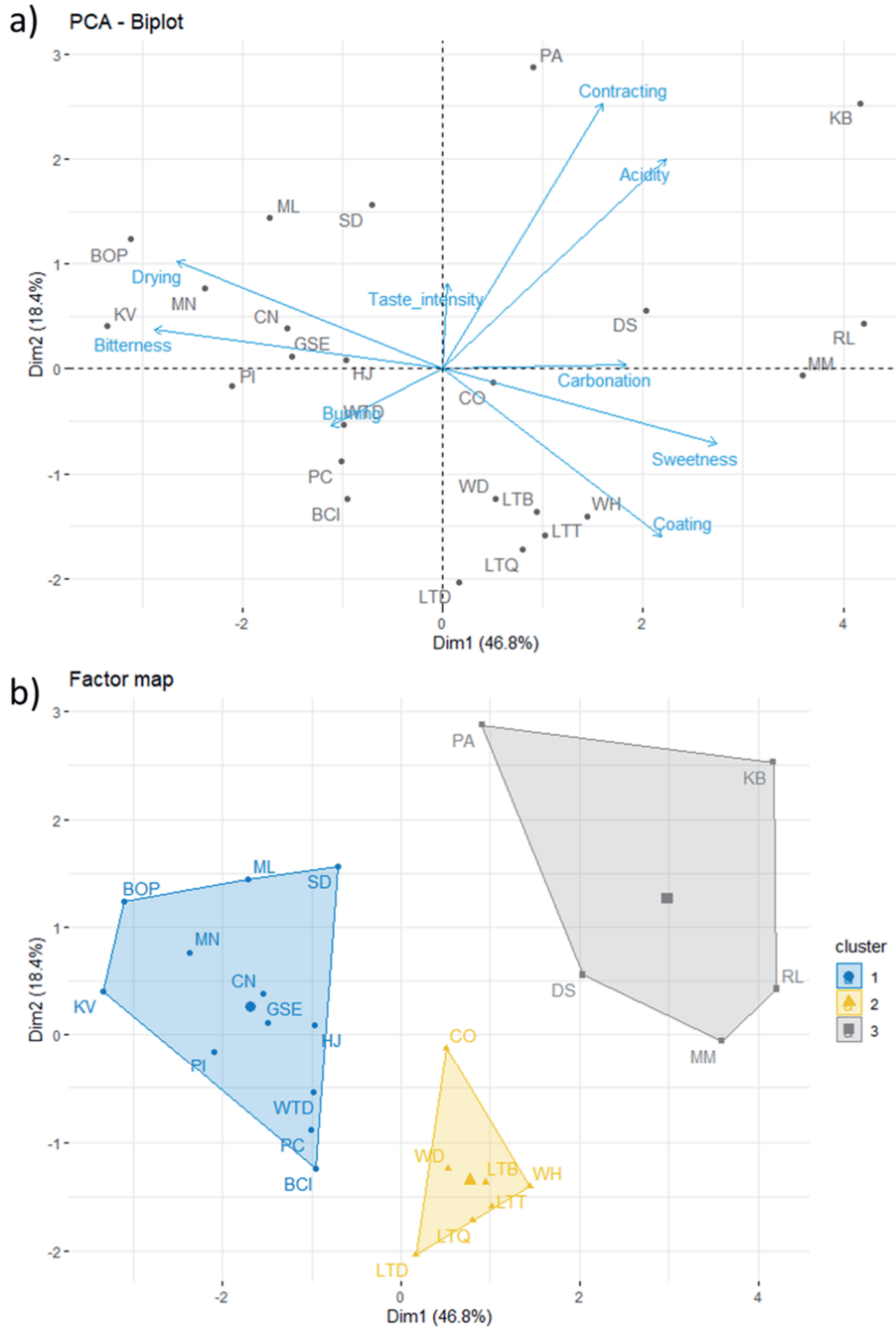

Figure 1. a) Principal Component Analysis biplot of among the sensory attributes after evaluation of 24 beer samples. b) Visualization of samples on the principal component map and colored according to different clusters.

Chemical and sensory relation of beer data

The beer samples were chemically analyzed. The chemical profiles of the non-volatile compounds observed in the beer samples are presented in table 3. Large differences between the chemical characteristics of the 24 beer samples were observed.

The pH values varied between 3.26 and 4.66, while RL had the lowest pH values and BCI the highest. Beers with pH values lower than 4 were categorized as 'high' (acidic), whereas beers with a pH higher than 4.5 were classified as the lower acidic beers. Titratable acidity showed the same trend among the beers. The beers with pH lower than 4 presented titratable acidity with values above 0.05 mol/L. An important notice is that RL had the lowest pH of 3.26, but presented a titratable acidity value of 0.050 mol/L. However, KB with a pH of 3.51 had a titratable acidity value of 0.080 mol/L. The lower values of titratable acidity were observed in the samples HJ and PI with values of 0.028 and 0.032 mol/L respectively. The rest of the samples fluctuated around values of 0.04 mol/L.

The carbon dioxide concentration showed a smaller fluctuation among the beer samples. CO had the highest CO_2 concentrations with a value of 7.52 mg/L. While HJ had the smallest concentration of 4.48 mg/L.

Beer samples KV, LTD and GSE scored high in polyphenol content, with concentrations of 543, 525 and 418 mg/L respectively. Samples RL, MM, CO and LTT had the lowest concentrations of polyphenols, with values of 25, 125, 156 and 173 mg/L respectively. Most of the beer samples fluctuated in values around 260 mg/L. Color was found to be related to the total polyphenol content. Beers with the darkest color had the highest concentrations of polyphenols, whereas the beers with lighter colors are the ones with lower TPC concentrations (Buiatti, 2008). KB had the highest total flavonoid concentration of 150 mg/L, with KV and WTD followed by 105 and 75 mg/L. Additionally, the RL sample had the lowest concentration of TF with values of 18.1 mg/L.

Both bitterness units (BU) and total iso-α-acids (TIA) presented a large variation among the different beer samples. The samples showed a range from 4.2 to 39.8 BU. GSE had the higher BU value of 39.8, while CN, SD, BOP and MN followed with values of 37.4, 33.6, 32.9 and 32.6 BU respectively. However, the highest TIA values were observed in MN and BOP samples with concentrations of 44.63 and 40.21 mg/L respectively. GSE and CN samples were followed with 38.67 and 33.19 mg/L, while RL and DS showed the lowest TIA concentrations of 2.90 and 1.63 mg/L respectively. Popular commercial beers have a concertation of TIA at 20-40 mg/L, while bitter beers such as India Pale Ale contain 50-80 mg/L (Ayabe et al., 2018). Finally, the ethanol concentration varied between 1.93 and 11.71 % v/v. DS and LTQ were the only samples that contained ethanol percentage above 10 %. Most of the beer samples varied between 4.4 and 7.0 %, while RL had the lowest ethanol percentage of 1.93 %.

Table 3. Chemical composition and characteristics obtained in 24 different beer samples.

Code	Total acidity (mol/L)	Total Flavonoids (mg/L)	Total polyphenols (mg/L)	Total sugars (mg/L)	Color (SRM)	pH	CO_2 (mg/L)	Ethanol (%)	Total Iso-α-acids (mg/L)	BU
PA	0.042 ± 0.002^{cd}	31.5 ± 0.5^{cd}	148.6 ± 0.9^{b}	603.8 ± 69.3^{a}	2.68 ± 0.01^{a}	3.95	5.19	4.75 ± 0.03^{c}	11.2 ± 0.1^{e}	13.7
KB	0.080 ± 0.001^{g}	150.2 ± 3.8^{m}	268.2 ± 3.4^{ef}	468.0 ± 72.4^{a}	16.73 ± 0.01^{q}	3.51	5.92	3.81 ± 0.12^{b}	>1.0	5.9
KV	0.042 ± 0.001^{cd}	105.0 ± 0.2^{l}	543.5 ± 15.2^{j}	452.8 ± 10.1^{a}	93.77 ± 0.01^{v}	4.33	4.57	6.01 ± 0.04^{efg}	27.9 ± 0.2^{m}	28.5
GSE	0.040 ± 0.001^{bcd}	51.2 ± 1.5ghi	418.2 ± 5.0^{i}	3275.2 ± 62.4^{a}	55.59 ± 0.01^{u}	4.14	5.88	8.39 ± 0.14^{jk}	38.6 ± 0.3^{p}	39.8
SD	0.038 ± 0.001^{bc}	51.9 ± 1.2hi	230.6 ± 10.7^{e}	178.5 ± 14.7^{a}	5.31 ± 0.01^{f}	4.36	6.95	6.79 ± 0.09^{hi}	29.8 ± 0.1^{n}	33.6
ML	0.041 ± 0.001^{cd}	20.2 ± 0.1b	236.0 ± 11.8^{e}	486.1 ± 52.5^{a}	5.14 ± 0.01^{e}	4.59	5.81	5.73 ± 0.17^{def}	22.8 ± 0.1^{k}	23.9
PC	0.032 ± 0.007^{a}	43.5 ± 0.9efg	233.1 ± 20.0^{e}	232.4 ± 27.0^{a}	12.69 ± 0.01^{o}	4.14	5.47	5.51 ± 0.02^{de}	17.9 ± 0.1^{h}	23.9
LTB	0.038 ± 0.001^{bc}	38.4 ± 0.6^{de}	252.8 ± 7.7^{e}	77.1 ± 25.0^{a}	5.91 ± 0.01^{i}	4.48	6.71	6.34 ± 0.05^{fgh}	13.4 ± 0.1^{f}	17.7
MM	0.054 ± 0.001^{f}	23.5 ± 0.2^{bc}	125.8 ± 6.5^{b}	25413.8 ± 669.0^{c}	2.94 ± 0.01^{b}	3.72	5.12	3.74 ± 0.11^{b}	2.3 ± 0.1^{bc}	5.7
RL	0.050 ± 0.001^{ef}	1.8 ± 0.4^{a}	25.1 ± 0.6^{a}	46698.8 ± 3301.1^{d}	3.48 ± 0.01^{d}	3.26	5.24	1.93 ± 0.01^{a}	2.9 ± 0.1^{c}	4.2
CO	0.039 ± 0.001^{bcd}	43.2 ± 1.4^{efg}	156.4 ± 14.9^{b}	615.3 ± 0.07^{a}	3.47 ± 0.01^{d}	4.04	7.52	8.77 ± 0.04^{k}	19.3 ± 0.1^{i}	19.2
LTT	0.031 ± 0.001^{a}	54.8 ± 3.4^{hij}	173.3 ± 12.6^{bc}	28.6 ± 21.4^{a}	5.74 ± 0.01^{h}	4.28	6.68	7.96 ± 0.11^{j}	17.6 ± 0.1^{h}	20.6
HJ	0.028 ± 0.001^{a}	57.3 ± 2.0^{ij}	233.7 ± 5.0^{e}	3459.7 ± 64.3^{a}	6.75 ± 0.01^{m}	4.4	4.48	5.23 ± 0.07^{cd}	13.5 ± 0.1^{f}	16.2
PI	0.032 ± 0.001^{a}	40.5 ± 0.9^{ef}	233.7 ± 9.5^{e}	2774.8 ± 17.74^{a}	5.93 ± 0.01^{ij}	4.47	4.67	5.68 ± 0.07^{def}	28.1 ± 0.1^{m}	27.4
WTD	0.037 ± 0.001^{b}	75.3 ± 1.6^{k}	343.7 ± 1.3^{gh}	378.3 ± 14.0^{a}	45.18 ± 0.01^{t}	4.29	6.8	6.94 ± 0.01^{hi}	25.6 ± 0.1^{l}	23.9
LTQ	0.039 ± 0.001^{bcd}	62.5 ± 1.0^{j}	333.7 ± 9.5^{gh}	52.3 ± 8.3^{a}	18.67 ± 0.01^{r}	4.42	6.61	10.33 ± 0.12^{l}	15.6 ± 0.1^{g}	18.4
MN	0.040 ± 0.001^{bcd}	48.5 ± 0.9^{fgh}	380.0 ± 8.6^{hi}	793.3 ± 60.2^{a}	11.48 ± 0^{n}	4.58	5.19	7.13 ± 0.24^{i}	44.6 ± 0.1^{r}	32.6
BOP	0.037 ± 0.001^{b}	48.9 ± 0.3^{fghi}	306.0 ± 7.2^{fg}	1102.1 ± 8.3^{a}	5.56 ± 0.01^{g}	4.57	4.53	6.64 ± 0.11^{ghi}	40.2 ± 0.1^{q}	32.9
BCI	0.038 ± 0.001^{bc}	22.7 ± 0.9^{b}	224.1 ± 1.7^{cde}	105.2 ± 5.2^{a}	5.94 ± 0^{j}	4.66	4.98	5.28 ± 0.11^{cd}	20.4 ± 0.1^{j}	20.6
CN	0.039 ± 0.002^{bcd}	35.6 ± 0.9^{de}	269.1 ± 3.1^{ef}	56.9 ± 2.0^{a}	6.50 ± 0.01^{l}	4.41	4.72	5.41 ± 0.23^{cde}	33.1 ± 0.1^{o}	37.4
WH	0.041 ± 0.001^{cd}	22.1 ± 1.2^{b}	228.7 ± 4.5^{de}	376.1 ± 33.7^{a}	6.31 ± 0.01^{k}	4.48	5.47	6.07 ± 0.01^{efg}	8.5 ± 0.2^{d}	11.4
DS	0.048 ± 0.001^{e}	41.7 ± 0.1^{ef}	176.9 ± 1.7^{bcd}	16636.2 ± 916.4^{b}	3.14 ± 0.01^{c}	3.93	5.39	11.71 ± 0.30^{m}	1.6 ± 0.1^{b}	5.7
LTD	0.043 ± 0.001^{d}	53.6 ± 1.1^{hi}	525.5 ± 12.2^{j}	118.8 ± 46.2^{a}	42.37 ± 0.1^{s}	4.47	6.67	6.89 ± 0.05^{hi}	10.4 ± 0.1^{e}	15.8
WD	0.042 ± 0.001^{cd}	24.6 ± 0.9^{bc}	267.4 ± 5.9^{ef}	78.3 ± 33.5^{a}	14.51 ± 0.01^{p}	4.44	5.86	5.44 ± 0.12^{cde}	8.6 ± 0.4^{d}	12.5

Remark: The values are presented in mean value ±standard error. The letters in each bar identify that the samples are significantly different according to Tukey's HSD test: $p < 0.05$

To investigate the relationships between sensory and chemical data, multiple factor analysis was applied. Means of the sensory descriptions and means of the chemical class loadings from 24 beer samples were used for the analysis.

The MFA plot explained 62.03% of the variance in the data with 45.25 % represented by dim-1 and 16.75 % by dim-2 (figure 2). The sensory attributes sweetness, coating and carbonation were plotted on the left side of the dim-1 and the upper portion of dim-2, while the acidity and contracting dimensions were placed on the lower portion of dim-2. On the opposite side, the burning sensation was plotted on the right side of dim-1 and the upper part of dim-2. Lastly, drying and bitterness were found on the right part of dim-1 and the lower side of dim-2.

The total acidity was associated with contracting attribute, while pH showed a negative correlation. The chemical analyses of total iso-α acid, pH and total polyphenol content highly correlated with drying and bitterness sensations. As expected, burning sensations correlated with the ethanol content and carbonation with the carbon dioxide concentration. Taste intensity correlated highly with total flavonoid content. Multiple factor analysis correlation circle for both sensory scores (green) and chemical analyses (red).

Similar trends have been observed by Rettberg et al. (2022). However, in this study, we did not find a correlation between sweetness and total sugar. Additionally, this research provided more information new into mouthfeel sensations since more chemical measurements and sensory attributes were tested.

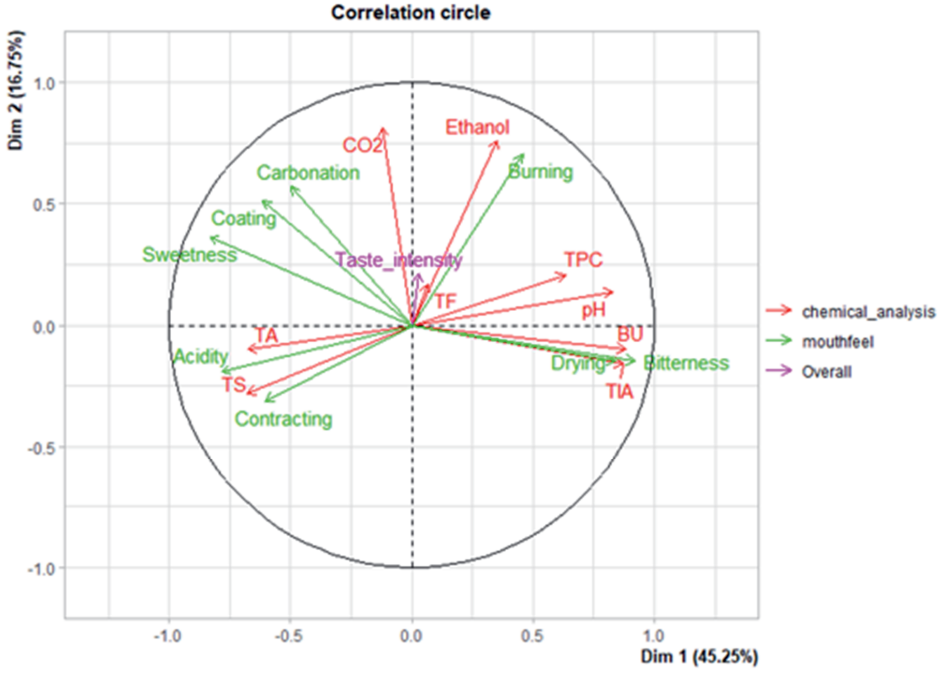

Figure 2. Multiple factor analysis biplot of the chemical analysis and sensory characteristics of 24 beer samples.

The outcome shows the ability to quantify mouthfeel sensations based on chemical analyses. Almost all sensory attributes correlated highly with chemical analyses. Only sweetness and coating were found not to correlate strongly with any chemical analysis. A likely explanation is that besides sweetness, other coating elements were perceived.

Discussion

Sensory characterization

The results obtained in this study suggested that three main categories can be identified to classify beers (figure 1). A total variance of 65.2 % was expressed by PCA, where three attribute groups of drying/bitterness, coating/sweetness and acidity/contracting were identified. The trained panelists were not able to distinguish the sensory attributes of drying and bitterness, which gave a coefficient correlation of 0.92 ($p < 0.05$). The coefficient correlation of 0.92 ($p < 0.05$) between drying and bitterness indicates a strong correlation between drying and bitterness sensations. Similar to the previous observation by Dietz et al. (2020), panelists were not able to differentiate these sensations even though they have a different origins. Bitterness is considered to belong to the basic tastes, recognized by the receptors located on the surface of the tongue for the reception of taste (Meilgaard et al., 1999; Saison et al., 2009; Schmelzle, 2009). While a drying (or often called astringent) sensation is associated with puckering or rough sensation across the oral cavity and often is explained by the loss of salivary lubrication (Bajec & Pickering, 2008). Despite the different origins, both sensations are related to the polyphenol content. The main difference that characterizes the two sensations is the degree of isomerization of polyphenols (Lesschaeve & Noble, 2005; McLaughlin et al., 2008a; Peleg et al., 1999). Polyphenols with larger molecular weight contribute to sensations of astringency, while lower molecular weight polyphenols contribute to the sensation of bitterness. Beer polyphenols originate from the hops and malt or can be formed during the (bio)chemical transformation in the brewing process (Jerkovic et al., 2005).

Sweetness and coating attributes also showed a high correlation with each other, with a correlation coefficient of 0.89 ($p < 0.05$). Sweetness is a basic taste, like bitterness, recognized by the gustatory receptors (Meilgaard et al., 1999; Saison et al., 2009; Schmelzle, 2009). On the other hand, coating is a tactile sensation associated with increased viscosity or the presence of a coating layer after consumption (Agorastos et al., 2020; Klosse, 2013). Those sensations are related to the presence of fat, polysaccharide or proteins in beer (Schmelzle, 2009).

The outcomes suggested that trained beer sommeliers were not able to recognize the basic taste from the tactile sensations. This indicates that molecules that correspond to the perception of basic tastes, such as sugars (sweetness) and polyphenols (bitterness), influence the tactile sensations that result in overall mouthfeel sensations. Additionally, the two different tactile sensations, coating and drying, were negatively correlated at -0.67, while sweetness and bitterness also gave a similar trend with a correlation coefficient of -0.76. Therefore, polysaccharides and sugars seem to be able to mask the polyphenols and vice versa.

The third group, contracting/acidity, did not correlate with the other two groups. Contracting and acidity were found to be significant ($p < 0.05$) and correlated with 0.89. Contracting and acidity are associated with “irritants” or “chemesthetic compounds” that cause a contraction in the mouth, as carbon dioxide (CO_2) does in beverages (e.g., sparkling or mineral water,

sparkling wines, beer, and soda). Organic acids and minerals are reported to enter the ion channel of taste cells directly (Gilbertson et al., 2000; McCleskey & Gold, 1999; Simons et al., 2019).
This study suggests that beers can be classified based on two main sensations, tactile and chemesthetic sensations. The tactile sensations are dominated by drying and coating while contracting is associated with the contracting dimension.

Relations between sensorial and chemical data

Sensory evaluation is time-consuming and expensive. Innovative tools and measurement techniques that could measure "taste" could potentially be used as a replacement for sensory panels. To achieve this goal the determination of relationships between sensory attributes and chemical analyses needs to be further established.
Beer consumption is a multisensorial experience to which many different components contribute. Therefore, this study, using multiple factor analysis (MFA), investigated the relations between chemical and sensorial data of 24 beer samples. The MFA plot explained 62.03 % of the variance in the data with 45.25 % represented by dim-1 and 16.75 % by dim-2 (figure 2).
Bitterness and drying were associated with the chemical measurements as total iso-α-acids (TIA), bitterness units (BU), pH and total polyphenols content (TPC). Beers in high polyphenol content and hop acid are known to have a hard/bitterness sensorial profile (Oladokun et al., 2016). TIA and BU were correlated with drying at 0.81 and 0.84, while bitterness was correlated at 0.85 and 0.88 respectively. TPC showed a smaller correlation of 0.59 and 0.61 regarding drying and bitterness sensory attributes.
The correlations among the chemical measurement and sensory scores indicate that TIA is the most sensitive method regarding the association between drying and bitter sensations. The higher correlation of TIA compared to BU regarding bitterness can be explained by the extra sensitivity of TIA for bittering compounds. BU method is the most frequently employed. BU method also detects humulinones which can be found in high concentrations and influence the BU value while are not bitter components (Parkin & Shellhammer, 2018). While TIA focused only on the isomerized bitter compounds (Fritsch & Shellhammer, 2007). This indicates the presence of humulinones which increases the UB values without significantly influencing the bitterness. Therefore, TIA measurements can express the bitterness of commercial beers more accurately than IBU.
Until now the magnitude to which polyphenols contribute to bitterness and astringency is not clear. McLaughlin et al. (2008) suggested that increasing 15-20 mg/L in total polyphenols leads to an increase of one unit in BU. A correlation between TPC values and bitterness sensation was found. However, the contribution of polyphenols seems lower compared to the iso-α-acids.
These results suggest that increased pH can be associated with higher bitterness (figure 2). This can be explained by the solubility of humulones (α-acid), lupulones (β-acids) and humulinic acid at elevated pH values. Therefore, the higher pH values may increase the concentration of humulones in the finished beer, which gives a more intensely bitter, or drying sensation (Vrzal et al., 2021).
As can be expected, ethanol content and a burning sensation showed a significant correlation ($p < 0.05$) of 0.81, and carbonation correlated with the carbon dioxide content with a value of

0.57. These results are in line with the hypotheses and previous research regarding the chemical association of burning and carbonation sensations in beer when ethanol and carbon dioxide were the only components responsible for those sensations (Clark et al., 2011). In contrast with previous outcomes, ethanol did not show to influence the sweetness or bitterness of the beer samples (King et al., 2013).

Total acidity in beverages is known to associate with the perceived acidity by consumers (Chidi et al., 2018). In our study, perceived acidity and total acidity were significantly ($p < 0.05$) correlated with 0.76, while perceived acidity and pH were negatively correlated with – 0.76. The contracting sensation was correlated with total acidity and pH 0.66 and -0.76 respectively. Therefore, the contracting and acidity sensations can be expressed by TA and pH values.

As mentioned, a positive correlation between sweetness and coating was not found. Both attributes showed a negative correlation with BU, TIA and pH measurements. It was hypothesized that total fermented sugars would be associated with the sweetness and coating sensations, however, the present results did not show this relationship. Since the TS analysis quantifies only small sugar units, instrumental analysis of carbohydrates, like dextrins, pentosans and β-glucan could potentially be a better prediction for the coating mouthfeel (Ferreira, 2009; Langenaeken et al., 2020). Therefore, small sugar molecules cannot contribute to the sweetness and coating sensation, while the investigation of larger sugar-based molecules, like carbohydrates, could be dominant for the prediction of those sensations.

Finally, previous studies indicate that carbon dioxide and ethanol induce the suppression of sweetness (Clark et al., 2011). However, in this study only BU, TIA and pH were shown to mask the sweetness of the beer.

This paper provides novel findings that could be translated into biomimetic-based devices such as the electronic tongue (Ghasemi-Varnamkhasti et al., 2010, 2011, 2015; Peres et al., 2009; Wei et al., 2009). More specifically, this research provided new relationships between chemical composition and sensory attributes. Additionally, it identifies the important sensorial parameters that are needed for capturing mouthfeel sensations. Literature supports that this work correlates well with human gustatory sensation (Kovacs et al., 2009; Lozano et al., 2007; Uchida et al., 2001).

Conclusion

Mouthfeel characteristics are important for beer classification and consumer acceptance. The mouthfeel model proposed by Klosse (2004), provides a basis to classify taste and was used for the first time in beer application. In this study 24 beer samples were analyzed both sensorially by an expert panel and chemically. A multifactor analysis was conducted to investigate the relationships between sensory and chemical data. Three different mouthfeel dimensions were found to separate the different beer samples by 19 trained panelists, drying, coating and contracting. Drying and coating came out as the opposite dimensions of mouthfeel. Contracting is mainly associated with chemesthetic and irritant sensations and affects the perception of mouthfeel. The three dimensions can be used for commercial beer classification. Furthermore, the instrumental data showed significantly high correlations between the chemical measurements and the three sensorial dimensions providing new insights into the prediction of mouthfeel. This research suggests the potential for an objective, data-driven analysis of mouthfeel characteristics via instrumental techniques. Such a computational

approach could reduce the subjectivity in taste analysis and allows for the development and optimization of biomimetic devices for mouthfeel prediction. Taste classification will aid consumers in finding the product of their preference.

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General discussion

Introduction

Around 85 per cent of the new food products fail within two years (Salnikova et al., 2019). Although failure rates may vary among various categories of food products, in many instances there appears to be a mismatch between product characteristics and consumer expectations. The key to a successful new product development strategy is to ensure that products meet people's sensory needs (Rudder et al., 2001). Developing a new product formulation and reformulation strategy requires a good understanding of taste and consumer preferences. The current dissertation aims to get a deeper understanding of chemical and physical interactions which are related to mouthfeel sensations. The studies in this dissertation address: the current mouthfeel terminology and existing mouthfeel models (**chapter 2**); the importance of saliva involvement in the mouthfeel sensations (**chapter 3**); the salivary lubrication changes upon the addition of polyphenols and variation of the model solution matrix (**chapter 4**); the effect of various phenols on salivary lubrication (**chapter 5**); the cationic influence on salivary proteins and lubrication (**chapter 6**); and a novel approach with the mouthfeel model towards beers classification based on sensorial and chemical data (**chapter 7**). In this chapter, the results of these studies are summarized, the theoretical and practical implications are discussed, and the remaining questions are identified. Last suggestions and recommendations for future research are given throughout the chapter.

Main findings

The first step in gaining a better understanding of taste is to scrutinize the terminology which is commonly accepted and used by different disciplines. To start with, it is important to make a distinction between taste, tasting and flavor. Tasting corresponds to taste perception, where a subject perceives taste based on interpretation in the brain after the transduction. On the other hand, taste refers to the presence of taste-related compounds that stimulate the sensations of the mouth. Flavor is the conjunction of taste, smell and texture, while mouthfeel is part of texture and refers to the tactile and chemesthetic sensations (Gawel et al., 2018; Guinard & Mazzucchelli, 1996; Yarmolinsky et al., 2009).

Sensory terms are words that are used by consumers or panelists to describe the sensations that occur during food consumption. Classification of food products based on sensorial terms is important for the communication between consumers, food designers and industry. Communication between different disciplines is often more effective by using sensory lexicons or wheels (Lawless & Civille, 2013). Attempts at categorizing mouthfeel terms categorization are summarized and current mouthfeel vocabularies and wheels are highlighted in **chapter 2**. However, the effectiveness of the mouthfeel lexicons and wheels was proven as too complex for the training panelists introducing errors in taste communication (DeMiglio et al., 2002; King et al., 2003). Therefore, an empirical model related to mouthfeel classification is introduced in **chapter 2**. The mouthfeel model developed by Klosse (2013), is a simplified model which aims to improve communication among disciplines while summarizing different categories of sensorial attributes under three dimensions.

The outcome of **chapter 2** suggests the need for a more concise and overarching model that is relatively easy to understand and improves communication among disciplines. For the future of research in flavor of foods and beverages, it is essential to have consensus on the definitions of relevant concepts and to have a model (classification) based on an approach that is generally accepted. A mouthfeel model is potentially a powerful tool for food producers and researchers alike since it can be used to classify food based on the differences in food composition. Generalist descriptors that can be used to describe mouthfeel in foods and beverages can improve the communication between diverse audiences and contribute to the understanding of taste, flavor and particularly mouthfeel.

Lastly, **chapter 2** reveals the ambiguity of terms that are regularly used in literature. This can be explained by different foci in research. Three classes of research related to mouthfeel are identified: (1) product oriented (molecular attributes), (2) product/human oriented (human interface: receptors, saliva, chewing, etc.) and (3) human oriented (perception, brain interpretation after swallowing). In the 'product' approach, the focus is on the food components and the physical characteristics of a product. In this approach, taste and texture are classified based on the molecular composition of a food product, without involving a human. In terms of classification of mouthfeel, it is the most objective method. The second approach ("product/human") classifies mouthfeel during human consumption. Food ingredients interact with saliva (chewing, dilution, enzymatic reaction, etc.) and receptors. Because saliva bathes the sensory receptors, food components are first in contact with saliva. Due to all kinds of human differences in receptors, saliva composition, and chewing and swallowing behaviors, this method is less objective. From this perspective, mouthfeel is the result of the interaction between food products and saliva. As a result, mouthfeel can be described as the interaction between taste and texture. The third category of analysis is the "human" approach. This type of research focuses on flavor perception after swallowing and processing in the brain. Spence (2017) introduced 'Gastrophysics' to describe this domain. It is defined as 'the scientific study of those factors that influence the human multisensory experience while tasting food and drink'. Under this new concept, it was introduced a new definition the "flave". Flave is the complete integration of all senses during tasting. There is no doubt that different research orientations result in different findings, which contributes to discrepancies in the results of sensory research in general. There is a need for an effective communication model for the classification of taste in food products. Communication between product specialists, researchers, sensory panelists, etc., will be improved by an objective, well-accepted model. It is likely that the most objective classification will be derived at the product level and will be based on real and measurable product characteristics.

Understanding mouthfeel sensations

Mouthfeel refers to the chemical-physical-textural sensations in the mouth during and after the consumption of food or beverages. Mouthfeel sensations are commonly defined as "the tactile, irritant and thermal sensations resulting from the activation of chemosensory and somatosensory receptors within the oral cavity by chemical stimuli" (Gawel et al., 2018).

Important tactile sensations have been defined by Klosse (2013) as the drying and coating sensations.

These sensations can be aroused by the presence of compositional factors such as polyphenols, polysaccharides, proteins, organic acids carbon dioxide, ethanol etc. Additionally, pH, ionic strength and matrix interference (masking) can also affect the tactile sensations. It is known that the bulk and surface properties of beverages are affected by product characteristics such as mentioned above. Therefore, rheological and tribological measurements have been used by researchers to characterize and detect changes in food products. An emerging tool that helps understand oral processing is tribology (Stokes et al., 2013). Tribology is the science of wear, friction and lubrication and includes how interacting surfaces and other tribo-elements behave in relative motion. Soft-tribology can mimic aspects of in-mouth lubrication by applying human saliva and soft surfaces. Therefore, uncovering the interactions that correspond to physical changes in the mouth is important for mouthfeel sensations.

Involvement of saliva in mouthfeel sensations

C**hapter 3** reviews the importance of saliva involvement in mouthfeel sensation. Recently, researchers are increasingly interested in the dynamic aspects of the oral sensation of food and beverages. Saliva is an important physiological component related to mouthfeel sensations. As a bio fluent possesses multiple functions in the oral cavity, where lubrication properties are the most significant for the mouthfeel sensations. Changes in salivary integrity and lubrication properties are associated with drying (astringency) or coating sensations (Mosca & Chen, 2017).

Salivary lubrication is associated with the presence of salivary proteins on oral surfaces. Salivary proteins are capable of forming salivary films (pellicles). The mucosal pellicle is a biofilm that covers the oral cavity (Odanaka et al., 2020). This film is unique among other biological lubricants since it resists wear and provides low friction (Boyd et al., 2021). Literature suggests that proline-rich proteins (PRPs) and mucins are the main salivary proteins responsible for oral lubrication (Boze et al., 2010; Sarni-Manchado et al., 2008; Cook et al., 2017; Gibbins et al., 2014). In addition, **chapter 3** provides further details regarding the interaction between salivary proteins and food ingredients that influence oral sensation. More specifically, the friction-based theory of drying sensations is mentioned in detail. It has been suggested that the aggregate formation of polyphenols and salivary proteins results in astringent sensations (Bajec & Pickering, 2008; Laguna et al., 2017). Two main interactions are responsible for the aggregate formation such as hydrophobic interactions and hydrogen bonds (Charlton et al., 2002; Jöbstl et al., 2004). Additionally, important parameters responsible for salivary protein aggregation by phenols are the ratio of protein types: polyphenols, molecular weight, pH, temperature, ionic strength and the type of polyphenols (Bajec & Pickering, 2008). However, until now the role of aggregate formation on lubrication loss is still limited investigated. Thus, one of the aims of this dissertation was to identify the chemical interactions and properties that are more important for oral lubrication. In the conclusion, a brief mention of the physical and chemical interactions responsible for the

coating sensations was summarized as well. The coating sensations occur upon consumption of food and beverages containing sugar and fat. It has been found that electrostatic, steric, and hydrophobic interactions are dominant in the adhesion and spreading of emulsion droplets on oral surfaces, with hydrophobic interaction being the most significant.

Uncovering the oral lubrication of beverages

In **chapter 4**, the effect of molecular weight, pH and the masking effect of mannoproteins in combination with gallic acid on the lubrication of saliva were studied. Oral lubrication is a dynamic phenomenon where different interactions take place between oral lubricants and surfaces. Soft-tribology applied for the understanding of oral lubrication incorporates the study of friction under soft surfaces, like polydimethylsiloxane (PDMS) elastomer which is known to mimic oral tissue. Therefore, a good approximation of oral friction can be established by the use of soft-tribology.

Astringency sensation and aggregate formation between polyphenols and salivary proteins have been suggested to depend on various parameters such as pH, molecular weight and matrix interferences (Carvalho et al., 2006; DeMiglio et al., 2002).

Human saliva was collected and interacted with different model solutions. A dynamic soft-tribological approach was applied to characterize the friction response for model solutions and their interactions with the ex-vivo salivary pellicle. To further investigate the interactions between the salivary proteins and polyphenols zeta-potential (electric potential in the interfacial double layer) and size distribution were used. It was found that the addition of gallic acid and tannins significantly reduces the lubrication properties of saliva. The induced friction depended on the polyphenol concentration. Tannins, as larger molecules, had a greater impact on salivary lubrication loss than gallic acid. Lowering pH increased the friction of the system due to stronger interactions between gallic acid and salivary proteins. A strong correlation was observed between the hydrodynamic diameter of aggregates and the friction values. Those outcomes suggest that the formation of aggregates determined the lubrication behavior of saliva.

Phenolic compounds which are present in beverages and foods have diverse molecular characteristics. Characteristics such as the degree of polymerization, galloylation, interflavanic bonds, and the number of hydroxyl moieties present in the B-ring of the flavanic nucleus are important parameters that can influence the astringency perception (García-Estévez et al., 2018). The effect of different phenolic components on saliva lubrication behavior was studied more in-depth in **chapter 5**. Four different phenols (gallic acid, catechin, epigallocatechin, epigallocatechin gallate) were selected. The interactions of phenols and salivary proteins were detected by soft-tribology, zeta sizer, SDS-page and tensiometer. These experiments showed that friction increases significantly with phenolic components with extra hydroxyl and galloyl groups. The salivary lubrication loss is explained by the aggregated salivary proteins which result in lower wetting salivary ability. More specifically, the aggregation of glycosylated proline-rich proteins by phenols were suggested to be associated with the loss of salivary lubrication. This chapter provides new insights into the interactions between saliva proteins

and polyphenols that dominate food and beverage consumption. Additionally, the outcome of this study shows that instrumental analysis can be a valuable tool to predict oral sensations. This is important for the design of new products or product reformulation.

However, not only polyphenols were found to be responsible for changes in lubrication behavior. Cations have also been suggested to associate with astringent sensations. Al^{3+} and Fe^{3+} were found to evoke an astringent sensation by crosslinking mucins in the mucosal pellicle (Biegler et al., 2016; Lim & Lawless, 2005). Additionally, the multivalent cations, and ionic strength can also affect the lubrication properties of saliva. **Chapter 6** summarizes the results related to the effects of cations and epigallocatechin gallate on salivary lubrication. The influence of cationic valences, molar concentrations and ionic strength on salivary lubrication was investigated in the presence of human saliva. Soft tribological measurements were used to characterize the friction response of the minerals to the ex-vivo salivary pellicle. Zeta-potential, SDS-page, size distribution, and viscosity measurements were conducted to help further explain the lubrication changes. Trivalent salts significantly reduced the lubrication properties of saliva, while divalent and trivalent salts did not show significant differences. The Mucins-Fe^{3+} complexes were responsible for the lower lubrication ability. However, at 150 mM ionic strength, KCl provided extra lubrication via hydration lubrication. Last, an antagonistic mechanism between trivalent salts and EGCG was profound. The chelating interactions between those components result in less binding to salivary proteins and less friction. This chapter provides insights into the effect of minerals present in beverages. As the changes in salivary properties have been linked with oral sensations, such information can be important for the design of new beverages or the reformulation of existing ones.

Translation of beer characteristics into mouthfeel sensations

The taste of beer is important for the acceptance of the product by consumers. Beer is a complex solution with different components that contribute to mouthfeel sensations. Currently, there is a limited association between the sensory properties of beer and its compounds. A better understanding of the relationship between sensory and chemical analysis could be useful in developing biomimetic-based devices for the food industry, such as electronic tongue (Ghasemi-Varnamkhasti et al., 2010, 2011, 2015; Peres et al., 2009; Wei et al., 2009). Therefore, the sensorial and chemical properties of commercial beers were investigated. C**hapter 7** shows the results of a study where twenty-four beer samples were evaluated by a trained panel of nineteen experienced beer sommeliers. Furthermore, the beers were chemically analyzed on total acidity (TA), total flavonoids (TF), total polyphenols (TPC), total sugars (TS), color, pH, carbon dioxide content, ethanol, bitterness units (BU) and total Iso-α-acids (TIA). The outcomes were analyzed by performing principal components analysis (PCA) and multiple factor analysis (MFA) analyses. The results show that panelists were able to classify the beers into three main sensorial groups. Those groups matched the mouthfeel model axes, drying, coating and contracting. A noteworthy finding was that drying and coating were negatively correlated, whereas chemesthetic groups seem to have a different effect on

mouthfeel sensations. The results of this study support the approach based on the mouthfeel model. A strong correlation was found between the sensory attributes and the chemical data. Thus, the findings of this study suggest the potential for measuring mouthfeel sensations using analytical methods.

The outcomes reported in the previously mentioned chapters suggest the need for a rethinking of mouthfeel definition and therefore adopting a new approach towards taste classification. Tastants and textural compounds are both important for mouthfeel characteristics since both are responsible for physical changes that lead to tactile sensations. Therefore, as can be seen in **chapter 7** sensory attributes corresponding to gustatory and trigeminal receptors could not be separated by the trained panelists which lead to an overlap of tastants and textural compounds. Thus, the mouthfeel model appears to be a useful tool for capturing the sensory characteristics of beverages.

Methodological considerations

Limitations

Several limitations should be considered when interpreting the results of the studies in this dissertation. The experiments reported in **chapters 3 to 6** were mainly focused on salivary lubrication which was measured with soft-tribology coupled with PDMS surface. PDMS is commonly used as a mimic elastomer of the tongue to reduce the natural changes among biological surfaces. Several manufacturers of tribometers recommend the use of PDMS as a tribomaterial since is easy to synthesize and shape, is low cost, the material does not swell in water and remains stable over a long period at a wide range of temperatures (Rudge et al., 2019). However, it is important to realize by using tribometers with synthetic surfaces to mimic biological processes parameters such as surface roughness, stiffness and hydrophobicity need to be taken into account. The PDMS characteristics as expected are not the same as with human tongue but despite those differences, the PDMS use is certainly already better than using a commonly used very hard surface such as steel.

Furthermore, frictional measurements are system-dependent, therefore every technique has limitations such as reciprocating or rotating movement, applied force, and velocities. Additionally, as system-dependent, friction cannot be compared with other tribo-system which makes it more challenging to link friction measurements with astringency perception.

Instrumental studies using saliva have some limitations as well. The most common parameters which influence the outcome of such a study are the type, amount and individual characteristics of saliva use. The studies in the experiments of this dissertation used unstimulated human saliva from Caucasian donors between the ages of 22 and 28 years. Saliva collected by a singular donor or saliva pooled from a small number of subjects does not represent the average of the naturally occurring differences between individuals over the day (Muñoz-González et al., 2019). Therefore, those studies cannot represent the general characteristics based on the general population. Lastly, demographic and age characteristics regarding oral lubrication cannot be

extracted from this dissertation which has been suggested to influence astringency perception (Bajec & Pickering, 2008).

The results of the beer experiment reported in **chapter 7** may be influenced by the selection of beer sommeliers for the beer sommelier for the sensorial evaluation of the beers. A consumer study is needed to further evaluate the beer classification based on mouthfeel terms. Additionally, a larger panel with variations based on age and ethnicity should be considered for getting a better illustration of the different parameters regarding mouthfeel classification. Further details about the importance of demographic characteristics on mouthfeel sensations will be discussed below.

Recommendations

Even though experiments using soft tribology have exponentially increased, there is no consensus on the accuracy regarding the measurements of friction on soft tribology and how to obtain desired correlations with in-vivo experiments. There are many challenges to establish a direct link between frictional data and sensory attributes perceived by human subjects.

For meaningful mouth-mimicking experiments, the following recommendations are suggested.

- Mimicking oral behavior can be optimized by using a synthetic surface that represents the oral surface as accurately as possible.
- Material properties such as surface roughness, stiffness and hydrophobicity need to be designed in the same range as the human tongue and palate.
- Soft tribology will greatly benefit from new methods that allow various sliding movements as food is not moving in a single direction during consumption.
- Having a better visual illustration of what is happening in the boundary layer of the sliding surfaces will be essential to understanding frictional sliding (Descartes et al., 2015; Suhina et al., 2015).

Using artificial saliva is an innovative approach to overcome the complexity of using human saliva. The application of artificial saliva on lubrication measurement will lead to little variation. This helps to establish a better understanding between the chemical interactions and their impact on oral lubrication. Artificial saliva has been reported to be stable reproducible and comparable among studies (Laguna et al., 2021). However, currently, the viscoelastic properties of artificial saliva are hardly comparable to those of human saliva. The main reason for this mismatch is that artificial saliva only mimics the ionic strength of saliva and incorporates mucin proteins. This is inadequate. Mucin alone cannot replicate the tribological properties of the human salivary pellicle (Sarkar et al., 2019). So the development of new artificial saliva which could elaborate on the properties of human saliva would be a valuable tool for future research.

Adding to saliva variation, papillae and tactile sensitivity could bring differences in the astringency sensations between people. Demographically, Asian consumers are reported to be more sensitive to tactile stimulations than Caucasians (Komiyama et al., 2007). The Asian

population presented higher fungiform papillae density than Europeans, therefore, fungiform papillae have been associated with tactile sensitivity (Olarte Mantilla et al., 2022). It appears that more evidence is needed to clarify the genetic basis between papillae density and oral sensitivity to astringency (Komiyama et al., 2007). Additionally, females are more sensitive to oral tactile stimulation than males, with the females presenting higher fungiform papillae density (Linne & Simons, 2017; Piochi et al., 2019). If there were a better understanding of such personal differences this information could potentially be used in product formulation.

Regarding **chapter 7**, a different sensory protocol could potentially be used to capture the sensorial attributes more accurately. Quantitative sensory analysis is based on the assessment of the perception of sensory attributes as "static". Sensory evaluation mostly from flavor and mouthfeel perspective is a dynamic phenomenon that alters during oral processing (Hutchings & Lillford, 1988). Therefore, quantitative analysis can capture the changes in the sensory attributes based on time. Dynamic methods provide insights about variations in perception intensity during time. Those sensory evaluation methods are closer to reality compared to static sensory methods because they provide information for every moment of the oral processing making the classification of food or beverages more sensitive (Dijksterhuis & Piggott, 2000).

Practical implications and future directions

Due to the high failure rate of new products on the market today, the characteristics of foods and the preferences of consumers need to be understood. To achieve such a goal, taste and flavor perception insights need to be taken into account. Examples of how to address such challenges are discussed below.

Importance of oral lubrication

Studies 3, **4** and **5** provide evidence of the effect of food ingredients on oral lubrication. These insights provide a new understanding of tactile sensations which are influenced by changes in oral friction changes and can affect consumer acceptance and prospective consumption (Silletti et al., 2007). Oral friction results from the aggregate formation of salivary proteins by food ingredients such as multivalent, polyphenols, etc (Panda & Chen, 2021). Different molecular properties such as molecular weight, degree of polymerization, galloylation, interflavanic bond and number of hydroxyl substitutes are important parameters that affect oral lubrication (García-Estévez et al., 2018).

In the present studies, the effect of astringent components and characteristics in oral lubrication was explored using soft tribology. The increase of astringent compound concentration and the lower pH increases friction. Higher molecular weight and the number of hydroxyl and galloy groups decrease the salivary lubrication ability. Glycosylated proteins (mucins and gPRPs) were responsible for the lubrication behavior of saliva and further aggregation of those proteins results in loss of lubrication. However, the current data are missing the direct link with sensory analysis, even though that oral friction is highly related to oral sensations (Mosca & Chen, 2017).

Additional experiments conducted in a complex matrix or real beverages could provide more insights into the antagonistic mechanism and interactions among food ingredients which affect oral lubrication. Furthermore, it would be valuable if more instrumental techniques could be optimized and compared for the accuracy of measuring oral sensations. Such an innovation would provide a useful tool for product designers.

Prediction of mouthfeel

Sensory evaluation is expensive, time-consuming and subjective. At the moment the food industry is seeking more innovative approaches to taste evaluation. Objective methods where mouthfeel can be quantified based on compound composition and characteristics are a novel way of evaluating the taste of food. **Chapter 7** shows how analytical methods can be associated with sensory characteristics. The results indicate that measuring mouthfeel characteristics based on instrumental analysis can be applied to the mouthfeel. The PCA in this study confirmed the dimensions of the mouthfeel and identified drying and coating as opposites, contracting forces interact on these dimensions, Valency and pH are likely to play a larger role than previously reported (**Chapter 6**). Further optimization of the current analytical methods is needed to achieve a higher correlation with sensory analysis.

Such studies can be used for the development of biomimetic-based devices such as the electronic tongue (Ghasemi-Varnamkhasti et al., 2010, 2011, 2015; Peres et al., 2009; Wei et al., 2009). Previous studies have documented the potential of such as possibility (Kovacs et al., 2009; Lozano et al., 2007; Uchida et al., 2001). However, despite the effort, those devices still need further development and optimization. The dimensions of the mouthfeel model could be used in the design of biomimetic devices that are used to evaluate taste.

Conclusion

Although textural sensations have been well-investigated there is still confusion regarding the definition, lexicon, and understanding of those sensations. More issues require further study such as 1) further optimization of the current method regarding oral sensations, 2) gaining a deeper understanding of the origin of tactile sensations, and 3) further optimizing the mouthfeel model in order to become applicable for commercial application. The current studies included in the dissertation provide only a first attempt at tackling these issues. The outcome will hopefully spur further investigation and discussion in an area of great interest for food science. The improved knowledge of textural sensations can be applied in the food industry, and it is likely to have a positive impact on the success rate of new food products.

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Appendices

Summary

Impact Addendum

Curriculum Vitae

Publications

Acknowledgements

Summary

A recent (2019) US report indicates that 85 % of new food-packaged products fail within two years from the market. It is believed that one of the main reasons behind this fact is the mismatch between consumers' expectations or preferences and the sensorial characteristics of new products. Therefore, understanding consumers' preferences and matching them with the desired sensorial properties is crucial for a successful strategy of new product design.

However, to understand and predict the sensorial properties of food products, many hurdles need to be taken. First, a well-accepted and defined lexicon among consumers, marketers and food researchers/designers needs to be established. Additionally, objective methods for sensorial characteristics evaluation must be further developed. Until now sensory evaluation is mainly conducted by subjective methods that are expensive, time-consuming and less reliable than the objective methods. However, developing and establishing objective methods has many challenges. The researchers need to gain a good understanding of the origin and different characteristics that arouse oral sensations. Furthermore, the development of chemical or physical analytical protocols needs to be established based on the related characteristics of oral sensations. Lastly, chemometric approaches are essential for the interpretation of potential patterns between chemical analysis and sensory attributes.

This thesis deals with uncovering the important characteristics of mouthfeel sensations that occur during consumption. Therefore, chapter 1 describes in detail the importance of mouthfeel sensations and disguising flavor, taste, mouthfeel and aroma. The oral processing and the importance of physical characteristics and changes which occur in the mouth during consumption are highlighted as well.

The current confusion and lack of a co-existing mouthfeel terminology in the literature are extensively reviewed in chapter 2. Additionally, an empirical mouthfeel model is introduced in the same chapter. The mouthfeel model is summarized by three axes, coating, contracting and drying. Where coating and drying axes are the subcategories of the tactile sensations when contracting summarizes the chemesthetic sensations. This simplified model provides classification among different food products based on the three axes.

To gain new insights into oral sensations, and more specifically into tactile sensations, chapter 3 highlights the importance of saliva involvement in oral sensations. As a bio fluent, saliva possesses unique physical properties due to salivary proteins. Therefore, this chapter reviews the salivary properties which are responsible for the lubrication properties, and the interaction of food components with the salivary proteins which leads to changes in the salivary film integrity and results in the arouse of tactile sensations. The properties of salivary lubrication and the interactions that occur during consumption. This is related to changes in the saliva composition of elderly people or as a result the use of pharmaceuticals. As salivary lubrication is intimately connected to the consumption and liking of foods and beverages these insights are likely to be useful in helping people to eat well. Research in this field merits further elaboration.

In chapter 4, the effect of molecular weight, pH and the addition of mannoprotein on the lubrication behavior of saliva was investigated. Human salivary proteins interacted with model solutions, containing tannic acid and gallic acid, under different conditions. The friction of the system was measured by soft tribology and hydrodynamic diameter by zetasizer. The results

of this study suggested that higher molecular weight and lower pH decreased the lubrication properties of saliva. Additionally, the presence of mannoproteins in gallic acid solutions resulted in less binding of the gallic acid into salivary proteins. A high correlation was observed between aggregate formation and friction which lead to the conclusion that aggregate salivary proteins could not provide their lubrication properties.

Chapter 5 describes a study about the effect of different phenols on salivary lubrication behavior. Four phenolic components were selected based on the difference between hydroxyl and galloyl groups. The lubrication properties were measured by soft tribology, aggregate formation of specific salivary proteins was investigated using SDS-page, while the wetting properties were recorded via tensiometer. The extra hydroxyl and galloyl groups decreased the salivary lubrication. The aggregate formation between phenols and salivary proteins lowers saliva wetting properties which can explain the lower lubrication properties. Finally, the glycosylated proline-rich proteins showed the highest association regarding lubrication loss.

The effect of cations on salivary lubrication was investigated in chapter 6. Cationic valences, concentrations and ionic strength were examined for their impact on the lubrication behavior of saliva. Additionally, the interactions among phenol, cation and salivary proteins were studied as well. Friction, hydrodynamic diameter, viscosity, and reduction of protein density were used for getting insights into the interactions between salivary proteins and cations. Trivalent salts were able to cross-link mucins and form aggregates which lead to lubrication loss. KCl at 150 mM ionic strength value was able to provide extra lubrication via hydration. Epigallocatechin gallate interacted with Fe^{3+} via chelating interactions which led to lower binding of Fe^{3+} to mucins and reduced the induced friction by Fe^{3}.

Commercial beers were evaluated based on mouthfeel sensorial and chemical characteristics in chapter 7. Chemometric tools were applied to discover potential patterns in the data. Panelists were able to classify the sensorial attributes into three main sensorial categories. The three categories are associated with the mouthfeel model axes. A clear correlation was observed between sensory and chemical data. Total polyphenol content, bitterness units, pH and total iso-α-acids were correlated with bitterness and drying. Carbon dioxide is correlated with carbonation, while ethanol is associated with burning. Last, pH and titratable acidity correlated with contracting. These outcomes indicate the mouthfeel model implication and the potential of objectively measuring mouthfeel.

Impact addendum

The success rate of new food products or food reformulation has been reported to range below 20 % at the moment. One of the main reasons for the high failure rate is the mismatch between consumer expectations or preferences and the food's sensorial properties. Therefore, to achieve a successful product design food industry and designers/researchers need to gain a better understating of the food sensorial properties, and their origin while developing evaluation methods that can capture those properties.

Understanding and being able to measure the sensorial properties of food is harder than it sounds. The absence of a well-defined lexicon among industry, food researchers/designers and consumers is still lacking. Additionally, the use of subjective evaluation methods, such as sensory analysis, can be influenced by psychological and physical factors, while those methods are expensive, time-consuming, low repeatable and reproducible.

Overcoming such problems first a common lexicon for effective communication among disciplines needs to be established, while the establishment of objective sensorial evaluation methods needs to be further developed. However, to effectively design objective evaluation methods, understanding the origin and the characteristics of oral sensations are important.

This dissertation aimed to expand the understanding of the origin of mouthfeel sensations, based on physical changes in human saliva and the application of those insights to improve the chemical measurement specialized in mouthfeel sensations. Lastly, the association of the sensory attributes with the chemical measurement was evaluated by chemometric analysis.

Understanding salivary lubrication

The present thesis is a series of studies on mouthfeel sensations regarding their origin and the different chemical properties/interactions which induce those sensations. Changes in salivary film integrity are important for the perception of mouthfeel sensations. Interaction of polyphenols with salivary proteins lowers the salivary lubrication ability, while larger molecular weights and lower pH further increase the friction of oral surfaces. Cationic characteristics, such as valences, ionic strength, et cetera, influence the lubrication properties of saliva as well. Trivalent cations lower salivary lubrication while monovalent cations at high ionic strength values provide extra hydration lubrication. The findings of this dissertation provide new insights into salivary lubrication which is responsible for the drying sensations during food consumption. The findings suggest that measuring both polyphenols and cations concentration and their molecular characteristics could potentially be used for the prediction of drying perception.

Chemometrics and mouthfeel sensations

Predicting or measuring “flavor” based on chemical measurements is a valuable tool for the food industry and researchers. To objectively quantify flavor, a good understanding of the components and their properties regarding oral sensations is a necessity. Therefore, by incorporating this knowledge of food components and their chemical properties, new

innovative chemical and chemometrics analyses can be further developed to quantify oral sensations. The results show that applied chemical analysis regarding sensory sensations is highly correlated with sensory attributes. Those results suggest that oral sensations can be potentially classified based on tactile sensations and chemometric sensations. Where tactile sensations, such as coating and drying are negatively associated with each other, while the chemometric sensations do not influence the tactile sensations.

The use of innovative chemical analysis and chemometric techniques can potentially be used to predict oral sensations. Therefore, biomimetic devices, like artificial tongues, can potentially be used to replace the existing subjective sensory evaluations, provided that these instruments are based on a useful model for taste classification.

Conclusion

In sum, this dissertation enhances the understanding of mouthfeel sensations, especially based on salivary lubrication changes. Additionally, the use of chemometric tools showed a high correlation between sensory attributes and chemical analysis. Although these implications warrant further investigation, they can provide food researchers, designers and industry with novel strategies to improve the success rate of the new products or product reformulation. Therefore, this dissertation provides a useful starting point for developing novel strategies and tools for predicting flavor properties.

Curriculum Vitae

Georgios Agorastos was born on July 6, 1991, in Thessaloniki (Greece). He graduated from secondary school in 2009, after which he completed a Bachelor of Science (diploma, 240 ECTS) in Food Technology in 2015 at the School of Agricultural Technology, Food Technology and Nutrition of International Hellenic University. He completed a Master of Science in Food Technology in 2019 at Wageningen University with the specialization in product design. In March 2019 he started his PhD project at the Faculty of Engineering at Maastricht University Campus Venlo. During his PhD project, he investigated the effect of physical changes on mouthfeel sensations under the supervision of Emo van Halsema, dr. Peter Klosse and prof.dr Aalt Bast. Furthermore, he was visiting PhD at the food physics and physical chemistry department at Wageningen University under the supervision of prof.dr. Elke Scholten.

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Agorastos, G., van Halsema, E., Bast, A., & Klosse, P. (2020). Review of Mouthfeel Classification. A New Perspective of Food Perception. *Journal of Food Science & Nutrition*, 1–10. https://doi.org/10.46715/jfsn2020.09.1000107

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Agorastos, G., van Uitert, E., van Halsema, E., Scholten, E., Bast, A., Klosse, P. (2023). The effect of cations and epigallocatechin gallate on salivary lubrication.

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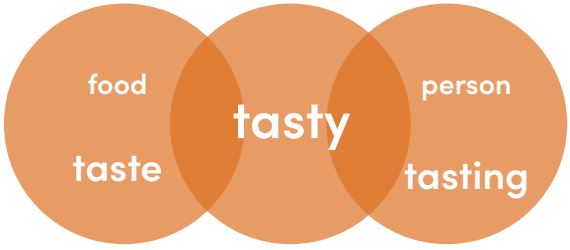
The Academy for Scientific Taste Evaluation

Dit proefschrift is geïnitieerd door de Stichting T.A.S.T.E. en mogelijk gemaakt door de provincie Limburg. Het onderzoek is uitgevoerd onder toezicht van de Universiteit Maastricht (campus Venlo) en gebruikmakend van de faciliteiten van de Brightlands campussen. Ook de Zuyd Hogeschool is met studenten en begeleiding betrokken geweest bij het onderzoek.

De stichting T.A.S.T.E. (The Academy for Scientific Taste Evaluation) is opgericht door dr. Peter Klosse om de consumptie van gezond en duurzaam voedsel te bevorderen. Een middel om dat te doen is smaak meetbaar te maken en sensorische meetmethoden op smaakgebied te verbeteren. Dat was de aanleiding voor dit promotieonderzoek. De resultaten laten zien dat het mondgevoelmodel een bruikbare basis vormt het meten van smaak. De resultaten van fysisch-chemische analyses in universiteits-laboratoria konden worden gecorreleerd aan de sensorische waarnemingen door smaakpanels. Een dergelijke instrumentele en objectieve benadering kan worden gebruikt voor de ontwikkeling van nieuwe, of de verbetering van bestaande voedingsproducten door ze bijvoorbeeld gezonder te maken. Het PhD onderzoek staat ter beschikking van bedrijven, overheden en onderwijsinstellingen.



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