Lead and Cadmium Contamination in Dairy Products and Its Repercussion on Total Dietary Intake

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An accurate and precise method is described for the direct determination of lead and cadmium in dairy products, using slurried samples in combination with electrothermal atomic absorption spectrometry. The samples are suspended in Triton X-100 and shaken with 10.0 g of zirconia spheres until a slurry is formed. The slurry preparation procedure and the graphite furnace conditions were optimized for both elements. The results obtained were validated against microwave acid sample digestion. The detection limit was 0.4 pg for Pb and 0.5 pg for Cd. The analytical application of this method was tested with 60 samples of 10 widely consumed dairy products. The mean values ranged from not detectable to 0.750 µg/g for Pb and from not detectable to 20.0 ng/g for Cd. Increased concentrations of lead and cadmium were detected in products packaged in glazed ceramic containers. Metal migration from the container was also tested. Because milk and dairy products are some of the most widely consumed foods in the human diet, they contribute a large fraction of the intake of trace elements. Strict control of trace element levels in these foods is therefore advisable.

INTRODUCTION

Environmental pollution is the main cause of heavy metal contamination in the food chain. Lead and cadmium are two potentially harmful metals that have aroused considerable concern. Atmospheric contamination, the excessive use of fertilizers and pesticides, and irrigation with residual waters are among the causes of contamination of raw foodstuffs. However, processing, packaging, and other technological processes used to bring foods to the consumer can significantly increase the total concentration of Pb and Cd (Concon, 1988; Creaser and Purchase, 1991). Milk and milk products usually contain very low concentrations of Pb and Cd, except when dairy animals have consumed contaminated fodders and feeds and when additional contaminants have been added during processing (Concon, 1988). Because large amounts of dairy products are consumed (Mahaffey et al., 1975), even small amounts of contaminants can lead to considerable concentrations of trace elements in an organism. The accumulation of heavy metals can have middle-term and long-term health risks, and strict periodic surveillance of these contaminants is therefore advisable. Milk and dairy products, which supply large amounts of protein, calcium, and vitamins, are unquestionably foods of choice for infant feeding, to provide the necessary energy and nutrients in appropriate amounts.

Electrothermal atomization atomic absorption spectrometry (ETA-AAS) has become one of the most popular and widely used techniques for lead and cadmium determination in quality control of foodstuffs (Littlejohn et al., 1985; AOAC, 1990; Cabrera et al., 1992). However, sample preparation, a critical stage in the analysis, is often complicated and can considerably increase the costs of analysis. Moreover, parts of the element under

study may be lost in the process, and other contaminants may be added. The most widely accepted approach currently seems to be the use of microwaves to accelerate acid digestion of the sample, which is placed in a closed recipient and subjected to high temperature and pressure (Kingston and Jassie, 1986; Stripp and Bogen, 1989; Cabrera et al., 1994). In recent years there has been growing interest in the direct analysis of suspensions or slurries of solids by atomic absorption spectrometric methods, which have the advantages of speed, ease of analysis, and reduction of blank levels. ETA-AAS has been used successfully to determine different elements in slurried food samples (Littlejohn et al., 1985; De la Guardia et al., 1986; Ebdon and Evans, 1987; Madrid et al., 1990; Haswell and Barclay, 1992).

This paper describes a simple and rapid method for lead and cadmium determination in dairy products, which combines a slurry procedure to minimize sample preparation with the sensitivity and selectivity of ETA-AAS. The parameters affecting the slurry preparation procedure, and the optimized time–temperature program for each element, are described. The accuracy, precision, and analytical selectivity were evaluated. The results obtained were validated with sample mineralization in a microwave acid digestion bomb. The advantages of our method make it useful for routine food control analyses. In addition, the proposed method was applied to 60 samples of 10 widely consumed dairy products, to determine possible sources of contamination and to evaluate the repercussion of these sources on total lead and cadmium dietary intake.

EXPERIMENTAL PROCEDURES

Apparatus. A Perkin-Elmer 1100B double-beam atomic absorption spectrophotometer equipped with a deuterium background corrector was used at a slit width of 0.7 nm, with a 12-mA hollow cathode lamp for Pb and an 8-mA hollow cathode lamp for Cd. The samples were atomized for Pb determinations at 283.3 nm under the conditions shown in Table 1 and for Cd determinations at 228.8 nm under the conditions shown in Table 2.

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Table 1. Furnace Conditions for Lead Determination in Slurries of Dairy Product Samples by Electrothermal Atomic Absorption Spectrometry

step	temp, °C	ramp time, s	hold time, s	gas flow rate, mL of Ar/min
dry	120	1	40	300
	130	1	20	300
char	800[a]	1	90	300
atomize	1800	1	3	stop
clean	2650	2	2	300
	20	1	1	300

[a] With 200 μg of $NH_4H_2PO_4$ + 10 μg of NH_4NO_3 as a chemical modifier, for a sample aliquot volume of 10 μL.

Table 2. Furnace Conditions for Cadmium Determination in Slurries of Dairy Product Samples by Electrothermal Atomic Absorption Spectrometry

step	temp, °C	ramp time, s	hold time, s	gas flow rate, mL of Ar/min
dry	100	20	10	300
	130	40	10	300
char	700[a]	30	30	300
atomize	2000	1	4	stop
clean	2650	1	2	300
	20	1	2	300

[a] With 200 μg of $NH_4H_2PO_4$ + 10 μg of NH_4NO_3 as a chemical modifier, for a sample aliquot volume of 20 μL

A Perkin-Elmer HGA-700 furnace and a pyrolytic graphite tube with a L'vov platform were used. The samples were injected manually with a Pipetman micropipet. Argon of 99.999% purity at 300 mL/min flow was used as the internal gas.

Slurries of the samples were prepared with a Rotaterm (Selecta) rotary shaker. Samples were digested with a microwave acid digestion bomb (Parr, Model 4782) heated in a Moulinex FM-460 microwave oven at 15–100% full power (600 W) in 25% increments.

Reagents. All solutions were prepared with ultrapure water (specific resistivity of 18 MΩ·cm) obtained by filtering distilled water through a Milli-Q purifier system (Millipore) immediately before use. Lead and cadmium standard solutions (1.00 ± 0.002 g) Titrisol (Merck) were used.

Triton X-100 (Serva, analytical grade), silicone antifoaming emulsion (Fluka), and blown zirconia spheres (Glen Creston, Stanmore, Middlesex, U.K.) were used to prepare sample slurries.

Samples were mineralized with nitric acid (65%) (Merck, Suprapure) and vanadium pentaoxide (Merck, analytical grade). Magnesium nitrate, ammonium dihydrogen phosphate, and ammonium molybdate (Merck, reagent grade) were used as chemical modifiers.

Material. To eliminate absorbance due to detergents and samples, all glassware and polyethylene sample containers were washed with tap water after each use, soaked in a 6 N HNO_3 solution (at least overnight), and rinsed several times in ultrapure water.

Samples. A total of 60 samples of 10 different dairy products were analyzed. Six different brand names of each product, representing the most widely accepted and frequently consumed articles in Spain, were selected for testing. To optimize the amount of material, previous assays were done to ensure that samples were homogeneous and representative of the product investigated.

Procedure. The samples were prepared according to two different procedures as described below.

Slurry Procedure. Portions of 1.0 g of homogenized sample were weighed in small polyethylene bottles, and then 10.0 g of blown zirconia spheres and 10.0 mL of 1.0% (v/v) Triton X-100 solution were added. The bottles were shaken for about 30 min in a flask shaker until a slurry was formed. The slurries were separated from the zirconia spheres using a Büchner funnel and were transferred into a calibrated flask. A few drops of 30% (v/v) silicone antifoaming emulsion in water was added before slurry dilution with deionized Milli-Q water. This grinding procedure ensured that 90% of the particles had
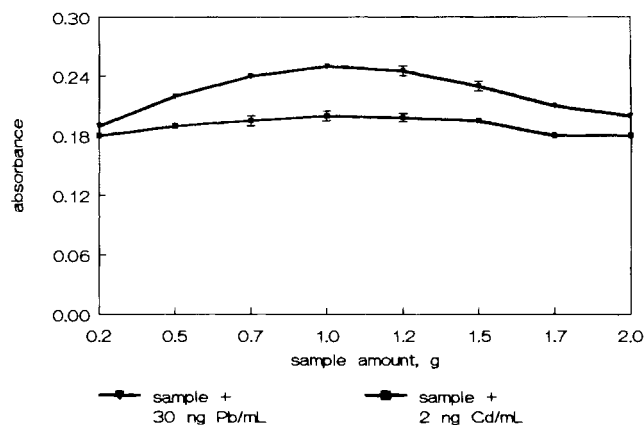

Figure 1. Effect of amount of sample on the slurry preparation procedure for Pb (top curve) and Cd (bottom curve) determination in dairy product samples by electrothermal atomic absorption spectrometry. Volume of 1.0% (v/v) Triton X-100 solution, 10.0 mL; slurry agitation time, 30 min.

a diameter of less than 25 μm (Madrid et al., 1989). This particle size was small enough to determine Pb and Cd by ETA-AAS.

Nitric Acid Mineralization in a Microwave Acid Digestion Bomb. Amounts of 0.200 g of lyophilized and homogenized sample were treated with 2.5 mL of 65% HNO_3 and 35 mg of V_2O_5 as a catalyst and then digested in a microwave acid digestion bomb. Mineralization was complete in 90 s with the oven at its highest setting. The digestion bomb was cooled by freezing at −18 °C for 30–40 min, and then the solution was diluted to a total volume of 25 mL in a glass volumetric flask with ultrapure water.

Sample Analysis. Lead and cadmium were directly determined by ETA-AAS in the same sample slurry and in the same digested sample solution of each dairy product analyzed. To determine Pb, a sample aliquot volume of 10 μL was injected into the tube and run under the optimized conditions shown in Table 1. For Cd quantifications, a sample aliquot volume of 20 μL was injected into the tube and run under the optimized furnace conditions given in Table 2. The graphite tube had been previously treated with saturated ammonium molybdate solution to avoid the formation of refractory carbides. The furnace conditions for each element were optimized on the basis of time–temperature studies using certified standards. To avoid possible errors due to inhomogeneity of the slurries and settling out, shaking of the flask is recommended before each sample is removed. All determinations were done in triplicate.

RESULTS AND DISCUSSION

Optimization of the Variables That Affect Slurry Preparation. The influence of sample amount on the preparation and stability of the slurry for Pb and Cd determination by ETA-AAS was investigated. Amounts ranging from 0.2 to 2.0 g of slurry of six different samples were tested for each element, using the same volume and concentration of Triton X-100 [10.0 mL of 1.0% (v/v) solution] to maintain constant viscosity of the medium. Optimum slurry agitation time was 30 min. To optimize this parameter, portions of samples of all dairy products considered in this study were tested under the conditions proposed above. We found that agitation times of 30–45 min were sufficient to produce a slurry that was homogeneous and stable for up to 48 h. The results, plotted in Figure 1, show that the optimum sample amount for both elements ranged from 0.7 to 1.5 g. Larger amounts of sample decreased slurry stability due to the higher concentration of solids, increased background in Cd determinations, and markedly reduced analytical precision. Moreover, larger samples required the addition to the slurry of higher
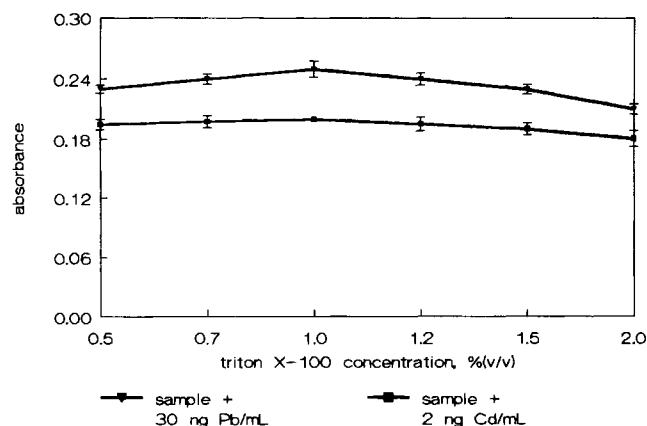
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Figure 2. Effect of concentration of Triton X-100 solution on the slurry preparation procedure for Pb (top curve) and Cd (bottom curve) determination in dairy product samples by electrothermal atomic absorption spectrometry. Volume of Triton X-100 solution, 10.0 mL; sample amount, 1.0 g; slurry agitation time, 30 min.
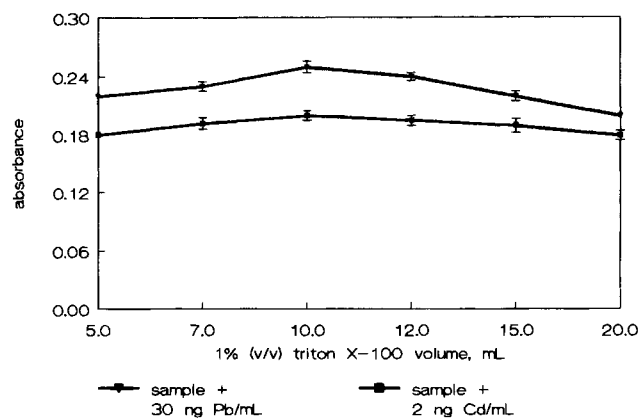

Figure 3. Effect of volume of Triton X-100 solution on the slurry preparation procedure for Pb (top curve) and Cd (bottom curve) determination in dairy product samples by electrothermal atomic absorption spectrometry. Concentration of Triton X-100 solution, 1.0% (v/v); sample amount, 1.0 g; slurry agitation time, 30 min.

volumes of Triton X-100, and sample sizes below 0.7 g were not representative of the real sample and yielded inaccurate values in Pb and Cd determinations.

The addition of a thixotropic thickening agent is necessary to produce a suspension that remains stable for several days and that can be sampled reproducibly (Madrid et al., 1989). In our experiments, Triton X-100 was used as a dispersing agent. To ascertain the influence of the Triton X-100 solution on slurry preparation for Pb and Cd determinations, we tested suspensions containing different concentrations and volumes of this solution. To determine the optimum concentration, 1.0 g of six portions of two different dairy products was treated with 10.0 mL of different aqueous solutions of Triton X-100 that ranged from 0.5 to 2.0% (v/v); the mixture was shaken for 30 min until the slurry had formed. The results (Figure 2) show an optimum concentration of 1% (v/v) for the determination of both elements. With concentrations lower than 0.5% (v/v) it was not possible to obtain a particle size suitable for the determination of these elements with ETA-AAS; moreover, precision declined notably, and stability of the slurry was approximately 5% shorter than the established duration. We found that concentrations of Triton X-100 of 2.0% or higher led to significant matrix interferences in the determinations, due to increased slurry viscosity.

To optimize the volume of 1% (v/v) Triton X-100 necessary for 1.0 g of sample, different volumes (range 5.0–20.0 mL) were tested. The results (Figure 3)show that a volume of 10.0 mL is sufficient to obtain a homogeneous, stable slurry with all dairy products sampled. Larger volumes caused chemical interferences and high background signals.

Optimization of Graphite Furnace Program. For lead determinations, mineralization of the matrix was completed after heating at 800 °C for 90 s. The atomization temperature that yielded maximum signals was 1800 °C for 3 s, with an integration time of 4 s. Stopping the flow of the argon purge gas at this point increased sensitivity without altering the lifetime of the tube. The furnace was cleaned by raising the temperature to 2650 °C, and the graphite tube was allowed to cool to 20 °C between each analysis (Table 1). The use of a L'vov platform gave more reproducible results. Different amounts of $NH_4H_2PO_4$–NH_4NO_3 were tested; the best results were obtained with 200 μg of NH_4H_2-PO_4 and 10 μg of NH_4NO_3 for a sample aliquot volume

of 10 μL. All analyses were done in peak area mode (integrated absorbance). The same procedure was followed for the blanks. Appropriate aqueous calibration graphs were prepared. Standard addition graphs were prepared for blanks and for samples, and from these the blank-to-sample slope ratios were calculated to compare the selectivity of the method. For the samples considered in this study, the slope ratio values obtained ranged from 1.00 to 1.10 and from 0.99 to 1.20 for Pb and Cd determinations, respectively. The standard addition method was therefore unnecessary, and consequently the analysis was much simplified.

For cadmium determinations by ETA-AAS, mineralization of the matrix was complete after heating at 700 °C for 30 s. The atomization temperature that yielded maximum signals was 2000 °C for 4 s, with an integration time of 5 s. Argon flow was stopped during the atomization stage to increase sensitivity, and this did not alter the usable life of the tube. The furnace was cleaned by raising the temperature to 2650 °C, and the graphite tube was cooled to 20 °C between analyses (Table 2). Different amounts of $NH_4H_2PO_4$ and NH_4H_2-PO_4–NH_4NO_3 were tested as chemical modifiers; the best results were obtained with 200 μg of $NH_4H_2PO_4$ and 10 μg of NH_4NO_3 for a sample aliquot volume of 20 μL. The samples were analyzed by the standard addition method: 0.0–1.0 ng Cd/mL was added to four aliquots of the slurried sample after analyte determination. Absorption was measured as the area under the absorbance peak. The same procedure was used for the blanks.

The proposed method was validated by comparison with Pb and Cd determinations in acid digested samples by ETA-AAS, using the same furnace conditions (Tables 1 and 2). The results obtained showed good agreement between slurry and acid digestion procedures for all samples (Table 4). Comparison of the results with the F-test revealed no significant differences at the 95% confidence level, so it was concluded that the slurry procedure can be applied as an alternative to sample acid digestion for analysis of Pb and Cd by ETA-AAS in dairy products and similar foodstuffs. In addition, the slurry procedure described above made multielemental determinations possible in the same suspension of the sample.

Analytical Characteristics. The detection limits were calculated according to IUPAC rules (Long and Winefordner, 1983). The sensitivity and precision of the analytical conditions were evaluated for each element.

Table 3. Analytical Characteristics for Lead and Cadmium Determination in Slurries of Dairy Product Samples by Electrothermal Atomic Absorption Spectrometry

| element	detection limit,[a] pg	characteristic mass[b]	accuracy,[c] %	precision (RSD),[d] %
lead	0.4	10.0	98.7 ± 0.5	4.5
cadmium	0.5	0.4	97.5 ± 0.7	6.0

[a] Detection limit calculated according to IUPAC rules. [b] Characteristic mass in pg/0.0044 A·s. [c] Results obtained by analyte recovery assays in five different samples. [d] Relative standard deviation for 10 replicate determinations on each of five different samples.

Accuracy was checked with recovery assays, by adding known amounts of analyte to five different randomly chosen samples and processing the mixtures as described above for experimental samples. Precision was also checked in 10 determinations of 5 different samples. The results are summarized in Table 3. The detection limits and sensitivity were suitable for the range of Pb and Cd concentrations encountered and are compatible with estimates given by other authors. Moreover, the analytical precision and accuracy were acceptable (Horwitz, 1982).

Lead and Cadmium Determination in Dairy Products. The proposed method was used to determine Pb and Cd contamination levels in 60 samples of 10 different widely consumed dairy products; the values obtained (mean ± standard deviation) for each type of sample are given in Table 4.

Although the Pb and Cd values found in the dairy product samples we analyzed do not represent an inminent toxicological risk, periodic determinations are advisable because of the large amounts of these products that are consumed (especially by children), in view of the potential medium- and long-term risks. For example, the annual consumption per person of dairy products in several European countries, according to data compiled by the European Economic Communities, was 201 kg in Ireland, 178 kg in Denmark, 162 kg in The Netherlands, 147 kg in the United Kingdom, and 113 kg in Spain (CEE, 1991). Pelus et al. (1994) reported that in France the mean consumption of dairy products is approximately 300 g/day, but large ranges in individual consumption are also described. In Spain, the major dairy products consumed by individuals are yogurt (7.6 kg/year) and cheese (5.8 kg/year); the mean consumption per person of powdered and condensed milk are 0.54 and 0.87 kg/year, respectively, and of the rest (curd, custard, creme caramel, whipped cream, ice cream) the mean consumption per person is 3 kg/year (Ministerio de Agricultura, Pesca y Alimentación, 1991).

On the basis of these data, the Pb and Cd levels found in the samples we analyzed imply that dairy products
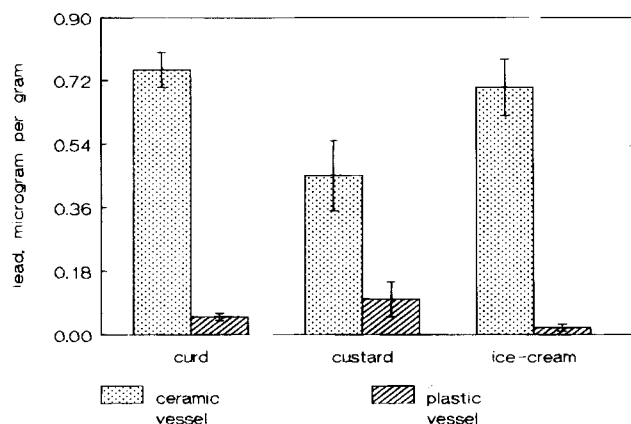
Figure 4. Comparison of lead contamination levels in dairy products packaged in plastic and glazed ceramic containers.

contribute a considerable fraction of the total dietary intake of these elements. The recommended daily dietary intake of lead had been established for adults at 400–450 μg (FAO–WHO, 1972). Children are considered a high risk group, for whom the recommended weekly intake is 25 μg/kg of body weight (FAO–WHO, 1989). The recommended daily dietary intake of cadmium had been established at 57–71 μg (FAO–WHO, 1972); approximately 5% of the Cd in the diet can be absorbed, although this percentage depends on the relative concentrations of Fe, Zn, and Cu (Concon, 1988; Fennema, 1993), among other factors.

It is important to emphasize that the highest levels of Pb and Cd found in the dairy products we analyzed were detected in foods packaged and sold in glazed ceramic containers. This was particularly evident in the results for Pb in curd, an acidic dairy product. Figure 4 shows the Pb contamination values in dairy products packaged in plastic and glazed ceramic containers. The ceramic containers, which are in contact with the food product during manufacture and throughout its shelf life until the moment of consumption, are made of a mixture of inorganic materials with a generally large proportion of clay and silicates, to which small amounts of organic materials are added before firing. These vessels are often vitrified and glazed and/or decorated with Pb- or Cd-containing pigments.

The influence of packaging processes and materials on heavy metal contamination of foods, particulary of dairy products, has been widely reported (Bruhn and Franke, 1976; Linder, 1985; Concon, 1988; McLaren and Meguid, 1988; Creaser and Purchase, 1991). A report from New Zealand compared the amount of lead contamination in several types of canned food, including baby food, available in retail shops. Differences were

Table 4. Lead and Cadmium Contamination Levels in Dairy Product, Determined in Slurried and Mineralized Samples by Electrothermal Atomic Absorption Spectrometry

	lead,[a] μg/g (mean ± SD)		cadmium,[a] ng/g (mean ± SD)	
sample	slurried	mineralized[b]	slurried	mineralized[b]
powdered milk	0.025 ± 0.025	0.026 ± 0.020	nd[c]	nd
condensed milk	0.075 ± 0.025	0.075 ± 0.030	nd	nd
children's milk	nd	nd	nd	nd
yogurt	nd	nd	nd	nd
curd	0.500 ± 0.250	0.475 ± 0.300	17.5 ± 2.5	18.0 ± 2.0
cream cheese	0.080 ± 0.020	0.080 ± 0.025	5.0 ± 5.0	5.5 ± 4.0
whipped cream	0.075 ± 0.050	0.078 ± 0.045	8.0 ± 4.0	8.0 ± 4.5
ice cream	0.250 ± 0.425	0.250 ± 0.450	nd	nd
custard	0.400 ± 0.300	0.450 ± 0.250	2.0 ± 1.0	2.0 ± 0.9
creme caramel	0.050 ± 0.030	0.050 ± 0.035	5.0 ± 2.5	4.8 ± 2.5

[a] Mean value of three replicate determinations on each of six different samples. [b] Nitric acid mineralization in a microwave acid digestion bomb. [c] Not detectable.

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found between lacquered and unlacquered cans. The authors concluded that in general the incidence of Pb contamination is significant and is mainly associated with lacquered cans. Metal contamination of foods has also been traced to the use of poor-quality glazed ceramic containers and enamel utensils. The glasslike surface on well-made plates and dishes should normally be unaffected by food and should not release its lead and other metals in contact with food, but coloring agents often contain Cd and Pb salts and may contribute to food contamination. Various factors such as the amount and type of organic acids present, the levels of nitrate, the amount of oxidizing or reducing agents, and the storage temperature determine the amount of corrosion (Reilly, 1980).

To determine Pb and Cd specific migration from these containers, we had used the technique proposed by the European Economic Community's technical health regulation (CEE, 1984). Containers used to package dairy products for retail sale were immersed in an aqueous solution of 4% acetic acid at a temperature of 22 ± 2 °C for 24 ± 0.5 h, and Pb and Cd were determined by AAS. Lead concentrations ranged from 0.2 to 1.0 μg/mL, and cadmium concentrations ranged from 0.25 to 0.50 ng/mL. The European Economic Community establishes a lead concentration limit of 0.400 μg/mL and a cadmium concentration limit of 0.03 μg/mL, referred to the extraction solution.

The results of our analyses show that many glazed ceramic containers are unsuitable for use in the packaging and sale of dairy products. The acidic nature of most such products, together with other factors such as light and storage temperature, favors the degradation of pigments used in ceramic vessels and increases the chances that Pb and Cd will be released into the food product. The future regulatory control of food contact materials will depend to an increasing degree on the use of positive lists of permitted substances supported by migration testing (Creaser and Purchase, 1991). Continuous surveillance of all processes during which contaminants can appear is the most effective approach to the prevention of contamination in foods and foodstuffs.

ACKNOWLEDGMENT

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