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Does chemical modification of tastants merely enhance their intrinsic taste qualities?

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The hypothesis that chemical modification of a sweet compound to create a new, sweeter compound merely enhances the intrinsic taste properties of the original sweetener is discussed. The detailed sensory profiles of sucrose and 4,1′,6′-trideoxy-4,1′,6′-trichlorogalactosucrose (sucralose) are examined to assess the validity of this hypothesis. These all show a remarkable and consistent conformity, which suggests that the sensory properties of sucralose are a direct consequence of those of its parent sweetener, sucrose. Structure–activity relationships of derivatives of sucrose further support this hypothesis, because the structural features of sucrose that are believed to be responsible for its ability to elicit a sweet taste are identical to similar structural features of sucralose. Copyright © 1996 Elsevier Science Ltd

INTRODUCTION

The initial event in sweet taste transduction is generally agreed to involve stereospecific binding of sweet compounds by a transmembrane protein receptor which subsequently undergoes a conformational transition. This transition then initiates a cascade of events ultimately leading to neurotransmitter release and nerve firing. The loose binding of sweet compounds by receptor proteins must involve forces such as hydrogen bonds, probably simultaneous hydrogen bonds formed between ligand and receptor, and receptor and ligand, as originally visualized by Shallenberger and Acree (1967) and expressed as a stereochemical AH,B arrangement, where A and B are electronegative atoms and H is a proton (Fig. 1). Thus, ligand structure will govern the ability of any compound to induce sweetness, and also its intensity.

Many examples serve to illustrate this observation:

1. D-Glucose is sweet, but D-mannose is bitter-sweet. Here, a single configurational change induces a bitter taste.
2. D-Glucose is approximately twice as sweet as D-galactose. Here, a single change of configuration influences perceived intensity, but has no apparent impact on taste quality.
3. D-Fructose in the pyranose conformation is believed to be substantially sweeter than sucrose, but in the furanose form is thought to be barely sweet. Here, conformation influences sweetness intensity, but not sweet taste quality.
4. D-Phenylalanine is sweet, but its L-enantiomer is not. Here, enantiomeric form controls the presence or absence of sweetness.
5. D-Phenylalanine is substantially sweeter than D-alanine. Here, relative intensity is governed by the degree of lipophilicity in each molecule.

This last example is an illustration of the importance of the Kier (1972) binding site (X) and its influence on sweetness intensity. This site, whose position relative to AH and B has been defined (Fig. 2), is clearly one controlling force influencing the sweetness intensity of compounds from within any given structural class. Thus, the sensory characteristics of compounds are influenced by the nature of their functional groups, the configuration of those groups, the conformation of the molecule, its degree of lipophilicity and the location of the lipophilicity relative to the functional groups responsible for sweetness.

There appears to be an optimum balance between lipophilicity and hydrophilicity in sweet molecules. With a lipophilic substituent of appropriate size, correctly positioned, that lipophilic substituent appears capable of enhancing the complete sensory characteristics of the unsubstituted molecule, rather than eliciting or inducing ‘new’ sensory effects. If so, it is only when the delicate balance of the relative ‘strengths’ of the saporous unit and lipophilicity are disrupted that the basic sensory profile is also disrupted and the qualitative changes induced in those basic sensory characters begin to

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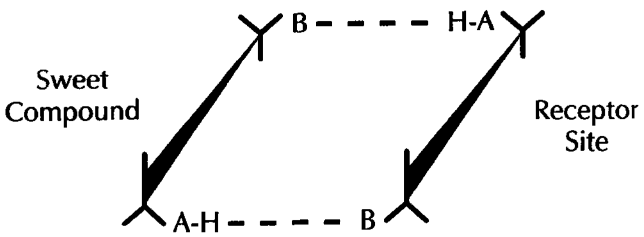

Fig. 1. The AH,B glucophore of Shallenberger & Acree (1967).

predominate. In other words, the only necessary requirement for sweet taste quality in a molecule is its fundamental (and functioning) AH,B system. The X site merely enhances this effect.

Intrinsically sweet compounds undergoing chemical modification which introduces tastes (e.g. bitterness) in addition to sweetness must, by definition, have upset either the balance of lipophilicity and hydrophilicity of the original sweet compound, or the substitution must be positioned outside the AH,B–X distance parameters defined by Shallenberger and Acree (1967) and Kier (1972). Therefore, the corollary to this is that chemical modification of an intrinsically sweet compound which produces a new, sweeter compound must merely be enhancing the basic sensory properties of the original compound. While this hypothesis is, of course, difficult to prove unequivocally, there is a consistency to the chemical and sensory literature which indicates that the premise has validity.

The most comprehensive sensory database to test this hypothesis can be found from examination of the comparative sensory properties of sucrose and 4,1′,6′-trideoxy-4,1′,6′-trichlorogalactosucrose (sucralose). Sucralose is the only commercial low-calorie sweetener that is prepared directly from a starting material that is also sweet and for which detailed sensory data are available. Additional insight into the validity of this hypothesis can be obtained by examination of the relevant literature on the relationships between molecular structure and sweet taste, some of which have already been mentioned.

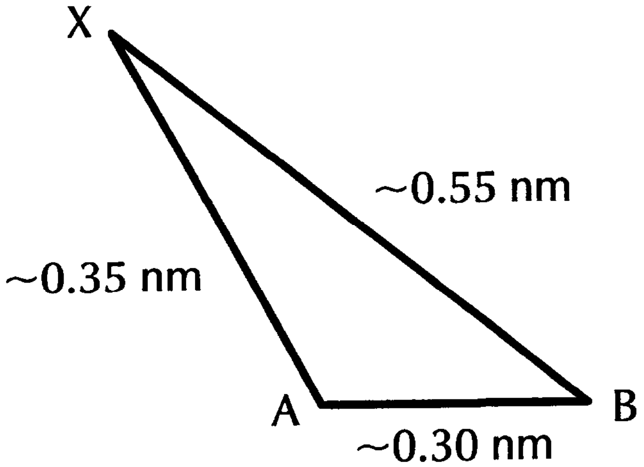

Fig. 2. The AH,B–X tripartite structural requirement for sweetness (Kier, 1972).

COMPARISON OF THE SENSORY PROPERTIES OF SUCROSE AND SUCRALOSE

Sucrose and sucralose display remarkably similar taste properties. Wiet and Beyts (1992) evaluated the taste properties of these sweeteners, as well as aspartame, acesulfame-K and saccharin, in water across a range of sweetness equivalencies. Magnitude estimation and non-ratio scaling procedures were used to quantify sweetness intensity, bitterness, sourness, body, sweet aftertaste and non-sweet aftertaste. Changes in the secondary taste attributes of these sweeteners as a function of sweetness intensity are presented in Fig. 3. Most notably, sucralose displayed extremely low levels of bitterness as a function of sweetness intensity, its curve paralleling very closely that of sucrose, and perceived body was also remarkably similar to that of sucrose.

Obviously, the taste profile of any sweetener is concentration-dependent, and sucralose is no exception. However, it is evident that the non-sweet taste characteristics of both sucralose and sucrose become more readily apparent as their concentrations increase, a finding in accord with that of Samundsen (1985) who reported that approximately 60% of consumers perceive a sweet, drying and slightly bitter aftertaste with sucrose.

Very similar observations have been recorded in a previously unpublished study which was designed to provide baseline flavor data on the taste of sucrose and sucralose at four concentrations using the A.D. Little 'Flavor Profile Analysis' technique. In this study, equi-sweet concentrations of the sweeteners were prepared in citrate buffer (pH 3.5) to mimic more closely the environment of soft drinks. Sucrose at 8% was perceived as primarily sweet, with faint caramelized sugar notes and some sour, drying characteristics. The equi-sweet sucralose solution (180 ppm) was also perceived as primarily sweet with slight drying and sour characteristics (Table 1).

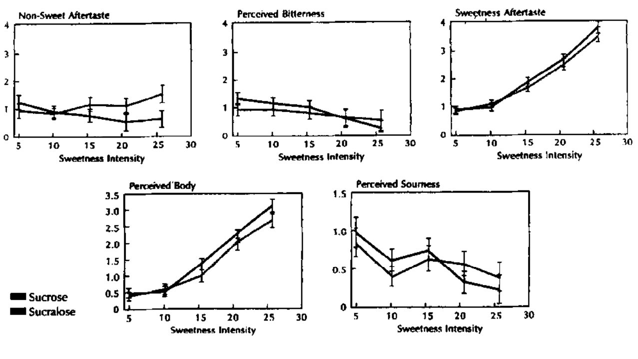

Fig. 3. Taste profiles of sucralose and sucrose.

At higher concentrations, a slightly different array of secondary taste characteristics became apparent (Table 2). Sucrose (12%) still exhibited some drying and caramelized notes and a sensation described as 'throat-catch'. Its aftertaste was largely sweet, with slight drying, astringent, sour and throat-catch notes. Sucralose (350 ppm) was perceived to deliver some drying and sour attributes, with a very slight rubbery taste. The aftertaste profile remained largely sweet with slight metallic/bitter and drying tastes emerging.

Both these studies indicate a close similarity between the taste profiles of sucrose and sucralose. Of the sweeteners included in these studies, sucralose was notable in lacking a 'synthetic sweetener sensation', and its taste profile was consistently judged as sucrose-like across the concentration range studied.

In addition to inducing a variety of sensory profile characteristics, sweeteners may also exhibit marked diversity in their time course of perceived sweetness. The sweet sensation may be elicited rapidly and dissipate quickly, or it may rise more slowly and have an extended, lingering sweet aftertaste. Comparisons of these temporal properties of sucrose and sucralose, measured using a computer-assisted time–intensity method, have been reported by Ketelsen *et al.* (1993).

In this study, two concentrations of sucrose and sucralose were compared. At both sweetness levels in water (5% and 9% sucrose, and 88 ppm and 252 ppm sucralose, respectively), the time–intensity curves of these sweeteners were similar, as measured across all time–intensity parameters (Figs 4 and 5). At 9% sucrose equivalence in citrate buffer, sucrose and sucralose again exhibited very similar time–intensity properties (Fig. 6).

This consistent pattern of conformity in the taste characteristics of these sweeteners has also been demonstrated in studies of persistence time (Shamil & Birch, 1992). Here, four concentrations of sucrose and sucralose in water were evaluated using a computer-assisted recording device. As concentration increases, an increase in persistence was recorded, as expected. In comparing concentration, sweetness intensity and persistence (Fig. 7), a strong resemblance was reported between the response surfaces of sucrose and sucralose. These authors conclude that sucralose approaches the 'ideal' sweet characteristics of sucrose, also noting that a majority of panelists described the taste of sucralose as 'clean' with no bitter aftertaste.

Using the method of survival analysis, in which a panel of 100 tasters was employed, Ketelsen *et al.* (1992) measured the residual sweetness at intervals after swallowing equi-sweet solutions of sucrose or sucralose. Forty seconds after swallowing a sucrose solution, the entire panel no longer perceived sweetness, whereas with

Table 1. Flavor profiles[a] of sucrose and sucralose determined by the A. D. Little Flavor Profile Analysis method

Characteristic	Sucrose (8.05%, w/v)	Sucralose (0.018%, w/v)
Sweet	200	200
Sour	50	100
Drying	100	100
Caramelized	25	0
Synthetic sweetener sensation	0	0
Bitter	0	0
Metallic	0	0

[a]0, not present; 25, threshold; 100, slight; 200, moderate; 300, strong.

Table 2. Flavor profiles[a] of sucrose and sucralose determined by the A. D. Little Flavor Profile Analysis method

Characteristic	Sucrose (12.0%, w/v)	Sucralose (0.035%, w/v)
Sweet	300	250
Sour	0	100
Drying	100	100
Caramelized	50	0
Synthetic sweetener sensation	0	0
Bitter	0	0
Metallic	0	0
Bitter aftertaste	0	25

[a]0, not present; 25, threshold; 100, slight; 200, moderate; 300, strong.

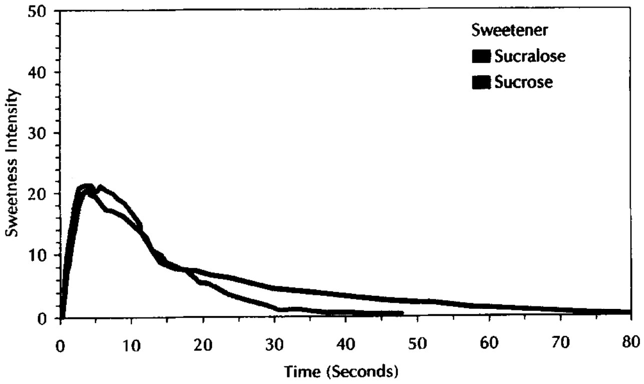

Fig. 4. Mean time–intensity curves for sweetness 5% sucrose equivalency in water.

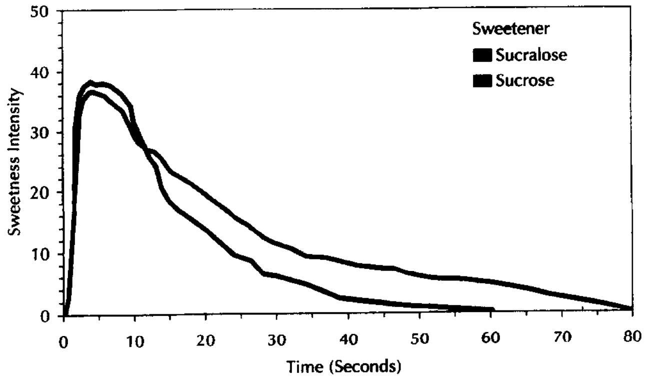

Fig. 5. Mean time–intensity curves for sweetness 9% sucrose equivalency in water.

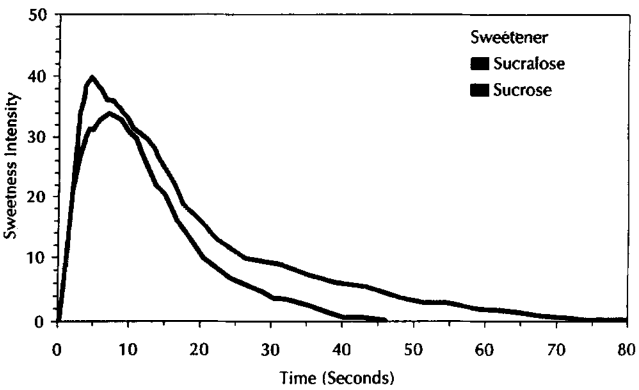

Fig. 6. Mean time–intensity curves for sweetness 9% sucrose equivalency in buffer.

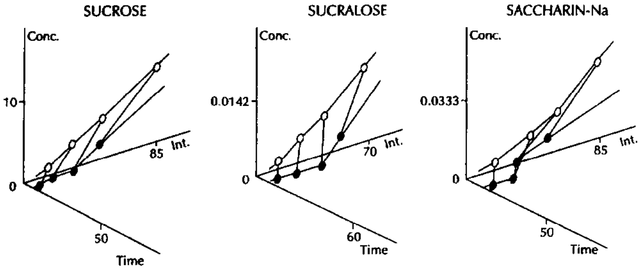

Fig. 7. Effects of concentration on sweetness intensity and persistence. From Shamil and Birch (1992).

sucralose at this point 20% of panellists still perceived a sweet aftertaste, a directional, but non-significant difference. In contrast, 40 s after swallowing, aspartame was still perceived as sweet by 35% of the panel, and this proved to be a statistically significant difference.

Collectively, these data all indicate that the temporal properties of sucrose and sucralose show remarkably close similarities across a wide sweetness equivalence range.

The sweetness synergy characteristics of sweeteners are important from a commercial standpoint, so are invariably examined. Although the mechanism(s) whereby sweetness synergy may be induced are not fully understood, one persuasive explanation is that synergy will be observed between blends of sweeteners which elicit different flavor profiles, and particularly if one sweetener in the blend exhibits a bitter aftertaste. Thus, saccharin and cyclamate blends are synergistic, as are blends of aspartame and acesulfame-K. Sucralose itself is synergistic with saccharin, acesulfame-K and cyclamate, but further synergy studies with sucralose have found none exhibited between that sweetener and sucrose (Beyts & Latymer, 1985). This provides further confirmation of the close similarity in overall sensory profiles of sucrose and sucralose and the lack of significant bitterness induced by sucralose.

All sweeteners exhibit curvilinear dose–response relationships. Carbohydrate sweeteners, including sucrose, exhibit linear dose–response relationships at what may be considered low to moderate concentrations and it is only as the concentrations reach high levels that this relationship departs from linearity. Thus, Moskowitz (1971) reports that sucrose departs from a linear dose–response function at concentrations above 35% (w/v). In contrast, although dose–response relationships for low calorie sweeteners are also curvilinear, they all tend to exhibit a linear relationship only up to much lower sweetness equivalencies than do sucrose and the other carbohydrates.

That being said, as part of a study examining the dose–response functions of a number of sweeteners, Wiet & Beyts (1992) used magnitude estimation to explore this relationship for sucrose (1.35–9.0%, w/v) and sucralose (20–150 ppm). Their findings are shown in Fig. 8. Across this sweetness range, the power functions for sucrose and sucralose were linear, with both sweeteners having exponents close to 1.0. These findings indicate that, for sucrose and sucralose, perceived sweetness intensity growth is commensurate with an increase in physical concentration across these low to moderate sweetness concentrations. In a similar study, DuBois *et al.* (1991) systematically determined the dose–response relationships for a wide range of sweeteners. Rather than employing a magnitude estimation technique, their chosen methodology was to match test solutions against sucrose standards. By definition, therefore, the dose–response relationship for sucrose was determined to be linear, whereas that for sucralose departed from linearity at a concentration of approximately 200 ppm.

In summary, therefore, the measured sensory characteristics of sucrose and sucralose are consistently reported to be very similar. Taste profiles of sucrose and sucralose, measured in water and citrate buffer, exhibit great similarity. The temporal characteristics of both sweeteners, evaluated by a variety of techniques, and measured in water and citrate buffer, are virtually identical. Sucrose and sucralose blends do not exhibit sweetness synergy, further indicating their taste profile correspondence, and their dose–response relationships at most practical sweetness intensities are also very similar. Therefore, although basically impossible to

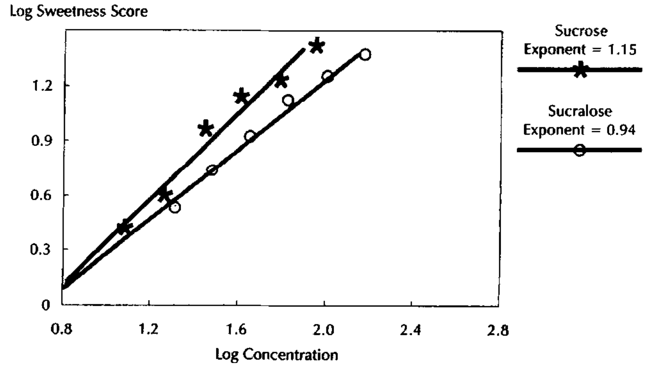

Fig. 8. Psychophysical sweetness functions.

prove, it seems a reasonable conclusion that the sensory characteristics of sucralose stem directly from its sucrose origins.

CONSUMER RESPONSES TO FOOD PRODUCTS SWEETENED WITH SUCROSE AND SUCRALOSE

The sensory comparisons of sucrose and sucralose discussed previously were all measured in simple, unflavored aqueous systems. As such, any sensory differences between sucrose and sucralose are likely to be fully exposed. Comparisons in fully formulated, flavored foods and beverages might therefore be expected to exhibit fewer significant differences in sensory properties between these two sweeteners.

In this study, consumer responses to a broad array of sucrose- and sucralose-sweetened products were determined. Products included coffee, hot tea, iced tea, cold cereal, cookies, coffee cake, apple pie and chocolate cake. Tests were conducted with a panel of 833 consumers, both males and females, aged between 18 and 65 years. Following tasting, consumers were presented with a series of competitive claims (e.g. 'tastes like sugar'), monadic claims (e.g. 'has a clean taste') and an aftertaste question ('has no unpleasant aftertaste'). Finally, an overall preference question was asked. The results of this study are summarized in Table 3.

No significant differences in overall ratings of sucralose and sucrose products were found for the following claims: 'tastes like sugar', 'tastes sweet like sugar', 'has a clean taste like sugar', and 'has a clean taste'. Aftertaste was not identified as a disliked attribute, with only 6.7% indicating an unpleasant aftertaste compared with sucrose at 4.4%. Finally, in response to the 'preference' question, sucralose and sucrose scored equally, each being preferred by 45.7% of the respondents (weighted totals across products: 8% 'no preference').

It was concluded from this study that the overwhelming majority of consumers perceive the taste of sucralose- and sucrose-sweetened products similarly. Additionally, products made with sucralose are equally preferred to similar products prepared with sucrose.

Table 3. Consumer reaction to foods and beverages sweetened with sucrose or sucralose

Claim	Agreement[a] (%)	
	Sucralose	Sucrose
Tastes like sugar	66.2	73.1
Tastes sweet like sugar	76.3	75.4
Has a clean taste like sugar	77.0	81.9
Has a clean taste	85.1	89.2
Has a pure taste	77.4	85.0
Has no unpleasant aftertaste	93.3	95.6

[a] Weighted totals across all products.

STRUCTURE–TASTE RELATIONSHIP CONSIDERATIONS

There are many examples where sweet taste intensity has been shown to be enhanced through an appropriate increase in lipophilicity within an already sweet molecule. As has been described, D-phenylalanine is substantially sweeter than D-alanine. To exemplify this point further, Verkade *et al.* (1946) described the relative sweetness intensities of 4-nitro-2-aminophenyl alkyl ethers. Changing the molecular size of alkyl substituent from methyl to ethyl and then to propyl yields ether compounds that are progressively more sweet. Increasing the size and lipophilicity of the alkyl substituent further to butyl then leads to a reduction in sweetness, the propyl substituent having generated optimum sweetness intensity. Similarly, the influence on sweetness intensity of halogen substitution in nitroamines has been reported by Blanksma *et al.* (1946). Here, as halogen size and lipophilicity increases from fluorine to chlorine, bromine and iodine, so sweetness intensity relative to sucrose also increases. The unsubstituted *m*-nitroaniline is also recorded (Moncrieff, 1951) as being sweet.

Other examples illustrate the impact of halogen substitution on the sensory properties of sweet compounds. Chlorination at the C6 position of D-tryptophan increases relative sweetness intensity from 25–40 times that of sucrose for D-tryptophan to around 1000 times for 6-chloro-D-tryptophan. Chlorination of saccharin to yield *p*-chlorosaccharin leads to an enhancement of saccharin's intrinsic bitter character and, due to mixture suppression, a reduction in relative sweetness intensity. As a result of these observations, the enhancement of sucrose sweetness and other sensory properties by selective chlorination has ample precedent.

Sweetness and bitterness are closely linked sensations. For example, many compounds that elicit sweetness also elicit bitterness, and compounds known to function effectively as inhibitors of sweet taste have recently been shown also to be able to inhibit bitterness (Kurtz & Fuller, 1993). Thus, it is entirely logical that any substituent enhancing sweetness will also enhance bitterness. Compounds such as sucrose and sucralose, whose concentrations in solution are adjusted so that both solutions elicit equal sweetness, would therefore also be expected to deliver equal bitterness as a side-taste. Conformity between the sweetness and bitterness of sucrose and sucralose has, in fact, been observed and has already been described (see Fig. 3); it suggests a strong physiological link between sucrose and sucralose and lends further support to the hypothesis that chlorine substitution is merely enhancing the intrinsic sensory qualities of sucrose.

Although a very useful concept in helping to rationalize the varying and often anomalous sensory characteristics of structurally analogous compounds, the AH,B–X glucophore concept neglects three-dimensional shape and volume considerations. However, advanced computer modeling techniques and calculations of molecular electrostatic and hydrophobicity potentials are now being used to overcome this limitation, and the results obtained are able to rationalize the dramatic range of sweetness intensities elicited by sucrose and sucralose (Lichtenthaler & Immel, 1993). A general conclusion of their work is that the lipophilic (X) part of the AH,B–X tripartite arrangement appears to be an entire flexible region rather than a specific point on the sweetness triangle. In sucrose and sucralose, this region is believed to encompass the outside area of the fructofuranose moiety. In contrast, the hydrophilic portions of these molecules are more compact, are located opposite the hydrophobic region, and appear to contain the AH,B couple (Lichtenthaler *et al.*, 1991). Thus, the structural features responsible for both the presence of sweetness in sucrose and sucralose, and their relative intensities, appear on the available evidence to be identical. Locating X on the fructose moiety of sucrose and sucralose, either as a point or, more reasonably, a region, makes location of AH and B at the glucosyl 2-OH and 3-OH groups of both sucrose and sucralose inevitable. These assignments are also in accord with other structure–taste relationships of sucrose and its derivatives, as has been summarized by Lee (1987). Therefore, it is an entirely logical conclusion that chlorination of sucrose to form sucralose is enhancing the effectiveness of the former's hydrophobic, X, region, thus having the consequence of merely enhancing sucrose's intrinsic taste qualities.

Similar studies seeking to assign AH,B–X aspects on sucrose and sucralose have been carried out employing advanced computer graphics techniques (Suami & Hough, 1992; Suami *et al.*, 1994). These studies confirm the importance of sites on the fructose moiety functioning as X, and conclude that two glucophoric bifunctional entities of the AH,B type are revealed for sucrose, one of which, the 3′-OH/2-O couple, is identified as the most probable AH,B site for sucralose.

DISCUSSION

Considerable progress has been made in developing logical and internally consistent relationships between molecular structure and sweet taste since the initial development of the AH,B concept and its extension to incorporate a hydrophobic (X) structural feature. These insights have helped to rationalize the varying and seemingly anomalous sensory characteristics of many structurally diverse compounds. The vital requirement of a structural AH,B couple in controlling the presence of sweetness itself is now universally acknowledged. The role of a suitably positioned hydrophobic region (X) in influencing resulting sweetness intensity is, on the basis of much evidence, incontrovertable. However, it is been unclear whether introduction of such a suitably positioned hydrophobic structure in an already sweet compound generates a wholly different range of sensory characteristics, or whether it merely enhances the intrinsic sensory properties of the original, unmodified compound. One difficulty in providing a satisfactory answer to this question has been the lack of suitably modified compounds. Now, with the availability of results from a broad range of sensory studies on sucrose and its structurally modified low-calorie version, sucralose, it has been possible to compare the sensory properties of these two sweeteners in some detail.

It is clear from an examination of these data that sucralose and sucrose elicit very similar sensory effects, with a broad range of sensory techniques having been utilized to compare the sweeteners. Magnitude estimation and non-ratio scaling procedures showed a remarkable conformity in sensory profiles, as did detailed flavor profile analyses. Sucrose and sucralose do not exhibit sweetness intensity synergy when blended, a finding consistent with the view that their taste profiles are very similar. Dose–response relationships, at practical use levels, show that perceived sweetness intensities grow as concentration increases according to very similar exponents. Time–intensity measurements also demonstrate great consistencies between the two sweeteners. All these findings are supported by extensive consumer studies using many sucrose- and sucralose-sweetened foods and beverages.

Thus, there is a pattern in the sensory evaluation of sucrose and sucralose wholly consistent with the proposal that the sensory profile of sucralose stems directly from that of its parent molecule, sucrose. Further evidence that this is the most likely explanation of the sensory qualities of sucralose may be derived from an examination of recent advanced computer-modeling studies. In these, assignment of probable AH,B and X sites on both sucrose and sucralose are consistent, and the assignments correlate with the reported structure–activity relationships of many other sucrose derivatives.

Consequently, we conclude that the taste qualities of sucralose probably stem directly from those of sucrose. We also consider it unlikely that the sensory relationships between sucrose and sucralose are unique to this 'pair' of sweeteners; we fully anticipate that similarly detailed sensory evaluation of other analogous pairs of sweeteners would yield essentially identical findings. For example, we expect that the taste profiles of D-alanine and D-phenylalanine would also exhibit great qualitative similarity.

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