

Physicochemical properties and vibrational spectra of small carbohydrates in aqueous solution and the role of water in their sweet taste

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Solution properties (intrinsic viscosity, apparent specific volume, hydration number and surface tension) were determined for three nutritive sugars (D-glucose, D-fructose and sucrose) and three polyols (xylitol, sorbitol and lactitol) in water and in 1% (w/w) aqueous salt (NaCl, KCl or MgCl₂) solution. Likewise, the laser-Raman spectra of these carbohydrates in water (10% (w/w)) or in salt solutions were recorded in the OH stretching region $(3800-2800 \text{ cm}^{-1})$ and analysed by comparison to the Raman band of water fitted to 4 Gaussian components. Microwave spectra of saturated sugar solutions were also obtained and interpreted in terms of polarity of the sugars. Comparison of the physicochemical results and Raman data in water, on the one hand, and in salt solution on the other, permitted determination of the influence of the biologically important cations (Na⁺, K⁺ and Mg²⁺) on the effect of sugars and polyols on water structure. This is also useful to support our three-step model of sweet taste chemoreception which relies on the fact that water mobility and the polarity of stimulant are the clue to the elucidation of the sweetness mechanism. Copyright 0 1996 Elsevier Science Ltd

INTRODUCTION

A combination of spectroscopic and macroscopic approaches was found (Mathlouthi *et al.,* 1993) useful to characterize the role of water structure in sweet taste chemoreception for pure sugars and sweeteners in water. Although no direct mention is made of water molecules and their association to the stimulus or to the receptor, many models of the sweet interaction take into account the hydrophilic and hydrophobic characters of the sweeteners (Daniel, 1989). The involvement of water molecules in the chains of hydrogen bonds that are important in molecular recognition of the taste of sugars was evoked earlier (Jeffrey, 1993). The hydration of electrolytes such as Na^{+} , K^{+} and Mg^{2+} is well documented (Robinson & Stokes, 1959; Seuvre, 1989) and their importance in a biological medium is well known.

The ions $Na⁺$ and $K⁺$ are involved in membrane potential change and very likely their distribution and the rate of their transfer across the receptor membrane are essential to the chemoreception of sweet taste. Mg^{2+} is one of the most abundant intracellular cations and its importance in biological mechanisms, especially enzyme activation and nucleic acid conformation (Flatman,

1988; Theophanides & Pelisson, 1986; Theophanides et *al.,* 1990), is well known.

We have applied, in this work, the double approach at microscopic (spectroscopy) and macroscopic (physicochemical) levels to the aqueous solutions of 3 sugars **(D**glucose, p-fructose and sucrose) and 3 polyols (xylitol, sorbitol and lactitol). The results are interpreted in terms of solute-solvent interactions and used to support the role of water structure in sweet-taste chemoreception according to our three-step model of the sweetness mechanism (Mathlouthi et *al.,* 1993).

MATERIALS AND METHODS

D-glucose and sucrose were Sigma products used without any further purification. **D-fructose**, xylitol and lactitol were a gift of Xyrofin France (Paris) and sorbitol a gift of Roquette Frères (Lestrem, France), all with a purity $> 99\%$. b-fructose was recrystallized in a water-ethanol mixture.

Macroscopic methods

Intrinsic viscosity was determined using the triple extrapolation procedure described elsewhere (Mathlouthi *et*

al., 1993). The equation of Huggins (1942) was used to calculate the interaction parameter (Huggins constant k'). B coefficients were derived from the Jones & Dole (1929) equation. The apparent specific volumes (V_2^0) were obtained from density measurements at $25^{\circ}C$ ($\pm 0.1^{\circ}C$) with a PAAR DMA45 densimeter. Surface tension was measured with a Prolabo (Paris) semi-automatic tensiometer (Tensimat-Densimat TD 2000). HPLC grade MilliQ doubly-distilled water (18.2 μ s) was used for all solution properties determinations except for surface tension which was obtained in Volvic slightly mineralized water.

Laser Raman spectra were recorded on a V45 model microspectrofluorometer (Dilor, France) equipped with an Olympus microscope comprising a $100 \times$ immersion objective. The monochromator consisted of a grating blazed at 600 grooves/mm and Raman signal detection was done with a two-dimensional (300×1152) pixels) CCD apparatus. Spatial resolution was about 8 cm^{-1} .

Excitation, was done by the 514.5 nm line of an Ar^+ laser (model 2065, Spectra Physics) operating at 500 mW.

Acquisition and treatment of data were done via an appropriate computing system and spectral decomposition was performed by the least-squares method.

Microwave spectra of saturated sugar solutions were obtained with a Proteon spectrometer (Société de Controle Moleculaire, Athis-Mons, France). These spectra are characterized by an amplitude obtained with an accuracy of ± 1 mV and a frequency (± 48 kHz). The increase in amplitude is generally attributed to an increase in polarity and that of frequency to an augmentation of packing (order).

in Table 1 for sugars (D-glucose, p-fructose and sucrose) and polyols (xylitol, sorbitol, lactitol), the solvent being pure water or 1% of salt (NaCl, KCl or MgCl₂). The value of intrinsic viscosity is between $2.30-2.40 \text{ cm}^3 \text{ g}^{-1}$ for sugars and 2.30–2.50 $\text{cm}^3 \text{.g}^{-1}$ for polyols. Huggins constant k' shows a slight decrease in presence of MgCl₂ for sugars and xylitol. A slight increase in $[\eta]$ for sucrose, fructose and xylitol is observed in presence of 1% MgCl₂ as well as a decrease in k'. As $[n]$ is the hydrodynamic volume derived from the flow time of the hydrated solute in the solvent and k' the descriptor of the ease of exchange between hydration water and bulk water, the influence of small amounts (physiology concentration) of salts is of relevance to the mechanism of sweetness. Magnesium cations are hexa-hydrated and their structure-maker effect is extended beyond the 6 water molecules of hydration to the bulk water (Theophanides *et al.,* 1990). Therefore, even with 1% concentration, the overall effect of Mg^{2+} is a decrease in mobility of water manifested by a slight increase in [n] and a decrease in k'. Although $Na⁺$ and $K⁺$ are involved in the sweet taste transduction cascade, the macroscopic effects given by $[\eta]$ and k' are not sensitive enough to the effects of these ions on water structure.

Apparent specific volumes (V_2^0) and *hydration number* (h) are listed in Table 2. All observed values of ASV are confined to the $0.50-0.70$ cm³.g⁻¹ range assigned to sweet taste (Shamil & Birch, 1987). Sugars, which have the same quality of sweetness, show an ASV of 0.61- $0.63 \text{ cm}^3 \text{.} \text{g}^{-1}$ whatever the solvent whereas ASVs of polyols vary from 0.62 for lactitol to 0.67 $cm³$.g⁻¹ for xylitol. Again, this densitometric method is not precise enough to reveal the small differences in solute-solvent interactions. The effect of 1% salt on the aqueous solvent is better revealed from hydration number determination. Indeed the negative hydration of K^+

RESULTS AND DISCUSSION

Intrinsic viscosity [v] and *Huggins constant* (k') are listed

Table 1. Intrinsic viscosity $[\eta]$ and Huggins constant k' of D-glucose, D-fructose, sucrose, xylitol, lactitol and sorbitol in water and 1% aqueous salt solutions

		sucrose		glucose		fructose		xylitol		lactitol		sorbitol
	$[\eta]$ $cm^3.2^-$	k'	[n] $cm3.g-1$	k	[7] $cm^3.g^{-1}$	\mathbf{k}'	$[\eta]$ $cm3.g-1$	k,	$[\eta] % \centering \includegraphics[width=0.9\columnwidth]{figures/fig_10.pdf} \caption{The graph α in the case of d-error of the network. The left is the number of~\alpha$-error of the network.} \label{fig:time}$ cm ³	\mathbf{k}'	$[\eta]$ $cm3$.g ⁻¹	\mathbf{k}'
H ₂ O	2.36	1.15	2.34	1.33	2.28	0.89	2.30	1.12	2.49	1.11	2.47	0.75
NaCl	2.37	1.14	2.36	1.08	2.32	0.88	2.28	1.13	2.45	1.33	2.45	0.84
KCI	2.36	1.18	2.35	1.11	2.29	0.74	2.28	1.12	2.45	1.29	2.38	1.04
MgCl ₂	2.40	1.07	2.34	1.12	2.31	0.72	2.37	0.82	2.50	1.30	2.42	1.04

Table 2. Apparent specific volume (V⁹) and hydration number (h) of sugars and polyols in water and 1% aqueous salt solutions

	B l.mol ⁻¹	B_{st} l.mol ⁻¹ sucrose	B_{st}/B	B l.mol ⁻¹	Bst 1 mol^{-1} glucose	B_{st}/B	B l.mol ⁻¹	B_{st} l.mol ⁻¹ fructose	B_{st}/B
H_2O	0.810	0.276	0.34	0.423	0.142	0.34	0.412	0.135	0.33
NaCl	0.813	0.269	0.33	0.426	0.143	0.34	0.418	0.140	0.33
KCI	0.807	0.269	0.33	0.423	0.139	0.33	0.413	0.131	0.32
MgCl ₂	0.822	0.290	0.35	0.422	0.140	0.33	0.417	0.141	0.34
		xylitol			lactitol			sorbitol	
H_2O	0.350	0.095	0.27	0.857	0.322	0.38	0.450	0.156	0.35
NaCl	0.347	0.092	0.27	0.844	0.311	0.37	0.446	0.148	0.33
KCl	0.347	0.089	0.26	0.844	0.305	0.36	0.434	0.139	0.32
MgCl ₂	0.360	0.105	0.29	0.859	0.325	0.38	0.440	0.147	0.33

Table 3. B coefficients and the ratio B_{st}/B for sugars and polyols in water and 1% aqueous salt solutions

Fig. 1. Ratio of viscosity coefficients B_{st}/B for sugars and polyols in water and in 1% (w/w) salt solutions.

(Samoilov, 1957) is shown in the slight decrease of h as compared to water for all sugars and polyols experimented. The increase in h in the presence of $MgCl₂$ is expected from the structure-maker effect of Mg^{2+} .

B coeficients were calculated using Jones & Dole (1929) and Einstein-Simha relations (Simha, 1940). The values of B, $B_{structure}$ and the ratio B_{size}/B are reported in Table 3. The coefficient B expresses intrinsic viscosity in dm^3 .mol⁻¹ and no appreciable change is observed as for $[\eta]$. The ratio B_{st}/B is more informative about the effect of the three halides on the structure of water in the presence of sugars and polyols. The representation of this ratio for the different solvents (see Fig. 1) shows, clearly, the tendency of decrease in the presence of KC1 and increase in the presence of $MgCl₂$ for this ratio.

Surface tension (γ) results are also represented graphically (see Fig. 2) and clearly show the opposite effects of KC1 on the one hand (chaotropic effect) and NaCl and $MgCl₂$ (structure-maker effect) on the other. As surface tension is a macroscopic manifestation of the cohesion of water molecules, these small differences in water mobility obtained from such an overall technique, will certainly be amplified and clearly demonstrated by use of microscopic methods.

From physicochemical results, we find that the accession of the sweet molecules to receptor site which necessitates a certain size of molecule and type of hydration is not greatly changed in the presence of 1% NaCl, KCl or MgCl₂. However, the water of hydration at the surface of the sugars or polyols seems to be qualitatively different. The time of residence of water molecules hydrating K^+ is shorter than in contact with another H_2O . $MgCl_2$ shows the opposite effect with long-living hydration $(6 H₂O)$ and a structure-maker

Fig. 2. Surface tension in mN/m for 10% sugars (D-glucose \triangle , D -fructose \bullet and sucrose \bullet) in water and in aqueous salt solutions.

Fig. 3. Experimental Raman spectrum of water in the 3800– 2800 cm^{-1} region and the calculated 4 components.

effect on the bulk water. These effects on water structure are better studied with laser Raman spectroscopy.

MICROSCOPIC METHODS

Laser-Raman spectra of aqueous solutions (10% (w/w)) of sugars were recorded in the 3800-2800 cm^{-1} range (Fig. 3). The solvents were either pure water or 1% (w/w) salt. Decomposition of the experimental Raman envelope was made using a 4 components model of water structure Luu *et al.* 1982. This model is just a basis of comparison of the effect of each of the solutes on water structure. The guidelines used to comment on Raman results are as follows.

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and the results of decomposition of the
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Table 4 Table 5. Compari The spectra of aqueous solutions at 1% (w/w) concentration of NaCl, KCl and $MgCl₂$ are shown in Fig. 4 and the results of decomposition of the experimental Raman band into 4 Gaussian components listed in Table 4 Table 5. Comparison of the frequencies and integrated intensities (area) of the OH vibration of water in presence and absence of salts yields the shifts in frequencies and change in weight (intensity) of each of the 4 components. The shifts in frequencies lie within the accuracy range $(\pm 8 \text{ cm}^{-1})$ for NaCl and KCl whereas the effect of 1% MgCl₂ on water structure is more evident from the data reported in Table 5. It is especially the case for component b assigned to amorphous solid species (Bounaaj, 1990) of water clusters which shows a significant shift towards higher frequencies $(+24 \text{ cm}^{-1})$. The less organised water associates corresponding to component c show a shift towards lower frequencies (-10 cm^{-1}) .

> Although the general rule for H-bonds is that these weak interactions are reflected in the OH-stretching region by frequency shifts towards lower wavenumbers, the perturbation of the structure of the hydrogen bonded species by a solute can be manifested by shifts either towards higher or lower frequencies Mathlouthi & Portmann, 1990. As concerns water, it is evident that free H_2O molecules show a band at the higher frequency

Fig. 4. Experimental Raman spectra of aqueous salt solutions (1% NaCl, 1% KCl and 1% MgCl₂) and the calculated 4 components a, b, c, d.

Table 4. Position of maximum (v), height of peak (I), area (integrated intensity), % total area and the assignment of the calculated components a, b, c and d of the Raman band of water in the 3800-2800 cm-'

component	ν (cm ⁻¹)	(arb. units)	area	area $(\frac{9}{6})$	assignments
a	3261	493	38496	42.61	quasi crystalline O O \sim 270 pm
	3426	394	24125	26.70	amorphous solid O O \sim 280 pm
c	3524	406	21842	24.18	amorphous liquid O O \sim 290 pm
	3666	159	5879	6.51	unassociated H ₂ O O O > 300 pm
total area			90342		

Table 5. Calculated frequencies (ν) and % area for the Raman band of water and the shifts in frequencies ($\Delta \nu$) and change in integrated intensity (\triangle area %) of the 4 components a, b, c and d for aqueous NaCl (1%), KCl (1%) and MgCl₂ (1%)

component	H_2O		$NaCl + sucrose$		$NaCl + glucose$		$NaCl + fructose$	
	$\boldsymbol{\nu}$ (cm^{-1})	area (%)	$\Delta \nu$ (cm^{-1})	Δ area (%)	$\Delta \nu$ (cm^{-1})	Δ area (%)	$\Delta \nu$ (cm^{-1})	Δ area (%)
a	3261	43	$+1$	-2	-7	-6	$+3$	-1
b	3426	27	$+7$	-2	-8	$+4$	$+9$	-1
c	3524	24	-3	$+5$	$+1$	$+2$	$+1$	$+6$
d	3666	7	$+3$	-3	$+3$	-2	$+4$	-3
component	H_2O		$KCl +$ sucrose		$KCl + glucose$		$KCl +$ fructose	
a	3261	43	$+6$	$+0$	$+0$	-2	$+5$	$+0$
b	3426	27	$+12$	-3	$+4$	-1	$+13$	-3
c	3524	24	$+0$	$+5$	$+6$	$+6$	-1	$+5$
d	3666		$+3$	-3	$+6$	-4	$+2$	-3
component	H_2O			$MgCl2 + sucrose$		$MgCl2 + glucose$	$MgCl2 + fructose$	
a	3261	43	$+2$	-1	$+7$	$+0$	$+3$	$+0$
b	3426	27	$+5$	-2	$+20$	-4	$+8$	-2
c	3524	24	$+1$	$+5$	$+1$	$+6$	-1	$+4$
d	3666	7	$+3$	-3	$+4$	-3	$+4$	-3

Table 6. Shifts in frequencies (Δv cm⁻¹) and changes in integrated intensities (Δ area %) of the 4 components of water in ternary solutions (10% sugar $+$ 1% salt): comparison to the Raman band of water

3666 cm-'. The three other water associates (components a, b, c) involve both donors and acceptors of H-bonds and the interpretation of their frequencies is therefore rather complex. If we assume that these species of water associates: a (quasi-crystalline); b (centered tetrahedra); and, c (chain-like), maintain their binding in presence of solutes but only become hindered in their motion, we can assign to this hindrance the increase in frequency for OH-stretching in the same species due to an increase in clustering and the decrease in frequency as a result of a decrease in clustering of the same type of water species. Thus the effect of $MgCl₂$ (1%) is to increase the clustering of the more organised species a and $b (+9)$ and $+24$ cm⁻¹) and to decrease that of the less organized species (-10 cm^{-1}) . The overall effect of Mg²⁺ cation on water is a structure maker effect due to the increase of the clustering of the most organised water associates.

The effects of binary mixtures (1% (w/w) salt $+10\%$ (w/w) sugar) on the Raman spectra of water are treated by the same method. After decomposition of the experimental envelopes of OH-stretching band into 4 components, the frequencies and integrated intensities are compared to that of pure water. The results are reported in Table 6. Although, the shifts in frequencies lie within the ± 8 cm⁻¹ range except for component b in NaCl-fructose solution, which shows a shift slightly higher ($+9$ cm⁻¹), it may be noticed that the behaviour of D-glucose and D-fructose are opposed (Mathlouthli, 1986). A decrease in frequency (-8 cm^{-1}) is observed in the presence of D-glucose. This behaviour is also observed with KCl. From Table 6, we see that, whereas no significant change in frequency is observed for D-glucose/ KC1 mixture, the component b shows a shift towards higher frequencies of $+13$ cm⁻¹ with D-fructose and $+ 12$ cm⁻¹ with sucrose in the presence of KCl. Another feature to be noticed is the opposite behaviour of **D**glucose in the presence of NaCl and KCl. The shifts of

frequencies are towards lower frequencies for component b (-8 cm⁻¹) in the presence of NaCl and towards higher frequencies $(+4 \text{ cm}^{-1})$ in the presence of KCl, but these shifts are moderate.

The largest perturbation of water structure for the ternary mixture (salt/sugar/water) is obtained for $MgCl₂/D-glucose/water.$ The component b shows up to 20 wavenumbers of increase in frequency (see Table 6) which is comparable to the effect of $MgCl₂$ alone on the spectrum of water (see Table 5). While D-fructose and sucrose compensate the tendency of $MgCl₂$ to increase the clustering of water associates, especially the tetrahedral swarms (component b), this is not the case for D-glucose.

As concerns the intensity of vibration, or the weight of each of the species in the overall energy of vibration, the most prominent fact is the decrease in the proportion of free H_2O molecules (component d). Indeed, although this species only constitutes 7% of the whole area of band for pure water, its decrease (-3%) is important on a relative basis ($\frac{3}{7} \approx -43\%$) and means that putting salts or sugars in water helps to reduce the proportion of non-associated water molecules.

If the comparison of the spectra of ternary solutions is made by reference to salt solutions, the differentiation of $MgCl₂$, on the other hand, and NaCl and KCl, on the other, becomes more evident; the results are summarized in Table 7. The shifts in frequencies towards lower wavenumbers are the largest for $MgCl₂/sugar$ mixtures than in the presence of NaCl or KCI, especially for component b which shows a decrease of -19 cm⁻¹ for sucrose and -16 cm⁻¹ for fructose. These shifts are opposite to what was observed for $MgCl₂$ alone (+24) cm^{-1}) (see Table 5). This means that, whereas both sugars and $MgCl₂$ are known to enhance the hydrogen bonding of water molecules, their specific effects on the energy of these bonds are opposite. Furthermore, the

component	NaCl		$NaCl + sucrose$		$NaCl + glucose$		$NaCl + fructose$	
	ν (cm^{-1})	area (%)	$\Delta \nu$ (cm^{-1})	Δ area $(\%)$	$\Delta \nu$ (cm^{-1})	Δ area (%)	$\Delta \nu$ (cm^{-1})	Δ area (%)
a	3258	41	$+4$	$+0$	-4	-4	$+6$	$+1$
b	3424	28	$+9$	-3	-6	$+3$	$+11$	-2
c	3526	26	-5	$+3$	-1	$+0$	-1	$+4$
d	3670	5	-1	-1	-1	$+0$	$+0$	-1
component	KCl			$KCl +$ sucrose		$KCl + glucose$	$KCl +$ fructose	
a	3261	41	$+6$	$+2$	$+0$	$+0$	$+5$	$+2$
$\mathbf b$	3434	27	$+4$	-3	-4	-1	$+5$	-3
c	3518	27	$+6$	$+2$	$+12$	$+3$	$+5$	$+2$
d	3669	5	$+0$	-1	$+3$	-2	-1	-1
component	MgCl ₂			$MgCl2 + sucrose$		$MgCl2 + glucose$	$MgCl2$ + fructose	
a	3270	47	-7	-5	-2	-4	-6	-4
b	3450	22	-19	$+3$	-4	$+1$	-16	$+3$
c	3514	26	$+11$	$+3$	$+11$	$+4$	$+9$	$+2$
d	3666	5	-3	-1	$+4$	-1	$+4$	-1

Table 7. Shifts in frequencies ($\Delta \nu$ cm⁻¹) and changes in integrated intensities (Δ area %) of the 4 components of water in ternary **solutions (10% sugar + 1% salt): comparison to the Raman band of the aqueous salt solution**

most organised species (a, b) are affected in a different way from the less organised clusters (component c). Again the results obtained with D-glucose are different from what was observed for sucrose or D-fructose mixed with 1% KCl or MgCl₂ (see Table 7).

Tables 6 and 7 show that the most comparable data are obtained for NaCl/sugar mixtures. These two solutes, although different in nature, seem to change the clustering of water molecules in the same way. Moreover, the taste of sodium chloride in dilute solutions might be sweet and an enhancement of sweetness is observed in NaCl/sucrose mixtures (Barisas *et al.,* 1995). KCl and $MgCl₂$ have different tastes, generally described as bitter or astringent. Their effects on water clustering are also different as may be observed from Raman results. KC1 seems to perturb water structure less than $MgCl₂$.

Microwave spectra

An example of microwave spectra is given in Fig. 5 and the results obtained for aqueous saturated solutions listed in Table 8. These results show a higher polarity

Fig. 5. Microwave spectrum of saturated aqueous sugar solution.

for fructose solution than sucrose and glucose. The degree of packing in these solutions follows the same order. Although saturated solutions correspond to a stable equilibrium of hydration, the way the solute molecules are arranged and the mobility of water around these solutes may be described by microwave spectroscopy. An increased chaotropic movement of water molecules corresponds to a higher amplitude of signal. The packing of solutes due to higher solubility of fructose is manifested by an increased frequency of resonance of the microwave field. These results are preliminary results. They will be extended to ternary solutions and other sweeteners in order to determine directly the hydrophilicity (polarity) of sweeteners.

CONCLUSION

We presented in the first ECRO sweetness symposium (Mathlouthi *et al.,* 1993) a three-step model of sweetness chemoreception. In this model the cascade of events underlying the sweet taste mechanism are: *accession* to site (first step); *stimulation* of receptor membrane, opening of ions channels and Na^+/K^+ transfer (second step); and, the *perception* of sweetness intensity and duration (third step). The effect of salts on the accession to site may be summarized from macroscopic results as

Table 8. Observed amplitudes and frequencies in the microwave spectra of saturated aqueous solutions of D-glucose, D-fructose and sucrose

Sugar	amplitude (mV)	frequency (MHz)		
glucose	979.2	2441.641		
fructose	989.0	2442.672		
sucrose	983.3	2441.907		

only slight as the size and shape of stimulants are only slightly modified. However, the slight detergent effect of KC1 (decrease in surface tension) should be related to its astringent-bitter taste, though it should be noted that the levels of salts used in this study are higher than is encountered in saliva.

The more evident differences originating from Na^+ , K^+ and Mg^{2+} ions are obtained with Raman spectra. A preponderant effect of Mg^{2+} was established and the differentiation of the sugars made clearer. Low levels of Na⁺ are known to enhance the sweetness of sucrose and other sweeteners (Barisas *et al.*, 1995). Na⁺ and K^+ are very likely involved in a 'transduction cascade'. Mg^{2+} is known to play an important part in biological mechanisms. These results of the physicochemical properties and Raman spectroscopy of aqueous solutions of nutritive sweeteners in the presence of small amounts of NaCl, KCl and MgCl₂ need to be completed by sensory studies. Then a correlation between the more or less important mobility of water induced by these ions will be made with their influence on sweetness intensity and persistence.

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