

## Determination of Mercury and Aluminum Concentrations in Aromatic Spices by Direct Mercury Analyzer and Inductively Coupled Plasma-Optical Emission Spectrometry

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**Abstract:** This study was aimed to determine the concentrations of mercury (Hg) and aluminum (Al) in aromatic spices widely consumed in Pakistan and worldwide. The Hg contents were analyzed by furnace-gold amalgamation direct mercury analyzer (DMA) while Al was determined by inductively coupled plasma-optical emission spectrometry (ICP-OES). The applied analytical techniques were validated by sensitivity, linearity, precision, spiking and analyzing Standard Reference Material, Spinach Leaves (NIST-1570a), obtaining satisfactory results in all cases. Concentrations of total mercury ranged from 4.44 ppm (turmeric) to 32.39 (cinnamon) while Al values varied from 48.70 ppm (cumin) to 1447 ppm (mango powder). In general contents of both Hg and Al were in accordance to the published literatures around the world on spices and very low compared to PTWIs specified by JECFA and EC. Their contribution to the overall intake by the subject aromatic spices was found very low and thus would not pose any threat to consumers.

**Key words:** Aromatic spices • Mercury • Aluminum • Direct mercury analyzer (DMA) • Inductively coupled plasma-optical emission spectrometry (ICP-OES)

### INTRODUCTION

The enchanting flavor and tanginess of spices make them pre-requisite in the preparation of palatable foods throughout the world. They are good preservatives and at times used for their several medicinal and pharmacological properties to fight diseases [1, 2]. According to FAO/WHO statistics, the global export of spices had spurt from 89, 21, 762 metric tons in 2007 to 1, 33, 26, 529 metric tons in 2010 [3]. Spices contain a variety of metal ions over a wide range of concentrations with important positive or negative physiological activities in humans and other animals. Among these the impacts of toxic elements like, Al, Pb, Hg, Cd and As on human health are of great interest, as these can be very damaging even at low levels when ingested over a long period of time [4-7].

Aluminum is the third most abundant element on earth and is widely distributed across the whole food chain [8]. It is a definite neurotoxin and is proposed to be

involved in the pathophysiology of neurodegenerative disorders such as Alzheimer's disease and Parkinsonism dementia [6, 9, 10]. Mercury is another very dangerous xenobiotic element especially its vapours and water soluble salts. It has the ability to accumulate in the internal organs of living organisms [11]. Various research studies on mercury have shown the adverse effects on humans such as tremors, impaired cognitive skills and sleep disturbance in workers with chronic exposure to mercury vapor [12]. In the past century, the anthropogenic inputs of mercury into the environment also significantly increased and therefore monitoring these inputs and estimating their many possible adverse impacts is very important, along with other toxic elements across the food chain [13].

A large variety of spices are used in Pakistan and in fact worldwide, in daily diet or as supplements to give flavor to dishes and for medicinal effects in health issues [14, 15]. Standard analytical techniques are always required to determine the concentrations of toxic elements

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in foods such as spices, since any increase causes serious disturbances in the whole human physiological system [15, 16-18]. In the past, micro-wave assisted combustion for sample digestion followed by analysis of Al using ICP-OES [19] and furnace-gold amalgamation method using direct mercury analyzer technique for analysis of Hg [13, 20] have been successfully applied to different foods.

In our previous studies on spices in the recent past, concentrations of various macro nutrients, minor, essential, trace and toxic trace elements have been determined in various spice varieties, obtaining variable amounts of analytes, largely dependent upon the type of spices [15]. The contents Hg and Al have already been studied in few spices from Ghana [21] and Turkey [14, 22], using atomic absorption spectroscopy and inductively coupled plasma atomic emission spectrometry (ICP-AES), respectively. However still there was no comprehensive study of Al and Hg involving large number of spice samples from several varieties widely consumed around the world. Therefore this study was designed to determine the contents of Hg and Al in aromatic spices widely consumed in Pakistan and worldwide. The furnace-gold amalgamation direct mercury analyzer and micro-wave assisted digestion followed by inductively coupled plasma-optical emission spectrometry techniques were evaluated as useful techniques to be applied for analyses of Hg and Al respectively, in spices. In order to know about the contamination levels if any, in the subject spice species, the provisional tolerable weekly intakes (PTWIs) recommended by Joint FAO/WHO Expert Committee on Food Additives (JECFA) and European Commission (EC) were used for both Hg and Al to compare our results and determine any toxicity from the subject food spices to consumers.

## Experimental

**Reagents and Materials:** All chemicals used in this research were of analytical reagent grade unless otherwise stated. For calibration curves of Al and Hg, standard stock solutions of 1000 mg/L were purchased from AnApure KRIAT Co, Ltd. Daejeon, Korea. The ultrapure deionised water (18.2 M $\Omega$  cm), was obtained from a Milli-Q Plus water purification system, Millipore, Bedford, MA, USA. The Standard Reference Material, Spinach Leaves, (NIST-1570a), was obtained from National Institute of Standards and Technology, Gaithersburg MD, USA. Analytical reagent grade concentrated HNO<sub>3</sub> (70%) was obtained from Dong Woo Fine-Chem Co., Ltd Iksan, Korea and was further purified by double sub-boiling distillation in quartz stills. The glass wares were first thoroughly washed with tap water using detergent, rinsed

with distilled water, then soaked in 10% v/v HNO<sub>3</sub> for at least 24 h and finally again rinsed extensively with Milli-Q water and dried in oven (HB-502M, Han Back, Korea), before use. All plastic containers, polypropylene ?asks, pipette tips, PTFE digestion tubes and reagents that came into contact with samples or standards were checked for contamination.

**Instrumentation:** A microwave combustion system, Multi-wave 3000 (Anton Paar GmbH, Graz, Austria), was used for digestion of the selected spice species. This system was having a total of 16 polytetrafluoroethylene-tetrafluoromethaxil (PTFE-TFM) digestion tubes (MF 100), enclosed in glass fiber reinforced polyetheretherketone (PEEK-GF) pressure jackets to withstand high pressures during digestion. Each digestion tube was having internal volume of 100 mL, capable of maximum pressure of 115 bar and temperature of 310°C. The instrument was programmable for power (600-1500 W), temperature and time. It was also having infrared and hydraulic sensors at the base of reaction vessels to ensure full control of selected temperature and pressure ranges during digestion of samples.

An automatic direct mercury analyzer (NIC MA-2000; Osaka, Japan) was used for the determination of total mercury contents in selected eleven spice species from Pakistan. This instrument was having separate sample preheating and decomposing furnaces, a mercury-gold amalgamation chamber and Silicon UV photo-detector. The auto sample changer (NIC BC-1) was also attached with the instrument.

The inductively coupled plasma-optical emission spectrometer (ICP-OES), used for the analysis of Al was a Varian Model 730-ES, simultaneous CCD, (Wyndmoor, PA, USA). This instrument was equipped with a SeaSpray concentric nebulizer and a cyclonic spray chamber (Glass Expansion, Pocasset, MA, USA). For analysis, a sample blank was scanned followed by a standard solution and a sample solution in the programmed wavelength range and the background correction wavelength was selected manually at appropriate background position for the analyte peak. Various important experimental conditions and instrumental configuration were the same as reported in Table 1.

**Preparation of Samples and Digestion:** A total of 520 samples of 11 selected spice species commercially available to consumers were collected during March to October, 2013, from local supermarkets all over Pakistan. The detailed descriptions of the spice species analyzed were as mentioned in Table 2.

Table 1: Operating conditions and measurement parameters of ICP-OES as applied for analysis of Al concentrations in spices

Spectrometer	ICP-OES (730-ES simultaneous CCD, Varian, USA)
RF generator (MHZ)	27.12
RF power (kW)	1.3
Nebulizer	SeaSpray
Spray chamber	Cyclonic
Plasma viewing	Axial
Processing mode	Area
Argon gas flow (L/min)	
Plasma	16.00
Auxiliary	1.5
Nebulizer	0.94
Read delay (sec)	30
Rinse (sec)	30
Replicates	3
Analyte, wavelength (nm)	Al(396.153)

Table 2: Description of aromatic spices analyzed

No.	Local name	English name	Botanical name	Family	Part used	Samples
1.	Ajwain	Bishop's weed	<i>Trachyspermum ammi</i>	Apiaceae	Fruits	48
2.	Kalwanji	Nigella seed	<i>Nigella sativa</i>	Ranunculaceae	Seeds	45
3.	Saunf	Fennel	<i>Foeniculum vulgare</i>	Apiaceae	Fruits	43
4.	Lowng	Clove	<i>Syzygium aromaticum</i>	Myrtaceae	Flower buds	45
5.	Dalchini	Cinnamon	<i>Cinnamomum verum</i>	Lauraceae	Bark	46
6.	Ellaichi	Green cardamom	<i>Elettaria cardamomum</i>	Zingiberaceae	Fruits	45
7.	Haldi	Turmeric	<i>Curcuma longa</i>	Zingiberaceae	Roots	47
8.	Dhania	Coriander	<i>Coriandrum sativum</i>	Apiaceae	Fruits	46
9.	Zira	Cumin	<i>Cuminum cyminum</i>	Apiaceae	Fruits	46
10.	Amchoor	Mango powder	<i>Mangifera indica</i>	Anacardiaceae	Fruits	45
11.	Anardana	Pomegranate	<i>Punica granatum</i>	Lythraceae	Seeds	46
	Total					502

All the samples were purchased in triplicate thus making a total of 1560 analyzed. First the samples were washed thoroughly with deionized distilled water and dried between layers of clean scientific tissue papers (Kimtech Science wipers, Seoul, Korea). For determination of moisture, each spice was dried at 60°C in oven, HB-502M, Han Back, Korea, until constant weight was achieved [23]. These were homogenized in a blender, MR 350 CA, Braun, Spain, properly labeled and stored in plastic bags at -20°C in refrigerator, MICOM CFD-0622, Samsung, Korea, until analysis.

For micro-wave digestion 0.25 to 0.5 g of each sample was accurately weighed directly into PTFE-TFM micro-wave digestion vessel. Then 7 mL concentrated HNO<sub>3</sub> (70%) and 1.0 mL H<sub>2</sub>O<sub>2</sub> was added and digested using microwave system. The combustion procedure was as follows: (1) 1000 W at 80°C for 5 min, (2) 1000 W at 50°C for 5 min, (3) 1000 W at 190°C for 15 min and (4) 0 W for 30 min to cool down. After cooling the contents of the tubes were transferred to 50 mL self standing polypropylene volumetric tubes with plug seal caps, Corning NY, Mexico. These were diluted to 25.0 g with ultrapure deionised water, labeled accurately and used for Al analysis [24].

**Analysis of Mercury:** Approximately 50 mg of each sample was weighted directly into the sample boat of mercury analyzer, along with catalysts; B (Al<sub>2</sub>O<sub>3</sub>) and M (Ca(OH)<sub>2</sub> + Na<sub>2</sub>CO<sub>3</sub>), above and below the samples. The samples were preheated for 60 s at 300°C, for dryness and then heated for 180 s at 850 °C to decompose mercury under a clean airflow, which was collected in a gold amalgamation tube. The tube was then further heated at 155°C to release mercury into UV-Photodetector and measured at 253.65 nm wavelength. Each sample was analysed in triplicate and Hg concentrations were reported on fresh weight basis in ppm, after considering the moisture contents [13, 20, 25].

**Calibration Procedure:** External calibration technique was followed for quantitative analysis. Standard calibration solutions of both Al and Hg were prepared by diluting their standard stock solutions in 19.6% (w/w) HNO<sub>3</sub> (the same percentage of acid present in the prepared samples). The calibration curves were built on eight different concentrations in such a way that content of analytes in the spice samples were within linear range of calibration curve. The correlation coefficients for all the calibration curves were at least 0.9996, showing good

linear relationship throughout the ranges of concentrations studied. The calibration standards were analyzed at regular intervals during analysis as samples to monitor the instrument drift. Also ultrapure deionised water blanks were frequently analyzed alongside samples to check for any loss or cross contamination. Sample blanks were also prepared by completion of the full analytical procedure above mentioned except samples and analyzed as blank [19].

**Quality Assurance:** Both the analytical methods applied for the determinations of Al and Hg in aromatic spices were validated by evaluation of the several quality parameters.

Sensitivity of the methods were determined through limits of detection (LOD) and limits of quantification (LOQ) for analyte elements which were calculated as three and ten times of the standard deviations of the blank divided by the slope of the analytical curve respectively [25]. The correlation coefficient ( $R^2$ ) values from the calibration curves were calculated to establish linearity [24, 26]. Precision was determined by the evaluation of percent coefficient of variation (CV%), using relative standard deviation of 06 repeated determinations of one sample [27]. For accuracy, the Standard Reference Material, Spinach Leaves (NIST-1570a), was analyzed for the determination of Al and Hg, measuring the recovery percentages. The analytical quality control was also verified by the recovery experiments for the both analyte elements, after spiking and measuring the recoveries (%) [19].

**Statistical Analysis:** Data were reported as mean  $\pm$  standard deviation of all replicate measurements for each sample. Significant differences ( $p < 0.05$ ) within means were analyzed by analysis of variance (ANOVA), following Tukey's honestly significant difference (HSD) test and Duncan test, in the SPSS (Statistical Package for Social Sciences) Software, Version 18, IBM, New York, USA.

## RESULTS AND DISCUSSION

The average concentrations and standard deviations of all replicate analyses obtained using ICP-OES for Al and DMA for Hg were reported as results. Table 3; gives methods validation parameters including linearity, sensitivity, precision, spiking and analysis of standard reference material, Spinach Leaves, (NIST-1570a), for Hg

and Al and Table 4; give average concentration ranges of Hg and Al determined in eleven spice varieties from Pakistan by ICP-OES and DMA.

**Analytical Methods Validation:** The important quality assurance parameters calculated for the analytical methods validation such as sensitivity, linearity, precision, spike recovery and SRM analysis proved that the analytical techniques followed in this study for Hg and Al, were fulfilling the specified criteria for analytical methods by Association of Official Analytical Chemists (AOAC) for analyses [24, 28]. The correlation coefficient ( $R^2$ ) values obtained from calibration curves were 0.9996 for both Hg and Al (Table 3), indicating a good linearity for the analytes. For sensitivity, the LOD and LOQ values were 0.002 ppb and 0.007 ppb for Hg and 0.450 ppm and 1.350 ppm for Al as shown in Table 3. These values allowed the determination of Hg and Al in the subject aromatic spices at the required levels. The values of correlation variance (CV%) calculated for precision were less than 3%; that is 2.21 (Hg) and 2.34 (Al) and the recovery of spiking tests were found to be 96.6% (Hg) and 97.6% (Al) (Table 3). These values were perfectly in accordance to the required criteria specified by AOAC for analysis [15, 28].

The percentage recoveries for the analysis of Standard Reference Material, Spinach Leaves, (NIST-1570-a) were 93.3% (Hg) and 101.3% (Al) (Table 3). The recovery values mean of the analyzed Hg and Al were within the interval of confidence ( $p < 0.05$ ) calculated for the certified values. Based on these results for the quality assurance parameters investigated, both the analytical techniques applied in this research to determine the contents of Hg by DMA and Al by ICP-OES in the selected aromatic spices were quite applicable and in accordance to the specified standard criteria of AOAC for analyses [19, 28].

**Concentrations of Hg and Al in Aromatic Spices:** The concentrations of Hg were closely related among the spice species analyzed while that of Al were found mostly variable largely dependent upon the subject spice varieties. The same trends of Hg and Al contents respectively, were also reported in literatures for spices from Ghana [21] and Turkey [22, 29, 30].

The concentrations of Hg ranged from 4.44 ppb (turmeric) to 10.54 ppb for mango powder. Cinnamon was the exception which gave very high Hg contents averaging 32.39 ppb, with minimum, maximum and median

Table 3: Validation of analytical method by linearity, sensitivity, precision, spike recovery and analysis of Standard Reference Material (NIST-1570-a), Spinach Leaves, for mercury and Al

Analyte	Linearity		Sensitivity			Standard reference material (NIST-1570-a), Spinach Leaves		
	coefficient (R <sup>2a</sup> )	LOD <sup>b</sup> (ppm)	LOQ <sup>c</sup> (ppm)	Precision (CV <sup>d</sup> %)	Spiking (%)	Certified value (ppm)	Observed value (ppm)	Recovery (%)
Hg <sup>e</sup>	0.9996	0.002	0.007	2.21	96.6	0.030±0.003	0.028±0.012	93.3
Al	0.9996	0.450	1.350	2.34	97.6	310±11	314±23	101.3

<sup>a</sup>) R<sup>2</sup> = Correlation coefficient

<sup>b</sup>) LOD = Limits of detection

<sup>c</sup>) LOQ = Limits of quantification

<sup>d</sup>) CV = Correlation variance

<sup>e</sup>) For Hg, LOD and LOQ are expressed in ppb

Table 4: Concentrations of Hg and Al in aromatic spices from Pakistan by DMA and ICP-OES

Analyte	Sample	Bishop's weed	Nigella seeds	Fennel	Clove	Cinnamon	Green cardamom	Turmeric	Coriander	Cumin	Mango powder	Pomegranate seeds
Hg (ppb)	Mean ±SD	7.19 <sup>b1</sup> ±1.81	10.23 <sup>b3</sup> ±3.98	6.67 <sup>b4</sup> ±2.42	7.65 <sup>b5</sup> ±2.08	32.39 <sup>a</sup> ±6.50	5.27 <sup>b2</sup> ±2.84	4.44±0.71	9.06±4.14	5.49±0.79	10.54±1.10	6.67 <sup>b6</sup> ±2.42
	Minimum	5.24	5.53	3.82	4.56	23.44	0.39	2.72	3.04	4.25	8.06	3.82
	Maximum	11.24	16.73	11.78	11.19	49.97	9.32	6.08	16.69	6.95	11.63	11.78
	Median	6.74	9.16	6.02	7.96	32.28	5.99	4.50	10.0	5.36	11.09	6.02
Al (ppm)	Mean ±SD	49.92±0.36	57.47±0.37	146.2±1.13	356.2±2.33	76.48±0.55	52.23±0.44	577.9±3.89	63.88±0.60	48.70±0.39	1447±7.60	1228±5.24
	Minimum	24.04	20.98	73.43	222.2	52.02	28.73	398.8	43.87	25.83	1026	810.4
	Maximum	93.52	102.5	240.3	509.2	112.9	80.92	728.3	103.4	103.1	2428	1639
	Median	44.70	44.31	115.7	342.9	72.47	53.70	600.1	56.40	36.56	1227	1290

<sup>1-6</sup> Values with different superscripts letters within row differ significantly (*p* < 0.05)

values of 23.44 ppb, 49.97 ppb and 32.28 ppb, respectively (Table 4). These values of Hg were lower than reported in literature (104 ppb) from India [30], while approximately the same compared to reported ( 1.30-24.93 ppb) for spices from Ghana [21]. The European Food Safety Authority (EFSA) has reported toxicity to mercury even at very low levels in member countries 31(EFSA 2004). The JECFA specified PTWI level for total Hg from all sources to be 1.6 ppb of body weight (bw), while EC specified 5 ppb of bw [32]. Thus keeping in view these specified limits and research findings, we can conclude that the Hg levels in the analyzed spice varieties from Pakistan were quite low and therefore would not pose any threat to consumers.

The concentrations of Al were the highest for mango powder (1447 ppm), followed by pomegranate seeds (1228 ppm), turmeric (577.9 ppm), clove (356.2 ppm), fennel (146.2 ppm), cinnamon (76.48 ppm), coriander (63.88 ppm), nigella seeds (57.47 ppm), green cardamom (52.23 ppm), bishop's weed (49.92 ppm) and cumin (48.70 ppm) (Table 4). These values of Al were lower than reported in literature for coriander (442 ppm), cumin (358 ppm), bishop's weed (317.2 ppm), fennel (278.9 ppm) and cinnamon (257.9 ppm) from Turkey [22, 29, 30]. The JECFA has developed a provisional tolerable weekly intake (PTWI) of 1 ppm bw, for aluminum from all sources [32]. From the present results the values of mango powder and pomegranate seeds are comparatively

higher. However keeping in view the facts that on the average all these analyzed spices are used in very small quantities, usually less than one gram per adult individual. Thus we can say that the contribution of Al from these edible spice varieties of spices is well below the specified PTWI.

## CONCLUSIONS

The furnace-gold amalgamation method using DMA for Hg analysis and micro-wave assisted digestion followed by ICP-OES analysis of Al was proved to be quite efficient analytical techniques for application to aromatic spice species. The concentrations of Hg were closely related for all analyzed spices while Al values were variable among the spices. The overall concentrations of both Hg and Al were below the standard recommended values of PTWIs by JECFA and EC. Any adverse health impacts to consumers from the daily intakes of the subject analyzed spices cannot be deduced under normal conditions.

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