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Heavy metals in agricultural cultivated products irrigated with wastewater in India: a review

Bibhabasu Mohanty[a,*] and Anirban Das[b]
[a] Department of Environmental Engineering, Sankalchand Patel University, Visnagar 384315, India
[b] Department of Chemistry, Pandit Deendayal Energy University, Gandhinagar 382007, India
*Corresponding author. E-mail: bibhabasu.mohanty@gmail.com

ABSTRACT

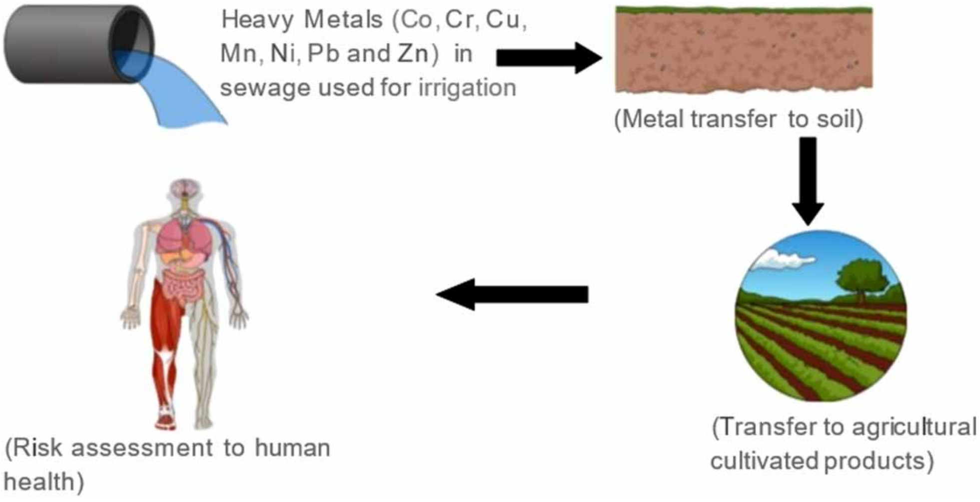
Indiscriminate industrialization and urbanization have negatively impacted our environment. One of the common environmental problems in semi-urban areas in India is the discharge of inefficiently treated municipal, industrial and domestic wastewater into the environment, resulting in the degradation of soil and water qualities. Depleting freshwater resources have led Indian farmers to look for easily available, cheaper, and nutrient-rich sources of irrigation water in the form of wastewater; however, this also led to increased pollutant transfer to the soils. Known as persistent pollutants, heavy metals such as cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb), mercury (Hg), and a few others are potentially hazardous due to their non-biodegradable nature, extended biological half-lives, and biological interactions. These heavy metals can bind to soil surfaces and then be absorbed by plant tissues. Vegetables supply micronutrients, antioxidants, vitamins, and other nutrients essential for human growth. Therefore, the consumption of vegetables/crops grown in wastewater-irrigated land poses a potential threat to humans. Since wastewater irrigation cannot be eliminated in the Indian (semi-)urban areas because of the ever-increasing demand for irrigation water, it is important to assess the impact of wastewater irrigation. This review article congregates the findings of studies from India wherein heavy metal contaminations in vegetables have been reported. An attempt was made to estimate the risk to human health because of the long-term consumption of vegetables cultivated in wastewater-irrigated lands from the Indian sites.

Key words: accumulation, contaminants, heavy metals, irrigation, nonbiodegradable, sewage

HIGHLIGHTS

- Heavy metals in vegetables produced from wastewater-irrigated croplands in India.
- Common sources contribute to heavy metals in soils.
- Geochemistry of heavy metals regulates the transfer factor more than plant physiology.
- Risk to human health associated with heavy metals.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Water conservation is being researched by scientists all over the world. With the increasing population, the demand for freshwater for agriculture and daily use is increasing to the point that it threatens sustainable development in some parts of the world. A decline in the supply of cleaner surface water or groundwater in many semi-urban areas of India has led farmers to seek out readily available and cheaper alternative sources of irrigation water in the form of municipal and industrial wastewater. Wastewater has been used for farming and aquaculture in many parts of the world, including India, for several decades (Amerasinghe *et al.* 2013). Although there are no comprehensive estimates of the overall agricultural area irrigated by wastewater, a study by the International Water Management Institute (IWMI) suggests that in India, roughly 73,000 ha of peri-urban agriculture areas are irrigated with wastewater (Amerasinghe *et al.* 2013). The most common practice in developing and developed countries is to discharge urban (treated and untreated) wastewater into the soil. In developed countries with strict environmental regulations, a significant portion of wastewater is treated efficiently before being used for irrigation. While in many developing countries, the regulations are only sometimes strictly followed, impacting the entire ecosystem adversely.

In India, higher crop production has tempted farmers to use wastewater enriched with vital nitrogen, phosphorous, and potassium (NPK) nutrients; however, as opposed to irrigation using groundwater or other cleaner water sources, this practice has resulted in possible pollutant transfers to crops and vegetation. Among these pollutants, heavy metals pose the greatest risk to the ecosystem and environment because of their toxicity, persistence, bioaccumulation, and nonbiodegradability. Heavy metal-contaminated wastewater can accumulate in the soil to levels beyond their permissible limit when used for irrigation for a long time. Therefore, the crops grown using wastewater irrigation can negatively affect human health.

Fruits and vegetables are essential for human health because they contain carbohydrates, proteins, vitamins, minerals, and fibers needed for human health (Boyd & Rajakaruna 2013). They also neutralize acidic substances that form during the digestive process. In addition to the essential metals, nonessential metals such as aluminum (Al), arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) have a less important role in the human body, and they can be toxic (Khan *et al.* 2008). Heavy metals such as Pb, As, Hg, Cd, and Cr are toxic and ultimately affect human health by creating various health-related issues like kidney failure, cancer, and neural and liver disorders (Kumari *et al.* 2016). The vegetables cultivated in contaminated soils may be exposed to higher levels of heavy metals than others grown in uncontaminated soils. The uptake of different heavy metals into different vegetable tissues depends on the chemistry of the metal and the plant physiology. In contrast, the toxicity of heavy metal depends on its concentration and oxidation state. Plant physiology also plays an important role in heavy metal uptake, e.g., leaves and roots have higher concentrations than stems and fruits (Wong *et al.* 1996; Yusuf *et al.* 2003; Sinha *et al.* 2005).

Since wastewater irrigation cannot be eliminated in the Indian peri-(semi-)urban areas because of the ever-increasing demand for irrigation water, the study also assesses, at least qualitatively and semiquantitatively, how safe it would be to use wastewater for agricultural uses in Indian sites. This article, therefore, compiles the findings of the studies that have focused on wastewater irrigation across India. Furthermore, it attempts to evaluate the impact of wastewater irrigation on the transfer of heavy metals to soils and then to the vegetation grown in each study area. Finally, the objective is to assess the risk to consumers due to heavy metal transfer from vegetation to irrigated soils to humans via the food chain.

2. MATERIALS AND METHODS

2.1. Design of the study

The study was designed based on a systematic review of research works (within India) focused on wastewater irrigation and the accumulation of heavy metals in cultivated products and in soils where the products are grown. The search was conducted in international databases such as Scopus and Sciencedirect, and a few times using the Google Scholar search engine. Keywords such as sewage farming, sewage farming in India, heavy metals, health effects, and transfer factor (TF) were used to narrow the search. Finally, 40 relevant research papers (out of 65) were considered for data analysis. This study covered the urban and rural regions across India where irrigation with wastewater has a long connection.

2.2. Collection of data

A systematic search was conducted to obtain research papers on wastewater irrigation and the accumulation of heavy metals in vegetables. Our search also included heavy metals in soils and the sources of irrigation water used. For selecting research papers, the study related to using wastewater for irrigation, experimental and review-based study, and paper related to the concentration of heavy metals in vegetables. Likewise, papers on transfer factors and health risk assessment were also included. From the finalized papers, a detailed study was done on the concentration of heavy metals in the agricultural cultivated product. For this review paper, data including sources of irrigation (waste)water, types of vegetables, nature of the vegetable (e.g., leafy versus nonleafy), location of the study area, and concentration of heavy metals were analyzed.

2.3. Analysis of data

Data were analyzed based on the concentration of heavy metals in different plant parts and also based on the TF of heavy metals, i.e., the ratio of the concentration of heavy metals in plants to the concentration of heavy metals in soil. Risk-assessment analysis, which indicates the health risk associated with different types of heavy metals and consumption of different types of agricultural cultivated products, was done by using the hazard quotient (HQ) index. Other indices (e.g., enrichment factor of soils and air accumulation factor of heavy elements in vegetables) were discussed wherever data were available.

3. DISCUSSION

3.1. Sources of heavy metals

Both natural and anthropogenic sources contribute to heavy metals in wastewater and irrigated soils. Natural sources are soil erosion, volcanic eruptions, urban runoffs, and aerosol deposition, while anthropogenic sources are identified as effluents from industrial and household activities.

The growth in population and its allied human activities have led to an increase in the concentration of heavy metals in wastewater. Hence, urbanized areas play an important role in the availability of metals in the sewage and soil (Buzier *et al.* 2011). The wastewater generated from households is a major component of urban wastewater. Such sources are food, detergents, cosmetics, sanitary fittings, kitchen wastewater, laundry, bath, and water closets (Moriyama *et al.* 1989). The possible sources of various heavy metals in domestic wastewater are listed in Table 1.

Industrialization has also resulted in heavy metal contamination in the soil. Toxic compounds accumulate in the soil due to rapid industrialization and disorderly urbanization, as well as long-term usage of enormous amounts of fertilizers and pesticides (Kishan *et al.* 2014; Kumar *et al.* 2015; Rodriguesa *et al.* 2017). The main sources of emissions induced by human activities are liner source (from vehicle transportation), surface source (household emission), and point source (fuel combustion, industrial exhaust through stack). Various heavy metals are generated from different industrial sources, such as chrome from the paint and metallurgical industry, nickel from steel, and cadmium from smelters. Road traffic also leads to the generation of certain heavy metals like lead. Tetraethyl lead was added to gasoline for many years, and its combustion generates Pb in the natural environment (Niec *et al.* 2013; Zhou *et al.* 2016), but this source is declining with time. Several investigations reported a significant amount of street dust in pollutants of urban soils along traffic routes (Zereini *et al.* 2007;

Table 1 | Sources of heavy metals in domestic wastewater

Metal	Sources	References
Pb	Sewer hookups and pipelines, wastewater from laundry	Comber & Gunn (1996), Meinzinger & Oldenburg (2009)
Cr	Cleaning of metal cookware	Sorme & Lagerkvist (2002)
Ni	Stainless steel materials, cleaning of cookware, feces,	Moriyama *et al.* (1989), Houhou *et al.* (2009)
Zn	Food, plumbing materials, detergents, toothpaste, shampoo, and deodorants	El Khatib *et al.* (2012)
Fe	Food coloring, diet supplements, steel and iron products, water supply pipes, paints, and cosmetics	Tjadraatmadja & Diaper (2006)
As	Medicines, washing products, glass, wood preservatives, garden products	Ismail *et al.* (2013)
Mn	Pesticides, food products like grains, nuts, tea leaves	Chino *et al.* (1991)
Cu	Copper piping corrosion	Meinzinger & Oldenburg (2009)
Cd	Washing powder, dish wash detergents	Chino *et al.* (1991)

Wiseman *et al.* 2015). Roads with heavy traffic could also be a source of pollution due to the generation of metal-enriched vehicular exhaust (Sternbeck *et al.* 2002; Wiseman *et al.* 2015).

Heavy metal contamination is a concern in soils near industrial districts (Murray *et al.* 2011; Sun & Chen 2018; Sung & Park 2018), adversely affecting nature. Mahmood & Malik (2014) reported that wastewater irrigation affects soil's physico-chemical properties. According to various Indian studies, when wastewater is used to irrigate crops, the quantity of heavy metals increases in the soil as well as in agricultural cultivated products (Rattan *et al.* 2005; Sharma *et al.* 2006; Singh & Kumar 2006; Singh & Agrawal 2007; Arora *et al.* 2008; Gupta *et al.* 2008a, 2008b; Tiwari *et al.* 2011).

The concentration of elements in the soil regulates the trace metal content of vegetables. Bedrock is a natural source of these metals in the soil. Metals are also introduced to agricultural soils in combination with organic and mineral fertilizers (mainly calcium and phosphates) (Antisari *et al.* 2015; Ye *et al.* 2015). Herbicides and fungicides are also sources of metals in the soils (Kabata-Pendias & Pendias 1999; Antisari *et al.* 2015; Ye *et al.* 2015).

The soil conditions play a very important role in the bioavailability of heavy metals and their uptake by plants. These conditions include soil pH, soil type, (soil) humidity, and the initial concentration of metal found in the soil (Kabata-Pendias & Pendias 1999; Jarup 2003; Leitzmann 2003). For example, granulometric soil tended to accumulate more cadmium in the soil (Wlasniewski & Hajduk 2012). They also reported that the cadmium content of the crops studied (wheat) was linked, to a small extent, to the features of the top layer of soil, including total and soluble cadmium levels in the soil. According to Kabata-Pendias & Pendias (1999), when the soil pH is acidic, it generally enhances crops' heavy metal absorption. Bielecka *et al.* (2009) reported that the pH of the soil is in the range of 7.1–8.1, and the presence of a higher concentration of organic matter (OM) lowers the bioavailability of metals like Cd in the soil. Hence, there is a decrease in the uptake of metal by plants.

3.2. Heavy metals in vegetables

Figure 1 indicates the different study locations of India whose findings have been covered in this article. Figures 2–8 indicate the mean concentration of various heavy metals, for example, Zn (Figure 2); Cu (Figure 3); Mn (Figure 4); Cr (Figure 5); Cd (Figure 6); Pb (Figure 7), and Ni (Figure 8) from those locations. Likewise, the concentration of heavy metals in leafy and nonleafy vegetables from several parts of India has been reported in the literature, and these are reported in Tables 2 and 3.

Heavy metal concentrations vary from below the detection limit to above the threshold limit depending upon the contamination source and the type of cultivated agricultural product. Singh & Kumar 2006 measured heavy metal concentrations in spinach and lady's finger cultivated in Delhi's peri-urban districts, contaminated by industrial effluents, sewage sludge, and vehicle exhaust. Spinach had heavy metal concentrations ranging from 7 to 50 mg kg^{-1} for Cu, 51 to 282 mg kg^{-1} for Zn, 1.4 to 9.0 mg kg^{-1} for Cd, and 1.7 to 9.2 mg kg^{-1} for Pb; the concentration of heavy metals in spinach was found to be higher than in lady's finger. This variance is attributed to variations in root interception of metal ions, the entrance of metal ions by mass flow and diffusion, translocation of metal ions from root to shoot, accumulation tendency, and retention capacity (Carlton-Smith & Davis 1983). Sharma *et al.* (2006) found Cu (0.55–10.30 mg kg^{-1}), Zn (29–469 mg kg^{-1}), Cd (1.55–6.90 mg kg^{-1}), Pb (9.0–28.0 mg kg^{-1}), Ni (4.05–15.00 mg kg^{-1}), and Cr (2.75–51.15 mg kg^{-1}) in the edible sections of crops

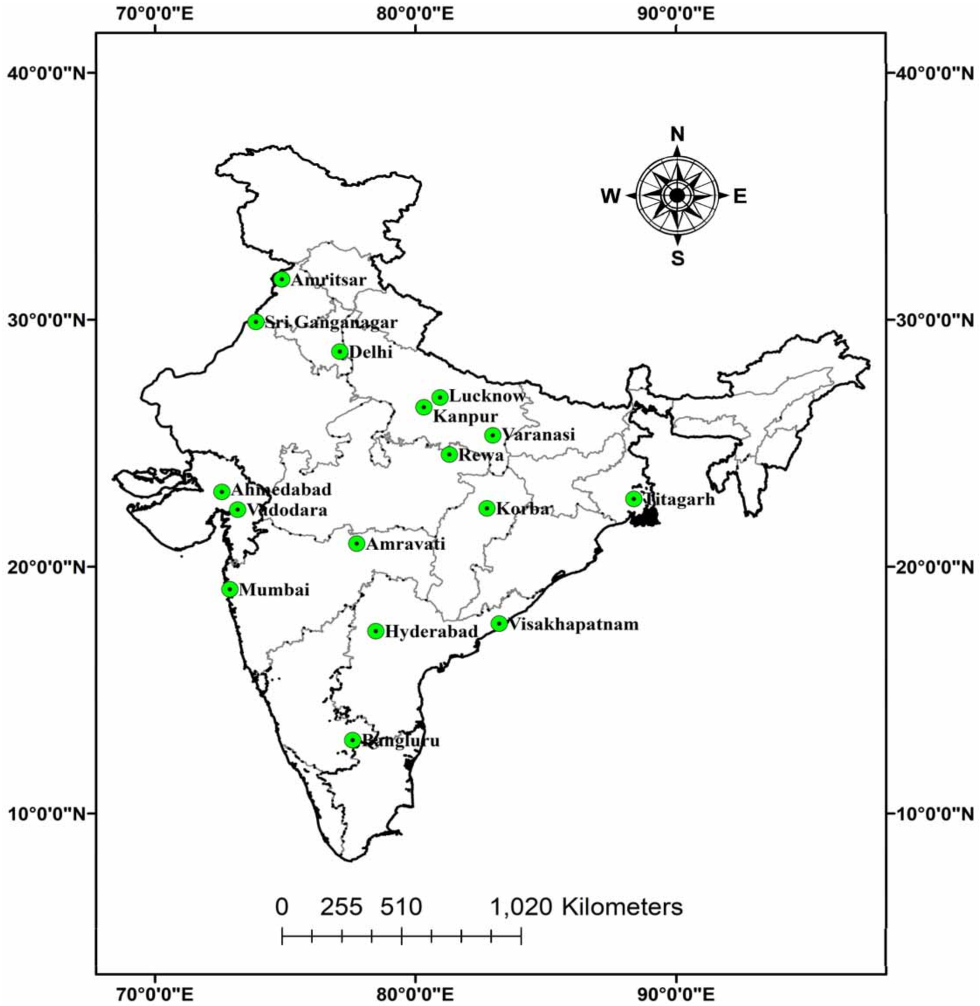


Figure 1 | Different study locations in India.

cultivated in late autumn in suburban Varanasi, India. Leafy vegetables (spinach, amaranthus, and cabbage) have higher levels of Cu and Cr than nonleafy vegetables (brinjal, lady's finger, and tomato). In the Dinapur site in Varanasi, higher levels of Cu (16.5 mg kg^{-1}), Pb (16.0 mg kg^{-1}), and Ni (7.5 mg kg^{-1}) were measured in the winter than in the summer: e.g., Cu (15.0 mg kg^{-1}), Pb (4.0 mg kg^{-1}), and Ni (3.0 mg kg^{-1}; Sharma *et al.* 2007). Since OM decomposes quickly in the summer, heavy metals are released into the soil solution for plant uptake. The risk to human health by heavy metals (Fe, As, Cr, Mn, Cu, Zn, Pb, Cd, and Hg) by the consumption of tomato, brinjal, amaranthus, spinach, and coriander obtained from the largest coal-burning basin, Korba, India, was evaluated by Ramteke *et al.* (2016). The soils of the studied area were enriched with As, Cd, and Pb; these authors calculated Al-normalized mean enrichment factors (with respect to crustal abundances) of 27 (As), 19 (Cd), and 11 (Pb). These enriched values were largely attributed to coal-burning activities. Among the different vegetables studied, a leafy vegetable like spinach was reported for the highest metal pollution of 16.2 mg kg^{-1} compared to a nonleafy vegetable like brinjal of 3.1 mg kg^{-1}. The higher biomass production of leafy vegetables may be the reason for higher metal pollution. Due to the higher accumulation of metals in spinach, high health risk index was reported. Similarly, high health risk index was reported for As.

To report the substantial buildup of heavy metals in vegetables irrigated with wastewater, Arora *et al.* (2008) conducted a study for Sri Ganganagar, Rajasthan, where different heavy metals like Fe, Mn, Cu, and Zn were assessed in vegetables irrigated with water from different sources. The range of various metals in wastewater-irrigated plants was 116 – 378 mg kg^{-1} (Fe), 12 – 69 mg kg^{-1} (Mn), 5.2 – 16.8 mg kg^{-1} (Cu), and 22 – 46 mg kg^{-1} (Zn). Mint and spinach had the highest mean

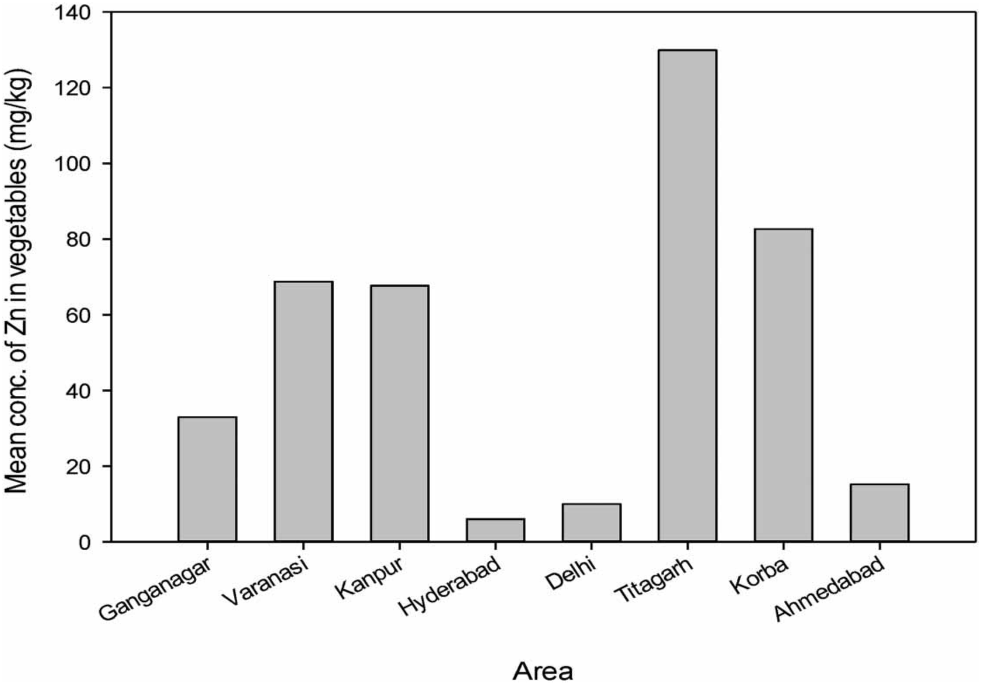


Figure 2 | Mean concentration of Zn in vegetable samples collected from different locations. WHO and Indian Standard for Zn safe limit are 60 and 50 mg kg^{-1}, respectively.

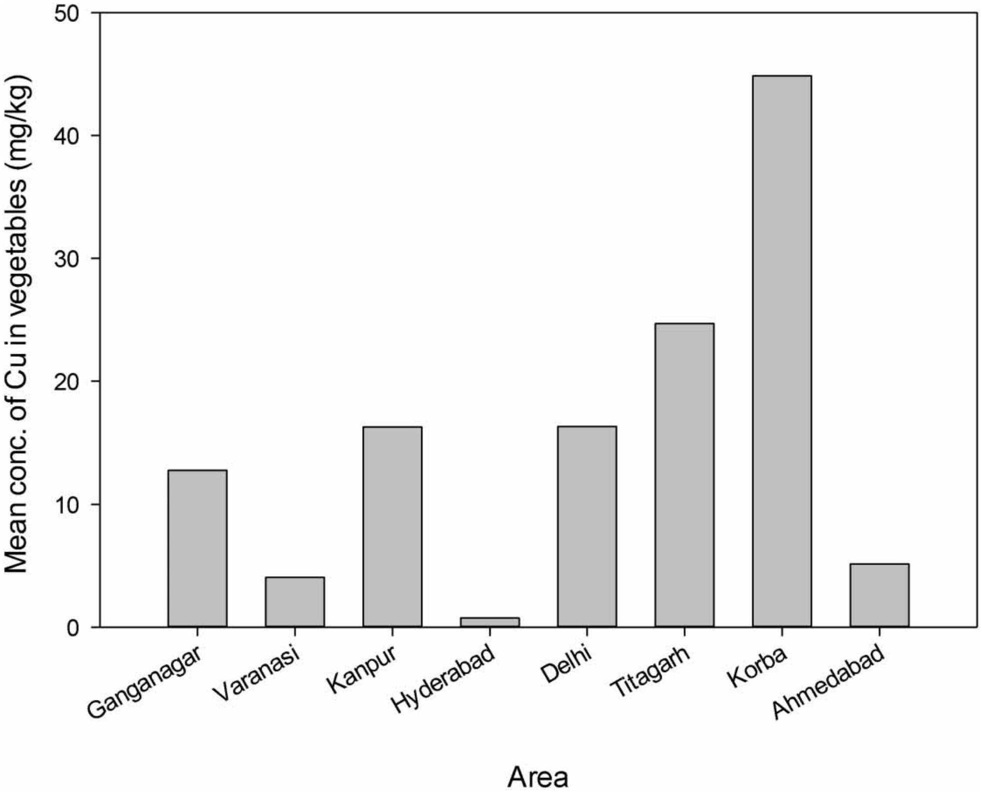


Figure 3 | Mean concentration of Cu in vegetable samples collected from different locations. WHO and Indian Standard for Cu safe limit are 40 and 30 mg kg^{-1}, respectively.

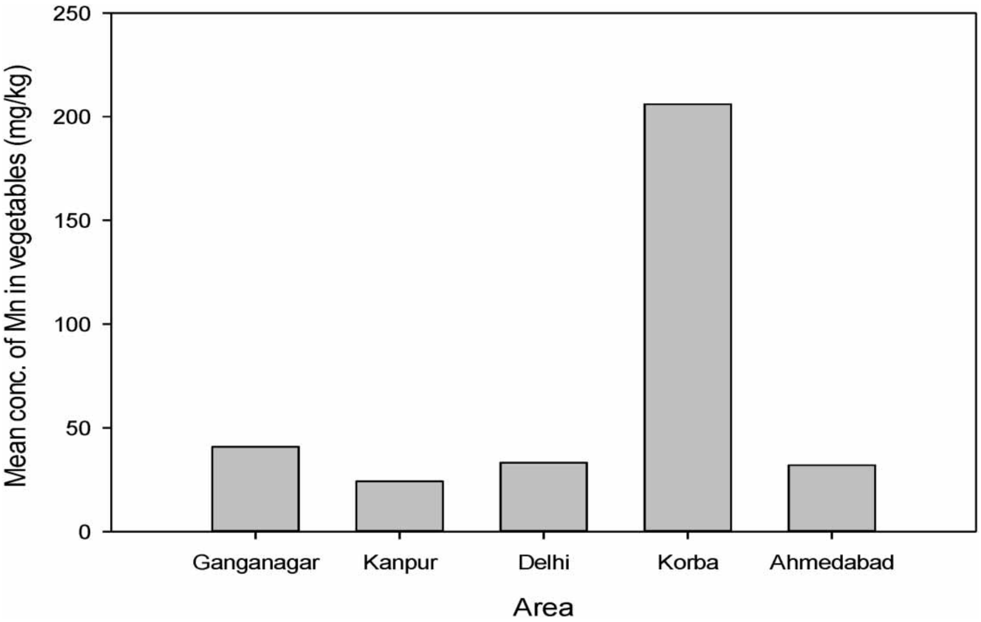


Figure 4 | Mean concentration of Mn in vegetable samples collected from different locations.

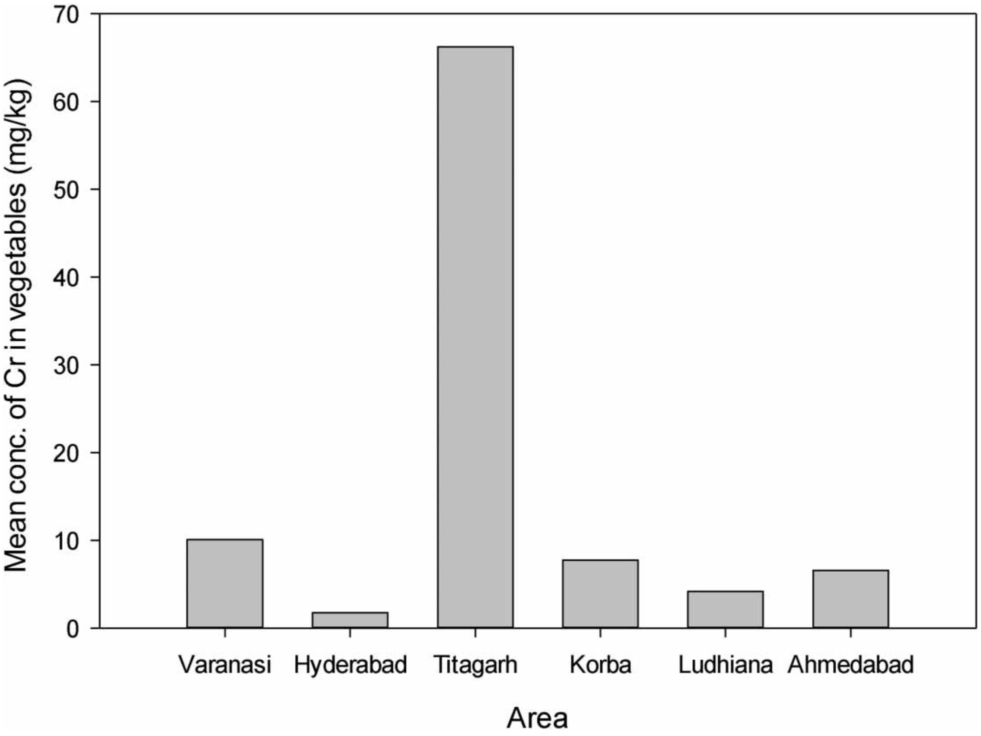


Figure 5 | Mean concentration of Cr in vegetable samples collected from different locations. Indian Standard for Cr safe limit is 20 mg kg^{-1}.

Fe and Mn levels, while carrots had the highest Cu and Zn levels. According to the findings of this study, both adults and children who eat vegetables cultivated in wastewater-irrigated soils consume a significant amount of these metals. However, the values of these metals were below the Food and Agriculture Organization of the United Nations (FAO) and World Health Organization (WHO)-recommended maximum tolerable levels (1999). However, continuous monitoring of levels of these metals in effluents and sewage, vegetables, and other food materials is required to prevent an increased buildup of such

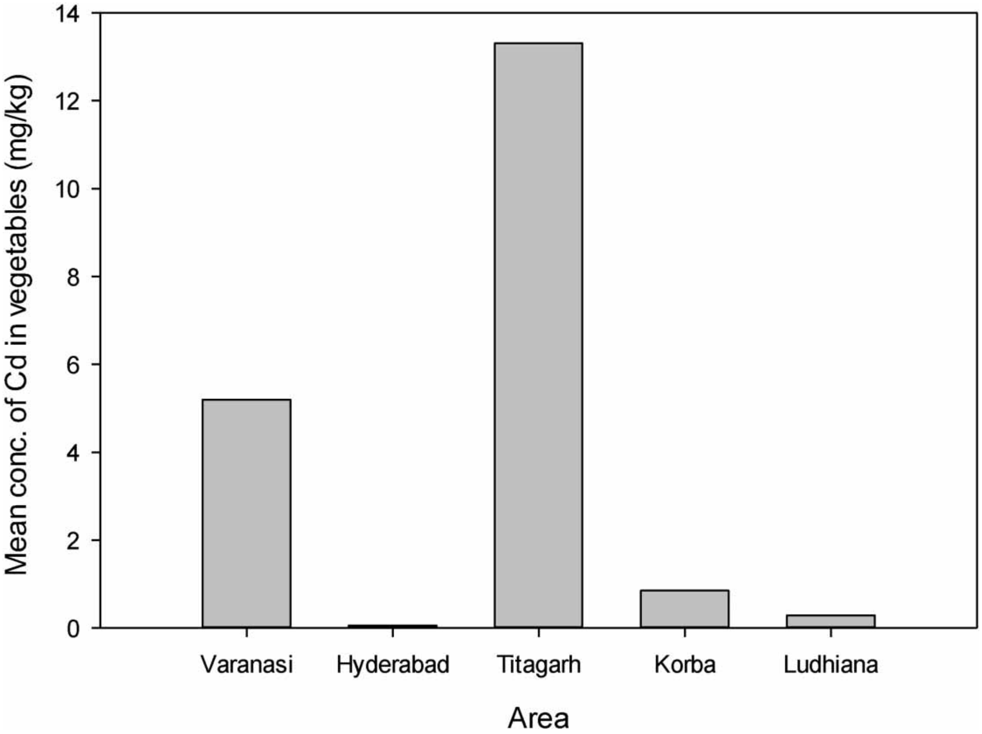


Figure 6 | Mean concentration of Cd in vegetable samples collected from different locations. WHO and Indian Standard for Cd safe limit are 0.2 and 1.5 mg kg^{-1}, respectively.

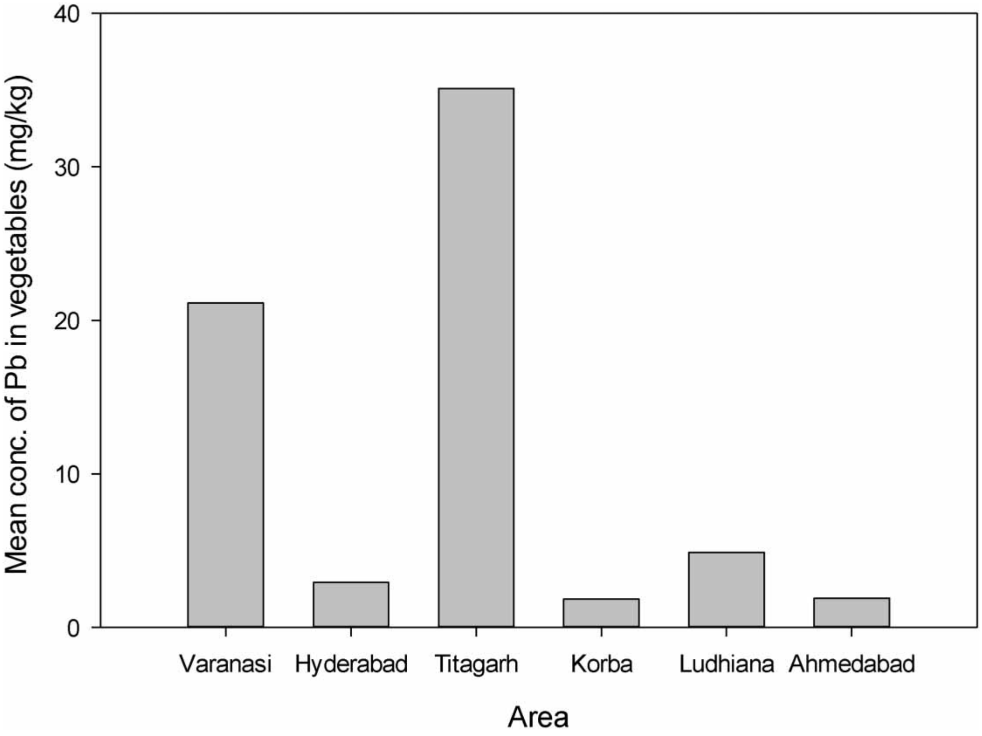


Figure 7 | Mean concentration of Pb in vegetable samples collected from different locations. WHO and Indian Standard for Pb safe limit are 5 and 2.5 mg kg^{-1}, respectively.

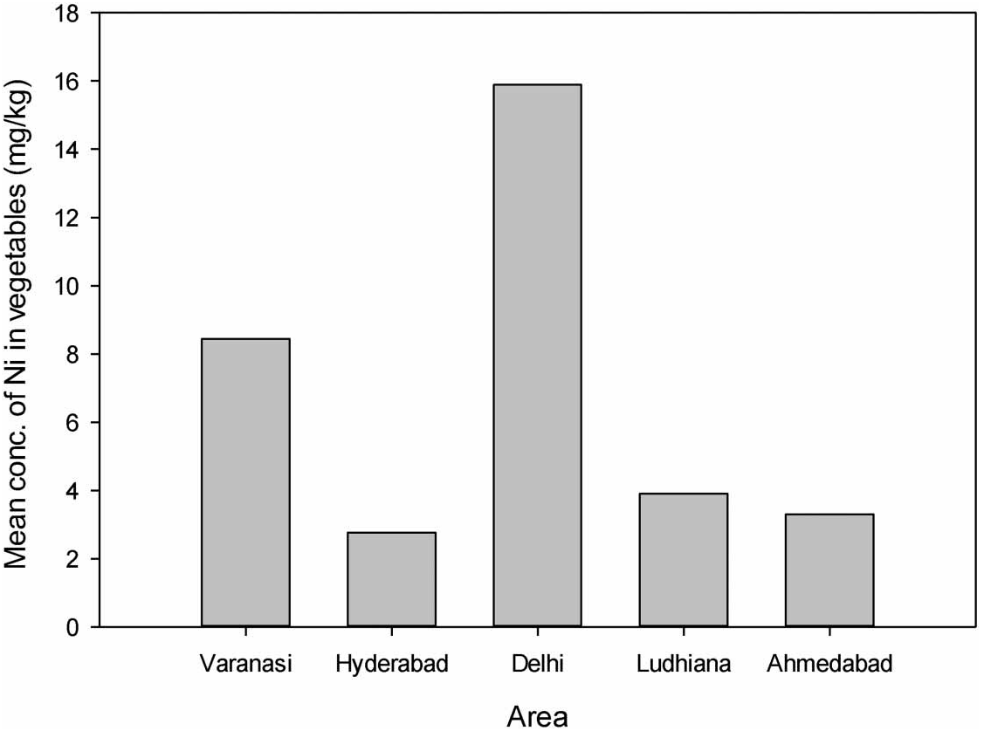


Figure 8 | Mean concentration of Ni in vegetable samples collected from different locations. Indian Standard for Ni safe limit is 1.5 mg kg^{-1}.

metals in the food system. A study for the stretch of the Musi River of Hyderabad was conducted by Chary *et al.* (2008), from which 12 soil and forage grass samples, 46 (spinach), 36 (amaranthus), 40 (mint leaves), 38 (coriander), 33 (brinjal), 30 (lady's finger), and milk samples were collected to analyze Zn, Cr, Cu, Ni, Co, and Pb in the samples. The concentrations of Zn (227–401 μg g^{-1}) and Pb (303–580 μg g^{-1}) were found above the permissible limits of Indian standards in collected soil samples (Zn 300 and Pb 250 μg g^{-1}). The wastewater generated from various anthropogenic sources like the electroplating industry, battery manufacturing, pharmaceutical waste, and disposal from domestic sources into the Musi River contributes to it. A fractionation study indicates that 30–35% of Zn and 25–30% each of Cr and Cu are associated with the mobile fraction of the soil and available for plant uptake. The acidic-to-moderate alkali nature of soil pH (5.9–7.3) and high organic carbon (4.9–6.2%) also favor this bioavailability. In the analyzed forage grass, transfer of heavy metals (see next section) from soil to plant was calculated for Zn (0.53–0.68), Cr (0.70–0.95), and Cu (0.76–0.84), and it indicates root uptake of such metals by forage grass. Metals like Zn, Cr, and Cu were also found in the analyzed milk sample from animals eating the forage grass, indicating a higher transfer. Among the analyzed vegetable samples, metals like Zn, Cr, Cu, and Pb are found to be the maximum. Leafy vegetables like spinach and amaranthus accumulate higher concentrations of metals; the high translocation and transpiration rate favors the transfer of metals from root to stem and to fruit in leafy ones. The heavy metal uptake by plants has shown that the metals can passively be transported through the xylem vessel from root to shoot. However, their redistribution into storage organs depends largely on the phloem, and heavy metals have low mobility in the phloem (Ghosh *et al.* 2012).

To evaluate the consequences of wastewater irrigation, Rattan *et al.* (2005) collected and analyzed soil, vegetable, millets, cereals, and fodder crops from agricultural land near Delhi. The concentrations of Zn, Cu, Fe, Mn, Ni, Pb, and Cd were determined, and it was found that the metal concentration in effluents was within the corresponding permissible Indian Standard limits. In the analyzed soil samples, Zn and Cu concentrations exceeded the phytotoxicity limits. The fractionation study of the soil sample reported that Zn (38.3%) and Cu (23.8%) were in the bio-available pool. The metal concentration in the analyzed vegetable samples indicates that the concentration of heavy metals was within the permissible limit in terms of phytotoxicity. In order to provide information regarding the presence of various heavy metals (Pb, Zn, Cd, Cr, Cu, and Ni), vegetables like lettuce, pudina, cauliflower, spinach, coriander, and radish were collected from the Titagarh town of

Table 2 | Concentration of heavy metals in the edible portion of leafy vegetables

Study area	Heavy metals	Sources of heavy metals	Types of sample vegetables	Key findings	References
Outskirts of Bombay city	Pb, Cd, Cu, and Zn	Atmospheric deposition	Amaranthus	The highest concentration of Cd	Tripathi *et al.* (1997)
Kanpur	Fe, Cr, Zn, Mn and Cu	Treated tannery wastewater is a source of pollution	Amaranth, spinach, coriander,	A higher accumulation of Cr in edible parts of leafy vegetables was observed	Sinha *et al.* (2006)
Amritsar, Punjab	Cd, Cu, Fe, Pb, and Co	Effluents from industrial units	Coriander, fenugreek, mint, spinach	Spinach was the most hazardous for cobalt, and copper was highest in spinach	Sharma *et al.* (2016)
Rewa, Madhya Pradesh	Fe, Zn, Cu, Pb, Cd, Mn, and Cr	Effluents from a cement factory	Spinach	Pb, Cd, and Cr were higher in spinach when compared to nonleafy plants	Chauhan (2014)
Sri Ganganagar district, Rajasthan	Fe, Mn, Cu, and Zn	Continuous and long-term irrigation with wastewater	Spinach, mustard, peppermint, coriander	Spinach has shown a higher accumulation of Fe, Mn, Cu, and Zn,	Arora *et al.* (2008)
Hyderabad	Zn, Cr, Cu, Ni, Co, and Pb	Sewage disposal to the river and use of river water as a source of irrigation	Forage grass, coconut, spinach, amaranthus, coriander leaves, mint leaves	High amounts of Zn followed by Cr and Ni in spinach and amaranthus	Chary *et al.* (2008)
Delhi	P, K, S, Zn, Cu, Fe, Mn and Ni	Sewage effluents are the primary source of pollution	Gobhi, Sarson, spinach, cauliflower	Spinach, gobhi, and sarson have a comparatively higher accumulation of heavy metals than nonleafy ones	Rattan *et al.* (2005)
Suburban of Varanasi	Cu, Zn, Cd, Pb, Ni, and Cr	Industrial effluents	Spinach, amaranthus, cabbage	Cu and Cr were higher in leafy vegetables	Sharma *et al.* (2006)
Lucknow	Cr, Ni, Zn, Cu, and Cd	Effluent from an electroplating industry	Spinach	Leafy vegetables have a greater accumulation	Pandey (2006)
Bangalore	Fe, Zn, Cu, Ni, Cr, Pb, and Cd	Sewage is the main source of pollution	Spinach	Heavy metal contamination in vegetables is much in leafy vegetables when compared to nonleafy vegetables	Lokeshwari & Chandrappa (2006)
Varanasi	Cd, Ni, Cu, Cr, Pb, and Zn	Sewage sludge	Spinach	Increase heavy metal concentration of Cd, Ni, and Zn	Singh & Agrawal (2007)
Northern India	Cd, Cr, Ni, and Pb	Treated sewage	Spinach, cabbage	Higher concentration of heavy metals in leaf when compared to other storage organs	Ghosh *et al.* (2012)
Titagarh, West Bengal	Pb, Zn, Cd, Cr, Cu, and Ni	Municipal wastewater irrigation	Lettuce, pudina, cauliflower, spinach, coriander	Pb, Zn, Cd, Cr, and Ni concentrations in all the leafy vegetables were beyond the safe limits	Gupta *et al.* (2008a, 2008b)
Korba, Chhattisgarh	Fe, As, Cr, Mn, Cu, Zn, Pb, Cd, and Hg	Industrial deposition	Amaranthus., spinach, coriander	High health risk index for As in all the tested vegetable	Ramteke *et al.* (2016)
Vadodara, Gujarat	As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn	Use of industrial effluent for irrigation	Spinach, coriander, cabbage, dill	Spinach and cabbage showed high accumulation and translocation of As, Cd, Cr, Pb, and Ni in their edible parts	Tiwari *et al.* (2011)

Table 3 | Concentration of heavy metals in the edible portion of nonleafy vegetables

Study area	Heavy metals	Sources of heavy metals	Types of sample vegetables	Key findings	Researcher
Visakhapatnam, Andhra Pradesh	Pb, Zn, Ni, and Cu	Deposition of metals due to emissions from industrial and transport sectors	Tomato, lady's finger, capsicum	Higher levels of Pb concentrations in vegetables grown in an industrial area	Srinivas *et al.* (2009)
Amba Nalla in Amravati City, Maharashtra	Pb and Cd	Wastewater from domestic sources	Tomato	Concentration more than the permissible limit	Mohod (2015)
Vadodara, Gujarat	As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn	Use of industrial effluent for irrigation	Radish, tomato, chili, brinjal and okra	Radish, tomato, and chili showed higher accumulation of As, Cd, Cr, Pb, and Ni in their edible parts	Tiwari *et al.* (2011)
Amritsar, Punjab	Fe, Co, Cu, Cd, and Pb	Wastewater drain	Radish, turnip	HQ higher than the safe limits in spinach	Sharma *et al.* (2016)
Delhi	P, K, S, Zn, Cu, Fe, Mn, and Ni	Sewage effluents are the primary source of pollution	Rice, wheat, sorghum, maize, oats, cucumber, radish	Ni has the greatest potential, followed by Zn, Fe, Mn, and Cu in the plants	Rattan *et al.* (2005)
Kanpur	Fe, Cr, Zn, Mn, and Cu	Treated tannery wastewater is a source of pollution	Garlic, potato, turmeric, bitter gourd, black mustards, bottle gourds, eggplant, chili, kidney bean, jackfruit, okra, cucumber, maize, wheat	Garlic, potato, and turmeric accumulate the lowest level of toxic metal	Sinha *et al.* (2006)

West Bengal, where the irrigation with wastewater is predominant (Gupta *et al.* 2008a, 2008b). In the untreated wastewater, the concentration of Pb (0.21–4.3 mg L^{-1}), Ni (0.00–4.2 mg L^{-1}), and Cu (0.07–6.3 mg L^{-1}) exceed the recommended level of irrigation water. Among the collected vegetable samples, the mean concentration of heavy metals in leafy ones accumulated a higher amount of Cu (34.5 mg kg^{-1}, spinach), Zn (171 mg kg^{-1}, lettuce, 154 mg kg^{-1}, spinach), Cr (96 mg kg^{-1}, spinach), Ni (mg kg^{-1}, spinach) compared to the nonleafy one.

A similar study was reported by Sharma *et al.* (2016) for Amritsar city, where the irrigation is with effluents generated from municipal and industrial sources. In the studied area among the vegetables, the spinach accumulated a higher amount of Co (123 mg kg^{-1}) and Cu (80 mg kg^{-1}) compared to Cd (0.6 mg kg^{-1}) and Pb (0.2 mg kg^{-1}). The consumption of leafy vegetables from the study area can thus be a threat to human health. The impact of mixed irrigation (treated and untreated wastewater) was studied by Sharma *et al.* (2007) for Varanasi city. Cu, Zn, Pb, Cr, Mn, and Ni were determined in spinach samples collected during summer and winter. The mean concentration of Mn (7.5–117 mg kg^{-1}) was reported as maximum, followed by Cu (10.95–28.55 mg kg^{-1}), Pb (3.09–15.74 mg kg^{-1}), Cr (5.37–27.83 mg kg^{-1}), Zn (2.22–41.51 mg kg^{-1}), Ni (1.81–7.57 mg kg^{-1}), and Cd (0.50–4.26 mg kg^{-1}). The accumulation of heavy metals by spinach mainly depended on the metal concentration in the soil and plant tissue's absorption. Depending on the season, Cd, Zn, Cr, and Mn concentrations were higher during summer, and Cu, Pb, and Ni concentrations were higher in winter. This variation in the concentration of metals in season may be due to soil properties such as pH, conductivity, OM, and $NO_3 - N$ content. Slightly higher values of soil properties were reported in summer (pH: 8.59 ± 0.06; electrical conductivity (EC): 0.33 ± 0.03 mS cm^{-1}, OM: 92 ± 6 g kg^{-1}, NO_3–N: 26 ± 8 mg kg^{-1}) compared to corresponding values in winter (pH: 8.13 ± 0.19, EC: 0.26 ± 0.08 mS cm^{-1}, OM: 86 ± 5 g kg^{-1}, NO_3–N: 20 ± 7 mg kg^{-1}) due to frequent use of wastewater for irrigation during summer. Another reason may be that metals like Cd, Cr, Mn, and Zn act as good carriers for low molecular weight organic molecules compared to Cu, Pb, and Ni. The high transpiration rate in summer also contributes to the availability of metals by the plant. When compared with the Indian permissible limit, the Pb, Ni, and Cd were found above the limit, which may be associated with the health to the population of the study area.

To evaluate the effect of mixed industrial effluent on agriculture, Tiwari *et al.* (2011) collected spinach, radish, tomato, chili, and cabbage from Vadodara city, and the concentrations of As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were determined. The physiochemical properties reported that industrial effluent was slightly acidic (6.14) with high EC (345 mS cm^{-1}), Total Dissolved Solids (TDS) (1,792 mg L^{-1}), chloride (1,085 mg L^{-1}), sulfate (6,450 mg L^{-1}), and nutrients ($NO_3 - N$: 199 mg L^{-1}, $PO_4 - P$: 85.7 mg L^{-1}, Total Carbon (TC): 2,450 mg L^{-1}) as compared to tube well water. Due to the continuous use of effluents for irrigation purposes, concentrations (mg kg^{-1}) of heavy metals like As (9.6), Cd (19.2), Cr (12.8), Cu (18.4), Fe (278), Mn (94), Ni (15), Pb (13), and Zn (85) in the soil enhance. Among all the analyzed samples, heavy metal concentrations were higher in leafy samples like spinach and cabbage than in nonleafy vegetables like radish, tomato, and chili. This study reported that the accumulation of heavy metals from soil to plant did not follow any particular pattern and differed with respect to metal, species, and different plant parts. For example, the maximum accumulation of metal was found in spinach in order of Fe > Mn > Zn in the shoot part compared to the root part, while radish accumulated Cd > Cr > Cu > Fe > Mn > Ni in the shoot part as compared to the root. The industrial effluent from electroplating industries adversely affects the growth, development, and tissue concentrations in analyzed spinach and radish samples for Lucknow city (Pandey 2006). The mean concentrations (all in μg g^{-1}) observed in spinach were Cr (302), Cu (81), Ni (155), and Zn (147), while the respective values in radish were 198, 41, 84, and 140. Plants tend to accumulate heavy metals depending on how much a plant is contaminated or how a plant is genetically diverse. In this study, leafy vegetables have greater accumulations of heavy metals than nonleafy vegetables.

Atmospheric deposition, industrial and urban activities can also transport heavy metals from the atmosphere into vegetables. The degree of heavy metals deposited on a vegetable depends on many factors, such as the presence of air pollutants, the nature of the traffic loads, and the period of exposure of the vegetables in the urban environment at the time of harvest (Agrawal 2003). A study by Tripathi *et al.* (1997) on the outskirts of Bombay city reported that inhalation and ingestion are the pathways through which heavy metals can enter the human body. Among the analyzed samples (cereals, pulses, vegetables, fruits), higher levels of Pb (439 μg kg^{-1}) in green gram, Cu (8,004 μg kg^{-1}) in red gram, and Pb (190 μg kg^{-1}) and Zn (8,420 μg kg^{-1}) in amaranth were reported in the study. For the studied area, a high concentration of Pb (1.12 μg/m^3) was found in the atmosphere, which was attributed to a nearby battery industry. A comparative study was conducted on a market and production sites to evaluate the possible effect of atmospheric depositions (Sharma *et al.* 2009) in Varanasi city of India. The authors studied spinach, cauliflower, and lady's finger for Cu, Zn, Cd, and Pb. Compared with the market site, the production exhibited increased concentrations of Cu, Zn, Cd, and Pb for cauliflower (114, 23, 103, 53%), spinach (36, 50, 100, 44%), and lady's finger (12, 32, 57, 17%), respectively. This was attributed to the generation of heavy metals during transportation and their atmospheric deposition. The average concentration of all the metals accumulated for cauliflower (25.8 μg g^{-1}) was maximum, followed by spinach (22.13 μg g^{-1}), and lady's finger (16.60 μg g^{-1}), the higher exposed area for cauliflower favors the higher accumulation. A similar type of observation was also reported by Chauhan (2014) for Rewa city of Madhya Pradesh, where atmospheric toxicity was linked to emissions from a cement factory. The mean concentration of metals like Pb (8.9 mg kg^{-1}), Cd (2.39 mg kg^{-1}), and Cr (5.25 mg kg^{-1}) exceeded the permissible limits for collected vegetable samples. Accumulation of higher concentrations of Pb in analyzed samples is due to industrial exhaust, road dust, and exhausts from diesel generators. The proposed effect of industrial and transport activity on different vegetables (tomato, lady's finger, cucumber) was also reported by Srinivas *et al.* (2009) for Visakhapatnam city. For the study area, a higher air accumulation factor was observed in analyzed tomato samples (Pb – 1.665), lady's finger (Pb – 1.297, Zn – 4.693), and *Hibiscus cannabinus* Linn (Pb – 1.913, Zn – 1.537) collected from an industrial area. Among all the metals, the average Pb concentration was reported higher (>2.5 μg g^{-1}) in the analyzed vegetable sample grown in an industrial area due to the emissions from industry and vehicles.

3.3. TF of heavy metals

The parameter TF helps quantitatively understand the accumulation of heavy metals from soil to different plant parts. TF is defined as the ratio of the concentration of heavy metals in plants to the concentration of heavy metals in soil. According to Mirecki *et al.* (2015), if the value for any metal is $TF > 1$, it indicates the accumulation of that metal by plants. TF ratio ~1 indicates lesser uptakes, and ratio < 1 indicates plants exclude the elements from uptake. Ghosh *et al.* (2012) found that with an increasingly heavy metal concentration in soil, there is also an increase in the TF, indicating a direct relationship between metal concentration in the soil and plants. The genotypic difference of plants also affects the transfer of heavy metals to a different part of the plant. The vegetables where roots are used for consumption are more efficient for transferring heavy metals over leafy ones, followed by crops where the edible part is fruits.

Various researchers across Indian cities have reported TF values for a leafy vegetable like spinach. For instance, for spinach, reported values ranged from 0.5 to 0.91 (Tiwari *et al.* 2011 from Vadodara), 0.20 to 0.82 (Sharma *et al.* 2008 from Varanasi), 0.10 to 0.25 (Lokeshwari & Chandrappa 2006 from Bangalore). A very high set of values (9 – 32) was reported by Rattan *et al.* (2005) in and around the capital city, Delhi, 1.01 (Mohanty *et al.* 2021 from Ahmedabad). For vegetables like cabbage, the range was from 0.50 to 0.65 (Tiwari *et al.* 2011), 0.26 to 1.36 (Sharma *et al.* 2008), 0.1 to 0.20 (Mohanty *et al.* 2021), for vegetables like brinjal the TF was 0.0–0.38 (Mohanty *et al.* 2021), 0.0–0.06 (Chary *et al.* 2008), 0.60–0.79 (Tiwari *et al.* 2011), and 0.26–1.36 (Sharma *et al.* 2008).

Metal uptake by plants from soil depends mainly upon the soil minerals (e.g., carbonates, oxides, and hydroxides), soil OM (e.g., humic acid and fluvic acid), soil pH, redox potential, temperature and humidity of the soil, precipitation, and dissolution of metal ion along with plant species. A study by Tiwari *et al.* (2011) highlighted that plants like tomatoes and cabbage transfer toxic metals like As, Cd, and Ni from the root to edible parts of the plant. Metals like Cr were accumulated and transferred in leafy vegetables like spinach from root to top. The TF may decrease time when the plants are grown in highly contaminated soils due to the continuous uptake and translocation in plant tissue (Mirecki *et al.* 2015). Tiwari *et al.* (2011) also reported higher transfer factors for metals like Cd, Cu, Fe, Mn, Ni, and Zn for the plants irrigated with tube well water compared to wastewater for the Vadodara area of Gujarat. This is due to the lower metal concentration in tube well-irrigated soil compared to wastewater-irrigated soil.

The TF varied within the same plant depending upon the metal accumulation capacity by different parts of the plant and even variants of the same species (Zhou *et al.* 2016). Leafy vegetables could absorb more heavy metals than fruit vegetables (Zhou *et al.* 2016). For example, Yang *et al.* (2009) reported the Cd concentration in a different group of plant species, and depending upon their Cd accumulation capacity following sequence in plant groups is observed; legumes < melon < alliums < root < solanaceous < leafy vegetables.

3.4. Risk-assessment analysis

The HQ was used to evaluate the risk to human health for the intake of metal-contaminated vegetables. HQ is the ratio between the exposure dose and an oral reference dose (RFD); for HQ < 1, there will be no potential risk associated, and for HQ > 1 it indicates a high risk associated with dietary consumption of metal-contaminated vegetables (Chien *et al.* 2002; Khaled & Muhammad 2016). Mohanty *et al.* (2021) modified the equation to calculate the HQ,

$$HQ_M = \frac{C_M \times (1 - f_{mositure}) \times C_R}{BW \times RFD_M} \times 10^{-3}$$

where C_M is the average metal consumption (μg g^{-1}) on a dry weight basis; f_{moisture} is the fraction of moisture content in vegetables; C_R is the consumption rate of vegetables in g day^{-1}; BW is the average weight of the body in kg, RFD is the reference dose of metal in mg day^{-1}.

Researchers from several parts of India reported different ranges of HQ. For Ahmedabad city of India, Mohanty *et al.* (2021) reported high health risks associated with metals like Cr (0.97 ± 0.01), Mn (1.01 ± 0.01) and Pb (6.15 ± 0.01). Rattan *et al.* (2005) reported that the HQ for Ni was higher than Zn and Cu for the samples collected from different parts of western Delhi. However, all the reported values were < 1, so fewer health hazards were associated with the consumption of the vegetables from that reported area. For the collected vegetable samples from Hyderabad, Chary *et al.* (2008) concluded that higher health risks are associated with Zn (1.9–5.3) and Cr (2.2–3.05). Among the collected leafy vegetables, higher HQ was reported for spinach (5.3), followed by amaranthus (4.3) and mint leaves (3.5). In a study by Singh *et al.* (2010), for vegetables irrigated with wastewater in the Varanasi area, the resulting HQ for the Cd, Pb, Ni, and Zn was more than 1; however, it was less than 1 for Cr. High risk is associated with vegetables like spinach, cabbage, and lady's finger cultivated in that area. For Amritsar city, Sharma *et al.* (2016) found that spinach had higher HQ values for Co (7.5 for adults and 12.5 for children) and Cu (5 for adults and 8 for children). The continuous use of wastewater for irrigation may enhance the health hazard and ultimately threaten health-related issues. Thus, continuous monitoring of vegetables for heavy metal concentration grown in the wastewater-cultivated area is required, and such vegetables should be avoided to reduce the health risk associated with them.

3.5. Health effects of metal-contaminated vegetables

Heavy metals are harmful because they are nondegradable over a long period and cause harmful effects when they accumulate in many parts of the body of the organism. Heavy metals have been shown to have mutagenic, teratogenic, neurotoxic, and carcinogenic effects. Metal toxicity causes various human diseases, including cardiovascular disease, kidney disease, and nervous system disorders (WHO 1996). Over the last several decades, exposure to cadmium, lead, and mercury has been evaluated in different areas. Metals like lead, arsenic, cadmium, and copper are cumulative poisons. These metals are known to be highly toxic to the environment and cause adverse effects on human health. Foods consisting of vegetables may include these metals because of environmental exposure.

Prolonged exposure to unsafe levels of heavy metals through foodstuffs may result in chronic heavy metal accumulation in the kidney and liver of humans, disrupting numerous biochemical processes and leading to cardiovascular, nervous, kidney, and bone diseases (WHO 1992; Waalkes *et al.* 1999; Jarup 2003). Heavy metal ions may cause chronic effects in humans through short- and long-term side effects. Heavy metals such as copper, zinc, manganese, cobalt, and molybdenum are micronutrients for humans and animals to be present in small amounts. Other metals such as Cr, Cd, and As are carcinogenic. The concentrations of Hg and Pb are related to severe developmental disabilities in infants. Pitot & Dragan (1996), Hartwig (1998), and Saplakoglu *et al.* (1997) stated that long-term ingestion of Cd caused renal, prostate, and ovarian cancers. When consumed over a long period, fruit and vegetables enriched with heavy metals such as Cd, Pb, Cu, and Zn can potentially be toxic to human health. This can contribute to pancreas, bladder, and prostate cancer risk. Lead can be very toxic to humans when absorbed through the digestive system. Neural disorder is one of the major health issues associated with it. The presence of Pb in the mother's body is associated with low birth weight, preterm birth, stillbirths, spontaneous abortions, and hypertension.

In the Indian context, a few studies, e.g., Singh & Kumar (2006) and Gupta *et al.* (2008a, 2008b), have linked farmers' health issues with contaminated soil and agricultural cultivated products. Heavy metals were a major concern in the capital city of Delhi, with multiple studies revealing high amounts of regularly consumed vegetables (Marshall *et al.* 2003; Singh & Kumar 2006). Similar issues were highlighted in Varanasi and the state of West Bengal, wherein the authors reported significant levels of helminth eggs on vegetables grown with sewage (Gupta *et al.* 2008a, 2008b). The findings of an International Water Management Institute (IWMI) study suggested that farmers in Delhi, Kolkata, and Hyderabad complained about skin irritations and respiratory difficulties. In contrast, farmers in industrial areas near Ahmedabad and Kanpur had noticeable skin diseases due to exposure to sewage combined with industrial effluent (Amerasinghe *et al.* 2013). Increased worm infections were also identified in sewage-fed vegetables in Hyderabad (Srinivasan & Ratna 2009). Farmers in Kanpur had higher levels of heavy metals and pesticides in their blood and urine (Singh *et al.* 2004).

4. CONCLUSIONS

We reviewed the published literature and compiled the findings of the studies on the impact of wastewater irrigation from (semi-)urban areas of India. Since the elimination of wastewater irrigation in the Indian context looks unlikely because of the ever-increasing demand for irrigation water, it is important to assess the impact of wastewater irrigation on the transfer of heavy metals to soils and then to the vegetation grown in those soils. Finally, the objective is to evaluate the risk to consumers (from HQ values) due to heavy metal transfer from vegetation to irrigated soils to humans via the food chain. It was found that high HQ values were reported for Mn and Pb (Ahmedabad), Zn and Cr (Hyderabad), and Co and Cu (Amritsar). Vegetables grown in industrialized and urban areas (e.g., in Varanasi) are exposed to high concentrations of heavy metals as a result of aerial deposition and irrigation with wastewater. The higher absorption capacity of leafy vegetables is known to accumulate higher amounts of toxic heavy metals than nonleafy ones.

Long-term consumption of contaminated foods may negatively impact human health due to increased intake and accumulation. Risk assessment based on heavy metal concentrations in the vegetables needs to be validated by clinical studies, which, at best, is very sparse in the Indian context. Metal absorption by different cell organs and membranes is another area where more studies must be carried out. Most heavy metals mentioned are harmful, but only after decades of exposure; they do show a detrimental effect. Therefore, frequent testing of heavy metals in plant tissues is recommended to avoid excessive growth in the human food chain. Finally, compliance with regulations for heavy metals from the industrial sectors should be strictly enforced to reduce the risk of heavy metal exposure to the public.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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