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Sweetness flavour interactions in soft drinks

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Sucrose can be substituted by intense sweeteners to lower the calorie content of soft drinks. Although the sweetness is kept at the same level as much as possible, the flavour of the product often changes. This change could be due to both the mechanism of sensory perception and interactive effects of the aroma compounds. Several types of interaction and some techniques for measuring interactive effects are reviewed. An example of psychological interaction is the influence of colour on flavour. Interactions of flavour molecules with the receptor can be affected by changes in their micro-environment. Molecular interactions play a role in the release of volatile compounds from aqueous solutions; release is increased by sugars and salts, and decreased by lipids and proteins. Intense sweeteners, such as aspartame and neohesperidine dihydrochalcone, interact with volatile compounds and modify the intensities of flavour attributes. The use of combinations of intense sweeteners can solve the flavour problems encountered with single sweetener applications. A quaternary model of Beidler's mixture equation was used to describe the sweetness of a light blackcurrant soft drink, containing the intense sweeteners saccharin, cyclamate, aspartame and acesulfam-K. The perceived sweetness of the light soft drink was lower than the sweetness of the original sucrose-sweetened soft drink. A proportional enhancement of the concentrations of the intense sweeteners was utilized to meet the sweetness of this classic soft drink. Consequently, the aroma attribute strawberry increased, while the currant and sour related attributes decreased. Copyright © 1996 Elsevier Science Ltd

INTRODUCTION

The taste of a classic soft drink is largely defined by its sweet taste from sucrose, while from a nutritional and marketing point of view there is a large and increasing demand for a reduced intake of sucrose. Therefore, the production of beverages containing less sucrose is of increasing importance to the beverage industry (O'Brien Nabors & Gelardi, 1991). Sucrose can be subsituted by intense sweeteners to lower its content in soft drinks. Due to this substitution, however, the flavour of a soft drink changes. To obtain acceptable light beverages of optimum quality, their flavour should be similar to that of classic soft drinks. These drinks can be considered as flavour standards for beverages containing single sweeteners and combinations of them. The quality of the flavour of a beverage can be changed by the properties of the intense sweeteners themselves or because of interactive effects of the sweeteners with aroma compounds. Interactions between intense sweeteners and aroma active compounds can lead to the selective release of these compounds during consumption. Information on this release is important for selecting the appropriate flavour compounds to be added to a beverage and the methods for dispersing or isolating them (Kinsella, 1988).

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TYPES OF INTERACTION

When sucrose is replaced by intense sweeteners, the sweetness of a soft drink is kept the same as much as possible, but its flavour often changes. Interactive effects of aroma active compounds as well as the mechanism of sensory perception could account for this result. Interactions can occur in the soft drink itself (physical and chemical interaction), at the periphery of the sense (receptor interaction) or in the brain (perceptual interaction).

Physical and chemical interaction

Several factors determine the rate of release of a volatile compound from an aqueous food during consumption.

Homologous series of volatile compounds in aqueous solutions were used for preliminary research on their behaviour in drinks (Buttery *et al.*, 1969, 1971). A low solubility in these solutions increased the volatility of a compound (Buttery *et al.*, 1971). Drinks are complex mixtures of water, carbohydrates, lipids, proteins and other organic compounds, and all of them can interact with and/or bind flavours. The release of volatile compounds depends on the concentration of the volatile, its disposition in the drink (free, entrapped, adsorbed, complexed), the composition (components) of the drink, the amount of saliva in the mouth and the influence of temperature on their partition coefficients. The properties of the volatile compound (functional groups, molecular size, shape, volatility, etc.) and the physical and chemical properties of the components in the drink determine the relative importance of these factors (Kinsella, 1988).

Carbohydrates as polysaccharides contribute often to the viscosity of a beverage and therefore influence the diffusion of small volatile compounds. The effects of polysaccharides are usually of minor importance in soft drinks, because their concentrations are rather low (Overbosch *et al.*, 1991). Mono- and disaccharides affect the volatility by altering the activity coefficients of volatile compounds (Land, 1978). At relatively high concentrations, these components lower the amount of bulk water by structuring water, which increases the effective concentration of some volatile compounds and therefore can enhance their volatility (Nawar, 1971; Wientjes, 1968).

Lipids can adsorb or dissolve hydrophobic aroma compounds. In drinks these lipids are present in a dispersed form and mostly exist in distinct regions (for example droplets). The concentrations of hydrophobic volatiles in the aqueous and vapour phases are reduced, because of their physical partition between the lipid and aqueous phases of the drink (Buttery *et al.*, 1971).

Interactions between proteins and volatile compounds in drinks mainly depend on the type, amount and composition of the proteins, and temperature, pH and ionic strength of the medium (Kim & Min, 1988). The hydrophobicity of the protein influences the binding of volatile compounds. Small apolar compounds can diffuse into hydrophobic regions for further binding reactions (Solms *et al.*, 1973). Some compounds, such as aldehydes, can be bound irreversibly in covalent bindings with free sulphide and amino groups in the protein (Overbosch *et al.*, 1991). Van Ruth *et al.* (1995*a*) found a decreased release of volatile compounds from rehydrated bell peppers as well as from French beans, due to the presence of the protein mucin in saliva. The influence of pH and ionic strength of the medium is mainly evident from the isoelectric point, at which the protein can precipitate. Changes in conformation and solubility of the protein strongly alter the affinity to bind with volatile compounds (Dumont & Land, 1986).

Similar to carbohydrates, salts can increase the release of volatile compounds from drinks, due to a 'salting out' effect (Land, 1978). Salts in saliva did not affect the flavour release of some rehydrated vegetables (Van Ruth *et al.*, 1995*a*), probably because of their low concentrations.

As binding of aroma molecules to receptors largely depends on their concentration in the vapour phase (nose) for olfaction and in the aqueous phase (saliva) for taste (Kinsella, 1988), the perception of these compounds will be influenced by their physical and chemical interactions with components of the soft drink.

Receptor interaction

Taste receptors are stimulated by physical interaction with tastant molecules. A stimulus molecule absorbs on to the surface of the receptor, which creates a disturbance of the neural response of the receptor. Beidler (1954) assumes a concept of receptor binding, analogous to the Michaelis–Menten model for the study of enzyme–substrate reactions, in which one stimulant combines with one receptor. Monod *et al.* (1965) proposed a more complex model, in which the stimulants have to fit perfectly to the receptor. When the donor, acceptor and hydrogen atoms of a sweet molecule are colinear in the Shallenberger and Acree AH, B system, an optimal binding for sweetness is possible (Shallenberger & Acree, 1967). Lawless & Stevens (1983) did not find a cross adaptation of the intense sweeteners aspartame, sodium saccharinate and neohesperidin dihydrochalcone to one another or sucrose. Other psychophysical studies on this subject have been reported by De Graaf & Frijters (1986). The results indicate that the perception of sweet taste could be ascribed to more than one receptor mechanism.

The perception of the sweet taste of intense sweeteners largely depends on their binding affinity to the hydrophobic part of the receptor; it is also influenced by changes in the micro-environment. The relative binding strength of sweet molecules alters because of these changes (Van der Wel *et al.*, 1987). For example, Hoopman *et al.* (1993) noticed a decrease in perceived sweet taste due to an increased ethanol concentration in the solution. Kurihara (1992) has demonstrated taste modifications for the curculin protein due to changes in the micro-environment of its receptor. The protein induces a sweet taste when it binds to a sweet taste receptor. The sweet taste of this protein disappears in the presence of divalent cations of saliva, and is regenerated by tasting water, probably by removing the cations. At acidic pH divalent cations of the saliva do not bind to the receptor, and the sweetness of curculin will last longer in the presence of acids, as in soft drinks.

Perceptual interaction

The consumption of a food implies a simultaneous stimulation of several senses, e.g. vision, taste and smell. The character and acceptability of a drink is largely determined by the integration of percepts of these senses. Von Sydow *et al.* (1974) ascribed the enhancement of aroma perception by sucrose in beverages to psychological interactions rather than to receptor or chemical interactions. Frank & Byram (1988) examined the influence of strawberry aroma on sweetness. It was demonstrated that the modification of taste perception by olfactory stimuli was both tastant- and odorant-dependent. The influence of the strawberry aroma on sweetness seems to be more olfactory than gustatory. Murphy & Cain (1980) and Algom *et al.* (1993) proposed an additive model of taste–smell integration, in which the overall intensity of taste–smell mixtures is equal to the sum of the intensities of the unmixed components. Taste and smell behave in an independent manner; however, percepts of them are often mixed up, whereby olfactory stimulation can evoke sensations of taste. The influence of the colour of a stimulus on its taste or smell is often reported (Maga, 1974; DuBose *et al.*, 1980; Johnson & Clydesdale, 1982). It is probable that colours are associated with particular tastes and smells. Booth (1994) reports that indications about calorie content and artificial nature on the label influence sweetness and overall preferences for a soft drink.

Besides the integration of several senses, the interaction between different stimuli of one sense can be found on a psychological level, e.g. the interaction of sucrose and NaCl (McBurney & Bartoshuk, 1973; Smith, 1974). Kroeze (1978, 1979) and Lawless (1979) have shown that the interaction of sweet and salty or bitter tastes does not occur at the periphery, but at a higher level in the transduction process.

KNOWN INTERACTIONS PERFORMED BY INTENSE SWEETENERS

Several studies report on the chemical reactions of intense sweeteners with volatile compounds. The data of Hussein *et al.* (1984) showed that aspartame reacts with aldehydes like benzaldehyde, cinnamaldehyde, citral, *n*-decanal and vanillin. Also Le Quéré *et al.* (1994) found a decrease in the concentrations of several aldehydes in diet orange soft drinks containing aspartame, whereas Tateo *et al.* (1988) proved aspartame to be reactive with carbonyl compounds. Le Quéré *et al.* (1994) observed the formation of new volatile compounds in diet orange drinks containing cyclamate. These new volatiles were found to be structurally related to sodium cyclamate.

Concerning flavour enhancement, Higginbotham (1983) mentions special properties of thaumatin. In soft drinks, certain flavours such as blackcurrant, would allow a replacement level of sucrose up to 50% as the enhancement of flavour masks the aftertaste of thaumatin. Baldwin & Korschgen (1979) found a significant higher sensory intensity of fruit-flavour in orange- and cherry-flavoured beverages sweetened with aspartame than in their standards sweetened with sucrose. The intense sweetener neohesperidine dihydrochalcone (NHDC) can improve the overall flavour profile and mouthfeel of certain soft drinks, even at very low concentrations ($\leqslant 5$ ppm) (Borrego & Canales, 1992). Lindley *et al.* (1993) recorded an intensification of fruity flavour attributes by addition of 1–4 ppm NHDC to sweet and non-sweet beverages. In these cases NHDC acts as a flavour enhancer and modifier rather than as a sweetener.

TECHNIQUES TO STUDY MOLECULAR INTERACTION

The binding of flavours to components of soft drinks may be studied by sensory analysis and/or instrumental techniques (Kinsella, 1988). In order to obtain meaningful correlations of the data, sensory and instrumental measurements need to be considered together (Taylor & Linforth, 1994).

Sensory analysis

In sensory analysis, the human subject is used as an instrument (Köster, 1975). People can be trained to adopt an analytical attitude and to use judgement procedures which are needed for the analysis of percepts (Kroeze, 1990). As molecular interactions would change the perception of a soft drink, sensory analysis can be used for their study. Sensory analysis comprises difference and descriptive tests (Meilgaard *et al.*, 1991; Punter, 1991). In descriptive analyses, assessors assign attributes to a type of beverage and then give intensity scores for these attributes for different products (Punter, 1991). The amount of aroma compounds reaching the olfactory epithelium is different for each person. It varies with the adsorption along the respiration track, the flow of air and the amount of saliva in the mouth, the time, the temperature and the profile of the mouth (Rothe, 1988; Overbosch *et al.*, 1991). Measurements of the actual profile sensed by the olfactory bulb might improve the correlation between instrumental and sensory measurements (Taylor & Linforth, 1994). A special type of sensory analysis is the time intensity measurement, which can be used to study aroma and taste release from soft drinks in the mouth. Assessors score the intensity of a particular attribute with time during consumption (Cliff & Heymann, 1993).

Headspace analysis

The release of volatiles from a beverage can be estimated by static headspace analysis of the product (Buttery *et al.*, 1969, 1971; Franzen & Kinsella, 1974). The volatile compounds of most beverages are present in extremely low concentrations and therefore provide a near ideal state of infinite dilution. Under such conditions Henry's law can be applied, which means that the concentration of a volatile compound in the static headspace is proportional to its concentration in the aqueous phase of a soft drink, at equilibrium. However, the volatile profile of the static headspace can be below the detection limit of gas chromatography. Therefore, techniques were developed for isolation and concentration of aroma compounds from soft drinks, which are purge-and-trap, dynamic headspace analysis, steam distillation, vacuum distillation and extraction with solvents (Taylor & Linforth, 1994). Oral vapour gas chromatography offers a possibility to analyse the release of volatiles in the mouth of human subjects, who had to follow instructions during eating (Legger & Roozen, 1994). *In vitro* mouth models can be used to obtain volatile profiles, which are closely related to the profiles of oral vapour gas chromatography, as Van Ruth *et al.* (1995*b*) found for rehydrated bell peppers. Oral conditions influencing the flavour release are saliva composition and volume, temperature, mouth volume and mastication.

The dynamic headspace techniques imply a transport of the volatile compounds by an inert gas, after which the volatiles are condensed and/or adsorbed on an appropriate medium (Taylor & Linforth, 1994). Subsequently, the volatile compounds can be analysed by gas chromatography, using different detection methods (Van Ruth & Roozen, 1994). These methods are limited to qualitative analyses, because each adsorbance material has a different affinity for each volatile compound (Wyllie *et al.*, 1978). The identity of the volatile compounds can be determined by gas chromatography combined with mass spectrometry, flame ionization detection (retention times and peak areas), and description of odours by assessors at the sniffing port (Acree *et al.*, 1984; Van Ruth & Roozen, 1994). This sensory technique is capable of associating flavour descriptors with chemical constituents (Acree *et al.*, 1984). Human subjects are used to detect aroma-active compounds at the outlet of a gas chromatograph. They differ in sensitivity with chemical detectors, and therefore they produce different chromatograms (Acree & Barnard, 1994). The number of assessors who smell an odour can be related to the concentration of odour-active volatile compounds at the sniffing port (Van Ruth *et al.*, 1995*c*), although an increase in concentration could also change the descriptions of an odour.

PRELIMINARY RESULTS OF A STUDY ON A LIGHT BLACKCURRANT SOFT DRINK

Substitution of sucrose by intense sweeteners demands a complete copy of taste and functional properties of sucrose. As none of the currently known sucrose substitutes possesses all of these qualities, manufacturers of soft drinks use combinations of intense sweeteners to solve flavour problems encountered with single sweeteners (Bakal, 1991; Houghton, 1988; Verdi & Hood, 1993). The sweetness of a light blackcurrant soft drink was evaluated as being lower than the sweetness of the classic soft drink. With the help of a quaternary model of Beidler's mixture equation the concentrations of the sweeteners in the light drink can be adjusted in order to meet the sweetness of the sucrose-sweetened soft drink. Descriptive sensory tests will allow a comparison of the classic and light soft drinks.

De Graaf & Frijters (1986) showed that Beidler's mixture equation can predict the intensity of a mixture of two substances. Beidler's mixture model describes the peripheral interaction between two taste substances; therefore, it is limited to mixtures of taste substances of similar taste qualities. Moskowitz *et al.* (1978) showed that psychological rules of sensory perception, which predict the responses to pure aqueous systems of sweeteners hold for complex beverages. According to Beidler's mixture model, the magnitude of the response of a mixture containing particular concentrations W, X, Y and Z (mM) of, respectively, substances A, B, C and D is given by:

$$R_{abcdWXYZ} = \frac{K_a W R_{sa} + K_b X R_{sb} + K_c Y R_{sc} + K_d Z R_{sd}}{1 + K_a W + K_b X + K_c Y + K_d Z} \quad (1)$$

in which R_{sa}, R_{sb}, R_{sc} and R_{sd} = maximum responses to substances A, B, C and D, respectively; K_a, K_b, K_c and K_d = association constants of substances A, B, C and D, respectively. Assuming that the responses to each mixture of a series of mixtures of substances A, B, C and D, containing the concentrations W (of A), X (of B), Y (of C) and Z (of D) (mM) are equal to the response magnitude R, the following equation can be derived [in analogy with the derivation by De Graaf & Frijters (1986) for a mixture of two substances]:

$$W + \frac{C_{ai}}{C_{bj}} X + \frac{C_{ai}}{C_{ck}} Y + \frac{C_{ai}}{C_{dl}} Z = C_{ai} \quad (2)$$

where C_{ai} (mM) is the concentration of substance A that evokes the response of magnitude R when substances B, C and D are not present. This is the point of subjective equality (PSE) for substance A. Similarly C_{bj}, C_{ck} and C_{dl} (mM) can be determined for substances B, C and D, respectively.

The combination of intense sweeteners in the light blackcurrant soft drink should approximate the sweetness of a 10% sucrose soft drink as closely as possible. The light soft drink contains the sweeteners Na-saccharinate, Na-cyclamate, aspartame and acesulfam-K. Saccharin is one of the cheapest sweeteners, however, the taste of this sweetener limits its use to a maximum dose (Mitchell & Pearson, 1991). Therefore the maximum concentration of Na-saccharinate in the light soft drink was kept at 0.18 mM. Several combinations of the other three sweeteners can be chosen to improve the taste and sweetness of the light soft drink.

The difference in taste between the various blackcurrant soft drinks was judged using quantitative descriptive analysis. This analysis was performed by a sensory panel of 12 selected and trained assessors (aged 20–23). A computer interactive interviewing system for composing questionnaires was used to gather survey information (Ci2 system, Sawtooth Software Inc., Ketchum, USA). In consultation with the panel, a vocabulary of 12 attributes was composed to describe the flavour of an assortment of blackcurrant soft drinks (Table 1). The sucrose-sweetened, light and adjusted light blackcurrant soft drinks were evaluated by tasting and the intensities of the attributes were marked on a

Table 1. Attributes describing the aroma of blackcurrant soft drinks (Bakker, 1995)

Attributes	
Strawberry	Sharp
Blackcurrant	Watery
Bitter	Tart
Refreshing	Sour
Metallic	Aftertaste
Musty	Sweet

120 mm visual analogue scale on a portable computer screen.

The four axes of the quaternary model were respectively presented by Na-saccharinate (W), Na-cyclamate (X), aspartame (Y) and acesulfam-K (Z). PSEs were determined for the four sweeteners, compared to a 10% sucrose blackcurrant soft drink. The PSEs are derived from equisweet values reported by O'Brien Nabors & Gelardi (1991) and they are presented in Table 2. Ketelsen *et al.* (1993) reported difficulties in determining PSEs for Na-saccharinate and acesulfam-K, due to their inherent bitterness and metal taste at that concentration. The PSE of aspartame was determined according to Bock & Jones (1968), using the method of constant stimuli (De Graaf & Frijters, 1986). As this PSE of 2.0 mmol/l approximates (sufficiently) the value calculated from O'Brien Nabors & Gelardi (1991), it was assumed that the other PSEs could be used as well.

The PSEs are the intercepts in the four-dimensional coordinate system representing the quaternary model by a tetrahedon. Combinations of the intensive sweeteners, on the peripheral triangles of this tetrahedon, predict equal sweetness to the 10% sucrose blackcurrant soft drink. Beidler's mixture equation for this tetrahedon was

$$W + 0.096X + 0.842Y + 0.640Z = 1.6 \qquad (3)$$

in which W, X, Y and Z are the concentrations (mmol/l) of Na-saccharinate, Na-cyclamate, aspartame and acesulfam-K, respectively. This equation represents an additive model for concentrations of sweeteners needed to obtain a particular sweetness. However, synergistic effects occurred when combinations of intense sweeteners were tried out. The average score was 41 on a visual analogue scale instead of 28 found for the sucrose-sweetened drink. The tetrahedon in the four-dimensional coordinate system was uniformly reduced to reckon with this synergy. After a reduction of 38%, the tetrahedon (Fig. 1) approximately fits with the sweetness of a 10% sucrose blackcurrant soft drink. The outcome of Beidler's mixture equation was then

$$W + 0.096X + 0.842Y + 0.640Z = 1.0 \qquad (4)$$

Beidler's mixture equation was found useful for sweetness prediction of binary mixtures of glucose and fructose by De Graaf & Frijters (1986). The response to intense sweeteners might be different; however, the quaternary model can provide a basis for the prediction of the

Table 2. Points of subjective equality (PSE) calculated from O'Brien Nabors & Gelardi (1991) for the intense sweeteners Na-saccharinate, Na-cyclamate, aspartame and acesulfam-K, related to a 10% sucrose-sweetened blackcurrant soft drink

Intense sweeteners	PSE (mmol/l)
Na-saccharinate (W)	1.6
Na-cyclamate (X)	16.6
Aspartame (Y)	1.9
Acesulfam-K (Z)	2.5

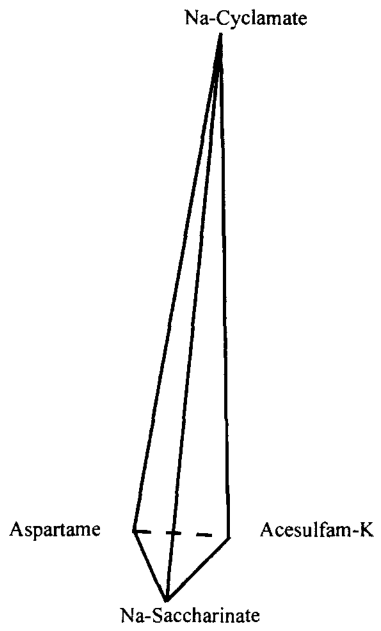

Fig. 1. Tetrahedon, representing the quaternary model of Beidler's mixture equation: $W + 0.096X + 0.842Y + 0.640Z = 1.0$.

sweetness of solutions. The sweetness of the light blackcurrant soft drink was predicted with the quaternary model of Beidler's mixture equation [equation (4)], which was 92% of the result for the sucrose-sweetened soft drink. A new combination of the intense sweeteners Na-saccharinate, Na-cyclamate, aspartame and acesulfam-K was used to meet the sweetness of the classic soft drink. The concentration of saccharin remained unchanged; the other sweeteners were increased about proportionally to their original ratios. In this way the limited shelf-life of aspartame (Homler *et al.*, 1991) and the bitterness of acesulfam-K, were taken into account.

Figure 2 illustrates the evaluation of the three soft drinks. Sensory data were subjected to Student's *t*-tests to determine significant differences between the three soft drinks. A significance level of $p < 0.05$ was used in this study. The light blackcurrant soft drink significantly differed from the classic soft drink for the attributes refreshing, sharp, tart, sour, aftertaste and sweet (Table 3). The flavour of the light soft drink clearly differs from the classic soft drink. When the new combination of intense sweeteners is applied, the composition of the flavour shifts from currant to strawberry, together with a decrease of the sour related attributes.

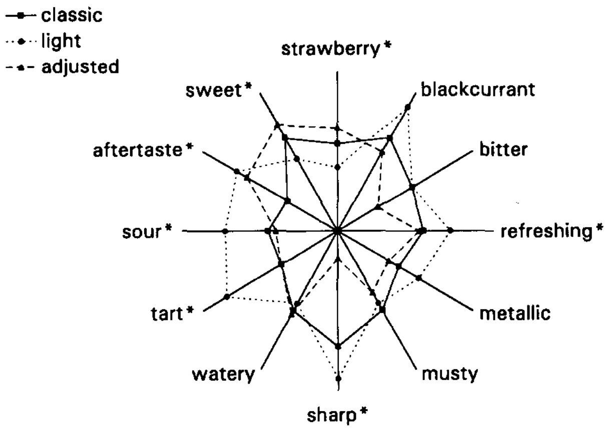

Fig. 2. Spider web diagram of scores for sensory attributes of a classic (sucrose-sweetened), light and adjusted light blackcurrant soft drink: * = significant differences ($p < 0.05$).

Table 3. Scores for sensory attributes of classic (sucrose-sweetened), light and adjusted light blackcurrant soft drinks (mean, $n = 12$)

	Mean score		
Attributes	Classic	Light	Adjusted light
Strawberry	22	16	26
Blackcurrant	27	36	23
Bitter	22	22	12
Refreshing	22	29[a]	21
Metallic	18	24	15
Musty	23	21	18
Sharp	29	37[a]	7[a]
Watery	23	21	24
Tart	17	33[a]	17
Sour	18	29[a]	16
Aftertaste	15	30[a]	27[a]
Sweet	27	21	31

[a]Significantly different from the classic soft drink ($p < 0.05$).

CONCLUSIONS

The study of sweetness flavour interactions in soft drinks demands a sound knowledge of the behaviour of combinations of intense sweeteners, because these combinations can solve the flavour problems encountered with single sweetener applications in beverages. A quaternary model of Beidler's mixture equation was used in a preliminary study to modulate the concentrations of intense sweeteners in a light blackcurrant soft drink, and to meet the sweetness of the classic soft drink. The model needs further elaboration of Beidler's mixture equation for its use in the future. For instance when the aroma of a light soft drink contributes additively to the sweetness of this drink, an extra term can be added to Beidler's mixture equation to denote the effect of aroma on sweetness.

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