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Determination of heavy metals in tomatoes cultivated under green houses and human health risk assessment

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RESEARCH ARTICLE

Abstract

The present work aims to assess the levels of some heavy metals in agricultural soils and Tomatoes in Jijel (Algeria). The soil samples were extracted by aqua regia and fluorhydric acid methods. The tomato's digestion was realised by the Hoening methods. Lead, Cadmium (Cd) and Zinc levels were measured using Atomic Absorption Spectrometry. According to the integrated pollution index, all the sites are slightly polluted by heavy metals. In tomatoes, lead (Pb) and Cd concentrations exceeded the standards set by the World Health Organization (WHO)/Food and Agriculture Organization (FAO). In general, the hazard indexes in all the studied area are less than 1, signifying that it is not risky for the people to consume these elements. However, the hazard quotients for Cd in sites 5, 6 and 7 are greater than 1, suggesting that inhabitants are experiencing a significant potential health risk especially from the consumption of tomatoes cultivated in these sites. To the best of our knowledge, this is the first study conducted on the agricultural soils of Jijel, and this could serve as a reference for future studies to monitor pollution in Jijel and its surrounding areas.

Keywords: health risk, heavy metals, pollution, soil, tomatoes

1. Introduction

The soil is a natural component of the earth consisting of water, air, living organisms, organic matter and mineral particles (Calvet, 2003; Wei and Yang, 2010). Among these mineral particles, there are heavy metals with a density higher than 5 g/cm^3. In environmental monitoring, trace elements are often called 'heavy metals'. While some are trace elements (copper [Cu], Zinc [Zn], nickel [Ni]) essential for the living beings, including humans, that become toxic at high concentrations, others are just heavy metals such as lead (Pb) and cadmium (Cd) with an unknown physiological role and pose a threat to the ecosystem and the living organism health if they exceed some threshold concentrations (Abrid *et al.*, 2013; Soubrand-Colin, 2004). Excessive use of fertilisers and pesticides, sewage irrigation, sewage sludge application and high atmospheric deposition cause a considerable increase in the concentrations of heavy metals in agricultural soils (Alghobar and Suresha, 2017; Cai *et al.*, 2009). Due to the adverse effects of heavy metals on the ecosystem and their potential threat to food safety, soil contamination by these elements is attracting a great attention from the scientific community (Lu *et al.*, 2012). The presence of these metals in agricultural soils is a growing concern because of their accumulation, transfer into soil solution and their ability to degrade the quality of groundwater and crops (Harmanescu *et al.*, 2011; Kelepertzis, 2014; Zheng *et al.*, 2013). One of the alarming facts is that crops cultivated in soils polluted with heavy metals tend to accumulate them in their edible and non-edible parts in quantities high enough to cause clinical symptoms to both animals and human beings. Moreover, the human body lacks a good mechanism for the elimination of heavy metals; thus, consumption of metal-rich plants would impose an intensified risk on human health (Alam *et al.*, 2003; Bhuiyan *et al.*, 2011).

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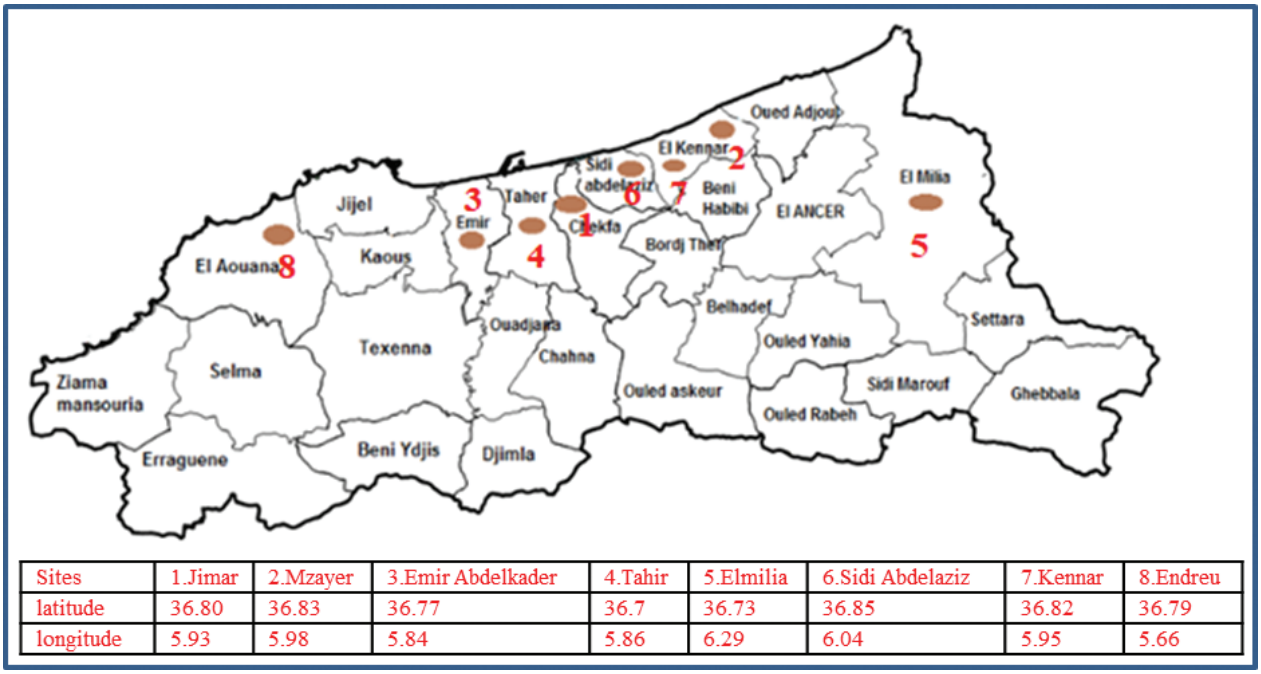
Since independence, Algeria has invested considerably in different economic sectors, notably agriculture. Unfortunately, the latter has not benefitted from any strategy to protect the environment (Kehila *et al.*, 2006; MATE, 2003). Particularly, soil pollution in Algeria is not well investigated and remains to be explored. Ignorance of the existent distribution and degree of soil pollution makes it impossible to devise adequate monitoring strategies and prevention measures against this pollution. The state of Jijel (North-East Algeria) is considered as one of the most fertile zones in North Africa due to the Mediterranean rainy climate. About 41.25% of the total area of the state (9,869,500 ha) is agricultural land, with high rate of greenhouse agriculture. In fact, in Algeria, Jijel is considered as the leader in production of several fruits and vegetables, including strawberries, peppers and tomatoes. These crops are meant for both local consumption and exportation to several parts of the world.

Due to the importance of agricultural activity in Jijel—nationally and internationally—the present study aims to assess the level of heavy metals (Pb, Cd and Zn) contaminating the agricultural soils and vegetables (Tomatoes) under greenhouses in Jijel (North Algeria) using two methods of extraction, aqua regia and fluorhydric acid, on the one hand, and to evaluate the potential health risks of Pb, Cd and Zn to local population through vegetable (tomato) consumption, on the other hand. The results of this study will provide a better understanding of the accumulation characteristics and health risks of heavy metals in a greenhouse vegetable production system.

2. Material and methods

The field study sites were located in agricultural zones where greenhouse culture is widely practised. Eight sites were selected; in each site, six composite soil samples (four samples and two controls) were collected at a depth of 10 to 15 cm using a stainless auger from the greenhouses used for tomato cultivation. The samples were stored in plastic bags. Crop samples were collected at each floor. The locations of sampling sites are shown in Figure 1.

The soil samples were dried at room temperature, crushed and ground to pass through 2 mm stainless steel sieve according to NF ISO 11464 standards. The soil pH was measured in water in accordance with ISO 10390 standards (Kouchou *et al.*, 2017). Conductivity value in 1:5 (V/V) was determined by conductimeter (Mathieu and Pieltain, 2003). Carbon content was determined with a calcimeter (Mathieu and Pieltain, 2003). The organic matter was measured by calcination at 500 °C at 5 h (ISO 10694). Cation exchange capacity (CEC) was estimated using Rhein methods (Mathieu and Pieltain, 2003). The 'pseudo-total' contents of metals were analysed using aqua regia extraction; samples were digested with a hydrochloric (37%) and nitric (70%) acids mixture in a ratio of 3:1 (V/V) (ISO 11466). Total contents were determined with



Sites	1.Jimar	2.Mzayer	3.Emir Abdelkader	4.Tahir	5.Elmilia	6.Sidi Abdelaziz	7.Kennar	8.Endreu
latitude	36.80	36.83	36.77	36.7	36.73	36.85	36.82	36.79
longitude	5.93	5.98	5.84	5.86	6.29	6.04	5.95	5.66

Figure 1. Map of the study area showing the sampling points.

hydrofluoric (40%) and perchloric (70%) acids (NF X31-147) (AFNOR, 1996). Samples of tomatoes were washed, mixed and heavy metal contents were determined according to Hoenig and Thomas (2002). Heavy metal concentrations were measured using Atomic Absorption Spectrometry flame AAS (SCIMAGZU AA—6200).

The Pollution Index (PI) was calculated according to Chon *et al.* (1998). It is defined as the average of the ratios of the metal concentrations in the soil samples to the threshold values.

$$PI = C_i / C_{0i}$$

Where: C_i is the concentration of a given *i*th element in soil samples (mg/kg); C_{0i} is its corresponding standards concentration (mg/kg).

To assess the global pollution status of the soil, the Integrated Pollution Index (IPI) can be used (Chen *et al.*, 2015; Liu *et al.*, 2013; Wei and Yang, 2010; Yu *et al.*, 2016; Zeng *et al.*, 2018). The latter is calculated according to the following equation:

$$\mathrm{IPI} = \sqrt{(p_{\mathrm{imax}}^2 + p_{\mathrm{iave}}^2)/2}$$

Where: P_{imax} is the maximum value of PI_i, and PI_{iave} is the arithmetic mean value of PI_i.

The soil quality concerning heavy metal safety were classified into five grades as follows: I—safety (IPI ≤ 0.7), II—warning (0.7 < IPI ≤ 1), III—light pollution (1 < IPI ≤ 2), IV—moderate pollution (2 < IPI ≤ 3) and V—heavy pollution (PI > 3) (Yu *et al.*, 2016; Zeng *et al.*, 2018).

The transfer coefficient (TF), also called Bioconcentration Factor (BCF), was calculated according to Bassey *et al.* (2014) by dividing the concentration of heavy metals in vegetables by the total heavy metal concentration in the soil.

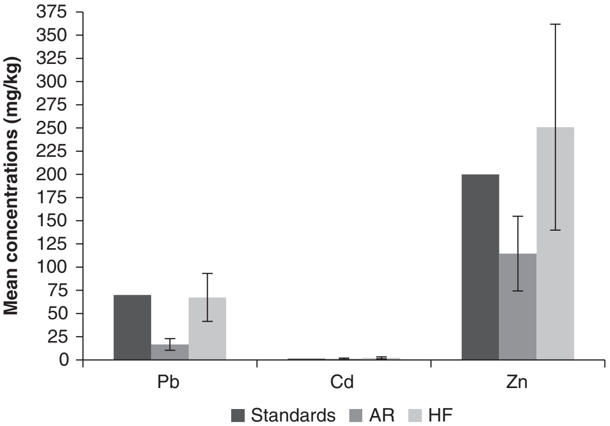


Figure 2. Mean levels of heavy metals extracted by aqua regia and HF comparing with standards

$$TF = C_{Plant} / C_{Soil}$$

Where, C_{Plant}: is the metal concentration in plant (mg/kg) and C_{Soil}: is the metal concentration in soil (mg/kg).

Human health risk assessment

Health risk assessment has been recognised as an important tool for identifying risk factors to human health associated with ingestion of heavy metals and providing evidence of risk to the decision-makers. However, estimated daily intake (EDI), hazard quotients (HQ) and hazard index (HI) were used to evaluate the toxicity of pollution and verify the risks it causes to humans (Balkhair and Ashraf, 2016; Han *et al.*, 2018; Hua *et al.*, 2017; Hub *et al.*, 2017; Shahid *et al.*, 2016; Zeng *et al.*, 2018).

The EDI of each metal through food consumption (tomatoes) and the HQ were calculated by the equations:

$$\mathrm{EDI}\ (\mathrm{mg.kg^{-1}.day^{-1}}) = C_v \times IR \times EF \times ED / BW \times AT$$

$$HQ = EDI / RfD = C_v \times IR \times EF \times ED / BW \times AT \times RfD$$

Where C_v is heavy metal concentrations in the edible parts of vegetables (mg/kg), IR represents the intake rates of selected dietary (tomatoes) considered to be 0.09 kg/person/day, EF is exposure frequency (365 days/year were considered due to daily intake of tomatoes by humans), ED is the exposure duration [76 years (Cherfi *et al.*, 2014)], BW is the average body weight [69.6 kg for adult (Atek *et al.*, 2010)], AT is the average time for non-carcinogens (365 days/year × number of exposure years, assuming 76 years in this study), RfD is the oral reference dose, which is 0.004, 0.001, 0.03 mg/kg for Pb, Cd and Zn, respectively (USEPA, 2017).

To assess the overall potential for non-carcinogenic effects posed by multiple metals, the HI was applied using the equation:

$$HI = HQ_{Pb} + HQ_{Cd} + HQ_{Zn}$$

When HQ or HI is lower than 1, there is no serious risk from exposure to heavy metals; however, if HQ or HI is equal to or higher than 1, the exposed population is likely to experience an adverse (non-carcinogenic) effect.

Statistical analysis was performed using Analysis of variance (ANOVA), and *T*-test ($p < 0.05$) was used to compare data from the different groups of soil samples distributed normally. However, if the data were non-normally distributed, non-parametric tests (Kruskal–Wallis and Mann–Whitney tests) were performed.

3. Results and discussion

The results of the statistical analysis of chemical properties in the studied area are presented in Table 1. The soil pH ranged from 6.61 to 7.8, from neutral to slightly alkaline. The average organic matter content in the plots studied was 3.57–9.51%, while CEC values of the samples have shown slight variations, with a mean value of 16.74 cmol/kg, thus indicating a greater than adequate CEC for agricultural use (Micó *et al.*, 2006). The Conductivity (CE) ranged from 82.72 to 606.50 μS/cm.

Table 1. Statistical values of physicochemical parameters in the topsoil of the study area.

	Mean	SD	Range	
			Min	Max
pH	7.15	0.31	6.61	7.80
EC (μS.cm^{-1})	288.79	162.60	82.72	606.50
OM (%)	5.53	5.17	3. 57	9.51
TC (%)	5.99	3.55	3.10	12.96
CEC (cmol.kg^{-1})	16.43	5.76	8.00	23.90
Humidity (%)	2.50	1.26	1.06	4.66

SD, standard deviation; CEC, cation exchange capacity; EC, Electric Conductivity; OM, Organic Matter; TC, Total Calcium carbonate.

pH is considered to be the main chemical parameter determining the bioavailability of heavy metals in soil (Brallier *et al.*, 1996); basically, the alkaline pH limits the passage of heavy metals from the solid phase to the solution of the soil and then to the plant (Jung and Thornton, 1996). In this study, the pH values were mostly neutral, and they are comparable to those found in the soils of the neighbouring areas such as Ghazaouet (North Algeria) (Kebir and Bouhadjera, 2011), Mamora and Casablanca (North Morocco) (Kassaoui *et al.*, 2009; Matech *et al.*, 2014).

The rate of organic matter in soil varies; generally, a rate of organic matter between 4 and 8% reflects good productivity and good mineralisation capacity (Badra and Qu, 2005). Electric Conductivity (EC) reflects soil salinity; high EC in the range of 600 μs/cm may affect salt-sensitive crops (Kassaoui *et al.*, 2009). In the present study, we registered values around 288.79 μs/cm; according to Durand (1983), these values reflect a negligible soil salinity.

The concentrations of three heavy metals (Pb, Cd and Zn) extracted by aqua regia and fluorhydric acid in this study are presented in Tables 2 and 3, respectively. The values of Pb, Cd and Zn (extracted by aqua regia) are 16.66 ± 6.26 mg/kg, 1.14 ± 0.73 mg/kg and 114.61 ± 40.27 mg/kg respectively and ranged from 9.47 ± 0.70 - 27.62 ± 2.65 mg/kg, 0.15 ± 0.05- 1.99 ± 1.8 mg/kg and 57.60 ± 40 - 173.20 ± 7.36 mg/kg. Pb, Cd and Zn (extracted by HF) contents ranged from 23.79 ± 3.12 – 111.00 ± 6.39 mg/kg, 0.38 ± 0.05 - 3.63 ± 0.07 mg/kg and 105.90 ± 1.27 - 434.10 ± 36.91 mg/kg with an average of 67.33 ± 25.80, 2.16 ± 1.06

Table 2. Heavy metal levels extracted by aqua regia of agricultural soils in study area.

	Pb (mg.kg^{-1})		Cd (mg.kg^{-1})		Zn (mg.kg^{-1})	
	Inside	Outside	Inside	Outside	Inside	Outside
1 JIM	22.89 ± 0.035*	7.50 ± 3.54	1.72 ± 0.25*	0.22 ± 0.12	118.62 ± 43.7	63.00 ± 4.24
2 MZY	27.62 ± 2.65*	13.12 ± 3.18	1.98 ± 1.86	0.19 ± 0.07	75.37 ± 2.65	75.50 ± 7.42
3 EMR	17.89 ± 0.16	12.62 ± 7.57	1.71 ± 0.39	0.80 ± 0.46	57.60 ± 40	55.50 ± 47.4
4 TAH	13.37 ± 0.88	18.25 ± 9.55	0.15 ± 0.01	0.65 ± 0.21	103.12 ± 18*	71.25 ± 3.89
5 ELM	25.62 ± 2.47	22.25 ± 3.54	1.15 ± 0.49	0.50 ± 0.49	97.00 ± 1.24	76.25 ± 7.07
6 SDA	10.62 ± 0.53*	15.25 ± 0.35	0.20 ± 0.05*	1.22 ± 0.49	106.25 ± 59.9	70.00 ± 4.95
7 KEN	13.12 ± 3.18	11.51 ± 0.00	1.40 ± 0.53	1.20 ± 0.25	164.00 ± 82	66.50 ± 0.00
8 END	9.47 ± 0.70	9.76 ± 1.7	1.45 ± 0.2	1.07 ± 0.2	173.20 ± 7.36*	98.91 ± 7.19
Mean ± SD	15.57 ± 7.02	13.78 ± 4.72	1.14 ± 0.73	0.81 ± 0.42	114.61 ± 40.27	72.11 ± 12.77
Normal content of heavy metals in soils (Bowen, 1979)	35		0.35		90	
Limit values	100^{a,b}, 60^{c}, 70^{d}, 50^{e}		2^{a}, 1^{b,c}, 1.4^{d}, 0.3^{e}		300^{a}, 200^{b,c,d}, 250^{e}	

SD, standard deviation; JIM, Jimar; MZY, Mzayer; EMR, Emir Abdelkader; TAH, Tahir; ELM, Elmilia; SDA, Sidi Abdelaziz; KEN, Kennar; END, Endreau. *T*-test was used to compare between the levels of heavy metals outside and inside sites.

*$P < 0.05$ compared to control (outside site).

^{a}WHO/FAO, ^{b}Hungarian governmental regulation number 10/2000 (2000), ^{c}Ministry of the Environment, Finland (MEF), (2007): ^{d}Canadian Council Of Ministers Of The Environment (CCME), 2007, ^{e}China National Environmental Protection Agency (CNEPA), 2006.

Table 3. Heavy metals concentrations extracted by HF of agricultural soils in study area.

HF	Pb (mg.kg^{-1})		Cd (mg.kg^{-1})		Zn (mg.kg^{-1})	
	Inside	Outside	Inside	Outside	Inside	Outside
1 JIM	111.00 ± 6.39*	69.35 ± 3.04	1.93 ± 0.1*	0.83 ± 0.25	394.80 ± 95.03	251.64 ± 125.75
2 MZY	61.23 ± 3.92	70.01 ± 1.42	2.12 ± 0.16	1.66 ± 0.08	165.00 ± 29.69	164.70 ± 3.81
3 EMR	103.33 ± 6.74	70.96 ± 19.14	1.24 ± 0.50	0.80 ± 0.27	105.90 ± 35.49	85.50 ± 36.13
4 TAH	70.55 ± 2.04	55.27 ± 6.04	0.38 ± 0.05	1.915 ± 0.47	203.70 ± 6.36	193.66 ± 57.27
5 ELM	94.50 ± 3.88*	43.94 ± 1.49	1. 97 ± 0.22	0.60 ± 0.003	213.93 ± 90.36	268.50 ± 112.42
6 SDA	42.10 ± 1.97*	101.73 ± 6.73	1.75 ± 0.08*	3.905 ± 0.13	434.10 ± 36.91	288.00 ± 155.56
7 KEN	73.83 ± 9.59*	23.83 ± 1.64	3.09 ± 0.31	3.445 ± 0.10	246.00 ± 110.30	152.70 ± 13.15
8 END	23.79 ± 3.12*	54.86 ± 1.21	3.63 ± 0.46	1.58 ± 0.05	243.30 ± 58.12	163.20 ± 6.78
Mean ± SD	67.33 ± 25.80	66.45 ± 28.89	2.16 ± 1.06	1.77 ± 1.22	250.84 ± 111	196.98 ± 68.62

SD, standard deviation; HF, hydrofluoric acid; JIM, Jimar; MZY, Mzayer; EMR, Emir Abdelkader; TAH, Tahir; ELM, Elmilia; SDA, Sidi Abdelaziz; KEN, Kennar; END, Endreau.
T test was used to compare between the levels of heavy metals of outside and inside sites
*$P < 0.05$ compared to control (outside site).

and 240.82 ± 97.84 mg/kg respectively, it is worthy to note that the concentration of Pb (sites 3, 4, 5 and 7), Cd (sites 1, 2, 5, 6, 7 and 8) and Zn (sites 1, 4, 5, 6 and 7) extracted by HF exceeds the Canadian limits. Figure 2 represent comparison between the booth extraction and threshold.

The present study showed that the concentrations of Pb and Zn extracted by aqua regia in all the studied sites do not exceed the limit of 70 mg/kg set by the Canadian Council of Ministers of the Environment (CCME). On the other hand, the majority of the sites (1, 2, 3, 7, and 8) are polluted by Cd when compared with the CCME standards. It can be seen that the values of Cd in site 5 is greater than the Chinese limits (CNEPA, 2006). Moreover, these values are comparable to those found in different regions in the world, such as China (Chen *et al.*, 2015) and Nigeria (Ahaneku and Sadiq, 2014). These results are also comparable to those reported in a study conducted on agricultural soils of Annaba, located 300 km east of Jijel (Maas *et al.*, 2010). While the concentrations of Pb and Zn extracted by aqua regia did not exceed the standard limits in all the studied sites, we noticed that they crossed the threshold limits of 70 and 200 mg/kg, respectively, in sites 1, 3, 4, 5 and 7 for Pb (CCME, 1999) and sites 1, 4, 5, 6 and 7 when extracted by hydrofluoric acid (HF). On the other hand, if we compare the Pb values in sites 1 and 2 with the lower limits set by the China National Environmental Monitoring Center (CNEMC) (50 mg/kg), we notice that these values exceed the threshold. By HF extraction, Cd also exceeded the standards limits of 1.4 mg/kg in sites 1, 2, 5, 6, 7 and 8 (CCME, 1999). However, it is found that Cd concentrations in all the sites exceed the CNEMC limits (0.3 mg/kg). In turn, Zn presented high concentrations that exceeded the threshold of 200 mg/kg in sites 1, 4, 5, 6 and 7. Site 6 represents the highest concentration (434.10 mg/kg) in the Constantine region; high concentrations of Zn up to 600 mg/kg have also been reported using the HF method (Naili *et al.*, 2016).

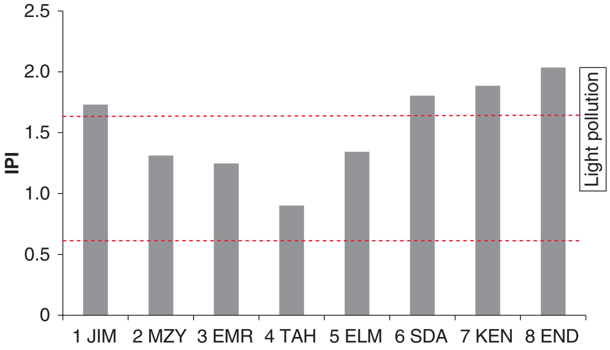


Figure 3. **IPIs of the studied area.**

Taking all together, we evaluated the PI and IPI. According to Chon *et al.* (1998), PI higher than 1 reflects soil pollution, Figure 3 presents the IPI values of the studied area.

The results of the IPI values show that all the sites are contaminated by heavy metals, with a slight pollution in all the sites, with the exception of sites 8 and 4 that present a moderate pollution and warning limit, respectively. According to the values of the IP, it is noted that pollution by Pb affects sites 3, 4, 5 and 7. As for Cd and Zn, the majority of the sites are contaminated by these elements with a PI higher than 1.

Overall, the values of Pb, Cd and Zn (extracted by HF) are close to those obtained using the same method in different regions of the world, such as Guangzhou in China

(Li *et al.*, 2009), Morocco (Matech *et al.*, 2014) and Congo (Mpundu *et al.*, 2013).

The significant difference between the yields of extraction of the two methods used in this study could be attributed to many factors, namely, the composition of the soil and the chemical nature of the extracted element (Baize, 1997). Scancar *et al.* (2000) showed a significant difference between the yields of extraction of heavy metals by aqua regia and HF. They attributed this difference to the presence of silicates or refractory aluminium, iron and manganese oxides to which the heavy metals could bound and consequently do not dissolve completely by aqua regia. In the same context, Santoro *et al.* (2017) reported in their study that when the soil was poor in silicate (SiO_2) aluminium (Al_2O_3) and iron oxides (Fe_2O_3), levels of heavy metals extracted by HF were similar to those extracted by aqua regia.

It can be seen that the concentrations of heavy metals varied from one site to another; this variation could be related to several factors such as the location of the site relatively to pollution sources, the nature of the soil and agricultural practices (Guero *et al.*, 2013). We noticed that in almost all of the studied sites, the Pb, Cd and Zn presented high concentrations in the cultivated soils, compared to the corresponding control soils (collected outside the greenhouses). This variation could be attributed to the use of fertilisers and pesticides. In fact, fertilisers are known to present one of the major sources of agricultural soil contamination by heavy metals (Benson *et al.*, 2014; Brigden *et al.*, 2002; Shahbazi *et al.*, 2017). In this regard, fertilisers used in the region of Jijel have been found to contain high amounts of heavy metals, particularly Cd (unpublished data), which is in agreement with our results discussed above.

However, we noticed that, particularly in the Sidi Abdelaziz (SDA) site, Pb and Cd presented the highest concentrations in soil outside the greenhouses (control), compared to that within the greenhouses; moreover, the outside levels exceeded the standard limits. These results could be explained by the fact that SDA agricultural soils are located alongside the highway, where the soils outside the greenhouses could be contaminated by the by-products of road traffic. In fact, it is well documented that the road traffic activities are the major source of heavy metal emission to roadside soils (Herath *et al.*, 2016; Lygren *et al.*, 1984; Parmentier and Garrec, 1994; Yan *et al.*, 2013; Zeng *et al.*, 2018; Zheng *et al.*, 2016). Furthermore, the SDA site is an agricultural village, characterised by increasing density of housing, which presents another source of pollution in the form of household wastes.

Table 4. Heavy metal levels in tomatoes.

	Lead (Pb)	Cadmium (Cd)	Zinc (Zn)
Site 1	1.21 ± 0.13	0.76 ± 0.02	7.24 ± 1.75
Site 2	1.76 ± 0.16	0.02 ± 0.00	6.00 ± 1.02
Site 3	0.46 ± 0.37	0.16 ± 0.03	8.50 ± 1.41
Site 4	1.18 ± 0.11	0.45 ± 0.18	6.88 ± 5.83
Site 5	1.25 ± 0.01	0.57 ± 0.01	6.24 ± 1.52
Site 6	ND	0.48 ± 0.04	3.75 ± 0.01
Site 7	0.73 ± 0.00	0.68 ± 0.17	9.25 ± 4.60
Site 8	ND	0.48 ± 0.00	4.75 ± 1.60
Mean	0.82 ± 0.36	0.45 ± 0.24	6.57 ± 1.81
Standard limit	0.1[a,b]	0.05[a,b]	50[c]

[a]European legislation limit values (mg/kg), [b]WHO/FAO permissible limits, [c]Indian standards.

Heavy metal levels in tomatoes, compared to standards, are presented in Table 4. Pb was detected in the majority of the samples except sites 6 and 8. Mean concentration of Pb was 0.82 ± 0.36 mg/kg and ranged from non-detectable to 1.76 mg/kg. Cd and Zn were detected in all the samples, and the mean concentrations were 0.45 ± 0.24 and 6.57 ± 1.81 mg/kg, ranging from 0.02 to 0.76 mg/kg and 3.75 to 9.25 mg/kg, respectively.

The uptake of heavy metals by plants and subsequently their accumulation along the food chain is a potential threat to animal and human health. Most of the daily human intake of heavy metals is due to the consumption of crops (Corguinha *et al.*, 2015). The risk of human exposure to heavy metals through food increases when plants are grown on soils contaminated with heavy metals (Abosede, 2017). In this regard, international guidelines such as those set by the European Union (EU), World Health Organization (WHO)/Food and Agriculture Organization (FAO) are used to set permissible levels of heavy metals in vegetables distended to human consumption. We quantified heavy metals in tomato, as it is an important component of people's diet in Jijel. The results indicated that Pb and Cd levels in tomato exceeded the permissible limits (0.1 and 0.05 mg/kg, respectively) in the samples corresponding to all the studied sites. Similar to our results, Singh and Kumar (2006) had shown that although the heavy metal load was low in the soils, it was higher in the vegetable samples. Zn, Pb and Cd in spinach and okra were in excess of the WHO limits. In the same context, Ray *et al.* (2010) recorded high levels of Cd and Pb (1.1 mg/kg) in cauliflower, cabbage and tomatoes, which were cultivated on anon-polluted soil. A research conducted in India showed that Pb and Cd levels in tomatoes exceeded the maximum permissible limit set by WHO/FAO (Mohod, 2015). The results from the present work and earlier reports (Balkhair and Ashraf, 2016; Banerjee and Gupta, 2017; Liu *et al.*, 2005; Muchuweti *et al.*, 2006) demonstrate that plants are frequently contaminated with heavy metals, thus posing a threat to public health.

Contrary to Cd and Pb, Zn levels in tomatoes in the present study were below the WHO/FAO and Indian permissible limits (60 and 50 mg/kg, respectively). The mean concentration recorded was 6.57 ± 1.81 mg/kg, ranging from 3.75 to 9.25 mg/kg. Similar studies (Ali and Al-Quahtani, 2012; Bvenura and Afolayan, 2012; Oteef and Fawy, 2015; Tasrina *et al*., 2015) showed that Zn levels in tomatoes were below the WHO/FAO and Indian permissible limits; these levels however were higher than those obtained in this study.

Efficiency of different plants in adsorbing metals is evaluated by soil to plant transfer factors of metals (TF); this is an important mean for human health risk assessment (Khan *et al*., 2009). It is an index for the bioavailability of metals to plants, reflecting thus, the risk of human dietary exposure to these elements. High transfer factors reflect a relatively poor retention in soils or a strong uptake efficiency of vegetables. Low transfer factor reflects a strong adsorption of metals to the soil colloids (Tasrina *et al*., 2015). According to Kloke (1984), a TF lower than 0.1 indicates that the plant is excluding the element from its tissues—the greater is the TF, the greater are the chances of vegetables for metal contamination (Khan *et al*., 2009). In this study, the TF of different heavy metals from soil to vegetable are presented in Table 5.

In all the studied sites (except site 3), the TF values obtained in tomatoes for Pb and Zn were within the norm levels of 0.01 to 0.1 suggested by Kloke *et al.* (1984). However, the TF values obtained for Cd were higher than 0.1. Kloke *et al.* (1984) suggested that a TF higher than 0.5 is an indicator of anthropogenic contamination of the site. Then, it has been suggested to lower the limit to 0.2 instead of 0.5, particularly in the case of leafy plants. In the present study, TF of Cd surpassed the value of 0.2 in almost all of the studied sites, indicating an anthropogenic contamination. In site 4, TF was higher than 1 (1.18). It has been suggested that TF values less than 1 signify that the plants absorb but do not accumulate heavy metals. A TF of above 1 signifies instead that plants are hyperaccumulators and indicates that the uptake of heavy metals in vegetables are higher than in the soil (Balkhair and Ashraf, 2016; Chopra and Pathak, 2015; Hellen and Othman, 2016). In this regard, high TF in site 4 could be related to the high exchangeable Cd content in this site compared to other sites. These values are in agreement with those found by Mohammed and Jimoh (2014).

Table 5. Transfer factors of heavy metal from Jijel soils into the vegetable samples.

	TF for heavy metals		
Location	**Pb**	**Cd**	**Zn**
1 JIM	0.01	0.7	0.02
2 MZY	0.03	0.01	0.04
3 EMR	0.004	0.12	0.08
4 TAH	0.02	1.18	0.03
5 ELM	0.012	0.29	0.03
6 SDA	*	0.27	0.01
7 KEN	0.01	0.22	0.03
8 END	*	0.13	0.02

TF, transfer coefficient; JIM, Jimar; MZY, Mzayer; EMR, Emir Abdelkader; TAH, Tahir; ELM, Elmilia; SDA, Sidi Abdelaziz; KEN, Kennar; END, Endreau.

*Soil/vegetable ratio was not calculated, one of the results was below detection limit.

The overall TF observation of heavy metals showed that TF values of Cd were the highest when compared to Pb and Zn values from all locations, which may be due to its high mobility relative to other elements as well as its low soil retention capacity (Gharaibeh *et al*., 2016; Mirecki *et al*., 2015).

Those metals that have a high transfer factor migrate to the edible part of the plant much more easily than do those with a low transfer factor TF. A Transfer factor of 0.1 indicates that a plant is excluding the element from its tissues, while a TF of 0.2 for vegetables indicates that a risk of metal contamination by anthropogenic activities, which in turn calls for environmental monitoring of the area (Bassey *et al*., 2014). TF values differ among locations and the plant species, and the difference in TF between locations may be related to soil nutrient management, and physical and chemical properties of the soil (Balkhair and Ashraf, 2016; Tasrina *et al*., 2015).

To assess the human health risks of each pollutant, it is essential to estimate the exposure pathways of the target organisms. Therefore, to evaluate the potential human health risks in the area, EDI, HQ and HI were calculated.

Consumers of tomatoes grown in the Jijel area may be exposed to contamination by the presence of heavy metals. Table 6 presents HQ, HI and comparison of EDI with tolerable daily intake (TDI)

Table 6. Estimated daily intake, hazard quotients and hazard index for heavy metal exposure from tomato consumption.

Heavy metals	EDI μg kg^{-1} day^{-1}	TDI[a] μg kg^{-1} day^{-1}	HQ	HI
Pb	2	3.6	0.51	0.434
Cd	0.7	1	0.76	
Zn	10.1	667	0.03	

TDI, tolerable daily intake; HI, hazard index; HQs, hazard quotients; EDI, estimated daily intake.

[a] WHO, 1993.

In this study, the means of EDI of Pb, Cd and Zn was below the TDI set by WHO/FAO. The HI values for adults decreased in the order of Zn > Pb > Cd. These results indicated that Cd was the main element contributing to the potential of health risk, followed by Pb. This is in agreement with the results reported for the greenhouse vegetable production (GVP) area in Nanjing City, Southeast China (Hu *et al.*, 2014). The mean HI of the three elements range from 0.24 to 0.54, and the maximum values are also below the safety threshold. In general, the HIs in all the studied areas are lower than 1, signifying that it is not risky for the consumers to consume the vegetables (tomatoes) cultivated in these areas. These results agree with results obtained in a study conducted in Algeria by Cherfi *et al.* (2014) on different crops, indicating that HI and HQ (Pb and Zn) are less than 1 in tomatoes, with the exception of potatoes, which had an HQ greater than 1, suggesting a potential health risk for consumers. Furthermore, the results obtained in this study are in agreement with an earlier research conducted by Han *et al.* (2018), Hua *et al.* (2017) and Hub *et al.* (2017). However, the HQ for Cd in sites 5, 6 and 7 are greater than 1, suggesting that the inhabitants are experiencing a significant potential health risk, especially from the consumption of tomatoes cultivated in these sites. Also, the fact that the HQ values of Pb and Cd are close to the threshold presents a source of uncertainty for the consumers of these products. However, the HQs of Zn are generally less than 1, signifying that it is not risky for people to consume these elements. The potential health risk of Zn is the lowest, which may be ascribed to its higher RfD. However, contrary to our study, the HI values for Pb and Cd were found to be higher than the safe threshold of 1 in previous studies (Balkhair and Ashraf, 2016; Zeng *et al.*, 2018; Zheng *et al.*, 2013).

4. Conclusion

These results show that the yield of extraction by HF was higher than the yield of Aqua regia, probably in relation to the nature of the soil and the presence of silicate. Such variables must be taken into consideration to avoid misinterpreted results in biomonitoring studies and hazard assessment. Heavy metal contents varied from one site to another. According to the IPI values, all the sites are contaminated by heavy metals with different degrees of pollution. However, we noticed that Pb, Cd and Zn presented high concentrations in the cultivated soils, compared to the corresponding control soils, reflecting an influence of agricultural activities. This must be taken into consideration to avoid any potential anthropogenic pollution in the future. In fact, Pb and Cd in tomatoes exceeded the permissible limit set by WHO/FAO and EU in all the samples; in few sites, TF was higher than 0.5, which could already indicate anthropogenic pollution. In general, the HIs in all the studied areas are less than 1, suggesting that it is not risky for people to consume the vegetables (tomatoes) cultivated in these areas. However, the HQ for Cd in sites 5, 6 and 7 are greater than 1, suggesting that inhabitants are experiencing a significant potential health risk solely from the consumption of tomatoes cultivated in these sites. Also, some attention should be paid to the lead and Cd content in the tomatoes whose HQ values approach the threshold value of 1. However, the HQs of Zn are generally less than 1, suggesting that it is not risky for the people to consume these elements.

To draw a representative image about soil and crop pollution in Algeria, further research is required to evaluate the concentration of additional heavy metals in different agricultural soils and vegetable crops in different states of the country. To the best of our knowledge, this is the first study conducted on the agricultural soils of Jijel, and this could serve as reference for future studies to monitor pollution in Jijel and the surrounding areas.

Conflict of interest

The authors declare no conflicts of interest with respect to research, authorship and/or publication of this article.

Compliance with ethical standards

This work has been conducted according to the ethical standards of scientific comity of University of Jijel.

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