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Heavy metals occurrence in Italian food supplements

P. Brizio[1] , A. Benedetto[2], S. Squadrone[3], R. Tarasco[4], S. Gavinelli[5], M. Pellegrino[6] and M. C. Abete[7]

[1] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY paola.brizio@izsto.it
[2] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY alessandro.benedetto@izsto.it
[3] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY stefania.squadrone@izsto.it
[4] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY renata.tarasco@izsto.it
[5] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY contaminanti.ambientali@izsto.it
[6] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY matteo.pellegrino@izsto.it
[7] Istituto Zooprofilattico sperimentale del Piemonte, Liguria e Valle d'Aosta, Via bologna, 148 ,10154 Torino, ITALY mariacesarina.abete@izsto.it

Abstract. In recent years a significant increase in food supplements consumption has been observed, maybe in the belief that they couldn't be dangerous for consumers health, even if they don't achieved medical effects. However, environmental pollution can cause heavy metals contamination that could exceed maximum levels established by European legislation. Aim of this work was to evaluate arsenic, cadmium, chromium, lead and mercury content in 12 food supplements seized in a Piedmont shop by the Italian authority against food adulteration. All metals were analysed after mineralization and dilution steps by ICP-MS, with the exception of mercury, detected by the direct analyser TDA-AAS. Only one sample exceed the European maximum limits for lead (3,00 mg/kg) but warning levels of chromium (over 3,00 mg/Kg) has been detected in three of them.

Key words: Heavy metals, food contaminants, food supplements

Introduction

A significant increase in food supplements consumption has been observed in the last years; this can mainly be due to performance enhancing or cosmetic, but more and more often it is an attempt to compensate for lack of nutrition in diet or exercise (Petroczi et al., 2011). Public opinion is supposed to believe that natural products can't represent an health risk and that, even if expected medical effects are not achieved, there are no adverse effects. However, as known, plants and other organism in polluted areas can be contaminated by heavy metals present in the environment from industrial or traffic emission and agricultural expedients . For these reasons the European Commission set in Regulation (EC) No 629/2008 maximum levels for certain contaminants in foodstuffs, particularly food supplements: 3,00 mg/Kg of lead (Pb), 0,10 mg/Kg of mercury (Hg) and 3,00 mg/Kg of cadmium (Cd).

No limits were established for arsenic or chromium, which is also included by the Commission Regulation (EC) No 1170/2009 and Regulation (EC) No 1925/2006 in the list of minerals that may be used in the manufacture of food supplements.

In November 2011, the Italian authority against food adulteration (NAS) seized in a Piedmont shop several commercialized food supplement to evaluate the heavy metal content. Analysis were performed by the chemical laboratory of the Istituto Zooprofilattico Sperimentale del Piemonte, Liguria e Valle d'Aosta, in Turin.

Materials and Methods

Six herbal samples (two of *Equisetum maximum* and *Solidago virgaurea,* one of *Crithmum maritimum* and

one of *Artemisia abrotanum*) were commercially available as air dried and powdered leaves, sprigs, cortex or flowers; five food supplements were sold as tablets made of different botanicals and botanical preparations mixtures and only one was a bulk mix.

All reagents used for analysis were of analytical-reagent grade.

Quantification of Hg was carried out by direct analyzer TDA-AAS (Thermal Decomposition Amalgamation and Atomic Absorption Spectrophotometry), without sample preparation. Determination of As, Cd, Cr and Pb was performed on a ICP-MS (Inductively Coupled Plasma-Mass Spectrometry) system after mineralization and dilution steps; samples weighted in Teflon digestion vessels were added of: 7 mL of concentrated nitric acid, 1,5 mL of 30%v/v hydrogen peroxide and 50 µL of concentrated hydrofluoric acid. Wet decomposition of samples were performed by a microwave digestion system.

All elements were quantify against standard calibration curve prepared at the day of analysis.

Limits of quantification (LOQ) were 0,01 mg/Kg for As and Cr, 0,02 mg/Kg for Cd and Pb and 0,03 mg/Kg for Hg.

Results and Discussion

Table 1 and Table 2 show mean, maximum and minimum heavy metals levels in single (N=6) and mixed (N=6) herbal products, while Figure 1 represent a comparison between them. Results reported to be <LOQ were handled by substitution and the value was imputed as one half of the limit of quantification (LOQ/2).

Table 1. Heavy metal content in single herbal samples

	mg/Kg in single herbal samples		
	Mean	Minimum	Maximum
As	0,06	0,01	0,20
Cd	0,07	0,01	0,15
Cr	1,25	0,26	3,55
Pb	0,78	0,08	1,68

Table 2. Heavy metal content in mixed herbal samples

	mg/Kg in mixed herbal samples		
	Mean	Minimum	Maximum
As	0,19	0,03	0,66
Cd	0,06	0,04	0,12
Cr	3,87	1,11	14,01
Pb	1,61	0,42	3,92

All samples analyzed presented Hg concentration under LOQ, while detectable levels of arsenic, cadmium, chromium and lead were found.

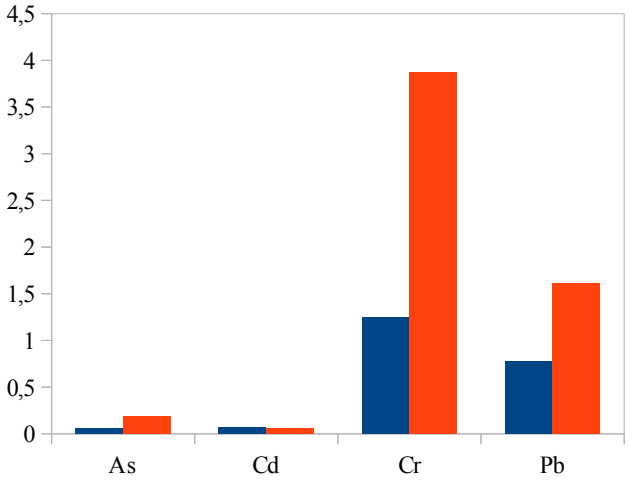

Fig. 1. Comparison between heavy metals content (mg/Kg) in single herbal (blue) and herbal mix (orange).

Only one of the food supplements exceed existing maximum levels established by European Legislation for lead, with a concentration of 3,92 mg/Kg; the same sample revealed also the higher chromium concentration (14,01 mg/Kg). This integrator was sold as tablets made of a mixture of *Solidago virgaurea, Actostaphyos, Agromonia eupatoria, Vaccinum Mirtillus, Pygeum africanum* and food gelatin; however we could not assign the contamination origin to one of the botanical components because other preparations containing one or more plants in common had lower metal levels. For example, one of the herbal sample of *Solidago virgaurea* had a content of chromium of 3,55 mg/Kg and of lead of 1,68 mg/Kg, while the other one had a concentration of chromium of 0,26 mg/Kg and of lead of 0,15 mg/Kg.

We also observed a big spread, particularly for chromium and lead, between sample types: herbal samples made of a single plant had higher metals concentration than those made of a mixture, probably due to the different contributors contamination.

Conclusion

Some types of heavy metals are natural essential components of enzymes and coenzymes, such as chromium; however an excessive and uncontrolled intake of this element in diet (estimates of daily intake by Codex 1995 range from 0,02-0,20 mg/day) could implicate serious consequences on health.

As seen, food supplements could contain heavy metals residues under legislation maximum levels, but for those metals without established limits, major control could be recommended in order to evaluate daily exposure and risk assessment of consumers.

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