

Heavy Metal Residues in the Milk of Cattle and Goats During Winter Season

Ijaz Javed · Ibadullah Jan · Faqir Muhammad ·
Zia-ur-Rahman · M. Zargham Khan · Bilal Aslam ·
Javed Iqbal Sultan

Received: 12 May 2008 / Accepted: 20 February 2009 / Published online: 5 March 2009
© Springer Science+Business Media, LLC 2009

Abstract Cadmium (Cd), chromium (Cr), nickel (Ni) and lead (Pb) residues (mg/L) were determined in the milk of cattle and goats. The milk samples of cattle from area 1 have higher levels of residues than area 2; Cd 0.089 ± 0.002 vs. 0.062 ± 0.01 Cr 1.14 ± 0.046 vs. 0.995 ± 0.017 Ni 23.38 ± 0.564 vs. 21.407 ± 0.275 Pb 21.781 ± 0.172 vs. 15.958 ± 1.00 . The residual levels of Cd (0.084 ± 0.003) and Pb (42.687 ± 0.051) have been found higher in goat milk. The Ni residues in cattle milk (22.395 ± 0.988) are higher than in goat milk (19.522 ± 0.011) while residues of Cr are non significantly different in both species.

Keywords Heavy metals · Milk · Cattle · Goats

Technological progress, various industrial activities and increased roadway traffic have caused a significant increase in environmental contamination. Ubiquitous presence of some metal pollutants, especially cadmium (Cd), chromium (Cr), arsenic (As), nickel (Ni), mercury (Hg) and

lead (Pb), facilitates their entry into the animal ration and food chain and thus increases the possibility of inducing toxic effects in humans and animals. Land application of sewage sludge, sewage water and industrial wastes gradually increases the toxic metals in the soil environment, which are increasingly taken by plants and subsequently transferred into the food chain causing severe damage to both animal and human health (Somasundaram et al. 2005). The use of sewage sludge in the pastures has been considered as a major source of intake of heavy metals by dairy cattle. Heavy metals like cadmium, lead, mercury and arsenic are the major toxic metals posing a threat to human health. In local studies, cadmium, lead, copper, zinc, iron, chromium and manganese are found at toxic levels in soils and vegetables (Ghafoor and Rasool 1999; Qadir et al. 2000). These studies guide to hypothesize that fodders grown on such soils will also accumulate these heavy metals, and animals reared on contaminated fodders will contain heavy metal residues in edible tissues such as milk. The general public eating such contaminated edible products may accumulate toxic levels of heavy metals.

Faisalabad is the third biggest city of Pakistan with a large number of industrial units which use large volumes of water in their manufacturing and supporting operations. All the domestic and industrial waste water is partly utilized for growing fodders which in turn are used for rearing animals in surrounding and rest of this is discharged in the river Chinab and Ravi through two main drains namely Paharang and Madduana drains (Hussain 2000). Therefore, the present study was proposed to assess the selected heavy metal residues in cattle and goat's milk to safeguard the local public health. This study will help to determine the potential risks from the toxic effects of heavy metals and to make recommendations for future implementations by the local health regulatory authorities.

I. Javed (✉) · I. Jan · F. Muhammad · Zia-ur-Rahman ·
B. Aslam
Department of Physiology and Pharmacology,
University of Agriculture, Faisalabad 38040, Pakistan
e-mail: sandhu_drijaz@yahoo.com

M. Zargham Khan
Department of Pathology, University of Agriculture,
Faisalabad 38040, Pakistan

J. I. Sultan
Institute of Animal Feed and Technology,
University of Agriculture, Faisalabad 38040, Pakistan

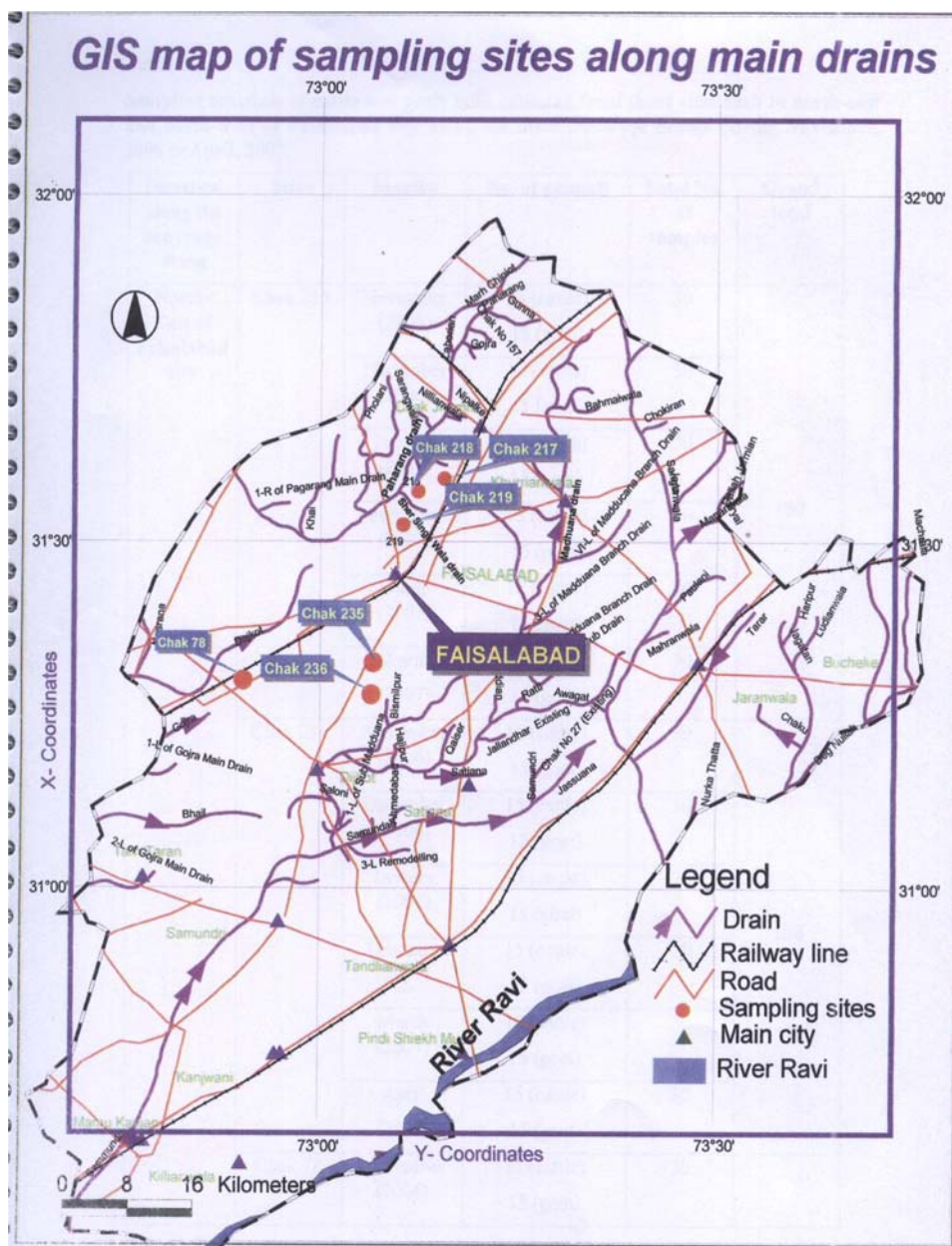
Materials and Methods

Milk samples were collected from two areas along the main sewerage drainage of Faisalabad city. Area 1 was selected in the North-East and Area 2 in the North-West of Faisalabad city. Fifteen animals were randomly selected from each of three sites in Area 1 (Chak No. 235 R.B., Chak No. 236 R.B. and Chak No. 78 R.B.) and similarly the same number of animals from other three sites in Area 2, (Chak No. 217 R.B., Chak No. 218 R.B. and Chak No. 219 R.B.). These three sites in each area were marked with a distance of almost 5 km in between each site (GIS map). Cattle were fed with fodder of the winter season, *Trifolium alexandrinum* (Berseem) and *Medicago sativa* (Lucerne),

cultivated in study areas while goats were used to graze. In these study areas sewerage water is pumped out of sewerage drains for irrigation purposes and there is no alternate source of water for both crops and animals (Fig. 1).

During 1 month, 15 milk samples, each from cattle and goats, were collected from one site. In this way, during the months from November, 2006 to April, 2007, a total of 180 × 6 milk samples of cattle and goats were collected. Milk samples were collected in 25 ml clean sterilized plastic bottles. Prior to milk collection, the udder was washed with water to avoid contamination. Following washing of the udder, the milk samples were collected in the bottle. The samples were kept frozen at -4°C until analysis.

Fig. 1 GIS map of sampling sites along main sewerage drains of Faisalabad, Pakistan



Collected milk samples were subjected to wet digestion by the method as described by Richards (1968). One gram of milk sample was taken into 100 ml digestion flasks and 10 ml of concentrated nitric acid (Riedel-de Haen®) was added. The contents of flask were heated for 20 min. After heating, the sample was cooled at room temperature, adding 5 ml Perchloric acid (Panreac Quimica SA) and was heated vigorously till the white fumes appeared and the sample volume reduced to 2–3 ml. The final volume was made to 50 ml by adding re-distilled water.

Heavy metals (Cd, Cr, Ni and Pb) concentration in milk was determined by atomic absorption spectrophotometric method as described by Licata et al. (2004) through graphite furnace atomic absorption spectrometry.

After the analysis of all the samples, mean \pm SE values were calculated. Student's *t*-test was used to analyze the significance of the results (Steel and Torrie 1996).

Results and Discussion

Mean \pm SE values (mg/L) of Cd, Cr, Ni and Pb in the milk of cattle and goats collected from Area 1 and Area 2 during the month of November, 2006 to April, 2007 have been presented in Tables 1 and 2 while the difference in concentrations of these residues is shown in Table 3.

The mean levels of Cd residues in cattle milk samples collected from Area 1 (0.089 mg/L) were higher than ($p < 0.05$) those collected from the Area 2 (0.062 mg/L) of

Table 1 Mean \pm SE values (mg/L) of Cd, Cr, Ni and Pb in the milk of cattle collected from Area 1 and Area 2 during the month of November, 2006 to April, 2007

Residues	Area 1	Area 2
Cd	0.089 \pm 0.002*	0.062 \pm 0.010
Cr	1.140 \pm 0.046*	0.995 \pm 0.017
Ni	23.382 \pm 0.564*	21.407 \pm 0.275
Pb	21.781 \pm 0.172*	15.958 \pm 1.000

* Significantly ($p < 0.05$) different from the respective value

Table 2 Mean \pm SE values (mg/L) of Cd, Cr, Ni and Pb in the milk of goats collected from Area 1 and Area 2 during the month of November, 2006 to April, 2007

Residues	Area 1	Area 2
Cd	0.081 \pm 0.010 ^{NS}	0.086 \pm 0.002
Cr	1.196 \pm 0.136 ^{NS}	1.107 \pm 0.020
Ni	19.511 \pm 0.174 ^{NS}	19.569 \pm 0.076
Pb	42.738 \pm 0.514 ^{NS}	42.894 \pm 0.076

^{NS} Non significantly ($p > 0.05$) different from the respective value

Table 3 Mean \pm SE values (mg/L) of Cd, Cr, Ni and Pb in the milk of cattle and goats collected from the study areas during the month of November, 2006 to April, 2007

Residue	Cattle milk	Goat milk
Cd	0.076 \pm 0.014*	0.084 \pm 0.003
Cr	1.066 \pm 0.074 ^{NS}	1.152 \pm 0.045
Ni	22.395 \pm 0.988*	19.522 \pm 0.011
Pb	18.870 \pm 2.912*	42.687 \pm 0.051

^{NS} Non Significantly ($p > 0.05$) different from the respective value

* Significantly ($p < 0.05$) different from the respective value

Faisalabad city. These values of present study are higher than their corresponding values in literature. Licata et al. (2004) and Tripathi et al. (1999) reported the levels of Cd in the milk of cattle as 0.0228 and 0.00007 mg/L, respectively. Different workers Baldini et al. (1990), Cerutti (1999) and Martino et al. (2001) found Cd concentration in cattle milk with the range between 0.0002 and 0.03 mg/L which is quite lower than the determined values in the present study (0.046–0.093 mg/L). Hussain (2000) analyzed the metals ion concentration in vegetables irrigated with Faisalabad city effluents and reported the Cd concentration in leaves and fruits as high as 1.5 and 1.00 mg/L, respectively. The goat milk was found richly ($p < 0.05$) contaminated with Cd than cattle milk, 0.084 vs. 0.076 mg/L, respectively. This value in the Area 1 was higher ($p < 0.05$) than the Area 2 by more than 0.08 mg/L. In present study the higher values of Cd residues in the milk of goats (0.084 mg/L) than in the cattle milk (0.076 mg/L) is also supported by the values as determined by Coni et al. (1996) in the milk of goat (0.15 mg/L) and by Licata et al. (2004) in cattle milk (0.0228 mg/L). However, 0.05 mg/L value of Cd residues in the milk of goat has been reported by Caggiano et al. (2005).

Mean Cr residue concentration in the milk samples of cattle collected from Area 1, 1.140 mg/L is significantly ($p < 0.05$) higher than its respective value, 0.995 mg/L from Area 2. Mean Cr concentration in cattle milk ranged from 0.995 to 1.140 mg/L. Chromium concentration in cattle milk was much higher than the literature values. The highest value of Cr residues in the milk of cattle 0.7 mg/L (Cerutti 1999) is 1.7 times less than its corresponding value in the present study. Various workers (Licata et al. 2004; Baldini et al. 1990; Cerutti 1999; Martino et al. 2001) determined the Cr concentration with the range, <0.0015–0.7 mg/L in the milk of cattle. However, in present study, the corresponding value of Cr in the milk of goats is non significantly ($p > 0.05$) different than that in cattle milk (1.152 vs. 1.066 mg/L). The values reported by Coni et al. (1996) as (0.03 mg/L) and Caggiano et al. (2005) as (0.15 mg/L) are far less than the current findings (1.126–1.196 mg/L). The higher value of Cr in goat milk than in literature may be due to the fact that goat has a particular

behavior of nibbling the emerging shoots, leaves, nodes and the tender part of the plants. Chromium deposits are much higher concentration in these parts (Hussain 2000).

The values of Ni residues in cattle milk from Area 1 are higher than those in the milk from Area 2 i.e. 23.382 and 21.407 mg/L, respectively. Anyhow, this difference in the related values of Ni in the milk of goats is negligible (19.511 vs. 19.569 mg/L). The Ni concentration in milk samples of cattle, contrary to cadmium and chromium, was comparatively higher than that in goat milk, 22.4 vs. 19.5 mg/L, respectively. Not much information regarding Ni residual levels in the milk is available in literature. However, Tuzen and Soylak (2004) reported a value below detection concentration of Ni in cattle and goat milk.

The levels of Pb in the milk from Area 1 were higher than the corresponding value in the Area 2 of Faisalabad city, i.e. 21.781 and 15.958 mg/L, respectively. Mean Pb concentration in the milk of cattle was 18.87 mg/L. The industrial byproducts and wastes might have heavy concentration of Pb drained through the effluents irrigating the Area 1. The available MRLs of Pb in cattle milk is 0.02 mg/L (Licata et al. 2004) which was surpassed by many times in all milk samples analyzed in the current study. Other workers (Baldini et al. 1990; Cerutti 1999; Simsek et al. 2000) have also reported Pb concentration in cattle milk, which was although higher than MRLs but much lower as compared to the present study. Almost similar Pb residual concentration has been found in the milk of goats both from Area 1 and Area 2 of Faisalabad city (42.738 vs. 42.894 mg/L). In the goat milk Pb concentration is almost double to that in cattle milk (42.894 vs. 18.870 mg/L). Coni et al. (1996) and Caggiano et al. (2005) have reported Pb residues which are much lower than the values of present study, 0.05 and 0.17 mg/L, respectively in goat milk. The higher levels of Pb in goat's milk contrary to cattle milk may be attributed to the difference in feeding behavior and raising system of goats and cattle.

In present study, the higher milk concentration of heavy metal residues can be attributed to the use of sewerage water for agricultural purposes. It has also been observed that the animals have direct access to this sewerage water for drinking. The uptake of heavy metal residues in the soil, vegetables, fodder and other herbage produced in the Area 1 and Area 2 of Faisalabad city along the main sewerage drains may have a definite role in the contamination of the milk composition. The standard residual risk in the milk of cattle and goats with reference to the concentration of heavy metals has not yet been established at the national level. However, it is certain that milk from various species might have a certain acceptable range of concentration for heavy metals. The current data suggests that the soil contamination have its subsequent effect on the herbage, which is ultimately reflected in the human and animals at their

body or the produce. The findings of the present study show that the Cd, Cr, Ni and Pb residues in the milk of cattle and goats collected from Area 1 are higher as compared to their related values in Area 2 of Faisalabad city. The difference may be attributed to the more industries in the Area 1 of Faisalabad and accordingly more contamination of sewerage water with the heavy metal rich effluents from these industries. The more concentration of Cd, Cr and Pb residues in the milk of goats as compared to the cattle may be due to the fact that during grazing, the goats have more chance to have access to the banks of main sewerage drains where possibly more heavy metal contaminated soil is available. Resultantly, the herbage of this soil will be richer in heavy metals (Ghafoor and Rasool 1999). However, in present findings, the higher concentration of Ni in the milk of cattle than in that of goat may be due to more uptake of Ni in the forage from the more rich contaminated soil with this particular heavy metal. Moreover, the higher Ni residues in cattle milk may also be attributed to the Ni rich industrial effluents, drained into the sewerage water, which ultimately increase the uptake of Ni to the fodder from the soil. Drastic measures are suggested to detoxicate and treat both the industrial and domestic effluents, which are destined to be used for agricultural purposes.

Acknowledgments We thank the Higher Education Commission, Government of Pakistan for funding this work.

References

- Baldini M, Coni E, Stacchini A, Stacchini P (1990) Presence and assessment of xenobiotic substances in milk and dairy products. *Ann Ist Super Sanita* 26:167–176
- Caggiano R, Serena S, Mariagrazia DE, Maria M, Aniello A, Maria R, Salvatore P (2005) Metal levels in fodder, milk, dairy products, and tissues sampled in ovine farms of southern Italy. *Environ Res* 99:48–57. doi:10.1016/j.envres.2004.11.002
- Cerutti G (1999) *Residui additive contaminanti degli alimenti*. 1st edn. Milano Tecniche Nuove
- Coni E, Bocca A, Coppolelli P, Caroli S, Cavallucci C, Marinucci MT (1996) Minor and trace element content in sheep and goat milk and dairy products. *Food Chem* 57:253–260. doi:10.1016/0308-8146(95)00216-2
- Ghafoor A, Rasool I (1999) Zinc, copper, iron and manganese in soils at different canal and water course sections in rice–wheat cropping zone. *Int J Agri Biol* 1:218–221
- Hussain IS (2000) Irrigation of crops with sewage effluent: implications and movement of Pb and Cr as affected by soil texture, Lime, Gypsum and organic matter. Ph. D thesis. Department of Soil Science, University of Agriculture Faisalabad, Pakistan
- Licata P, Trombetta D, Cristani M, Giofre F, Martino D, Calo M, Naccari F (2004) Levels of “toxic” and “essential” metals in samples of bovine milk from various dairy farms in Calabria, Italy. *Environ Int* 30:1–6. doi:10.1016/S0160-4120(03)00139-9
- Martino FAR, Sanchez MLF, Sanz-Medel A (2001) The potential of double focusing-ICP-MS for studying elemental distribution patterns in whole milk, skimmed milk and milk whey of different

- milks. *Anal Chim Acta* 442:191–200. doi:[10.1016/S0003-2670\(01\)01170-9](https://doi.org/10.1016/S0003-2670(01)01170-9)
- Qadir M, Ghafour A, Murtaza G (2000) Cadmium concentration in vegetables grown on urban soils irrigated with untreated municipal sewage. *Environ Dev Sustain* 2:11–19. doi:[10.1023/A:1010061711331](https://doi.org/10.1023/A:1010061711331)
- Richards LA (1968) *Diagnosis and improvement of saline and alkaline soils*, 1st edn. IBH Publications Company, New Delhi Agri. Handbook No. 60
- Simsek O, Gultekin R, Oksuz O, Kurultay S (2000) The effect of environmental pollution on the heavy metal content of raw milk. *Nahrung/Food* 44:360–363
- Somasundaram J, Krishnasamy R, Savithri P (2005) Biotransfer of heavy metals in Jersey cows. *Indian J Ani Sci* 75:1257–1260
- Steel RGD, Torrie JH (1996) *Principles and procedures of statistics. A biometrical approach*, 3rd edn. McGraw Hill Book Company, New York, pp 336–352
- Triphathi RM, Raghunath R, Sastry VN, Krishnamoorthy TM (1999) Daily intake of heavy metals by infants through milk and milk products. *Sci Total Environ* 227:229–235. doi:[10.1016/S0048-9697\(99\)00018-2](https://doi.org/10.1016/S0048-9697(99)00018-2)
- Tuzen M, Soylak M (2004) Column system using diaion HP-2MG for determination of some metal ions by flame atomic absorption spectrometry. *Anal Chim Acta* 504:325–334. doi:[10.1016/j.aca.2003.10.043](https://doi.org/10.1016/j.aca.2003.10.043)