

ORIGINAL ARTICLE

EVALUATION OF TOXIC HEAVY METALS IN AYURVEDIC SYRUPS SOLD IN LOCAL MARKETS OF HAZARA, PAKISTAN

Bibi Hajra, Iftikhar Qayum*, Shaukat Orakzai, Fida Hussain, Uzma Faryal, Aurangzeb

Department of Biochemistry, *Director Medical Research, Women Medical College, Abbottabad-Pakistan

Background: Herbal and Ayurvedic preparations, widely used in Pakistan and the developing world, present serious risk of heavy metal toxicity related to their medicinal content and prolonged use by patients. The objective of this study was to find out the concentration of heavy metals in Herbal & Ayurvedic liquid preparations commonly used for treatment of different diseases, from local markets of Hazara. **Methods:** The cross sectional survey of traditional herbal & Ayurvedic medicine shops included ten liquid preparations selected from local shops of Mansehra and Abbottabad after interviewing the shopkeepers; so as to select the most commonly sold preparations along with their indications. All samples were analysed on standard Atomic Absorption Spectroscopy for qualitative and quantitative study of toxic heavy metals (Mercury, Iron, Zinc, Lead, Manganese and Arsenic). **Results:** Toxic levels of Mercury were present in seven syrups, i.e., (Kashneeze, Akseer e Pachas, Tankar, Sharbat e folad, Urosinal, Akseer e Jigar and Amrat dhara) while Arsenic was present only in Urosinal. Iron, Zinc, Manganese and Lead were present in permissible limits in all syrups. **Conclusion:** Mercury and Arsenic are present in local Herbal & Ayurvedic liquid preparations far beyond the permissible limits as proposed by the International Regulatory Authorities for health drugs while the rest of metals, i.e., Zinc, Manganese, and Iron are within the therapeutic limits.

Keywords: Herbal bottle preparations, Heavy metal toxicity, Mercury, Arsenic, Atomic absorption spectrophotometer, Quality control

J Ayub Med Coll Abbottabad 2015;27(1):183-6

INTRODUCTION

Commercially prepared herbal drugs are commonly used in various communities of the world. There are different preparations of herbal medicines available namely, Homeopathic, Chinese, and Ayurvedic. Herbal medicines are not only used widely in the developing countries but they are also among European countries.¹ There are many side effects that have been reported after the use of these medicines which make them controversial.^{2,3} In the rural areas of Pakistan, these herbal products are very commonly used because they are easily available at a lower price.⁴

In general, it is thought that herbal preparations are without side effects but this is not proven, as harmful quantities of heavy metals are present in herbal preparations.⁵ Heavy metals, which include Zinc, Cadmium, Lead, Copper, Mercury and Arsenic are extremely toxic even in very minute amounts.⁶ The first case of heavy metal poisoning due to Ayurvedic medicine was reported in United Kingdom in 1978.⁷ There are many factors which leads to the poisoning due to heavy metals. Signs and symptoms of the toxicity of depend upon the metal involved, amount of metal ingested and whether the effect was acute or chronic. How much this toxicity will affect any individual depends the age as younger adults can ingest lead in greater quantities as

compared to children, so they are more prone to develop complications.⁸

This study was done to determine the level of toxic heavy metals in selected syrups of Pakistan used in Hazara division of Khyber Pakhtunkhwa province for treatment of different ailments.

MATERIAL AND METHODS

The study was carried out from February till August 2010. Syrups samples were obtained from all different accessible shops of Abbottabad and Mansehra. A total of 10 different commonly used syrups were taken as shown in table-1.

The samples were labelled and kept under room conditions for qualitative and quantitative analysis. 4ml of each sample was taken in microwave digestion vessels. In every vessel 10ml of nitric acid was transferred and the vessels were gently swirled. The samples were taken out from oven and to each sample 1ml of H₂O₂ was added and kept for 5 minutes. The samples were again placed in microwave oven for digestion at 90 % power for 15 minutes. This was followed by cooling of samples for 5 minutes. 20 ml of concentrated HCl was added to each sample and left for 5 minutes at room temperature. Further digestions of samples were carried out at 30% power for 10 minutes. After digestion the samples were rinsed with deionized water. Deionized water was used to dilute these samples. These were thoroughly mixed and an aliquot of 60ml was transferred to Nalgen bottles for

determination of heavy metals. Atomic Absorption spectrophotometer was used for estimation of Lead, Mercury, Zinc, Manganese, Arsenic and Iron with the adjustment of their wavelength 217 nm, 2.375 nm, 2.138 nm, 2.79.5 nm, 193.7 nm and 248.3 nm respectively and then reading was noted from the computer attached to atomic absorption spectroscopy.⁹

The levels of heavy metals detected were compared to reference ranges for permissible levels (Parekh) as shown in table-2.

RESULTS

Seven of ten samples contained heavy metals in excess of permissible levels; of these Mercury was found in all seven samples, while one sample contained both Mercury and Arsenic beyond permissible levels (Table 3).

In the remaining three samples (Surficol, Taifex and Rooh e Kafour), all the six heavy metals, i.e. Arsenic, Manganese, Lead, Mercury, Iron and Zinc were within permissible limits (Table 3).

Among the toxic heavy metals mercury ranked at the top. In our sample the quantity of mercury ranged from 0–76.16 ppm. The tolerable limit of Mercury in the body is less than 15 ppm. Toxic levels were detected in seven samples, with the highest concentration found in Urosinal (76.16 ppm) followed by Sharbat e folad (49.0 ppm), Kashneeze (27.38 ppm), Amrat Dhara (26.90 ppm), Akseer e Paichas (26.20 ppm), Tankaar (19.37 ppm), Akseer e Jigar (19.0ppm). Arsenic is known as King of poison or poison of Kings. Most common cases of acute toxicity in adults are due to arsenic. The concentration of Arsenic in this study ranged from 3.99–16.73 ppm. The highest concentration of Arsenic was found in Urosinal (16.73 ppm) as compared to the tolerable limit of Arsenic (0.5–15 ppm).

Table-1: List of common syrups sold in local Market and their common indications

Names of Syrups	Common usage (indications)
S1	Headache, cough, temperature
S2	diarrhoea, cramps
S3	Constipation
S4	Physical strength
S5	Cough and flu
S6	Kidney problems
S7	Hepatitis
S8	Typhoid
S9	Vomiting , cholera
S10	Pneumonia, stomach ache,

Table-2: Permissible level (in ppm) of heavy metals in the body

Heavy metals	Value in ppm
Zinc	400
Arsenic	0.5–15
Manganese	12–20
Iron	153–185
Lead	0.1–28
Mercury	Less than 15

Table-3: Concentration of different heavy metals (ppm) in ten sampled syrups including detections beyond permissible levels

Syrups	Zinc (ppm)	Manganese (ppm)	Arsenic (ppm)	Mercury (ppm)	Lead (ppm)	Iron (ppm)
S1	0.47	0.004	3.997	27.388*	7.598	11
S2	0.833	0.034	10.40	26.20*	9.62	13.61
S3	0.084	0.482	4.167	19.377*	18.217	2.867
S4	0.0337	0.351	8.08	49.0*	8.40	0.45
S5	0.004	3.14	8.495	5.80	8.28	0
S6	0.558	6.90	16.73*	76.16*	4.19	0
S7	5.09	0.026	9.58	19.1*	8.83	9.74
S8	0.303	0.236	10.11	0	10.38	5.78
S9	0.154	3.67	5.620	0	9.640	0
S10	7.1	5.56	6.380	26.90*	10.89	2.257

S1=Kashneeze; S2=Akseer e Paichas; S3=Tankaar; S4=Sharbat e Folad; S5=Surficol; S6=Urosinal; S7=Akseer e Jigar; S8=Taifex; S9=Rooh e Kafoor; S10=Amrat Dhara.

*Metals detected beyond permissible levels

DISCUSSION

In the present study, out of the 6 heavy metals tested, only 2 heavy metals are present in the 10 selected syrups i.e. Mercury and Arsenic. Mercury was found in 7 syrups and Arsenic only in Urosinal. Hina B *et al* showed similar results in their study¹⁰, a picture of occurrence of heavy metals in some selected known herbal drugs purchased from different areas of Karachi. The concentration of heavy metals detected in these drugs are in the range of, 65.68–1652.89 µg for Iron 1.6–4.91 for Cadmium,0.65–120.21 for Copper, 3.26–30.46 for Lead 83.74–433.76 for Zinc, 1.61–186.75 for Chromium.

Brain kidneys and developing foetus are at risk of permanent damage due to exposure to high levels of mercury, i.e. toxic level. Following effects such as shyness, tremors, irritability, memory problems and change in hearing or vision are seen with long term exposure.¹¹ diarrhoea, nausea, lung damage and vomiting increase in blood pressure or heart rate, skin irritation and eye rashes are the effects of short term exposure.¹¹ Methyl mercury and Mercuric chloride are determined as possible human carcinogen by EPA.

An exposure to mercury is the second most common cause of toxic metal poisoning. There is serious Public health concern over Mercury exposures, because of contamination of fish with elemental mercury content of dental amalgams and the methyl mercury. It has long been a topic or political and medical debate. Although the toxicology of mercury is complex, there is evidence that antioxidant have protective role in the prevention of renal and neurological damage caused by mercury toxicity.¹²

ATSDR of the U.S department has reported that third most commonly found toxic heavy metals is mercury.¹³The people taking mercury containing

syrups (Kashneze, Akseer e paichas, Tankar, Sharbat e folad, Akseer e jigar, Rooh e kafour and Amrat dhara) should avoid taking wheat flour, mutton, egg white and egg yolk as these nutrients contain exceeding levels of Mercury (Qureshi, *et al* 1982).¹⁴ Vegetables, like potato, peas, tomato, and spinach should be avoided while taking these syrups, as these nutrients also contain exceeding levels of Hg (Qureshi, *et al* 1982).¹⁴

Most important heavy metals involved in environment pollutants are mercury, arsenic and lead. When heavy metals present in water and soil are in minute amounts they can accumulate in food chain and cause serious health problem to humans and can also affect rest of organisms. Their presence in atmosphere, water and soil even in trace amount can cause serious problem to all organisms. Factors which affect the threshold of dietary toxic metal in soil crop system include organic matter content, soil type, minerals and soil PH, along with this, interaction of root-microbes and soil plants play major role in regulating heavy metals movement from soil to the edible parts of crops.¹⁵

Arsenic exists both in organic and inorganic states. Inorganic Arsenic is a known carcinogen and causes cancer of skin, lungs and bladder. There are symptoms of acute exposure of Arsenic can give symptoms with rapid onset of headache, nausea and severe gastrointestinal irritation that accompanied by intense diarrhoea, and vomiting.¹⁶

Amster E *et al*¹⁶ also evaluate the extent of Arsenic contamination, in commercially available kelp by analysing nine samples randomly obtained from local health food stores, eight out of the nine samples showed detectable level of a Arsenic higher for certain food products.

A study conducted in Bangladesh also showed unsafe levels of Arsenic in tube well water.¹⁷ 33% of the tube wells had Arsenic concentrations greater than the WHO drinking water guidelines.

Manganese, Zinc and Iron in our study were in permissible limits in syrups and was consistent with other studies (M Saeed *et al* 2010).¹⁸ Similar results have been shown by Samudralwar *et al* (1996)¹⁹ who observed that heavy metals like Zinc, Manganese, Sodium, Iron and Copper were found in Ayurvedic medicine.

Rai V *et al* (2008)²⁰ also studied Dashmoola roots which are used for manufacturing of Ayurvedic medicines. From India Forty different Dashmoola samples were collected for testing of Chromium, Mercury, Arsenic, Cadmium, Nickel and lead. From this study it was observed that almost all samples contained heavy metals in toxic levels. In South Asia Saper RB *et al* (2004)²¹ concluded the ayurvedic herbal products which were available in grocery

stores, contained harmful level of Arsenic Mercury and Arsenic.

Our data was consistent with studies of Ayurvedic HMPs sold outside the United. In India out of 22 Ayurvedic HMPs purchased, 41% contained Arsenic and 74% contain Mercury and Lead. Lead, Mercury and Arsenic were found 30% Ayurvedic HMPs in England. Heavy metals are also present in traditional medicines which are in use in Mexico, Africa China, Malaysia.²¹

Heavy metals unfortunately found in water, air and food supply. Cases of severe acute Arsenic, Mercury, Lead and Cadmium poisoning is rare, however when there is poisoning non-toxic treatment is essential. Since in 1950 DSMA (dimecaptosuccinic acid) which is rare sulphhydryl-containing water soluble non-toxic chelator. It is orally administered and has been used in antidote to heavy metal toxicity. More recent research showed this compound efficacy and safety. Clinical use has established that the primer metal chelation compound, based on oral dosing and urinary excretion as compare to other chelating substances.²²

CONCLUSION

Most of the syrup preparations contained Mercury beyond permissible limits; its presence in toxic levels could be due to various process/solvents/excipients used in the manufacturing. However, this level can be reduced or eliminated strictly by following the guidelines of current Good Manufacturing Practice (cGMP) or other quality control parameters for specified herbal products. Public awareness campaign would help to decrease the usage of these syrups till such time that they are certified as safe products.

Conflict of Interest: None

ACKNOWLEDGEMENTS

The help of Fida Hussain, Uzma Faryal, Javeria Saqib from the Department of Biochemistry, Quaid-i-Azam University Islamabad for laboratory procedures is gratefully acknowledged. For help in sample collection and processing, we acknowledge Tahira Durrani, Department of Biochemistry at Women Medical College Abbottabad.

REFERENCES

1. Silano M, De Vincenzi M, De Vincenzi A, Silano V. The new European legislation on traditional herbal medicines: main features and perspectives. *Fitoterapia* 2004;75(2):107-16
2. Saxe TG. Toxicity of medicinal herbal preparations. *Am Fam Phys* 1987;5:135-42.
3. Shamala G. Concern over safety of herbal remedies. *The Sun* 2001: p.15.

4. Ahmad SS, Hussain SZ. Ethno medicinal survey of plants from salt range (Kallar Kahar) of Pakistan. *Pak J Bot* 2008;40(3):1005-11.
5. Chan K. Some aspects of toxic contaminants in herbal medicines. *Chemosphere* 2003;52:1361.
6. Tonguc O. Determination of Heavy metals in some moss species around thermic power stations. *Turk J Biol* 1998;28:171-80.
7. Brearley RL, Forsythe AM. Lead poisoning from Aphrodisiacs: potential hazard in immigrants. *Br Med J* 1978;23-30;2(6154):1748-9.
8. Samara S. Heavy metal toxicity. (Internet article). Updated May 6, 2011. Cited on: October 13, 2011. Available from: <http://emedicine.medscape.com/article/814960-overview>
9. Kalra YP, Maynard DG, Radford FG. Microwave digestion of tree foliage for multi-media analysis. *Can J Res* 1989;19:981-5.
10. Hina B, Rizwani GH, Naseem S. Determination of toxic metals in some herbal drugs through drugs through atomic absorption spectroscopy. *Pak J Pharm Sci* 2011;24(3):353-8.
11. Martin S, Griswold W. Human Health Effects of Heavy Metals. Manhattan: Center for Hazardous Substance Research. 2009 Contract No.: 15.
12. Patrick L. Mercury Toxicity and Antioxidants: part 1: Role of Glutathione and Alpha Lipoic Acid in the Treatment of Mercury Toxicity. *Altern Med Rev* 2002;7(6):456-71.
13. Ozuah PO. Mercury poisoning. *Curr Probl Pediatr* 2000;30:91-9.
14. Qureshi IH, Zaidi JH, Fatima I, Waheed S. Trace elements nutritional status of some Pakistan food items. In: Rahman MA (Ed.) *Biochem for development: Proceeding of 6th federation of Asian and Oceanian Biochemistry Symposium No. 171, Karachi, Pakistan 1987;2-4 Nov. Lahore: Pakistan Society of Biochemists: 1988. pp. 201-12.*
15. Islam E, Yang X, He Z, Mahmood Q. Assessing potential dietary toxicity of heavy metals in selected vegetables and food crops. *J Zhejiang Univ Sci B* 2007;8(1):1-13.
16. Amster E, Tiwary A, Schenker M.B. Case report: potential Arsenic toxicosis secondary to herbal kelp supplement. *Environ Health Perspect* 2007;115(4):606-8.
17. Frisbie SH, Mitchell EJ, Mastera LJ, Maynard DM, Yusuf AZ, Siddiq MY *et al.* Public health strategies for western Bangladesh that address Arsenic, Manganese, Uranium, and other toxic elements in drinking water. *Environ Health Perspect* 2009;117:410-6.
18. Saeed M, Muhammad N, Khan H, Khan SA. Analysis of toxic heavy metals in branded Pakistani herbal products. *J Chem Soc Pak* 2010;32(4):471-5.
19. Samandralwar DL, Garg AN. Minor and trace elemental determination in the Indian herbal and other medicinal preparations. *Biol Trace Ele Res* 1996;54(2):113-21.
20. Rai V, Kakkar P, Singh J, Misra C, Kumar S, Mehrotra S. Toxic metals and organochlorine pesticides residue in single herbal drugs used in important ayurvedic formulations. *Environ Monit Assess* 2007;143(1-3):273-7. .
21. Saper RB, Kales SN, Paquin J, Burns MJ, Eisenberg DM, Davis RB *et al.* Heavy metal content of ayurvedic herbal medicine products. *JAMA* 2004;292(23):2868-73.
22. Miller AL. Dimercaptosuccinic (DMSA), a non-toxic, water soluble treatment for heavy metal toxicity. *Altern Med Rev* 1998;3(3):200-7.

Address for Correspondence:

Bibi Hajra, Department of Biochemistry, Women Medical College, Murree Road, Nawanshehr, Abbottabad 2208-Pakistan.

Cell: +92 313 361 4617

Email: hajra.khan1982@gmail.com