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Assessment of Potentially Toxic Elements and Associated Health Risk in Bottled Drinking Water for Babies

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Abstract: Potentially toxic elements are chemical pollutants which are dangerous to human health, especially for babies and children. Because their presence has been detected in baby food and baby drinking water, exposure to these elements is mainly due to ingestion. For this reason, the main objective of this study was quantification of 12 potentially toxic elements, including Ba, Co, Cu, Zn, Mn, Ni, Li, Fe, Pb, Cd, Cr, Sb, by ICP–MS, from 19 samples of bottled baby water. Based on the levels obtained, a health risk assessment was performed of the risk caused by their consumption, as well as an analysis of the quality of the samples. Excep iron, the values obtained for all other metals were below the limits imposed by the legislation in force. The risk analysis shows that Hazard index values were included in Risk Class 1, with a very low hazard level. The order of Cancer Risk values is as follows, Cd < Cr < Ni < Pb. As a general conclusion, we can say that the samples can be intended for consumption by children and infants.

Keywords: baby water; potentially toxic elements; health risk assessment; water quality

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1. Introduction

Heavy metals represent a group of potentially toxic elements, metals or metalloids with a density higher than 5 g/cm^3, which are able to induce toxicity even at low level of exposure [1,2]. Potentially toxic elements contamination represents a high concern due to their inherent toxicity, non-degradability, bio-accumulation and persistence in the environment and frequent detection in different matrices [1,3].

Environmental pollution with these chemical contaminants is due to both natural processes and anthropogenic activities. Natural sources include volcanic eruptions, forest fires, weathering of minerals, soil erosion, biogenic sources, airborne dust, while anthropogenic activities which are the major cause of pollution with these elements are burning of fossil fuels, mining operations, smelting, metallurgical processes, industrial discharges, agriculture, vehicular emissions [4–6].

Potentially toxic elements can be stored and bioaccumulated in the soil and can have negative consequences on human health or the environment, as they may be mobilized from the soil, or not, depending on the characteristics of the soil [4].

Moreover, more than 99% of these elements entering in aquatic systems and can be accumulated and stored in sediments [6], leading to various degrees of contamination of groundwater.

Chemical contaminants from sediments can affect water quality and aquatic ecosystem, when their concentrations exceed imposed limits, directly or indirectly having a negative impact for agricultural production and people's lives [1,7].

Soil contamination could lead to drinking water pollution, which can be transferred to human food chain which brings bioaccumulation and bio-magnification [6].

Humans are exposed to potentially toxic elements through ingestion of contaminated food or water, by inhalation of atmospheric particles enriched with metals, or absorption through the skin [8,9]. Even if potentially toxic elements reach the body at trace levels, they can accumulate and causes disorders like, neurological disease, hormone imbalance, respiratory problems, endocrine disorders, infertility, cardiovascular problems, kidney diseases, digestive problems [8].

The quality of drinking water is very important, being one of the major factors which can affect human health and, for this reason, is strongly inter-related with human heath [10,11].

The risk due to exposure to polluted water can be calculated through health risk assessment, and the results obtained may require measures to reduce the risk caused by contaminants. The most commonly used health risk assessment model is the one recommended by United State Environmental Protection Agency (USEPA) [10]. This model includes a carcinogenic and non-carcinogenic analysis, based on parameters like estimated daily intake (EDI), target hazard quotient (THQ), hazard index (HI) and cancer risk (CR) [12].

The quality of water can be evaluated by parameters like water quality index (WQI), entropy water quality index (EQWI) [10], water pollution index (WPI), comprehensive pollution index (CPI), organic pollution index (OPI), trace metal pollution index (TPI) [13], contamination factor (C_f), degree of contamination (mC_d), pollution load index (PLI) [8].

The health risk assessment of the water quality helps quantify the risk of exposure to polluted water, and, in this mode, by quantitative description of the hazards, decisions related to water source protection and management can be implemented [10].

In a study conducted by Beal, 2020 [14] on a series of baby food products and baby bottled water purchased from 15 retail chains in major cities in the United States, it was observed that 95% of products contain at least one toxic element (arsenic, lead, cadmium, mercury).

Moreover, in the study conducted by Decharat and Pan-in, 2020 [15] on a number of 48 samples of bottled water intended for consumption in schools, it was observed that lead and cadmium levels were between 2.5–18 μg/L, respectively 0.16–1.3 μg/L. In the case of lead, only 4 samples exceeded the 10 μg/L limit imposed by the WHO guide [16], while cadmium did not exceed the 3 μg/L limit. As a concerning Hazard index for lead and cadmium, the values were less than 1, but if other elements had been analyzed, the HI levels would probably have been higher.

In the study conducted by Peleka et al., 2021 [17], by estimating the hazard quotients based on the values of potentially toxic elements analyzed, for the infants and children population categories, HQ values were higher than 1 for both lead and cadmium, which show a relatively high hazard due to the consumption of the samples by these age groups.

The presence of potentially toxic elements in bottled water may also be due to contamination of raw water, as well as due to the packaging material, because some elements may remain in plastic packaging as catalyst or reaction residues, such as Pb-, Cd-, Ba-, Ca-Zn dicarboxylates and other, which can migrate into the packaged product under certain conditions [18]. Thus, in a study conducted by Allafi, 2020 [19], it was observed that the increase in temperature positively influences the migration of antimony from PET. The same was observed in other studies [20–24]. In addition to temperature, storage time can influence the migration of some elements [20,22–25], but also pH, as observed in the study by Chapa-Martinez et al., 2016 [20].

Due to these mentioned aspects, the main aim of this research is evaluation of the chemical quality of 19 samples of bottled drinking water for baby through quantification of 12 potentially toxic elements (Ba, Co, Cu, Zn, Mn, Ni, Li, Fe, Pb, Cd, Cr, Sb), as well as carcinogenic and non-carcinogenic health risk assessment for age group infants and children, based on the mentioned elements levels.

2. Materials and Methods

2.1. Sample Collection

The bottled baby water samples were purchased from local supermarkets and hypermarkets in Romania and included 10 samples of mineral water, 8 samples of spring water and 1 sample of table water. Also, 5 baby water are of Romanian origin and 14 samples are imported from France (3), Italy (1), Croatia (1), Greece (4), Hungary (1), Austria (1), Germany (1), Poland (1) and Bulgaria (1). The samples were bottled in polyethylene terephthalate (PET), with high density polyethylene screw cap and maintained at room temperature prior analysis.

2.2. Sample Preparation

The analysis of potentially toxic elements content in bottled water were performed according EPA3010A standard [26], and include that 300 mL of well mixed sample, initial acidified with a few drops of HNO_3, were evaporated to dryness on a water bath. After evaporation, the samples were treated with 2.5 mL of concentrated HNO_3, evaporated to dryness, and then, another 2.5 mL of concentrated HNO_3 were added. The extract with HNO_3 is covered with ribbed watch glass and left on the water bath for 15 min. The solution is then transferred to a 50 mL volumetric flask with ultrapure water and made up to the mark. Each of water samples was analyzed in triplicate to ensure data quality.

2.3. Equipment

Potentially toxic elements detection was performed using an Inductively Coupled Plasma–Mass Spectrometer NexION 300Q (Perkin Elmer Inc., Waltham, MA, USA). Also, for evaporation of water samples, a water bath GFL 1032 (Gesellschaft für Labortechnik, Germany), was used. ICP–MS operating conditions are listed in Table 1.

Table 1. ICP–MS operating conditions.

Parameter Description	Current Value
Sample introduction	Spray chamber
Radio Frequency Generator	Free running type, 40 MHz
Radio Frequency power	1000 W
Nebulizer Gas Flow	0.89 L/min
Auxiliary Gas Flow	1.20 L/min
Plasma Gas Flow	16 L/min
Sample Uptake Flow	1.0 mL/min
Dual Detector Mode	Pulse
Torch Z position	0.00 mm
Analytical masses	Standard mode
Ba, Co, Cu, Zn, Mn, Ni, Li, Fe, Pb, Cd, Cr, Sb mass	138, 59, 63, 66, 55, 27, 60, 7, 57, 208, 111, 52, 121
Scanning mode	Peak hopping
Number of replicates	3
Wash	Time (45), speed (±rpm)–24
Sample flush	Time (35), speed (±rpm)–24
Read delay	Time (15), speed (±rpm)–20

2.4. Analytical Method

For quantification of analytes, calibration curves for all the tested elements were performed, using a Multielement Standard Solution 6, with a concentration of 100 mg/L, from which standard solution of 10 µg/L, 20 µg/L, 30 µg/L, 40 µg/L and 50 µg/L were obtained. The validation of the method was performed according to Magnusson and Ornemark, 2014-Eurachem Guide [27] and DIN EN ISO/IEC 17025:2018 [28]. The correlation coefficients of calibration curves obtained for a working range between 10 µg/L and 50 µg/L were higher than 0.996. Detection limits, estimated as LOD = 3 SD/b, where SD is standard deviation obtained after 10 replicates measurements of blank samples and b in the slope of the calibration curve, were 0.07 μg/L for Sb and Pb, 0.09 μg/L for Ba, Cu, Mn, Ni and Cd, 0.10 μg/L for Co and Cr, 0.11 μg/L for Li, 0.13 μg/L for Zn and 0.15 μg/L for Fe. To check the measurement precision, standards' reference solutions spiked at 10 μg/L were used, the repeatability obtained being between 2.18 and 3.86. To assess the quality and accuracy of the results, standard reference solutions and a certified reference material (CRM) SPS-SW2 Surface water were used. Recovery rates for each element were between 91.50% and 108.40%, being considered as acceptable. The mean extended compound uncertainty (U) of the tested elements was of 10.31%.

2.5. Estimation of Potentially Toxic Elements Content

Estimation of potentially toxic elements content was performed using Equation (1).

$$C = [(\text{Reading} \times F) - M) \times V]/m, \tag{1}$$

where C is analyte concentration, in μg/L, Reading represent concentration obtained from the calibration curve, F is dilution factor, M is blank sample, V is sample volume, in mL (50 mL) and m is sample weight, in mg.

2.6. Health Risk Assessment

Risk assessment represent a method of estimation of risk of exposure to a pollutant and its probability to produce harmful effects on human health. This includes carcinogenic and non-carcinogenic analysis [29].

2.6.1. Non-Carcinogenic Analysis

For non-carcinogenic estimation, the model proposed by PHA Guidance, 2005 [30] was used, considering as the main source of exposure, ingestion. In this case, the evaluation was performed for 2-year children, with a weigh of 10 kg and an ingestion rate of 1 L/day. This analysis involves calculations of parameters, like exposure dose (D), hazard quotient (HQ) and hazard index (HI). The calculation of these indices was done using Equations (2)–(4).

$$D = (C \times IR \times EF)/BW, \tag{2}$$

where D is exposure dose, in mg/kg/day, C is contaminant concentration, in mg/L, IR is intake rate of water, in L/day, EF is exposure factor (unitless) and BW is body weight, in kg.

$$HQ = D/RfD, \tag{3}$$

where HQ is hazard quotient (unitless), D is exposure dose, in mg/kg/day and RfD is reference dose, in mg/kg/day. The reference dose represents the tolerable daily intake of the metal via ingestion. The values of this parameters are presented in Table 2. Hazard quotient represent the ratio between contaminant exposure and the respected reference values, specify for each exposure way (ingestion, inhalation or dermal contact) [16].

$$HI = HQ_{Pb} + HQ_{Cd} + HQ_{Cr} + HQ_{Cu} + HQ_{Zn} + HQ_{Mn} + HQ_{Ni} + HQ_{Ba} + HQ_{Co} + HQ_{Li} + HQ_{Fe} + HQ_{Sb}, \tag{4}$$

where HI is hazard index (unitless), and HQ is hazard quotient (unitless) of all heavy metals tested. Hazard index represent the total potential risk produced by chemicals in a mixture [31].

2.6.2. Carcinogenic Analysis

This analysis represents the probability that a chemical could produce a form of cancer, after exposure of a specified dose [29]. The model used for estimation of carcinogenic analysis, was the one proposed by PHA Guidance Manual, 2005 [30].

The carcinogenic analysis evaluation involves estimating the parameter cancer risk (CR), which has been calculated using Equation (5). This parameter, has been estimated for Pb, Cd, Cr and Ni, because only these have the set CSF values.

$$CR = D/CSF, \tag{5}$$

where CR represent cancer risk (unitless), D is exposure dose in mg/kg/day and CSF is Cancer Slope Factor, in mg/kg/day. The values of CSF for oral exposure are presented in Table 2.

For the 4 elements studied, total cancer risk (TCR) was calculated, which means the result of exposure to multiple heavy metals [32], using Equation (6).

$$TCR = Cr_{Pb} + Cr_{Cd} + Cr_{Cr} + Cr_{Ni}, \quad (6)$$

where TCR is total cancer risk (unitless), Cr represent cancer risk (unitless) for the four elements tested.

Table 2. RfD and CSF values for oral ingestion.

Element	RfD_{oral} (mg/kg)	CSF_{oral} (mg/kg/day)
Ba	7.0×10^{-2} [a]	-
Co	2.0×10^{-2} [b]	-
Cu	3.7×10^{-2} [b]	-
Zn	3.0×10^{-1} [b]	-
Mn	4.7×10^{-2} [c]	-
Ni	2.0×10^{-2} [b]	8.4×10^{-1} [a]
Li	2.8×10^{-2} [d]	-
Fe	7.0×10^{-2} [e]	-
Pb	3.6×10^{-3} [b]	8.50 [a]
Cd	5.0×10^{-4} [b]	6.10 [a]
Cr	3.0×10^{-3} [b]	4.1×10^{1} [a]
Sb	3.5×10^{-4} [f]	-

[a] [29]; [b] [33]; [c] [34]; [d] [35]; [e] [36]; [f] [37].

3. Results

3.1. Estimation of Potentially Toxic Elements Content

Toxic elements (metals) concentrations of the tested water samples are presented in Table 3. All the samples were analyzed in triplicate, the values being reported as mean value and standard deviation.

With the exception of Fe, all the tested elements are below the maximum imposed limits by national and international legislation in force. In case of Fe, only 4 samples were lower than imposed limit (200 µg/L).

3.2. Health Risk Assessment

3.2.1. Non-Carcinogenic Analysis

The values of exposure dose (D) and hazard quotient (HQ) are presented in Table 4. The ascendent trend of exposure dose, based on potentially toxic elements concentrations, is as following Cd > Cr > Sb > Co > Mn > Pb > Cu > Ni > Li > Zn > Ba > Fe. In case of HQ, the values decrease in order Cd > Co > Cr > Sb > Zn > Mn > Ni > Ba > Li > Pb > Cu > Fe. The interpretation and description of HQ and HI is presented in Table 5.

Except Fe, the estimated levels of hazard quotient were lower than 1. This means that repeated exposure of children does not cause side effects. However, in case of Fe, only one sample had the value lower than 1. This implies the appearance of some non-carcinogenic side effects, after repeated exposure to tested samples.

Regarding the hazard index values, obtained for hazard quotient of all analyzed elements, ranged between 9.10×10^{-1} and 2.42×10^{1}, with an average value of 7.10. Of the total of 19 samples, the sample that had the value HQ < 1 also had the value HI < 1. The rest of samples recorded HI values greater than 1. This means that after repeated exposure, some non-carcinogenic side effects can appear.

Table 3. Potentially toxic elements levels in baby drinking water samples.

Element	Heavy Metals Levels (µg/L)			Detection Rate (%)	Law No. 311/2004 (µg/L) [38]	Directive EU 2020/2184 (µg/L) [39]
	Min	Max	Mean			
Ba	<0.09	$1.68 \times 10^{1} \pm 0.32$	3.87 ± 0.19	68.42	-	-
Co	<0.10	$2.50 \times 10^{-1} \pm 0.01$	$7.0 \times 10^{-2} \pm 0.01$	47.37	-	-
Cu	<0.09	$1{,}73 \pm 0.03$	$9.60 \times 10^{-1} \pm 0.02$	100	100	2000
Zn	$9.60 \times 10^{-1} \pm 0.07$	4.47 ± 0.22	2.04 ± 0.07	100	5000	-
Mn	<0.09	4.17 ± 0.20	$3.50 \times 10^{-1} \pm 0.04$	42.11	50	50
Ni	$3.10 \times 10^{-1} \pm 0.01$	2.25 ± 0.06	1.02 ± 0.04	100	20	20
Li	<0.11	7.09 ± 0.43	1.69 ± 0.08	84.21	-	-
Fe	$6.24 \times 10^{1} \pm 6.13$	$1.69 \times 10^{3} \pm 3.92 \times 10^{1}$	$4.95 \times 10^{2} \pm 1.82 \times 10^{1}$	100	200	200
Pb	$1.10 \times 10^{-1} \pm 0.01$	1.79 ± 0.02	$4.30 \times 10^{-1} \pm 1.0 \times 10^{-2}$	100	10	5
Cd	<0.09	< 0.09	-	0	5	5
Cr	<0.10	$1.60 \times 10^{-1} \pm 0.01$	1.0×10^{-2} 0.00	10.53	50	25
Sb	<0.07	$1.30 \times 10^{-1} \pm 0.00$	$3.0 \times 10^{-2} \pm 0.00$	31.58	5	10

Table 4. Exposure dose (D), Hazard quotient (HQ) and Cancer risk (CR) from baby drinking water samples.

Element	Exposure Dose ($\mu g \cdot kg^{-1} \cdot day^{-1}$)			Hazard Quotient			Cancer Risk		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
Ba	0.00	1.68×10^{-3}	3.87×10^{-4}	0.00	2.39×10^{-2}	5.52×10^{-3}	-	-	-
Co	0.00	2.50×10^{-5}	7.95×10^{-6}	0.00	1.25×10^{-3}	4.19×10^{-4}	-	-	-
Cu	3.80×10^{-5}	1.75×10^{-4}	9.6×10^{-5}	4.25×10^{-3}	4.73×10^{-2}	2.54×10^{-2}	-	-	-
Zn	9.60×10^{-4}	4.47×10^{-4}	2.04×10^{-4}	3.20×10^{-4}	1.49×10^{-3}	6.18×10^{-4}	-	-	-
Mn	0.00	4.17×10^{-4}	3.51×10^{-5}	0.00	9.07×10^{-3}	7.62×10^{-4}	-	-	-
Ni	3.10×10^{-5}	2.25×10^{-4}	1.06×10^{-4}	1.55×10^{-3}	1.13×10^{-2}	5.12×10^{-3}	2.60×10^{-5}	1.89×10^{-4}	8.61×10^{-5}
Li	0.00	7.28×10^{-4}	1.72×10^{-4}	0.00	2.60×10^{-2}	6.13×10^{-3}	-	-	-
Fe	6.20×10^{-3}	1.69×10^{-1}	4.93×10^{-2}	8.86×10^{-1}	2.41×10^{1}	7.05	-	-	-
Pb	1.10×10^{-5}	1.79×10^{-4}	4.36×10^{-5}	3.06×10^{-3}	4.97×10^{-2}	1.21×10^{-2}	9.35×10^{-5}	1.52×10^{-3}	3.63×10^{-4}
Cd	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cr	0.00	$1.6\,0 \times 10^{-5}$	1.32×10^{-6}	0.00	5.33×10^{-3}	4.39×10^{-4}	0.00	6.56×10^{-4}	5.39×10^{-5}
Sb	0.00	1.30×10^{-5}	1.63×10^{-6}	0.00	4.33×10^{-3}	5.44×10^{-4}	-	-	-

Table 5. Hazard quotient, hazard index, cancer risk and total cancer risk classification and description (adapted from [14]).

Hazard Quotient	Description	Hazard Index	Description	Cancer Risk	Description	Total Cancer Risk	Description
≤1	repeated exposure not cause side effects	≤1	repeated exposure not cause side effects	$<1 \times 10^{-6}$	tolerable	$<1 \times 10^{-6}$	insignificant
>1	repeated exposure causes a potential risk	>1	repeated exposure can produce side effects	1×10^{-6}–1×10^{-4}	tolerable range	1×10^{-5}	acceptable level
-	-	-	-	$>1 \times 10^{-4}$	intolerable	$>1 \times 10^{-4}$	harmful

3.2.2. Carcinogenic Analysis

The values of cancer risk are presented in Table 4. The interpretation and description of this parameter is shows in Table 5. The results obtained in case of Pb were in a tolerable range, while Cd values are tolerable, due to low detection in the samples. Regarding Cr, 11% of samples were in a tolerable range and the rest of the samples were tolerable. Concerning Ni, 89% of results were in tolerable range and 11% were classed as intolerable. This means that, after repeated exposure to intolerable samples, Ni can produce some carcinogenic side effects.

The ascendent trend of cancer risk, estimated for the 4 studied elements, is as following Cd > Cr > Ni > Pb.

The levels of CR_{total} were between 1.69×10^{-4} and 1.58×10^{-3}, and a mean value of 5.03×10^{-4}. As can be seen, all the results were in tolerable range. This means that after repeated exposure to tested samples, heavy metals analyzed can't produce carcinogenic side effects in babies and children.

4. Conclusions

In this study, an Inductively Coupled Plasma Mass Spectrometry (ICP–MS) analytical method was used for detection and quantification of 12 potentially toxic elements from 19 samples of baby drinking water. The obtained results were below the maximum imposed limits by National and European legislation, with exception of Fe, where only 4 samples were below the limit. Cadmium was not detected in any sample. Based on tested elements levels, a carcinogenic and non-carcinogenic analysis was performed. The results of hazard quotient were lower than 1, except Fe, where only 1 sample had the value below the reference value. If in case of Fe the results demonstrated that after repeated exposure, some non-carcinogenic side effects could appear, the rest of elements can't produce adverse effects. Hazard index values were higher than 1, except only one sample. This means that after repeated exposure, can appear some adverse effects, but non-carcinogenic. The levels of carcinogenic risk of the four tested elements increase in the following order Cd > Cr > Ni > Pb, while total cancer risk results were between 1.69×10^{-4} and 1.58×10^{-3}, being framed in tolerable range. From the point of view of tested elements and health risk assessment, the samples can be intended for consumption by children and babies. In order to determine exactly whether the samples endanger the health of children, further, more detailed studies are needed, to detect as many potential contaminants as possible, for which a risk assessment can be performed.

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