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Simultaneous Differential Pulse Polarographic Determination of Cadmium, Lead, and Copper in Milk and Dairy Products

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The contents of potentially toxic elements lead and cadmium and the essential element copper in various milk and dairy products consumed in Turkey were determined by differential pulse polarography (DPP), primarily to assess whether the intakes comply with recommended desired concentrations for essential and permissible levels for toxic elements. A simple and rapid DPP method has been developed for the simultaneous determination of cadmium, lead, and copper in samples. Using the differential pulse mode, half-wave peak potentials as $E_{1/2}$ were −0.58, −0.40, and −0.07 V for cadmium (Cd), lead (Pb), and copper (Cu), respectively. Marketed formulations of dairy products have been analyzed by calibration and standard addition methods. Recovery experiments were found to be quantitative. The linear domain ranges were 0.00−674.28 μg/L for Cd ($R^2 = 0.9999$), 0.19−2.94 mg/L ($p < 0.01$) for Pb ($R^2 = 0.9997$), and 0.41−133.46 μg/L for Cu ($p < 0.01$) ($R^2 = 0.9999$). The studies have shown that the method is a rapid, reproducible, and accurate determination of these elements in milk and dairy products and can be used in the analysis of marketed formulations in the milk and dairy industry.

KEYWORDS: **Milk; dairy; differential pulse polarography (DPP); Cd; Pb; Cu**

INTRODUCTION

Environmental pollution is the main cause of heavy metal contamination such as cadmium (Cd), lead (Pb), and copper (Cu) in the food chain. All foodstuffs, especially products of animal origin, present the problems of hygienic safety with increasing environmental pollution (*1*). Therefore, it is of prime importance to determine the concentrations of essential and toxic elements in foods. Milk is an important food of animal origin as it has all of the nutrients necessarry for a healthy diet. Furthermore, milk and dairy products are the main or even the only foods for some consumer groups, such as infants and the elderly.

The need for regular monitoring of toxic and essential trace elements in milk and dairy foods has led to an increasing demand for adequately sensitive and selective analytical techniques with multielement capabilities. Current techniques capable of multielement determination, highly sensitive neutron activation techniques, atomic emission spectrometry with inductively coupled plasma excitation (ICP-AES), X-ray fluorescence (XRF), and atomic absorption spectrometry (AAS), are very expensive, time-consuming, and often do not offer adequate sensitivity for reproducible determination at trace to ultratrace concentrations of multielements in biological materials (*2*).

Voltammetric techniques such as differential pulse polarography (DPP), anodic stripping voltammetry (ASV), cathodic stripping voltammetry (CSV), and adsorption voltammetry (AV) require relatively inexpensive instrumental analysis methods and are capable of accurate multielement determination at trace to ultratrace levels (*3*−*5*).

DPP has become accepted as one the most powerful electroanalytical tools for trace element analysis of food and biological matrices due to its extreme sensitivity, selectivity, and capability of multielemental analysis (*3*, *6*).

The aims of this study were (1) to develop an analytical method for determination of toxic elements Pb and Cd and essential trace element Cu in milk and dairy products by using DPP and (2) to assess the concentration ranges of these elements in a wide variation of milk and dairy products consumed in Turkey.

MATERIALS AND METHODS

Research Material. Milk and dairy samples including different brands of pasteurized milk, whole, low-fat, or light (%0 w/w fat) ultrahigh-temperature (UHT) milks, UHT-treated chocolate, banana or strawberry milks, UHT milks fortified with calcium or vitamins, UHT lactase-treated milk, whole plain and apricot yogurts, pasteurized yogurt drinks (ayran), kasar cheese, cream cheese, pickled white cheese, powdered milk, cream, concentrated cream (kaymak), and plain and chocolate puddings were collected in the local supermarkets in İzmir,

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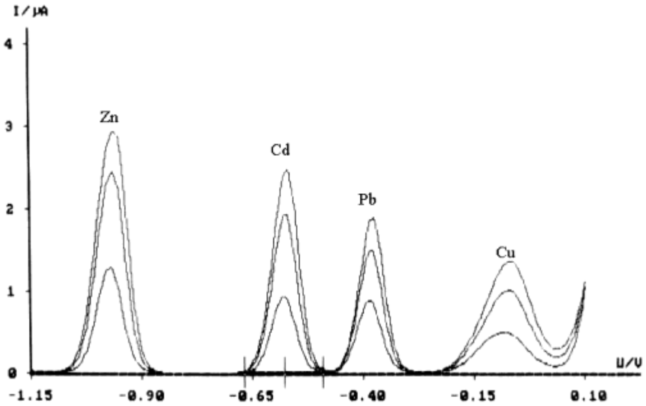

Figure 1. Differential pulse polarogram (DPP) of standard mixture.

Turkey, in October 2002. The production date was the same for all brands, and all samples were prepared for electrochemical analysis at the same time.

Sample Preparation for Electrochemical Analysis. Milk samples were weighed as aliquots of milk and dairy product (5 mL of milk or ayran and 2 g of yogurt, cheese, cream, kaymak, powdered milk, or pudding) into 8 mL Pyrex tubes and centrifuged during 5 min at 4000*g* and then the lipid phases were precipitated. The upper phase including lipids was removed. The lower layer containing the minerals was transferred to another Pyrex tube, 1−2 drops of of concentrated hydrochloric acid (HCl) was added, and then the mixture was again centrifuged at 4000*g* during 5 min. Supernatants including the elements Cd^{2+}, Pb^{2+}, and Cu^{2+} were kept for up to 5 min at 4 °C prior to transfer to Pyrex polarographic cells for electrochemical analysis. All determinations were done in triplicate and represent three samples of each product. All samples were measured three times.

Apparatus. A polarographic analyzer (Metrohm 746 VA trace analyzer) together with a capillary hanging mercury drop electrode (HMDE) and Lenseis LY 1600 model recorder was used for all electroanalytical measurements. A platinum wire was used as the counterelectrode, and Ag/AgCl was used as the reference electrode.

Reagents. All of the reagents used were of analytical reagent grade (Merck, KGaA, Darmstadt, Germany). The mercury used in the HMDE was obtained from Merck. In all research, double-distilled water was used and contaminated mercury was cleaned by passing it successively through a dilute HNO_3 and water column in the form of fine droplets. The collected mercury was dried between filter paper sheets. Prior to use, a differential pulse polarogram of that mercury was recorded to confirm the absence of impurities.

A 0.1 M stock matrix standard solution including cadmium, lead, copper, and zinc was prepared by dissolving $Cd(NO_3)_2$, $Pb(NO_3)_2$, and $Cu(NO_3)_2$ in water, respectively. In these stock solution, concentrations of individual elements were as follows: Cd^{2+}, 112.4 mg L^{-1}; Pb^{2+}, 108.3 mg L^{-1}; and Cu^{2+}, 106.5 mg L^{-1}. Dilute solutions were prepared daily from the above-mentioned standard stock solution.

Electroanalytical Determination. For quantitative determination of cadmium, lead, and copper in milk and dairy products, the polarogram of the individual metal ions in the standard mix solution was obtained using a standard addition procedure as shown in **Figure 1**. For the sample analysis, 1.25 mL of buffer (pH 4.64) as electrolide solution including 0.75 mL of 1.5 M potassium chloride (KCl) and 0.50 mL of 0.5 M sodium acetate ($NaCH_3COO$) in the Pyrex polarographic cell was added to 20 mL of bidistilled water, and then 100 μL of sample was added to this solution. The cell contents were deaerated by passing pure nitrogen (N_2) gas (99.999%) for ∼300 s at a flow rate 100 mL/min to obtain the inert medium. After the first measurement, 0.10 mL of standard mixture was added to the cell. Then the polarographic cell was exposed to N_2 gas for 15 s to perform the second measurement. With the application of the successively standard addition procedure, cadmium(II), lead(II), and copper(II) in milk and dairy products were determined. For each sample, total analysis time was 10 min.

ICP-AES Analysis. For six products, including Nestle Mis UHT milk, Pınar UHT chocolate milk, Pınar UHT milk, Sütaş whole yogurt, Pınar powdered milk, and Eker concentrated cream (kaymak), the DPP method was confirmed by ICP-AES (Varian-El97103728, Palo Alto, CA). The sample preparation and the analytical ICP-AES procedure of milk and dairy samples were performed according to the reported method concerning biomass Cd, Pb, and Cu detection by Tokuşoğlu and Ünal (*7*). A mixture of macro- and microelement standards (1000 mg/L) was prepared, and working standard solutions were obtained by serial dilutions of the standards. Matrix standard solutions were used for calibration plotting for macro- and microelements. Their R^2 values were 1.0. The ICP-AES method for milk and dairy foods was validated by using Nestle Mis UHT milk ($R^2 = 0.9998$). The operating conditions of the ICP-AES were as follows: operating power, 1.2 kW; coolant argon flow rate, 7.5 L/min; plasma argon flow rate, 0.8 L/min; burner type, Minitorch; nebulizer type, Meinhard; sample flow rate, 2.3 mL/min; radio frequency, 27.20 MHz. Data were obtained from three samples of each product, and each was analyzed in triplicate using ICP-AES ($n = 3$).

DPP Method Validation with Standard Reference Food. DPP method validation was performed with infant formula (Milupa, Milumil 2 follow-on formula) as standard reference food. As a certified standard reference food material, Milumil includes 230 μg of Cu per 100 g of powder and contains no Pb or Cd (Milupa, Istanbul, Turkey). We reported that 100 g of Milumil powder contained 230.03 ± 0.02 μg of Cu and included no amount of Pb and Cd ($R^2 = 0.9999$).

Statistical Analysis. Data were analyzed with Statistica for Windows ’98 edition, ver. 6.0, StatSoft Inc., Tulsa, OK (*8*) by one-way analysis of variance (Kruskal−Wallis ANOVA) with element contents of milk and dairy products as the source of variance.

RESULTS AND DISCUSSION

A simple and rapid DPP method was developed for the simultaneous determination of cadmium, lead, and copper in milk and dairy products consumed in Turkey. The standard addition method was used to determine Cd, Pb, and Cu in the samples.

The standard solution was scanned from −700 to 20 V versus a Ag/AgCl electrode. Using the differential pulse mode, peak half-wave potentials as $E_{1/2}$ were −0.58, −0.40, and −0.07 V for cadmium, lead, and copper, respectively. The polarogram of the individual metal ions in the standard mix solution is shown in **Figure 1**. The differential pulse polarograms of pasteurized milk (A), kasar cheese (B), natural yogurt (C), and cream (D) samples are presented in **Figure 2**.

Simultaneous DPP determination of Cd, Pb, and Cu in 34 milk and dairy products was reported. The DPP analysis results of these samples are shown in **Table 1**.

Lead and cadmium are two potentially harmful metals that have aroused considerable concern. From an overall point of view of the data, the general examination reveals that significant differences occurred for Cd and Pb in commercial milk and dairy products consumed in Turkey.

Two different brands of pasteurized milk (Pınar and Nestle Mis) contained similar concentrations of Cd, and significant differences were not found between their Cd contents according to the result of the Kruskal−Wallis test ($p > 0.01$) (**Table 1**). However, Sütaş pasteurized milk had a significantly higher cadmium content (591.92 ± 0.03 μg L^{-1}) than found in other brands of pasteurized milks ($p < 0.01$) (**Table 1**). The level of lead, 0.98 mg L^{-1}, for Pınar pasteurized milk was lower and significantly different from the other two brands of pasteurized milk samples ($p < 0.01$) (**Table 1**). Nestle Mis pasteurized milk had the highest content of lead (2.09 ± 0.04 μg L^{-1}) (**Table 1**). In natural UHT milk samples, the largest variations were obtained for both Cd and Pb concentrations, ranging from 0.03 ± 0.01 to 674.28 ± 0.16 μg L^{-1} and from 0.15 ± 0.07 to 2.94 ± 0.05 mg L^{-1}, respectively. UHT milks also containing lower

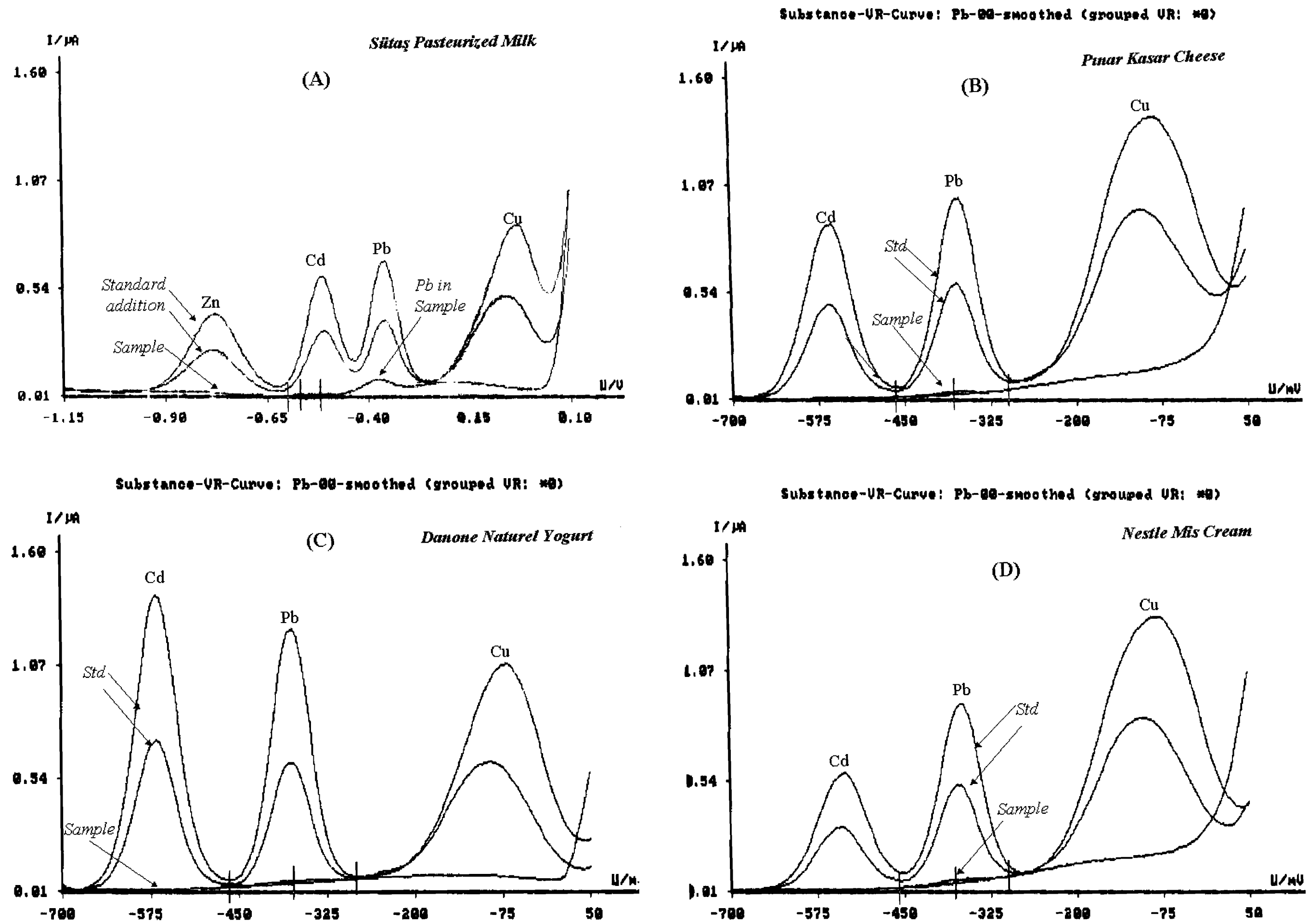

Figure 2. Differential pulse polarograms (DPP) of pasteurized milk (A), kasar cheese (B), natural yogurt (C), and cream (D) samples.

Cd concentrations generally had lower Pb levels. The highest concentrations for both of the two toxic elements were found in Sek UHT milk, probably due to the high content of Pb and Cd in raw milk used in the production and undesired contaminations during manufacturing processes. The levels of Pb and Cd were found to be least in Tansaş UHT milk. Martino et al. (*9*) also reported similar concentrations of Cd (0.50 ± 0.19 μg L^{-1}) in UHT cow's milk, whereas Pb content was significantly lower (2.1 ± 0.3 μg L^{-1}) than obtained for UHT milk in this study. According to the Turkish Food Codex (*10*), the acceptable highest levels of Pb in milk and milk powder were reported as 0.02 and 0.2 mg kg^{-1}, respectively. Hence, the Pb contents of all drinking milks analyzed in this study were found to be extremely high.

The differences in Pb concentration were higher than those found for Cd concentrations between different brands of market milks including pasteurized, natural, or flavored UHT milks (**Table 1**). These differences may be due to raw milk quality or contamination during processing or packaging, although the precise cause is difficult to identify because of the variety of factors involved.

Atmospheric contamination, the excessive use of fertilizers and pesticides, and irrigation with residual waters are among the causes of contamination of raw foodstuffs with Pb and Cd. However, processing, packaging, and other technological processes used to bring dairy products to the consumer can significantly increase the total concentrations of Pb and Cd. In other words, the levels of Pb and Cd in milk and dairy products can rise when dairy animals have consumed contaminated fodders and feeds and when additional contaminants have been added during processing (*11*, *12*). Hence, the variations observed in Pb and Cd concentrations may reflect different environmental situation (i.e., local contamination), which can have a negative impact on toxic element levels in water and feed. Reilly (*13*) showed that the Pb content of milk was 3−4 times normal values when cows consumed grass with 50−100-fold the usual Pb level. Boudene (*14*) also found that Cd concentrations were low except in milk from animals fed with contaminated feed or forage.

On the other hand, processing or packaging at dairy products may raise concentrations of some elements. As a result of contact with the equipment employed (e.g., mechanical milkers, metallic containers, and tanker lorries), elements present in the equipment or container such as Cd and Cu can be released into the milk (*15*). Clay, ceramic, or plastic containers used for dairy products can also release metals (*16*).

Strawberry milk had the highest levels of both Pb (1.61 ± 0.10 mg L^{-1}) and Cd (512.57 ± 0.17 μg L^{-1}) among the flavored UHT milks (**Table 1**). In addition, the concentrations of Cd in UHT-treated strawberry, banana, and chocolate milks were significantly higher than those concentrations found in the same brand of their natural UHT milks, probably due to the possible contamination formed by the flavor addition process. Similarly, Danone apricot yogurt contained significantly higher concentrations of Pb and Cd than found in Danone natural yogurt ($p < 0.01$) (**Table 1**). Significant differences were also found among the yogurt samples dealing with the Pb and Cd contents ($p < 0.01$). Danone natural yogurt contained the least concentrations of Pb and Cd, whereas, Tikveşli yogurt had the highest levels of these toxic elements. There were no significant

Table 1. Cadmium (Cd), Lead (Pb), and Copper (Cu) Contents of Milk and Dairy Products Consumed in Turkey[a]

no.	sample	Cd (μg L^{-1})	Pb (mg L^{-1})	Cu (μg L^{-1})
1	Pınar pasteurized milk	23.40 ± 0.18	0.98 ± 0.15	92.67 ± 0.10
2	Nestle Mis pasteurized milk	25.43 ± 0.09	2.09 ± 0.04	90.06 ± 0.26
3	Sütaş pasteurized milk	591.92 ± 0.03	1.88 ± 0.06	89.66 ± 0.38
4	Pınar UHT milk	43.28 ± 0.03	1.04 ± 0.13	91.72 ± 0.21
5	Nestle Mis UHT milk	35.17 ± 0.02	0.15 ± 0.07	63.56 ± 0.16
6	Ülker İçim UHT milk	405.94 ± 0.21	1.18 ± 0.09	43.56 ± 0.22
7	Sek UHT milk	674.28 ± 0.16	2.94 ± 0.05	67.89 ± 0.15
8	Sütaş UHT milk	10.70 ± 0.05	1.58 ± 0.22	90.34 ± 0.31
9	Tansaş UHT milk	0.03 ± 0.01	0.19 ± 0.08	94.83 ± 0.18
10	Yörsan UHT milk	521.58 ± 0.26	1.89 ± 0.19	88.65 ± 0.56
11	Pınar low-fat UHT milk	31.06 ± 0.07	0.79 ± 0.05	74.46 ± 0.11
12	Pınar skimmed (%0 fat) UHT milk	89.72 ± 0.05	1.01 ± 0.07	50.92 ± 0.18
13	Nestle low-fat UHT milk	499.42 ± 0.15	0.56 ± 0.13	52.56 ± 0.27
14	Pınar UHT chocolate milk	258.62 ± 0.09	0.29 ± 0.03	95.83 ± 0.09
15	Nestle Nesquik UHT banana milk	124.10 ± 0.06	0.13 ± 0.02	59.35 ± 0.28
16	Nestle Nesquik UHT strawberry milk	512.52 ± 0.17	1.61 ± 0.10	60.27 ± 0.52
17	Nestle calcium-fortified UHT milk	0.00 ± 0.00	1.83 ± 0.19	44.33 ± 0.20
18	Nestle vitamin-fortified UHT milk	0.08 ± 0.02	1.22 ± 0.11	65.40 ± 0.42
19	Ülker İçim lactase-treated milk	52.20 ± 0.08	0.79 ± 0.04	56.93 ± 0.27
20	Sütaş pasteurized yogurt drink (ayran)	13.90 ± 0.04	0.24 ± 0.02	22.08 ± 0.06
21	Sakıpağa pasteurized yogurt drink (ayran)	491.87 ± 0.18	0.28 ± 0.07	30.72 ± 0.15
no.	**sample**	**Cd (μg kg^{-1})**	**Pb (mg kg^{-1})**	**Cu (μg kg^{-1})**
22	Danone natural yogurt	30.13 ± 0.08	0.39 ± 0.03	108.84 ± 0.32
23	Danone apricot yogurt	453.92 ± 0.13	1.04 ± 0.09	118.23 ± 0.45
24	Sütaş whole yogurt	389.52 ± 0.19	0.89 ± 0.15	122.13 ± 0.29
25	Sakıpağa whole yogurt	396.46 ± 0.04	1.66 ± 0.11	110.35 ± 0.26
26	Tikveşli whole yogurt	671.97 ± 0.34	1.82 ± 0.10	112.30 ± 0.43
27	Pınar kasar cheese	284.55 ± 0.08	1.05 ± 0.21	111.54 ± 0.20
28	Pınar cream cheese	335.10 ± 0.19	0.69 ± 0.05	105.27 ± 0.25
29	Yörsan pickled white cheese	471.10 ± 0.22	1.78 ± 0.16	100.89 ± 0.27
30	Pınar powdered milk	0.00 ± 0.00	0.19 ± 0.10	0.41 ± 0.08
31	Nestle Mis cream	75.93 ± 0.07	0.83 ± 0.09	78.20 ± 0.14
32	Eker concentrated cream (kaymak)	39.59 ± 0.06	0.31 ± 0.04	98.56 ± 0.38
33	Pınar Kido plain pudding	61.32 ± 0.23	1.26 ± 0.09	78.77 ± 0.23
34	Danone chocolate pudding	478.90 ± 0.06	1.38 ± 0.10	133.46 ± 0.25

[a] Data are expressed as the average of six determinations ± standard deviation corresponding to triplicate extraction each measured duplicate.

differences between the lead contents of Sütaş (0.24 ± 0.02 mg L^{-1}) and Sakıpağa (0.28 ± 0.07 mg L^{-1}) pasteurized yogurt drinks ($p > 0.01$) (**Table 1**), but the high Cd content of Sakıpağa yogurt drink was found to be significantly different from that of the other brand of yogurt drink ($p < 0.01$) (**Table 1**).

From the point of view of the cheese samples, the highest concentrations of Pb and Cd were found in pickled white cheese, probably due to the ripening process in cans and the difference of manufacturing process. The contents of these toxic elements in Quartirolo cheese were lower than values of Pb and Cd obtained for our cheese samples (*17*). It is noteworthy that UHT milks fortified with calcium or vitamins contained no or very little content of Cd. However, the Pb concentrations were found as 1.83 ± 0.19 and 1.22 ± 0.11 mg L^{-1} in UHT milks fortified with calcium and vitamins, respectively. Pınar milk powder had no cadmium and 0.19 ± 0.10 mg kg^{-1} of lead content. Similar findings regarding Pb and Cd contents in milk powder were found in earlier studies (*16*, *18*). The levels of Pb and Cd of Eker concentrated cream (kaymak) were relatively lower compared to many of the other dairy pruducts (**Table 1**), but the levels were higher than those reported by Cabrera et al. (*16*) for whipped cream. The levels of toxic elements observed in this study were generally higher than those concentrations found in a previous study reporting the Cd and Pb contents of Spanish dairy products were between 0 and 28.98 ng g^{-1} and between 0 and 0.211 μg g^{-1}, respectively (*18*).

Copper levels in milk and dairy products were found to be more constant than contents of Cd and Pb (**Table 1**). They ranged from 0.41 ± 0.08 μg L^{-1} for Pınar milk powder to 133.46 ± 0.25 μg kg^{-1} for Danone chocolate pudding. Although cow's milk is a poor source of Cu, there can be some differences among concentrations of this essential element between dairy products depending on the metal release from mechanical milkers and metallic milk containers and/or processing equipment employed (*17*). As shown in **Table 1**, the concentrations of Cu varied for different dairy products, with somewhat higher values for yogurt and cheese samples, possibly because of contamination during the manufacturing process and/or different compositional character of these products. Coni et al. (*17*) reported that the elements such as Cu and Pb show a general trend of increase during cheese manufacture. In their study, the contents of Cu and Pb in raw and cheese samples rose from 0.613−4.50 to 1.51−19 μg g^{-1} and from 0.049−0.075 to 0.076−0.115 μg g^{-1}, respectively, during the manufacture of Quartirolo cheese. The concentrations of Cu in our cheese samples were found to be much lower than findings of Coni et al. (*17*). On the other hand, it has been observed that curdling in cheesemaking increases concentrations of Cu and Cd in the curds compared to raw milk because of their preference for binding to casein and fat (*15*). Our findings also showed high Cu and Cd levels in cheese samples. Furthermore, these levels can be increased by release of metals from containers and tools, with which milk and intermediate products come into contact during the process. In addition, the acidity of cheese and yogurt compared to milk may affect the metal release from containers during storage. Fresno et al. (*19*) also reported similar Cu contents in cow's milk cheeses.

On the basis of the current results, it is possible to state that milk and dairy products analyzed in this study do not contain adequate Cu to match the recommended dietary allowance for

Cu (*20*). However, the Pb and Cd levels found in the samples imply that dairy products contribute a considerable fraction, even the whole amount, of the total dietary intake of these elements.

The mean consumption of dairy products is ~220 g per day in Turkey, but large variations in individual consumption are also described. The major dairy products consumed by individuals are yogurt and yogurt drink (35 kg per year), liquid milk (30 kg per year), and cheese (12.5 kg per year); the mean consumptions per person of butter and powdered milk are 1.3 and 0.5 kg per year, respectively (*21*).

The recommended daily dietary intake of lead had been established for adults at 400−450 μg (*22*). Children are considered a high-risk group, for whom the recommended weekly intake is 25 μg/kg of body weight (*23*). The recommended daily dietary intake of Cd had been established at 57−71 μg (*22*). According to Recommended Dietary Allowances (RDA), the recommended daily intakes of Pb and Cd are $\leq$0.02 mg/L for adult males and females (*24*). No RDA has been established for copper (*24*).

Taking into account these values, it is important to emphasize that the levels of Pb and Cd found in the milk and dairy product samples can represent an imminent toxicological risk. Therefore, precautions should be taken to decrease the values of these toxic elements because of the large amounts of milk and dairy products that are consumed, especially by children.

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