Short communication

Assessment of heavy metal contamination in raw milk for human consumption

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Abstract

The presence of heavy metals in various farm inputs, including feed, fertilizer, water and environment leads to excretion of the residues in animals' milk. Because consumption of milk contaminated with heavy metals poses serious threats to consumers' health, a study was conducted in 2012 – 2013 in Pakistan to evaluate the concentration of heavy metals in unprocessed raw milk procured in different seasons from Jhang city, Punjab, Pakistan. A total of 400 milk samples was collected from milk shops and dairy farms, and the contents of heavy metals, i.e. copper (Cu), lead (Pb), cadmium (Cd) and chromium (Cr) were analysed via atomic absorption spectrometry. Results indicated that the average concentrations of Cu, Pb and Cd were significantly higher than International Dairy Federation Standards. Within a season, heavy metal levels differed significantly among samples. For each type of milk source, there was a significant difference in mean concentration of heavy metals between summer and winter. Based on these findings, there is a critical need to set legal limits and surveillance for heavy metals in an animal's milk.

Keywords: Consumer, health metals, milk, season

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In developing countries such as Pakistan, the rapid increase in industrial and agricultural activities has contributed to increased levels of heavy metals in the environment, for example in the air, soil and water (Pack *et al*., 2014). Heavy metals accumulate in tissues of dairy animals and ultimately excrete in milk because of their non-biodegradable and persistent nature (Burger & Elbin 2015; Meli *et al*., 2015). Consumption of adulterated milk by consumers results in various health problems, and is a most important concern in the food industry (Singh *et al*., 2010). Globally, there has been many reports on heavy metal intake by humans through food contamination (Muchuweti *et al*., 2006). Heavy metals are compounds that can affect human health if taken in large quantities. These are all naturally present at low levels in the environment. Ingestion (through drinking or eating) and inhalation (through breathing) are the main routes of human exposure. Long-term exposure to lower levels of Cd and Cr leads to stomach irritation, kidney disease, lung damage and nervous manifestations. Additionally, they are known to be human carcinogens. Toxicity by Pb can result in decreased performance, and damage to the brain, kidneys and sperm-producing organs (Martin & Griswold, 2009). Copper is essential for optimal innate immune function. However, long-term exposure to copper can damage the liver and kidneys (Copper, 2005). Raw milk and dairy products provide a quick and easy way to supply many nutrients in daily diets. Accordingly, in many rural areas of the world, milk is considered as the major and perhaps only source of nutrition, especially for children. In Pakistan, cattle and buffalo are the main milk-producing animals, with approximately 18 706 and 32 180 thousand tons of milk produced in fiscal year 2014-15, respectively (Anonymous, 2015). There are a few studies on the amount of chemical contaminants in unprocessed and raw milk that is available at conventional milk shops in various cities in Pakistan. Thus, the aim of this study was to assess and quantify residues of selected toxic heavy metals, including Cu, Pb, Cd and Cr, in raw milk sold in street shops and dairy farms of Jhang, the milk production belt of Pakistan.

A total of 400 milk samples was collected from milk shops and dairy farms in Jhang, Punjab, Pakistan. The city was purposively selected because its dairy production and marketing are representative of the whole country as it is considered the milk belt in Pakistan. From Jhang city map, 100 streets were randomly selected and geo-referenced in Quantum GIS Lisboa, version 1.8.0. In each street, one shop was selected. Systematic sampling was performed to sample bulk milk from non-commercial dairy farms located within a 3-km buffer around wastewater drains in peri-urban areas of the city. The sampling was done in winter (n = 200) and summer (n = 200) from November 2012 to July 2013. From each selected shop, 250 mL unprocessed liquid milk was purchased, of which 25 mL was sampled in clean sterilized screw-capped glass tubes. Immediately after collection, milk samples were transported to the laboratory by placing the tubes in ice packs, and were stored at −20 ºC until further analysis. Wet digestion was performed for sample extraction. Briefly, 1 mL milk sample was taken into a 50 mL digestion flask containing 10 mL nitric acid (HNO_3). Later the flask was heated on a hot plate for 5 minutes and the fumes were allowed to evaporate until 1 mL solution was left. Then the contents of the flasks were cooled down and 5 mL per-chloric acid ($HClO_4$) was added. The contents were heated again and allowed to evaporate until 1 mL solution was left. Finally, the left-over solution was diluted with distilled water to 25 mL (Richards, 1968). The concentration of metals in samples was determined by an atomic absorption spectrometer with Zeeman-effect background correction (AAS-Perkin Model: AA analyst 300). The data were analysed in SPSS version 21. Wilcoxon signed rank and chi-square tests were performed to evaluate and compare the concentrations of heavy metals in raw milk samples.

It is evident from the findings (Table 1) that median concentrations of Cu, Pb and Cd were higher (P <0.05) than standards set by the International Dairy Federation (IDF 1979). However, a permissible level for Cr was not available to enable such comparison.

Table 1 Median and interquartile range of concentration of measured metals in milk samples stratified by season and source

Source of milk	Metal	Season	Median (mg/kg)	Interquartile range (mg/kg)	IDF ML (mg/kg)
HH & dairy farms near drain	Cu	Summer	0.938	0.729	0.01
		Winter	0.0	0.25	
	Cd	Summer	0.092	0.092	0.0026
		Winter	0.0	0.0	
	Pb	Summer	0.227	0.909	0.02
		Winter	1.25	1.5	
	Cr	Summer	12.4	5.6	------
		Winter	4.25	1.0	
Milk shops	Cu	Summer	1.354	3.229	0.01
		Winter	0.5	0.5	
	Cd	Summer	0.092	0.079	0.0026
		Winter	0.25	0.25	
	Pb	Summer	0.455	1.136	0.02
		Winter	2.0	1.75	
	Cr	Summer	8.4	10.0	------
		Winter	3.75	3.0	

HH: Households.
IDF ML: Maximum level according to IDF standard (1979).

In each season, Cu, Pb and Cr levels differed (P <0.05) in milk samples collected from shops and dairy farms near wastewater drain canals. For each type of milk source, there were differences (P <0.05) in mean concentrations of Cu, Pb and Cr between samples collected in summer and winter. The concentration of Cu was greater (P <0.05) in milk samples from urban shops and in summer. In winter, the proportion of abnormal samples was higher in samples collected from milk shops. All samples collected near wastewater drains in summer contained Cu above the permissible level. Possible sources of Cu in milk samples could be feed contamination, water contamination and copper alloys used in milk collection equipment. Concentration of Cr was higher in summer, especially in milk from dairy farms located near wastewater drainage canals as the animals were being reared in closed barns and fed on sewage-grown fodders. There is no legal standard for the proportion of samples containing ‘unsafe’ levels of Cr. Relatively high levels of Cd were found in milk samples from shops in both seasons. Interestingly, samples from farms did not contain detectable limits of Cd in winter, despite a high level measured during summer. Similarly, the proportion of samples containing an abnormal level of Cd was greater in summer in milk from both shops and farms. Concentration of Pb was higher in winter from urban shops. The proportion of samples with Pb above safe limits was higher in winter, irrespective of the source (farm/shop). However, all the milk samples collected from urban shops in winter had a Pb level above the safe limit. The difference in proportion of abnormal samples in summer and winter may be attributed to various factors, such as seasonal variations in heavy metal contents of soil, wastewater, fodder and particulate matter. Moreover, the uptake of these metals by plants is influenced by soil pH and agriculture practices such as the use of fertilizers. The amount of soil ingestion by animals and vegetation types in different seasons also contributes to variable degrees of heavy metal exposure (Singh *et al*., 2010). The present results are in accordance with a previous study from Faisalabad (Pakistan), which showed high levels of Cd, Cr, Ni, Pb, and Hg in milk of goats and cattle (Aslam *et al*., 2011). Heavy metals are added to the environment in large quantities through atmospheric deposition, solid waste disposal, sludge application and wastewater irrigation. Animal feed, drinking water and environmental exposure, for example irrigation of agriculture land with sewage and industrial wastewater, might be sources of heavy metals in animal products such as milk (Awasthi *et al*., 2012). Consumption of milk from cattle and buffalo reared in the polluted sites leads to long exposure to these environmental heavy metals, which results in considerable human health hazards (Kar *et al*., 2015). Milk and dairy products could also become contaminated during manufacturing and packaging (Abdulkhaliq *et al*., 2012). The sources of heavy metals are multiple and their entry into the dairy chain depends on biological variables such as the rate of absorption into the animal body. While this study confirms seasonality, explanation of the inconsistent behaviour of metals in different seasons would require concurrent sampling of possible sources of exposure.

The present findings conclude that, in Jhang, Punjab, Pakistan, raw milk from urban shops and dairy farms near wastewater drains contained undesirable level of Cr, Cd, Pb and Cu during the experimental period. Their mean concentrations in raw milk were higher than permissible limits. Consequently, consumption of adulterated milk by consumers will result in health problems. Hence, there is a need to set legal limits, at least for heavy metals in milk and its products. Pastures polluted with industrial by-products play a key role in contamination of milk and its products with high level of heavy metals. Appropriate risk communication and mitigation strategies are required for farms near wastewater drainage. Detection of levels above the permissible limits in most samples warrants continuous monitoring of these contaminants and a policy for their control. Milk and milk products used for human consumption should be monitored regularly and tested for contamination by toxic heavy metals, as well as for levels of essential trace elements.

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Authors’ Contributions

SR and AK carried out the samples collection and processing; MZ and AHS performed analytical technique and finalized the data; M.A. Idrees analysed the data; QUN, RA and GS helped in drafting; TA co-principal investigator and MY was principal investigator of the project and approved this article for publication.

Conflict of Interest Declaration

The authors declare that they have no conflict of interest.

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