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SHORT COMMUNICATION

Measurement of concentrations of six metals in Indian traditional medicine preparations and Sindoor powder

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Abstract

Background: Ayurveda is one of the traditional medical practices that is originated from India where it is still widely practiced. This study is an attempt to determine the concentration of 6 selected metals, namely chromium, cobalt, nickel, arsenic, mercury, and lead in 19 samples of Ayurvedic herbal medicines and 7 Sindoor powders sent by physicians for analysis.
Methods: In this study, ICP-MS as direct analysis of a 1 in 100 dilution of the tested materials was employed which gives an estimate of the solubility of the metal constituents of the materials tested in 0.5% nitric acid.
Results: The highest individual metal values found per gram in the tested materials were: chromium 3.2 microgram/g, cobalt 3.1microgram/g, arsenic 2811 microgram/g, mercury 1320 microgram/g, and lead 8329 microgram/g. Assuming only a 1 g intake/day of any single material tested, lead content exceeded in 10/26 (38%) of the preparations above the ANSI 173 oral permitted daily limit (PDE). Likewise, mercury and arsenic contents exceeded the oral PDE in 6/26 (23%). Some of these folk medicines had high levels of more than one element in it. The lead content in 3 of the 7 Sindoor powders surpassed the guideline. However, the nickel content did not exceed the PDE in the 19 samples tested.
Conclusions: Our data shows that, many of Ayurvedic medicine preparations tested still contain toxic amounts of arsenic, mercury, and lead. Sindoor powder which is traditionally and religiously used by many Indian women at their forehead also contains heavy metals like lead. All these materials can pose serious health risks to their users

Keywords: Herbal medicine, Heavy metal poisoning, Lead poisoning

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INTRODUCTION

The composition of Indian traditional medicines can be from vegetable, animal, and mineral products (1, 2). In Ayurveda, which is one of the major forms of Indian traditional medicines, the contents are classified into two major types: herbal-only and Rasa Shastra preparations. Rasa Shastra is an ancient process of adding metals, minerals or gems to herbal preparations. The added metals are usually lead, mercury, and arsenic (3, 4). It is essential to note that about 35-40 % of all Ayurveda medicines involve some amount of metal (1, 2). Many studies have already revealed that ayurvedic products result in serious health risks due to their metal content (5, 7).

Sindoor is an essential cosmetic item for married women In India, who religiously and traditionally apply it on the forehead and at the parting of their hairs. Nowadays, Sindoor is made of chemical dyes, synthetic materials, and lead salts. Some of the manufacturers add crude red lead (Lead (II, IV) oxide) as well. The red colour may also be derived from mercuric sulphide (HgS). When traces of these powders are ingested or inhaled, they can produce toxic symptoms. It is not known how much of these compounds can be absorbed through the skin. (8).

In this study, we have measured the soluble metal content levels of arsenic, cobalt, lead, chromium, nickel, and mercury in Ayurvedic medicines and Sindoor powders sent by our clinicians.

METHODS

***Study Details*:** Nineteen Ayurvedic medicines and seven Sindoor powder samples were used in the present study. These medicines were collected from patients and sent for metal analysis by clinicians. They were gathered over a period of 42 months, from June 2012 to November 2015. The clinical inquiry was whether the metal content of these materials could be responsible for the patients' symptoms. These medicines were presented in capsule or "ball" forms. No reliable or complete clinical information was provided about these preparations.

Methods: Metal analysis was performed using an inductively coupled plasma mass spectrometer (ICP-MS0, X Series 2) (Thermo Fisher Scientific, USA). A direct analysis technique was used, namely 10 mg of each compound from the surface of medicine balls, or the contents of individual capsules, was

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tested. Sindoor powders were weighted directly from the sample. These samples were mixed with 10 ml of 0.5% nitric acid (Fisher Scientific, Trace Metal grade, 69% Nitric acid). This mixture was vortexed for 30 seconds and then left for 24 hours at room temperature to dissolve, after which it was centrifuged at 1800g for 5 minutes. Then 1.0 ml of the supernatant or 0.5ml of internal quality control was diluted to 10 ml with 0.5% nitric acid and then aspirated into the analyser, where it was measured in triplicate, and the average value was reported (when the % relative standard deviation of the triplicates was <10%).

The standards used were Certipur reference material (ICP Multi Element Standard solution XXI, ref 2.74473.0100), and Certipur reference material (Mercury ICP Standard, ref. 1.08623.0100) (both from EMD Chemicals Inc, Phil. USA).

RESULTS

Daily variations of the metal concentrations were estimated using BioRad Lyphochek Quantitative Urine Control (normal) (Ref. 376 Bio-Rad Laboratories India Pvt. Ltd., California, and USA). Concentration values obtained and expressed as mean ± SD from 33 to 39 runs: chromium 2.5 ±1.51; cobalt 0.44 ±0.21; nickel 4.57 ±1.61; arsenic 42.36 ±5.35; mercury 3.33 ± 2.17; lead 42.31 ±5.11 µg/L.

The estimated metal content of the preparations for metals, is shown in Figure 1. The results were expressed as µg/g of the preparation. The lowest reported value was < 0.1µg/g, and the highest values for different metals were as follows: chromium 3.2 µg/g; cobalt 3.1µg/g; arsenic 2811 µg/g; mercury 1320 µg/g; lead 8329 µg/g. These concentrations can be compared to the ANSI 173 guidelines in table 1. The limits mentioned in ANSI 173 guideline is the reference provided for the daily intake limits, when heavy metals are consumed orally. There are no regulatory limits for cobalt and one estimation for permitted daily exposure (PDE) of chromium is 2 mg/day (9). Assuming only a 1 g intake/day of any single preparation tested, then 10/26 (38%) of the preparations exceeded the PDE limit for lead and 6/26 (23%) exceeded the daily limit for mercury and arsenic. The median (Interquartile range) concentrations of different metals in the herbal medicine samples analysed were as follows: lead; 6.15µg/g (0.93-21.2), mercury; 9µg/g (0.55-81.7), arsenic; 1.9µg/g (0.25-43.8), chromium; 0.87µg/g (0.43-1.3), and cobalt; 0.4µg/g (0.25-1.2). Some preparations had high levels

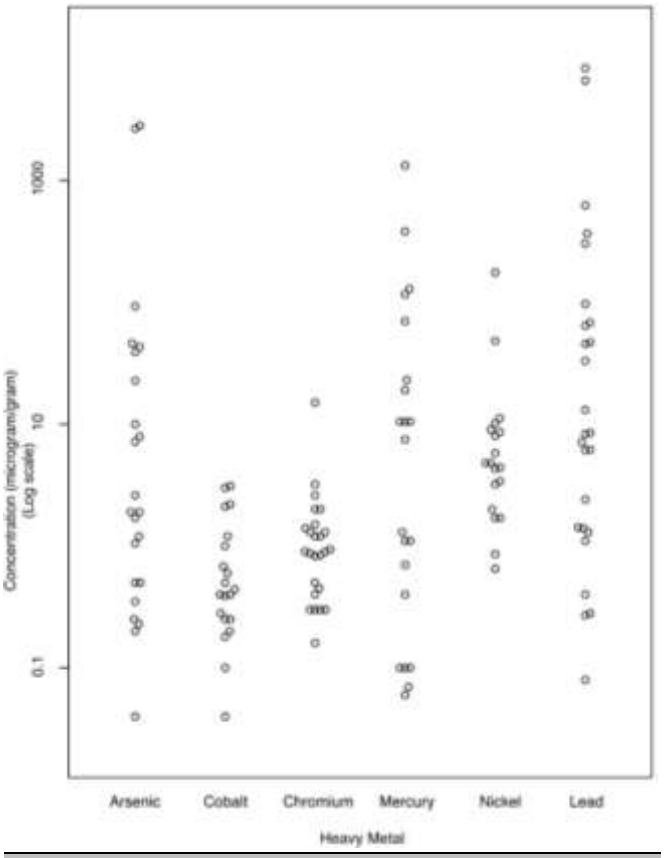

Figure 1. Log scale concentration of 6 metal components in µg/g of 26 compounds received in the laboratory.

of more than one element. The nickel content of 19 of these preparations was also determined. It ranged from 0.65 to 175.6 with median of 4.6 µg/g. The PDE limit for nickel, as suggested by Food Standards Agency (FSA) is, 250 µg/day (10). The nickel content did not exceed this PDE value in any of the tested samples.

Moreover, seven Sindoor preparations were included in the 26 mixtures analysed. The highest levels of each metal in the Sindoor preparations tested were as follows: chromium 3.2 µg/g; cobalt 3.0 µg/g; nickel 10.20 µg/g; arsenic 9.9 µg/g; mercury 0.40 µg/g, and lead 624 µg/g. Three of the 7 preparations exceeded the guideline value for the lead.

Table 1. Guideline values for limits of arsenic and other toxic metals in herbal preparations

Guideline	Arsenic	Mercury	Lead	Units
ANSI 173 – ref (4)	10	20	20	µg/day
JECFA-ref (4)	150	50	250	µg/day
WHO 2007 –ref (10)	<5	<10	<10	mg/kg

Key: Guideline values used by the American National Standards Institute (ANSI)/National Sanitation Foundation (NSF) International Dietary Supplement Standard 173 (ANSI 173), listed in ref 4.
The Joint FAO/WHO Expert Committee on Food Additives (JECFA) is an international expert scientific committee that is administered jointly by the Food and Agriculture Organization of the United Nations (FAO) and the World Health Organization (WHO). They give acceptable daily intakes for a 70-kg adult, also quoted in ref 4.
WHO guidelines for medicinal plants, quotes Malaysian guidelines for finished herbal products, ref 10.

DISCUSSION

Guideline values for metal content of herbal medicines were quite inconsistent in their established limits for each toxic metal (refer to table 1) (11). Like other groups, we also found that lead poisoning is the most common poisoning associated with ayurvedic medications (4, 5). Other heavy metal poisoning reported with these traditional medicines including Ayurveda are chronic arsenic poisoning and mercury poisoning. (3, 4). We can compare relative values between the medicines in the study meant for ingestion and the Sindoor powders meant only for scalp application. We found that Ayuverdic medicines had up to 13x higher values of lead than Sindoor powders, 36X higher values of mercury, and 355X higher values of arsenic.

Contamination of herbal materials with toxic substances, such as lead or arsenic, can be attributed to other causes than voluntary adding, which include environmental pollution, the conditions in which the plants are dried, processed, and stored. Gasser et al. (2009) showed that mercury and arsenic levels measured in the medicines when compared to the values from the original dried herbs suggest that additional amounts of these metals were deliberately added during manufacture (11). Though Ayurveda claims a unique method of detoxification and calcination of metals, called Shodhan" and "Maran" (1, 2), heavy metal toxicities are reported with many of these medicines (3, 4, 5, 12, 13). Despite a wealth of literature demonstrating the high metal content of folk medicines in India, China, Tibet, Africa etc., our data shows that many of the preparations (up to 40% for lead and 24% for mercury and arsenic) still contain heavy metals much above the recommended PDE. Ingestion of these materials may produce heavy metal toxicity from these metals, while ingestion of the Sindoor powder has the risk of lead poisoning. Lead and other metals from contaminated Sindoor powders can also enter into human's body by respiratory route or via the skin, which can also lead to toxicity (14).

LIMITATIONS

For analysis, we used a direct analysis, infusion technique, rather than a digestion procedure which would have given the total recoverable analyte content. This simple procedure aimed at giving an estimate of the readily soluble metal constituents of the material tested. It will give a lower estimate of the metal content of the materials analysed. Also, scraping the surface of "ball" tablets may not be representative of the interior. Often our patients brought multiple capsules or tablets (maximum 8). We have only assumed a minimum intake of 1g/per day of any given ball or capsule, even though most of the balls/ capsules were > 1 g in weight, again giving a lower estimate for the amount ingested by the patient.

Furthermore, since this was a retrospective study, clinical information regarding the patients was insufficient to correlate the metal content of the medicines and the symptoms of the patient.

CONCLUSION

Our data shows that, despite a wealth of literature demonstrating the high metal content of folk medicines, many Ayurvedic medicine preparations tested still contain toxic amounts of arsenic, mercury, and lead. Users of these medicines may be at risk of these heavy metal toxicities. Traditionally and religiously used Sindoor powder by Indian women for the local forehead application also contain high levels of lead. Thus, proper analysis of both Ayurvedic and other traditional medicines along with Sindoor powders is a necessity from the perspective of the general health of the population.

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REFERENCES

1. Sen S, Chakraborty R. Revival, modernization and integration of Indian traditional herbal medicine in clinical practice: Importance, challenges and future. J Tradit Complement Med. 2017 Apr;7(2):234–44.
2. Chopra A, Doiphode VV. Ayurvedic medicine. Core concept, therapeutic principles, and current relevance. Med Clin North Am. 2002 Jan;86(1):75–89
3. Abdalla AA, Smith RE. Determination of Mercury in Ayurvedic Dietary Supplements That Are Not Rasa Shastra Using the Hydra-C Direct Mercury Analyzer. Int J Anal Chem. 2013;2013:1–4.
4. Saper RB, Phillips RS, Sehgal A, Khouri N, Davis RB, Paquin J, et al. Lead, Mercury, and Arsenic in US- and Indian-Manufactured Ayurvedic Medicines Sold via the Internet. JAMA. 2008;300(8):915–23.
5. McElvaine MD, Harder EM, Johnson L, Baer RD, Satzger RD. Lead poisoning from the use of Indian folk medicines. JAMA. 1990 Nov 7;264(17):2212–3.
6. Lynch E, Braithwaite R. A review of the clinical and toxicological aspects of "traditional" (herbal) medicines adulterated with heavy metals. Expert Opin Drug Saf. 2005 Jul;4(4):769–78.
7. Saper RB, Kales SN, Paquin J, Burns MJ, Eisenberg DM, Davis RB, et al. Heavy metal content of ayurvedic herbal medicine products. JAMA. 2004 Dec 15;292(23):2868–73.
8. Samariya S, Dwivedi S, Panigrahi D, Joshi H. Formulation and Evaluation of Herbal Sindoor Using Different Natural/Herbal Ingredients. International J Pharm Teach Pract. 2013 Sep 1;4: 752–4.
9. World Health Organization (WHO) guidelines for assessing quality of herbal medicines with reference to contaminants and residues. Geneva: World Health Organization; [Internet]. 2007 [cited 2021 Apr 29]. Available from: https://apps.who.int/iris/bitstream/handle/10665/43510/9789241594448_eng.pdf?sequence=1&isAllowed=y
10. Guideline on the specification limits for residues of metal catalysts or metal reagents. European Medicines Agency. Committee for medicinal products for human use (CHMP) [Internet] London, 21 February 2008. [cited 2021 Apr 29]. Available from: https://www.ema.europa.eu/en/documents/scientific-guideline/guideline-specification-limits-residues-metal-catalysts-metal-reagents_en.pdf
11. Gasser U, Klier B, Kuhn AV, Steinhoff B. Current findings on

the heavy metal content in herbal drugs. Pharmeuropa Scientific Notes 2009-1: 37-50

12. Afshari R, Bellinger DC. Chemical Risk without Concurrent External Exposure! Asia Pac J Med Toxicol. 2019 Jun 1;8(2):30–3.
13. Jafri L, Ullah Khan N, Baig MA, Ali N, Majid H, Habib Khan A. The Clinical Spectrum of Plumbism; an Experience from a Tertiary Care Hospital in Karachi, Pakistan. Asia Pac J Med Toxicol. 2017 Dec 1;6(4):118–22.
14. Lin CG, Schaider LA, Brabender DJ, Woolf AD. Pediatric lead exposure from imported Indian spices and cultural powders. Pediatrics. 2010;125(4):e828–e835.