人間に対する糖類の甘味度の物理化学的検定に関する一提案(農芸化学科)

A Proposal for the Physicochemical Determination of the Sweetness of Sugars to Man

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One of significant problems in the field of sensory physiology would be the establishment of a means for the quantitative expression of the responses induced by a given intensity of a given mode of stimulus (4). In human gustation the number of basic qualities of sensations is limited to four; sweetness, bitterness, sourness, and saltiness. Although the recognition of an individual quality is relatively easy to be carried out, the quantitative evaluation of a given mode of taste, in general, requires much practictices. For example, an examiner memorizes the strength of the sweet sensatition of the standard solutions in the case of the evaluation of the sensation of sugars. Then, it would taste a given sample and would identify an equivalent concentration among the standard solutions. Since adaptation in taste sensations occurs rapidly, the examiner would be required to taste the standard solutions and the sample solutions alternatively to acquire a reliable evaluation of the strength of the sweetness. The repetitive evaluation of the intensity of the taste of a sample by this method may not necessarily bring out an accuracy in the sweetness scoring: A larger error could result from the variations in not only the skilfulness of the examiner but also its intrinsic sensitivity to the particular mode of the gustatory sensations; i.e. ' taste blindness '.

In order to remove such a difficulty in the evaluation of taste intensity, an instrumental determination of the strength of taste is argently desired. Nevertheless, in man the test mentioned above is the major method of the quantitative evaluation of tastes, available in an ordinary situation. In other animals, on the contrary, the instrumental determination of ' taste sensation ' is the major method accompanied with behavioral responses. In general, peripheral nervous responses and behavioral responses in other animals are considered as the sensations equivalent to the taste sensation of man: The taste may be evaluated in various stages of the physiological and behavioral responses of animals, including man.

The intensity and mode of taste expressed at various levels of the physiological or behavioral responses may carry significant information as to the elucidation of these biological responses related to taste sensation. Unfortunaly, even with a great effort utilizing these levels of taste responses, the structure-response relationship of the taste compounds has been left in confusion quite often, even in determining the sequences in the order of

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effectiveness among the compounds with a similar mode of taste.

I now report an effective method of an instrumental evaluation of the sweetness of sugars. The technique is extremely simple and may also be extended toward other modes of taste fruitfully.

The instrumental determination of the sweetness of chemical compounds could be carried out by the evaluation of their physicochemical property only if such a property of the compounds were known to be correlated to their sweetness.

The important charcteristics of sweetness inducing compounds may be summarized in the following three points (5). The first point is that a hydrogen donor group and a hydrogen acceptor group must exist in a pair within a single molecule. The second point is that the hydrogen donor and the hydrogen acceptor must be located in a distance of 2.5−4.0 A. And the third point is that the hydrogen bond forming groups should exist in *cis*−conformation.

From the above information and the fact that the most of sweet compounds can form molecular complexes, it is very reasonable to assume that a proper selection of complex forming agents may be used for an evaluation of the sweetness of the compounds if we assume that the strength of the sweet sensation of a sugar is dependent on that of the bonds responsible for the complex formation.

Although sugars cannot be isolated electrophoretically, at the presence of boric acid they can be separated readily, since molecular complexes can be formed between sugar molecules and boric acid. Electrophoretic mobilities of the complex are dependent on pH of the solutions used: The mobilities are greater at the higher values of pH (1). Therefore, the relationship between the electrophoretic mobilities of sugars and their sweetness was examined using the literature values of the electrophoretic mobility (1) at pH 7 and the sweetness determined by several workers (3,5) in human responses. The selection of pH 7 was made due to the fact that at higher values in pH sugar molecules may undergo cunformational modifications and the pH of the saliva of man is known to be around 7. As a result, it became evident that the higher electrophoretic mobility of the complexes gave the greater sweetness of the sugars. The relationship can be expressed as the following equation:

$$\log S = a \log M + b, \qquad 1)$$

where S is the sweetness of the sugars relative to sucrose, M their electrophoretic mobilities expressed in $cm/V \cdot sec \times 10^5$, a and b constants, 1.25 and 0.76, respecively (with $n = 13$, $r = 0.775$, and $t_r = 4.07 > t_{.01} = 3.11$). This can be related to the concentration (C) of the sugars since it is established that the sweetness of sugars is related to the concentration of the sugars as follows (5):

$$S = d \cdot [C]^{0.5}, \qquad 2)$$

where S is the absolute sweetness of the sugars, d a constant, and C the concentration of the sugars tested.

Our problem here is to express S with measurable parameters only. First, let us formulate 2) in terms of the threshold concentration of sugars. If we put the sweetness and concentration of a sugar as S° and C°, respectively, we have :

$$S^\circ = d\ [C^\circ]^{0.5}. \qquad 3)$$

By rearrangement we get :

$$d = S^\circ / [C^\circ]^{0.5}\ . \qquad 4)$$

By a definition :

$$S^\circ = C^\circ / C^\circ_{suc}, \qquad 5)$$

where C°_{suc} is the threshold concentration of sucrose. The substitution of 5) into 4) leads to the following equation :

$$d = [C^\circ]^{0.5} / [C^\circ_{suc}]\ . \qquad 6)$$

Since

$$S = S^\circ, \qquad 7)$$

from 1) and 5) we obtain :

$$C^\circ / C^\circ_{suc} = b\ M^a, \qquad 8)$$

or by rearrangement we get :

$$C^\circ = b\ C^\circ_{suc}\ M^a\ .. \qquad 9)$$

The substitution of 9) into 6) leads to :

$$d = (b\ M^a / C^\circ_{suc})^{0.5}\ . \qquad 10)$$

Now, let us substitute 10) into 2) . Then, we get :

$$S = (b\ C\ M^a / C^\circ_{suc})^{0.5}. \qquad 11)$$

Since $C^\circ_{suc} \sim 0.01$ M, we finally obtain :

$$S = 10\ (b\ M^a\ C)^{0.5}\ . \qquad 12)$$

Since a and b are the constants given in this article and M can be evaluated by the electrophoretic experiment, it becomes now possible to determine the sweetness of the sugars as a function of C, expressed in Moles per Liter.

For the convenience of using the concept of sweetness, expressed in 12) it is advisable to adopt the unit of sweetness. If we assume the sweetness of sucrose at a concentration of 1 M as the unit of sweetness, then, from 2) we have :

$$S^{1M}_{suc} = d^{1M}_{suc} = d_{suc}\ . \qquad 13)$$

If we define 13) as 1 *hedes*, a Greek meaning sweetness, we obtain the sweetness of sugars in terms of *hedes* as follows :

$$S / S^{1M}_{suc} = 10\ (b\ M^a\ C)^{0.5} / d_{suc}. \qquad 14)$$

Using the constants evaluated in this article, 14) becomes :

$$S / S^{1M}_{suc} = 8.7\ (M^{1.25}\ C)^{0.5} / d_{suc}. \qquad 15)$$

Now, at the first time, it became possible to express the sweetness of sugars by knowing only the concentration of the compounds in addition to their electrophoretic mobilities at the presence of boric acid at pH 7. By a use of such a technique one can evaluate a "universal sweetness" of a sugar without being influenced by variations in the sensitivity of tasting individuals.

A direct consequence of the acceptance of such a relationship in the quantitative expression of the sweetness of sugars would be a possibility in relating the mode of the molecular interaction involved in the formation of complexes between sugar and boric acid to the molecular mechanism of the induction of the sweetness by the sugars. First, the strength of the complex forming bonds is directly related to the strenght of the sweetness of the sugars. It is not certain at this time whether the mobility of the sugars in complexed forms under the effect of an electric field near the receptor membranes may have a certain significance in the induction of sweet sensations or not. Nevertheless, the ability of forming a complex may be a good means for the evaluation of the sweetness inducing forces. Secondly, the complex formation between the sugars and other complex forming agents should decrease the sweetness of the sugars as long as the complex formation is maintained : An effective concentration of the sugar molecules that can induce the sweetness is decreased due to the complex formation. Such a means may be used as the gustatory repellent against pests, since the sugars are known to have a role of an arrestant factor for insects (2). In applying 14) to animals other than man, it is quite possible that the constants a and b can be different from the values reported in this work.

Furthermore, an application of the present technique to other modes of taste would be fruitful in carrying out an instrumental evaluation of the intensity of the taste and in achieving the deeper understanding of the mechanisms of the early processes of gustation in chemical terms.

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人間に対する糖類の甘味度の物理化学的検定に関する一提案

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要　　約

人間に対する糖類の甘味度は，糖類とホウ酸の分子間化合物が，pH 7において示す電気泳動度の対数値に正の相関関係にあることを認めた。この関係と，糖類の甘味度と糖溶液の濃度との関係式から，糖濃度のみを未知な項とし，糖類の電気泳動速度を決定することにより，糖類の甘味度を推定できる式を導いた。また，現在甘味度を示す単位が存在せず不便なため，ショ糖1モルの甘味に相当する甘味を1 *hedes*（ギリシャ語で甘味を意味する）と定義することにより，甘味の定量化を可能にする式を導いた。この式の応用により，新甘味剤の開発研究，甘味発現機構の研究および，糖類は昆虫に対し固着剤として作用することから，甘味発現阻害機構による接触忌避剤の開発研究が可能であることを示した。

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