Evaluation of bottled water quality by determining nitrate concentration

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ABSTRACT

The presence of nitrate in sources of drinking water is a matter of concern because of its potential risk for human health. In many countries like Argentina, an increasing proportion of the population chooses to consume bottled water, among other reasons, for lack of water access. The present study was conducted (a) to evaluate the quality of bottled waters by determining nitrate concentration, (b) to relate bottled water quality with water access, (c) to analyze public awareness about bottled water quality and consumption habits of the population in the urban area of Buenos Aires. Two locations were selected, Ciudad Autónoma de Buenos Aires (C.A.B.A.) and Malvinas Argentinas in Buenos Aires Province (PBA), with percentages of water access of 99.6% and 8.8%, respectively. Random samples from both locations ($n = 100$) were analyzed. A survey was conducted in order to inquire about perception of population on bottled water quality and their consumption habits. In C.A.B.A., no sample exceeded the 45 mg/L limit value in force in Argentina, while in Malvinas Argentinas, 34% of the brands analyzed showed values above it. The survey revealed that 71.7% of people consume bottled water. While people in C.A.B.A. do so mainly out of habit, safety is the priority in PBA.

Key words | bottled water, nitrate contamination, perceptions, water access, water quality

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HIGHLIGHTS

- Nitrate concentration in bottled water was determined in two neighborhoods in the urban area of Buenos Aires, C.A.B.A. and Malvinas Argentinas, with significant differences in their access to water.
- Nitrate contamination of bottled water was much higher in the area with the least access to drinking water.
- Nitrate values exceeded in many cases 100 mg/L.
- A survey carried out shows a high consumption pattern for bottled water.
- The reasons for choosing bottled water were different in the two study areas.

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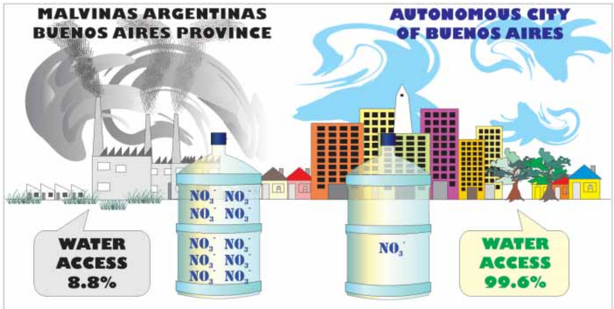
GRAPHICAL ABSTRACT



INTRODUCTION

Improvement in water access is a central objective in plans for future development, for example, in the Millennium Development Goals of the World Health Organization. Moreover, in 2010 the right to water and sanitation was considered by the United Nations General Assembly in Resolution A/RES/64/292 as a human right, since it is a requirement for the realization of all human rights.

Meanwhile, consumption of bottled water has continued to increase over time worldwide. In some cases, bottled water can be used as a substitute for tap water when problems with its supply occur. Numerous studies related increase of bottled water consumption in response to water quality alerts (Dupont & Jahan 2012; Pennino et al. 2017; Allaire et al. 2019; Cohen et al. 2017).

However, the reasons for the increased sales of bottled water go far beyond the lack of provision of municipal water supply. First of all, even in the absence of tap water problems, people choose bottled water because they consider it to be safer (De Queiroz 2013; Levêque & Burns 2017; Robak & Bjornlund 2018). Second, it has been said that the reason for the choice has to do with a matter of taste (Doria 2006; York et al. 2011; Font-Ribera et al. 2017).

These arguments are difficult to sustain rationally, particularly the issue of safety. In many countries, such as the United States or Argentina, where this study was carried out, drinking bottled water does not have the same strict standards as tap water (Levêque & Burns 2018; Litter et al. 2019). In addition, the evaluation frequency is different and so is the communication of standard non-compliance. Incidents involving municipal water must be mandatorily disclosed (EPA 2009). The bottled water industry takes advantage of this negative publicity from its main competitor.

Contrary to popular belief, there are numerous reports of contamination due to bottled water, which include the most diverse contaminants in very different regions. As an example, the presence in bottled water of nitrate in Argentina (Gallego 2006), bromate in the UK (Gillingham & Noizet 2007), arsenic in Chile (Daniele et al. 2019), turbidity in Malawi (Chidya et al. 2019), pesticides in Mexico (Díaz et al. 2009), endocrine disruptors in Sudafrika (Anech Hahn et al. 2018), and microplastics in many countries (Mason et al. 2018) can be cited. Even outbreaks of infectious diseases due to mineral water have been reported, surprisingly, a cholera outbreak in Portugal in 1974 (Blake et al. 1974) and, more recently, a norovirus outbreak in Spain (Blanco et al. 2017).

In some cases, concern regarding bottled water is not due to what it has but due to what it does not have. The example is lack of fluoride (Rosinger et al. 2018) or hardness (Klevay 2018), both considered beneficial for health.

Regarding taste, it would be more related to the chemical composition than to the origin (Harmon et al. 2018). Although it is true that tap water with relatively high trihalomethane content can be unpleasant (Font-Ribera et al. 2017), the differences are not always discernible. In a blind taste test carried out in Germany, Debeller et al. (2018) found that consumers were not able to discriminate between tap and bottled water.

Some researchers suggest that in addition to safety and taste there are other factors involved in the selection of some kinds of water, including psychological and behavioral issues, such as the social status or the lifestyle people have or intend to demonstrate (York *et al.* 2011; Etale *et al.* 2018).

In opposition to the debatable advantages listed, bottled water has two very notable disadvantages. The first is its cost (Cohen *et al.* 2017; Komarulzaman *et al.* 2017) and the second its environmental impact (Van Der Linden 2015).

Ciudad Autónoma de Buenos Aires (C.A.B.A.) is the capital of Argentina. It is completely surrounded by the Province of Buenos Aires (PBA), forming an urban conglomerate where more than 31% of the country's population lives (INDEC 2010). It is common to refer to the entire area as the urban area of Buenos Aires, including C.A.B.A. and neighboring areas in the province also called 'Greater Buenos Aires'. Social inequalities in the area are remarkable. Access to water, for example, is close to 100% in C.A.B.A., with a level of control comparable to that of any developed country, while at less than 20 km away in Greater Buenos Aires the population may depend on the service of small companies, cooperatives or individual wells, with an often questionable quality level. Relating this to the consumption of bottled water, it can be foreseen that although in some areas it may be a choice, in others it is a necessity.

The water origin is also different according to the area. C.A.B.A. and the most privileged areas receive potable water from the Río de la Plata, while in the periphery the origin is mainly from underground sources (AySA 2020). This different origin is the reason that the possible pollutants present in each area may also be different.

In the urban area of Buenos Aires, one of the contaminants of major concern in groundwater is nitrate. It has an anthropogenic origin. In urban areas, nitrate comes mainly from septic wells from the population without access to sanitation systems. Due to its extreme solubility, nitrate reaches groundwater, where it accumulates, being able to reach very high concentrations (Zendehbad *et al.* 2019). The World Health Organization (WHO) recommends a guide level of 50 mg/L for nitrate in drinking water (WHO 2017). The Argentine Food Code (ANMAT 2020) established a limit of 45 mg/L, in response to concerns related to the risk for methemoglobinemia in infants less than six months of age who are fed infant formula mixed with water (Greer & Shannon 2005; Fossen Johnson 2019). In addition to this acute effect, nitrate has been suggested to be associated with an increase of colorectal cancer risk, due to endogenous transformation into carcinogenic N-nitroso compounds (Schullehner *et al.* 2018). Thyroid disease and neural tube defects were also associated with nitrate ingestion (Weyer *et al.* 2014; Ward *et al.* 2018). If the concentration limit is exceeded, the water must undergo proper treatment to meet the requirements. Treatments to remove nitrate are based on ion exchange or reverse osmosis. Some ceramic filters have also been proposed for economic removal in low-income households (Shivaraju *et al.* 2019).

Nitrate is one of the few chemical contaminants capable of producing acute toxicity, methemoglobinemia, in the population at risk (WHO 2017). Furthermore, it has been observed in previous studies that it can also be used as a quality indicator of bottled water (Gallego 2006). Since bottled water can come from an underground source, it is expected that if it is not suitably treated it may exceed the nitrate values required by legislation, due to the ubiquity of nitrate contamination in the area. Thus, the presence of a high concentration of nitrate would indicate bad practice in the elaboration.

The present study was conducted (a) to evaluate the quality of different commercial bottled waters from different locations by determining nitrate concentration, (b) to relate bottled water quality with water access in the different locations, (c) to analyze public awareness about bottled water quality and consumption habits of the population in the urban area of Buenos Aires.

This study was conducted within the framework of a project-based learning experience carried out with students of training course for assistants in the chair of Public Health and Environmental Hygiene, Faculty of Pharmacy and Biochemistry, Buenos Aires University. A description of the educational aspects of the proposal can be found in the quote that appears in the bibliography as González *et al.* (2020).

MATERIALS AND METHODS

Selection of the type of samples and working area

Large volume bottles (20 L) present in shops in the selected areas were chosen for the study.

The selection of the working area was carried out taking into account the data published by the last national population census (INDEC 2010), whose data are shown in Figure 1.

C.A.B.A. and Malvinas Argentinas, with 99.6% and 8.8% of water access, were chosen to carry out the study. In C.A.B.A., communes 2 and 3, both near the Faculty of Pharmacy and Biochemistry, but with very different characteristics, were selected. In Malvinas Argentinas, the areas classified as 'central area' according to the municipal cadastre map were chosen. These areas correspond to seven commercial zones located around the seven railway stations present in the municipality: Tortuguitas, Tierras Altas, Grand Bourg, Pablo Nogués, Los Polvorines, Villa de Mayo, and Sordeaux.

Randomization of sampling sites

A random sampling plan was made for the two zones. Municipal cadastre maps were used in both cases. The number of total blocks in the working area was counted and a number was assigned to each of them. Employing a random numbers table, the blocks where a sample would be taken were selected. In both cases it was decided to work with a total of 100 samples.

In C.A.B.A., the number of samples taken from commune 2 and commune 3 were proportional to the number of blocks that each commune has. It was planned to take four samples per selected block, one from each side. In the same way in Malvinas Argentinas, the number of samples from each locality was proportional to the size of the area. It was planned to take only one sample per block due to the lower presence of shops compared to C.A.B.A. The distribution of samples is shown in Table 1.

Sampling, storage and analysis of samples

The samples were taken from May to September 2018. They were collected in glass tubes with plastic screw caps, using 2 mL/L of concentrated sulfuric acid as a preservative.

Samples were processed within 48 hours. The UV screening method for nitrate (APHA/AWWA/WEF 2017), simple and precise but not specific, was chosen. Absorbance at 220 and 275 nm was measured (Metrolab UV 1700 Spectrophotometer). Potassium nitrate of analytical grade (Merck, Darmstadt, Germany) was employed for the realization of the standard curve. The concentration was expressed in all cases in terms of nitrate.

To confirm positive results, a commercial kit was employed (MQuant® Merck, Darmstadt, Germany). The test is specific and is based on the reduction of nitrate

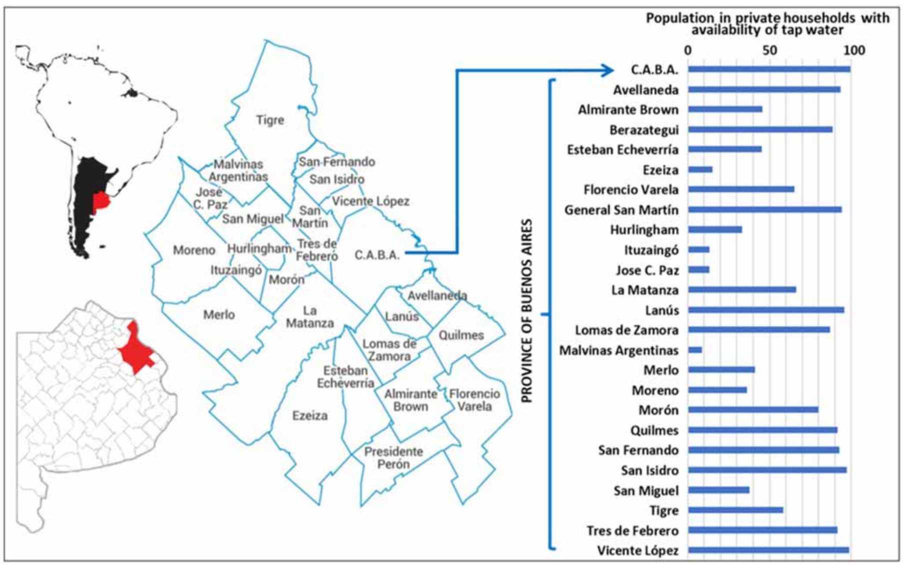


Figure 1 | Location of the sampling area and water access.

Table 1 | Distribution of the samples in the study areas

Work area	Locality	Number of blocks	Number of samples	Samples per block
C.A.B.A.	Commune 2	54	40	4
	Commune 3	80	60	4
	Total	134	100	
Malvinas Argentinas	Tortuguitas	21	18	1
	Tierras Altas	2	2	1
	Grand Bourg	37	32	1
	Pablo Nogués	6	5	1
	Los Polvorines	14	12	1
	Villa de Mayo	9	8	1
	A. Sourdeaux	26	23	1
	Total	115	100	

to nitrite and its subsequent reaction with an aromatic amine to form a diazonium salt, which in turn, reacts with N-(1-naphthyl)-ethylene-diamine to form a red-violet azo dye.

Survey design and data collection

A questionnaire survey was designed using the Google Formulary tool. The owners of the shops where the samples were taken were invited to answer it. In addition to this, the students who were part of the project also distributed it among their contacts.

First of all, people were asked about their bottled water consumption and why they prefer or not to consume it. The rest of the questions were intended to investigate their perceptions of bottled water and how it is considered in comparison to tap water. People were also asked about the reasons that guided their choice of a certain product and knowledge of its characteristics. Age, level of education and place of residence were also collected by the survey (Supplementary material 1, Survey formulary).

RESULTS

Nitrate concentration in bottled waters

A total of 100 samples from 36 different brands were taken at C.A.B.A. They were identified as brand 1C to 36C. None of them exceeded the nitrate value required by legislation (Figure 2). The highest value obtained was 38 mg/L for brand 16C. The samples of brand 12C and 27C yielded results of 23 mg/L and 19 mg/L, respectively. The three brands came from bottling companies located in municipalities in the neighboring Greater Buenos Aires: brand 12C from Morón, 16C from Lanús, and 27C from La Matanza (Figure 2). All the other brands had a nitrate concentration below 12 mg/L and, in most cases, below the detection limit of 5 mg/L (Supplementary material 2, Nitrate concentration of water samples).

The situation was totally different in the municipality of Malvinas Argentinas, where 100 samples were also analyzed. Of a total of 41 brands (identified as 1MA to 41MA) there were 14 that exceeded the 45 mg/L limit value. That represents 34.1% of brands that did not comply with the legislation. Six of them (14.6% of the total) had a nitrate concentration that exceeded 100 mg/L (Figure 2).

For four other brands there were some bottles that had nitrate concentrations below the limit, and bottles that exceeded it. In all cases the differences were substantial and could not be attributed to measurement uncertainty. In brand 6MA, one bottle was below 5 mg/L and another had a nitrate concentration of 87 mg/L. The same extreme differences were found for brands 6MA (<5–87), 22MA (<5–173), 23MA (9–91), and 34MA (<5–87). Brand 16MA and brand 17MA also showed an important variation between bottles, but at values close to or above the limit: brand 16MA was between 42 and 180 mg/L and brand 17MA between 81 and 115 mg/L.

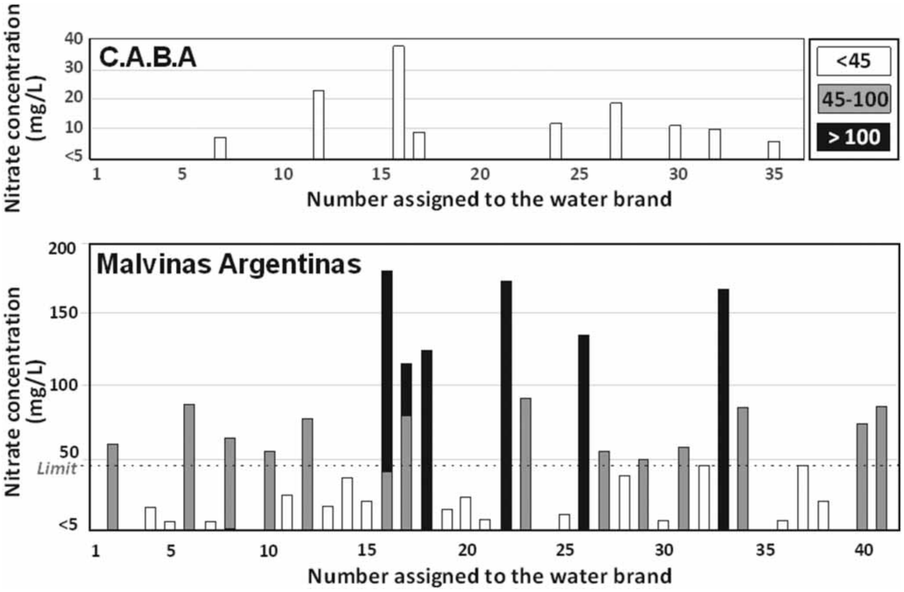


Figure 2 | Nitrate concentration in the different bottled water brands analyzed in C.A.B.A. and Malvinas Argentinas. Each water brand was identified with a number and a letter: 1C to 36C in C.A.B.A.; 1MA to 41MA in Malvinas Argentinas.

Consumption habits and public perception of water quality

A total of 364 people answered the survey. The results show that a large majority of the population consumes bottled water: 71.7% of the participants consume it while only 28.3% do not. The opinion that it is better than tap water is shared by 82.4% of the respondents. A summary of the results is shown in Figure 3 (Supplementary material 3, Survey data).

Among people who do not consume bottled water, the majority do so because they have access to tap water (60.2%). The other principal reasons mentioned are: the high price of bottled water (14.6%), its lack of quality (8.7%) or the fact of having a water purifier at home (1.9%).

The reasons for choosing bottled water are mainly because it is considered healthier (38.7%). Almost to the same extent, people say it is out of habit (37.5%) and, to a lesser extent, out of necessity (23.8%). People who consume bottled water (261 persons) valued especially its taste (34.4%). Other valued characteristics were mainly safety (27.0%) and purity (25.6%). It is remarkable that most people did not know the origin of the bottled water they consume. Also, practically half of the respondents indicated that they did not read the label on the product.

There were 57.7% of respondents residing in C.A.B.A. and 42.3% in the PBA. Consumption data of bottled water were very similar when discriminated by geographical location. Among the people residing in C.A.B.A., the percentage that consumes bottled water was 71.9% and 71.4% in the PBA. Similarly, the perception that bottled water is better is quite similar: 79.5 in C.A.B.A.; 86.0% in PBA.

The only differences that can be pointed out between the two districts are the reasons for the consumption of bottled water and the most valued characteristics of the product. In C.A.B.A., the main reason for consuming bottled water has to do with the habit (44.4%) and then for health reasons (34.4%). In the PBA, the order is reversed, health reasons are in first place (44.5%) and habit in second (28.2%). Also, the most valued characteristics for bottled water that stand out are different in the two jurisdictions. In C.A.B.A., the order is taste (36.7% of the responses), followed by purity (25.0%) and safety (20.8%). In the PBA, on the other hand, safety was the predominant reason with 34.7% of the responses, followed by taste (31.6%) and purity (26.3%).

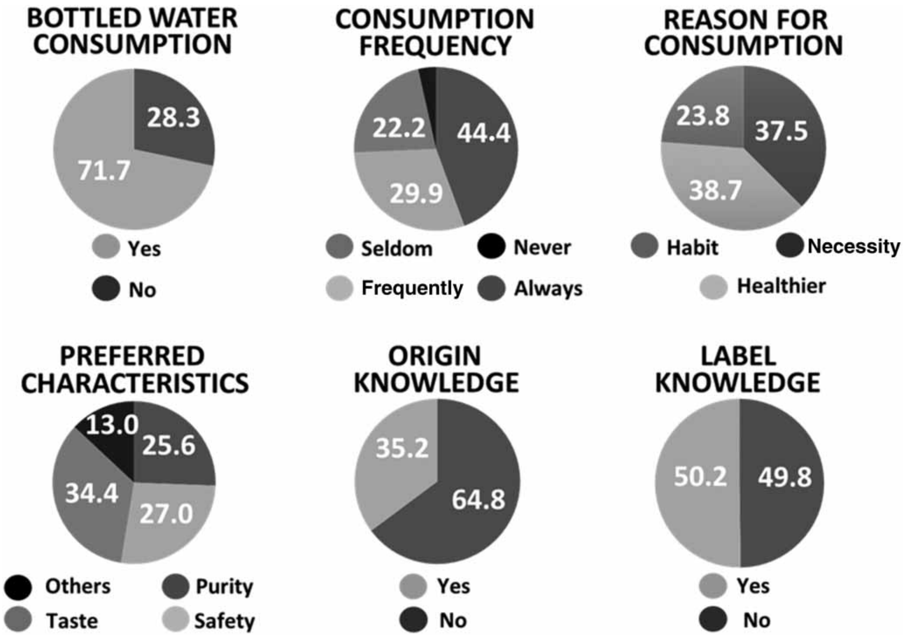


Figure 3 | Analysis of the survey about consumption habits and public perception of water quality.

DISCUSSION

It is very common in Argentina to have 20 L water dispensers for consumption, both at home or in shops, so these large water bottles were chosen to be tested. Commercial venues instead of residences were selected to take the samples due to easier access. It must be considered that the same bottled water brands are also consumed by people in their homes. This was corroborated in the field work, particularly in Malvinas Argentinas, where owners of the shops lived nearby and bought the water for their homes from the same provider. There are many different small bottlers that distribute the containers, often only in a given municipality or even in a small section of it. Each municipality is responsible for product control unless the brand is approved for sale throughout the country. Registration and control requirements in these cases are more rigorous (ANMAT 2020).

C.A.B.A. and Malvinas Argentinas were selected for the study because they have the most extreme values in water access: in C.A.B.A. almost everyone has access to safe water, in Malvinas Argentinas almost no one. This extreme inequality is even more remarkable considering that both districts are separated by a short 45-minute local train journey within the same urban area.

Drinking water in the urban area of Buenos Aires is provided by a state company, AySA. In most of the network the water comes from the La Plata river, while in the peripheral areas there are still underground water wells in service. The most remote areas, such as Malvinas Argentinas, are not even reached by the network and people get water from their own wells, without any quality control.

Groundwater nitrate contamination is very common in the area (Heredia *et al.* 2000). In fact, frequent violations of the maximum nitrate levels were the reason for the nationalization of Aguas Argentinas, the private company that previously provided water service, through Decree 303/2006. On the other hand, nitrate level in La Plata river is low, below 10 mg/L (Giannuzzi *et al.* 2012).

Given the widespread presence of nitrate contamination in wells, it is highly probable that, since bottled water can also come from a ground source, it may also be contaminated. Of course, there are treatments to reduce nitrate, but if these treatments do not exist or are not properly carried out, contamination will appear.

Analysis of the obtained results demonstrates a completely different situation in the two areas under study. In C.A.B.A., no sample exceeded the nitrate limit value. Only three samples have slightly high nitrate values, typical of waters that come from an underground source. On the other hand, in Malvinas Argentinas, 34.1% of the brands do not comply with the legislation. This value reaches 43.9% if the 9.8% of brands for which some but not all of its bottles exceeded the limit is added to the sum.

The fact that different bottles of the same brand have different degrees of contamination can be explained by the progressive saturation of the treatment system. In cases where these differences are extreme, the adulteration of the product cannot be ruled out. In a previous study (Gallego 2006), a single contaminated sample of a first-line brand was detected, for which ten other analyzed containers showed an undetectable value.

Six of the samples presented concentrations higher than 100 mg/L. The highest values obtained were for 16MA: 180 mg/L; for 17MA: 115 mg/L; for 18MA: 127 mg/L; for 22MA: 173 mg/L, for 26MA: 131 mg/L, and for 33MA: 167 mg/L. These are extremely high nitrate concentrations, that could pose a high health risk, particularly for babies (Greer & Shannon 2005; Fossen Johnson 2019). It is very likely that, lacking access to safe water, people use the bottled water they buy for those uses that are considered more critical. Among them is, undoubtedly, the preparation of a baby's bottle.

In a previous work we found a high percentage of nitrate-contaminated water in the PBA (Gallego 2006). However, its presence in bottled water has not been frequently reported in the literature. In Italy, in a study carried out on 37 samples of bottled water, an average value for nitrate of 6 mg/L was obtained (Cidu *et al.* 2011). Weyer *et al.* (2014) in the United States, analyzed 261 samples of bottled water from Texas and Iowa and did not obtain any value above 26.6 mg/L. Results obtained in developing countries, although somewhat worse, are also not comparable to those obtained in this study. Alimohammadi *et al.* (2018) found only a sample for a total of 71 tested that exceeded the limit, with a value of 50.1 mg/L. Other studies carried out in Chile on ten samples (Daniele *et al.* 2019), in Iran, on 21 samples (Moazeni *et al.* 2014), and in Ghana, on 118 samples (Dzodzomenyo *et al.* 2018) did not report any sample above the values required by legislation.

There may be two reasons for these differences. One is that the water source in the places where the studies were carried out does not present nitrate contamination. This may be an explanation for arid regions, where the presence of individual wells is probably not as frequent, but not for the United States, for example, where wells exist and nitrate concentration of groundwater is also a problem (Greer & Shannon 2005). The second explanation is that, despite this contamination, there are stricter controls on the bottled water manufacturing facilities that prevent access of contaminated water to the market. The latter seems to be the reason for the differences between the two districts in this study as well. No sample in C.A.B.A. showed nitrate contamination, although some of the brands detected came from the PBA. It should be highlighted that C.A.B.A. has laboratories for bromatological analysis of international level, while few municipalities in the PBA have such infrastructure.

The survey carried out shows an even higher consumption pattern for bottled water than in other countries: 71.7% of the respondents consume the product and 44.4% of them say they do so continuously. The highest percentages reported in the literature are 54% for Barcelona (Font-Rivera *et al.* 2017), 42% for France (IFEN 2000), 43% in Quebec (Levallois *et al.* 1999), 37% in Appalachia, USA (Levêque & Burns 2017). It must be said that a quantitative comparison is not possible, since the number of cases in this study is low and there was no random selection of participants, so the presence of some type of bias cannot be ruled out. However, these numbers undeniably show a relatively high consumption of the product.

The reasons for the choice and the most valued characteristics of bottled water are similar to those reported in other countries, and have already been presented in the Introduction. But there is a difference to highlight in the results between the two districts, which seems to be related to the lack of access to water in the PBA. The reason for consumption in C.A.B.A. is predominantly a matter of habit and the most appreciated characteristic is taste, while in the PBA, the reason is that it is healthier and the main characteristic sought for is safety.

Despite the high consumption of bottled water and the perception of the respondents about its quality, 64.6% do not know its origin, while another 50% admit that they do not consult the information on the labels when purchasing water for consumption. This acquires great relevance if it is taken into account that the Argentine Food Code

(ANMAT 2020) in its article 983 establishes that bottled drinking water 'is that which is marketed packaged in bottles, or other suitable containers, provided with regulatory labeling, and which origin may be from an underground or from a public supply'. Therefore, the quality of the bottled water is strongly conditioned by its origin and by the responsibility of the bottler. In any case, the existence of a label is of little use if the supervisory authority does not efficiently control that it really corresponds to the product.

CONCLUSIONS

Nitrate is a major contaminant in water, one of the few capable of causing acute toxicity problems. But, in addition to this, nitrate could work as an indicator of bottled water quality. Since it is very easy to determine and its presence is an indicator of inadequate treatment for water, it can be a hint about the presence of other contaminants.

It is worrying that precisely in the area where access to water is a problem and bottled water is purchased for safety reasons, the quality is worse. Nitrate contamination of bottled water was much higher in the area with the least access to drinking water. The only reason that seems plausible to explain this, is that, just as there is no water network in the area, there are also no adequate controls on the product.

York *et al.* (2011) said that the motives leading to bottled water consumption are difficult to understand, but that it appears to be a choice reflecting taste and lifestyle. As an example, water consumption has increased globally, especially in western countries (Quian 2018) paradoxically, where the best tap water quality exists. In the present work, it seems to be clear that the forces that govern the consumption of bottled water in one or other case are different. People who lack access to safe water only have that option.

What characterizes developing countries is not the lack of advances present in the developed world, but inequality in their access: advances exist, but they are not for everyone. C.A.B.A. has a water quality and control standards for its bottled water similar to those of the first world. Only 45 minutes away, drinking water can be a dangerous experience, even if you have paid for what was supposed to be a human right.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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