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Trace Element Occurrence in Vegetable and Cereal Crops from Parts of Asia: A Meta-data Analysis of Crop-Wise Differences

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Abstract

In the present study, a systematic review along with a meta-analysis was conducted based on relevant studies from 11 Asian countries (1999–2022, *Scopus*, *PubMed*, *MEDLINE*, *ScienceDirect*, and *Google Scholar*) to evaluate the crop-wise differences in the accumulation of trace element (TE) in the edible part of different crops (vegetables: leafy (LV), root (RV), fruit (FV); cereal crops: rice (RIC), wheat (WHE), maize (MAZ)). Based on the median concentration of the compiled data, the TE accumulation in different vegetable crops was ranked in the decreasing order of Fe > Zn > Mn > Cu > Ni > Cr > Pb > Co > Se > Cd > As, and in cereal crops, this is followed as Fe > Zn > Cu > Ni > Cr > Co > Pb > As > Se > Cd > Hg. A clear difference was found between vegetable categories, with a higher accumulation of most of the elements in LV, especially spinach, coriander, radish leaves, mustard, amaranthus, and pakchoi than other vegetable types. Root vegetables displayed higher bioconcentration factors (BCF) than the other two vegetable types. For cereal crops, higher metal contents were found in WHE followed by RIC and MAZ, but RIC had relatively higher BCF for certain metals (As, Cd, Cu, Cr, Ni) and WHE dominated for the remaining metals. When compared with the prescribed safe limits of the non-essential metals (As, Cd, and Pb), this study revealed that the majority of the vegetable and cereal crop contaminations were from Bangladesh, China, India, Iran, and Pakistan.

Keywords Vegetable and cereal crops · Metal occurrence · Bioaccumulation · Asian countries

Introduction

Vegetables, grains, and legumes are essential foodstuffs in Asian countries as they are rich in nutrients and are a significant source of proteins, carbohydrates, minerals, and antioxidants, which help to reduce various kinds of chronic diseases and improve human body function [1, 2]. Therefore, there is a growing demand for the cultivation of these crops in these regions. However, vegetable crops grown in contaminated areas are one of the greatest threats to food quality and human health as they can readily accumulate a wide variety of chemical pollutants (beyond their recommended limits), commonly termed as "trace elements" (TEs), in their edible and inedible (fodder) parts and enter into the food chain [3–5]. These TEs are now considered a significant health concern worldwide, especially in developing and underdeveloped countries [6, 7]. Some of the TEs, specifically Fe, Mn, Cr, Cu, Zn, and Se, are essential for human health at specific concentrations, but they may become toxic at higher doses. In contrast, elements like As, Cd, Pb, and Hg are non-essential elements that are included in the top 20 list of dangerous substances by the United States Environmental Protection Agency (USEPA) and the Agency for Toxic Substances and Disease Registry (ATSDR) [8]. These elements can be very harmful even in trace quantities in any food item when ingested over a longer duration [8–13] and can cause various health diseases like anemia, bone diseases, gastrointestinal cancer, kidney failure, decrease in immunological defenses, tissue damage, osteoporosis, skin lesions, cardiovascular diseases, diabetes, and blood and lung cancers, etc. [14–16]. These TEs also decline crop yield by inhibiting metabolic processes [17]. Many

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studies have been carried out on the origin and distribution of TE within plants for over a century. This work presents an inventory of TEs in major crops from some Asian countries.

The accumulation of TE in agricultural soil may be the consequence of both geogenic and anthropogenic factors (Fig. 1). The latter is more pronounced in developing countries due to the continued growing population, resulting from the need for food security, rapid urbanization, industrialization, hazardous waste dumping, coal combustion in power plants, vehicular traffic, automobile exhausts, land-use change, and excessive/unsystematic application of pesticides, fertilizers, sewage, and irrigation with wastewater or polluted groundwater [18–22]. Urban wastewater, some with industrial effluents, is used for crop production in many densely populated Asian countries like China, India, Pakistan, and Bangladesh as a compromise measure for lack of freshwater and water scarcity [23]. Irrigation with contaminated groundwater or surface water receiving agrichemical runoff is another major factor in metal-crop composition, especially in Southeast Asian countries, where groundwater bodies can be severely contaminated with toxic metals [18, 24–26]. Long-term use of contaminated irrigation water and soil amendments leads to a high accumulation of TE in soils, which can be subsequently transferred to the edible and fodder parts of crops via various uptake and translocation mechanisms (Fig. 1), see Supplementary Note 1, and ultimately end up in the food chain [26–33]. Thus, poor irrigation water quality and soil amendments threaten sustainable agriculture and human health in developing countries like India, China, and Bangladesh [24, 26]. Crops requiring higher irrigation frequency accumulate greater metal concentrations than crops with fewer water requirements [34]. Moreover, the accumulation of TE in food crops is affected by soil types and soil physicochemical properties (e.g., temperature, moisture, pH, redox potential, clay mineral content, organic matter, Fe/Al oxides, cation exchange capacity, and speciation) which in turn control solubility and bioavailability of metals in the soil-rhizosphere [35–39]. This is significantly varied between elements. For example, the bioaccumulation of As increases significantly under reducing conditions, whereas that of Cd decreases [40–42] due to changes in solubility when soil redox potential drops [41, 42]. Also, metal accumulation in crops significantly varies depending on the plant varieties/species, their ability to take up metals, and soil–plant uptake factors [43]. Thus, the occurrence of TE in plants depends on their soil bioavailability and the composition of irrigation water and on the plant's ability to uptake and sequester TE. Therefore, it is crucial to determine the TE accumulation potential of different crop/plant species. Understanding these differences will be helpful when describing the TE

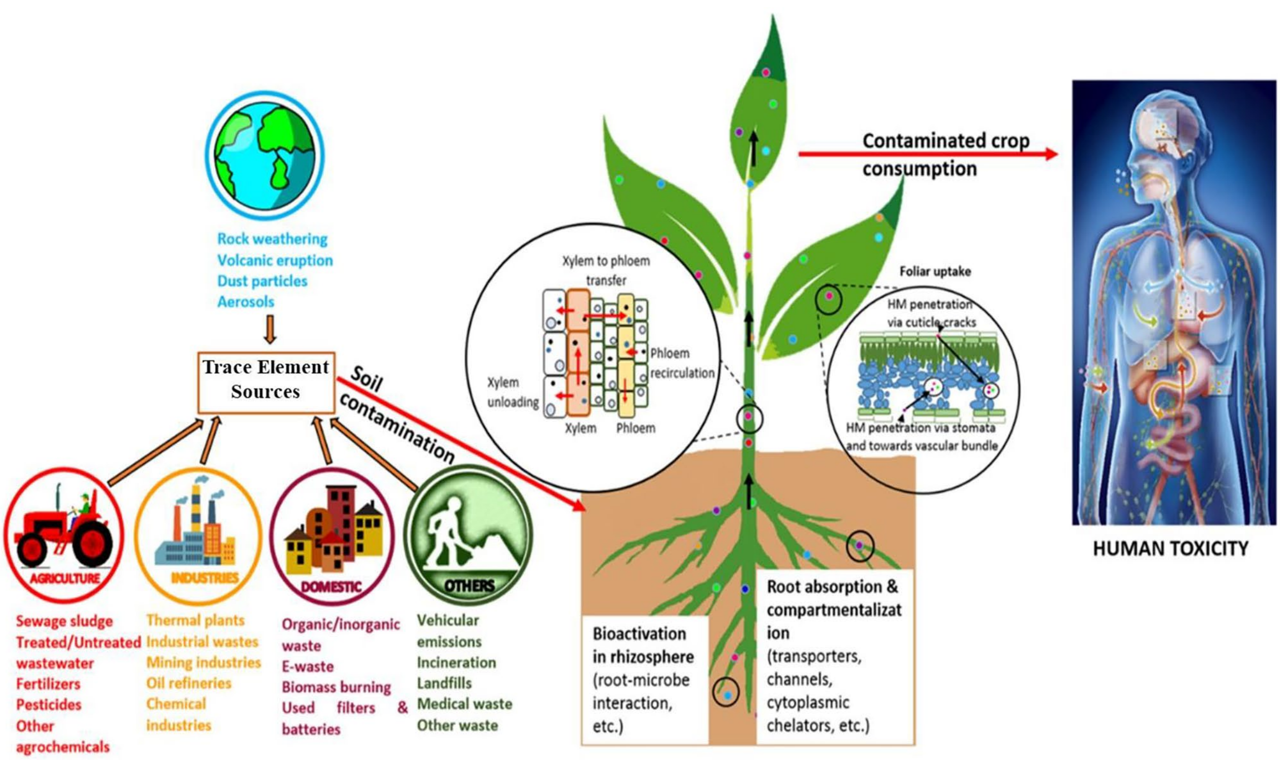


Fig. 1 Schematic diagram shows various natural and anthropogenic sources of trace element contamination in agricultural soils and the major processes involved in the uptake, translocation, and sequestration of metals in crops (see Supplementary Note 1)

contamination status of food crops, which can help predict health risks and dietary planning.

There have been numerous studies from Asian countries on the relative importance of irrigation water types and soil amendments concerning their impact on crop-metal concentrations in edible and non-edible parts of plants. Some recent reviews also concentrate on this topic which have mostly focused on a specific crop type, either only vegetables [44, 45], or wheat (*Triticum aestivum*) [46], or rice (*Oryza sativa*) [47, 48], or maize (*Zea mays*) [49]. However, to the best of our knowledge, recent systematic reviews are scarce on the TE accumulation on a wide variety of crops (vegetables and cereal crops) in the Asian context; this study strives to fill this gap. Both descriptive and meta-analyses are important for synthesizing the information extracted from original studies and determining comprehensive conclusions on this topic [47]. In view of the above-mentioned point, we compiled an extensive database focusing on the Asian context (mostly major populated developing countries) from 1999 to 2022 and used basic statistics and meta-analysis to address the current status of TE contamination in edible parts of vegetables and grains and their corresponding soils and irrigation water and to examine the crop species differences in TE accumulation in them. The discussion is limited for irrigation water and soils due to the paucity of information provided in the referenced works. We further summarize the various factors responsible for this contamination and highlight the key variables. This work divides crops into leafy, root, and fruit vegetables and grains such as rice (*O. sativa*), wheat (*T. aestivum*), and maize (*Z. mays*), which include many of the essential food staples. The information provided in this paper will be helpful for policymakers to make sustainable agricultural irrigation policies and develop approaches to limit TE accumulation in crops to ensure future food security.

Methodology

Data Collection and Processing

A literature search was conducted using several electronic databases, i.e., Scopus, PubMed, MEDLINE, ScienceDirect, and Google Scholar. Relevant articles were then searched using the phrases "heavy metal accumulation in vegetable crops," "metal accumulation in food crops," "potentially toxic elements in food crops," and "metal accumulation in grains/rice/wheat." Search results were screened based on field and market-based studies in the Asian context, covering Bangladesh, China, India, Iran, Malaysia, Pakistan, Saudi Arabia, South Korea, Taiwan, Thailand, and Vietnam. The vegetable data were divided into three major categories based on agronomic classification: leafy vegetables (LV), root vegetables (RV), and fruit vegetables (FV). Similarly, grains were categorized as wheat (WHE), rice (RIC), and maize (MAZ). Detailed information for vegetable crops and grains from all primary studies is listed in Supplementary Tables S1 and S2, respectively. The number of studies and from which countries are given in the sections below. Only a few of the included studies contained usable information on TEs in irrigation water or soils, or both together (Supplementary Tables S1 and S2).

Bioconcentration factor (BCF) was calculated by computing the ratio of the concentration of each metal in the edible part of vegetables/grains and the corresponding soil concentration to determine the accumulation potential for different TEs (Eq. 1). This was calculated only for those specific crop records which had soil data. If an author used the soil data in a study for multiple crop types, the same data was counted for each crop.

$$\mathrm{BCF} = \frac{\text{Concentration of element in vegetables/crops}}{\text{Concentration of element in soils}} \quad (1)$$

Statistical and Meta-data Analysis

The crop data were subjected to basic and multivariate statistical analysis utilizing Microsoft Excel, SPSS, and R Studio. The normality of the data was checked by using the Kolmogorov–Smirnov test. Since most variables do not follow a normal distribution, Spearman's rank correlation coefficients were calculated to investigate the association between the target metals in soils and crops. Kruskal–Wallis (K–W) test, a non-parametric version of the ANOVA, was performed using SPSS for all the metals to evaluate the differences among vegetable and cereal species. Meta-analysis was applied on the concentrations of TE in different vegetable crops. This was done by the MedCalc statistical software. The statistical meta-analysis was utilized for the total heterogeneity effect using the I^2 and Q tests. Under $I^2 > 50\%$ and $p < 0.05$ conditions, we looked for a significant heterogeneity [50].

Basic Summary of the Compiled Data

The whole crop dataset (mostly average TE concentrations in crop wise from each study) was compiled from over 300 studies comprising over 1300 records of elements such as, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, and Zn (very limited Hg) (Supplementary Tables S1 and S2). Due to significant variations in consumption of these types of crops across/within Asian countries, the 40 vegetable crops were grouped into 3 categories: fruit vegetables (FV), leaf vegetables (LV), and root vegetables (RV); no attempt was made to compare individual crop types due to the large disparity in numbers of analyses or lack of data from one study/country to another. The highest number of records was for FV (52%), followed by LV (37%) and RV (11%). The vegetable data was compiled from 122 studies containing 838 records and 7713 separate analyses (individual analyses of either a crop, water, or soil). Certain crops belonging to the same family were grouped, resulting in 28 vegetable crops. Crops belonging to the same family like "green cabbage (*B. oleracea var. capitata*)," "Chinese cabbage (*Brassica rapa*)," and "cabbage" (*B. oleracea var. capitata*) were grouped into "cabbage," and "Indian spinach (*Amaranthus sp*)", "amaranth (*Amaranthus sp*)," and "red amaranthus (*Amaranthus sp*)" were clubbed as "amaranth"; "long beans (*Vigna unguiculata, sub. Sesquipedalis*)", "flat beans" (*Phaseolus coccineus*), "Indian beans (*Lablab purpureus*)," "beans (*Phaseolus vulgaris*)," and "common beans (*Phaseolus vulgaris*)" were grouped together as "beans"; "chilly (*Capsicum annuum*)" and "capsicum (*Capsicum annuum*)" were grouped into "chilly"; "lettuce," "long leaf lettuce," "iceberg lettuce," "leaf lettuce," and "Romaine lettuce" were all grouped as "lettuce" (*Lactuca sativa*). Using this scheme resulted in a total of 28 vegetable crop classifications (see Supplementary Table S1 for descriptions of the 3-letter crop abbreviations used in this paper: AMR, ARM, BIG, BNS, BOG, BRN, CBB, CRR, CBR, CLF, CLL, CRN, GRL, KAL, LTT, LEK, MNT, MST, OKR, ONN, PRS, PKC, PTT, RDH, RDL, SPN, TMT, and TNP). These vegetables were collected from 8 countries (number of studies): Bangladesh (21), China (20), India (36), Iran (14), Pakistan (19), Saudi Arabia (5), South Korea (6), and Vietnam (1; not included in the discussion). The top 4 countries represent 79% of the total studies compiled. More than 80% of the studies on vegetables were field-based, and the remaining were from a market-based survey. In grains, the major groups are rice (*O. sativa*), wheat (*T. aestivum*), and maize (*Zea mays*). The grain data were collected from 201 studies containing 526 records and 2887 separate analyses, of which 68% of data were from field studies and the other 32% were from market-based trials. Studies on rice account for 71% of the total, 20% for wheat, and 9% for maize, indicating the importance of rice in the Asian diet relative to wheat or maize, although all these are important to the Asian diet. In addition to the countries that reported the TE concentration in vegetables, published works from Malaysia, Taiwan, and Thailand (11 countries in total) were also included for grains. The number of studies from different countries included China (67), Bangladesh (29), India (25), Iran (23), Malaysia (5), Pakistan (19), Saudi Arabia (4), South Korea (18), Taiwan (5), Thailand (2), and Vietnam (8).

The data for associated soil and irrigation water were reported only in some records (Supplementary Tables S1 and S2). Of all the studies, 52% were done on crops, 37% on soil, and 11% on irrigation water. Irrigation water was separated into 6 types (% of each in all sources): "PPT," precipitation (2%); "SUR," surface water from canals and rivers or when uncontaminated groundwater or tube well water is mixed with them (15%); "UGW," uncontaminated groundwater (7%); "SWW," sewage, wastewater, including urban/domestic sources (38%); "IWW," industrial wastewater including mining, processing, etc. (25%); and "UNK," when no information is provided (12%) (Supplementary information: Fig. S1); market studies only reported data for 3, and of those, approximately 80% were unknown sources; any study with an "unknown" source of water was not considered in this work. Also, the entire data from each study, including the "outliers," have been considered for analysis as these data were gleaned from peer-reviewed published articles, and hence, the extremely low or high values are real. These outliers give weight to possible problems which require action for specific areas that need new policies on crop and dietary intake relevant to potentially toxic elements.

Results and Discussion

Statistical Analyses of the Occurrence of Trace Elements in Vegetable Crops

The statistical summary of the TE in the three vegetable categories is listed in Table 1. Overall, the concentration of TE varies widely in the studied vegetable species. The mean values for several elements are significantly different, which also have a higher relative standard deviation (RSD), indicating these elements are asymmetrically distributed. Considering this, median values are a statistically more robust measure of central tendency than means; thus, former values were used for further comparison purposes. A notable difference in the median concentrations of TE between and within the three vegetable categories was observed (Table 1). In LV, the median concentration of As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, and Zn was 0.45 mg kg^{-1}, 0.48 mg kg^{-1}, 0.42 mg kg^{-1}, 4.08 mg kg^{-1}, 6.31 mg kg^{-1}, 157.84 mg kg^{-1}, 34.29 mg kg^{-1}, 4.00 mg kg^{-1}, 2.24 mg kg^{-1}, 0.64 mg kg^{-1}, and 27.93 mg kg^{-1}, respectively. Based on these values, the TE in LV followed the order of Fe > Mn > Zn > Cu > Cr > Ni > Pb > Se > Cd > As > Co. Similarly, the median concentration in RV for As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, and Zn was as follows: 0.29 mg kg^{-1}, 0.29 mg kg^{-1}, 1.00 mg kg^{-1}, 1.97 mg kg^{-1}, 4.92 mg kg^{-1}, 68.94 mg kg^{-1}, 15.34 mg kg^{-1}, 2.58 mg kg^{-1}, 1.08 mg kg^{-1}, 0.76 mg kg^{-1}, and 21.90 mg kg^{-1}, respectively. The order of these metals in RV was Fe > Zn > Mn > Cu > Ni > Cr > Pb > Co > Se > Cd > As. Likewise, the median concentrations for As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn in FV were 0.50 mg kg^{-1}, 0.41 mg kg^{-1}, 0.84 mg kg^{-1}, 1.80 mg kg^{-1}, 6.43 mg kg^{-1}, 131.07 mg kg^{-1}, 14.93 mg kg^{-1}, 2.50 mg kg^{-1},

Table 1 Descriptive statistics of the average trace element concentrations from all studies in crops (all types) according to crop categories (for detailed information with individual study, see Supplementary Tables S1, S2)

LV (*n* = 422)	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	Zn
Nbr. of obs	105	335	36	195	312	84	113	179	370	9	278
Min (mg kg^{-1})	0.01	0.001	0.01	0.04	0.01	4.31	0.99	0.001	0.001	0.45	0.03
Max (mg kg^{-1})	16.0	70.0	57.6	322.0	100.0	2669.1	368.1	350.0	780.0	59.0	950.0
Q1 (mg kg^{-1})	0.16	0.16	0.09	1.17	1.34	74.75	16.58	0.96	0.32	0.55	8.33
Median (mg kg^{-1})	0.45	0.48	0.42	4.08	6.31	157.84	34.29	4.00	2.24	0.64	27.93
Q3 (mg kg^{-1})	2.08	1.63	5.06	9.90	14.51	497.68	73.10	14.35	8.81	0.67	58.95
Mean (mg kg^{-1})	1.79	2.35	8.20	19.15	10.63	400.93	58.97	21.14	15.90	7.10	53.18
SD	3.06	5.87	17.18	44.76	13.54	527.44	67.03	50.74	65.08	18.35	101.44
RSD (%)	171	249	209	234	127	132	114	240	409	258	191
RV (*n* = 186)	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	Zn
Nbr. of obs	61	126	18	68	118	54	73	74	146	4	117
Min (mg kg^{-1})	0.01	0.001	0.13	0.07	0.001	0.97	0.08	0.06	0.001	0.56	0.001
Max (mg kg^{-1})	8.20	30.00	53.20	78.02	35.00	1666.0	277.0	62.7	59.0	0.83	139.05
Q1 (mg kg^{-1})	0.14	0.09	0.46	0.84	1.23	32.58	5.65	0.62	0.34	0.68	7.20
Median (mg kg^{-1})	0.29	0.29	1.00	1.97	4.92	68.94	15.34	2.58	1.08	0.76	21.90
Q3 (mg kg^{-1})	0.81	0.99	4.97	5.99	11.17	148.73	29.45	6.78	4.42	0.81	39.58
Mean (mg kg^{-1})	0.97	1.34	10.09	7.37	7.28	147.85	33.26	7.31	4.37	0.73	29.00
SD	1.48	3.84	19.06	14.79	7.74	268.60	51.87	13.10	9.52	0.10	29.37
RSD (%)	153	286	189	201	106	182	156	179	218	14	101
FV (*n* = 235)	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	Zn
Nbr. of obs	82	187	15	124	174	50	63	115	196		139
Min (mg kg^{-1})	0.01	0.01	0.01	0.03	0.03	0.10	0.06	0.03	0.001		0.05
Max (mg kg^{-1})	12.00	13.00	9.00	548.0	201.7	1660.0	144.0	50.0	78.20		701.40
Q1 (mg kg^{-1})	0.21	0.09	0.50	0.59	1.18	15.62	3.81	0.83	0.43		3.57
Median (mg kg^{-1})	0.50	0.41	0.84	1.80	6.43	131.07	14.93	2.50	1.49		20.60
Q3 (mg kg^{-1})	1.79	1.47	4.33	8.71	12.49	349.00	32.81	10.09	6.00		34.45
Mean (mg kg^{-1})	1.36	1.39	2.61	15.86	10.43	217.82	28.36	7.35	6.63		37.64
SD	2.26	2.63	3.16	65.23	18.06	280.79	36.92	10.35	14.16		83.67
RSD (%)	165	189	121	411	173	129	130	141	214		222
ALL VG (*n* = 838)	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	Zn
Min (mg kg^{-1})	0.01	0.001	0.01	0.03	0.001	0.10	0.06	0.001	0.001	0.45	0.001
Max (mg kg^{-1})	16.0	70.0	58.0	548.0	202.0	2669.0	368.0	350.0	780.0	58.0	950.0
Mean (mg kg^{-1})	1.44	1.88	7.48	16.03	9.92	279.54	43.69	14.05	10.98	5.14	43.84
Median (mg kg^{-1})	0.40	0.41	0.76	3.00	6.09	125.62	21.93	3.10	1.66	0.64	22.84
Rice (*n* = 371)	As	Cd	Co	Cr	Cu	Hg	Fe	Ni	Pb	Se	Zn
Nbr. of obs	251	199	32	109	117	11	27	76	170	8	106
Min (mg kg^{-1})	0.001	0.001	0.001	0.01	0.001	0.001	3.44	0.01	0.001	0.03	0.01
Max (mg kg^{-1})	3.27	17.00	49.89	19.98	123.56	0.77	536.10	46.34	45.75	1.82	121.76
Q1 (mg kg^{-1})	0.10	0.03	0.01	0.18	1.90	0.01	11.90	0.21	0.11	0.06	11.70
Median (mg kg^{-1})	0.17	0.08	0.03	0.65	3.74	0.01	17.20	0.62	0.25	0.17	20.50
Q3 (mg kg^{-1})	0.26	0.29	0.48	1.96	6.74	0.02	74.20	2.26	0.98	1.52	32.00
Mean (mg kg^{-1})	0.25	0.36	3.02	2.16	7.27	0.08	89.01	2.70	1.50	0.69	22.92
SD	0.34	1.42	8.95	3.94	14.19	0.22	147.74	6.11	4.86	0.77	19.82
RSD (%)	133	397	297	183	195	274	166	226	324	112	86
Wheat (*n* = 106)	As	Cd	Co	Cr	Cu	Hg	Fe	Ni	Pb	Se	Zn
Nbr. of obs	49	74	12	54	70	19	25	43	71	6	59
Min (mg kg^{-1})	0.001	0.01	1.04	0.01	0.01	0.001	0.06	0.09	0.01	0.01	0.03
Max (mg kg^{-1})	15.1	2.4	28.0	23.0	23.4	0.2	431.0	40.0	23.8	0.4	169.2
Q1 (mg kg^{-1})	0.05	0.03	1.06	0.18	2.39	0.00	17.98	0.24	0.12	0.03	13.92

Table 1 (continued)

LV ($n=422$)	As	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Se	Zn
Median (mg kg^{-1})	0.20	0.11	1.12	0.47	5.08	0.01	41.10	1.18	0.31	0.15	25.50
Q3 (mg kg^{-1})	0.57	0.89	4.01	1.11	8.24	0.02	70.28	3.76	1.72	0.25	34.81
Mean (mg kg^{-1})	1.59	0.42	4.35	2.46	5.63	0.03	81.56	3.14	1.99	0.16	25.74
SD	3.78	0.56	7.35	4.57	4.52	0.05	117.39	6.36	4.26	0.13	23.79
RSD (%)	238	132	169	186	80	183	144	203	214	86	92
Maize ($n=49$)	As	Cd	Co	Cr	Cu	Hg	Fe	Ni	Pb	Se	Zn
Nbr. of obs	24	35	1	20	16	9	2	27	22		15
Min (mg kg^{-1})	0.01	0.001	15.13	0.03	0.19	0.00	37.35	0.08	0.04		0.21
Max (mg kg^{-1})	2.0	1.3	15.1	10.2	43.9	0.0	99.8	10.3	18.3		59.0
Q1 (mg kg^{-1})	0.03	0.05	0.00	0.13	1.98	0.00	52.96	0.40	0.08		20.10
Median (mg kg^{-1})	0.13	0.10	0.00	0.45	2.97	0.01	68.58	1.00	0.20		34.47
Q3 (mg kg^{-1})	0.29	0.17	0.00	1.35	6.24	0.01	84.19	1.20	0.59		38.75
Mean (mg kg^{-1})	0.26	0.19	15.13	1.35	6.14	0.01	68.58	1.36	1.52		30.08
SD	0.44	0.28	0.00	2.30	10.02	0.01	31.23	2.02	3.97		15.10
RSD (%)	165	144	0	170	163	110	46	149	261		50
ALL GN ($n=526$)	As	Cd	Co	Cr	Cu	Hg	Fe	Ni	Pb	Se	Zn
Min (mg kg^{-1})	0.001	0.0001	0.005	0.009	0.004	0.002	0.056	0.007	0.003	0.013	0.011
Max (mg kg^{-1})	15.10	17.00	49.89	23.00	124.0	0.77	536.0	46.34	45.75	1.82	169.0
Mean (mg kg^{-1})	0.46	0.36	3.64	2.16	6.62	0.04	84.80	2.58	1.63	0.46	24.44
Median (mg kg^{-1})	0.17	0.09	0.34	0.56	4.10	0.01	25.40	0.86	0.25	0.17	21.78

n total number of crop reports, *Nbr. of obs* number of reports for which each elemental data was reported, *Min* minimum, *Max* maximum, *Q1* first quartile, *Q3* third quartile, *SD* standard deviation, *RSD* relative standard deviation, *blank space* data not available, *LV* leafy vegetables, *RV* root vegetables, *FV* fruit vegetables, *RIC* rice, *MAZ* maize, *WHE* wheat, for all countries: *ALL VG* all vegetable crops, *ALL GN* all grains

1.49 mg kg^{-1}, and 20.60 mg kg^{-1}, respectively. The order of TEs in FV was Fe > Zn > Mn > Cu > Ni > Cr > Pb > Co > As > Cd. Overall, the median concentration of TE varied widely ranging from 0.40 mg kg^{-1} (As) to 125.62 mg kg^{-1} (Fe) in the studied vegetable crops with the order of Fe > Zn > Mn > Cu > Ni > Cr > Pb > Co > Se > Cd > As.

Meta-analyses (I^2, Q-statistic, and Tau^2 values) were determined for all the metals individually among vegetable types as follows: As ($I^2=77.24\%$, $Q=105.463$, Tau$^2=0.30$), Cd ($I^2=84.44\%$, $Q=179.90$, Tau$^2=0.18$), Co ($I^2=66.19\%$, $Q=50.2882$, Tau$^2=0.33$), Cr ($I^2=57.54\%$, $Q=61.23$, Tau$^2=0.48$), Cu ($I^2=95.18\%$, $Q=580.7$, Tau$^2=-0.33$), Fe ($I^2=89.90\%$, $Q=217.87$, Tau$^2=-0.16$), Mn ($I^2=94.64\%$, $Q=429.18$, Tau$^2=0.46$), Ni ($I^2=84.42$, $Q=154.0807$, Tau$^2=0.16$), Pb ($I^2=88.94\%$, $Q=253.16$, Tau$^2=-0.014$), and Zn ($I^2=91.19\%$, $Q=317.8$, Tau$^2=0.34$) (Supplementary Table S3; Fig S2). These results show that a significant degree of heterogeneity occurs for Cd, Cu, Fe, Mn, Ni, Pb, and Zn, while the remaining metals, As, Co, and Cr, showed moderate heterogeneity. A higher relative standard deviation (in most cases > 100) also indicates a wide variety of concentrations. Kruskal–Wallis test (Supplementary Table S4) also indicated that there was a significant variation ($p<0.05$) of most of these elements between the vegetable categories, except Se, which has $p>0.05$.

Comparison of Trace Elements in Leafy vs. Root vs. Fruit Vegetables

For a better understanding of the TE accumulation in different vegetable categories, a comparison using median values (Table 1), boxplots (Fig. 2), and the reported concentration values was performed (Supplementary Table S5). Clear differences in concentrations in the edible parts of the three vegetable classes were observed with higher in LV compared to RV and FV (Fig. 2). Among all the studied elements, As, Cd, and Pb are of particular interest due to their highly toxic nature even at low concentrations. As, Cd, and Pb occurred in 25 (RV = 7, LV = 10, FV = 8), 28 (RV = 7, LV = 13, FV = 8), and 28 (RV = 7, LV = 13, FV = 8) vegetable crops, respectively.

The overall As concentration ranged from 0.01 to 16 mg kg^{-1} with a median value of 0.40 mg kg^{-1} (Table 1). The highest concentration was present in spinach (concentration; source country; citation) (16 mg kg^{-1}; India; [51]) along with other LV such as pakchoi (9.15 mg kg^{-1}; China; [52]), coriander (8.58 mg kg^{-1}; India; [51]), and mustard (5.87 mg kg^{-1}; China; [52]). The same LV also had the highest median concentration in the order of coriander (2.05 mg kg^{-1}) > pakchoi (1.97 mg kg^{-1}) > mustard (1.074 mg kg^{-1}) > spinach (0.90 mg kg^{-1}). Other LV such

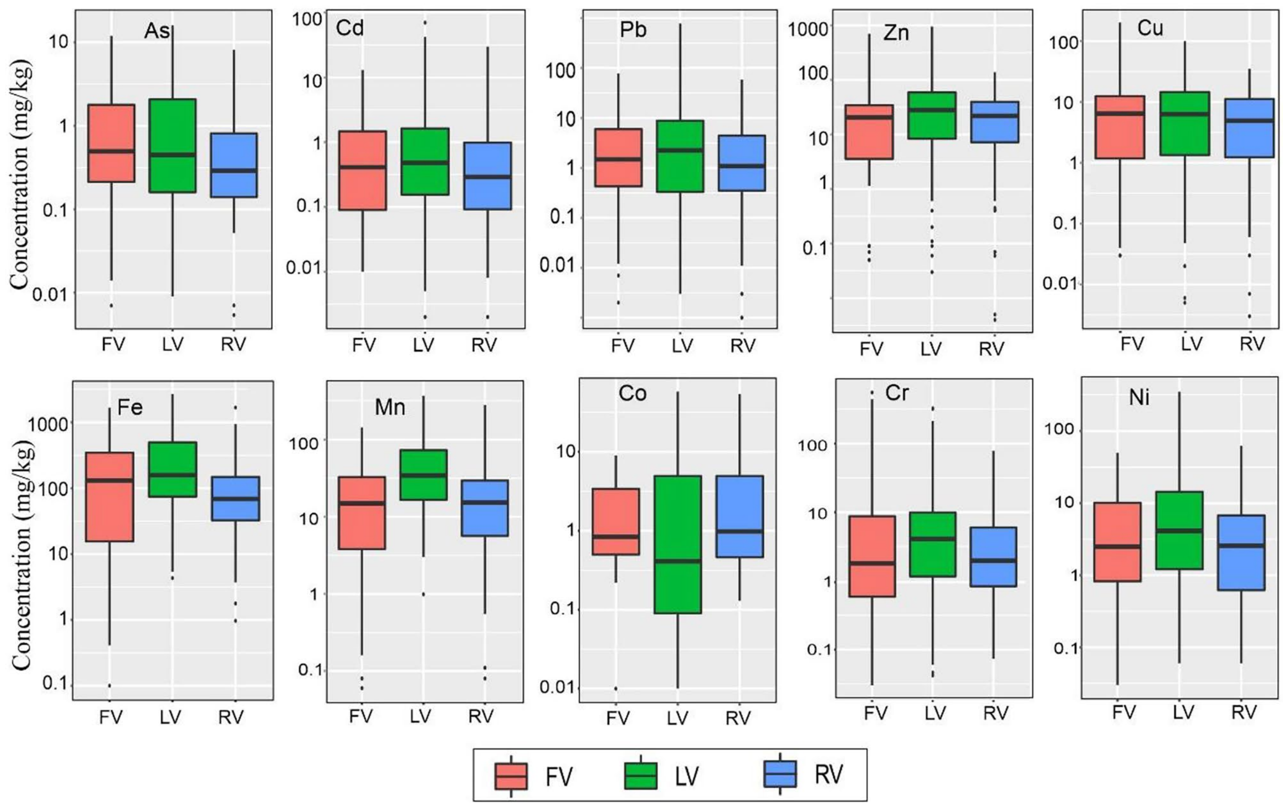
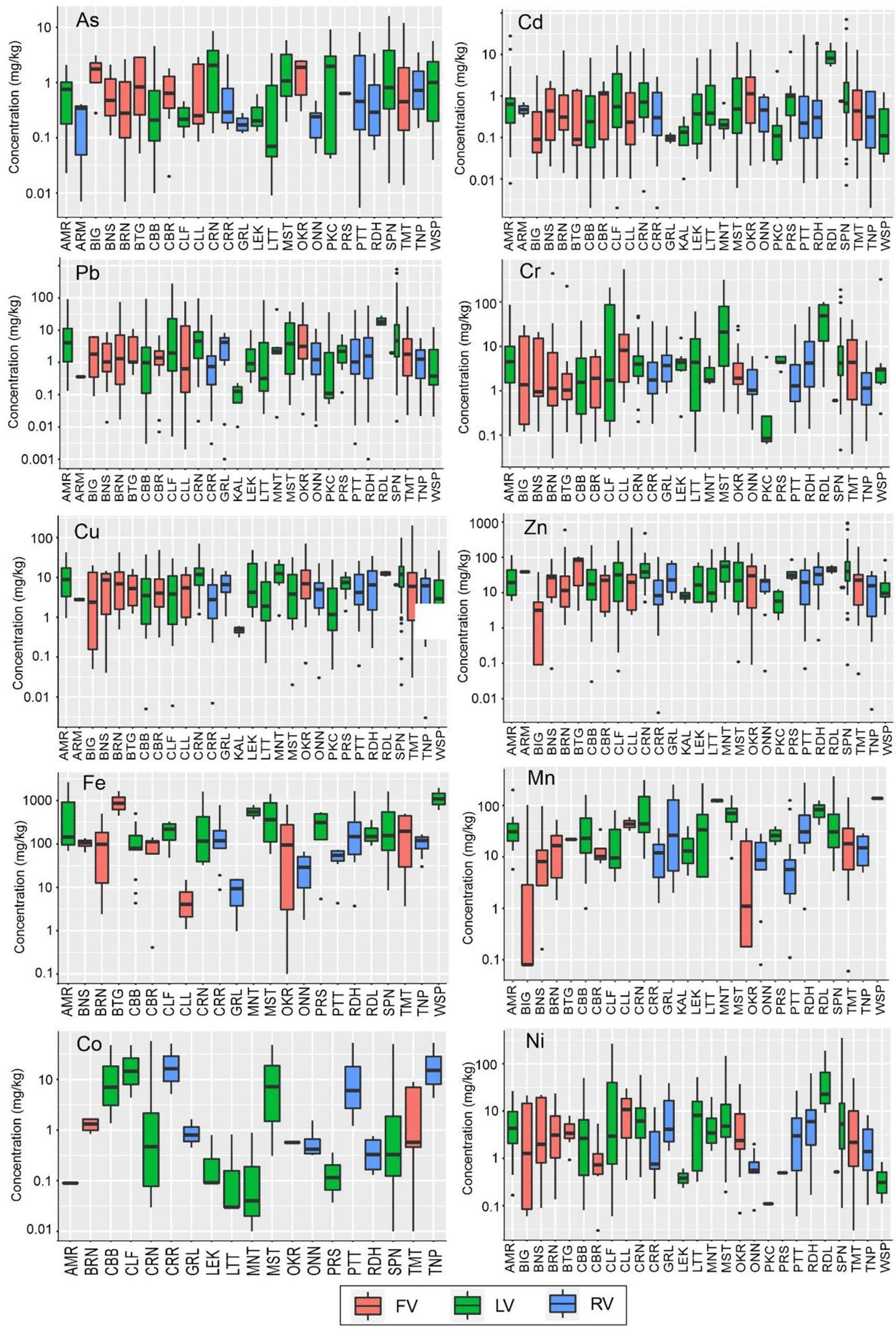


Fig. 2 Box plots showing the differences in trace element accumulation between three vegetable categories (LV, RV, and FV)

as cauliflower, leek, amaranth, lettuce, and cabbage contained relatively lower As concentrations. In FV, tomato (12 mg kg^{-1}; India; [51]) and bitter gourd (3.1 mg kg^{-1}; Bangladesh; [53]) had the maximum As content, whereas the median values were higher in okra (1.88 mg kg^{-1}) and bitter gourd (1.775 mg kg^{-1}). Low As accumulators among the FV were cucumber, brinjal, and beans. Of the three vegetable categories, As accumulation in RVs was the least, with the maximum and median concentration found to be higher in potato (8.2 mg kg^{-1}, Pakistan, [54]) and turnip (3.51 mg kg^{-1}, Pakistan, [55]) and lower in garlic, arum, onion, and carrot (Fig. 3; Supplementary Table S5).

For Cd, the overall concentration ranged from 0.001 to 70 mg kg^{-1} with a median value of 0.41 mg kg^{-1}. Higher levels of Cd were also observed in LV, followed by RV and FV. Elevated Cd levels were found to be in spinach (70 mg kg^{-1}, India, [56]), amaranthus and mustard (28 mg kg^{-1} and 20 mg kg^{-1}, respectively, India, [57]), and leaves of radish (19.18 mg kg^{-1}, India, [58]). However, on the basis of median values, radish leaves had the highest Cd level of 8.2 mg kg^{-1}, followed by parsley (0.98 mg kg^{-1}), coriander (0.71 mg kg^{-1}), and amaranthus (0.63 mg kg^{-1}). Low Cd concentration LV include kale, mint, and pakchoi. Similar to As, top accumulating RV included potato and radish (30 mg kg^{-1} and 18.92 mg kg^{-1}, respectively, India, [59] and [60], respectively) and carrot (2.91 mg kg^{-1}, Iran, [61]) and turnip (1.34 mg kg^{-1}, Saudi Arabia, [62]), whereas low concentrations were recorded in garlic, arum, and onion. Contrary to the reported levels, arum (0.51 mg kg^{-1}) and onion (0.48 mg kg^{-1}) had the highest median concentrations. Among FV, tomato (13 mg kg^{-1}, India, [63]), okra, brinjal, and chilly (13 mg kg^{-1}, 12.6 mg kg^{-1}, and 11.8 mg kg^{-1}, respectively, India, [64]) showed the maximum levels, whereas bottle gourd, cucumber, beans, and bitter gourd exhibited the least. In terms of median, okra (1.135 mg kg^{-1}) and cucumber (1.13 mg kg^{-1}) were the top accumulators of Cd in this category (Fig. 3; Supplementary Table S5).

Pb was recorded at a range of 0.001 to 780 mg kg^{-1} with a median concentration of 1.66 mg kg^{-1}. Similar to As and Cd, Pb was found to be high in LV such as spinach and cauliflower (780 mg kg^{-1} and 280 mg kg^{-1}, respectively, [56]) and coriander and cabbage (96.1 mg kg^{-1} and 94.1 mg kg^{-1}, respectively, [65]), all reported from India and the lowest concentration in kale (0.23 mg kg^{-1}). However, based on median values, LV such as radish leaves and pakchoi showed the maximum and minimum concentrations. FV followed LV in Pb concentration with significantly higher concentrations in chilly (78.2 mg kg^{-1}, China, [66]) and brinjal and okra (76.4 mg kg^{-1} and 73.3 mg kg^{-1}, respectively, India, [64]) and minimum concentration in bitter gourd which on


As
Cd
Pb
Cr
Cu
Zn
Fe
Mn
Co
Ni
Concentration (mg/kg)
FV
LV
RV

◂**Fig. 3** Box plots showing the differences in trace element accumulation in individual vegetable category (LV, leafy; RV, root; FV, fruit) and individual crop (AMR, amaranth; ARM, arum; BIG, bitter gourd; BNS, beans; BTG, bottle gourd; BRN, brinjal; CBB, cabbage; CLF, cauliflower; CLL, chillies; CRN, coriander; CRR, carrot; GRL, garlic; KAL, kale; LEK, leek; LTT, lettuce; MNT, mint; MST, mustard; OKR, okra; ONN, onion; PKC, pakchoi; PRS, parsley; PTT, potato; RDH, radish; RDL, radish leaf; SPN, spinach; TMT, tomato; TNP, turnip; WSP, water spinach)

the other hand had the highest median concentration among the FV. RV had the lowest Pb content among the three vegetable categories, with maximum concentration in radish and potato (59 mg kg^{-1} and 43 mg kg^{-1}, respectively, India, [67] and [59], respectively) and carrot (30 mg kg^{-1}, Pakistan, [68]) and the lowest concentration in arum (0.36 mg kg^{-1}). However, based on the median concentration, garlic and turnip had the highest concentration with the lowest in carrot (Fig. 3; Supplementary Table S5).

The overall range and median concentrations for the remaining elements (Co, Cr, Cu, Fe, Mn, Ni, Se, and Zn) are mentioned in Table 1 from which it is evident that LV had the maximum concentration of all the elements except Cr and Cu, which were higher in FV (Fig. 2). However, based on the median values, Co and Zn in RV and Fe in FV were found to be the highest, while the remaining elements were in LV. Wide variation of metal accumulation between different vegetable species and even within the same group of crops was observed. This could be due to plant physiology differences that influence the absorption and accumulation capabilities and/or geochemical forms of metals in soil and different climatic conditions [69]. Leaves are a significant site for photosynthesis with a rapid development rate and higher transpiration potential resulting in higher accumulation in LV than in other vegetable categories [70–73]. In addition, leaves are the first recipient of contaminants from the atmosphere, while the roots act as a protective barrier for the movement (translocation) of metals into other parts [74]. Furthermore, the large surface area of the edible parts of LV is more susceptible to metal accumulation from the soil, rainwater, and atmosphere particles, particularly in urban and industrial areas [75–77]. LV such as spinach, mustard, coriander, and radish leaves; RV such as potato, turnip, radish, carrot, and onion; and FV like tomato, chilly, okra, brinjal, and bitter gourd were found to accumulate higher amounts of the TE, particularly the toxic ones, and hence can be considered to be unfit for consumption. RV are the first to be exposed to TE from soil and irrigation water, and hence, they can be expected to contain higher levels of metals. Similarly, higher translocation of fluid in FV can lead to metal accumulation. Also, the crops with greater accumulation of the three toxic elements were mostly reported from India and China, along with Pakistan, Bangladesh, and Iran (Fig. 4). Such elevated levels in the crops of these countries might be due to higher elemental availability due to local geological/mineralogical (geogenic: erosion of rapidly uplifted mafic basement rocks and subsequent increases in exposure and erosion) settings and certainly to anthropogenic contributions.

Statistical Analyses of the Occurrence of Trace Elements in Grains

The descriptive statistics of the TE in the major grain crops, RIC, WHE, and MAZ, are listed in Table 1. Overall, the TE concentration in all three types of grain varied widely, ranging from 0.0001 mg kg^{-1} (Cd) to 536 mg kg^{-1} (Fe). For RIC, the concentrations of As, Cd, Pb, and Hg vary from 0.001 to 3.27 mg kg^{-1}, 0.001 to 17 mg kg^{-1}, 0.001 to 45.75 mg kg^{-1}, and 0.001 to 0.77 mg kg^{-1}, respectively. The median concentrations for these elements were 0.17 mg kg^{-1} (As), 0.08 mg kg^{-1} (Cd), 0.25 mg kg^{-1} (Pb), and 0.01 mg kg^{-1} (Hg). Similarly, the overall range (median value) for other elements, i.e., Co, Cr, Cu, Fe, Ni, Se, and Zn, was as follows: 0.001 to 49.89 mg kg^{-1} (0.03 mg kg^{-1}), 0.01 to 19.98 mg kg^{-1} (0.65 mg kg^{-1}), 0.001 to 123.56 mg kg^{-1} (3.74 mg kg^{-1}), 3.44 to 536.10 mg kg^{-1} (17.20 mg kg^{-1}), 0.01 to 46.34 mg kg^{-1} (0.62 mg kg^{-1}), 0.03 to 1.82 mg kg^{-1} (0.17 mg kg^{-1}), and 0.01 to 121.76 mg kg^{-1} (20.50 mg kg^{-1}), respectively. Based on the median concentrations, the overall decreasing order of the TEs for RIC was Zn > Fe > Cu > Cr > Ni > Pb > As > Se > Cd > Co > Hg.

In the case of WHE, the overall concentration of As, Cd, Pb, and Hg ranged from 0.001 to 15.1 mg kg^{-1}, 0.01 to 2.4 mg kg^{-1}, 0.01 to 23.8 mg kg^{-1}, and 0.001 to 0.2 mg kg^{-1}, respectively. Based on the median concentration, the order of the elements was 0.31 mg kg^{-1} (Pb), 0.20 mg kg^{-1} (As), 0.11 mg kg^{-1} (Cd), and 0.01 mg kg^{-1} (Hg). Similarly, the overall range (median value) in WHE for other elements, i.e., Co, Cr, Cu, Fe, Ni, Se, and Zn, was as follows: 1.04 to 28 mg kg^{-1} (1.12 mg kg^{-1}), 0.01 to 23 mg kg^{-1} (0.47 mg kg^{-1}), 0.01 to 23.4 mg kg^{-1} (5.08 mg kg^{-1}), 0.06 to 431 mg kg^{-1} (41.10 mg kg^{-1}), 0.09 to 40 mg kg^{-1} (1.18 mg kg^{-1}), 0.01 to 0.4 mg kg^{-1} (0.15 mg kg^{-1}), and 0.03 to 169.2 mg kg^{-1} (25.50 mg kg^{-1}), respectively. Based on the median values, the decreasing order of TEs in WHE was Fe > Zn > Cu > Ni > Co > Cr > Pb > As > Se > Cd > Hg. Similarly, in the case of MAZ, the concentration (median value) of As, Cd, and Pb varied from 0.01 to 2 mg kg^{-1} (0.13 mg kg^{-1}), 0.001 to 1.3 mg kg^{-1} (0.10 mg kg^{-1}), and 0.04 to 18.3 mg kg^{-1} (0.20 mg kg^{-1}), respectively. The overall order of the metals on the basis of the median values was Fe > Zn > Cu > Ni > Cr > Pb > As > Cd > Hg > Co. The overall order of metals in grains, in decreasing order, is Fe > Zn > Cu > Ni > Cr > Co > Pb > As > Se > Cd > Hg.

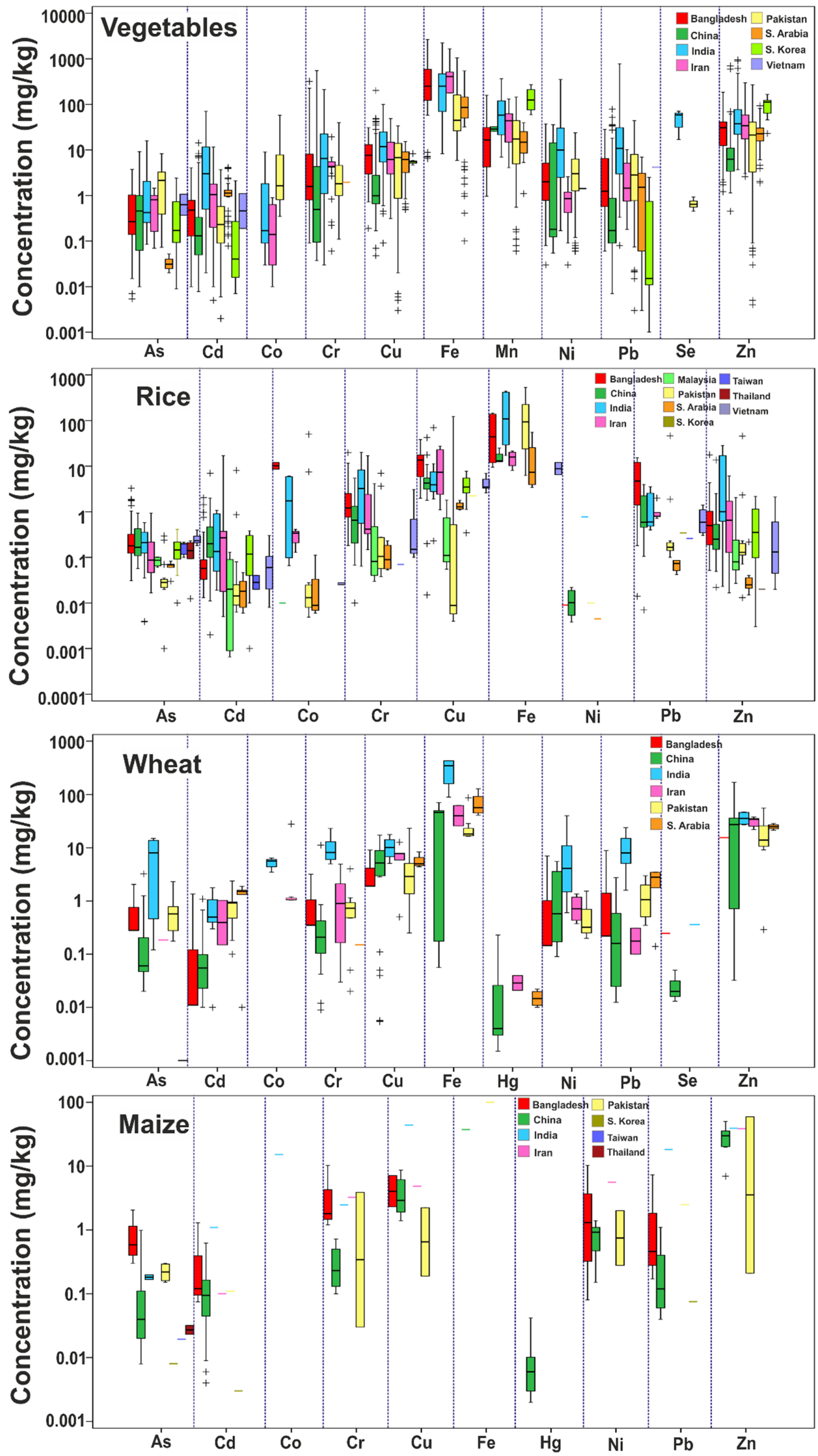


Fig. 4 Concentrations (mg/kg) of trace elements in all crop types in different countries

Trace Element Variability Between Grain Types and Place of Origin

A comparison between the three grain types on the basis of their reported concentration and calculated medians was made to understand the metal-accumulation variability between them. The number of individual observations for the three toxic elements (As, Cd, Pb) was found to be relatively high for all the three crops under study, with the maximum number of observations for RIC than the other two. Considering the overall statistics for the three crops, As was the highest in WHE at 15.1 mg kg^{-1}, while Cd and Pb were in RIC with concentrations of 17 mg kg^{-1} and 45.75 mg kg^{-1}, respectively (Table 1; Fig. 5). However, based on the median values, all these three elements were found to be maximum in WHE (As: 0.2 mg kg^{-1}, Cd: 0.11 mg kg^{-1}, Pb: 0.33 mg kg^{-1}). The details of other elements, including their concentration range and median values for the three grain crops discussed, are mentioned in Table 1.

To determine the origin nation and the underlying variables causing the accumulation, individual grain crops were also examined based on the reported concentrations. RIC was mostly studied for As contamination in all of the selected countries, with predominance in Bangladesh. The overall range of As in RIC was 0.001–3.27 mg kg^{-1} with the maximum concentration reported from Bangladesh [78] and a median value of 0.17 mg kg^{-1}. Other studies from Bangladesh have also recorded similar As levels in RIC [74, 79–83]. The presence of As in RIC samples (as well as other crops) in Bangladesh could be due to the hydrogeological conditions causing extensive release and mobilization of this toxic metal in the groundwater that is very commonly used for irrigation purposes [80]. Other countries such as Iran [84], China [85–88], and India [89] reported rice-As concentration above the permissible limit of 0.2 mg kg^{-1} [90]. Cd ranged from 0.001 to 17 mg kg^{-1} in RIC with the highest concentration in Iran [91], and the median concentration was 0.08 mg kg^{-1}. Elevated levels in Iranian rice grains were also reported by other works [92–95]. Likewise, other nations such as Bangladesh [74, 78, 96–98], China [85, 99–101], India [102, 103], and Pakistan [104, 105] also recorded Cd levels above the safe limit of 0.2 mg kg^{-1}. For Pb, the concentration varied from 0.001 to 45.75 mg kg^{-1} with the highest concentration reported from Pakistan [105] and a median of 0.25 mg kg^{-1}. Mahfooz et al. [106] also quantified Pb at 2.33 mg kg^{-1} in Pakistan. Bangladesh [79, 81, 96], China [52, 70, 85], India [102, 103, 107], Iran [84, 92, 93], and Malaysia [31] were a few countries that reported Pb above the prescribed limit of 0.4 mg kg^{-1}. Also, the median concentrations of As, Cd, and Pb were well within the permissible limit, while the third quartile (Q3) values exceeded the limit for Cd and Pb.

In the case of WHE, As ranged from 0.001 to 15.1 mg kg^{-1}, whereas the median value was 0.2 mg kg^{-1}. The maximum concentration was reported in Uttar Pradesh, India [51]. Only one other study from India recorded As above 0.5 mg kg^{-1}, which is the recommended limit by WHO/FAO [90]. Very few countries, such as China [108], Pakistan [109, 110], and Bangladesh [78] reported As above the safe limit. Cd levels varied from 0.01 to 2.4 mg kg^{-1} with the highest concentration reported in WHE of Pakistan [104] and the median value of 0.11 mg kg^{-1}. Various studies from Pakistan indicated similar Cd levels [111, 112]. Likewise, Saudi Arabia [62], India [102, 113], Bangladesh [78], China [108, 114], and Iran [115] also had Cd levels above the permissible limit of 0.2 mg kg^{-1}. For Pb, the concentration ranged from 0.01 to 23.8 mg kg^{-1} with the maximum value quantified in India [102] and a median concentration of 0.31 mg kg^{-1}. Chandra et al. [113] and Kumar et al. [51] also found similar Pb concentrations in WHE. Other countries that reported levels above the permissible limit of 0.4 mg kg^{-1} were Bangladesh [78], China [108, 114, 116–119], Pakistan [106, 111, 112], and Saudi Arabia [62]. The median of As, Cd, and Pb in wheat was within the recommended limit by WHO/FAO [90], while the Q3 values for all three elements were higher than the permissible limit.

For MAZ, the overall concentration of As varied from 0.01 to 2.04 mg kg^{-1}, and the median concentration was 0.13 mg kg^{-1}. The maximum concentration of As was reported from Bangladesh [78]. Islam et al. [120] and Rahman and Islam [83] have also found As above the WHO/FAO [90] limit. Only China documented As above the safe limit in MAZ of the remaining countries. Cd concentration was also found to be maximum in Bangladesh [78] with an overall range of 0.001 to 1.3 mg kg^{-1} with a median value of 0.1 mg kg^{-1}. Exceedingly high levels were also recorded in India [103] and China [121–123]. Pb concentration in MAZ was the highest in India [103] with an overall range of 0.04 to 18.3 mg kg^{-1}, whereas the median concentration was 0.2 mg kg^{-1}. Other studies that reported Pb above the prescribed limit of 0.4 mg kg^{-1} were [78] and [83] (Bangladesh), [106] (Pakistan), and [124] (China). Similar to RIC and WHE, the median concentration of As, Cd, and Pb in maize was within the safe limit while only the Q3 value of Pb exceeded the limit.

Comparison Between Vegetable and Grain Crops

Since vegetable and grain crops make up an important share of the Asian diet, an evaluation of the differences in the concentration of TEs in these food crops is essential to help determine the crop category that is highly prone to contamination. Considering all studies taken together, the median concentrations of all the studied TEs were higher in vegetables than in grains (median vegetable/median grains (in mg kg^{-1})): As (0.40/0.17), Cd (0.41/0.09), Co (0.76/0.34), Cr (3/0.56), Cu (6.09/4.10), Fe (125.62/25.4), Ni (3.10/0.86),

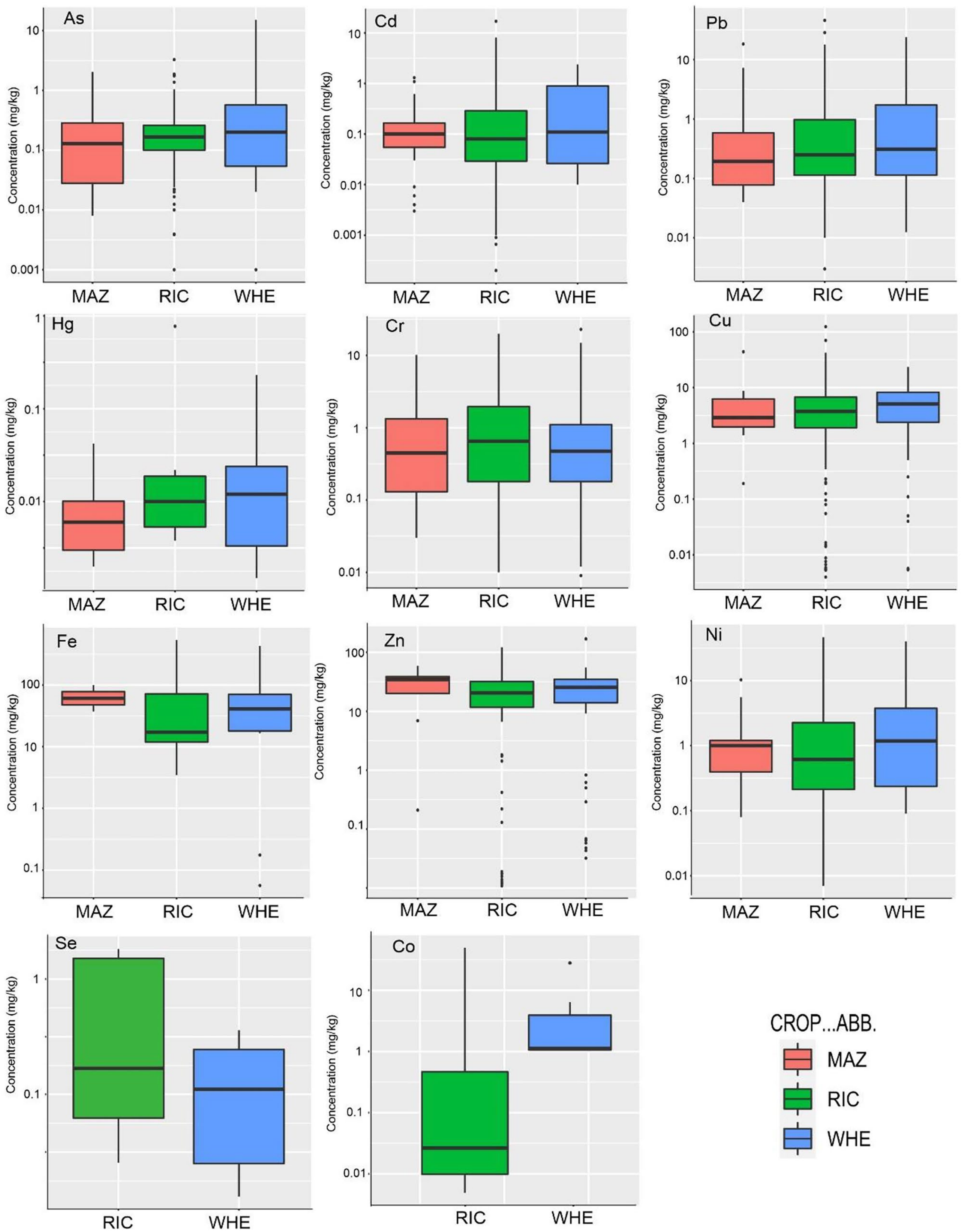


Fig. 5 Box plots showing the distribution of trace element accumulation in rice (RIC), maize (MAZ), and wheat (WHE) in all countries. The box represents the 25th–75th percentiles, and the whiskers represent the 10th–90th percentiles

Pb (1.66/0.25), Se (0.64/0.17), and Zn (22.84/21.78). The data for Mn and Hg were not studied for grains and vegetables, respectively; therefore, a comparison between the two crop categories was not made. Median values for vegetables ranged from 250 to 1900% higher compared to grains, except Zn and Co. Furthermore, all three vegetable classes and the three grain crops were compared on the basis of their Q1, median, and Q3 values. All these three values for Cd, Cr, Fe, Mn, and Ni were found to be higher in LV than in other crop categories. The median for Pb and Q3 value for the remaining TE except Se was also the maximum in LV, whereas the median values for As and Cu were higher in FV, Co in WHE, Se in RV, and Zn in MAZ which were the highest than other crop classes. Overall, this shows that most of the TEs have a higher tendency to accumulate in vegetable crops, especially LV, relative to grains, which may mean they may be more important than grains regarding TEs human uptake. However, the data comparing vegetable categories or vegetables with grains cannot be taken literally when evaluating potential TEs in the food chain. Significant variations in study types within and between countries or authors and laboratories, anthropogenic vs. geogenic settings, urban vs. rural locations, and more will definitely impact results, but the largest lack of control when considering TEs in the food chain comes when considering the social, economic, familiar, religious, and geographic controls on dietary intake and differences in foodstuffs. The above comparisons are a starting point, but must be used with caution in any attempt to make or alter policy related to food security or its potential impact on health.

Bioconcentration Factors (BCF) for Trace Elements in Crops

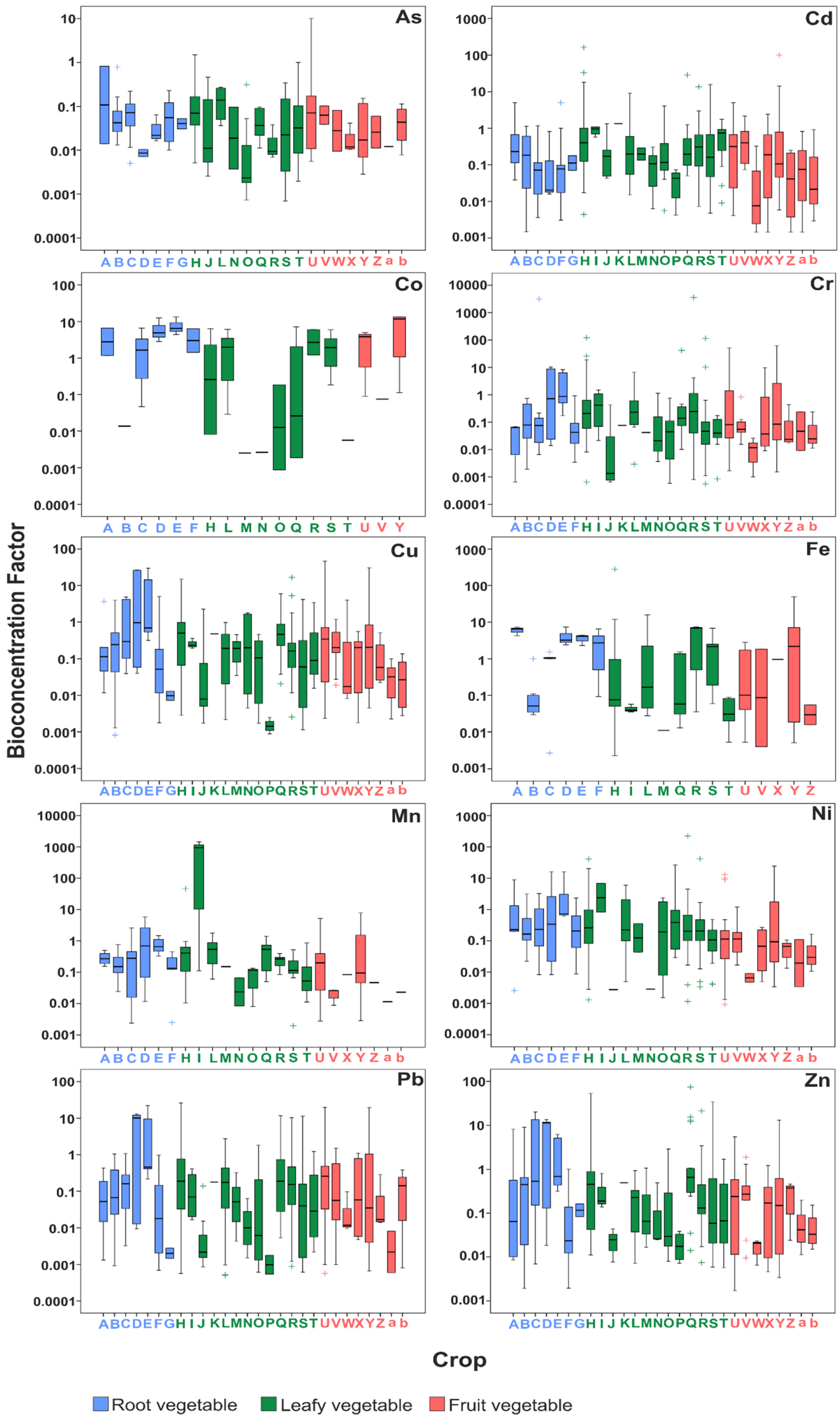
Bioconcentration factor (BCF) is a unitless component used to determine the uptake of metals and other elements from the soil by crops or to certain parts of a crop [125]. BCF is directly and indirectly proportional to the crop and soil concentration, respectively [126]. For the purpose of this work, BCF (BCF = crop: soil) was used to describe the transfer or mobility of metals from contaminated soils and waters to selected vegetables (Fig. 6) and cereal crops (Fig. 7). The pooled average BCF for all the vegetables and cereal crops decreased in the order of Cr > Mn > Fe > Co > Ni > Zn > Cd > Cu > Pb > Se > As and Co > Zn > Cd > Fe > Cu > Cr > Se > As > Ni > Pb > Hg, respectively. The mean BCF values for vegetables ranged from 0.16 to 27.08, of which Cd, Co, Cr, Cu, Fe, Zn, Mn, and Ni were above 1, indicating soil is the potential source of metal uptake and accumulation. As and Se had BCF values below 0.5, implying factors other than soil, such as contaminated irrigation water, are responsible for their accumulation in crops. However, this fact may not be valid for Se due to the insufficient data collected. Pb had a BCF of 0.93, indicating that soil played a major role in its accumulation. In the case of cereal crops, the average BCF values for all the TE ranged from 0.065 to 0.69. Only Co and Zn had BCF values greater than 0.5, indicating that soil concentrations determine their uptake in plants.

Metal-wise boxplots for the three vegetable categories and three cereal crops are shown in Figs. 6 and 7. BCF values for most vegetables have large interquartile ranges (IQR) and significant skewness and a significant number of outliers in some cases (e.g., Cd, Cr, Cu, Ni, Pb, Zn), which are likely indicative that anthropogenic soil contamination (atmospherically or through irrigation and fertilizer application or effluent association with urban/industrial centers) in some specific locations is significantly contributing to metal accumulation. Overall, root vegetables have higher BCF for As, Co, Cr, Cu, Fe, Pb, and Zn than the other two vegetable categories. The most probable reason for this could be that root vegetables are the first ones to come in contact with soil metals and therefore accumulate more metals. Even though the BCF values obtained for some leafy vegetables are < 1, the leafy vegetables have accumulated metals to a greater extent compared with the other vegetables. Considering all the grain crops, essential elements particularly Cr, Cu, and Zn exhibited relatively higher BCF than other elements, whereas among the three non-essential TE, Pb had the minimum. This could be due to the presence of husk which protects the grains from Pb deposition from the atmosphere. For RIC, WHE, and MAZ, the IQR were relatively smaller than for any vegetable with very few outliers for certain metals. RIC had higher BCF for Cr, Cd, Pb, Zn, Cu, and Ni while WHE was higher in the remaining metals except for Fe which was maximum in MAZ.

For all TE, Spearman's correlation coefficients were calculated between BCF value and corresponding soil concentrations to understand the relationships (Supplementary Table S6) better. As mentioned earlier, the soil concentrations and BCF for both grain and vegetable crops had a negative correlation for all the metals which is possibly due to the control of other factors including soil physicochemical properties such as pH and Eh, organic matter content, cation exchange capacity (CEC), soil texture, plant species, which govern their phytoavailability (for details see Supplementary Note 2) and/or metal contamination from other sources, as discussed below.

Probable Sources of Trace Element Contamination in Vegetable Crops

The wide range of concentrations of TE in soils, crops, or irrigation water (Supplementary Table S7, Fig. S4) along with extreme outliers makes average (mean) comparisons difficult, and not significant in some cases. The use of median (Q2) values was chosen as a better descriptor of relative differences between populations and elemental concentrations; the


As
Cd
Co
Cr
Cu
Fe
Mn
Ni
Pb
Zn
Bioconcentration Factor
Crop
Root vegetable
Leafy vegetable
Fruit vegetable

◂**Fig. 6** Box plots showing the BCF of trace elements in LV, RV, and FV (A turnip, B radish, C potato, D onion, E garlic, F carrot, G arum, H spinach, I radish leaves, J pakchoi, K parsley, L mustard, M mint, N leek, O lettuce, P kale, Q coriander, R cauliflower, S cabbage, T amaranth, U tomato, V okra, W cucumber, X chilly, Y brinjal, Z bottle gourd, a bitter gourd, b beans). The box represents the 25th–75th percentiles, and the whiskers represent the 10th–90th percentiles

skewness and kurtosis of the data show extreme asymmetry. These differences are to be expected considering agroclimatic settings, crop management systems, and the different geogenic and anthropogenic factors. Of all analyses (all studies, all elements), soil exhibited significantly highest median values than either crops or water (Fig. S4). This trend points to the importance of a crop relative to the soil in which it was grown, or the quality of the irrigation water, when it comes to interpreting health issues and policies pertaining to agricultural practices. The source of irrigation water can have a dramatic impact on the contamination of agricultural fields and edible crops. This is more pronounced in developing nations as water scarcity has forced farmers to rely on industrial/sewage wastewater for agricultural activities. It was impossible for us to know the origins or level of pollution in most of the waters, other than the sparse analytical results, which were not provided in the vast majority of the studies. For this reason, water concentrations are simply reported based on their average elemental concentrations compared for the category of irrigation water focusing on the trace elements such as, Cd, Cr, Cu, Ni, and Pb. Considering all the data (Fig. S3), SUR water had the highest median values for As, Co, Cr, Fe, Mn, Ni, Pb, and Zn; SWW was highest in Cd; Se was the same in SUR, SWW, and UGW. When duplicates were removed (unique data points), IWW was highest in Cu, Fe, Pb, and Zn; SUR in As, Cd, Mn, and Ni; UGW in Co and Cr; Se remained the same as when all data were considered ($n=2$). Interestingly, SUR had the highest median values for both all data and unique data sets. This may be related to the fact that SUR water is susceptible

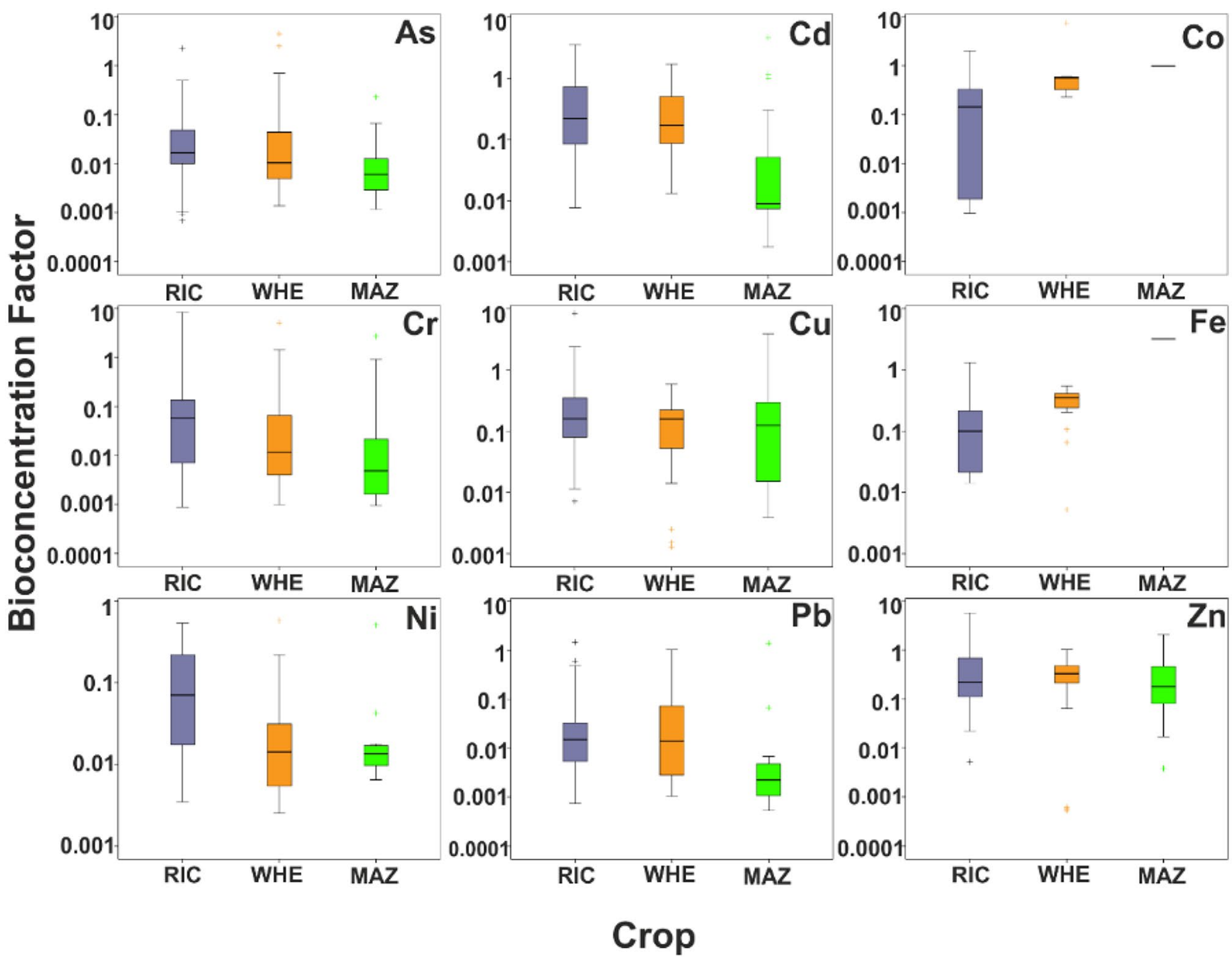


Fig. 7 Box plots showing the BCF of trace elements in rice (RIC), maize (MAZ), and wheat (WHE). The box represents the 25th–75th percentiles, and the whiskers represent the 10th–90th percentiles

to acquiring pollution loads from point and non-point sources. Asian countries dispose of waste materials from rural and urban areas into local surface waters, and it is seldom treated in any way. Massive populations live adjacent to waterways, and this gives rise to significant disposal of virtually all chemicals related to human activity and their associated TEs. Several studies compiled in this study also reported the contamination of crops with toxic TEs due to irrigation with IWW [127–129] and SWW [63, 64, 67, 85, 104, 130, 131]. Other than industrial activities, long-term agrochemical application has also been documented extensively as a major source of TE in soils and crops across the countries under investigation for this work [126, 127, 132–135]. Atmospheric deposition by coal-fired thermal power plants, burning fossil fuels, brick kilns, resuspended road dust, and industrial and automobile emissions are also other important sources of TEs in crops in some cases [136, 137]. Mining and related activities are equally responsible for heavy metal contamination of crops as reported by [86, 138–140]. Metal-contaminated groundwater is one of the major irrigation sources in Bangladesh, Pakistan, and India and has supplied TE to crops [80, 89, 141–143]. These findings indicate that the practice of irrigating vegetables with untreated wastewater/contaminated groundwater by local farmers or vegetables grown in the vicinity of industrial/mining areas can cause a high accumulation of TEs in soils and vegetables, thereby posing a health risk to the population.

Limitations

Data collected for this work came from online databases and therefore is limited to the data provided in each study. There was little consistency in methods from one study to another, and soil and water data were only reported in some studies. These constraints impacted our ability to thoroughly report on some aspects of TEs in crops. However, we could still group important spheres of data, which provide an interesting picture of TE uptake into crops from parts of Asia. Also, considering the limitation in online data availability and search strategy, this study may not give a complete picture of country-wise differences in metal accumulation. Some studies reported extremely high concentrations (which can be an outlier) for certain vegetables that have been reported only once in the literature; they are still included in these discussions even though they skew the data averages. We consider this critical for reporting all values for any given metal; there is no statistical evidence that exceptionally high or low values are outliers, considering the vast differences in irrigation water and soil geochemistry throughout Asia. Very few papers were collected for the maize (*Z. mays*), and for rice (*O. sativa*), the majority of studies were determined to be only As. The referenced studies followed different methodologies and analytical techniques. The TE concentrations in irrigation water were lacking in many studies, as were some soil parameters such as pH, EC, CEC, soil organic matter, clay content, and bioavailable fractions that are important to plant accumulation.

Conclusions

In the present study, a comprehensive database, along with a meta-analysis and critical review of TE accumulation in different vegetable and cereal crops in the Asian context has been presented. The results show considerable variability among crop types for metal accumulation. Among the three different categories of vegetables and grain crops, LV tend to accumulate more metals than other crops. Based on the total concentration, spinach was the highest accumulator of the three non-essential toxic elements (As, Cd, Pb). In contrast, according to the calculated median values, coriander (As) and radish leaves (Cd, Pb) were the top accumulators. Other LV that reported high concentrations of these three elements were mustard, amaranthus, and pakchoi. The top accumulators of these elements in FV were tomato, okra, brinjal, chilly, bottle gourd, bitter gourd, and cucumber, and in RV were turnip, potato, radish, arum, and carrot. These crops also reported higher levels of the other essential elements in their tissues irrespective of their category. The concentration of these metals was lower in grain crops than in vegetables. Based on the total concentration values, wheat reported higher As whereas Cd and Pb in rice were the highest among the three grain crops. However, based on the median concentrations, wheat has the highest concentration of the three non-essential elements than rice and maize. Thus, growing these prolific accumulator crops in a contaminated environment is a serious concern since vegetables, particularly leafy ones, can provide an easy entry into the food chain to these dreaded metals. Furthermore, the median concentration for As was well within the recommended limit, which was not the same for Cd and Pb in the three vegetable classes; thus, this is a major concern for the human consumer in Asian countries, whereas, in the case of cereal crops, all three elements had median values below the prescribed limit. On further analysis, the crops with greater accumulation of the three toxic elements were mostly reported from India and China, along with Pakistan, Bangladesh, and Iran. Studies of TE accumulation in wheat grains are relatively fewer than in rice, with the former reportedly having higher levels of the three non-essential toxic elements. It appears that metal accumulation in crops depends more upon their concentration in irrigation water than in soil. This may be a function of the ability of soils to complex metals in any of the soil exchangeable fractions, which is not the case for water. For irrigation, surface water that might be contaminated from various sources, as well as industrial wastewater and contaminated groundwater, are preferred in Asian countries. However, long-term use of them leads to the accumulation of TE in soils and subsequent contamination of food crops, and in some cases, this contamination can be higher than the recommended limits. Hence, there is a heightened need for policy related to wastewater use in irrigation. Also, there is a need for regular monitoring of TE from effluents and sewage before using for irrigation to prevent their excessive buildup in the food chain and to preserve the health of communities within the vicinity of the contaminated area. Therefore, farmers should be provided with requisite information regarding toxic element content in the soil and the hierarchy of potential metal uptake by different crops that can be grown in their respective zones. Eco-toxicity studies on the consumption of contaminated vegetables are focused more on human health and rarely on livestock and other animals, which should be a higher consideration considering the use of fodder and its TE concentrations.

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Data Availability Data will be available on request.

Compliance with Ethical Standards

Conflict of Interest I hereby declare that there are no known conflicts of interest associated with this publication.

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