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Analysis of Lead in Spices Obtained from Bulk Food Stores

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Abstract

Over the past decade, spices have been frequently recalled due to high levels of lead. Infants, young children, and the developing fetus can be adversely affected by chronic exposure to amounts of lead that may not result in noticeable symptoms of lead poisoning. The United States Food and Drug Administration (USFDA) has called attention to contaminants in spices in a recently published white paper. The USFDA concludes that a primary source of lead contamination in spices appears to be from the use of tainted irrigation water in the countries in which the spices originate. At present, the USFDA does not have a specific limit on the acceptable amount of lead in spices, but does set a limit of 100 parts per billion (ppb) in candy and the United States Environmental Protection agency (USEPA) states that lead levels of 15 ppb in drinking water require treatment. In this work, we analyze lead levels using inductively coupled plasma-mass spectrometry (ICP-MS) of spices obtained from bulk food stores in Northwestern Pennsylvania. Based on the results from similar studies of spices from bulk food stores in other regions of the United States, there is a strong likelihood that spices obtained from bulk food stores in Northwestern Pennsylvania may have levels higher than those deemed acceptable by the USFDA and USEPA.

Keywords: lead contamination, spices, inductively coupled-mass spectrometry

1. Introduction

Lead in consumables and drinking water has been regulated since the lead poisoning prevention policies were enacted in the early 1970s;[1] however, spices have not received as much attention from regulatory agencies even with evidence showing elevated lead levels in spices.[2] Along with headlines about drinking water contamination in municipalities such as Flint, Michigan, there have been several reports of lead contaminated spices.[3-5] Spices have been referred to as the last frontier in food safety.[6] As an example, a recent study found lead contamination in spices, herbal remedies, and ceremonial powders in the homes of children with elevated blood lead levels.[7]

In 2017, the USFDA produced a white paper calling attention to pathogens and other materials, including heavy metal contaminants, present in spices. In this white paper, the USFDA indicates that at least 12% of spices brought to the United States are contaminated.[8] According to the United State Department of Agriculture (USDA), 95% of spices consumed in the U.S. are imported[9] and grown in countries, specifically in southern and Southeast Asia, polluted by leaded gasoline, smelters, battery-manufacturing plants, irrigation with industrial wastewater, and mines.[10-11]

For children, there is no safe blood lead level as even low levels of lead can adversely affect growth and development.[12] Adults are also not immune from the adverse effects associated with lead contamination as reproductive issues for both men and women, cardiovascular and nerve disorders, memory and concentration problems, and muscle and joint pain.[12] In the US, there is no specific criteria for the limit for lead contamination in spices. The USFDA has set a limit of 100 ppb for products that children will eat, such as candy.[7]

Traditional atomic absorption (AA) spectroscopy, graphite furnace AA spectroscopy, and ICP-MS are all analytical techniques that have been used to analyze food products, including spices, for lead.[13-16] However, improving the method using ICP-MS is desirable because it is capable of detecting metals at concentrations as low as 1 parts per trillion (ppt) and offers wide elemental coverage and fast analysis time compared with techniques such as atomic absorption spectroscopy. In brief, ICP-MS uses an inductively coupled plasma, an ionization source that completely decomposes a sample into its constituent elements and transforms those elements into ions, created by an argon torch that rests in an induction coil. A nebulizer introduces the sample through a central tube within the argon torch where the sample is ionized. These ions are then sent to a quadrupole mass analyzer where the mass to charge ratios are measured and their relative abundance within the sample are measured.[17]

This preliminary study aims to assess the levels of lead in spices, specifically, curry and turmeric, obtained from bulk food stores in northwestern Pennsylvania and to determine if ICP-MS is sensitive enough to detect lead in these spice samples.

2. Methodology

2.1 Chemicals

All chemicals were used as received. Nitric acid for acid digestion of spice samples was of trace metal grade from Fischer Scientific, Inc. Water and 30% hydrogen peroxide were ACS reagent grade from Ricca Chemical Company. A 1.000±0.003 mg/L lead standard for ICP-MS used for spiking samples was from Millipore-Sigma. Spices analyzed were obtained from bulk food stores in northwestern Pennsylvania.

2.2 Acid digestion of spice samples and sample preparation

All sample vessels, glassware, and plasticware were cleaned by soaking in 10% (v/v) nitric acid for at least 24 hours and rinsed abundantly with reagent grade water before use. Spice samples (1 gram) were digested with repeated additions of nitric acid and 30% hydrogen peroxide with heating at 95°C±5°C in an automated QBlock hot block (HORIBA, LTD.) according to USEPA Method 3050B.[18] For ICP-MS analysis, digestate volumes was reduced with heating, filtered, and diluted with reagent grade water to a final volume of 100-mL. All samples were run in duplicate, including a curry samples spiked with 5 ppb lead to assess recovery. A method blank, a sample that does not contain spice that was carried through the acid digestion and aforementioned sample preparation procedure, was used to evaluate contamination during preparation and analysis.

2.3 ICP-MS instrumentation

Thermo Fisher Scientific X-Series II ICP-MS was determine lead concentrations in digestates. A seven point calibration curve with concentrations between 5 ppb and 200 ppb was prepared. Specifically, the standards were prepared from a mixed ICP stock solution (Agilent Technologies). The stock solution was diluted using ultra-pure (18.2 M ohm cm^{-1}) water acidified with ICP-MS grade nitric acid.

3. Data

3.1 ICP-MS analysis of spice samples

This preliminary study demonstrates that ICP-MS is capable of detecting lead in the ppb range in the spice samples analyzed. Table 1 summarizes the lead concentrations in the spice samples and reagents used for sample preparation. Lead concentrations reported are the sum of lead-206, -207, and -208 isotopes as recommended by the USFDA when performing elemental analysis on food and related products.[19]

Table 1. Lead concentrations in samples and reagents

Sample	Pb concentration (ppb) Sum ^{206}Pb, ^{207}Pb, ^{208}Pb	Average Pb concentration (ppb)
Sample 1. Curry	2.447	1.978
Sample 2. Curry	1.509	
Sample 3. Curry – matrix spike	8.228	7.734
Sample 4. Curry – matrix spike	7.240	
Sample 5. Turmeric	14.809	13.419
Sample 6. Turmeric	12.028	
Method blank	0.270	–
30% Hydrogen peroxide	<0.000	–
Nitric acid	0.378	–
Water	0.579	–

Samples (3 and 4) were spiked with a lead standard to determine analyte recovery and a method blank was used to evaluate contamination during sample preparation and analysis. As mentioned in the *Methodology* section, duplicate curry samples were each spiked with 5 ppb lead and a +14% percent recovery of the lead spike in curry samples was achieved. This percent recovery meets the standard acceptable criteria for percent recovery of 100% ± 20%.[20] The lead concentration in spiked curry samples after ~5 ppb spike are subtracted from the total concentration is similar to those concentrations found in the non-spiked curry samples. The method blank concentration is well-below the concentrations of the spice samples, which indicates that the samples were not contaminated with lead from the laboratory environment during preparation or analysis. Cumin was also analyzed, but results in this preliminary study were inconclusive as one of the duplicate samples had a concentration of 9.053 ppb and the other 0.463 ppb. Analyst error as well as challenges with a lack of sample homogeneity may have resulted in the lead concentration differences observed.

3.2 Lead levels in recipes with turmeric and curry

The USFDA does not have a specific limit on acceptable lead concentration in spices, but sets limit at 100 ppb for candy and USEPA sets a limit at 15 ppb for drinking H_2O. To get a sense of lead levels in recipes with curry and turmeric, estimates of lead concentration in one serving of tikka masala sauce and coconut curry turmeric chicken were made. A half teaspoon of turmeric and curry are both approximately one gram, which is the amount of spice digested in this study. Three servings of tikka masala sauce requires one-fourth teaspoon of turmeric and one-half teaspoon of curry, and three servings of coconut curry turmeric chicken uses 1.13 teaspoons of turmeric and 2.25 teaspoons of curry. Table 2 summarizes lead levels in one serving of these recipes if the samples analyzed were used to prepare dishes with curry and turmeric.

Table 2. Estimated lead concentration in selected recipes containing curry and turmeric

	Pb concentration (ppb) in 3 servings of Tikka Masala Sauce	Pb concentration (ppb) in 3 servings of Coconut Curry Turmeric Chicken
Curry	~2 ppb	~10 ppb
Turmeric	~4 ppb	~30 ppb
Total	~6 ppb	~40 ppb

One serving of each of these recipes would have lead levels (tikka masala sauce and coconut curry turmeric chicken, ~2 ppb and 13 ppb, respectively), below limits that the USFDA and USEPA have set for candy and drinking water. Since lead levels in one serving are below the limits set by the USFDA and USEPA candy and drinking water, this initial analysis of curry and turmeric from a bulk food store in northwestern Pennsylvania indicate that, in moderation, the spices would be likely be safe to consume.

4. Conclusion

ICP-MS is a sensitive technique capable of detecting ppb lead levels in curry and turmeric purchased from bulk food stores in northwestern Pennsylvania. Our initial results warrant continuing to develop and improve a method for detecting lead in spices, and investigating the extent of the treat imposed by lead in commercially available spices.

5. Future Work

The sample preparation method used in this current work will be repeated and improved including using a microwave digestion system to increase efficiency in sample preparation. Initial ICP-MS analysis showed that lead levels in one-gram spice samples were just below the lowest concentration of the calibration standards. For future experiments, the calibration standards will need to be lowered to at least 0.5 ppb or the amount of spice digested will need to be increased (EPA Method 3050B allows up to 2-grams to be digested). More data will allow the limit of detection for the ICP-MS to be calculated from the mean and standard deviation of the replicate blank readings. One analyst will perform each step to ensure consistency in the method. The reagent grade water had higher than expected lead levels; therefore, a neater source of water will be used in future experiments.

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