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Coffee flavour: an overview

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ABSTRACT: This paper is a concise review of the research on coffee flavour to serve as a rapid reference on the subject. It covers the process of roasting coffee beans, the volatile and non-volatile components generated by the process and the chemical reactions responsible for their formation. Volatile compounds significant on the determination of coffee aroma are given according to the most recent research. Finally, the paper discusses the chemical indexes used over the years to characterize coffee flavour deterioration and estimation of shelf-life. Copyright © 2004 John Wiley & Sons, Ltd.

KEY WORDS: coffee flavour; volatiles; non-volatiles; chemical indexes

Introduction

Coffee is a tropical plant that grows at 600–1800 m above sea level.[1] It is native of Ethiopia, from where it spread first to India and then to Indonesia, Brazil, Colombia and Central America.[2] What we commonly know as 'coffee' is a beverage prepared by brewing roasted, ground beans. The plant produces red cherry-like fruits containing two seeds, which, after being separated from the fruit pulp, are known as 'green coffee'. They are packed in sacks and transported to consuming countries. When received, they are blended with green beans from other origins and roasted to produce the characteristic flavour and colour associated with coffee beverage.[3]

Roasting

Green coffee beans lack the colour and characteristic aroma of roasted coffee; both are formed during the roasting process. Coffee oil, which comprises about 10% of the roasted beans, carries most of the coffee aroma. The aroma is made up of a complex mixture of volatile compounds, whereas non-volatiles determine sourness, bitterness and astringency.[4]

The roasting process can be roughly divided into three phases:[2]

1. An initial drying phase, evidently endothermic, during which moisture is eliminated. The smell of the beans changes from green to peasy to bread-like, and the colour turns yellowish.
2. The actual roasting phase, during which a number of complex pyrolytic reactions take place. The chemical composition of the beans is drastically modified, with release of large amounts of carbon dioxide and the formation of the many hundreds of substances associated with coffee aroma and taste. The beans change colour to a dark brown. Initially the process is exothermic. Pyrolytic reactions reach a maximum of 190–210 °C, when the process becomes endothermic with the release of volatile compounds. The overall reaction becomes exothermic again at about 210 °C.
3. A final rapid cooling phase to stop the final exothermic part of the roasting operation, using air or water as the cooling agent.

The quantity of heat transferred to the beans is a very influential parameter of the roasting process. This can be controlled by the roasting temperature and time. The colour of the beans is directly correlated to the final roasting temperature: the higher the temperature, the darker the coffee, so that colour can be used to define the end of the operation. The degree of roasting is usually described as being 'light', 'medium' or 'dark'. Roasting time may be as long as 40 min, as is often the case in Brazil, or as short as 90 s in fast roasting to produce high-yield coffees. Roasting time influences the reactions within the bean: longer roasting periods produce a bitter coffee lacking a satisfactory aroma, whereas very short roasting periods may be insufficient for the completion of

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all pyrolytic reactions, resulting in coffee with underdeveloped organoleptic characteristics.[2]

Chemical Composition of Roasted Coffee

The chemical reactions that take place during roasting have not been completely elucidated. This results from the extreme difficulty of reproducing or simulating in the laboratory all the reactions that take place inside a bean, since:

1. Not all the active precursors of aroma, colour and taste present in green coffee have been identified.
2. The very large number of precursors give rise to complex reactions, difficult to isolate, since intermediate products react further.
3. Most reactions take place within intact bean cells, which have very thick walls. These walls are comparable to autoclaves, where the pressure can only be guessed.[2]

Roasting produces a net loss of dry matter, primarily as gaseous carbon dioxide, water and volatiles products of pyrolysis. There is a considerable degradation of polysaccharides, sugars, amino acids and chlorogenic acids, and a moderate relative increase in organic acids and lipid content. Roasting also produces high levels of caramelization and condensation products. Caffeine and trigonelline (N-methyl nicotinic acid) concentrations remain almost unchanged.[5]

Non-volatile Components

As aforementioned, chemical components of roasted coffee can be grouped into volatile and non-volatile substances, some of the former being responsible for the aroma and the latter for the basic taste sensations of sourness, bitterness and astringency. The non-volatile compounds in roasted coffee are:[2,5,11]

1. Caffeine, which contributes to the strength, body and bitterness of brewed coffee.
2. Trigonelline and its two non-volatile derivatives: nicotinic acid (of nutritional value) and N-methylnicotinamide.
3. Proteins and peptides that did not undergo Maillard reaction.
4. Polysaccharides, namely cellulose, hemicellulose, arabinogalactan and pectins, that play an important role in the retention of volatiles and contribute to coffee brew viscosity.
5. Humic acids or melanoidins, as final products of the Maillard reaction between amino acids and monosaccharides; they are the brown-coloured substances that impart to roasted coffee its characteristic colour.
6. Carboxylic acids, mainly citric, malic and acetic, responsible for sourness.
7. Chlorogenic acids, mainly cinnamic, caffeic, ferulic, isoferulic and sinapic, and their main degradation product, quinic acid, all responsible for astringency.
8. Lipids, including triglycerides, terpenes, tocopherols and sterols, that contribute to brew viscosity.
9. Minerals, with potassium being the most abundant (around 40% w/w). Metals such as manganese (10–50 ppm), iron (15–40 ppm) and copper (2–5 ppm) may catalyse reactions occurring during roasting and storage.

Carbon Dioxide

Carbon dioxide is quantitatively the most important non-aroma-contributing volatile compound in roasted coffee. It is generated by pyrolysis and the Strecker degradation reaction.[6] The amount is dependent on the degree of roast and can be up to 10 ml/g coffee.[7] This gas is released slowly in whole beans but rapidly after grinding. Because it has a zero dielectric constant, carbon dioxide is very soluble in the coffee matrix. Its loss from the bean is most likely due to relationships between temperature and the glass transition state in the system, although this later physical effect has not been explored.

After coffee grinding, much of the carbon dioxide produced during roasting is released. A suitable degassing time (2–8 h) must be applied before packaging using hermetic containers. Efficient degassing of roasted ground coffee is critical when it is packaged in flexible bags without vacuum. If the time for degassing is too short, the package can swell, losing appearance and even bursting. To solve this, gas valves can be placed in the wall of plastic bags to allow the release of carbon dioxide.[8]

It has been reported that 45% of the carbon dioxide is released during the first 5 min after grinding.[9] One work in the literature states that 1.21 ml/g carbon dioxide and 0.002 ml oxygen/g roasted ground coffee (Arabica) were released during the first hour after grinding.[10] As particle size decreases, the release of carbon dioxide is facilitated because of greater surface-to-volume ratios. A decrease of particle size from 1000 to 500 μm has been reported to double the release of the gas.[7] Pressurization with nitrogen and carbon dioxide itself helps to prolong the shelf-life of espresso coffee, because it maintains carbon dioxide inside the coffee cells, avoiding cell wall damage and further flavour release.[11]

Volatiles

The mechanisms of formation of coffee aroma are extremely complex and there is clearly a wide range of interactions between all the routes involved. The major mechanisms include:[2,12,13]

1. Maillard reaction (non-enzymatic browning): a reaction between nitrogen-containing substances on the one hand (proteins, peptides, amino acids, serotonine and trigonelline) and reducing carbohydrates, hydroxy-acids and phenols on the other, to form aminoaldoses and aminoketones by condensation.
2. Strecker degradation: a reaction between an amino acid and an α-dicarbonyl with the formation of an aminoketone that condenses to form nitrogen heterocyclic compounds or reacts with formaldehyde to form oxazoles.
3. Breakdown of sulphur amino acids, viz. cystine, cysteine and methionine, that are transformed into mercaptans, as well as thiophenes and thiazoles, after reacting with reducing sugars or intermediate products of the Maillard reaction.
4. Breakdown of hydroxy-amino acids, viz. serine and threonine, able to react with sucrose to form mostly alkylpyrazines.
5. Breakdown of proline and hydroxyproline, that react with intermediate Maillard products; the former gives pyridines, pyrroles and pyrrolyzines, whereas the latter forms alkyl-, acyl- and furfurylpyrroles.
6. Degradation of trigonelline, forming alkyl-pyridines and pyrroles.
7. Degradation of the quinic acid moiety, forming phenols.
8. Degradation of pigments, mostly carotenoids.
9. Minor lipid degradation, primarily diterpenes.
10. Interaction between intermediate decomposition products (mostly unknown).

Aroma Volatiles in Roasted Ground Coffee

Active research on the volatile composition of coffee has taken place in the last 25 years with the advent of gas chromatography–mass spectrometry. The principal classes of aroma compounds in roasted ground coffee are listed in Table 1.

Table 1. Classes of volatile compounds identified in roasted coffee

- Sulphur compounds
  - Thiols[14]
  - Hydrogen sulphide[15]
  - Thiophenes (esters, aldehydes, ketones)[16]
  - Thiazoles (alkyl, alcoxy and acetal derivatives)[17]
- Pyrazines
  - Pyrazine itself[18]
  - Thiol and furfuryl derivatives[19]
  - Alkyl derivatives (primarily methyl and dimethyl)[20]
- Pyridines
- Methyl, ethyl, acetyl and vinyl derivatives[14]
- Pyrroles
  - Alkyl, acyl and furfuryl derivatives[14,21,22]
- Oxazoles[23]
- Furans
  - Aldehydes, ketones, esters, alcohols, acids, thiols, sulfides and in combination with pyrazines and pyrroles[24]
- Aldehydes and ketones
  - Aliphatic and aromatic species[25]
- Phenols[26]

Significant Contributors to Aroma

The identification of new volatiles in roasted coffee may well become an exercise in futility because the central issue is to determine the chemicals effectively responsible for coffee aroma.[12] Gas chromatography–olfactometry (GCO) techniques, such as AEDA (aroma dilution extraction analysis) and CharmAnalysis™ have allowed the individualization of those key odorants by directly sniffing the chromatographic eluent. From the list of around 80 compounds proposed by Holscher and Steinhart,[26] Semmelroch *et al.*,[13] using AEDA, selected only 14 compounds to reproduce coffee aroma based on odour activity values (OAV), i.e. the ratio of concentration to odour threshold (Table 2). Deibler, Acree and Lavin[27] used CharmAnalysis™ to signal 30 potent odorants in brewed coffee, of which 18 compounds were identified by comparing their mass spectra, odour activity and Kovát's retention indices with those of authentic standards (Table 3).

Semmelroch and Grosch[28] quantified these potent odorants in ground roasted Arabica and Robusta coffee and in the corresponding brews, finding that a change in aroma takes place from roasted to brewed coffee due to a shift in the concentration of the odorants. The polar odorants, e.g. 4-hydroxy-2,5-dimethyl-3(2*H*)-furanone, were preferentially extracted, whereas the yield of the unpolar ones, e.g. 3-isobutyl-2-methoxy pyrazine, (*E*)-β-damascenone and 2-furfurylthiol, was low. On the basis of the quantitative data obtained, these authors formulated synthetic mixtures (aroma models) for the Arabica and Robusta brews. The models smelled clearly coffee-like and reproduced the differences in the odour profile of the original brews.

For roasted Arabica coffee, Czerny, Mayer and Grosch[29] carried out sensory assessments of model systems of 27 odorants in an oil/water mixture. An expert sensory panel evaluated the changes in the overall odour after omission of one or more odorants. These omission experiments indicated that 2-furfurylthiol, 4-vinylguaiacol, several alkylpyrazines, furanones, acetaldehyde, propanal, methyl propanal and 2- and 3-menthylbutanal had the greatest impact on the flavour of ground coffee. In contrast, the absence of 2,3-butanedione, 2,3-pentanedione, β-damascenone and vanillin in the aroma model according to the corresponding

Table 2. Odour activity values (by AEDA) and formation mechanisms of volatiles in the aroma of roasted ground Arabica coffee[13]

Compound	Odour activity value	Formation mechanism
(*E*)-β-Damascenone	2.7×10^5	Carotene degradation
2-Furfurylthiol	1.7×10^5	Maillard reaction
3-Mercapto-3-methylbutylformate	3.7×10^4	Maillard reaction
5-Ethyl-4-hydroxy-2-methyl-3(2*H*)-furanone	1.5×10^4	Maillard reaction
4-Hydroxy-2,5-dimethyl-3(2*H*)-furanone	1.1×10^4	Maillard reaction
Guaiacol	1.7×10^3	Phenol degradation
4-Vinylguaiacol	1.1×10^3	Phenol degradation
Methional	1.2×10^3	Maillard reaction
2-Ethyl-3-dimethylpyrazine	1.6×10^2	Maillard reaction
2,3-Diethyl-5-methylpyrazine	95	Maillard reaction
3-Hydroxy-4,5-dimethyl-2(5*H*)-furanone	74	Maillard reaction
Vanillin	48	Phenol degradation
4-Ethylguaiacol	32	Phenol degradation
5-Ethyl-3-hydroxy-4-methyl-2(5*H*)-furanone	21	Maillard reaction

Table 3. Aroma occurrences resulting from CharmAnalysis[27]

Compound	OAV (standardized)	Sensorial description
Sotolon	81	Toast
β-Damascenone	98	Fruit
2-Furfurylthiol	100	Toast
4-Vinylguaiacol	62	Cloves
2-Methyl-3-furanthiol	89	Nuts
Vanillin	71	Vanilla
Guaiacol	77	Plastic
Furaneol	58	Caramel
Methional	43	Potato
3-Methoxy-2 isobutyl pyrazine	38	Plants
2,4,5-Trimethylthiazole	38	Plastic
Abhexon	44	Honey
4-Ethyl guaiacol	31	Spice
5-Methyl-6,7-dihydrocyclopentapirazine	22	Cotton candy
2-Ethyl-3,5,-dimethylpyrazine	25	Burnt
cis-2-Nonenal	27	Toast
2-Isopropyl-3-methoxypyrazine	14	Green
2,3,5-Trimethylpyrazine	15	Toast

omissions was not noticed by the assessors. This study confirmed for the most part the compounds previously identified by Semmelroch *et al.*[13] with the exception of (*E*)-β-damascenone, the compound with the highest odour activity value. Although the degradation of carotenoids that generates β-damascenone has been regarded as having a major importance on tea flavour and is even suggested to play an important role in coffee aroma as well,[30] sensory studies did not validate this possibility.

In a follow-up study, Mayer, Czerny and Grosch[31] quantified the most potent odorants in brewed medium-roasted Arabica coffee. In accordance with previous results,[28] the more polar compounds yielded much larger amounts than the unpolar ones (>75% vs. <25%, respectively). An aroma model was prepared based on these results for the brew. In triangle tests, the model containing all 24 odorants was compared with a set of models missing one or more compounds using the aforementioned omission tests. These experiments indicated that the aroma of the brew was mainly caused by some alkylpyrazines, furanones and phenols, and by 2-furfurylthiol, methional, and 3-mercapto-3-methylbutyl formate. The higher impact of methional and formate and the lower aroma activity of 4-vinylguaiacol were in contrast to the results previously obtained for ground coffee of the same provenance and roast degree.

Finally, Mayer and Grosch[32] quantified 22 potent odorants in the headspace of roasted Arabica coffee powder, and a model mixture was prepared accordingly. When evaporated, the similarity of the aroma of the model to that of the roasted coffee headspace was scored 2.6 on a scale of 0 (no similarity) to 3.0 (identical). The model included odorants such as acetadehyde, methylpropanal, 2- and 3-methylbutanal, 2,3-butadione, 2,3-pentadione, 2-furfurylthiol, 2-ethyl-3,5-dimethylpyrazine and 2,3-diethyl-5-methylpyrazine.

Chemical Indicators of Sensory Deterioration

The onset of sensory staleness is used to determine the end of shelf-life of roasted ground coffee.[9] *Staleness* in coffee is defined as 'a sweet but unpleasant flavour and aroma of roasted coffee which reflects the oxidization of many of the pleasant volatiles and the loss of others; a change in the flavour and the acid constituents causing a partial bland tone'.[33] Since the late 1950s when the gas chromatographic work on coffee aroma was initiated, a great evolution in analytic and data processing techniques has occurred, and different staling indexes have been developed and correlated to sensory deterioration.

'M/B' Aroma Index

Raymond *et al.*[34] defined the M/B aroma index as the quotient of the concentrations of methylfuran and 2-butanone. This ratio decreased from 2.6 to 0.1 within 4 days in roasted ground coffee dissolved in water at 23 °C and 30 °C, which led to the suggestion that the M/B index could be used as an indicator of staling. However, no sensory results were presented and, in fact, this was a storage study of a 'coffee solution' and not the roasted ground coffee itself. Later Vitzthum and Werkhoff[35] found a linear correlation ($r^2 = 0.96$) between M/B and a sensory index reported on a 1–5 scale (5 = best, 1 = worst) for the aroma of roasted ground coffee stored at 20 °C in air (unstated moisture).

Further studies focused on the influence of the degree of roasting and origin of coffee on the M/B index. Kwasny and Werkhoff[36] observed that the M/B index determined in one kind of coffee might not be directly applied to others. An empirical exponential function was used to correlate the M/B index with degree of roasting:

$$\text{M/B} = a\ (\text{degree of roasting})^{b}$$

Robusta	a = 31.3	b = 0.59	($r = -0.9999$)
Salvador	a = 20.5	b = 0.49	($r = -0.9898$)
Kenya	a = 21.9	b = 0.52	($r = -0.9900$)
Colombia	a = 28.4	b = 0.60	($r = -0.9951$)

Furthermore, Spadone and Liardon[37] reported that the M/B index decreases continuously irrespective of storage temperature or oxygen partial pressure in the package (metal cans at 5% moisture, unstated basis). However, correlations between M/B index and sensory evaluation of brewed coffee prepared from the stored samples did not show a linear trend, thus undermining the validity of the parameter.

Flavour Quality Index (FQI)

Looking for a better index, Spadone and Liardon[37] applied multiple linear regression to results of volatile composition of roasted ground coffee vs. time of storage, and defined a flavour quality index (FQI) based on five key volatiles, viz. hexanal, vinylpyrazine, pyrrol, furfurylmethylketone and pyridine, with a good fit of their sensory data ($r^2 = 0.87$).

$$\begin{aligned}\text{FQI} = 6.53 &+ 0.027\,[\text{hexanal}] - 0.08\,[\text{vinylpyrazine}]\\ &- 0.04\,[\text{pyrrol}] - 0.022\,[\text{furfurylmethylketone}]\\ &- 0.001\,[\text{pyridine}]\end{aligned}$$

Nevertheless, the equation was only an experimental fit and did not have a theoretical basis.

'M/M' Aroma Index

Vitzthum and Werkhoff[35] defined the M/M aroma index as being the ratio of the concentrations of methanol to 2-methylfuran. This index showed an inverse linear relation with the M/B index and has been used to follow coffee staling in industrial settings.

'S' Aroma Index

Radtke and Piringer[38] defined the 'S' aroma index as the sum of the concentrations of 2-methylpropanal, 3-methylbutanal, diacetyl and 2-methylfuran, determined by gas chromatographic analysis of the static headspace of 10 g coffee dissolved in 40 ml water at room temperature. They also found a linear correlation between the index and sensory evaluation by a panel of experts.

Sulphur Compounds as Aroma Index

Steinhart and Holscher[39] applied headspace profile analysis in combination with a computer-aided discriminant analysis to evaluate aroma freshness of roasted whole coffee beans in non-air-tight packs at 20 °C. Their results indicated that aroma freshness of whole roasted coffee is mainly determined by certain low-boiling components, namely low-molecular Seight sulphur compounds, Strecker aldehydes and α-dicarbonyls. The loss of aroma freshness was due to the loss of certain aroma impact volatiles, mainly methanethiol, which then could be used as an indicator of freshness. The heavier components, such as furfurylmercaptan, will remain in the coffee and will cause an 'ageing' note. In addition, an increase of dimethylsulphide, the oxidation product of methanethiol, was observed, although no 'true' staling compound was found.

Summary

Past research on coffee flavour has built a significant knowledge base. About 900 volatile compounds have been identified, although less than 20 of them have been found to be relevant to coffee aroma. In addition, chemical indicators of sensory deterioration have been proposed and validated. Future research efforts will probably focus on the elucidation of mechanistic and kinetic parameters for the formation and deterioration of key aroma compounds. This would translate into practical guidelines for producing and maintaining a good coffee aroma.

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