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Evaluation of Lead and Cadmium Heavy Metal Residues in Milk and Milk Products Sold in Ogbomoso, Southwestern Nigeria

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Abstract: The concentrations of lead (Pb) and cadmium (Cd) heavy metals in cow milk, goat milk butterfat, soft cheese and yoghurt samples were evaluated. Forty samples (eight of each sample) were analyzed using the atomic absorption spectrophotometer. There were detectable residual concentrations of Pb and Cd in all the 40 samples. The range of 0.0025 to 0.0061 ppm of Pb concentrations in the samples was within the Maximum Residue Limit (MRL) of 0.0125 to 0.0175 ppm of Pb in milk and dairy products. There was a significantly ($P < 0.05$) higher mean Pb concentration of 0.0061 ± 0.0025 ppm in the soft cheese samples. The residual concentrations of Pb in the cow milk samples were not significantly ($P > 0.05$) different from that of the goat milk samples. Residual concentrations of Cd were higher in soft cheese samples (0.0048 ± 0.0007 ppm) and in goat milk samples (0.0045 ± 0.0005 ppm Cd) and these two concentrations exceeded the MRL of 0.0035 ppm of Cd recommended by India regulations. The residual concentration of Cd in goat milk samples (0.0045 ± 0.0005 ppm) was significantly ($P < 0.05$) higher than in the cow milk samples (0.0021 ± 0.0007 ppm). The results of this study showed that all the milk and milk products samples analyzed contained residues of Pb and Cd heavy metals. The observation of residual Cd concentrations above the MRLs in the soft cheese samples and in the goat milk samples is of public health concern and could cause health hazards to consumers.

Key words: Lead, cadmium, heavy metals, milk, milk products

INTRODUCTION

Milk and its products are considered as a nearly complete food, since they are good sources of protein, fat and major minerals (Belewu, 2006). Tona *et al.* (2000) reported Ca, P, Mg, K and Na major minerals in the milk of Bunaji cows. Church *et al.* (1974) explained that major or macro minerals are those which occur in larger amounts such as Ca, 1.33%; P, 0.74%; Na, 0.16% and S, 0.15% in the animal body tissues. They further stated that in contrast, the minor or trace elements may be present in the animal and plant tissues only at concentrations of a few ppm, examples are Co, Se and Mo. That other minerals like Pb, Cd and F may be very toxic and result from contamination of the environment by man and may cause reduced productivity or death. Somer (1974) stated that it has been noticed that most of the heavy metals are members of the micro elements which are required in small doses for proper functioning of animals. Aslam *et al.* (2011) defined heavy metals as the elements with density more than 5gcm^{-3} , atomic weight between 63.55 and 200.59 and specific gravity greater than 4. The authors then mentioned that although heavy metals normally remain in ground water and soil, however in certain areas their levels might

increase and they could tend to accumulate to toxic levels in human and animal tissues, deriving food from water and soils. Chitmanat and Traichaiyaporn (2010) explained that living organisms normally require some of these heavy metals up to certain limits but if excess accumulation occurs it could lead to severe detrimental effects. The mineral content of milk depends on numerous factors such as genetic characteristics, the stage of lactation, environmental conditions, the type of pasture and soil contamination (Gambelli *et al.*, 1999). Ward and Savage (1994) also mentioned that foodstuffs grown on contaminated soil or irrigated with impure water accumulate metal contents and could be sources of heavy metals exposure to animals and humans. Again, livestock reared on contaminated fodder become continuous sources of heavy metal residues in edible tissues and milk (Aslam *et al.*, 2011). Lead is toxic to the blood, the nervous, gastric and genital systems and the accumulation of lead produces damaging effects in the hematopoetical, hematic, renal and gastro-intestinal systems (Correia *et al.*, 2000). Cadmium is also a toxic metal when released into the environment at a high level. Cadmium toxicity may originate through the dumping of industrial wastes in the rivers as well as the

application of phosphate fertilizers in plants (Venugopal and Luckey, 1978). Santhi *et al.* (2008) reported that at relatively low concentrations, heavy metals could cause adverse effects and this calls for the immediate attention of health regulatory authorities and researchers. Tripathi *et al.* (1999) stated the recommended safe value of Pb concentration as 0.0125 to 0.0175 ppm and that for Cd as 0.0028 to 0.0035 ppm in milk and milk products. Anastasio *et al.* (2006) reported that the Maximum Residue Limit (MRL) of Pb concentrations for bovine milk was 0.0200 ppm but that there was no specific MRL for Cd in milk and dairy products. Aslam *et al.* (2011) stated that high levels of heavy metal residues in milk of cattle and goat above the Maximum Residue Limits (MRLs) must be regarded as a potential health hazard in animals and human beings. That this must draw the attention of food regulatory bodies to adopt measures that could bring the heavy metal residues in food chain up to permissible values not injurious to human health. This study was undertaken to evaluate to what extent lead and cadmium heavy metals may be present in raw cow and goat milk and the processed butterfat, soft cheese and yoghurt sold for human consumption in Ogbomoso, Nigeria.

MATERIALS AND METHODS

Sampling procedures, processing and analysis: A total of forty samples (eight samples each) of cow milk, goat milk butterfat, soft cheese and yoghurt were obtained from a Fulani nomadic farm in Abogunde village, Ogbomoso, Nigeria. The butterfat, soft cheese and yoghurt used were processed from the White Fulani cows' milk. Twenty mls each of cow milk, goat milk and yoghurt and 20g each of butterfat and soft cheese samples were put in well labeled, sterile, plastic bottles and kept in a cooler with ice pack and then carried immediately to the laboratory for analysis.

Analysis of Pb and Cd in milk and milk product samples using the Atomic Absorption Spectrophotometer (AAS): The milk and milk products samples first needed to be brought into clear solution for analysis by the AAS. Thus the samples were first digested with strong acids where the organic portions of the samples were destroyed and the minerals left in a clear solution.

Digestion: Amounts of 0.5g of each sample were weighed into a set of digestion tubes and 10mls each of perchloric and nitric concentrated inorganic acids were dispensed into the sample tubes. The samples were then digested on the digestion block at 120°C for 2 hours, until the organic substances were completely decomposed. At the end of the digestion, the samples were allowed to cool to room temperature.

Analysis using the AAS: Digested samples were made up to the 50mls volume with deionized water and then transferred into centrifuge tubes and shaken for 10 minutes. The shaken solutions were transferred to the centrifuge machine and centrifuged at the rate of 4500 rpm for 5 minutes. Finally, the supernatants were placed in duplicates in a set of pyrex glass vials to be analyzed in the AAS, for the determination of Pb and Cd levels through comparisons with known standard concentrations (AOAC, 2005).

Calculation of results by the software: The software generates the standard curve or equations from the signals of the working standard versus the standard concentrations of the unknown analytes. The results were then expressed in ppm.

Statistical analysis: Samples were analyzed in duplicates. The standard deviations of treatment means were calculated. All statements of differences were based on significance at $P < 0.05$.

RESULTS

Concentrations of Pb in milk and milk products: The residual concentrations of Pb in cow milk, goat milk butterfat, soft cheese and yoghurt samples are presented in Table 1. Detectable residual concentrations of Pb were observed in all the 40 milk and milk product samples and the ranges of values were 0.0025 to 0.0061 ppm of Pb. The residual concentration of Pb was significantly ($P < 0.05$) higher in the soft cheese samples (0.0061 ± 0.0025 ppm of Pb) than in the cow milk, goat milk butterfat and yoghurt which were not significantly ($P > 0.05$) different one from the other. The butterfat, soft cheese and yoghurt samples were processed from the milk of White Fulani cows. The residual concentrations of Pb in the cow milk

Table 1: Concentrations of Pb in milk and milk product samples

Samples	Mean concentration of Pb (ppm)
Cow milk	0.0030±0.0016 ^a
Goat milk	0.0035±0.0005 ^a
Butterfat	0.0026±0.0012 ^a
Soft cheese	0.0061±0.0025 ^a
Yoghurt	0.0025±0.0009 ^a

Values are means±standard deviation;

^{a,b}values with different superscripts along the column are significantly ($P < 0.05$) different

Table 2: Concentrations of Cd in milk and milk products samples

Samples	Mean concentration of Cd (ppm)
Cow milk	0.0021±0.0007 ^a
Goat milk	0.0045±0.0005 ^a
Butterfat	0.0016±0.0003 ^a
Soft cheese	0.0048±0.0007 ^a
Yoghurt	0.0019±0.0008 ^a

Values are means±standard deviation;

^{a,b}values with different superscripts along the column are significantly ($P < 0.05$) different

Table 3: Regulations for MRLs for Pb and Cd in milk and dairy products

Lead (Pb)	0.0125-0.0175 ppm	India regulations	Tripathi <i>et al.</i> (1999)
Cadmium (Cd)	0.0028-0.0035 ppm	India regulations	Tripathi <i>et al.</i> (1999)
Lead (Pb)	0.0200 ppm	EU regulations	Anastasio <i>et al.</i> (2006)

MRLs: Maximum residue limits

samples (0.0030 ± 0.0016 ppm) was not significantly different ($P > 0.05$) from that of goat milk samples (0.0035 ± 0.0005 ppm).

Concentrations of Cd in milk and milk products: The residual concentrations of Cd in cow milk, goat milk butterfat, soft cheese and yoghurt samples are shown in Table 2. Detectable residual concentrations of Cd were observed in all the 40 milk and milk product samples and the ranges of values were 0.0019 to 0.0048 ppm of Cd. The residual concentrations of Cd in soft cheese samples (0.0048 ± 0.0007 ppm) and goat milk samples (0.0045 ± 0.0005 ppm) were similar ($P > 0.05$), both of which were also significantly ($P < 0.05$) higher than Cd concentrations in cow milk (0.0021 ± 0.0007 ppm) butterfat (0.0016 ± 0.0003 ppm) and yoghurt samples (0.0019 ± 0.0008 ppm) which were similar ($P > 0.05$) in residual concentrations (Table 2). The residual concentration of Cd in goat milk samples (0.0045 ± 0.0005 ppm) was significantly ($P < 0.05$) higher than in the cow milk samples (0.0021 ± 0.0007 ppm). Regulations for the Maximum Residue Limits (MRLs) for Pb and Cd in milk and dairy products are as presented in Table 3.

DISCUSSION

Concentrations of Pb in milk and milk products:

The detection of residual concentrations of Pb in all the 40 milk and milk products samples might be due to the contamination of the soil, fodder and water on which the lactating ruminants were nourished (Ward and Savage, 1994; Gambelli *et al.*, 1999; Aslam *et al.*, 2011). The range 0.0025 to 0.0061 ppm of Pb concentrations in the milk and milk products samples obtained in this study were within the MRL of 0.0125 to 0.0175 ppm of Pb. Also, these values were lower than the values of 0.0167-0.0232 ppm Pb concentrations in cow milk reported by Aslam *et al.* (2011). The higher mean concentration of Pb in the soft cheese samples than the others might imply that the heating and addition of coagulant during cheese processing, did not eliminate the Pb residue but caused increased residual Pb concentration of 0.0061 ppm in the soft cheese samples. Similarly, Anastasio *et al.* (2006) observed highest residual Pb concentration in cheese (0.390 ppm) processed from the raw milk of sheep (0.180 ppm of Pb). Anastasio *et al.* (2006) attributed higher concentrations of Pb in cheese than in the raw milk and other milk products

to contamination during the cheese making process. Processing of the cow milk into butterfat and yoghurt also did not eliminate

Pb residues. In this study, mean concentration of Pb in cattle milk was similar to the concentration in goat milk. This result was however contrary to the observation of higher levels of Pb in goat milk (0.0434 ppm) than that in cattle milk (0.0199 ppm of Pb) reported by Aslam *et al.* (2011).

Concentrations of Cd in milk and milk products: The detection of residual concentrations of Cd in all the 40 milk and milk products samples might be due to the contamination of the soil, fodder and water on which the lactating ruminants were nourished. Similarly, Aslam *et al.* (2011) reported that livestock reared on contaminated fodder become continuous sources of heavy metal residues in edible tissues and milk. The residual concentration of Cd of between 0.0019 and 0.0048 ppm in milk and milk products were higher than the range of 0.00012 to 0.00017 ppm of Cd found in the milk of cattle by Aslam *et al.* (2011). The significantly ($P < 0.05$) higher residual concentration of 0.0048 ppm Cd in soft cheese samples exceeded the MRL of 0.0035 ppm recommended by Tripathi *et al.* (1999). This might be due to the heat treatment and addition of coagulant during cheese processing. The significantly ($P < 0.05$) higher mean concentration of 0.0045 ppm of Cd in goat milk samples was also above the MRL and was higher than in the cow milk butterfat and yoghurt samples. This might be because goats are known to feed on wider varieties of forage trees, grasses, herbs and other non conventional feedstuffs than cattle (Ogungbesan *et al.*, 2010). The mean concentration of Cd observed in the current study was not in agreement with the results of Aslam *et al.* (2011) where the Cd residue concentration in goat milk (0.000145 ppm) was similar to that in cattle milk (0.000147 ppm). The Cd concentrations in the soft cheese and goat milk samples should be regarded as potential health hazard for consumers.

Conclusion: The results of this study showed that all the milk and milk products samples analyzed contained residues of Pb and Cd heavy metals. The observation of residual Cd concentrations above the MRLs in the soft cheese samples and in the goat milk samples is of public health concern and could cause health hazards to consumers.

Care needs to be taken in the choice of coagulants to be used for cheese processing. Sources of heavy metal

contamination such as, environmental contamination of soil, water and fodder should be checked by livestock farmers and government environmental control agencies. There is the need for continuous monitoring by health regulatory authorities in Nigeria in order to ensure that the milk and milk products sold do not cause any health hazard.

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