



Heavy metals in vegetables collected from production and market sites of a tropical urban area of India

Rajesh Kumar Sharma^a, Madhoolika Agrawal^{a,*}, Fiona M Marshall^b

^aEcology Research Laboratory, Department of Botany, Banaras Hindu University, Varanasi 221 005, India

^bSPRU, The Freeman Centre, University of Sussex, Brighton, BN1 9QE, United Kingdom

ARTICLE INFO

Article history:

Received 13 January 2007

Accepted 15 December 2008

Keywords:

Vegetables
Heavy metals
PFA
EU
Daily intake

ABSTRACT

Vegetables (*Beta vulgaris* L., *Abelmoschus esculentus* L. and *Brassica oleracea* L.) from the production and market sites of India were tested for Cu, Cd, Zn and Pb. At market sites, the mean concentration of Cu in cauliflower, and of Zn and Cd in both palak and cauliflower had exceeded the PFA standard. Zn at the production sites also exceeded the PFA standard in cauliflower. Cd concentration in vegetables tested from both production and market sites was many folds higher than the EU standard. In contrast, Pb in vegetables tested from both production and market sites was below the PFA limit, but was considerably higher than the current EU and WHO standards. Heavy metals accumulation in vegetables tested are higher at market sites than those at the crop production sites. The contributions of these vegetables to dietary intake of Cu, Zn, Cd and Pb were 13%, 1%, 47% and 9% of provisional tolerable daily intake, respectively. The study concludes that the transportation and marketing systems of vegetables play a significant role in elevating the contaminant levels of heavy metals which may pose a threat to the quality of the vegetables with consequences for the health of the consumers of locally produced foodstuffs.

© 2008 Elsevier Ltd. All rights reserved.

1. Introduction

Heavy metal contamination of vegetables cannot be underestimated as these foodstuffs are important components of human diet. Vegetables are rich sources of vitamins, minerals, and fibers, and also have beneficial antioxidative effects. However, intake of heavy metal-contaminated vegetables may pose a risk to the human health. Heavy metal contamination of the food items is one of the most important aspects of food quality assurance (Marshall, 2004; Radwan and Salama, 2006; Wang et al., 2005; Khan et al., 2008). International and national regulations on food quality have lowered the maximum permissible levels of toxic metals in food items due to an increased awareness of the risk these metals pose to food chain contamination (Radwan and Salama, 2006).

Rapid and unorganized urban and industrial developments have contributed to the elevated levels of heavy metals in the urban environment of developing countries such as China (Wong et al., 2003) and India (Tripathi et al., 1997; Khillare et al., 2004; Marshall, 2004; Sharma et al., 2008a,b). Heavy metals are non-biodegradable and persistent environmental contaminants, which may be deposited on the surfaces and then absorbed into the tissues of vegetables. Plants take up heavy metals by absorbing them from deposits on the parts of the plants exposed to the air from polluted

environments as well as from contaminated soils (Khairiah et al., 2004; Jassir et al., 2005; Kachenko and Singh, 2006; Singh and Kumar, 2006; Sharma et al., 2008a,b). A number of studies have shown heavy metals as important contaminants of the vegetables (Singh et al., 2004; Marshall, 2004; Sinha et al., 2006; Singh and Kumar, 2006; Sharma et al., 2006, 2007, 2008a,b). Heavy metal contamination of vegetables may also occur due to irrigation with contaminated water (Singh et al., 2004; Sharma et al., 2006, 2007; Singh and Kumar, 2006).

Emissions of heavy metals from the industries and vehicles may be deposited on the vegetable surfaces during their production, transport and marketing. Jassir et al. (2005) have reported elevated levels of heavy metals in vegetables sold in the markets at Riyadh city in Saudi Arabia due to atmospheric deposition. Recently, Sharma et al. (2008a,b) have reported that atmospheric deposition can significantly elevate the levels of heavy metals contamination in vegetables commonly sold in the markets of Varanasi, India.

Prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases (WHO, 1992; Jarup, 2003). Some heavy metals such as Cu, Zn, Mn, Co and Mo act as micronutrients for the growth of animals and human beings when present in trace quantities, whereas others such as Cd, As, and Cr act as carcinogens (Feig et al., 1994; Trichopoulos, 1997), and Hg and Pb

* Corresponding author. Tel.: +91 542 2368156; fax: +91 542 2368174.

E-mail address: madhoo58@yahoo.com (M. Agrawal).

are associated with the development of abnormalities in children (Gibbes and Chen, 1989; Pitot and Dragan, 1996). Hartwig (1998) and Saplakoglu and Iscan (1997) have reported that long-term intake of Cd caused renal, prostate and ovarian cancers.

Monitoring and assessment of heavy metals concentrations in the vegetables from the market sites have been carried out in some developed (Jorhem and Sundstroem, 1993; Milacic and Kralj, 2003), and developing countries (Parveen et al., 2003; Jassir et al., 2005; Radwan and Salama, 2006), but limited published data are available on heavy metals concentrations in the vegetables from the market sites of India (Agrawal, 2003; Marshall, 2004; Tripathi et al., 1997; Sharma et al., 2008a,b). Comparison of heavy metal contamination due to atmospheric deposition at production and market sites is, however, not available in the literature till date.

The present study presents data on heavy metals (Cu, Zn, Cd and Pb) concentrations in some key Indian vegetables such as palak (*Beta vulgaris* L. var *bhenghalensis* cv allgreen H1 Family: Chenopodiaceae), lady's finger (*Abelmoschus esculentus* L. Family: Malvaceae) and cauliflower (*Brassica oleracea* var *botrytis* Family: Brassicaceae) grown locally in suburban and rural areas and sold in urban open markets. It was hypothesized that atmospheric depositions in urban areas may increase the levels of heavy metals during transport and marketing, leading to significant contamination of vegetables at the market sites than that at the production sites. Observed concentrations of Cu, Zn, Cd and Pb in the vegetables were also compared with Prevention of Food adulteration (PFA) act (30, 50, 1.5 and 2.5 µg/g, respectively; Awasthi, 2000) and European Union (EU) (0.1 and 0.3 µg/g, respectively, for Cd and Pb) standards of food contamination. The contribution of the heavy metal contamination through dietary intake of the vegetables tested is also assessed based on the average daily consumption of the vegetables.

2. Materials and methods

2.1. Study area

The present study was carried out in (market sites; in between 0 and 4 km from the city centre) and around (production sites; in between 4 and 15 km from the city centre) Varanasi, India, during September 2004 to March 2005. Fresh samples of three key Indian vegetables (palak, lady's finger and cauliflower) were collected simultaneously from the production and market sites. A reconnaissance survey was conducted to identify locally grown vegetables and their marketing area in the city. Based on the survey, it was found that palak, cauliflower and lady's finger are grown at a large scale at various production sites around Varanasi city (Fig. 1; Table 1). Eleven, eight and four production sites are selected for cauliflower, lady's finger and palak, respectively, based on their production areas and frequency of marketing. Some of the production sites are either located in the vicinity of brick kiln industries (sites 7, 10, 12, 14, 17, 25, 24 and 28) or located close to the national highway (sites 17 and 18). These vegetables being perishable are sold in open markets of the city. Five market sites were identified based on the supply of these vegetables from their production sites, the traffic load, industrial activities, and residential and commercial areas (Fig. 1; Table 1). The market sites such as 1, 2, 4 and 5 are located in a zone dominated by heavy traffic on a narrow road (1000–1500 vehicles run per hour during 10.00 am to 7.00 pm) and dense population. Site 3 is located in an industrial area having a large number of small-scale industries (manufacturing of tires, fabric paintings, cements, small-scale workshops, etc.) and heavy traffic on a narrow road. The site name and position no. of the production and market sites are listed in Table 1, and their relative positions are shown in Fig. 1.

2.2. Sampling and analysis

Samples of lady's finger (September and October), cauliflower (November to February) and palak (November, January and March) were collected (40–45, 50–60 and 70–80 days after seed sowing of palak, cauliflower and lady's finger, respectively) during their growing seasons from three fields of each production site and simultaneously from three outlets of each market site (2 kg each for palak and lady's finger, and one intact inflorescence head of cauliflower), respectively, in pre-distilled water-washed polyethylene bags. The samples of each vegetable were of the same size and age group. The samples were immediately brought back to the laboratory, and uneatable portions of the vegetables were removed and the edible por-

tion was chopped into small pieces. The samples were then oven dried at 80 °C and then crushed using a stainless steel blender and passed through a 2 mm sieve. The resulting fine powder was kept at room temperature before analysis. Soil samples were collected from three fields at each production site by digging out a monolith of 10 cm × 10 cm × 20 cm size. The soil samples were brought back to the laboratory, air dried, crushed and passed through a 2 mm mesh size sieve and were stored at room temperature before analyses.

Samples of both soils and vegetables (1.00 ± 0.001 g each) were placed into 100 ml beakers separately, to which 15 ml of tri-acid mixture (70% high purity HNO₃, 65% HClO₄ and 70% H₂SO₄ in 5:1:1 ratio) was added. The mixture was then digested at 80 °C till the solution became transparent (Allen et al., 1986). The resulting solution was filtered and diluted to 50 ml using deionized water and was analyzed for concentrations of Cu, Zn, Cd and Pb using an atomic absorption spectrophotometer (Model 2380, Perkin-Elmer, Norwalk, CT, USA). The measurements were made using a hollow cathode lamp of Cu, Zn, Cd and Pb at wavelengths of 324.8, 213.9, 288.8 and 213.3 nm, respectively. The slit width was adjusted for all the heavy metals at 0.7 nm. The values of the detection limits for Cu, Zn, Cd and Pb were 0.001, 0.0008, 0.0005 and 0.01 µg/ml, respectively. Standard solutions were frequently run to check the sensitivity of the instrument.

2.3. Health risk assessment and daily intake of heavy metals through vegetables

An assessment of the health risk posed in human beings by the consumption of contaminated vegetables was made by comparing the concentration of the contaminants recorded from both production and market sites with national and international safe limits. The daily intake of heavy metals through the consumption of the vegetables tested was calculated according to the given equation (Cui et al., 2004):

$$\text{Daily intake of heavy metals (}\mu\text{g/day)} = [\text{Daily vegetable consumption} \\ \times \text{vegetable heavy metal concentration}]$$

Daily vegetable consumption was obtained through a formal survey conducted in the study area. An interview of 25 persons of 30–50 years age group and of 60–70 kg body weight was conducted at each market site regarding their daily consumption rate of vegetables tested. Each person represents a household having ≥5 individuals, and thus a total of 125 persons were effectively interviewed. An average consumption rate of each vegetable per person per day was calculated. The percent contributions to dietary intake of heavy metals by the urban population through the consumption of the vegetables tested in this study were calculated by dividing the daily consumption rates of the heavy metals with the values of provisional tolerable daily intake (Joint FAO/WHO Expert Committee on Food Additives, 1999).

2.4. Quality assurance

Appropriate quality assurance procedures and precautions were taken to ensure the reliability of the results. Samples were carefully handled to avoid contamination. Glasswares were properly cleaned, and reagents were of analytical grades. Deionized water was used throughout the study. Reagent blank determinations were used to correct the instrument readings. For validation of the analytical procedure, repeated analysis of the samples against internationally certified plant standard reference material (SRM-1570) of National Institute of Standard and Technology were used, and the results were found within ±2% of the certified values.

2.5. Statistical analysis

The recorded data were subjected to two-way analysis of variance (ANOVA) to assess the influence of different variables on the concentrations of heavy metals in the vegetables tested. ANOVA for each vegetable was performed separately using variables such as months and sites, as numbers of sites and month for each vegetable tested varied. All the statistical analyses were computed with SPSS software version 12.

3. Results and discussion

3.1. Levels of heavy metals in soil

The concentrations of heavy metals (µg/g) in agricultural soil collected from different crop production sites ranged from a minimum of 9.8 to a maximum of 19.3 for Cu, 83–133 for Zn, 0.6–2.3 for Cd and 2.9–10.3 for Pb (Table 2). The upper limits of Cu, Zn and Cd in the soil during the present study were higher than the values reported for uncontaminated soil (Cu; 15, Zn; 100 and Cd; 1 µg/g), but were lower for Pb (50 µg/g) (Temmerman et al., 1984). The crop production sites which were in the vicinity of brick kilns or

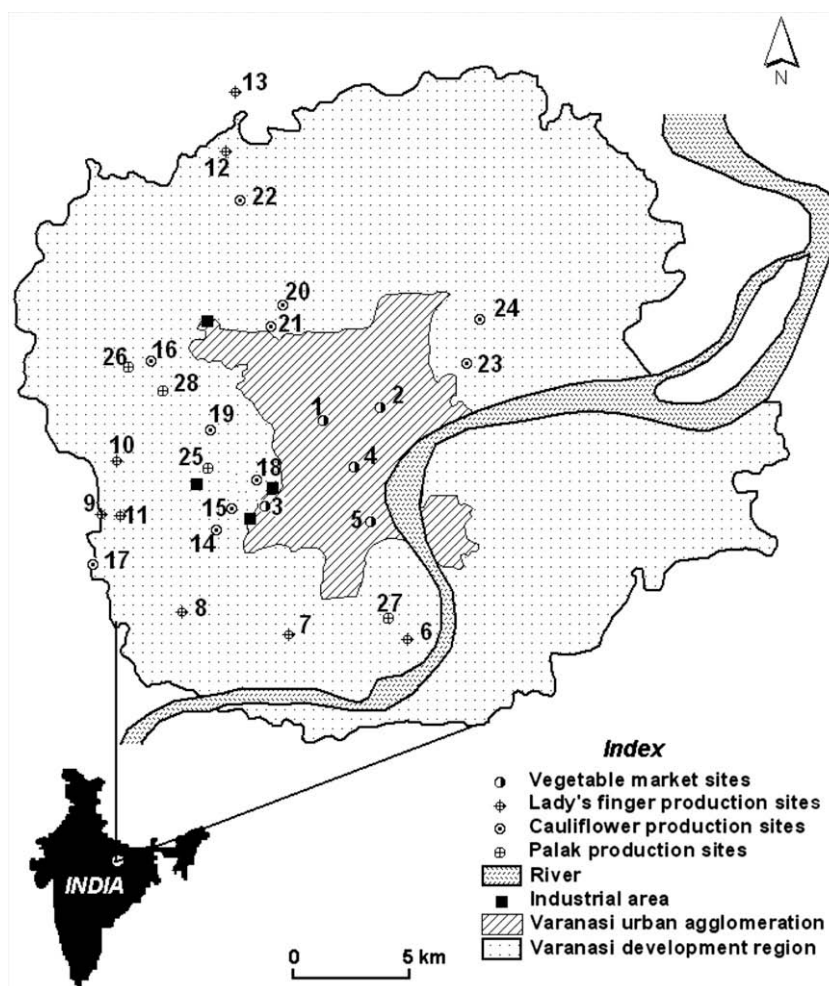


Fig. 1. Map of Varanasi showing the relative positions of different production and market sites.

Table 1

List of both market and production sites (names and no.) used to label Fig. 1 in the present study.

Market sites ^a		Production sites					
		Lady's finger		Cauliflower		Palak	
Sites name	No.	Sites name	No.	Sites name	No.	Sites name	No.
Chanduasatti	1	Banpurwan	6	Nakain	14	Lohta	25
Kabirchaura	2	Akhari	7	Madhwan	15	Daniyapur	26
Chandpur	3	Lathwan	8	Derekhu	16	Tikari	27
Rathyatra	4	Parampur	9	Karnadandi	17	Rohania	28
Lanka	5	Nakainpur	10	Lakhanpur	18		
		Ramraipur	11	Bharathara	19		
		Bhatpurwan	12	Chuppeypur	20		
		Murdi	13	Parmanandpur	21		
				Bhusaula	22		
				Kotwa	23		
				Rustampur	24		

^a Market sites are common for all the test vegetables.

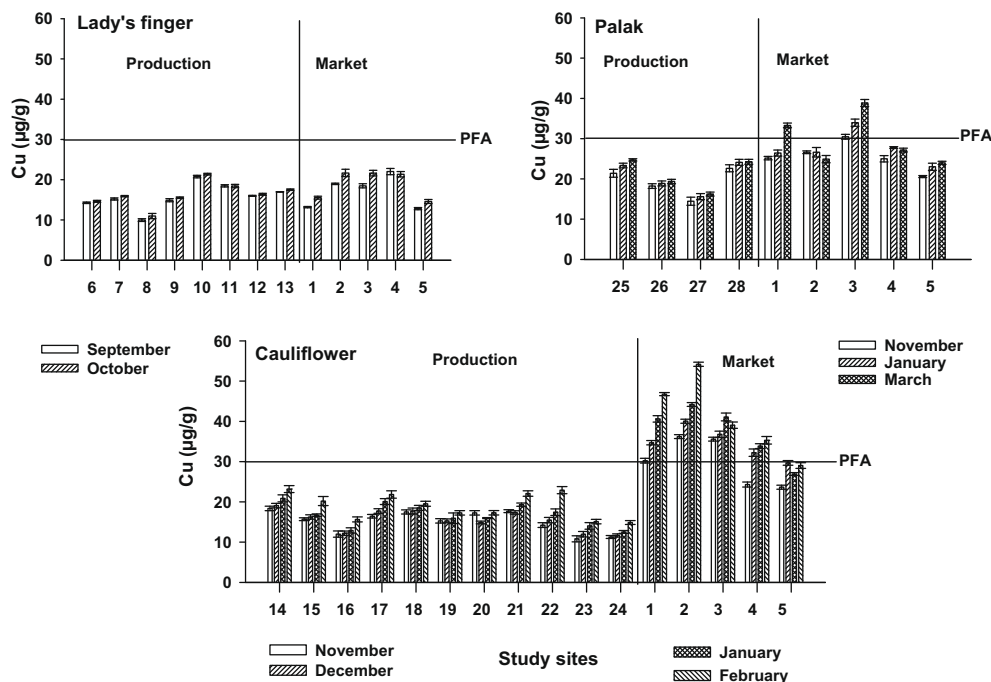
Table 2

Mean, median, minimum and maximum, and standard deviation of heavy metal concentrations in agricultural soil of the production sites of Varanasi ($n = 66$).

Heavy metals	Mean	Median	Minimum	Maximum	Standard deviation
Copper	14.59	14.25	9.80	19.30	2.79
Zinc	109.09	110.00	83.00	133.00	11.41
Cadmium	1.27	1.16	0.60	2.30	0.43
Lead	6.39	5.70	2.90	10.30	2.40

Table 3Heavy metal concentrations ($\mu\text{g/g}$ dry wt) in the test vegetables from production and market sites of Varanasi (mean, minimum and maximum).

Heavy metals	Production sites			Market sites		
	Palak	Lady's finger	Cauliflower	Palak	Lady's finger	Cauliflower
Copper						
Mean	20.27	16.07	16.63	27.59	18.02	35.72
Min	12.80	9.50	9.80	20.10	12.20	21.20
Max	25.60	21.80	24.10	40.10	27.10	56.30
Zinc						
Mean	38.40	34.69	51.52	57.56	45.96	63.63
Min	30.10	29.60	38.60	42.30	32.50	25.20
Max	45.50	39.20	63.30	92.30	66.20	94.30
Cadmium						
Mean	0.98	0.90	1.26	1.96	1.41	2.57
Min	0.40	0.50	0.60	0.80	0.10	0.80
Max	1.50	1.20	2.10	3.80	2.60	4.30
Lead						
Mean	1.00	0.88	1.02	1.44	1.03	1.56
Min	0.70	0.30	0.20	1.10	0.20	0.90
Max	1.40	1.20	1.80	1.90	2.56	2.40

**Fig. 2.** Concentration of Cu in the test vegetables collected from different production and market sites of Varanasi.

near the national highway showed higher concentrations of heavy metals than those having no specific sources of emission. But even the upper limits of heavy metal concentrations were below the upper permissible limits of PFA standards (6, 300, 270 and 500 $\mu\text{g/g}$, respectively, for Cd, Zn, Cu and Pb; Awasthi, 2000). Other studies conducted in Varanasi at sites having long-term uses of wastewater for irrigation showed higher mean concentrations of Cu, Cd and Pb in the soil (20.35, 2.8 and 15.57 $\mu\text{g/g}$, respectively; Sharma et al., 2007) than the concentrations recorded (14.59, 1.27 and 6.39 $\mu\text{g/g}$, respectively) during the present study. However, the mean concentration of Zn recorded during the present study (109.09 $\mu\text{g/g}$) was higher than the value (43.56 $\mu\text{g/g}$) reported by Sharma et al. (2007). The higher concentration of Zn in the soil may also be ascribed to the use of Zn in fertilizers and metal-based pesticides apart from ash from brick kilns. Singh et al. (2004) have reported 3–15-fold higher concentration of Cu, Zn, Cd and Pb in wastewater-irrigated areas of Kanpur and Varanasi

as compared to those reported in the present study. By comparison with our study, Krishna and Govil (2007) also reported higher concentrations of Cu (137.7 $\mu\text{g/g}$) and Zn (139 $\mu\text{g/g}$) in the soil collected from industrial areas of Surat, western India.

3.2. Levels of heavy metals in vegetables

The heavy metal concentrations varied between the production sites and vegetables. The levels of Zn in lady's finger, palak and cauliflower, respectively, varied from 29.6 to 39.2, 30.1 to 45.5, and 38.6 to 63.3 $\mu\text{g/g}$, of Cu varied from 9.5 to 21.8, 12.8 to 25.6 and 9.8 to 24.1 $\mu\text{g/g}$, of Cd varied from 0.5 to 1.2, 0.4 to 1.5, and 0.6 to 2.1 $\mu\text{g/g}$, and of Pb varied from 0.3 to 1.2, 0.7 to 1.4 and 0.2 to 1.8 $\mu\text{g/g}$, respectively, at the production sites (Table 3).

Within the production sites, the mean concentrations of the heavy metals in lady's finger, cauliflower and palak, respectively, were recorded maximum at sites 10, 14 and 25 for Cu (Fig. 2), at

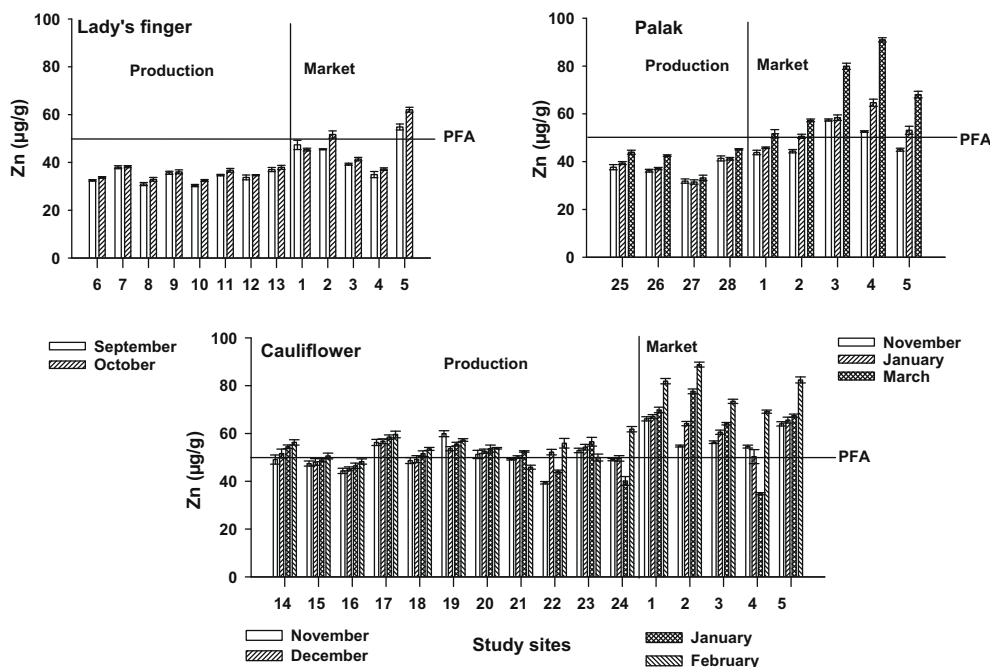


Fig. 3. Concentration of Zn in the test vegetables collected from different production and market sites of Varanasi.

sites 7, 24 and 28 for Zn, at sites 10, 17 and 28 for Cd and at sites 12, 17 and 28 for Pb (Figs. 2–5). The higher concentrations of heavy metals in the vegetables tested at sites 10, 12, 17 and 28 may be ascribed to their location in the vicinity of brick kiln industries or proximity to national highway.

The mean concentration of Zn was highest in all the vegetables tested followed by that of Cu, Cd and Pb at the production sites as well as at the market sites (Table 3). The mean concentration of Cu (µg/g) was recorded minimum in lady's finger (18.02) and maximum in cauliflower (35.72) at the market sites (Table 3). Zn con-

centrations (µg/g) ranged from 32.5 to 66.2, 42.3 to 92.3 and 25.2 to 94.3, respectively, in lady's finger, palak and cauliflower at market sites (Table 3). The mean concentrations of Cd (µg/g) were 1.41, 1.96 and 2.57, and of Pb were 1.03, 1.44 and 1.56, respectively, in lady's finger, palak, and cauliflower collected from the market sites (Table 3).

Within the market sites, the mean concentration of Cu was recorded maximum at site 2 in lady's finger and cauliflower, and at site 3 in palak (Fig. 2). Mean Zn concentration was recorded maximum at sites 2, 4 and 5, respectively, in cauliflower, palak and

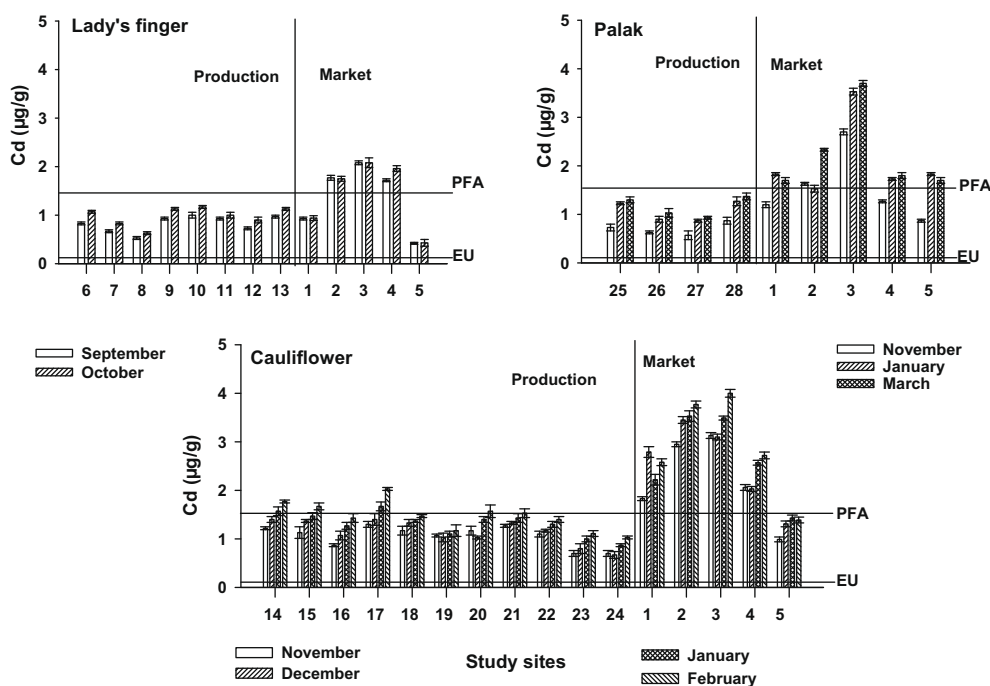


Fig. 4. Concentration of Cd in the test vegetables collected from different production and market sites of Varanasi.

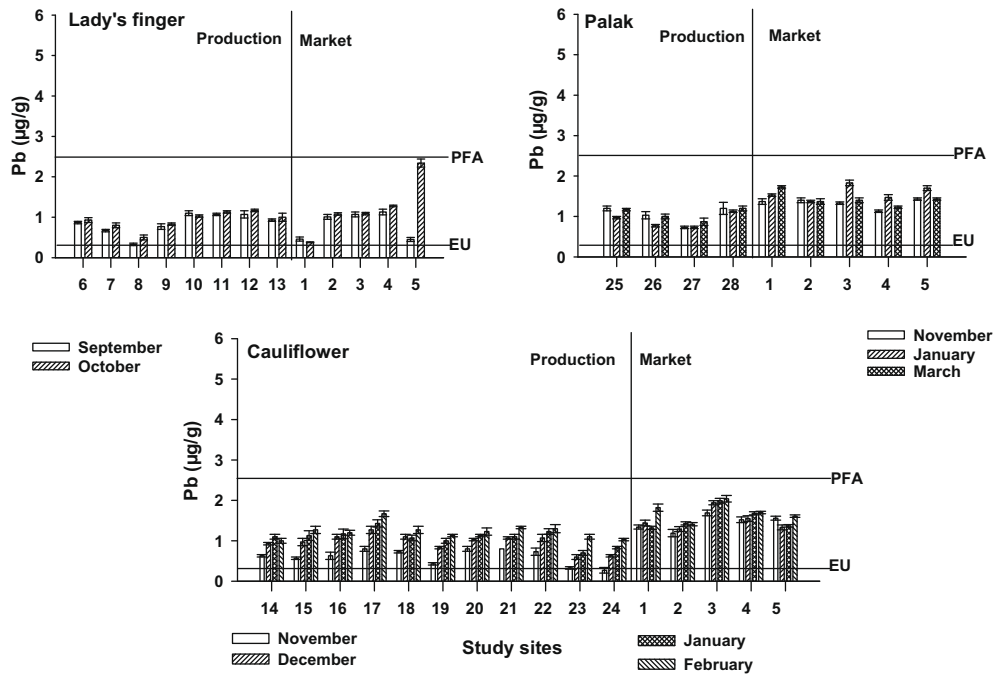


Fig. 5. Concentration of Pb in the test vegetables collected from different production and market sites of Varanasi.

lady's finger (Fig. 3). The mean concentrations of Cd and Pb between the market sites were, respectively, recorded maximum at

sites 3 and 5 in lady's finger, at site 3 in cauliflower, and at sites 1 and 3 in palak (Figs. 4 and 5).

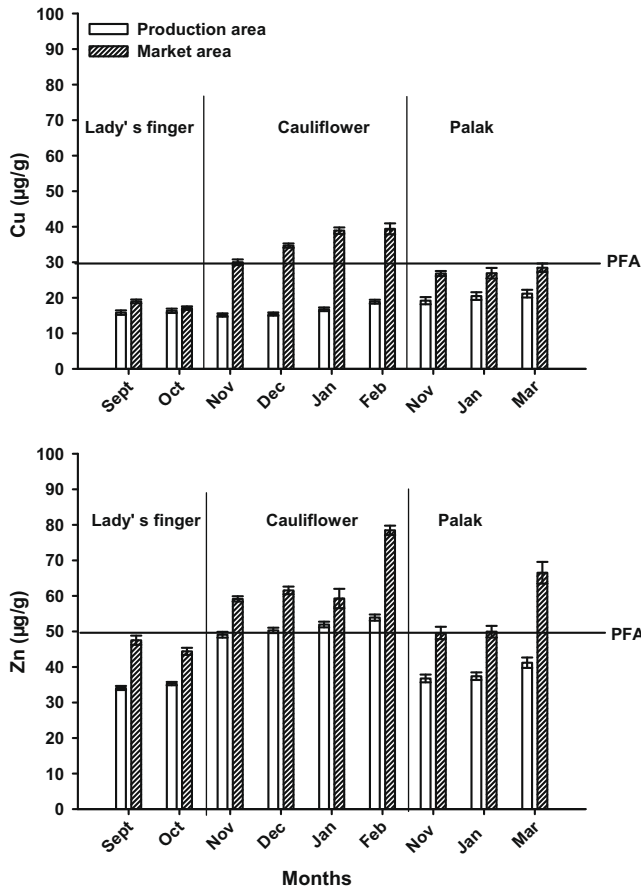


Fig. 6. Monthly mean concentrations of Cu and Zn in the test vegetables from production and market areas of Varanasi.

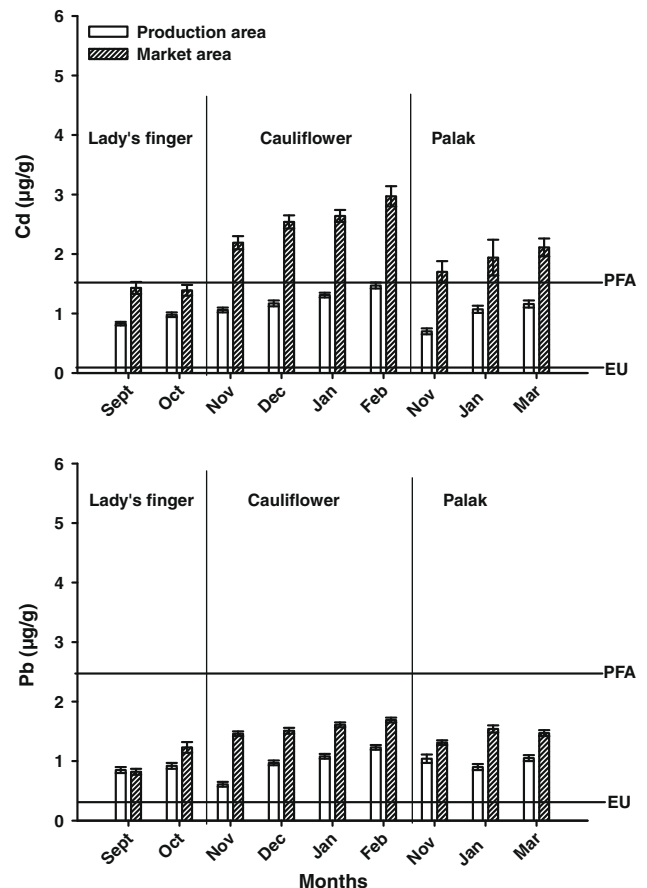


Fig. 7. Monthly mean concentrations of Cd and Pb in the test vegetables from production and market sites of Varanasi.

Table 4

Results of two-way ANOVA for heavy metal concentrations in the test vegetables collected from different production and market sites.

Vegetables/variables	df	Heavy metals			
		Cu	Zn	Cd	Pb
Palak					
Market sites					
Months	2	43.56***	523.70***	264.36***	31.49***
Sites	4	119.09***	242.56***	663.92***	16.64***
Month × site	8	11.65***	35.62***	22.73***	8.57***
Production sites					
Months	2	8.06**	37.59***	59.09***	6.54**
Sites	3	91.45***	94.84***	24.77***	21.12***
Month × site	6	0.50 ^{NS}	2.60 [†]	0.86 ^{NS}	1.02 ^{NS}
Cauliflower					
Market sites					
Months	3	244.01***	432.52***	178.95***	46.14***
Site	4	197.27***	267.51***	750.51***	80.60***
Month × site	12	18.23***	93.32***	13.24***	8.94***
Production sites					
Months	3	71.94***	19.60***	70.54***	183.36***
Sites	10	63.38***	40.33***	48.15***	30.62***
Month × site	30	1.67 [†]	5.10***	1.39 ^{NS}	1.66 [†]
Lady's finger					
Market sites					
Months	1	36.58***	39.68***	2.10 ^{NS}	221.88***
Sites	4	114.73***	236.74***	403.42***	141.10***
Month × site	4	4.14**	12.46***	2.43 [†]	182.11***
Production sites					
Months	1	12.75**	14.76**	57.76***	7.05 [†]
Sites	7	221.60***	28.99***	34.63***	39.05***
Month × site	7	0.53 ^{NS}	0.61 ^{NS}	0.80 ^{NS}	0.86 ^{NS}

df = Degree of freedom. NS = not significant.

*** $p < 0.001$.

** $p < 0.01$.

[†] $p < 0.05$.

The heavy metals concentration in the vegetables tested obtained from the production and market sites were compared. The concentrations of Cu, Zn, Cd and Pb elevated by 36%, 50%, 100% and 44% in palak, by 12%, 32%, 57% and 17% in lady's finger and by 114%, 23%, 103% and 53% in cauliflower, respectively. This result suggests that all the vegetables tested were contaminated maximally by Cd during their transport and marketing in the contaminated environment of Varanasi. The percent increase in the levels of heavy metals was recorded maximum in cauliflower (except for Zn) followed by palak and then lady's finger at all the months of sampling (Figs. 6 and 7). The results further showed that the atmospheric depositions have contributed more than 50% of Zn and Cd to palak, Cd in lady's finger and Cu, Cd and Pb in cauliflower. The monthly variations in the concentrations of heavy metals in vegetables tested at the production sites may be due to their different uptake rate. The levels of all the heavy metal in all the vegetables tested collected from the market sites were higher than those of heavy metals in the respective vegetables collected from the production site (Figs. 6 and 7). This might be due to heavy metal depositions on the vegetables during transport and marketing in more polluted urban environment of Varanasi city. The concen-

trations of heavy metals in different vegetables at the market sites varied with the length of exposure and vegetable types. The concentrations of Cu, Cd and Pb were higher in cauliflower than in palak. Results suggest that cauliflower having a higher exposed area of inflorescence has greater capacity to adsorb heavy metals from the atmosphere.

Results of two-way ANOVA showed that sites and months have significant effects on the levels of heavy metals in vegetables collected from market and production sites (Table 4). This result clearly showed that the location of study sites and growing periods of vegetables influenced the levels of heavy metals in the vegetables. However, month did not show any significant effect on the level of Cd concentration in lady's finger from market sites. This may be ascribed to a small variation in Cd deposition on lady's finger during the sampling months (Table 4). Interaction of month × site did not influence the levels of all the heavy metals in lady's finger, Cd in cauliflower, and Cu, Cd and Pb in palak collected from the production sites (Table 4).

The variations in the concentrations of the heavy metals in vegetables observed during the present study may be ascribed to the physical and chemical nature of the soil of the production sites, absorption capacities of heavy metals by vegetables, atmospheric deposition of heavy metals, which may be influenced by innumerable environmental factors such as temperature, moisture and wind velocity, and the nature of the vegetables, i.e. leafy, root, fruit, exposed surface area, hairy or smoothness of the exposed parts (Zurera et al., 1989). The variations in the concentrations of heavy metals in the vegetables tested may also be ascribed to the variations in the anthropogenic activities such as brick kiln activities, addition of phosphate fertilizers or use of metal-based pesticides around production sites and urban industrial activities at market sites. The production sites showing higher levels of heavy metals are either located in the areas having a number of brick kiln industries (sites 7, 10, 12 and 14) or located close to national highways (17 and 28).

The average values of heavy metals in the vegetables tested collected from production sites are lower than the PFA standard (Awasthi, 2000). At market sites, the mean concentration of Cu in cauliflower, and of Zn and Cd in both palak and cauliflower had exceeded the PFA standard. Zn at production sites also exceeded the PFA standard in cauliflower. Cd concentrations in vegetables tested from both production and market sites were many folds higher than the EU standard (Marshall, 2004). In contrast, Pb in vegetables tested from both production and market sites was below the PFA standard, but was many folds above the EU standard (Table 3). Cu concentration exceeded the PFA standard at all the market sites in cauliflower, and at sites 1 and 3 in palak (Fig. 2). Zinc concentration exceeded the PFA standard in cauliflower at most of the production and market sites (Fig. 3). Cadmium level in all the vegetables tested exceeded the EU standard (0.1 µg/g) at all the production sites, but was lower than PFA standard (1.5 µg/g) in both lady's finger and palak (Fig. 4). The average values of Cd in all the vegetables tested at market sites were higher than both the PFA and EU standards. Pb concentration was, however, below

Table 5

Daily intake of heavy metals by urban population of Varanasi through consumption of the test vegetables.

Vegetables (g/person/day)	Moisture Content (%)	Cu (mg/day)	Zn (mg/day)	Cd (µg/day)	Pb (µg/day)
Palak (13.6)	90	0.04	0.08	2.67	1.96
Lady's finger (21.96)	85	0.06	0.15	4.61	3.37
Cauliflower (41.09)	80	0.29	0.52	21.15	12.84
Total		0.39	0.75	28.43	18.17
PTDI ^a		3	60	60	214

^a Joint FAO/WHO Expert Committee on Food Additives (1999).

the PFA standard, but was many folds higher than the EU standard at all the study sites (Fig. 5).

3.3. Estimation of daily intake of heavy metals

Formal interviews conducted in the urban areas of Varanasi showed that the average consumptions of fresh palak, lady's finger and cauliflower per person per day are 13.6, 21.96 and 41.09 g, respectively; the corresponding dry weights were 1.36, 3.27 and 8.23 g, respectively. If the levels of heavy metal contamination (Table 3) recorded in this study are representative of contaminant concentrations in the vegetables tested and consumed by the interview sample then the contribution of each of the heavy metals to the dietary intake would be 0.04 mg, 0.08 mg, 2.67 µg and 1.96 µg for palak, 0.06 mg, 0.15 mg, 4.61 µg and 3.37 µg for lady's finger and 0.29 mg, 0.52 mg, 21.15 µg and 12.84 µg/day for cauliflower, respectively, for Cu, Zn, Cd and Pb (Table 5).

From the estimated daily intake of the studied heavy metals through the consumption of palak, lady's finger and cauliflower, it can be suggested that the consumption of average amounts of these contaminated vegetables does not pose a health risk for the consumers as the values obtained are below the FAO/WHO limits for heavy metals intake based on the body weight of an average adult (60 kg body weight). Provisional tolerable daily intakes (PTDIs) for Cu, Zn, Cd and Pb are 3 mg, 60 mg, 60 µg and 214 µg, respectively (Joint FAO/WHO Expert Committee on Food Additives, 1999). The present study showed that the contributions of these vegetables to daily intake of Cu, Zn, Cd and Pb were 13%, 1%, 47% and 9% of provisional tolerable daily intake (PTDI), respectively.

4. Conclusions

The present study has generated data on heavy metal pollution in and around an Indian city and associated risk assessment for consumer's exposure to the heavy metals. The proposed hypothesis that the transportation and marketing of vegetables in contaminated environment may elevate the levels of heavy metals in vegetables through surface deposition has been proved through this study. The consumption of vegetables directly from production areas might be less hazardous to human health in comparison to those from polluted open market areas. Heavy metals have a toxic impact, but detrimental impacts become apparent only when long-term consumption of contaminated vegetables occurs. It is therefore suggested that regular monitoring of heavy metals in vegetables and other food items should be performed in order to prevent excessive buildup of these heavy metals in the human food chain. Appropriate precautions should also be taken at the time of transportation and marketing of vegetables.

5. Conflict of interest statement

The authors declare that there are no conflicts of interest.

Acknowledgments

The present research work is an output of a collaborative research project led by Fiona Marshall (F.Marshall@sussex.ac.uk, University of Sussex) entitled "Enhancing food chain integrity: quality assurance mechanisms for air pollution impacts on fruit and vegetable systems" which was funded by Department for International Development (DFID), United Kingdom, for the benefit of developing countries (RNRRS project number R8160). The views expressed are not necessarily those of DFID. Further details can be found at <http://www.pollutionandfood.net>. Rajesh Kumar Sharma

is thankful to DFID, U.K., and CSIR, New Delhi, India, for providing Junior and Senior Research Fellowship, respectively.

References

- Agrawal, M., 2003. Enhancing food chain integrity: quality assurance mechanisms for air pollution impacts on fruit and vegetable system. Final Technical Report II submitted to Department of International Development, UK, R 7530.
- Allen, S.E., Grimshaw, H.M., Rowland, A.P., 1986. Chemical analysis. In: Moore, P.D., Chapman, S.B. (Eds.), *Methods in Plant Ecology*. Oxford: Blackwell Scientific Publication, London, pp. 285–344.
- Awasthi, S.K., 2000. Prevention of food Adulteration Act No. 37 of 1954. Central and State Rules as Amended for 1999, third ed. Ashoka Law House, New Delhi.
- Cui, Y.J., Zhu, Y.G., Zhai, R.H., Chen, D.Y., Huang, Y.Z., Qiu, Y., Liang, J.Z., 2004. Transfer of metals from soil to vegetables in an area near a smelter in Nanning, China. *Environ. Int.* 30, 785–791.
- Feig, D.I., Reid, T.M., Loeb, L.A., 1994. Reactive oxygen species in tumorigenesis. *Cancer Res.* 54 (Suppl.), 1890–1894.
- Marshall, 2004. Enhancing food chain integrity: quality assurance mechanism for air pollution impacts on fruits and vegetables systems. Crop Post Harvest Program, Final Technical Report (R7530). <<http://www.sussex.ac.uk/spru/1-4-7-1-11-1.html>>.
- Gibbes, H., Chen, C., 1989. Evaluation of issues relating to the carcinogens risk assessment of chromium. *Sci. Total Environ.* 86 (1), 181–186.
- Hartwig, A., 1998. Carcinogenicity of metal compounds: possible role of DNA repair inhibition. *Toxicol. Lett.* 102, 235–239.
- Jarup, L., 2003. Hazards of heavy metal contamination. *Br. Med. Bull.* 68, 167–182.
- Jassir, M.S., Shaker, A., Khaliq, M.A., 2005. Deposition of heavy metals on green leafy vegetables sold on roadsides of Riyadh city, Saudi Arabia. *Bull. Environ. Contam. Toxicol.* 75, 1020–1027.
- Joint FAO/WHO Expert Committee on Food Additives, 1999. Summary and conclusions. In: 53rd Meeting, Rome, June 1–10, 1999.
- Jorhem, L., Sundstroem, B., 1993. Levels of lead, cadmium, zinc, copper, nickel, chromium, manganese and cobalt in foods on the Swedish market, 1983–1990. *J. Food Comp. Anal.* 6, 223–241.
- Kachenko, A.G., Singh, B., 2006. Heavy metals contamination in vegetables grown in urban and metal smelter contaminated sites in Australia. *Water Air Soil Pollut.* 169, 101–123.
- Khairiah, T., Zalifah, M.K., Yin, Y.H., Aminath, A., 2004. The uptake of heavy metals by fruit type vegetables, grown in selected agricultural areas. *Pak. J. Biol. Sci.* 7 (2), 1438–1442.
- Khan, S., Cao, Q., Zheng, Y.M., Huang, Y.Z., Zhu, Y.G., 2008. Health risk of heavy metals in contaminated soils and food crops irrigated with waste water in Beijing, China. *Environ. Pollut.* 152 (3), 686–692.
- Khillare, P.S., Balachandran, S., Meena, B.R., 2004. Spatial and temporal variation of heavy metals in atmospheric aerosols of Delhi. *Environ. Monit. Assess.* 90, 1–21.
- Krishna, A.K., Govil, P.K., 2007. Soil contamination due to heavy metals from an industrial area of Surat, Gujarat, Western India. *Environ. Monit. Assess.* 124 (1–3), 263–275.
- Milacic, R., Kralj, B., 2003. Determination of Zn, Cu, Cd, Pb, Ni and Cr in some Slovenian foodstuffs. *Eur. Food Res. Technol.* 217, 211–214.
- Parveen, Z., Khuhro, M.I., Rafiq, N., 2003. Market basket survey for lead, cadmium, copper, chromium, nickel and zinc in fruits and vegetables. *Bull. Environ. Contam. Toxicol.* 71, 1260–1264.
- Pilot, C.H., Dragan, P.Y., 1996. Chemical carcinogenesis. In: Casarett, Doulls (Eds.), *Toxicology International Edition*, fifth ed. McGraw Hill, New York, pp. 201–260.
- Radwan, M.A., Salama, A.K., 2006. Market basket survey for some heavy metals in Egyptian fruits and vegetables. *Food Chem. Toxicol.* 44, 1273–1278.
- Saplakoglu, U., Iscan, M., 1997. DNA single-strand breakage in rat lung, liver and kidney after single and combined treatments of nickel and cadmium. *Mutat. Res.* 394 (1), 133–140.
- Sharma, R.K., Agrawal, M., Marshall, F.M., 2006. Heavy metals contamination in vegetables grown in wastewater irrigated areas of Varanasi, India. *Bull. Environ. Contam. Toxicol.* 77, 311–318.
- Sharma, R.K., Agrawal, M., Marshall, F.M., 2007. Heavy metals contamination of soil and vegetables in suburban areas of Varanasi, India. *Ecotox. Environ. Saf.* 66, 258–266.
- Sharma, R.K., Agrawal, M., Marshall, F.M., 2008a. Heavy metals (Cu, Cd, Zn and Pb) contamination of vegetables in Urban India: a case Study in Varanasi. *Environ. Poll.* 154, 254–263.
- Sharma, R.K., Agrawal, M., Marshall, F.M., 2008b. Atmospheric depositions of heavy metals (Cd, Pb, Zn, and Cu) in Varanasi city, India. *Environ. Monit. Assess.* 142 (1–3), 269–278.
- Singh, K.P., Mohon, D., Sinha, S., Dalwani, R., 2004. Impact assessment of treated/untreated wastewater toxicants discharged by sewage treatment plants on health, agricultural, and environmental quality in wastewater disposal area. *Chemosphere* 55, 227–255.
- Singh, S., Kumar, M., 2006. Heavy metal load of soil, water and vegetables in peri-urban Delhi. *Environ. Monitor. Assess.* 120, 71–79.
- Sinha, S., Gupta, A.K., Bhatt, K., Pandey, K., Rai, U.N., Singh, K.P., 2006. Distribution of metals in the edible plants grown at Jajmou, Kanpur (India) receiving treated tannery wastewater: relation with physicochemical properties of the soil. *Environ. Monit. Assess.* 115, 1–22.

- Temmerman, L.O., Hoenig, M., Scokart, P.O., 1984. Determination of "normal" levels and upper limit values of trace elements in soils. *Z. Pflanzenernähr. Bodenkd.* 147, 687–694.
- Trichopoulos, D., 1997. Epidemiology of cancer. In: DeVita, V.T. (Ed.), *Cancer, Principles and Practice of Oncology*. Lippincott Company, Philadelphia, pp. 231–258.
- Tripathi, R.M., Ragunath, R., Krishnamurty, T.M., 1997. Dietary intake of heavy metals in Bombay City, India. *Sci. Total Environ.* 208, 149–159.
- Wang, X., Sato, T., Xing, B., Tao, S., 2005. Health risk of heavy metals to the general public in Tianjan, China via consumption of vegetables and fish. *Sci. Tot. Environ.* 350 (1–3), 28–37.
- WHO, 1992. Cadmium. *Environmental Health Criteria*, vol. 134, Geneva.
- Wong, C.S.C., Li, X.D., Zhang, G., Qi, S.H., Peng, X.Z., 2003. Atmospheric depositions of heavy metals in the Pearl River Delta, China. *Atmos. Environ.* 37, 767–776.
- Zurera, G., Moreno, R., Salmeron, J., Pozo, R., 1989. Heavy metal uptake from greenhouse border soils for edible vegetables. *J. Sci. Food Agric.* 49, 307–314.