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Spatial distribution of Pb, As, Cd contents in Chinese raw milk and risk assessment for human health

X. W. Zhou[1,2], X. Y. Qu[1], N, Zheng[1], C. Y. Su[1], J. Q. Wang[1]*

[1] Ministry of Agriculture-Key Laboratory of Quality & Safety Control for Milk and Dairy Products, Institute of Animal Science, Chinese Academy of Agricultural Sciences, Beijing 100193, P. R. China; [2] Statistics, Informatics and Applied Modeling lab, AGROBIOCHEM dep., Gembloux Agro-Bio Tech, University of Liège, Gembloux, Belgium.

INTRODUCTION

Milk and dairy products contain protein, fat and essential elements necessary for human health, especially for body metabolism, growth and development. However, heavy metals such as Pb, As, and Cd may contaminate raw milk and involve serious systemic health problems when they are in excessive concentration in human body (Oliver, 1997). Milk may contaminant due to local environmental contamination (Kazi et al., 2009). Drinking water contamination is the most important route of As exposure, whereas food is another source, because agricultural products can accumulate As from contaminated soil and water (Ohno et al., 2007). The objective of this study was first to investigate the spatial distributions of Pb, As and Cd in raw milk in the 10 main milk producing areas in China. Second, the health risk related to the content heavy metals in milk was evaluated.

MATERIALS & METHODS

Raw milk samples (n=997) were collected in ten main milk producing areas in China in May and August 2016. Milk samples were stored in 200 mL polypropylene bottles at -20 ° C after collection. All the reagents were of analytical pure. Water used in the analytical progressing was obtained from a Milli-Q Plus water purification system (18.2 MΩ cm) (Millipore, USA). Nitric acid (65 %) and hydrogen peroxide (30 %) used in sample digestion were obtained from Merck (Germany). Working standard solutions were prepared from multi-element stock standard solution (10 mg/L, SPEX, USA). All the vessels were immersed in 20% HNO3 (v/v) for at least 12 h and rinsed with ultrapure water before analysis. Samples were measured by ICP-MS after microwave-assisted acid digestion. Limit of detection (LOD) for the analytical method has been detailed in previous study (Zhou et al., 2016). Samples spiked with standard solution were used to assess the accuracy of the method. Mean recovery of analytical method of Pb, As and Cd in milk was 97.1%, 94.4% and 95.5%, respectively (Table 1). The results of recovery accord with precision of quantitative methods. Non-normal distribution Turkey's HSD tests and Spearman correlations were calculated using SPSS 17.0. The spatial variability of Pb, As and Cd was performed using ArcGIS 10.2.

RESULTS

Mean values of Pb, As and Cd in raw milk samples are presented in table 1. Values above LOD were used for mean value calculation. Element concentrations below LOD were replaced by half the value of the respective detection limits. Levels of Pb in 13 milk samples (1.30% of collected samples) were upper than maximum residue limit (MRL) imposed by the European Union (0.02mg/kg). However, all samples were below the Chinese MRL (0.1mg/kg). Pb, As and Cd showed different spatial patterns. Spatial distribution of Pb was shown in figure 1.

Contents of Pb in raw milk in area E (43° 55′-45° 45′ N, 84° 11′-87° 67′ E) was significantly ($p<0.05$) higher than other in other studied areas. Contents of As and Cd in area J (31° 21′-34° 28′ N, 115° 09′-118° 75′ E) were significantly ($p<0.05$) higher （Table 2）. Content of Cd in milk was significantly ($p<0.01$) correlated with Pb and As contents. In addition, Pb in raw milk was significantly ($p<0.01$) correlated with As. Health risk assessment of heavy metals for adults was determined by the calculation of hazard index. The quotient value was < 1 for Pb, As and Cd in raw milk, which indicated that there is potentially no health risk.

CONCLUSION

The result indicated significant correlations among Pb, Cd and As in raw milk. Samples located in area E were close to the mines, which may explain the high level of Pb observed in this region. Further studies are required to clarify the relationships between the contamination of raw milk by heavy metals and the herd environment.

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Table 1. Descriptive statistics of elements in raw milk (n=997).

	Positive rate (%)	Spike recovery (%)	Mean ± SD	P25	Median	P75
Pb	68.5	97.1	2.64±5.67	0.14	0.80	1.91
As	46.5	94.4	0.61±1.43	0.05	0.05	0.25
Cd	77.4	95.5	0.07±0.32	0.01	0.03	0.06

Mean values in the table was calculated with values above the LOD.
Concentrations expressed in μg/L.

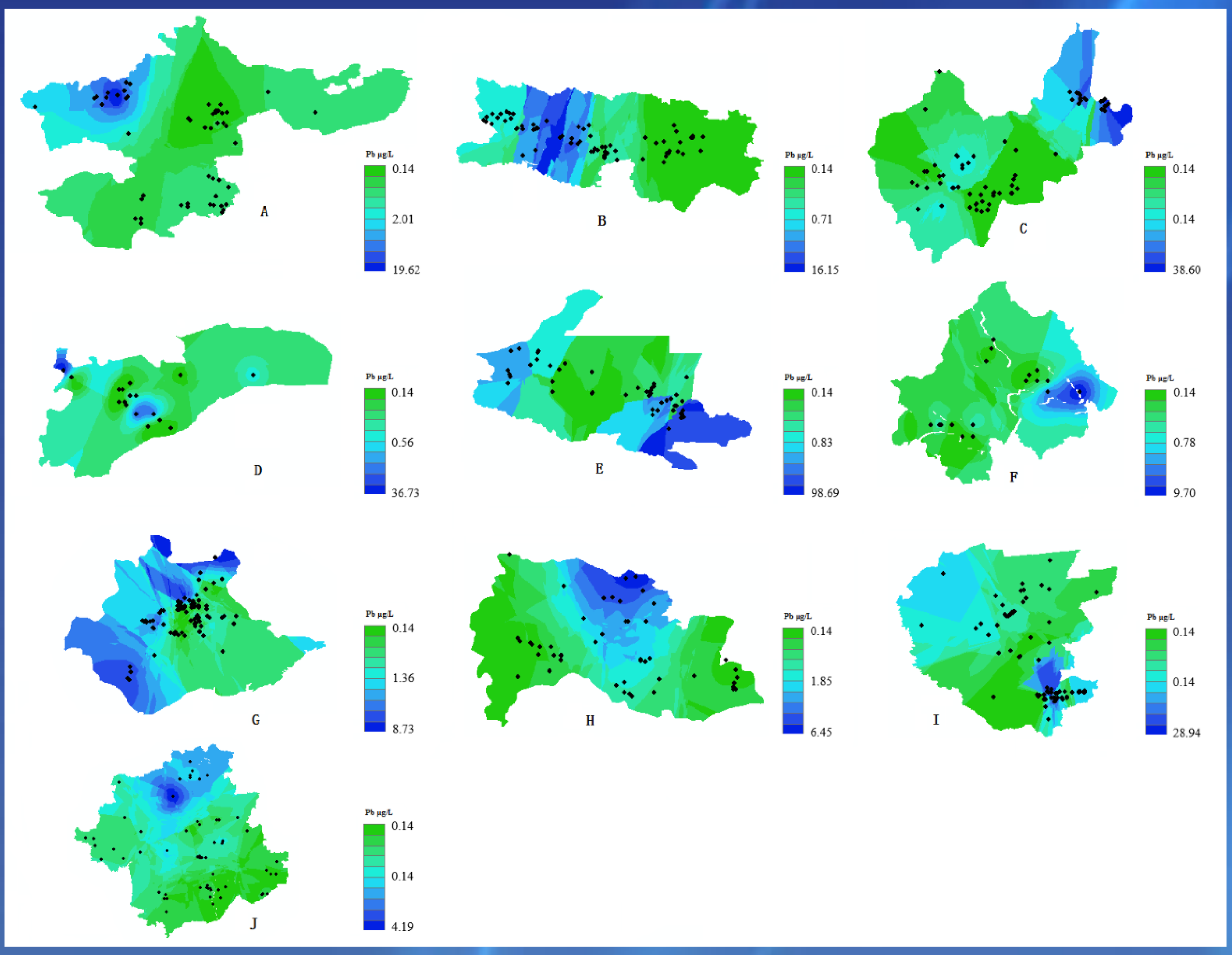

Figure 1. The distribution of Pb in raw milk samples in areas A-J.
Dots on the map are the locations of sampling.

Table 2. Concentrations of Pb, As and Cd in raw milk from different areas.

	A	B	C	D	E	F	G	H	I	J
Pb	2.33^{abc}	2.10^{ab}	3.61^{ab}	4.46^{bc}	4.75^{c}	1.73^{ab}	2.09^{abc}	2.61^{ab}	0.94^{a}	1.85^{ab}
As	0.44^{a}	1.05^{ab}	0.62^{a}	0.33^{a}	1.10^{a}	0.24^{a}	0.29^{a}	0.37^{a}	0.36^{a}	1.19^{b}
Cd	0.09^{ab}	0.04^{a}	0.12^{ab}	0.07^{ab}	0.06^{ab}	0.03^{ab}	0.06^{ab}	0.04^{ab}	0.04^{ab}	0.16^{b}

Values in the table are average. Concentrations expressed in μg/L.
Within a column, values with a different letter means significant difference ($p < 0.05$)