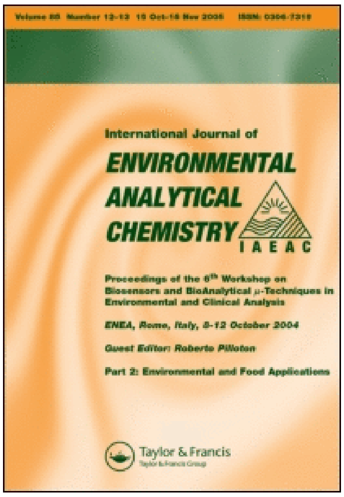


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# Trace Element Analysis of Food Spices by Instrumental Neutron Activation Analysis. I. Trace Elements in Food Spices Belonging to Umbelliferae, Myrtaceae, Piperaceae, Zingiberaceae and Lauraceae Families

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# TRACE ELEMENT ANALYSIS OF FOOD SPICES BY INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS. I. TRACE ELEMENTS IN FOOD SPICES BELONGING TO UMBELLIFERAE, MYRTACEAE, PIPERACEAE, ZINGIBERACEAE AND LAURACEAE FAMILIES

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Spices are extensively used in oriental and Latin American cooking and some of these are also used in pharmacy and perfumery. Therefore, concentrations of essential and toxic elements have been measured by instrumental neutron activation analysis (INAA) in seven different spices consumed in the area of Islamabad/Rawalpindi. Cumin and caraway seeds were found to contain relatively high amounts of essential as well as toxic elements. The comparison of our values with Canadian and Egyptian data shows variation in trace element contents of black pepper and cumin seeds. The studies show that these food spices are additional sources of trace element intake.

KEY WORDS: Spices, trace elements, toxic elements, essential elements, INAA.



## INTRODUCTION

There is increased interest in the amounts of various trace elements present in foodstuffs.[1–4] It has been established that essential trace elements, e.g. Fe, Zn, Mn, Co, Cu, Cr, I, Ni, Sn, V and Ca, are vitally important for biochemical systems, whereas toxic elements, e.g. As, Hg and Se, if present in relatively high amounts adversely affect these systems.[5,6] Toxic substances are either naturally occurring toxic elements and compounds or toxic compounds that are synthesized industrially. The danger accompanying the naturally occurring toxic elements and compounds depends upon their distribution in the environment. In the natural cycle their distribution remains relatively constant, mainly due to natural biological processes that affect both degradation and synthesis and they do not pose any significant threat to public health. But in the industrial processes, the toxic elements and compounds may enter the environment and affect the natural action of organisms in such a way that the balance between degradation can no longer be maintained.[7,8] Since food is the main source of intake of these elements, it is necessary to assess the adequacy and safety of the diet by monitoring the concentration of toxic and essential elements in various food items of daily consumption.

**Table 1** Botanical classification and nomenclature of the food spices analyzed

| *Family* | *Botanical name*[12] | *Common name* |
|---|---|---|
| Umbelliferae | Cuminum cyminum | Cumin |
| | Carum carvi | Caraway |
| Zingiberaceae | Elettaria cardamomum | Cardamom |
| | Amomum subulatum | Amomum |
| Myrtaceae | Eugenia caryopyllata | Clove |
| Lauraceae | Cinnamomum zeylanicum | Cinnamon |
| Piperaceae | Piper nigrum | Black pepper |

In continuation of our previous work,[9,11] seven commonly used food spices consumed in the area of Islamabad/Rawalpindi have been analysed. These spices which belong to the Umbelliferae, Myrtaceae, Piperaceae, Zingiberaceae and Lauraceae families are listed in Table 1, along with their botanical names and respective families.

Cumin and caraway have excellent flavour and are especially popular in oriental and Latin American cooking due to their aroma and taste. They can also be considered as medicinal plants for stomach pains. Cardamom and amomum have a very sweet fragrance and are commonly used in some dishes as well as in medicinal carminatives. Cloves, which are dried unopened flower buds, are very commonly used in oriental dishes and also as medicine for various forms of gastric irritability and dyspepsia. The oil distilled from cloves is mainly used in pharmacy and perfumery. Cinnamon is the inner bark of a tree which is light brown in colour and has a delicately fragrant aroma and sweet flavour. It is used in cooking and also has medicinal value. Black pepper is one of the earliest spices known and is the most widely used spice in the world today. It has limited uses in medicine as a carminative and a stimulant of gastric secretions.

Neutron activation analysis has been extensively employed[13–15] for trace element measurements because of its sensitivity and accuracy. Food spices were analysed employing this multi-element analysis technique for the determination of 18 toxic, essential and non-essential elements.



## EXPERIMENTAL

### *Sampling and Sample Preparation*

The specimen of food spices were collected randomly from local markets of Rawalpindi/Islamabad area in sufficient quantities. All samples were thoroughly washed with distilled deionized water and air dried in a clean fumehood. After weighing, each food spice was oven-dried at 60 °C for 72 h. Each item was then re-weighed to determine the moisture content for conversion of dry-weight to wet-weight values. The moisture contents were found to be 5–7%. All spices were analysed directly in the form they are usually used in cooking foods. Representative samples were stored in pre-cleaned polyethylene capped bottles.

**Table 2** Optimum experimental conditions and nuclear data[16] employed for the analysis

| *Irradiation time* | *Cooling time* | *Isotope* | *Half life* | *γ-peak used (keV)* |
|---|---|---|---|---|
| 2 min | 30 min | $^{38}Cl$ | 37.20 m | 1642.4, 2167.5 |
| | 2 hours | $^{56}Mn$ | 2.58 h | 846.6 |
| | | $^{42}K$ | 12.40 h | 1524.7 |
| | | $^{24}Na$ | 15.00 h | 1368.5 |
| 24 hours | 2 days | $^{82}Br$ | 35.40 h | 776.5 |
| | | $^{76}As$ | 26.30 h | 559.1 |
| | | $^{122}Sb$ | 2.70 d | 564.1 |
| | 2 weeks | $^{51}Cr$ | 27.80 d | 320.1 |
| | | $^{59}Fe$ | 44.60 d | 1099.3, 1291.6 |
| | | $^{46}Sc$ | 83.90 d | 889.3, 1120.5 |
| | | $^{65}Zn$ | 243.80 d | 1115.5 |
| | | $^{60}Co$ | 5.26 y | 1173.2, 1332.5 |
| | | $^{134}Cs$ | 2.04 y | 795.8 |
| | | $^{86}Rb$ | 18.60 d | 1078.8 |
| | | $^{181}Hf$ | 42.50 d | 482.0 |
| | | $^{152}Eu$ | 13.20 y | 1408.0 |
| | | $^{203}Hg$ | 46.60 d | 279.2 |
| | | $^{75}Se$ | 120.00 d | 264.5, 135.9 |



### *Irradiation*

Irradiations were carried out at a thermal flux of $2 \times 10^{13}\,n\,cm^{-2}\,sec^{-1}$ in a 5 MW swimming pool type research reactor. Each sample, weighing about 250 mg, was taken in triplicate and heat-sealed in pre-cleaned polyethylene vials for short irradiation and in silica vials for long irradiation. The samples along with appropriate amount of NBS SRM-1567 Wheat Flour and IAEA SRM H-9 Mixed Human Diet were irradiated for 2 to 60 min in pneumatic tube facility of PARR-I and in the reactor core for longer irradiations.

After appropriate cooling, the irradiated samples and standards were transferred to pre-weighed polyethylene vials and re-weighed to determine the exact weight. The gamma-ray spectra of the samples were measured for varying times ranging from 2 min to 16 h employing a 4 k series 85 Canberra multichannel analyzer coupled with ORTEC coaxial Ge(Li) detector. The system has a resolution of 2.1 keV for 1332.5 keV peak of $^{60}Co$ and peak/compton ratio of 40:1. The data from MCA was transferred through a serial port to the central computer facility and was processed employing various computer programmes developed by our group.

## RESULTS AND DISCUSSION

The optimized conditions for the non-destructive analyses of spices were established and are listed in Table 2 along with the nuclear data. The concentrations

**Table 3** Analysis of NBS and IAEA reference materials (concentrations in μg/g)[a]

| Element | Wheat flour (SRM-1567) | | Mixed human diet (H-9) | |
|---|---|---|---|---|
| | Our values | NBS values | Our values | IAEA values |
| Fe | 18.1 ± 1.0 | 18.3 ± 1.3 | 32.9 ± 2.0 | 33.5 ± 2.2 |
| Mn | 8.7 ± 0.5 | 8.5 ± 0.5 | 12.1 ± 0.9 | 11.8 ± 0.8 |
| Co[b] | 18 ± 1 | — | 50 ± 6 | 43 ± 5 |
| Zn | 9.6 ± 0.5 | 10.6 ± 1.0 | 27.3 ± 1.5 | 27.5 ± 1.8 |
| Na | 8.4 ± 0.3 | 8.0 ± 1.5 | 8120 ± 581 | 8100 ± 690 |
| K | 1368 ± 50 | 1360 ± 40 | 8295 ± 620 | 8300 ± 664 |
| Hg[b] | 1.0 ± 0.7 | 1.0 ± 0.8 | 5.0 ± 1.0 | 5.0 ± 1.0 |
| Se | 1.2 ± 0.2 | 1.1 ± 0.2 | 0.12 ± 0.01 | 0.11 ± 0.01 |
| As[b] | 5 ± 1 | (6) | 90 ± 30 | 88 ± 32 |
| Br | 9.8 ± 1.0 | (9) | 8.1 ± 0.6 | 7.5 ± 0.68 |
| Cl | — | — | 12590 ± 1570 | 12500 ± 1500 |
| Cs | — | — | 0.03 ± 0.002 | (0.025) |
| Rb | 0.9 ± 0.07 | (1) | 8.2 ± 0.5 | 8.0 ± 0.6 |

[a]Values in parentheses are uncertified.
[b]Concentrations in ng/g.



were determined on dry-weight basis. According to the optimum conditions studied, radionuclides of Cl, Mn, K and Na were measured employing short irradiations, whereas long irradiations along with appropriate cooling times were employed for the measurements of radionuclides of Br, As, Sb, Cr, Fe, Sc, Zn, Co, Cs, Rb, Hf, Eu, Hg and Se. The peaks of all the elements investigated, with the exception of Zn and Hg, were well resolved and interference-free. The full energy peak areas of 1115 keV from $^{65}Zn$ and 279.2 keV from $^{203}Hg$ were determined, after subtracting contributions from $^{46}Sc$ and $^{75}Se$ respectively, as described elsewhere.[10]

The precision and accuracy of our method was checked by analyzing NBS SRM-1567 Wheat Flour and IAEA Mixed Human Diet (H-9) employing the above mentioned experimental conditions. Our values are in fairly good agreement with certified values as shown in Table 3.

Table 4 presents the results as averages of four or more determinations with standard deviations around the mean values. The Fe and Mn contents are higher in caraway seeds and clove, respectively, while the concentrations of Cr and Zn are higher in cumin seeds and cardamom, respectively, than in the other spices. The content of Co is significantly higher in cinnamon as compared to other spices. Cumin and caraway seeds are not only rich in essential elements, but also contain higher amounts of toxic elements.

The data on trace element concentration in food spices are scarce, and few references are available. The comparison of our values with the literature values[17–19] given in Table 5 reveals that Fe and Mn contents in black pepper and cumin seeds are in reasonable agreement with Canadian values reported by Ila *et al.*,[17] while Egyptian values reported by Sherif *et al.*[18,19] are much lower. The concentration of Zn in our black pepper and cumin seeds is less by a factor of 2

**Table 4** Trace element concentration in some food spices (in $\mu g/g$ on dry weight basis)

| Element | *Cumin (seeds)* | *Caraway (seeds)* | *Cardamom (seeds)* | *Amomum (seeds)* | *Clove (buds)* | *Cinnamon (bark)* | *Black pepper* |
|---|---|---|---|---|---|---|---|
| Cr | 5.9±0.4 | 1.6±0.2 | 1.3±0.1 | 2.1±0.1 | 0.5±0.03 | 0.4±0.03 | 0.7±0.04 |
| Mn | 46.4±1.8 | 66.6±2.1 | 359±9.0 | 202±5.00 | 989±26 | 746±12.0 | 73.0±2.6 |
| Fe | 825±25 | 1058±58 | 115±4.0 | 263±6.0 | 70±3.8 | 67±2.5 | 97.9±5.0 |
| Co[a] | 31±2 | 42±3 | 13±1 | 31±3 | 27±2 | 305±13 | 17±1 |
| Zn | 41.5±2.0 | 23.8±1.0 | 62.0±2.0 | 34.9±3.0 | 9.3±0.7 | 9.7±0.8 | 13.0±1.0 |
| Na | 4246±165 | 492±19 | 206±10 | 90±3 | 2359±54 | 67±3 | 73±2 |
| K | 21500±1800 | 19500±1000 | 30700±1900 | 18200±900 | 19850±1300 | 4100±180 | 12300±1400 |
| As | 0.86±0.07 | 0.42±0.03 | 0.05±0.003 | 0.13±0.01 | 0.29±0.01 | 0.13±0.01 | 0.13±0.01 |
| Se | 0.85±0.05 | 0.17±0.02 | 0.32±0.02 | 0.36±0.02 | 0.17±0.02 | 0.12±0.01 | 0.05±0.003 |
| Hg[a] | 28±2 | 49±2 | 26±2 | 10±1 | 4±1 | 10±1 | 5±1 |
| Sb[a] | 56±3 | 9±1 | 18±2 | 13±1 | 28±2 | 25±2 | 4±1 |
| Cl | 8027±218 | 2715±113 | 6727±335 | 1498±13 | 4297±110 | 96±6 | 3745±288 |
| Br | 4.9±0.3 | 9.5±0.6 | 14.8±0.5 | 0.7±0.03 | 4.9±0.4 | 0.5±0.03 | 13.0±1.0 |
| Hf | 0.55±0.06 | 0.28±0.03 | 0.19±0.02 | 0.4±0.02 | 0.35±0.03 | 0.1±0.01 | 1.5±0.2 |
| Rb | 13.0±1.0 | 7.6±0.3 | 63.9±2.4 | 28.6±1.5 | 32.5±2.8 | 26.0±2.0 | 12.3±0.9 |
| Cs[a] | 117±5 | 260±10 | 59±3 | 63±4 | 70±2 | 250±20 | 64±3 |
| Sc[a] | 260±30 | 390±40 | 30±3 | 140±10 | 30±2 | 30±2 | 20±1 |
| Eu[a] | 35±3 | 50±2 | 26±2 | 23±2 | 25±2 | 20±2 | 13±2 |

[a]Concentrations in ng/g.

Table 5 Comparison of data on trace elements in spices (concentrations in $\mu g/g$)

| Element | Cumin seeds | | | Black pepper | | |
|---|---|---|---|---|---|---|
| | *Canada*[b] | *Pakistan* | *Egypt*[c] | *Canada*[b] | *Pakistan* | *Egypt*[c] |
| As | — | 0.86 | 0.1 | — | 0.13 | 0.36 |
| Sb[a] | — | 56 | 10 | — | 4 | 100 |
| Cl | 2049 | 8027 | — | 2960 | 3745 | — |
| Br | 25.2 | 4.9 | — | 55 | 13 | 0.1 |
| Fe | 701 | 825 | 0.2 | 155 | 98 | 0.4 |
| Mn | 35 | 46 | 0.1 | 42 | 73 | 0.1 |
| Zn | 101 | 42 | 0.3 | 29 | 13 | 0.3 |
| Cr | 1.6 | 6 | 0.02 | — | 0.7 | 0.03 |
| Co[a] | 234 | 30 | 100 | 58 | 20 | 300 |
| K | 18200 | 21500 | — | 14500 | 12300 | — |
| Na | 814 | 4246 | — | 686 | 73 | — |

[a]Concentrations in ng/g.
[b]Values taken from ref. 17.
[c]Values taken from refs. 18 and 19.

compared with the Canadian values, whereas Egyptian values are lower than our values. The Cr contents are higher and the Co concentration is very low in our cumin and black pepper seeds. As and Sb concentrations are higher in Pakistani cumin seed and Egyptian black pepper. The concentration of Cl in Pakistani cumin seeds is higher by a factor of 4 compared with the Canadian value, while in black pepper the values are similar. The contents of Br reported by Ila *et al.* are 5 times higher than our values. This variation may be due to different geographical and environmental conditions.

## CONCLUSION

The present paper provides base-line values of certain essential and toxic elements in some food spices. The cumin seeds contain higher amounts of toxic elements as compared to the rest of the food spices. Our studies show that these food spices are appreciable sources of trace element intake. The results reported in the present work may prove useful not only in the fields of nutrition and medicine, but in other areas such as geochemistry and food technology in particular. Further studies are desired to evaluate other food spices.

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