

**RISK ASSESSMENT OF POTENTIALLY TOXIC TRACE ELEMENTS
THROUGH THE CONSUMPTION OF TAP WATER AND
BLACK COFFEE IN YEREVAN**

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Abstract

This study aims at assessing non-carcinogenic and carcinogenic risks of trace elements (Pb, As, Cd, Hg, Mo, Cu, Fe) through the consumption of tap water and black coffee in Yerevan. The obtained results indicate that the estimated daily intake and non-carcinogenic risk values of the studied elements do not exceed the acceptable level. Nonetheless, the carcinogenic risk values of Pb exceed the risk limit of 1×10^{-6} in case of black coffee consumption.

Keywords and phrases

Water, coffee, trace element, estimated daily intake, risk.

ԴԱՎԻԹ ՊԻՊՈՅԱՆ

սննդագիտության դոկտոր (Իտալիա)

ՀՀ ԳԱԱ Էկոլոգանոոսֆերային հետազոտությունների կենտրոն

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ՍԵՂԱ ՍՏԵՓԱՆՅԱՆ

շրջակա միջավայրի մագիստրոս

ՀՀ ԳԱԱ Էկոլոգանոոսֆերային հետազոտությունների կենտրոն

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տեխնիկական գիտությունների թեկնածու

ՀՀ ԳԱԱ Էկոլոգանոոսֆերային հետազոտությունների կենտրոն

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**ՀԱՎԱՆԱԿԱՆ ԹՈՒՆԱՎՈՐ ՏԱՐԴԵՐԻ ՌԻՍԿԻ ԳՆԱՀԱՏՈՒՄԸ ԵՐԵՎԱՆՈՒՄ ԾՈՐԱԿԻ
ՋՐԻ ԵՎ ՍԵՎ ՍՈՒՐՃԻ ՍՊԱՌՄԱՆ ԴԵՊՔՈՒՄ**

Համառոտագիր

Սույն հետազոտության նպատակն է գնահատել Երևանում ծորակի ջրի և սև սուրճի սպառման միջոցով հավանական թունավոր տարրերի (Pb, As, Cd, Hg, Mo, Cu, Fe) ոչ

քաղցկեղածին և քաղցկեղածին ռիսկը: Ստացված արդյունքները ցույց են տալիս, որ ուսումնասիրված տարրերի օրական ընդունման հաշվարկված չափաքանակի և ոչ քաղցկեղածին ռիսկի արժեքները չեն գերազանցում սահմանված թույլատրելի շեմը: Այնուամենայնիվ, սև սուրճի սպառման դեպքում կապարի քաղցկեղածին ռիսկը գերազանցել է 1×10^{-6} շեմը:

Բանալի բառեր և բառակապակցություններ

Չուր, սուրճ, հետքի տարր, գնահատված ամենօրյա ընդունում, ռիսկ:

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ОЦЕНКА РИСКА ПОТЕНЦИАЛЬНО ТОКСИЧНЫХ ЭЛЕМЕНТОВ ПРИ ПОТРЕБЛЕНИИ ВОДОПРОВОДНОЙ ВОДЫ И ЧЕРНОГО КОФЕ В ЕРЕВАНЕ

Аннотация

Целью исследование является оценка канцерогенного и неканцерогенного риска потенциально токсичных элементов (Pb, As, Cd, Hg, Mo, Cu, Fe) при потреблении водопроводной воды и черного кофе в Ереване. Полученные результаты показывают, что значения суточного поступления и неканцерогенного риска для изучаемых элементов не превышают допустимый уровень. Тем не менее, при потреблении черного кофе значения канцерогенного риска для свинца превышает 1×10^{-6} уровень.

Ключевые слова и фразы

Вода, кофе, микроэлемент, предполагаемая суточная доза, риск.

Introduction

Water and coffee are the two most commonly consumed beverages in the world [10]. Access to safe drinking water is essential to health and sustainable life. Water helps perform important chemical reactions and maintain important cell shapes [5]. Being a mountainous country, Armenia has abundant water resources, and the entire population has access to safe drinking water source [1, 3]. Coffee is the second largest consumed beverage in the world after water, and its market is expected to grow by 10% annually. Coffee is consumed by around 40% of the world's population, and the average consumption is 0.9 kg/capita in 2020 [15, 16].

Drinking water may contain essential trace elements, including Na, K, Cl, Fe, Cu, Mo; however, it can also contain potentially toxic trace elements, like Pb, Hg, Cd and As, which can be toxic to human health even at low levels of exposure [23]. Pb and Cd are rarely present in tap water, but due to the water distribution system, which includes pipes, storage tanks or service connections to home, chemical contamination may occur [24]. Although coffee contains antioxidants that support the body's natural

cell defences, it may also contain trace elements, since coffee plants take up heavy metals from various sources [15].

In Armenia, trace elements have been found in various food items: such as fruits and vegetables, cereals and honey. Moreover, health risk assessment has been conducted mostly for the Yerevan population, the capital of Armenia, where approximately one-third of the nation resides. The investigation results indicated that there is a potential carcinogenic risk due to the consumption of these products [11-14].

Armenians use tap water for drinking and cooking, particularly for coffee preparation. Since tap water and black coffee are the major beverages of the Armenians' diet, the content of trace elements in those products can have a substantial contribution to total human exposure. Up till now, investigation of tap water and black coffee consumption habits and their related health risks of trace elements has never been conducted among Yerevan population. Therefore, this study aimed to assess the dietary exposure and the health risk of trace elements (Pb, As, Cd, Hg, Mo, Cu, Fe) through the intake of tap water and black coffee in Yerevan.

Materials and Methods

Sample collection, treatment and analysis

The sample collection was done as a part of a Total Diet Study (TDS) conducted from 2018 to 2019 in Yerevan. All the sample collection, preparation and treatment procedures were done according to TDS approaches. To ensure the representativeness, tap water samples were obtained from all 12 administrative districts of Yerevan, while the most commonly consumed coffee samples were taken from supermarkets. Coffee samples were subject to kitchen preparation (similar to household preparation mostly used by Yerevan population). Afterwards, the samples were pooled and sent for laboratory analysis [25]. Overall, 2 representative groups of pooled samples of tap water and black coffee were included in the study.

The levels of seven trace elements (Pb, As, Cd, Hg, Mo, Cu, and Fe) were determined with Tandem ICP-mass spectrometry (ICP-MS/MS) in accordance to STB ISO 17294-2-2007 standard.

Data collection and statistical analysis

TDS is used for estimating population dietary exposure to both beneficial and harmful chemical substances across the entire diet [25]. Food consumption data were collected at the individual level using a 24-hour dietary recall (24HR). The data analysis and statistical tests were conducted using SPSS software (version 22.0). Several homogeneity tests were conducted to check for the normal distribution of consumption data, and K-means cluster analysis method was applied to reveal any possible homogenous groups of consumers [2].

Estimated Daily Intake (EDI)

To assess the dietary exposure to trace elements, the estimated daily intake (EDI) was calculated with the following formula:

$$EDI = \frac{C \times IR \times EF \times ED}{BW \times AT}$$

where C is the mean concentration of each trace element (mg/kg), IR is the rate of ingestion of tap water and black coffee (kg/day), EF is the frequency of exposure (365 days/year), ED is the duration of exposure (63.6 and 69.7 years for males and females, respectively, taking into account mean life expectancy from 8 years of age). BW is the body weight (kg) (mean body weights of 70 and 60 kg, for males and females, respectively), AT is the time over which the dose is averaged. For evaluating the carcinogenic risk, AT of 70 years (25550 days) was considered [21].

Non-Carcinogenic Risk Assessment

To assess the non-carcinogenic risk posed by chronic exposure to trace elements, target hazard quotient (THQ) was calculated according to the formula below [14]:

$$THQ = \frac{EDI}{RfD}$$

where *RfD* is the reference dose (mg/kg bw/day), an estimate of a toxicity threshold value [20]. The *RfD* values for Pb, Hg and Cu were taken as 0.0035, 0.00057 and 0.01 mg/kg/bw/day, respectively, using provisional tolerable weekly intake (PTWI) for Pb and Hg and dietary reference intake (DRI) for Cu [4, 7, 8]. The *RfD* values for As, Cd, Mo and Fe were 0.0003, 0.001, 0.005, and 0.7 mg/kg/bw/day, respectively [17, 18, 19, 22].

Carcinogenic risk assessment

The carcinogenic risk for trace elements (Pb, As and Cd) was estimated according to the formula below [9]:

$$CR = \sum_{k=1}^n EDI_k \times SF_k$$

where CR is the carcinogenic risk that can be developed by an individual over a lifetime, EDI_k (mg/kg/day) and SF_k (mg/kg/day)⁻¹ are the estimated daily intake and the cancer slope factor, respectively for elements starting from *k* to *n*. *SF* is the slope factor for estimating the probability of an individual developing cancer from exposure to the contaminant over a lifetime. *SF* values for Pb, As and Cd were taken as 0.5, 1.5 and 0.38 (mg/kg/day)⁻¹ [13].

Results and Discussion

Levels of trace elements

The contents of trace elements in tap water and black coffee are presented in Figure 1.

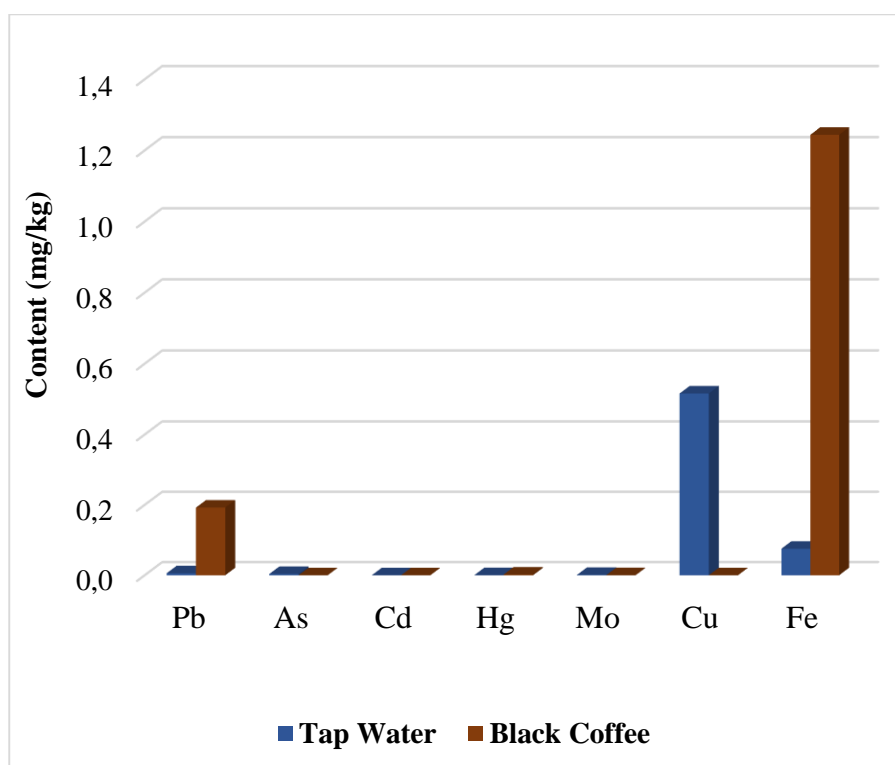


Fig. 1. Trace element content in tap water and black coffee

The obtained results indicate that tap water samples had relatively high levels of Cu and Fe ($5.16\text{E-}01$, $7.54\text{E-}02$ mg/kg, respectively), while coffee samples had high levels of Pb and Fe ($1.93\text{E-}01$ and $1.24\text{E+}00$ mg/kg, respectively). Compared with the World Health Organization (WHO) permissible levels of trace elements in drinking water, none of the trace elements exceeded the allowable levels [24]. In comparison with the available maximum allowable concentrations of Technical Regulations of Customs Union (TR CU) where Armenia is a member state, contents of trace elements did not exceed the established levels in tap water [6]. It is noteworthy that there are no WHO or Eurasian Economic Community (EAEC) established maximum allowable levels for the studied trace elements in prepared coffee.

Water and Coffee consumption

The 24HR study of water and coffee consumption indicated that both form a significant part of the Armenian diet. Water is consumed daily in Yerevan. Black coffee is consumed habitually by approximately 71% of the population. Based on Kolmogorov-Smirnov and Shapiro-Wilk normality tests, the consumption data did not have a normal distribution ($p = 0.000$, $p < 0.05$). To get a normal distribution K-means cluster analysis was conducted, which revealed 3 cluster groups of tap water consumers and 2 cluster groups of black coffee consumers. The daily consumption values of tap water and black coffee are summarized in Figure 2 and 3, respectively.

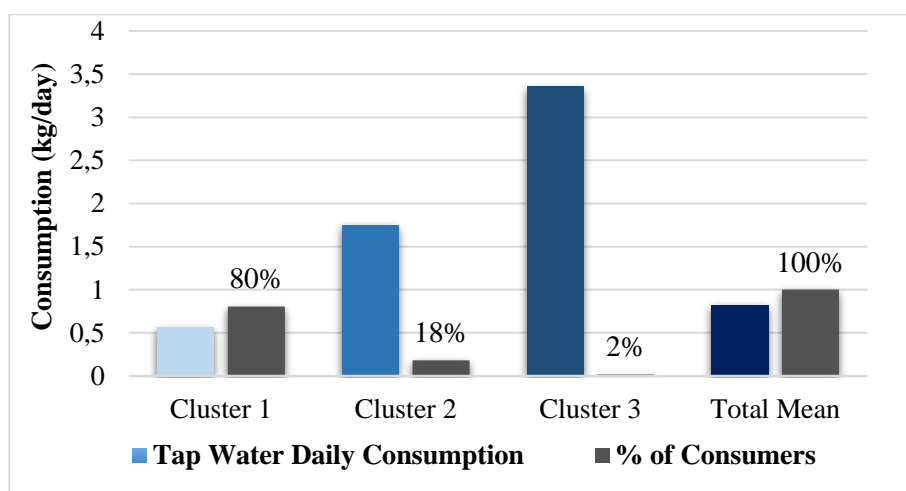


Fig. 2. Consumption of tap water

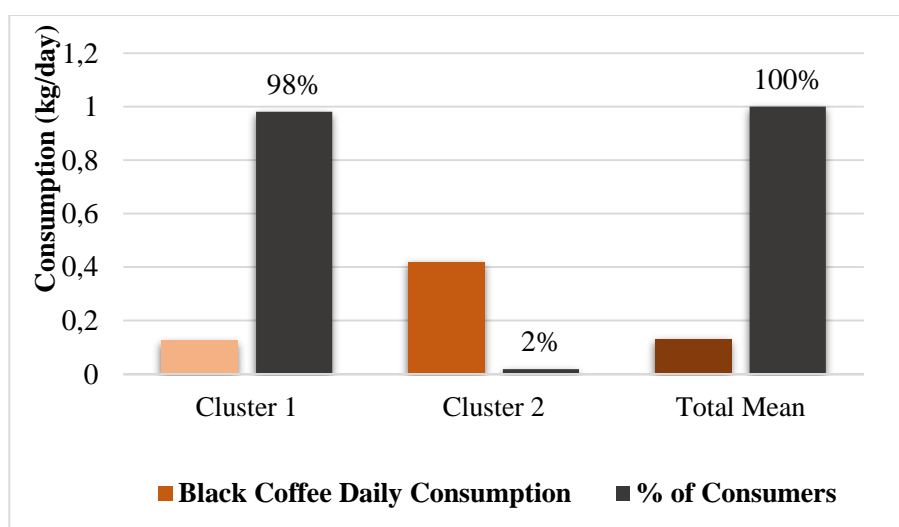


Fig. 3. Consumption of black coffee

The mean of daily tap water consumption is 822g. It is noteworthy that 80% of the population consumes 560g of water per day, which is less than the general recommended value of at least 2 litres of water per day [26]. The mean of daily black coffee consumption is 132g, which is equal to two cups of coffee. 98% of the studied population consumes precisely that amount of coffee per day.

Estimated Daily Intake of Trace Elements

To carry out the health risk assessment, the estimated daily intake of trace elements was calculated (Table 1) and compared with the health-based guidance values (HBGVs) established by international organizations.

Table 1. EDI of trace elements (mg/kg/day)

Beve- rage	Gender	EDI of trace elements						
		Pb	As	Cd	Hg	Mo	Cu	Fe
Tap Water	Male	6.19E-05	3.79E-05	1.39E-06	3.42E-06	1.16E-05	5.51E-03	8.05E-04
	Female	7.91E-05	4.85E-05	1.77E-06	4.37E-06	1.49E-05	7.04E-03	1.03E-03
Black Coffee	Male	3.30E-04	8.57E-08	8.57E-08	3.48E-06	4.11E-07	8.57E-08	2.13E-03
	Female	4.22E-04	1.10E-07	1.10E-07	4.45E-06	5.26E-07	1.10E-07	2.73E-03

The EDIs of all the trace elements were within their reference values. In general, in case of tap water, the highest contribution in EDIs had Cu (86%) and Fe (12%), while in case of black coffee, Fe (87%) and Pb (13%). The rest of the trace elements were identified in minimal quantities and therefore, had nearly 0% contribution in EDI.

Non-carcinogenic risk assessment

To conduct a non-carcinogenic risk assessment and to identify possible human health risks posed by chronic exposure, target hazard quotient (THQ) was estimated (Figure 4).

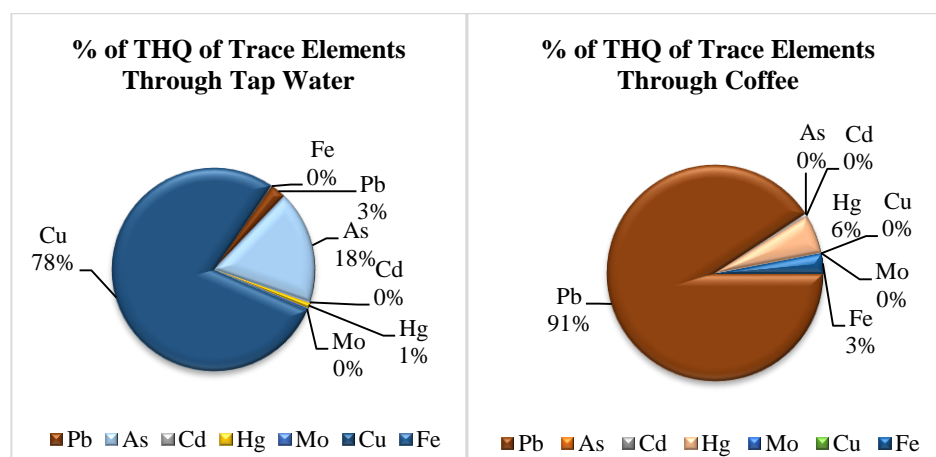


Fig. 4 THQ of trace elements (shown as a %)

Generally, a THQ value of less than one indicates that the exposed population is unlikely to experience adverse health effects. While, in the case of THQ of higher than one, there may be a concern for potential non-carcinogenic effects [13]. The THQ values for Pb, As, Cd, Hg, Mo, Cu and Fe were below one, indicating no risks of

chronic toxic effects through tap water and black coffee consumption. Overall, relatively higher levels of THQ values were reported for Pb, As and Cu in case of tap water and black coffee.

Carcinogenic risk assessment

In this study, carcinogenic risk (CR) was estimated for Pb, As and Cd, classified by the International Agency for Research on Cancer (IARC) as human carcinogens. Overall, according to the US Environmental Protection Agency, a carcinogenic risk of less than 1×10^{-6} or between 1×10^{-6} and 1×10^{-4} is considered acceptable for humans, while CR of 1×10^{-4} is the threshold limit and is considered unacceptable [21]. The results of carcinogenic risk assessment are summarized in Table 2.

Table 2. *CR of trace elements*

Food Type	Trace Elements	CR for Trace Elements		Cumulative CR of all Trace Elements	
		Male	Female	Male	Female
Tap Water	Pb	3.09E-05	3.96E-05	8.84E-05	<i>1.13E-04</i>
	As	5.69E-05	7.28E-05		
	Cd	5.27E-07	6.74E-07		
Black Coffee	Pb	<i>1.65E-04</i>	<i>2.11E-04</i>	<i>1.65E-04</i>	<i>2.11E-04</i>
	As	1.29E-07	1.64E-07		
	Cd	3.26E-08	4.16E-08		

Note: *The values exceeding the threshold level are in bold and italics.*

In the case of tap water, the CR values of Pb and As were within the acceptable limit, and the CR value of Cd was within the safe limit. However, the cumulative CR value exceeded the acceptable risk level (1×10^{-6} - 1×10^{-4}) for females, indicating that tap water consumption is possibly carcinogenic.

In case of black coffee, the CR value of As was within the acceptable limit, the CR value of Cd was within the safe limit, while the CR value of Pb exceeded the possible risk level. The cumulative CR value also exceeded the potential risk level of 1×10^{-4} for both male and female, indicating that black coffee consumption is possibly carcinogenic.

Overall, in the case of a combined consumption of tap water and black coffee, there is an unacceptable level of possible carcinogenic risk posed by Pb.

Conclusion

The results of the study highlight the importance of investigating dietary exposure to trace elements through water and coffee consumption. In Armenia, water and coffee are consumed daily (coffee is consumed by 71% of the population); therefore, both have an essential contribution to the intake of investigated trace elements. For all the studied trace elements, none of the EDI exceeds the established HBGV value. However, carcinogenic risk assessment indicated that only the values of Pb exceed the risk limit in case of tap water and black coffee consumption for both male and female and therefore, are indicative of a possible carcinogenic risk. Future studies are needed

to investigate, whether the possible carcinogenicity of black coffee is attributable to the use of tap water in coffee. Moreover, to get a holistic view of total carcinogenic risk, it will be beneficial to conduct further investigations taking into consideration also other commonly consumed food products.

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ԴԱՋԱՐ ԳԱԼՈՅԱՆ

Երկրաբանական գիտությունների թեկնածու
 տիեզերքի մասին գիտությունների դոկտոր (Ֆրանսիա)
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Համառոտագիր

Աշխատանքի նպատակն է հստակություն մտցնել Սևանի օֆիոլիտային համալիրի
 Դալի գետահովտի տեղամասում առկա հրաբխամագմատիկ առաջացումների