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An Investigation of Heavy Metal Content in Commercially Available Powdered Spices and Recipe Mixes in Karachi

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Abstract. In Pakistani cuisine, there is a broad usage of commercially available powdered spices to enhance aroma and flavour of the food, most of Pakistanis use commercially available powder spices both branded and un-branded. The branded powder spices and recipe mix powder spices are available in aluminum laminated bags that enclosed in cardboard boxes whereas the un-branded powder spices and recipe mix powders are packed in polyethene bags. The recent study to examine the concentrations of heavy metals (Cu, Ni, Fe, Co, Mn, Cd and Pb) in sixteen samples of spices and recipe mix powders that were purchased from local markets and super stores located in Karachi city of Pakistan. Each sample was first digested by wet digestion and then the levels of heavy metals in samples were analyzed through flame atomic absorption spectrophotometer (FAAS). The data obtained was then compared with the permissible limits and recommended by Food and Agriculture Organization (FAO) and World Health Organization (WHO) for the samples studied. The average concentrations of heavy metals as Cu ranges from (0.0345-0.345) ppm, Fe ranges from (0.242-5.76) ppm, Ni ranges from (0-1.653) ppm, Pb ranges from (0-3.221) ppm, Co ranges from (0.05-1.529) ppm, Cd ranges from (0-0.551) ppm, Mn detected in the range of (1.195-7.018) ppm and As detected in the range from (0-0.005) ppm. The order of maximum level of heavy metals detected in spice samples are Mn>Fe>Pb>Ni>Co>Cu>Cd>As. Most of these values are within the maximum permissible limits as recommended by FAO/WHO and can be considered acceptable for all the samples studied, while in a few samples the levels of certain heavy metals are exceeding the allowable limits by FAO and WHO.

Keywords: spices, flame atomic absorption spectroscopy, wet digestion, heavy metals, world health organization (WHO), food and agriculture organization (FAO).

Introduction

Spices have rich sources of naturally occurring bioactive chemicals for numerous uses for people. The primary bioactive substances found in spices are proteins, alkaloids, flavonoids, carotenoids, fats, oils, phenols, minerals and vitamins, etc. (Hussain *et al*., 2019). Based on botanical analogies, families or plant components, the spices and herbs are categorized. Hot spices include dried white and black peppercorns, cilantro, ginger root, mustard seeds and chili or hot pepper (Adugna *et al*., 2023). Since ancient times, spices have been utilized in both pharmaceutical medication research and traditional medicine, known as ethnomedicine (Puvaca, 2022; Karahan *et al*., 2020). Many herbs and spices are either cultivated naturally in different places or are grown on small farms. Over the past three decades, the usage of spices has increased dramatically in most parts of the world, partly due to their medical effects (Oladeji *et al*., 2023). Spices also contain macro and micro-nutrients (Islam *et al*., 2022). The heavy metals found in spices and medicinal herbs are vital to living cells but could have dangerous impacts on human health, so it is essential that we understand the concentration of these metals in food (Brima, 2017). Even at low concentrations, heavy metals are not necessary for human health because they are hazardous and bio accumulate. In contrast to chemical concentration in the environment, bioaccumulation is the gradual increase in concentration of chemicals within a biological organism (Kandić *et al*., 2023; Savoca and Pace, 2021). The

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potential causes that contributes in heavy metal toxicity are contamination of irrigation water, fertilizers and metal-based insecticides, industrial emissions, harvesting, transportation and storage. Vegetables and crops cultivated in those soils that are contaminated with heavy metals accumulate more than those grown in un-contaminated soils (Yaradua *et al*., 2019). Plants may absorb metal contaminants from the soil through their vascular systems and roots, an accumulation of metals in the soil can endanger not only the ecosystem but also people, plants and animals. High metal concentrations in a plant can impair stomata resistance, increase oxidative stress and impede the formation of chlorophyll (Keshvari *et al*., 2021; Sulaiman and Hamzah, 2018). Heavy metal residues may also find their way into and become absorbed into spices and medicinal plants due to the high prevalence of heavy metals in the environment (Oladeji *et al*., 2024). According to their roles in biological processes, heavy metals are classified as either non-essential or essential components of living organisms. Essential metals are required for metabolic functions in living things. Zn, Mn, and Fe are important elements, while Pb and Cd are regarded as dangerous metals and physiologically unneeded (Jadaa and Mohammed, 2023). Though micronutrients like Zn, Mn, Cu, Cr, Ni and Fe are required for living beings but could be alarming at elevated levels due to metal toxicity (Alaqouri *et al*., 2020). The brain and kidney are the organs most commonly affected by heavy metal toxicity but other organs may also show symptoms. For instance, arsenic is known to cause cancer, lead exposure can cause hypertension and cadmium exposure can cause renal toxicity (Yaradua *et al*., 2020).

This has provided a direction to scientists all over the world to study heavy metals as contaminants in spices to evaluate their detrimental effects and to establish their acceptability for human consumption. From this point of view, the current study is designed to find out the heavy metal content in locally available samples of powdered spices and recipe mix.

Materials and Method

Sample collection. A total of 16 samples of powder spices (Table 2) out of which 8 samples were branded and 8 were un-branded chosen for the current investigation and acquired from local markets and stores. The branded spice and recipe mix samples were placed inside cardboard boxes and packaged in aluminum laminated polythene bags, while un-branded samples of powder spices and recipe mix were packed in polythene bags.

Reagents and standards. Analytical grade 65% nitric acid, deionized water, hydrogen peroxide, standard solutions of Cu, Fe, Ni, Mn, Co, Pb, Cd and As, all chemicals were purchased from Merck, Germany.

Experimental procedure. Each sample of spice (1 g) is weighed by using an analytical balance which is then transferred to a beaker then 5 mL of 65% nitric acid was added into the process of digestion for 20 min at 120 °C on a hot plate. Then it was allowed to cool till attained room temperature, finally, the digested sample was shifted to a 50 mL volumetric flask and diluted with deionized water upto the mark very carefully (Al-Keriawy *et al*., 2023). To determine the metal content in these samples by flame atomic absorption spectrophotometer (Model no. Perkin Elmer Aanalyst 400) and the radiation source was double-beam standard hollow cathode lamps made of the appropriate metal which were outfitted with a standard burner and air-acetylene supply. Concerning general recommendations, all the instrumental conditions used for the metal determination were established (Table 3): Cu (324.75 nm), Ni (232.00 nm), Fe (248.33 nm), Co (240.73 nm), Mn (279.48 nm), Cd (228.8 nm) and Pb (283.31 nm).

Results and Discussion

It is considered acceptable for heavy metals to exist in nature and in food that humans and animals consume within permissible ranges. In this study, the levels of heavy metals in branded and un-branded spices from local and mega markets of Karachi, Pakistan. The World Health Organization (WHO) and the Food and Agriculture Organization (FAO) have set maximum permissible limits for heavy metal concentrations. These limits are shown in (Table 1), which defines acceptable levels of heavy metals in spices. Significant differences were found between the concentrations of heavy metals in the samples analyzed and their limits. Flame atomic absorption spectroscopy (FAAS) was used to analyze numerous heavy metals, including Cu, Fe, Ni, Mn, Co, Pb, Cd and As in both branded and un-branded powder spices and recipe mix. Summary of the findings are illustrated in (Table 2) and (Figs. 1-4). The same solvent was used in the analysis and sample preparation, each element was given a distinct wavelength, as indicated in (Table 2). Among the branded and un-branded spice samples study the highest value of Mn detected is 7.018 ppm and which is greater than 2.0 ppm found in un-

Table 1. FAO/WHO determined acceptable limits for heavy metals in spices, expressed in ppm

Heavy metals.	Permissible limits	References
Copper	3.0 ppm	(Kohzadi *et al*., 2019)
Iron	300 ppm	(Russom *et al*., 2019)
Nickel	1.63 ppm	(El-Rahman, 2019)
Manganese	2.0 ppm	(Kohzadi *et al*., 2019)
Cobalt	0.2 ppm	(Kulhari *et al*., 2013)
Lead	5.0 ppm	(El-Rahman, 2019)
Cadmium	0.2 ppm	(El-Rahman, 2019)
Arsenic	1.0 ppm	(El-Rahman, 2019)

branded white pepper powder, while lowest is 1.195 ppm in branded garlic powder (Fig. 2) whereas Mn detected in the branded garlic powder is within the range, the permissible range of Mn being 2.0 ppm (Table 1). An excessive level of Mn can lead to central nervous system toxicity, whereas a Mn insufficiency causes illnesses (Russom *et al*., 2019). The highest value of Fe detected in the branded and un-branded spice samples is 5.76 ppm in un-branded kofta masala and lowest is 0.242 ppm in un-branded corn flour powder (Fig. 1) both of which are within the permissible range of Fe 300 ppm (Table 1). Iron deficiency leads to various health problems, such as anemia, as it is one of the essential minerals for human health (Marinescu *et al*., 2020; Russom *et al*., 2019). The highest value of Cu detected in the branded and un-branded spice samples is 0.345 ppm in branded karahi masala and lowest is 0.035 ppm in branded corn flour both are within the permissible range of Cu (3.0 ppm) (Table 1) and (Fig. 1). Cu poisoning can cause serious poisoning, although on the one hand, copper is important for the antioxidant defense system (Russom *et al*., 2019), the highest value of Pb detected in the branded and un-branded spice samples is 3.221 ppm in un-branded kofta masala and lowest is 0 ppm (Fig. 3) in usually most of the spice samples, the highest obtained value is within the permissible range of Pb (5.0 ppm) (Table 1). Pb has been determined to be the most hazardous environmental contaminant. It interacts with several biomolecules and has a detrimental impact on numerous developmental processes as well as the neurological, reproductive, gastrointestinal, renal, cardiovascular and immunological systems (Russom *et al*., 2019) the highest value of Ni detected in the branded and un-branded spice samples is 1.653 ppm in branded haleem masala and lowest is 0 ppm in un-branded corn flour (Fig. 2), the highest value is slightly greater than the permissible range of Ni (1.63 ppm) (Table 1). Since the body absorbs it at a very low rate, its toxicity does not occur frequently (Russom *et al*., 2019) the highest value of Cd detected

Table 2. Concentrations of heavy metals in branded and un-branded powder spices and recipe

Spice samples	Cu Conc. (ppm)	Fe Conc. (ppm)	Ni Conc. (ppm)	Mn Conc. (ppm)	Co Conc. (ppm)	Pb Conc. (ppm)	Cd Conc. (ppm)	As Conc. (ppm)
Branded fish masala	0.131	3.488	0.051	3.281	0.125	0.061	0.01	0
Un-branded fish masala	0.084	1.483	1.011	3.113	0.084	0.189	0.025	0.005
Branded garlic powder	0.067	0.894	0.156	1.195	0.063	0.529	0.02	0
Un-branded garlic powder	0.049	1.731	0.182	3.491	0.076	0.102	0	0.005
Branded white pepper	0.136	2.482	0.072	6.649	0.056	0.013	0	0.005
Un-branded white paper	0.134	1.315	0.029	7.018	1.529	0.012	0	0.005
Branded coriander powder	0.152	2.760	0.076	3.717	0.07	0.518	0.033	0.005
Un-branded coriander powder	0.162	1.563	0.139	2.562	0.107	1.352	0.01	0.005
Branded karahi masala	0.345	0.936	0.102	3.582	0.083	2.417	0.028	0.005
Un-branded karahi masala	0.060	2.527	0.031	3.137	0.062	0	0	0
Branded kofta masala	0.142	3.153	0.089	2.18	0.135	0.017	0.551	0
Un-branded kofta masala	0.255	5.760	0.048	4.268	0.323	3.221	0.011	0
Branded corn flour	0.035	5.487	0.053	1.799	0.05	0	0	0
Un-branded corn flour	0.106	0.242	0	1.911	0.107	0.386	0	0.005
Branded haleem masala	0.231	1.071	1.653	3.437	0.097	0.441	0.043	0
Un-branded haleem masala	0.066	2.114	0.046	3.417	0.091	0.218	0.025	0

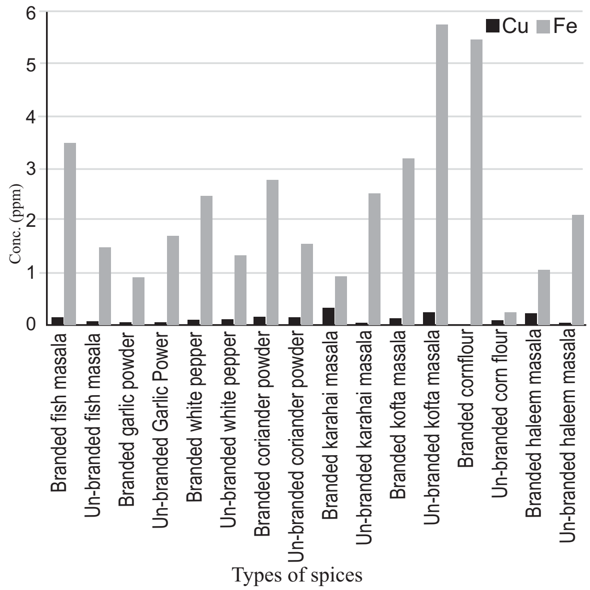

Fig. 1. Comparing the concentration of Cu and Fe for studied samples in ppm.

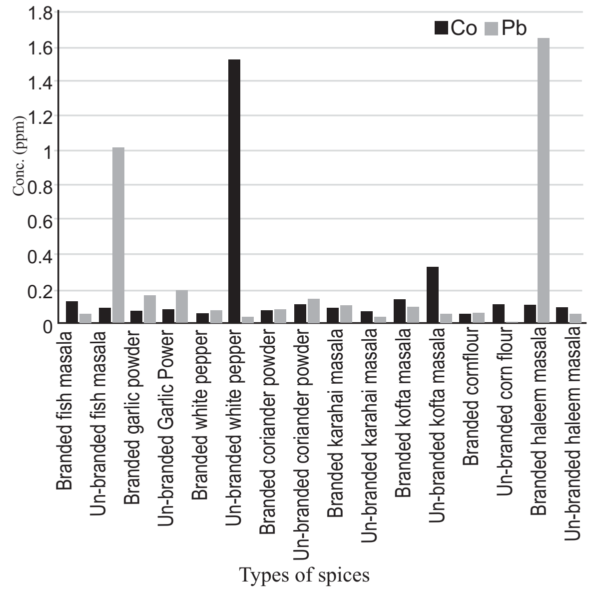

Fig. 3. Comparing the concentration of Co and Pb for studied samples in ppm.

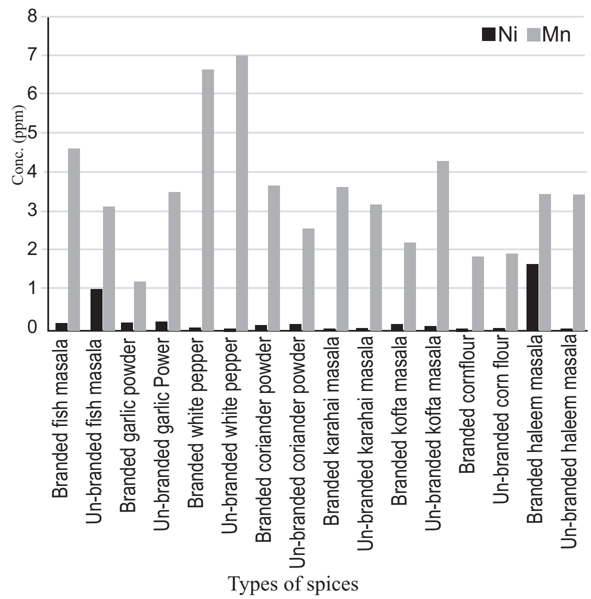

Fig. 2. Comparing the concentration of Ni and Mn for studied samples in ppm.

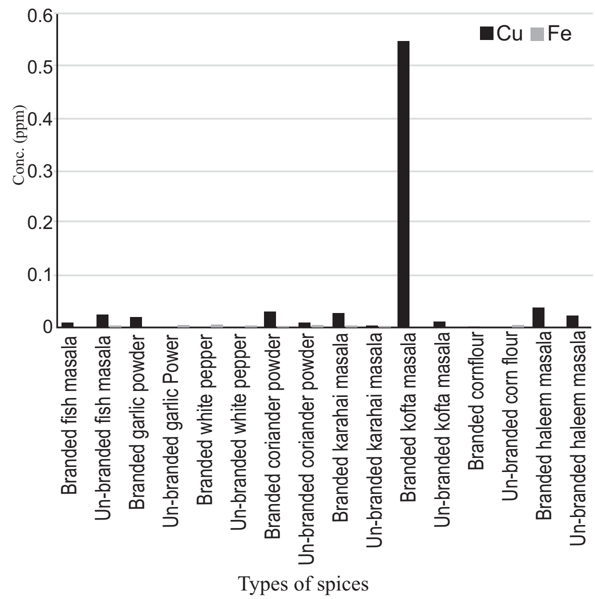

Fig. 4. Comparing the concentration of Cd and As for studied samples in ppm

in the branded and un-branded spice samples is 0.551 ppm in branded kofta masala and lowest is 0 ppm (Fig. 4) the highest value obtained is greater than the permissible range of Cd (0.2 ppm) (Table 1). Cd is categorized as human carcinogenic the increased concentration of Cd in meals can pose health risks (Russom *et al*., 2019) half samples of branded and un-branded spice powders and recipe mixes showed maximum 0.05 ppm levels of As and half of the samples showed 0 level of As (Fig. 4), all detected values are within the permissible range of As (1 ppm) (Table 1) the usage of specific pesticides and fertilizers may be

Table 3. Instrumental conditions for FAAS

Metal	Flame type	Slit width (nm)	Wavelength (nm)	Lamp
Cu	Air-acetylene	2.7/0.8	324.75	HCL*
Fe	Air-acetylene	1.8/1.35	248.33	HCL
Ni	Air-acetylene	1.8/1.35	232	HCL
Mn	Air-acetylene	1.8/0.6	279.48	HCL
Co	Air-acetylene	1.8/1.35	240.73	HCL
Pb	Air-acetylene	2.7/1.05	283.31	HCL
Cd	Air-acetylene	2.7/2.3	228.80	HCL
As	Air-acetylene	2.7/2.3	193.70	HCL

*HCL=Hollow cathode lamp

the cause of the elevated level of arsenic. Increased levels of arsenic in food and spices can damage the bladder, liver, lungs and skin. Its less severe side effects may result in blood vessel damage, nausea or vomiting (El-Rahman, 2019), the highest value of Co detected in the branded and un-branded spice samples is 1.529 ppm in un-branded white pepper powder and lowest is 0.05 ppm in un-branded corn flour powder (Fig. 3). The highest value of Co detected in the spice sample is greater than the permissible range of Co (0.2 ppm) (Table 1). Consuming cobalt from spices has no negative effects on health. The body requires very little cobalt, despite the fact that it is hazardous at high concentrations. Co is present in the body in an active physiological form as vitamin B_{12} (Russom *et al*., 2019).

Low levels of heavy metals are detected in the powder spices and recipe mix which are within the permissible limits of FAO/WHO and are safe to use. A very few of the studied samples showed detection of some heavy metals like Co, Cd and Mn detected slightly more than their allowable limits. The sample in which Cd was detected is branded Kofta masala the level of Cd is 0.551 ppm whereas the permissible limit is 0.2 ppm being carcinogenic Cd must be not present more than its allowable limit.

The excessive level of Mn can contribute in toxicity of central nervous system among the studied sample un-branded white pepper powder showed elevated level of Mn 7.018 ppm, so it's important to note that the un-branded spices and recipe mix powders are more risky to use, while branded ones are fit to use. The manufacturers must emphasize on the removal of heavy metals and storage of their raw materials and products.

Future of research scope. The consumers should know what they are consuming in their diet. Are they eating healthy food? A homemade food that is prepared by using spices that are already contaminated with heavy metals is not suitable for health as it will pose serious health risks. So, contamination of food with heavy metals must be known and obviously, manufacturers should focus on eliminating the levels of heavy metals from their products. With the help of such research, consumers and manufacturers both gain awareness regarding the presence of heavy metals in foodstuffs like spices and it is an attention-grabber topic as heavy metals have bio-accumulation and pose health risks, so it opens a gate for research.

Conclusion

The levels of heavy metals in the powdered spices are alarming as they pose serious health risks for their consumption hence, consumers should be aware of their daily intake of chemicals in their diet. Although the levels of heavy metals detected in most of the branded spices and recipe mixes are low and within the permissible limits, one cannot ignore the effects in cases where they exceed the threshold. The health impacts of each studied metals are Cu being essential nutrient, but excessive exposure can cause liver damage, kidney damage and neurological problems. Ni can cause allergic reactions, lung cancer and neurological damage. Fe essential nutrient but excessive exposure can cause gastrointestinal problems, liver damage and cancer. Co can cause cardiovascular disease, neurological damage and thyroid problems. Mn being essential nutrient but excessive exposure can cause neurological damage, respiratory problems and reproductive issues. Cd can cause kidney damage, bone disease and cancer. Pb can cause neurological damage, developmental delays and organ damage. Consumers are recommended to check product labels, check warnings or certifications indicating safe metal levels.

It is essential to be aware of the potential health impacts of these metals and take proactive steps to minimize exposure. By staying informed and taking precautions, you can reduce your risk and maintain a healthy lifestyle. Choose suppliers that test for metal contaminants and provide certification. Store spices properly keep spices in a cool, dry place to prevent moisture absorption, which can increase metal levels. Consume spices in moderation, as excessive consumption can lead to increased metal exposure. Encourage suppliers and regulatory bodies to conduct regular metal testing.

Conflict of Interest. The authors declare that they have no conflict of interest.

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