

Ion specificity in determining physico-chemical properties of drinking water

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Abstract

Drinking water plays a key role in regulating the daily salt intake, thus potentially affecting (either in positive or in negative direction) the incidence of diet-related pathologies, including nephrolithiasis. Being regarded as wholesome, bottled water is increasingly preferred to tap water. However, its physico-chemical characterization, as reported on the label, is usually limited to pH and conductivity measurements, along with ion content. Other parameters, which also are likely to influence the processes in which water is involved in the body, are usually neglected. In this work, the surface tension and density of tap water and two bottled mineral waters produced in Italy are investigated. These parameters are critically determined by the ion content. Density is connected to the total ion amount, while surface tension is differentially affected by the presence of specific ions: sulfate increases its value, while bicarbonate keeps it almost constant. These results furnish a reliable scientific basis for future investigation on the connection between the physico-chemical properties of drinking water and its effect on health. Particularly, since interfacial phenomena play a key role in kidney stone formation, our results suggest that dietary enrichment of selected ions due to water intake could affect nephrolithiasis.

Keywords: bottled mineral water; labeling; surface tension; density; ion content.

Practical Application: This paper highlights the importance of implementing analytical tests on tap water as well as on bottled mineral waters. Parameters different from those routinely reported on the labels could be important in determining the water effect on the human health. These parameters depend in different way on the ion content. A deeper investigation on these points, joined to a wide dissemination of the results is essential to drive a correct choice of water to be drunk from the consumer to optimize the beneficial effects.

1 Introduction

Diet is considered to be a major risk factor for idiopathic calcium stone formation (Robertson, 2016; Prezioso et al., 2015), and is likely responsible for nephrolithiasis growing incidence in the last decades. Preventing recurrent urolithiasis, by adopting correct dietary habits, could lead to a significant cost savings for the national health care systems (Lotan et al., 2013). Many dietary factors have been implicated in this disease, including an insufficient fluid intake (Robertson, 1987), high or low intakes of calcium (Curhan et al., 1993), high intakes of oxalate (Robertson & Peacock, 1980) and sodium (Massey & Whiting, 1995), low intakes of magnesium (Reungjui et al., 2002) and potassium (Domrongkitchaiporn et al., 2006). As a matter of fact, drinking water plays a key role in regulating all these factors. Water effect is further enhanced by the fact that for all nephrolithiasis patients, regardless of the underlying cause of the disease, a keystone of conservative management is an increased water intake to achieve a daily urine output of two liters (Borghesi et al., 1999; Frank & Vries, 1966). Indeed, patients prefer a change in the quantity water intake to another dietary or pharmaceutical intervention to contrast the high probability of recurrence of nephrolithiasis (Hughes, 2007; Trinchieri et al., 2000). Moreover, they pay particular attention to the quality of drinking water (McCaughey et al., 2012).

Many people are now preferring bottled water to tap water. By 2016, bottled water volume achieved a new record, exceeding 48 billion liters, almost 15 billion liters higher than it had been in 2007 (Rodwan, 2017). Worldwide, there are thousands of companies bottling water. The impressive increase in bottled water consumption has to be ascribed to i.) consumers' apprehension over attaining safe and accessible drinking water; ii.) consumers' complaints about tastes and odors from municipal water due to additives such as fluoride or chlorine; iii.) manufacturers' advertising of bottled water as "pure" water, perfectly suited to kids and old people; and last, but not least, iv.) consumers' conviction that natural mineral waters exert beneficial, and even therapeutic, effects on human health (Salehi et al., 2014; Semerjian, 2011; Güler & Alpaslan, 2009; Ward et al., 2009). With respect to the last point, the connection with nephrolithiasis would appear obvious. Physico-chemical parameters of drinking water, and particularly hardness, fixed residue and ions concentration, are expected to impact on the risk of renal stones. In the case of bottled water, all these parameters are by law listed on the label, and significantly drive the consumer choice (Kermanshahi et al., 2010).

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From the scientific viewpoint, drinking water quality has been demonstrated to affect urine characteristics, including pH, the ratio of calcium crystal formation initiators and inhibitors, the concentration of ions (Schwartz et al., 2002). In turn, these play a fundamental role in regulating the processes leading to the formation of stones in the kidneys and urinary tract (Evan et al., 2015; Coe et al., 2010). On this basis, the relationship between nephrolithiasis and water physico-chemical features is object of active current research (Rabi et al., 2013). Most of the studies focus on the total quantity of dissolved electrolyte in drinking water (Bellizzi et al., 1999; Chatterjee et al., 2012) or, more specifically, on the effects of distinct electrolytes (Panhwar et al., 2013; Siener et al., 2004). Despite the researchers efforts, the effect of water hardness on nephrolithiasis remains unclear (Schwartz et al., 2002; Coen et al., 2001; Rodgers, 1997); on the other hand, it is now generally accepted that bicarbonate-rich water have a positive effect (Karagülle et al., 2007; Bertaccini & Borghesi, 2009), probably due to increased pH which increases solubility of uric acid and urinary citrate concentration, with inhibition of calcium crystal formation and aggregation. The presence of other solutes, such as humous (humic and fulvic) acids, has also been analyzed and connected with the degradation of stones in the urinary tract (Davydova et al., 2014). In contrast, analysis of collective physico-chemical parameters of water is usually limited to electrical conductivity and pH measurements (Siener et al., 2004). Investigation on other parameters is missing.

The nucleation and growth of stone-forming minerals is related to the interfacial energy between the liquid (urine) and the solid (the growing stone) (Wu et al., 1999; Russo et al., 2015). In turn, the interfacial energy is determined by the surface tension of the two phases put in contact (Marmur & Valal, 2010; Muñoz-García et al., 2015). The liquid surface tension is determined by the solutes content that is related to diet (Paßlack et al., 2014) and, especially in a regime of increased water intake, to drinking water (Toxqui & Vaquero, 2016; Schwartz et al., 2002). Thus, surface tension of drinking water is likely to be related to nephrolithiasis. However, scientific investigation on this specific parameter is missing.

In the present work, we present surface tension data (IUPAC symbol: γ) collected on tap water as well as on two bottled mineral waters commercially available in Italy. One of these, *Acqua Rocchetta*, has been reported to exert a significant detoxicant activity on the body (Paolo et al., 2000), and hypothesized to be beneficial in preventing nephrolithiasis. The other mineral water, chosen as a reference, presents very similar solid residue, thus allowing the relevance of the different salt composition (at almost constant total amount) to be highlighted. Moreover, to complement the surface tension data, we report density data (IUPAC symbol: ρ) acquired on the same samples.

2 Materials and methods

2.1 Water sampling

Tap water was sampled in Piazza Dante, a square in center of Napoli (Italia), in July 2017. Two brands of mineral water were considered. The former one is *Acqua Rocchetta* (whose spring is located in Gualdo Tadino, in central Italy); the latter one has been

chosen to allow a comparison and, in the following sections, will be indicated as Reference Mineral Water. The physico-chemical parameters of tap water, as reported on the official website of the local public water supplier (ABC – *Acqua Bene Comune*) and those of the two mineral waters, reported on the labels as required from the current legislation (Council of the European Union, 1980), are shown in Table 1.

A total of 10 water bottles for each considered brand were purchased in spring 2017 at 10 different supermarkets throughout the city of Napoli (Italia). The bottles, with a capacity of 0.5 liters, were made of PET (polyethylene terephthalate) and had a PE (polyethylene) screw cap. From each bottle, four water samples were analyzed by tensiometry and densimetry, and each measurement was repeated in triplicate.

2.2 Surface tension measurements

Surface tension measurements were performed using apparatus and procedures described in details in previous studies (Ortona et al., 2006). Briefly, the surface tension, γ , was measured by the De Nouy ring method using a KSV Sigma 70 digital tensiometer equipped with an automatic device to set the time between two consecutive measurements and to select the rising velocity of the platinum ring. Mineral water samples were put in the tensiometer circular vessel, thermostatted at 25.00 ± 0.01 °C. Instrument accuracy was checked to be better

Table 1. Physicochemical parameters reported on the official website of the neapolitan public water supplier ABC (*Acqua Bene Comune*) for tap water and on labels of the investigated bottled mineral waters.

	Tap Water	Acqua Rocchetta	Reference Mineral Water
Solid Residue at 180 °C / (mg/L) ^a	536	174.1	162
Specific Electric conductivity at 20 °C / ($\mu\text{S}/\text{cm}$) ^b	765	276.3	251
pH at the spring temperature	7.4	7.56	7.9
Ca ⁺⁺ / (mg/L)	126.0	57.36	35.9
Na ⁺	35	4.13	2.0
Mg ⁺⁺	31.3	3.23	12.6
K ⁺	17.5	0.35	0.5
Sr ⁺⁺	not reported	0.23	not reported
HCO ₃ ⁻	503	182.1	148
SO ₄ ⁻	55	6.75	19.3
Cl ⁻	50	6.78	2.4
NO ₃ ⁻	31	1.10	4.4
F ⁻	0.69	0.13	not reported
SiO ₂	not reported	4.17	not reported
NO ₂ ⁻	0.03	not reported	<0.002

^aThis value represents the mass of inorganic and organic substances that remains after complete evaporation of one liter of water at 180 °C, according to the Italian D. Lgs. 31/2001 (Italy, 2001); ^bThe specific electric conductivity of ultrapure water is 0.055 $\mu\text{S}/\text{cm}$.

than 0.10 mN m^{-1} by measuring γ for double distilled and degassed water (surface tension of ultra-pure water at 25°C is 71.97 mN m^{-1}) before and after each session of measurements.

2.3 Density measurements

Density measurements were performed using apparatus and procedures described in details in previous studies (Molisso et al., 2010; Vergara et al., 2002; Ambrosone et al., 1997). Briefly, densities, ρ , of water samples were determined with an Anton Paar DMA5000 density meter at the temperature (25.000 ± 0.005) $^\circ\text{C}$, using double distilled water and air at known pressure and humidity as references. According to the specifications declared for this instrument the repeatability is equal to $10^{-6} \text{ g}\cdot\text{cm}^{-3}$.

2.4 Statistical treatment of data

The statistical analyses were performed using the Graph-Pad Prism version 7.00 (GraphPad Software). D'Agostino and Pearson normality test was performed and groups showing normal distribution were analyzed by the t-test, while groups showing not-normal distribution were analyzed by Mann-Whitney U test; two-tailed $P < 0.05$ was chosen as significance threshold.

3 Results and discussion

Experimental surface tension data are shown in Figure 1. The average values are summarized in Table 2. Tap water presents a surface tension, γ , higher than that of double distilled and degassed water (72.40 mN m^{-1} vs. 71.97 mN m^{-1}). The datum obtained for *Acqua Rocchetta* ($72.04 \pm 0.08 \text{ mN m}^{-1}$) coincides, within the experimental uncertainty, with that of double distilled

water. The datum obtained for the Reference Mineral Water ($72.33 \pm 0.09 \text{ mN m}^{-1}$) is similar to that observed for tap water.

It is interesting to analyze these data on the basis of the salt contents reported in Table 1. As a general consideration, the presence of inorganic salts increases the surface tension of aqueous mixtures (Chen et al., 2017). This is interpreted considering that ions tend to be surrounded by hydrating water molecules, thus avoiding residing at the air/water interface; this means that fewer ions are found at the interface than in the water bulk. As a consequence, the interface tends to shrink, and this results in an increase of the surface tension (Dutcher et al., 2010). This is confirmed by the values obtained for tap water and for the Reference Mineral Water, showing values higher than that of double distilled and degassed water (see Table 2). One could expect surface tension to positively correlate with solid residue values. Indeed, the solid residue which remains after complete evaporation of one liter of water at 180°C is mostly constituted by inorganic salts, see Table 1, so that it can be considered a coarse index of the ion content in the considered water samples. Similar considerations hold for the electric conductivity, which also is a measure of dissolved electrolytes. However, the positive correlation is not confirmed by our results: *Acqua Rocchetta*, which has a similar (slightly higher, indeed) solid residue and specific electric conductivity values with respect to the Reference Mineral Water and, overall, a higher salt content (see Table 1), presents a lower surface tension. Thus, our data show that the surface tension is affected by the presence and amount of specific ions rather than their total amount. Perusal of Table 1 reveals that: i) concerning the cations, *Acqua Rocchetta* presents a high content of sodium and calcium while the magnesium content is low; ii) concerning the anions, *Acqua Rocchetta* presents a high content of bicarbonate and a low content of sulfate.

Magnesium and sulfate are among the most “structure makers” ions (kosmotropes), considered to be able to induce ordering in water beyond their hydration shells (Tobias & Hemminger, 2008), while large singly charged ions, with low charge density, such as bicarbonate, act as “structure breakers” (chaotropes) (Du et al., 2008). Kosmotropes prefer to reside in the bulk water, where ordered structures can be fully formed in all directions. This driving force is much minor for chaotropes, which can remain at the interface, thus being much less effective in increasing the solution surface tension. This interpretation is fully in line with our experimental evidences.

Experimental density data are shown in Figure 2. The average values are summarized in Table 2. Tap water present a density (0.997877 g/cm^3) higher than that of double distilled water (0.997045 g/cm^3). The datum obtained for *Acqua Rocchetta* (0.997294 g/cm^3) is only slightly different from the datum obtained for the Reference Mineral Water (0.997274 g/cm^3) and

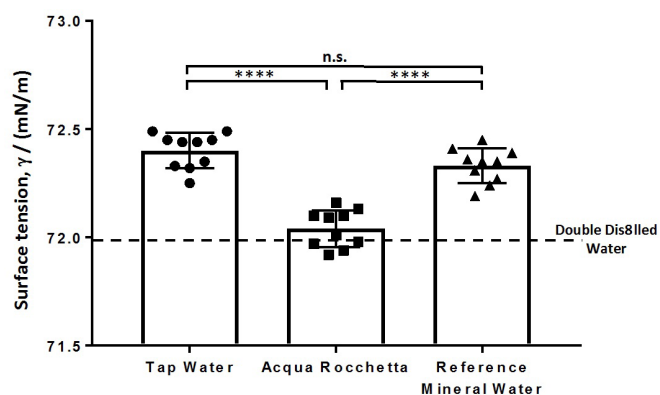


Figure 1. Surface tension, γ , of tap water and bottled mineral waters at 25°C . Bars represent the mean value \pm standard deviation (**** $P < 0.0001$; n.s. means no significant difference).

Table 2. Surface tension, γ , and density, ρ , of the investigated tap and mineral waters at 25°C .

	Degassed and Doubly Distilled Water	Tap Water	Acqua Rocchetta	Reference Mineral Water
Surface tension, γ /(mN/m)	71.97 ^a	72.40 ± 0.08	72.04 ± 0.08	72.33 ± 0.09
Density, ρ /(g/cm ³)	0.997045 ^b	0.997877 ± 0.000005	0.997294 ± 0.000008	0.997274 ± 0.000005

^aFrom Ortona et al. (2006); ^bFrom Ambrosone et al. (1996).

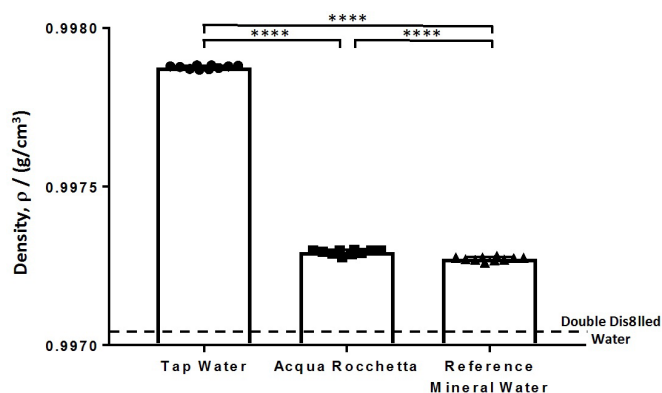


Figure 2. Density, ρ , of tap water and bottled mineral waters at 25 °C. Bars represent the mean value \pm standard deviation (**** $P < 0.0001$).

both of them are significantly higher than the density of double distilled and degassed water.

Even in this case, the experimental evidences can be related to the ion content. When a salt is dissolved in water, the solution density usually increases. This evidence is ascribed to the electrostriction phenomenon: under the electric field exerted by ions, the hydrogen bonds between water molecules break down leading to a molecular crowding and a diminution of the void spaces (Marcus, 2011). Inspection of Table 1 shows that the density increment well correlates with the solid residue and the specific electric conductivity of the investigated samples. Tap water, whose solid residue is much higher than those of bottled waters, presents a much higher density. In the same manner, similar densities correspond to similar solid residue values, the slightly higher density observed for *Acqua Rocchetta* being in line with its solid residue which slightly higher than that of the Reference Mineral Water. Thus, it seems that the ions total amount is the fundamental properties in regulating the density of mineral water; specific effects exerted by selected ions, reported in the literature (Mazzini & Craig, 2017), play a minor role.

4 Conclusions

Studies concerning nephrolithiasis disease distribution demonstrate that, besides a specific genetic predisposition, environmental factors, and specifically dietary habits, are important (Miller et al., 2007). A correct nutritional management of stone formers patients is effective in preventing or at least delaying kidney stones formation and recurrence, thus reducing the need of drugs and the consequent risks of intolerance and side effects (Prezioso et al., 2015). Particularly, the quantity and quality of drinking water seems to play a relevant role in contrasting nephrolithiasis. However, the water characteristics that determine this beneficial effect remain poorly understood.

In the present study we investigated the surface tension and density of two bottled mineral waters with similar solid residue. We found similar density values, indicating that this property is mainly regulated by the total amount of ions. Interestingly, this is not true for surface tension: one of the water brands presents a value very close to that of neat water, while a significant surface

tension increase is observed for the other one. This evidence has been interpreted in terms of the different ions present in the two mineral waters. Particularly, we propose sulfate and magnesium to cause a surface tension increase. At the molecular level, this could be connected to the effect of ions on the structure of water, which is related to the subtle balance between ion–water and water–water interactions. Tap water, whose ion content is much higher with respect to those of the considered bottled waters, present both higher density and higher surface tension.

As a general conclusion of our work, it is to be remarked that, among the various features of drinking water, the importance of solubilized ions cannot be overstated. Beyond their total amount, the specific nature is vital in controlling a wealth of physical, chemical and biological properties. The ion specificity in driving a given physico-chemical phenomenon in aqueous solution is largely variable: some phenomena seem to nonspecifically depend on the total ionic strength, while others are only induced by selected ions. In these cases, the order of ions effectiveness is hardly predictable (Imperatore et al., 2014). Our results demonstrate that the surface tension of water depends on the presence of specific ions. Being interfacial phenomena fundamental in crystal growth underlying kidney stone formation, it can be reasonably speculated that enrichment of urine in selected ions due to water intake could affect nephrolithiasis.

Although far from conclusive, this study shows that small variations in ions content and composition could significantly affect physico-chemical properties of drinking water. The extent to which this variation reflects in the healthy effect of a given mineral water brand remains to be established. In this respect, surface tension is a perfect candidate to be further investigated as a key property to be related to the insurgence of nephrolithiasis.

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