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A Systematic Review and Meta-analysis to Investigate the Correlation Vegetable Irrigation with Wastewater and Concentration of Potentially Toxic Elements (PTES): a Case Study of Spinach (*Spinacia oleracea*) and Radish (*Raphanus raphanistrum* subsp. *sativus*)

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Abstract

Water shortage and stress around the world lead to the reuse of wastewater in many sectors while the recycling of water in agriculture as one of the most consumed sectors can boost the contamination of crops by potentially toxic elements (PTEs). Therefore, this study was aimed to investigate the correlation between the accumulation of PTEs (Fe, Zn, Cr, Ni, Cu, Pb, As, Cd, and Se) in edible parts of spinach and radish plants and sewage irrigation by the aid of a meta-analysis. Moreover, the non-carcinogenic risk (N-CR) and carcinogenic risk (CR) for health risk assessment of consumers were assessed through actual total target hazard quotient (TTHQact) and carcinogenic risk (CRact). After the screening process, 51 articles with 75 studies were included. According to findings, the rank order of PTEs in spinach and radish were Fe > Zn > Cr > Cu > Ni > Pb > Cd > As > Se and Fe > Zn > Cr > Ni > Cu > Pb > As > Cd > Se, respectively. PTE adsorption by edible parts of spinach (leafy vegetable) was higher than radish. The health risk assessment shows that residents in Iran, India, and China are at N-CR while the population of Iran, India, and Pakistan are facing CR.

Keywords Spinach · Radish · Irrigation · Wastewater · Meta-analysis · Potentially toxic elements

Introduction

Today, human beings face two serious challenges in water resources management: water scarcity and its related pollution consequences [1]. In recent decades, with a rapid increase in population and subsequently their water demand, one-third of renewable water is consumed. It is also estimated that during 1962–2011, 54% of global freshwater was declined [2].

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However, climate change and non-uniform temporal and spatial distribution of water on the planet can boost the problem [3]. For instance, many semi-arid and arid regions access to surface water just in rainy seasons, while irrigation water is needed for dry seasons [4]. Since in many regions, the agriculture sector is known as the largest water consumer [5], utilizing of treated domestic and industrial effluents for irrigation purposes is a common practice in some countries such as France, the USA, and Spain. However, agricultural irrigation by untreated wastewater in many developing countries is also observed [6–8].

Indeed, wastewater reuse in the agriculture sector has some benefits and drawbacks. On the one hand, the wastewater reuse in addition to supplying needed water has other advantages like recharging aquifers, surface, and groundwater storage and providing needed nutrients and organic matter for plants [9, 10]. On the other hand, since treated/untreated wastewater contains toxic substances like potentially toxic elements (PTEs), irrigation by wastewater leads to soil contamination and subsequently vegetable pollution which finally faces consumers with many health problems [9, 10]. PTEs due to their unique properties, such as non-biodegradability, long biological half-lives, and accumulative potential in different parts of crops, can enter the food chain and make health issues [11]. Little concentrations of PTEs can damage the nervous, enzymatic, endocrine, immune, skeletal, and circulatory systems. Others can make disorders in the lung, kidney, and liver and some are carcinogenic [12, 13].

Substances such as carbohydrates, proteins, vitamins, minerals, and dietary fibers, vegetables are considered as one of the most consumed sources in the human diet, around the world [14, 15]. Vegetables can adsorb PTEs in their parts. Leafy vegetables like spinach are ranked as highly contaminated [16]. However, since the root is the first part of vegetables exposed to contaminated water, it can also accumulate a high level of contamination [17]. In a study conducted by Ahmad et al. [18] in Sargodha, Pakistan, the concentration of PTEs in spinach irrigated by treated wastewater exceeded the maximum limits. In another study conducted in Khushab City, Pakistan [19], the same authors also observed this situation for Mn, Ni, and Pb in radish and spinach. Therefore, quantitative assessments, especially for heavy metal concentrations in vegetables irrigated by wastewater, are yet necessary to monitoring and assessing their health risk [20, 21].

However, there have been many studies around the world that exhibit the importance of the issue [17]; no study was conducted to pluralize PTE accumulation in radish and spinach vegetables irrigated by wastewater. In this study, for the first time, the PTE concentrations in the edible portion of the radish and spinach irrigated by wastewater were investigated by the aid of a meta-analysis

Material and Method

The Search Strategy and Protocol of the Study

The strategy of the search was conducted based on the Cochrane protocol (Fig. 1) [17, 22–24] in Web of Science, Embase, PubMed, and Scopus databases up to 31 September 2019. The strategy targeted for the articles that investigated PTE concentration in spinach and radish vegetables irrigated by wastewater. The used terms for the search process are described in Table 1S.

Inclusion/Exclusion Criteria

In this quantitative review, the studies were selected based on specific characteristics: (i) the studies with English language, (ii) the studies that measured heavy metal concentration in spinach and radish, and (iii) the studies that surveyed vegetables irrigated by wastewater. However, there were also characteristics that caused the exclusion of articles: (i) books, clinical trial studies, review articles, (ii) the studies where the source of irrigation was diluted, (iii) the studies where sludge was used with wastewater. In the papers that showed their results as a figure, GetData Graph Digitizer software (ver.2.24) was used for measuring the results. The papers with low-quality figures were also excluded.

Data Extraction

The extracted properties of papers were the year of study, country, and measurement statistics (sample size, mean ± SD, and range of PTE concentration). In order to accurate analysis, all measurement units were transformed into the dry-weight unit (mg/kg).

Meta-analysis and Statistical Analysis

In this study, the meta-analysis was done by STATA software version 14.0 (Stata Corporation, College Station, USA). The statistical parameters of average and standard error (SE) were applied to investigate the pooled concentration of PTEs [25, 26]. Table 2S bears all methodological equations in the study. Also, the statistical model of random effects (REM) was utilized for analysis. Indeed, the model is preferred when heterogeneity is above 50%. Chi-square test (I^2) as a determinant of heterogeneity percent was obtained at > 50% in the current study [27, 28].

Health Risk Assessment

Equations 2–7 (Table 2S and Table 3S) are utilized to calculate the non-carcinogenic risk (N-CR).

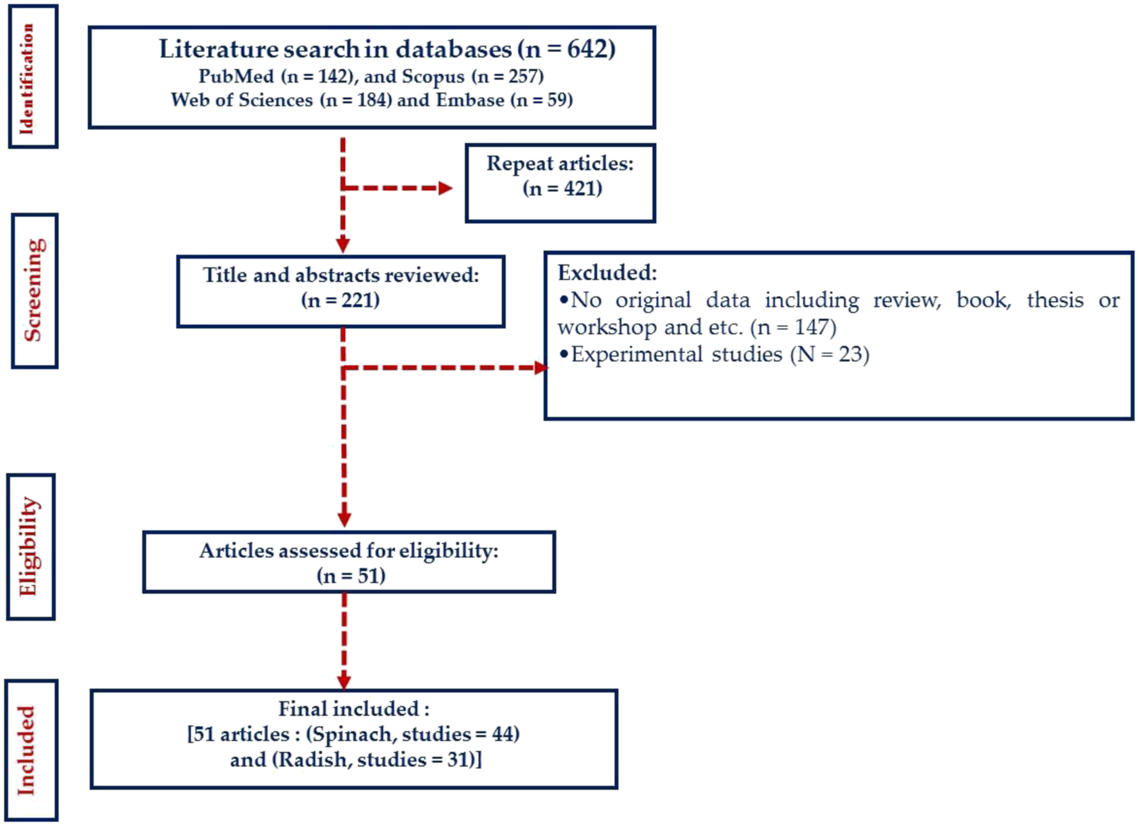

Fig. 1 Flow chart search of studies

Results and Discussion

Characteristic of Studies

After searching among the databases of Scopus, Web of Science, PubMed, and Embase up to 31 September 2019, 642 articles were retrieved. Exclusion of articles was conducted as follows: during the steps of removal of duplicate articles (*n* = 421) via EndNote software (version X9); removal based on the articles' title and abstract (*n* = 170); and removal based on the articles' full text (*n* = 0). Finally, 51 articles with 75 studies (spinach studies = 44; radish studies = 31) were downloaded and reviewed (Fig. 1).

Meta-analysis Findings

Generally, PTE concentration in vegetables irrigated by wastewater depends on many factors including wastewater types (domestic, industrial, or both), wastewater contents, wastewater treatment efficiency, irrigation time, soil type and its characteristics, plant characteristic (type, age, and parts), and PTE properties [29–31]; however, these pollutants can also be accumulated by another source like atmospheric deposition [32, 33]. An atmospheric pollutant can translocate to the plant through both the absorption of irrigation water and adsorption on the leaf surface [34]. Mentioned factors cause the variation of metal concentration in plants around the world (Tables 4S and 5S).

As demonstrated in previous studies, industrial wastewater depending on the product manufacturing process and utilized raw materials has a wide range and high content of metals than domestic [35–37]. According to Alemu et al., after surveying the suitability of treated tannery wastewater on vegetable growth, irrigation by high Cd content effluents causes the high accumulation of tomato vegetables [35]. In another study, the effect of vegetable irrigation by wastewater of three manufacturers in Sari, Iran, was investigated [36]; the areas irrigated by chrome chemical wastewater had more than two times Cr concentration. Moreover, treatment processes have an effective role in affluent content. Indeed, updated and high-efficiency units in wastewater treatment plants can significantly reduce the output pollutants [38–41]. Sometimes, the strict laws of countries can result in a further reduction in the effluent pollutants from treatment plants [42]. Long-term and repeated irrigation with wastewater significantly promotes of PTE penetration into soil and plants [43]. In a study done in Titagarh, West Bengal, India [44], long-term irrigation by wastewater causes a notable adsorption of PTEs by some vegetables while the soil type and bioavailability were the most important factors on the adsorption of PTEs by plants. The bioavailability is also affected by soil pH, cation exchange capacity (CEC), redox potential, and organic content, whereas

Table 1 The non-carcinogenic risk due to ingestion vegetables (spinach and radish) content of PTE

		Spinach						Radish						
		Afghanistan	Pakistan	Iran	Kenya	China	India	Afghanistan	Pakistan	Iran	Iraq	China	India	South Korea
Adults	As	3.16E−03	3.54E−02	1.38E+00		6.35E−02	7.04E-01		4.97E−03			7.70E−02		
	Pb		1.09E−02	1.16E−01		1.25E−01	2.58E−01	1.22E−03	6.41E−03	3.55E−04	7.54E−03	1.36E−02	4.08E−02	
	Cd		2.11E−02	6.56E−02					1.84E−03	4.26E−04	2.64E−03	1.39E−02		
	Zn	1.53E−03	1.40E−03	4.00E−02	7.18E−05	1.28E−02	4.28E−03	5.81E−04	4.40E−04			4.43E−03	1.35E−03	1.10E−03
	Cu	3.33E−03	2.80E−03	9.52E−02	2.16E−04	1.71E−02	9.01E−03	4.39E−04	5.88E−04		2.39E−03	5.31E−03	1.98E−03	5.84E−04
	Fe		2.30E−03				1.21E−02		2.36E−04				1.42E−03	
	Ni		7.80E−03	2.15E−02		5.31E−02	4.92E−02		6.78E−04		3.56E−03	2.77E−02	1.41E−02	
	Cr	9.49E−06	1.00E−04	7.09E−04		1.57E−04	2.97E−04		1.87E−05			1.72E−04	1.72E−04	
	Se		7.00E−04						2.30E−05					
	TTHQ	8.03E−03	8.18E−02	1.72E+00	2.88E−04	2.72E−01	1.04E+00	2.24E−03	1.52E−02	7.81E−04	1.61E−02	1.42E−01	5.98E−02	1.69E−03
Children	As	1.48E−02	1.65E−01	6.45E+00		2.96E−01	3.29E+00		2.32E−02			3.59E−01		
	Pb		5.08E−02	5.40E−01		5.83E−01	1.20E+00	5.69E−03	2.99E−02	1.66E−03	3.52E−02	6.36E−02	1.90E−01	
	Cd		9.84E−02	3.06E−01					8.57E−03	1.99E−03	1.23E−02			
	Zn	7.12E−03	6.60E−03	1.87E−01	3.35E−04	5.96E−02	2.00E−02	2.71E−03	2.05E−03			2.07E−02	6.30E−03	5.15E−03
	Cu	1.55E−02	1.33E−02	4.44E−01	1.01E−03	7.98E−02	4.20E−02	2.05E−03	2.75E−03		1.12E−02	2.48E−02	9.22E−03	2.73E−03
	Fe		1.06E−02				5.66E−02		1.10E−03				6.65E−03	
	Ni		3.65E−02	1.00E−01		2.48E−01	2.30E−01		3.17E−03		1.66E−02	1.29E−01	6.58E−02	
	Cr	4.43E−05	3.00E−04	3.31E−03		7.31E−04	1.39E−03		8.73E−05			8.03E−04	8.05E−04	
	Se	3.75E−02	3.30E−03		1.34E−03				1.07E−04					
	TTHQ	7.50E−02	3.85E−01	8.03E+00	2.69E−03	1.27E+00	4.84E+00	1.04E−02	7.10E−02	3.65E−03	7.53E−02	5.99E−01	2.79E−01	7.87E−03

Table 2 The actual non-carcinogenic risk due to ingestion vegetables (spinach and radish) content of PTE

Country	Adults		Children		Adults	Children
	Spinach	Radish	Spinach	Radish	TTHQ act	TTHQ act
Afghanistan	8.03E−03	2.24E−03	7.50E−02	1.04E−02	1.03E−02	8.54E−02
Pakistan	8.18E−02	1.52E−02	3.85E−01	7.10E−02	9.70E−02	4.56E−01
Iran	1.72E+00	7.81E−04	8.03E+00	3.65E−03	1.72E+00	8.03E+00
Kenya	2.88E−04		1.71E−03		2.88E−04	1.71E−03
China	2.72E−01	1.42E−01	1.27E+00	5.99E−01	4.14E−01	1.87E+00
India	1.04E+00	5.98E−02		2.79E−01	1.10E+00	2.79E−01
Iraq		1.61E−02		7.53E−02	1.61E−02	7.53E−02
South Korea		1.69E−03		7.87E−03	1.69E−03	7.87E−03

the increase in pH, CEC, and organic matter of soil leads to the decreasing of PTE mobility; increasing of redox potential results in the conversion of a soluble form of PTEs to insoluble [45–47]. Kabata-Pendias [48] argued that CEC has an inverse correlation with PTEs' bioavailability and influenced by soil clay content. Soil type can affect the translocating of PTEs from irrigation water into plants. For instance, it is proved that calcareous soils have generally less bioavailability for Zn and Ni [43]. All soils naturally contain a variety of PTEs, depending on the soil type. Ultramafic rocks and the soil derived from these rocks have a high content of Cr and Ni, or gabbro and basalt have a high content of Cu and Zn. In addition, Fe is one of the ten major elements which constitute more than 99% of the total content of the earth's crust [49, 50].

According to the results, the PTE ranking in spinach was Fe (374.12 mg/kg dw) > Zn (77.95 mg/kg dw) > Cr (22.43 mg/kg dw) > Cu (21.18 mg/kg dw) > Ni (19.11 mg/kg dw) > Pb (18.95 mg/kg dw) > Cd (4.76 mg/kg dw) > As (3.02 mg/kg dw) > Se (0.56 mg/kg dw) (Figs. 1s–9s). Also, the PTEs of Fe (116.55 mg/kg dw) > Zn (50.51 mg/kg dw) > Cr (21.81 mg/kg dw) > Ni (12.32 mg/kg dw) > Cu (9.05 mg/kg dw) > Pb (3.84 mg/kg dw) > As (0.73 mg/kg dw) > Cd (0.25 mg/kg dw) > Se (0.04 mg/kg dw) had the highest concentrations in radish, respectively (Figs. 10s–18s). The global average ranking of some metals in topsoil is Zn > Cr > Pb > Ni > Cu > Cd [50, 51] in which, considering Fe, ranking seems to be in alignment with the first three metals for both vegetables. Therefore, it can be claimed that soil type is one of the most influential parameters in PTE accumulation (Table 1).

Table 3 The actual carcinogenic risk due to ingestion vegetables (spinach and radish) content of PTE

Country	Spinach	Radish	CRact
Afghanistan	6.10E−07		6.10E−07
Pakistan	6.87E−06	9.59E−05	1.03E−04
Iran	2.67E−04		2.67E−04
Kenya			
China	1.23E−05	9.90E−06	2.22E−05
India	1.36E−04		1.36E−04
Iraq			
South Korea			

Nevertheless, the PTE concentration in the topsoil does not always reflect the concentration in the plant. With respect to the occurrence of various adsorption and physio-chemical mechanisms in plants, the concentration of PTEs in their parts is miscellaneous [45, 47]. For example, much of the fruit and seed tissue is composed of phloem, which can be used to prevent the accumulation of PTEs [41]. Based on the findings of Barman et al., no particular pattern for metal adsorption from soil to plants was observed [52]. For instance, Zn and Cu have an antagonistic correlation which results in more Zn accumulation in soil, depending sensitively on pH [53, 54].

In this study, it is found that edible part of spinach as a leafy vegetable has more PTE concentration than radish which was in concordance with the findings of a previous study [17] related to the cases of Cd, Cr, Fe, and Zn in the edible part of tomato than the onion. This can be attributed to pollutant adsorption on plant surfaces, or pollutant translocation from root to shoot and leaves, [47]. Of course, there is the hypothesis of PTE precipitation from the air to the leaf surface. However, because of the utilization of standard methods for sample digestion in most studies (washing the samples with distilled water), this hypothesis is likely to be rejected.

Health Risk Assessment

The actual total target hazard quotient (TTHQact) ≤ 1 is acceptable for the N-CR, but when the TTHQact > 1, N-CR in the exposed population is unacceptable [55]. The ranking of counties by TTHQact for adults was Iran (1.72E+00) > India (1.10E+00) > China (4.14E−01) > Pakistan (9.70E−02) > Iraq (1.61E−02) > Afghanistan (1.03E−02) > South Korea (1.69E−03) > Kenya (2.88E−04) and for children Iran (8.03E+00) > China (1.87E+00) > Pakistan (4.56E−01) > India (2.79E−01)

> Afghanistan (8.54E−02) > Iraq (7.53E−02) > South Korea (7.87E−03) > Kenya (1.71E−03) (Table 2).

TTHQact in Iran and India for adults and Iran and China for a group of children was > 1, which leads to a considerable N-CR for consumers (Table 2). If CRact > 1.00E−04, residents are at acceptable carcinogenic risk (CR), but when CRact < 1.00E−06, residents are not at acceptable risk [56]. Also, if 1.00E−06 < CRact < 1.00E−04, cancer threatens the consumers [56]. The countries ranking by CR due to As in spinach were Iran (2.67E−04) > India (1.36E−04) > China (1.23E−05) > Pakistan (6.87E−06) > Afghanistan (6.10E −07) and due to radish was Pakistan (9.59E−05) > China (9.90E−06) (Table 3). The ranking of countries by actual CR was Iran (2.67E−04) > India (1.36E−04) > Pakistan (1.03E −04) > China (2.22E−05) > Afghanistan (6.10E−07) (Table 3). CR act in Iran, India, and Pakistan due to ingestion spinach and radish content of As was higher than 1.00E−4; therefore, consumers are at the considerable cancer risk. Estimated TTHQact and CRact were different in the countries because of different concentrations of PTEs in spinach and radish, number of PTEs analyzed, ingestion rate, type of PTEs, exposure frequency, and exposure duration [57–59].

Conclusion

The principal objective of this work was the meta-analysis of the data-associated studies on metal accumulation in spinach and radish vegetables irrigated by wastewater and the following estimation of non-carcinogenic and carcinogenic risks in the consumers. In addition, N-CR for analyzed metals (except As) and CR for As based on countries were assessed. According to the results, the ranking of Fe > Zn > Cr > Cu > Ni > Pb > Cd > As > Se for spinach and the ranking of Fe > Zn > Cr > Ni > Cu > Pb > As > Cd > Se for radish were obtained. The health risk assessment results allowed us to conclude that adults and/or children residents in Iran, India, and China encounter unacceptable N-CR. Also, adults and/or children residents in Iran, India, and Pakistan are at the unacceptable CR. The results of the current study showed that the consumption of vegetables such as spinach and radish irrigated with wastewater can endanger the health of consumers in some countries; therefore, some actions are needed for the reduction of PTE contents in irrigation wastewater to standard levels.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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