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TRACE ELEMENT ANALYSIS OF FOOD SPICES BY INAA

II. Solanaceae, Liliaceae, Zingiberaceae and Apiaceae Families

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Extensive use of food spices in oriental and Latin American cooking, pharmacy, and perfumery entails the evaluation of trace element contents in them. Instrumental neutron activation analysis (INAA) was used to determine the concentration of 18 trace elements (essential, toxic and non-essential) in six different spices consumed in Rawalpindi/Islamabad area. Turmeric powder and ginger powder were found to contain relatively high amounts of essential elements and low amounts of toxic elements. The comparison of our values with Canadian and Egyptian data shows variation in trace element contents of red pepper, turmeric, coriander and ginger. The estimated dietary intake through food spices show that these may be considered appreciable sources of trace element intake due to their extensive use. With the present studies the investigation of trace element contents of 13 food spices has been completed.

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

INTRODUCTION

The field of trace elements is one of the most looked for and interesting areas of nutrition.¹ Trace element levels have been measured extensively in individual food articles and integrated human diets.²⁻⁴ Essential trace elements are vitally important for various metabolic processes. On the other hand, toxic elements if present in relatively higher amounts adversely affect these processes.^{5,6} Therefore, from the nutritional view point the focus of interest is the adequacy of the daily intake of all essential elements by the population at large. As far as toxicologists are concerned their interest is in the intake of amounts of toxic elements. Being food the main source of intake of these elements,⁷ it becomes imperative to monitor the concentrations of toxic and essential elements in various food items of daily consumption.

Various foodstuffs have been analysed, for establishing base-line levels of trace elements, in our laboratory.^{4,8-10} A set of seven food spices was analysed and reported earlier.¹¹ In the present work another set of six commonly used food spices consumed in Rawalpindi/Islamabad area have been analysed. The list of these spices is given in Table 1, along with their botanical names and respective families.

Table 1 Botanical classification and nomenclature of the food spices analyzed

| <i>Family</i> | <i>Botanical name</i> ¹² | <i>Common name</i> |
|---------------|-------------------------------------|--------------------|
| Solanaceae | Capsicum (cayenne) | Red pepper |
| Liliaceae | Allium sativum | Garlic |
| | Allium cepa | Onion |
| Zingiberaceae | Curcuma longa | Turmeric |
| | Zingiber officinale | Ginger |
| Apiaceae | Coriandrum sativum | Coriander |

Red pepper, garlic, onion and ginger are all pungent spices. Red pepper is used to flavour many foods and as condiment. Garlic has a powerful aroma and its bulb contains an antibiotic, allium; it has antiseptic properties and is an expectorant and intestinal antispasmodic. Onion which has a characteristic aroma and sharp taste is used as a spice for many foods particularly meats, vegetables and salads. Turmeric is a perennial herbaceous plant. Its tuberous rhizomes or underground stems have been in use from a remote period as a condiment, a dye and medically as an aromatic stimulant. It is the ingredient of curry powder, relishes, pickles, spiced butters for vegetables, fish and egg dishes, etc. Ginger is an aromatic rhizome and is used as a spice flavouring food and medicine. This spice has a pleasant slightly biting taste. Coriander may be one of the oldest spices known since 5000 BC¹² and is used to flavour many foods particularly curries and confectionery. In the past it was used as a carminative medicine, but is now only used to mask unpleasant tastes and odours of drugs.

Food spices were analysed for the determination of 18 toxic, essential and non-essential elements, employing neutron activation analysis (INAA). This technique has been extensively used for this purpose^{13,14} due to its high precision, accuracy and sensitivity.

EXPERIMENTAL

Sampling and sample preparation

All six spices were taken in powder form as used in cooking foods. The specimens were collected as previously¹¹ from local markets of Rawalpindi/Islamabad area in sufficient quantities. Each of the powdered food spices was dried as described earlier¹¹ to eliminate moisture contents, if any. Representative samples were stored in pre-cleaned polyethylene capped bottles.

Neutron irradiations

Samples, each weighing about 250 mg, were taken in triplicate and heat sealed in pre-cleaned polyethylene and silica vials for short and long irradiations respectively.

A 5 MW swimming pool type research reactor (PARR-I) was used for neutron irradiations of the samples. The thermal neutron flux at the irradiation site was of the order of $2 \times 10^{13} \text{ n cm}^{-2} \text{ sec}^{-1}$. The samples along with appropriate amounts of NBS Orchard Leaves (SRM-1571) and IAEA Mixed Human Diet (SRM H-9) were irradiated in pneumatic tube facility of PARR-I for short irradiations (2–60 min) and in the reactor core for longer irradiations. The irradiated samples and standards were transferred to pre-weighed polyethylene vials and re-weighed to determine the exact weight.

Gamma-ray measurement and analysis

The gamma-ray spectra of the samples and standards were measured, after appropriate cooling (30 min to 2 weeks, see Table 2), for varying times ranging from 2 min to 16 h employing a 4 k series 85 Canberra multichannel analyzer (model 8503) coupled with ORTEC coaxial $30 \text{ cm}^3/\text{Ge}(\text{Li})$ 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, transferred from MCA to the central computer facility, were processed employing indigenously developed programmes. 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

RESULTS AND DISCUSSION

The optimized conditions for the non-destructive analyses of spices were the same as described earlier¹¹ and are listed in Table 2 along with the nuclear data¹⁵ for ready

Table 2 Optimum experimental conditions and nuclear data¹⁵ employed for the analysis

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

reference. Accordingly the radionuclides of short half-lives, i.e. ^{38}Cl , ^{56}Mn , ^{42}K and ^{24}Na were measured employing short irradiations and the radionuclides of relatively long half-lives, i.e. ^{82}Br , ^{76}As , ^{122}Sb , ^{51}Cr , ^{59}Fe , ^{46}Sc , ^{65}Zn , ^{60}Co , ^{134}Cs , ^{86}Rb , ^{181}Hf , ^{152}Eu , ^{203}Hg and ^{75}Se , were measured employing long irradiations along with appropriate cooling times. The precision and accuracy of the method was rechecked¹¹ by analyzing the NBS Orchard Leaves (SRM-1571) and IAEA Mixed Human Diet (H-9). Our values are in fairly good agreement with certified values as shown in Table 3.

The concentrations were determined on dry weight basis and the results, as averages of at least four determinations with standard deviations around mean values, are given in Table 4. The amounts of Mn and Co are comparable in turmeric and coriander and that of Cr and Zn in garlic and turmeric. The concentration of Fe is comparable in garlic and ginger. Turmeric contains exceptionally higher amounts of Fe, whereas Mn and Co are higher in ginger and garlic respectively. The As contents are significantly higher in red pepper and onion, while Sb and Hg contents are higher in turmeric and onion, respectively, than the other spices. Turmeric and ginger are not only rich in essential elements but also contain relatively low amounts of toxic elements.

Since very little information in literature is available on trace element data in food spices, our values were compared with the data of only two workers.¹⁶⁻¹⁸ Table 5 reveals that the amount of Fe in our turmeric and that of Mn in red pepper are reasonably close to Canadian values reported by Ila *et al.*¹⁶ The Zn and Cr contents are less by a factor of 2-3 and Co contents are less by a factor of 9-15 in our red pepper and turmeric than Canadian values. The Cl, Br, Na, and K contents in our red pepper are in reasonably good agreement with Canadian values, but in turmeric with the exception of K the concentrations of these elements are lower than

Table 3 Analysis of NBS and IAEA reference materials (concentrations in $\mu\text{g g}^{-1}$)^a

| Element | Orchard leaves (SRM-1571) | | Mixed human diet (H-9) | |
|-----------------|---------------------------|-------------|------------------------|--------------|
| | Our values | NBS values | Our values | IAEA values |
| Fe | 298 ± 15 | 300 ± 20 | 32.9 ± 2.0 | 33.5 ± 2.2 |
| Mn | 92 ± 3 | 91 ± 4 | 12.1 ± 0.9 | 11.8 ± 0.8 |
| Co ^b | 197 ± 7 | (200) | 50 ± 6 | 43 ± 5 |
| Zn | 23 ± 4 | 25 ± 3 | 27.3 ± 1.5 | 27.5 ± 1.8 |
| Na | 85 ± 4 | 82 ± 6 | 8120 ± 580 | 8100 ± 690 |
| K | 14595 ± 400 | 14700 ± 300 | 8295 ± 620 | 8300 ± 664 |
| Hg ^b | 149 ± 20 | 155 ± 15 | 5.0 ± 1.0 | 5.0 ± 1.0 |
| Se | 0.1 ± 0.01 | 0.08 ± 0.01 | 0.12 ± 0.01 | 0.11 ± 0.01 |
| As ^b | 9 ± 2 | 10 ± 2 | 90 ± 30 | 88 ± 32 |
| Br | 9.8 ± 1.0 | (10) | 8.1 ± 0.6 | 7.5 ± 0.68 |
| Cl | 678 ± 30 | (690) | 12590 ± 1570 | 12500 ± 1500 |
| Cs | 0.04 ± 0.002 | (0.04) | 0.03 ± 0.002 | (0.025) |
| Rb | 11 ± 2 | 12 ± 1 | 8.2 ± 0.5 | 8.0 ± 0.6 |

^a Values in parentheses are uncertified.

^b Concentrations in ng g^{-1} .

Table 4 Trace element concentration in some food spices (in $\mu\text{g g}^{-1}$ on dry weight basis)

| Element | Red pepper | Garlic | Onion | Turmeric | Ginger | Coriander |
|-----------------|--------------|--------------|-------------|--------------|--------------|-------------|
| Cr | 0.69 ± 0.04 | 1.47 ± 0.06 | 0.46 ± 0.07 | 1.53 ± 0.09 | 0.89 ± 0.07 | 0.28 ± 0.02 |
| Mn | 13.70 ± 0.9 | 12.5 ± 1.1 | 7.50 ± 0.6 | 29.20 ± 1.8 | 358 ± 10 | 23.30 ± 0.6 |
| Fe | 84 ± 4 | 140 ± 12 | 44 ± 3 | 1039 ± 31 | 145 ± 9 | 359 ± 27 |
| Co ^a | 23 ± 2 | 83 ± 3 | 17 ± 1 | 33 ± 3 | 18 ± 2 | 32 ± 2 |
| Zn | 22 ± 1.0 | 24 ± 1.4 | 19.3 ± 1.1 | 20.1 ± 1.3 | 28.2 ± 2.0 | 36.9 ± 1.5 |
| Na | 1310 ± 24 | 389 ± 18 | 888 ± 14 | 760 ± 8 | 443 ± 13 | 817 ± 25 |
| K | 19400 ± 1600 | 9600 ± 600 | 11300 ± 500 | 26000 ± 2800 | 12900 ± 1100 | 17200 ± 500 |
| As ^a | 250 ± 10 | 13 ± 2 | 270 ± 40 | 110 ± 10 | 12 ± 1 | 110 ± 10 |
| Se | 0.20 ± 0.02 | 0.13 ± 0.01 | 0.22 ± 0.02 | 0.18 ± 0.02 | 0.31 ± 0.02 | 0.15 ± 0.01 |
| Hg ^a | 11 ± 1 | 6 ± 1 | 42 ± 2 | 19 ± 2 | 6 ± 1 | 5 ± 1 |
| Sb ^a | 20 ± 2 | 110 ± 10 | 33 ± 2 | 130 ± 10 | 39 ± 3 | 27 ± 2 |
| Cl | 4341 ± 278 | 1362 ± 32 | 2647 ± 115 | 1080 ± 60 | 579 ± 23 | 3877 ± 182 |
| Br | 8.4 ± 0.5 | 2.4 ± 0.2 | 3.0 ± 0.2 | 1.2 ± 0.2 | 2.1 ± 0.2 | 4.6 ± 0.4 |
| Hf | 0.37 ± 0.02 | 0.03 ± 0.002 | 0.19 ± 0.01 | 0.43 ± 0.02 | 0.07 ± 0.003 | 0.41 ± 0.04 |
| Rb | 5.1 ± 0.2 | 2.7 ± 0.1 | 0.65 ± 0.02 | 10.1 ± 1.0 | 2.7 ± 0.6 | 1.8 ± 0.2 |
| Cs ^a | 50 ± 2 | 63 ± 4 | 40 ± 2 | 198 ± 8 | 24 ± 2 | 60 ± 3 |
| Sc ^a | 33 ± 2 | 42 ± 2 | 22 ± 2 | 260 ± 30 | 42 ± 4 | 220 ± 20 |
| Eu ^a | 27 ± 2 | 38 ± 2 | 19 ± 2 | 39 ± 1 | 44 ± 3 | 25 ± 2 |

^a Concentration in ng g^{-1} .

the values reported by the Canadians. Egyptian values, reported by Sherif *et al.*,^{17,18} for Cr, Mn, Zn and Fe contents in coriander and ginger are much lower (by a factor of 10–3000), while the contents of Co, As and Sb are higher by a factor of 7–10 than our values. This variation may be attributed, to some extent, to different geographical and environmental conditions but the Egyptian values for Cr, Mn, Zn and Fe are exceptionally low. It may be noticed that Sherif *et al.*,^{17,18} have not checked the precision and accuracy of their method employing any of the standard reference

Table 5 Comparison of data on trace elements in spices (concentrations in $\mu\text{g g}^{-1}$)

| Element | Red pepper | | Turmeric | | Coriander | | Ginger | |
|-----------------|---------------------|----------|---------------------|----------|--------------------|----------|--------------------|----------|
| | Canada ^b | Pakistan | Canada ^b | Pakistan | Egypt ^c | Pakistan | Egypt ^c | Pakistan |
| As | — | 0.25 | — | 0.11 | 0.34 | 0.11 | 0.1 | 0.01 |
| Sb ^a | — | 20 | — | 130 | 100 | 27 | 150 | 39 |
| Cl | 4554 | 4341 | 3142 | 1080 | — | 3877 | — | 579 |
| Br | 10.89 | 8.4 | 21.34 | 1.2 | — | 4.6 | 0.1 | 2.1 |
| Fe | 1815 | 84 | 970 | 1039 | 0.1 | 359 | — | 145 |
| Mn | 23 | 13.7 | 142 | 29.2 | 0.1 | 23.3 | 0.2 | 358 |
| Zn | 54 | 22 | 75 | 20.1 | 0.2 | 36.9 | 0.2 | 28.2 |
| Cr | 1.71 | 0.69 | 2.19 | 1.53 | 0.03 | 0.28 | 0.01 | 0.89 |
| Co ^a | 304 | 23 | 289 | 33 | 200 | 32 | 200 | 18 |
| K | 24700 | 19400 | 23900 | 26000 | — | 17200 | — | 12900 |
| Na | 1163 | 1310 | 1128 | 760 | — | 817 | — | 443 |

^a Concentration in ng g^{-1} .

^b Values taken from ref. 16.

^c Values taken from refs. 17 and 18.

Table 6 Dietary intake values of trace elements through spices (expressed in µg/week, person)

| Element | Cumin ^a seeds | Caraway ^a seeds | Cardamom ^a seeds | Amonum ^a seeds | Clove ^a buds | Cinnamon ^a bark | Black ^a pepper | Red pepper powder | Garlic powder | Onion powder | Turmeric powder | Ginger powder | Coriander powder | Total intake | Weekly requirement ^c / tolerance ¹⁹ |
|-----------------|-----------------------------|-------------------------------|--------------------------------|------------------------------|----------------------------|-------------------------------|------------------------------|-------------------------|------------------|-----------------|--------------------|------------------|---------------------|-----------------|---|
| Cr | 2.8 | 0.8 | 0.9 | 1.0 | 0.7 | 0.2 | 1.5 | 3.3 | 7.0 | 12.0 | 0.7 | 2.7 | 1.4 | 35 | 0.07-1.4 |
| Mn | 22 | 31 | 255 | 95 | 1395 | 351 | 155 | 66 | 60 | 197 | 14 | 1074 | 112 | 3827 | 3.5-35 |
| Fe ^c | 0.4 | 0.5 | 0.08 | 0.12 | 0.1 | 0.03 | 0.2 | 0.4 | 0.7 | 1.2 | 0.5 | 0.4 | 1.7 | 6.3 | 70-140 |
| Co ^b | 15 | 20 | 9 | 15 | 38 | 143 | 36 | 110 | 398 | 446 | 16 | 54 | 154 | 1454 | 1-12 |
| Zn | 20 | 11 | 44 | 16 | 13 | 5 | 28 | 106 | 115 | 507 | 9 | 85 | 177 | 1136 | 56-105 |
| Na ^c | 2.0 | 0.2 | 0.15 | 0.4 | 3.3 | 0.3 | 0.15 | 6.3 | 1.8 | 23.3 | 0.4 | 1.3 | 3.9 | 43.5 | 805-21000 |
| K ^c | 10.1 | 9.2 | 21.8 | 8.6 | 28.0 | 1.9 | 26.1 | 93.1 | 46.1 | 296.6 | 12.2 | 38.7 | 82.6 | 675 | 2100-35000 |
| As | 0.4 | 0.2 | 0.04 | 0.06 | 0.41 | 0.06 | 0.28 | 1.2 | 0.06 | 7.09 | 0.05 | 0.04 | 0.53 | 10.42 | 2800 |
| Se | 0.4 | 0.08 | 0.23 | 0.17 | 0.24 | 0.06 | 0.11 | 0.96 | 0.62 | 5.8 | 0.08 | 0.93 | 0.72 | 10.4 | 1400 |
| Hg ^b | 13 | 23 | 18 | 5 | 6 | 5 | 11 | 53 | 29 | 1103 | 9 | 18 | 24 | 1317 | 280 |
| Sb ^b | 26 | 4 | 13 | 6 | 39 | 12 | 8 | 96 | 528 | 866 | 61 | 117 | 130 | 1906 | — |
| Cl ^c | 3.8 | 1.3 | 4.8 | 0.7 | 6.0 | 0.05 | 7.9 | 20.8 | 6.5 | 69.5 | 0.5 | 1.7 | 18.6 | 142.2 | 1400-3500 |
| Br | 2 | 5 | 11 | 0.5 | 7 | 0.5 | 28 | 40 | 12 | 79 | 1 | 6 | 22 | 214 | 7 |

^a Intake values estimated from data taken from ref. 11.

^b Values expressed in ng/week, person.

^c Values expressed in mg/week, person.

All values expressed on dry weight basis.

Weekly ingestion of each of the spices was estimated on the basis of survey of the food habits of the residents of Rawalpindi/Islamabad area and taken [in grams (dry weight)/week, person, from left to right in the table] as:0.47, 0.47, 0.71, 0.47, 1.41, 0.47, 2.12, 4.8, 4.8, 26.25, 0.47, 3.0 and 4.8, respectively.

materials, instead they used self-made standards for the determination of different trace elements.

The weekly dietary intake values for essential and toxic elements through spices (present work and our work reported in Ref. 11) were estimated and are given in Table 6 along with the suggested weekly requirement/tolerance limits.¹⁹ The estimated intake of Fe through coriander is maximum in comparison with the rest of the varieties, whereas intake of Cr through cinnamon is minimum. The intake of Zn and Co through onion is higher by a factor of 2–100 and 1–50 respectively, than rest of the spices. The total estimated intake of Cr, Mn, Fe, Co and Zn corresponds to 2.5%, 11%, 4.5%, 0.01% and 1% respectively, of the upper limits of the suggested weekly requirements. The estimated intake of toxic elements is well below the tolerance limits.

CONCLUSION

With the present studies the investigations on trace element contents of almost all the food spices commonly consumed have been completed. This paper provides the base-line values of certain essential and toxic elements in another set of six food spices. The turmeric powder and ginger powder are not only rich in essential elements but also contain relatively lower amounts of toxic elements. Estimation of weekly dietary intake shows that food spices are appreciable sources of trace element intake. The intake of toxic trace elements is within the safe limits recommended by joint WHO/FAO expert committees.³ Comparing all the spices investigated it can be seen that cumin, clove, caraway/turmeric, cinnamon and cardamom are rich in Cr, Mn, Fe, Co and Zn, respectively. Each spice has its characteristic taste, flavour and other properties such as consumption of red pepper causes water retention in body whereas spicy food consumed by teenagers is partly responsible for acne and sore spots in mouth. Garlic is considered helpful to control the cholesterol level in blood. Work in the direction of correlation between different properties/characteristics of spices and trace elements would not only be desirable but needed too. The results reported in the present work may prove useful in the fields of nutrition, medicine, geochemistry and food technology.

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