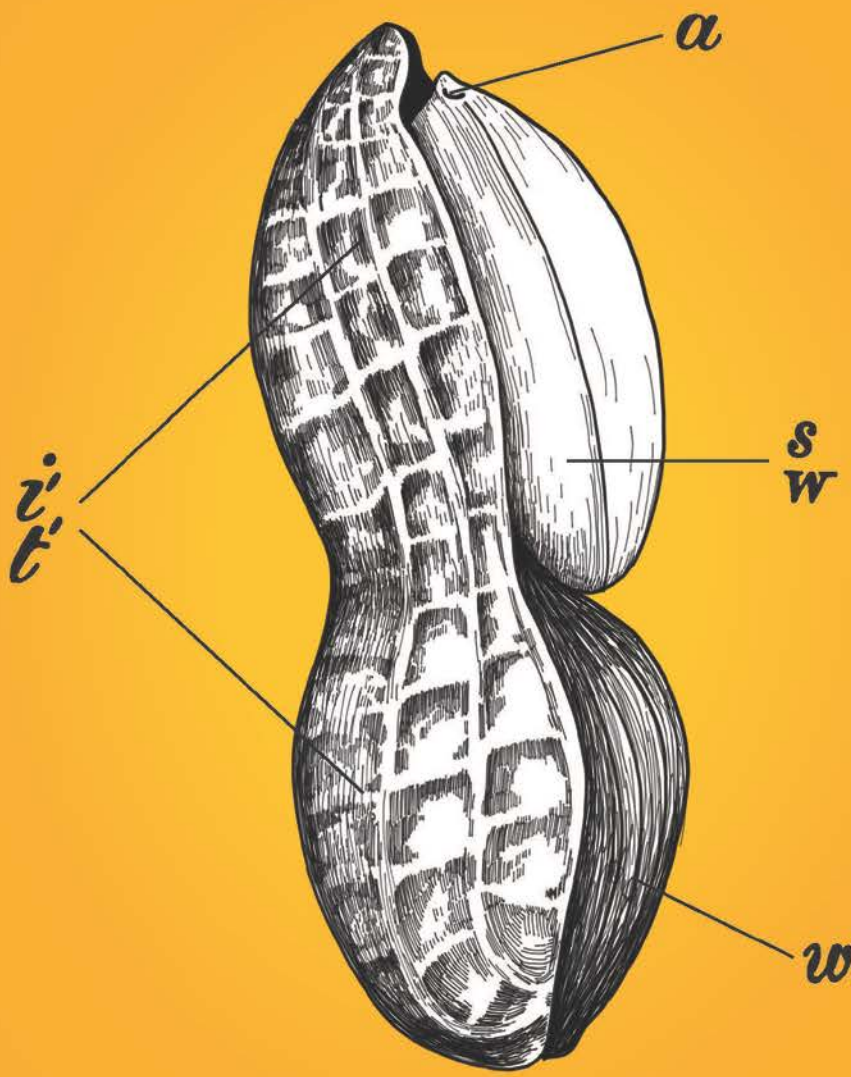
Dimitrios Lykomitros

Multivariate relationships between instrumental attributes, microstructure, sensory profiles and consumer preference in roasted peanuts (*Arachis spp*)



2018 Dimitrios Lykomitros | Multivariate relationships between instrumental attributes, microstructure, sensory profiles and consumer preference in roasted peanuts (*Arachis spp*)

Propositions

1. People can tell if a peanut is fresh.
   (this thesis)

2. The only difference between flavour “defect” and flavour “character” is the concentration of the responsible compound.
   (this thesis)

3. Identifying a correlation is more useful than proving causation.

4. Practical applicability is more important than statistical power.

5. Easier access to information has brought about the second dark age.

6. Food is a too complex subject for laymen to understand.

Propositions belonging to the thesis, entitled

Multivariate relationships between instrumental attributes, microstructure, sensory profiles and consumer preference in roasted peanuts (*Arachis spp*).

Dimitrios Lykomitros

Wageningen, 4th of April 2018

Multivariate relationships between instrumental attributes, microstructure, sensory profiles and consumer preference in roasted peanuts (*Arachis spp*).

Dimitrios Lykomitros

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Dimitrios Lykomitros

Thesis

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in the presence of the
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To Bianca

Abstract

The objective of this research was to determine how raw material and process technology selection can affect the organoleptic characteristics of roasted peanuts, and further identify which of those characteristics drive liking with European consumers. Twelve different raw peanuts of various market types, origins and grades were treated by eleven different process (maceration in water, aqueous glucose and at different pH followed by frying or baking), resulting in 134 unique samples, which were profiled by a sensory panel (SPECTRUM, DSA) and analysed for colour (CIELAB), fatty acid composition (FAMEs-GC-MS), headspace volatile composition (DHS-GC-MS, SPME-GC-MS, and GC-MS-O), sugar profile (ion chromatography), and textural characteristics (large deformation compression).

Principal Component Analysis, Canonical Variate Analysis and General Linear Model regressions were used to identify differences in sensory attributes, fatty acid and headspace volatile profiles, and to relate them to raw materials and process conditions. Process selection had a large impact on the final sensory characteristics. Specifically, baking reduced 'roasted peanut' and 'dark roast' and increased 'raw bean' aromas compared to frying. Maceration significantly increased 'roasted peanut' and 'dark roast', and reduced 'sweet', 'raw bean' aromas, and sweetness. 'Crispy', 'crunchy' and 'hardness' attributes were significantly rated higher in the presence of glucose in the medium, while the effect of pH was minor. The microstructure was further probed with confocal microscopy and X-ray tomography. The degree of alveolation was similar in differently processed macerated peanuts, even though sensory attributes were significantly different. Quantitative data on alveolation showed that microstructure disruption through steam generation cannot explain all the texture differences among processed peanuts.

Correlations between sensory and instrumental attributes were also explored using Partial Least Squares Regression. Several headspace volatile compounds which positively or negatively correlated to 'roasted peanut', 'raw bean', 'dark roast' and 'sweet' attributes were identified. It was also determined that sensory texture attributes can be predicted from instrumental measurements, but a multivariate approach using both hardness and toughness data from different probe geometries was necessary.

26 of the most varied samples were hedonically rated by consumers in The Netherlands, Spain and Turkey (n>200 each). Preference map models revealed that the drivers of liking are similar across the three countries. Sweet taste, 'roasted peanut', 'dark roast' and 'sweet' aromas and the colour b* value were related to increased liking, and 'raw bean' aroma and bitter taste with decreased liking. The colour coordinates, sucrose content, several pyrroles and low levels of hexanal and 2-heptanone were strong predictors of both preference and perceived freshness.

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1 Introduction

1.1 Definitions, nomenclature and conventions

Several terms are commonly used when referring to peanuts, whose meaning may not be obvious to those not familiar with the crop. Firstly, the term *peanut* itself is misleading, as the *Arachis Spp* are not botanically nuts, but legumes. Eighty one species of *Arachis* have been identified, including the domesticated *Arachis Hypogaea L.* and *Arachis Fastigiata*, while new species are being discovered in the tropics (Stalker, Tallury, Seijo, & Leal-Bertioli, 2016).

Peanuts, also known as *groundnuts*, are generally divided into *market types*. Peanuts of the same market type generally exhibit similar morphological characteristics, such as general kernel size, skin colour and number of kernels in a pod, but it is not a taxonomical classification and several genetically different plants can be members of the same type (Woodroof, 1983). The most common market types include *Spanish* (smaller kernels with reddish-brown skin, predominantly used in candy and peanut butter making due to their slightly higher oil content), *Runner* (the most common type generally due to higher crop yields), *Virginia* (largest kernels, most often consumed in the shell), and *Valencia* (three or more kernels in a pod, generally sweeter than other types) (American Peanut Council, 2014). Valencia and Spanish types are examples of the *Arachis Fastigiata* species, whereas Runner and Virginia of the *Arachis Hypogaea* species. A visual representation of the terminology hierarchy can be seen in **Figure 1**.

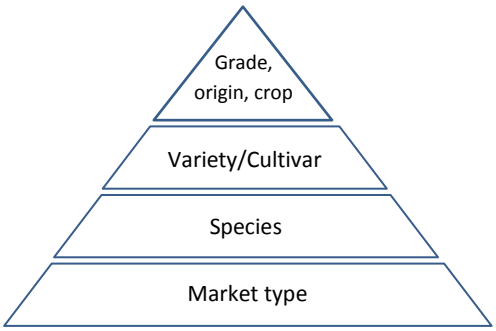

Figure 1: Hierarchy of terms used in the peanut industry. Terms on the bottom of the pyramid resolve larger differences compared to terms on the top of the pyramid.

Third in the hierarchy of classification is 'variety', which is the term used commercially to describe the specific hybrid and is related to the plant genome. Hybridization can be done within or across the different *Arachis* species, although a

considerable compatibility limitation exists due to the different chromosome structure across several species (Stalker et al., 2016). Over fifteen thousand hybrids are known to exist, and hundreds more are released yearly by commercial breeders, mainly focusing on pathogen and drought resistance and increased yield (Stalker et al., 2016). One of the most commercially successful traits bred is the 'high oleic' trait, which is responsible for a higher oleic to linoleic acid ratio. The trait can now be found in several varieties, and although it has been found not to significantly affect the organoleptic properties of the peanut (Isleib et al., 2015), it does offer a significantly longer shelf life due to higher oxidative stability of the oleic acid and potential health benefits (Braddock, Sims, & O'Keffe, 1995; Davis, Dean, Faircloth, & Sanders, 2008; Derbyshire, 2014).

'Grade' is the term used to refer to the size of the kernel, and it is defined based on the dimensions of the sieve openings through which the kernels can and cannot pass through. Grades range from Extra Jumbo, Jumbo, Extra Large, Large, Medium, Small to Extra Small. As expected, whether or not a kernel will pass through a sieve is not only dependent on the size of the kernel, but also the shape (rounder vs elongated kernels). For this reason the United States Department of Agriculture (USDA) has defined grades separately for each market type (USDA, 1997c, 1997a, 1997b). The US definitions are commonly used by other countries since the USA is large exporter of peanuts, but to avoid confusion the term 'count' is also employed. 'Count' is simply defined the number of whole kernels in a 100 gram sample.

Finally, the term 'origin' is used to denote the location where the peanuts where grown (state/province, country), and 'crop year' to denote the year of harvest (Woodroof, 1983). The later can be somewhat misleading, as depending on the variety and the local climate conditions, two harvests per season are sometimes possible (e.g. Nicaragua).

1.2 Commercial significance and existing research focus areas

Peanuts are one of the five most important oil seeds in the world, and are grown in six continents (Fletcher & Shi, 2016), with large appeal in both developed and developing markets (Euromonitor, 2010a, 2010b; USDA, 2015a) both for their organoleptic properties, as well as their nutritional content. Peanuts are consumed both directly (as a snack or food ingredient) and indirectly (crushed for edible oil extraction) **Figure 2**.

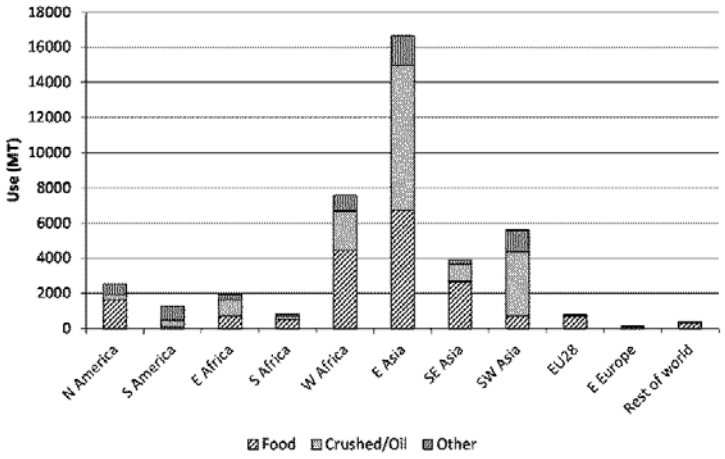

Figure 2: Annual peanut use around the world (2010-2013 average) (USDA, 2017). Other: exports, losses and non-reported uses.

Over the last several decades, an extensive amount of research has been done on the crop, but the main focus has been in developing new varieties with increased production yield (the most notable step change was a 52% yield improvement when Cultivars with Procumbent Growth HABIT (CPGH) were introduced in Argentina in 1975) (Haro, Baldessari, & Otegui, 2013; Htoon et al., 2014), or increased tolerance to environmental stress factors (droughts (de Sousa, de Azevedo, Fernandes, de Araújo Viana, & Silva, 2014; Junjittakarn, Girdthai, Jogloy, Vorasoot, & Patanothai, 2014), poor soil quality (Sarathi Patra & Chandra Sinha, 2014), extreme temperatures (Paulucci, Medeot, Dardanelli, & de Lema, 2011), and disease resistance (Hollis, 2014)). Furthermore, there has been a large amount of research done with regards to peanut oil extraction, aimed at identifying methods for improving extraction yield (Lihua Jiang, Di Hua, Zhang Wang, & Shiying Xu, 2010; Osuji, Brown, & South, 2010; Russo & Webber, 2012). More recently emphasis has also been placed on shelf life stability (Mozingo, O'Keefe, Sanders, & Hendrix, 2004; V. Nepote, Olmedo, Mestrallet, & Grosso, 2009; Shakerardekani, Karim, Ghazali, & Chin, 2013), resulting in the development of 'high oleic' varieties; peanuts that contain a larger ratio of oleic to linoleic acid (Davis et al., 2008; Derbyshire, 2014), making them more resistant to oxidative degradation (Braddock et al., 1995; de Godoy et al., 2014; Mozingo et al., 2004; Valeria Nepote, Mestrallet, & Grosso, 2006). Coincidentally, this novel fatty acid profile also offers nutritional advantages (Derbyshire, 2014; Moreira Alves et al., 2014) and the peanut industry has capitalized on the increased consumer desire for healthier oils that has developed over the last decade (Anonymous, 2007).

1.3 A brief overview of the growing cycles and processing flow

In order to understand peanut quality, a brief discussion of the growing cycle is required. The peanut plant forms perfect/hermaphroditic flowers, which self-pollinate (Stalker et al., 2016). Generally only one flower per day reaches anthesis, and as a result all flowers in a given plant will be pollinated on a different day. Several days after fertilization, the flowers develop stalk-like structures called 'pegs' which the plant directs towards the ground. The pegs penetrate the soil and after 60-80 days develop into pods containing 2-3 peanut kernels (Stalker et al., 2016). As a consequence of reaching anthesis on different days, each pod within a plant will reach maturity at a different time, and so at the time of harvest there will be a distribution of pod maturity levels. This has large implications on the kernel quality and organoleptic characteristics (Kim & Hung, 1991; Sanders, 1989; Sanders, Vercellotti, Blankenship, Crippen, & Civille, 1989; Sanders, Vercellotti, Crippen, & Civille, 1989; Williams, Ware, Lai, & Drexler, 1987).

The field to pack journey of peanuts includes several steps, namely harvest, drying, storage, shelling, grading, blanching and roasting (Cowart, Powell, Locke, Starling, & Takash, 2016), all of which can have an impact on the final product quality and characteristics. The operations are rarely done by the same entity, so several commercial transactions take place through the chain (Archer, 2016). An overview of the product transformations for ready to eat roasted peanuts can be found in **Figure 3**.

Figure 3: Visual overview of major peanut transformations in the supply chain.

The chain begins in the field where the entire plant is removed from the ground, exposing the pods to sunlight and thusly arresting the vegetative growth (Cowart et al., 2016). The pods have relatively high moisture content, and unless they are dried to ~10% w/w moisture, mould damage and off-flavours will ensue (Woodroof, 1983). Depending on environmental conditions, this drying (referred to as *'curing'*) can occur on the field under the sun over 2-3 days, or mechanically in specially designed kilns (generally at 35°C) (Cowart et al., 2016). Grading follows, where quality characteristics such as foreign bodies, damaged kernels and off-flavours are quantified (as defined by the local relevant regulatory body) and the market value is determined. The pods, at this point referred to as *'farmer stock',* can be stored in bulk silos (usually at the farm) for up to 12 months. Temperature

or humidity abuse during storage will have a detrimental effect on the organoleptic properties of the peanuts (Cowart et al., 2016).

The pods are consequently shelled, after the removal of foreign bodies by gravity tables, aspirators and magnets (Cowart et al., 2016). This is accomplished by crushing between rollers whose gap has been carefully set to crush the pod but not the kernels (Woodroof, 1983), and the shells are removed by aspirators and shaker tables. The resulting kernels are referred to as '*seeds*', because they are still viable and will germinate if planted.

The seeds are further *blanched*, a process by which the testa (skin) of the seed is removed. Inadvertently, some of the intrinsic enzymes will also be inactivated, but contrary to what the name implies, this is not the primary aim of this operation. There are several methods to loosen the skin, including dry heat and warm water soaking, generally followed by abrasion between rolling brushes (Woodroof, 1983). The unit operation method and conditions will have a significant impact on the finished product breakage (split cotyledons), moisture content, shelf life and organoleptic properties (texture, flavour and appearance), but most commonly peanuts are dry-blanched in 85 °C hot air for approximately 30 min (Sanders, Adelsberg, Hendrix, & McMichael, 1999; Schirack, Drake, Sanders, & Sandeep, 2006a, 2006b). The blanched peanuts are at this stage ready for roasting, and can be stored for several months in cool, dry and dark storage, or several years in frozen storage (Woodroof, 1983).

Table 1: Nutritional composition of major peanut types and overall average (raw, per 100 grams). Data from (USDA, 2015b).

Nutrient	Units	All Types	Valencia	Spanish	Virginia
Water	g	6.5	4.3	6.4	6.9
Energy	kJ	2374.0	2385.0	2386.0	2356.0
Protein	g	25.8	25.1	26.2	25.2
Total lipid (fat)	g	49.2	47.6	49.6	48.8
Ash	g	2.3	2.2	2.0	2.6
Carbohydrate, by difference	g	16.1	20.9	15.8	16.5
Fibre, total dietary	g	8.5	8.7	9.5	8.5
Minerals					
Calcium	mg	92.0	62.0	106.0	89.0
Iron	mg	4.6	2.1	3.9	2.6
Magnesium	mg	168.0	184.0	188.0	171.0
Phosphorus	mg	376.0	336.0	388.0	380.0
Potassium	mg	705.0	332.0	744.0	690.0
Sodium	mg	18.0	1.0	22.0	10.0
Zinc	mg	3.3	3.3	2.1	4.4
Copper	mg	1.1	1.2	0.9	1.1
Manganese	mg	1.9	2.0	2.6	1.7
Selenium	µg	7.2	7.3	7.2	7.1

A final roasting step is required to develop the flavour and ensure microbial safety, as peanuts are particularly prone to Salmonella since they grow underground (Cowart et al., 2016). A maceration step in various media (such as spices in water) varying from a few seconds to several hours can precede the roasting step if desired. Roasting can be done in oil (*'oil roasting'*) in batch or continuous fryers, in hot air (*'dry roasting')* in batch, continuous or drum ovens or even in heated salt or sand (Woodroof, 1983). In Europe, the most common processes are oil and dry roasting, and the majority of the nuts are consumed blanched, although skin-on peanuts are also consumed in the east (Prusak, Schlegel-Zawadzka, Boulay, & Rowe, 2014). The peanuts can be finally salted, packed, and distributed for retail.

1.4 Chemical composition

The average composition of peanuts can vary substantially between market types, variety, growing environment, storage condition, maturity level and processing conditions (Davis & Dean, 2016), but a 20 year average nutritional compositions can be found in **Table 1**. **Table 2** lists the average amino acid and fatty acid compositions, components known to be associated to flavour development during processing.

1.5 Gaps and opportunities in current literature

Given the differences in their chemical composition, it is not surprising that different peanut varieties also exhibit differences in their sensory profile, and several studies have made specific comparisons. Organoleptic differences between different grades of the same cultivar (Pattee, Isleib, Gorbet, & Geisbrecht, 2002), cultivars grown in the same region (Ng & Dunford, 2009), different genotypes (Baker et al., 2003) and different origins (Bett et al., 1994) when roasted by the same process to the same degree have all been previously reported. As part of the Uniform Peanut Performance Test (UPPT) the USDA, ARS, Market Quality and Handling Research Unit in Raleigh, NC, USA has been analysing and logging the sensory profiles of new peanut cultivars since 2001. Since the same expert panel, lexicon, roasting and testing protocol have been observed, comparisons of the sensory profile of cultivars can be accurately made (Isleib et al., 2015). **Figure 4** shows these data for 2008 (194 observations), and demonstrates the magnitude of differences observed. No quantitative comparison of the texture characteristics of different cultivars could be found in the literature, but there are several qualitative

comparisons indicate a large variety to variety difference (American Peanut Council, 2014; Woodroof, 1983).

Table 2: Average fatty acid and amino acid composition of major peanut types (raw, per 100 grams). Data from (USDA, 2015b).

Nutrient	Units	All Types	Valencia	Spanish	Virginia
Fatty acids					
Fatty acids, total saturated	g	6.3	7.3	7.1	6.4
14:0	g	0.0	0.0	0.0	0.0
16:0	g	5.2	5.4	5.7	5.0
18:0	g	1.1	1.2	1.3	1.3
Fatty acids, total monounsaturated	g	24.4	21.4	22.3	25.6
16:1 undifferentiated	g	0.0	0.0	0.0	0.1
18:1 undifferentiated	g	23.8	20.9	21.8	24.7
20:1	g	0.7	0.5	0.5	0.5
Fatty acids, total polyunsaturated	g	15.6	16.5	17.2	14.9
18:2 undifferentiated	g	15.6	16.5	17.2	14.7
18:3 undifferentiated	g	0.0	0.0	0.0	0.0
Fatty acids, total trans	g	0.0	0.0	0.0	0.0
Amino Acids					
Tryptophan	g	0.3	0.2	0.3	0.2
Threonine	g	0.9	0.9	0.9	0.9
Isoleucine	g	0.9	0.9	0.9	0.9
Leucine	g	1.7	1.6	1.7	1.6
Lysine	g	0.9	0.9	0.9	0.9
Methionine	g	0.3	0.3	0.3	0.3
Cystine	g	0.3	0.3	0.3	0.3
Phenylalanine	g	1.4	1.3	1.4	1.3
Tyrosine	g	1.0	1.0	1.1	1.0
Valine	g	1.1	1.1	1.1	1.1
Arginine	g	3.1	3.0	3.1	3.0
Histidine	g	0.7	0.6	0.7	0.6
Alanine	g	1.0	1.0	1.0	1.0
Aspartic acid	g	3.1	3.1	3.2	3.1
Glutamic acid	g	5.4	5.2	5.5	5.3
Glycine	g	1.6	1.5	1.6	1.5
Proline	g	1.1	1.1	1.2	1.1
Serine	g	1.3	1.2	1.3	1.2

It is therefore clear that different raw materials can deliver different sensory profiles if processed in the same manner. It is intuitive that a given raw material processed at different conditions (e.g. degree of roast, or fry versus bake) will demonstrate different organoleptic properties. However, little data can be found in the literature on the relative impact of material to process selection, and by extension, on whether altering the process conditions can overshadow differences in the raw material characteristics, as the majority of the work has mainly been descriptive in nature.

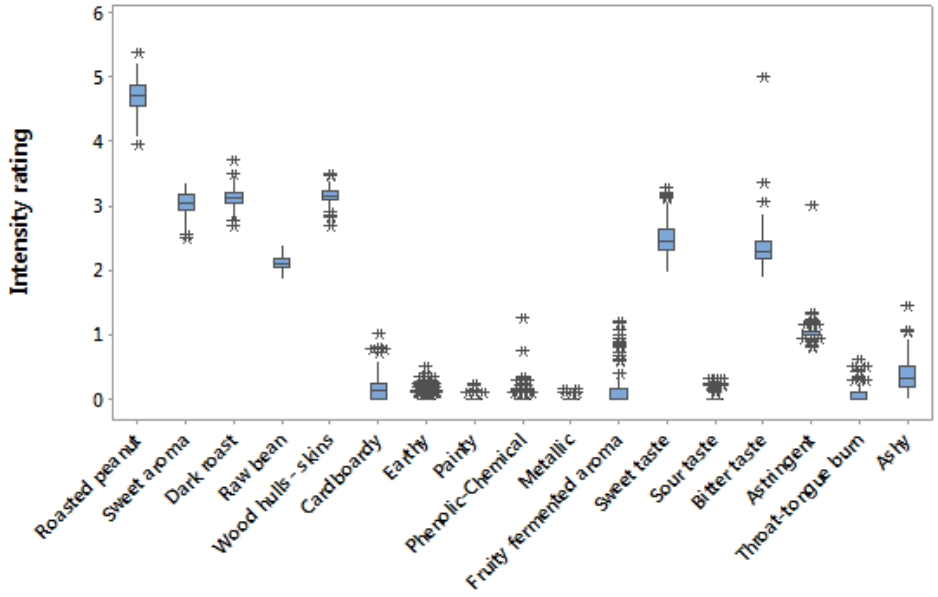

Figure 4: Variation observed in the sensory attribute intensities of different cultivars (data from 2008 UPPT). Horizontal line: median, box: 1st (Q1) to 3rd (Q3) quartile, upper whisker: Q3+1.5(Q3-Q1), lower whisker: Q1-1.5(Q3-Q1), *: outliers (defined as the observations outside the whisker range).

Past research has mostly focused on characterizing the flavour and/or texture of specific cultivars, growing locations and maturity levels (Bett et al., 1994; Isleib et al., 2015; Pattee, Beasley, & Singleton, 1965; Pattee et al., 2002; Sanders, Vercellotti, Crippen, et al., 1989; Walczyk et al., 2013; Young, Pattee, Schadel, & Sanders, 2004; Young, Sanders, Drake, Osborne, & Civille, 2005; Young & Schadel, 1993), as well as novel processing techniques such as microwave or infrared roasting (Davis et al., 2010; Kumar, Debnath, & Hebbar, 2009). In order to increase the statistical power of their analysis, most studies focused on a small number of materials or processes (commonly less than 3), resulting in high statistical power but narrow experimental space range. Conclusions drawn based on these studies are therefore statistically powerful, but limited in application to the specific cultivars or processes studied.

With regards to improving consumer liking, the vast majority of the literature has taken the ‘contrast and select’ approach. Numerous materials or processes have been characterized, and the best performer was recommended. This approach, although powerful, is not very practical: sourcing only a specific variety/origin/grade can result in increased purchasing costs (not open market, limited supply) and volatile supply (vulnerable to the geopolitical factors associated

with the supplier). In addition, it raises the question of what to do with the remainder of the crop. As mentioned above peanuts are also used for oil extraction and animal feed, but these applications often favour varieties specifically bread for that purpose (e.g. high oil content, low cell wall strength for easy oil extraction, as mentioned above). A restricted sourcing strategy therefore, may result in increased waste in the overall supply chain. Food waste has been identified as a major issue to be tackled by the food industry (FAO, 2011). Using processing technology to compensate for the shortcomings of raw materials in order to deliver a disable project appears to be an approach of great practical value, and yet it is very rarely encountered in the literature. This concept was central to the design and execution of this thesis.

Although peanut flavour and texture has been extensively studied since the 1960s, there still significant gaps in the literature. First and foremost, there is still no alignment on what constitutes peanut flavour. Even though significant progress has been made on identifying key aroma compounds (Chetschik, Granvogl, & Schieberle, 2010; Da Conceicao Neta, 2010), not all the responsible aroma compounds have been yet identified. This is likely due to the large differences occurring between cultivars and process settings (a case of a 'moving target' as key aroma compounds may vary across cultivars and processing methods), as well as potential synergistic and antagonist effects between aroma active compounds, which reconstitution studies struggle to identify (Chambers & Koppel, 2013). Examples, such as the case of the raspberry fruit where different subspecies contain different key aroma compounds (Aprea, Biasioli, & Flavia Gasperi, 2015), suggest that a wide range of cultivars should be analysed. However, most peanut flavour studies focus on a handful of cultivars of the Runner type. Furthermore, fingerprint studies on peanut flavour have not yet been published, and as such there is little published information on any potential antagonistic or flavour masking effects, even though experience shows that defects and other off-flavours do reduce the perceived peanut flavour intensity. Perhaps more importantly, there is an even greater lack of understanding on sensory attributes other than 'peanut aroma'. Very limited literature could be identified on other important attributes of snack peanuts, such 'roast aromas'. Similarly, although flavour-colour correlations are well established in the peanut industry (Pattee, Sanders, Isleib, & Giesbrecht, 2001) there is little published evidence on whether the relationship holds across different raw materials and process conditions.

Likewise for texture, past research has focused on comparisons between materials or the development of texture during roasting. Certain processes such as dry roasting and pre-boiling in water have been well studied (Davis et al., 2010; Shi et al., 2017), but done across a very limited number of raw materials and without comparisons to other processes. As a result, it is not known whether material-process interactions exist, if for example certain varieties or market types are better suited for blistering or not.

Due to the lack of wide research with a wide scope, there is little information that can be systematically leveraged to modify a specific sensory attribute. Isolated optimization studies and roast level range tests have been published (McDaniel, Price, Sanders, & Davis, 2011) but there is no clear guidance available on what process is required in order to decrease or increase the intensity of the 'breakdown' sensory attribute, for example. It is clear therefore, that although a lot of high quality and focused research has been published on the subject, there is a need for a large scale texture and flavour study that evaluates several raw materials and process technologies simultaneously. Furthermore, the availability of computing power and specialist software today particularly lends itself to mining such large datasets, something that was not possible a few decades ago.

Technological developments can also aid in the study of microstructure. Even though it has been claimed that alveolation in peanuts due to steam generation during roasting is responsible for the crunchy texture (Dean, Davis, Hendrix, Debruce, & Sanders, 2014), no quantitative evaluation could be performed prior to the development of powerful image analysis software and CT X-ray imaging.

To this day, limited research has been done in understanding the consumer drivers for preference, and even less on how these drivers relate to the different process technologies (Lee & Resurreccion, 2004; McDaniel, White, Dean, Sanders, & Davis, 2012) or raw material origins (Onemli, 2012; Walczyk et al., 2013; Young et al., 2005). Product differentiation in the market revolves around subjective claims of 'freshness' and 'minimally processed' in the developed markets, and local availability in developing markets. Since manufacturers have turned their focus on 'soft' claims (i.e. marketing) and distribution, product differentiation has been neglected and so there is very little in the way of new products other than incorporation of nuts as inclusions in other categories. The result has been the commoditization of the peanut category, as well as a consumer shift to 'more exciting' snacks, often of lower nutritional qualities such as snacks made of peanuts or peanut containing snacks (Euromonitor, 1989).

Furthermore, no research has been published on the peanut preference of the European consumer, even though peanut consumption is significant ins Europe. As a result, it is not clear how one could improve consumer liking. Similarly, even though it makes intuitive sense that perceived freshness is an important attribute for the consumer, no published research has attempted to identify if specific product attributes can increase it, as in the case of adding 'bits' in orange juice for instance (Zhang, Lusk, Mirosa, & Oey, 2016).

1.6 Thesis aims and potential applications

The present thesis therefore aims at three objectives:

1) First and foremost, to identify the drivers of liking and perceived freshness of snack peanuts with European consumers. To achieve this, samples exhibiting a large organoleptic variance are necessary.
2) The second objective therefore, is to understand how different processes and materials can be utilized in order to deliver significantly different sensory profiles.
3) Finally, the third objective is to identify correlations between sensory and instrumental attributes, in order to better describe said samples (while not restricted to 'peanut aroma' attribute only).

The practical applications of these three objectives are numerous and varied: Firstly, by understanding what the consumer wants, processors can better meet those needs. This can be in the form of a better liked peanut, or by enabling the offer of differentiated products in cases where consumer segmentation is observed (i.e. different population groups prefer different product designs). Besides the obvious financial benefit to the processor, consumer satisfaction can also be expected to increase. Peanuts are nutritionally advantaged over several alternative snacks due to their low sugar content and fatty acid profile, but could also aid in weight loss due to their strong satiating effect and thermogenesis in certain population groups (Moreira Alves et al., 2014). As a result any activity that would make it more likely for consumers to select peanuts over other snacks could benefit the population health.

Achieving the second objective and unlocking understanding of the process-material interaction can also offer significant advantages. In the first instance it complements the first objective: knowing what consumers like has no value unless one knows how to deliver it through process and material selection. Secondly, by understanding how processing can be leveraged to adjust or compensate for shortcomings of raw materials, allows a reduction in both cost (less restrictive procurement specifications) and food waste (substandard raw materials can potentially be salvaged). Finally, it gives the processor the toolbox needed to be able to control the product design. This allows the creation of new products (e.g. 'extra crunchy'), or the improvement of current ones. An example of the later would be restoring crunchiness of baked peanuts through a maceration pre-treatment, allowing the production of a baked peanut with the organoleptic properties of a fried one, but without the negative health connotations.

The third objective offers more tactical advantages in a manufacturing environment, by identifying 'shortcuts' in quality control. If, for example, sensory

texture can be modelled by instrumental data, maintaining and running costly expert sensory panels for product evaluation will be less needed. Similarly, identifying correlations between quality characteristics, such as colour and flavour offers additional advantages: a colourimeter is not only significantly less costly and easier to operate than a GC-MS or expert taste panel, but it is also much faster. This creates opportunities in automation, as colour reading can be practically done in real time, and could therefore be used as an input in a control loop system.

1.7 Thesis outline

This thesis can be logically divided into four focus areas: i) flavour and colour study, ii) texture study, iii) understanding consumer preference, and iv) general discussion.

i. Chapters 2 and 3 are focused on the flavour aspect of peanuts. Chapter 2 focuses on the relative impact that raw material and process selection can have on the flavour, colour and fatty acid profile of the finished product. The impact of kernel maturity and market type is further investigated using targeted contrasts. With regards to process, the impact of maceration, baking and the topical application of various oils on flavour and colour attributes are discussed. Finally, the correlation between colour and flavour in roasted peanuts is also investigated. Chapter 3 is focused deeper on the correlation between headspace volatiles and sensory flavour attributes, investigating both linear and logarithmic relationships. In addition, fingerprints with several positive and negative correlations are identified for four important flavour attributes (‘roasted peanut aroma’, ‘dark roast aroma’, ‘raw beany aroma’, ‘sweet aroma’).
ii. Chapter 4 is solely focused on texture, and it investigates how different raw materials and processes lead to different texture characteristics, and how this is related to changes in the microstructure of the samples. Particular focus is placed on the impact of maceration as a pre-processing step, while a short discussion can also be found on the modelling of sensory by instrumental textural attributes. Finally, quantitative data on alveolation are presented, which suggest that the currently accepted mechanism of texture development as a consequence of microstructure disruption due to steam generation is not sufficient to fully explain textural differences, and that a secondary mechanism likely also exists.
iii. Chapter 5 introduces consumer data to the research, and an analysis of the drivers of liking for snack peanuts by consumers in three different European countries is given. In addition to liking, a discussion on the

consumer perception of freshness is also given and modelled back to headspace volatiles concentration data.

iv. Finally, Chapter 6 summarizes and integrates the flavour, colour, texture and consumer analyses, and discusses possible applications of this research. The validity of some of the models developed, methodological limitations and proposed future research are also briefly discussed.

1.8 References

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2 Effect of raw material and processing technology on flavour, colour and fatty acid composition of peanuts.

Adapted from:

Lykomitros, D., Fogliano, V., & Capuano, E. (2016). Flavour of roasted peanuts (*Arachis Hypogaea*) - Part I: Effect of raw material and processing technology on flavour, colour and fatty acid composition of peanuts. Food Research International, 89, 860–869.

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Abstract

Flavour and colour of roasted peanuts have a strong impact on consumer acceptability. They can be influenced by raw material and processing technology. Raw peanuts of various market types, origins and grades were processed by different technologies to produce 134 unique samples, which were profiled by a sensory panel and analysed for colour and fatty acid composition. Principal Component Analysis, Canonical Variate Analysis and General Linear Model regression were used to identify differences in flavour, colour and fatty acid profiles, and to relate them to raw materials or process conditions. Data showed that raw material selection is key for flavour, but processing is also significant. Specifically, maceration significantly increased 'roasted peanut' and 'dark roast' aromas, reducing 'sweet', 'raw bean' aromas, and sweetness. It also influenced colour and the fatty acid profile. Baking reduced 'roasted peanut' and 'dark roast' and increased 'raw bean' aromas compared to frying, and impacted colour development.

Keywords: Peanut, fatty acid, colour, flavour, processing, sensory

Highlights:

- Processing can have a large impact on the flavour characteristics of roasted peanuts.
- The effect can sometimes overshadow differences due to raw materials.
- Maceration in aqueous media can increase roasted and reduce raw aromas.
- Virginia type peanuts develop roasted aromas to a greater extent than Runner type.
- Processing affects the fatty acid profile regardless of raw material.

2.1 Introduction

Peanut is an important world crop for both developing and developed markets (USDA, 2015, 2017) both for its organoleptic properties, and its nutritional content. Peanuts can be consumed in several forms, however roasted whole peanuts enjoy a large and increasing market share (USDA, 2015). Genetic makeup and environmental and storage conditions are factors known to affect peanut flavour development, (Neta, Sanders, & Drake, 2010) however little information is available on the effect of industrial processing. The available studies about peanut flavour can be mostly divided into two categories: Those which attempted to identify the volatile compounds responsible for certain flavour attributes (Neta et al., 2010; Schirack, Drake, Sanders, & Sandeep, 2006a) and those which described and compared the flavour profile of peanuts from different origins, market types, varieties and grades(Bett et al., 1994; Isleib et al., 2015; Pattee, Isleib, Gorbet, & Geisbrecht, 2002; Walczyk et al., 2013; Young, Sanders, Drake, Osborne, & Civille, 2005). The importance of raw materials was recognized early on, and has given rise to the term 'market type', coined by growers to categorize peanuts with similar characteristics not necessarily following botanical taxonomy (Woodroof, 1983). Although the importance of processing on flavour development is generally recognized, little research has been published in the field. Some studies have focused on flavour development during processing, comparing treated versus raw peanuts (Chetschik, Granvogl, & Schieberle, 2008, 2010; Liu et al., 2011; Muego-Gnanasekharan & Resurreccion, 1993). One study comparing different processes previously published focused on the nutritional content and on a limited number of raw materials (Chukwumah, Walker, Vogler, & Verghese, 2007). In addition to their impact on nutrition, peanut processing technologies (roasting, microwaving and combination) have also been compared with regards primarily to food safety, specifically Salmonella inactivation (Smith, Perry, Marshall, Yousef, & Barringer, 2014).

The aim of the present research was to study the impact a wide range of process technologies can have on peanut flavour formation, and more specifically, whether this impact is significant enough to compensate for the differences driven by the source material. In other words we aim to verify whether selection of an appropriate process technology and processing conditions is able to yield final products having similar sensory features, thus minimizing the differences driven by the raw material. This would allow peanut processors to utilize a wider range of raw materials (e.g. more varieties or origins) whilst ensuring a consistent finished product. This can not only have large cost reduction implications (less selective procurement in a highly commoditized market), but also reduce food waste (fewer materials rejected), a major concern according to the Food and Agriculture Organization of the United Nations (FAO, 2011).

2.2 Materials and Methods

2.2.1 Peanut samples

Peanuts of three market types (Valencia, Virginia and Runner) and different grades (sizes) were sourced from different origins spanning from USA to China and Australia (Canon Garth Ltd, London, UK). A cross-section of common varieties, both with and without the high oleic trait was selected, while the majority of the groundnuts were from the 2009 crop. **Table 3** provides an overview of the raw materials used in the study.

Table 3. Overview of the raw materials used in the study and their basic characteristics.

code	origin	type	variety	grade	count per 100 grams	high oleic	crop year	incoming moisture content	incoming split kernels
A	USA – Texas	Runner	Flavorunner 458	medium	141/177	yes	2008	7%	1%
B	USA-Texas	Runner	Flavorunner 458	jumbo	134/148	yes	2008	7%	3%
C	USA – Georgia	Runner	Georgia Green	medium	173	no	2008	6%	2%
D	USA – Georgia	Runner	Georgia Green	jumbo	137	no	2008	6%	2%
E	Argentina	Runner	Granoleic	jumbo	134/148	yes	2009	7%	3%
F	South Africa	Valencia	CN Natals	small	177	no	2008	5%	2%
G	Argentina	Runner	Tegua	medium	141/177	no	2008	5%	4%
H	China	Runner	Hsuji	medium	141/177	no	2007	5%	5%
I	USA – Virginia	Virginia	mixed	extra large	106	no	2008	7%	3%
J	USA – Virginia	Virginia	mixed	medium	148	no	2008	7%	1%
K	Australia	Virginia	Middleton	extra large	71/92	yes	2008	6%	6%
L	Australia	Virginia	Middleton	medium	120/141	yes	2008	6%	4%

The peanuts were procured shelled and raw, with typical incoming moisture content ranging from 5 to 7% w/w, sorted for defective kernels and foreign material and stored at a commercial storage facility at ambient temperature. Dry blanching was used to remove the testa of the seeds, as it is one of the most commonly used industrial blanching methods (Schirack, Drake, Sanders, & Sandeep, 2006b). All seeds were blanched within a window of 3 weeks to minimize variations due to ageing, at approximately 85 °C for 30 minutes, (Sanders, Adelsberg, Hendrix, & McMichael, 1999) followed by mechanical removal of the testa (Steinweg-Handelsveem BV, Oosterhout, NL). The blanched kernels were kept at -15 °C until further processed, generally within less than 6 months. Frozen storage has been shown to not affect the organoleptic properties of the nuts, and as such is commonly practiced (Woodroof, 1983).

The blanched seeds were consequently processed in a variety of methods in a fractional factorial design, as shown in **Table 4**. Several processing methods were investigated, but small deviations were allowed in order to ensure all samples had a final moisture content between 2.0% and 2.7% w/w. The same procedure was followed for all samples that were macerated, changing the medium, temperature and time as described in **Table 4**: 12 kilograms of the blanched peanuts were placed in a stainless container, and 20 kilograms of the appropriate maceration medium was added. An immersion heater with stirrer (Scheffers Apparatenbouw, Zaandam, NL) was used for temperature control, and after the designated time the peanuts were promptly removed from the maceration medium, and the corresponding thermal process was applied. All baking was done using an Aeroglide C1 12-16 REX continuous oven (Cary, NC), at a 1.5 cm bed depth. The oil roasted samples were fried in a batch fryer in 5kg batches (30L fry oil capacity, De Kuiper (De Kwakel-Uithoorn,NL)). An initial temperature dip of the frying oil of up to 7 degrees Centigrade was observed upon addition of the peanuts but the oil temperature recovered in 4 minutes or less. After all heat treatments the samples were spread in open mesh trays and cooled under forced air to room temperature. Topical application of the oils for the appropriate samples was done in coating drum (S-1050, Walter Brucks, Bruggen, DE). The samples were finally sealed in metalized bags and flushed with nitrogen to an in bag residual oxygen content of less than 2% v/v.

In order to get a better understanding of dry roasting, one of the most common process technologies employed in the industry, more than one baking condition was included in the design. The total heat treatment was conceptually divided into two zones of equal time, and the temperature of each zone was set either high (155°C) or low (135°C). The total baking time was adjusted so that the final moisture content was approximately 2%, and was evenly split between the two zones. As a result, it was possible to employ a (2 level – 2 factor) Placket-Burman design for the dry roasted samples, where roasting to the same degree at higher or lower temperatures could be resolved (processes D-G in **Table 4**). Given the commercial importance of dry and oil (frying) roasting, all raw materials were processed by these methods.

All samples were salted to 1% NaCl w/w (Cargill, MO), and the salt was applied as 5.5M aqueous solution prior to the thermal processing step, or topically after the application of sunflower (High Oleic Sunflower Oil, Cargill, MO) or aromatic roasted peanut oil at 2% w/w (Aromatic Roasted Peanut Oil 100E, NutrIn, Washington DC). Aromatic Roasted Peanut Oils (ARPO) are unrefined mechanically expelled peanut oils, and as such exhibit a strong characteristic roasted peanut odour (Liu et al., 2011). Generic cider vinegar procured from a local supermarket (Albert Heijn, Zaandam, NL), powdered dextrose (Brouwmarkt, Almere, NL), and $CaOH_2$ (Merck, Kenilworth, NJ) were used for the acidified, sweetened and alkalized

samples respectively. All the reagents used in this research were obtained from Sigma-Aldrich (St Louis, MO), unless otherwise specified.

Table 4. Overview of the roasting processes applied.

code	process	key process parameters	applied to materials [a]
A	aqueous acid maceration, dry roasting	acidified to pH 4 with acetic acid, 30 min at 20 °C, roasted at 145 °C for 18 min	A,C,E[b],F[b],H,J[b],L
B	cold (long) aqueous maceration, dry roasting	potable water, 90 min at 20 °C, roasted at 145 °C for 18 min	A,C,E[b],F[b],H,J[b],L
C	aqueous dextrose maceration, dry roasting	2.5% w/w dextrose solution, 30 min at 20 °C, roasted at 135 °C for 26 min	A,C,E[b],F[b],H,J[b],L
D	dry roasting (low temperature long time)	continuous convection oven 135 °C 16 min	A,B,C,D,E[b],F[b],G[b],H,I,J[b],K,L
E	dry roasting (high temperature short time)	continuous convection oven 155 °C 9 min	A,B,C,D,E[b],F[b],G[b],H,I,J[b],K,L
F	two temperature zone dry roasting (high-low)	continuous convection oven 155 °C 5 min /135 °C 5 min	A,B,C,D,E,F,G,H,I,J,K,L
G	two temperature zone dry roasting (low-high)	continuous convection oven 135 °C 5 min /155 °C 5 min	A,B,C,D,E,F,G,H,I,J,K,L
H	oil roasting (frying)	fried in high oleic sunflower seed oil at 150 °C for 4,5 min	A,B,C,D,E,F,G,H,I,J,K,L
J	aqueous alkaline maceration, dry roasting	alkalized to pH 10 with $CaOH_2$, 30 min at 20 °C, roasted at 145 °C for 18 min	E,F,J
K	aqueous dextrose maceration, oil roasting	2.5% w/w dextrose solution, 30 min at 20 °C, fried in high oleic sunflower seed oil at 150 °C for 7 min	E,F,J
M	cold (short) aqueous maceration, dry roasting	potable water, 30 min at 20 °C, roasted at 145 °C for 18 min	A,C,E[b],F[b],H,J[b],L
X	topical aromatic roasted peanut oil application	2% w/w aromatic roasted peanut oil spray after any process (A-M).	
Z	topical sunflower oil application	2% w/w high oleic sunflower seed spray after any process (A-M).	

[a] Codes in **Table 3**. [b] Additional samples prepared with post treatments X and Z.

2.2.2 **Sensory Analysis**

Five hundred grams of each of the samples was ground to a paste with a food processor (Cuisinart DLC- 7 with cutting blade DLC-001, Cuisinart, E Windsor, NJ) by processing for 3 minutes, while stopping to scrape the sides at 1.5, 2.5 and 3 minutes. Paste was preferred over whole nuts to ensure sample uniformity and address individual kernel maturity difference, as will be discussed below.. Descriptive Sensory Analysis (DSA) was performed in duplicate (every sample was seen by each panellist twice) at room temperature by a highly trained DSA panel at the USDA, ARS, Market Quality and Handling Research Unit (Raleigh, North Carolina, USA). Panellists (n=10, 3:7 male:female, mean age: 33, age range: 20-55, >250hours experience on peanut DSA panels each) utilized the Spectrum ™ Method (15 point scale) to evaluate all samples and used water and non-salted crackers as palate cleansers (Meilgaard, Civille, & Carr, 1999). Details on the methods, lexicon

and attribute definitions have been previously published, (Johnsen, Civille, Vercellotti, Sanders, & Dus, 1988; Sanders, 1989; Schirack et al., 2006a) but the lexicon can be found in **Table 5**.

Table 5. Flavour sensory attributes as obtained from the expert panel. Lexicon and method defined in (Johnsen et al., 1988; Sanders, Vercellotti, Crippen, & Civille, 1989; Schirack et al., 2006a).

attribute	description
roasted peanut	the aroma associated with medium roast peanuts (3-4 on USDA colour chips), and having fragrant character such as methyl pyrazine
sweet aroma	the aromas associated with sweet material such as caramel, vanilla, molasses, fruit (specify type)
dark roast	the aroma associated with dark roasted peanuts (4+ on USDA colour chips) and having very browned or toasted character
raw beany	the aroma associated with light roast peanuts (1-2 on USDA colour chips) and having legume like character (specify beans or pea if possible)
woody, hulls, skins	the aromas associated with base peanut character (absence of fragrant top notes) and related to dry wood, peanut hulls and skins.
cardboard	the aroma associated with somewhat oxidized fats and oils and reminiscent of cardboard
earthy	the aroma associated with wet dirt and mulch.
painty	the aroma associated with linseed oil, oil based paint.
phenolic/chemical	aroma associated with chemical/plastic/band aid
fruit fermented	the aroma associated with over ripe or sweet fermenting fruit
ashy	the aroma associated with ash-tray without tobacco notes
total off note	intensity rating of total off notes
sweet	the taste on the tongue associated with sugars
sour	the taste on the tongue associated with acids.
bitter	the taste on the tongue associated with bitter agents such as caffeine or quinine.
salty	the taste on the tongue associated with sodium ions.
tongue, throat burn	the chemical feeling factor on the tongue and throat associated with burning (benzoate).
metallic	the chemical feeling factor on the tongue described as flat, metallic and associated with iron and copper.
astringent	the chemical feeling factor on the tongue, described as puckering/dry and associated with tannins or alum.

2.2.3 **Fatty acid profile analysis**

The fatty acid profile of each sample was analysed with an Autosystem XL gas chromatographer fitted with a flame ionization detector (Perkin Elmer, Waltham, MA) and a BPX70, 50 meter by 0.32 mm internal diameter column (SGE Ltd, Milton Keynes, UK). The samples were pre-treated in accordance to method BS EN ISO 5509:2001 to produce the fatty acid methyl esters, and the chromatographic analysis was performed in accordance to method ISO 15304:2002 (BSI, 2001; ISO, 2002).

2.2.4 Colour and moisture analysis

For the colour measurements a 250 gram sample was equilibrated at 20 °C for 24 hours, and consequently placed in a 95mm diameter petri dish, taking care to ensure no portion of the dish is visible through the kernels. The petri dish was viewed from above by the colorimeter and the L*, a* and b* parameters of the CIELAB system were obtained (automatic averaging of three measurements), using a Hunter Lab CR400 colorimeter (Reston, VA). The procedure was repeated in triplicate, with the peanut sample being redistributed in the petri dish between measurements to ensure different kernels were viewed by the instrument for each measurement. A 100 gram sample of each material was ground in a mini food processor (Kenwood, Havant, UK), and three grams of the ground samples was analysed for moisture content with a Leco TGA701 thermogravimetric analyser (St. Joseph, MI).

2.2.5 Statistical analysis

Principal Component Analysis (PCA, Pearson's method) was run on the sensory flavour attributes using XLSTAT 2015.5 (Addinsoft, Paris, FR on MS Excel 2010, Microsoft, Redmond, WA). As colour was quantified instrumentally and not by the sensory panel, the colour attributes were not included in the principal component analysis, but were only added as supplementary variables and superimposed on the biplots for completion. A Principal Component Analysis (PCA, Pearson's method, XLSTAT) was also run on the fatty acid profiles. The XLSTAT procedure for Principal Component Analysis automatically mean-centres and auto scales the input data.

Two-way (processes v raw materials) ANOVA was run on SAS (proc GLM, SAS Institute, Cary, NC), to compare two market types (Virginia and Runner) and grades (size) (medium and jumbo). For the market type comparison, the medium grade Virginia samples were pooled (materials J and L,**Table 3**) and contrasted to the pooled medium grade Runner samples (materials C, G and H). Similarly, for the grade comparison, jumbo Runner samples were pooled (materials B and D) and contrasted to the pooled medium grade Runner samples (materials A and C). The null hypothesis tested was that the difference between the pool sample means was zero.

To get an overview of the impact of process and raw materials on the flavour sensory attributes, two Canonical Variates Analyses (CVA) were also run on XLSTAT using the flavour attributes as the X matrix (forward model selection,

threshold to enter model α=0.2, equal within class covariance), one with raw material and one with process as the classification variable.

Finally, to provide insight into the impact of process on flavour sensory attributes, general linear model regressions were run in XLSTAT for key attributes (roasted peanut, sweet, dark roast and raw bean aromas, sweet taste and the L*, a* and b* CIELAB colour parameters). The analysis was limited to these attributes as they were less likely to be contributed by raw material defects, and had a large enough range in magnitude to allow the regressions to be run. The models included the following categorical independent variables with the levels given in parentheses: maceration (yes, no), baking (Baked, Fried) and topical application (none, high oleic sunflower seed oil, aromatic roasted peanut oil), and the general form can be seen in **Equation 1**.

$$sensory\ attribute_i = a_i \times (no\ maceration) + b_i \times (fried) + c_i \times (topical\ ARPO) + d_i \times (no\ topical) + e_i$$

Equation 1

The analysis was repeated a second time on a reduced sample set in order to get a better understanding of the most common process: baking. In this case, only the (2 level – 2 factor) Placket-Burman roasted samples (processes D-H, **Table 4**) where selected, without any topical oil application, and the categorical dependent variables included zone one oven profile (High, Low) and zone 2 oven profile (High, Low) (**Equation 2**). Tukey's HSD test (α=0.05) was used for comparing the means.

$$sensory\ attribute_i = a_i \times (high\ zone\ 1\ temperature) + b_i \times (high\ zone\ 2\ temperature) + c_i$$

Equation 2

2.3 Results and discussion

2.3.1 Sensory profiles of processed peanuts

The PCA model developed on the peanut sensory data is shown in **Figure 5**. Two components are sufficient to graphically represent the data (cumulative variance explained 73.5%). The figure shows that sensory profiles generally differ in two dimensions: sweet to bitter (sweet taste and aroma in top left quadrant, bitter taste and other off-flavours in bottom right quadrant) and effect of roast (roasted peanut and dark roast aroma in bottom left, raw bean aroma in top right quadrant). The CIELAB colour parameters are also mapped on the degree of roast continuum, with a* and b* correlated directly and L* inversely to the roasted

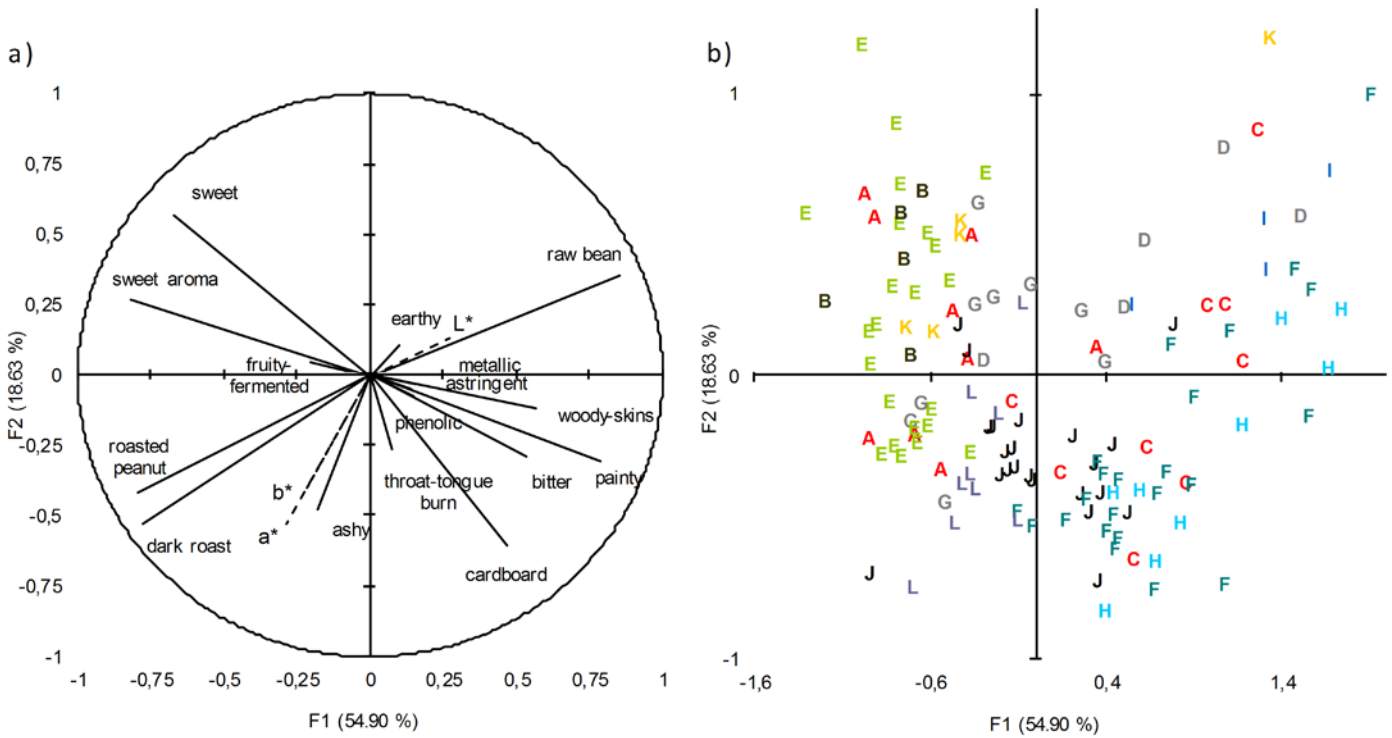

Figure 5. Left (a). Factor loadings for PCA of sensory profiles. Dotted vectors denote supplementary data not used in the calculation of the PCA space (CIELAB parameters). Right (b). PCA scores of sensory profiles of all samples coded for raw material (**Table 3**). Two principal components resolve 73.5% of the data variance.

aromas. The same figure also shows that although clusters by raw material are present, there is a fair amount of overlap. This suggests that although raw material does play a role in the final sensory profile of the product, the process type and conditions could in certain cases overshadow that effect.

2.3.2 Effect of raw material on flavour profile

Sensory data of the Flavorunner 458 and Georgia Green varieties were pooled, and the attributes for the medium grade were contrasted versus the attributes of the jumbo grade. The results are reported in **Table 6**. In addition, the contrast between medium grade Runner and Virginia market types is also included in the same table.

Table 6. Summary of ANOVA contrasts of sensory attributes of jumbo vs medium grades ([B+D]-[A+C] in **Table 3**) and Virginia vs Runner market types ([L+J]-[C+G+H] **Table 3**). Positive contrasts indicate higher scores for jumbo and Virginia in the appropriate columns.

sensory attribute	contrast estimate jumbo v medium	contrast estimate Virginia v Runner
roasted peanut	-0.099	0.310 [a]
sweet aroma	0.010	0.102 [a]
dark roast	0.037	0.285 [a]
raw beany	0.026	-0.222 [a]
woody, hulls, skins	0.082 [a]	-0.029
cardboard	0.267 [a]	-0.178 [a]
earthy	-0.018	-0.015 [b]
painty	0.130	-0.317 [a]
phenolic/chemical	-0.003	0.008
metallic	0.004	0.002
fruit fermented	0.002	-0.005
sweet	-0.019	0.053
sour	0.007	-0.006
bitter	0.075 [b]	0.076 [a]
astringent	-0.007	0.020 [b]
tongue, throat burn	0.055 [b]	0.002
ashy	0.029	0.089 [a]

[a] significant at P<0.05
[b] significant at P<0.15

Several studies have reported differences in the chemical composition of kernels of the same variety, origin and crop driven by a kernel size difference (Sanders, 1989; Sanders et al., 1989; Williams, Ware, Lai, & Drexler, 1987). This seemingly surprising observation has been explained on the basis of maturity: smaller kernels are often a mix of fully mature small kernels and immature large-to-be kernels. In contrast, large kernels can only by fully mature (Sanders et al., 1989).

The compositional differences reported in the scientific literature are indeed consistent with compositional changes observed during plant development, i.e. the conversion of starches to sugars and production of nitrogen containing compounds. Studies on sensorial characteristics between grades have been published, (Ng & Dunford, 2009; Pattee et al., 2002; Pattee, Yokoyama, Collins, & Giesbrecht, 1990) but most have focused on the peanuts from the same plot and processed in the same way in order to increase the power of the comparison. Furthermore, mostly large differences in grades (e.g. small vs jumbo) have been investigated.

From **Table 6** emerges that even with pooled varieties and origins, there are still significant differences in sensory attributes between the jumbo and medium grades and that those differences are large enough to still be observable across different process conditions. This observation has a large practical significance, as commercial peanut roasters (especially in Europe) often have a flexible sourcing strategy in order to minimize procurement costs, and different varieties and grades may be used at different times of the year, or even in the same package (Prusak, Schlegel-Zawadzka, Boulay, & Rowe, 2014).

The significantly higher woody and cardboard notes, as well as the significant higher bitter and throat burn notes could be attributed to the relative more immature, medium grade kernels (Sanders, 1989; Sanders et al., 1989). Pattee et al further observed that immature (generally smaller) kernels are also more susceptible to developing off-flavours if mishandled (Pattee et al., 1990). This implies that the higher woody and cardboard notes detected here on the medium grade may not be intrinsic to the material, but generated disproportionately faster during processing, especially during maceration which involves elevated water activity and temperature that can lead to faster lipid degradation (Civille & Dus, 1992; St Angelo, 1996).

In the second contrast (**Table 6**), where Virginia and Runner market types are compared, Virginia peanuts scored consistently higher on attributes associated with roasting (roasted, dark roast aromas) and sweetness (as previously observed (Chetschik et al., 2010)) and lower on attributes associated with insufficient roasting and/or defects (raw beany, painty, earthy and cardboard aromas). Virginias have also significantly higher scores for bitter taste and ashy aroma, attributes that are associated with scorching. Since all samples were roasted to similar final moisture contents, this observation suggests that Runner types are slower to develop a roasted flavour, and should therefore be roasted in a more aggressive time-temperature profile, in order to deliver roasted flavours similar to Virginia type. This could be an indication that Runner peanuts contain a lower concentration of precursors for Maillard or thermal degradation reactions attributed to the production of roasted peanut flavour (Branch, Worthington, Chinnan, Heaton, & Nakayama, 1988).

A contrast between varieties with and without the high oleic trait was not possible due to collinearity with origin. However, flavour attributes of high oleic

peanuts have been reported to be very similar to conventional cultivars (Isleib et al., 2015; Isleib, Pattee, Sanders, Hendrix, & Dean, 2006).

Figure 6 shows the Canonical Variate Analysis (CVA) scores plot and group clusters for the raw material classification. CVA is a type of discriminant analysis often used for classification (Huberty & Olejnik, 2006). The approach is similar to PCA, but instead of the principal components being selected for maximizing the explained variance, the factors are selected so that they maximize the difference between the class means. The resulting maps are a convenient way of visualizing the importance of different variables on classification. In other words, one can see which traits (attributes) are important for characterizing the classes, i.e. raw materials in **Figure 6**.

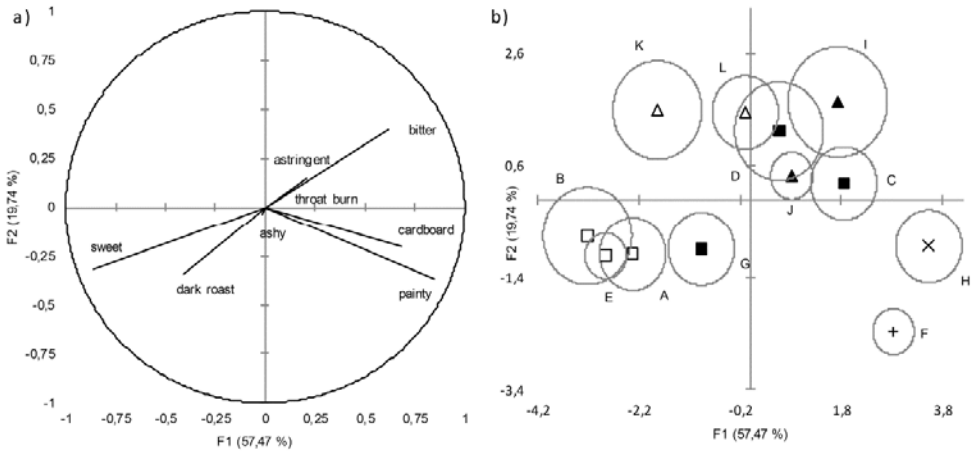

Figure 6. Left (a). Canonical Variate Analysis space of flavour sensory attributes for raw material classification. Only attributes that satisfied the α=0.2 variable selection criterion are shown. Right (b). Analysis of sensory attributes with raw material used as a classifier, with 95% confidence ellipses. Empty shapes: high oleic trait, filled shapes: low oleic trait, squares: Runner, triangles: Virginia, +: Valencia, x: Chinese Runner. Codes for individual materials can be found in **Table 3**. Two components resolve 77.2% of the data variance.

It can be seen that the space resolves the samples well on the sweet (bottom left) to bitter (top right) continuum. The dominant role of sweetness in distinguishing between raw materials has also be noted by others, and shown to be caused by variety and growing conditions (Pattee, Beasley, & Singleton, 1965). Pattee et al found sweetness to be mainly caused by simple sugars (inositol, glucose, fructose, sucrose, raffinose, stachyose), as well as other unidentified compounds (Pattee, Isleib, Giesbrecht, & McFeeters, 2000). Defects (cardboard and painty) are mapped in the bottom right quadrant. The CVA map (**Figure 6b**) shows a clustering of the market types and high oleic traits. The presence of clustering of

high oleic and non-high oleic raw materials is somewhat surprising, as it has been widely reported in the literature that the flavour differences between the two are not significant (Isleib et al., 2015, 2006). The high and low oleic clusters seem to be best separated on the sweet to bitter continuum, with the high oleic peanuts being generally sweeter. Given that sweetness is highly dependent on genetics (Pattee et al., 1965) it is possible that the observed contradiction with the literature is an artefact of the selected varieties. Valencia is isolated to the bottom right, the high oleic Runners in bottom left and the Virginias in top semicircle. Non high-oleic Runners are dispersed, but the Chinese are isolated in the bottom right quadrant. This could be due to the Hsuji variety naturally exhibiting more of these aromas or, most likely due to a higher incidence of flavour defects as appears from comparison of the left and the right panel of **Figure 6**. Defects are mostly caused by limitations in infrastructure (moulding, heat stress, oxidation (Pattee et al., 1965)). This often manifests as an origin effect, because a country of origin has a specific infrastructure (e.g. availability/quality of storage facilities), which could explain the coordinates of the Chinese cluster on the plot. However, it has been noted that this effect cannot easily be separated from the effect of different growing conditions, as every country of origin also has different climatic and geological conditions (Bett et al., 1994). In fact, growing conditions can often have a larger effect on the final composition of peanut kernels than genetics (Onemli, 2012; Pattee, Isleib, & Gorbet, 2004). A concrete conclusion cannot be drawn in this case because in this study we only have one variety from China, which is unique to China.

2.3.3 **Effect of process on sensory profiles**

A CVA using process technology as the classifier paints a different picture (**Figure 7a**). The sample space is mainly explained by degree of roast, from dark roast in the top left to raw beany in the bottom right. The top right and bottom left quadrants appear to resolve mainly defects, with the notable exception of the bitter and sweet tastes (top right). Looking at the process classifications (**Figure 7b**) logical clusters emerge: Oil roasting (frying) is somewhat isolated around x=0. All the dry roasted samples are clustered in the right, along the bitterness, sweetness and raw bean vectors. This suggests that differentiation amongst these attributes may be possible to a degree by varying the baking conditions (using a two-zone roasting step (processes G,F) vs a single zone roast (processes D,E). The fact that the confidence ellipses somewhat (but not entirely) overlap for these classes suggest that the extend would be small, but still potentially of use to peanut processors. This observation prompted additional analysis, which showed that the effect is in fact not significant across all raw materials. This analysis is discussed below.

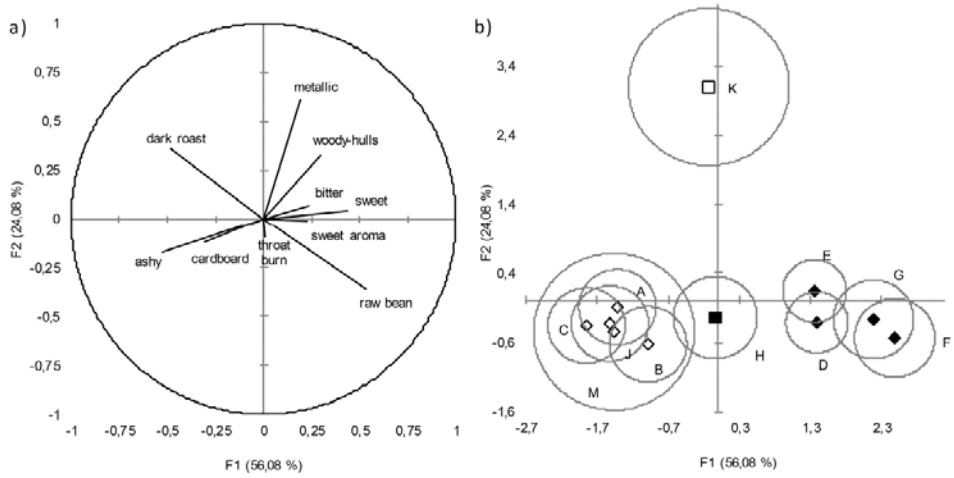

Figure 7. Left (a). Canonical Variate Analysis space of flavour sensory attributes for process technology classification. Only attributes that satisfied the α=0.2 variable selection criterion are shown. Right (b). Analysis of sensory attributes with process used as a classifier, with 95% confidence ellipses. Diamonds: dry roasted, squares: oil roasted (fried). Filled shapes: non-macerated, empty shapes: macerated. Codes for individual processes can be found in **Table 4**. Two components resolve 80.2% of the data variance.

Dextrose maceration (oil roasted) is the most differentiating process (**Figure 7**), yielding samples high in dark roast aroma but also metallic off notes, regardless of the starting raw material. Finally, all dry roasted maceration process had a similar but smaller impact on the flavour profiles, mainly driving dark roast, ashy and cardboard aromas. However, it is important to consider the magnitude of these changes: a statistically significant increase in an off-flavour, for example, may be of little practical importance if the magnitude of the attribute is still very low. Indeed, the magnitude of off flavours such as earthy, phenolic/chemical, metallic, fruity fermented and ashy aromas was below 1 on a 15 point scale (data not shown), which would be barely perceivable by untrained consumers. As a result, processes such as dextrose maceration can still be recommended to improve the dark roast aroma despite of its impact on metallic taste, because although statistically significant, the magnitude is too small to cause concern.

Figure 8 shows the main effect plots of maceration, frying/baking and oil topical application on roasted peanut, sweet, dark roast and raw bean aromas, sweet taste and the CIELAB colour parameters. It is worth considering why the models were selected to only include three main effects (bake/fry, maceration and topical application) and not attempt to account for more variance by including additional parameters to the model (such as market type, origin, variety, high oleic trait or grade). Firstly, the design of the study was only a fractional factorial, and so

multicollinearity concerns prevent one from resolving the effect of all these factors (for instance, only one material is from China). Secondly, accounting for more variance would indeed increase statistical power, but at a loss of generality and in danger of overfitting. This was not the intent of this research, as several studies with a balanced designs have already been published, precisely contrasting flavour differences between varieties, grades of the same varieties, growing origins of the same variety, or expression of the high oleic trait within a type (Bett et al., 1994; Isleib et al., 2015; Pattee et al., 2002; Sanders, 1989; Walczyk et al., 2013; Young et al., 2005). By building a simple model with only a few parameters related to process, all the variance related to raw materials is pooled in the error. This allows evaluating whether a process variable has an effect pronounced enough to overshadow raw material differences, something of value to processors dealing with several different raw materials.

Figure 8 shows that baking imparts a weaker roasted peanut and dark roast aroma (and stronger raw bean aroma). This could be explained by the lower temperature and heat transfer rates encountered in baking (vs frying) and consequently to the lower rates of the Maillard and caramelization reactions. Interestingly, sweetness was not seen to significantly vary when comparing frying to baking, even though sweetness is known to develop during heating via the breakdown of complex sugars into simple sugars (inositol, glucose, fructose, sucrose, raffinose, stachyose, etc.) (McDaniel, White, Dean, Sanders, & Davis, 2012; Pattee et al., 2000). The fact that sweetness is not significantly affected (whereas flavour formation is), could be explained as follows: Complex sugar degradation to monosaccharides is a sequence of monomolecular reactions, whereas flavour generation comprises of reactions that are (at least) bimolecular and therefore slower and dependent on molecular mobility. In addition, once simple sugars have been formed they can be consumed in Maillard and caramelization reactions, further contributing to an increase in roasted flavour but not sweetness. This agrees with the findings of Pattee et al, who proposed that sweet taste in peanut is mostly due to genetics and growing conditions (Pattee et al., 1965, 2000). An alternative explanation is that the changes in sugar content were too small for the sensory panel to detect in the roasting ranges investigated.

Maceration appears to have a mixed effect on flavour: it increases roasted peanut and dark roast aromas, and decreases sweet and raw bean aroma and sweet taste. This can be explained by two mechanisms: Firstly, maceration appears to increase the formation of roasted notes (increase roasted peanut and dark roast, and decrease raw bean aromas), likely due to its impact on the Maillard reaction: elevated moisture content can increase molecular mobility, and therefore the reaction rate (Fennema, 1996). Increasing the concentration of reagents (dextrose

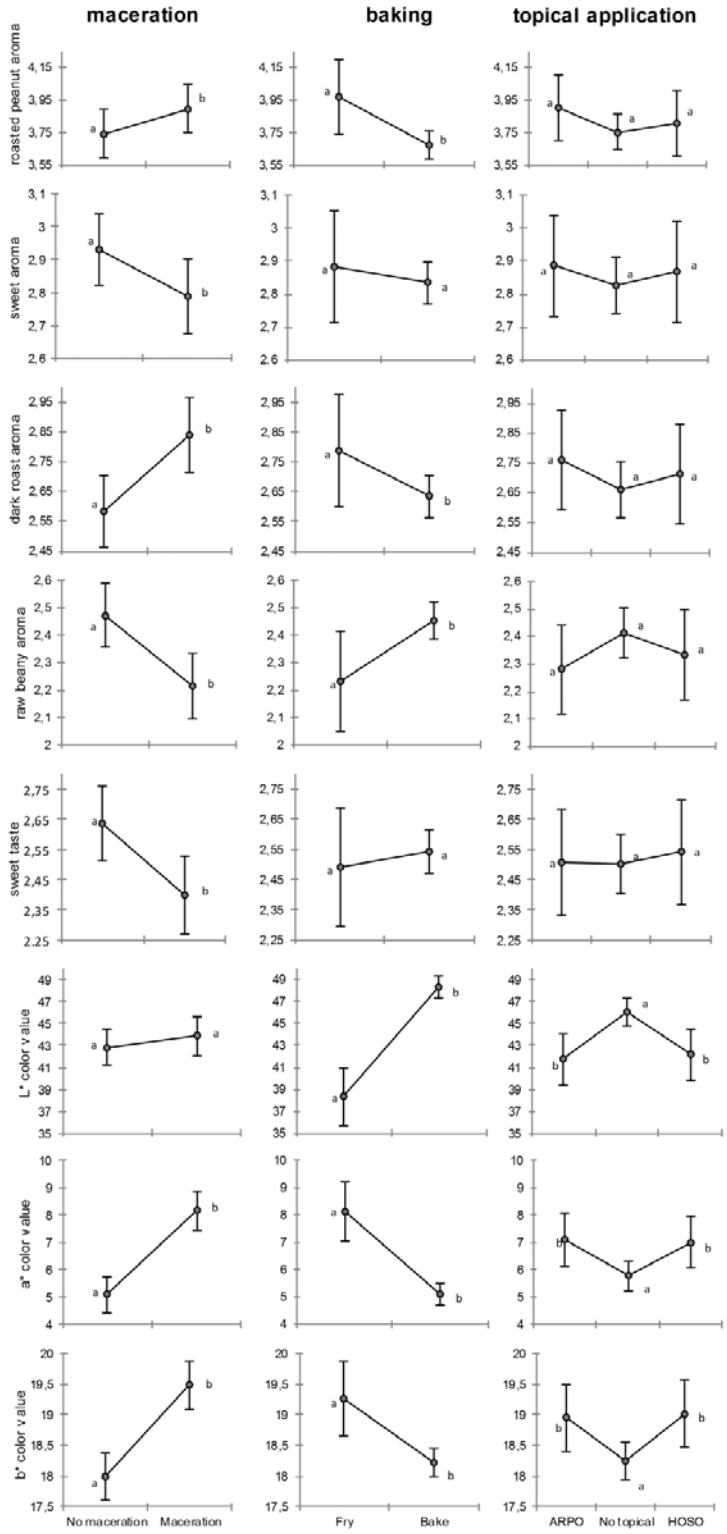
maceration
baking
topical application
roasted peanut aroma
sweet aroma
dark roast aroma
raw beany aroma
sweet taste
L* color value
a* color value
b* color value
No maceration
Maceration
Fry
Bake
ARPO
No topical
HOSO

Figure 8. Impact of peanuts process on sensory attributes: main effects plot with 95% confidence intervals. Significance of means difference was calculated using Tukey's HSD test (α=0.05). (HOSO: High Oleic Sunflower Oil, ARPO: Aromatic Roasted Peanut Oil).

maceration) or changing the pH (alkali and acidic maceration processes), will also affect the Maillard reaction kinetics. The reaction consumes intrinsic reducing sugars and generates polymerized non volatiles, both of which could be responsible for the reduction in sweetness and the increase in bitterness observed. Secondly, presence of water has been shown to increase the free amino acid concentration after roasting, which could also increase bitterness and manifest as a reduction in sweetness (Chiou, Chang, Tsai, & Ho, 1991). An aqueous environment is known to promote lipid oxidation, which can also suppress perceived roasted notes (Civille & Dus, 1992; St Angelo, 1996). This however, was not seen to overpower the positive effect of maceration on roasted flavour development, allowing the process to remain practical in a manufacturing environment.

It is not surprising that the application of an Aromatic Roasted Peanut Oil (ARPO) increases sweet, roasted peanut and dark roast aromas (as it is a flavouring agent itself), but it is surprising that the effect was not statistically significant at the level applied. However, topical oil application (HOSO or ARPO) had an impact on colour, but this is likely due to the physical effect wetting the surface.

Finally, colour was also affected by processing, even though the final moisture content of all samples was very similar. Browning of peanuts has been argued to be mostly caused by Maillard and to a lesser degree by caramelization reactions at the surface (Neta et al., 2010; Pattee, Giesbrecht, & Young, 1991). Both reactions are temperature dependent, and so as expected frying (higher temperature than baking) yields a darker product (lower L*). Maceration did not significantly affect the L* value, suggesting the effect is mostly temperature dependent. An additional contributor to the darker colour of the fried samples was the presence of surface oil, which tends to fill surface imperfections that would otherwise diffuse light. This effect is also evident in **Figure 8**, where the samples with topical ARPO or HOSO have statistically significant lower L values than untreated peanuts. The a* (red-green) and b* (blue-yellow) CIELAB values are also significantly different for both pre-treatment (maceration) and roasting (bake/fry). The analysis shows that maceration generally yields a more orange-brown colour (higher a* and b*). The macerating media had various pH values and reducing sugar contents, as previously described. These factors will have an impact on the evolution of the Maillard reaction, leading to melanoidins of a different degree of conjugation and therefore colour. The data also show that baking reduces the orange-brown colour (lower a* and b*) commonly associated with roasted goods when compared to frying, something that can be attributed to lower kinetic rates for the Maillard reaction due to the lower roasting temperatures employed in the

baking treatments. As in the case of the L* value, surface oil is also likely contributing to the observed a* and b* value differences.

To evaluate if even smaller process changes such as the oven temperature-time profile is sufficient to drive flavour difference in the finished product as hinted in **Figure 7**, the Plackett Burman subset (only baked, non-macerated samples D-G in **Table 4**) was analysed (data not shown). The analysis showed that different baking profiles had no effect on flavour and colour significant enough to over shadow the impact of different raw materials across the range investigated.

2.3.4 **Effect of processing on peanut fatty acid profile**

An overview of the range of the fatty acid profiles observed can be found in **Table 7**. An examination of the principal component analysis of the fatty acid profiles (**Figure 9**) shows oleic acid (C18:1cis) content (bottom left quadrant) to be inversely related to linoleic (C18:2cis), palmitic (C16:0), stearic (C18:0, 'tallow' odour (Burdock, 2010)), arachidic (C20:0) and behenic (C22:0) acids (top right quadrant). This is expected as high oleic varieties have been bred for increasing the oleic to linoleic acid ratio in order to improve shelf life, (Davis, Dean, Faircloth, & Sanders, 2008; Derbyshire, 2014; Riveros, Mestrallet, Nepote, & Grosso, 2009) and thus high oleic samples are mapped mainly on the bottom left quadrant, and conventional on the top right quadrant. Most of these fatty acids have little or no odour (Burdock, 2010), and it is thus not surprising that no significant flavour differences have been reported between high oleic and conventional peanuts in sensory (Isleib et al., 2015, 2006) or consumer tests (Riveros et al., 2009).

Table 7. Overview of the range in fatty acid profiles for all 134 samples analysed.

fatty acid	average (% w/w)	standard deviation	minimum (% w/w)	maximum (% w/w)
C14:0	0.04	0.08	0.00	0.43
C16:0	8.87	2.62	4.89	14.82
C16:1trans	0.03	0.01	0.00	0.05
C16:1cis	0.05	0.02	0.00	0.09
C17:0	0.07	0.04	0.00	0.24
C17:1cis	0.03	0.04	0.00	0.11
C18:0	2.56	0.72	1.61	4.78
C18:1cis	58.33	16.10	38.26	81.85
C18:2cis	22.77	13.79	3.25	39.37
C18:3cis	0.03	0.04	0.00	0.11
C18:4cis	0.16	0.46	0.00	2.38
C20:0	1.22	0.22	0.87	1.85
C20:1cis	1.40	0.75	0.00	2.84
C22:0	2.75	0.34	1.64	3.52
C22:1cis	0.13	0.13	0.00	1.00
C24:0	1.53	0.28	0.60	2.25

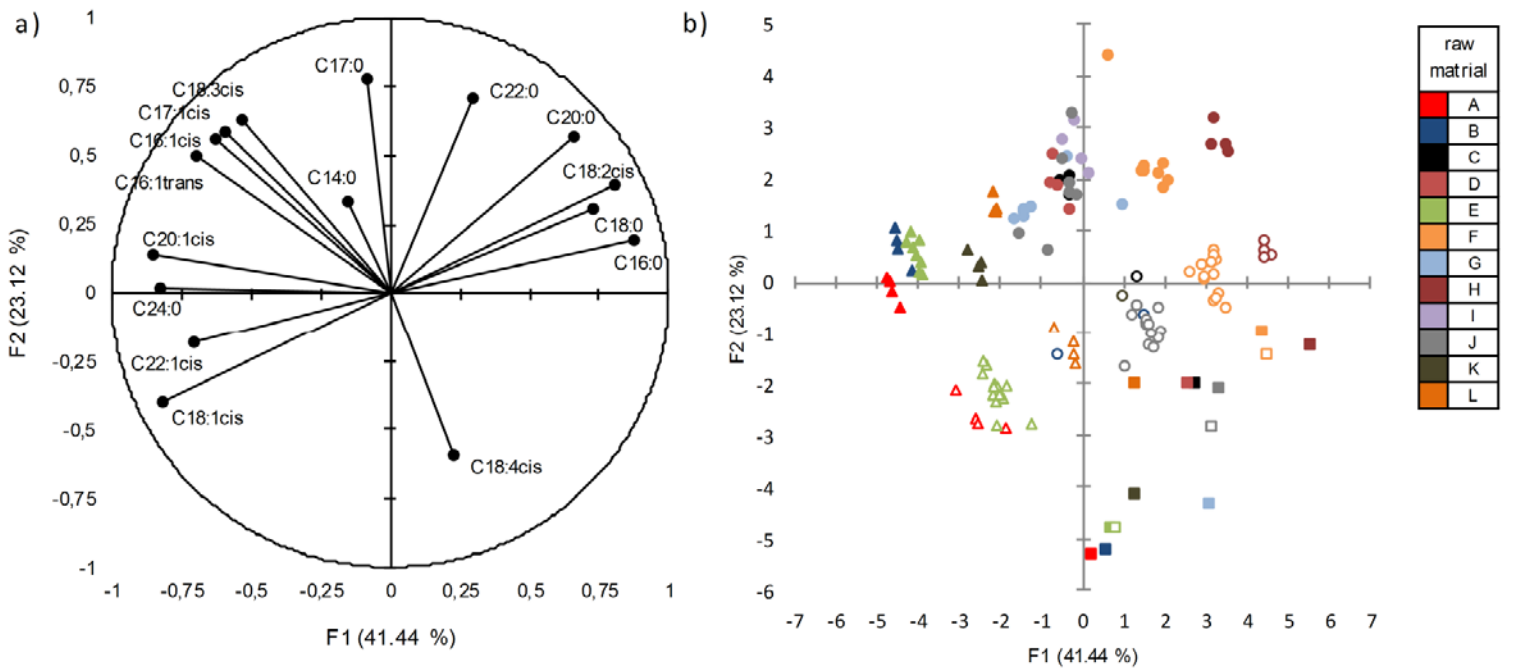

Figure 9. left (a) PCA scores for the fatty acid profiles. Right (b). Principal component loadings of fatty acid profiles. triangles: high oleic, circles: non high oleic, squares: fried, empty shapes: macerated, filled shapes: non macerated. Colours represent different raw materials, key in **Table 3**. Two principal components explain 64.6% of variance.

It is clear therefore that a significant variation in the fatty acid profile can be attributed to the raw material (genetic makeup and growing conditions, as previously reported (Davis et al., 2008; Onemli, 2012)). However, raw material selection alone is not sufficient to explain all the variance in fatty acid profiles observed: Fried samples are clustered in the bottom right quadrant, an area related to high stearidonic acid (C18:4cis) concentration, contributed mainly by the frying oil (high oleic sunflower oil). This is also shown in **Figure 9**, where samples from the same raw material (colour coding) are only loosely clustered together. Macerated samples skew away from the more saturated regions (**Figure 9**, bottom right & top left quadrants). This could be explained on the basis of lipid hydrolysis and/or lipid oxidation reactions (primarily of linolenic acid) occurring at elevated rates while the kernels are in an aqueous environment (St Angelo, 1996). Processing conditions therefore, and in particular maceration, appear to affect the fatty acid composition of peanuts. Branch and co-workers also noted a similar effect, but determined that it did not affect the shelf life of the peanuts (Branch et al., 1988). Manufacturers would have to compensate with a higher roast level to mask some of the oxidation by-products created during maceration (Civille & Dus, 1992), or use lipid oxidation resistant high oleic peanuts (Braddock, Sims, & O'Keffe, 1995; Davis et al., 2008). The minimum water quantity possible should be used for maceration, as excess water can extract water soluble flavour precursors and thereby reduce the peanut flavour of the finished product (Muego-Gnanasekharan & Resurreccion, 1993).

2.4 Conclusions

In this study, the effect of raw material and processing on peanut flavour has been investigated. Roasted peanut samples from a wider range of raw materials and processes than previously encountered in the literature have been produced (134 unique samples). To avoid overfitting, the models developed include as few parameters as possible, so that the main effects identified as significant are more likely to be significant across most market types, varieties, origins and grades of raw materials. Based on this wide sample set we have showed that, although the choice of raw material is clearly very important in determining the final flavour attributes and fatty acid composition of roasted peanuts, peanut processors can induce significant changes to the flavour, colour and fatty acid profile by manipulating the process conditions, and in particular by introducing a maceration step prior to roasting. However, if limited within baking (with no maceration) and moderate temperature ranges (±10 °C), only minor flavour modification can by induced, and the effect of raw material variety will be dominant.

Process settings that allow for the adjustment of finished product flavour attributes can be hugely valuable to food processors because they make flavour

optimization of the product possible, whether it is for the entire market, or a market segment (for example by creating line extensions such as 'extra dark roast' or 'mild flavour'). Being able to do so without relying on a restrictive raw material procurement strategy however, is even more beneficial due to the potential cost savings and reduced food waste. From an academic perspective, obtaining insights from large data sets of a seemingly simple category can highlight the power of novel data analysis techniques, as well as identify effects that can later be explained by first principles. Finally, insights in the relationship between sensory attributes, chemical composition and process conditions can also spark ideas for future research; the most interesting to us being to develop a roasting process that yields peanuts with a significantly extended shelf life, given that processing can affect both the flavour intensity and fatty acid composition.

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3 Correlation of volatile compounds to sensory aroma characteristics.

Adapted from:

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Abstract

Flavour and colour of roasted peanuts are important research areas due to their significant influence on consumer preference. The aim of the present study was to explore correlations between sensory attributes of peanuts, volatile headspace compounds and colour parameters. Different raw peanuts were selected to be representative of common market types, varieties, growing locations and grades used in Europe. Peanuts were roasted by a variety of processing technologies, resulting in 134 unique samples, which were analysed for colour, volatile composition and flavour profile by expert panel. Several headspace volatile compounds which positively or negatively correlated to 'roasted peanut', 'raw bean', 'dark roast' and 'sweet' attributes were identified. Results demonstrated that the correlation of CIELAB colour parameters with roast related aromas, often taken for granted by the industry, is not strong when samples of different raw materials are subjected to different processing conditions.

Keywords: Peanut, Maillard, sensory, instrumental analysis, raw material, aroma

Highlights:

- The chemical fingerprint of several sensory flavour attributes was determined.
- Both positively and negatively correlated headspace volatiles were identified.
- A logarithmic transformation on the concentrations was used to increase the fit.
- Colour is correlated to roasted flavour but the correlation is not universal.

3.1 Introduction

Flavour and colour are important attributes affecting the consumer preference of whole roasted peanuts (Lee & Resurreccion, 2006; Young, Sanders, Drake, Osborne, & Civille, 2005), and are therefore commercially significant areas of research. Flavour development in peanuts, however, is a complicated topic as it depends on several factors including the genetic makeup and growing conditions of the plant, post-harvest handling, processing and storage conditions (Neta, Sanders, & Drake, 2010).

Significant work has been done over the last 40 years in identifying the flavour components of roasted peanuts by quantifying volatile compounds by various GC-MS techniques, (Chetschik, Granvogl, & Schieberle, 2008, 2010) but most of it has used a smaller number of processes or peanut types. Recently the focus has been on flavour active compounds (Chetschik et al., 2008, 2010; Schirack, Drake, Sanders, & Sandeep, 2006a) which are generated in peanuts through a variety of common pathways, including the Maillard reaction, Strecker degradation, thermal degradation of sugars, and lipid oxidations (Neta et al., 2010). Roasted peanut flavour has been mostly attributed to heterocyclic compounds, such as alkylpyrazines (Williams et al., 2006) or more specifically to O-heterocyclic and N-heterocyclic compounds (Liu et al., 2011). In general, in older investigations pyrazines appeared to be the most important compounds produced during roasting in peanuts, (Baker et al., 2003; Ho, Lee, & Chang, 1981; Ho C.T., Jin Q.Z., Lee M.H., & Chang S.S., 1983) even though more recently this has been disproven using recombination studies (Chetschik et al., 2010; Da Conceicao Neta, 2010).

To identify correlations between food analytical data and sensory attributes is of major importance from a scientific standpoint and has practical applications for the food industry. Several studies have tried to correlate volatile compound concentration to sensory response. Some were focused on quality defects (Reed, Sims, Gorbet, & O'Keefe, 2002; St Angelo, 1996) while others specifically on roasted peanut flavour (Baker et al., 2003; Chetschik et al., 2010; Da Conceicao Neta, 2010; Liu et al., 2011), and two of these studies were also able to validate their findings with aroma reconstitution studies (Chetschik et al., 2010; Da Conceicao Neta, 2010). The correlation between volatiles and sensory attributes is arguably of larger practical value than identifying character impact compounds. For example, it has been argued that the total 'roasted peanut' flavour is reduced during storage not because of the depletion of the relevant aroma active compounds, but because of the creation of flavour-masking low molecular weight aldehydes from lipid oxidation, although this was not proven by recombination studies (Warner, Dimick, Ziegler, Mumma, & Hollender, 1996). The fingerprinting approach therefore, can provide incremental information to GC-O studies by identifying potential flavour antagonists, but a recombination study would still be required to definitively prove the effect.

Potential synergistic or antagonistic effects cannot be detected by GC-O,

and such effects are often reported. For instance, the (sensory) odour detection threshold is usually much larger in complex food matrices versus water (Belitz, Grosch, & Schieberle, 2004). Specific antagonistic examples include 2,5-dimethyl-3-methoxypyrazine which has an 'earthy' character, but in red wine it also shown to dramatically reduce 'cherry' and 'red berry' notes, in addition to inducing an 'earthy' note (Botezatu & Pickering, 2012). Synergistic effects have also been documented: panellists did not detect beany notes in pure hexanal nor trans-2-nonenal, but did report beany notes in a blend of the two (Bott & Chambers, 2006). Analogously, a mixture of 4-hexanolide, (E)-2-hexenyl hexanoate, (Z)-3-hexenol, and indole (all at subthreshold levels, and thus odourless in isolation) can exhibit an 'astringent' or 'heavy' odour (Ito & Kubota, 2005). For this reason, a fingerprint approach can represent a complementary approach to GC-O for identifying potential flavour contributing molecules (Chetschik et al., 2008, 2010; Schirack et al., 2006a), although both methods require validation with a recombination study if the objective is to prove causation.

In addition to flavour, colour is also an important attribute in consumer preference of roasted peanuts (Lee & Resurreccion, 2006). Given that colour and flavour develop through similar pathways (Maillard and caramelization reactions), it is not surprising that a correlation between colour parameters and roasted aromas has been identified (Lee & Resurreccion, 2006; McDaniel, White, Dean, Sanders, & Davis, 2012; Pattee, Giesbrecht, & Young, 1991). To our knowledge however, little research has been done to explore the colour - flavour relationship across different market types, varieties and origins of peanuts.

The aim of this research was to study correlations between volatile compounds, sensory and colour attributes of roasted peanuts. To that aim, several raw materials and processes were employed to produce a diverse sample set, and the intent was to evaluate whether previous findings are also applicable across this wider range of peanut varieties and process technologies. Moreover, we aimed at evaluating several flavour sensory attributes simultaneously. In the process of pursuing these objectives we employed two original approaches in peanut flavour research, namely logarithmically transforming the GC-MS data prior to modelling, and considering compounds that negatively correlate with the flavour attributes under investigation.

3.2 Materials and Methods

3.2.1 Peanut samples

The peanuts samples used in this research are identical to those presented in Chapter 2. The full experimental detail on sample preparation has also been published in the complementary part of this research, but the procedure is also summarized below.

Shelled, raw peanuts of different market types (Valencia, Virginia and Runner), grades (sizes) and origins (USA, China, Argentina and Australia) were sourced from Canon Garth Ltd (London, UK). The peanuts were dry blanched to remove the testa at approximately 85 °C for 30 minutes, (Sanders, Adelsberg, Hendrix, & McMichael, 1999) followed by mechanical abrasion (Steinweg-Handelsveem BV, Oosterhout, NL), and kept at -15 °C until further processed, generally within less than 6 months. The blanched seeds were consequently processed according to a fractional factorial design, and 134 roasted peanut samples were obtained (**Table 8**), ensuring that the final moisture content for all treatments was between 2 and 2.7% w/w. The process can be visualized as having three parts: a pre-treatment, a thermal treatment, and a post treatment. The pre-treatment consisted of maceration of raw peanuts in various media (43% w/w) at 20°C for 30 to 90 minutes, and was only applied to a subset of the samples (**Table 8**). The thermal treatment consisted of baking in an continuous oven (Aeroglide C1 12-16 REX, Cary, NC) at temperature between 135°C and 155°C or fried in a batch fryer (30L model, , De Kuiper (De Kwakel-Uithoorn,NL)) at 150°C for the time required to reach the target moisture content. To better resolve the impact of dry roasting in particular on flavour, a (2 level – 2 factor) Plackett-Burman design was employed. The total heat treatment was conceptually divided in two zones of equal time, and the temperature for each zone was fixed at high (155°C) or low (135°C). The total baking time was adjusted so that the final moisture content was approximately 2%, and was evenly split between the two zones.

Finally, the post treatment consisted of salting in a coating drum (S-1050, Walter Brucks, Bruggen, DE). All samples were salted to 1% NaCl w/w (Cargill, MO, USA), and either High Oleic Sunflower Oil (HOSO, Cargill, MO) or Aromatic Roasted Peanut Oil (ARPO, 100E, NutrIn, Washington DC) was applied at 2% w/w to a subset of the samples (**Table 8**). ARPO are unrefined mechanically expelled peanut oils, with a strong characteristic roasted peanut odour (Liu et al., 2011).

All reagents used in this research were obtained from Sigma-Aldrich (St Louis, MO), unless otherwise specified.

3.2.2 Dynamic Headspace Gas Chromatography-Mass Spectrometry (DHS-GC-MS)

Volatile compounds in peanut samples were analysed by DHS-GC-MS. Heptanone-d5 and 1,2-dichlorobenzene were used as internal standards and C5 to C25 n-alkanes used as standards for the retention index determination. A one hundred gram sample of peanut was ground in a mini food processor (Kenwood, Havant, UK), and 2 grams were added into a 20mL sample vial (Thermo Fisher,

Table 8. Overview of the full experimental design. Letters indicate which raw materials were processed in the respective process-topical treatment combination. More information on raw materials and process conditions can be found in the complementary part of this research (Lykomitros, Fogliano, & Capuano, 2016).

process	no topical	ARPO topical [a]	HOSO topical [b]
aqueous acid maceration, dry roasting	A,C,E,F,H,J,L	E,F,J	E,F,J
cold (long) aqueous maceration, dry roasting	A,C,E,F,H,J,L	E,F,J	E,F,J
aqueous dextrose maceration, dry roasting	A,C,E,F,H,J,L	E,F,J	E,F,J
dry roasting (low temperature long time)	A,B,C,D,E,F,G,H,I,J,K,L	E,F,J,G	E,F,J,G
dry roasting (high temperature short time)	A,B,C,D,E,F,G,H,I,J,K,L	E,F,J,G	E,F,J,G
two temperature zone dry roasting (high-low)	A,B,C,D,E,F,G,H,I,J,K,L		
two temperature zone dry roasting (low-high)	A,B,C,D,E,F,G,H,I,J,K,L		
oil roasting (frying)	A,B,C,D,E,F,G,H,I,J,K,L		
aqueous alkaline maceration, dry roasting	E,F,J		
aqueous dextrose maceration, oil roasting	E,F,J		
cold (short) aqueous maceration, dry roasting	A,C,E,F,H,J,L	E,F,J	E,F,J

A: Runner, Flavorunner 458, M grade, High Oleic, USA – Texas
B: Runner, Flavorunner 458, Jumbo grade, High Oleic, USA-Texas
C: Runner, Georgia Green, M grade, Low Oleic, USA – Georgia
D: Runner, Georgia Green, Jumbo grade, Low Oleic, USA – Georgia
E: Runner, Granoleic, Jumbo grade, High Oleic, Argentina
F: Valencia, CN Natals, S grade, Low Oleic, South Africa
G: Runner, Tegua, M grade, Low Oleic, Argentina
H: Runner, Hsuji, M grade, Low Oleic, China
I: Virginia, mixed varieties, XL grade, Low Oleic, USA – Virginia
J: Virginia, mixed varieties, M grade, Low Oleic, USA – Virginia
K: Virginia, Middleton, XL grade, High Oleic, Australia
L: Virginia, Middleton, M grade, High Oleic, Australia

[a] Aromatic Roasted Peanut Oil applied as topical spray; [b] High Oleic Sunflower seed Oil applied as topical spray. Details on both topically applied oils in the complementary part of the research (Lykomitros et al., 2016).

Waltham, MA), with 1µL d5-heptanone (100µg/mL in methanol) internal standard and 400µl of Millipore water, prior to sealing with caps (Gerstel, Mülheim an der Ruhr, DE). A vial with 1µL of the C5-C25 n-alkanes standard (100µg/mL each in methanol) was also analysed under the same conditions to enable the calculation of the Linear Retention Index values. Finally, 1µL of the 1,2-dichlorobenzene (13.06µg/mL in ethanol) internal standard was injected directly on each Tenax trap used for the volatile collection. All additions/injections were made using a micro syringe (Thermo Fisher, Waltham, MA) that was first rinsed with purge & trap grade methanol in triplicate, followed by one rinse with the standard.

The GC-MS setup consisted of a Gerstel Automated Dynamic Headspace Agilent Technologies 7890A GC system with 5975C inert MSD – triple axis detector (Mülheim an der Ruhr, DE), and a DB-5ms 60m x 0.32mm ID, 1 µm film thickness

column from Agilent (Welmington, DE). The sample vials where incubated at 37 °C, and purged with 2400 ml of Helium gas (rate 40ml/min), and the volatiles trapped on a Gerstel Tenax TA trap at 37 °C. For the chromatography, Helium gas was used at 2.5 mL/min, while the column temperature profile was 40 °C for 7.5 minutes, 4 °C/min to 210 °C, followed by 15 °C/min to 320 °C for 8 minutes. The desorption temperature was set to 30 °C ramped to 320 ^{O}C at 400 ^{O}C/min, with a five minute hold in a Gerstel Thermal Desorption Unit with a 1:10 split ratio. The mass spectrometer source temperature was to 230 °C, and the quadrupole temperature to 150 °C. The ionization voltage was set at 70 eV and a scan range from m/z 33 to 400.

Identification of peaks was made using the Agilent Technologies MSD Productivity ChemStation Software (Welmington, DE), which references the NIST 2005 mass spectral database (Boulder, CO) and Linear Retention Indices. Where identification of peaks was not possible, the peak was marked as 'unknown' and the retention index was noted. Finally, the same software was used to compare the resolved peaks against that of the d5-heptanone internal standard using a response factor of 1, in order to provide the semi-quantification data. To obtain a measure of reproducibility, a random sample was measured in sextuplicate and the Coefficient of Variation for twenty five typical volatiles was calculated. The median coefficient of variation was 0.24.

3.2.3 Colour and moisture measurements.

The colour was measured by a Hunter Lab CR400 colorimeter (Reston, VA, USA), in triplicate. Two hundred and fifty grams of whole peanuts were dispersed in a 95mm diameter petri dish, and the colour values were obtained. The peanuts were redistributed in the petri dish between the replications. The moisture content was determined by a Leco TGA701 thermogravimetric analyser (St. Joseph, MI, USA) in duplicate with a 3 gram sample, taken from a quantity of 100 grams of peanuts previously ground in a mini food processor (Kenwood, Havant, UK).

3.2.4 Sensory Analysis.

Five hundred grams of each of the samples was ground to a paste (DLC-7/DLC-001 Cuisinart, E. Windsor, NJ) and Descriptive Sensory Analysis (DSA) was performed in duplicate by a ten member (3:7 male:female, average age: 33, age range: 20-55) trained DSA panel (minimum 250 hours experience in DSA panels on peanuts for each member) at the USDA, ARS, Market Quality and Handling

Research Unit (Raleigh, North Carolina, USA), using the Spectrum ™ Method (15 point scale). Details on the methods, lexicon and attribute definitions have been previously published, (Johnsen, Civille, Vercellotti, Sanders, & Dus, 1988; Sanders, Vercellotti, Crippen, & Civille, 1989; Schirack et al., 2006a). The sensory attributes used were: roasted peanut aroma, sweet aroma, dark roast aroma, raw beany aroma, woody/hulls/skins aroma, cardboard aroma, earthy aroma, painty aroma, phenolic/chemical aroma, fruit/fermented aroma, ashy aroma, total off notes, sweet taste, sour taste, bitter taste, tongue/throat burn sensation, metallic sensation and astringent sensation.

3.2.5 Statistical analysis.

Principal Component Analysis (PCA) was used to analyse the sensory variation within the samples (eigenvectors were calculated from the covariance matrix). Where appropriate, volatile concentrations and colour attributes were superimposed on the biplots as secondary variables but not included in the computation of the eigenvectors. Pearson's correlations were calculated to visualize correlations between chromatographic, sensory and colour data. However, in the case of the colour analysis the data set was reduced to only include samples made by the four roasting methods or frying with no topical oil/ARPO applied, for reasons that will be discussed below.

Partial Least Squares Regression (PLSR) models were developed to identify correlations between instrumental (X matrix) and sensory (Y matrix) measurements. A maximum of 4 components were included in each PLS model and the confidence interval was set at 95%. The Variable Importance for the Projection (VIPs) values were calculated to estimate the importance of each variable in the PLS projection (Shaffer, 2002). Variables with VIP>1 are considered highly influential in the model whereas variables with 0.8<VIP<1 were considered moderately important for the PLS model. PLSR models were developed with both raw, untransformed chromatographic data and after logarithmic transformation of the data. All the statistical analyses were performed using XLSTAT (XLSTAT-Sensory package, Addinsoft, Paris, FR) on MS Excel 2010 (Microsoft, Redmond, WA, USA). The general form of the models can be seen in **Equation 1.**

$$Attribute_i = constant_i + \sum_{103}^{j=1}\left(concentration_j \times coefficient_{i,j}\right)$$

Equation 1

Where, i= the i^{th} attribute, j= the j^{th} compound (GC-MS peak), $concentration_j$ = the concentration (or log transformed concentration) of the j^{th} compound, $coefficient_{i,j}$

= the model coefficient of the j^{th} compound in the i^{th} model (sensory attribute) and $constant_i$ = the constant of i^{th} model (sensory attribute)

3.3 Results and discussion:

3.3.1 Sensory and volatile profile of peanut samples.

Figure 10 summarizes the intensities of all sensory attributes for the 134 roasted peanut samples. A wide range in the values of the response variable (flavour attributes in this case) is preferable in regression analysis. Variability was especially large for roasted peanut, sweet, dark roast, raw bean, cardboard and painty aromas as well as sweet and bitter taste. We decided to include all the attributes in the PCA, but focus our regression analysis only on the first four attributes, namely roasted peanut, sweet, dark roast and raw bean aromas. This is because painty and cardboard aroma are mostly caused by lipid oxidation (St Angelo, 1996) and sweet and bitter taste is likely better correlated to non-volatile compounds which were not measured in this research.

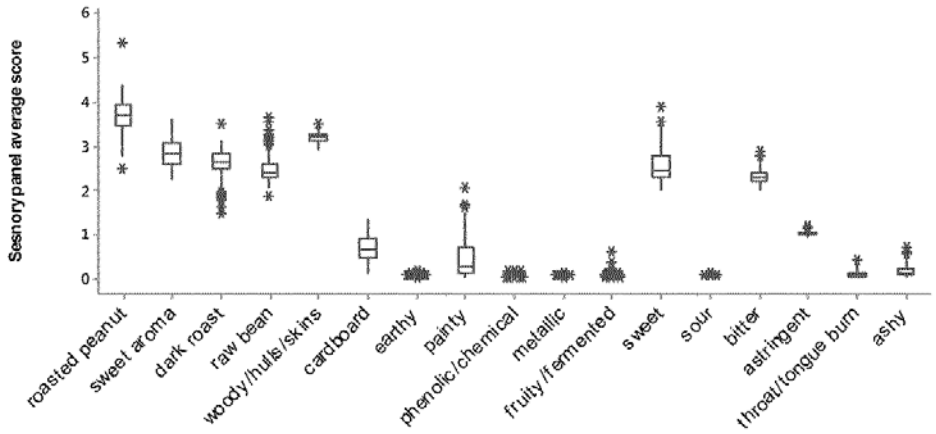

Figure 10. Box plot of sensory attributes scores from the expert panel. Horizontal line: median, box: 1st (Q1) to 3rd (Q3) quartile, upper whisker: Q3+1.5(Q3-Q1), lower whisker: Q1-1.5(Q3-Q1), *: outliers (defined as the observations outside the whisker range).

Overall, in the GC-MS analysis 103 peaks were quantified and used in the statistical analyses. In **Table 9** the results are summarized, by listing the 45 compounds with the highest observed concentrations, plus an additional 28 compounds that were highly correlated with the flavour attributes in question. For simplicity, the 30 remaining compounds that were present at very low

concentrations and very weakly correlated to the flavour attributes are not listed in tables and figures. 1-methyl-1H-pyrrol (associated with sweet and woody odour) and 2,5-dimethylpyrazine (associated with sweet, malty odours) are two of the most reported aromatic components in roasted peanuts (Braddock, Sims, & O'Keffe, 1995; Ho et al., 1981; Ng, Dunford, & Chenault, 2008; Schirack et al., 2006a; Schirack, Drake, Sanders, & Sandeep, 2006b; Williams et al., 2006) and were also among the compounds detected at the highest concentrations in this study. In fact, some past researchers went further to suggest that 2,5-dimethylpyrazine is the compound with the single highest correlation with roasted peanut aroma, followed by 2,3,5-trimethylpyrazine and 2,3-dimethylpyrazine (Baker et al., 2003), which were also detected in this research, even though the flavour activity of 2,5-dimethylpyrazine has been since disproven (Chetschik et al., 2010). 2-methylpyrazine, 2,6-dimethylpyrazine, 2,3,5-trimethylpyrazine, and ethylpyrazine often cited in peanut shelf life studies (Mei, Qi, Chang-sheng, Chang, & Feng-hong, 2011; Reed et al., 2002; Warner et al., 1996) can also be found in the same table. Benzeneacetaldehyde, benzaldehyde, 1-pentanol, and N-methylpyrrole related to the 'sweet' aroma (Braddock et al., 1995; Ho et al., 1981; Ho C.T. et al., 1983; Ng & Dunford, 2009) were also detected in this study. Finally, simpler aldehydes such as 2-methylpropanal, 3-methylbutanal, pentanal, octanal, hexanal and nonanal as previously reported (Didzbalis, Ritter, Trail, & Plog, 2004; Neta et al., 2010; Ng & Dunford, 2009) were also detected in this study, but, as also noted elsewhere, they were not always associated with off flavours (Chetschik et al., 2010; Da Conceicao Neta, 2010).

Table 9. List of the 45 compounds with the highest concentrations in roasted peanuts and additional 28 compounds that were identified as highly correlated with the tested aroma attributes. Each compound appears at its highest concentration in the sample set.

concentration relative to internal standard	compound	odour [a]	major ions	quantitation ion	factor (total ion count area/ quantitation ion area)	Linear Retention Index	previously reported odour active in peanuts [b]
97084	hexanal	green, grassy	44, 56, 72, 82, 100	82	29,11	800	cde
18489	1-methyl-1H-pyrrole	nutty, sweet [f]	81, 53, 39, 66	TIC	1	740	
10689	1-hexanol	woody, sweet, green fruity	56, 43, 69, 84	56	4,07	868	
9656	3-methyl-butanal	almond, apple, acrid	44, 58, 71, 86	TIC	1	656	cd
8351	2,5 dimethyl pyrazine	nutty, cocoa, green	108, 42, 81, 52, 66	81	16,54	914	
8335	pentanal	powerful acrid	44, 58, 71	58	6,86	699	

concentration relative to internal standard	compound	odour [a]	major ions	quantitation ion	factor (total ion count area/ quantitation ion area)	Linear Retention Index	previously reported odour active in peanuts [b]
6733	methyl-pyrazine	nutty, cocoa, green, roasted	94, 67, 39, 53	94	2,7	825	
5371	toluene		91, 65, 39, 51	91	2,86	770	g
5158	undecane		57, 43, 71, 85	71	9,48	1099	c
4882	octane		85, 59, 71	85	6,99	799	
3081	3-hexen-2-one	metallic, acrid [f]	83, 55, 98, 43	83	4,67	797	
2556	butyrolactone	sweet, buttery	42, 56, 86	42	3,01	911	
2381	trimethyl-pyrazine	baked potato, roasted nut	122, 42, 81	81	21,74	1003	ce
2218	dimethyl-disulfide	onion- like	94, 79, 45, 61	94	2,85	747	c
2216	benzaldehyde	sweet, strong bitter almond	105, 106, 77, 51	106	4,41	969	g
2199	1-pentanol	fusel-like sweet	42, 55, 70	70	9,37	767	
2009	2-octenal (E)	fatty, green	41, 55, 70, 83, 97	97	54,31	1060	
1969	1-octen-3-ol	earthy, mushroom-like	57, 43, 72, 85, 99	57	2,51	979	cd
1891	benzeneacetaldehyde	harsh green	91, 120, 65, 51, 39	91	2,14	1051	g
1451	2-heptenal (E)	pungent green, fatty [f]	83, 41, 55, 70, 97	70	21,59	959	c
1274	2-pentyl-furan	fruity	81, 138, 53	81	2,32	991	
1156	nonanal	fatty, orange/rose	41, 57, 70, 82, 98	70	22,4	1105	cg
1113	p-xylene/ 1,3 dimethyl-benzene		91, 106, 77, 63, 51	91	2,92	874	
860	2,3-pentanedione	honey-like, quinone	43, 57, 100	100	11,95	698	cde
814	2,3 dihydro-benzofuran		120, 91, 119, 65	91	6,28	1214	
787	ethyl-pyrazine	peanut butter, musty, nutty, woody	107, 108, 80, 53	107	2,69	918	e
770	heptanal	penetrating oily, harsh	70, 44, 55, 81, 96	70	9,02	902	cg
628 [h]	2,2,4,6,6-pentamethyl-heptane		57, 71, 85, 99	71	29,1	993	

concentration relative to internal standard	compound	odour [a]	major ions	quantitation ion	factor (total ion count area/ quantitation ion area)	Linear Retention Index	previously reported odour active in peanuts [b]
615	3(2H)-furanone, dihydro-2-methyl	wintergreen	43, 72, 100	72	4,59	807	
604	p-xylene		91, 106, 77, 63, 51	91	2,74	897	
493	2-heptanone	banana, cinnamon	43, 58, 71	58	3,98	889	
484	1-chloro pentane		55, 70, 42	70	6,89	757	
439 [h]	2,3- butanediol		45, 57, 75, 90	45	1,75	780	g
431	2,3 dimethyl-pyrazine	nutty, cocoa like	108, 67, 40, 52	67	4,1	920	e
390	3-methyl-1-butanol	fusel oil, whiskey characteristic pungent	55, 70, 42	70	7,39	736	
324	1-octanol	fresh, orange-rose, sweet	56, 70, 84, 41	84	28,27	1069	
297	ethylbenzene		91, 106, 65	91	2,67	865	
287	1-(acetyloxy)- 2-propanone		43, 86, 116, 73, 57	43	1,44	863	
269	unknown 2		98, 83, 69, 55	98	2,2	750	
252	unknown 8		45, 57	57	4,69	806	
234	unknown 9		133, 134, 54	133	3,5	1101	
221	dimethyl-trisulfide	fresh onion	126, 78, 45, 64	126	3,12	1320	cd
221	2-methoxy-4-vinylphenol	spicy, apple, rum	150, 135, 107, 77	150	4	980	d
217	2-hexenal	grassy [i]	41, 55, 69,83, 98	83	11,12	855	i
203 [h]	unknown 6		57	57	2,25	803	
194	3-carene / 1s-alpha-pinene	sweet, pungent, turpentine	93, 77, 121, 136	93	5,01	1016	
186	maltol	caramel, butterscotch	126, 71, 55, 97	97	14,72	1115	g
182	diethyl phthalate		149, 177	149	2,27	1595	

concentration relative to internal standard	compound	odour [a]	major ions	quantitation ion	factor (total ion count area/ quantitation ion area)	Linear Retention Index	previously reported odour active in peanuts [b]
176	unknown 7		68, 42, 111, 57, 87	68	7,58	803	
173	unknown 5		43, 55, 70, 83	70	14,64	791	
163	dihydro-4-methyl-3(2H) furanone		42, 56, 100, 70	100	22,34	961	
161	2,4 -decadienal	oily, orange	81, 67, 95	81	2,52	1324	dg
150	6-aminoindoline		134,107, 66	134	4,82	1206	
143	alpha-ethylidene-benzenacetaldehyde	green, floral, woody	146, 117, 115, 91	146	8,29	1279	
140 [h]	hydroxydihydromaltol	roasted [j]	43, 101, 55, 72, 144	144	6,95	1150	
136	3-methoxy-pyridine		109, 66, 79, 94	109	2,61	1001	
112	5-methyl-2-furancarboxaldehyde	sweet, spicy, caramel	110, 53, 81	110	3,49	963	
107	acetophenone	sweet, pungent, medicinal	105, 77, 51	105	3,34	1074	g
105	1-(methylthio)-propane		90, 61, 45	90	5,09	764	
98	unknown 13		121, 93, 65	93	3,17	1323	
96	1- (2-furanyl)-ethanone		95, 110	95	2,12	910	
96	2-pentanone	ethereal, fruity	43, 86, 58, 71	43	2,12	686	
94	5-methyl-2-furanmethanol	cooked sweet potato, honey	95, 112, 41	95	6,76	950	
91	propanoic acid, 2-oxo, methyl ester	caramel [f]	43, 102	102	15,85	728	
84	pantolactone		71, 43, 57	71	3,69	1043	
79	2,5 diethyl-pyrazine	nutty, hazelnut [f]	135, 136, 121, 108	135	4,97	1095	
70	1H-pyrrole-2-carboxaldehyde		95, 66, 39	95	3,89	1009	
65	benzenemethanimine		104, 105, 77, 51	104	3,08	1018	

concentration relative to internal standard	compound	odour [a]	major ions	quantitation ion	factor (total ion count area/ quantitation ion area)	Linear Retention Index	previously reported odour active in peanuts [b]
64	3-methylthio-propanal	strong onion, meat	48, 104, 61, 76	48	4,55	908	cd
63	2/3-methyl-1H-pyrrole		80, 81, 53	80	3,1	836	
56 [h]	1-nitro-hexane		43, 55, 73, 85	55	6,65	1046	
50	2,3 dimethyl-1H-pyrrole		94, 95	94	2,77	866	
49	phenol	sickeningly sweet, irritating	94, 66, 39, 55	94	2,97	974	

[a] All odour descriptions from Burdock, (Burdock, 2010) unless otherwise stated; [b] compounds found in the literature to be odour active in peanuts by means of GC-olfactometry; [c] (Chetschik et al., 2008); [d] (Chetschik et al., 2010); [e] (Didzbalis et al., 2004); [f] odour descriptions from (FAO, 2015); [g] (Schirack et al., 2006a) ; [h] compounds were not detected in over 15% of the sample set; [i] Odour description from (Neta et al., 2010); [j] Odour description from (Cutzach et al., 1997) TIC: Total Ion Count. Quantitation Ion: fragment ion used for integration.

In **Figure 11**, the PCA scores plot of the sensory profile is reported, where the loadings for the GC-MS data have also been superimposed as secondary variables. The first two components explained approximately 82% of the variance.

As expected, a correlation between certain attributes is apparent in **Figure 11**. Attributes associated with intense thermal treatment (dark roast, roasted peanut, sweet and ashy aromas) are clustered together in the left hemicycle (negative scores on the first component), diametrically opposing the raw beany aroma. Painty, an attribute not considered related to roasting (Sanders et al., 1989), is essentially independent as seen by the almost orthogonal direction with respect to the roast related attributes. The remaining attributes are not resolved well within the two principal components plotted, likely due to the limited variance in panellists scores (**Figure 10**).

A clustering of the volatile compounds was also visible in **Figure 11**. Most of the low molecular weight aldehydes and ketones are clustered on the top right quadrant, correlating well with the painty attribute. These chemicals are often associated with lipid oxidation pathways, which are also known to be associated with painty off flavours (St Angelo, 1996). The sensory attributes associated with dark roast, roasted peanut, sweet, raw beany and painty aromas, appear to correlate fairly well with several volatile compounds, found in the top left quadrant, largely pyrroles, pyrazines and furans. These appear to be negatively correlated with linear aldehydes and ketones (top right quadrant), providing

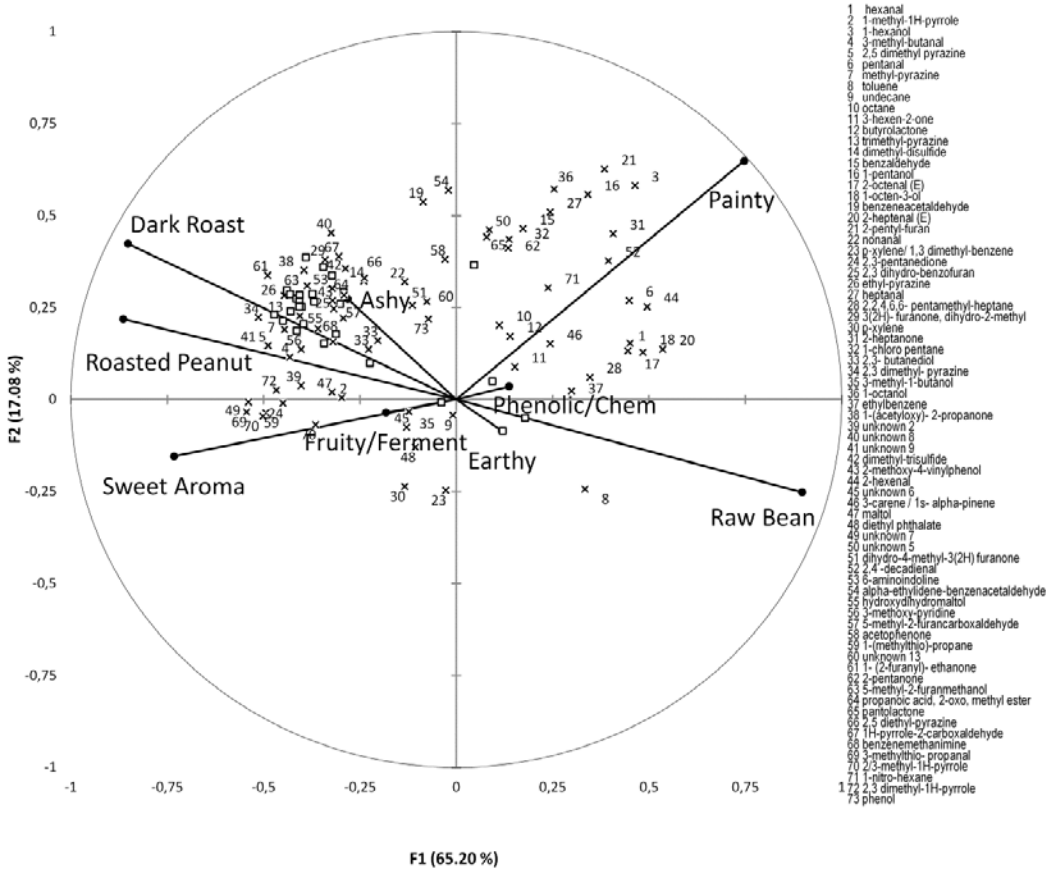

Figure 11. First and second component of the PCA scores and loadings plot of sensory scores and volatile content of processed peanuts. 82.3% of the variation explained by the two components. x: compounds included in **Table 9**. Squares: compounds excluded from **Table 9** due to low concentration or weak correlation to sensory attributes.

further supporting evidence for the hypothesis that these lipid oxidation by-products may react and deplete the compounds associated with roasted aromas (Williams et al., 2006), as will be described below.

3.3.2 Correlations between flavour attributes and volatile headspace compounds.

To better understand the link between sensory responses and volatile compound fingerprint, PLS Regressions were run. Four sensory attributes, namely roasted peanut, sweet, dark roast and raw bean aromas were modelled with

R^2=0.43, R^2=0.56, R^2=0.66 and R^2=0.70 respectively. The roasted peanut attribute had a poor R^2 coefficient, and the model should not be considered well-fitting, but the observations are nonetheless included here, as they are in good agreement with the literature (Baker et al., 2003; Braddock et al., 1995; Ho et al., 1981; Ho C.T. et al., 1983; Schirack et al., 2006b; Xiao et al., 2014). Better fitting models were obtained by transforming the concentrations as will be discussed below, but a brief discussion of non-transformed PLSR models is included to allow comparisons with other studies. In general, aldehydes correlated with reduced roasted/peanut attributes and increased raw/beany aromas (as also reported elsewhere (Didzbalis et al., 2004; Pattee, Beasley, & Singleton, 1965)), while the opposite is true for most pyrazines and pyrroles (**Figure 12**).

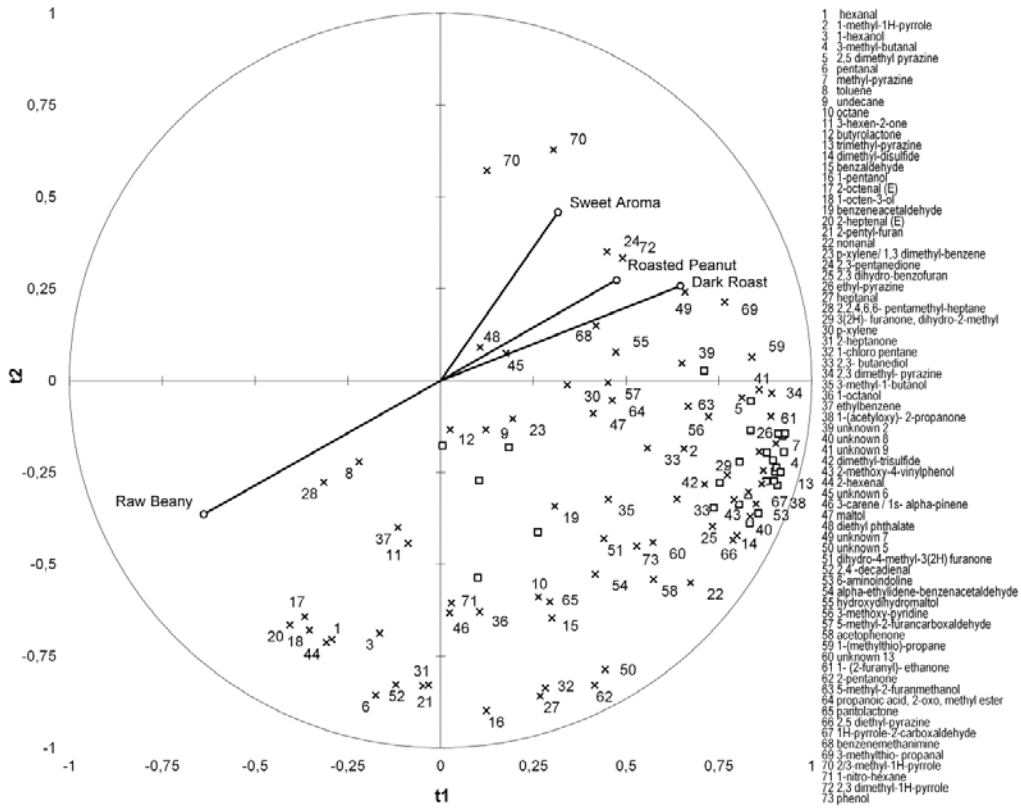

Figure 12. Two dimensional graphical representation of the correlations between key flavour attributes and volatile components, as determined by the PLS regression. Crosses: compounds included in **Table 9**. x: compounds excluded from **Table 9** due to low concentration or weak correlation to sensory attributes.

Toluene correlated negatively with the roasted peanut aroma. This compound was previously identified to have a negative effect to roasted and

peanut aromas, (Smith, Perry, Marshall, Yousef, & Barringer, 2014) but it appears to occur at a subthreshold level. Interestingly, maltol (butterscotch/caramel aroma (FAO, 2015)) correlates highly with roasted peanut aroma. This is the first time that such correlation is reported although maltol's occurrence in roasted peanuts has been known for decades (Ho et al., 1981). 2/3-methyl-1H-pyrrole (experimental setup could not confidently resolve if the peak was 2-methyl-1H-pyrrole or 3-methyl-1H-pyrrole) also correlates highly with roasted notes, as previously noted (Xiao et al., 2014). These two compounds also appear to be related to the sweet aroma attribute, even though 2,5-dimethylpyrazine is also significantly correlated (in agreement with previous research (Baker et al., 2003)). Looking at the sum of the 'peanut' and 'roasted' aromas, the most important positively correlated compounds appear to be 2/3-methyl-1H-pyrrole, 5-methyl-2-furancarboxaldehyde, benzeneacetaldehyde, 2,3 dimethyl-1H-pyrrole, 2,5 dimethyl pyrazine, 5-methyl-2-furanmethanol, and maltol, while toluene, ethylbenzene, octane, benzaldehyde and butyrolactone are negatively correlated.

3.3.3 Correlations between flavour attributes and volatile headspace compounds: improved fit.

In addition to the linear PLSR, we also evaluated a logarithmic transformation on the DHS-GC-MS data (biplot shown in **Figure 13**) and the standardized coefficients of the models for raw beany, dark roast, sweet and roasted peanut aromas are shown in **Figure 14** (only compounds with a standardized coefficient larger than 0.05 in absolute value are shown, for clarity) **.** The standardized model coefficients, also known as β coefficients, are a measure of the relative weight of each variable in the model. Being relative, the standardized coefficients are preferred to comparing the unstandardized model coefficients, which are not scaled. By applying the logarithmic transformation, the R^2 value for the four models (prediction of roasted peanut, sweet, dark roast and raw bean aromas) increased by approximately 50%: from R^2=0.43, R^2=0.56, R^2=0.66 and R^2=0.70 to R^2=0.69, R^2= 0.71, R^2=0.83, and R^2=0.84 respectively. The high R^2 coefficients of the derived models suggest that the transformation was a good choice.

Transforming the data is acceptable as dynamic headspace analysis is a method that does not pre-concentrate head space volatiles, and can therefore provide meaningful quantitation data on concentration , even if advanced quantitation methods such as isotope labelling is not employed (Snow & Slack, 2002). The use of a logarithmic transformation specifically, can also be justified: The increased weight the logarithmic transformation places on compounds found

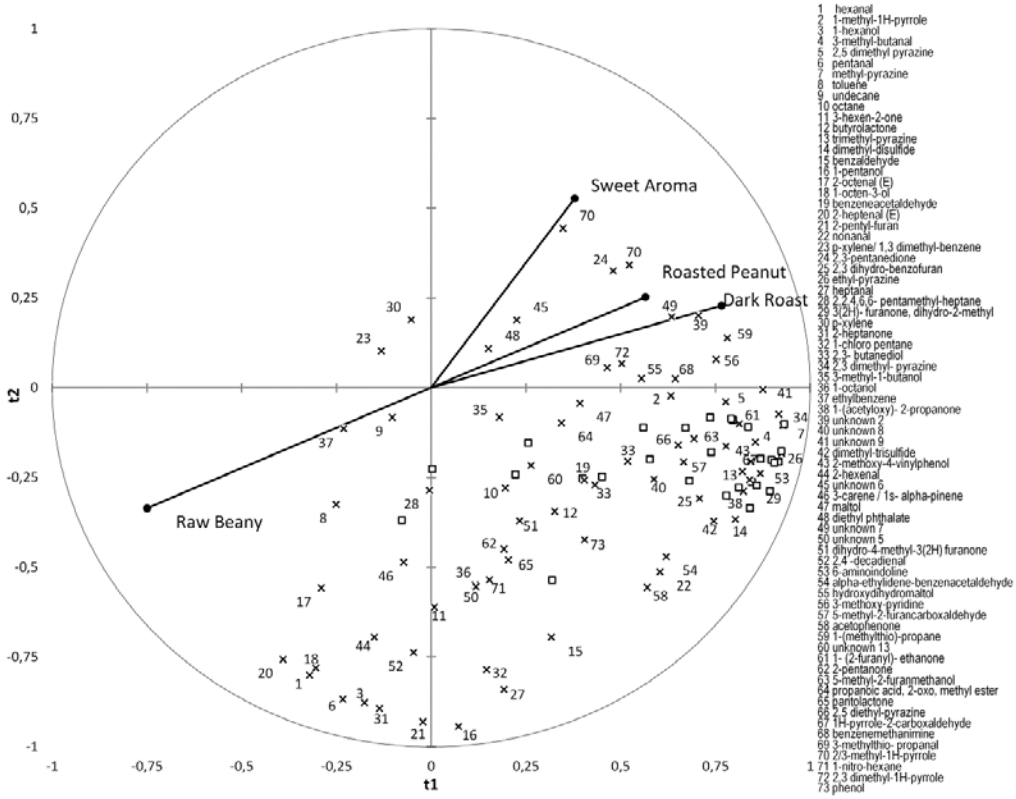

Figure 13. Two dimensional graphical representation of the correlations between key flavour attributes and log concentration of volatile components, as determined by the PLS regression. x: compounds included in **Table 9**. Squares: compounds excluded from **Table 9** due to low concentration or weak correlation to sensory attributes.

in lower concentration can be advantageous, as not all compounds have the same odour threshold: mono, di and trimethylpyrazines often have fairly high odour thresholds, whereas substituting methyl with ethyl groups greatly reduces this threshold (Liu et al., 2011). The logarithmic transformation was preferred over linear or other transformations (such as the auto scaling built into the covariance method of PCA/PLSR) that could also increase the weight of compounds occurring at lower concentration (Chambers & Koppel, 2013). The reason for this is that odour thresholds commonly relate to the partition behaviour of volatile compounds which is described by the logarithm of the n-octanol/water partition coefficient (Abraham, Gola, Cometto-Muniz, & Cain, 2002).

Interestingly, even though in the transformed PLSR models the compounds identified in the linear PLSR models are mostly still significant (and specifically 2,5-dimethylpyrazine), the relative value of their standardized coefficients significantly

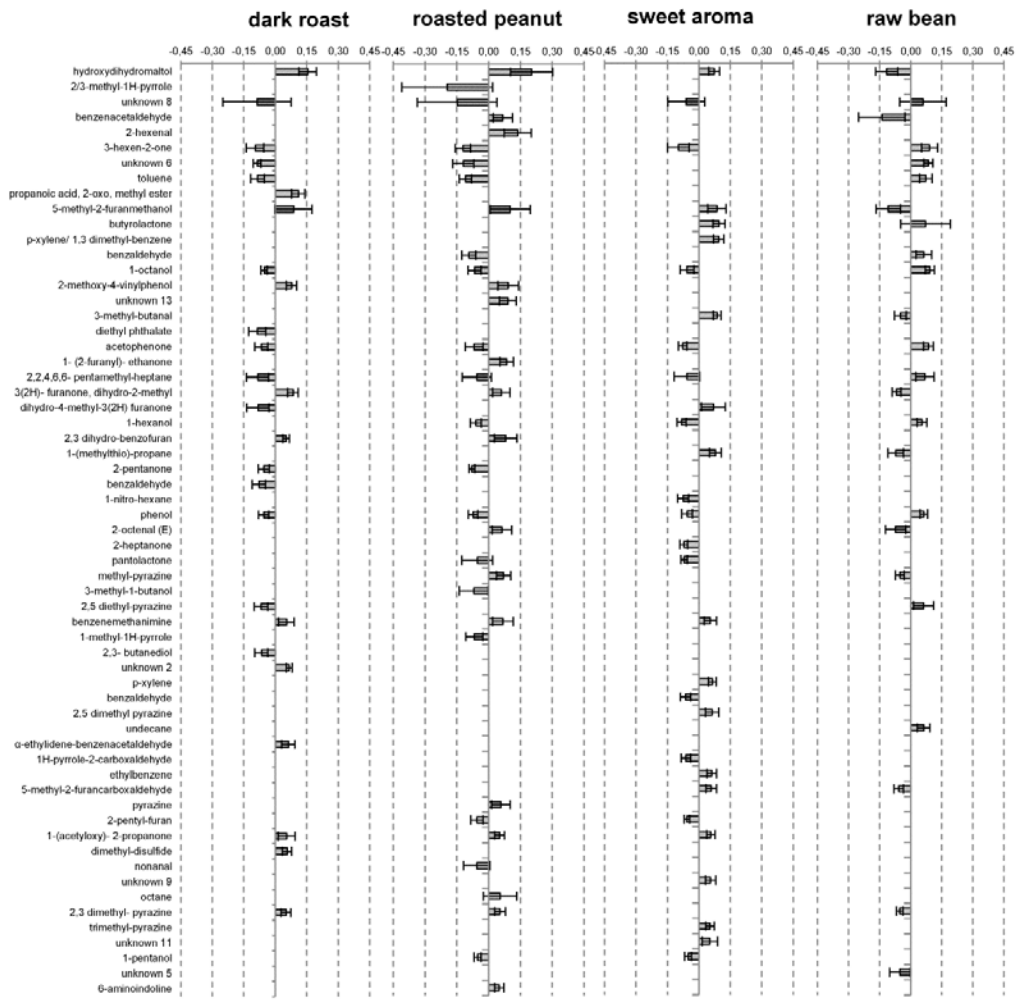

Figure 14. Standardized coefficients (with standard deviations) of volatile compounds quantified by DHS-GC-MS in the PLS model developed for the dark roast, roasted peanut, sweet and raw bean aromas. Only coefficients larger than 0.05 are shown.

dropped. This could be due to pyrazines contributing less to the sensory profile at higher concentrations, due to their ability to easily bring on adaptation to panellists (highly volatile and persist in nasal mucosa (Linforth, Baek, & Taylor, 1999)), given their highly polar nature and relatively low volatility (quickly dissolve and remain dissolved on mucous membranes in the nasal cavity), or due to the low odour activity of some of the abundant compounds (2,5-dimethylpyrazine, 2,3-dimethylpyrazine, 2-ethyl-3-methylpyrazine and methylpyrazine (Chetschik et al., 2008, 2010)). When the logarithmic transformation was applied, 3,5-dihydroxy-6-methyl-2,3-dihydropyran-4-one (Hydroxydihydromaltol) appears to be highly

correlated to all four attributes (roasted peanut, dark roast, sweet and raw bean aromas). This has not been observed in the surveyed published literature.

For the 'raw bean' attribute, most ketones and aldehydes were positively correlated, with benzeneacetaldehyde ('harsh green' (Burdock, 2010)) hydroxydihydromaltol (3,5-dihydroxy-6-methyl-2,3-dihydropyran-4-one, 'roasted aroma' (Cutzach, Chatonnet, Henry, & Dubourdieu, 1997)) and 5-methyl-2-furanmethanol (cooked sweet potato/honey (Hui & Sinha, 2011)) having the largest negative coefficients and octanol ('fruity/orange' (Burdock, 2010)) and 3-hexen-2-one ('metallic/acrid' (FAO, 2015)) having the largest positive coefficients. For the 'sweet aroma' attribute, most ketones were negatively correlated, 3-hexen-2-one and hexanol (odourless) had the greatest negative impact, whereas butyrolactone ('sweet, caramel' (Burdock, 2010)) p-xylene/ 1,3 dimethyl-benzene and 3-methyl-butanal ('malty' (Burdock, 2010)) had the greatest positive impact.

For the 'dark roast aroma' the largest positive coefficients were for hydroxydihydromaltol (3,5-dihydroxy-6-methyl-2,3-dihydropyran-4-one, 'roasted aroma'), 2-oxo-propanoic acid , methyl ester ('caramel' (FAO, 2015)) and 5-methyl-2-furanmethanol ('cooked sweet potato/honey' (Hui & Sinha, 2011)) whereas the largest negative coefficients were for 3-hexen-2-one diethyl phthalate and toluene. Finally, for 'roasted peanut' aroma there was a surprising negative correlation with a pyrrole (2/3-methyl-1H-pyrrole), even though most pyrroles have roasted odours (Neta et al., 2010). A possible explanation is that this was either a degradation or a lipid oxidation intermediate by-product, and therefore correlated with other off-flavours (regardless if it's actual aroma). Another interesting observation is the positive correlation of 2-hexenal ('grassy' (FAO, 2015)), although low molecular weight aldehydes have been previously seen to be related to peanut flavour (Da Conceicao Neta, 2010). 2-hexanal is a lipid oxidation by-product and it is likely that as roasting causes some degree of lipid oxidation, it is indeed correlated with roasted peanut aroma at low levels (which in these models are weighed higher due to the transformation). 5-methyl-2-furanmethanol ('cooked sweet potato/honey' (Hui & Sinha, 2011)) is also strongly correlated with this attribute, while hydroxydihydromaltol had again very significant positive correlation.

3.3.4 Aroma active compounds versus aroma fingerprint – compound correlations.

A comparison between the type of results obtained by aroma active compound characterization studies (see for instance the studies by Chetschik and co-workers (Chetschik et al., 2008, 2010) utilizing GC-O or some type of aroma dilution analysis) and PLS regression results is warranted. Several findings are in line with those studies, however some are not. These inconsistencies can be

explained in two ways: Firstly, aroma characterization studies focus on demonstrating causality of general flavour active compounds, whereas a correlation study searches for the connection between the volatile fingerprint and *specific* flavour attributes (Chambers & Koppel, 2013), and a discussion on how the two approaches account synergistic and antagonistic effects was given earlier It is possible therefore, that some compounds may indeed be responsible for the character of roasted peanut meal, but not necessarily correlate with specific sensory attributes such as 'sweet aroma' as sweet aroma does not necessarily have a peanut character. The importance of these compounds therefore, would not be evident in this study and vice versa, false positives may be present. In other words, correlation does not necessarily imply causation. However, information on correlations (positive or negative) can be very valuable to manufacturers: the presence of a compound may not directly cause an increase or a decrease to the intensity of a flavour attribute, but if the correlation is known it can be a useful quality diagnostic.

We propose that the correlation data is complementary to the definitive identification of the aroma active compounds when it comes to distinguishing between flavour attributes or understanding antagonistic effects as discussed below.

Secondly, due to the laborious nature of reconstitution studies, a very limited number of varieties or samples can be analysed. Our research instead focused on breadth by looking at several material-process combinations and in this way compounds relevant to specific varieties or processes can be detected (this is widely discussed in the complementary part of the present research). This can also be observed in **Table 9**, where several compounds (marked with '^{C_7}') abundant in some samples, where not detected at all in over 15% of the sample set. In other words, different raw materials and processes can develop unique compounds which can be responsible for variety or process specific aromas. With regards to raw materials, Aprea et al noticed a similar effect in raspberries (Aprea, Biasioli, & Flavia Gasperi, 2015), where large qualitative and quantitative differences in the odour active compound profile was observed in different varieties, crop years, and post-harvest treatments. In peanuts, large organoleptic and volatile profile differences have also been observed due to processing (microwaving) (Schirack et al., 2006b).

Finally, specific compounds may be very important for specific samples, but the effect may be diluted by larger differences or similarities between different peanut market types and processing technologies, when one is analysing a larger sample set.

3.3.5 Compounds negatively correlated to flavour attributes.

In both the linear and transformed PLSR models (**Figure 14**) several compounds were negatively correlated to flavour attributes. This could be explained in three ways. Firstly, compounds could exhibit receptor antagonism and thus the mere presence of some may decrease the odour potency of others as in the examples given above (R. S. T. Linforth & Taylor, 2010). Secondly, it is plausible that strong 'green' aromas overpower the roasted notes and change the overall aroma balance, resulting in the panellists not recognizing the sum as a 'roasted' flavour, even though the individual components can still be detected. Indeed, a similar case has been reported, (Civille & Dus, 1992) where oxidized and non-oxidized peanut pastes from the same raw material were seen to organoleptically differ in roasted peanut and sweet aromas, in addition to the cardboard and painty attributes as one would expect from oxidation alone. Warner and co-workers suggested that low molecular weight aldehydes formed during lipid oxidation are particularly prone to cause this effect due to their pungent aroma (Warner et al., 1996).

Finally, it is possible that certain compounds are negatively correlated with specific sensory attributes because their presence is related to a chemical reaction in the sample which is responsible for creating or depleting other aroma active compounds. It has been proposed that lipid radicals and hydroperoxides that are formed during lipid oxidation, can degrade heterocyclic nitrogen compounds, (Williams et al., 2006) which are in turn known to be associated with roasted flavours (Liu et al., 2011; St Angelo, 1996). As a result, presence of free radicals or fragments of heterolytic nitrogen compounds, for example, would be identified as negatively correlated to a specific flavour attribute, even if themselves are odourless. Analogously if a chemical compound is a precursor of an odour-active compound, it may be negatively correlated with that attribute. This information is particularly useful in quality monitoring, where the chemical fingerprint of a 'good' or 'bad' sensory attribute provides more information than identifying the odour active compounds.

3.3.6 Colour as a predictor for roasting level.

A PCA was run on the sensory profiles with the colour and DHS-GC-MS data superimposed. The analysis was performed on a reduced sample set comprising only of dry roasted and fried samples (Plackett-Burman design plus frying). The samples represent a wide range of peanut types, varieties and origins, as well as roasting conditions. Other samples were excluded, because they had been specifically created to disrupt the colour - flavour relationship, by manipulating the

Maillard reaction kinetics (by changing for example the pH or reducing sugars concentration). The loadings plot can be seen in **Figure 15**, where some correlations are immediately obvious: L* value is inversely correlated to roasted attributes and positively correlated to the raw bean aroma, as one would expect (low L* values are associated to dark colours).

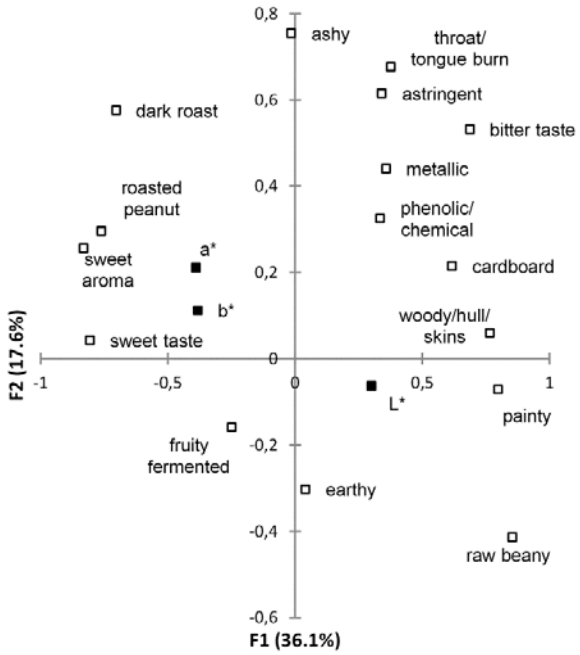

Figure 15. Principal Component Analysis of sensory profiles (squares) with average colour values superimposed (x) for the reduced data set (fried and roasted processes only). 53.7% of the variation explained by the two components.

This correlation is also apparent in the Pearson's correlation (similarity) matrix (**Table 10**). Interestingly, even though highly significant ($P<0.05$), the correlation coefficients for L*, a* and b* values against 'roasted peanut' are -0.44, 0.55 and 0.43 respectively when more than one varieties/processes are taken into consideration. In addition, the strong correlation between 2,5-dimethylpyrazine and L* colour value (light-dark) that has been previously reported when only a limited pool of raw materials was investigated (Baker et al., 2003), is still significant but with a magnitude of only 0.52. In addition, a* and b* also appear to be highly correlated to 2,5-dimethylpyrazine (Pearson's coefficients = 0.60 and 0.32), and in fact, the a* value has an even higher coefficient than the L* value.

Table 10. Pearson's correlation matrix for the reduced data set (fried and roasted processes only)., filtered for correlations with Pearson's correlation coefficient ≥0.4 (coefficients ≥0.65 have been highlighted in bold). Only values that are different from 0 with a significance level alpha=0.05 are listed.

Variables	roasted peanut	sweet aroma	dark roast	raw beany	colour L*	colour a*	colour b*
roasted peanut	**1**	**0.653**	**0.766**	**-0.761**	-0.441	0.552	0.426
sweet aroma	**0.653**	**1**	**0.732**	**-0.761**		0.350	
dark roast	**0.766**	**0.732**	**1**	**-0.878**	-0.370	0.506	0.282
raw beany	**-0.761**	**-0.761**	**-0.878**	**1**	0.381	-0.511	-0.424
woody/hull/skins	-0.546	-0.519	-0.393	0.589		-0.341	-0.366
painty	-0.594	-0.573	-0.506	**0.701**			-0.498
sweet taste	0.438	**0.823**	0.536	**-0.651**			
bitter taste	-0.374	-0.492		0.347	0.262	-0.209	
ashy			0.449	-0.387			0.275
colour L*	-0.441		-0.370	0.381	**1**	**-0.922**	-0.346
colour a*	0.552	0.350	0.506	-0.511	**-0.922**	**1**	0.532
colour b*	0.426		0.282	-0.424	-0.346	0.532	**1**
3-methyl-butanal	0.542	0.613	0.561	-0.577	-0.315	0.355	
1-hydroxy-2-propanone	0.493	0.429	0.554	-0.543	-0.442	0.533	0.312
2-methyl-butanal	0.535	0.560	**0.659**	-0.592	-0.450	0.501	
2-pentanone		-0.300		0.328			-0.444
2,3-pentanedione	0.494	**0.683**	0.583	**-0.660**		0.288	0.351
pentanal	-0.326	-0.347	-0.306	0.423			-0.551
3-hydroxy-2-butanone	0.533	0.394	0.565	-0.560	-0.568	**0.663**	0.465
pyrazine	0.568	0.421	0.639	-0.619	-0.528	0.638	0.461
1-methyl-1H-pyrrole			0.525	-0.447			
2-methyl-2-butenal (E)	0.549	0.466	**0.688**	-0.633	-0.486	0.544	
dimethyl-disulfide	0.301	0.282	0.552	-0.394		0.312	
unknown 2		0.320	0.568	-0.510		0.314	0.411
pyrrole	0.474	0.384	**0.701**	-0.621	-0.370	0.484	0.320
1-chloro pentane							-0.409
4,5 dimethyloxazole	0.454	0.339	**0.691**	-0.632	-0.430	0.556	0.406
1-(methylthio)-propane	0.509	0.600	0.505	-0.598	-0.604	**0.654**	0.359
1-pentanol	-0.282	-0.312		0.428			-0.455
octane		-0.411	-0.314	0.394			-0.439
hexanal		-0.302	-0.281	0.389			-0.481
unknown 7	0.499	0.448	**0.715**	**-0.700**	-0.422	0.540	0.499
unknown 8	0.543	0.465	0.598	-0.541	-0.571	0.646	0.283
3(2H)- furanone, dihydro-2-methyl	0.556	0.566	0.607	-0.565	-0.546	0.632	
methyl-pyrazine	0.635	0.566	**0.736**	**-0.713**	-0.505	0.610	0.341
furfural	0.501	0.385	0.539	-0.556	-0.627	**0.691**	0.487
2/3-methyl-1H-pyrrole	0.450	0.520	0.644	-0.638	-0.357	0.423	0.370
2-furanmethanol	0.497	0.461	0.606	-0.507	-0.344	0.430	

Variables	roasted peanut	sweet aroma	dark roast	raw beany	colour L*	colour a*	colour b*
2-hexenal	-0.417	-0.430	-0.390	0.501			-0.564
1-(acetyloxy)- 2-propanone	0.593	0.581	0.573	-0.538	-0.527	0.587	
ethylbenzene	-0.402			0.323	0.445	-0.469	-0.414
2,3 dimethyl-1H-pyrrole	0.298	0.299	0.636	-0.567		0.328	0.337
1-hexanol	-0.349	-0.269		0.477			-0.366
p-xylene/ 1,3 dimethyl-benzene					0.397	-0.406	-0.305
2-heptanone		-0.342		0.392			-0.435
styrene	0.303				-0.626	**0.658**	0.486
heptanal		-0.267		0.355			-0.467
2(5H) furanone	0.531	0.444	0.554	-0.472	-0.363	0.411	
3-methylthio- propanal	0.554	0.646	0.470	-0.583	-0.462	0.537	0.345
1- (2-furanyl)- ethanone	0.617	0.504	**0.651**	-0.592	-0.423	0.513	
2,5 dimethyl pyrazine	0.590	0.547	**0.666**	-0.642	-0.520	0.595	0.324
ethyl-pyrazine	0.604	0.506	**0.651**	-0.607	-0.569	0.624	
2,3 dimethyl- pyrazine	0.598	0.519	**0.712**	**-0.664**	-0.498	0.562	
ethenyl-pyrazine	0.541	0.472	0.618	-0.522	-0.424	0.481	
5-methyl-2-furanmethanol	0.503	0.559	0.463	-0.491	-0.343	0.383	
2-heptenal	-0.396	-0.409	-0.375	0.478			-0.547
dihydro-4-methyl-3(2H) furanone							-0.419
1-octen-3-ol	-0.342	-0.309	-0.299	0.409			-0.471
2-pentyl-furan	-0.339			0.408			-0.476
2-ethyl-6-methyl-pyrazine	0.298	0.295	0.409	-0.362			
3-methoxy-pyridine		0.295	0.489	-0.379			
trimethyl-pyrazine	0.469	0.442	0.575	-0.502	-0.312	0.374	
2-ethyl-5/6-methyl pyrazine	0.578	0.476	0.636	-0.561	-0.498	0.559	
1H-pyrrole-2-carboxaldehyde	0.355		0.537	-0.364			
benzenemethanimine	0.456	0.557	0.497	-0.493	-0.352	0.417	0.268
2-ethenyl-6-methyl-pyrazine	0.524	0.487	0.608	-0.509	-0.320	0.377	
2-ethenyl-5-methyl pyrazine	0.558	0.517	0.598	-0.544	-0.474	0.514	
2-octenal	-0.321	-0.331	-0.295	0.366			-0.463
1-(1H-pyrrole-2-yl)-ethanone	0.458	0.376	0.466	-0.366			
1-octanol	-0.264			0.444			-0.407
3-ethyl-2,5 dimethyl-pyrazine	0.574	0.544	**0.695**	-0.622	-0.389	0.468	
2,6-diethyl-pyrazine	0.539	0.472	0.615	-0.517	-0.330	0.395	
2-ethyl-3,5 dimethyl-pyrazine	0.576	0.470	0.638	-0.531	-0.353	0.432	
2,3 dimethyl- 5-ethyl-pyrazine	0.582	0.468	0.645	-0.543	-0.405	0.478	
2,5 diethyl-pyrazine	0.402	0.275	0.373	-0.260	-0.306	0.343	
unknown 9	0.554	0.550	**0.665**	-0.617	-0.383	0.474	0.310
maltol	0.371	0.279	0.362	-0.338	-0.404	0.443	
hydroxydihydromaltol	0.442	0.526	0.430	-0.458	-0.337	0.419	
2,3-diethyl-5-methyl-pyrazine	0.547	0.468	0.602	-0.512	-0.345	0.411	
3,5-diethyl-2-methyl-pyrazine	0.544	0.469	0.594	-0.501	-0.299	0.372	
6-aminoindoline	0.504	0.371	0.626	-0.470	-0.323	0.404	

Variables	roasted peanut	sweet aroma	dark roast	raw beany	colour L*	colour a*	colour b*
2,3 dihydro-benzofuran	0.457	0.317	0.454	-0.366	-0.286	0.340	
alpha-ethylidene-benzenacetaldehyde	0.347		0.450	-0.265	-0.276	0.337	
2-methoxy-4-vinylphenol	0.619	0.375	**0.666**	-0.588	-0.632	**0.710**	0.391

Several researchers have reported correlations between colour and flavour when it comes to roasted peanuts (Lee & Resurreccion, 2006; McDaniel et al., 2012; Pattee et al., 1991; Schirack et al., 2006b). Some have explained the relationship on the basis of colour development in peanuts to be mainly due to Maillard reaction, and only partly by caramelization (Pattee et al., 1991), while Maillard reaction is also the major contributor to roasted peanut flavour (Schirack et al., 2006b). Given the common pathway therefore, a relationship between colour and flavour is expected.

McDaniel and co-workers found that this relationship is disrupted when a large roasting temperature range (40 °C range) is investigated: differences in quality attributes (e.g. antioxidant activity and free sugars) and flavour were observed even if the peanuts are roasted to the same final surface colour (McDaniel et al., 2011, 2012). It is clear therefore, that there are some limits to the range over which the relationship holds, and in this study the limits were set to be as relevant as possible to industrial peanut processors: as many raw materials as possible, several common processing methods, and fairly narrow roasting temperature range (20 °C range). In this setup, we also observed that keeping the final moisture content constant is not enough to ensure a constant roasted aroma development (McDaniel et al., 2011, 2012).

Although colour parameters can be a particularly convenient quality control tool for controlling roasting level in a production environment (for instance, measurement of colour is quicker and less expensive than measurement of 2,5-dimethylpyrazine by GC-MS), care should be taken before extrapolating across all raw materials. Colour parameters correlation with 'roasted peanut' aroma is particularly interesting, as this attribute has been seen to correlate well with consumer preference (Sanders et al., 1989). Smith and co-workers, studied the relationship between colour and roasting method (microwave, oven and combination of the two), and concluded that peanuts roasted to the same L* value by different methods, showed no significant differences in most sensory attributes (Smith et al., 2014). In our study, using various raw materials but only two processes, the relationship between L* and 'roasted peanut' was also significant, but weaker. We concluded that the colour – flavour relationship mostly depends on the chemical composition of the material, and only to a small degree on the processing conditions. In other words, although colour is an excellent indicator of roasting level, it may not be as sensitive in resolving small differences as a

chromatographic technique, and should therefore be used for quality control in setups with limited variation in raw materials and process conditions, rather than as an absolute measure of roasting level (Smyth et al., 1998).

3.3.7 Correlations between off - flavours and volatile headspace compounds.

An effort was also made to develop PLSR models for the rest of the sensory attributes, but it was unsuccessful (generally R^2<0.35, most attributes R^2<0.1). We have three hypotheses to explain why modelling 'defect' sensory attributes was not successful: Firstly, even though 134 samples were analysed, there were still not enough exemplars of off-flavours in the sample set, and therefore the signal to noise ratio was too small for the statistics to resolve. This can be expected, as there was no effort to include purposefully defective or aged samples in the sample set.

Secondly, since the compounds often associated with solvent-type aromas have a very low threshold, it is possible that they were detected by the panellists (in the case of 'painty' aroma for example) but not by the specific DHS-GC-MS setup. For this reason, for future research the authors recommend a pre-concentration method, such as the use of Solid Phase Micro Extraction (SPME) rather than a dynamic head space analysis, or the incorporation of additional sample preparation steps. Indeed, some success in identifying compounds responsible for flavour defects using a pre-methylation step and SAFE has been reported (Didzbalis et al., 2004; Nepote, Olmedo, Mestrallet, & Grosso, 2009). Alternatively, GC-O techniques can also be leveraged, as the human nose is often still more sensitive for certain compounds than MS or other detectors.

Thirdly, off-flavours ('fruity/fermented' in particular) are often generated by individual abused kernels, and not uniformly produced throughout the sample. For this reason the use of samples larger than 300 grams when analysing for off flavours, to account for the unusually high sampling error has been proposed (Whitaker, Slate, Greene, Hendrix, & Sanders, 2007).

3.4 Conclusions

The purpose of this study was to identify compounds correlated to the sensory attributes in a large variety of samples and processes. Analysing 134 samples, rather than focusing on a handful, provided interesting insights to the peanut category as a whole, and the conclusions are applicable to several raw

materials and process technologies. This expanded scope however, does not come without trade-offs in statistical power, as in order to keep the number of samples manageable several factors (growing area, crop year, variety etc.) cannot be fully accounted for by the use of a full factorial design. We chose to indeed sacrifice depth over breadth, however, for two reasons. Firstly, a number of high quality published studies focusing on depth already exist. Secondly, even though the peanut category is highly commoditized and different lots, varieties, and origins are often blended, little research has been made on the 'average peanut' flavour. For instance, an effect that is statistically significant but of small magnitude is of little interest to the producers and growers, as it will be masked by other, larger effects when peanut lots or crops are blended. Information on 'average peanut flavour' can be used to help guide future researchers in which areas to focus.

This research analysed the relationship of four flavour sensory attributes ('roasted peanut', 'sweet', 'dark roast' and 'raw bean' aromas) to headspace volatile compounds through PLS regression. It was shown that although the technique is most commonly applied to untransformed concentration data, the model quality could be significantly improved by a logarithmic transformation. Several compounds commonly reported in high concentration (such as 1-methyl-1H-pyrrol and 2,5-dimethylpyrazine) were also found in abundance but their correlation coefficients were relatively low (as for several pyrazines). It was found that several compounds are highly correlated to more than one sensory attribute. Five of the most significant compounds across all model attributes included hydroxydihydromaltol (not previously reported in peanuts), 2/3-methyl-1H-pyrrole, benzeneacetaldehyde, 2-hexenal and 3-hexen-2-one, but the correlation was not always positive. The negative correlations where mostly between aldehydes and ketones against aromatics associated with roasting ('dark roast', 'roasted peanut', 'sweet aroma' and furans and pyrroles against 'raw bean' aroma), and have not been previously reported. Finally, the widely reported correlation between L* colour value and roasted aroma was shown to be significantly weaker across a range of raw materials and processes, suggesting that although colour is an excellent quality control tool for a production environment, the relationship is highly dependent on the specific raw material and process.

The next logical step for the future is to use recombination studies to definitively prove the positive or antagonistic effect on flavour sensory attributes of the compounds short listed here. In addition, improved analytical techniques such as two dimensional gas chromatography that use consecutive separation columns, coupled with advanced fingerprinting tools such as sensomics promise exciting prospects in both reducing the detection thresholds and in better identifying candidate compounds, making the recombination studies more focused and easier to undertake (Cordero et al., 2010; Cordero, Kiefl, Schieberle, Reichenbach, & Bicchi, 2015; Kiefl et al., 2012).

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4 A comprehensive look at the effect of processing on peanut (Arachis spp) texture.

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Abstract

Relationships between process and peanut texture have been only studied in *Hypogaea* species, and focused on very limited processing conditions. In this study, 94 samples were prepared from a combination of 12 raw materials (*Arachis Hypogaea* and *Fastigiata* cultivars) and 11 roasting conditions (maceration in water, aqueous glucose and at different pH followed by frying or baking). Texture was analysed by a trained sensory panel (Spectrum method) and large deformation compression tests (TA/XT2), and the microstructure probed with confocal microscopy and X-ray tomography. The impact of maceration on 'crispy', 'crunchy' and 'hardness' sensory attributes was significantly larger adding glucose in this step, while the effect of pH was minor. The relationship held for both fried and baked peanuts as well as for both *A. Hypogaea* and *Fastigiata* subspecies. The degree of alveolation was similar in differently processed peanuts, even though sensory attributes were significantly different. Maceration in different media can yield large textural changes in both peanut species, for both baking and frying. Maceration in glucose solutions can induce much larger textural changes than maceration in water. Quantitative data on alveolation show that microstructure disruption through steam generation cannot explain all the texture differences among processed peanuts.

Keywords: maceration, peanut, microstructure, texture, melanoidins, X-ray tomography

Highlights:

- Maceration prior to roasting increases crunchiness regardless of variety and roasting method.
- Maceration medium pH has small effect on texture development, but addition of glucose has a large effect.
- Microstructure disruption through steam generation only partly contributes to texture changes.
- Maillard reaction products may contribute to textural changes (mainly hardness but potentially crunchiness) during roasting.
- The 'blister fry' process also functions on Runner and Spanish (Arachis Fastigiata) type peanuts.
- Several instrumental attributes are simultaneously needed to model sensory texture attributes.

4.1 Introduction

Peanut is an important world crop widely cultivated for edible oil production and human consumption (Woodroof, 1983). It can be consumed in many forms, but whole roasted peanuts are a significant segment of the world market (USDA, 2015a). Texture, together with flavour, are the most important drivers of consumer liking for roasted peanuts (Lee & Resurreccion, 2006; Miyagi & Ogaki, 2014).

Past studies on the texture of peanuts have mainly focused on the impact of the raw material, including varietal and maturity differences (Pattee, Beasley, & Singleton, 1965; Young, Sanders, Drake, Osborne, & Civille, 2005). For over sixty years effort has been made to determine which raw materials deliver the preferred texture to the finished product and which should be avoided, even though a restrictive raw material procurement strategy can lead to increased costs (prevents sourcing from open market) and more waste (more materials rejected). The development of texture during processing with regards to microstructure changes has also been studied with qualitative electron microscopy techniques (Young, Pattee, Schadel, & Sanders, 2004; Young & Schadel, 1993), with most studies concluding that crunchiness and crispiness increase during roasting solely due to the disruption of the microstructure, caused by the generation of steam upon heating. Others(Nader, Afif, & Louka, 2016) have also demonstrated that water absorption during maceration can lead to a crispier texture due to larger amount of steam generated. Most available research is focused on contrasting raw versus roasted peanuts, rather than the difference between process conditions (Davis et al., 2010). One study has previously compared the impact of baking vs frying on microstructure (Young & Schadel, 1993), but no connection to texture was made. The study concluded that similar type of microstructure disruption occurs during frying and baking, albeit faster during frying due to the higher heat transfer rates. A study comparing roasting profiles using pulsed infrared radiation and conventional roasting providing the same final colour has also been published (Kumar, Debnath, & Hebbar, 2009), but the finished product moisture content was not controlled.

Recently, a robust comparison of dry roasting, oil roasting and ‘blister frying’ has been published for one specific cultivar (Jumbo Georgia O6G) (Shi et al., 2017). Blister frying was defined as pre-boiling in water (to increase the moisture content of the kernels) followed by frying, yielding a very crunchy product with a blistered appearance, popular in the southern United States. The study focused on flavour development and oxidative stability, but the texture was also qualitatively investigated by scanning electron microscopy (SEM). The results agree with prior literature, specifically that texture development is attributed to microstructure disruption due to steam generation (Shi et al., 2017).

All identified texture studies were run on Runner or Virginia peanuts (*Arachis Hypogaea*) but none on *Arachis Fastigiata* (Valencia or Spanish types). Similarly, all studies that included maceration have been done with water at various time-temperature combinations followed by frying, but there has been no account of the effect of the pH of the macerating solution or its sugar (glucose) content, nor of the effect of maceration when followed by baking for roasting. Finally, there has not been any published quantitative description of the degree of alveolation, as microstructure has only been qualitatively described by electron micrographs.

Our group recently published a study(Lykomitros, Fogliano, & Capuano, 2016a, 2016b) on the flavour development and correlation with sensorial characteristics in a large dataset of different varieties of peanuts processed in different ways. In this paper the primary objective was to study the impact of raw material and process conditions on the texture sensory attributes of peanuts. The aim was to identify processing parameters that can be adapted by peanut roasters to adjust the texture of a product from a given raw material, focusing specifically on non-common treatments, such as macerations in different media and pH followed by baking and frying. The impact of the raw material was also of interest, and the processes were applied to several peanut cultivars of both *A. Hypogaea* and *Fastigiata* species. To this purpose 12 raw peanuts were processed with a variety of technologies and conditions, resulting in 94 unique samples. The microstructure of a selected sample subset was investigated by confocal microscopy and X-ray tomography, in order to complement information from published scanning and transmission electron micrographs.

4.2 Materials and Methods

4.2.1 Peanut samples

Twelve different raw peanuts were sourced (Canon Garth Ltd, London, UK), including different market types (Valencia, Virginia and Runner), origins (China, USA, Australia, Argentina) and grades (Small, Medium, Extra Large, Jumbo) (**Table 11**). The peanuts were dry blanched at commercial scale (Steinweg-Handelsveem BV, Oosterhout, NL) by heating at 85°C for 30 minutes and subjecting to mechanical abrasion to remove the testa. The blanched peanuts were subsequently stored at -15°C until further processed and analysed, generally in less than 6 months.

Table 11. Overview of processing methods and raw materials used. Roasting times varied so that final moisture content was approximately 2 g $100g^{-1}$.

process code	Applied to material	key process parameters
α	A,C,E,F,H,J,L	macerated in potable water at pH 4 (acetic acid), 30 min at 20 °C, roasted at 145 °C
β	A,C,E,F,H,J,L	macerated in potable water, 90 min at 20 °C, roasted at 145 °C
γ	A,C,E,F,H,J,L	macerated in potable water with 2.5% w/w glucose, 30 min at 20 °C, roasted at 135 °C
δ	A,B,C,D,E,F,G,H,I,J,K,L	baked continuous impingement oven 135 °C
ε	A,B,C,D,E,F,G,H,I,J,K,L	baked continuous impingement oven 155 °C
ζ	A,B,C,D,E,F,G,H,I,J,K,L	baked continuous impingement oven 155 °C /135 °C
η	A,B,C,D,E,F,G,H,I,J,K,L	baked continuous impingement oven 135 °C /155 °C
θ	A,B,C,D,E,F,G,H,I,J,K,L	fried in high oleic sunflower seed oil at 150 °C
ι	E,F,J	macerated in potable water at pH 10 ($CaOH_2$), 30 min at 20 °C, roasted at 145 °C
κ	E,F,J	macerated in potable water with 2.5% w/w glucose, 30 min at 20 °C, fried in high oleic sunflower seed oil at 150 °C
λ	A,C,E,F,H,J,L	macerated in potable water, 30 min at 20 °C, roasted at 145 °C

A: Runner, Flavorunner 458, M grade, High Oleic, USA – Texas
B: Runner, Flavorunner 458, Jumbo grade, High Oleic, USA-Texas
C: Runner, Georgia Green, M grade, Low Oleic, USA – Georgia
D: Runner, Georgia Green, Jumbo grade, Low Oleic, USA – Georgia
E: Runner, Granoleic, Jumbo grade, High Oleic, Argentina
F: Valencia, CN Natals, S grade, Low Oleic, South Africa
G: Runner, Tegua, M grade, Low Oleic, Argentina
H: Runner, Hsuji, M grade, Low Oleic, China
I: Virginia, mixed varieties, XL grade, Low Oleic, USA – Virginia
J: Virginia, mixed varieties, M grade, Low Oleic, USA – Virginia
K: Virginia, Middleton, XL grade, High Oleic, Australia
L: Virginia, Middleton, M grade, High Oleic, Australia

Key: Raw material code: Market type, grade, High oleic trait, geographical source.

The raw peanuts were roasted at different time-temperature combinations by baking (dry roasting) or frying (oil roasting) in high oleic sunflower seed oil (HOSO - Cargill, MO). Pre-treatments included maceration in potable water, and aqueous solutions of vinegar (pH 4) (Albert Heijn, Zaandam, NL), 2.5g $100g^{-1}$ powdered glucose monohydrate (Brouwmarkt, Almere, NL), and $CaOH_2$ (pH 10, 0.01g $100g^{-1}$) (Merck, Kenilworth, NJ). Two post treatments (application of HOSO or Aromatic Roasted Peanut Oil at 2g $100g^{-1}$ each) were also applied to a subset of samples. The topical application of a small amount of oil is unlikely to have a textural impact, and so these samples were treated as replicates. This resulted in 94 unique roasted peanut samples. Details on the raw materials and process conditions are shown in **Table 11**. Additional information can be found elsewhere (Lykomitros et al., 2016a).

Effort was placed on maintaining the final moisture content as constant as possible, as moisture content is known to be a major driver of texture (McDaniel, White, Dean, Sanders, & Davis, 2012). The baking or frying time for each treatment

and sample therefore varied, in order to ensure a final moisture content of 2g $100g^{-1}$. All the reagents used in this research were obtained from Sigma-Aldrich (St Louis, MO), unless otherwise specified.

For the confocal and X-ray tomography micrographs two separate samples were prepared from the 'Granoleic' variety (Runner type, Jumbo size, from Argentina), both fried at 150°C in HOSO, one first macerated at 20°C with potable water for 30min, and the other without maceration. The fried, non-macerated sample was selected as the control. Two additional samples were prepared for the X-ray tomography from the same raw material, one fried and one baked, both macerated in aqueous glucose.

4.2.2 Texture and moisture measurements

Three hundred grams of each sample were first equilibrated at 20°C for 24h in airtight containers, before being split into two portions, one for moisture and one for texture analysis. The moisture content was determined by grinding a 100g sample of each material in a mini food processor (Kenwood, Havant, UK) and analysing 3g in a Leco TGA701 thermogravimetric analyser (St. Joseph, MI) at 113° C until constant weight was observed.

The large deformation mechanical properties were analysed using a TA-XT2 Texture Analyser (Stable Micro Systems, Godalming, UK) with a 25kg load cell and two different probes: The 'Volodkevitch Bite Jaw' (VBJ) and a the 'P/2' probe. The VBJ probe is designed to resemble an incisor tooth, and therefore simulate biting, whereas the P/2 probe is a 2mm diameter cylinder that provides compression and puncture data (Stable Micro Systems, 2015).

The 'hardness' (peak force [N]) and 'toughness' (positive area under the force-deformation curve [mJ]) were recorded. The probe speed was set at 2.00 mm/s, and the end point was set at 2mm and 3mm deformation after a trigger force of 0.05N was first detected for the P/2 and VBJ probes respectively. The pre- and post-test probe speeds were set at 1.00 mm/s and 10mm/s, respectively. The data acquisition rate was set at 500 points per second. Twenty peanut halves (single cotyledon) from each sample were individually measured putting the flat side down for each probe and the results averaged.

4.2.3 Sensory texture profiles

Three hundred grams of each sample was equilibrated at room temperature for 24h and Descriptive Sensory Analysis (DSA) was performed by a trained panel at the USDA, ARS, Market Quality and Handling Research Unit (Raleigh, North Carolina, USA). The panel consisted of ARS affiliates (10 panellists, 7 female, mean age 33), with a large experience in the sensory profiling of peanuts (minimum 250 hours judging experience per person; median 500 hours). The panellists were further calibrated over 3 two-hour sessions over three consecutive days, using a randomly selected subset of the sample set. Samples were evaluated in duplicate by every panellist in a randomized order (10 samples per session, approx. 90min), identifiable only by a random three digit code, using the Spectrum ™ method (15 point scale, Sensory Spectrurm, Inc., Chatham, NJ, USA). Non-salted crackers and water were used as palate cleansers (Meilgaard, Civille, & Carr, 1999). Based on panel experience, two replicates were deemed to demonstrate sufficient reproducibility, but a third replication was introduced in cases were the difference between the averages exceeded 1 point. This resulted in a mean Coefficient of Variation (across all samples and attributes) of 0.22 and a median CV of 0.18. An analysis of variance for each attribute confirmed the panel was indeed well trained (in all four cases F-test statistic for sample effects yielded $P<0.01$, while for effect of panellist was $P>0.6$).

The profiling took approximately 4 weeks to complete. The development and validation of the lexicon has been previously published in detail by the same long standing panel (Grosso & Resurreccion, 2002; Johnsen, Civille, Vercellotti, Sanders, & Dus, 1988), and has been widely accepted as comprehensive and used by several studies (Dean, Davis, Hendrix, Debruce, & Sanders, 2014; Lee & Resurreccion, 2006; Young et al., 2005). Attributes include: ‘crispy’ (degree (volume) to which the sample makes a high-pitched sound (incisors)), ‘crunchy’ (degree (volume) to which a sample makes a low pitched sound (molars)), ‘hardness’ (amount of force required initially to bite/fracture the sample using the molars) and ‘breakdown’ (degree to which the sample breaks apart using the molars on the first bite). The corresponding flavour analysis has been published elsewhere (Lykomitros et al., 2016a).

4.2.4 Imaging

An SP5 Leica Laser Scanning Confocal Microscope (Leica Microsystems, Buffalo Grove, IL) fitted with a Leica HC PL Fluotar 10.0x0.30 DRY objective was used to obtain the confocal micrographs. Fluorescence from neutral lipids was

detected in the green channel at 559-601nm, using Nile Red stain at an excitation wavelength of 514nm (Jose & Burgess, 2006). The red channel shows fluorescence collected from 650 to 682nm using Nile Blue stain excited at a wavelength of 233 nm. The Nile Blue stain at this wavelength highlights cell nuclei and peptides (Jose & Burgess, 2006). The micrographs were taken at 512x512x8bit resolution, with 486V to 448V at the PhotoMultiplier Tube (PMT). Due to the crumbly nature of the samples, micro slices were impossible to obtain, and so thicker slices (~1mm) where prepared and optical sections were scanned with the confocal microscope (~80 sections), which were further combined into a single image per channel using the Fiji/Image J open source software (Z projection, MAX emission) (Schindelin et al., 2012).

For the 3D imaging, a Phoenix v|tome|x m X-ray tomographer (General Electric, Wunstorf, DE) was used. A 240 kV X-ray source with a tungsten target was employed. The images were recorded by a GE DXR detector array with 2024 × 2024 pixels (pixel size 200µm), located 815mm from the X-ray source. X-rays were produced with a voltage of 80 kV and a current of 120 µA. The sample was placed 40.78 mm from the X-ray source, resulting in a spatial resolution of 10.0 µm. A full scan consisted of 1800 projections over 360°, with the first image omitted. The resulting projection was the average of 3 images where every image is obtained over 250 ms exposure time. GE reconstruction software (Wunstorf, DE) was used to calculate the 3D structure via back projection, and was further analysed using the Avizo 9.2.0 imaging software (FEI, Hillsboro, OR): The 3D data was first filtered by a non-local means filter to reduce the noise in the greyscale dataset, followed by separating the images of the sample and the sample holder using different segmentation tools into a 3D labelled image. The air cell volume distribution was consequently measured by the software for each sample. Air cells with volume less than 0.0001mm^3 were ignored, as they were likely imaging artefacts. Finally, a cross section of the greyscale and labelled image was exported to provide a visual.

4.2.5 **Statistical analysis**

Unless otherwise stated, all statistical analysis was performed using XLSTAT 2015.5 (Addinsoft, Paris, FR on MS Excel 2010, Microsoft, Redmond, WA). Firstly, a Principal Component Analysis was run on the sensory scores (using the covariance matrix) in order to visualize the range. A Canonical Variate Analysis (CVA) was further run on the texture sensory attributes (forward model selection, threshold to enter model α=0.2, equal within class covariance), using 'process' for classification. The motivation for this analysis was to visually determine the relative importance of process versus material selection on texture, and specifically if processing can overshadow the effect of raw material.

Finally, to quantify the effect of process (baking vs frying), pre-treatment (maceration yes/no) and their interaction on the texture sensory attributes, general linear model regressions were run based on the model shown in **Equation 1**.

$$sensory\ attribute_i = a_i \times (no\ maceration) + b_i \times (fried) + c_i \times (no\ maceration \times fried) + d_i$$

Equation 1

i is the ith sensory attribute, and a_i, b_i,c_i, and d_i are the corresponding parameters for the ith sensory attribute. 'no maceration' and 'fried' are binary model factors that assume the values 0 or 1 for 'yes' or 'no', respectively.

4.3 Results and discussion

4.3.1 Impact of processing on texture

Figure 16 illustrates the results of the four texture instrumental parameters obtained. A wide range of responses were observed for all four parameters. The VBJ probe is less precise compared to P/2 2mm puncture cylinder probe, with relative standard deviation across all sample measurements of 20.4% versus 11.5%. Probes similar to the VBJ have also been successfully used with peanuts by other authors who reported similar values (Miyagi & Ogaki, 2014), while puncture tests have been successfully used to quantify texture in pecans (Ocon & Anzaldua-Morales, 1995). A discussion on probe selection can be found in the supplementary material.

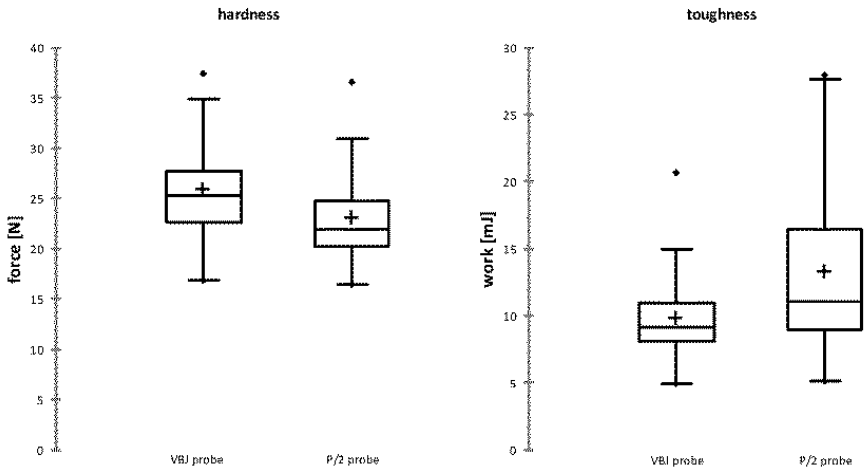

Figure 16. Overview of large deformation texture data for VBJ and P/2 probes. Left panel: hardness; right panel: Toughness. Diamonds: Min/Max values, Crosses: mean, Line: median, Box: Q2 and Q3, whiskers: <Q1-1.5(Q3-Q1); >Q3+1.5(Q3-Q1).

Table 12 summarizes the range of sensory scores observed across all samples and attributes. A larger variation in texture was observed for 'crispy', 'crunchy' attributes compared to 'hardness' and 'breakdown'. **Figure 17** displays the PCA scores plot for the sensory texture with the samples coded for the raw material they were produced from. The figure shows that significantly different texture profiles can be derived from the same raw material through different processing, something best demonstrated by observing the scores for raw material E (Runner, Granoleic): This sample can be seen to exhibit a widely different texture profile depending on the process it underwent. However, the effect of the material is still present, as samples made from a given material are not entirely randomly dispersed in the texture space. This is not surprising, as textural differences between different market types and maturity levels have been reported (Kim & Hung, 1991; Woodroof, 1983), and can be attributed to the significant compositional differences between peanut types: for instance, Spanish type peanuts consist on average of 10% more dietary fibre compared to Virginia type peanuts (USDA, 2015b), while the sugar content of peanuts is known to vary based on genetics and growing conditions (Pattee, Isleib, Giesbrecht, & McFeeters, 2000). The sensory attributes were further modelled against the large deformation instrumental attributes using Partial Least Square regression, and the results can be found in the supplementary material.

Table 12. Range and descriptive statistics of sensory scores for all samples. Scores were obtained using the Spectrum ™ method (15 point scale).

	crispiness	crunchiness	hardness	breakdown
minimum	1.12	2.49	3.90	6.33
maximum	5.97	7.88	8.07	8.89
range	4.86	5.40	4.18	2.57
average	1.94	4.00	5.68	7.88
CV*	36%	24%	14%	6%

* coefficient of variation

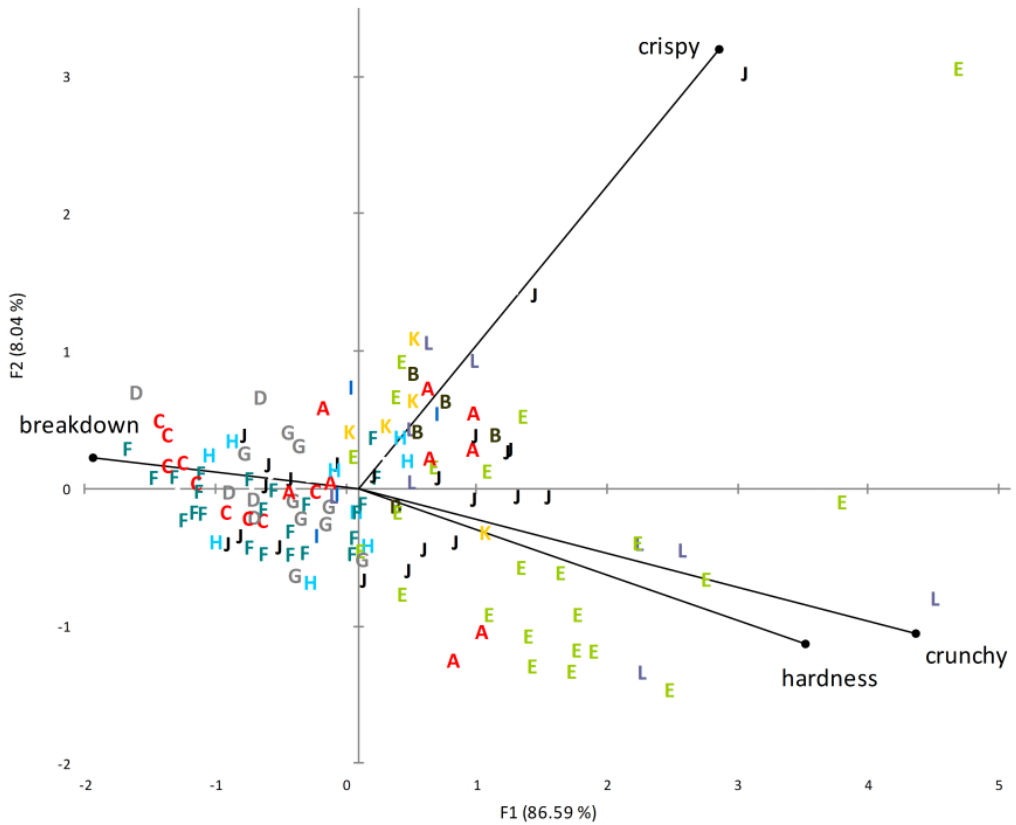

Figure 17. Principal Component Analysis scores plot of the sensory texture space. Two principal components account for 94.6% of the variance. The raw material used in each sample is colour and letter coded in the chart (key in **Table 11**).

The Canonical Variate Analysis reported in **Figure 18** was run to evaluate the differences in texture caused by the different processes. Panel B describes the texture sensory space: the bottom left quadrant is mainly characterized by high 'breakdown' scores, whereas 'hardness', 'crunchy' and 'crispy' increase towards the top right quadrant. Panel A, shows the clusters of products labelled by process. All maceration processes (processes α,β,γ,ι,κ,λ in **Table 11**) fall in the top quadrants, suggesting that aqueous maceration leads to a relative increase of the 'crispy', 'hardness' and 'crunchy' attributes, for all raw materials. A significant interaction between maceration in glucose and oil roasting (process κ) can also be seen, resulting in a much higher score for 'crispy'. Conceptually, Canonical Variate Analysis (a type of Discriminant Analysis) can be understood as being similar to PCA, but rather than selecting eigenvectors that maximize differentiation between samples, the components are selected so as to maximize the differences between groups of samples with a common classifier (process in this case).

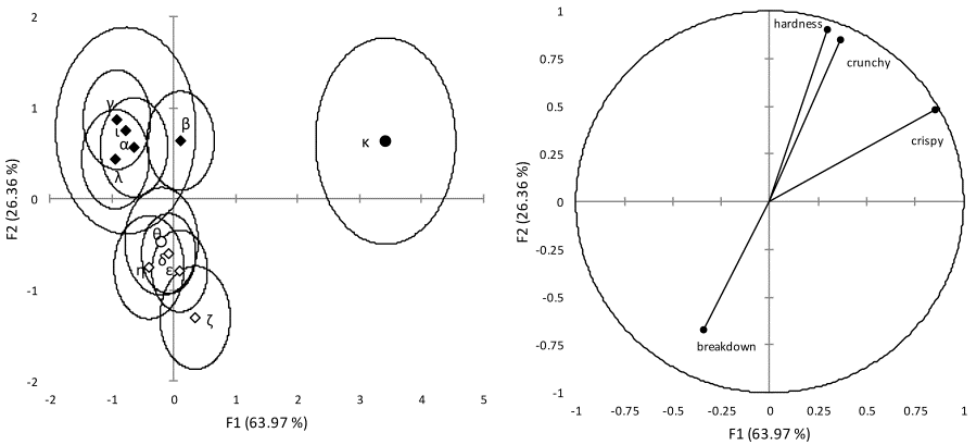

Figure 18. Left (Panel A): Canonical Variate Analysis of sensory attributes with process used as a classifier, with 95% confidence ellipses. Diamonds: dry roasted (baked); Circles: oil roasted (fried); Filled shapes: macerated; Empty shapes: non-macerated. Codes for individual processes can be found in **Table 11** (Processes γ and κ employed glucose maceration). Two components resolve 90.3% of the data variance. The significantly larger impact of process (margination) versus raw material is evident. Right (Panel B). Canonical Variate Analysis sensory space of texture attributes for process technology classification.

General Linear Model Regressions were further run to quantify the effect of processing on the four sensory attributes. The process treatments modelled included baking (vs frying), maceration (yes-no) and their interaction, and are displayed graphically in **Figure 19**. The model parameters were kept as few as possible to retain generality of conclusions: factoring for country of origin, market type, and so on could have further improved the model fit, but the interest was

more in determining main effects that are significant across a large variety of raw materials and process conditions rather than risking overfitting. The models confirm that maceration in any medium significantly increases 'crunchy', 'crispy' and 'hardness', while the opposite is true for the 'breakdown' attribute. This has been previously reported only for water. Similar trends were observed for frying (vs baking), but the effect was only statistically significant for 'crispiness'. Examining the interaction plots, it is apparent that although maceration has a larger impact when followed by frying (as in 'blister frying') on all attributes, the effect is also significant when maceration is followed by baking (but to a smaller magnitude), something not previously reported. In addition, as the effect of baking (vs frying) and maceration (vs no treatment) are more or less opposite, the data suggest that one could potentially employ a maceration step prior to a dry roasting process in order to obtain similar textural attributes of fried products. Further optimization is required to determine the exact baking conditions, but this insight could allow peanut roasters to produce baked peanuts with the same textural characteristics as fried.

The observation that maceration followed by frying can affect texture has been widely documented. What is surprising is that the magnitude is large enough to surpass the effect of different peanut market types and varieties (not previously observed as all published and market examples of fry blistered peanuts employ Virginia type kernels only). However, it was noted that glucose macerated and fried samples had similar crunchiness but increased hardness and significantly less 'blistered' appearance compared to water macerated and fried samples.

4.3.2 **Microstructure and texture**

Figure 20 displays 2D X-ray tomography planes for selected samples, while **Figure 21** displays the air cell distributions for the entire sample (not only the plane displayed in **Figure 20**), and provides for the first time quantitative data on microstructural changes in roasted peanuts. **Figure 20** panels A-D show the grayscale images, while panels E-H have the closed air cells only highlighted in blue. The control fried peanut (A) has some large air cells mainly in the centre, and a few small mainly on the perimeter. In the macerated-fried samples (B-C), the number of small air cells is significantly larger, and they are dispersed throughout the cotyledons. The macerated-fried sample was prioritized in this investigation because it exhibited one of the highest sensory scores for 'crunchy' and 'crispy'. Little difference in the microstructure between panels B and C is evident (water vs glucose maceration), suggesting that glucose does not have a significant role in microstructure disruption. Finally, panel D displays a glucose macerated baked sample, where a marginally smaller number of air cells are observed compared to the fried samples, albeit somewhat concentrated to the perimeter of the cotyledon. This suggests that microstructure disruption by steam generation is not

the only mechanism that impacts texture during roasting of macerated peanuts, as the samples displayed here have similar microstructure disruption, but different textural characteristics.

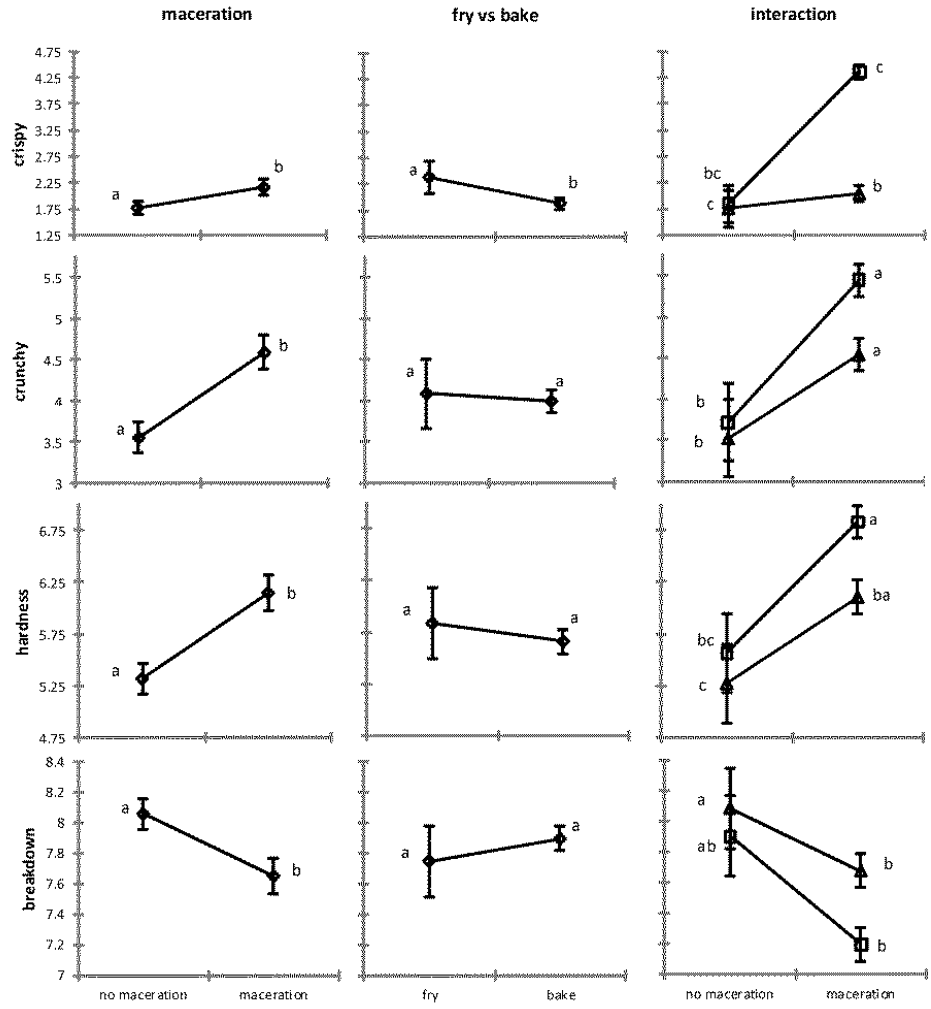

Figure 19. Impact of peanut process on sensory texture attributes: main effects (maceration; roasting) and interaction (roasting x maceration) plot (model shown in Eq. 2) with 95% confidence intervals. Diamonds: main effects; Squares: fry; Triangles: bake. The right most column shows that the impact of maceration on the sensory attribute depends on which roasting method is employed (fry (squares) or bake (triangles)). Significance of means difference was calculated using Tukey's HSD test (α=0.05).

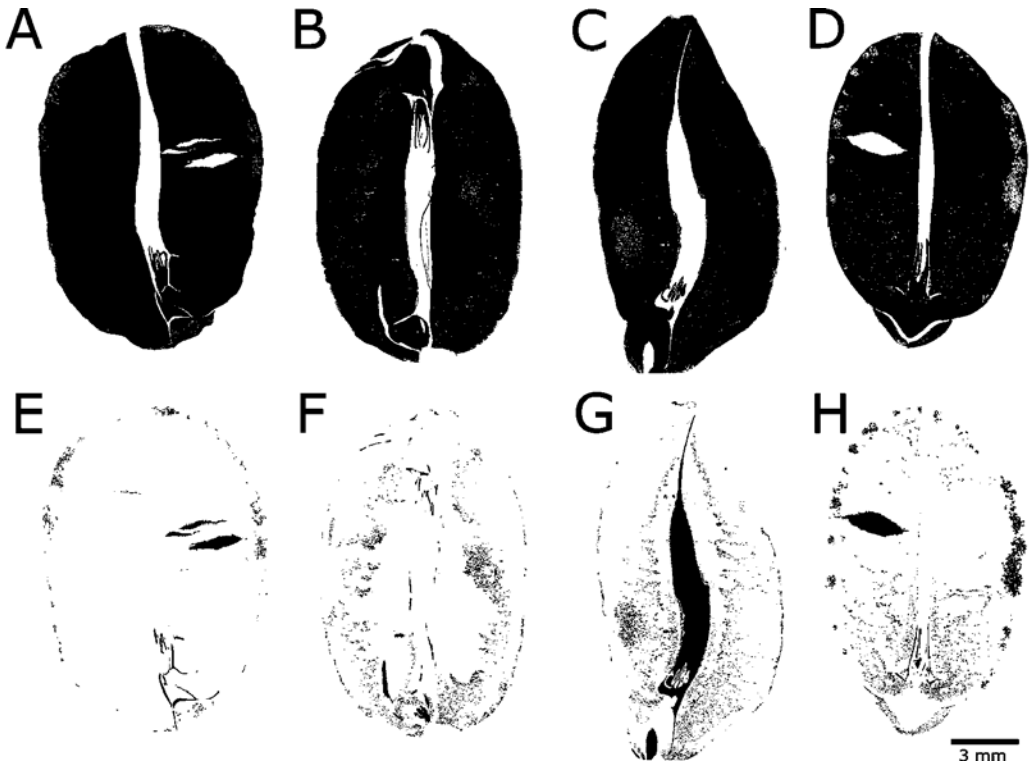

Figure 20. Selected X-ray tomography 2D projections. A: Fried control, B: Water macerated and fried ('blister fry'), C: Glucose macerated and fried, D: Glucose macerated and baked. Panels A-D show the actual projections (peanut matrix in black, air cells in white), while panels E-H show the closed air cells only, highlighted in black (air cells in black, peanut matrix not shown). It can be seen that macerated samples (B, C and D) exhibit a significantly larger number of closed small air cells than control (A), This is better visible in panel F G and H respect to control E. The differences induced by maceration procedure is independent by the maceration medium composition and by the processing (fried or baked).

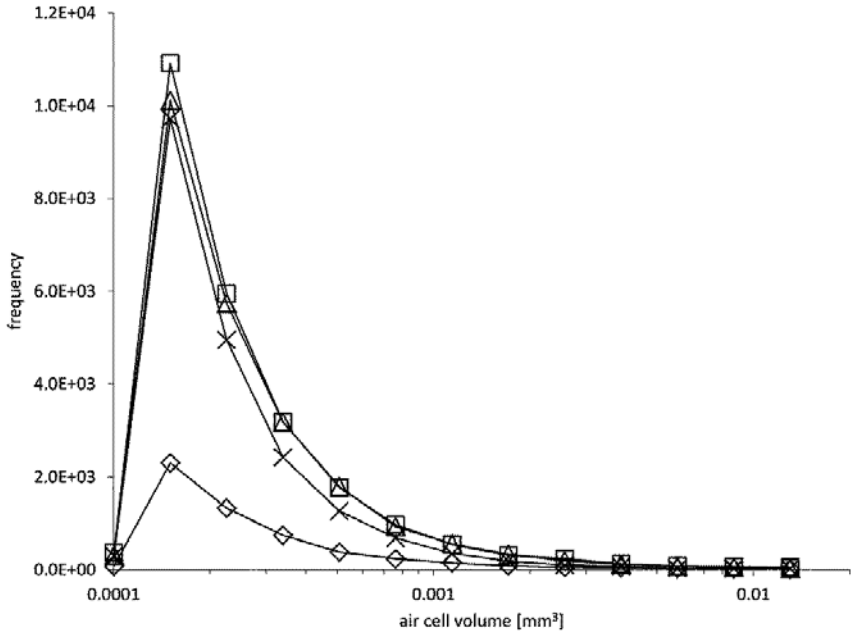

Figure 21. Quantitative data on air cell volume distributions of the samples displayed in **Figure 20**. Diamond: Fried control, Square: Water macerated and fried, X: Glucose macerated and fried, Triangle: Glucose macerated and baked.

Indeed, others have also reported that the roasting method (fry vs bake) does not have a significant impact on the microstructure disruption, but only on the kinetics by which it is achieved (Young & Schadel, 1993). Others(Shi et al., 2017) have also noticed similar microstructure disruption of the parenchyma cells regardless of maceration and roasting method, albeit the epidermal cells were more damaged by the 'blister fry method'. Compared to these previously published qualitative microscopy images, the data presented in **Figure 21** represents a quantitative measure of the number of pores created upon roasting and somewhat of the microstructural change induced by roasting. In addition, the X-ray tomography data presented here provide a more holistic view, as X-ray tomography analyses the entire kernel, rather than the small loci that electron microscopy can resolve. Despite of the very small difference in microstructure disruption however, baked samples are consistently less crispy (**Figure 19**) (Shi et al., 2017), providing additional evidence that a secondary mechanism is likely contributing to texture development.

Figure 22 shows the confocal micrographs of a control fried sample (Panels A-C) and a macerated (high initial moisture content, 'blister fry') fried sample (Panels D-F). To avoid interactions with the stains, only samples macerated in water were investigated with confocal microscopy. Panels A and D highlight lipids (in green), while panels B and E highlight proteins (in red). The composite images are

shown in panels C and F, where loci with both the green and red channel activated appear as yellow. The significantly higher disruption of the microstructure (diagonal fissures) on the macerated (high crunchiness and hardness) sample (Panels D-F) is clearly visible also in the confocal micrographs. Lipids appear less uniformly dispersed than the control, and tend to aggregate in droplets throughout the structure of the kernel and in particular around the fissures. This suggests that not only the cells but also oleosomes undergo a larger damage by adopting a maceration procedure prior to heating.

The microscopy observations reported in the present paper support the theory that microstructure disruption due to steam generation is a key contributor to crispy/crunchy texture development, in line with the evidence from electron microscopy found in the literature. Other researchers also noted that when the initial moisture content prior roasting was higher, a more extensive disruption of the microstructure took place, due to the larger amount of steam produced with roasting (Dean et al., 2014; Debruce, Dean, & Sanders, 2009; Idrus & Yang, 2012).

However, the mastication sound associated with crispiness and crunchiness is generated by micro fracture of layered or cellular materials, and the amplitude is related to the strength and flexibility of these layers or cells (Luyten, Plijter, & Vliet, 2004). Thus, processes that increase heterogeneity by developing layering or increasing the local mechanical moduli, will also increase perceived crunchiness and crispiness. Indeed, microstructure disruption in peanuts has been previously associated with increased crispiness and crumbliness, and it was directly correlated to lower chewing requirement (McKiernan & Mattes, 2010). The amplitude of the mastication sound however, is related to the strength of these heterogeneous layers, something not influenced by steam generation. Any process induced chemical interaction that increases said strength can therefore make a key contribution to the peanut texture.

Here it is proposed that Maillard reaction between proteins and reducing sugars may contribute to peanut texture (mainly hardness, but potentially also crispiness and crunchiness). Moisture and heat provide the means for macromolecule unfolding and mobility, enabling chemical interactions that lead to increased local mechanical moduli. While most literature has focused on the effect of steam generation, one other study has also proposed that some chemical interaction is also contributing to texture development when SEM images showed that material near the fissures appeared to be denser and amorphous, and described it as being in ‘a glassy state’ (Miyagi & Ogaki, 2014).

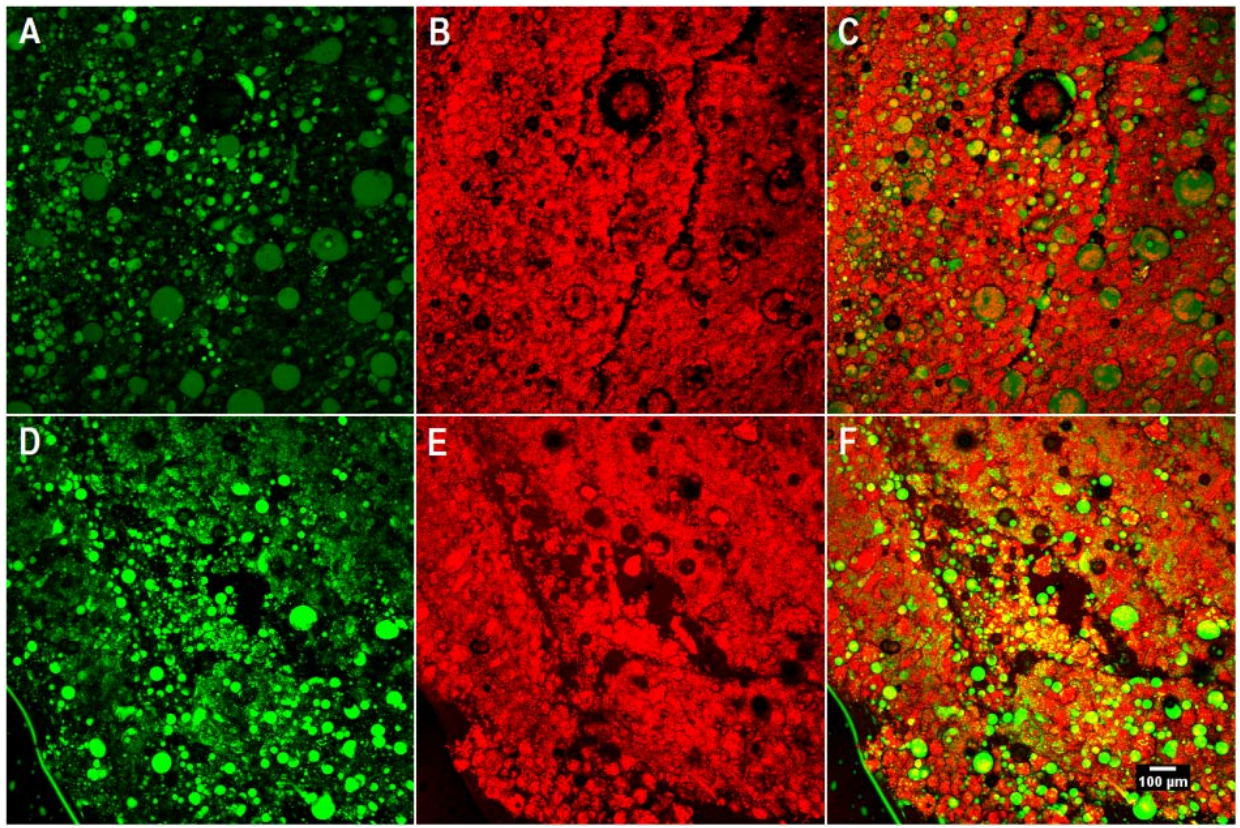

Figure 22. Confocal Micrographs of control fried (panels A-C) and macerated fried (Panels D-E) peanuts. Panel A: control, lipids. Panel B: control, proteins. Panel C: control, both lipids + proteins. Panel D: macerated, lipids. Panel E: macerated, proteins. Panel F: macerated, lipids + proteins. More extensive cellular disruption and lipid pooling is evident in the macerated sample (D-F) versus the control (A-C).

Indeed, comparison of samples C and D in **Figure 20** (produced by processes γ and κ; see **Table 11** and the Canonical Variate Analysis in **Figure 18**) showed that microstructure disruption alone (steam generation) cannot fully explain the observed texture differences between samples. A preliminary sensory evaluation of the samples produced for the X-ray tomography showed significantly higher hardness (but not crunchiness) for samples macerated in glucose solutions (sample C in **Figure 20**) compared to samples macerated in water ('blister fried') (sample B in **Figure 20**) despite the number and distribution of air cells not being significantly different between the two samples. It is proposed that the development of texture in macerated peanuts is due to two mechanisms: Firstly, the microstructure disruption due to steam generation, as widely accepted and documented ('blister fry'). Secondly, by cross linking between proteins, reducing carbohydrates or oxidized lipids leading to the formation Maillard reaction products. Melanoidins in particular, the brown polymers which are the final product of the Maillard reaction, are known to be present in large amounts in roasted nuts (Açar, Gökmen, Pellegrini, & Fogliano, 2009). Indeed has been shown that in protein bars high molecular weight Maillard reaction products can result in texture hardening (Zhou, Guo, Liu, Liu, & Labuza, 2013). It is likely that the presence reducing sugars (endogenous or added in the maceration medium) increases the rate of Maillard reaction, and therefore leads to a higher production of melanoidins which in turn affects the texture (hardness). The extent of this effect is not entirely independent of the raw material, as the type and amount of naturally occurring sugars and amino acids (Maillard reagents) is determined by genetics, growing conditions and maturity of the groundnut (Pattee et al., 2000).

The contribution of Maillard reaction to peanut texture can also explain why the impact of maceration is higher in fried compared to baked peanuts. Indeed, for constant end moisture, higher roasting temperatures lead to darker colour peanuts (McDaniel et al., 2012), suggesting more Maillard reaction products. More research is required to definitively demonstrate the contribution of Maillard reaction to peanut texture.

4.4 Correlations between instrumental and sensory texture attributes

4.4.1 Probe geometry selection for large deformation compression testing of peanuts

The peculiar geometry of the peanut kernels requires a careful selection of a suitable probe. The peanut kernel consists of two cotyledons, connected at the germ (Woodroof, 1983), and so any mechanical compression test on the whole

kernel will essentially test the strength of the germ. Furthermore, the cotyledons themselves are heterogeneous (Young et al., 2004), and of an irregular shape with no flat surfaces, so any parallel plate test will also be more likely to test the stress points, and not the bulk of the material. Some researchers have attempted to standardize the shape of nuts (pecans) by trimming with blades (Anzaldua-Morales & Brusewitz, 1999; Surjadinata, Brusewitz, & Bellmer, 2001), but in the current study some of the crunchiest samples would crumble if a cut was attempted. Finally, since a distribution of different maturity levels in the sample is unavoidable (Williams, Ware, Lai, & Drexler, 1987), and since maturity level is known to affect microstructure and therefore texture (Kim & Hung, 1991; Young et al., 2004), enough repetitions had to be made to minimize the signal to noise ratio (Vivar & Brennan, 1980). The selected probes (VBJ and P/2) were shown to fit the sample geometry and provided repeatable results.

In cases where the sample has small, non-uniform physical dimensions and large piece to piece variability, bulk compression probes such as the Kramer shear cell have been seen to obtain good results by mechanically averaging out sample to sample variation (Hung & Chinnan, 1989; Lee & Resurreccion, 2006). However as noted elsewhere, although a Kramer Cell could offer lower coefficients of variation (load cell of 400 Kilograms used), individual compressions offer a better insight into 'hardness' as perceived by consumers (Davis, Price, Smyth, Drake, & Sanders, 2009). The Kramer cell approach was evaluated but abandoned, as the load on the instrument would quickly exceed even the largest available load cell (50KG), due to the hard nature of some of the macerated samples.

4.4.2 Modelling of sensory texture parameters from instrumental parameters

A separate field in peanut texture studies includes the search for correlations between instrumental data and sensory attributes. Such examples can be found, but often the primary focus of the research lies on characterization of a particular process (Idrus & Yang, 2012; Miyagi & Ogaki, 2014; Young et al., 2005) or raw material (Kim & Hung, 1991), and thus the correlation was developed for a restricted sample set. In other cases, the primary focus was on storage changes and so the lexica and instrumental methods were optimized to resolve smaller stimuli ranges (Lee & Resurreccion, 2004, 2006).

Partial Least Squares regression was run in order to model the textural sensory attributes ('crispy', 'crunchy', 'hardness' and 'breakdown') with the attributes from the instrumental analysis ('hardness' and 'toughness' for each of the two probes). The model used is shown in **Equation 2**.

$$Attribute_i = constant_i + \sum_4^{j=1}\left(instrumental\ attribute_j \times coefficient_{i,j}\right)$$

Equation 2

Where i= the i^{th} sensory attribute, j= the j^{th} instrumental textural attribute, $coefficient_{i,j}$ = the model coefficient of the j^{th} instrumental attribute in the i^{th} model (sensory attribute) and $constant_i$ = the constant of i^{th} model (sensory attribute).

The models between the instrumental and sensory attributes were developed (four response variables) and are shown in **Figure 23** (R^2= 0.35, 0.71, 0.68 and 0.57 for 'crispy', 'crunchy', 'hardness' and 'breakdown' respectively). It is immediately obvious that the sensory attributes 'crunchy', 'crispy' and 'hardness' are fairly correlated, suggesting that even though the panel was well trained, there may still be some confusion between the terms (Tunick et al., 2013). It was decided to keep both terms as several trained panels have concluded that both terms are needed (Grosso & Resurreccion, 2002; Lee & Resurreccion, 2006; Meilgaard et al., 1999). **Figure 23** also shows that hardness measured by the VBJ probe is highly correlated to hardness measured by the P/2 probe, suggesting that the two attributes provide similar information on texture. Conversely, toughness measured by the P/2 probe is practically orthogonal to the other instrumental attributes, meaning it provides incremental information capable of resolving differences amongst the samples, but is not significantly correlated to any of the sensory textural attributes. This agrees with Varela and co-workers, who proposed that although humans evaluate texture using mainly their molars (almonds were evaluated by consumers in that case), better instrumental correlations can be obtained using probes that resemble incisor teeth, such as the VBJ probe (Varela, Salvador, & Fiszman, 2008).

The standardized model coefficients for each of the four regression models are reported in **Figure 24**. The standardized coefficients, or 'β-coefficients', are a measure of the relative weight of the model parameters, and as such if the confidence interval includes zero the parameter is not significant. Interestingly, all instrumental attributes were significant in predicting all sensory attributes, with the exception of toughness measured by the P/2 probe. This suggests that the different attributes measured by the different probes, all contribute to resolving sensory attributes (also evident in **Figure 23**, where the different instrumental attributes are seen to not be entirely correlated). This is likely the reason why some studies have been unable to model sensory response by compression test data, using only one instrumental parameter at a time (Lee & Resurreccion, 2006).

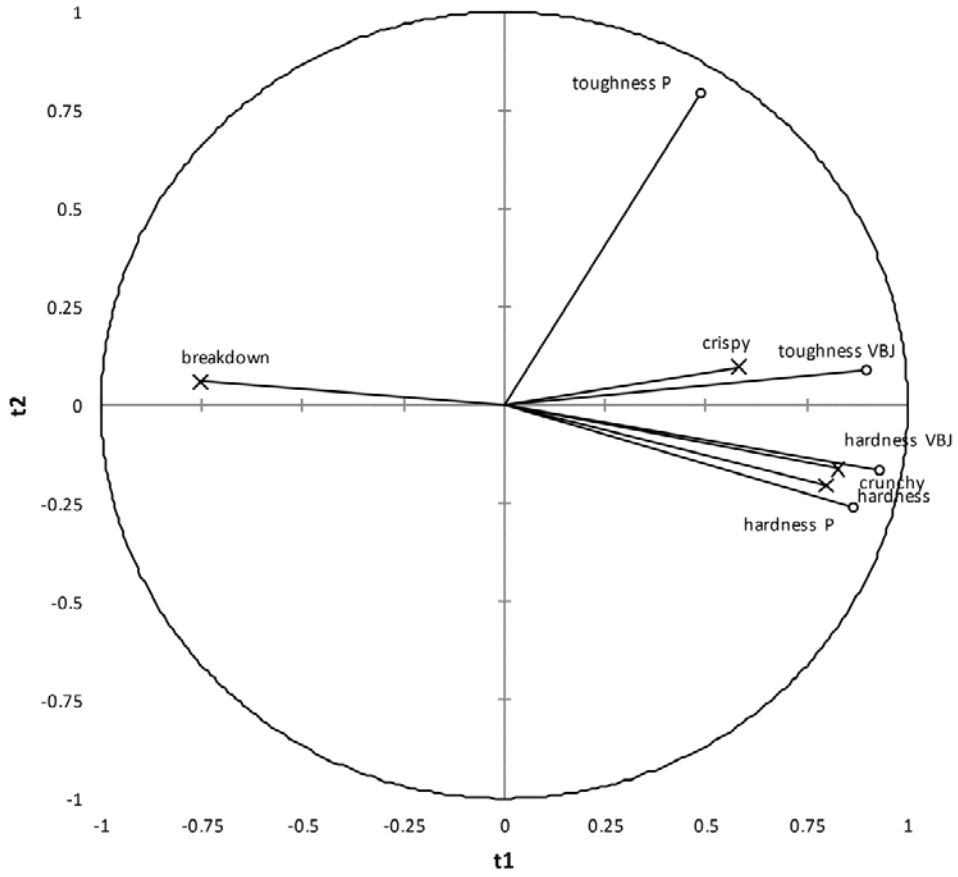

Figure 23: Graphical representation of the first two dimensions of the Partial Least Square Regression between sensory (X) and instrumental (O) attributes. Instrumental attributes have been obtained with the P/2 probe (P) and the Volodkevitch Bite Jaw probe (VBJ).

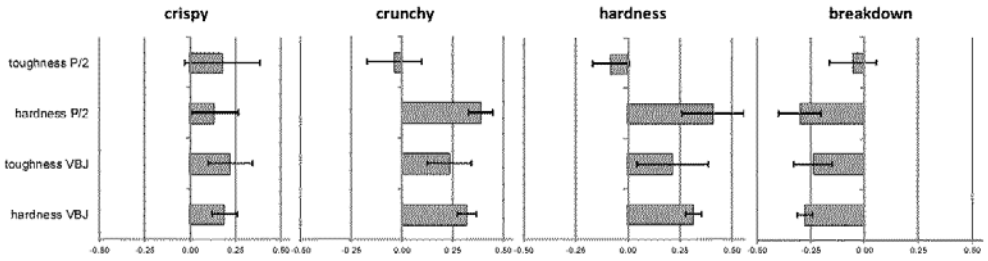

Figure 24. Standardized coefficients (with 95% confidence interval) of the Partial Least Square Regression models between instrumental and sensory texture parameters.

The sign of the PLS coefficients (**Figure 24**) are consistent with correlations seen on the PCA analysis (**Figure 23**); 'breakdown' is inversely correlated to 'crispy', 'crunchy' and 'hardness', and indeed the coefficients follow the same trend. **Figure 24** also shows that the maximum force measurements (hardness) have generally a larger weight in the model than compression work (toughness) for both probes. This is not surprising considering roasted peanuts are generally not 'rubbery' and tend to fracture rather than deform, particularly in the case when the force is applied over a small area (as with the P/2 probe). Consequently, model coefficients for P/2 toughness were not significant. Finally, the low R^2 value for the 'crispy' model is also reflected by low standardized coefficients. The poor correlation obtained for 'crispy' can be attributed to two factors: Firstly, as discussed above the response variable itself (sensory 'crispy') is often confused with 'crunchy'. Secondly, it has been reported that crispiness often correlates better with acoustic measurements (Davis et al., 2010; Varela, Salvador, & Fiszman, 2009), and this sensory panel also defined it as an acoustic attribute. However, acoustic measurements require bespoke and often sophisticated experimental setup, which is not always available particularly in a manufacturing environment. A methodology that relies on equipment currently found in quality control laboratories would have an obvious advantage. Furthermore, given that sound is generated by micro-fractures during large deformation (Luyten et al., 2004) they could also be detectable by stress-deformation in addition to acoustic methods. For these reasons, only a simple deformation test was used.

The results suggest that an acceptable correlation of most sensory texture attributes of peanuts can be obtained from large deformation tests, with the exception of crispiness. Consequently, the relative simplicity, minimal training requirements and low cost of this instrumental method compared to a sensory panel, instrumental texture measurements represent a valuable tool for peanut processors.

4.5 Conclusions

This study demonstrated that, although the choice of raw material has a significant effect on texture of the final peanut, the type of process is highly significant and can often overshadow the impact of raw material. Specifically, it was shown that maceration in aqueous glucose media followed by roasting (both frying and baking, and regardless of the peanut type) can significantly increase the

perceived hardness, crunchiness and crispiness, without inducing a blistered appearance as in the case of conventional 'blister frying'. This may allow processors to fine tune the textural characteristics of their end product, rather than settle for the 'all or nothing' approach of 'blister frying' versus conventional roasting (very crunchy, hard and blistered, vs standard roasted peanuts). Furthermore, these findings suggest that process can be used to compensate for suboptimal raw materials, providing economic benefits and help reduce food waste, an area food processors are urged to invest in. Finally, this study showed agreement with the existing theory that the textural changes induced by maceration are caused by steam generation upon heating, but further suggested a possible secondary mechanism: textural (mainly hardness) development though formation of melanoidins or other products of interaction between cellular components.

4.6 Acknowledgement

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5 Drivers of preference and perception of freshness in roasted peanuts (arachis spp) for European consumers.

Adapted from:

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Abstract: Roasted peanuts are a popular snack in Europe, but their drivers of liking and perceived freshness have not been previously studied with European consumers. Consumer research to date has been focused on US consumers, and only on specific peanut cultivars. In this study twenty-six unique samples were produced from peanuts of different types, cultivars, origins and with different process technologies (including baking, frying and maceration). The peanut samples were subjected to sensory (expert panel, Spectrum ™) and instrumental analysis (colour, headspace volatiles, sugar profile, large deformation compression tests and graded by size) and were hedonically rated by consumers in The Netherlands, Spain and Turkey (n>200 each). Preference Mapping on mean liking (PREFMAP) models revealed that the drivers of liking are similar across the three countries. Sweet taste, roasted peanut, dark roast and sweet aromas and the colour b* value were related to increased liking, and raw bean aroma and bitter taste with decreased liking. Further Partial Least Square Regression (PLSR) modelling of liking and perceived freshness against instrumental attributes showed that the colour coordinates in combination with sucrose content and a select few headspace volatiles were strong predictors of both preference and perceived freshness. Finally, additional PLSR models focusing on the headspace volatiles only, showed that liking and ‘‘fresh’’ attributes were correlated with the presence of several pyrroles in the volatile fraction, and inversely related to ‘‘stale’’ as well as to hexanal and 2-heptanone.

Keywords: Peanut, consumer preference, volatiles, liking, freshness

Highlights:

- The drivers of liking for Dutch, Turkish and Spanish consumers are both similar to each other and to US consumers.
- Colour is the most practical predictor of preference because it also correlated to flavour and texture.
- ‘Stale’ is better understood by consumers as a quality description attribute than ‘fresh’.
- Several pyrroles are highly correlated to consumer liking (but not pyrazines).
- Hexanal and 2-heptanone are correlated to ‘stale’ and decreased liking.

5.1 Introduction

Peanuts are very widely consumed crop due to their nutritional profile, cultural heritage, and low cost relative to other nuts (He, Fletcher, & Rimal, 2005). Studies on the drivers of peanut consumption can be divided into two categories, namely consumer or product focused. Consumer focused studies attempt to characterize peanut *consumers* based on socioeconomic, demographic, pricing or other factors (such as health consciousness)(He et al., 2005; He, Florkowski, & Elnagheeb, 1998; Jolly, Hinds, Lindo, Ham, & Weiss, 2001; Moon et al., 1999; Nelson, Jolly, Hinds, Donis, & Prophete, 2005). In the second category, studies have focused on *product* attributes, all executed exclusively with US consumers and with a single peanut type (Runner). The impact of sensory flavour attributes on preference has been probed in several studies, and 'roasted peanut' and 'fresh peanut' intensities were identified as drivers of liking, and 'bitterness' as a driver of dislike (Young, Sanders, Drake, Osborne, & Civille, 2005). In another study, the colour (L value) was the only attribute correlating to US consumer preference (Lee & Resurreccion, 2006b). The negative impact of shelf life on consumer preference, specifically the reduction in flavour intensity and crispiness due to moisture uptake over storage, has also been documented (Lee & Resurreccion, 2006a). Finally, four studies have attempted to correlate hedonic scores, sensory attributes and instrumental analyses. Three were focused on a single instrumental attribute (hexanal and force compression tests, respectively (Grosso & Resurreccion, 2002; Miyagi & Ogaki, 2014; Nader, Afif, & Louka, 2016), and the third was focused on oxidation during the shelf life (Nepote, Olmedo, Mestrallet, & Grosso, 2009). Recently a study has been published on sensory and headspace volatile drivers of liking for US consumers (Wang, Adhikari, & Hung, 2017). To the best of our knowledge, there has been no study simultaneously combining flavour, texture and appearance sensory and instrumental attributes, nor incorporating several peanut market types and process conditions. Moreover, no consumer study with European consumers nor incorporating perception of freshness was identified in the literature.

Given the recent strong consumer interest on 'freshness' across practically all categories (Sloan, 2015) it is reasonable to hypothesize that in addition to aroma, flavour, appearance and texture, perceived freshness could also be significant driver of consumer liking. The specific sensory attributes that signal a reduction in freshness depends on the category, with examples ranging from sourness (titratable acidity) in apples (Iwanami et al., 2017) to leaf turgidity in salad greens (Dinnella, Torri, Caporale, & Monteleone, 2014). To the consumer, 'freshness' usually implies 'recently prepared', a quality that consumers cannot objectively quantify.

In peanuts, lack of freshness for US consumers has been mainly linked to rancid flavour, as determined by hexanal content (Grosso & Resurreccion, 2002) as well as to a reduction in roasted peanut aroma (an effect termed 'flavour fade') (Lee & Resurreccion, 2006a; Williams et al., 2006). Even though 'freshness' has been seen to be a key driver of consumer liking of peanuts (Young et al., 2005), no published study could be identified attempting to quantify it's drivers in a defect free, non-aged sample. In cases where an obvious defect is absent, the perception of freshness may depend on the product design and/or subjective and often culturally relevant 'meta'-characteristics, such as type of packaging, and sale channel (e.g. nut roaster vs supermarket). Examples of where freshness is related to the product design and not the actual age of the product include orange juice, where perceived freshness was related to closeness to the orange fruit (Zhang, Lusk, Mirosa, & Oey, 2016) and ready to eat salads where the assortment of leafy greens in the salad correlated with perceived freshness (Dinnella et al., 2014).

The objective of this study was to identify which are the drivers of liking and perceived freshness of snack peanuts for European consumers, and to evaluate if these are different between consumers of the three countries tested (Netherlands, Spain and Turkey). To achieve this, the 'external preference mapping' (Burgard & Kuznizki, 1990) and Partial Least Squares methodologies were employed with 26 roasted peanut samples of distinct and varied organoleptic characteristics, analysed instrumentally and by trained sensory panel, and tested by consumers in the three countries.

5.2 Materials and methods

5.2.1 Sample preparation

Peanuts were procured raw (Canon Garth Ltd, London, UK) and were dry blanched to remove the testa (85°C, approximately 30min at Steinweg-Handelsveem BV, Oosterhout, NL). Twenty six unique and varied samples were prepared **(Table 13**). Where applicable, maceration was performed in a stainless steel container at a ratio of 12Kg blanched kernels per 20Kg maceration medium. Samples were thermally processed in a convection oven (Batch nut roaster Wolverine-Proctor, Glasgow, UK), an impingement continuous oven (Aeroglide C1 12-16 REX, Cary, NC), or a batch fryer (30L, De Kuiper, De Kwakel-Uithoorn,NL), for the time necessary to reach a final moisture content of approximately 2% w/w. Finally, all samples were salted to 1% w/w NaCl (Cargill, MO). The salt was applied in a 5.5M aqueous slurry prior to thermal process, or post roasting with the addition of 2% w/w spray of High Oleic Sunflower seed Oil (HOSO) (Cargill, MO), or 2% w/w Aromatic Roasted Peanut Oil (ARPO) (100E, NutrIn, Washington, DC).

Table 13: Overview of experimental design.

raw material						process technology		
Origin	**Type**	**Variety**	**Grade**[a]	**Count per 100 grams**	**High oleic**	**Maceration**	**Thermal treatment**	**Topical application**
Argentina	Runner	Tegua	M	141/177	N	none	Fry 150°C	None
Argentina	Runner	Granoleic	J	134/148	Y	Water, 90min	Convection bake 145°C	HOSO [b]
Argentina	Runner	Granoleic	J	134/148	Y	1.5% w/w dextrose, 30min	Convection bake 135°C	HOSO
Virginia USA	Virginia	mixed	M	148	N	1.5% w/w dextrose, 30min	Convection bake 135°C	HOSO
Argentina	Runner	Granoleic	J	134/148	Y	1.5% w/w dextrose, 30min	Fry 150°C	HOSO
Argentina	Runner	Granoleic	J	134/148	Y	Acetic acid pH 4, 30min	Convection bake 145°C	HOSO
S. Africa	Valencia	CN Natals	S	177	N	Water, 30min	Convection bake 145°C	None
Virginia, USA	Virginia	mixed	M	148	N	Water, 30min	Convection bake 145°C	None
Virginia, USA	Virginia	mixed	M	148	N	Water, 30min	Convection bake 145°C	ARPO [c]
Georgia, USA	Runner	Georgia Green	M	173	N	none	Impingement bake 180°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 180°C	HOSO
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 180°C	ARPO
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 180°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 180°C	None
S. Africa	Valencia	CN Natals	S	177	N	none	Impingement bake 180°C	None
Australia	Virginia	Middleton	XL	71/92	Y	none	Impingement bake 180°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 145°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 145°C	HOSO
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 145°C	ARPO
Texas, USA	Runner	Flavorunner 458	M	141/177	Y	none	Impingement bake 145°C/180°C	None
Georgia, USA	Runner	Georgia Green	M	173	N	none	Impingement bake 145°C/180°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 145°C/180°C	None
Virginia, USA	Virginia	mixed	M	148	N	none	Impingement bake 145°C/180°C	None
Texas, USA	Runner	Flavorunner 458	J	134/148	Y	none	Impingement bake 145°C/180°C	None
Argentina	Runner	Granoleic	J	134/148	Y	none	Impingement bake 145°C/180°C	None
China	Runner	Hsuji	M	141/177	N	none	Impingement bake 145°C/180°C	None

[a] Grade key: J: Jumbo, XL: extra-large, L: Large, M: Medium, S: small. [b] High Oleic Sunflower seed Oil. [c] Aromatic Roasted Peanut Oil.

Instrumental, sensory and consumer testing were run concurrently in order to minimize ageing effects, and typically completed between 3 and 6 weeks from the production day. The samples were packaged in 200g high oxygen and light barrier metalized bags, flushed with nitrogen gas and stored at room temperature in light proof corrugated cardboard boxes.

5.2.2 Sensory analysis

Flavour and texture profiles were obtained using Descriptive Sensory Analysis (DSA) in duplicate, with water and non-salted crackers provided as palate cleansers. Samples were equilibrated at room temperature for 24h and 400g were used for the texture panel. For the flavour and aroma profile, 600g were ground to a paste with a food processor (Cuisinart DLC- 7 with cutting blade DLC-001, Cuisinart, E Windsor, NJ) by processing for 3 minutes, scraping the sides at 1.5, 2.5 and 3 minutes. The same equilibration and grinding procedure was also used for the physical and chemical analyses. Flavour and aroma attributes were thus evaluated on peanut paste in order to ensure sample uniformity and address potential individual kernel maturity differences or defects (Sanders, Vercellotti, Crippen, & Civille, 1989; Schirack, Drake, Sanders, & Sandeep, 2006a). Texture was evaluated in whole peanuts, while no appearance attributes were analysed by the panel.

A trained panel (USDA, ARS, Market Quality and Handling Research Unit (Raleigh, NC), 7f:3m, mean age 33 y.o., with a long experience in peanut profiling experience) utilized the Spectrum ^{TM} method (Meilgaard, Civille, & Carr, 1999). Three sessions of two hours each preceded the analysis for language and sample calibration. Ten samples were analysed per 90min session, and each sample was seen by all panellists at least twice, with a third replicate being introduced in cases where the panel averages differed by more than one point. Indeed, the setup resulted in good reproducibility ($P<0.01$ for sample effects, $P>0.6$ for panellist effects; F-statistic), and the analysis took two weeks to complete. The panel has been operational since 1988, and so the lexicon had been previously defined (Johnsen, Civille, Vercellotti, Sanders, & Dus, 1988), and continuously refined over the last 35 years (Lee & Resurreccion, 2006b; Schirack et al., 2006a). An overview of the attributes used in this study is reported in **Table 14**.

Table 14: Summary of texture and flavour attributes as obtained from the expert panel.

sensory attribute	description
crispy sounds	degree (volume) to which the sample makes a high-pitched sound [a]
crunchy sounds	degree (volume) to which a sample makes a low pitched sound [z]
hardness	amount of force required initially to bite/fracture the sample using the molars [a]
breakdown	degree to which the sample breaks apart using the molars on the first bite [a]
roasted peanut	the aroma associated with medium roast peanuts (3-4 on USDA colour chips), and having fragrant character such as methyl pyrazine [b]
sweet aroma	the aromas associated with sweet material such as caramel, vanilla, molasses, fruit (specify type) [b]
dark roast	the aroma associated with dark roasted peanuts (4+ on USDA colour chips) and having very browned or toasted character [b]
raw beany	the aroma associated with light roast peanuts (1-2 on USDA colour chips) and having legume like character (specify beans or pea if possible) [b]
woody, hulls, skins	the aroma associated with base peanut character (absence of fragrant top notes) and related to dry wood, peanut hulls and skins [b]
cardboard	the aroma associated with somewhat oxidized fats and oils and reminiscent of cardboard [b]
earthy	the aroma associated with wet dirt and mulch [b]
painty	the aroma associated with linseed oil, oil based paint [b]
phenolic/chemical	aroma associated with chemical/plastic/band aid [c]
fruit fermented	the aroma associated with over ripe or sweet fermenting fruit [c]
ashy	the aroma associated with ash-tray without tobacco notes [c]
total off note	intensity rating of total off notes [c]
malty	the aroma associated with malted milk
peppery	the aroma associated with black peppercorns
sweet	the taste on the tongue associated with sugars [b]
sour	the taste on the tongue associated with acids [b]
bitter	the taste on the tongue associated with bitter agents such as caffeine or quinine [b]
salty	the taste on the tongue associated with sodium ions [b]
tongue, throat burn	the chemical feeling factor on the tongue and throat associated with burning (benzoate) [c]
metallic	the chemical feeling factor on the tongue described as flat, metallic and associated with iron and copper [b]
astringent	the chemical feeling factor on the tongue, described as puckering/dry and associated with tannins or alum [b]

[a] (Lee & Resurreccion, 2006b) [b] (Johnsen et al., 1988) [c] (Schirack et al., 2006b)

5.2.3 Headspace volatile composition analysis

The headspace volatiles where profiled using SPME-GC-MS-O (Agilent Technologies 7890A GC system with 5975C inert MSD – triple axis detector, a Gerstel MPS2XL auto sampler, and a dB5-MS semi-polar capillary column, 60m/0.32mm diameter/1µm film thickness (Mülheim an der Ruhr, DE)). 0.2g of each peanut paste was added to a sample vial (Fischerbrand FB67515; caps Gerstel 093640-040-00). One µL each of 1,2-Dichlorobenzene (13.06µg/mL) and heptanone-d5 (100µg/mL) in methanol internal standards were introduced and left to equilibrate for 2 hours, before 8mL of saturated NaCl solution was added and left on an agitator table (40rpm) for 8 hours at 20°C. Unless otherwise stated, all

materials in this study were sourced from Sigma-Aldrich (St Louis, MO). The vials where further agitated for 5 minutes at 60°C by the auto sampler before the SPME fibre was introduced (2cm PDMS/DVB/Carboxen fibre) and further agitating at 250rpm for 50min. The analytes were desorbed at 260°C, while the temperature profile started at 40°C for 7.5 minutes and ramping up to 200°C at 4°C/min, followed by a sharp ramp to 320°C to clear the column at the end of each run. Helium gas (2.5mL/min) was used as the solvent, and the ionization voltage was set to 70eV, with a scan range from m/z 33 to 400. A flow splitter diverted part of the eluent stream to a sniff port, where an operator recorded the character and intensity of the odor (4 point scale). The process was repeated twice, by two different operators, and the intensity ratings were averaged. A C5-C25 n-alkane sample was run prior to each sample, to assist in calculating the Linear Retention Indices. The MSD Productivity ChemStation software (Welmington, DE) was used to compare the retention indices to the NIST 2015 mass spectral database (Boulder, CO) and return the tentative identification of the compounds. Semi-quantification was obtained by comparing the peak area of the 146 m/z ion vs the internal standard. All samples were analysed in duplicate, and the median coefficient of variation across replicates for all peaks was 0.07.

5.2.4 Sugar profile analysis

The fructose, glucose and sucrose contents were determined by ion chromatography. A Dionex ICS 3000 chromatographer was used, fitted with the CarboPac 20 analytical column 3mmx150mm, CarboPac PA20 guard (3mmx30mm) and Borate trap (4mmx50mm) columns, a ICS-3000 eluent generator and an ICS-3000 dual pump (Thermo-Fisher, Breda, BE). The eluent was 52 mM NaOH at 0.39 ml/min. The column and detector temperatures were set to 25°C. Two electrochemical detectors were used: the gold working electrode and the pH Ag/AgCl combination reference electrode, while the EC wave form was set to carbohydrate mode. One gram of the paste sample was stirred in 50mL of water and placed in a sonic bath for 10min, before being centrifuged at 9000rpm for 20min. 0.2ml of the supernatant was diluted with 10 ml of water, and 20µL of the solution was injected into the column. Quantification was performed by comparing to previously prepared five point calibration curve of glucose, fructose (0.05, 0.2, 0.5, 1, 5 mg/L), sucrose (0.1, 0.4, 1, 2, 10 mg/L) and combined fructose, glucose and sucrose (1:1:2 mg/L) solutions, with the aid of the Chromeleon data analysis software (ver 6, Thermo Fisher, Breda, BE) in duplicate.

5.2.5 Large deformation texture analysis

Textural characteristics were derived using large deformation compression tests with a TA-XT2 Texture Analyser (Stable Microsystems, Godalming, UK) with two probes used separately (P/2 and Volodkevitch Bite Jaw (VBJ) ('hardness'-peak force in newtons- and 'toughness'- work of deformation in millijoules). Twenty single cotyledons from each sample were analysed (flat side down) at a compression rate of 2.00 mm/s, the start point defined by a trigger force of 0.05N and the end point set at 2mm and 3mm deformation for the P/2 and VBJ probes respectively, at a data acquisition rate of 500 points per second. The measurements were consequently averaged. The median coefficient of variation across replicates for all samples for the hardness was 0.14 and 0.15 and for the toughness 0.27 and 0.26 for the VBJ and P/2 probes respectively.

5.2.6 Colour and size analysis

The CIELAB colour parameters were obtained using a Hunter Lab CR400 colorimeter (Reston, VA). A 250 gram sample was placed on a 95mm diameter petri dish, and measured by the instrument from above in triplicate, with the kernels being redistributed between measurements. To obtain the moisture content, a three gram ground sample was placed in a Leco TGA701 thermogravimetric analyser (St. Joseph, MI), and heated to 113°C until a constant weight was observed. Finally the size distribution was determined by manually sieving a 500g sample through sieves with progressively smaller rectangular-shaped openings, and calculating the weight percentage on each sieve. The fraction not passing through a sieve opening of 20x8mm was designated as 'large'. Of the remainder fraction, the peanuts not passing through a sieve of 20x6.2mm was designated as 'small', while the fraction passing through 20x6.2mm openings was mainly split peanuts (breakage).

5.2.7 Consumer testing

Consumer testing was carried out in three countries (Spain, The Netherlands and Turkey), selected for their geographic dispersion. Two-hundred-ten untrained consumers were interviewed in each country, equally split between different cities: for Spain, Madrid and Barcelona, for Turkey, Istanbul and Ankara, and for The Netherlands, Amsterdam and Utrecht. The respondents were filtered

to ensure 50:50 gender split, 30:30:40 18-34yo:34-54yo:55+yo age split, 100% non-rejecters of salted flavour, and 100% peanut users defined as 'used once in last 3 months', of which 50% had consumed peanuts in the last month.

To accommodate for the large number of samples the sample set was divided into five sets of six samples evaluated on consecutive days, each containing five unique samples and one 'dummy'/'warm-up', identical to all sets. The presentation order was randomized across respondents and sessions, with the exception of the warm-up sample which was presented first in all but the first day to anchor (Macfie, 2007). Twenty five grams of each sample was presented to the subjects in a white plastic plate, identifiable only by a 3 digit code.

The survey ballot included hedonic questions in the areas of appearance, flavour and texture, in order to provide the subjects with options for describing the difference between samples, and prevent 'sensory dumping' (Lawless & Heymann, 1998), and was translated to the local language. Although more questions appeared on the ballot, responses for only one hedonic ('How well do you like the sample overall?' (9 point scale)) and one multiple choice ('Check All That Apply) questions were included in the analysis. The CATA question was 'Which of the following words do you connect with the product you just tasted?', and included the following options: 'Looks tasty', 'Oily', 'Matte appearance', 'Shiny appearance', 'Has an unusual shape', 'Has an unusual size', 'Variable colour', 'Dark roast', 'Bland', 'Burnt', 'Fresh', 'Stale/old', 'Raw', 'Chewy', 'Dry', 'Hard', 'Smooth texture', 'Gritty texture', 'Sweet', 'Bitter', 'Sour', 'Salty'. However, the analysis was focused on the 'Fresh' and 'Stale/old' attributes, hereafter referred to as 'fresh' and 'stale'.

5.2.8 Statistical analysis

To aid in the sample set selection from the pool of 134 process-material combinations previously described (Lykomitros, Fogliano, & Capuano, 2016a), a combination of statistical (Agglomerative Hierarchical Clustering of flavour and texture profiles; Covariate, Ward Linkage, Euclidean Distance centroids selected) and empirical algorithms (expert opinion; ensure representation of all grades and process methods) (data not shown), were used. This was performed in order to ensure large organoleptic differentiation of the sample set as it facilitates consumer differentiation and can improve the quality of the preference maps (Lawless & Heymann, 1998).

Pearson's correlation matrix and the Cronbach's alpha coefficient were calculated to visualize the similarity in consumer preference between the three countries. The standard External Preference Map on mean liking methodology was

used to relate sensory attributes to consumer preference for each country (Macfie, 2007; Meilgaard et al., 1999). The sample space used for the consumer preference map was obtained by running a Factor Analysis (Pearson's correlation matrix) on the sensory profiles, instrumental colour and size analytical results, followed by a VariMax rotation of the first three factors (hereafter indicated as D1, D2 and D3). The average consumer liking scores for each country were modelled against the three rotated factors identified above to determine the vector of consumer liking. This was done using a quadratic model (**Equation 1**), and the model variables were reduced by a stepwise selection algorithm (entry P=0.15, exit P=0.15, confidence interval=95%).

$$Liking_j = a_j \times D1 + b_j \times D2 + c_j \times D3 + d_j \times D1 \times D2 + e_j \times D2 \times D3 + f_j \times D1D3 + g_j \times D1^2 + h_j \times D2^2 + k_j \times D3^2 + intercept_j$$

j= j[th] country. a,b,c,d,e,f,g,h,k, and the intercept are the model parameters corresponding to each combination of factor 1 (D1), 2 (D2) and 3 (D3).

Equation 1

Consumer segmentation for each country was tested separately by running Agglomerative Hierarchical Clustering analyses (Euclidean distance, Ward's method, centred and reduced, Entropy truncation) on the panellist scores for each sample.

In addition to the preference map, Partial Least Squares (PLS) regressions were used to model overall liking, 'freshness' and 'stale' against headspace volatiles (**Equation 2**) and all the instrumental attributes (**Equation 3**). Only 3 factors were included in the PLS models, while the data was auto-scaled and centred.

$$Attribute_i = constant_i + \sum_{44}^{j=1} \left(concentration_j \times coefficient_{i,j} \right)$$

i= i[th] response attribute, j= the j[th] compound (GC-MS peak), $concentration_j$ = the concentration of the j[th] compound, $coefficient_{i,j}$ = the model coefficient of the j[th] compound in the i[th] model and $constant_i$ = the constant of i[th] model (response attribute).

Equation 2

$$Liking = constant + \sum_{56}^{j=1} \left(Magnitude_j \times coefficient_j \right)$$

j= the j^{th} instrumental attribute, $Magnitude_j$ = the magnitude of the j^{th} instrumental attribute, $coefficient_{i,j}$ = the model coefficient of the j^{th} instrumental attribute.

Equation 3

The continuous (response) variables for 'freshness' and 'stale' were obtained by counting the number of times the attributes were respectively checked for each sample, across all consumers from all countries (Ares, Deliza, Barreiro, Giménez, & Gámbaro, 2010). For clarity of representation, only parameters with a β coefficient > 0.05 in absolute value were reported. All statistical analyses were run on XLSTAT 2017.1 (Addinsoft, Paris, FR on MS Excel 2010, Microsoft, Redmond, WA).

5.3 Results and discussion

An overview of the sensory panel ratings for all the samples is shown in **Figure 25**. The attribute means and attribute by sample interaction significance are listed in **Table 15**. The resulting profiles where highly repeatable (median coefficient of variation between replicates across all samples and sensory attributes was 0.06). A moderate range in the scores of 'roasted peanut', 'sweet', 'dark roast', 'raw bean', 'wood hulls', 'painty', off-flavors', and 'malty' aromas, 'sweet' and 'bitter' tastes and 'crispy', 'crunchy', 'hardness' and 'breakdown' texture attributes is observed, making these attributes a good choice for explanatory variables in the preference models. Several attributes associated with off-flavors, such as 'earthy', 'phenolic-chemical', 'metallic', 'fruity-fermented', 'peppery', 'ashy' as well as 'astringent', 'throat-tongue burn' and sour taste exhibit small magnitude and little variation amongst the samples. This is not surprising considering that there were no defect exemplars in the sample set, and those attributes were thus excluded from the sub-sequent analysis.

The average hedonic scores for all samples ranged from 3.8 to 6.6 on 9pt scale (**Table 16**). This range is consistent with other published peanut studies evaluating a similar number of samples (Young et al., 2005). **Table 17** shows the Pearson's correlation coefficient of the average preference of each sample across the three countries. It is immediately obvious that the order of preference is very similar across the three countries, and indeed the linear regression correlation matrix confirms that the liking order is highly correlated (all correlation coefficients >0.9), with the Dutch and Turkish consumers being the most dissimilar (coefficient

0.904). Due to the high similarity, the data from all three countries were also pooled in the subsequent analysis to show overall trends. To test for consumer

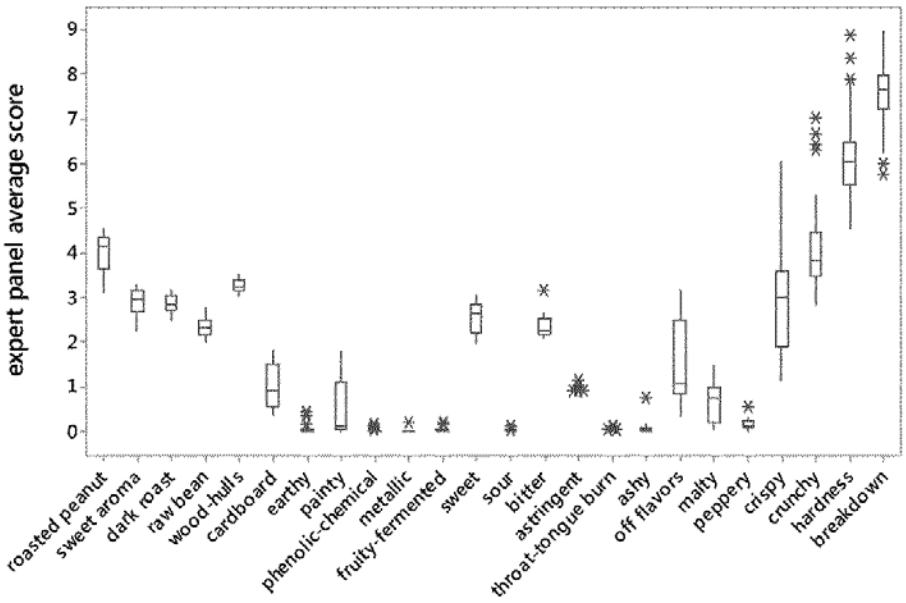

Figure 25: Overview of expert panel sensory scores for the 26 unique peanut samples. Horizontal line: median, box: 1st (Q1) to 3rd (Q3) quartile, upper whisker: Q3+1.5(Q3-Q1), lower whisker: Q1-1.5(Q3-Q1), *: outliers (outside the whisker range).

segmentation, a hierarchical cluster analysis was run for each country. Three clusters were identified for Turkey and Spain, and 4 for the. Although some differences between the sample preferences of the different clusters were detected, they were relative small in magnitude (**Figure 26**). The assumption that the drivers of liking did not significantly vary across the consumer clusters was validated by comparing to models of individual preference (Macfie, 2007) (data not shown). The comparison revealed that the drivers are similar across the clusters, but there are some minor differences in their optima and relative weight. However, the research objective was to determine country and Europe-wide drivers of liking, and for these reasons no clustering was carried forward to the rest of the analysis.

In this research, three sets of preference models were developed. Firstly, a conventional external preference map was developed to link preference data to sensory attributes. Secondly, PLSR was used to introduce additional instrumental analysis attributes into the model. Finally, a separate model focused on the headspace volatile composition only, and how it is related to consumer liking.

Table 15: Mean intensity scores of all sensory attributes from all samples as derived by the expert panel.

	A	B	C	D	E	F	G	H	I	J	K	L	M
roasted peanut[a]	3.1 ^{j}	4.3 ^{abcd}	3.4 ^{ij}	3.3 ^{ij}	3.7 ^{fgh}	3.4 ^{hi}	3.7 ^{fgh}	3.7 ^{fg}	3.3 ^{ij}	4.2 ^{bcd}	4.4 ^{abc}	4.3 ^{abcd}	4.1 ^{cde}
sweet aroma	2.3 ^{j}	3.3 ^{a}	2.3 ^{ij}	2.6 ^{ghi}	2.5 ^{hi}	2.7 ^{efgh}	2.7 ^{efgh}	2.7 ^{fgh}	2.5 ^{hi}	3.0 ^{bcd}	3.2 ^{abc}	2.8 ^{def}	3.1 ^{abc}
dark roast	2.7 ^{fghi}	3.0 ^{abcd}	2.6 ^{ghi}	2.7 ^{fghi}	2.8 ^{efgh}	2.6 ^{hi}	2.9 ^{abcde}	2.8 ^{defg}	2.6 ^{ghi}	3.0 ^{abcde}	3.0 ^{abcd}	2.8 ^{efgh}	3.1 ^{ab}
row beany	2.6 ^{bcd}	2.2 ^{klmno}	2.5 ^{defg}	2.7 ^{ab}	2.5 ^{cdef}	2.6 ^{abc}	2.3 ^{fghijk}	2.5 ^{bcde}	2.8 ^{a}	2.2 ^{hijklm}	2.1 ^{lmno}	2.4 ^{efgh}	2.2 ^{jklmno}
wood, hulls, skins	3.5 ^{ab}	3.2 ^{bcdefgh}	3.4 ^{abc}	3.3 ^{abcdefg}	3.4 ^{abc}	3.3 ^{bcdefgh}	3.4 ^{abc}	3.4 ^{abcd}	3.5 ^{a}	3.1 ^{efgh}	3.2 ^{cdefgh}	3.2 ^{cdefgh}	3.3 ^{abcdef}
ardboard	1.8 ^{a}	0.5 ^{g}	1.5 ^{abc}	1.5 ^{abc}	1.1 ^{abcdefg}	1.5 ^{abcd}	1.8 ^{a}	1.7 ^{a}	1.6 ^{ab}	0.5 ^{g}	0.7 ^{efg}	1.1 ^{abcdefg}	1.4 ^{abcde}
earthy	0.0 ^{c}	0.0 ^{c}	0.1 ^{abc}	0.4 ^{ab}	0.1 ^{abc}	0.0 ^{c}	0.1 ^{bc}	0.0 ^{c}	0.1 ^{bc}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}
painty	1.4 ^{ab}	0.0 ^{c}	1.7 ^{a}	1.7 ^{a}	1.1 ^{b}	1.1 ^{b}	1.0 ^{b}	1.1 ^{b}	1.8 ^{a}	0.1 ^{c}	0.1 ^{c}	0.1 ^{c}	0.2 ^{c}
phenolic-chemical	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.1 ^{ab}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.2 ^{a}
metallic	0.0 ^{b}	0.0 ^{b}	0.2 ^{a}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
fruit fermented	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.2 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}
sweet	2.0 ^{j}	2.8 ^{abcde}	2.2 ^{ij}	2.3 ^{hi}	2.2 ^{ij}	2.2 ^{ij}	2.2 ^{hij}	2.2 ^{ij}	2.2 ^{ij}	2.7 ^{def}	2.9 ^{abcd}	2.4 ^{ghi}	2.5 ^{fgh}
sour	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
bitter	3.2 ^{a}	2.2 ^{e}	2.6 ^{b}	2.5 ^{bc}	2.7 ^{b}	2.6 ^{b}	2.6 ^{bc}	2.6 ^{b}	2.5 ^{bc}	2.2 ^{e}	2.2 ^{e}	2.3 ^{de}	2.6 ^{b}
astringent	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	0.9 ^{b}	1.0 ^{b}	1.0 ^{b}	1.2 ^{a}	0.9 ^{b}	1.0 ^{ab}	0.9 ^{b}	1.0 ^{b}
tongue, throat burn	0.1 ^{a}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
ashy	0.1 ^{bc}	0.0 ^{c}	0.0 ^{c}	0.1 ^{bc}	0.1 ^{bc}	0.1 ^{bc}	0.1 ^{bc}	0.1 ^{bc}	0.1 ^{bc}	0.0 ^{c}	0.1 ^{bc}	0.0 ^{c}	0.8 ^{a}
total offnote	3.0 ^{ab}	0.5 ^{ef}	3.2 ^{a}	2.8 ^{ab}	2.0 ^{bcd}	2.5 ^{ab}	2.7 ^{ab}	2.5 ^{ab}	2.8 ^{ab}	0.4 ^{f}	0.9 ^{ef}	1.1 ^{def}	2.2 ^{abc}
malty	0.3 ^{def}	1.5 ^{a}	0.2 ^{def}	0.2 ^{def}	0.1 ^{f}	0.2 ^{def}	0.1 ^{ef}	0.2 ^{ef}	0.1 ^{f}	0.8 ^{bc}	1.1 ^{ab}	0.6 ^{cd}	0.5 ^{cde}
peppery	0.3 ^{ab}	0.3 ^{ab}	0.0 ^{b}	0.1 ^{b}	0.2 ^{b}	0.6 ^{a}	0.3 ^{ab}	0.0 ^{b}	0.1 ^{b}	0.1 ^{b}	0.3 ^{ab}	0.3 ^{ab}	0.1 ^{b}
crispy	3.4 ^{de}	5.5 ^{b}	2.0 ^{hij}	1.5 ^{jkl}	3.5 ^{de}	4.0 ^{c}	1.7 ^{ijk}	1.4 ^{kl}	1.6 ^{ijk}	6.1 ^{a}	5.6 ^{ab}	1.1 ^{l}	2.4 ^{gh}
crunchy	3.9 ^{efghi}	6.3 ^{b}	3.5 ^{ijk}	3.5 ^{ijk}	5.3 ^{c}	4.4 ^{de}	3.5 ^{hijk}	3.3 ^{jkl}	3.2 ^{kl}	7.0 ^{a}	6.4 ^{b}	2.8 ^{l}	3.6 ^{ghijk}
hardness	6.8 ^{c}	8.9 ^{a}	5.4 ^{ghi}	5.0 ^{hij}	6.4 ^{cd}	6.2 ^{cde}	5.6 ^{efghi}	4.9 ^{ij}	5.1 ^{hij}	8.4 ^{ab}	7.9 ^{b}	4.5 ^{j}	5.5 ^{fghi}
breakdown	6.8 ^{ijk}	5.8 ^{m}	7.9 ^{cdefg}	8.1 ^{cde}	6.9 ^{hij}	7.4 ^{gh}	8.0 ^{cdef}	9.0 ^{a}	8.2 ^{bcd}	6.0 ^{lm}	6.3 ^{jkl}	8.8 ^{ab}	8.3 ^{bc}

[a] Different letters within each row denote significant differences in the means between samples (P <0.05, Fisher LSD).

Table 15, Continued.

	N	O	P	Q	R	S	T	U	V	W	X	Y	Z
roasted peanut[a]	3.5 ^{ghi}	4.3 ^{abcd}	4.5 ^{a}	4.1 ^{de}	4.4 ^{ab}	4.3 ^{abcd}	4.3 ^{abcd}	4.4 ^{ab}	4.5 ^{a}	4.1 ^{de}	4.1 ^{cde}	4.3 ^{abcd}	3.9 ^{ef}
sweet aroma	2.7 ^{efgh}	3.2 ^{ab}	3.2 ^{abc}	2.8 ^{defgh}	3.3 ^{a}	3.2 ^{ab}	3.1 ^{abc}	3.1 ^{abc}	3.1 ^{abc}	2.9 ^{cde}	3.0 ^{bcd}	3.2 ^{ab}	2.8 ^{defg}
dark roast	2.5 ^{i}	3.1 ^{abc}	3.0 ^{abcde}	2.8 ^{efgh}	3.2 ^{a}	3.1 ^{ab}	3.1 ^{ab}	2.9 ^{cdef}	3.0 ^{abcde}	2.8 ^{defg}	2.8 ^{defg}	2.8 ^{efgh}	2.9 ^{bcde}
row beany	2.6 ^{bcd}	2.0 ^{o}	2.1 ^{lmno}	2.4 ^{efghij}	2.0 ^{no}	2.1 ^{mno}	2.1 ^{mno}	2.3 ^{hijkl}	2.2 ^{ijklmn}	2.4 ^{efgh}	2.3 ^{ghijkl}	2.4 ^{efgh}	2.4 ^{efgh}
wood, hulls, skins	3.1 ^{gh}	3.2 ^{cdefgh}	3.2 ^{cdefgh}	3.3 ^{bcdefgh}	3.2 ^{cdefgh}	3.2 ^{cdefgh}	3.4 ^{abcde}	3.2 ^{defgh}	3.2 ^{defgh}	3.1 ^{gh}	3.1 ^{fgh}	3.0 ^{h}	3.4 ^{abcd}
ardboard	0.9 ^{bcdefg}	0.5 ^{g}	0.5 ^{g}	1.3 ^{abcdef}	0.8 ^{cdefg}	0.8 ^{defg}	0.6 ^{g}	0.6 ^{fg}	0.4 ^{g}	0.8 ^{cdefg}	0.9 ^{bcdefg}	0.9 ^{cdefg}	1.3 ^{abcde}
earthy	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.0 ^{c}	0.2 ^{abc}	0.0 ^{c}	0.1 ^{bc}	0.0 ^{c}	0.0 ^{c}	0.4 ^{a}
painty	0.0 ^{c}	0.1 ^{c}	0.1 ^{c}	0.2 ^{c}	0.0 ^{c}	0.2 ^{c}	0.1 ^{c}	0.1 ^{c}	0.0 ^{c}	0.1 ^{c}	0.0 ^{c}	0.0 ^{c}	0.1 ^{c}
phenolic-chemical	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
metallic	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
fruit fermented	0.0 ^{b}	0.0 ^{b}	0.2 ^{a}	0.0 ^{b}	0.1 ^{ab}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}
sweet	3.0 ^{ab}	2.6 ^{defg}	2.8 ^{bcde}	2.6 ^{efg}	2.8 ^{abcde}	2.7 ^{cdef}	2.7 ^{cdef}	2.9 ^{abcde}	3.0 ^{abc}	3.0 ^{ab}	2.9 ^{abcd}	3.1 ^{a}	2.4 ^{ghi}
sour	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{a}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}
bitter	2.2 ^{e}	2.3 ^{de}	2.2 ^{e}	2.3 ^{cde}	2.2 ^{e}	2.2 ^{e}	2.2 ^{e}	2.2 ^{e}	2.2 ^{e}	2.1 ^{e}	2.2 ^{e}	2.1 ^{e}	2.5 ^{bcd}
astringent	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}	0.9 ^{b}	1.0 ^{b}	1.0 ^{b}	1.0 ^{b}
tongue, throat burn	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.1 ^{ab}	0.0 ^{b}	0.0 ^{b}	0.0 ^{b}
ashy	0.0 ^{c}	0.1 ^{bc}	0.1 ^{bc}	0.0 ^{c}	0.1 ^{bc}	0.1 ^{bc}	0.2 ^{b}	0.1 ^{bc}	0.1 ^{bc}	0.1 ^{bc}	0.0 ^{c}	0.1 ^{bc}	0.1 ^{bc}
total offnote	1.0 ^{def}	0.7 ^{ef}	0.9 ^{ef}	1.5 ^{cde}	0.9 ^{ef}	1.1 ^{def}	0.8 ^{ef}	0.8 ^{ef}	0.4 ^{f}	1.0 ^{ef}	0.9 ^{ef}	0.7 ^{ef}	2.0 ^{bcd}
malty	1.0 ^{b}	0.8 ^{bc}	1.1 ^{ab}	0.6 ^{cd}	1.1 ^{ab}	0.8 ^{bc}	1.1 ^{ab}	0.9 ^{bc}	0.8 ^{bc}	0.9 ^{bc}	1.0 ^{b}	0.9 ^{bc}	0.1 ^{f}
peppery	0.1 ^{b}	0.1 ^{b}	0.1 ^{b}	0.0 ^{b}	0.1 ^{b}	0.1 ^{b}	0.1 ^{b}	0.0 ^{b}	0.1 ^{b}	0.2 ^{b}	0.0 ^{b}	0.3 ^{ab}	0.2 ^{b}
crispy	5.7 ^{ab}	2.9 ^{fg}	3.1 ^{ef}	2.1 ^{hi}	2.9 ^{f}	3.5 ^{de}	3.3 ^{def}	3.8 ^{cd}	2.3 ^{h}	3.4 ^{de}	2.3 ^{h}	1.8 ^{ijk}	3.5 ^{de}
crunchy	6.7 ^{ab}	4.1 ^{efgh}	4.2 ^{ef}	3.6 ^{ghijk}	3.9 ^{efghi}	4.8 ^{cd}	4.2 ^{efg}	3.5 ^{ijk}	3.6 ^{hijk}	3.8 ^{fghij}	4.1 ^{efgh}	3.7 ^{ghijk}	3.8 ^{fghijk}
hardness	7.8 ^{b}	5.9 ^{defg}	6.1 ^{cdefg}	5.9 ^{defg}	6.0 ^{defg}	6.7 ^{c}	6.2 ^{cde}	6.1 ^{cdef}	5.9 ^{defg}	6.1 ^{cdef}	6.0 ^{defg}	5.7 ^{efgh}	6.2 ^{cde}
breakdown	6.3 ^{klm}	7.5 ^{efg}	7.6 ^{defg}	7.8 ^{cdefg}	8.0 ^{cdef}	7.3 ^{ghi}	7.5 ^{fg}	7.8 ^{cdefg}	7.5 ^{efg}	7.7 ^{defg}	7.5 ^{efg}	7.8 ^{cdefg}	7.8 ^{cdefg}

[a] Different letters within each row denote significant differences in the means between samples (P <0.05, Fisher LSD).

Table 16. Mean liking scores across the three countries.

	Netherlands			Spain			Turkey		
Sample	Liking	Checked Fresh	Checked Stale	Liking	Checked Fresh	Checked Stale	Liking	Checked Fresh	Checked Stale
A[a]	4.3 [l]	7%	18%	3.8 [n]	3%	29%	3.8 [m]	12%	14%
B	4.6 [k]	11%	8%	4.7 [l]	4%	18%	4.4 [l]	17%	7%
C	4.6 [kl]	11%	28%	4.3 [m]	9%	30%	4.4 [l]	18%	19%
D	5.0 [ij]	16%	30%	4.9 [jkl]	10%	26%	4.6 [jkl]	15%	15%
E	4.8 [jk]	11%	33%	4.7 [kl]	9%	24%	4.6 [kl]	21%	13%
F	5.1 [i]	14%	20%	5.0 [ijk]	7%	25%	4.7 [ijkl]	23%	12%
G	5.1 [i]	11%	31%	5.0 [ijk]	12%	21%	4.9 [ijk]	23%	17%
H	5.2 [i]	20%	25%	5.0 [ij]	18%	19%	4.9 [hij]	23%	17%
I	5.0 [ij]	13%	28%	4.9 [jk]	13%	25%	4.7 [ijkl]	23%	21%
J	5.5 [h]	21%	5%	5.3 [h]	9%	10%	5.0 [hi]	26%	5%
K	5.5 [h]	21%	6%	5.2 [hi]	6%	11%	5.0 [hi]	23%	7%
L	5.6 [fgh]	25%	13%	5.7 [fg]	15%	12%	5.6 [fg]	29%	9%
M	5.5 [gh]	23%	14%	5.4 [h]	14%	10%	5.3 [gh]	35%	5%
N	5.5 [gh]	27%	10%	5.5 [gh]	23%	10%	5.4 [fg]	38%	5%
O	5.6 [fgh]	30%	11%	5.8 [efg]	17%	7%	5.6 [fg]	36%	8%
P	5.8 [def]	28%	5%	6.0 [de]	17%	8%	6.0 [de]	35%	4%
Q	6.1 [cd]	36%	9%	6.2 [cd]	12%	15%	6.2 [abcd]	37%	8%
R	5.7 [efgh]	28%	12%	5.8 [ef]	12%	10%	5.6 [f]	44%	7%
S	6.1 [bcd]	33%	6%	6.3 [bc]	12%	7%	6.3 [abcd]	38%	4%
T	6.0 [cd]	36%	3%	6.0 [cde]	18%	6%	6.2 [bcd]	40%	3%
U	5.9 [cde]	32%	7%	6.0 [cde]	26%	6%	6.1 [cd]	46%	4%
V	5.8 [efg]	34%	8%	6.0 [de]	21%	9%	6.0 [de]	46%	6%
W	5.7 [efgh]	28%	12%	5.9 [ef]	26%	6%	5.7 [ef]	39%	9%
X	6.4 [ab]	45%	2%	6.5 [ab]	31%	3%	6.5 [ab]	49%	6%
Y	6.2 [abc]	47%	5%	6.5 [ab]	26%	2%	6.4 [abc]	46%	5%
Z	6.4 [a]	45%	5%	6.6 [a]	29%	6%	6.6 [a]	54%	3%

[a] Different letters within each column denote significant differences in the means between samples (P <0.05, Fisher LSD).

Table 17. Pearson's correlation coefficients of overall liking scores across countries. Cronbach's alpha = 0.973.

	NL	ESP	TR
NL	1	0.921	0.904
ESP	0.921	1	0.942
TR	0.904	0.942	1

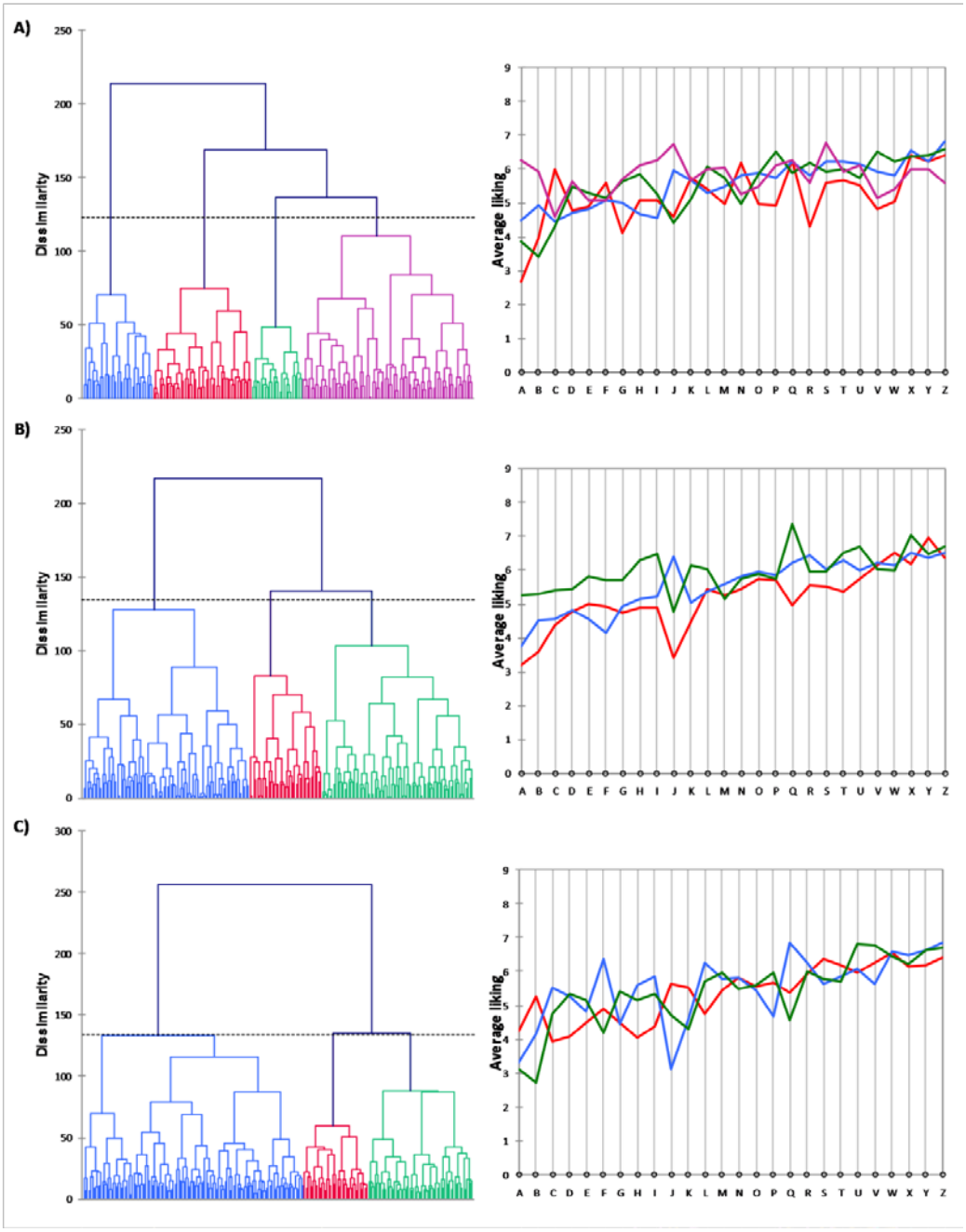

Figure 26: Consumer segmentation details: Dissimilarity dendrograms (left) and profile plots (right) showing the average liking for each sample by each consumer cluster. Different colours denote different consumer clusters. Panel A: Netherlands, Panel B: Spain, Panel C: Turkey.

5.3.1 External preference map

The VariMax rotated factor analysis that summarizes the sensory, colour and size data can be seen in **Figure 27**, with three factors capturing 78.4% of the variation. The VariMax rotation was used in order facilitate the interpretation of the factors, as it is designed to maximize the sum of variances of the loadings, so that eigenvectors are either very, or not at all correlated with principal factors (Lawless & Heymann, 1998). The figure shows that factor D1 captures mainly flavour and taste sensory attributes and the b* CIELAB colour value, D2 captures mainly sensory texture and the remaining colour values, and D3 mainly size and 'ashy' aroma.

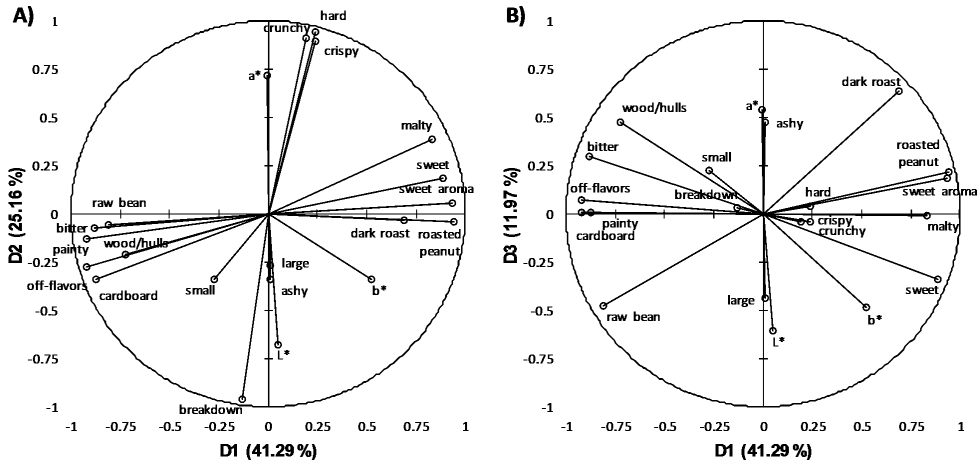

Figure 27. Graphical representation of the first 3 VariMax-rotated factors used in the preference map (78.4% of variation captured). Panel A: Factors D1 and D2, 66.5% variance captured; Panel B: Factors D1 and D3, 53.3% variance captured.

This grouping makes intuitive sense: D1 separates 'favourable' from 'unfavourable' aromatics, with the colour b* value correlating with roasted aromatics,. D2 separates 'hardness', 'crispiness' and 'crunchiness' against 'breakdown' D3 resolves physical characteristics (size), which are orthogonal to most other attributes. Finally, 'ashy' and size are inversely correlated since small kernels are more prone to scorching during roasting due to their higher surface to volume ratio''.

The correlation between colour and flavour (shown in D1) has previously observed in whole peanuts (Lykomitros et al., 2016a) as well as peanut butter (Tomlins, Rukuni, Mutungamiri, Mandeya, & Swetman, 2007). Low L* values (dark) and high a* (reddish) CIELAB values are correlated with higher melanoidin

production from Maillard reaction and lower finished moisture content. Both parameters are associated with increased crispiness/crunchiness (Açar, Gökmen, Pellegrini, & Fogliano, 2009; Pattee, Sanders, Isleib, & Giesbrecht, 2001), and it is thus not surprising these CIELAB values are also resolved by the 'texture' factor (D2).

The three liking models are shown in **Figure 28** and their parameters reported in **Table 18**.

It can be seen that the three models are of different orders. D1 (flavour) and D2 (texture and colour) are significant in the linear or quadratic power in all models, suggesting texture and flavour are important drivers of liking in all countries. **Figure 28** shows that 'roasted' and 'sweet' aromas are correlated with consumer liking in all countries, particularly so in the Netherlands and Turkey (panels A and C). This is not surprising as both roasted and sweet aromas are generally considered pleasant, while the relationship has also been previously reported with American consumers (Grosso & Resurreccion, 2002; Lee & Resurreccion, 2006a; Wang et al., 2017; Young et al., 2005). The same figure shows that 'crispy', 'crunchy', 'hardness' and 'breakdown' are very much correlated with each other. The correlation between 'crispiness' and 'crunchiness' has also been previously reported (Wang et al., 2017), suggesting that either the attributes are collinear (i.e. peanuts that are crunchy are also crispy, hard and with slow breakdown) or that the terms are not entirely distinguished by the expert panel. A significant impact of texture (D2) on liking can be expected, since crunchy and crispy texture has been linked to increased appeal and enjoyment in several food categories for western consumers (Tunick et al., 2013).

However, all texture attributes cross the same contour lines more than once in **Figure 28(A,C)**, suggesting that optimum hardness, crispiness, crunchiness and breakdown exist. Interestingly, factor D3 (size) is significant for the Netherlands and Spain but not for Turkey **(Table 18**), which is likely the reason for the slight dissimilarity of Turkey demonstrated in **Table 17**. No link between size and preference was found in the surveyed scientific literature, although anecdotal references for British consumers have been made by peanut brokers (Prusak, Schlegel-Zawadzka, Boulay, & Rowe, 2014). Finally, flavour and texture (factors D1 and D2) in The Netherlands and texture and colour (factor D2) in Turkey appear at the second power with a negative coefficient, liking exhibits an optimum for some flavour and texture intensities. In other words, more intense flavour or texture does not always lead to higher liking scores. This contradicts earlier studies with US consumers, where liking was seen to be linearly correlated to peanut aroma intensity (Young et al., 2005), however it is in good agreement with a more recent US consumer study where over-roasting was shown to be a dissatisfying factor (Wang et al., 2017).

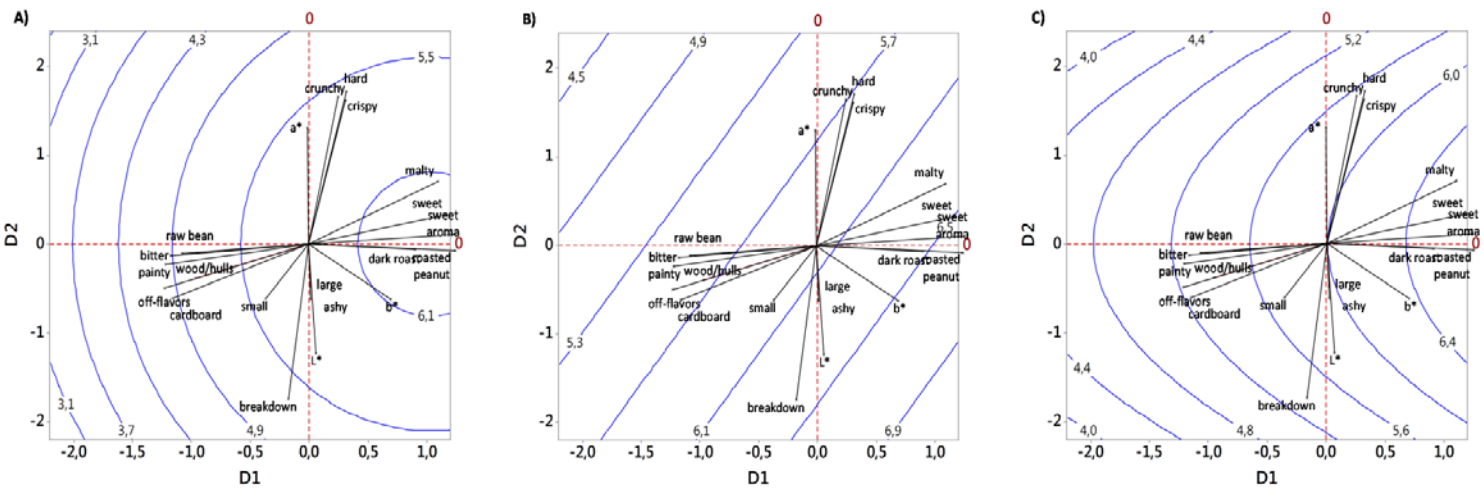

Figure 28. Two-factor visualization (D1 and D2, D3 fixed at -2.0) of the preference models for each of the three countries. Contours: hedonic scores; vectors: sensory and appearance attributes. Panel A: Netherlands; Panel B: Spain; Panel C: Turkey.

Table 18. Preference map (Liking against VariMax factors) model coefficients and significance (only parameters significant at P=0.1 are shown). The model is shown in Equation 1.

		Netherlands			Spain			Turkey	
Coefficient	**Value**	SE [a]	P-value	**Value**	SE [a]	P-value	**Value**	SE [a]	P-value
Intercept	**5.915**	0.109	< 0,0001	**5.497**	0.055	< 0,0001	**5.572**	0.083	< 0,0001
D1	**0.274**	0.075	0.001	**0.491**	0.056	< 0,0001	**0.602**	0.071	< 0,0001
D2	-	-	n.s. [b]	**-0.269**	0.084	0.004	-	-	n.s.
D3	-	-	n.s.	**-0.261**	0.060	<0.001	**-0.210**	0.073	0.008
D1×D2	-	-	n.s.	-	-	n.s.	-	-	n.s.
D2×D3	-	-	n.s.	-	-	n.s.	-	-	n.s.
D1×D3	**-0.143**	0.079	0.086	-	-	n.s.	-	-	n.s.
$D1^2$	**-0.269**	0.081	0.003	-	-	n.s.	-	-	n.s.
$D2^2$	**-0.159**	0.038	<0.001	-	-	n.s.	**-0.176**	0.047	0.001
$D3^2$	-	-	-	-	-	n.s.	-	-	n.s.

[a] Standard Error, [b] Not significant at P=0.1

5.3.2 Sensory and instrumental drivers of liking

The overall liking was modelled against all chemical and physical instrumental attributes by PLSR (R^2=0.90) (model in **Figure 29**; β coefficients in **Figure 30**, only the pooled data from all three countries presented). It can be seen that three of the largest drivers for liking are the CIELAB colour parameters. Colour does not only drive liking directly (L* value was also found to be a driver of liking for both peanut butter and roasted peanuts with American consumers (Lee & Resurreccion, 2006b; Pattee et al., 2001)), but has also been shown to be highly correlated to (presumably) pleasant 'roasted', 'peanut', 'sweet' and low 'raw peanut' aromatics (Chetschik, Granvogl, & Schieberle, 2010; Lee & Resurreccion, 2006b; McDaniel, White, Dean, Sanders, & Davis, 2012; Pattee, Giesbrecht, & Young, 1991; Schirack et al., 2006a). This likely because the Maillard Reaction is the main pathway for both colour (Pattee et al., 1991) and flavour (Schirack, Drake, Sanders, & Sandeep, 2006b) development. As discussed above this relationship is likely to exhibit an optimum L* value above which liking starts to drop (under-roasting/raw), but given that PLSR only includes linear parameters and that most of the samples were not be excessively pale/light roasted, the effect appears as a positive, *linear* relationship in this model. The Partial Least Squares methodology has been recommended for analysing the relationship between hedonic scores of a relatively few samples, against a large number of instrumental attributes (such as headspace volatiles) (Tenenhaus, Pagès, Ambroisine, & Guinot, 2005).

Sucrose content and large kernels are also important contributors to liking. These observations are consistent with the sensory attribute correlations discussed

in **Figure 28** and **Table 18**, as well as published research for US consumers (Wang et al., 2017; Young et al., 2005). The impact of instrumental texture on liking appears underestimated compared to the role sensory texture plays on preference (**Table 18**), something also previously observed (Lee & Resurreccion, 2006b). This can be explained by the lack of a single instrumental attribute that highly correlates with sensory texture.

The forty-four headspace volatiles that were detected at the sniff port or had been previously correlated with specific sensory attributes in peanuts (Lykomitros, Fogliano, & Capuano, 2016b) are shown in **Table 19**. Interestingly, five of the compounds (propanoic acid, 2-oxo, methyl ester, 1-(acetyloxy)-2-propanone, pantolactone, 1-nitro-hexane and 4H-pyran-4-one, 2,3-dihydro-3,5-dihydroxy-6-methyl (hydroxydihydromaltol)) previously found to be present in relatively high concentrations or correlated with at least one sensory attribute were not detected in this study. This is likely due to the methodological difference between the two assays (Dynamic Headspace vs Solid Phase Micro Extraction), but it is also possible that these compounds were simply not present in the 26 samples analysed. 1-nitro-hexane and hydroxydihydromaltol especially, have been previously shown to be absent from several cultivar-process combinations (Lykomitros et al., 2016b).

5.3.3 Headspace volatile drivers of liking, 'fresh' and 'stale'

Since headspace volatile composition was identified as the largest instrumental driver after the colour parameters, separate models were developed focusing exclusively on the volatile profile. PLSR models for overall liking, 'fresh' and 'stale' were developed for each country and shown in **Figure 31**. The models for all three countries are very similar, and so the consumer data was also pooled and the three agglomerated models were developed (marked 'all' in the figure), obtaining a good fit (R^2= 0.86, 0.848 and 0.855). The standardized coefficients of the agglomerated models can be found in **Figure 32**. It is clear that liking and 'fresh' are very highly correlated, as their model coefficients are almost identical, as previously seen with US consumers (Young et al., 2005). Although inversely related, 'stale' and 'fresh' are not exact opposites of each other, especially for Dutch consumers.

Liking (and 'fresh') is correlated with 2,5/3-dimethyl-1H-pyrrole, 2,3 pentanediol, 2/3 methyl pyrrole, and (E)-2-methyl-6-(1-propenyl)-pyrazine, while being inversely related to several aldehydes and ketones. Aldehydes are often associated to lipid oxidation, and have been shown to decrease consumer liking in peanuts (Nepote et al., 2009). Hexanal formation has been attributed to the oxidation of linoleic acid (St Angelo, 1996) and its content has been shown to be one of the main driver of dissatisfaction for US consumers, with an acceptability

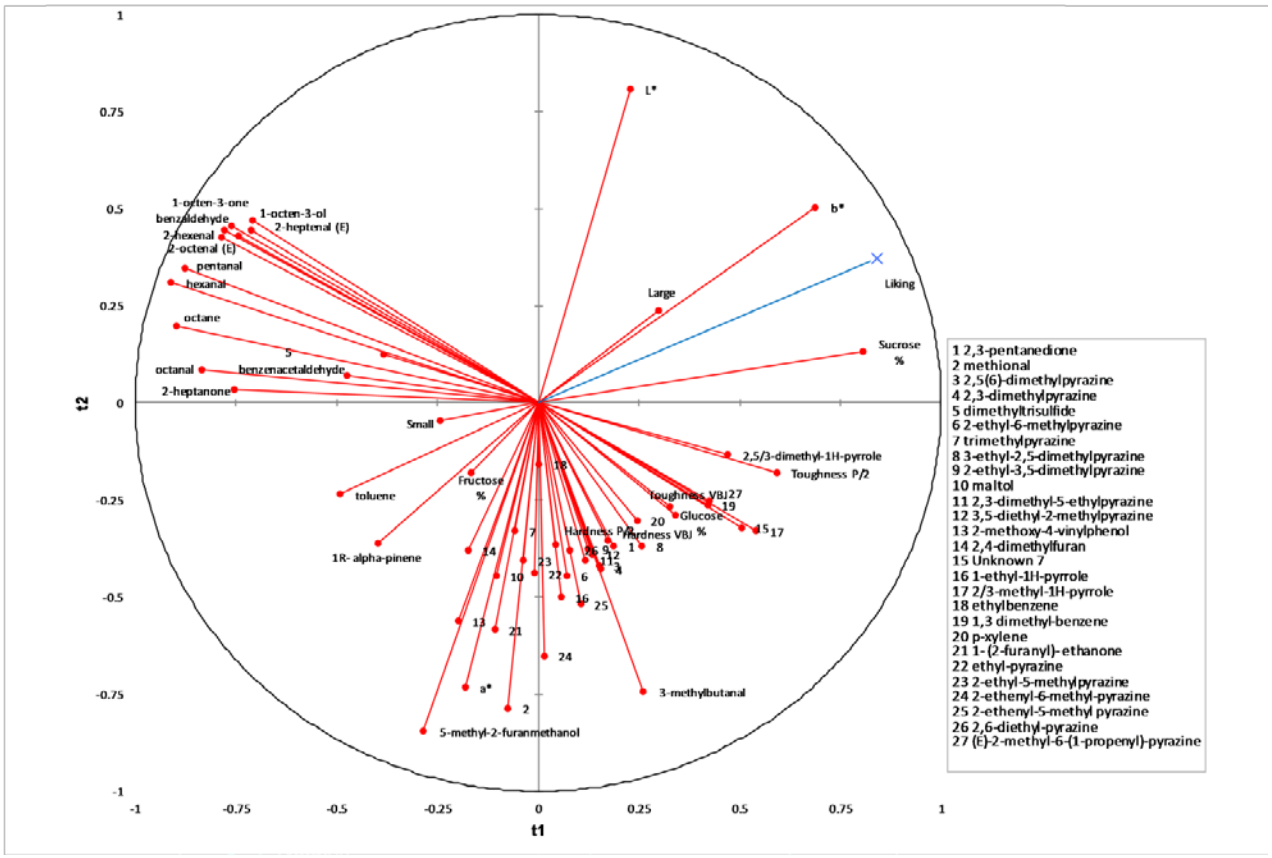

Figure 29: Two dimensional graphical representation of the correlations between liking and all instrumental attributes as determined by the PLS regression (combined data from all countries). Circles: explanatory variables, x: response variable (liking).

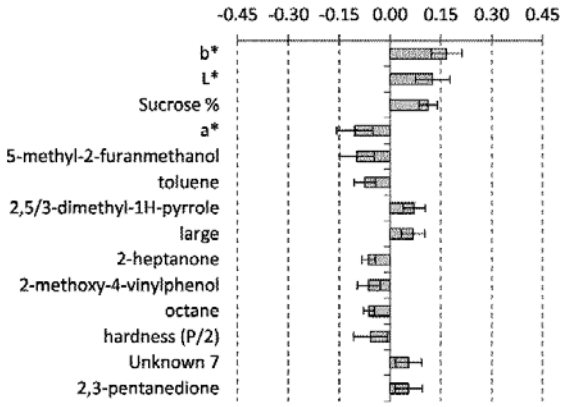

Figure 30: Standardized β coefficients (with standard errors) of instrumental attributes of the PLS model for liking (all countries pooled). Only coefficients larger than 0.05 are shown.

threshold of 7.40 μg/g (Grosso & Resurreccion, 2002). Pyrazines appear to play a very limited role in consumer liking. This is consistent with recombination studies that showed limited aroma activity in peanuts (Chetschik et al., 2010), even though some references suggest that pyrazines (2,5-dimethyl pyrazine in particular) were drivers of liking in roasted peanuts (Baker et al., 2003; Wang et al., 2017). 'Stale' is correlated with several aldehydes, ketones and other volatiles, particularly 2-heptanone, hexanal, 5-methyl-2-furanmethanol and octane. No study of 'stale' as an attribute was identified in the literature, but the identified compounds have been linked to lipid oxidation (St Angelo, 1996), particularly during storage of peanuts (Grosso & Resurreccion, 2002; Lee & Resurreccion, 2006a).

Given that 'fresh' and liking are collinear in this sample set, and that all samples had similar age and were essentially defect free, it is not possible to infer if 'fresh' is a driver of liking, or a description assigned by consumers to the samples they prefer. With peanuts, there does not appear to a specific product design that is consistently perceived as fresh, unlike the orange juice and salad examples previously discussed. In contrast, 'stale' appears to be a well understood descriptor by the European consumer, and it is mainly related to lipid oxidation defects. The use of the attribute 'stale' should be therefore favoured over 'fresh' in consumer research on product quality, as it relates better to physical characteristics and offer more incremental information to hedonic ratings.

Finally, it can be seen that relative order of the headspace volatile drivers of liking shown in **Figure 32** is somewhat different than those shown in **Figure 29**. This is likely because the later model includes colour parameters, which share some

Table 19. The selected headspace volatiles used in the PLS models. More detail (Linear Retention Indices (LRI), major ions, quantification ions and quantification factors) has been previously published (Lykomitros et al., 2016b).

Compound	Odour character reported by operators	Mean odour intensity [a]	Maximum concentration observed relative to internal standard (μm/ml)
hexanal	green, grassy	3.3	7250
benzaldehyde	bread, biscuit, coconut	2.0	3807
benzenacetaldehyde	sweet, caramel, honey, candied	3.6	1671
2,5(6)-dimethylpyrazine	green, sesame, nutty, nasturtium	3.3	1642
2-heptenal (E)			1408
2-ethyl-5-methylpyrazine			1257
trimethylpyrazine	musty, earthy	2.9	1193
3-ethyl-2,5-dimethylpyrazine	musty, green, earthy	2.9	1183
2-octenal (E)			759
1-octen-3-ol			710
3-methylbutanal	malt, bread	3.0	530
pentanal	green, grassy	2.7	479
2-ethyl-6-methylpyrazine	roasted, nutty	2.9	394
2-hexenal			354
2-methoxy-4-vinylphenol	clove, spice	2.5	336
octanal	fresh, soapy, green, fruity	3.4	321
1R- alpha-pinene			247
octane			244
ethyl-pyrazine			191
toluene			185
(E)-2-methyl-6-(1-propenyl)-			183
2-heptanone			165
2-ethyl-3,5-dimethylpyrazine	musty, earthy, potato	2.4	158
3,5-diethyl-2-methylpyrazine	musty, earthy	2.5	133
2,6-diethyl-pyrazine			128
2,3-pentanedione	butter, cheese, clotted cream	2.1	125
2-ethenyl-6-methyl-pyrazine			108
1,3 dimethyl-benzene			96
2,3-dimethylpyrazine	nutty, biscuit, popcorn, pie crust	3.5	81
dimethyltrisulfide	garlic, roast onion, pickle	2.9	66
2,3-dimethyl-5-ethylpyrazine	raw potato, raw peanut, hazelnut	3.0	57
p-xylene			50
2-ethenyl-5-methyl pyrazine			38
ethylbenzene			29
1-octen-3-one			29
1- (2-furanyl)- ethanone			21
2/3-methyl-1H-pyrrole [b]			17
2,5/3-dimethyl-1H-pyrrole [b]			13
maltol			10
Unknown 7 [c]			9
methional	potato, instant mash	2.6	8
1-ethyl-1H-pyrrole			7

Compound	Odour character reported by operators	Mean odour intensity [a]	Maximum concentration observed relative to internal standard (μm/ml)
5-methyl-2-furanmethanol			6
2,4-dimethylfuran			1

[a] 4 point scale (1: barely detected, 2: weak but identifiable, 3: medium strength, 4: high strength) [b] Experimental setup could not distinguish between the listed compounds [c] Compound could not be identified: Major ions 68, 42, 11, 57, 87, Linear Retention Index 803.

Maillard reaction pathway with certain flavour compounds (Pattee et al., 1991). As a result, the PLSR procedure prioritizes colour attributes as they can explain more variance than individual compound concentrations.

5.4 Conclusions

In this work, it was shown the attributes that drive preference for roasted peanuts with Dutch, Spanish and Turkish consumers, were not only very similar to each other, but also to US consumers. Sweet taste (related to the intrinsic sucrose content), sweet aroma and roasted peanut aroma were shown to drive liking, while raw and painty aromas and bitter taste drive dislike. The preference map models suggest that an optimal intensity for flavour, colour and textural attributes. Even though it makes intuitive sense (an excessively dark or light roast is likely undesirable), this has not been previously reported. Texture and colour were also shown to play an important, but secondary role on preference. Appearance characteristics had limited impact, with kernel size being of moderate importance and breakage (split cotyledons) not having a significant impact.

Liking and ‘fresh’ where concepts virtually indistinguishable in the mind of the consumer. It is possible that the strong correlation between ‘fresh’ and liking is casual rather than causal, and that ‘fresh’ is probably dependent on other factors related to the cultural heritage of each country and not to the actual quality of the product. Both were correlated to the presence of pyrroles such as 2,5/3-dimethyl-1H-pyrrole and 2/3 methyl pyrrole, and inversely correlated to small molecular weight aldehydes and ketones. Finally, pyrazines had no significant correlation with liking. ‘Stale’ was clearly correlated to lipid oxidation products such as 2-heptanone and hexanal, and as expected was inversely related (but not the exact opposite) to liking and ‘fresh’. As a result, ‘stale’ appears to be better associated with the physical condition of peanut samples by European consumers compared to ‘fresh’. The results suggest that there are no category wide freshness cues that could be incorporated into a product design to increase the perception of

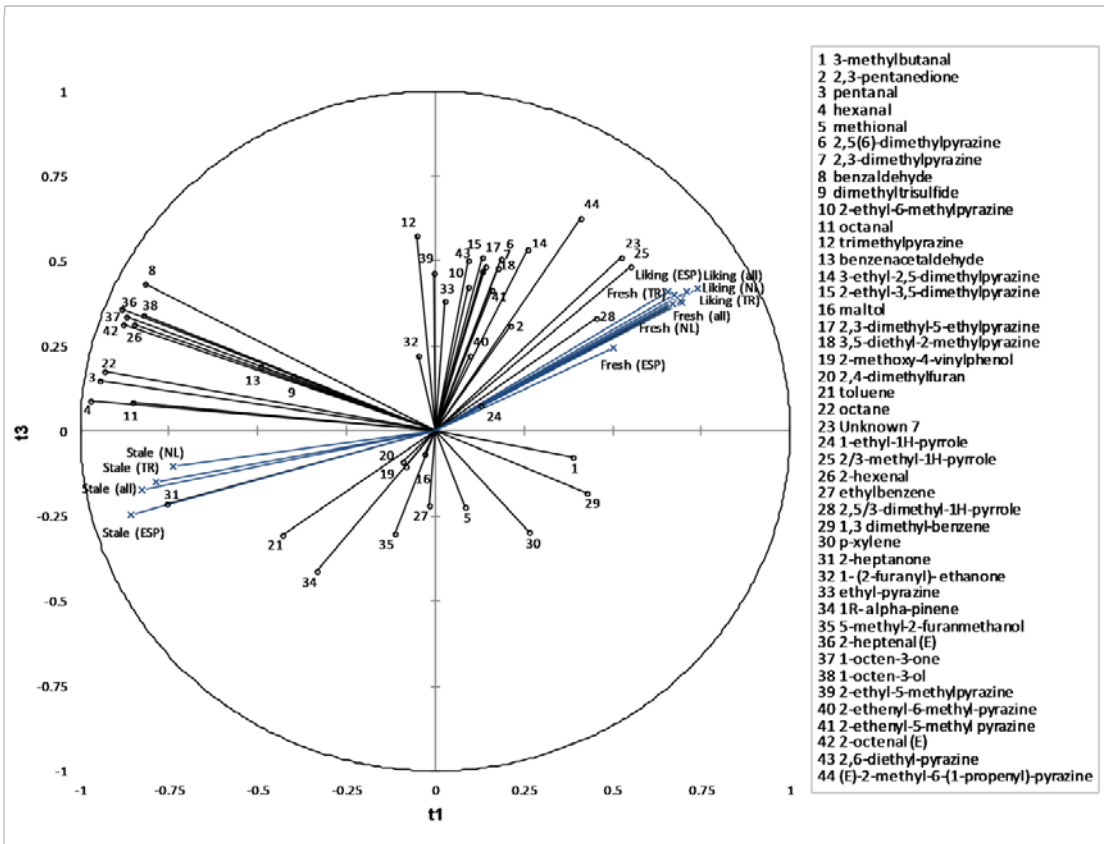

Figure 31: Two dimensional graphical representation of the correlations between liking, 'fresh' and 'stale' and volatile components, as determined by the PLS regression. (NL): Dutch consumers, (ESP): Spanish consumers, (TR): Turkish consumers: (all): pooled consumers. X: response variables, o: explanatory variables.

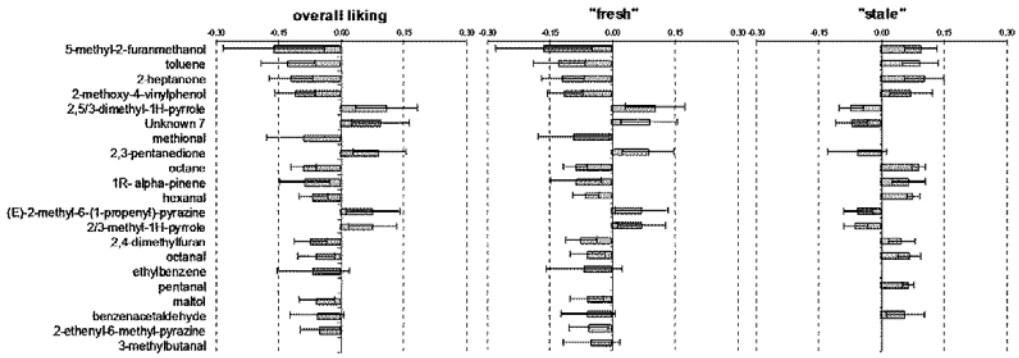

Figure 32: Standardized β coefficients (with standard errors) of volatile compounds quantified by SPME-GC-MS in the PLS models for liking, 'fresh' and 'stale'. Only coefficients larger than 0.05 are shown.

freshness, and that the 'fresh' seems to be mainly related to the degree of lipid oxidation in a sample. In this study, no attempt was made to resolve the possible meta-characteristics that could drive perceived freshness, such as packaging, on-pack information or marketing concept.

Finally, it was shown that fairly simply instrumental analysis, mainly CIELAB colour values and sucrose content, can provide significant insight into consumer preference for peanuts. It was argued that colour is such an important attribute because it is itself highly correlated to flavour and texture attributes, as colour, flavour and texture are known to develop through the same mechanism (Maillard reaction). Commonly used large deformation tests where not seen to be significantly incremental when the samples have a standardized finished moisture content and the colour parameters are already included in the preference model.

5.5 Acknowledgements

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6 General discussion

6.1 Overview of the experimental design

The general research outline is summarized in **Figure 33**. Twelve different raw materials were processed in eleven different ways (including pre and post processing steps, such as maceration and topical oil application) to yield 134 unique samples. The samples were consequently analysed instrumentally for their physical and chemical characteristics, and by an expertly trained sensory panel for their sensory profiles (aroma, flavour, taste, texture and aftertaste). Of the 134 samples, the 26 most differentiated samples were further tasted by consumers in three countries (The Netherlands, Spain and Turkey) and hedonic ratings where obtained.

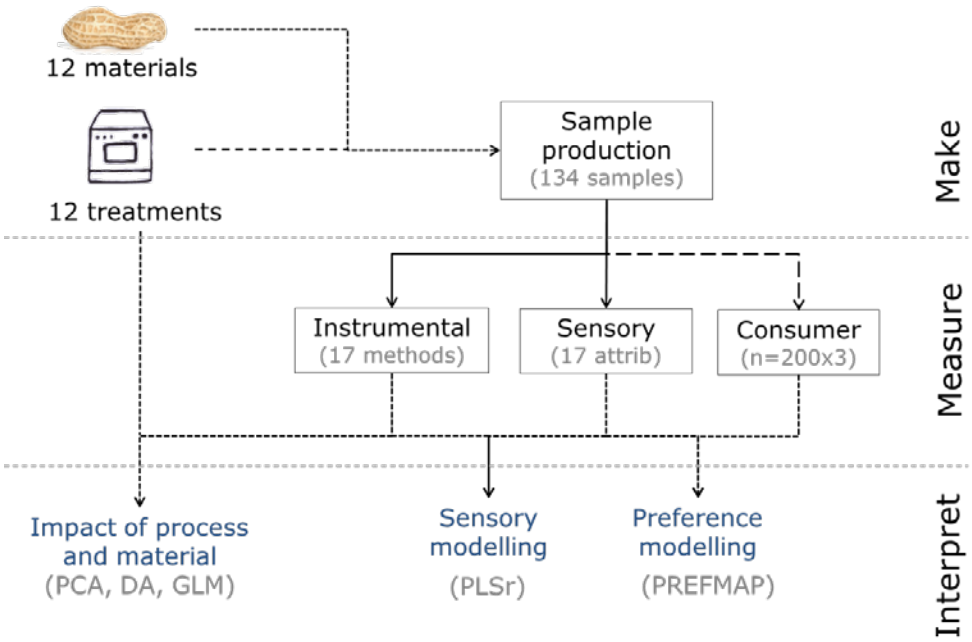


Figure 33: General research outline, and thesis chapter overview. PCA: Principal Component Analysis, DA: Discriminant Analysis (specifically Canonical Variate Analysis), GLM: General Linear Model, PLSr: Partial Least Squares Regression, PREFMAP: Preference map of liking means.

The data analysis can be divided in three areas: i) quantification of the raw material and process impact (analysis of process, material and sample instrumental data with multivariate methods such as Principal Component Analysis, Canonical Variate Analysis, and General Linear Model regression), ii) sensory modelling

(Partial Least Squares regression modelling of sensory attributes against instrumental data), and iii) preference modelling (using the PREFMAP procedure to relate sensory and instrumental attributes to mean hedonic scores (Macfie, 2007)).

6.1.1 Sample selection and preparation

As discussed in Chapter 1, genetics, kernel maturity, growing conditions, post-harvest treatment (drying at the farm and storage), blanching and processing (including pre-treatments such as maceration, and post treatments such as the application of aromatic oils and salt) all have an impact on the flavour and texture of peanuts, and therefore potentially consumer liking. It is obvious that the factors are too great in number for a study to be able to tackle them simultaneously, and so compromises and assumptions must be made for practical reasons, which are discussed in this section. The guiding principle was to evaluate as many different types of materials and process as possible, while ensuring that common materials and processes are investigated to a somewhat deeper level. The design resulted in 134 unique peanut samples (this was the sample set used in Chapters 2,3 and 4), while the restrictions described below ensured it was practical to execute. As a result, the study has one of the largest sample sets encountered in the literature on peanut sensory research.

With regards to the raw material selection, some of the most common peanut growing regions were selected: USA (Texas, Georgia and Virginia states), Argentina, South Africa, China and Australia. The market type was restricted to Runner, Valencia and Virginia, and some of the most common cultivars and grades (kernel sizes) from each type were selected. The design is more biased towards the Runner type as it is the one most commonly used for snack peanuts. Not every producing country grows the same cultivars, and so a full factorial design where every cultivar is grown in every location was not possible without bespoke cultivation, something beyond the resources available for this study. To reduce the number of raw materials further, materials from only one crop year were included in the study. Similarly, no attempt was made to control for the harvesting method, on-field drying or farmer stock storage method. The blanching method was controlled, but limited to only one method, applied to all samples (approx. 80°C heating in hot air for 30min, followed by mechanical abrasion, performed in the same facility). The result was the selection of twelve different raw materials, and the full details are shown in **Table 20.**

Table 20: The raw materials used in the research, and some of their major characteristics.

type	variety	origin	grade	count per 100 grams	high oleic	incoming moisture content	incoming split kernels
Runner	Flavorunner 458	USA – Texas	medium	141/177	yes	7%	1%
Runner	Flavorunner 458	USA-Texas	jumbo	134/148	yes	7%	3%
Runner	Georgia Green	USA – Georgia	medium	173	no	6%	2%
Runner	Georgia Green	USA – Georgia	jumbo	137	no	6%	2%
Runner	Granoleic	Argentina	jumbo	134/148	yes	7%	3%
Runner	Tegua	Argentina	medium	141/177	no	5%	4%
Runner	Hsuji	China	medium	141/177	no	5%	5%
Valencia	CN Natals	South Africa	small	177	no	5%	2%
Virginia	mixed	USA – Virginia	extra large	106	no	7%	3%
Virginia	mixed	USA – Virginia	medium	148	no	7%	1%
Virginia	Middleton	Australia	extra large	71/92	yes	6%	6%
Virginia	Middleton	Australia	medium	120/141	yes	6%	4%

Similarly, with processing emphasis was placed on frying and baking (dry and oil roasting) at several temperature and time combinations. Maceration in different media (glucose content and pH levels) and times was added to the design as a pre-treatment. Maceration was selected because it is a simple industrial process commonly applied to plant materials, but not commonly encountered in snack peanuts. A full description of the process and process by material combinations investigated can be found in Chapter 2, but an overview is given in **Table 21**.

The final challenge was to determine the sample subset for the consumer sets (used in Chapter 5). The objective was to reduce the sample size as much as possible in order to facilitate a consumer test design where all respondents evaluate all the samples, while ensuring the samples were as organoleptically differentiated as possible in order to maximize the power of the preference map algorithm (Macfie, 2007). To this effect, a combination of statistical and empirical approaches were followed: Firstly, agglomerative hierarchical cluster analysis was used to classify the sensory data presented in Chapters 2-4 by similarity, and the centroids were selected as representatives of all the flavour and texture sample groups. The process was summarized in Chapter 5, and to better illustrate the sensory variation the dendrograms are shown in **Figure 34**. To ensure the design objective of wide scope but high resolution for common processes and materials, an additional 12 samples were empirically selected by an expert technical panel, so that the following sets were also represented in the design: i) at least one variety processed by all dry roasting conditions), ii) at least one sample from each grade (kernel size) and iii) at least one instance of the same sample at high and low breakage (split cotyledons).

Table 21: Overview of processes employed in this research.

Type of process	process	key process parameters [a]
Pre-treatment: Maceration	aqueous acid maceration, dry roasting	acidified to pH 4 with acetic acid, 30 min at 20 °C, roasted at 145 °C
Pre-treatment: Maceration	long aqueous maceration, dry roasting	potable water, 90 min at 20 °C, roasted at 145 °C
Pre-treatment: Maceration	aqueous dextrose maceration, dry roasting	2.5% w/w dextrose solution, 30 min at 20 °C, roasted at 135 °C
Pre-treatment: Maceration	aqueous alkaline maceration, dry roasting	alkalized to pH 10 with $CaOH_2$, 30 min at 20 °C, roasted at 145 °C
Pre-treatment: Maceration	short aqueous maceration, dry roasting	potable water, 30 min at 20 °C, roasted at 145 °C
Pre-treatment: Maceration	aqueous dextrose maceration, oil roasting	2.5% w/w dextrose solution, 30 min at 20 °C, fried in high oleic sunflower seed oil at 150 °C
Roasting	dry roasting (low temperature long time)	continuous convection oven 135 °C
Roasting	dry roasting (high temperature short time)	continuous convection oven 155 °C
Roasting	two temperature zone dry roasting (high-low)	continuous convection oven 155 °C /135 °C
Roasting	two temperature zone dry roasting (low-high)	continuous convection oven 135 °C /155 °C
Roasting	oil roasting (frying)	fried in high oleic sunflower seed oil at 150 °C
Post-treatment: oil spray	topical aromatic roasted peanut oil application	2% w/w aromatic roasted peanut oil spray
Post-treatment: oil spray	topical sunflower oil application	2% w/w high oleic sunflower seed spray

[a]: Roasting time varied per sample so that the final moisture content was approximately 2% w/w.

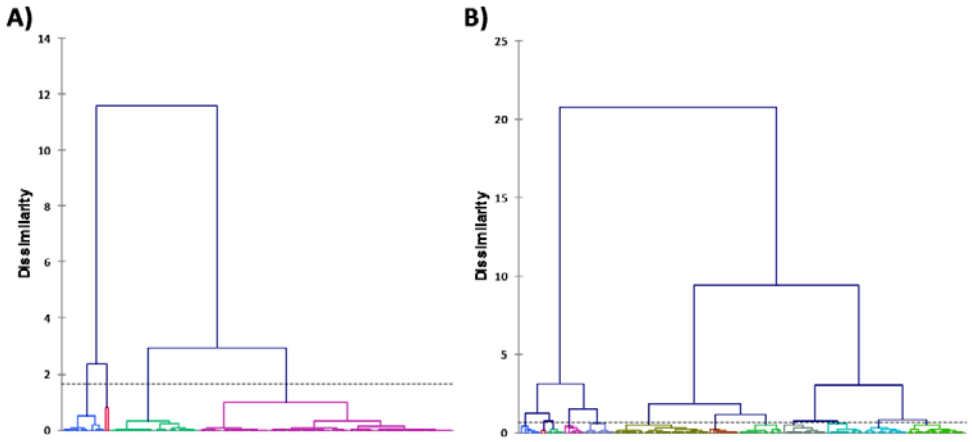


Figure 34: Agglomerative hierarchical cluster analysis of texture (Panel A) and flavour (Panel B) sensory profiles (covariate, Ward Linkage, Euclidean Distance, XLStat). The different colours denote the different clusters.

This resulted in a total of 26 unique samples to be evaluated by consumers, well dispersed in the texture and flavour space as visually demonstrated in **Figure**

35. Larger quantities (30Kg) of these 26 samples were separately produced, and the instrumental and sensory analyses were repeated on these fresh samples.

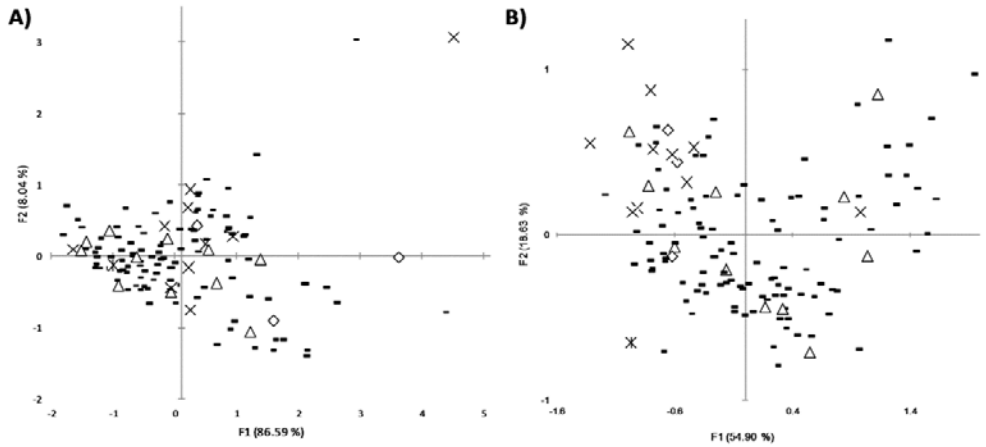


Figure 35: The 26 selected samples for consumer testing shown in the texture (Panel A, left) and flavour (Panel B, right) sensory space. PCA space for texture defined in Chapter 4, PCA space for flavour defined in Chapter 1. ◊: texture cluster centre elements, ∆: flavour cluster centre elements, *: centre elements for both texture and flavour clusters, X: samples empirically selected by technical panel, -: samples excluded from the consumer testing subset.

6.1.2 Sample and data analysis

The aim of this work was to identify correlations between sensory and instrumental attributes and consumer liking. To do so, a large number of instrumental analysis was performed on all samples, measuring a variety of attributes including the chemical composition (Gas Chromatography- Mass Spectroscopy headspace volatile analysis, sugar profiling by ion chromatography and fatty acid profiling by Fatty Acid Methyl Ester gas chromatography), physical structure (textural properties by large deformation stress-strain tests, moisture content by thermal balance, and microstructure by confocal microscopy and X-ray Computer Tomography), and macro characteristics (kernel size distribution by screen analysis, CIELAB colour parameters by colorimeter).

The sensory profiles were obtained through a highly trained sensory panel (USDA ARS, North Carolina) using the Spectrum ^{TM} and Descriptive Sensory Analysis method (additional panel details in Chapter 4). This methods provide ratings of flavour, taste, aftertaste and mouthfeel attributes in a standardized 15-point scale

(Meilgaard, Civille, & Carr, 1999). A summary of the sensory attributes measured is shown in **Table 22**. To ensure homogeneity, peanut paste (prepared by grinding) was used to evaluate the flavour and aroma attributes. Whole peanuts were used for the texture profiles.

Finally, consumer liking data were obtained from consumers in three different countries (Netherlands, Spain and Turkey), using a minimum of 200 consumers per country (details can be found in Chapter 5).

The multidimensional data was analysed with Principal Component Analysis when a description of the multivariate landscape was required (e.g. to describe the sensory space) and Canonical Variate Analysis when classification was required (e.g. do demonstrate whether sensory differences can be explained by raw material or process selection) (Burgard & Kuznizki, 1990). Wherever a main effect or hypothesis was to be tested, a General Linear Model regression was the method of choice. Partial Least Squares Regression was used to build the predictive models of the sensory from the instrumental attributes. Finally, the external preference mapping methodology on liking means (up to quadratic models) was used to identify the drivers of consumer liking (Liggett, 2010). Details on all methods and procedures are provided in the appropriate chapter

6.2 Thesis main findings by research objective

A schematic that summarizes how the literature gaps connect with the research objectives, the main findings, and the potential application of these findings can be found in **Figure 36**. The findings and research applications sections discuss the topics listed in the figure in more detail.

The primary objective of this research was to determine what drives preference in snack peanuts for European consumers. Interestingly the drivers are very similar for Dutch, Spanish and Turkish consumers, and consistent with what has been previously reported for Americans (Young, Sanders, Drake, Osborne, & Civille, 2005). Some consumer segmentation was detected within each country, but the segment means where not dramatically different from each other. Light colour (better interpreted as ‘not too dark’), sweetness and the concentration of several pyrroles were the main drivers of liking, while hexanal and 2-heptanone were drivers of disliking and perceived staleness. CIELAB colour parameter b*(blue-yellow) mainly (but also L* (lightness) and a* (green-red) to a lesser degree) was by far the most significant driver of liking. This is not surprising as the colour parameters are highly correlated to roasted aromas themselves (as discussed in Chapter 2), even though the literature has mainly focused on the L* value.

An investigation of the perceived 'fresh' and 'stale' attributes showed that they are not design driven as in certain products (e.g. orange juice (Zhang, Lusk, Mirosa, & Oey, 2016)), but mainly related to the oxidation state of the peanut lipids. The data suggests that liking and freshness are practically synonymous for the consumer, although this research design cannot resolve whether the effect is causal. Perceived staleness however, appears to be a more distinct attribute to the consumer, and it is less strongly correlated to the inverse of liking, suggesting that the attribute carries incremental information. The data therefore demonstrated that inquiring about perceived staleness ion consumer tests may be advantageous over perceived freshness.

The second objective of this thesis was to provide a better understanding of the impact of raw material and process technology and their interaction on the organoleptic profile and consequently consumer preference of snack peanuts. The aspiration has been to discover processing conditions that can compensate for the differences among raw materials so that a wider range of raw materials can be used, thereby reducing food waste and procurement costs. The main finding was that process can often have a larger effect than raw material selection on both flavour and texture. More specifically, it was determined that maceration in aqueous media has a significant impact on flavour (increase 'roasted' and decrease 'raw beany' aromas, Chapter 2 & 3), texture (increase crunchiness and/or hardness (if dextrose is included), Chapter 4) and colour (degree of browning, depending on pH of the medium, Chapter 2), while the effect is significantly larger than the differences observed among raw materials. Interactions between maceration treatments and roasting methods were also observed, suggesting that the treatment can be leveraged to produce dry roasted peanuts with the organoleptic properties of fried peanuts. Maceration however also affects the fatty acid composition due to hydrolysis, and so it is best used with high oleic cultivars. With regards to raw materials, it was demonstrated that the 'blister fry' type process which imparts the characteristic blistered appearance and crunchy/hard texture also works on Runner and Spanish types (both *Arachis Hypogaea* and *Fastigiata* sub species), something not previously seen (Chapter 4).

The third objective was to gain understanding of the compositional changes induced by processing and contrast their magnitude to those caused by raw material selection. With regards to flavour and aroma, multivariate predictive models correlating the chemical fingerprint to four sensory attributes was successfully developed in Chapter 3 ('roasted peanut aroma', 'dark roast aroma', 'raw bean aroma' and 'sweet aroma'). It was demonstrated that a logarithmic transformation of the headspace volatile concentration data significantly improved

Table 22: Sensory attributes used by the expert panel. (Johnsen, Civille, Vercellotti, Sanders, & Dus, 1988; Lee & Resurreccion, 2006b; Sanders, Vercellotti, Crippen, & Civille, 1989; Schirack, Drake, Sanders, & Sandeep, 2006)

	attribute	Description
Aroma/Flavour	roasted peanut	the aroma associated with medium roast peanuts (3-4 on USDA colour chips), and having fragrant character such as methyl pyrazine
Aroma/Flavour	sweet aroma	the aromas associated with sweet material such as caramel, vanilla, molasses, fruit (specify type)
Aroma/Flavour	dark roast	the aroma associated with dark roasted peanuts (4+ on USDA colour chips) and having very browned or toasted character
Aroma/Flavour	raw beany	the aroma associated with light roast peanuts (1-2 on USDA colour chips) and having legume like character (specify beans or pea if possible)
Aroma/Flavour	woody, hulls, skins	the aromas associated with base peanut character (absence of fragrant top notes) and related to dry wood, peanut hulls and skins.
Aroma/Flavour	cardboard	the aroma associated with somewhat oxidized fats and oils and reminiscent of cardboard
Aroma/Flavour	earthy	the aroma associated with wet dirt and mulch.
Aroma/Flavour	painty	the aroma associated with linseed oil, oil based paint.
Aroma/Flavour	phenolic/chemical	aroma associated with chemical/plastic/band aid
Aroma/Flavour	fruit fermented	the aroma associated with over ripe or sweet fermenting fruit
Aroma/Flavour	ashy	the aroma associated with ash-tray without tobacco notes
Aroma/Flavour	total off note	intensity rating of total off notes
Taste	sweet	the taste on the tongue associated with sugars
Taste	sour	the taste on the tongue associated with acids.
Taste	bitter	the taste on the tongue associated with bitter agents such as caffeine or quinine.
Taste	salty	the taste on the tongue associated with sodium ions.
Texture	Crispy	degree (volume) to which the sample makes a high-pitched sound (incisors)
Texture	Crunchy	degree (volume) to which a sample makes a low pitched sound (molars)
Texture	hardness	amount of force required initially to bite/fracture the sample using the molars
Texture	breakdown	degree to which the sample breaks apart using the molars on the first bite
Mouthfeel	tongue, throat burn	the chemical feeling factor on the tongue and throat associated with burning (benzoate).
Mouthfeel	metallic	the chemical feeling factor on the tongue described as flat, metallic and associated with iron and copper.
Mouthfeel	astringent	the chemical feeling factor on the tongue, described as puckering/dry and associated with tannins or alum.

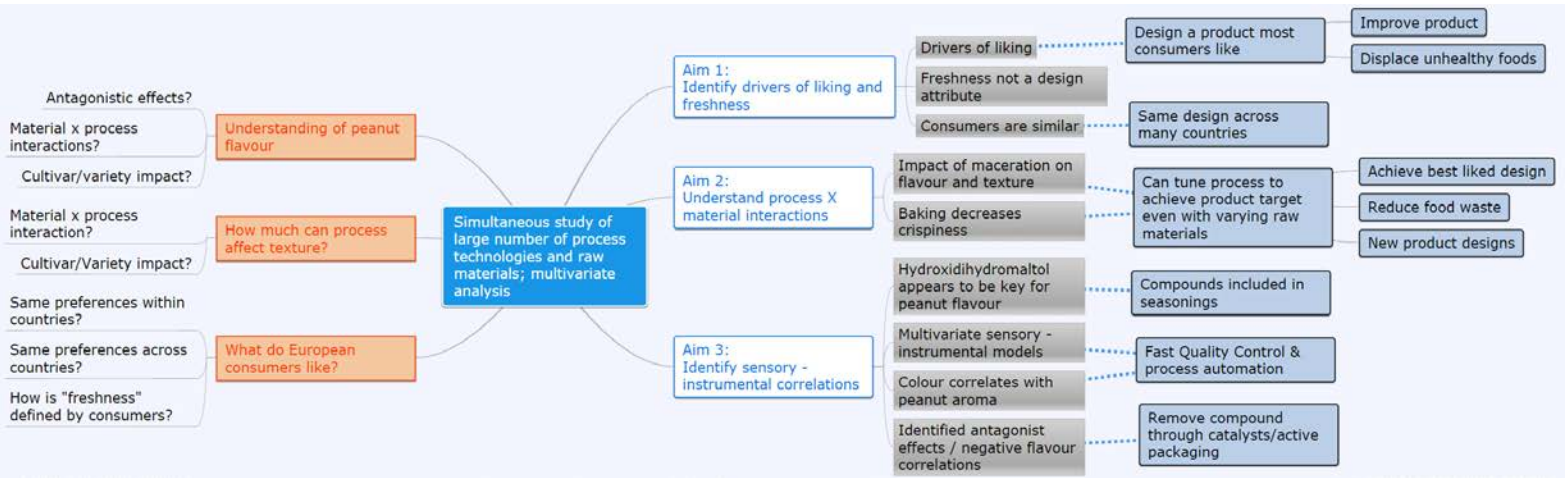


Figure 36: A schematic demonstrating the overall information workflow of this thesis. The orange boxes list the main knowledge gaps, which give rise to the three research objectives (blue frames). The grey boxes list the main research findings, and the shade blue boxes on the right list the potential applications of those findings.

the model fits. Somewhat surprisingly, but in agreement with recent recombination studies on Runner type peanuts (Chetschik, Granvogl, & Schieberle, 2010; Da Conceicao Neta, 2010) pyrazines were seen to be less correlated to roasted peanut aromas. Compounds highly correlated to sensory attributes (but not always with a positive coefficient) included hydroxydihydromaltol (not previously reported in peanuts), 2/3-methyl-1H-pyrrole, benzeneacetaldehyde, 2-hexenal and 3-hexen-2-one.

The headspace volatiles identified to be correlated to sensory attributes in this study are in general agreement with previous work, with the exception of 3,5-dihydroxy-6-methyl-2,3-dihydropyran-4-one (hydroxydihydromaltol). Hydroxydihydromaltol (CAS# 28564-83-2) has not been previously reported on peanuts, but has been identified in other 'toasted' aromas, such as in oak wine barrels (Cutzach, Chatonnet, Henry, & Dubourdieu, 1997). In this study it was found in only some samples (~85% of samples), but when present, it was highly correlated to several roasted aromas. To better understand if the presence of the compound is related to a specific material or process, the concentration was plotted per raw material (across all treatments) and treatment (across all raw materials) in **Figure 37**. The figure suggests that although the Granoleic variety tends to produce more hydroxydihydromaltol, maceration in any medium and particularly in aqueous glucose can significantly increase the concentration of hydroxydihydromaltol in the finished product.

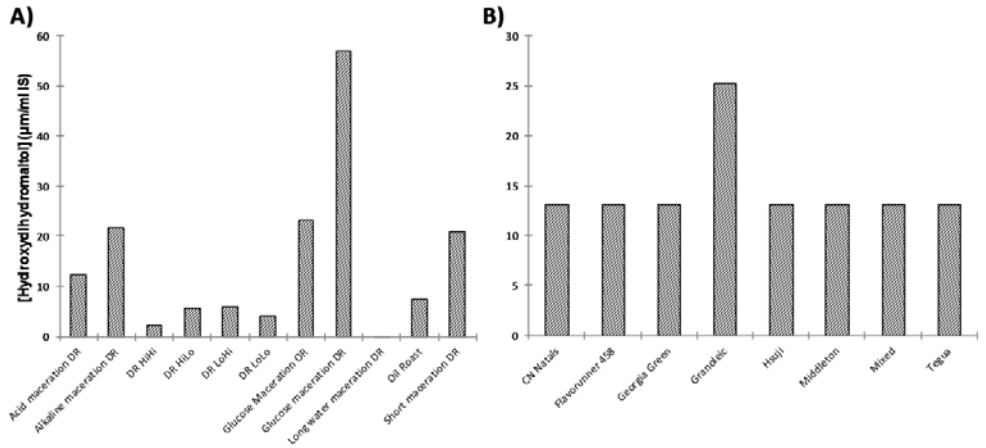


Figure 37: Main effect means of hydroxydihydromaltol concentration (µm/ml Internal Standard). A: means of the different processes, B: means of the different varieties.

It was also observed that flavour and colour attributes were highly correlated, something that can be used to simplify quality control and product optimization in an industrial production environment. However, the correlation

was not as strong when maceration processes were applied, and the b* value was more strongly correlated than the L* value, on which most literature is focused.

With regards to texture, most of the observed changes could be attributed to microstructure modification. The current dogma is that maceration increases the water content of the interior of the peanut, which in turn causes more steam to be generated during roasting, leading to more extensive microstructure disruption (Shi et al., 2017; Young, Pattee, Schadel, & Sanders, 2004). In this research, the extent of microstructure disruption was for the first time quantitatively described (as the number and size of air cells measured by X-ray computer tomography). The analysis demonstrated that although maceration in different media results in a similar air cell distribution, there are significant sensory differences between the samples. Given that the effect was most pronounced in the presence of glucose (a reducing sugar) in combination with oil roasting (highest temperature process), the proposed hypothesis was that Maillard or other molecular interactions increase the local mechanical moduli. Consequently a secondary pathway was proposed in Chapter 4, wherein in addition to the steam microstructure disruption, molecular (most likely Maillard) interactions lead to the increase of local mechanical moduli. Potential local interactions near the air cell sites had been previously cited (Miyagi & Ogaki, 2014) while the impact of melanoidins and other Maillard by products on texture is well known (Açar, Gökmen, Pellegrini, & Fogliano, 2009), but this was the first instance this secondary pathway of texture development in peanuts was proposed.

Interestingly, it was further observed that Virginia type peanuts develop with roasting both flavour and texture faster and to a greater degree than Runner or Valencia market types.

6.2.1 **Methodological contributions**

With regards to methodology, three techniques used in this research have not been observed in the literature: the logarithmic transformation of the headspace volatiles (in peanut flavour research), the discussion of volatiles with negative coefficients in sensory attributes models and the use of PLSR on CATA count data for evaluating consumer response.

Transformations of all or part of the X data matrix is a very common practice in regression, as it allows linear algorithms to model non-linear responses (Neter, Kutner, Nachtsheim, & Wasserman, 1996). However, even though linear transformations (as in the case of autoscaling in factor analysis) are fairly common, no reference was found in the surveyed peanut flavour literature employing a non -

linear transformation prior to the regressions. The logarithmic transformation is of specific interest, as discussed in Chapter 3, because it places higher weight on volatiles with low headspace concentration. Currently, the human nose is still more sensitive at detecting many odour compounds than GC-MS instrumentation, and so it is likely that several potentially important compounds such as mono, di and trimethylpyrazines will only give a small signal (Liu et al., 2011). Linear regression has been shown to underestimate the correlation coefficient of compounds that appear in lower concentrations versus those appearing in higher concentrations (Chambers & Koppel, 2013).

The fact that the psychometric function (the function connecting physical stimulus, such as concentration, to human response, such as perceived odour intensity) is often logarithmic (O'Mahony, 1986) and that sensory thresholds are related to partition coefficients which are also logarithmic (Abraham, Gola, Cometto-Muniz, & Cain, 2002) are further indicators to the potential usefulness of the logarithmic transformation. Even though it can be seen in both Chapter 3 and **Table 23** that the logarithmic transformation greatly improved the fit of the sensory-headspace volatile models, the same improvement was not observed in Chapter 5. There are likely two reasons for this: Firstly, liking does not necessarily follow the psychometric function, because liking is both multimodal (vision, tactile, odour and taste) and also related to other factors (e.g. previous experience, cultural heritage and others). Secondly, the headspace profile in Chapter 5 was analysed with a Solid Phase Micro Extraction Gas Chromatography Mass Spectroscopy (SPME-GC-MS) setup, and not Dynamic Headspace Gas Chromatography Mass Spectroscopy (DHS-GC-MS) (Snow & Slack, 2002). It is proposed therefore, that the logarithmic transformation on headspace volatile data is a useful treatment for improving the fit of linear models against sensory attributes, especially when the headspace volatile is obtained by DHS-GC-MS.

To the second point, it is likely that any regression analysis of real-life data will produce both positive (correlated) and negative (inversely correlated) model parameters. However, in the area of headspace volatile fingerprinting, most researchers have only focused on positive correlations, presumably because their ultimate goal was to determine causation. Both conventional wisdom and the mechanics of flavour reconstitution studies assume that the aromas are caused by certain compounds, not by their absence. Even though this may be technically true for model systems, in whole food systems antagonistic effects (the presence of one compound reduces the perceived intensity of an aroma attribute, either by antagonizing for the same receptors or by having a strong, 'masking' aroma of their own (Linforth & Taylor, 2010; Warner, Dimick, Ziegler, Mumma, & Hollender, 1996)) and chemical interactions (one compound chemically reacts and modifies an odorous compound (Chapter 5)) are not uncommon. As a result, it is proposed that there is value in studying both positive *and negative* correlations in sensory-volatile compound models, but this has not been observed in the surveyed literature.

Finally, this research also demonstrated a novel approach for analysing sensory data, wherein categorical Check All That Apply (CATA) data could be modelled against a large number of instrumental attributes. CATA questions are easy to answer and can be used to probe a wide range of attributes in a ballot without fatiguing the respondent and are thus particularly useful in research with a large number of samples or long questionnaires. The drawback however, is that the response is binary (respondent checked or did not check) and as a result it has limited value as a response variable for common modelling techniques that require continuous response variables. This can be resolved by transforming it to a probability and using logistic regression, but the output becomes harder to interpret. However, Ares and Jaeger have noted that given a large enough sample size (>40) the number positive responses can be summed, and the response effectively converted to a continuous variable with a range from 0 to 1 using **Equation 1** (Ares & Jaeger, 2013).

$$Response_{continuous} = \frac{\sum_{i=1}^{n} Checked_i}{n}$$

Equation 1

Where, i is the i^{th} respondent, $Checked_i$=1 if the i^{th} respondent check the answer, and n is the total number of respondents. Further, Tenenhaus et al have demonstrated that Partial Least Square Regression (PLSR) is an excellent tool for relating hedonic scores to a large number of instrumental attributes (such as headspace volatiles) (Tenenhaus, Pagès, Ambroisine, & Guinot, 2005). This research demonstrated that the two approaches can be combined, and that **Equation 1** can be used to generate continuous response variables for consumer attributes ('stale' and 'fresh' in this case) which are consequently fed into a PLSR algorithm, to return meaningful correlations between said consumer and instrumental attributes. This is a simple but powerful approach to quickly scan for the drivers of multiple consumer attributes, something not previously possible due to the excessively long questionnaire this would require.

Finally, this research has demonstrated the value in taking advantage of modern computing power availability to revisit the study of seemingly simple commodities. Even though multivariate approaches were available when most of the foundations in peanut research were laid over the last 60 years, they were rarely used before 2000 due to their large computational needs. There are several instances in this research that a multivariate approach has delivered results where univariate approaches failed, such as the successful PLSR modelling of sensory textural attributes based on large deformation data (Chapter 4) when a one-variable-at-time regression previously failed (Lee & Resurreccion, 2006b).

6.2.2 **Research applications**

This research has several practical applications related to the control of flavour, texture and consumer preference that nut roasters can leverage, outlined in **Figure 36**.

In the area of flavour, this research has identified headspace volatiles that are highly correlated with desirable and undesirable attributes. As discussed below causality cannot be inferred without reconstitution studies, but the correlations point to the most likely candidates for such an exercise. Compounds which are proven to be causing some of the desirable attributes can consequently be added to the product, either directly in the form of a flavouring, or indirectly by providing the required reagents in the cases where the formation mechanism is known. Similarly, compounds proven to reduce liking could be controlled by the use of active packaging. This can be done by removing reagents or catalysts, as in the case of multi-layered packaging containing green tea extracts shown to scavenge free radicals and reduce hexanal build up in chocolate peanuts (Carrizo, Taborda, Nerin, & Bosetti, 2016), or directly by absorbing the offending compounds, as in the case of cyclodextrins in EVOH films, shown to reduce the presence of aldehydes (especially hexanal) in fried peanuts (Lopez-de-Dicastillo et al., 2012; Lopez-de-Dicastillo, Catala, Gavara, & Hernandez-Munoz, 2011).

With regards to texture, the results offer even more near term applications to producers. The impact of moisture addition on improving the textural profile of peanuts was demonstrated. ‘Blister fry’ peanuts have been available for a long time (Shi et al., 2017), but this study showed that by manipulating the maceration medium, increasing the crunchiness and introducing ‘blisters’ can be decoupled and texture can be manipulated independently of the visual appearance. This allows recovering the crispy texture in soft varieties/lot of peanuts that would have otherwise been discarded, resulting in both savings for the producers and a reduction in food waste (an area of focus (FAO, 2011)). Finally, as discussed below, the results could potentially lead to a single step wet blanching-roasting process that could improve the textural quality of undesirable raw materials.

The identified correlations between colour, flavour, texture and preference also have other applications. The results show that due to this correlation, colour is a good proxy for ensuring constant quality. Colour can be accurately, quickly and economically measured, allowing production managers to ensure that their product is within specification without resource consuming sensory panels or headspace volatile analysis. This can potentially be taken a step further, by automating the operation of the fryer or oven using a closed loop control system feeding from a colour sensor as shown in **Figure 38**.

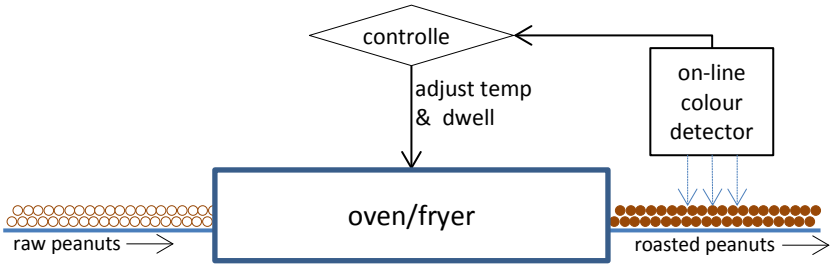


Figure 38: A schematic of a closed loop colour control system.

With regards to preference, the research identified the attributes that drive consumer liking and disliking; in other words, the desirable and undesirable sensory attributes. One can argue that the analysis objective is a reduction in the dimensionality of the dataset: starting form a large number sensory attributes, the few attributes affecting consumer preference have been identified. This information can be used to make product optimization easier, by reducing the number of factors one needs to evaluate. Consumer tests with a narrower scope, number of samples and ultimately cost can be therefore run to determine the optima for the different attributes, using simpler diagnostic surveys such as Just About Right (JAR) questions (Lawless & Heymann, 1998).

The discovery that the drivers of consumer preference in the Netherlands, Spain and Turkey are very similar also has large practical implications: These three countries have different national cuisines, eating habits and flavour preferences, and yet when it comes to roasted peanuts their preference is remarkably similar. The reason for this is unclear: perhaps peanuts are seen as a commodity, and consumers do not have strong preferences other than avoiding raw or rancid off-flavours. Perhaps consumers in all three areas have similar tastes because they have all been exposed to the same, non-diverse supply of peanuts over the last six centuries (originally imported from the Americas as part of the Columbian exchange (Sokolov, 1993)). What is important however, is that their preference is similar, which implies that the same product design could be marketed in all three areas.

Finally, the knowledge created by this study allows for the development of new products. These can be based on product characteristics determined by processing technology such as ‘extra crispy’ macerated peanuts, or ‘dark roast’ glucose infused peanuts. The impact of baking vs frying has been characterized, and coupled with the other processes identified that control texture, a baked peanut could be produced with a similar texture to a fried one, but with a lower oil content and therefore superior perceived nutritional qualities. Additionally, the identified drivers of liking can spawn new marketing concepts. An example of this could be a range of ‘extra-large peanuts’. Since kernel size is a driver of liking, procurement can source a range of large peanuts, and marketing can create a

concept around the offer. The results presented here suggest that consumers in Europe are likely to find the offer appealing.

6.3 Technical considerations on modelling

6.3.1 Statistical power vs overfitting: The case for breadth vs depth

It was clear that several models would have to be built in order to achieve the aims of the thesis, and so a consideration of some of the technicalities of statistical modelling needed to be made in order to ensure the experimental design best served the research objectives.

Statistical modelling aims to explain the observed variance between the experimental treatments (response variables) by attributing it to factors (explanatory variables) (Ott & Longnecker, 2001a). Any remaining, unexplained variable is referred to as the 'model error'. Experimental error (random, but not systematic) will contribute to the model error, as it introduces noise to the data. In addition, the model error will be larger if factors that were significant are not included in the model ('under-specification of the model' (Ott & Longnecker, 2001b)). An underspecified model, in other words, contains fewer terms than are needed to fully model the response variable. On the other extreme, an over-specified model contains additional, irrelevant parameters that do not contribute to resolving the variance caused by the different treatments. This can cause several issues, the most important of which is 'over-fitting'.

In over-fitting, the additional model factors are actually modelling the data set error (Hastie, Tibshirani, & Friedman, 2009). As a result, the model diagnostics (e.g. P-value, and R^2) will keep improving, but the additional apparent model power is meaningless as it is merely modelling the dataset error (Harrell, 2016). Overfitting manifests as several non-important factors appearing as significant in the model, and extreme cases have been demonstrated where random numbers have been successfully modelled given enough model parameters (Hastie et al., 2009). One can only definitively demonstrate that a model is not over-specified if it can pass a validation process, but a good practice is to try to keep the number of model parameters to a minimum, and select them based on some a priori hypothesis (Ott & Longnecker, 2001b).

To avoid over-specifying, the General Linear Models developed in Chapters 2 and 4 only included the parameters related to the process conditions under investigation. Additional characteristics such as variety and origin were kept out of the models. It is very likely that the model P and R^2 values would be significantly

improved if additional sample parameters were included (such as market type, origin or variety), but this would spread the degrees of freedom over more model parameters. For the same reasons, the preference models developed in Chapter 5 were run on principal factors and not raw variables. The intent was to greatly reduce the number of model parameters by pre-treating the 'process' variables with a factor analysis. The factor analysis greatly reduced the dimensionality of the data from 20 (sensory attributes, colour and size parameters) to 3 factors. In addition, although the factors where not exactly orthogonal due to the VariMax rotation, they are significantly less auto-correlated than the raw variables.

Finally, there is also a non-Bayesian argument as to why using potentially less powerful models with fewer parameters is practically advantageous. In a less statistically powerful model, the effect of a treatment needs to be larger before the corresponding parameter is identified as significant (leading to reduced type I errors/false positives). This offers practical advantages, because only largely significant effects will be identified. In the peanut category there is little practical value to knowing that a statistically significant but very small in magnitude difference exists between two treatments. This is both because it is likely that the exact comparison will not be relevant in the future (as discussed in Chapter 1, hundreds of varieties are released every year, and differences can be expected between crop years) and because consumers are not as concerned with only minor organoleptic differences as seen in Chapter 5. It can be seen therefore, that although a powerful model would be able to identify even small differences between treatments, in an industrial environment this information would essentially be irrelevant.

6.3.2 Model fit and assumptions

Table 23 shows a summary of 45 models developed in this thesis. It can be seen that the model fit metrics range from good to excellent, and more detailed discussion of each model can be found in the appropriate chapter. The models fall under several types, each with different mechanics and quality metrics, but in general they all make three basic assumptions: Independent and normally distributed data, and additive effects (Ott & Longnecker, 2001a; Tenenhaus et al., 2005). All three assumptions are satisfied reasonably well (data not shown).

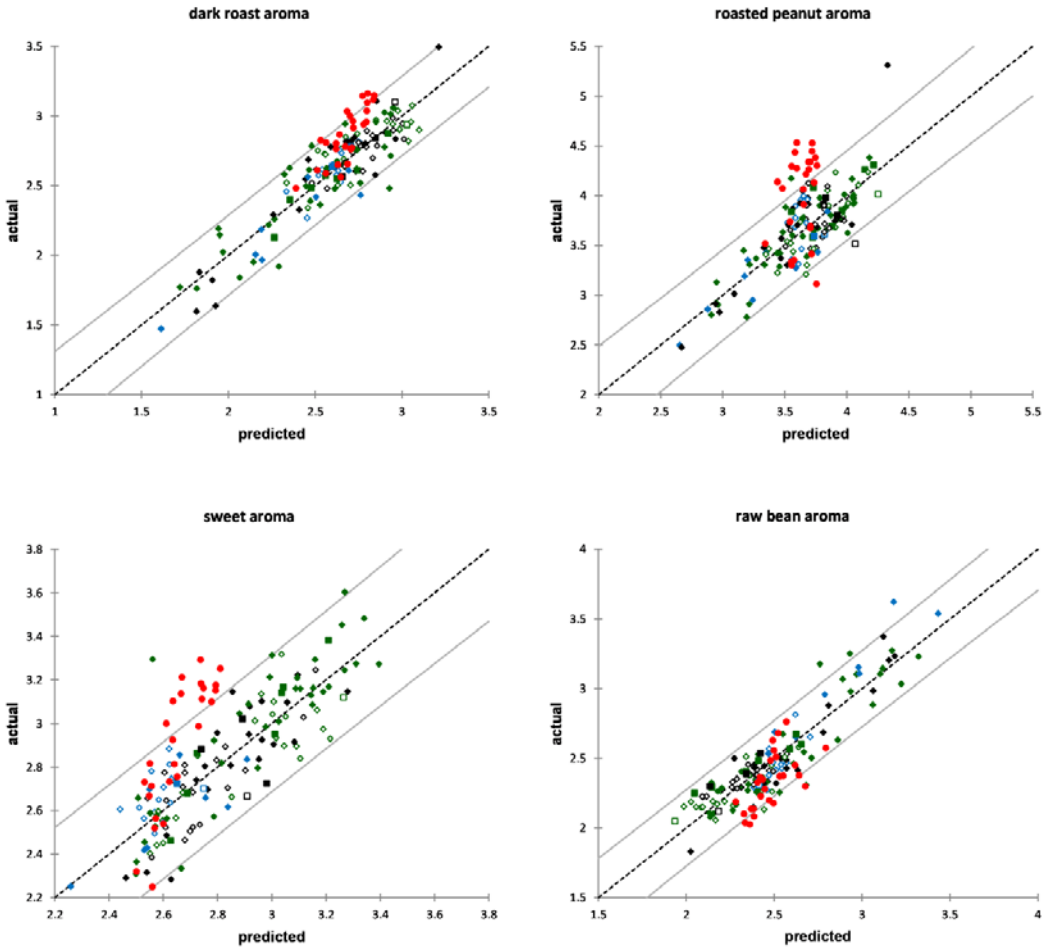
Table 23: Summary of all the models developed, including significance, fit and predictive quality. a: F statistical probability of all model parameters being zero, b: Analysis of variance, c: partial least squares regression, d: General linear model.

response variable	explanatory variables	appears in chapter	model type	Pr>F [a]	R^2	Q^2
L*	process conditions	2	ANOVA [b]	<0.001	-	-
a *	process conditions	2	ANOVA	<0.001	-	-
b *	process conditions	2	ANOVA	<0.001	-	-
dark roast aroma	process conditions	2	ANOVA	<0.001	-	-
raw bean aroma	process conditions	2	ANOVA	<0.001	-	-
roasted peanut aroma	process conditions	2	ANOVA	0.015	-	-
sweet aroma	process conditions	2	ANOVA	0.094	-	-
sweet aroma	process conditions	2	ANOVA	0.005	-	-
dark roast aroma	headspace volatiles	3	PLSR [c]	-	0.639	0.294
raw bean aroma	headspace volatiles	3	PLSR	-	0.661	0.372
roasted peanut aroma	headspace volatiles	3	PLSR	-	0.383	0.162
sweet aroma	headspace volatiles	3	PLSR	-	0.501	0.278
dark roast aroma	log headspace volatiles	3	PLSR	-	0.827	0.599
raw bean aroma	log headspace volatiles	3	PLSR	-	0.837	0.612
roasted peanut aroma	log headspace volatiles	3	PLSR	-	0.678	0.352
sweet aroma	log headspace volatiles	3	PLSR	-	0.710	0.545
breakdown	instrumental texture	4	PLSR	-	0.572	0.555
crispy	instrumental texture	4	PLSR	-	0.347	0.346
crunchy	instrumental texture	4	PLSR	-	0.710	0.690
hardness	instrumental texture	4	PLSR	-	0.680	0.666
breakdown	process conditions	4	ANOVA	<0.001	-	-
crispy	process conditions	4	ANOVA	<0.001	-	-
crunchy	process conditions	4	ANOVA	<0.001	-	-
hardness	process conditions	4	ANOVA	<0.001	-	-
fresh (pooled)	headspace volatiles	5	PLSR	-	0.848	0.471
fresh (ESP)	headspace volatiles	5	PLSR	-	0.778	0.313
fresh (NL)	headspace volatiles	5	PLSR	-	0.857	0.517
fresh (TR)	headspace volatiles	5	PLSR	-	0.822	0.442
liking (pooled)	headspace volatiles	5	PLSR	-	0.862	0.492
liking (ESP)	headspace volatiles	5	PLSR	-	0.841	0.436
liking (NL)	headspace volatiles	5	PLSR	-	0.867	0.530
liking (TR)	headspace volatiles	5	PLSR	-	0.828	0.429
stale (pooled)	headspace volatiles	5	PLSR	-	0.855	0.576
stale (ESP)	headspace volatiles	5	PLSR	-	0.908	0.680
stale (NL)	headspace volatiles	5	PLSR	-	0.759	0.394
stale (TR)	headspace volatiles	5	PLSR	-	0.800	0.469
liking (pooled)	instrumental and sensory	5	PLSR	-	0.939	0.648
liking (ESP)	sensory and appearance rotated factors	5	GLM [d]	<0.001	0.864	-
liking (NL)	sensory and appearance rotated factors	5	GLM	<0.001	0.786	-
liking (TR)	sensory and appearance rotated factors	5	GLM	<0.001	0.816	-
liking (pooled)	sensory and appearance attributes	6	PLSR	-	0.938	0.705
liking (ESP)	sensory and appearance attributes	6	PLSR	-	0.924	0.531
liking (NL)	sensory and appearance attributes	6	PLSR	-	0.857	0.560
liking (TR)	sensory and appearance attributes	6	PLSR	-	0.908	0.642

6.3.3 **Sensory – instrumental models**

The confidence intervals of the model parameters and predictions were derived using the jack-knife ('Leave One Out (LOO)') method (iteratively calculated by using all but one datum per iteration, thereby also minimizing the bias (Abdi & Williams, 2010)). However, for models with a large number of parameters (as in this research), it has been noted that the jack-knife derived variance is often overestimated (Tenenhaus et al., 2005). This can explain the relative high predicting power of the models despite somewhat large standard error of the β-coefficients reported in the previous chapters. To test the robustness of the sensory vs instrumental PLS models, an external cross validation was also performed. The validation set was obtained from the samples prepared for the

consumer testing, as they had not been previously included in the model. **Figure 39** shows actual versus predicted for all sensory models with the 95% confidence intervals, as well external validation data where relevant data points are marked in in red.



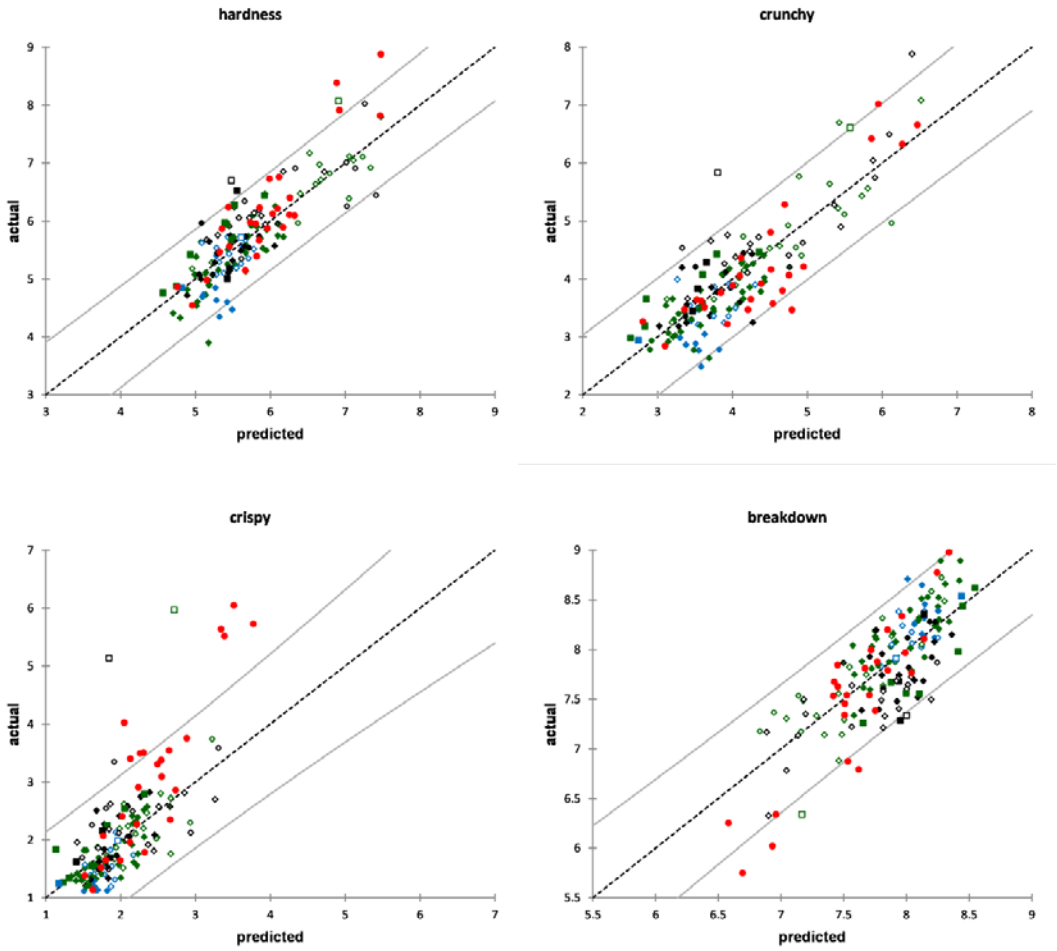


Figure 39: Actual versus predicted (95% confidence interval of the fit) for all sensory attribute models built with Partial Least Squares Regressions. The 95% confidence interval of the fit was derived by jack-knifing (Leave One Out). Diamonds: dry roasted, squares: oil roasted (fried). Filled shapes: non-macerated, empty shapes: macerated. Colour denotes market type: Black: Virginia, Blue: Valencia, Green: Runner. The external validation samples are marked with red circles.

At a first glance, it is obvious that that the flavour models are better fitting than the texture models, as there significantly fewer points outside the 95% confidence intervals. For flavour, the deviations are quite small in general (outliers are close to the 95% confidence curves), and are mostly caused by Virginia and Valencia type peanuts, rather a specific process technology. In contrast for texture, most of the poorly fitting samples seem to be due to maceration processes, rather than a specific cultivar or market type.

The plots are consistent with the model metrics presented in **Table 23**: the 'crispy' model has the poorest predictive power, most likely due to the reasons discussed in Chapter 4 regarding lack of acoustic analytic methods. As with most models, the prediction error tends to get larger at extreme high or low values, an effect known as 'shrinkage' (Harrell, 2016). The training samples however were developed to cover as wide sensory space as possible, and so it can be expected that most samples will be well within the experimental space, and the need for extrapolation will rarely arise. With the vast majority of the validation samples lying within the 95% confidence interval therefore, it can be claimed that the models fit rather well.

For the flavour attribute models, there are significantly more samples outside the 95% confidence interval which is to be expected for two reasons: Firstly, as previously discussed, different GC-MS setups were used (DHS vs SPME). Unfortunately, the SPME equipment was not available for the earlier part of the project, and as a result the flavour sensory-analytical correlations had to be calculated with DHS data. Secondly, only some of the headspace volatiles were analysed in the validation set. As discussed in Chapter 5, only odorous compounds (as identified by the GC-MS-O analysis) and certain compounds highly correlated to sensory attributes (as identified in Chapter 3) were analysed (total of 44 compounds). In contrast, the flavour models included all (103) compounds identified in Chapter 3. As a result the predictive power of the flavour models cannot be fully validated, as not all the model inputs are available to make the predictions. This problem was partially overcome by using the 'imputation' technique (estimate value of missing parameters with the mean concentration for that compound found in the training samples). This technique essentially converts the model parameters for which information is missing into an adjusted intercept (Harrell, 2016).

Even with these two issues however, the predictive power is moderately good, with 'raw bean' and 'dark roast' aromas being the two best performing models.

6.3.4 Preference models

The predicted versus residual plots for the preference models can be seen in **Figure 40**. In this case there is only one (the same) outlier in all models, which corresponds to a dry roasted extra-large Virginia peanut. This was the largest peanut included in the study, which suggests that the preference models are likely under-estimating the importance of the size for consumer preference in all three countries.

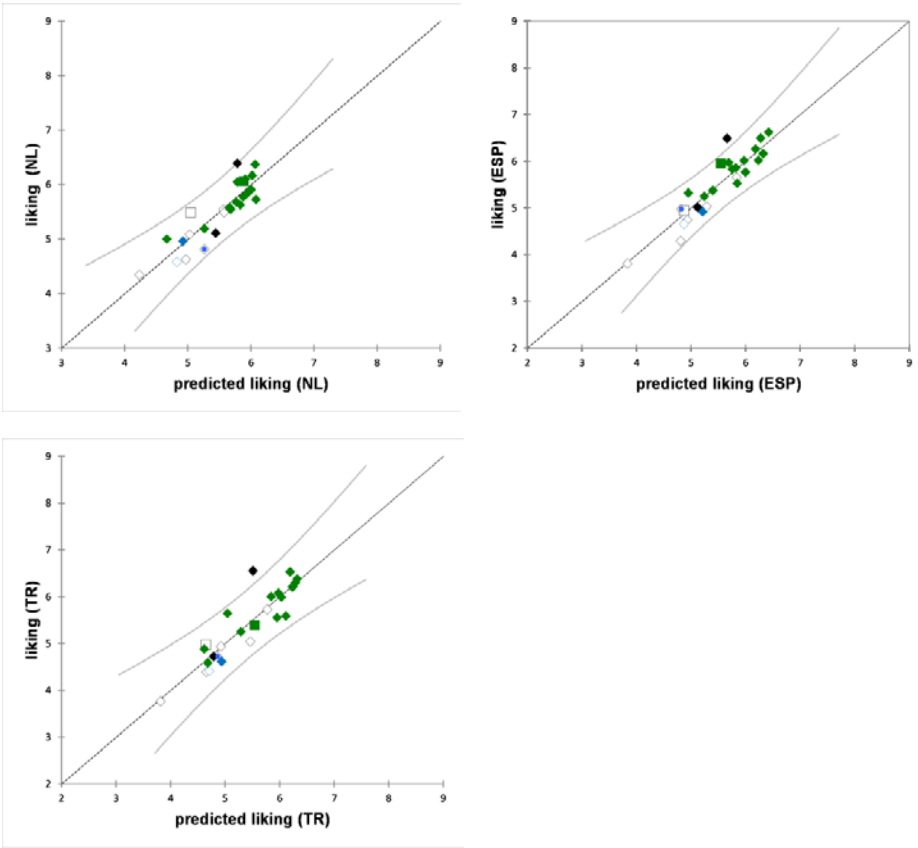


Figure 40: Actual versus predicted (with 95% confidence interval) for all consumer liking models (NL: Netherlands, ESP: Spain, TR: Turkey). Diamonds: dry roasted, squares: oil roasted (fried). Filled shapes: non-macerated, empty shapes: macerated. Colour denotes market type: Black: Virginia, Blue: Valencia, Green: Runner.

Cross validating the preference model is not as straight forward, primarily because the model is not meant to be predictive. The preference map approach aims at resolving the attributes that are responsible for consumer liking and disliking, not to predict it (Endrizzi, Gasperi, Rødbotten, & Næs, 2014). Furthermore, it is well understood that absolute hedonic scores are only meaningful in tests where only one sample is presented ('monadic' tests), since context (the rest of the samples presented) and presentation order (which sample is seen first) have a large impact on the hedonic rating (Lawless & Heymann, 1998). In addition, the number of samples included in a test also affects the absolute scores: the more samples are tested in one test, the lower the mean absolute hedonic scores (Vickers, Christensen, Fahrenholtz, & Gengler, 1993). Consequently,

even though mathematically the model can be used to predict the hedonic scores of validation samples, the prediction has no practical meaning because it will be assuming the sensory context of the training set.

Nevertheless, three approaches can be used to indirectly validate the model: One can compare the conclusions against literature, against preference models obtained with a different methodology, and finally one can perform a simple conventional cross-validation, but focus on the relative scores rather than absolute hedonic scores. Firstly, even though no preference studies for European consumers have been previously published, the conclusions presented here do agree with observations made by others on US consumers (Lee & Resurreccion, 2006a; Young et al., 2005) as discussed in Chapter 5. Secondly, in order to ensure none of the conclusions was a methodological artefact, the preference mapping exercise was repeated using a PLS regression (Liggett, 2010). The results were reasonably similar (R^2>0.9), considering PLS is a linear procedure (PREFMAP returned some quadratic models). It can therefore be deduced that there are no methodological artefacts in the preference mapping results.

Finally, a simplified external validation can be run to evaluate the model predictive power. This was done by using the model to predict the expected mean score and 95% confidence interval of two samples, and comparing the difference of the expected mean score to the actual mean score. Due to limited resources, only 2 samples were tested with n=32 consumers, only in the Netherlands. As a result, only the model for the Netherlands could be validated with this approach. The two samples selected were both fried Runners: a Flavorunner 458 (coded 'V1') and a Georgia Green (coded 'V2') cultivar (**Table 24**). The samples were selected to be of the same market type and process technology, and have predicted mean liking scores just over the 95% significance threshold. The same samples were consequently consumer tested (randomized presentation order; blind coded, with n=21 employees of PepsiCo Nederland, Maarssen, Netherlands and n=11 students and staff of Wageningen University, Wageningen, Netherlands). The predicted and actual scores can be seen in **Table 24**.

As discussed above, it is the preference order, rather than the absolute score that is relevant, and it can be seen that the model has successfully predicted that sample V1 is significantly preferred over V2 (p=0.025, 1 tailed paired t-test). As expected (Vickers et al., 1993), due to the much smaller number of test samples (2 vs 26), the absolute value of the scores are significantly higher to those described in Chapter 5. The validation test should only be taken as an indication of the model applicability, as it relies on relative small number of respondents and test samples and was performed on a different demographic than what was used to derive the preference models (Female: Male = 45:55 vs 50:50 and 18-34:35-54:55+ years old = 43:47:10 vs 30:30:40).

Table 24: Cross validation sample details, predicted and actual relative scores.

Code	Material	Process	Predicted mean score (9pt scale)	95% CI [a]	Validation test score (9pt)	Difference of prediction mean scores	Difference of actual mean scores
V1	Runner, Flavorunner 458, medium, high oleic, Texas, USA.	Oil roasting (fry) in high Oleic Sunflower Seed oil, 150°C, 4.5min	5.88	(5.72, 6.03)	7.28	0.63 [b]	0.48 [b]
V2	Runner, Georgia Green, jumbo, low oleic, Georgia, USA		5.49	(5.29, 5.70)	6.66		

[a] Confidence interval of the mean prediction, [b]: significant at P=0.05.

6.4 **Methodological limitations**

For practical and resource availability reasons, one can expect several compromises to be necessary in any exploratory research with several treatments and raw materials. Indeed, such compromises have also been made here and can be divided into two categories: experimental design (sample selection) and compositional analysis methods.

With regards to the experimental design, compromises had to be made in order to keep the number of samples manageable. As discussed in Chapter 1, this lead to an unbalanced selection of materials (more Runner than other market types) but also as summarized in Chapter 2 an unbalanced experimental design (not all materials were treated by all processes). This was an acceptable approach as the intent was to cover as many treatments as possible. However, maceration has emerged as a particularly promising treatment after the analysis, and so there would have been value in applying the process to all raw materials. It would also we worthwhile to perform more combinations of maceration conditions and frying, so that better comparisons with recent literature on 'blister fry' peanuts could be made (Shi et al., 2017). In addition, a more complete design around maceration treatments (particularly a comparison with more raw materials of water macerated-fried and dextrose macerated-fried) could have provided sufficient evidence to prove the hypothesis proposed in Chapter 4 on the role of melanoidins on the development of texture.

Margin for improvement also exists with respect to the analytical methodology. In Chapters 2-3, more robust correlations could have been obtained if a pre-concentration such as SPME (Solid Phase Micro Extraction) would have been employed in the headspace volatile analysis. This is particularly true for

solvent-type off flavour attributes which were not adequately modelled (R^2<0.1), which are often associated with very low sensory threshold compounds. SPME is a technique wherein an absorbent fibre is introduced into the sample headspace and left to equilibrate for several hours (Belitz, Grosch, & Schieberle, 2004). As a result the technique can somewhat compensate for the inadequate detection threshold of the MS detector for certain compounds, as the sample is essentially pre concentrated. This technique was leveraged for the volatile analysis of the consumer tested samples, but it was not available in our laboratory at the time the first part of the study was executed. This mismatch of volatile analysis methodologies is also preventing direct comparison of the results presented in Chapters 2 and 5, as well as the use of the Chapter 2 samples to externally validate the preference models developed in Chapter 5.

Use of the LC-MS-TOF (Liquid Chromatography Mass Spectrometry Time Of Flight) technique could also help develop better preference models. The technique can provide a profile of the non-volatile components of a sample (Belitz et al., 2004), but creating the method is complicated and time consuming, and the instrument was not available due to its very high cost. The preference models developed show the importance of sweet and bitter tastes and the texture analysis points to the potential importance of melanoidins content in texture, both areas where information on the non-volatile composition of the samples could have added great value.

With regards to the texture discussion in Chapter 4, it is clear that particularly for the 'crunchy' attribute, a better instrumental method is required. Large scale deformation tests were shown to provide a significant but not a complete description of texture instrumentally, and so additional acoustic analysis is recommended in future research. The 'crunchy' and 'crispy' sensory attributes are defined as acoustic attributes (Grosso & Resurreccion, 2002), and more recent literature has shown that acoustic data can improve the sensory-instrumental correlation of texture attributes (Tunick et al., 2013).

Finally, it is worth repeating that almost all the modelling techniques used in this research are linear, while most responses are likely not linear. Arguably, this is not as critical as it may appear if the objective is to identify drivers or correlations, rather than dwell on the exact coefficients of the models. In addition, by avoiding extrapolations and restraining the use of the models within the experimental range, a linear model may be an acceptable estimate of a non-linear response for a limited range.

6.5 **Future research**

It is important to remind the reader that the sensory-instrumental models presented here are intended to highlight correlations, not prove causation. There is significant benefit to identifying cross-modal correlations (Chambers & Koppel, 2013) not only because it may provide insight into common formation pathways (as in the case of colour and roasted aroma being correlated suggests a common Maillard pathway (McDaniel, White, Dean, Sanders, & Davis, 2012)), but because it also identifies proxies for fast and economical quality control tools (it is significantly faster and less costly to measure colour, than to run a headspace volatile or flavour profile panel in industrial setting) (Smyth et al., 1998). However, if causation needs to be proven, a full reconstitution study needs to be run, where the contribution of every compound can be quantified. Such a study has been run on Runner type peanuts (Chetschik et al., 2010; Da Conceicao Neta, 2010), but the results of this thesis here suggest it may be worth repeating with different market types (i.e. species of *Arachis*): Chapter 3 showed that several compounds which were in fact correlated to sensory attributes were not present in all samples, while Aprea et al noticed a similar effect in raspberries (Aprea, Biasioli, & Flavia Gasperi, 2015), where large qualitative and quantitative differences in the odour active compound profile was observed in different varieties, crop years, and post-harvest treatments. It is therefore possible that the current understanding of odour active compounds in peanuts is incomplete, as it is based on *Arachis Hypogaea* only, and not *Arachis Fastigiata* or other commonly cultivated species. In addition, reconstitution studies have not been done on samples of different crop years or processed with different technologies, even though large organoleptic and volatile profile differences have also been observed with these factors (Schirack et al., 2006).

With regards to further research that follows through the results presented here, there are three main areas: further investigate and validate some of the key results, extend focus to process pre-treatments such as blanching, and evaluate the meta factors affecting preferences (such as packaging, pricing, label information and sale channel).

Key results include the highly correlated nature of hydroxydihydromaltol (3,5-dihydroxy-6-methyl-2,3-dihydropyran-4-one) to ‘roasted peanut’, ‘dark roast’ and ‘sweet aroma’ sensory attributes, and the potential impact on melanoidins formation on texture development. Hydroxydihydromaltol has a strong ‘roasted aroma’ (Cutzach et al., 1997), but it only appears in less than 85% of the samples analysed. In addition, it appears in low but significant levels, resulting in the correlation to only be significant when the concentration data was logarithmically transformed. These two reasons could explain why it has not been previously identified in correlation or reconstitution studies, and so more research is needed to determine its relevance in the roasted odour character of peanuts. For texture,

melanoidins formation does appear to be a secondary mechanism for the development of texture during roasting, but further research is needed to definitively prove or disprove the effect. A preliminary Differential Scanning Calorimetry analysis was inconclusive (data not shown).

Pre-hydration of peanuts was shown to improve the texture of peanuts. However, moisture uptake is slow, and must be followed by an energetically expensive dehydration process. At the same time, there are wet blanching methods, in which the peanuts are immersed in water followed by a gentle drying (Woodroof, 1983). Further research is required to determine if a wet blanching process *combined* with aggressive roasting, could offer the similar texture modification as maceration produced in this study. If this is true, processors can leverage this combined blanching-roasting process to produce an acceptable finished product using particularly soft lots of raw peanuts.

With regards to the preference modelling additional work is also required. The research presented here identified the attributes that drive preference for European consumers. Now that the number of potentially important attributes has been reduced, additional consumer research is needed with a smaller number of samples, differing only on the critical attributes. The resulting smaller number of factors will allow evaluation of more levels, and so smaller differences between populations, and/or non-linear response optima can be resolved. For example, this research showed the b*CIELAB colour value is a driver of liking, but a follow up research of similar samples roasted to different b* values will more accurately show what is the optimal b* value. Finally, it is worth investigating the impact of meta factors such as pricing, packaging style and information on pack, as it has been seen to affect both the preference (He, Fletcher, & Rimal, 2005; Jolly, Hinds, Lindo, Ham, & Weiss, 2001; Lagerkvist, 2013; Nelson, Jolly, Hinds, Donis, & Prophete, 2005) and perception of freshness (Dinnella, Torri, Caporale, & Monteleone, 2014; Sääksjärvi, van den Hende, Mugge, & van Peursem, 2015). Often seen as a 'commodity' category, one may find that these factors have a larger impact on consumer preference than the product characteristics.

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7 Summary

Twelve different raw materials were processed in eleven different ways (including pre and post processing steps, such as maceration and topical oil application) to yield 134 unique samples. The samples were consequently analysed instrumentally for their physical and chemical characteristics, and by an expertly trained sensory panel for their sensory profiles (aroma, flavour, taste, texture and aftertaste). Of the 134 samples, the 26 most differentiated samples were further tasted by consumers in 3 countries (NL, ESP, TR) and hedonic ratings where obtained. Some of the most common peanut growing regions were selected: USA (Texas, Georgia and Virginia states), Argentina, South Africa, China and Australia. The market type was restricted to Runner, Valencia and Virginia, and some of the most common cultivars and grades (kernel sizes) from each type were selected. Similarly, only the most common processing methods were selected (dry and oil roasting), at several temperature and time combinations. Maceration in different media (glucose content and pH levels) and times was added to the design as a pre-treatment. The design resulted in 134 unique peanut samples (this was the sample set used in Chapters 2,3 and 4), while the aforementioned limitations ensured it was practical to execute. As a result, the study has one of the largest sample sets encountered in the literature.

The aim of this work was to identify correlations between sensory and instrumental attributes and consumer liking. To do so, a large number of instrumental analyses was performed on all samples measuring a variety of attributes, including the chemical composition (Gas Chromatography- Mass Spectroscopy headspace volatile analysis, sugar profiling by ion chromatography and amino acid profiling by Fatty Acid Methyl Ester gas chromatography), physical structure (large deformation stress-strain tests, moisture content analysis, confocal microscopy and X-ray Computer Tomography), and macro characteristics (kernel size distribution, CIELAB colour parameters). The sensory profiles were obtained through a highly trained sensory panel, while consumer liking data were obtained from consumers in three different countries (Netherlands, Spain and Turkey), using a minimum of 200 consumers per country.

The multidimensional data was analysed with Principal Component Analysis and Canonical Variate Analysis. Whenever a main effect or hypothesis was to be tested, a General Linear Model regression was the method of choice. Partial Least Squares Regression was used to build models relating the sensory with the instrumental attributes. Finally, the external preference mapping on liking means methodology (up to quadratic models) was used to identify the drivers of consumer.

The primary objective of this research was to determine what drives preference in snack peanuts for European consumers. Interestingly the drivers are very similar for Dutch, Spanish and Turkish consumers, and consistent with what has been previously reported for Americans (Chapter 5). Some consumer segmentation was detected within each country, but the segment means where not dramatically different from each other. Light colour (better interpreted as 'not too dark'), sweetness and the concentration of several pyrroles were the main drivers of liking, while hexanal and 2-heptanone were drivers of disliking and perceived staleness. CIELAB colour parameter b* mainly (but also L* and a* to a lesser degree) was by far the most significant driver of liking. This is not surprising as the colour parameters are highly correlated to roasted aromas themselves.

An investigation of the perceived 'fresh' and 'stale' attributes showed that they are not design driven, but mainly related to the oxidation state of the peanut lipids. The data suggests that liking and freshness are practically synonymous to the consumer, although this experimental setup cannot resolve whether the effect is causal. Perceived staleness however, appears to be a better distinguishable attribute for the consumer, and it is less strongly correlated to the inverse of liking, suggesting that the attribute carries incremental information.

The second objective of this thesis was to provide a better understanding of the impact of raw material and process technology and their interaction on the organoleptic profile of snack peanuts (Chapters 2 for flavour and 4 for texture). The aspiration has been to discover processing conditions that can compensate for the differences between raw materials so that a wider range of raw materials can be used, thereby reducing food waste and procurement costs. To this end, it was determined that maceration in aqueous media has a significant impact on flavour (increase 'roasted' and decrease 'raw beany' aromas), texture (increase 'crunchiness' and/or 'hardness' (if dextrose is included)) and colour (degree of browning, depending on pH of the medium), while the effect is significantly larger than the differences observed between raw materials. Interactions between maceration treatments and roasting methods were also observed. An example application this enables is the production dry roasted peanuts with the organoleptic properties of fried peanuts. Maceration however also affects the fatty acid composition due to hydrolysis, and so it is best used with high oleic cultivars. With regards to materials, it was demonstrated that 'blister fry' type processes also work on Runner and Spanish types (both *Arachis Hypogaea* and *Fastigiata* sub species), something not previously seen.

The third objective was to gain understanding of the compositional changes induced by processing and contrast their magnitude to those caused by different raw materials. With regards to flavour and aroma (Chapters 2-3), the chemical fingerprint of four sensory attributes was successfully determined ('roasted peanut aroma', 'dark roast aroma', 'raw bean aroma' and 'sweet aroma'). It was

demonstrated that a logarithmic transformation of the headspace volatile concentration data significantly improved the model fits, an approach not previously seen on peanut flavour research. Somewhat surprisingly, but in agreement with recent recombination studies on Runner type peanuts, pyrazines were seen to be less correlated to roasted peanut aromas. Compounds highly correlated to sensory attributes (but not always with a positive coefficient) included hydroxydihydromaltol (not previously reported in peanuts), 2/3-methyl-1H-pyrrole, benzeneacetaldehyde, 2-hexenal and 3-hexen-2-one. It was also observed that flavour and colour attributes were highly correlated, something that can be used to simplify quality control and product optimization in an industrial production environment. However, the correlation was not as strong when maceration processes were applied. In general the b* value was determined to be more significant than the L* value, which is the one more frequently referenced on the literature. Finally, the fatty acid profile was seen to be highly affected by maceration processed, mainly driven by increased saturation though lipid oxidation due to the aqueous environment.

With regards to texture (Chapter 4), most of the observed changes could be attributed to microstructure modification. Increased alveolation due to steam generation was linked to increased crunchiness and crispiness, a mechanism that is widely accepted. However, quantitative data on the degree alveolation were for the first time published in this research, which suggest that a secondary mechanism is also likely in effect. The analysis demonstrated that although maceration in different media results in a similar air cell distribution, there are significant sensory differences between the samples. Given that the effect was most pronounced in the presence of glucose (a reducing sugar) in combination with oil roasting (highest temperature process), the proposed hypothesis was that Maillard or other molecular interactions increase the local mechanical moduli. Interestingly, it was observed that Virginia type peanuts develop with roasting both flavour and texture faster and to a greater degree than Runner or Valencia types.

8 Dankwoord

‘Life-long learning, that’s what it is’. This was the first response I received from Fre Pepping when I contacted Wageningen for information on the PhD programme. And it sure has been! I want to thank Dr Edoardo Capuano for supervising my work. His attention to detail and the many interesting discussions this has led to have not only greatly improved the quality of my research, but have also taught me a lot about both the field and my personal strengths and weaknesses. I can honestly say that it was a pleasure working with you! A big thank you is also due to Prof Vincenzo Fogliano not only for helping to focus the research, but for taking a chance on an ‘industry guy’ to begin with!

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Dimitris.

9 About the author

Curriculum Vitae

Dimitris was born in Athens, Greece in 1980. After graduating from the Athens College International Baccalaureate (Athens, Greece) in 1998 he received a BSc in Food Science from the University of Leeds (Leeds, UK), with an Industrial Placement at the Slough campus of M&M Mars in the UK. While in the UK, his research focused on emulsion and chocolate rheology. In 2004 he attended Cornell University (NY, USA) earning an MSc in Food Engineering and Sensory Science, in the field of biopolymer rheology. In 2004 Dimitris joined the Research and Development function of Frito-Lay (TX, USA) and in 2008 transferred to the European Sector Research and Development division of PepsiCo International, in Maarssen, Netherlands. Dimitris has worked in several areas over the past 14 years at PepsiCo, launching numerous products in various countries and been awarded 6 international patents. In his spare time, Dimitris enjoys cooking, brewing (is a Certified Culinary Scientist since 2007) and motorcycle touring.

Publications

Peer reviewed scientific journals

1. **Lykomitros, D.**, Fogliano, V., & Capuano, E. (2016a). Flavour of roasted peanuts (Arachis hypogaea) - Part I: Effect of raw material and processing technology on flavour, colour and fatty acid composition of peanuts. *Food Research International*, *89*, 860–869.
2. **Lykomitros, D.**, Fogliano, V., & Capuano, E. (2016b). Flavour of roasted peanuts (Arachis hypogaea) — Part II: Correlation of volatile compounds to sensory characteristics. *Food Research International*, *89*, 970–881.
3. **Lykomitros, D.**, Den Boer, L., Hamoen, R., Fogliano, V., & Capuano, E. (2018). A comprehensive look at the effect of processing on peanut (Arachis spp) texture. *Journal of the Science of Food and Agriculture*. Advance online publication. DOI: 10.1002/jsfa.8920.
4. **Lykomitros, D.**, Fogliano, V., & Capuano, E. (2018). Drivers of preference and perception of freshness in roasted peanuts (arachis spp) for European consumers. *Journal of Food Science.* Advance online publiocation. DOI: 10.1111/1750-3841.14095.

International patents

1. Anand, A., Jacoby, B. P., **Lykomitros, D**., Puppala, V., & Rao, V. N. M. (*granted*:2012). 'Fruit and vegetable snacks' *US 8192784 B2*.
2. Anand, A., Hargrove, R.S., **Lykomitros, D**., & Rao, V. N. M. (*granted*:2012). 'Process for producing nut-based expandable pellets and nut-based snack chips'. US 8119181 B2.
3. Barnette, M., Guatam, A., Keller, L., **Lykomitros, D.**, Morales, J., & Richey, S. (*granted*:2011). 'Extruded legume snack food' *RU 2423874*.
4. Lawson, G., **Lykomitros, D**., & Smith, R. T. (*granted*:2016). 'Method for producing a crunchy food product' *CA 2715650 C*.
5. **Lykomitros, D.**, Graham, D. W., & Jacoby, B. P. (*application*:2009). 'Vegetable Containing Food Product and Method of Making' *WO/2009/155428*.
6. **Lykomitros, D.**, O'Sullivan, K., Rao, V. N. M., & Del Mercado, B. V. (*granted*:2013). 'Method for making a coated food product having a heat susceptible coating'. *US 8394437 B2*.
7. **Lykomitros, D.**, & Ripberger, D. R. (*granted*:2011). 'Rice snack chip with high rate of visual inclusions made on tortilla sheeting equipment'. *US 8080273 B2*.
8. Vera-Nunez, D. V., Han, J., **Lykomitros, D.**, Campbell, J., Mcgarvey, R., & Stewart, C. (*application*:2013). 'Production of expanded nuts'. WO/2015/099665
9. **Lykomitros, D.**, Vera-Nunez, D., Van Temmen, R., Nielen, L. (*application:*2013) 'Production of enhanced nuts and legumes'. WO2013072387 (A2).

Overview of completed training activities

Category A: Discipline specific activities (courses, workshops, symposia, summer schools, conferences etc.) > 11 ECTS					
Name of the course/meeting	**Organizing institute (s)**	**City**	**Country**	**Year**	**Presenter**
Annual Global Research Forum	Pepsico	Shanghai	CN	2014	Oral
Process Scale up	Leon Levine & associates	Leicester	GB	2015	
Culinology on demand sessions	Research Chefs Association	webinar		2014	
Culinology annual meeting	Research Chefs Association	Denver	US	2015	
Annual Global Research Forum	Pepsico	London	GB	2015	Poster
Food Flavour	University of Nottingham	Nottingham	GB	2015	
Tortilla Chip University	Pepsico	Plano, TX	US	2015	
Dairy fundamentals	Pepsico	Webinar		2014	
Hands on Preference mapping	Hal MacFie Consulting	Paris	FR	2015	
16th Food Colloids Conference	WUR/TNO	Ede	NL	2016	
Advanced Consumer product testing	Hal MacFie Consulting	Bristol	GB	2016	
Fundamentals of Frying	Pepsico	Warsaw	PL	2016	
Applied Biocatalysis	VLAG, ENTEG and GBB	Groningen	NL	2017	

Category B: General courses (e.g. PhD week, writing and presenting courses, statistics, etc.) > 6 ECTS					
Name of the course	**Organizing institute**	**City**	**Country**	**Year**	
Facilitation for success	Nigel Allfrey associates	Hamburg	DE	2014	
Unleashing Innovation Summit 2014	Global executive events	Amsterdam	NL	2014	Panel
The inclusive leader	Pepsico	Athens	GR	2014	
Innovation fusion conference	European Networking Group	Berlin	DE	2015	Oral
Triz Level 1 certification: Creative technical problem solving	ICG T&C	Utrecht	NL	2016	
Managing remote teams	Marcus Evans	London	UK	2015	
LUX executive summit Europe	Lux executive summit	Amsterdam	NL	2015	
New Product Development Process and Implementation Overview	Pepsico	Maarssen	NL	2014	
Experience Design Seminar, 4th Ed	Jakajima BV	Eindhoven	NL	2015	
Coaching Manager	Pepsico	Maarssen	NL	2016	

Category C: Optionals (participation in discussion groups, PhD excursions, MSc courses, etc.) > 8 ECTS				
Name of the course	**Organizing institute**	**City**	**Country**	**Year**
Preparation of research proposal				2014
SIAL Innovation Trade show	SIAL	Paris	FR	2014
CS1156x Machine Learning	CaltechX	Online		2014
MITx_6.00.1 Introduction to programming using Python	MITx	Online		2015

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