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INSIGHT INTO THE RISKS OF MILK CONSUMPTION, POTENTIALLY CONTAMINATED WITH HEAVY METALS AND POLYCYCLIC AROMATIC HYDROCARBONS

Andreea Laura Banica^{1,2}, Cristiana Radulescu^{1,3,4*}, Radu Lucian Olteanu³, Ioana Daniela Dulama², Raluca Maria Stirbescu²

¹Politehnica University of Bucharest, Doctoral School of Chemical Engineering and Biotechnology, 060042 Bucharest, Romania ²Valahia University of Targoviste, Institute of Multidisciplinary Research for Science and Technology, 130004 Targoviste, Romania ³Valahia University of Targoviste, Faculty of Sciences and Arts, 130004 Targoviste, Romania

⁴Academy of Romanian Scientists, 050044 Bucharest, Romania

*Corresponding author: <u>radulescucristiana@yahoo.com</u>

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Abstract: The study aimed to identify the health risks associated with consuming processed milk contaminated with heavy metals (HMs) and polycyclic aromatic hydrocarbons (PAHs) for adults and children. The risk analyses were performed by applying specific exposure and toxicity indices, which allowed a differentiated comparison between age groups and types of milk. The results highlighted significant variations in the risk level depending on the contaminant and consumer category, with higher values observed in the case of adults. At the same time, the statistical correlations performed between the risk indices suggested the existence of interdependence between exposure to heavy metals and polycyclic aromatic hydrocarbons, highlighting the potential cumulative effect of these emerging contaminants.

Keywords: health risk, heavy metal, ingestion, milk intake, polycyclic aromatic hydrocarbon, risk indexes

Over the last century, the rapid growth of the economy and globalization of industrialization have led to significant environmental pollution [1-3]. This pollution poses numerous threats to both human health and environmental safety. Heavy metals are inorganic contaminants that are non-biodegradable, highly toxic, and persistent in the environment. They also have the potential to bioaccumulate, which further exacerbates their harmful effects [4]. Copper, zinc, arsenic, lead, mercury, chromium, nickel, and cadmium are classified as hazardous substances due to their documented ecotoxicological impact [5-8]. These contaminants infiltrate food chains through biotic absorption, posing long-term threats to terrestrial and marine ecosystems, and implicitly to humans. These metals contaminate crops and aquatic fauna, posing a serious risk to human health through their accumulation in the food chain [9, 10]. Despite numerous studies on the persistence, complexity, toxicity, and behavior of these metals within the food chain, there remains a lack of comprehensive mapping of pollution levels in each region [11-13].

Polycyclic aromatic hydrocarbons (PAHs) primarily result from human activities such as burning fossil fuels, oil, coal, and wood, but they can also originate from natural processes like forest fires. These compounds are extremely toxic and include carcinogens, teratogens, and mutagens. PAHs can bioaccumulate and are semi-volatile, meaning they can evaporate from water or soil into the atmosphere. They can also adhere to particulate matter (PM), allowing them to travel long distances before being deposited in cold regions and at high altitudes. As temperatures rise, these compounds can re-enter the atmosphere and continue to migrate over vast distances [14-16].

Food can be contaminated with heavy metals and polycyclic aromatic hydrocarbons in various ways through exposure to environmental factors: animals are usually exposed to emerging contaminants by inhalation or ingestion, but contamination can also occur through the food chain, or food can also become contaminated during processing/preparation (i.e., smoking or grilling meat) [17 - 19]. Additionally, these contaminants are widely recognized as environmental contaminants present in air, water, and soil.

The toxic effects of heavy metals and polycyclic aromatic hydrocarbons on human health are primarily determined by the duration and route of exposure, and secondarily by the volume or concentration to which a person is exposed, as well as the relative toxicity of these emerging compounds [20]. The skin, lungs, pancreas, esophagus, bladder, colon, and other organs are prone to developing tumors as a consequence of long-term exposure to heavy metals and polycyclic aromatic hydrocarbons; also, long-term exposure to PAHs can increase the risk of cardiovascular diseases (i.e., atherosclerosis, hypertension, thrombosis, and myocardial infarction) [20, 21].

The primary objective of this study was to assess the level of risk associated with the consumption of processed milk, both conventional and organic, that is contaminated with heavy metals and polycyclic aromatic hydrocarbons. This evaluation focused on different age groups, specifically adults and children. Additionally, the study aimed to identify potential statistical correlations that could establish links between the risk indices associated with the two emerging contaminants.

MATERIALS AND METHODS

Materials

To achieve the two objectives of this study, previous research conducted by Banica *et al.* [22] on 16 milk samples (both conventional and organic) provided valuable results. Figure 1 shows the selection criteria for these samples.

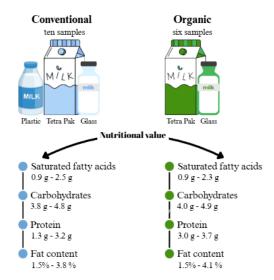


Figure 1. Presentation of conventional and organic milk samples as described by Banica et al. [22] in their research

Methods

Numerous methods and tools for data analysis have been developed in the field of statistics. Statistical analysis and data interpretation are essential aspects of research. To facilitate this, several statistical software packages are available, with IBM SPSS being one of the most used [23, 24]. IBM SPSS Statistics v. 26 was employed for the statistical analysis. Principal component analysis (PCA), scatterplot matrix, Pearson correlation, and pairwise comparisons were performed. These four statistical models were applied to both heavy metals and polycyclic aromatic hydrocarbons.

DATA ANALYSIS

Food is essential for maintaining a healthy lifestyle and ensuring the harmonious growth and development of humanity [25]. Researchers are currently focusing on emerging contaminants such as pesticides [26], microplastics [22, 27 - 29], heavy metals [30 - 32] and polycyclic aromatic hydrocarbons [22, 33, 34] due to their impact on the food industry, humans and ecotoxicological effects, as well as the speed with which they pollute terrestrial and marine ecosystems [35 - 37]. Ingestion of emerging contaminants is the main route of human exposure, and risk factors are presented in the following sections. The pollution load index (PLI) for heavy metals, the estimated daily intake

(EDI), and the health risk index (HRI) were calculated for heavy metals in previous research by Banica et al. [22].

Intake risk of heavy metals

The estimate of the chronic daily ingestion dose (CDI_{ng}) to heavy metals was established based on Equation (1) adopted after the research conducted by Bucur (Popa) *et al.* [38], and after the research conducted by Aralu *et al.* [39], and the indicators and their description are presented in Table 1. CDI_{ng} was calculated for both adults and children.

$$CDI_{ng} = C_{HM} \cdot \frac{I_{ng}R \cdot E_f \cdot E_d \cdot C_m}{B_w \cdot A_t} \tag{1}$$

where: $I_{ng}R$ is the ingestion rate (expressed as kg·day⁻¹), E_f is the exposure frequency (expressed as day·year⁻¹), E_d is the exposure autonomy (expressed in years), C_m is the conversion factor, B_w is the body weight (expressed in kg), and A_t is the average exposure time (expressed in days).

Table 1. Values of standardized indicators based on which CDI_{ng} was calculated (adapted from Peirovi-Minaee et al. [40])

Indianton	Crowbal	Measurement	Age category (years)			
Indicator	Symbol	unit	Adults	Children		
Ingestion rate	$I_{ng}R$	L·day ⁻¹	0.750	0.625		
Exposure frequency	$E_{\rm f}$	days·year ⁻¹	365			
Exposure duration	E _d	years	70	10		
Conversion factor*	C _m	-	0.0085			
Body weight	B_{w}	kg	70	14		
Average time	A_{t}	days	$A_t = E_f \cdot E_d$			

^{*} Conversion factor for fresh mass converted to dry sample mass

The hazard quotient for the ingestion of heavy metals from food was calculated as the ratio between CDI_{ng} and R_fD (oral reference dose) according to Equation (2) adapted from Farahmandkia *et al.* [41].

$$HQ = \frac{CDI_{ng}}{R_f D} \tag{2}$$

The oral reference dose expressed in mg·kg⁻¹·day⁻¹ for each determined metal (Cr, Mn, Ni, Cu, Zn, Sr, Cd, and Pb) is presented in Table 2.

Table 2. Oral reference dose for the analyzed metals [22, 42]

	Cr	Mn	Ni	Cu	Zn	Sr	Cd	Pb
$R_{\rm f}D$	1.500	0.140	0.020	0.040	0.300	0.600	0.001	0.035

The risk index or hazard index (HI) was calculated based on Equation (3), which represents the sum of HQ [43]. Both HQ and HI were calculated for adults and children.

$$HI = \sum HQ \tag{3}$$

The lifetime risk of developing cancer (CR) due to exposure to carcinogens (heavy metals) was established based on Equation (4), which was adopted after research conducted by Sushila *et al.* [44].

$$CR = CDI_{ng} \cdot CSF \tag{4}$$

where *CSF* represents the cancer slope factor and was estimated for Cd, Cr, Ni, and Pb to be 0.38 mg·kg⁻¹·day⁻¹, 41 mg·kg⁻¹·day⁻¹, 0.84 mg·kg⁻¹·day⁻¹, and 0.0085 mg·kg⁻¹·day⁻¹, respectively.

The total carcinogenic risk (CR_t) represents the sum of the carcinogenic risk for each heavy metal accumulated in a sample (milk sample) and was established by applying Equation (5):

$$CR_{t} = \sum_{n=1}^{4} CR = CR_{Cd} + CR_{Cr} + CR_{Ni} + CR_{Pb}$$
 (5)

Intake risk of polycyclic aromatic hydrocarbons

The average daily dose (ADD) of PAHs was determined using Equation (6) [45].

$$ADD = \frac{C_{PAH} \cdot I_{ng}R}{B_{w}} \tag{6}$$

where: C_{PAH} represents the concentration of PAHs in the sample (ng·L⁻¹), $I_{ng}R$ represents the ingestion rate of the contaminated product (L·day⁻¹) and was calculated for adults (I_{ng}R = 0.750 L·day⁻¹) and children (I_{ng}R = 0.625 L·day⁻¹), and B_w is the body mass established in the present study for adults at 70 kg and 14 kg for children.

After establishing the average daily dose, HQ and HI were calculated based on Equations (2) and (3), where R_fD has regulated values for three PAHs and are presented in Table 3. Based on Equation (1), CDI_{ng} was also calculated for PAHs, where C_{PAH} represents the concentration of the PAHs determined in the sample.

Table 3. Oral reference dose for three PAHs according to the US EPA IRIS [46 – 48]

PAH	R_fD	Reference
Naphthalene	0.02	[44]
Fluorene	0.04	[45]
Benzo[a]pyrene	0.0003	[46]

In the present study, daily exposure to PAHs through ingestion of contaminated food was calculated. The equivalent toxicity coefficient (TEQ) has been calculated using Equation (7) [49, 50].

$$TEQ = \sum C_{PAH} \cdot TEF_{PAH} \tag{7}$$

where TEF_{PAH} represents the equivalent toxicity factor for each PAH determined according to the research carried out by Nisbet and LaGoy [51] and presented in Table 4.

Table 4. Equivalent toxicity factor for the 13 PAHs determined in milk samples [51]

PAHs	TEF Factor
Naphthalene	0.001
Acenaphthene	0.001
Fluorene	0.001
Anthracene	0.01
Fluoranthene	0.001
Pyrene	0.001
Benzo[a]anthracene	0.1
Chrysene	0.001
Benzo[b]fluoranthene	0.1
Benzo[k]fluoranthene	0.1
Benzo[a]pyrene	1.0
Dibenzo[a,b]anthracene	5.0
Benzo[g , b , i]perylene	0.01

RESULTS AND DISCUSSION

Risk of heavy metal ingestion

Metals identified in processed milk have both positive and negative roles on human health. Cd, Pb, Cu, and Cr are considered the most toxic metals by the US Environmental Protection Agency (USEPA) [52-54]. Zinc, together with magnesium, selenium, phosphorus, and potassium, is a metal that is naturally found in milk composition [54]. In the study by Banica *et al.* [22], heavy metals were determined in processed milk samples (conventional and organic milk), and the risk levels were evaluated in this section. In the specialized literature, there are no minimum and maximum limits for CDI_{ng}, but the authors of this research propose several risk levels according to Table 5.

Table 5. Risk levels for CDI_{ng} following heavy metal ingestion

Risk level	$\mathbf{CDI}_{\mathbf{ng}}$
Very low risk	>10-6
Low risk	10-6
Medium risk	10-5
High risk	10-4
Very high risk	≤10 ⁻³

The chronic daily exposure dose by ingestion of heavy metals was calculated for children (Figure 2a) and adults (Figure 2b). The CDI_{ng} values determined for adults are higher than those calculated for children. The chronic daily exposure dose by ingestion of heavy metals determined in the milk samples analyzed in the present study presents a very high risk for adults, and the concentration of heavy metals in milk presents a worrying risk for human health. The daily chronic exposure of children to zinc highlights a very high risk. The high level of risk is associated with the abundance of zinc in the milk samples. The CDI_{ng} values for chromium and copper associate the analyzed milk samples with a medium risk level. Manganese and nickel, through the chronic daily exposure dose established in the analyzed milk samples, associate the analyzed samples with a low to

medium risk level. The CDI_{ng} values calculated for lead place the analyzed milk samples in the low and medium risk levels, as do the two conventional milk samples (L_4 and L_5), for cadmium.

When a heavy metal is present in the air, soil, and/or water, it is absorbed by plants and crops that subsequently become food for cattle and other animals, and ultimately it can reach the human body [55-57]. The route of heavy metals in the food chain from soil to humans can explain their presence in food, but only clinical studies can demonstrate their reactions in the human body [57, 58].

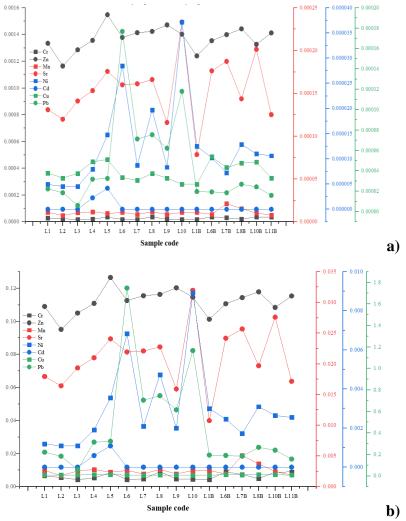


Figure 2. Daily dose of chronic exposure through ingestion of heavy metals for: a) children and b) adults

The Hazard Quotient (HQ) and the Hazard Index (HI) of heavy metal consumption were used to assess the carcinogenic risk for both adults and children based on the analyzed processed milk samples. The HI was calculated by summing the concentrations of eight heavy metals identified in the samples. The resulting values are presented in Table 6 for conventional milk and in Table 7 for organic milk.

Table 6. Risk quotient and hazard index for heavy metal consumption in conventional milk

Samples	Cr	Mn	Ni	Cu	Zn	Sr	Cd	Pb	HI			
HQ Adults												
L_1	9.97·E ⁻³	3.70·E ⁻⁴	2.37·E ⁻⁵	3.96·E ⁻⁴	3.27·E ⁻²	1.08·E ⁻²	0.00	7.75·E ⁻³	6.20·E ⁻²			
L_2	8.33·E ⁻³	2.55·E ⁻⁴	2.19·E ⁻⁵	3.45·E ⁻⁴	2.85·E ⁻²	9.86·E ⁻³	0.00	6.43·E ⁻³	5.38·E ⁻²			
L_3	6.42·E ⁻³	3.57·E ⁻⁴	2.19·E ⁻⁵	3.92·E ⁻⁴	3.15·E ⁻²	1.16·E ⁻²	0.00	2.10·E ⁻³	5.24·E ⁻²			
L_4	7.92·E ⁻³	3.95·E ⁻⁴	3.83·E ⁻⁵	5.11·E ⁻⁴	3.33·E ⁻²	1.26·E ⁻²	5.94·E ⁻⁷	1.10·E ⁻²	6.58·E ⁻²			
L_5	1.28·E ⁻²	3.32·E ⁻⁴	7.10·E ⁻⁵	5.35·E ⁻⁴	3.80·E ⁻²	1.44·E ⁻²	1.09·E ⁻⁶	1.13·E ⁻²	7.75·E ⁻²			
L_6	6.56·E ⁻³	3.70·E ⁻⁴	1.37·E ⁻⁴	3.48·E ⁻⁴	3.38·E ⁻²	1.32·E ⁻²	0.00	6.12·E ⁻²	1.16·E ⁻¹			
L_7	6.97·E ⁻³	2.93·E ⁻⁴	4.19·E ⁻⁵	3.21·E ⁻⁴	3.47·E ⁻²	1.33·E ⁻²	0.00	2.47·E ⁻²	8.02·E ⁻²			
L_8	1.35·E ⁻²	3.95·E ⁻⁴	9.47·E ⁻⁵	3.92·E ⁻⁴	3.49·E ⁻²	1.37·E ⁻²	0.00	2.61·E ⁻²	8.91·E ⁻²			
L_9	7.10·E ⁻³	2.93·E ⁻⁴	4.01·E ⁻⁵	3.45·E ⁻⁴	3.61·E ⁻²	9.55·E ⁻³	0.00	2.15·E ⁻²	7.50·E ⁻²			
L_{10}	6.83·E ⁻³	3.70·E ⁻⁴	1.79·E ⁻⁴	2.81·E ⁻⁴	3.44·E ⁻²	1.92·E ⁻²	0.00	4.08·E ⁻²	1.02·E ⁻¹			
				HQ (Children							
L_1	4.16·E ⁻⁵	1.54·E ⁻⁶	9.87·E ⁻⁸	1.52·E ⁻⁶	4.00·E ⁻⁴	7.85·E ⁻⁵	0.00	7.84·E ⁻⁷	5.24·E ⁻⁴			
L_2	3.47·E ⁻⁵	1.06·E ⁻⁶	9.11·E ⁻⁸	1.32·E ⁻⁶	3.49·E ⁻⁴	7.19·E ⁻⁵	0.00	6.51·E ⁻⁷	4.59·E ⁻⁴			
L_3	2.68·E ⁻⁵	1.49·E ⁻⁶	9.11·E ⁻⁸	1.50·E ⁻⁶	3.86·E ⁻⁴	8.47·E ⁻⁵	0.00	2.13·E ⁻⁷	5.01·E ⁻⁴			
L_4	3.30·E ⁻⁵	1.65·E ⁻⁶	1.59·E ⁻⁷	1.96·E ⁻⁶	4.07·E ⁻⁴	9.20·E ⁻⁵	2.28·E ⁻⁹	1.12·E ⁻⁶	5.37·E ⁻⁴			
L_5		1.38·E ⁻⁶	2.96·E ⁻⁷	2.05·E ⁻⁶	4.65·E ⁻⁴	1.05·E ⁻⁴	4.17·E ⁻⁹	1.14·E ⁻⁶	6.28·E ⁻⁴			
L_6	2.73·E ⁻⁵	1.54·E ⁻⁶	5.69·E ⁻⁷	1.34·E ⁻⁶	4.14·E ⁻⁴	9.61·E ⁻⁵	0.00	6.19·E ⁻⁶	5.47·E ⁻⁴			
L_7		1.22·E ⁻⁶	1.75·E ⁻⁷	1.23·E ⁻⁶	4.24·E ⁻⁴	9.68·E ⁻⁵	0.00	2.50·E ⁻⁶	5.55·E ⁻⁴			
L_8	5.64·E ⁻⁵	1.65·E ⁻⁶	3.95·E ⁻⁷	1.50·E ⁻⁶	4.27·E ⁻⁴	9.97·E ⁻⁵	0.00	2.64·E ⁻⁶	5.89·E ⁻⁴			
L_9		1.22·E ⁻⁶	1.67·E ⁻⁷	1.32·E ⁻⁶	4.42·E ⁻⁴	6.97·E ⁻⁵	0.00	2.18·E ⁻⁶	5.46·E ⁻⁴			
L_{10}	2.85·E ⁻⁵	1.54·E ⁻⁶	7.44·E ⁻⁷	1.08·E ⁻⁶	4.21·E ⁻⁴	1.40·E ⁻⁴	0.00	4.13·E ⁻⁶	5.97·E ⁻⁴			

In studies conducted by Badeenezhad *et al.* [59] on freshwater and by Farahmandkia *et al.* [60] on milk, they suggested that HQ and HI values less than 1.0 represent a tolerable non-carcinogenic risk, and a value greater than 1 indicates a considerable risk. Conventional milk samples analyzed for adults and children do not represent a hazard or risk to consumers, and the obtained values are very low.

Table 7. Risk quotient and hazard index for heavy metal consumption in organic milk

Samples	Cr	Mn	Ni	Cu	Zn	Sr	Cd	Pb	HI			
HQ Adults												
L_1B	6.56·E ⁻³	3.70·E ⁻⁴	6.01E ⁻⁵	2.81·E ⁻⁴	3.04·E ⁻²	6.46·E ⁻³	0.00	6.83·E ⁻³	5.09·E ⁻²			
L_6B	1.35·E ⁻²	2.93·E ⁻⁴	4.92E ⁻⁵	5.62·E ⁻⁴	3.32·E ⁻²	1.45·E ⁻²	0.00	6.70·E ⁻³	6.88·E ⁻²			
L_7B	1.11·E ⁻²	7.14·E ⁻⁴	3.46E ⁻⁵	4.51·E ⁻⁴	3.43·E ⁻²	1.54·E ⁻²	0.00	6.43·E ⁻³	6.84·E ⁻²			
L_8B	7.38·E ⁻³	5.23·E ⁻⁴	6.19E ⁻⁵	4.99·E ⁻⁴	3.54·E ⁻²	1.18·E ⁻²	0.00	9.32·E ⁻³	6.50·E ⁻²			
$L_{10}B$	1.33·E ⁻²	3.44·E ⁻⁴	5.28E ⁻⁵	5.07·E ⁻⁴	3.25·E ⁻²	1.66·E ⁻²	0.00	8.40·E ⁻³	7.16·E ⁻²			
$L_{11}B$	1.34·E ⁻²	2.68·E ⁻⁴	5.10E ⁻⁵	3.45·E ⁻⁴	3.46·E ⁻²	1.03·E ⁻²	0.00	5.52·E ⁻³	6.45·E ⁻²			
			Н	IQ Childre	en							
L_1B	2.73·E ⁻⁵	1.54·E ⁻⁶	2.50·E ⁻⁷	1.08·E ⁻⁶	3.72·E ⁻⁴	4.71·E ⁻⁵	0.00	6.91·E ⁻⁷	4.50·E ⁻⁴			
L_6B	5.64·E ⁻⁵	1.22·E ⁻⁶	2.05·E ⁻⁷	2.16·E ⁻⁶	4.06·E ⁻⁴	1.06·E ⁻⁴	0.00	6.77·E ⁻⁷	5.73·E ⁻⁴			
L_7B	4.61·E ⁻⁵	2.98·E ⁻⁶	1.44·E ⁻⁷	1.73·E ⁻⁶	4.20·E ⁻⁴	1.12·E ⁻⁴	0.00	6.51·E ⁻⁷	5.84·E ⁻⁴			
L_8B	3.07·E ⁻⁵	2.18·E ⁻⁶	2.58·E ⁻⁷	1.91·E ⁻⁶	4.33·E ⁻⁴	8.63·E ⁻⁵	0.00	9.43·E ⁻⁷	5.55·E ⁻⁴			
$L_{10}B$	5.52·E ⁻⁵	1.43·E ⁻⁶	2.20·E ⁻⁷	1.94·E ⁻⁶	3.98·E ⁻⁴	1.21·E ⁻⁴	0.00	8.50·E ⁻⁷	5.79·E ⁻⁴			
$L_{11}B$	5.58·E ⁻⁵	1.12·E ⁻⁶	2.13·E ⁻⁷	1.32·E ⁻⁶	4.24·E ⁻⁴	7.51·E ⁻⁵	0.00	5.58·E ⁻⁷	5.58·E ⁻⁴			

Considering the aforementioned research by Badeenezhad *et al.* [59] on freshwater and Farahmandkia *et al.* [60] on milk and the levels imposed by them for HQ and HI, organic milk samples are classified as having a very low risk level. The organic milk samples

tested have levels below 1, indicating that they pose no danger to consumers, including both adults and children.

Risk of Developing Cancer (Table 8) and Total Carcinogenic Risk (Table 9) represent the assessment of the lifetime risk of developing cancer following exposure to heavy metals. Exposure to heavy metals, regardless of their abundance in food products, can cause disorders in the human body, which can lead to the development of various types of cancer, neurological disorders, etc. [59].

Table 8. Risk of developing cancer from heavy metals in analyzed milk samples (conventional and organic)

	CR											
Samples	(Cr	I	Ni		Cd	Pb					
_	Adults	Children	Adults	Children	Adults	Children	Adults	Children				
L_1	2.73·10-1	1.14·10 ⁻³	9.95·10-4	4.14.10-6	0.00	0.00	1.88·10-3	1.90·10 ⁻⁷				
L_2	2.28·10-1	9.49·10 ⁻⁴	9.18·10-4	3.83·10 ⁻⁶	0.00	0.00	1.56·10-3	1.58·10 ⁻⁷				
L_3	1.75·10-1	7.31.10-4	9.18·10-4	3.83.10-6	0.00	0.00	5.10-10-4	5.16.10-8				
L_4	2.17·10-1	9.02·10 ⁻⁴	1.61·10-3	6.69·10 ⁻⁶	2.26.10-4	8.65·10 ⁻⁷	2.68·10-3	2.71·10 ⁻⁷				
L_5	3.51·10-1	1.46·10-3	2.98·10-3	1.24·10 ⁻⁵	4.14.10-4	1.59·10 ⁻⁶	2.74·10-3	$2.77 \cdot 10^{-7}$				
L_6	1.79·10 ⁻¹	7.47·10-4	5.74·10 ⁻³	2.39·10 ⁻⁵	0.00	0.00	1.49·10-2	1.50·10-6				
L_7	1.90·10 ⁻¹	7.93·10 ⁻⁴	1.76·10-3	7.33·10 ⁻⁶	0.00	0.00	6.00·10 ⁻³	6.06·10 ⁻⁷				
L_8	3.70·10-1	1.54·10-3	3.98·10-3	1.66·10-5	0.00	0.00	6.35·10-3	$6.42 \cdot 10^{-7}$				
L_9	1.94·10 ⁻¹	8.09·10 ⁻⁴	1.68·10-3	$7.01 \cdot 10^{-6}$	0.00	0.00	5.23·10-3	5.29·10 ⁻⁷				
L_{10}	1.87·10-1	7.78·10-4	7.50·10 ⁻³	3.12·10-5	0.00	0.00	9.92·10-3	1.00·10 ⁻⁶				
L_1B	1.79·10-1	7.47·10-4	2.52·10-3	$1.05 \cdot 10^{-5}$	0.00	0.00	1.66·10-3	1.68·10 ⁻⁷				
L_6B	3.70·10-1	1.54·10 ⁻³	2.07·10-3	8.61·10 ⁻⁶	0.00	0.00	1.63·10-3	1.64·10 ⁻⁷				
L ₇ B	3.02·10 ⁻¹	1.26·10-3	1.45·10-3	6.06·10-6	0.00	0.00	1.56·10-3	1.58·10-7				
L ₈ B	2.02·10-1	8.40·10 ⁻⁴	2.60·10-3	1.08·10-5	0.00	0.00	2.26·10-3	2.29·10 ⁻⁷				
$L_{10}B$	3.62·10-1	1.51·10 ⁻³	2.22·10-3	9.24·10 ⁻⁶	0.00	0.00	2.04·10 ⁻³	2.06·10-7				
$L_{11}B$	3.66·10 ⁻¹	1.52·10 ⁻³	2.14·10 ⁻³	8.93·10 ⁻⁶	0.00	0.00	1.34·10-3	1.35·10 ⁻⁷				

Table 9. Total carcinogenic risk from heavy metal ingestion from analyzed milk samples (conventional and organic)

Commiss		CR _t
Samples	Adults	Children
L_1	2.75·10 ⁻¹	1.14·10 ⁻³
L_2	2.30·10 ⁻¹	9.53·10 ⁻⁴
L_3	1.77·10 ⁻¹	7.35·10 ⁻⁴
L ₄	2.21·10 ⁻¹	9.10·10 ⁻⁴
L_5	3.57·10 ⁻¹	1.48·10 ⁻³
L_6	2.00·10 ⁻¹	7.72·10 ⁻⁴
L_7	1.98·10 ⁻¹	8.01·10 ⁻⁴
L_8	3.80·10 ⁻¹	1.56·10 ⁻³
L_9	2.01·10 ⁻¹	8.17·10 ⁻³
L_{10}	2.04·10 ⁻¹	8.10·10 ⁻⁴
L_1B	1.83·10 ⁻¹	7.57·10 ⁻⁴
L_6B	3.73·10 ⁻¹	1.55·10 ⁻³
L ₇ B	3.05·10 ⁻¹	1.27·10 ⁻³
L_8B	2.06·10 ⁻¹	8.51·10 ⁻⁴
$L_{10}B$	3.66·10 ⁻¹	1.52·10 ⁻³
$L_{11}B$	3.69·10 ⁻¹	1.53·10 ⁻³

Farahmandkia *et al.* [60] reported in their study an acceptable limit for heavy metals ranging from 10^{-6} to 10^{-4} , according to EPA guidelines. The calculated carcinogenic risk for adults and children had alarming values for chromium, and in the case of adults, nickel, cadmium, and lead presented values ranging from 10^{-3} to 10^{-1} , which can endanger the lives of consumers. The summation of the carcinogenic risk of the eight heavy metals led to the establishment of the total carcinogenic risk (CR_t). The determined values range from $1.98 \cdot 10^{-1}$ to $3.80 \cdot 10^{-1}$ for adults, while the calculated values for children ranged from $1.27 \cdot 10^{-3}$ to $9.10 \cdot 10^{-4}$. The analyzed milk samples exceeded the maximum CRt values for adults, and some samples also exceeded recommended values for children, according to the guidelines suggested by Farahmandkia *et al.* [60] for CR.

Risk of polycyclic aromatic hydrocarbon ingestion

Every day, 88 % of polycyclic aromatic hydrocarbons (PAHs) are ingested through the consumption of contaminated food, while 12 % are absorbed through inhalation. PAHs, which are classified as toxic substances, are listed as priority pollutants by the International Agency for Research on Cancer (IARC), and they may pose a significant health risk to consumers [61-65].

In the study conducted by Banica *et al.*, 16 different PAHs were analyzed in 16 milk samples, both conventional and organic. The research, detailed by Banica *et al.*, used high-performance liquid chromatography with fluorescence detection to quantitatively determine the levels of PAHs. Table 10 presents the coding of the 13 PAHs included in the risk analysis.

Table 10. Coding of the 13	polycyclic aromatic hy	drocarbons used in this stu	dy [22]

Code	Polycyclic aromatic hydrocarbon
1	Naphthalene
3	Acenaphthene
4	Fluorene
5	Anthracene
6	Fluoranthene
7	Pyrene
8	Benzo[a]anthracene
9	Crysene
10	Benzo[b]fluoranthene
11	Benzo[k]fluoranthene
12	Benzo[a]pyrene
13	Dibenzo[a,h]anthracene
14	Benzo[g,h,i]perylene

The average daily dose of polycyclic aromatic hydrocarbons was calculated for the 16 processed milk samples analyzed in the previous study by Banica *et al.* [22]. The ADD was calculated for adults and children, and the results are presented in Figure 3 for conventional milk and Figure 4 for organic milk. The literature indicates that in milk samples, the amount of polycyclic aromatic hydrocarbons identified is relatively low [66]. From the results obtained for the average daily dose, children are more exposed to polycyclic aromatic hydrocarbons than adults. The highest ADD values were determined for anthracene (kg·day⁻¹) and benzo[a]anthracene (kg·day⁻¹) for conventional milk. In the

case of children, benzo[b]fluoranthene has the highest ADD values, ranging from 0.6913 kg·day⁻¹ to 5.7838 kg·day⁻¹. In the study conducted by Rawash *et al*. [67], the authors reported ADD values of 0.61 - 1.22 mg·day⁻¹ for all age groups. In a study conducted in Italy by Zhao *et al*. [68] on raw milk, ADD values ranged from 1.75 ng·day⁻¹ to 22.96 ng·day⁻¹. The ADD values presented in this study are much higher than the results reported by Rawash *et al*. [67], but on average lower than those reported by Zhao *et al*. [68].

Samples code	1	3	4	5	6	7	8	9	10	11	12	13	14
	ADD Adults [ng-kg ⁻¹ -day ⁻¹]												
L1	0.0286	0.0000	0.0276	0.0874	0.0103	10.0040	0.0586	0.0028	0.0071	0.0026	0.0000	0.0000	0.0000
L2	0.0229	0.0940	0.0173	0.0816	0.0047	0.0026	0.0223	0.0000	0.0008	0.0008	0.0000	0.0000	0.0000
L3	0.0286	0.0000	0.0099	0.0790	0.0122	10.0035	0.0336	0.0011	0.0027	0.0011	0.0000	0.0001	0.0000
L4	0.0227	0.0000	0.0282	0.0762	0.0102	0.0726	0.0000	0.0019	0.0035	0.0011	0.0000	0.0000	0.0000
L5	0.0000	0.0000	0.0270	0.1213	0.0048	0.0069	0.1145	0.0108	0.0690	0.0177	0.0029	0.0005	0.0000
L6	0.0565	0.0000	0.0254	0.1279	0.0156	10.0029	0.1112	0.0113	0.0660	0.0179	0.0000	0.0004	0.0000
L7	0.0228	0.0000	0.0274	0.0724	0.0027	0.0031	0.0219	0.0000	0.0012	0.0009	0.0002	0.0001	0.0000
L8	0.0233	0.0000	0.0320	0.0728	0.0035	10.0044	0.0238	0.0000	0.0008	0.0018	0.0000	0.0001	0.0021
L9	0.0357	0.0000	0.0248	0.0819	0.0113	0.0000	0.0382	0.0012	10.0037	0.0012	0.0000	0.0000	0.0000
L10	0.0259	0.0000	0.0267	0.0795	0.0045	0.0000	0.0311	0.0016	0.0055	0.0007	0.0000	0.0001	0.0000
					ADD	Cildren [ng	kg-1-day-1						
L1	0.1193	0.0000	0.1150	0.3640	0.0429	0.0165	0.2442	0.0117	5.7838	0.0109	0.0000	0.0000	0.0000
L2	0.0955	0.3916	0.0720	0.3399	0.0196	0.0107	0.0930	0.0000	0.6913	0.0032	0.0000	0.0002	0.0000
L3	0.1190	0.0000	0.0413	0.3292	0.0508	10.0146	0.1399	0.0044	2.2138	0.0047	0.0000	0.0005	0.0000
L4	0.0947	0.0000	0.1175	0.3175	0.0426	0.3026	0.0000	0.0081	2.8525	10.0046	0.0000	0.0000	0.0000
L5	0.0000	0.0000	0.1125	0.5053	0.0202	0.0286	0.4770	0.0450	5.6359	0.0738	0.0121	0.0022	0.0000
L6	0.2353	0.0000	0.1057	0.5331	0.0650	0.0120	0.4635	0.0471	5.3918	0.0748	0.0000	0.0018	0.0000
L7	0.0949	0.0000	0.1142	0.3016	0.0113	0.0128	0.0912	0.0000	0.9975	0.0038	0.0007	0.0002	0.0000
L8	0.0970	0.0000	0.1331	0.3031	0.0144	10.0184	0.0990	0.0000	0.6913	0.0077	0.0000	0.0004	0.0087
L9	0.1486	0.0000	0.1035	0.3412	0.0469	0.0000	0.1591	0.0049	3.0450	0.0051	0.0000	0.0000	0.0000
L10	0.1077	0.0000	0.1114	0.3314	0.0188	0.0000	0.1298	0.0068	4.4888	0.0030	0.0000	0.0005	0.0000

Figure 3. The average daily dose of polycyclic aromatic hydrocarbons ingested through the consumption of conventional milk

In the case of organic milk, the ADD values (Figure 4) for acenaphthene and benzo[g,h,i] perylene were 0.0000 ng·kg⁻¹·day⁻¹. ADD values for benzo[b]fluoranthene are much higher in children, as are the other polycyclic aromatic hydrocarbons. The ADD values for organic milk samples are higher than the results reported by Rawash $et\ al.\ [67]$, but lower than the values reported by Zhao $et\ al.\ [68]$ in their study.

Samples code	1	3	4	5	6	7	8	9	10	11	12	13	14
					ADD C	ildren [ng	·kg-1-day	1					
L1B	0.0486	0.0000	0.0351	0.0839	0.0032	0.0000	0.0277	10.0003	0.0015	0.0009	0.0000	0.0000	0.0000
L6B	0.0156	0.0000	0.0230	0.0731	0.0076	0.0009	0.0347	0.0011	0.0025	0.0000	0.0000	0.0000	0.0000
L7B	0.0205	0.0000	0.0262	0.0842	0.0026	0.0024	0.0470	0.0000	0.0069	0.0023	0.0005	0.0001	0.0000
L8B	0.0085	0.0000	0.0237	0.0794	0.0028	0.0042	0.0419	0.0000	0.0049	0.0005	0.0000	0.0000	0.0000
L10B	0.0196	0.0000	0.0251	0.0912	0.0022	0.0000	0.0732	0.0042	0.0076	0.0004	0.0002	0.0000	0.0000
L11B	0.0159	0.0000	0.0271	0.0677	0.0078	0.0000	0.0249	0.0000	0.0021	0.0000	0.0000	0.0000	0.0000
					ADD C	ildren [ng	·kg-1-day	1					
L1B	0.2024	0.0000	0.1463	0.3496	0.0133	0.0000	0.1154	0.0014	1.2250	0.0036	0.0000	0.0000	0.0000
L6B	0.0650	0.0000	0.0960	0.3045	0.0316	0.0036	0.1445	0.0044	2.0125	0.0000	0.0000	0.0000	0.0000
L7B	0.0855	0.0000	0.1093	0.3510	0.0110	0.0101	0.1957	0.0000	5.6263	0.0096	0.0022	0.0004	0.0000
L8B	0.0356	0.0000	0.0986	0.3306	0.0116	0.0174	0.1745	0.0000	3.9813	0.0021	0.0000	0.0000	0.0000
L10B	0.0817	0.0000	0.1046	0.3800	0.0092	0.0000	0.3052	0.0175	6.2388	0.0016	0.0010	0.0002	0.0000
L11B	0.0662	0.0000	0.1128	0.2820	0.0326	0.0000	0.1036	0.0000	1.7063	0.0000	0.0000	0.0000	0.0000

Figure 4. Average daily dose of polycyclic aromatic hydrocarbons ingested through the consumption of organic milk

The hazard quotient and hazard index of polycyclic aromatic hydrocarbons, as shown in Table 11, are vital parameters for evaluating the risk level to human health. HQ was calculated for naphthalene, fluorene, and benzo[a]pyrene, which have an oral reference dose (R_fD) regulated by USEPA IRIS [46-48].

The results obtained and presented for HQ and HI, calculated for adults and children in conventional and organic milk samples, are below the threshold of one, indicating no significant risk to human health, even for vulnerable populations.

Table 11. Hazard quotient and hazard index of polycyclic aromatic hydrocarbons ingestion through the consumption of conventional and organic milk

	1	4	12	HI	1	4	12	HI	
Samples		Adı	ults	I	Children				
L_1	5.73·10 ⁻⁴	1.10·10 ⁻³	0.00	1.68·10-3	2.39·10 ⁻³	4.60·10 ⁻³	0.00	6.99·10 ⁻³	
L_2	4.59·10 ⁻⁴	6.91.10-4	0.00	1.15·10 ⁻³	1.91.10-3	2.88.10-3	0.00	4.79·10 ⁻³	
L_3	5.71.10-4	3.96·10 ⁻⁴	0.00	9.68·10-4	2.38·10 ⁻³	1.65·10 ⁻³	0.00	4.03·10 ⁻³	
L_4	$4.55 \cdot 10^{-4}$	1.13·10 ⁻³	0.00	1.58·10 ⁻³	1.89·10 ⁻³	$4.70 \cdot 10^{-3}$	0.00	6.59·10 ⁻³	
L_5	0.00	1.08·10 ⁻³	$8.71 \cdot 10^{-7}$	1.08·10 ⁻³	0.00	$4.50 \cdot 10^{-3}$	3.63·10 ⁻⁶	4.50·10 ⁻³	
L_6	1.13·10 ⁻³	1.01·10 ⁻³	0.00	2.14·10 ⁻³	$4.71 \cdot 10^{-3}$	$4.23 \cdot 10^{-3}$	0.00	8.93·10 ⁻³	
L_7	4.56.10-4	1.10.10-3	$4.82 \cdot 10^{-8}$	1.55·10 ⁻³	1.90·10 ⁻³	4.57·10 ⁻³	2.01·10 ⁻⁷	6.47·10 ⁻³	
L_8	4.65.10-4	1.28·10 ⁻³	0.00	1.74·10 ⁻³	1.94·10 ⁻³	5.33·10 ⁻³	0.00	7.26.10-3	
L ₉	7.13·10 ⁻⁴	9.93·10 ⁻⁴	0.00	1.71.10-3	$2.97 \cdot 10^{-3}$	4.14.10-3	0.00	7.11.10-3	
L_{10}	5.17·10 ⁻⁴	1.07·10 ⁻³	0.00	1.59·10 ⁻³	$2.15 \cdot 10^{-3}$	$4.46 \cdot 10^{-3}$	0.00	6.61.10-3	
L_1B	9.72·10 ⁻⁴	1.40·10 ⁻³	0.00	2.38·10 ⁻³	$4.05 \cdot 10^{-3}$	5.85.10-3	0.00	9.90·10 ⁻³	
L_6B	3.12·10 ⁻⁴	9.21.10-4	0.00	1.23·10 ⁻³	1.30·10 ⁻³	3.84·10 ⁻³	0.00	5.14·10 ⁻³	
L ₇ B	$4.11 \cdot 10^{-4}$	1.05·10 ⁻³	1.58·10 ⁻⁷	1.46·10 ⁻³	1.71·10 ⁻³	4.37·10 ⁻³	$6.56 \cdot 10^{-7}$	6.08·10 ⁻³	
L ₈ B	1.71.10-4	9.46·10 ⁻⁴	0.00	1.12·10 ⁻³	7.12·10 ⁻⁴	3.94·10 ⁻³	0.00	4.65·10 ⁻³	
$L_{10}B$	3.92·10 ⁻⁴	1.00·10 ⁻³	$7.39 \cdot 10^{-8}$	1.40·10 ⁻³	1.63·10 ⁻³	$4.19 \cdot 10^{-3}$	3.08·10 ⁻⁷	5.82·10 ⁻³	
$L_{11}B$	3.18.10-4	1.08·10 ⁻³	0.00	1.40·10 ⁻³	1.32·10 ⁻³	4.51·10 ⁻³	0.00	5.83·10 ⁻³	

For the other ten polycyclic aromatic hydrocarbons, the risk coefficient and risk index could not be established. In the study conducted by Sadighara *et al.* [69], in which the health risk of benzo[*a*]pyrene was assessed, in the case of children, adolescents, and adults, it did not exceed the value of 1, so the samples analyzed do not represent a risk to the health of consumers. In the study conducted by Oni *et al.* [70] on deep water samples, the HQ values do not pose a risk to consumers, the values being < 1.0. If the HQ values were lower than 1.0, implicitly, the HI values were < 1.0 for adults and children. So, the risk index values due to the ingestion of polycyclic aromatic hydrocarbons were <1.0, and then the conclusion is that there is no danger to the lives of consumers, children, or adults. The equivalent toxicity quotient from polycyclic aromatic hydrocarbons was another alternative to determine the risk to consumer health, regarding potentially contaminated dairy products. The determinative TEQ values are presented in Figure 5 for conventional and organic milk samples.

Naphthalene was absent in only one sample (L_5) out of all the samples analyzed, which means that the TEQ is 0, and the maximum value for the other samples was 0.0053 ng·L⁻¹ (L_6). Acenaphthene and benzo[g,h,i]perylene were identified in two different samples (L_2 and L_8), and the TEQ was 0.0088 ng·L⁻¹ and 0.0019 ng·L⁻¹, respectively. Fluorene, anthracene, and fluoranthene were determined in all processed milk samples analyzed, the values obtained were between 0.0009 - 0.0030 ng·L⁻¹, 0.0676 - 0.1194 ng·L⁻¹, and 0.0003 - 0.0015 ng·L⁻¹ for conventional milk, respectively, while in

organic milk, the TEQ values determined were between 0.0022 and 0.0030 $\rm ng\cdot L^{-1}$, 0.0632 - 0.0851 $\rm ng\cdot L^{-1}$, and 0.0002 - 0.0007 $\rm ng\cdot L^{-1}$.

Sample code	1	2	4	5	6	7	8	9	10	11	12	13	14
•						[ng-L	1						
L1	0.0027	0.0000	0.0026	0.0815	0.0010	0.0004	169	0.0026	0.0661	0.0245	0.0000	0.0000	0.0000
L2	0.0021	0,0088	0.0016	0.0761	0.0004	0.0002	0.2084	0.0000	0.0079	0.0071	0.0000	0.0200	0.0000
L3	0.0027	0.0000	0.0009	0.0737	0.0011	0.0003	03134	0.0010	0.0253	0.0105	0.0000	0.0550	0.0000
L4	0.0021	0.0000	0.0026	0.0711	0.0010	0.0068	0.0000	0.0018	10.0326	0.0104	0.0000	0.0000	0.0000
L5	0.0000	0.0000	0.0025	0.1132	0.0005	0.0006	1.0684	0.0101	0.6441	0.1653	0.2710	0.2450	0.0000
L6	0.0053	0.0000	0.0024	0.1194	0.0015	0.0003	1.0382	0.0105	0.6162	0.1675	0.0000	0.2050	0.0000
L7	0.0021	0.0000	0.0026	0.0676	0.0003	0.0003	0.2043	0.0000	0.0114	0.0086	10.0150	0.0250	0.0000
L8	0.0022	0.0000	0.0030	0.0679	0.0003	0.0004	3.2217	0.0000	0.0079	0.0172	0.0000	0.0400	0.0019
L9	0.0033	0.0000	0.0023	0.0764	0.0011	0.0000	3564	0.0011	0.0348	0.0114	0.0000	0.0000	0.0000
L10	0.0024	0.0000	0.0025	0.0742	0.0004	0.0000	2907	0.0015	0.0513	0.0068	0.0000	0.0550	0.0000
L1B	0.0045	0.0000	0.0033	0.0783	0.0003	0.0000	2585	0.0003	0.0140	0.0081	0.0000	0.0000	0.0000
L6B	0.0015	0.0000	0.0022	0.0682	0.0007	0.0001	3237	0.0010	10.0230	0.0000	0.0000	0.0000	0.0000
L7B	0.0019	0.0000	0.0024	0.0786	0.0002	0.0002	03 384	0.0000	0.0643	0.0215	0.0490	0.0500	0.0000
LSB	0.0008	0.0000	0.0022	0.0741	0.0003	0.0004	3908	0.0000	0.0455	0.0048	0.0000	0.0000	0.0000
L10B	0.0018	0.0000	0.0023	0.0851	0.0002	0.0000	0.6836	0.0039	D.0713	0.0035	0.0230	0.0200	0.0000
L11B	0,0015	0.0000	0.0025	0.0632	0.0007	0.0000	2320	0.0000	0.0195	0.0000	0.0000	0.0000	0.0000

Figure 5. Equivalent toxicity coefficient of polycyclic aromatic hydrocarbons identified in conventional and organic milk

Benzo[*a*]anthracene was identified in eight conventional milk samples and three organic milk samples, being absent in the others. The highest TEQ value for benzo[*a*]anthracene was 0.0068 ng·L⁻¹, determined in sample L₄. Chrysene was absent in sample L₄, while TEQ values were the highest compared to the other polycyclic aromatic hydrocarbons determined. The TEQ values of chrysene for the ten processed milk samples in which it was identified ranged from 0.0010 ng·L⁻¹ to 0.0105 ng·L⁻¹. Samples L₅ and L₆ had values higher than 1. For the 16 and, respectively, 14 conventional and organic milk samples in which benzo[*b*]fluoranthene and benzo[*k*]fluoranthene were identified, TEQ values ranged from 0.0079 ng·L⁻¹ to 0.6441 ng·L⁻¹, and 0.0035 ng·L⁻¹ to 0.1675 ng·L⁻¹, respectively. Benzo[*a*]pyrene and dibenzo[*a*,*h*]anthracene were identified in four and nine samples, respectively, where the average TEQ values were 0.0895 ng·L⁻¹ and 0.0794 ng·L⁻¹. Goswami *et al.* [71] obtained a TEQ value for powdered milk of 0.158 ng·g⁻¹ of analyzed sample, much higher than those obtained in this study.

Statistical correlations between the determined risk parameters

Statistical analysis was performed to identify potential correlations between risk indices associated with the consumption of milk (conventional and organic) contaminated with heavy metals and polycyclic aromatic hydrocarbons. Statistical models must respond to two well-defined hypotheses:

H1: Are the risk indices significantly correlated with each other?

H2: Are the risk indices calculated for both adults and children different?

Principal component analysis (PCA) was applied to identify the correlation of risk indices for both adults and children (Figure 6). The three main components explained most of the variability in the data obtained, allowing visualization of the latent structure of the relationships between the variables.

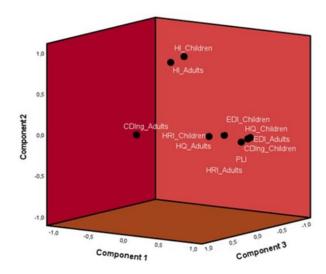


Figure 6. Distribution of variables in the principal component space after Varimax rotation (PCA) for risk indices associated with heavy metal ingestion

The distribution of variables in the three-dimensional space highlighted clear differences between the three components formed. Thus, Component 1 distinctly separated the risk indices HI Children and HI Adults from the rest of the variables, indicating that the hazard index (HI) captures a specific and relatively independent dimension of risk, which is not significantly correlated with the other indices included in the statistical analysis. Component 2 differentiated the indices CDI_{ng}_Adults, HQ_Adults, and HRI_Children from the central cluster, suggesting the existence of specific variations between adults and children in terms of intake and risk associated with heavy metals ingestion. The relatively isolated positioning of these indices suggests that this component mainly reflects intergroup differences (adults versus children), in the context of food consumption and risk response. In contrast, Component 3 brought together a compact core of correlated variables - HQ_Children, EDI_Children, EDI_Adults, CDIng_Children, PLI, and HRI_Adults, indicating a strong convergence between daily milk consumption, estimated daily intake, and risk indices associated with heavy metal ingestion. Component 3 suggests that these indices describe the magnitude of chronic exposure and composite risk in a coherent and convergent manner, with a high degree of similarity between adults and children for the variables under analysis.

Scatterplot Matrix analysis allows for the simultaneous assessment of bivariate relationships between multiple variables, providing a graphical and compact representation of all pairs formed within the statistical model. In this study, the method was used to explore the relationships between risk indices associated with heavy metal ingestion for both adults and children, depending on the metal (Cd, Cr, Cu, Mn, Ni, Pb, Sr, and Zn) identified in the analyzed milk samples (Figure 7).

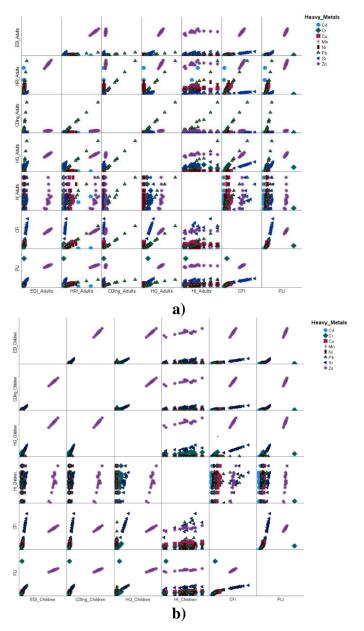


Figure 7. Scatterplot Matrix of risk indices associated with heavy metal ingestion for: a) adults and b) children

For both groups (adults and children), clear trends of positive association are observed between indices determined for the same exposure category (e.g., EDI and CDIng). In both sets of graphs, certain heavy metals, such as Pb, Sr, and Zn, appear to be concentrated in areas with higher values, suggesting a greater contribution to the total exposure. Also, the distribution of points indicates the existence of nonlinear relationships in certain pairs of variables, especially between HI and other risk indices, which may reflect the influence of other factors (e.g., heavy metal concentration and frequency of dairy product consumption). Values located outside the main group suggest the presence of samples with atypical risk profiles, which could be investigated separately. Comparing adults and children, the general pattern of correlations is similar, indicating that exposure is comparable between the two population groups, but the effects associated with heavy

metal ingestion may differ depending on the metal, the age of the consumer, the duration of exposure, etc. In the case of children, risk indices appear more concentrated towards higher values for some indices (e.g., EDI_Children and CDI_{ng}_Children).

Pairwise Comparisons allows for the rapid identification of pairs that present significant differences, facilitating the highlighting of relationships between the risk indices analyzed. In this study, the method was applied to compare the risk indices calculated for adults and children (Figure 8), to detect possible positive/negative variations related to the age of consumers, the degree of exposure to heavy metals, and the risks associated with the consumption of milk contaminated with heavy metals.

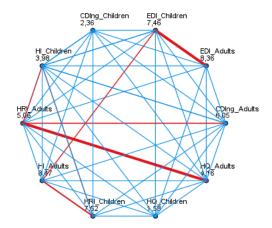


Figure 8. Pairwise comparisons between risk indices for adults and children, associated with heavy metal ingestion following the consumption of conventional and organic milk

The analysis of the network of correlations between the exposure and risk indices highlighted the existence of central nodes, acting as pivotal variables, as well as peripheral nodes, less integrated in the relational structure. The indicators EDI_Adults and HI_Children (8.36 and 8.47) and EDI_Children and HRI_Children (7.46 and 7.52) presented the highest values, confirming their role as central variables in the network. They presented positive correlations with most of the risk indices subjected to the analysis, confirming their status as essential parameters in the exposure modeling. The indices HQ_Adults (1.76) and HQ_Children (1.58) presented the lowest scores, which indicate a less central, but still relevant role as linking variables between EDI-CDI_{ng} and HRI-HI. The risk indices HI_Adults (3.82) and HI_Children (3.98) clustered together, showing a medium relationship, but relatively separate from the cluster dominated by EDI, HI, CDI_{ng}, and HRI. This confirms that HI describes a distinct dimension of risk, which does not completely overlap with the other risk indices. The differences between adults and children are notable: for adults, both EDI and CDI_{ng} show higher values, indicating a more robust correlation structure in risk modeling.

Pearson correlation analysis indicated the existence of strong and significant relationships between several risk indices. High values of the correlation coefficient (under > 0.9) indicate strong correlations between risk indices. Medium correlations are represented by r values greater than 0.6 but less than 0.9. In contrast, certain risk indices showed moderate correlations ($r \ge 0.5$) with the other indices, but weak correlations were also highlighted under r was less than or equal to 0.4. Moderate and weak correlations suggest

that the risk indices subjected to statistical analysis describe distinct dimensions of risk compared to the other indices. The graphical representation in the form of a heat map (Figure 9) highlights these groupings, emphasizing the existence of well-defined clusters between the risk indices subjected to analysis.

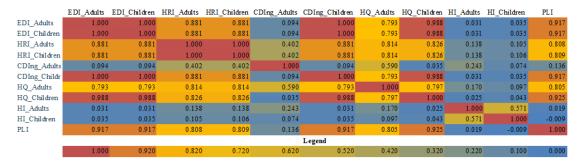


Figure 9. Heat map of the matrix of Pearson correlation coefficients for the risk index associated with heavy metal ingestion

Pearson correlation analysis graphically represented by a heat map (Figure 10) indicated the existence of a strong correlation relationship (r > 0.9) between CDI_{ng}_Adults and CDI_{ng}_Children that may suggest a degree of risk for both adults and children. Medium correlations are represented by r values greater than 0.6 but less than 0.9. In contrast, the Peason heat map was dominated by moderate ($r \ge 0.5$) and weak (r < 0.5) correlations between risk indices associated with the consumption of milk (conventional and organic) contaminated with polycyclic aromatic hydrocarbons.

	ADD_Children	ADD_Adults	CDIng_Children	CDIng_Adults	HQ_Adults	HQ_Children	TEQ	HI_Adults	HI_Children
ADD_Children	1.000	0.122	0.122	0.123	-0.074	-0.074	0.251	-0.126	-0.126
ADD_Adults	0.122	1.000	0.122	0.999	0.161	0.161	0.549	0.026	0.026
CDIng_Children	0.122	0.122	1.000	0.999	0.161	0.161	0.549	0.026	0.026
CDIng_Adults	0.123	0.999	0.999	1.000	0.164	0.164	0.554	0.028	0.028
HQ_Adults	-0.074	0.161	0.161	0.164	1.000	0.123	-0.123	0.718	0.718
HQ_Children	-0.074	0.161	0.161	0.164	0.280	1.000	-0.123	0.718	0.718
TEQ	0.251	0.549	0.549	0.554	-0.123	-0.123	1.000	-0.154	-0.154
HI_Adults	-0.126	0.026	0.026	0.028	0.718	0.718	-0.154	1.000	0.122
HI_Children	-0.126	0.026	0.026	0.028	0.718	0.718	-0.154	0.123	1.000
				Le	gend				
1.000	0.900	0.800	0.700	0.600	0.500	0.400	0.300	0.200	0.100

Figure 10. Heat map of the matrix of Pearson correlation coefficients for the risk index associated with polycyclic aromatic hydrocarbon ingestion

Scatterplot Matrix analysis allowed the simultaneous evaluation of bivariate relationships between several variables (risk indices), providing a graphical representation of the correlation mode according to the polycyclic aromatic hydrocarbons identified in the analyzed milk samples. A Scatterplot Matrix analysis was performed based on the risk indices for both adults and children (Figure 11).

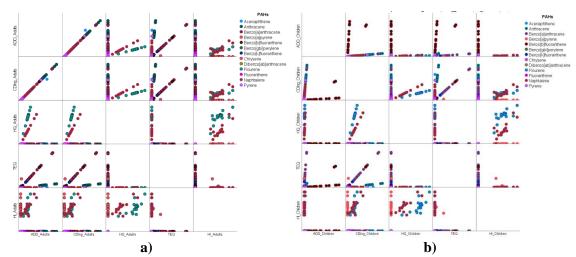


Figure 11. Scatterplot Matrix of risk indices associated with the ingestion of polycyclic aromatic hydrocarbons for: a) adults, and b) children

The Scatterplot Matrix for adults (Figure 11a) showed positive linear correlations for the 13 PAHs in the ADD and CDI_{ng} risk indices, indicating a link and dependence between the two risk indices. Values outside the main group suggest the presence of samples with atypical risk profiles, which could be investigated separately. Comparing adults and children, the general pattern of correlations is similar, indicating that exposure is comparable between the two groups of consumers, but the effects associated with the ingestion of PAHs may differ depending on the daily dose of the product consumed (750 mL for adults and 650 mL for children) and the age of the consumer. In both children and adults, the risk indices appear to be more concentrated towards higher values for some risk indices (e.g., ADD, CDI_{ng} , HQ, etc.).

To identify latent structural and group variables based on their common behavior, the data were subjected to a principal component analysis (PCA) with Varimax orthogonal rotation. The results are presented in Table 12, which clearly highlights the loadings of each variable on the extracted components, facilitating the interpretation of the main risk dimensions.

Table 12. Rotated factor loadings (Varimax) obtained through PCA for the analyzed risk indices

Risk indicators	Component					
RISK Illuicators	1	2				
HQ Children	0.926	0.071				
HQ Adults	0.926	0.071				
HI Adults	0.922	-0.079				
HI Children	0.922	-0.079				
CDI _{ng} Adults	0.122	0.975				
CDI _{ng} Children	0.120	0.974				
ADD Adults	0.120	0.974				
TEQ	-0.188	0.705				
ADD Children	-0.164	0.236				

Principal component analysis (Figure 12) with Varimax orthogonal rotation led to the extraction of two major components, which cumulatively explain a significant proportion of the total variation in the data.

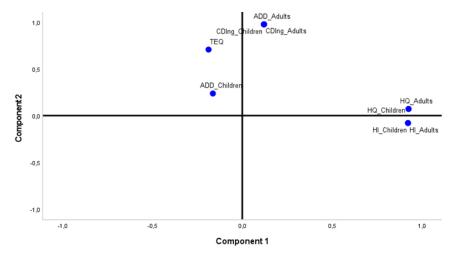


Figure 12. Distribution of variables in the principal component space after Varimax rotation (PCA) for risk indices associated with polycyclic aromatic hydrocarbon ingestion

The analysis of the pairwise correlation network (Figure 13) between the exposure and risk indices highlighted the existence of central nodes (ADD_Children and CDIng), acting as pivot variables, as well as peripheral nodes, less integrated in the relational structure (CDIng_Adults and ADD). The statistical model developed highlights a higher exposure of children and a high risk following the ingestion of polycyclic aromatic hydrocarbons, compared to adults.

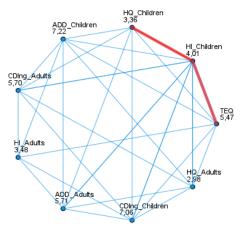


Figure 13. Pairwise comparisons between risk indices for adults and children, associations of polycyclic aromatic hydrocarbon ingestion following the consumption of conventional and organic milk

CONCLUSIONS

In line with the growing interest in the assessment of the human effects of contaminated food consumption, the results provided in this study show that a staple food category (i.e., milk) contains heavy metals and polycyclic aromatic hydrocarbons with potential toxic responses in the case of adults and children. The calculated risk indices PLI, EDI, HRI, CDIng, HQ, HI, CR, and CRt for heavy metals and ADD, CDIng, HQ, HI, and TEQ for PAHs demonstrated that exposure levels depend substantially on the product, but also on the duration of exposure (expressed in years), body mass (expressed in kg), age of consumers (expressed in years), and the recommended daily intake of a product. These findings highlight the importance of specific strategies to monitor and reduce contaminants present in foods with a high risk to consumers.

The statistical models performed in this study allowed testing and confirming the two established hypotheses: hypothesis **H1** (*Are the risk indices significantly correlated with each other?*) was confirmed by PCA analysis and pairwise comparison analysis, while hypothesis **H2** (*Are the risk indices calculated for both adults and children different?*) was also confirmed by the statistical models performed. EDI_Adults and CDI_{ng}_Adults had higher values for pairwise comparisons compared to the corresponding indicators for children, suggesting a more robust structure in adults in the case of heavy metals. Pairwise comparisons in the case of polycyclic aromatic hydrocarbons, ADD_Children and CDI_{ng}_Children were highlighted as the core of the network. Scatterplot Matrix performed on the risk indices calculated for children showed more positive linear correlations compared to the statistical model performed for adults in the case of heavy metals. In the case of polycyclic aromatic hydrocarbons, the statistical model developed for adults showed more positive linear values.

This study provides comprehensive results of the cumulative risk posed by contaminants (heavy metals and polycyclic aromatic hydrocarbons), highlighting the importance of different assessments and techniques based on product type and age category. The study also underlines the need to develop a regulatory and monitoring framework for contaminants in consumption milk, with a focus on reducing exposure of children and other vulnerable groups (e.g., the elderly). The study shows how useful statistical models are for identifying and describing the links between risk indices and for strengthening evidence-based food safety regulations.

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