Original article

Heavy metal residues in milk and dairy products produced in Northern Cyprus

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Abstract. *Background:* The present study aimed to determine the heavy metal concentrations (^{75}As, ^{111}Cd, ^{208}Pb, ^{65}Cu, ^{200}Hg) in milk and dairy products produced in Northern Cyprus with a view to evaluating the daily metal intake and potential health risks. *Method:* The study was conducted in a total of 616 individuals between the ages of 18 and 65, and the average daily consumption of milk and dairy products, in addition to body weight, were determined. Heavy metal concentrations in raw milk (cow, sheep, goat) and the most commonly consumed packaged milk products (UHT milk, yogurt, halloumi, lor, kashar, and white cheese) were determined by inductively coupled plasma-mass spectrometry (ICP-MS). Samples were taken from each product in triplicate, resulting in the analysis of a total of 27 samples. For evaluation of the potential health risks, the Health Risk Index (HRI) and Total HRI (THRI) were calculated using the results of packaged dairy product consumption, daily intake of metal, and oral reference level (RfD) (HRI<1, THRI<1 safe limits). Moreover, heavy metal concentrations were compared with maximum permissible limits (MPL). *Results*: As residue was detected only in raw sheep's milk samples (0.33±0.58 μg/kg). The average As, Cd, Pb, Cu, and Hg residues in packaged products were 4.55±6.19 (range: 0.00-25.00 μg/kg), 14.44±20.65 (range: 0.00-58.00 μg/kg), 6.83±19.03 (range: 0.00-81.00 μg/kg), 237.38±215.22 (range: 0.00-738.00 μg/kg), and 4.61±6.37 (range: 0.00-24.00 μg/kg), respectively. In addition, the calculated HRI and THRI values were below 1. *Conclusions:* According to the HRI and THRI results, no possible health risks were determined with respect to consumption of the analyzed products. Cd residue was higher than the MPL in yogurt, halloumi, and UHT milk samples, while Pb residue was higher than the MPL in halloumi samples, and Cu residue was higher than the MPL in yogurt, halloumi, white cheese, kashar, and lor samples. It is important to control all steps that may cause heavy metal contamination during the processing of milk.

Keywords: Milk consumption, heavy metals, public health nutrition, food safety, potential health risk

Introduction

Cyprus is an island known as an archaeological copper deposit, where mining activities took place in the past. Approximately 10 million tons of waste, including heavy metals, is thought to contaminate the soil surrounding disused copper mines (1). A study conducted by Akun et al., in 2010 supports this view, and in certain regions across the island, a high amount of lead (*Pb*), cadmium (*Cd*), and arsenic (*As*) contamination has been found. In particular, the level of arsenic across the island is higher than the limit determined by the European Productivity Agency (EPA) (2). Heavy metal pollution in the soil poses a risk to food production. *As*, *Pb*, *Cd*, and mercury (*Hg*) are among the heavy metals that substantially threaten food safety. Moreover, although necessary for human nutrition, copper (*Cu*) has toxic effects when consumed in excessive amounts (3,4).

Plants grown in soil or watered containing high concentrations of heavy metals may easily absorb these metals from soil and/or water and store them in their edible parts (1,5). Heavy metals accumulate in the tissues of mammals consuming these plants (6), such as in the mammary glands, which are an active part of the body, and pass to the milk (7). Thus, heavy metals can enter the human body by consumption of meat and milk obtained from animals fed with these plants or directly from the plants themselves (6). Prolonged natural or anthropogenic heavy metal contamination shows potentially carcinogenic, mutagenic, teratogenic, and endocrine-disrupting effects that cause long-term health impacts. Therefore, it is important to determine the heavy metal concentration in high-risk food groups and take the necessary measures (8). Milk and dairy products are a high-risk food group in terms of heavy metal contamination. Access to milk and dairy products is cheaper and easier as compared with many food sources (7,8); therefore, it is imperative to produce, preserve, and sell milk and dairy products under hygienic conditions. Heavy metals can pass directly to milk and dairy products from the animal, as well as from the equipment used during milk storage and processing. During the production of certain types of cheese, the corrosion that develops due to the acidic environment caused by the fermentation of milk increases the risk of contamination (7,9,10). Moreover, depending on the method employed in the production phase and the amount of milk used, the heavy metal concentration in different dairy products may vary (6,11,12).

In studies conducted in different countries (10,13-20), heavy metal residues have been identified in milk and dairy products; however, no such study has been performed in Northern Cyprus. The present study aimed to evaluate the heavy metal concentration, daily metal intake, and potential health risks associated with milk and dairy products produced in Northern Cyprus.

Materials and methods

Study population

The study population consisted of 616 male (308) and female (308) aged between 18 to 65 adults (mean age 38.0±12.5). Eligible participants were selected by stratified random sampling method from different cities (Nicosia, Famagusta, Kyrenia, Guzelyurt, Iskele) in North Cyprus between the November of 2018 and June of 2019. Total population of the country between 18-65 ages is 200,107. Since reaching the whole research universe will be difficult in terms of time, cost and control, sample selection was determined with a 95% confidence level and 4% sampling error to represent the study universe.

This study was conducted according to the guidelines laid down in the Declaration of Helsinki and all procedures involving human subjects were approved by the Ethics Committee at the Eastern Mediterranean University (date:13.12.2017, decision no: ETK00-2017-0263). Written informed consent was obtained from all subjects.

Milk and dairy product consumption

A survey including demographic information of participants and the frequency and habits of milk and milk product consumption was applied using the one-on-one interview technique. The daily consumption of milk and dairy products by individuals was determined using one-month retrospective consumption frequency survey (21).

Heavy metal analysis

According to the survey results, the most commonly consumed packaged products (full-fat UHT milk, full-fat yogurt, halloumi, kashar, lor and white cheese) and also unprocessed raw milk (cow, goat, and sheep) were analyzed in terms of their *As*, *Cd*, *Pb*, *Cu*, and *Hg* concentrations. The products were obtained from a supermarket in the capital city Nicosia, while the unprocessed raw milk was obtained from the collection center of all companies that produce packaged milk and dairy products. A total of nine products were analyzed, including unprocessed raw cow's, sheep's, and goat's milk, packaged ultra-high-temperature (UHT) full-fat milk, full-fat yogurt, lor (type of traditional curd cheese), halloumi, kashar, and white cheese. Three different samples of each product were analyzed (3 different packages of the same brand were analyzed for each product). As a result, 27 samples were analyzed totally. The heavy metal content of milk may differ depending on the seasonal differences, therefore products that were collected in the same season (October) were analyzed to avoid seasonal differences. All products were stored at 4°C until the time of analysis. For heavy metal analysis, 0.5 g each sample was placed in a Teflon container and digested with acid [7 ml nitric acid (HNO_3, %65) and 1 ml hydrogen peroxide (H_2O_2, %30)] in a microwave oven (Milestone START D) [Operation Parameters as follows; Power 1000W, Ramping temperature (10:00 min.), Temperature (200°C), Holding time (20:00 min.)]. Samples were diluted to 50 mL with distilled water for standardization and subsequently placed in an Agilent 7500ce series inductively coupled plasma mass spectrometer (ICP-MS) for heavy metal analysis (*As, Cd, Pb, Cu*, and *Hg*) (22). Using the certified reference standard, a standard solution was prepared for each element of interest (High-Purity Standards, Charleston, U.S.). For the quantitative analysis of dairy products, calibration graphics were created using standards at the concentrations determined for each element. The working concentration for *As, Cu, Cd* and *Pb* was 5.0, 10.0, 30.0, 50.0, 100.0 µg/L whereas it was 0,5, 1.0, 2.0, 3.0 µg/L for *Hg*. All samples were duplicated, read three times, and the mean results were calculated. Since there are no country-specific recommended maximum limit values for heavy metal residues, the *Pb* residue in the products was compared to the maximum permissible limit (MPL) specified by the Codex Alimentarius commission while the *As* and *Hg* residue levels were compared to the MPL specified by Arianejad et al, and the *Cd* and *Cu* residues were compared to the MPL specified by Sobhanardakani. The MPL values specified for *As*, *Cd*, *Pb*, *Cu*, and *Hg* were 140.0 µg/kg, 2.6 µg/kg, 20.0 µg/kg, 10.0 µg/kg, and 500.0 µg/kg, respectively (23-25).

Body weight measurement

The body weight of each individual, without shoes and wearing lightweight clothing, was measured after at least three hours of fasting using a Tanita SC 330 device sensitive to 0.1 kg (26).

Health risk assessment

The potential harmful effects of heavy metals consumed with foods were evaluated using the Health Risk Index (HRI). An HRI value greater than 1 is an indication of potentially negative effects on health (25,27,28). To calculate the HRI level, an individual's Daily Intake of Metal (DIM) must be known. The DIM is calculated by multiplying the heavy metal level in food (µg/kg) by the average daily amount of food consumed (kg) and a conversion factor (0.085), followed by division by the average body weight (kg) (25,27,28). The HRI is calculated by the ratio of the DIM determined by the heavy metal content of the food of interest to the oral reference level (RfD). For RfD, values determined by The Joint FAO/WHO Expert Committee on Food Additives (JECFA) and United States Environmental Protection Agency (US-EPA) were used. The RfD values determined for *As, Cd, Pb, Cu*, and *Hg* were 0.3 µg/kg Bw/day, 1.0 µg/kg Bw/day, 3.5 µg/kg Bw/day, 40.0 µg/kg Bw/day and 0.6 µg/kg Bw/day, respectively (29,30). Moreover, by calculating the HRI values, the Total HRI (THRI) value was obtained for each metal (25,27).

$$DIM = (C_{metal} \times C_{Factor} \times D_{nutrient\ intake}) \div BW$$

C_{metal} = Heavy metal level in food (µg/kg)

$D_{nutrient\ intake}$ = Amount of food consumed (kg)

C_{factor} = Conversion factor

BW = Average body weight (kg)

RfD: Oral reference intake level

Statistical analysis

Evaluation of the data was carried out using the Statistical Package for Social Sciences SPSS 20.0 program. The average daily consumption of milk and dairy products by the participants and the heavy metal levels in the analyzed products were determined using descriptive statistics. In statistical comparisons, compatibility with a normal distribution was examined by the Kolmogorov-Smirnov test. Since the dataset did not show a normal distribution, the Kruskal-Wallis test was used for multiple comparisons, in which the distribution of heavy metal residues in milk and dairy products was examined. Moreover, the Mann-Whitney

Table 1: Heavy Metal Concentrations (μg/kg) in the Most Commonly Consumed Packaged Products

		As	Cd	Pb	Cu	Hg
	n	X̄±SD (Min-Max)	X̄±SD (Min-Max)	X̄±SD (Min-Max)	X̄±SD (Min-Max)	X̄±SD (Min-Max)
Full-fat UHT milk	3	2.33±2.51 (0.00-5.00)	5.00±8.66 (0.00-15.00)	2.66±4.61 (0.00-8.00)	N.D	3.66±3.21 (0.00-6.00)
Full-fat yogurt (g)	3	6.00±6.00 (0.00-12.00)	36.33±16.66 (23.00-55.00)	3.00±3.60 (0.00-7.00)	15.66±16.56 (0.00-33.00)	6.00±4.00 (2.00-10.00)
Halloumi cheese (g)	3	12.33±11.37 (3.00-25.00)	44.33±13.50 (31.00-58.00)	35.33±39.62 (10.00-81.00)	591.33±137.24 (466.00-738.00)	15.00±9.00 (6.00-24.00)
Kashar cheese (g)	3	0.33±0.57 (0.00-1.00)	0.33±0.57 (0.00-1.00)	N.D	270.06±308.92 (235.00-289.00)	2.00±2.64 (0.00-5.00)
White cheese (g)	3	5.66±1.15 (5.00-7.00)	0.66±1.15 (0.00-2.00)	N.D	300.66±132.09 (182.00-443.00)	1.00±1.73 (0.00-3.00)
Lor cheese (g)	3	0.66±0.57 (0.00-1.00)	N.D	N.D	246.00±47.28 (212.00-300.00)	N.D
Total	18	4.55±6.19 (0.00-25.00)	14.44±20.65 (0.00-58.00)	6.83±19.03 (0.00-81.00)	237.38±215.22 (0.00-738.00)	4.61±6.37 (0.00-24.00)
P-value		0.097	*0.019	*0.025	*0.012	*0.039

*$P < 0.05$
(N.D. = not detectable)

U test was employed for pairwise comparisons. The Spearman correlation test was performed to understand the relationship between heavy metal residues in the milk and dairy products. Statistical significance was set at $p<0.05$.

Results

Heavy metal concentrations of the analyzed products were divided into two groups, raw milk and packaged products. It was determined that the concentration of heavy metals in raw milk was undetectable, with only a low amount (0.33±0.58 μg/kg) of *As* residue in sheep's milk. Examination of the packaged products showed that no *Cd*, *Pb,* or *Hg* residues were detected in lor samples, no *Pb* was detected in white cheese or kashar samples, and no *Cu* residue was detected in UHT milk samples. Moreover, it was found that *As* residue in kashar and lor samples, and *Cd* residue in white cheese and kashar samples were negligible (table 1). Comparison of the heavy metal concentrations in the most commonly consumed products determined that *Cd*, *Pb*, and *Hg* residues in halloumi samples were high, whereas *Cu* residues in yogurt samples were low as compared with other products ($P<0.05$).

The mean age and body weight of the individuals were 38.0±12.5 years and 73.2±13.7 kg, respectively. The daily consumption of the most commonly consumed packaged products and heavy metal analysis are given in table 2. Considering the mean consumed amounts, the most commonly consumed products were found to be full-fat UHT milk, full-fat yogurt, halloumi, lor, kashar, and white cheese.

The HRI values of consumed products and the THRI values of heavy metals are given in table 2. Since raw milk is not consumed directly, the HRI values of raw milk were not calculated. Because of that only packaged products were calculated. The HRI values calculated for UHT milk, yogurt, halloumi,

Table 2. Daily Consumption of the Most Commonly Consumed Packaged Dairy Products (n = 616) and Daily Intake of Metal (DIM, µg) and Health Risk Index (HRI) Values Calculated According to the Amount of Heavy Metal Ingested by Consuming Packaged Products

	X̄±SD (Min-Max)		As	Cd	Pb	Cu	Hg	THRI
Full-fat UHT Milk (ml)	81.7±145.1 (3.3-1000.0)	DIM	$2.18x10^{-4}$	$4.73x10^{-4}$	$2.56x10^{-4}$	N.D	$3.51x10^{-4}$	$1.25x10^{-3}$
		HRI	$7.26x10^{-4}$	$4.74x10^{-4}$	$7.31x10^{-5}$	N.D	$8.76x10^{-5}$	
Full-fat Yogurt (g)	50.7±62.8 (1.2-337.5)	DIM	$3.53x10^{-4}$	$2.14x10^{-3}$	$1.77x10^{-4}$	$9.23x10^{-4}$	$3.53x10^{-4}$	$3.19x10^{-3}$
		HRI	$1.18x10^{-3}$	$2.14x10^{-3}$	$5.04x10^{-5}$	$2.31x10^{-5}$	$8.83x10^{-5}$	
Halloumi cheese (g)	28.5±26.6 (0.8-125.0)	DIM	$4.07x10^{-4}$	$1.47x10^{-3}$	$1.17x10^{-3}$	$1.96x10^{-2}$	$4.97x10^{-4}$	$3.47x10^{-3}$
		HRI	$1.36x10^{-3}$	$1.47x10^{-3}$	$3.34x10^{-4}$	$4.89x10^{-4}$	$1.24x10^{-4}$	
K*as*har cheese (g)	11.9±17.1 (0.7-100.0)	DIM	$4.16x10^{-6}$	$4.14x10^{-6}$	N.D	$3.74x10^{-3}$	$2.76x10^{-5}$	$1.08x1^{0-}4$
		HRI	$1.38x10^{-5}$	$4.14x10^{-6}$	N.D	$3.74x10^{-3}$	$6.91x10^{-6}$	
White Cheese (g)	9.9±16.8 (0.8-150.0)	DIM	$6.61x10^{-5}$	$8.12x10^{-5}$	N.D	$3.49x10^{-3}$	$1.16x10^{-5}$	$2.93x10^{-4}$
		HRI	$2.21x10^{-4}$	$8.12x10^{-5}$	N.D	$8.7x10^{-5}$	$2.91x10^{-6}$	
Lor cheese (g)	12.0±40.8 (1.2-300.0)	DIM	$1.01x10^{-5}$	N.D	N.D	$3.55x10^{-3}$	N.D	$1.12x10^{-4}$
		HRI	$3.37x10^{-5}$	N.D	N.D	$8.8x10^{-5}$	N.D	

(N.D. = not detectable)

kashar, white cheese, and lor were between $2.91x10^{-6}$ and $2.14x10^{-3}$, and the THRI values were between $1.08x10^{-4}$ and $3,47x10^{-3}$.

Discussion

There are many studies evaluating the heavy metal content of milk and dairy products (10,13-20). The main objective of this study was to determine the content of milk and dairy products in Northern Cyprus and to estimate the potential human health risk factors from individual consumption. In the present study, it was determined that heavy metals in raw cow's, sheep's, and goat's milk were at undetectable levels, and only *As* (0.33 ± 0.58 µg/kg) residue was determined in sheep's milk (data not shown). Similar studies conducted in different countries have reported that *As* and *Hg* levels are undetectable in cow's milk samples in Japan and China (31). In a study examining goat's and sheep's milk in Italy, it was found that there were residues of 0.16±0.14 and 0.24±0.10 mg/kg *As*, 0.71 and 0.34±0.10 mg/kg *Cu*, and 0.06±0.02 mg/kg *Pb* (32). In another study examining cow's and goat's milk in Nigeria, it was found that there were 30.0±1.6 and 3.5±0.5 µg/kg *Pb* and 2.1±0.7 and 4.5±0.5 µg/kg *Cd* residues in milk (33). Environmental conditions, water, and feed used on farms are the main factors that affect the heavy metal content in raw milk. Using uncontaminated water and feed under hygienic conditions significantly reduces the risk of heavy metal contamination in raw milk (34).

In the present study, it was determined that the heavy metal residue in packaged products was higher than that in the milk used for production ($P<0.05$). Increasing the processing steps during the production of products increases the risk of heavy metal contamination from the equipment used. Raw milk according to the type of product to be produced; It goes through many different stages such as clarification, separation, standardization, homogenization, filtration, pasteurization, sterilization, deodorization, fermentation, scalding, maturation, packaging, preparation, cooling, storage and distribution. The structure and quality of the tools and equipments used in these stages are very important. During these stages, coating of the water or milk used in the production phase of the tanks with lead lining, polishing pots or cans with lead and using lead pipes, the use of copper alloy equipment, the use of cadmium-plated metal equipment are among the main causes of equipment-borne contamination. In addition, during the production of certain types of cheese, the corrosion that develops due to the acidic environment caused by the fermentation of milk increases the risk of heavy metal contamination (7,9,10,16). Moreover, the heavy metal concentration in dairy products, especially cheese, is associated with an increase in dry matter values. Maas et al. associated higher heavy metal concentrations in Comte cheese as compared with the milk used for its production with an increase in dry matter and processing stages (13). A study by Enb et al. supports these results, demonstrating that the heavy metal concentration is increased by 5.9 to 9.0 times in butter as compared with cow's milk, 6.7 to 9.2 times in samna, and 1.3 to 1.5 times in cream (10).

Comparison of the heavy metal concentrations in packaged products showed that the levels of heavy metal residues differed according to the product (table 1). The use of different ratios of cow's, goat's, and sheep's milk, and the fact that they are produced using different methods and equipment, may lead to variation in heavy metal levels in products. Comparison of the average residue values in the products with the MPL showed that *Cd* in yogurt, halloumi, and UHT milk samples, *Pb* in halloumi samples, and *Cu* in yogurt, halloumi, white cheese, kashar, and lor samples were higher than the MPL (16,23-25).

Lead, which is considered one of the most important toxic heavy metals that threatens human health, was detected only in UHT milk (2.66±4.61 μg/kg), yogurt (3.00±3.60 μg/kg), and halloumi (35.33±39.62 μg/kg). Although the average *Pb* residue in yogurt and UHT milk samples was below the safety limit, the average residue level found in halloumi samples was higher than the MPL (20 μg/kg) (23). *As* a result of non-environmentally friendly industrialization, lead contaminates foods through the air, water, and soil (35,36). Coating the inner side of tanks used to transport water or milk with lead, polishing pots or cans with lead, and using lead pipes increase the risk of contamination (16). In comparison with the present study, similar studies have detected a higher amount of *Pb* residue in UHT milk produced in Egypt and Pakistan and in yogurt produced in South Korea and Iran. In their study in Egypt, Abou-Arab et al. reported an average of 20.00±20.00 μg/kg Pb in UHT milk, while Akhtar et al. reported an average of 190±137 μg/kg *Pb* in UHT milk produced in Pakistan (37, 38). Khan et al. reported an average of 7.54±1.76 μg/kg *Pb* in yogurt produced in South Korea, and Shahbazi et al. reported an average of 7.54±1.76 μg/kg *Pb* residue in yogurt produced in Iran (39,40). In a study by Ayar et al. conducted in Turkey kashar, white cheese, yogurt, lor, and milk samples *Pb* residue levels were above the MPL (16). It has also been reported in studies carried out in different cities in Turkey that *Pb* residue in kashar samples is higher than the MPL (41-43). In another study conducted in Greece, *Pb* residue was not detected in kashar samples (44), which is in accordance with the results of the present study.

Cadmium, another important contaminant, has been determined at certain levels in packaged products, with the exception of lor, while the average *Cd* levels detected in halloumi (44.33±13.50 μg/kg), yogurt (36.33±16.66 μg/kg), and UHT milk (5.00±8.66 μg/kg) samples were found to be above the MPL (2.6 μg/kg), and the *Cd* levels determined in kashar (0.33±0.57 μg/kg) and white cheese (0.66±1.15 μg/kg) samples were at safe levels (25). *Cd* is a metal commonly found in nature, usually carried around industrial sites by air and water. Food is contaminated through the soil, water, and feed. Moreover, the use of *Cd*-coated metal equipment, irrigation of agricultural areas with sewage water, and the use of fertilizers and feeds containing *Cd* in production areas are the main factors that increase the risk of contamination (16,45). In the present study, *Cd* residue found in white cheese and kashar was low, almost undetectable. Similarly, in a study conducted in Turkey, packaged and unpackaged white cheese, kashar, and milk, had almost no *Cd* residue (11). In a study conducted in another city in Turkey, there was no *Cd* residue in white cheese samples (46). On the contrary, in a study conducted by Ayar et al. in Turkey, the average *Cd* residue levels were higher in white cheese (12.00±16.00 mg/kg), kashar (29.00±23.00 mg/kg), and lor samples (23.00±36.00 mg/kg). However, the average *Cd* residue (9.00±11.00 µg/kg) in the analyzed yogurt samples has been reported to be lower as compared with those determined in the present study (16). Similarly, a lower amount of *Cd* residue (1.36±0.022 µg/kg) was found in yogurt samples examined in South Korea (39).

A positive correlation w*as* found between *Cd* and *Hg* residues in packaged products (P<0.05, r=0.485). Similar to *Cd*, the highest *Hg* residue was found in halloumi samples (15.00±9.00 µg/kg) and there were no *Hg* residues detected in lor samples. Since these metals contaminate foods through similar routes and are common in nature, the levels of residues detected were thought to be similar. A previous study determined that yogurt, UHT milk, and kashar samples had an average of 6.00±4.00, 3.66±3.21, and 2.00±2.64 µg/kg *Hg* residue, while that in white cheese samples could be regarded as negligible (1.00±1.73 µg/kg). *Hg* residues in all products were found to be lower than the specified limit level (500 µg/kg) (24). Christophoridis et al. reported that kashar samples in Greece contain no *Hg*, and Demirözü-Erdinç and Saldamlı showed that white cheese samples in Turkey contain no *Hg* (44,47). In a study carried out in Iran, it was found that there was 23.8±10.9 and 27.2±11.6 µg/kg *Hg* residue in yogurt and cheese samples, respectively (48).

Although there were almost negligible levels of *As* residue in kashar and lor samples, a certain amount of *As* residue was found in packaged products (table 1). Moreover, despite the amount of arsenic in the soil on the island being higher than the limit level determined by EPA (2), *As* residue in the products analyzed within the scope of the present research was found to be below the limit level (140 µg/kg) (24). It was determined that the average *As* residue level in UHT milk, yogurt, halloumi, and white cheese samples was 2.33±2.51, 6.0±6.0, 12.33±11.37, and 5.66±1.15 µg/kg, respectively. Since contamination generally occurs through water, controlling the water used in product production and on farms reduces the risk of *As* contamination. Moreover, while the contamination of food with *As* has been at higher levels in recent years due to the drugs used in agriculture, the risk of contamination has decreased since the use of such insecticides, herbicides, and pesticides has been banned (49). Similar studies in the literature show that the *As* residue in analyzed products can be deemed negligible or not high-risk (11,44,46). İstanbulluoğlu et al. (2013) reported that, although the amount of *As* was found at higher levels in unpackaged milk, kashar, and white cheese samples as compared with those in packaged products, these levels pose no risk. Further, Christophoridis et al. (2019) reported that there was no *As* residue in kashar samples, while Öksüztepe et al. (2013) reported no *As* residue in white cheese samples.

Although no *Cu* residue was detected in milk samples used for product production, the *Cu* residue in packaged products other than UHT milk was found to be very high. The mean *Cu* residue values in yogurt, halloumi, kashar, white cheese, and lor samples were 15.66±16.56, 591.33±137.24, 270.06±308.92, 300.66±132.09, and 246.00±47.28 µg/kg, respectively. These values were higher than the MPL (10.0 µg/kg) (25). *Cu* contamination was thought to occur from the copper-alloy equipment used during the production phase, since the *Cu* residue in the products was much higher than that in the raw milk used for product production. Although stainless steel boilers are more advantageous in terms of health, manufacturers generally prefer copper boilers because they are cheaper. Despite copper boilers being tinned well at certain intervals, copper contamination cannot be prevented, and *Cu* contamination occurs in the products (50). In a study by Öksüztepe in Turkey, the average *Cu* residue in cheese samples was determined to be 340.00±60.00 mg/kg (46). Similarly, in kashar samples examined in different cities in Turkey, the average *Cu* residue level was higher than that obtained in the present study (41,42,43,49). It was found that kashar samples in Greece also have higher *Cu* residue levels as compared with those obtained in the present study (44). Moreover, *Cu* residue levels in yogurt samples in South Korea and Iran were found to be higher as compared with those obtained in the present study. Khan et al. reported an average of 157.60±0.13 µg/kg *Cu* residue in yogurts in South Korea, while Shahbazi et al. reported an average of

399.00±125.00 μg/kg *Cu* residue in yogurts in Iran (39,40). These results are in accordance with those of the present study, in which the *Cu* residue in cheese samples was high.

The HRI values calculated for UHT milk, yogurt, halloumi, kashar, white cheese, and lor were between 2.91×10^{-6} and 2.14×10^{-3}, and the THRI values were between 1.08×10^{-4} and $3,47 \times 10^{-3}$ (below 1), and it was determined that the consumed products do not pose any health risks. Similarly, in a study by Sobhanardaki in Iran, heavy metal levels in raw and pasteurized cow's milk were determined, and the calculated HRI values posed no health risks (25).

Weak positive correlations were found between *As* and *Hg* ($r=0.489$, $P=0.9$) and between *Cd* and *Hg* ($r=0.485$, $P=0.041$) in packaged dairy products; a moderate correlation was found between *Pb* and *Hg* ($r=0.629$, $P=0.005$); and a strong positive correlation was found between *Cd* and *Pb* ($r=0.757$, $P=0.00$). It has been reported that there is a positive correlation between *Hg* and *Cd* levels in packaged milk (11). Moreover, Sobhanardaki found that there is a positive correlation between *Cd* levels and *Cu* and *Pb* levels in pasteurized milk (25). Accordingly, it can be argued that *Cd*, *Hg*, and *Pb* contaminate milk and dairy products in a similar manner.

There are some limitations in the present study. Firstly, only *As, Cd, Pb, Hg,* and *Cu* residues were evaluated due to the high cost of analysis and limited budget; however, it is very important to consider other heavy metals such as; chromium, nickel, cobalt etc. because that may pose potentially carcinogenic, mutagenic, teratogenic, and endocrine-disrupting effects that cause long-term health impacts. Heavy metal analysis of other dairy products, in addition to products with the highest daily consumption, will contribute to a better assessment of the potential health risks. Moreover, increasing the number of samples and the number of brands will be advantageous in terms of determination of heavy metal residues. Within scope of the study only the most preferred brands of products were analyzed. Only one brand is not enough to generalize the products. Because different brands may have different heavy metal contents according to the methods and equipments that were used during productions steps. Since the public doesn't consume the only one brand, analyzing all brands consumed by the public may be more accurate in assessing potential health risks. Unfortunately, the relationship between biochemical parameters of surveyed individuals and heavy metal intake could not be evaluated. Comparing the parameters of diseases with daily consumption of heavy metals may be more effective for realizing the relations between the diseases and heavy metals. Since there are no country-specific MPL values for heavy metal residues, the levels of heavy metals were evaluated according to the MPL values given in similar studies. It is recommended that the foods produced and consumed are maintained under control at certain intervals by determining the country-specific MPL values against the negative effects of heavy metals. Moreover, by increasing the variety and number of samples analyzed in subsequent studies, it will be beneficial to evaluate other heavy metals that may pose a risk to human health, and to evaluate the relationship between the intake of heavy metals and human health in more detail with respect to biochemical parameters in the blood.

Conclusion

In conclusion, it was determined that the heavy metal concentration in milk used for the production of products was undetectable, and only a small amount of *As* residue was detected in sheep's milk. However, it was observed that the heavy metal concentration increased with the number of milk processing stages. In comparison with the raw milk used for product production, the amount of heavy metals in packaged products was higher. The highest level of heavy metals among the packaged products was found in halloumi samples. *Cd* residues in yogurt, halloumi, and UHT milk samples, *Pb* in halloumi samples, and *Cu* in yogurt, halloumi, white cheese, kashar, and lor samples were higher than the MPL. Considering these results, it is recommended that all steps during milk processing be checked. Moreover, evaluation of the heavy metal residue in products with the highest daily consumption showed that the heavy metal residue was not present at a level that negatively affected health. Nevertheless, to avoid the negative effects of heavy metals, it is recommended that foods with a high level of heavy metal residues be identified and consumed at a minimum level. The present study can be a guide for future comprehensive studies with a high number of samples.

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