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Quality of bottled water consumed in the state of São Paulo: analysis of inorganic substances

Qualidade da água envasada consumida no estado de São Paulo: análise de substâncias inorgânicas

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ABSTRACT

Introduction: The availability of different brands of bottled water on the national market, combined with the growing consumption by the population, makes it important to assess the quality of its chemical composition. **Objective:** To quantify inorganic chemical substances (nutrients and toxic metals) in samples of bottled water from national and imported sources consumed in the State of São Paulo, in compliance with legislation and compendiums; verify the parameters of inorganic elements indicated on labeling, as well as the recommended daily intake. **Method:** Determinations were carried out by inductively coupled plasma mass spectrometry. **Results:** The evaluation indicated that 93.5% of the total of the 107 samples analyzed presented concentrations of inorganic chemical elements in accordance with the maximum values allowed in national legislation for all analytes (96.5% for national and 81.0% for imported), thus being considered suitable for human consumption. Discordant results were obtained for 6.5% for the elements chromium (2.3%) and selenium (1.2%) in national water and chromium (19.0%) in imported water. Levels above the recommended limits by the international compendia for sodium intake were quantified in an imported water sample that could cause illness in the body. Furthermore, the variability observed between the experimental results and values declared in the chemical composition tables (labeling) can lead consumers to purchase a product containing inadequate information, thereby harming their daily intake needs. **Conclusions:** The results highlight the importance of establishing and maintaining continuous monitoring programs in the country to monitor the sanitary quality of food consumed by the population and thus protect population's health.

KEYWORDS: Bottled Water; Inorganic Chemicals; Water Quality Control; ICP-MS

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RESUMO

Introdução: A disponibilidade de diferentes marcas de água envasada no mercado nacional, aliada ao crescente consumo pela população, acarreta a importância em avaliar a qualidade de sua composição química. **Objetivo:** Quantificar substâncias químicas inorgânicas (nutrientes e metais tóxicos) em amostras de água envasada de procedências nacional e importada, consumidas no estado de São Paulo, em atendimento à legislação e aos compêndios; verificar os parâmetros de elementos inorgânicos apontados em rotulagem, bem como a ingestão diária recomendada. **Método:** As determinações foram realizadas por espectrometria de massas por plasma indutivamente acoplado. **Resultados:** A avaliação indicou que 93,5% do total de 107 amostras analisadas apresentou concentrações de elementos químicos inorgânicos de acordo com os valores máximos permitidos em legislação nacional para todos os analitos (96,5% para nacional e 81,0% para importada), sendo consideradas adequadas para o consumo humano. Resultados discordantes foram obtidos para 6,5% para os elementos cromo (2,3%) e selênio (1,2%) para água nacional, e cromo (19,0%) para água importada. Níveis acima dos limites recomendados por compêndios internacionais para a ingestão de sódio foi quantificado em amostra de água importada, podendo acarretar enfermidades ao organismo. Além disso, a variabilidade verificada entre os resultados experimentais e valores declarados nas tabelas de composição química (rotulagem) pode conduzir o consumidor a adquirir produto contendo informação inadequada, acarretando prejuízo à sua necessidade diária de ingestão. **Conclusões:** Os resultados gerados apontam para a importância do estabelecimento e da manutenção de programas de monitoramento contínuos no país, a fim de fiscalizar a qualidade sanitária de alimentos consumidos pela população e, assim, proteger a saúde da população.

PALAVRAS-CHAVE: Água Envasada; Substâncias Químicas Inorgânicas; Controle de Qualidade da Água; ICP-MS

INTRODUCTION

Water is characterized as a fundamental element for the body's health due to the presence of mineral salts in its chemical composition, which are essential for maintaining the proper balance of various biochemical processes and physiological mechanisms in the body. Its consumption is extremely significant in terms of hydration, regulating the transportation of nutrients that are easily absorbed by the gastrointestinal tract, and stimulating intestinal transit[1,2]. Low intake can lead to health problems, with children being particularly vulnerable[3].

However, the presence of toxic inorganic chemical substances in drinking water can have deleterious effects on health, due to their high toxicity and lack of known biological function in the body, causing acute or chronic intoxication and gastrointestinal, renal, central nervous system, and vascular disorders, as well as carcinogenicity, among others[4,5]. Fetal and early childhood exposure to these contaminants can lead to neurological problems and developmental disabilities with permanent consequences, due to the fragility of the immune system[6].

In recent decades, anthropogenic factors related to urbanization, industrialization, and economic development have resulted in the increasing generation and discharge of pollutants into water bodies (industrial effluents, sewage, illegal waste disposal, leaking landfill leachate), contaminating aquifers with toxic chemicals and thus making them unfit for consumption[7,8].

Consequently, the current concern about the quality of the water consumed, due to possible water pollution, lack of basic sanitation policies, variations in socio-economic standards, age group characteristics, race/ethnicity, marketing strategies, among others, are reasons that lead a large part of the world's population to consume bottled water, as one of the alternatives in the prevention of diseases related to the use of public water supplies, as well as a nutritional supplement that integrates the diet[9,10,11].

Following the global trend[9,10,11] and considering the increasing consumption of bottled water by the Brazilian population[12,13], it is important to establish and implement health surveillance monitoring programs to evaluate its quality in relation to the maximum permitted values (MPV) in legislation[14,15], in order to ensure that the competent government bodies can take action when inadequate results are obtained[16].

In view of the above, the objectives of this study were: to assess the levels of inorganic chemical substances (Ag, Al, As, Ba, Be, Ca, Cd, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Tl, and Zn) in samples of bottled water sold in 19 municipalities in the state of São Paulo, in compliance with the requirements established by legislation; analyze compliance with the chemical composition by comparing the concentrations of the elements with the values declared on the label; and assess the recommended daily intake.

METHOD

Samples

A total of 107 samples of bottled water from different brands and batches, sealed and packaged in polyethylene terephthalate (PET), sachet, or glass containers with a capacity of 200 to 800 mL, were evaluated between 2018 and 2022.

The samples were collected by professionals from the Health Surveillance Groups in the state of São Paulo, in compliance with the Programa Paulista (São Paulo Program), distributed as follows: national origin (40 packs of non-carbonated bottled water and one pack of artificially carbonated bottled water) and imported origin (one non-carbonated sample). And, to complement the research, the sample also included private purchases of the product not covered by the Program, distributed as follows: national origin (39 with non-carbonated composition and six artificially carbonated samples) and imported (nine with non-carbonated composition and 11 naturally carbonated).

The purchase was made at different retail outlets in 19 municipalities located in the state, which include: the São Paulo Metropolitan mesoregions (São Paulo and the micro-regions of São Paulo, Mogi das Cruzes, Osasco, Santos, and Guarulhos); Litoral Sul Paulista (Registro and Itanhaém micro-regions); Vale do Paraíba Paulista (São José dos Campos micro-region); Campinas (Campinas micro-region), and Macro Metropolitana Paulista (Jundiaí micro-region).

The sample set consisted of:

Bottled *water of national origin*: 86 bottles of water distributed in natural still water (75); natural artificially carbonated with added carbon dioxide gas (seven); added salts still water (three); and added salts artificially carbonated (one). The salt-added water samples were produced from deep abstraction, followed by purification treatments using chlorination, reverse osmosis, vaporization, condensation, filtration, and ozonation, with the subsequent addition of food-grade salts permitted by law[14,15]. The natural water samples came directly from sources located in the states of São Paulo, Minas Gerais, Paraná, Bahia, Rio de Janeiro, and Rio Grande do Sul.

Bottled *water from imported sources*: 21 bottles of natural mineral water, distributed among 11 different brands, labelled non-carbonated (10) and naturally carbonated (11). The samples came directly from natural sources in the following countries: France, the United States, Norway, Portugal, Spain, Mexico, and Italy.

The containers were opened at the time of preparation and acidified with ultrapure nitric acid to a final concentration of 0.2% (v/v)[17] and homogenized, without any pre-treatment. Samples containing gas were degassed prior to acidification in the containers themselves.

Analytical Instrumentation

Determinations of metals and semi-metals in bottled water were carried out using an inductively coupled plasma mass spectrometer/ICP-MS (model Elan DRC II, Perkin Elmer), operated with ultra-high purity (UHP) argon gas (grade 5.0, 99.999%), in standard mode.

Reagents and materials

The analytical procedure for preparing solutions and handling samples was carried out using ultrapure water (resistivity 18.2 MΩ cm) obtained from the Milli-Q® purification system (Integral 10 model, Merck Millipore) and ultrapure nitric acid (HNO_3 65%, Suprapur, Merck), and performed in an ISO class 5 laminar flow unit, installed in an environment with ISO class 7 particulate matter control (clean area).

Standard solutions of the analytes under study Al, Ag, As, Ba, Be, Ca, Cd, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Tl, and Zn (Inorganic Ventures and NSI Lab Solutions) and the internal standards Ge, In, Re, and Sc (Sigma-Aldrich) were diluted for use according to the analytical methodology employed.

Analytical methodology

The analytical methodology was established in accordance with the recommendations described in EPA method 200.8[17]. All the tests were accredited by the General Coordination for Accreditation of the National Institute of Metrology, Quality and Technology (CGCRE/Inmetro)[18].

The concentration ranges of the inorganic chemicals used in the analytical curve prepared in HNO_3 solution to a final concentration of 0.2% (v/v)[(17)] for the analytes under study were: 0.0004-0.02 mg L^{-1} (Be); 0.1-4.0 mg L^{-1} (Na, Mg, K, Ca); 0.02-1.0 mg L^{-1}) (Ba, Cu, Mn); 0.001-0.05 mg L^{-1} (Ag, Al, As, Cr, Ni, Pb, Se, Tl, Zn); and 0.0005-0.025 mg L^{-1} (Cd).

Accuracy was assessed by analyzing NIST SRM 1643f *Trace elements in water* and NIST SRM 1640a *Trace elements in natural water* certified reference materials; and external quality control was carried out through the laboratory's periodic participation in national interlaboratory programs for the determination of metals and semi-metals in drinking water: Proficiency Testing Program/Metals in drinking water matrix and in raw water (PEP TOQ - Sabesp), Proficiency Testing Program in water/Measurement of metals in water (Inmetro), and Interlaboratory Program in Treated Water (Instituto Senai de Tecnologia - PEP-IST Ambiental).

RESULTS

In the course of the research, new legislation came into force for bottled water[15]. The MPVs remained unchanged when compared to the repealed legislation[14], making it possible to infer the same correspondence for the levels of inorganic chemical substances.

Table 1 shows the compliance of the quality standard for the chemical substances As, Ba, Cd, Cr, Cu, Mn, Ni, Pb, and Se and the nutrients Ca, K, Mg, and Na (provided for in the legislation only for water with added salts) in different types of bottled water in compliance with the legislation[14,15]. Analytes not recommended by legislation[14,15] were also analyzed: (i) Ag, Al, Be, Tl, and Zn for water samples with or without added salts, (ii) and the Ca, K, Mg, and Na group of elements for water without added salts.

Table 2 shows the statistical evaluation carried out, represented by the median and interquartile range (IQR) of the concentrations obtained for the inorganic chemical substances, considering the types of water analyzed. Since the data did not show a normal distribution, they were treated as non-parametric. The evaluation was carried out using the median and IQR, considering the analytes for which more than 50% of the results were quantified in a given type of water.

In order to verify the quality of bottled water consumed in the state of São Paulo, the maximum concentrations obtained for the inorganic elements, which may exceed the limits recommended by legislation, were compared with values suggested by international compendia (regulations and bodies), shown in Table 3. In addition, the results were also compared with concentration values reported in scientific literature, as shown in Table 4.

DISCUSSION

Legislation and compendia

The general overview of the evaluation of inorganic substances in all the bottled water samples from national and imported sources (Table 1) showed that, of the total of 107 analyzed, 93.5% had concentrations of inorganic chemical elements in accordance with the MPVs in national legislation[14,15] for this category of product.

Table 1. Limit of quantification of the method and minimum and maximum values of the concentration of inorganic chemical substances, considering the types of water analyzed, expressed in mg L^{-1}.

Analito	LQ	National still water (N = 75)		National sparkling water (N = 7)		National added salts (N = 4)		Imported still water (N = 10)		Imported sparkling water (N = 11)	
		[Minimum]	[Maximum]	[Minimum]	[Maximum]	[Minimum]	[Maximum]	[Minimum]	[Maximum]	[Minimum]	[Maximum]
Cr	0.0010	NQ (43)*	0.0160	0.007	0.0730	NQ (75)*	0.0720	NQ (70)*	0.035	0.016	0.0760
Mn	0.0250	NQ (73)*	0.0300	NQ (71)*	0.0260	NQ (75)*	0.0260	NQ (90)*	0.027	NQ (73)*	0.0410
Ni	0.0010	NQ (92)*	0.0020	NQ (100)*		NQ (50)*	0.0040	NQ (70)*	0.005	NQ (45)*	0.0070
Cu	0.0020	NQ (85)*	0.0260	NQ (71)*	0.0260	NQ (50)*	0.0080	NQ (90)*	0.005	NQ (91)*	0.0650
The	0.0010	NQ (80)*	0.0080	NQ (86)*	0.0020	NQ (100)*		NQ (80)*	0.005	NQ (55)*	0.0090
If	0.0010	NQ (91)*	0.0370	NQ (86)*	0.0020	NQ (100)*		NQ (80)*	0.002	NQ (73)*	0.0060
Ba	0.0050	NQ (28)*	0.4500	NQ (14)*	0.1800	NQ (75)*	0.0080	NQ (60)*	0.120	NQ (73)*	0.3100
Be	0.0004	NQ (68)*	0.0032	NQ (86)*	0.0013	NQ (75)*	0.0014	NQ (100)*		NQ (73)*	0.0010
Al	0.0020	NQ (68)*	0.0550	NQ (71)*	0.0100	NQ (100)*		NQ (80)*	0.005	0.004	0.0410
Zn	0.0010	NQ (76)*	0.0160	NQ (86)*	0.0090	NQ (100)*		NQ (100)*		NQ (100)*	
Ag	0.0020	NQ (89)*	0.0040	NQ (86)*	0.0020	NQ (100)*		NQ (50)*	NQ (100)*	NQ (100)*	
Cd	0.0005	NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*	
Pb	0.0010	NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*	
Tl	0.0010	NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*		NQ (100)*	
In	0.0230	0.2	116	0.4	119	0.1	17	2	51	10	1.351
K	0.0300	NQ (3)*	40	0.2	3	2	65	NQ (30)*	1	0.2	50
Ca	0.1200	NQ (4)*	37	0.2	18	3	86	0.8	122	3	178
Mg	0.02	NQ (89)*	19	NQ (14)*	10	1	40	0.1	32	0.2	58

Source: Prepared by the authors, 2023.
* The number in parenthesis corresponds to the percentage of samples that obtained an NQ (not quantified) result for a given analyte.

Table 2. Median and interquartile range (IQR) of the concentrations obtained for the inorganic chemical substances, considering the types of water analyzed, expressed in mg L^{-1}.

Analito	National still water	National sparkling water	Imported still water	Imported sparkling water	Water with added salts
	Median [IQR]				
Cr	0.001 [<LQ*-0.004]	0.017 [0.011-0.038]	**	0.044 [0.025-0.066]	**
Ni	**	**	**	0.003 [<LQ-0.005]	0.002 [<LQ*-0.004]
Cu	**	**	**	**	0.004 [<LQ*-0.008]
Ba	0.032 [<LQ*-0.068]	0.044 [0.031-0.138]	**	**	**
Al	**	**	**	0.012 [0.005-0.024]	**
Ag	**	**	0.002 [<LQ*-0.004]	**	**
In	7.760 [1.69-17.07]	12.20 [1.20-17.93]	6.880 [5.05-8.87]	191.550 [34.99-645.9]	3.850 [0.15-10.05]
K	1.420 [0.90-1.95]	0.93 [0.52-2.04]	0.650 [0.13-0.86]	2.410 [0.43-28.07]	30.910 [2.85-60.33]
Ca	8.930 [1.89-18.01]	6.50 [5.53-17.18]	16.920 [0.96-32.45]	109.080 [15.43-157.41]	42.880 [3.24-83.29]
Mg	**	2.22 [0.60-5.57]	2.990 [1.42-6.73]	3.110 [2.51-23.94]	19.500 [1.57-37.90]

Source: Prepared by the authors, 2023.
*LQ: Limit of quantification of the method.
** Blank spaces indicate concentrations below the LQ in more than 50% of the results.

When we consider only the group of bottled water of national origin (86), we found that 96.5% of the results were in line with the legislation[14,15], with levels below the MPVs for the elements As, Ba, Ca, Cd, Cu, K, Mg, Mn, Na, Ni, and Pb. As for the group of imported origin made up of 21 samples, 81.0% of the analyses complied with the legislation[14,15] for As, Ba, Ca, Cd, Cu, K, Mg, Mn, Ni, Pb, and Se. In this way, these samples can be classified as suitable for human consumption with regard to the elements analyzed. elements analyzed.

Table 3. Maximum levels of inorganic chemical substances obtained from national and imported bottled water samples consumed in the state of São Paulo, and values referenced in international compendia (organizations and regulations), expressed in mg L^{-1}.

Reference	Analito																	
	Cr	Mn	Ni	Cu	The	If	Cd	Ba	Pb	Be	Al	Zn	Ag	Tl	In	Mg	K	Ca
National*	0.073	0.030	0.004	0.026	0.008	0.037	0.0005	0.447	0.001	0.0032	0.055	0.016	0.004	0.001	119.46	39.55	65.25	86.05
Imported*	0.076	0.041	0.007	0.065	0.009	0.006	0.0005	0.313	0.001	0.001	0.041	0.001	0.017	0.001	1.351.00	58.48	50.13	178.30
RDC[14,15]	0.050	0.500	0.020	1.000	0.010	0.010	0.003	0.700	0.010	-	-	-	-	-	600	65.00	500.00	250.00
WHO[19]	0.050	0.400	0.070	2.000	0.010	0.040	0.003	0.700	0.010	-	-	-	-	-	-	-	-	-
EPA[20]	0.100	-	-	1.300	0.010	0.050	0.005	2.000	0.015	0.004	-	-	-	0.002	-	-	-	-
FDA[21]	0.100	0.050	0.100	1.000	0.010	0.050	0.005	2.000	0.005	0.004	0.200	5.000	0.100	0.002	-	-	-	-
England[22]	0.050	0.050	0.020	2.000	0.010	0.010	0.005	1.000	0.010	-	0.200	-	-	-	200	-	-	-
Bulgaria[23]	0.050	0.050	0.020	2.000	0.010	0.010	0.005	-	0.010	-	0.200	4.000	-	-	200	80.00	-	150.00
Spain[24]	0.050	0.050	0.020	2.000	0.010	0.010	0.005	1.000	0.010	-	0.200	-	-	-	200	-	-	-
Turkey[25]	0.050	0.500	0.020	1.000	0.010	0.010	0.003	1.000	0.010	-	0.200	-	-	-	200	50.00	-	150.00
Chile[26]	0.050	2.000	-	1.000	0.010	0.010	0.010	1.000	0.050	-	-	5.000	-	-	-	-	-	-
Korea[27]	0.050	0.050	-	1.000	0.010	0.010	0.005	-	0.010	-	0.200	3.000	-	-	-	-	-	-
Romania[28]	0.050	0.050	20.000	0.100	0.010	0.010	0.005	-	0.010	-	0.200	5.000	-	-	200	-	-	-
Italy[29]	0.050	0.500	20.000	1.000	0.010	0.010	0.003	1.000	0.010	-	-	-	-	-	-	-	-	-
Mexico[30]	0.050	0.400	0.020	1.000	0.010	0.010	0.003	0.700	0.010	-	-	-	-	-	-	-	-	-
Slovenia[31]	0.050	0.050	0.020	2.000	0.010	0.010	0.005	-	-	-	0.200	-	-	-	200	-	-	-

Source: Prepared by the authors, 2023.
RDC: Resolution of the Collegiate Board (national legislation); WHO: World Health Organization; EPA: United States of America Environmental Protection Agency; FDA: U. S. Food and Drug Administration.
* Maximum concentration values of nutrients and toxic metals obtained experimentally in this study.

Table 4. Parameters of inorganic chemical substances in bottled water available in the literature.

Literature	Elements		Reference
	Suitable	Inadequate	
Iran[2]	Ca, Cu, Mg, Mn, Na, Zn	-	EPA
Chile[32]	As (70%), Ba, Ca, Cd, Cr, Cu, K, Mn, Mg, Na, Ni, Pb, Se	As (30%)	Chile Regulation
Bulgaria[33]	Al, Ag, As, Ba, Be, Ca, Cd, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Tl, Zn	-	WHO, EPA, Regulation n° 9
Slovenia[34]	Al, As, Ba, Ca, Cd, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Zn	-	Slovenian Regulation, EC, EPA, WHO
Nigeria[35]	Ca, Cd, Cr, Cu, K, Mn, Na, Pb, Zn	-	WHO
China[36]	Al, As, Ba, Cr, Cu, Mn, Ni, Pb, Zn	-	WHO, GB 5749-2006
Norway[37]	Al, Ca, Cu, Mg, Mn, Na	-	WHO
Philippines[38]	As, Cd, Cr, Pb	-	WHO
Romania[6]	Ba, Cd, Cr, Cu, Mn, Ni, Pb, Zn	-	Law 311/2004, Directive EU 2020/2184
Nepal[39]	Ag, Ca, Cd, Cu, K, Mg, Na, Ni, Pb, Zn	-	IBWA
Italy[40]	As, Cd, Cr, Pb, Ni	-	Italian Legislation
Rwanda[41]	Al, As, Ba, Ca, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Zn	-	WHO, Rwanda Standard Board
Morocco[42]	Al, As (88%), Cd, Cr, Ni, Pb	As (12%)	WHO, Moroccan Food Water Standards
Croatia[43]	Al, As, Ba, Cd, Cr, Cu, K, Mn, Na, Pb, Zn	-	WHO, Croatian Regulations
Brazil[44]	Al, As, Ba, Ca, Cd, Cr, Cu, K, Mg, Mn, Na, Ni, Pb, Se, Zn	-	RDC no. 274/2005
Brazil[45]	Ca, K, Mg, Na	-	RDC no. 274/2005

Source: Prepared by the authors, 2023.
EPA: United States of America Environmental Protection Agency; WHO: World Health Organization; EC: EU Drinking Water Directive; GB 5749-2006: Guide values established by the Ministry of Health of the People's Republic of China; EU: European Union; IBWA: International Bottled Water Association; RDC: Resolution of the Collegiate Board.

These results indicate: the absence of water pollution deriving from environmental contamination of natural occurrence (geological area) or by anthropogenic action in natural water catchment sources; the absence of migration of inorganic chemical substances from packaging to the product; the use of food-grade salts with proven purity for water of the type added with salts, among others; prove the adequate quality of the product offered on the market.

On the other hand, 6.5% of the total of 107 samples evaluated indicated inadequate results for human consumption in relation to the Cr and Se parameters.

Excessive intake of inorganic chemical elements can cause potential adverse health effects. Hexavalent Cr (VI) has been associated with the incidence of lung cancer, as well as diseases related to the liver, kidneys, gastrointestinal tract, and circulatory systems, among others[46]. The effects of Se toxicity can lead to health problems such as muscle tremors, fragility and hair/nail loss, neurological and gastrointestinal disorders[47].

Concentration values above the MPVs set by the legislation[14,15] were achieved for the set of bottled water of Brazilian origin in two samples for the Cr element, both artificially carbonated. One was produced from purified water, followed by chlorination, reverse osmosis, vaporization, condensation, filtration, and ozonation, plus the addition of calcium ($CaCl_2$), magnesium ($MgCl_2$), and potassium ($KHCO_3$) salts; and the other was extracted from a source located in the state of São Paulo. Se was quantified in a still sample obtained from a natural source located in the state of Rio Grande do Sul.

In Brazil, high levels of Cr, identified in water supply sources and river basins in the Southeast, may be associated with natural occurrence in the soil, erosion, leaching, industrial and agricultural activities, and the discharge of urban effluents[48,49,50,51]. On the other hand, the element Se was quantified in water and fish, as a result of environmental and ecological factors that regulate the distribution of the element in the southern region of the country, which may lead to the enrichment of water by this substance[52,53].

In addition to the geology of the source, other factors can affect the conformity of the chemical composition of bottled water when subjected to industrialized processes in salt-added products. The reasons may be associated with: the production and filling process, the type of water used which does not meet the water potability standard established by the Ministry of Health, failure to decontaminate machinery, the inferior quality of the food-grade salts used in the addition process permitted by law[14,15], among others[38,54]. In addition, considering that one of the samples was obtained through an industrial process using reverse osmosis, the occurrence of Cr in disagreement with the legislation can be attributed to the industrialization process[38,54].

In general, the low percentage of inadequate results achieved for the elements Cr (2.3%) and Se (1.2%) can be classified as one-off events and should not be indicative of evidence of contamination characteristic of natural actions or anthropogenic activities, and so these results can also be in line with the literature (Table 4).

As for the imported group of 21 samples, 19.0% exceeded the limits of national legislation[14,15] for Cr. The higher concentration values corresponded to four sparkling samples produced by countries located on the European continent: Spain (natural source), Italy (natural source), France (natural source), and Norway (described on the label as having added salts).

In Europe, the occurrence of Cr among other elements (Pb, Zn, Cu, and Ni), whether from natural origin in soil, water, and groundwater, or from anthropogenic industrialization activities, is an emerging public health problem in countries located on this continent, which can affect food production, water supply, and consequently the human health of the exposed population[55,56,57,58].

Labeling (nutritional information)

The chemical composition table described in nutritional labeling is one of the fundamental requirements for complying with the legislative parameters for packaged food products offered on the market, as well as being an instrument for promoting public health and guaranteeing consumers' rights to choose.

The legislation for inorganic substances in bottled water for labeling describes the obligation to state "contains sodium" when the product contains more than 200 mg/L of the substance[14,15]. And in addition to this category of product, Ordinance No. 470 of November 25, 1999, which establishes the basic characteristics of the labels on mineral and table drinking water packages, recommends that the label should include, among other information, the "chemical composition, expressed in milligrams per liter, containing at least the eight predominant elements, in ionic form"[59].

Thus, the evaluation of the nutritional information contained on the labels only for cations, referring to the group of bottled water of national origin (86), pointed to the description of the major elements Ca, K, Mg, and Na in all the samples (100%), as well as the presence of the declaration for the element Ba in 53% of the total evaluated. For the group of samples of imported origin, of the 21 samples analyzed, only the inorganic substances Ca (81%), K (57%), Mg (71%), and Na (81%) were declared.

In this way, the evaluation of the results obtained with the values declared on the label (Table 2) indicated that the median concentrations varied between the types of water analyzed, especially for the Na and Ca analytes in the imported samples taken from naturally gaseous sources, coming from the European countries of Spain and Italy. Variability of three orders of magnitude was also observed in the IQR amplitude for the same elements (615 mg L^{-1} for Na and 141 mg L^{-1} for Ca).

The high Na concentrations found can possibly be attributed to the natural geological composition of countries located on the European continent[60-62]. This factor is reflected in international guidelines[63], which classify a concentration of less than 500 mg L^{-1} as low mineral content, while a concentration of more than 1,500 mg L^{-1} is classified as water rich in mineral salts.

In the national bottled water samples, less variability can be seen in the medians reported (Table 2), and all were artificially carbonated. Unlike European waters, Brazilian waters have few dissolved solids and can therefore be classified as "very low mineralization" (dissolved solid content < 50 mg L^{-1}), with little geological influence on the composition of this type of product[64].

In the group of national water samples with added salts (Table 3), there was an amplitude of two orders of magnitude in the IQR for Na, K, Ca, and Mg. However, these values are in line with the legislation[14,15] for these elements.

Thus, the comparison between the labels on products of national origin and the concentrations of the analytes studied revealed inconsistencies between the declared values and those obtained experimentally. For the group of samples of national origin, only for those obtained from natural sources, of the total of 82 samples analyzed, the variation ranges for the analytes were: Ba (-99.4 to +545.5%), Na (-92.2 to +571.0%), Mg (-589.6 to +2041.1%), K (-82.1 to +33.8%), and Ca (-98.8 to +352.1%).

This variability points to the need for manufacturers to periodically carry out analytical control of the product and adapt it to nutritional labeling, thus making up-to-date and reliable composition tables available on labels, so that consumers can perceive and choose the product according to their needs[38].

This action by producers aims to correct the possible difference that can occur in the chemical composition of the water produced over time, as a result of possible soil contamination events that can reach the water table, seasonal interference, contamination during bottling, among others[45,62,64,65,66].

In the imported bottled water samples, the chemical composition of the nutrients also showed differences between the concentrations obtained and the respective label descriptions for Na (-18.2 to +43.5%), Mg (-62.7 to +18.1%), K (-38.9 to +17.1%), and Ca (-60.6 to +51.3%). The lower variability observed when compared to national samples may indicate more effective analytical control of labeling by manufacturers in these countries.

The assessment of labeling was also a relevant factor, considering only the set of samples of bottled water with added salts of national origin. The legislation[14,15] recommends that "the addition of food-grade salts should not exceed the MPVs for Ca (250 mg L^{-1}), Mg (65 mg L($^{-1}$)), K (500 mg L($^{-1}$)), and Na (600 mg $L^{(-1)}$)) and should contain at least 30 mg L^{-1} of all added salts". Analysis of the label description with the experimental data indicated that the manufacturers complied with this requirement for the elements Ca, Mg, K, and Na.

Concentration levels ranging from 0.10 to 7.54 mg L^{-1} were determined in samples of national bottled water that were labeled as Na-free. The designation of absence of Na may induce consumers who pay attention to the label description as an attribute when choosing to buy, to purchase a product with information that differs from its real chemical composition.

In addition, the lower concentration values obtained for the elements Ca, K, Mg, and Na in samples of bottled water of national origin, when compared to the concentrations obtained in samples of European origin (Table 3), may be related to the geology in which the national water resources are located, which, in turn, are less rich in macronutrients with low mineralization[64,65].

Data from the literature has also indicated discrepant values for the chemical elements declared on the label, as well as for other parameters, suggesting that this is a recurring problem which therefore requires greater attention and monitoring by the competent inspection bodies[67,68].

Recommended daily intake

The growing trend in the consumption of bottled water in the country, associated with the search for a healthy lifestyle, has led to greater concern among the population about the quality of the product available on the market, since consumers may pay attention to the reliability of the label as an attribute in the purchasing decision[12,13,61].

As the largest constituent of the human body, water is one of the components that contributes to the absorption of nutrients and possibly inorganic chemical contaminants in the body[1,2]. Inadequate intake of the macronutrient Na may be associated with harmful effects on the body, such as an increased risk of chronic non-communicable diseases[33,69,70].

Although national legislation for bottled water does not recommend the insertion of the daily reference value for inorganic elements on labeling[14,15], it was found that the highest concentrations were obtained for the nutrients Ca, K, Mg, and Na when compared to the toxic inorganic substances evaluated (Table 3).

Samples of national bottled water without added salts showed maximum levels of 119.5 mg L^{-1} for Na (natural, still), 16.5 mg L^{-1} for Mg (natural, still), 39.7 mg $L^{(-1)}$ for K (natural, still), and 35.7 mg $L^{(-1)}$ for Ca (natural, still). Imported water had higher values than national water, corresponding to 1,351.0 mg L^{-1} for Na (natural, sparkling, originating in Spain), 58.5 mg $L^{(-1)}$ for Mg (natural, sparkling, originating in Italy), 50.1 mg L^{-1} for K (natural, sparkling, originating in Spain), and 178.3 mg $L^{(-1)}$ for Ca (natural, sparkling, originating in Italy).

A comparison between the maximum concentrations and the World Health Organization (WHO) recommendations for daily nutrient intake of 2,000 mg for Na, 3,510 mg for K, 1,000 mg for Ca, and 320 mg for Mg[71,72,73], combined with fluid consumption for adult individuals (2.2 L for females and 2.9 L for males), indicated that excessive intake can only be attributed to the Na element in a sample of natural sparkling bottled water of Spanish origin. The concentration values exceeded 95.9% and 48.6% for male and female adults, respectively, considering only the intake of this product as food. For children, the situation is more critical, as the levels proposed should be adjusted to lower levels than those recommended for adults, based on children's energy requirements[71,72,73,74].

In addition to the possible presence of high sodium content in water, other factors may be related to its excessive daily consumption. These sources of intake of this nutrient can come from foods containing sodium naturally, industrialized foods (processed/ultra-processed, which include table salt as a preservative), routine use of medication/nutritional supplements, which can lead to the promotion of disorders and worsening of the population's health. The main illnesses include chronic kidney disease, hypertension, cardiovascular, and brain diseases. These diseases can show a possible increase in the morbidity and mortality rate of individuals affected by such anomalies and can be a major cause for concern for Public Health, leading to serious economic consequences for countries in the fight against chronic non-communicable diseases[69,70,71,74,75,76].

Finally, a comparison of the concentrations of the inorganic chemical substances obtained in this study with the values suggested by international organizations (U.S. Environmental Protection Agency - EPA, World Health Organization - WHO and U.S. Food and Drug Administration - FDA) with limits recommended by regulations in various countries (Table 3) showed that the levels found for the analytes As, Ba, Ca, Cd, Cu, K, Mg, Mn, Na, Ni, Pb, and Se in national bottled water samples were in agreement, and for the inorganic substances As, Ba, Ca, Cd, Cu, K, Mg, Mn, Na, Ni, Pb, and Se in imported water samples.

Concentration levels similar to those obtained in bottled water samples for most nutrients and inorganic contaminants have also been reported in recent literature, indicating the same inference with the results achieved in this study, as shown in Table 4, and demonstrating that the results obtained prove the quality of the bottled water offered on the market, which is considered suitable for consumption by the population.

CONCLUSIONS

The results generated in this research in the determination of inorganic chemical substances in bottled water of national and imported origin - 93.5% of the total of 107 samples evaluated were in accordance with the legislation - pointed to the importance of establishing and maintaining continuous monitoring programs in the country, in compliance with the MPVs in legislation.

In addition, the variability found between experimental results and the values declared in the chemical composition tables of national products can induce consumers to purchase products containing inadequate information, which can harm their daily intake needs. Consequently, it is important to develop monitoring actions in compliance with the labeling legislation for this category of product, which is a useful and effective tool for encouraging industries to periodically update the nutritional components of their products.

It should be considered that the significant consumption by the Brazilian population is directly associated with the quality of the product as being essential to the health of the organism and as an alternative in the prevention of diseases related to the use of public water supply, and allied to factors such as: the scarcity of basic sanitation policies in the country, the increase in purchasing power, market marketing strategies, reinforce the need to implement national quality control programs in order to protect the health of the population.

In addition, the results of the research are intended to stimulate laboratories working in the field of analytical chemistry to develop and validate analytical methodology for monitoring programs, used as an auxiliary tool in conjunction with the competent authorities, in the decision-making and actions of the health surveillance system, providing the establishment of public policies in the inspection of the sanitary quality of food, in order to protect the health of the population, with a view to promoting Public Health in the country.

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Authors' Contribution

Buzzo ML - Conception, planning (study design), acquisition, analysis, data interpretation, and writing of the study. Arauz LJ - Acquisition, analysis, data interpretation, and writing of the paper. Carvalho MFH, Mataveli LRV - Analysis. All the authors approved the final version of the paper.

Conflict of Interest

Authors have no potential conflict of interest to declare, related to this study's political or financial peers and institutions.