Heavy Metals and Salmonella in Herbs and Spices

The purpose of this study was to determine the levels of arsenic, cadmium, lead, mercury, and *Salmonella* species in 15 popular and commonly consumed spices, and to assess any associated health risk(s). In addition, we investigated possible adulteration, specifically the addition of bulking and/or coloring agents, in four of the 15 spices that are commonly adulterated in these ways. We tested five to 10 products each of basil, black pepper, chili, coriander, cumin, curry powder, garlic powder, ginger, oregano, paprika, saffron, sesame seed, thyme, turmeric, and white pepper. We tested two to three samples of each product, for a total of about 330 samples. The products tested were selected based on sales data and data from shopper surveys of stores in New York, New Jersey, and Connecticut. We purchased the samples between October 2020 and January 2021 from stores and online from these three states. We made an effort to include nationally available, private/store, and culturally relevant products, with each sample representing a unique lot of each product.

SAMPLE PREPARATION

The samples were either masked or transferred into brown polyethylene jars, blind coded them to preserve their identities, and shipped them overnight to two independent, accredited laboratories for the analyses. At the heavy metals lab, sample preparation or homogenization was performed in fume hoods known to be free from trace-metal contamination. At the second lab, microbiological (Salmonella) tests, botanical identification tests, and the test for coloring agents or Sudan dyes were performed according to specified methods. Water, sample containers, and other materials used for the analyses were monitored for contamination, where appropriate, to account for any biases in s ample results.

TESTING

Analysis for Arsenic, Cadmium, Lead, and Mercury by Triple Quadrupole Inductively Coupled Plasma Mass Spectrometry (IC-QQQ-MS), With Collision Cell Reaction (CRC)

The samples were prepared and analyzed in accordance with the Association of Official Analytical Chemists (AOAC) Method 2015.01.

Arsenic Speciation Analysis by Ion Chromatography-Inductively Coupled Plasma-Collision Reaction Cell-Mass Spectrometry (IC-ICP-CRC-MS)

Select samples with high total arsenic results from each spice category were analyzed for total inorganic arsenic and three organic arsenic species—monomethyl arsonic acid (MMA), dimethyl arsinic acid (DMA), and trimethylarsine oxide (TMAO).

Analysis for *Salmonella*

Samples were tested for the presence of *Salmonella* following the Food and Drug Administration's Bacteriological Analytical Manual (FDA BAM) method (Chapter 5: *Salmonella*).

Botanical Identification Analyses

The identification of specific plant species in the tested samples was conducted by deoxyribonucleic acid (DNA) extraction and polymerase chain reaction (PCR) analysis, using specific primers for known bulking agents or adulterants.

Analysis for Sudan Dyes

The samples were prepared and analyzed for seven Sudan dyes by liquid chromatography tandem mass spectrometry (LC-MS/MS) in accordance with the American Spice Trade Association (ASTA) Method 28.

Sample analyses for chemical contaminants were precluded by at least a five-point calibration curve spanning the entire concentration range of interest. Calibration curves were performed at the beginning of each analytical day and verified during analysis. The testing conformed to the quality control criteria and performance requirements set in cited official methods, as well as to those under the lab's ISO 17025 accreditation.

RISK ASSESSMENT

We estimated daily consumption of the spices using the ¼ teaspoon or 0.5 grams per eating occasion FDA Center for Food Safety and Applied Nutrition (CFSAN) reference amount customarily consumed (RACC) of spices and herbs, our test results, and the average body weights of adults and children 1 to under 6 years old. We used the recommended body weights from the Environmental Protection Agency (2011 Exposure Factors Handbook). For heavy metals test results below the method detection limit (MDL), we used the method from Xue, et al. (2010), to estimate the average concentration of a product. If the metal was detected in any of the two or three samples of a product, then test results for that product that were below the MDL were assumed to have a concentration of half the MDL. If the metal was not detected in any of the samples tested of a product, we assumed a concentration of zero for all the samples of that product.

Non-Cancer Health Risks From Estimated Daily Intakes of Inorganic Arsenic, Cadmium, Lead, and Methyl Mercury

SELECTED HEALTH-BASED EXPOSURE AND OTHER LIMITS			
Heavy Metal	**Source**	**Endpoint and Basis for Limit**	**Value** (Unit)
Inorganic Arsenic	EPA (1991) (currently under review)	Non-cancer oral reference dose based on hyperpigmentation, keratosis, and possible vascular complications from chronic human exposures	0.3 ug/kg-day (upper limit)
		Lower bound of suggested range whereby 0.8 ug/kg/day was the upper bound	0.1 ug/kg-day (lower limit)
Cadmium	EFSA (2009)	Based on a tolerable intake of 2.5 ug/kg bw/week, to keep the population U-Cd concentration below 1 ug/g creatinine by the age of 50 years	0.36 ug/kg bw/day
Lead	OEHHA (2017)	OEHHA Proposition 65 Maximum Allowable Dose Level (MADL) for Chemicals Causing Reproductive Toxicity (total lead, oral exposure)	0.5 ug/day
	FDA Interim Reference Levels (IRLs)	In 2018 FDA derived the IRLs from the CDC reference value of 5 ug/dL of BLL to result in 3 ug/day for children and 12.5 ug/day for women of childbearing age or WOCBA, to serve as useful benchmarks in evaluating the potential for adverse effects in dietary lead.	3 ug/day (children) 12.5 ug/day (WOCBA)
Methyl Mercury	EPA (Updated 2001, currently under review)	Non-cancer oral reference dose based on developmental neuropsychological impairment	0.1 ug/kg-day
Salmonella	FDA		Absence in 25g

We compared our estimated daily intakes to health based limits in the above table using the following equation: *Hazard Quotient (HQ) = Estimated Daily Intake (or Exposure Dose)/Reference Dose.*

If the HQ is greater than 1, then adverse health effects are possible. As evidence suggests that impacts on neurodevelopment are a sensitive endpoint for each of inorganic arsenic, cadmium, and lead, we summed the HQs for the three heavy metals, to provide an estimate of the total hazard associated with a specific exposure pathway, which is known as a hazard Index (HI). As with HQ, if HI is greater than 1, then adverse health effects are possible.