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Metal Contamination and Health Risk from Consumption of Organically Grown Vegetables Influenced by Atmospheric Deposition in a Seasonally Dry Tropical Region of India

Ashima Singh · Jitendra Pandey

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Abstract Pot-culture experiments showed that organically grown *Vicia faba,* influenced by atmospheric deposition, accumulated (µg g^{-1}) 0.088–3.246 Cadmium, 0.19–42.48 Chromium, 0.0124–30.43 Copper, 0.075–4.28 Lead and 0.63–67.68 Zinc. Similar trends appeared for *Abelmoschus esculentus.* At high deposition sites, Cadmium, Lead and Zinc exceeded the safe limits of Prevention of Food Adulteration standards. Health risk index for Cadmium, Copper and Lead exceeded the safe limits of United States Environmental Protection Agency. The study suggests that atmospheric deposition could substantially elevate metal levels in organically grown vegetables in 2011.

Keywords Atmospheric deposition · Health risk · Metal · Organic farming

Rising demand of food by burgeoning human population coupled with agriculturally-driven food chain associated health risks have become a subject of global concern. In this context, organic farming has emerged as one of the fastest growing sectors of environment friendly sustainable agricultural practice (Pandey and Singh 2012). Metals are among the most widely known contaminants of natural and agro-ecosystems. Consumption of such contaminated foodstuffs may lead serious health risk in long-term future. Soil irrigation with waste water has been cited as the principal source of contamination (Xue et al. 2012). Atmospheric deposition has now been recognized as one of the important sources (Azimi et al. 2004; Pandey et al. 2009a, b). Particularly for developing countries such as ours, this problem is rapidly increasing due to fastened urban-industrial growth and lack of efficient control devices (Pandey et al. 2009a, b). Thus, although waste water irrigation can be avoided, air–borne metals may continue to contaminate agriculture far from sources of emission. In organic farming, the net accumulation of metals may partly be offset by reduced soil transfer mediated by organic supplements (Pandey and Pandey 2009a). However, since a substantial part of air-borne metals can be absorbed directly through aerial parts (Pandey and Pandey 2009b), despite all efforts to raise safe and hazard-free agricultural produce, atmospheric deposition may obviate the goals of organic farming. There is however, a general dearth of studies in India explicitly addressing this issue. The present study was an effort to investigate atmospheric deposition and associated metal contamination of organically grown vegetables in a seasonally dry tropical region of India. For this purpose, we used pot-culture experiments, in which soil from the same stock is used in earthen pots enabling some control over root environment and biotic interference (Pandey and Agrawal 1994). Attempts were also made to assess possible daily intake and associated health risks to consumers.

Materials and Methods

The data presented here are the results of three consecutive years (2008–2011) of study conducted at selected urban and sub-urban sites of Varanasi (25°18′N lat, 83°01′E long and 76.19 m above msl). The climate of the region is characterized by three distinct seasons, a hot and dry summer (March–June), a humid monsoon season (July–October) and a cold winter season (November–February). The region

A. Singh · J. Pandey (✉)
Environmental Science Division, Centre of Advanced Study in Botany, Banaras Hindu University, Varanasi 221005, India
e-mail: jiten_pandey@rediffmail.com

receives about 1100 mm average rainfall. During the study period, mean monthly maximum temperature ranged from 27.5 to 45.4°C and minimum temperature from 9.5 to 25.7°C. The alluvium soil of the region is highly fertile light textured sandy loam (pH 6.8–7.9).

For the present study, we considered four zones with three sub-sites each selected on the basis of emission sources. The study zones were Banaras Hindu University campus (BHU; consisted of educational and residential areas), Sunderpur (SPR; with residential and shopping complex sites), Cantonment (CNT; with heavy traffic and industrial emission) and Ramnagar (RNR; heavy traffic and industrial emission zone). We considered two most commonly used dietary vegetables, Lady's finger (*Abelmoschus esculentus* L.) and Bakla (*Vicia faba* L.) in pot- culture experiments using recommended dose of nitrogen, phosphorus and potassium (NPK), farmyard manure (FYM) and vermicompost (VC) as per standard agricultural practices. Three sets of each amendment per species were used at each deposition sub-site.

Bulk atmospheric deposition was collected using bulk samplers maintained at a height of 2 m to avoid contamination of re-suspended particulates and was devised with PVC needles on top to avoid bird nesting (Pandey and Pandey 2009a, b). Tri-acid mixture (70 % high purity HNO_3, 65 % $HClO_4$, and 70 % H_2SO_4; 5:1:1) was used for digestion of particulates and soil samples (Allen et al. 1986). Edible portions were separated from the finally harvested vegetables and properly washed to remove surface dust. Cleaned plant samples were chopped into small pieces, oven dried at 80°C, ground in a stainless steel blender and passed through a 2 mm sieve. Samples were digested in tri-acid mixture (Allen et al. 1986) and filtered using Whatman No. 42 filter paper. Digested filtrates were analyzed for Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb) and Zinc (Zn) using an Atomic Absorption Spectrophotometer (Perkin-Elmer, A Analyst 800, USA).

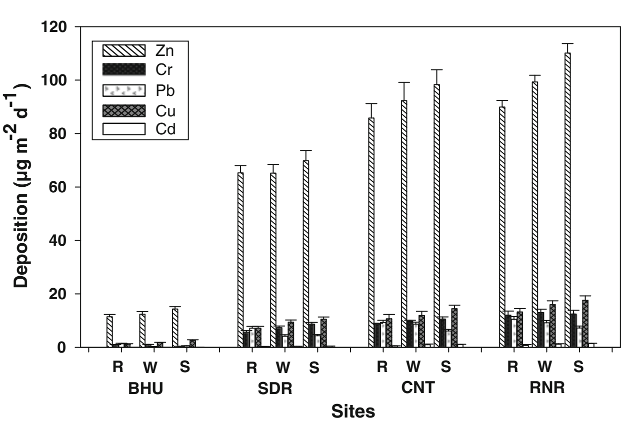

Fig. 1 Atmospheric deposition (µg m^{-2} day^{-1}) of heavy metals at different study sites of Varanasi. Values are mean (n = 9) ±1SE

The detection limits (µg ml^{-1}) were: 0.0005 (Cd), 0.002 (Cr), 0.001 (Cu), 0.01 (Pb) and 0.0008 (Zn). The precision of metal analysis was recovered by repeated analysis against fortified NIST-SRM, 1570 for all the metals. A quality control has been performed using acidified water blanks for checking the contamination during field collection and sample treatments. Blank and drift standards (Sisco Research Laboratory Pvt. Ltd., India) were run after five reading to calibrate the instrument.

Contribution of atmospheric and soil sources to vegetal metal was estimated in terms of air accumulation factor (AAF) and transfer factor (TF) following Voutsa et al. (1996). Daily intake of metal (DIM) was determined considering metal concentration in plants and per person per day consumption of vegetables in study areas. Health risk index (HRI) was estimated using DIM and the reference oral dose of each metal (USEPA 2006). At HRI below 1, the exposed population is considered to be safe.

Data were subjected to three-way analysis of variance (ANOVA) to address significant effects on vegetal heavy metal. Correlation models were used to test relationship between variables. The statistical analyses were done using SPSS (version 16).

Results and Discussion

Atmospheric deposition was recorded highest at RNR, the values being 1.10, 12.45, 15.6, 8.96 and 99.77 µg m^{-2} day^{-1} for Cd, Cr, Cu, Pb and Zn respectively (Fig. 1). Deposition of all metals was lowest at BHU. Deposition of Zn was found highest followed by Cr, Cu, Pb and Cd (Fig. 1). Deposition recorded in the present study, although was comparable to those measured at some urban and sub-urban locations of the world (Wong et al. 2003), was higher than those observed at Great Lake site (Sweet et al. 1998), at Chesapeake and Delaware Bay sites (Kim et al. 2000), and in northern England (Lawlor and Tipping 2003).

We found elevated level of metals in soil at sites receiving high atmospheric deposition (Table 1). At RNR, (highest deposition site), soil metal (µg g^{-1}) ranged from 13.36 to 26.78 for Cd, 14.52 to 36.86 for Cr, 45.52 to 76.49 for Cu, 119.35 to 139.98 for Pb and 400.45 to 456.28 for Zn (Table 1). Almost 100 % of soil samples at RNR showed Cd, Cr, Cu, Pb and Zn levels exceeding safe limits (Temmerman et al. 1984). As shown in previously reported results (Pandey and Pandey 2009a, b), we found significant positive relationship between deposition input and soil metal levels ($R^2 = 0.63$–0.86, $p < 0.0001$), indicating deposition-linked soil contamination.

Metals in edible part of vegetables showed a trend corresponding to the gradient of deposition (Fig. 2). In *V. faba*, concentrations (µg g^{-1}) ranged from 0.088 to 3.246 for Cd,

Table 1 Concentration of heavy metals ($\mu g\ g^{-1}$) in amended soils at different sites. Values are mean (n = 9) ±1SE

	Zn	Cr	Pb	Cu	Cd
Preharvest	109.85 ± 5.26	10.46 ± 2.53	52.32 ± 4.70	16.58 ± 0.42	3.18 ± 0.39
Site I					
NPK	96.95 ± 1.46	4.03 ± 0.69	45.61 ± 2.70	10.27 ± 0.94	2.27 ± 0.24
FYM	110.83 ± 2.19	10.42 ± 1.88	49.48 ± 2.19	13.35 ± 1.28	2.64 ± 0.19
VC	111.46 ± 1.66	10.95 ± 1.38	50.17 ± 1.32	13.88 ± 0.79	2.88 ± 0.34
Site II					
NPK	116.76 ± 2.11	8.74 ± 1.47	83.17 ± 2.00	22.39 ± 1.20	6.17 ± 0.40
FYM	125.98 ± 2.24	12.44 ± 1.23	90.38 ± 2.57	29.45 ± 0.59	7.86 ± 0.28
VC	126.50 ± 2.87	12.83 ± 1.69	91.29 ± 1.75	29.92 ± 0.77	8.19 ± 0.32
Site III					
NPK	390.95 ± 3.04	17.95 ± 1.29	114.54 ± 1.89	51.13 ± 1.30	9.96 ± 0.29
FYM	398.74 ± 2.55	19.49 ± 1.72	119.44 ± 2.27	60.56 ± 1.74	13.76 ± 0.86
VC	399.92 ± 3.64	19.78 ± 2.28	119.75 ± 2.53	61.17 ± 1.89	13.88 ± 0.68
Site IV					
NPK	406.97 ± 2.89	16.6 ± 1.20	122.79 ± 1.98	47.45 ± 1.11	14.62 ± 0.72
FYM	418.34 ± 2.27	25.48 ± 1.95	127.62 ± 2.33	54.38 ± 1.23	17.38 ± 0.23
VC	420.68 ± 2.20	25.76 ± 1.96	125.58 ± 1.80	55.44 ± 1.35	18.55 ± 0.98

0.19 to 42.48 for Cr, 0.0124 to 30.43 for Cu, 0.075 to 4.28 for Pb and 0.63 to 67.68 for Zn (Fig. 2). Concentration was highest for Zn followed by Cu, Cr, Pb and Cd. The overall trend was common at all sites. In *A. esculentus* also, metal levels were generally consistent with their atmospheric supplies (Fig. 2). With respect to treatment, although overall trends were generally similar, the consistency and magnitude of accumulation did vary between crops as well as among the metals. For instance, compared to *V. faba*, *A. esculentus* showed high tissue accumulation of Zn (2.76 to 90.73) and Cu (0.367 to 53.78), whereas Cr was comparatively higher in *V. faba.* At RNR, in comparison to BHU site, vegetal metal concentration increased by 6–29 fold in unamended soil, 6–32 fold in NPK, 4–48 fold in FYM and 5–64 fold in VC amended soil.

Treatment-wise variation in metal concentration could be due possibly to soil amendment associated modification in root environment. Crops generally retrieve nutrients and trace elements from soil and organic supplements added to the root environment may reduce metal uptake due to complex formation and chelation effects (Walker et al. 2003). Further, absorption through aerial parts also contributes substantially (Voutsa et al. 1996). We found significant positive relationship between atmospheric input and metal concentration in *V. faba* ($R^2 = 0.26$–0.77, $p < 0.002$–0.0001) and *A. esculentus* ($R^2 = 0.30$–0.88, $p < 0.005$–0.0001). As shown in previously reported results (Pandey and Pandey 2009a), the increase in vegetal metal was related to the increase in atmospheric deposition along the gradient.

Except for Cr, vegetal metal was significantly related to soil metal levels in *V. faba* ($R^2 = 0.27$–0.62, $p < 0.001$–0.0001) and *A. esculentus* ($R^2 = 0.24$–0.59, $p < 0.002$–0.0001). The correlations however, were stronger with atmospheric input indicating that air driven input significantly adds to vegetable metal concentration. Metal concentration was comparatively lower in organically grown crops (Fig. 2) indicating that organic amendment may partly offset the effect of atmospheric deposition. Organic amendments in soil generally reduce root uptake of metals by complex formation and chelation (Walker et al. 2003), and as also reported here, VC is generally found superior (Chanda et al. 2011). Atmospheric deposition showed both direct and indirect influence on vegetal metal enrichment through air and soil. Air accumulation factor (AAF) was found higher in vegetables grown in amended soils indicating atmosphere to be the major contributor of metal enrichment in organically grown vegetables. Nutrients added in soil as NPK or organic supplements promote plant growth and in turn, enhance aerial uptake (Pandey and Pandey 2009a). Due probably to this reason, despite low overall concentration, metal accumulation was faster in organically grown crops (5–64 fold increase) than those grown in unamended soil (6–29 fold increase at same time intervals). At higher deposition sites, most metals in organically grown crops exceeded the prevention of food adulteration (PFA) standards (Awasthi 2000). Cadmium in both the vegetables was although lower than PFA standards (1.5 $\mu g\ g^{-1}$) yet was many fold higher than EU standard (0.10 $\mu g\ g^{-1}$). Thus, the organic amendment in soil did not fully able to overcome the effect of atmospheric input as far as metal accumulation in crops is concerned. Air-driven input of heavy metal could override the ameliorating effects of organic residues.

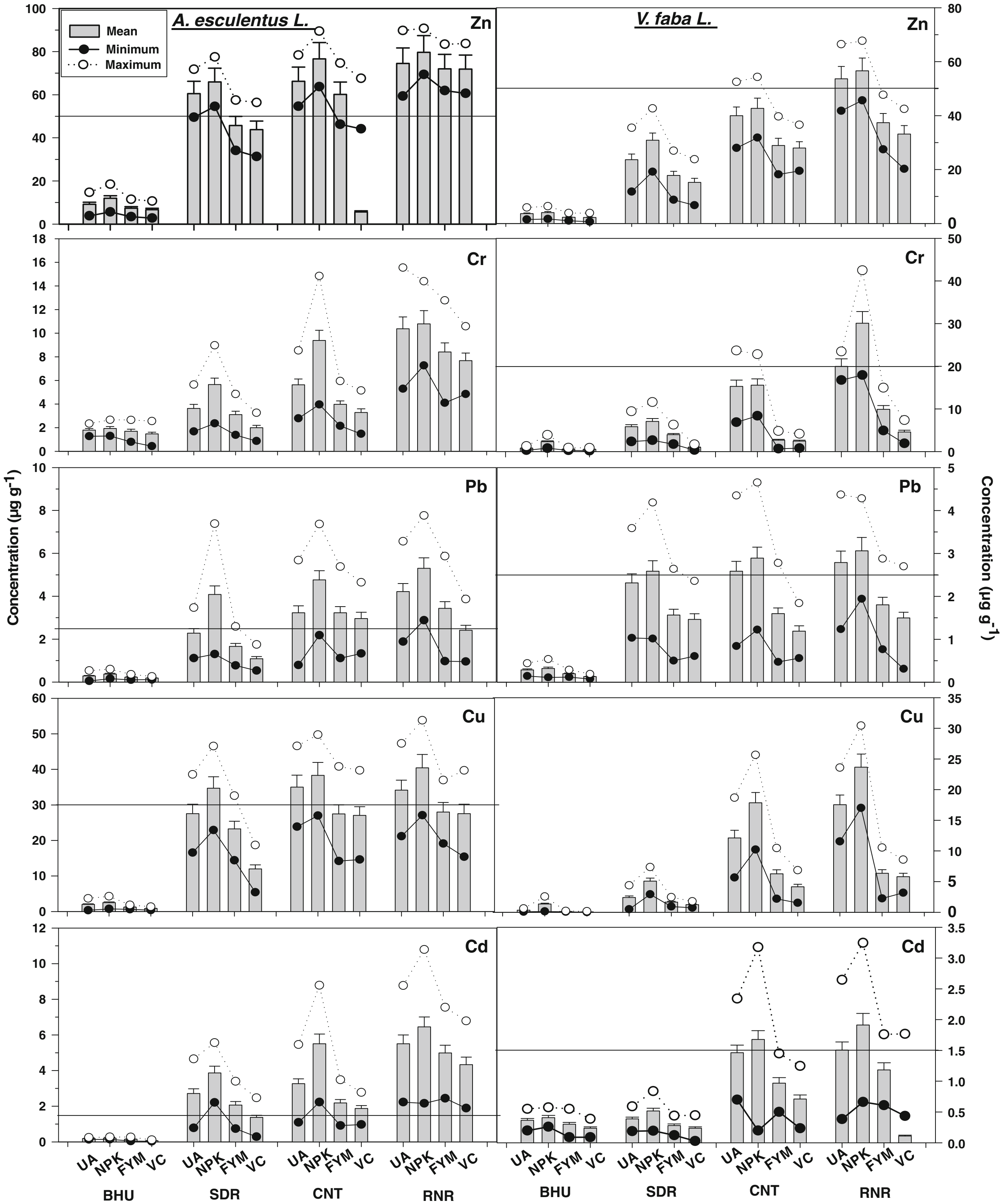

Fig. 2 Heavy metal concentration (μg g^{-1}) in edible part of vegetables grown at study sites. Values are mean (n = 9) ±1SE

At highest deposition site (RNR), daily intake of metal (DIM) for Cd, Cr, Cu, Pb and Zn was 0.0043, 0.0492, 0.0406, 0.0069 and 0.1372 mg person^{-1} day^{-1} respectively, for *V. faba* and 0.0173, 0.0305, 0.1063, 0.0125 and 0.2436 mg person^{-1} day^{-1} respectively, for *A. esculentus* (Fig. 3). At this site, corresponding to DIM, the HRI was 4.342, 0.0328,

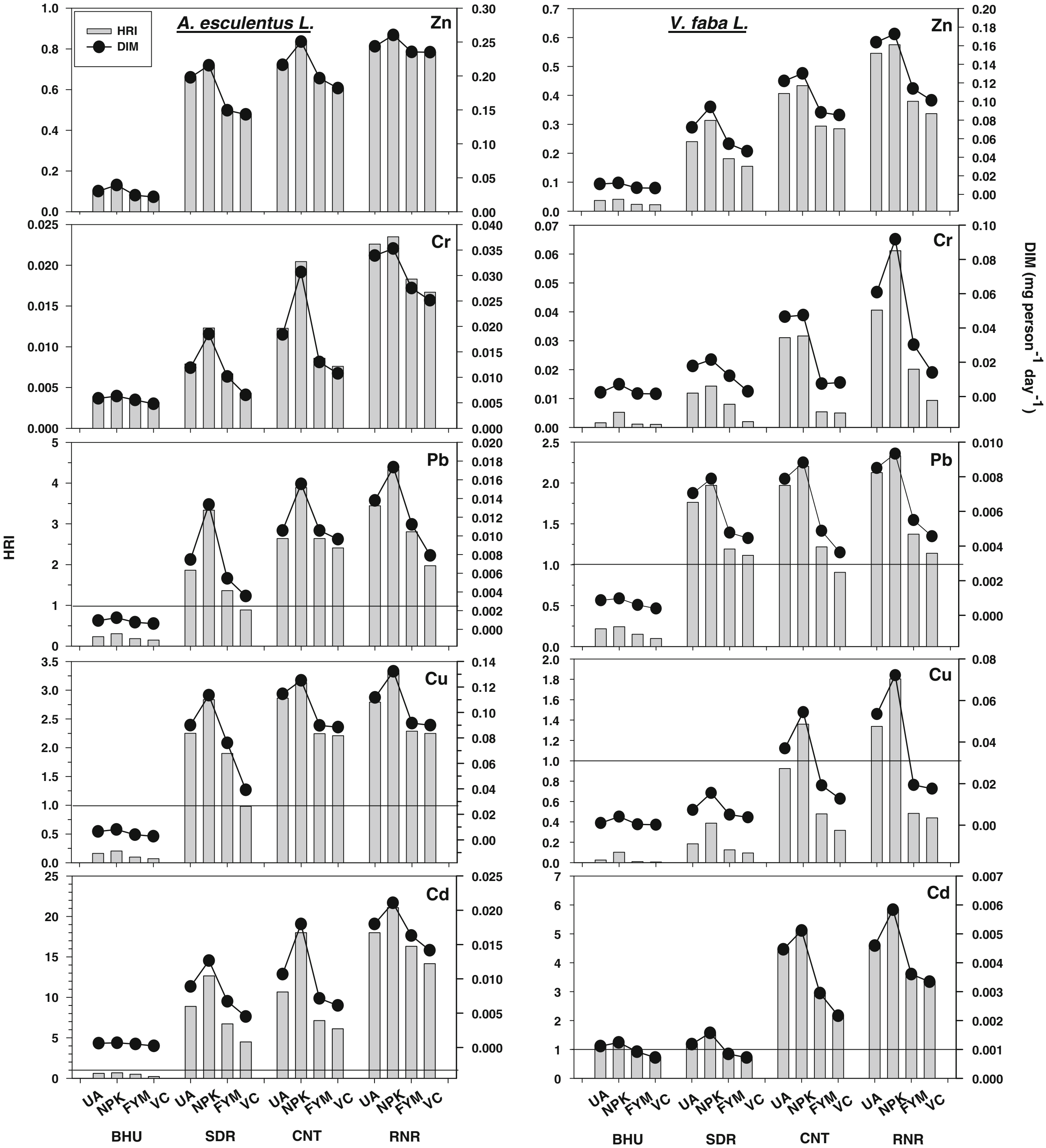

Fig. 3 Daily intake of metal (mg person^{-1} day^{-1}) and health risk index for local consumers of contaminated vegetables

1.016, 1.743 and 0.4593 for Cd, Cr, Cu, Pb and Zn respectively, for *V. faba* and 17.39, 0.0203, 2.659, 3.139 and 0.8120 respectively, for *A. esculentus* (Fig. 3). Although DIM was lower than Provisional Tolerable Daily Intake (PTDI) for Cd, Cr, Cu, Pb and Zn (Joint FAO/WHO Expert Committee on Food Additives, 1999), high values of HRI of Cd, Cu and Pb show that daily consumption of such contaminated vegetables can pose health risk to local consumers. For Cd, Cu and Pb the HRI exceeded the established standards (Zhuang et al. 2009).

Thus, despite the fact that organic supplements (FYM, VC) could reduce accumulation of metals, the two-way effect (pedo-atmospheric) of atmospheric metal supplies may obviate one of the major objectives of organic farming in the long-term future. We conclude that even where irrigation-linked and other sources of contamination are completely avoided, atmospheric deposition, which is expected to rise in future, particularly in developing countries, may continue to undermine the success of organic farming in long-run.

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