# Investigations Of Heavy Metals In Commercial Spices Brands

Hifsa Mubeen, Ismat Naeem*, Abida Taskeen and Zeb Saddiqe.

Lahore College for Women University, Lahore
Jail Road Lahore, Pakistan.
Tel: 92-42-9203801-9/245
Email: ismat4_naeem@yahoo.co.in

**Abstract:** In present study concentrations of some heavy metals such as iron(Fe), copper(Cu), chromium(Cr), lead (Pb), cadmium(Cd) and cobalt (Co) present in common spices of two brands widely used in Pakistan coded as A & B were determined using atomic absorption spectroscopy. The studies showed differences in metal concentrations in different spices samples. The concentration of Fe ranged from 144.5 to 1260 $mgkg^{-1}$ on dry weight basis, where as that of Cu was ranged from 9 to 44 $mgkg^{-1}$ to 3.05 mg $kg^{-1}$. The concentration level of Cr was from 115 to368 mg $kg^{-1}$. Concentration of Co and Cd varied little and ranged from 11.5 to 15 $mgkg^{-1}$ and 0.5 to $mgkg^{-1}$ respectively. While variable levels of Pb were detected from 54 to 70 mg kg-1. Daily intake limit was calculated and compared with MRL (minimum risk level) values given by ATSDR (2001). Results showed that concentrations of Cr and Pb of all spices samples under study were much lager than those of MRL values. Thus intake of these spices can cause accumulation of these hazardous metals in body. Metal to metal correlation study showed strong correlation between. Pb, Cu and Co. [New York Science Journal. 2009;2(5):20-26]. (ISSN: 1554-0200).

**Keywords:** Heavy metals, spices, atomic absorption spectroscopy.

**Abstract:**
In present study concentrations of some heavy metals such as iron(Fe), copper(Cu), chromium(Cr), lead (Pb), cadmium(Cd) and cobalt (Co) present in common spices of two brands widely used in Pakistan coded as A & B were determined using atomic absorption spectroscopy. The studies showed differences in metal concentrations in different spices samples. The concentration of Fe ranged from 144.5 to 1260 $mgkg^{-1}$ on dry weight basis, where as that of Cu ranged from 9 to 44 $mgkg^{-1}$. The concentration level of Cr was from 115 to368 mg $kg^{-1}$. Concentration of Co and Cd varied little and ranged from 11.5 to 15 $mgkg^{-1}$ and 0.5 to 2$mgkg^{-1}$ respectively. While variable levels of Pb were detected from 54 to 70 mg kg-1. Daily intake limit was calculated and compared with MRL (minimum risk level) values given by ATSDR (2001). Results showed that concentrations of Cr and Pb of all spices samples under study were much lager than those of MRL values. Thus intake of these spices can cause accumulation of these hazardous metals in body. Metal to metal correlation study showed strong correlation between. Pb, Cu and Co.

**Keywords:** Heavy metals, spices, atomic absorption spectroscopy.

## Introduction:

Trace metals composition of foods is of interest because of their essential or toxic nature [(1)]. The accumulation of heavy metals can have middle-term and long term health risks, and strict periodic surveillance of these contaminants is therefore advisable [(2)]. Micronutrients constitute a small fraction of the entire diet but play important roles in different metabolic processes [(3)]

Food composition data is important in nutritional planning and provides data for epidemiological studies [(4)]. Environmental pollution is the main cause of heavy metal contamination in food chain. The trace metal contents of individual foods varies and is dependent upon the trace metals introduced in the growing, transport, processing and fortification of food [(5)]. The other technological processes used to bring the food to the consumer can significantly increase the total trace metal contents of the food [(6,7)]

Spices are dried parts of plants, which have been used as diet components often to improve color, aroma, palatability and acceptability of food. Most of these are fragrant, aromatic and pungent. Natural food spices such as pepper and mustard have been reported to contain significant quantities of some trace metals [(8)]. These trace metals in spices and medicinal plants play vital role as structural and functional components of

metalloprotiens and enzymes in living cells [9]. The addition of spices –that may be contaminated with trace and heavy metals- to food as a habit may result in accumulation of these metals in human organs. Subjecting to trace and heavy metals above the permissible affect the human health and may result in illness to human fetus, abortion and preterm labor, and mental retardation to children. Adults also may experience high blood pressure, fatigue and kidney and brain troubles.

Pakistan has a high diversity of plants used as spices, herbs, and traditional medicines. Several herbs and spices are either produced on small farmlands or naturally grow in different regions. There is little information available about the safety of those plants and their products in respect to heavy metal contamination. In Pakistan majority of population use more spices than any other beverages. Due to the use of enormous amount of spices daily, it is important to know the toxic metal contents in these The objective of this work is to estimate the levels of some heavy metals i.e. lead, cadmium, cobalt, iron, copper, and chromium that may be present in two major spices brand available in local markets in Pakistan. Also, the levels of investigated metals were compared with recommended with recommended levels by the International Organizations.

**Materials and methods:**

**Sample collection and processing:**

Various food taste enhancers (seasoning and culinary condiments) and spices including chili powder, black pepper powder, tumeric powder, and different mix spices e.g. garam masala powder, chat masala mix, quorma masala mix and biryani masala mix of brand A & brand B were purchased from liberty market Lahore. These food condiments represented the most widely used taste enhancers in Pakistan. Spices from open market were also purchased coded as C. A total of fifty one samples were collected (three of each type) and analyzed.

**Analysis:**

The samples were carefully opened and dried to constant weight.1g sample was digested with 20mL of 2:1 $HNO_3$ /$HClO_4$ (Anal grade), and heated until evolution of white fumes. Where necessary more acid mixture was added and the sample digested until evolution of white fumes marking the end of the digestion process. The digests were filtered into standard 50mL volumetric flask and made up to mark with distilled water. This was subsequently analyzed for Pb, Cd, Cu, Cr, Co and Fe by air-acetylene flame atomic absorption spectrometry (Hitachi Z – 5000) by the standard calibration technique. Calibration standards were prepared by dilution of the high purity commercial metal standards (Applichem) for atomic absorption analysis. Adequate quality assurance measures were carried out to ensure reliability of results. Glassware was properly cleaned and reagents ($HNO_3$, $HClO_4$ and distilled water) were of analytical grade. Spikes and blanks were also introduced. Results reported are average of duplicates.

The daily intake ($mgkg^{-1}day^{-1}$) was calculated based on these suppose

1) The human weight is 50 kg and

2) The human intake from spices per day is 20 g.

The daily intake ($mgkg^{-1}day^{-1}$) = metal concentration in spice × 20/1000 /50 (Eq.1)

**Results and Discussion:**

Fifty one samples of spices available in markets of Pakistan were analyzed for iron, copper, chromium, cobalt, cadmium and lead. Of the total samples analyzed all the six metal were detected in each sample. Iron was present in highest amount followed by Cr and Pb. Metal to metal correlation showed that only three elements Pb, Cu and Co were strongly correlated in spices samples. Significant correlation was found ($r>0.5$) between Pb & Cu and Pb and Cu.

**Iron:**

As revealed by analytical results ( see Table 2) iron content of spices samples ranged between 144.5 to $1260mgkg^{-1}$. The highest mean level of Fe was found in sample C-1 chili powder from open market. On the other hand, lowest mean value was found in sample B-1. The samples B-4 and B-2 and C-3 were relatively richer in Fe concentration.

Although there was a high content of Fe in all the samples, but daily intake was less then MRL (minimal risk level) value (see Fig: 1). So Fe intake from spices has no effect on health.

Fe is an essential element .It is a constituent of active site of various reproductive hydrogenases, most frequently associated with sulfur containing ligands. Fe together with heamoglobin and ferrodoxin plays a

central role of metabolism. Fe facilitates the oxidation of carbohydrates, proteins and fat to control body weight which is an important factor in some diseases (diabetes)[10,11].

**Copper:**

As revealed by analytical results (see Table 2) copper content of spices samples ranged between 9 to 44mgkg$^{-1}$ The highest mean level of copper was found in sample A-1 and lowest mean value was found in sample A-3. In all other samples the concentration was close to 15 to 25mgkg$^{-1}$. Daily intake was much less then MRL (minimal risk level) value in all samples (see Fig 2). Copper intake from spices has no effect on health.

Although copper is an essential element in trace amount but can be toxic at excess level. Copper build can result in a tendency for hyperactivity in autistic children. An excess of copper can cause oily skin loss of skin tone (due to ability to block vitamin C) and cause a dark pigmentation of skin specially, around face. It can attribute to hair loss specially, in women[12].

**Chromium:**

In case of chromium highest mean concentration was found in sample B-4 and lowest mean concentration in B-7. Chromium particularly Cr (III) plays an important role in the body function in trace amount but it is toxic in excess amount. Cr (VI) is toxic and have no role in body. Daily intake values were less than MRL values for Cr (III) in all samples while higher than those for Cr (VI) (see Fig 3). MRL values are not a bright line for health risk but as the distance of experimental daily intake increases from MRL increase risk level also increases. So all the spices samples under study are source of Cr accumulation in body and are thus health hazards.

**Cobalt:**

In case of cobalt there was a small variation in concentration for all samples ranging from 11.5 to 15mgkg$^{-1}$. Daily intake values were found to be much lower than MRL values (see Fig 4). So there is no effect on health due to intake of cobalt from spices. Although cobalt is toxic at elevated levels, however the body needs in small amount. Co in the form of vitamin $B_{12}$ is in active physiological form.

**Cadmium:**

There was a little variation in case of cadmium. The concentration ranged between 0.5 to 2 mgkg$^{-1}$ while permissible limit of Cd is 6 mgkg$^{-1}$ for all foods in Pakistan[13]. Daily intake values were much lower than MRL values (see Fig 5). So there was no harm by intake of cadmium from spices under study.

**Lead:**

As revealed by the analytical data high concentration of Pb was found in A-1 (70mgkg$^{-1}$). In other samples concentration of Pb ranged between 54 to57.5 mgkg$^{-1}$ i.e. greater than permissible standard limit of Pb (0.3mgkg$^{-1}$) for herbs. Daily intake was much higher than MRL values (see Fig 6). It means intake of spices under this study can cause Pb accomulation in body. It has been reported to competitively inhibit Pb uptake in cells [14]. Pb is a heavy metal poison which forms complexes with oxo-groups in enzymes to affect virtually all steps in the processes of heamoglobin synthesis and porphyrin metabolism [15]. Toxic levels of Pb in man have been associated with encephalopathy seizures and mental retardation [16].

**Conclusion:**

On the basis of results it can be concluded that the spices of two widely used brands in Pakistan and spices from open markets of Pakistan are not only source of trace metals but also source of contamination of toxic heavy metals especially Pb(II) & Cr(III). Thus excess use of these spices in foods is health hazard.

**Acknowledgement:**

The authors want to acknowledge Higher Education Commission for the research grant awarded to Prof. Dr. Ismat Naeem for stable funding required to carry out this research work.

**Table 1: Instrumental parameters used by FAAS (Hitachi Z - 5000).**

| Metal | Wavelength (nm) | Slit width (nm) | Air:acetylene ($Lmin^{-1}$) | Upper measureable limit ($mgL^{-1}$) |
|---|---|---|---|---|
| Fe | 248.3 | 0.2 | 2.0 | 20 |
| Cu | 324.8 | 1.3 | 2.2 | 30 |
| Cr | 359.3 | 1.3 | 2.8 | 100 |
| Co | 240.7 | 0.2 | 2.2 | 10 |
| Cd | 228.8 | 1.3 | 2.0 | 6 |
| Pb | 283.3 | 1.3 | 2.2 | 200 |

**Table 2:** Levels of lead, cadmium, cobalt, iron, copper, and chromium present in spices samples of two brands available in local markets in Pakistan.

| **Spices Sample** | **Fe ($mgkg^{-1}$)** | **Cu ($mgkg^{-1}$)** | **Cr ($mgkg^{-1}$)** | **Co ($mgkg^{-1}$)** | **Cd ($mgkg^{-1}$)** | **Pb ($mgkg^{-1}$)** |
|---|---|---|---|---|---|---|
| A1 | 716±3.1 | 44±0.8 | 207.5±2.2 | 15.5±0.5 | 2±0.08 | 70.5±0.8 |
| A2 | 536.5±2.1 | 20±1.7 | 134±1.3 | 12±0.45 | 1±0.1 | 57.5±5.2 |
| A3 | 416±0.7 | 9±1.1 | 175±0.8 | 11.5±0.3 | 1±0.1 | 55.5±0.8 |
| A4 | 183±4.1 | 21.5±0.5 | 162.5±0.7 | 11.5±0.5 | 0.5±0.05 | 56.5±3.4 |
| A5 | 577±2.3 | 15±0.8 | 205±1.2 | 11.5±0.02 | 1±0.02 | 55.5±0.9 |
| A6 | 372±4.5 | 18±0.09 | 204.5±1.5 | 11.5±0.1 | 1±0.1 | 56.5±1.7 |
| A7 | 835±2.2 | 16±0.6 | 192.5±0.9 | 13±0.09 | 0.5±0.4 | 57.5±1.3 |
| B1 | 144.5±4.3 | 26.5±1.3 | 152±1.5 | 11.5±0.05 | 1±0.2 | 57.5±1.5 |
| B2 | 523.5±2.2 | 26.5±1.1 | 183.5±2.2 | 11.5±0.1 | 1±0.1 | 57.5±2.5 |
| B3 | 405±0.7 | 13.5±1.1 | 175.5±0.6 | 11.5±0.8 | 1±0.05 | 57.5±1.2 |
| B4 | 1181.5±5.2 | 24±1.8 | 368.5±1.8 | 14±0.2 | 1±0.08 | 57.5±1.1 |
| B5 | 619±0.7 | 16±0.6 | 192.5±1.7 | 13±0.2 | 0.5±0.1 | 57.5±0.5 |
| B6 | 352±3.2 | 19.5±1.8 | 226±2.5 | 12±0.7 | 1±0.05 | 55.5±1.8 |
| B7 | 699.5±1.3 | 15.5±1.8 | 115±2.2 | 11.5±0.5 | 1±0.05 | 57.5±2.4 |
| C1 | 1260±4.9 | 21.5±1.2 | 123±2.5 | 12±0.3 | 1±0.0.3 | 56.5±1.3 |
| C2 | 1024±2.4 | 26.5±1.9 | 165±3.1 | 11.5±0.8 | 1±0.05 | 54.5±1.5 |
| C3 | 1171±1.2 | 20.5±1.3 | 150±1.7 | 12±0.6 | 0.8±0.011 | 54±1.4 |

**Table 4: Multiple Metal Correlation Coefficient Matrixes for Various Metals in Spices Samples**

| | Fe | Cu | Cr | Co | Cd | Pb |
|---|---|---|---|---|---|---|
| Fe | 1.0 | 0.0328 | 0.0605 | 0.1239 | 0.0394 | 0.0313 |
| Cu | | 1.0 | 0.0673 | 0.4585 | 0.3466 | 0.5686 |
| Cr | | | 1.0 | 0.2475 | 0.0029 | 0.0364 |
| Co | | | | 1.0 | 0.3232 | 0.561 |
| Cd | | | | | 1.0 | 0.363 |
| Pb | | | | | | 1.0 |

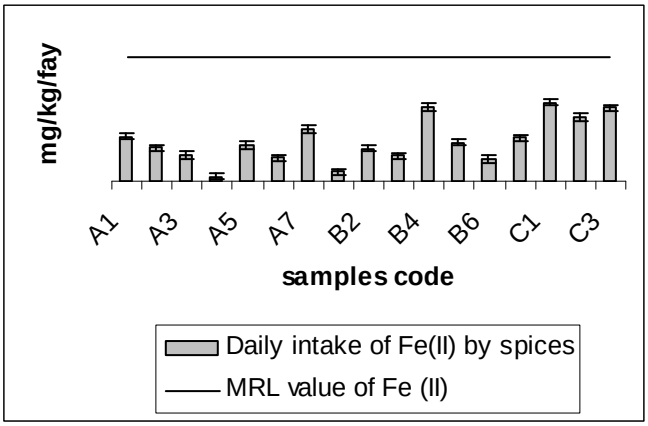


Fig 1: Comparison of MRL values with daily intake to assess health effect of Fe(II) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

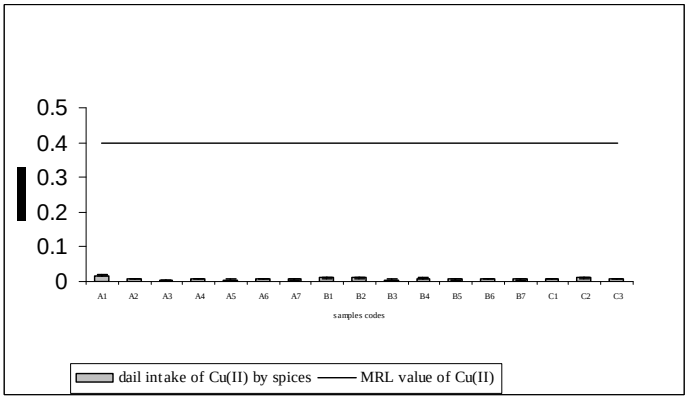


Fig 2: Comparison of MRL values with daily intake to assess health effect of Cu(II) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

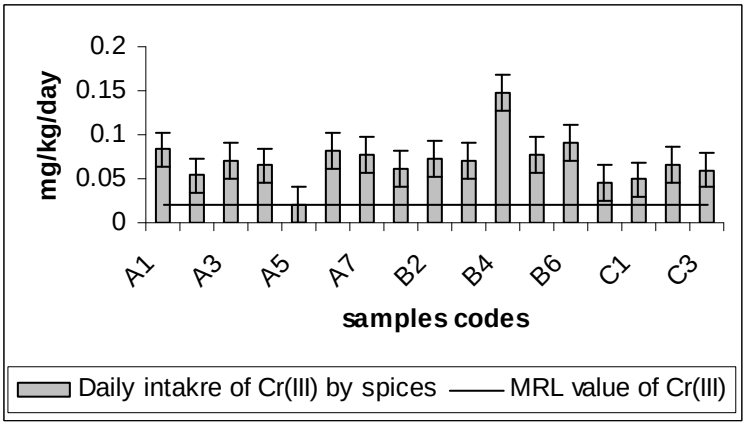


Fig 3: Comparison of MRL values with daily intake to assess health effect of Cr(III) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

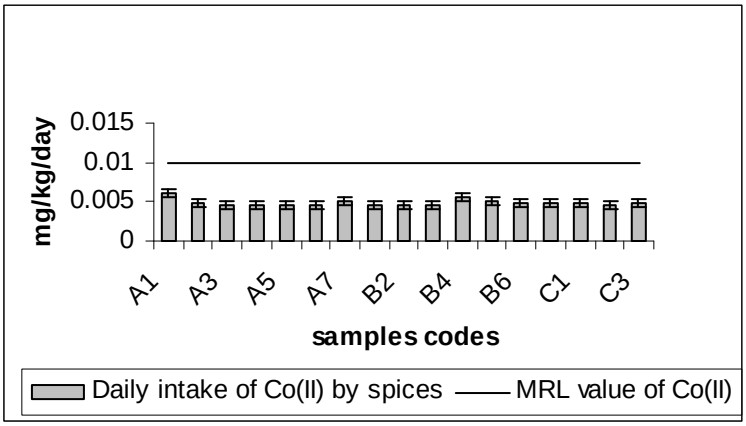


Fig 4: Comparison of MRL values with daily intake to assess health effect of Co(II) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

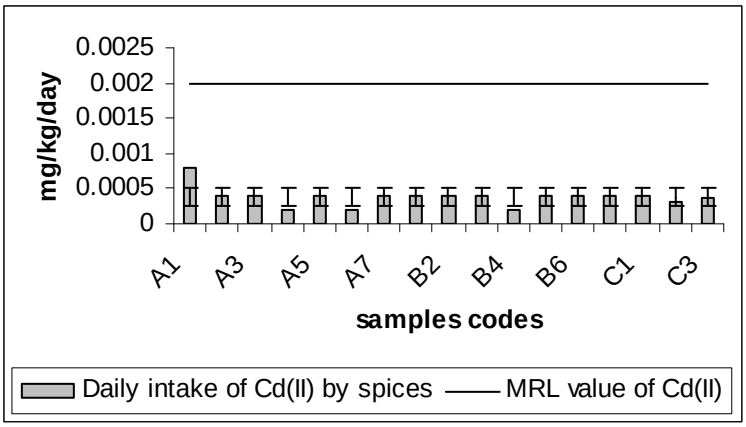


Fig 5: Comparison of MRL values with daily intake to assess health effect of Cd(II) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

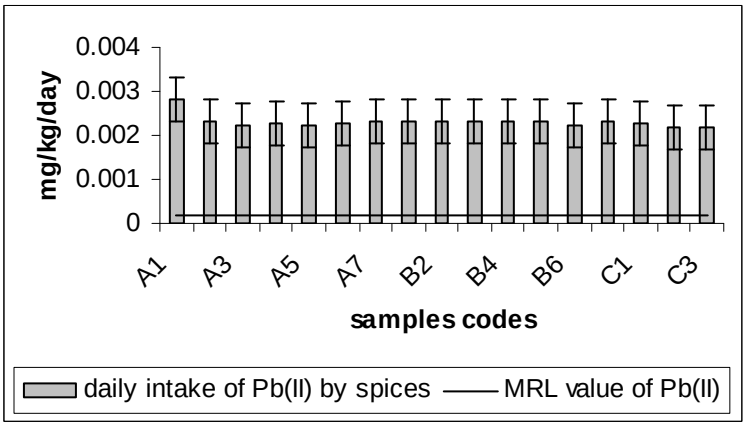


Fig 6: Comparison of MRL values with daily intake to assess health effect of Pb(II) assuming daily intake of spices being studied is 20g & wt. of human body is 50kg.

**References:**


1. Onianwa PC, Adetola IG, Iwegbue CMA, Ojo MF, Tella OO. *Food Chem.* 1999; 66: 275-279.
2. Cabrera C, Lorenzo ML, Lopez CM. *J. Agri. Food Chem*.1995; 43: 1605-1609
3. Akhter P, Akram M, Orfi SD, Ahmad N. *Nutr.* 2002; 18: 274-278.
4. Bruce A. and Bergstrom L. *Food Nutr. Bull.* 1983; 5: 24-29.
5. Anderson R.A, . Bryden NA, Polansky MM. *Biol. Trace Elem. Res.* 1992; 32: 117-121.
6. Cabrera C, Lloris F, Gimenez R, Olalla M, Lopez C. *The Science of The Total Environ.* 2003; 308: 1-14.
7. Tsoumbaris, P. and Tsoukali-Papadopoulou, H. *Bulletin of Environmental Contamination and Toxicology*.1994; 53 (1): 61-66.
8. Gupta KK, Bhattacharjee S, Kar S, Chakrabarty S, Thakar P, Bhattacharyya G and Srivastava SC. *Comm. Soil Plant Anal*. 2003; 34: 681-693.
9. Ansari TM, Ikram N, Najam-ul-Haq M, Fayyaz O, Ghafoor I and Khalid N. *J. Biol. Sci.* 2004; 4: 95-99.
10. Rajukar N.S. and Perdeshi B.M *Appl.Radiat. Isot* 1997; 48: 1059
11. Ya Sakolnik M.Elsevier, Amsterdam,1948; 25
12. Gupta U.Copper in the Environment. J.O.Nriagu ed. 255, John Wiley and Sons Inc., New York, 1975
13. Walker JM. 19881. Proceedings No 2 “Cadmium Accumulations in Australtin Agriculture: National Symposium, Canberra, 1-2 March 1988”, Australian Government Publishing Service, Canberra, 176-85.
14. Lou M, Garray R, Alda J. *J.Physiol.* 1991; 443: 123-126.
15. Ademorati CMA 1996. *Environmental Chemistry and Toxicology Pollution by Heavy metals*. Foludex press Ibadan.1996; 171–172.
16. Schumann K. *Z. Ernahrungswiss* 1990 ; 29: 54–73.


4/5/2009