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Availability of essential trace elements in Indian cereals, vegetables and spices using INAA and the contribution of spices to daily dietary intake

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Abstract

Indian diet is primarily vegetarian and consists of various cereals and vegetables along with spices, often used in the preparation of curries. The nutritive potential of each ingredient, in terms of trace element contents, has been evaluated using instrumental neutron activation analysis (INAA). Four minor (Na, K, P and Cl) and 16 trace elements (Br, Co, Cr, Cs, Cu, Fe, Hg, Mn, Mo, Rb, Sb, Sc, Se, Sr, Th and Zn) have been determined in six cereals, nine vegetables and 20 spices and condiments, including two betel leaves. None of the carbohydrate-rich cereals or potato was rich in any of the essential elements but leafy vegetables showed higher contents of Fe and other nutrients. Fe/Zn is well correlated with Fe contents in cereals and spices. Out of various spices, cinnamon was most enriched in Fe, Co, Cr, Na, K, P and Zn, whereas turmeric and curry leaves were found to be particularly rich in Se. Cumin and mustard seeds were rich in Cu. Some environmental contaminants, such as Hg, Cr, Br and Th, were also present in significant amounts. An attempt has been made to evaluate the contribution of essential elements (Cr, Cu, Fe, Mn, P, Se and Zn) in spices to the daily dietary intake (DDI) through an Indian vegetarian diet. For a typical mixture of six commonly used spices, contributions of Cr, Fe, Mn and Zn, were found to be 7.5% of DDI in each case.

Keywords: Cereals; Vegetables; Spices; Essential nutrients; Instrumental neutron activation analysis; Daily dietary intake; Toxic contaminants

1. Introduction

It is now well recognized that several trace elements are essential constituents of enzymes and play a vital role in human metabolism. All the nutrient elements are primarily supplied through diet. However, this may change, depending on age, sex, health status, geographical and climatic conditions (O' Dell & Sunde, 1997; Prasad, 1993). For all the elements essential for metabolism, there exists a range of intake over which their supply is adequate for the body. However, beyond this range, deficiency and toxic effects are observed (Merian, 1991). Therefore, it is essential to determine elemental contents of food items and to estimate their daily dietary intake (Jansen, Kandall, & Jansen, 1990). WHO and the Indian Council of Medical Research (ICMR) have recommended selective studies of individual foodstuffs as an important step in the estimation of dietary intake of trace elements (ICMR, 1987; WHO, 1985). The American Chemical Society has published a monograph on antioxidant properties of spices often used in the preparation of vegetable curries (Risch & Ho, 1997).

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The Indian vegetarian diet primarily consists, typically of bread baked at home, boiled rice and curries prepared from pulses (lentils) and vegetables, including spices added during cooking. Though bread is often made from wheat flour, other cereals, such as maize, barley, millet and sorghum, are also used, depending on the region or socio-economic status. Indian curry is spiced with salt, red chilies and other spices, such as turmeric & coriander powder, depending on individual taste and other considerations. Besides, condiments such as cloves, black pepper, cardamom and bay leaves are also used to spread over cooked/fried rice and curry preparations. Several workers have discussed the constituents of these spices in terms of their volatile oils and other vitamin contents (Gopalan et al., 1999; Pruthi, 1999). Pruthi (1999) has extensively reviewed various characteristics and determination of chemical and toxic constituents of Indian spices. Gopalan et al. (1999) have compiled selected elemental data for various food items, including cereals, vegetables and spices used in different parts of India. Crosby (1977) reviewed determination of metals in food items from all parts of world. Ila and Jagam (1980) reported 24 elements in six Indian spices. Abou-Arab and Abou-Donia (2000) have determined heavy metal contaminants in Egyptian spices.

Several workers have analyzed individual food items of their respective countries or regions and calculated the contributions of trace elements to the total dietary intakes (Bro, Sandstrom, & Heydorn, 1990; Cunningham & Stroube, 1987; Dermelj et al., 1996; Laiyan, Ying, Wei, & Xi, 1991; Liu, Chung, Chuang, Wang, & Aras, 1991; Mannan, Waheed, Ahmed, & Qureshi, 1992; Sarmani, Wood, Hamzah, & Majid, 1993; Wieteska, Zioiek, & Drzewienkka, 1996). Cunningham and Stroube (1987) analyzed many food composites as part of a study conducted by the Food and Agricultural Organization of the UN. Laiyan et al. (1991), Mannan et al. (1992) and Sarmani et al. (1993) have estimated intake of essential and toxic trace elements in Chinese, Pakistani and Malaysian foodstuffs, respectively. Wieteska et al. (1996) have determined eight elements (Al, Ca, Cd, Cu, Fe, Mg, Pb and Zn) in Polish vegetables. Miyamoto, Kajikawa, Zaidi, Nakanishi, and Sakamoto (2000) determined 26 trace elements in food spices and pulses of different origins by NAA and PAA. Dermelj et al. (1996) and Favaro et al. (2001) have employed NAA for the determination of some essential and toxic elements in food articles in Slovenian and Brazilian nursery diets. In an extensive study, Srikumar, Kallgard, Ockerman, and Akesson (1992) investigated the change in trace element status by switching from a mixed to a lacto-vegetarian diet in Sweden. As part of a IAEA Coordinated Research Programme (CRP), intakes of essential and toxic elements have been estimated in day-to-day foodstuffs (Cortes-Toro et al., 1994).

In view of such studies from other parts of world, we have also analyzed raw as well as cooked, Indian vegetarian diets, including dietary components, by instrumental neutron activation analysis (INAA) and compared daily dietary intake (DDI) with the Recommended Dietary Allowances (RDAs) (Samudralwar & Garg, 1994; Singh & Garg, 1997). In continuation we report here the analyses of six cereals, nine vegetables and 20 spices and condiments, along with two betel leaves, for 20 elements, by employing INAA. The contribution of spices to the daily dietary intake has been estimated on the basis of a typical Indian vegetarian diet formulation (Gopalan et al., 1999). Four biological standard reference materials (SRMs) of plant origin including our data on participation in the intercomparison study of spinach, IAEA-331, are also presented for quality assurance and data validation.

2. Materials and methods

2.1. Sample preparation

All the food articles including cereals, vegetables, spices and condiments, in raw form, were procured from the local market. The quality of the cereal grain was so chosen as that consumed by the middle-income group. In each case, 3–5 samples were collected at intervals of a few days or from different stores, so as to have representative samples as far as possible. The seed type items, such as rice, wheat, fenugreek and black pepper, were wipe cleaned with tissue paper, dried under an IR lamp at ~80 °C and crushed to powder in a food processor/mixer and then in an agate mortar. Leafy and other vegetables were thoroughly washed with distilled water so as to remove all the dirt and surface contamination. All the samples were dried in an oven at <80 °C for overnight and crushed to powder in an agate mortar to a uniform particle size (50 mesh). All the replicate samples of different food products were mixed well for homogeneity and stored in BOROSIL glass bottles. These were then irradiated in a ^{60}Co Gamma Chamber-900 at a dose of 30 kGy to avoid biodegradation according to IAEA recommendations (IAEA, 1984). SRMs of rice flour (SRM 1568), citrus leaves (SRM 1572), both from NIST, USA, and Bowen's kale (Bowen, 1985) were dried before use. Spinach (CRM 331), from Analytical Quality Control Services (AQCS) at the Seibersdorf Laboratory of the International Atomic Energy Agency (IAEA), Vienna, was analysed as part of a inter-comparison study (IAEA, 1997). A synthetic multielemental comparator standard was prepared by spiking 2–5 μg each of AR/GR or high purity grade salts (mostly nitrates and in some cases oxides) in aqueous/nitric acid solution on a Whatman No. 42 filter paper strip.

2.2. Irradiation and counting

About 30–40 mg aliquots each of SRMs and samples, in dried form, were packed in alkathene or high purity Al-foil (Superwrap) and irradiated, together, in a batch of 5–15 at a thermal neutron flux of $10^{11}/l0^{13}$ n/cm^2/s for 10 min and 1 d in the APSARA and CIRUS reactors, respectively, at the Bhabha Atomic Research Centre (BARC), Mumbai. Care was taken, that in each batch, 2–4 SRMs, along with synthetic multielemental standards, were packed along with 4–10 samples. For short irradiation (10 min) counting was followed on an 80 cm^3 coaxial HPGe detector (EG&G ORTEC) and 4k MCA at the Radiochemistry Division of BARC, Mumbai. Long irradiated (1 d) samples were airlifted to Nagpur and counted on a 113 cm^3 HPGe detector (PGT, Germany) coupled to a 4k MCA (Nucleonix, India) at different intervals of 2 day, 4 day, 2 week, 4 week and 8–10 weeks for up to 3 months. In each case, either duplicate analyses were performed or elemental concentrations from different irradiation periods or from different photo peaks of the same radionuclide (e.g., ^{24}Na,^{42}K, ^{82}Br) were determined in short as well as long irradiations, whereas Sc and Co were determined from two different photo peaks. Elements determined from different irradiation and counting schedules are the same as described earlier (Garg, Kumar, Reddy, & Nair, 2005). Blank correction was done for ^{28}Al activity or other interfering activities due to trace elements on Al foil. After the end of two weeks, aluminium wrapping was removed and the sample transferred to glass vials. Phosphorus was determined by counting β^- activity from ^{32}P on an end window GM counter (ECIL, India) using a 27 mg/cm^2 Al-filter after a delay of ~3 weeks, as described earlier (Weginwar, Samudralwar, & Garg, 1989). In all cases, statistically significant counts ($>10^4$) were collected, so that statistical error was $<\pm 1$–2%.

3. Results and discussion

3.1. General

Elemental concentrations were calculated in SRMs and samples using multielemental synthetic standards as comparators. For some elements, such as Na, K, Cl, Br and P, however, Bowen's kale (Bowen, 1985), a widely analyzed botanical standard, was used as comparator. Mean concentrations of 20 elements calculated on the basis of replicate analyses, different irradiations and γ-ray energy photo peaks of different SRMs, are listed in Table 1. A comparison of our data with the certified/literature values (in parentheses) shows good agreement, within ±5–10% in most cases. Standard deviations, in most cases, were small and well within ±10%. We had participated in the intercomparison study for the certification of spinach (CRM 331) for the determination of trace elements (IAEA, 1997). Most elements compare well with the IAEA estimated mean values and those of Becker (1995). Relative standard deviations are <±5–10% for most cases, suggesting a high order of precision. Therefore, it is presumed that all the elemental concentrations in the samples should be accurate and precise within <±10%. Elemental concentrations in various cereals and vegetables are listed in Tables 2 and 3,

Table 1
Elemental concentrations in standard reference materials and inter-comparison study

Element	Bowen's Kale	Citrus leaves (NIST 1572)	Rice flour (NIST 1568)	Spinach (IAEA 331)
Ba (μg/g)	4.86 ± 0.66 (4.86)	–	–	6.72 ± 0.05 (13.2) [6.94]
Br (μg/g)	25.0 ± 3.1 (24.9)	8.03 ± 0.40 (8.2)	0.92 ± 0.03 (1)	34.3 ± 1.0 (–) [37.91]
Cl (mg/g)	3.23 ± 0.20 (3.56)	(–)	–	6.23 ± 0.28 (–) [6.64]
Co (ng/g)	65 ± 13 (63)	24 ± 2 (20)	20 ± 1 (20)	341 ± 9 (382) [390]
Cr (μg/g)	0.37 ± 0.05 (0.37)	0.74 ± 0.06 (0.8)	0.16 ± 0.01 (–)	2.12 ± 0.11 (1.92)
Cu (μg/g)	4.77 ± 0.04 (4.89)	18.1 ± 0.9 (16.5 ± 1)	–	11.1 ± 0.3 (–) [12.2]
Fe (μg/g)	120 ± 8.6 (119.3)	108 ± 4 (90)	8.9 ± 2.1 (8.1)	289 ± 6
Hg (ng/g)	164 ± 32 (171)	82 ± 4 (80)	–	269 ± 22 (293) [259]
K (mg/g)	21.2 ± 2.0 (24.4)	–	1.13 ± 0.02 (1.28 ± 0.01)	28.2 ± 0.3 (29.1) [29.03]
Mn (μg/g)	15.9 ± 2.0 (14.8)	–	–	–
Mo (μg/g)	1.71 ± 0.28 (2.27)	1.12 ± 0.05 (–)	1.58 ± 0.15 (1.46)	–
Na (mg/g)	2.30 ± 0.05 (2.37)	0.13 ± 0.01 (0.160)	0.014 ± 0.002 (0.007)	17.0 ± 0.2 (18.2) [18.2]
P (mg/g)	4.83 ± 0.34 (4.48)	1.28 ± 0.03 (1.30)	2.63 ± 0.17 (–)	5.63 ± 0.91 (–) [5.18]
Rb (μg/g)	55.5 ± 2.6 (53.4)	4.53 ± 0.16 (4.84)	6.24 ± 0.04 (6.14)	12.9 ± 0.1 (13.1) [12.59]
Sb (ng/g)	62 ± 13 (68)	35 ± 5 (10)	1 ± 1 (0.5)	12 ± 1 (13.3) [14.0]
Sc (ng/g)	8.0 ± 1.0 (9.48)	10.5 ± 0.9 (40)	2 ± 1 (–)	68 ± 17 (53.3) [56.1]
Se (ng/g)	131 ± 2 (134)	26 ± 2 (25)	381 ± 29 (380)	127 ± 3 (117) [117]
Sr (μg/g)	76.2 ± 9.4 (75.7)	94.6 ± 8.4 (100)	18.1 ± 3.2 (–)	59.8 ± 12.3 (48) [55.6]
Th (ng/g)	9.4 ± 0.7 (10.4)	17.0 ± 2.8 (–)	12 ± 3 (–)	53 ± 10 (48)
Zn (μg/g)	33.3 ± 2.1 (32.3)	32.2 ± 1 (29)	18.0 ± 0.1 (–)	81.8 ± 0.4 (80.3) [82]

Values in parentheses ( ) are literature values (Becker, 1995) whereas in [ ] are from IAEA estimated means (IAEA, 1997).

Table 2
Elemental contents in cereals (flour-form)

Element	Barley	Rice	Maize	Millet (Bajra)	Wheat	Sorghum (Jowar)
	H. vulgare	*O. sativa*	*Z. mays*	*P. pyphoides*	*T. aestivum*	*S. vulgare*
Br (μg/g)	64.2	17.6	36.4	42.5	27.6	40.4
Ca (mg/g)	0.42	0.29	0.50	0.24	0.16	0.21
Cl (mg/g)	2.29	0.52	0.57	1.13	1.42	1.33
Co (ng/g)	77.2	102	42.6	190	79.3	124
Cr (μg/g)	3.59	1.64	2.01	1.88	0.69	1.50
Fe (μg/g)	167	124	46.6	178	43.8	194
K (mg/g)	3.84	0.64	2.39	2.20	1.90	3.68
Hg (ng/g)	55	13	121	96	20	77
Mn (μg/g)	82.8	28.1	3.35	32.7	29.7	23.1
Na (mg/g)	390	37	48	37	362	47
P (mg/g)	4.26	2.37	3.54	3.20	3.29	3.78
Rb (μg/g)	15.2	3.18	3.37	3.60	2.67	4.31
Sc (ng/g)	72	36	18	56	6	41
Se (ng/g)	625	346	149	167	329	175
Sr (μg/g)	43.5	35.7	26.5	46.4	54.3	50.9
Th (ng/g)	114	50	140	47	71	45
Zn (μg/g)	54.1	26.1	14.5	25.2	20.3	24.7

Table 3
Elemental contents in some common vegetables

Element	Potato	Cauli flower	Cabbage	Chaulai leaves	Green peas	Onion	Ghui[a]	Ghol[a]	Pagur[a]
	S. tubirosam	*B. ieraceaapitata*	*B. oieraceacapitata*	*Amaranthus*	*P.sativum*	*A.cipa*			
Br (μg/g)	36.8	49.8	80.1	128	29.8	45.4	198	76.4	257
Ca (mg/g)	0.10	2.95	3.62	18.1	0.21	16.1	10.1	42.1	2.53
Cl (mg/g)	1.37	ND	1.66	3.21	1.64	1.61	ND	ND	ND
Co (ng/g)	178	680	157	218	99.7	332	30	574	536
Cr (μg/g)	1.64	3.59	0.50	1.58	1.45	1.34	0.92	2.48	0.42
Fe (μg/g)	69.7	653	54.4	1060	111	958	545	1440	4410
K (mg/g)	19.8	7.23	11.3	21.1	9.33	6.51	30.4	8.41	10.5
Hg (ng/g)	18	32	39	95	26	97	140	113	80
Mn (μg/g)	6.68	ND	15.7	67.0	28.5	33.1	ND	ND	ND
Na (mg/g)	0.18	0.09	3.76	1.16	0.02	0.56	6.48	4.72	0.58
P (mg/g)	2.37	4.89	3.84	3.87	4.10	10.0	8.94	8.64	2.09
Rb (μg/g)	13.7	21.1	43.8	36.2	12.0	12.0	20.4	6.12	6.13
Sc (ng/g)	12.9	265	11	470	14	253	108	846	2
Se (ng/g)	325	210	38	362	150	180	164	116	352
Sr (μg/g)	45.8	64.1	30.3	119	49.3	131	98.6	152	210
Th (ng/g)	77	496	32	563	20	151	117	637	19
Zn (μg/g)	21.2	16.0	11.1	25.7	37.5	16.	190	19.2	243

[a] These leafy vegetables are particularly used in the countryside of central India.

respectively. Spices and condiments analysed in this study have been classified as seeds and other plant parts and their mean elemental contents are listed in Tables 4 and 5, respectively. Also included in Table 5 are data for the two varieties of betel leaves commonly chewed in the Indian subcontinent after meals.

Indian diet is primarily vegetarian and consists of home-made bread from flours of cereals such as wheat, barley, sorghum, millet and maize depending on the region, climatic conditions, socio-economic status and food habits, boiled rice, curries prepared from pulses and a variety of vegetables (sometime fried in oil), which are heavily spiced. In addition, however, it also consists of milk and seasonal fruits, along with oil used for frying/cooking. Whereas the main dietary constituents remain almost the same throughout India, actual composition may differ widely, depending on geography, sex and socioeconomic status. For example, in northern and western parts of India, diet is primarily wheat bread-based whereas, in southern and eastern parts, it is boiled rice with curries prepared from pulses. Other cereals, such as barley, maize, millet and sorghum, are used occasionally and in the countryside only. A cursory look at the elemental contents in Tables 2–5 shows wide variations in elemental contents. In general, these can be broadly classified as cereals, leafy vegetables and seed spices, including condiments. These are discussed separately in the following lines.

Table 4
Elemental contents in seed spices used in preparation of vegetable curries

Element	Cumin *C. cyminun*	Omum *T. supermum*	Mustaed *B. juncea*	Coriander seeds *C. sativun*	Fenugreek seeds *T. foenum*	Black pepper *P. nigrum*	Cardmom (small) *E. carda momum*	Cardmom (large) *A. sabulatum*	Fennel *F. vulgare*	Red Chilies *A. capsicum*
Br (μg/g)	32.9	29.6	3.31	2.37	3.47	0.06	8.38	26.0	21.0	83.5
Cl (mg/g)	ND	1.64	0.56	4.23	2.02	2.48	1.24	ND	4.04	Ca;12.8
Co (ng/g)	21	28	22	14	16	33	40	28	29	465
Cr (μg/g)	0.23	0.60	0.22	0.33	1.64	ND	0.11	ND	0.12	5.54
Cs (μg/g)	ND	0.92	0.88	0.51	0.72	0.44	0.27	1.83	0.46	ND
Cu (μg/g)	5.64	2.00	7.84	0.10	0.36	ND	ND	ND	2.86	ND
Fe (μg/g)	71.1	127	67.0	102	266	76.0	94.7	96.9	51.8	287
Hg (ng/g)	ND	ND	ND	104	154	ND	ND	ND	ND	WD
K (mg/g)	16.5	26.6	2.63	19.1	10.1	14.6	27.0	24.0	26.6	52.0
Mn (μg/g)	ND	125	40.8	38.9	20.9	73.3	205	ND	107	ND
Mo (μg/g)	ND	ND	ND	0.41	0.23	ND	ND	ND	ND	ND
Na (mg/g)	4.06	0.17	0.34	0.35	0.43	0.30	0.74	0.04	2.90	1230
P (mg/g)	1.17	1.06	2.35	1.03	2.05	1.93	0.56	3.28	2.85	11.7
Rb (μg/g)	12.7	18.0	17.2	4.74	16.9	16.0	16.3	79.9	36.1	24.7
Sb (μg/g)	31	11	17	29	16	26	33	21	22	ND
Sc (ng/g)	16	22	8	9	6	16	29	24	15	86
Se (ng/g)	51	71	106	58	65	116	260	143	89	35
Sr (μg/g)	52.6	54.6	51.6	39.4	41.6	51.8	151	87.8	66.9	110
Th (ng/g)	ND	ND	ND	49	64	ND	ND	ND	ND	167
Zn (μg/g)	56.3	63.3	58.7	33.6	46.5	37.1	104	41.1	42.2	105

ND, not detected, detection limits for Cs, Hg, Th and Mo being 0.2 μg/g, 50 ng/g, 20 ng/g and 0.1 μg/g, respectively.

Table 5
Elemental contents in some spices and condiments (flavouring agents)

Element	Turmeric *C. longa*	Cinnamon *C. zeylanicum*	Bay leaves *C. tamala*	Cloves *S. arom aticum*	Coriander leaves *S. sativum*	Ginger *S. officinale*	Garlic *A. sativum*	Betel Kapuri *P. betel*	Leaves bangla	Sweet neem *A. Indica*
Br (μg/g)	12.8	3.54	4.53	4.50	9.71	6.44	1.77	72.3	76.5	64.1
Cl (mg/g)	ND	6.03	0.78	2.80	7.10	0.99	1.34	ND	ND	5.43
Co (ng/g)	20	93	8	12	31	16	17	62	82	740
Cr (μg/g)	0.22	9.14	ND	0.24	1.15	0.46	0.70	0.64	1.05	2.27
Cs (μg/g)	1.19	1.77	0.80	0.44	ND	ND	ND	ND	ND	ND
Cu (μg/g)	0.12	ND	0.20	2.16	1.92	<1	<1	ND	ND	ND
Fe (μg/g)	45.0	2390	19.6	17.1	140	71.8	1380	177	513	1920
Hg (ng/g)	ND	ND	ND	ND	360	198	91	1290	1970	133
K (mg/g)	29.6	33.8	7.13	13.9	53.2	24.0	16.0	24.36	57.2	22.6
Mn (μg/g)	ND	83	632	1060	115	675	8.9	ND	ND	86.9
Mo (μg/g)	ND	ND	ND	ND	0.64	3.41	1.79	ND	ND	ND
Na (mg/g)	0.25	1.81	0.73	2.68	2.01	0.80	0.71	7.09	3.82	1.02
P (mg/g)	0.99	2.39	0.45	0.61	2.01	5.73	2.11	2.97	2.63	8.1
Rb (μg/g)	10.6	11.5	14.8	5.92	25.6	10.9	4.19	49.7	9.14	15.6
Sb (μg/g)	11	29	194	55	42	117	22	35	41	ND
Sc (ng/g)	12	58	6	12	12	6	8	8	9	730
Se (ng/g)	500	68	12	110	129	60	127	262	156	670
Sr (μg/g)	157	75.0	50.5	44.8	3.4	1.6	2.9	139	94.6	147
Th (ng/g)	ND	ND	ND	ND	64	113	64	86	89	710
Zn (μg/g)	101	104	32.7	30.3	201	49.8	78.6	41.7	53.4	89.4

ND, not detected, detection limits for Cs, Hg, Th and Mo being 0.2 μg/g, 50 ng/g, 20 ng/g and 0.1 μg/g respectively.

3.2. Cereals

Table 2 shows that no single cereal is enriched in all the essential elements. In general, wide variation is observed for all the elements in six cereals (as shown in bar plots in Fig. 1), which are mostly carbohydrate-rich. Liu et al. (1991) have proposed *rice enrichment factor* in the Taiwanese diet to account for trace element intake. Iron varies in a wide range of 43.8–194 μg/g, with the highest concentration in sorghum (194 μg/g) but barley, used mostly in the countryside, has highest contents of Na, K, Cr, Mn, P, Se and Zn. It is interesting to note that most commonly consumed cereals, wheat and rice flours do not contain any of the nutrient elements in significant amounts. However, maize and sorghum are enriched in Ca (0.50 mg/g) and Fe (0.194 mg/g),

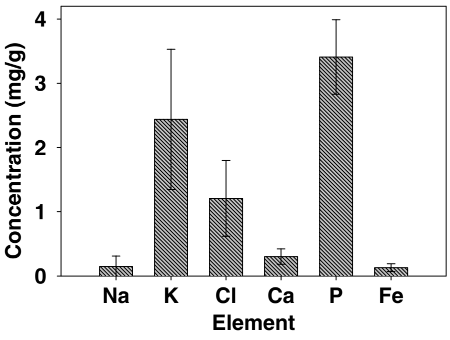

Fig. 1. Variation of mean elemental concentrations in six cereals.

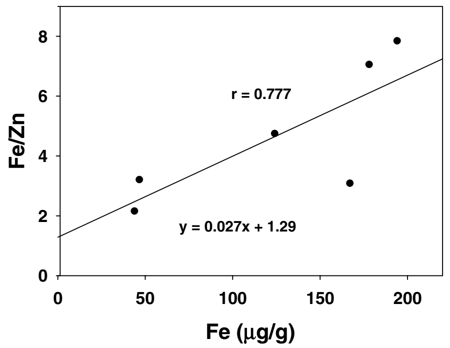

Fig. 2. Correlation of Fe vs. Fe/Zn in six cereals.

respectively, compared to other cereals analyzed in this study. In general, K content is higher than Na, Ca and P in plant leaves (Garg et al., 2005) but in the present study of cereals P, content is found higher than K and Ca. However, the P/K ratio is almost constant in a close range of 1.03–1.73 except in rice where it is highest (3.70). On the other hand, millet, widely used in the western countryside, has only average amounts of most elements. K/Na ratio varies in a wide range of 5.25–78.3, being highest for sorghum. A linear correlation of Fe concentration vs. Fe/Zn ($r = 0.777$) is illustrated in Fig. 2, suggesting increasing Fe with decreasing Zn content (Prasad, 1993). Se is well known for its anticarcinogenic action (Schrauzer, 1992) and its deficiency may be a cause of concern, as reported for Canadian (Morris et al., 2001) and Turkish (Aras, Nazli, Zhang, & Chatt, 2001) population groups. Giray and Hincal (2004) have reported DDI of Se in Turkey to be 44 μg/day compared to the RDA of 55 μg/day and correlated with a possible link with oxidative DNA damage. Earlier we have observed correlation of Se dietary intake with the occurrence of breast cancer (Singh & Garg, 1997, 1998). Incidentally, maize flour has significantly higher contents of Hg (121 ng/g), and Th (140 ng/g), which are environmental contaminants. Our values for some essential elements (Ca, Fe and P) are comparable with those reported in a compilation from the National Institute of Nutrition, India (Gopalan et al., 1999).

3.3. Vegetables

India is a tropical country and several leafy and other vegetables are available in different parts throughout the year. Some of the vegetables, such as potato, onion, cabbage, cauliflower and green peas, are well known in other parts of world as well. However, other leafy vegetables, such as *chaulai*, *ghui*, *poi*, *pagur* and *sweat neem* (curry leaves), are known mostly in the countryside of central and southern parts of India. It is a common belief that most leafy vegetables are, in general, especially enriched in Fe as is our own observation (0.545–4.41 mg/g). Table 3 shows that most of the green leafy vegetables, especially *chaulai* are particularly rich in Ca, Na, K, P, Fe, Co, Se and Zn, all essential nutrients. On the other hand, carbohydrate-rich potato and, to some extent, cabbage, the most commonly-consumed vegetables throughout the world, contain below average amounts of most nutrient elements. Cauliflower, another common vegetable, is enriched in Co, Rb and Se. Onion is particularly rich in Ca, Co, Fe and P, though its Hg, Sr and Th contents are also high. Another common vegetable, green pea is not enriched in any of nutrient elements. Curry leaves (sweet neem) and coriander leaves are not actually vegetables but their tender green leaves are commonly spread over dishes to add flavour in most curry preparations. Curry leaves are particularly enriched in Co, Fe, Mn, P, Se and Zn whereas coriander leaves are rich sources of Mn, Se and Zn. Some electrolytic and structural elemental ratios, such as K/Na, K/P and Ca/P, vary in a wide range of 1.78–18.2, 0.97– 5.42 and 0.94–4.87, respectively. Se has found importance in glutathione peroxidase (GPx) mechanism (Schrauzer, 1992) and gained importance in the prevention of breast cancer (Singh & Garg, 1998). In a recent review by Kohrle et al. (2000), it has been suggested that selenocysteine acts as an essential component of the catalytic cycle where it may be involved in redox reactions. Also, Se binds with methionine in bacteria, yeast and higher plants. However, its probability of being found in inorganic selenate form is much less. Thus, *chaulai* leaves could probably be a good source of Se in bioavailabe form, which could be easily digested by the body system. It may be noted that Br, Hg, Sr and Th, all environmental contaminants, are also present in significant amounts in several vegetables. These could have originated from industrial emissions, which must have been deposited on our samples while growing in the fields. Mannan et al. (1992) and Bro et al. (1990) have also found significant amounts of several toxic elements, such as Hg, Pb, Cd, Br and Sb in Pakistani and Danish diets and attributed these to industrial emissions. Similarly Yukawa et al. (1997) have found Th and U in Japanese diets where the internal dose was estimated along with intake of essential nutrients in homogenized diet samples. In view of this, the Th content observed in our samples can be justified though we could not determine U.

3.4. Elemental content of seed spices

Unlike the western world, vegetable preparations in India are mostly consumed in fried form, with a lot of oil (mustard, groundnut and coconut) and spices. Most spices, except garlic and ginger, are in seed form. Out of various spices, cumin, omum or mustard seeds are used in primary frying of vegetables in oil. Coriander and fenugreek seeds form the base material in a typical spice mixture. Red chilies and cardamom (large) may be used, either as such or in powder form during cooking. Cardamom (small) and fennel seeds are mostly used as flavouring agents or mouth fresheners due to their rich aroma. It is observed that two varieties of cardamom (large and small) exhibit wide variation in elemental contents, e.g. K/P ratio for small cardamom is large (48.2) compared to a small value (7.32) for larger variety whereas K/Na for smaller variety is small (36.5) compared to a large value (600) for the larger variety. On the other hand, mustard and fenugreek seeds show almost comparable and much smaller K/P ~4.8. Most spices, along with oil, are also used for pickling of mango, lemon and other fruits and seasonal vegetables.

Each of these spices seems to be enriched in one or the other, of the essential/nutrient elements. Table 4 shows that cardamom (both types, small as well as large) is enriched in several essential elements (e.g. Co, Mn, Se and Zn) though omum seeds are also rich in Br, Fe and Se. We have attempted to correlate Fe/Zn and Fe/Co vs. Fe contents in nine seed spices, as shown in Figs. 3 and 4 and observed linear correlations, with $r = 0.916$ and 0.951, respectively. Mustard seeds are rich in Cu (7.84 μg/g) while fenugreek seeds are noticeably rich in Fe (266 μg/g) and Cr (1.64 μg/g). It may be mentioned that fenugreek seeds have hypoglycaemic and hypocholesterolaemic properties in experimental animals and in diabetic patients (Ribes, Sauvaire, Costa, Baccou, & Loubatieres Mariani, 1986; Sharma, Raghuram, & Rao, 1991). These are often recommended for lactating mothers and diabetics (Sharma, 1993). Coriander seeds are not rich in any particular element though, in general, these have above average amounts of Fe and Cl. Though black pepper is widely used as a hot spice, its trace element contents are below average.

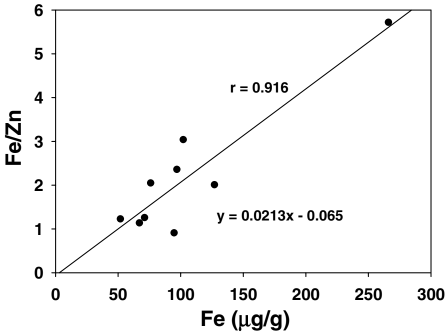

Fig. 3. Correlation of Fe vs. Fe/Zn in nine seed spices.

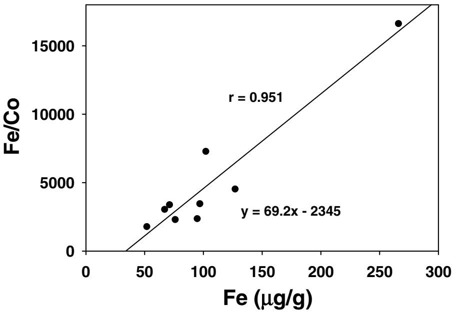

Fig. 4. Correlation of Fe vs. Fe/Co in nine seed spices.

A comparison of our elemental data with an earlier study of six spices (five of them cumin, fenugreek, mustard, black pepper and turmeric being common with this study) by Ila and Jagam (1980) shows that most elemental concentrations are in comparable ranges. Remarkably, our values for Br, Cl, K and Zn match well with those of Ila and Jagam (1980). However, Fe and Co concentrations are somewhat lower in our samples, which may be attributed to local soil conditions where these spices are grown. Miyamoto et al. (2000) have also reported 26 elements in various food spices and pulses of Indian and Pakistani origin. It has been shown that high elemental correlations exist in some sets of elements, reflecting characteristics of chemical nature of elements and/or their physiological functions. Some of these are rich in essential as well as toxic elements. By and large our elemental concentrations in various spices are comparable with those observed by other workers (Ila & Jagam, 1980; Miyamoto et al., 2000).

3.5. Elemental contents in other spices and condiments

Turmeric (root) is particularly rich in Se (500 ng/g) and is perhaps a good source/supplement for this element, which otherwise is an important nutrient (Koller & Exon, 1986; O' Dell & Sunde, 1997) and medicinally important (Kohrle et al., 2000). Cinnamon (bark of a tree) is found to be rich in Fe (2.39 mg/g), Co (93 ng/g), Cr (9.14 μg/g) and to some extent in Zn (104 μg/g). While the importance of iron need not be emphasized, high Co content may be indicative of its role in vitamin B_{12} metabolism and Cr may be correlated with glucose tolerance factor (Anderson, 1989). In addition, cloves (dried fruit) are especially rich in Mn (1080 μg/g) and coriander leaves in Zn (201 μg/g). Zinc is an important constituent of several enzymes and plays vital role in clinical, biochemical and immunological effects (Fell & Lyon, 1994; O' Dell & Sunde, 1997; Sirover & Loeh, 1976). Interestingly, tender coriander leaves, spread over curry preparations during season of its availability, are also enriched in several nutrient elements compared to seeds (Cu, Fe, Mn, P, Se and Zn). Also, fresh green coriander leaves are crushed with other spices and made into a paste, which is often used, as a dressing on potatoes or typical Indian ketchup. Incidentally, bay leaves exhibit a high content of Sb (194 ng/g), which is an environmental contaminant (Merian, 1991). The two types of betel leaves (called *pan* which, along with catchu and betel nuts, are often chewed after meals) have higher contents of Br and Hg, which are known carcinogens (Sirover & Loeh, 1976). This may be essentially due to pollution of local soil. However, Sarmani et al. (1993) have reported dietary intake of up to 30 μg/d of Hg for fish-consuming populations in Malaysia. Perhaps, at such low level, or as that found in spices, species, consumption of Hg may not be so harmful (WHO, 1989).

3.6. Contribution of spices to dietary intake

According to recommendations of the Indian Council of Medical Research (ICMR, 1987), a balanced vegetarian diet of an Indian adult consists of approximately 400 g cereals, 50 g pulses and 150 g vegetables. Besides, an average Indian also consumes approximately 25 g spices everyday as part of vegetable or other preparations (Singh & Garg, 1997). A typical spice mixture may have the composition: 6–8 g coriander seeds, 2–4 g cumin/omum/mustard seeds, 2 g each of turmeric, red chilies and fenugreek seeds, and 1 g fennel and other spices, all in powder form. Thus, daily dietary intake (DD1) of essential trace elements, by way of consumption of spices, may be broadly estimated as given in Table 6. It is observed that Cr, Fe, Mn and Zn are contributed to the extent of 7.5% by various spices, whereas Cu, P and Se are contributed at less than 5% of the total dietary intake (ICMR, 1987). It may be mentioned that most middle and higher income groups use these spices. However, for lower income groups, the daily dietary intake of some of these elements may be considerably low. Since India is a vast country, with widely varying food habits and income groups, the exact role of spices in the daily dietary intake of nutrient elements through an Indian vegetarian diet needs further investigation. Many of these spices are used in Asia, Africa, Middle East, South America, China, Mexico and other countries, as well (Miyamoto et al., 2000). Ila and Jagam (1980) have suggested that trace element contents in spices may be correlated with their taste. It may be concluded that spices, not only add flavour to food, but also enhance its nutritive value by providing several essential nutrient elements in bioavailable form (Pruthi, 1999; Risch & Ho, 1997; WHO, 1985).

Table 6
Elemental contents in some common vegetables and spices

Element	DDI (total)	DDI (spices)	% of DDI	Recommended Dietary Allowances (RDA)[a]
Cr (μg/d)	250	17.5	7.0	50–200
Cu (mg/d)	4.0	0.02	0.5	1.5–3
Fe (mg/d)	28	2.1	7.5	10–18
Mn (mg/d)	8.1	0.06	7.5	2–5
P (g/d)	1.2	0.04	3.5	1.0
Se (μg/g)	45	1.8	4.0	50–100
Zn (mg/g)	12	0.86	7.2	15

Contribution of the spices to the total DDI (daily dietary intake) of trace elements and comparison with RDA.

[a] Food and Nutrition Board (1989).

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