

## CRITICAL REVIEW

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## Heavy metal contamination in wastewater-irrigated vegetables: assessing food safety challenges in developing Asian countries

Navneet Kaur, <sup>a</sup> Jagdev Singh, <sup>a</sup> Neeta Raj Sharma, <sup>a</sup> Simranpreet Kaur Natt, <sup>b</sup> Anand Mohan, <sup>a</sup> Tabarak Malik <sup>\*c</sup> and Madhuri Girdhar <sup>\*d</sup>

Vegetables are crucial for human nutrition, providing essential micronutrients and beneficial compounds. Heavy metal contamination of vegetables irrigated with wastewater poses a significant public health risk in developing Asian countries. This review analyses recent research on heavy metal accumulation in vegetables across India, Bangladesh, Pakistan, and China. Studies consistently report concerning levels of cadmium, lead, chromium, arsenic, nickel, and mercury in vegetables, often exceeding international safety standards. Leafy vegetables consistently show higher heavy metal accumulation compared to fruit and root vegetables. Within plant structures, roots generally contain higher heavy metal concentrations than edible parts, though this varies depending on the metal and plant species. Many studies report health risk indices exceeding safe limits, indicating potential non-carcinogenic and carcinogenic risks from chronic dietary exposure, with children at higher risk. The review highlights inadequate regulatory frameworks and enforcement mechanisms. A multi-faceted approach is urgently needed, encompassing improved wastewater treatment, best agricultural practices, rigorous monitoring, and public awareness campaigns. Future research directions are identified, including long-term health impact studies, development of cost-effective remediation techniques, and exploration of sustainable alternatives to wastewater irrigation. While wastewater irrigation addresses immediate water scarcity, it poses significant long-term food safety and public health risks. Integrated policies balancing water scarcity, agricultural productivity, and health risks are essential. This review underscores the pressing need for coordinated efforts from policymakers, researchers, and health officials to safeguard public health and ensure sustainable agriculture in developing Asian countries facing increasing urbanization and water scarcity.

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### Environmental significance

Heavy metal contamination of vegetables irrigated with wastewater is a critical environmental and public health issue in developing Asian countries. Understanding this problem is crucial for ensuring food safety and sustainable agriculture in water-scarce regions. This review reveals widespread contamination of vegetables with toxic metals like cadmium, lead, and arsenic, often exceeding international safety standards. Leafy vegetables consistently show higher accumulation compared to other types. Health risk assessments indicate potential carcinogenic and non-carcinogenic risks from chronic dietary exposure. These findings highlight the urgent need for improved wastewater treatment, agricultural practices, and regulatory enforcement to mitigate risks. Insights gained can inform integrated policies and interventions to balance water scarcity, agricultural productivity, and public health across developing regions facing similar challenges globally.

## 1. Introduction

Vegetables are essential to human nutrition, providing vital micronutrients and other beneficial compounds.<sup>1</sup> These

include minerals such as iron, sodium, copper, magnesium, zinc, calcium, iodine, and potassium, as well as vitamins, antioxidants, and important metabolites.<sup>2,3</sup> People consume vegetables both in their raw state and after cooking.<sup>4</sup> However, if vegetables contain toxic metals, they may present serious health hazards to consumers.<sup>5</sup> Research has extensively documented heavy metal (HM) contamination in food chains worldwide.<sup>6-9</sup> Plants can absorb HMs from the soil, often accumulating them at higher concentrations than found in their environment.<sup>10-12</sup> The primary factor contributing to this metal buildup in plants is the presence of HM pollutants in the soil.<sup>13,14</sup>

<sup>a</sup>School of Bioengineering and Biosciences, Lovely Professional University, Phagwara, Punjab, India

<sup>b</sup>Punjab Agricultural University, Ludhiana, Punjab, India

<sup>c</sup>Department of Biomedical Sciences, Institute of Health, Jimma University, Jimma, Ethiopia. E-mail: tabarak.malik@ju.edu.et

<sup>d</sup>Division of Research and Development, Lovely Professional University, Phagwara, Punjab, India. E-mail: madhuri.girdhar89@gmail.com

Globally, wastewater production is estimated to be 359.4 billion m<sup>3</sup> per year, with South Asia contributing approximately 25.6 billion m<sup>3</sup> – just 7% of the global total despite hosting around 24% of the world's population.<sup>15</sup> In India, the estimated wastewater generation in 2020–21 was approximately 14.46 billion m<sup>3</sup> per year in rural areas and 26.41 billion m<sup>3</sup> per year in urban centers, totaling about 40.8 billion m<sup>3</sup> per year.<sup>16,17</sup> A 2002 report stated that Pakistan produces an estimated 4.369 billion m<sup>3</sup> of wastewater annually, comprising 3.060 billion m<sup>3</sup> from municipal and 1.309 billion m<sup>3</sup> from industrial sources.<sup>18</sup> In Bangladesh, annual domestic and industrial wastewater generation is approximately 4.87 billion m<sup>3</sup> and 0.45 billion m<sup>3</sup>, respectively.<sup>19</sup> China discharged 55.7 billion m<sup>3</sup> of wastewater in 2021, of which 28.7% originated from industrial sources.<sup>20</sup> While data vary across years and sources, they collectively highlight the substantial and growing volume of wastewater in developing Asian countries. Thus, urbanization and industrialization in many developing Asian nations have led to the proliferation of wastewater irrigation for agricultural purposes, particularly in peri-urban areas.<sup>21–26</sup> This has led to the accumulation of HMs in farmlands, which can then be absorbed by vegetable crops, posing a serious health hazard to consumers.<sup>25,27–29</sup>

The toxicity of food due to HM contamination poses a serious threat to human well-being and is a significant food safety problem globally.<sup>30</sup> HMs can be classified depending on their toxicity levels into low toxicity (Mo, Mn, and Fe),<sup>31,32</sup> average toxicity (Zn, Cr, Cu, W, Co, V, and Ni)<sup>33</sup> and high toxicity (As, U, Pb, Sb, Hg, Cd, and Ag).<sup>34</sup> While some trace elements like iron, manganese, zinc, and copper are vital nutrients for plants and animals with lower health risks at elevated levels, others such as cadmium, nickel, arsenic and lead can be detrimental even in small amounts.<sup>5,35,36</sup> Ingesting these metals beyond safe thresholds can potentially harm human and animal well-being.<sup>37,38</sup> Plants, particularly vegetables, absorb these elements through their root systems, leading to accumulation in different tissues.<sup>39</sup> Excessive intake of such metals by humans and animals can result in various health issues, including disrupted endocrine function, cancer, skeletal damage, and impairment of the circulatory, nervous, excretory and cardiovascular systems.<sup>40,41</sup> The persistent nature of HMs, their extended biological presence, and high potential for uptake and bio-accumulation make them particularly dangerous to all living organisms, including humans.<sup>42</sup> Prolonged exposure to toxic metals through dietary sources, even in minute quantities, is believed to cause gradual buildup in the hepatic and renal systems of humans, livestock, and bird species.<sup>43,44</sup>

The quality of irrigation water is a crucial factor, as sewage<sup>45</sup> or industrial wastewater<sup>46,47</sup> often contains much higher concentrations of HMs, such as mercury, arsenic, chromium, nickel, zinc, lead, copper and cadmium, compared to clean water.<sup>48</sup> Approximately 11% of global cultivated land is irrigated using untreated wastewater.<sup>49</sup> Furthermore, an estimated 10% of the world's population consumes produce grown with wastewater irrigation.<sup>50</sup> Due to its high content of organic matter and nutrients, wastewater is often utilized in agriculture as a method of wastewater management.<sup>51</sup> While HM

concentration in wastewater is typically low and often within acceptable limits, prolonged irrigation with wastewater has led to an increase in HM levels in soil over time.<sup>52,53</sup> Consequently, farmlands irrigated with this contaminated water over extended periods have become heavily polluted with HMs, which are then taken up by the vegetables grown in these soils. HM uptake by plants from soil is affected by multiple factors.<sup>45</sup> These include the nature of the metal sources, seasonal variations, loading rates, soil pH levels, oxidation-reduction potential, soil composition, organic content, various forms of bioavailable metal fractions, and overall metal concentrations in the soil.<sup>2,54</sup>

The global context of this issue cannot be overstated (Fig. 1). However, the situation in developing Asian countries is particularly acute due to a confluence of factors. Rapid industrialization, coupled with inadequate waste management infrastructure and often lax environmental regulations, has exacerbated the problem in these nations.<sup>55</sup> The United Nations' Sustainable Development Goals define food security as a state where every individual has consistent access to adequate and safe nutrition for a healthy, active lifestyle.<sup>56</sup> However, developing countries like India often prioritize increasing food quantity and access in their efforts to achieve food security, often overlooking practices that ensure food quality and safety. These include cultivating crops on polluted land and irrigating with untreated wastewater that could alter food composition. Such practices risk introducing non-essential elements and harmful substances into the food supply.<sup>57</sup> The production of fruits and vegetables has been on the constant rise, as shown in Fig. 2, but the quality of such vegetables due to the use of wastewater has declined. Addressing HM contamination in vegetables is not just a matter of environmental concern but is crucial for achieving sustainability goals and ensuring food security and public health in developing regions.

The World Health Organization (WHO) has established safety guidelines for the maximum permissible concentration of HMs in various edible items, including vegetables.<sup>59,60</sup> These guidelines serve as a global benchmark for food safety. However, numerous studies conducted in wastewater-irrigated areas have reported that vegetables grown in these conditions

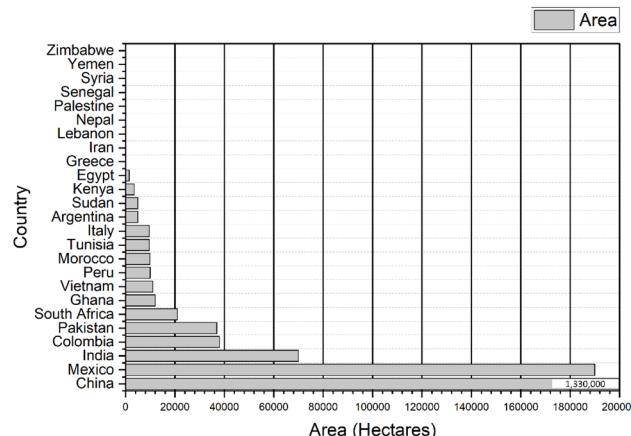


Fig. 1 Area under wastewater irrigation in various countries.<sup>76</sup>

often exceed these safety limits.<sup>34,61</sup> This widespread non-compliance with international safety standards raises serious concerns about the long-term health impacts on consumers who rely on these vegetables as a staple part of their diet.

There are various sources of HM contamination of agricultural fields, as shown in Fig. 3, of which HMs usually enter through irrigation with contaminated water. The use of wastewater for irrigation in developing Asian countries is driven by a complex set of factors that highlight the challenging trade-offs between immediate agricultural needs and long-term health and environmental concerns.<sup>29,69,70</sup> Water scarcity is a primary driver, with many regions facing severe shortages that make wastewater an attractive alternative for irrigation. Additionally, wastewater often contains high levels of nutrients, which can enhance crop yields and reduce the need for chemical fertilizers.<sup>71</sup> Farmers favour wastewater irrigation due to its nutrient content and constant availability.<sup>72</sup> This nutritional benefit makes wastewater irrigation an economically attractive option for many small-scale farmers who view it as a free or low-cost irrigation source. In some peri-urban areas, wastewater may be the only readily available water source for irrigation, leaving farmers with little choice but to use it despite the potential risks.<sup>73</sup> While wastewater irrigation offers these apparent benefits, it also presents significant risks, particularly in terms of HM contamination. Balancing these competing interests poses a significant challenge for policymakers and agricultural practitioners in developing Asian countries.

The bioaccumulation of these metals in vegetables is not a straightforward process but depends on a variety of factors. Soil properties play a crucial role, with factors such as clay content, cation exchange capacity, organic matter content, and pH all influencing the bioavailability of HMs.<sup>74</sup> Different vegetable species have varying capacities for uptake of HMs and their translocation. In general, compared to fruit vegetables, leafy vegetables like spinach and lettuce tend to acquire higher quantities of HMs.<sup>21</sup> Vegetables accumulate varying levels of HMs based on agronomic conditions and farming practices. These diverse products converge in common markets across

villages, towns, and cities, potentially exposing unsuspecting consumers to significant health risks.<sup>42</sup>

The duration of irrigation with wastewater is another critical factor. The long-term use of wastewater for irrigation leads to increased accumulation of HMs in soils and, consequently, in vegetables.<sup>57</sup> The source of wastewater also plays a significant role, with industrial effluents typically containing higher concentrations of HMs compared to domestic wastewater.<sup>57</sup> Climate and environmental factors, including temperature, rainfall, and atmospheric deposition, can influence HM mobility and uptake.<sup>71</sup> Additionally, agronomic practices such as the use of fertilizers and pesticides can affect HM dynamics in soil-plant systems.<sup>75</sup>

The current status of HM contamination in vegetables due to wastewater irrigation varies across developing Asian countries but they share common challenges, including rapid urbanization, inadequate wastewater treatment infrastructure, and the need to balance food security with environmental and health concerns.

By identifying knowledge gaps and proposing future research directions, this review seeks to enhance our understanding and management of HM contamination in vegetables grown using wastewater irrigation in developing Asian countries. By evaluating the extent of contamination across different vegetables and geographic regions, the review seeks to identify patterns and trends that can inform targeted interventions. Assessing the associated health risks based on reported contamination levels and established safety standards is crucial for prioritizing public health measures. This review holds significant implications for various stakeholders. Policymakers can use the findings to inform the development and refinement of food safety regulations, environmental protection policies, and urban planning strategies to minimize the risks associated with wastewater irrigation. By addressing the critical issue of HM contamination in vegetables grown using wastewater irrigation, this review contributes to the broader goals of ensuring food security, promoting public health, and advancing sustainable agricultural practices in developing Asian countries.

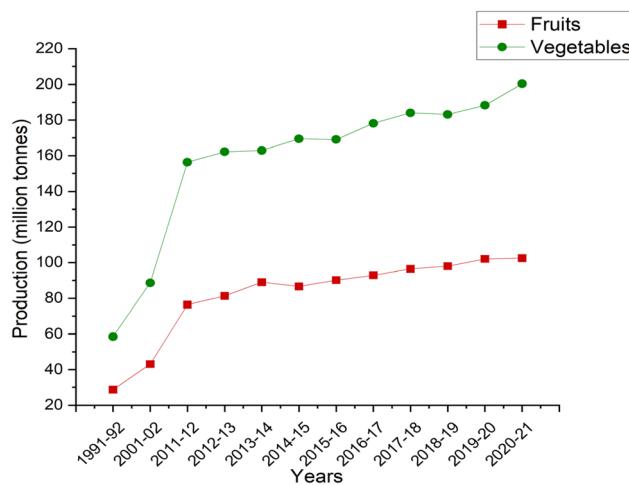


Fig. 2 Year wise increase in production of fruits and vegetables.<sup>58</sup>

## 2. Heavy metal determination in various vegetable samples

The determination of HMs in vegetable samples from developing Asian countries has highlighted both the prevalence of these contaminants and the methodologies employed for their detection. Studies conducted in countries such as India, Bangladesh, Pakistan, and China have focused on urban and peri-urban areas, agricultural regions, and industrial zones, reflecting a comprehensive approach to understanding the extent of HM contamination. In these studies, various digestion methods were employed to prepare vegetable samples for analysis. The most commonly used method involved nitric acid ( $\text{HNO}_3$ ), either alone or in combination with other acids like hydrofluoric acid (HF), sulfuric acid ( $\text{H}_2\text{SO}_4$ ), perchloric acid ( $\text{HClO}_4$ ), and hydrochloric acid (HCl). The analysis of digested samples



### Sources of heavy metal pollution in agricultural fields

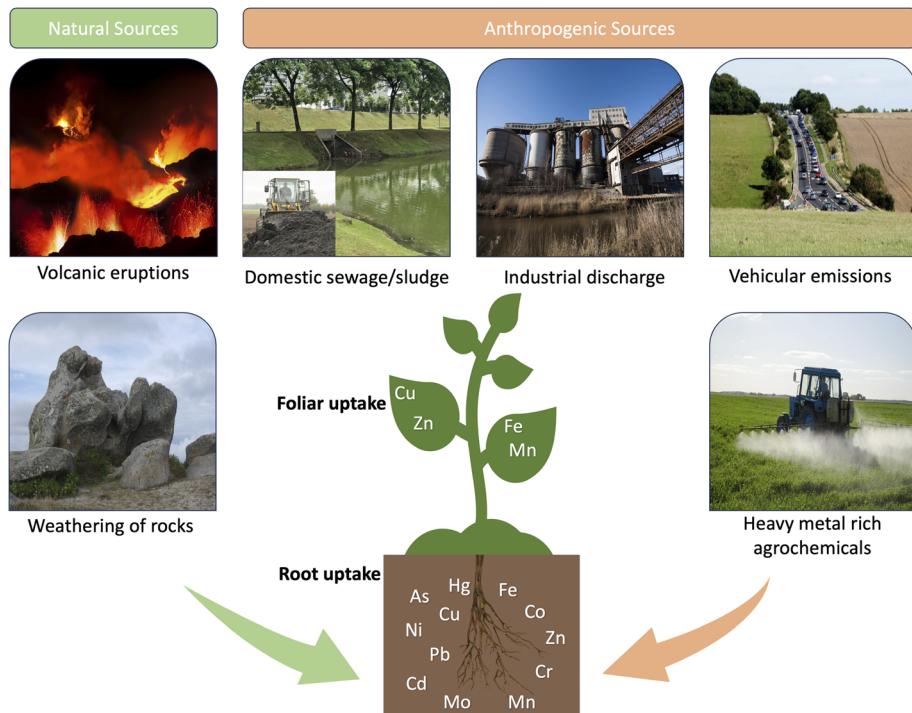


Fig. 3 Different sources of HM pollution in agricultural fields. All the images used in the figure are reused under the terms of the Creative Commons license CC BY 2.0 <https://creativecommons.org/licenses/by/2.0/>.<sup>62–68</sup>

predominantly used Atomic Absorption Spectroscopy (AAS), Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES), and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) whereas Atomic Fluorescence Spectroscopy (AFS), Atomic Fluorescence Morphological Analyzer (AFMA) and Automatic Mercury Analyzer (AMA) were rarely used. These were used to identify various HMs such as aluminum (Al), arsenic (As), boron (B), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), selenium (Se), and zinc (Zn) (Table 1).

A wide variety of vegetables were analyzed across these studies, with spinach, tomato, cabbage, radish, and brinjal being the most frequently tested. These vegetables, commonly consumed in the regions studied, serve as key indicators of HM contamination due to their varying abilities to absorb and accumulate metals from the soil. Results obtained from these studies are listed in Table 2 and discussed in the following sections.

### 3. Contamination of vegetables irrigated with wastewater in India

Several recent studies have highlighted the concerning levels of HM pollution in vegetables that are irrigated with wastewater in different parts of India. A study conducted in Jhansi, North India, found that vegetables irrigated with wastewater/grown near industrial areas, particularly leafy varieties like spinach

and fenugreek, accumulated hazardous levels of cadmium, manganese, and lead. The target hazard quotients for these metals exceeded unity, indicating significant non-carcinogenic health risks for regular consumers.<sup>82</sup> Research in peri-urban and urban areas of Delhi analyzed six vegetables for 5 HMs (Pb, Hg, Cr, Cd and As). The study found that Cd concentrations exceeded permissible levels in most vegetables, with spinach and okra showing the highest metal pollution index and transfer factor values.<sup>70</sup> Similarly, another research study in peri-urban and urban areas of Delhi and Dehradun revealed the ubiquitous presence of toxic elements like cadmium and lead in vegetables, often exceeding permissible limits set by the World Health Organization (WHO) and the Food and Agriculture Organization (FAO). Vegetables, especially spinach and okra, were identified as major contributors to dietary exposure of HMs. Cadmium and lead levels in vegetables, particularly in spinach samples, were found to be dangerously high. The health risk assessments concluded that children are at a higher risk compared to adults, with vegetables being the primary contributors to this risk.<sup>89</sup> Another research study conducted in peri-urban regions of Lucknow city found that while HM levels in irrigation water and agricultural soil were within permissible limits, they exceeded these limits in vegetables. Iron levels were particularly high, being 5 to 60 times above permissible limits in almost all samples. The hazard index values for vegetables from all studied peri-urban areas were found to be more than 1 (threshold level), indicating probable adverse non-carcinogenic health hazards.<sup>90</sup>



Table 1 Sampling sites and processing methods for HM analysis of vegetables in developing Asian countries

Country	Sampling site	Vegetables analyzed	Digestion	Analysis method (metals)	Metals detected	References
India	Bangalore, Karnataka	Spinach, coriander	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (20 : 1)	AAS	Pb, Cu, Zn, Cd, Cr, Mn	77
	Nashik, Maharashtra	Coriander, spinach, onion, cauliflower, brinjal, cabbage, tomato, cucumber, potato, carrot	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Pb, Cd, As, Cu	78
	Dehradun, Uttarakhand	Beet, French beans, spinach, cauliflower	$\text{HNO}_3 + \text{HClO}_4$ (2 : 1)	AAS	Pb, Ni, Zn, Cd, Cu, Cr	79
	Patna, Bihar	Spinach, red spinach, beans, okra, cabbage, cauliflower, beetroot	$\text{HNO}_3$	AAS	Hg, Zn, Pb, Cu, Mn	80
	Haridwar, Uttarakhand	Spinach	$\text{HNO}_3 + \text{HClO}_4$ (4 : 1)	AAS	Zn, Mn, Fe, Cd, Cr, Cu	81
	Jhansi, Uttar Pradesh	Spinach, fenugreek, brinjal, green chili	$\text{HNO}_3 + \text{HClO}_4$ (3 : 1)	AAS	Zn, Pb, Mn, Ni, Cu, Co, Cd	82
	Lucknow, Uttar Pradesh	Tomato, spinach, cucumber, cabbage, green pepper, mint, bitter gourd, beet root, cauliflower	$\text{HNO}_3 + \text{HClO}_4$ (2 : 1)	ICP-OES	Cr, Cd, Pb, Ni, Cu, Co, Zn, Mn, Fe	42
	New Delhi (urban and peri-urban areas)	Spinach, mustard, okra, tomato, carrot, potato	$\text{HNO}_3 + \text{HClO}_4$ (5 : 1) and $\text{HNO}_3 + \text{H}_2\text{SO}_4$ (4 : 1) for Hg	ICP-OES	As, Cd, Cr, Pb, Hg	70
	Lucknow (peri-urban regions), Uttar Pradesh	Tomato, spinach	$\text{HNO}_3 + \text{HClO}_4$ (2 : 1)	ICP-OES	Ni, Pb, As, Cr, Cd	83
	Faridabad (Agra Canal), Haryana	Spinach, radish, coriander, garlic	$\text{HClO}_4 + \text{HNO}_3 + \text{HClO}_4$ (5 : 1 : 1)	ICP-OES	As, Pb, Cr, Mn, Cu, Zn, Ni	84
	Solapur, Maharashtra	Cabbage, carrot, fenugreek, garlic, ginger, potato, radish, onions, sorghum, sugarcane, apple, brinjal, broad beans, chickpea, cucumber, cream beans, green peas, mung beans, okra, orange, papaya, red gram, tamarind, tomatoes	$\text{HNO}_3 + \text{HCl} + \text{H}_2\text{O}_2$	ICP-MS, automatic mercury analyzer	Pb, Cd, As, Hg	85
	Bhadohi, Uttar Pradesh	Spinach, radish, garlic, cabbage, brinjal	$\text{HNO}_3 + \text{HClO}_4 + \text{HCl}$	ICP-OES		
	Ladakh	Spinach, cabbage	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (2 : 1)	ICP-MS	Zn, Cu, Cd, Ni, Cr	86
	Prakasam, Andhra Pradesh	Potato, amaranthus	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (2 : 1)	ICP-MS	Zn, Se, Pb, Fe, Cu, Co, Cd, B, Al, As	87
	Delhi & Dehradun (urban and peri-urban areas)	Tomato, potato, onion, okra, cabbage, cauliflower, spinach	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (2 : 1)	ICP-MS, automatic mercury analyzer	Cr, As, Cd, Hg, Pb	88
	Lucknow (peri-urban regions), Uttar Pradesh	Tomato, spinach	$\text{HNO}_3 + \text{HClO}_4$ (2 : 1)	ICP-OES	Co, Mo, Mn, Cu, Zn, Se, As, Ni, Hg, Cr, Cd, Pb	89
	Bokaro, Jharkhand	Cabbage, Indian spinach, tomato, radish, carrot, onion	$\text{HNO}_3 + \text{HClO}_4$ (5 : 1)	AAS	Cu, Fe, Zn, Mn, Co	90
	Yamuna Floodplain, New Delhi	Radish, brinjal, cauliflower, spinach	$\text{HNO}_3 + \text{HF}$ (5 : 1)	AAS	Co, Pb, Hg, As, Cr	91
					Zn, Mn, Co, Ni, Pb, Cr	92



Table 1 (Cont'd.)

Country	Sampling site	Vegetables analyzed	Digestion	Analysis method (metals)	Metals detected	References
Bangladesh	Kolkata, West Bengal	Red spinach, mustard, fenugreek, pok Choi, coriander, bathua, spinach, capsicum, M. spinach, lettuce, green amaranthus	$\text{HNO}_3 + \text{H}_2\text{O}_2 + \text{HCl}$ (15 : 5 : 1)	ICP-MS	Pb, Cd, Ni	93
	Jessor district	Bottle gourd, taro, drumstick leaf, stem amaranth, brinjal, ash gourd, snake gourd, elephant foot, potato, okra, eddoe, plantain, green papaya, ghatkol, Indian spinach	$\text{HNO}_3$	ICP-AES, ICP-MS	As, Cd, Cu, Pb, Zn	94
	Dhaka Export Processing Zone	Green cabbage, brinjal, chilli, okra, tomato	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Zn, Pb, Cr, Cd, Fe, Ni, Cu	95
	Chittagong	Green arum leaves, jute leaves, water spinach, bottle gourd, wax gourd, sweet gourd	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Chromium, copper, lead	96
	Pakshi, Pabna	Potato, red amaranthus, spinach, amaranthus, carrot, cabbage, tomato, brinjal	$\text{HNO}_3$	AAS	Ni, Cr, Cd, Co, Cu, Pb, Fe, Zn, Mn, Hg, As	97
	Savar Upazila, Dhaka	Stem amaranth, green papaya, Indian spinach, red amaranth, radish, bottle gourd, pointed gourd, coriander, jute Lettuce, cucumber, tomato, coriander	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (9 : 1)	AAS	Pb, Ni, As	98
	Dhaka Metropolitan City Area	Indian spinach, sponge gourd, bitter gourd, bottle gourd, okra, papaya, beans, brinjal, chili, cucumber	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (3 : 1)	ICP-MS	Pb, Cd, Cr	99
	Tangail	Red amaranth, spinach, jute, bottle gourd, mustard green, water spinach Brinjal, lady's finger, water spinach, red spinach, green spinach	$\text{HNO}_3 + \text{H}_2\text{O}_2$ (3 : 1)	AAS	Pb, Cd, As, Cu, Ni, Cr	100
	Dhaka (Buriganga River)	Bottle gourd, esculent, papaya	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Cd, Cr, Cu, Ni, Pb, Zn	101
	Savar Upazila, Dhaka	Cauliflower, bitter gourd, radish, pumpkin, apple gourd	$\text{HNO}_3 + \text{HClO}_4$ (5 : 1)	AAS	As, Mn, Ni, Cd, Ph, Fe, Cu	102
	Dhaka-Mymensingh Highway	Spinach, mustard, coriander, mint, fenugreek, lettuce, chenopodium, carrot, radish, beetroot, turnip	$\text{HNO}_3 + \text{HClO}_4 + \text{HCl}$	AAS	Fe, Mn, Cu, Zn, Cd, Pb, Cr, Ni	103
	Gujranwala	Okra, onion, green pepper, pumpkin	Wet digestion	AAS	Cd, Ni, Cr, Pb, Mn, Fe, Cu, Zn	104
	Sargodha district	Cauliflower, tinda, spinach, cabbages, carrot, taro, radish, turnip, lettuce Spinach, mustard, radish, green beans, carrot, potato	$\text{HNO}_3 + \text{HCl}$ (3 : 1)	ICP-OES	Fe	105
	Mingora, Swat	Cauliflower, round gourd, fenugreek, lucerne, carrot, spinach, radish, turnip, mustard, cabbage, pea, coriander, onion, brinjal	$\text{HNO}_3 + \text{HCl}$ (1 : 3)	AAS	Fe, Pb, Cr, Ni, Mn, Co	106
	Khyber Pakhtunkhwa region	Cauliflower, tinda, spinach, cabbages, Spinach, mustard, radish, green beans, carrot, potato	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Pb, Cu, Zn, Ni, Cd, Cr, Fe, Mn	107
	Lahore	Cauliflower, round gourd, fenugreek, lucerne, carrot, spinach, radish, turnip, mustard, cabbage, pea, coriander, onion, brinjal	$\text{HNO}_3 + \text{HCl}$ (3 : 1)	ICP-OES	As, Cd, Pb	108
	Vehari	Cauliflower, tinda, spinach, cabbages, Spinach, mustard, radish, green beans, carrot, potato	$\text{HNO}_3 + \text{HClO}_4$	AAS	As, Cr, Cd, Cu, Fe, Ni, Mn, Pb, Zn	109

Table 1 (Cont'd.)

Country	Sampling site	Vegetables analyzed	Digestion	Analysis method (metals)	Metals detected	References
Punjab		Spinach, mustard leaves, lettuce, tomato, radish, cabbage, okra, onion, chilli, brinjal, bottle gourd (round), garlic, fenugreek, cauliflower, zucchini, mint, coriander, potato, sugar beet, turnip	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (10 : 1 : 4)	AAS	Ni, Cd, Pb, Cr	110
Bhakkar		Spinach, cabbage, cauliflower, turnip, radish, taro, wax gourd, carrot, lettuce	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Fe, Pb, Mn, Ni, Cd, Co	111
Kirri Shamoza		Lettuce, carrot, tinda, radish, turnip, cabbage, spinach	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Zn, Mn, Cu, Fe, Pb, Cd, Ni, Cr	112
Dera Ghazi Khan, Punjab		Brinjal, red corn, white corn, tomato, luffa, apple gourd, cabbage, spinach	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	Cu, Fe, Zn, Ni, As, Cd, Cr, Mn, Pb	113
Layyah, Punjab		Radish, turnip, spinach, fenugreek, carrot	Wet digestion	AAS	Cd, Zn, Fe	114
Sargodha district		Cauliflower	$\text{HNO}_3 + \text{H}_2\text{O}_2$	AAS	Pb, Cu, Co, Mn, Ni, Cd, Cr, Zn, Fe	115
Faisalabad and Multan		Spinach, lettuce, cabbage, cauliflower, okra	$\text{HNO}_3 + \text{HClO}_4$ (3 : 1)	AAS	Cd, Pb	116
Sargodha		Carrot, cauliflower, pea, potato, radish, spinach	$\text{HNO}_3 + \text{aqua regia}$ (70% conc. $\text{HNO}_3$ and 65% $\text{HClO}_4$ ; 2 : 1) (1 : 3)	AAS	Pb	117
Bahawalpur		Cauliflower, cabbage, brinjal, green chilli, turnip, spinach, potato, radish, carrot, radish pods	$\text{HNO}_3 + \text{H}_2\text{SO}_4 + \text{HClO}_4$ (5 : 1 : 1)	AAS	As, Cd, Cr, Pb, Ni	118
China	Pearl River Delta, South China Chenzhou city, Suxian district	Chinese leaf mustard, loose-leaf lettuce, pak choi, Chinese cabbage, lettuce White radish, carrot, sweet potato, white caitai, red caitai, brinjal, red pepper, tomato, bitter gourd, towel gourd, cucumber, pumpkin, mater convolvulus, edible amaranth, cabbage, pai-tsai, Chinese cabbage, spinach, caraway, lettuce, asparagus beans, kidney bean Cabbage, coriander, chlorella, lettuce, Chinese lettuce, asparagus leaves, celery, spinach, radish, tomato	$\text{HNO}_3 + \text{HClO}_4 + \text{H}_2\text{O}_2$ (87 : 13 : 10) Dry ashing method	AFMA, AAS ICP-OES, AFS	Hg, As, Pb, Cr, Cd As, Zn, Cu, Cd, Pb	119 36
Xi River, Shenyang city, Northern China		Carrot, potato, Chinese cabbage, spinach, shallot, Chinese chives, cabbage, celery, Chinese lettuce, garland chrysanthemum, coriander, leafy lettuce, rape, cucumber, kidney beans, pepper, winged beans, brinjal, zucchini, tomato	Boiled with $\text{HNO}_3$	ICP	Cd, Cu, Pb, Zn	120
Baiyin city, Gansu province			$\text{HNO}_3 + \text{H}_2\text{O}_2$ (8 : 1)	ICP-MS	Zn, Pb, Cu, Cr, Cd, As	121

Table 2 Range of various HMs (mg kg<sup>-1</sup>) at different locations in developing Asian countries. BDL: below detection limits

Country	Sampling site	As	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn	References
India	Nashik (Maharashtra)	<b>0.50–2.30</b>	BDL– <b>3.30</b>		2.20–						BDL– <b>9.70</b>		78
	Dehradun, Uttarakhand	<b>17.51–</b> <b>23.19</b>	<b>15.30–</b> <b>57.18</b>	30.80– 33.49–	23.65– 33.49				65.69– <b>80.72</b>	49.54– 86.69	48.12– <b>161.86</b>		79
	Patna, Bihar			2.17–36.3		0.145– 0.705	13.09– 143.36		9.42– 0.06–4.09	9.42– 30.08	24.66– <b>183.72</b>		80
	Jhansi (North India)	0.00–2.23	0.08–1.63		3.45– 15.33		3.56– 59.47		0.06–4.09	0.00– <b>8.03</b>	5.89– 57.14		82
	New Delhi (urban and peri-urban areas)	0.056– <b>0.964</b>	0.009– <b>0.332</b>	0.008– 0.731			BDL– <b>0.911</b>			0.040– 2.215			70
	Lucknow (peri-urban regions)	<b>13.73–</b> <b>2392.05</b>	<b>5.90–</b> <b>503.86</b>	<b>7.14–</b> <b>215.02</b>					<b>19.96–</b> <b>82.62</b>	<b>1.66–</b> <b>18.99</b>			83
	Prakasam, Andhra Pradesh	0.269 ± <b>0.265</b>	<b>0.371 ±</b> <b>0.383</b>	18.111 ± 24.542		0.019 ± 0.026			1.409 ± <b>1.761</b>				88
	Ladakh	0.02–0.25	0.03–0.06	0.00– 0.009		0.07–0.66	1.07– 14.99			0.05– <b>0.56</b>	0.02–2.33		87
	Faridabad (Agra Canal)	<b>0.11–0.68</b>		3.41– 28.67	4.92– 21.62			107.67– 368.12	7.25– 58.00	4.00– 12.33	24.12– 126.13		84
	Solapur, Maharashtra	0.001– 0.013	0.001– <b>0.37</b>		65.58– 868.53	<b>1460.28–</b> <b>27363.74</b>		298.17– <b>11667.87</b>		0.001– <b>1.7441</b>			85
	Lucknow (peri-urban regions)		0.947– <b>249.2786</b>			0.000– 0.135		8.550– 101.000	0.090– 3.330	0.020– 8.580	119.33– <b>988.248</b>		90
	Delhi & Dehradun (urban and peri-urban areas)	0.003– <b>0.757</b>	0.014– <b>1.070</b>	0.022– 0.832	0.070– <b>9.360</b>	2.400– 24.800					15.000– 59.000		89
	Bokaro, Jharkhand	0.01–0.04	<b>0.79–2.18</b>		0.80–2.90		0.006– <b>0.05</b>			0.50– <b>2.07</b>			91
	Yamuna floodplain, New Delhi		BDL– <b>2.2</b>	BDL– <b>173.1</b>			1.4–206.2	0.2– <b>19.0</b>	BDL– <b>27.9</b>	15.7–78.3			92
	Kolkata, West Bengal	0.075– <b>0.285</b>							0.12–0.34	0.112– <b>1.33</b>			93
	Dhaka Export Processing Zone Chittagong	<b>0.82–4.85</b>		1.19– <b>11.84</b>	7.16– 37.73	108.57– <b>560.44</b>			2.41– <b>26.63</b>	2.41– <b>26.34</b>	11.43– 67.06		95
	Savar Upazila, Dhaka	0.002– 0.076		<0.1	<0.1–0.75	BDL	6.444– 136.3	<0.03	0.99– 5.720	0.072– 0.840	0.026– 1.596		98
	Pakshi, Pabna	<0.1							<0.1– 0.109	1.069– 0.188	0.119– 0.111–		97
Bangladesh		0.016– <b>1.31–3.89</b>		0.022– 0.094	0.094– <b>2.1–33.16</b>	2.97– 25.45			0.840	1.596– 0.840	1.206– 11.305		99
		0.14– <b>4.09</b>		2.1–33.16		25.45				0.066– 0.066			100
		<b>1.00–3.05</b>			11.20– <b>510.31</b>	125.58– 18.25			374.23	374.23	5.83– <b>8.75</b>		102
	Savar Upazila, Dhaka	<b>0.84–1.45</b>											

Table 2 (Contd.)

Country	Sampling site	As	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Pb	Zn	References
Pakistan	Dhaka-Mymensingh Highway	0.001		<b>7.98-9.02</b>	9.86-17.6	183.4-387.6		16.9-387.6	2.02-2.92	0.001	28-80.7	103	
	Gujranwala	0.15- <b>3.06</b>	0.86- <b>4.32</b>	<b>31.1-78.2</b>	148-261	79.2-107	3.81-9.61	<b>1.05-6.29</b>	51.8-68.3	104			
	Vehari	<b>BDL-3.7</b>	<b>0.0-3.7</b>	<b>19-55</b>	<b>5-81</b>	<b>228-1125</b>	14-119	0.0-7.8	<b>25-38</b>	<b>57-148</b>	109		
	Lahore	0.477- <b>3.50</b>	0.149- <b>0.944</b>	<b>1.8-7.5</b>	<b>6-38</b>	11.7-27	7.2-39.3		1.065- <b>7.83</b>		108		
	Khyber Pakhtunkhwa region							12.4-60.4	<b>15.6-61.2</b>	<b>7-41</b>	7.9-34.9	107	
	Mingora, Swat			0.07-0.14	0.43-2.30	1.70-		0.08-0.97	0.20-0.39	<b>0.67-3.58</b>		106	
Punjab	Kirri Shamozaï Bhakkar	<b>0.70-11.0</b> <b>0.67-8.0</b>	1.01- 14.89	<b>BDL-3.43</b>	<b>75.89</b>	<b>1.22-4.61</b>	10.6-33.5	<b>12.7-329.8</b>	<b>2.90-63.1</b> 6.89-85.0	<b>5.28-66.0</b> 4.29- <b>67.90</b>	<b>5.0-44.0</b> <b>0.42-500</b>	4.90-23.0	112
	Dera Ghazi Khan, Punjab	0-0.001	0-0.18						0-2.0		<b>0.42-500</b>	111	
	Sargodha district	1.153- <b>1.389</b>	0.037- 0.095	0.61- 0.892	0.625- 0.921	1.165- 2.399		1.268- 1.816	0.561- 0.652		0.565- 0.585	115	
	Faisalabad and Multan	0.19-5.36							0.21- <b>15.34</b>		0.218	116	
	Sargodha											117	
	Bahawalpur	0.0009- 0.07	0.14-0.25	<0.01					0.01-0.08	<b>0.15-0.35</b>	<b>9.071</b>	118	
China	Pearl River Delta, South China	0.033- 0.063	0.027- 0.060	0.095- 0.23				0.0014- 0.0026		0.055- 0.06		119	
	Suxian district	0.014- <b>1.780</b>	0.002- <b>2.918</b>			0.155- 3.125			0.004- 0.361	1.151- 54.65		36	
	Xi River, Northern China	0.0056- 0.0365		0.1529- 0.3287		0.1529- 2.54-14.3			0.0017- 0.06	0.5931- 2.488		120	
	Baiyin city, Gansu Province	<b>0.12-2.87</b>	<b>0.06-6.47</b>	<b>0.82-16.8</b>					<b>0.32-7.8</b>	<b>16-172</b>		121	
	Permissible limits	0.5 <sup>85</sup>	0.2 <sup>59</sup>	50 <sup>59</sup>	2.3 <sup>59</sup>	73.3 <sup>59</sup>	425.5 <sup>59</sup>	0.03 <sup>85</sup>	500 <sup>59</sup>	10 <sup>60,110</sup>	0.3 <sup>59</sup>	59, 60, 85, 102 and 110	
	Oral reference dose	0.0003 <sup>87</sup>	0.001 <sup>87</sup>	0.0003 <sup>87</sup>	1.5 <sup>84</sup>	0.04 <sup>87</sup>	0.7 <sup>87</sup>	0.14 <sup>84</sup>	0.02 <sup>84</sup>	0.004 <sup>87</sup>	0.3 <sup>87</sup>	84 and 87	

Vegetables watered with untreated wastewater had a notable buildup of HMs, according to a study conducted on the Agra Canal in Faridabad. The metals were ranked according to the extent of accumulation as Mn > Zn > Ni > Cu > Cr > Pb > As. Metal bioaccumulation varied considerably between leafy and non-leafy vegetables. The health risk index for manganese, nickel, and lead was above 1, indicating significant health hazards for consumers of these wastewater-irrigated vegetables.<sup>84</sup> A study found that vegetables in the Yamuna River floodplain in Delhi had varying degrees of trace element contamination; leafy vegetables had higher concentrations of Mn, Co and Ni, while fruit and inflorescence vegetables had higher levels of Pb, Zn and Cr. It also found that the highest translocation efficiency from soil to vegetables was for Cr. Most significantly, the research indicated potential carcinogenic risks due to consumption of all studied vegetables, with higher carcinogenic and non-carcinogenic risks for cauliflower and brinjal.<sup>92</sup> Significant HM contamination was found in the soil, vegetables, and grain crops near the Bokaro thermal power station in Jharkhand. The study discovered that the levels of Cd and Pb in every vegetable and grain crop sampled were higher than allowed. Cd had the highest transfer factor for HMs due to its great mobility. Adults were at significant risk of non-carcinogenic health problems due to high concentrations of Cr, Cd and Pb in grain crops and vegetables. Moreover, carcinogenic risks associated with Pb and Cr were also identified for adults.<sup>91</sup> A study focusing on areas receiving untreated wastewater from carpet industries in northern India revealed high concentrations of HMs in irrigation water, crops and soils. Notably, Cd levels in all tested vegetables exceeded both national and international safety limits.<sup>86</sup>

A study carried out in the industrialised city of Solapur, Maharashtra, examined the presence of HMs in twenty-four distinct varieties of fruits and vegetables. Varying levels of Pb, Cd, As, and Hg were found, with vegetables generally showing higher metal accumulations than fruits. Garlic and potato were identified as having the highest HM accumulation. Concerningly high human health risk index values were found in several vegetables, indicating possible negative health impacts from their consumption.<sup>85</sup> A comprehensive study conducted across a 100 km radius around Lucknow, India, revealed significant contamination of toxic HMs (Cd, Cr, Pb, and Ni) and metalloid (As) in vegetables. Notably, the edible parts of vegetables contained significantly higher levels of these elements than the maximum allowable concentration. The study highlighted a high bioaccumulation factor for Cd and Ni, indicating their entry into the food chain primarily through contaminated irrigation water. Although ecological risk indices showed moderate risk, As, Ni, Cd and Cr showed high carcinogenic risk factors, suggesting potential long-term health hazards for consumers.<sup>83</sup> A study in Haridwar, India, focused on the impact of integrated industrial wastewater irrigation on spinach cultivation. While integrated industrial wastewater irrigation increased crop yield and growth, it also led to greater build-up of HMs in spinach tissues. Prediction models based on soil characteristics were developed to evaluate HM uptake by spinach, which could be

valuable for policymakers in formulating guidelines for controlled irrigation using industrial effluents.<sup>81</sup>

Another study in Kolkata revealed high levels of Pb, Cd, and Ni in commercially grown vegetables from 8 zones irrigated with wastewater and affected by solid waste dumping. Leafy vegetables—particularly red spinach, bathua, coriander, and mustard—showed higher HM accumulation (Pb: up to 1.3 mg kg<sup>-1</sup>; Cd: up to 0.2 mg kg<sup>-1</sup>; Ni: up to 0.3 mg kg<sup>-1</sup>) compared to fruit vegetables. Spinach, capsicum, and Malabar spinach from several zones demonstrated TF values > 1 for Cd and Pb, suggesting their potential as hyperaccumulators and their role in enhancing dietary exposure to toxic metals. Health risk assessments identified Dhapa and Chhelegoalia as regions of greatest concern, with red spinach, mustard and coriander showing hazard index values greater than 1. Notably, the roots of most vegetables retained more heavy metals than shoots or fruits, although exceptions were observed where shoot concentrations surpassed root levels, particularly for Pb in coriander and spinach.<sup>93</sup>

These studies collectively emphasize the widespread issue of HM contamination in vegetables due to wastewater irrigation in India. They highlight the variability in metal accumulation across different vegetables, with leafy vegetables generally showing higher levels of contamination. These findings underscore the urgent need for regular monitoring of soil, water, and vegetables in areas using wastewater for irrigation, as well as the implementation of proper wastewater treatment methods before agricultural application to mitigate potential health hazards.

#### 4. Contamination of vegetables irrigated with wastewater in Bangladesh

Like many developing countries, Bangladesh is facing significant issues related to the problem of HM pollution in crops that are irrigated with wastewater. This problem stems from the widespread use of untreated or partially treated wastewater for crop irrigation, a practice that has far-reaching implications for food safety and public health. Numerous studies have delved into this matter, revealing alarming levels of HM accumulation in various vegetables across different regions of Bangladesh.

A comprehensive investigation in the Tangail district of Bangladesh evaluated the concentrations of Pb, Cu, Ni, Cr, As and Cd in rice and vegetable crops irrigated with industrial effluents. The analysis yielded mean concentrations of 16.26, 16.11, 13.99, 2.28, 1.86, and 7.93 mg kg<sup>-1</sup> for Cr, Ni, Cu, As, Cd, and Pb respectively in the examined samples. The study identified differential accumulation patterns among crop species, with okra exhibiting the highest metal concentrations, followed by chili, bitter gourd, papaya, brinjal, beans, bottle gourd, rice, cucumber, sponge gourd, and Indian spinach, in descending order of accumulation. Estimated daily intake values were calculated for the analyzed metals. With the exception of Cu, all metals examined exceeded the maximum tolerable daily intake, indicating potential consumer health risks. Furthermore, target



hazard quotients for Pb, Cd, As, Cu and Ni were determined to be greater than 1, suggesting significant non-carcinogenic risks for both children and adult populations consuming these vegetables. The study also evaluated the target carcinogenic risk for As and Pb, which exceeded the United States Environmental Protection Agency (USEPA) threshold level of  $10^{-4}$ ,<sup>122,123</sup> indicating a heightened cancer risk linked with eating these contaminated vegetables.<sup>100</sup>

Another study focused on the translocation and bioaccumulation of trace metals from industrial effluents to locally grown vegetables in the heavily industrialized area along the riverbank of the Buriganga River in Dhaka. This research analyzed soil samples and various vegetables, including red amaranth, spinach, jute leaf, bottle gourd, mustard green, and water spinach. In the vegetables, chromium and lead concentrations were found to be around  $20 \text{ mg kg}^{-1}$  and  $0.5 \text{ mg kg}^{-1}$ , respectively, significantly exceeding the permissible limits set by WHO/FAO (Cr) at  $2.3 \text{ mg kg}^{-1}$  and lead (Pb) at  $0.3 \text{ mg kg}^{-1}$ .<sup>60</sup> The investigation revealed that metal concentrations in the analyzed vegetables exceeded the established safety thresholds, with chromium. These findings provide evidence for the translocation of HMs from contaminated agricultural land to the edible portions of the vegetables. Hazard index calculations were performed for all vegetables in the study, yielding values exceeding 1, which indicates severe health risks associated with their consumption. Moreover, the total cancer risk values for Cr, nickel (Ni), and Pb in the vegetables were approximately  $1 \times 10^{-3}$ , surpassing the USEPA threshold of  $1 \times 10^{-6}$ . This suggests a significant carcinogenic risk linked to the consumption of these contaminated vegetables.<sup>101</sup> A separate study examined HM contamination in vegetables cultivated in industrially polluted regions across Bangladesh. This research quantified the concentrations of Fe, Ni, As, Cd, Cu, Mn and Pb in various crops and vegetables. The metal concentrations were found to be in the following consistent order: Fe > Mn > Ni > Cu > Pb > Cd > As. Target hazard quotient and target carcinogenic risk approaches were used in health risk assessments, and the results showed that consumption of these metal-contaminated vegetables could increase carcinogenic and non-carcinogenic risks for both adult and child populations.<sup>102</sup>

The level of HM pollution in vegetables grown along Bangladesh's main traffic route, the Dhaka–Mymensingh Highway, was evaluated in a recent study. 45 vegetable samples were analysed and collected from five distinct sampling sites with varied land use patterns adjacent to the highway. The analysis revealed the following average concentrations (in  $\text{mg kg}^{-1}$ ) for bottle gourd: Cu (14.9), Fe (281.2), Mn (387.6), Zn (49.0), Cr (10.1), and Ni (2.92). Esculent exhibited concentrations of Cu (17.6), Fe (183.4), Mn (107.2), Zn (80.7), Cr (7.98), and Ni (2.34). The average concentrations of Mn, Cr and Zn in these vegetables exceeded the established acceptable limits. The target hazard quotient for Cr in bottle gourd was found to be 9.52, suggesting a substantial non-carcinogenic risk. The target carcinogenic risk was highest for Cr in bottle gourd at 0.014.<sup>103</sup> A separate study focused on industrial areas of Dhaka, examining water, soil, and vegetables for HM contamination. The investigation determined that the order of metal contents in

contaminated irrigation water was Fe > Cu > Zn > Cr > Pb > Ni > Cd, with a similar pattern observed in arable soils. Notably, the mean concentrations of Cu, Fe, and Cd in irrigation water and Cd content in soil significantly exceeded the recommended thresholds. Although the majority of the vegetables under investigation showed HM buildup below the Joint FAO/WHO Expert Committee on Food Additives' (1999)<sup>124</sup> maximum allowable threshold, cadmium levels were higher.<sup>95</sup>

These investigations collectively underscore the critical issue of HM contamination in vegetables irrigated with wastewater in Bangladesh. The consistent findings of elevated contamination of toxic metals such as cadmium, chromium, lead, and arsenic in various vegetables cultivated across different regions of the country raise significant concerns regarding food safety and public health.

## 5. Contamination of vegetables irrigated with wastewater in Pakistan

Recent investigations into HM accumulation in vegetables due to wastewater irrigation have been conducted across diverse regions of Pakistan. These studies have revealed concerning contamination levels and potential health risks associated with this practice.

A survey experiment implemented in Multan and Faisalabad identified significant levels of Cd and Pb in soils subjected to wastewater irrigation. However, the contamination remained below the established allowable limits in both soil and water values. Among the plant species examined, Cd and Pb accumulation was highest in *Zea mays*. The study quantified the translocation factor from soil to root, with *Brassica oleracea* demonstrating the highest factor for Cd (7.037) and *Zea mays* for Pb (6.383). For Cd, target hazard quotient values exceeded non-carcinogenic thresholds in the majority of vegetables analyzed, with *Allium cepa* exhibiting the highest value (5.256). In contrast, target hazard quotient levels for Pb were generally below non-carcinogenic limits, with exceptions observed in *Allium cepa*, *Solanum lycopersicum*, and *Solanum tuberosum*.<sup>116</sup>

Another study near a sugar mill in the Sargodha district examined HM contents in water–soil–coriander samples. The mean concentrations of all metals except manganese exceeded permissible limits in sugar mill wastewater. In coriander specimens, only Cd had higher mean levels in both groundwater and sugar mill wastewater irrigated sites. Health risk index values indicated high risks from Ni, Cu, Fe, Mn, Zn and Cd in coriander irrigated with wastewater.<sup>125</sup> Research in Chenab Nagar, Chiniot, found that Cr and Co concentrations in crops irrigated with wastewater exceeded WHO recommended levels. All studied vegetables had unsafe Cd concentrations compared to FAO limits. Coriander, sponge gourd, and cauliflower also had unsafe Cr levels, while cauliflower showed unsafe Pb concentration. The highest health risk index values were associated with Cd and Cr for all vegetable samples.<sup>126</sup> A study in Faisalabad analyzing samples of okra, pumpkin, brinjal, spinach, tomato, potato and cabbage found that 27% of samples had Cd levels, 50% had Pb levels, and 63% had Hg



levels above European Commission standards. This indicates widespread HM contamination in vegetables from this region.<sup>127</sup>

In the Vehari district, a study found that soil samples exceeded limit values for several potentially toxic elements, including arsenic, cadmium, copper, chromium, manganese, iron, and zinc. Analysis of soil risk indices, specifically the potential ecological risk index and pollution load index, demonstrated severe contamination levels within the investigated area. These indices provide quantitative measures of the environmental impact and cumulative effects of multiple HMs in the soil ecosystem. Some vegetables exhibited high metal accumulation indices, signifying potential health hazards.<sup>109</sup> Research in the Sargodha district focused on iron (Fe) contamination in leafy and root crops irrigated with wastewater, canal water, and tube well water. While Fe concentrations in crops were within WHO permissible limits, the pollution load index indicated soil contamination. This study emphasized the need for regular monitoring to prevent excessive Fe accumulation in the food chain.<sup>105</sup>

A comprehensive study conducted in Dera Ghazi Khan investigated the presence of HMs in wastewater, soil, and diverse plant species. The findings revealed a predominance of chromium (Cr) and lead (Pb) contamination in soil samples. Analysis of plant tissues indicated that the concentrations of Cr, nickel (Ni), manganese (Mn), and Pb surpassed the safety thresholds established by WHO/FAO. Health risk index calculations were performed to assess potential consumer exposure. The results indicated elevated risk levels associated with the consumption of several crops, notably spinach, wheat, brinjal, tomato, and red corn. For the adult population, copper (Cu), zinc (Zn), and Cr were identified as presenting heightened risk factors.<sup>113</sup> An investigation was conducted in Mingora, located in the Swat region, to compare the accumulation of HMs in both soil and vegetable samples under two distinct irrigation conditions. The study contrasted the effects of industrial wastewater irrigation with those of tube well water irrigation. Soil and vegetables irrigated with IWW showed higher concentrations of HMs, often exceeding WHO limits. The metal transfer factor was particularly high for lead in ladyfinger and green pepper and for manganese in pumpkin. Health index values indicated that long-term consumption of these vegetables poses a greater threat to adults than children.<sup>106</sup> Another study conducted in Punjab, Pakistan, from 2016 to 2019 examined contamination of nickel, cadmium, lead, and chromium levels in vegetables irrigated with wastewater across eight districts. The results indicated varying concentrations of chromium, with the highest levels observed in contamination found in the Gujranwala district, with 44% of vegetable samples contaminated with nickel, 87% with cadmium, 97% with lead, and 88% with chromium. Analysis showed that leafy vegetables like mustard leaves, spinach, and lettuce accumulated higher levels of HMs compared to other vegetables. The study concluded that cultivation of vegetables with untreated wastewater should be prohibited due to the significant contamination levels observed.<sup>110</sup>

An investigation was conducted in the peri-urban regions of Bhakkar, Pakistan, to assess the accumulation of HMs in vegetables irrigated with wastewater. The study's findings revealed significant concentrations of cadmium, cobalt, nickel,

manganese, lead, and iron in both the soil and the vegetable samples. Although the HM content in the vegetables did not exceed the limits recommended by the WHO, the data suggest potential environmental and health concerns that warrant further examination. The daily intake of these metals through vegetable consumption was considerable for both adults and children. Lead and cadmium were identified as posing the highest health risks, with the study concluding that regular consumption of wastewater-irrigated vegetables may present significant health hazards.<sup>111</sup> Research conducted in Kirri Shamzai, Pakistan, investigated eight harmful metals (copper, iron, zinc, manganese, lead, cadmium, nickel, and chromium) in water, soil, and vegetable samples. The results showed that vegetables grown on wastewater-irrigated land had significantly higher HM concentrations compared to those grown with freshwater irrigation, often exceeding USEPA and WHO limits. Principal component analysis identified lead, copper, and chromium as the main contaminants affecting water quality and posing health risks. The study found that leafy vegetables accumulated higher quantities of HMs compared to root and fruit vegetables, with some vegetables deemed harmful for human consumption due to high health risk index values.<sup>112</sup>

Results reported in a study conducted near Multan found that the levels of lead and cadmium are higher than FAO/WHO standards in okra and brinjal samples.<sup>128</sup> Similarly, research in Dera Ismail Khan reported high lead accumulation in two varieties of *Raphanus sativus* irrigated with sewage water, with the local variety showing higher bioaccumulation than the exotic one.<sup>129</sup>

A health risk assessment in Bahawalpur's urban and peri-urban areas found elevated levels of Cd, Cr, Pb, and Ni in wastewater used for irrigation, despite compliant pH and total dissolved solids levels. Vegetables grown in sandy clay loam soils showed metal accumulation factors  $>1$  for Ni, Cd, and Cr. Although health risk index values ranged between 0.313 and 0.515—within safe limits—target hazard quotient (THQ) values exceeded 1 for all vegetables, indicating potential non-carcinogenic health risks.<sup>118</sup> A field study evaluating wastewater irrigation with organic and mineral fertilizers found significantly elevated Pb levels in soils and six vegetables irrigated solely with wastewater, often exceeding WHO limits. However, combining wastewater with organic (e.g., cow and poultry manure, leaf litter, and sugarcane bagasse ash) or mineral fertilizers reduced Pb accumulation in soil and crops. Manure was particularly effective, lowering Pb bioavailability by complexing with organic matter and raising soil pH. Daily Pb intake and health risk index were highest in wastewater-only treatments, with spinach (1.022) exceeding the safe threshold. All other treatments remained below the HRI limit of 1, indicating that health risks only arose under unamended wastewater use. The study underscores the effectiveness of integrated nutrient management, especially organic amendments, in mitigating Pb uptake and improving food safety in wastewater-irrigated systems.<sup>117</sup>

These investigations collectively demonstrate the pervasive HM accumulation in wastewater-irrigated vegetables across diverse Pakistani regions. The findings consistently indicate that leafy vegetables exhibit elevated HM concentrations

relative to other vegetable categories. While some studies found contamination levels below permissible limits, others reported alarming concentrations of toxic metals, particularly lead and cadmium. The variation in results across different regions underscores the need for localized assessments and targeted interventions to reduce health hazards linked to ingestion of wastewater-irrigated produce in Pakistan.

## 6. Contamination of vegetables irrigated with wastewater in China

Over the past two decades, researchers have extensively investigated HM contamination in vegetables irrigated with wastewater across various Chinese regions. These studies have provided crucial insights into pollution levels, potential health risks, and strategies for mitigation.

In the early 2000s, a study examined copper toxicity in vegetables grown in polluted environments. The research found that *Brassica chinensis* (cabbage), *Brassica chinensis* (pichkoi), and *Apium graveolens* var. dulce had copper bioaccumulation levels of 19.4, 5.5, and 30.9 mg kg<sup>-1</sup> respectively, far exceeding what is considered safe.<sup>130</sup> A decade later, researchers investigated HM accumulation of cadmium, lead, mercury, chromium and arsenic in six different leafy vegetables near the Pearl River in South China. Their findings showed that among the vegetables studied, lettuce, Chinese cabbage, pakshi, and Chinese leaf mustard pakshi were less prone to absorbing HMs compared to other leafy varieties.<sup>119</sup> Further research in 2016 looked at 22 different vegetable species grown in contaminated fields in the Suxian district. The study revealed a clear pattern of HM accumulation, with leafy vegetables showing the highest levels, followed by stalk vegetables, root vegetables, solanaceous vegetables, legume vegetables, and finally melon vegetables, which had the lowest accumulation.<sup>36</sup>

Another study analyzed vegetables grown in soils with a 50-year history of treated wastewater irrigation. Among the 20 common vegetable species examined, researchers observed significant variations in their ability to accumulate HMs, particularly cadmium, with more than 100-fold difference in Cd concentration between the highest- and lowest-accumulating vegetables. Their health risk assessment indicated that spinach, Chinese lettuce, and Chinese chives posed potentially severe health risks with a health risk index >5, while vegetables like tomatoes, kidney beans, potatoes, and cabbage showed minimal risk (health risk index <1).<sup>121</sup> A comprehensive study in northern China, specifically the Xi River sewage irrigation area of Shenyang city, highlighted cadmium as a major pollutant. The metal exceeded standard levels by 1.82 times and accumulated significantly in the soil. The researchers also noted that different vegetables had varying BCFs, with leafy vegetables showing the highest tendency, followed by rhizome and solanaceae vegetables. Moreover, the carcinogenic risk for cadmium in some vegetable consumption was higher and warranted attention.<sup>120</sup> A recent study examined agricultural soils near Hongfeng Lake in Guizhou Province. The researchers identified mercury as the main pollutant, with arsenic and lead as secondary concerns. Interestingly, while arsenic

posed the primary non-carcinogenic risk, chromium was found to be the main carcinogenic threat.<sup>131</sup>

These studies collectively highlight the complex nature of HM contamination in vegetables irrigated with wastewater across China. They emphasize the importance of considering factors such as soil characteristics, crop selection, and regional differences when developing strategies to reduce health risks associated with HM accumulation in vegetables. The research underscores the need for continuous monitoring and targeted interventions to ensure food safety in areas where wastewater irrigation is practiced.

## 7. Comparative analysis of heavy metal contamination in developing countries

The examination of metal concentrations across India, China, Bangladesh, and Pakistan reveals significant variations and critical contamination issues, as shown in Table 2, emphasizing the urgent need for targeted interventions and enhanced regulatory measures. The severity of contamination varies by metal and location, but certain areas emerge as hotspots for multiple contaminants. In India, the Yamuna Floodplain in New Delhi and peri-urban areas of Lucknow show alarmingly high levels of several metals. Studies evaluating the HM contamination in urban and peri-urban areas of India consistently showed high levels of HMs in vegetable samples, signifying the extent of this contamination in the food of the urban population. In Pakistan, regions like Vehari and Bhakkar are similarly affected. Bangladesh faces severe contamination near the Dhaka Export Processing Zone and in the Tangail district. The limited data from China also indicate significant contamination in the Suxian district and Baiyin city, Gansu Province. In India, Lucknow's peri-urban regions report alarmingly high levels of As, Cd, Cr, Cu, Mn and Ni, which far exceed the permissible limits and had the highest concentrations among all the other locations studied. The highest levels of Co (Kirri Shamozai), Fe (Dera Ghazi Khan) and Pb (Bhakkar) were found in Pakistan and the highest mercury levels were in Dhaka, Bangladesh, as shown in Table 2.

Lead pollution was particularly severe, with numerous locations exceeding the 0.3 mg kg<sup>-1</sup> limit. India's Dehradun (49.54–86.69 mg kg<sup>-1</sup>) and Patna (9.42–30.08 mg kg<sup>-1</sup>) showed high levels, while Pakistan's Bhakkar region reported an extreme range of 0.42–500 mg kg<sup>-1</sup>. In all the studies across India and Pakistan, lead levels exceeded the permissible limits. These levels reflect severe environmental pollution, with potential long-term health effects on the population. Similarly, Baiyin city in China reports concerning levels of Pb concentrations (0.32–7.8 mg kg<sup>-1</sup>), surpassing the safety threshold of 0.3 mg kg<sup>-1</sup>. In Bangladesh, lead contamination is notable in the Tangail district, exceeding safety thresholds even at a lower value of 0.84 mg kg<sup>-1</sup>.

Arsenic contamination is widespread, with levels surpassing the 0.5 mg kg<sup>-1</sup> limit in multiple locations. In Bangladesh, Savar Upazila (0.84–1.45 mg kg<sup>-1</sup>) and Tangail (1.31–3.89 mg kg<sup>-1</sup>) showed elevated concentrations. China's Suxian district (0.014–1.780 mg kg<sup>-1</sup>) and Baiyin city (0.12–2.87 mg kg<sup>-1</sup>) also exceeded



safe levels. India presented severe contamination, particularly in peri-urban Lucknow ( $13.73\text{--}2392.05\text{ mg kg}^{-1}$ ), while Pakistan's Vehari (BDL- $3.7\text{ mg kg}^{-1}$ ) and Lahore ( $0.477\text{--}3.50\text{ mg kg}^{-1}$ ) also reported concerning levels. Cadmium pollution is equally troubling, with the  $0.2\text{ mg kg}^{-1}$  threshold frequently surpassed. Bangladesh's Dhaka Export Processing Zone reported levels of  $0.82\text{--}4.85\text{ mg kg}^{-1}$ , while China's Baiyin city showed concentrations of  $0.06\text{--}6.47\text{ mg kg}^{-1}$ . India's peri-urban Lucknow again emerged as a hotspot with  $5.90\text{--}503.86\text{ mg kg}^{-1}$ , and Pakistan's Kirri Shamoza registered  $0.70\text{--}11.0\text{ mg kg}^{-1}$ . Chromium contamination exceeds the  $2.3\text{ mg kg}^{-1}$  limit in several areas. Bangladesh's Tangail ( $2.1\text{--}33.16\text{ mg kg}^{-1}$ ) and Dhaka Export Processing Zone ( $1.19\text{--}11.84\text{ mg kg}^{-1}$ ) showed high levels. In India, the Yamuna floodplain in New Delhi (BDL- $173.1\text{ mg kg}^{-1}$ ) and Lucknow's peri-urban regions ( $7.14\text{--}215.02\text{ mg kg}^{-1}$ ) displayed severe contamination. Pakistan's Punjab region reported levels up to  $75.89\text{ mg kg}^{-1}$ . Copper levels above the  $73.3\text{ mg kg}^{-1}$  threshold were observed in Lucknow ( $65.58\text{--}868.53\text{ mg kg}^{-1}$ ) and parts of Pakistan, including Gujranwala ( $31.1\text{--}78.2\text{ mg kg}^{-1}$ ) and Vehari ( $5\text{--}81\text{ mg kg}^{-1}$ ). Iron contamination surpassed the  $425.5\text{ mg kg}^{-1}$  limit in several locations, with Lucknow again showing extreme levels ( $1460.28\text{--}27363.74\text{ mg kg}^{-1}$ ) and Pakistan's Vehari reporting  $228\text{--}1125\text{ mg kg}^{-1}$ . Mercury pollution exceeded the  $0.03\text{ mg kg}^{-1}$  limit in parts of India and Pakistan. While most areas in Bangladesh and China showed levels within safe limits, India's Patna, Bihar ( $0.145\text{--}0.705\text{ mg kg}^{-1}$ ) and Pakistan's Dera Ghazi Khan, Punjab ( $2.3\text{--}18.8\text{ mg kg}^{-1}$ ) reported significantly elevated concentrations. Manganese levels in most locations remained below the  $500\text{ mg kg}^{-1}$  threshold, except for Lucknow's peri-urban regions, which reported an astounding range of  $298.17\text{--}11667.87\text{ mg kg}^{-1}$ . Nickel contamination surpassed the  $10\text{ mg kg}^{-1}$  limit in several areas, including Bangladesh's Savar Upazila ( $17.66\text{--}30.33\text{ mg kg}^{-1}$ ) and Tangail ( $1.41\text{--}37.52\text{ mg kg}^{-1}$ ), India's Lucknow ( $19.96\text{--}82.62\text{ mg kg}^{-1}$ ), and Pakistan's Kirri Shamoza ( $5.28\text{--}66.0\text{ mg kg}^{-1}$ ) and Bhakkar ( $4.29\text{--}67.90\text{ mg kg}^{-1}$ ). Zinc concentrations also surpassed the  $99.4\text{ mg kg}^{-1}$  threshold in several areas, with Lucknow again showing the highest levels ( $119.33\text{--}988.248\text{ mg kg}^{-1}$ ).

The data presented paint a deeply concerning picture of widespread HM contamination across various regions in South Asian countries. The levels of contamination for multiple metals far exceed permissible limits in many locations, posing significant risks to human health and the environment. The data highlight the urgent need for comprehensive environmental monitoring and stringent regulatory measures to address contamination. There is a pressing need for comprehensive action plans to address contamination, safeguard public health, and ensure environmental safety.

## 8. Heavy metal accumulation patterns in vegetable crops: a comparative analysis of plant types and tissues

Research on HM contamination in vegetable crops reveals a consistent trend of higher metal accumulation in leafy vegetables compared to fruit vegetables such as brinjal or

chili,<sup>36,82,132</sup> suggesting higher HM accumulation capacity by leaves than fruits.<sup>133-135</sup> This phenomenon can be attributed to several physiological and morphological factors. Leafy vegetables, in particular, demonstrate significantly higher metal uptake, as shown in Fig. 4, for various heavy metals,<sup>84,85,121</sup> hypothesized to be due to their elevated transpiration and translocation rates,<sup>136</sup> facilitating enhanced HM absorption *via* roots and subsequent translocation to foliar tissues. Secondly, a significant amount of absorbed HMs in fruit plants is deposited in organs other than the edible parts, particularly in the leaves. Moreover, the larger surface area of leaves renders vegetables more susceptible to physical contamination from atmospheric deposition and soil particles.<sup>137,138</sup> Notably, members of the Brassicaceae family exhibit hyperaccumulator properties, consistently showing the highest total metal concentrations among studied crops.<sup>101,139</sup> Leafy vegetables such as spinach (*Spinacia oleracea*), fenugreek (*Trigonella foenum-graecum*), and coriander (*Coriandrum sativum*) consistently display higher metal content compared to non-leafy varieties.

Upon examining the distribution of HMs within plant structures, roots generally contain higher concentrations of HMs and metalloids compared to edible parts, as shown in Fig. 5.<sup>102,116,140</sup> This observation aligns with the well-documented

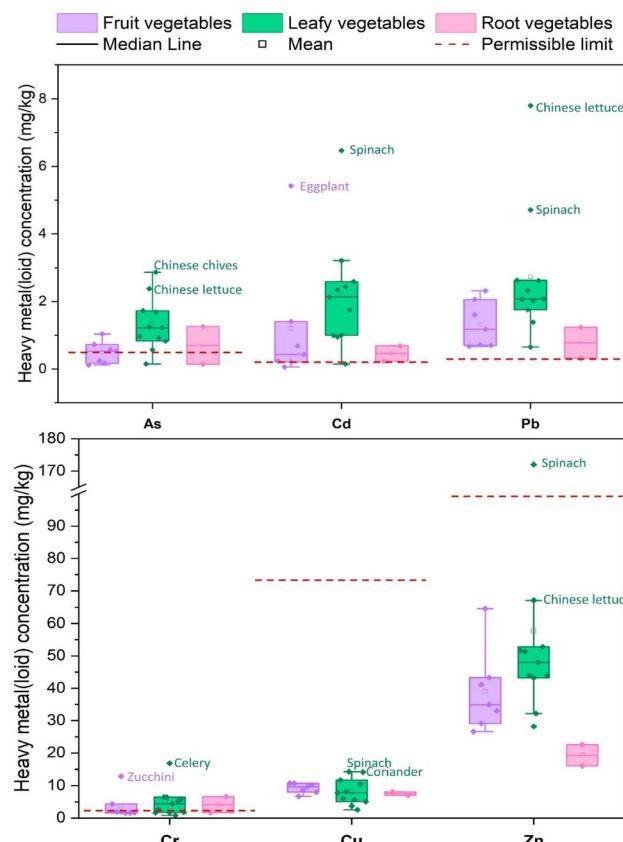


Fig. 4 Heavy metal(lloid) concentration in the edible parts of different types of vegetables, *i.e.*, fruit (cucumber, kidney beans, pepper, winged beans, brinjal, zucchini, and tomato), leafy (Chinese cabbage, spinach, shallot, Chinese chives, cabbage, celery, Chinese lettuce, garland chrysanthemum, coriander, leafy lettuce, and rape) and root vegetables (carrot and potato).<sup>121</sup>



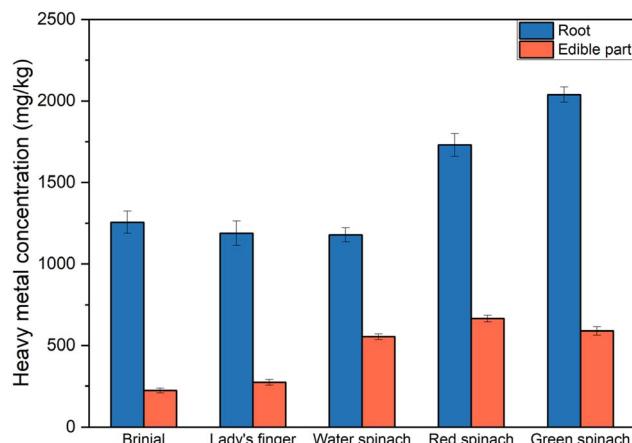


Fig. 5 Total heavy metal(lloid) concentration (As, Cd, Cr, Pb, Fe, Ni, and Mn) in the root and edible parts of various vegetables.<sup>102</sup>

phenomenon of high metal sequestration in root tissues coupled with limited root-to-shoot translocation for most metal(lloid)s.<sup>141,142</sup> Consequently, vegetables where fruit parts constitute the edible portion may present a comparatively lower risk when cultivated in metal(lloid)-contaminated soils, as opposed to root vegetables. These results underscore the importance of species-specific and tissue-specific analyses in assessing the potential health risks associated with HM contamination in vegetable crops. Although different vegetable crops have varying capacities for accumulating heavy metal(lloid)s from soils to their edible sections, crop selection may be utilised to mitigate the potential health concerns associated with eating vegetables grown in contaminated soils.<sup>121</sup> The data indicate that the accumulation of HMs varies significantly among plant species, attributed to differences in their accumulation capacities, rhizosphere characteristics, and interactions with soil properties affecting metal bioavailability. Thus, establishment of an effective and simpler metal transport model for prediction will help ensure global food safety and assess the effectiveness of various management techniques.

## 9. Conclusion

The analysis reveals widespread HM contamination in vegetables irrigated with wastewater across developing Asian countries, notably India, Bangladesh, Pakistan, and China, highlighting a critical public health and environmental concern. Cadmium, lead, chromium, arsenic, nickel, and mercury are the most commonly reported contaminants, with leafy vegetables such as spinach, amaranth, and cabbage, showing consistently higher HM accumulation than fruit and root vegetables. This trend, observed across multiple studies in different countries, suggests a common pattern of bioaccumulation that warrants particular attention.

Regional variability in contamination reflects complex interactions among soil properties, irrigation practices, and industrial pollution sources. Many studies report health risk indices above safe limits, highlighting significant non-carcinogenic and carcinogenic risks, especially for vulnerable

populations like children. These findings point to significant gaps in regulatory oversight and the enforcement of environmental and food safety standards. Mitigating these risks demands a comprehensive, interdisciplinary approach. Key strategies include upgrading wastewater treatment infrastructure, promoting best agricultural practices tailored to local conditions, and establishing routine monitoring systems for soil, water, and crop contamination. Public awareness and education on the risks of wastewater-irrigated vegetables and safe food handling are also critical. Future research should prioritize cost-effective remediation technologies, alternative irrigation sources, and long-term health impact studies.

While wastewater irrigation offers a practical response to water scarcity in the region, it poses serious, long-term risks to food safety and public health. Addressing this challenge requires coordinated action from policymakers, researchers, agricultural practitioners, and public health authorities. Only through integrated and sustained efforts can we ensure agricultural productivity without compromising human health, thereby advancing both food security and environmental sustainability in rapidly urbanizing, water-stressed regions of Asia.

## 10. Research gaps and future directions

Despite the growing body of research on HM contamination in vegetables, several significant knowledge gaps remain, pointing to critical areas for future research. One of the most pressing needs is for comprehensive, longitudinal studies to assess the long-term health impacts of consuming vegetables with elevated HM concentrations. While the acute toxicity of many HMs is well-documented, the effects of chronic, low-level exposure through dietary intake are less understood. Such studies would provide valuable insights into the cumulative health risks associated with long-term consumption of contaminated vegetables and help in developing more accurate risk assessment models. The combined effects of multiple HMs on human health, known as the cocktail effect, are not fully understood and require further investigation.

From a remediation perspective, there is a pressing need for research required to develop and validate economically viable soil remediation methods that effectively reduce HM contamination and limit vegetable uptake. While various remediation methods have been proposed, many are too expensive or impractical for widespread implementation in developing countries. Research into affordable, scalable solutions that can be readily adopted by farmers in resource-limited settings is crucial.

The search for sustainable alternatives to wastewater irrigation is another important area for future research. Studies on the feasibility and effectiveness of using treated greywater, implementing rainwater harvesting systems, or developing drought-resistant crop varieties could provide valuable alternatives to the use of contaminated water for irrigation. Future investigations should encompass a multifaceted approach, examining not only the technological feasibility but also the



socioeconomic determinants that may impact the implementation and widespread acceptance of these proposed alternatives.

Studies examining the effectiveness of existing policies and barriers to their implementation are essential for improving regulatory frameworks. Additionally, research on effective strategies for raising public awareness about the risks of HM contamination and promoting behavioural change is necessary.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Author contributions

Navneet Kaur: data curation, formal analysis, investigation, visualization, writing – original draft, and writing – review & editing. Jagdev Singh: conceptualization, formal analysis, investigation, visualization, and writing – original draft. Anand Mohan: conceptualization, validation, and writing – review & editing. Neeta Raj Sharma: writing – review and editing. Simranpreet Kaur Natt: writing – review and editing. Tabarak Malik: conceptualization, supervision, resources, and writing – review & editing. Madhuri Girdhar: conceptualization, supervision, and writing – review & editing.

## Conflicts of interest

The authors declare no competing interest.

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