J. Med. Toxicol. (2013) 9:21–24
DOI 10.1007/s13181-012-0231-5

TOXICOLOGY INVESTIGATION

Heavy Metal Contaminants in Yerberia Shop Products

Michael Levine · Jason Mihalic · Anne-Michelle Ruha · Robert N. E. French · Daniel E. Brooks

Published online: 5 May 2012

Abstract Complementary and alternative medications, including the use of herbal medications, have become quite popular in the USA. Yerberias are found throughout the southwest and specialize in selling Hispanic herbal products. The products sold in these stores are not regulated by any governmental agency. Previous reports have found Ayurvedic medications contain high levels of lead, mercury, and arsenic. The primary purpose of this study is to examine the prevalence of heavy metal contaminants sold at Yerberia stores in the southwest. Yerberias in the Phoenix, Arizona area were identified via search of an on-line search engine using the words "Yerberia Phoenix." Every second store was selected, and products were purchased using a standard script. The products were subsequently analyzed for mercury, lead, and arsenic. The main outcome is the prevalence of heavy metal content in over-the-counter "cold" medications purchased at a Yerberia. Twenty-two samples were purchased. One product contained pure camphor (2-camphone) and was subsequently not further analyzed. Of the 21 samples analyzed, lead was found in 4/21 (19.4 %). Arsenic and mercury were in 1/21 (4.8 %) each. Because two samples contained two heavy metals, the total prevalence of heavy metals was 4/21 (19.4). Heavy metal contaminants are commonly encountered in over-the-counter herbal "cold" medications purchased at Yerberias in the southwest.

Keywords Heavy metal · Yerberia · Contaminant · Toxicity · Herbal

M. Levine · A.-M. Ruha · R. N. E. French · D. E. Brooks
Department of Medical Toxicology, Banner Good Samaritan Medical Center,
Phoenix, AZ, USA

M. Levine
Section of Medical Toxicology, Department of Emergency Medicine, University of Southern California,
Los Angeles, CA, USA

J. Mihalic
Office of Public Health Chemistry,
Arizona Department of Health Services,
Phoenix, AZ, USA

M. Levine (✉)
Department of Emergency Medicine,
University of Southern California,
1200 North State Street #1101,
Los Angeles, CA 90033, USA
e-mail: Michael.Levine@bannerhealth.com

Introduction

Complementary and alternative medication (CAM) is a heterogeneous term used to describe various practices used to treat or prevent disease [1]. The prevalence of CAM in the USA is high, with nearly 40 % of adults, and 11 % of children having used CAM within the previous year [1]. The consumption of natural and herbal products is one of the most commonly encountered forms of CAM [1].

Yerberias are stores found throughout the southwestern United States that specialize in the sale of Hispanic herbal remedies. These stores sell non-prescription products. Thus, these products are not regulated by the Food and Drug Administration (FDA) as medications, but rather as dietary supplements. While dietary supplements are somewhat regulated by the Center for Food Safety and Nutrition, the regulation standards are typically much less than that required by the FDA for prescription medications. As a general rule, the products sold in these stores originate from a wide variety of countries, including Mexico, and those in Central and South America. Due to their lack of stringent regulation, such products may contain impurities. Previous studies have found elevated concentrations of lead, mercury, and arsenic among unregulated medicinal products purchased in the USA. The majority of these studies have involved Ayurvedic medications, which originate from South East Asia and India [2–5]. The prevalence of heavy metal-contaminated products sold at Yerberias, however, has not been previously examined. The primary purpose of this study is to examine the prevalence of lead, mercury, and arsenic in over-the-counter cold medications purchased at Yerberias.

Methods

Collection of Products

Using an on-line search engine, a list of Yerberias was compiled by searching the words "Yerberia Phoenix." Every other store on the list in Phoenix, Arizona was selected. The final study population comprised of 11 stores. An investigator entered each of these selected Yerberias, and using a standardized scripted sentence and asked for a "medicine for a cold." Two internationally manufactured products were purchased at each store. The products were selected on the advice of the Yerberia employee. After purchase, the products were de-identified and analyzed by the Arizona Department of Health Services laboratory.

Each product was assigned a unique study identification number, inventoried, weighed, and analyzed for the presence of mercury, lead, and arsenic. The analysis required product digestion, followed by analysis. Lead and arsenic contents were identified via graphite furnace atomic absorption. Samples that were determined to be of a low organic matrix were analyzed for mercury via atomic absorption spectrophotometry. Samples that were determined to be of a high organic matrix, and hence an explosion risk, were analyzed for mercury via inductively coupled plasma mass spectroscopy (ICP-MS).

Detection Limits

The limit of quantization (LOQ) for the high organic matrix mercury samples is 0.033 mg/kg. The LOQ for the low organic matrix mercury samples is 0.05 mg/kg. Lead and arsenic were each able to be detected at 0.5 mg/kg sample.

Consent

Because the study did not involve human subjects, it was exempt from needing approval from the institutional review board.

Lead and Arsenic Analysis

The samples were digested using a modified hot block procedure of nitric acid and hydrogen peroxide based on Environmental Protection Agency (EPA) method 3050B, using a SCP Science DigiPrep digestion unit which has been previously described [6]. Samples were weighed dry to 0.50±0.01 g with a final post-digestion volume of 50 mL. Samples were digested in duplicate with matrix spike quality control as well as method blanks, fortified blanks, and a lead/arsenic control.

During digestion, a 0.50-g aliquot of the sample was placed into a 50-mL digestion tube, and 10 mL of 1:1 nitric acid was added. The digestion block was heated to 95±5°C, and the samples were heated for 15 min before being removed and allowed to cool. An addition of 2.5 mL concentrated nitric acid was added followed by 30 min of heating with subsequent cooling. Once cool, 1 mL of deionized water and 1.5 mL 30 % hydrogen peroxide were added to the sample, which was then heated for 15 min before being allowed to cool. A second 1-mL aliquot of 30 % hydrogen peroxide was added, and the sample was heated for 15 min, then cooled. This hydrogen peroxide process was repeated as needed per sample matrix. The sample volume was then reduced to 5 mL, cooled, and filtered using Whatman filters, before being brought to a final volume of 50 mL using deionized water.

Samples and quality controls were analyzed for both lead and arsenic using a Perkin-Elmer AAnalyst 600 graphite furnace atomic absorbance spectrometer and EPA method 200.9, Revision 2.2 [7]. Analysis was accomplished separately using a single element (lead or arsenic) electrodeless discharge lamp (EDL). The digested sample and method controls were individually placed onto an instrument platform prior to being introduced into the furnace chamber. Once in the chamber, the sample was purged with a gas mix of 95/5 (argon/hydrogen) to dryness. The sample was then charred to an ash-like consistency and cooled. Atomization occurred in a stopped flow atmospheric environment of 95/5 (argon/hydrogen) gas by the rapid heating of the furnace to a temperature that atomized the analyte (arsenic or lead) from the pyrolytic graphite surface of the instrument platform into an atomized cloud. The cloud then absorbed element-specific atomic emission produced by the EDL for both arsenic and lead. This signal was subsequently measured and corrected for background interferences. The corrected absorbance is directly related to the concentration of the analyte and was plotted against a calibration curve to determine both the arsenic and lead concentrations of each sample.

Mercury Analysis: Low Organic Matrix

The samples were analyzed using a Milestone DMA-80 Direct Mercury Analyzer based on EPA method 7473 [8].

An aliquot of the sample was weighed and introduced into the DMA-80 analyzer. The sample was subsequently dried and underwent thermal and chemical decomposition under a continuous flow of oxygen. The decomposed products were carried via the oxygen flow to a catalyst bed, where oxidation was completed. Using this method, different species of mercury are converted to elemental mercury vapor, which is trapped onto a gold amalgamator. After flushing the system with oxygen to remove any decomposition by-products, the amalgamator was rapidly heated to release the mercury vapor. This vapor was carried through the absorbance cells, positioned in the light path of a single wavelength atomic absorption spectrophotometer. A photosensitive detector measured the amount of light absorbed by the sample vapor at a wavelength of 253.7 nm. The sample absorbance values were compared to the absorbance values from prepared calibration standards to calculate the mercury content of the samples.

Mercury Analysis: High Organic Matrix

Due to a potential explosion hazard, the above method was deemed too unsafe to be performed on samples with a high organic matrix. As a result, samples with a high organic matrix were analyzed using a different technique. The samples were diluted in a 1:10 ratio. One gram of sample was then placed in a 15-mL tube, and deionized water was added to a final weight of 10 g. The samples were analyzed using a Perkin-Elmer Elan DRCe ICP-MS, using standard analytic methods.

Results

Twenty-two samples were purchased and submitted to the state health department's laboratory for analysis. Evaluation of one sample revealed camphor (2-camphanone), and thus was not analyzed as heating the sample was felt to be too high of an explosion risk. As such, a total of 21 samples were subsequently analyzed for mercury, lead, and arsenic. These products included a combination of pills, liquids, and two topical ointments.

Of the 21 samples analyzed, 4 (19.4 %) contained lead. The maximal concentration of any samples was 1.6 mg/kg. Among the lead-containing samples, the mean (± standard deviation) lead content was 1.19 (±0.46) mg/kg. Mercury was detected in 1 of the 21 (4.8 %) samples at a concentration of 0.059 mg/kg. Arsenic was detected in 1 of the 21 (4.8 %) samples at a concentration of 0.54 mg/kg. Because two samples contained two different heavy metals, the overall prevalence of heavy metal contamination was 4/21 (19.4 %).

Discussion

This study examined the prevalence of heavy metal contaminations in over-the-counter "cold" medications purchased at local Yerberias in Phoenix, Arizona. The use of complementary and alternative medications to treat acute presentations of common medical conditions is quite frequent among both adult and pediatric patients [9–11]. Furthermore, use of nonprescription medications in doses exceeding the manufacturer's recommendations is not uncommon [12]. The pattern of use of medications purchased at Yerberias is unknown. Therefore, this study strictly examined the prevalence of heavy metal contamination. While some products gave specific dosing recommendations, others had no dosing recommendations, and some included vague statements, such as use “a small amount.” Because it is not known how patients will use the medications, it is impossible to determine if the amount of heavy metals contained in these products would be potentially toxic. As a result, the authors are not able to make conclusions as to the potential health hazards of such contaminants. Nonetheless, these medications represent a potential source of exposure to toxic metals, and toxicity has previously been reported from various Ayurvedic medications [13–15].

Previous studies have examined the content of heavy metals in other cultural medicines. Saper and colleagues reported the prevalence of lead, mercury, and arsenic from Indian-manufactured Ayurvedic medications sold over the internet in the USA [3]. Ayurvedic medications include herbal medications and *rash shastra*, the latter of which is a product containing herbal medications and metals, minerals, or gems [3]. Their study found a 20.7 % prevalence of heavy metals in Ayurvedic medications, with lead comprising the majority of the heavy metal contaminants. Thus, their results are similar to those observed in the current study. Importantly, however, some Ayurvedic medications are intended to contain heavy metals (e.g. *rash shastra*). In contrast, none of the products purchased at the Yerberias were expected to contain heavy metals.

Saper and colleagues also examined heavy metal contaminants among Ayurvedic herbal medications from South East Asia purchased in the metropolitan Boston region [2]. Again, the overall prevalence of heavy metal contamination in their study (20 %) was similar to the current study. However, in Saper's study, the overall concentration of heavy metals was much greater than observed in this study.

The United States General Accountability Office conducted a study of 40 herbal dietary supplements and found heavy metal contaminants including lead, cadmium, and arsenic in nearly all samples, although similar to this study, the concentrations were not very elevated [16].

The Food and Agricultural Organization/World Health Organization Joint Expert Committee on Food Additives has determined the maximal acceptable content of various heavy metals in foods. While the acceptable content varies depending on the specific foods, the maximal concentration of lead, arsenic, and mercury are 0.01–1.5, 0.01–0.5, and 0.001–0.1 mg/kg, respectively [17]. Thus, the amount of heavy metal contaminants in the samples in this study is present, but not markedly elevated. The true toxicity risk, therefore, would depend on how much of the product is used.

The study has several limitations. Because not every product available in the store was tested, the products examined may have introduced a selection bias. It is possible that certain products with higher concentrations of heavy metals are not displayed on the shelf, and as such, the purchased products may have underestimated the prevalence of contaminants. Furthermore, as previously stated, this study was not designed to determine if the amount of contaminants would represent an actual source of toxicity. Specifically, if a product is taken for a short time span (e.g., for the duration of a viral illness), it is probable that a patient would not ingest enough of the product to result in substantial harm. In addition, this study only examined the prevalence of lead, mercury, and arsenic. Other toxic heavy metals could have potentially been present, but simply not tested. However, based on previous studies, this study examined only lead, mercury, and arsenic to permit a direct comparison with other studies in the literature. Lastly, the testing simply examined the prevalence of heavy metal contamination. Many metals, including arsenic, can exist in several different states, each with different degrees of toxicity. This study did not seek to identify the valence of identified arsenic. Similarly, mercury was identified in one sample, but this study did not identify whether the mercury was present in the elemental, inorganic, or organic form.

Conclusion

Lead, mercury, and arsenic are common contaminants in over-the-counter products purchased at Yerberias. The prevalence of contamination found in this study (19.4 %) is similar to that reported with studies of Ayurvedic products.

Conflict of Interest There are no financial, litigational, or other conflicts of interest involved in the preparation of this manuscript.

References

1. Barnes PM, Bloom B, Nahin R (2008) Complementary and alternative medicine use among adults and children: United States, 2007. National Health Statistics reports; no 12. Hyattsville, MD. National Center for Health Statistics
2. Saper RB, Kales SN, Paquin J et al (2004) Heavy metal content of ayurvedic herbal medicine products. JAMA 292:2868–2873
3. Saper RB, Phillips RS, Sehgal A et al (2008) Lead, mercury, and arsenic in US- and Indian manufactured ayurvedic medicines sold via the internet. JAMA 300:915–923
4. Garvey GJ, Hahn G, Lee RV et al (2001) Heavy metal hazards of Asian traditional remedies. Int J Environ Health Res 11:63–71
5. Ernst E (2002) Heavy metals in traditional Indian remedies. Eur J Clin Pharmacol 57:891–896
6. Method 3050 B. Acid digestion of sediments, sludges, and soils. Environmental Protection Agency. http://www.epa.gov/wastes/hazard/testmethods/sw846/pdfs/3050b.pdf. Accessed 23 Oct 2011
7. Method 200.9, Revision 2.2 (1994) Determination of trace elements by stabilized temperature graphite furnace atomic absorption. Environmental Protection Agency. http://water.epa.gov/scitech/methods/cwa/bioindicators/upload/2007_07_10_methods_method_200_9.pdf. Accessed 23 Oct 2011
8. Method 7473 (2007) Mercury in solids and solutions by thermal decomposition, amalgamation, and atomic absorption spectrophotometry. http://www.epa.gov/sam/pdfs/EPA-7473.pdf. Accessed 23 Oct 2011
9. Ottolini MC, Hamburger EK, Loprieato JO et al (2001) Complementary and alternative medicine use among children in the Washington, DC area. Ambul Pediatr 1:122–125
10. Sidora-Arcoleo K, Yoos HL, Kitzman H et al (2008) Don't ask, don't tell: parental nondisclosure of complementary and alternative medicine and over-the-counter medication use in children's asthma management. J Pediatr Health Care 22:221–229
11. Yu SM, Ghandour RM, Huang ZJ (2004) Herbal supplement use among US women, 2000. J Am Med Wom Assoc 59:17–24
12. Heard K, Sloss D, Weber S et al (2006) Overuse of over-the-counter analgesics by emergency department patients. Ann Emerg Med 48:315–318
13. Khandpur S, Malhotra AK, Bhatia V et al (2008) Chronic arsenic toxicity from Ayurvedic medicines. Int J Dermatol 47:618–621
14. Dunbabin DW, Tallis GA, Popplewell PY et al (1992) Lead poisoning from Indian herbal medicine (Ayurveda). Med J Aust 157:835–836
15. Moore C, Adler R (2000) Herbal vitamins: lead toxicity and developmental delay. Pediatrics 106:600–602
16. Herbal dietary supplements: examples of deceptive or questionable marketing practices and potentially dangerous advice (2010) United States Government Accountability Office. Report GAO-10-662 T. http://www.gao.gov/assets/130/124774.pdf. Accessed 23 Mar 2012.
17. Codex general standard for contaminants and toxins in foods (1994). http://www.codexalimentarius.net/download/standards/17/CXS_193e.pdf. Accessed 13 Nov 2011.