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Risk assessment of heavy metals via consumption of vegetables collected from different supermarkets in La Rochelle, France

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Abstract In this study, a food survey was carried out with two purposes: (1) to investigate the levels of nickel (Ni), zinc (Zn), and copper (Cu) in various vegetables randomly collected in supermarkets of La Rochelle and (2) to assess the potential health risk for consumers by estimating the daily intake (*EDI*) and the target hazard quotient (*THQ*) for each heavy metal. The concentrations of Ni, Cu, and Zn in selected foodstuffs were detected within the following ranges: (3.2–9.6), (25.2–104.7), and (10.8–75.6) mg/kg (DW), respectively. Results showed that metals are more likely to accumulate in fruit vegetables (8.8, 63.8 and 47.8 mg/kg DW for Ni, Cu, and Zn, respectively), followed by leafy vegetables (6.5, 60.9 and 42.6 mg/kg DW for Ni, Cu, and Zn, respectively) and finally root vegetables (5.4, 40.0 and 27.3 mg/kg DW for Ni, Cu, and Zn, respectively). The levels of the metals match with those reported for similar vegetables from some other parts of the world. For all foodstuffs, *EDI* and *THQ* were below the threshold values for Cu (*EDI* 11.30; *THQ* 0.283) and Zn (*EDI* 6.86; *THQ* 0.023), while they exceeded the thresholds for Ni (*EDI* 20.71; *THQ* 1.035), indicating an obvious health risk over a life time of exposure.

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Keywords Heavy metals · Vegetables · Consumption survey · Health risks assessment · Dietary intake · Target hazard quotient (*THQ*)

Introduction

Human exposure to heavy metals through food, air, and water has increased dramatically during the past century, as a result of different anthropogenic activities such as industrialization, mining, wastewater irrigation, sludge application, used of agrochemicals, and vehicular emission (Cherfi et al. 2015; Jarup 2003; Li et al. 2014; Liao et al. 2011; Khan et al. 2014, 2015a). Thus, the heavy metal pollution has become of great concern because of food safety issues, potential health risks, and its detrimental effects on soil ecosystems (Jarup 2003; Waqas et al. 2014; Li et al. 2014). As a result, the monitoring of heavy metal contents in foods including fruits and vegetables is becoming a vital necessity. Indeed, fruits and vegetables are important components of human diet across the world both in terms of quantities consumed and nutritional value (Khillare et al. 2012; Khan et al. 2015b). They are rich sources of proteins, vitamins, minerals, and fibers and have also beneficial antioxidative effects (Hu et al. 2013; Minkina et al. 2012). Regular surveys and monitoring programs of heavy metal contents in foodstuffs have been carried out for

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decades in many countries (Becker et al. 2011; Cherfi et al. 2014, 2015; Hu et al. 2013; Khan et al. 2008; Khillare et al. 2012; Parveen et al. 2003; Pennington and Young 1990; Rodriguez-Iruretagoiena et al. 2015; Roychowdhury et al. 2003; Song et al. 2009; Tuzen et al. 2007; Wang et al. 2005; Yang et al. 2011). Among these metals harmful for humans, nickel (Ni) is known as carcinogenic metal and an environmental and occupational pollutant. Chronic exposure to Ni has been connected with lung cancer, cardiovascular disease, neurological deficits, developmental deficits in childhood, and high blood pressure (Chervona et al. 2012; De Brouwere et al. 2012). Other metals such as copper (Cu) and zinc (Zn) are essential for important biochemical and physiological functions and necessary for maintaining health throughout life (Li et al. 2014). Even though Zn is an essential requirement for a healthy body, excess Zn can be harmful; indeed, excessive absorption of this element can suppress Cu and Fe absorption. In human body, free Cu causes toxicity; indeed, as a transition element (valence of either Cu^{+} or Cu^{++}), Cu is a key participant in numerous enzyme reactions that generate reactive oxygen species (ROS), molecules such as superoxide, hydrogen peroxide, the hydroxyl radical, and others. These ROS can damage all kinds of molecules such as proteins, lipids, DNA, etc. (Brewer 2010).

Previously several studies have focused on vegetable contamination with toxic heavy metals, mostly collected from agricultural fields (Cherfi et al. 2015; Khan et al. 2008, 2010; Liao et al. 2011; Parveen et al. 2003). However, the present study provides data on Ni, Cu, and Zn contents in some selected vegetables sold in supermarkets of La Rochelle city (France). This study aimed to investigate the concentrations of Ni, Cu, and Zn in the vegetables collected from supermarkets and figure out whether they meet the agreed international requirements or not. The study also focuses on the health risks through the estimated daily intake (*EDI*) and the target hazard quotient (*THQ*) of these metals via consumption of vegetables.

The *THQ*-based risk assessment method, recognized as being valid and useful, does not provide a quantitative estimate on the health risks; it provides an indication of the risk level due to pollutant exposure (Chien et al. 2002; Wang et al. 2005). The THQ_i is a ratio of determined dose of a pollutant to a Reference Dose (R_fD): If the ratio is less than 1, there is no obvious risk from the substance over a lifetime of exposure. Conversely, if THQ_i is higher than 1, the toxicant may produce an adverse effect. The higher the THQ_i value, the higher the probability of experiencing long term carcinogenic effects (Song et al. 2009).

Materials and methods

Sampling, preparation, and analysis

Samples of 12 vegetables (zucchinis, tomato, green peppers, eggplant, salad, cabbage, fennel, potato, onion, carrot, leek, and turnip) were collected during the first 2 weeks of October 2014 from three supermarkets surrounding the city of La Rochelle in order to quantify the Zn, Cu, and Ni contents in their edible parts. Approximately, 4 kg of each vegetable was randomly collected in different boxes. These vegetable samples were washed and cleared of rotten and damaged parts, grinded and homogenized in a mixer grinder, and then oven-dried at 105 °C for 24 h to get the moisture content. Two grinded samples (3 g, each) for each food item were dry-ashed for 3 h in porcelain crucibles at 450 °C in a muffle furnace. The ash was then digested with 10 mL of concentrated nitric acid solution, filtered using Whatman filter paper No. 41 and diluted to 50 mL into a volumetric flask (Andrade Korn et al. 2008; Haswell 1991). The analysis for heavy metals of interest was performed using a flame atomic absorption spectrophotometer (Perkin Elmer, A Analyst 200). The detection limits of the apparatus were 1.5 μg/L for Zn and Cu and 6 μg/L for Ni. All measurements were made under the following conditions: air-acetylene flame, spectral bandwidth 0.5 nm, lamp current 10 mA, air flow rate 10 L/min, and acetylene flow rate 2.5 L/min. Five separate readings were made for each solution at the wavelengths (nm): 213.86 for Zn, 324.75 for Cu, and 232 for Ni. The means of these measurements were used to calculate the concentrations.

All chemical reagents used were of analytical grade. Stock solutions of diverse metals were prepared from the high purity compounds (99.9 %) purchased from Sigma-Aldrich (St. Louis, MO, USA).

Health risk assessment: estimated daily intake–target hazard quotient

In order to evaluate the health risks through consumption of vegetables by the local inhabitants, the estimated daily intake (*EDI*: μg of the selected heavy metal/day kg body wt.) and the target hazard quotient (*THQ*) were calculated using Eqs. (1)–(3).

Index *i* indicates the foodstuffs categories consumed referring to 12 vegetables in this study.

$$EDI_i = \frac{C.F_{IR}}{BWa} \tag{1}$$

$$EDI = \sum\nolimits_{i=1}^{12} EDI_i = \sum\nolimits_{i=1}^{12} \frac{C.F_{IR}}{BWa} \tag{2}$$

$$THQ_i = \frac{EFr \times ED_{tot} \times F_{IR} \times C}{R_f D \times BWa \times AT_n} \cdot 10^{-3} \tag{3}$$

In Eq. (1), EDI_i is the estimated daily intake by individual foodstuff, *C* the measured concentration of the targeted metal in individual vegetable (μg/kg FW), F_{IR} the individual food ingestion rate of the selected dietary (g FW/person day (Volatier 2000)) and *BWa* is the average body weight (77 kg for men between 18 and 65 years of age (De Saint Pol 2007)).

EDI given by Eq. (2) is the total estimated daily intake of each heavy metal through consumption of all foodstuffs.

In the Eq. (3) established by the Environmental Protection Agency (US EPA 2000a; b), *EFr* is the exposure frequency (365 days/year), ED_{tot} the exposure duration (78.4 years for men (Bellamy and Beaumel 2013)), R_fD the oral reference dose (μg heavy metal/day kg body wt.), AT_n the averaged exposure time for non-carcinogens (365 day/year for ED_{tot}) and 10^{-3} is the unit conversion factor (Chien et al. 2002; Hu et al. 2013; Song et al. 2009; Wang et al. 2005).

The total target hazard quotient (*TTHQ*) and the multi-metal (combined) target hazard quotient (*CTHQ*) were also calculated by using Eqs. (4) and (5), respectively.

$$TTHQ = \sum_{i=1}^{12} THQ_i \tag{4}$$

$$CTHQ = \sum_{j=1}^{3} THQ_j \tag{5}$$

Where *j* represents the index of heavy metal referring to Cu, Zn, and Ni.

The *TTHQ*, total *THQ*, gives an evaluation of health risks for each studied heavy metal through consumption of all concerned foodstuffs (i.e., the sum of individual metal THQ_i).

The *CTHQ*, combined *THQ*, evaluates the risks of the three studied metals together (i.e., all of Cu, Zn, and Ni) in each vegetable separately. In this study, *CTHQ* is estimated for each food item separately as the mathematical sum of individual THQ_i of all studied metals (Eq. (5)). Indeed, it has been reported that exposure to two or more pollutants may result in additive and/or interactive effects (Wang et al. 2005).

Results and discussions

Concentrations and dietary daily intake of heavy metals

Table 1 presents the concentrations of Ni, Cu, and Zn (mg/kg DW) in the selected vegetables collected from supermarkets of La Rochelle, France. The levels of these targeted heavy metals, based on sample fresh weight (mg/kg FW), in similar vegetables reported from the other parts of the world are given in Table 2. The estimated values of EDI_i and *EDI* (calculated through Eqs. (1) and (2)) through ingestion of vegetables are presented in Table 3.

In Table 1, the metal concentrations (mg/kg DW) found in the studied vegetables range between (3.24 in turnip–9.63 in tomato) for Ni, (25.23 in potato–104.68 in tomato) for Cu, and (10.83 in potato–75.60 in zucchinis) for Zn. The results in Table 1 show that the concentration order of heavy metals in the analyzed vegetable samples is Cu > Zn > Ni. The mean concentration of Cu is found to be equal to one and half times that of Zn and eight times that of Ni. Results show also that among the selected vegetables, the highest concentrations of heavy metals were noticed in fruit vegetables: respectively in tomato, salad, zucchinis, and green peppers.

Figure 1 shows the distribution of heavy metals by vegetable category: fruit vegetables, root vegetables, and leafy vegetables. For all studied heavy metals, it can be concluded from Fig. 1 that the accumulation seems to be favored in fruit vegetables, followed by leafy vegetables, and finally root vegetables. The arithmetic mean concentrations of metals (mg/kg DW) were [fruit vegetables 8.79, leafy vegetables 6.51, root vegetables 5.36] for Ni (Fig. 1a), [fruit vegetables 63.78, leafy vegetables 60.94, root vegetables 40.04] for Cu (Fig. 1b) and [fruit vegetables 47.80, leafy vegetables 42.56, root vegetables 27.35] for Zn (Fig. 1c).

Table 1 Concentration of Ni, Cu, and Zn (mg/kg dry weight) in vegetables

Foodstuffs	Moisture (%)	Concentrations of metals (mg/kg dry wt.)					
		Ni	SD[a]	Cu	SD[a]	Zn	SD[a]
Zucchinis	95.56	9.33	1.92	54.75	11.55	75.60	0.93
Green peppers	94.33	7.06	1.09	51.34	7.83	39.48	1.16
Fennel	94.51	4.95	1.65	51.06	11.01	37.44	1.75
Turnip	93.60	3.24	1.46	29.78	8.40	20.36	0.69
Salad	96.64	7.94	2.15	82.21	13.86	46.44	1.36
Carrot	88.80	5.97	0.46	30.38	4.76	35.56	0.56
Eggplant	93.70	9.12	1.28	44.36	6.78	24.41	1.27
Leek	91.53	6.43	0.97	44.08	4.47	36.56	0.79
Cabbage	91.22	6.63	0.55	49.55	8.11	43.81	0.34
Potato	83.59	6.41	0.52	25.23	7.32	10.83	0.17
Tomato	95.59	9.63	1.12	104.68	20.01	51.71	1.75
Onion	91.24	4.74	0.52	70.71	8.14	33.46	1.61

[a] Standard deviation

From a literature review, there is empirical evidence that legumes accumulate low amounts, root vegetables moderate amounts and leafy vegetables high amounts of trace metals (Alexander et al. 2006; Finster et al. 2004;

Table 2 Levels of nickel, copper, and zinc in vegetables (mg/kg fresh weight) from the French market compared with previously published results from other parts of the world

	Present study			India[a]			Algeria [b]		USA[c]		Spain[d]			Sweden [e]		Pakistan[f]			China[g]		
	Ni	Cu	Zn	Ni	Cu	Zn	Cu	Zn	Cu	Zn	Ni	Cu	Zn	Cu	Zn	Ni	Cu	Zn	Ni	Cu	Zn
Potato	1.05	4.14	1.78	0.3	0.94	4.28	0.80	3.32	0.51	2.2	–	–	–	0.77	2.77	0.83	0.1	–	0.05	1.03	3.77
Onion	0.42	6.19	2.93	0.49	1.09	4.72	1.98	4.06	0.41	1.8	–	–	–	–	–	–	0.09	0.83	–	–	–
Leek	0.54	3.73	3.10	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.07	0.51	2.72
Turnip	0.21	1.91	1.30	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Carrot	0.67	3.40	3.98	–	–	–	1.94	1.81	0.52	2.5	–	–	–	–	–	–	–	–	–	–	–
Tomato	0.42	4.62	2.28	–	–	–	5.77	5.94	0.58	1.3	1.02	13.8	55	–	–	4.15	2.31	2.45	0.03	0.41	1.05
Green peppers	0.40	2.91	2.24	–	–	–	0.33	0.81	0.74	1.4	–	–	–	–	–	–	–	–	–	–	–
Eggplant	0.57	2.80	1.54	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	0.04	0.77	1.70
Zucchinis	0.41	2.43	3.35	–	–	–	0.40	1.33	–	–	–	–	–	–	–	–	–	–	–	–	–
Fennel	0.27	2.80	2.06	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Cabbage	0.58	4.35	3.85	–	–	–	–	–	0.12	0.9			–	–	–	–	–	–	0.05	0.25	2.42
Salad	0.27	2.76	1.56	–	–	–	0.31	1.75	0.27	1.6	–	–	–	–	–	–	2.08	–	–	–	–

[a] Roychowdhury et al. 2003

[b] Cherfi et al. 2014

[c] Pennington and Young 1990

[d] Rodriguez-Iruretagoiena et al. 2015

[e] Becker et al. 2011

[f] Parveen et al. 2003

[g] Song et al. 2009

Table 3 Estimated daily intake (*EDI*$_i$, *EDI*, *Multi-metal EDI*) of heavy metals through consumption of vegetables

Categories	Foodstuffs	F_{IR}[a] (g/day person)	EDI$_i$ (μg/day kg body wt.)			Total multi-metal EDI	
			Cu	Zn	Ni		
Root vegetables	Potato	82.19	4.41	1.89	6.84	13.14	21.56
	Carrot	26.03	1.15	1.34	2.02	4.51	
	Onion	12.33	0.99	0.47	0.76	2.22	
	Leek	8.49	0.41	0.34	0.71	1.46	
	Turnip	2.74	0.07	0.05	0.12	0.23	
Fruit vegetables	Tomato	41.10	2.46	1.21	5.14	8.81	12.78
	Zucchinis	14.52	0.46	0.63	1.76	2.85	
	Green peppers	4.66	0.18	0.14	0.43	0.74	
	Eggplant	2.19	0.08	0.04	0.26	0.38	
Leafy vegetables	Salad	20.00	0.71	0.40	2.06	3.18	4.53
	Cabbage	6.30	0.36	0.31	0.54	1.21	
	Fennel	1.10	0.04	0.03	0.07	0.14	
EDI (μg/day kg body wt.)			11.30	6.86	20.71	38.88	
PTDI[b] (μg/day kg body wt.)			500	1000	5		

[a] Food ingestion rate (Volatier 2000 (INCA))

[b] Provisional tolerable daily intake (FAO/WHO 2001)

Säumel et al. 2012). However, it is good not to generalize, because the capacity for uptake and accumulation of trace metals depend on many other factors including plant species, type of contaminant, soil conditions, and characteristics such as pH, electrical conductivity, and organic carbon (Khan et al. 2014; Waqas et al. 2014). Therefore, heavy metals vary among the parts of a given plant and among varieties of the same crop species (Alexander et al. 2006; Finster et al. 2004; Säumel et al. 2012). Thus far, little is known about the involved mechanisms in the metal uptake, transfer, and redistribution within vegetables (Säumel et al. 2012). Many studies are still ongoing and will provide additional details in this field in the coming years. Furthermore, in our case, the collected products for the study purposes (three leafy vegetables, four fruit vegetables, and five root vegetables) were those intended for citizen's consumption, regardless of their origin; our market basket contained vegetables from different regions of France, Spain, Morocco, and other countries for some items (USA, Italy, etc.).

From Table 2, it can be concluded that the metal content in vegetables of this study are generally comparable for Ni and Zn with other countries: USA (Pennington and Young 1990), India (Roychowdhury et al. 2003), Algeria (Cherfi et al. 2014), Sweden (Becker et al. 2011), and Pakistan (Parveen et al. 2003). However, Ni levels are higher in Spain (Rodriguez-Iruretagoiena et al. 2015) and Pakistan (Parveen et al. 2003) for tomato while they are much lower in China (Song et al. 2009) for all commodities. Besides, Zn levels in this study have showed a divergence from other countries: lower in potato, onion, and salad than similar item analyzed by Roychowdhury et al. (2003), Cherfi et al. (2014), Becker et al. (2011), and Song et al. (2009) while they are higher in carrot, zucchinis, green pepper, and cabbage than samples analyzed by Cherfi et al. (2014) and Pennington and Young (1990).

On the other hand, the levels of Cu in the present study were higher, especially for potato and onion, than those in other countries: USA (Pennington and Young 1990), India (Roychowdhury et al. 2003), Algeria (Cherfi et al. 2014), Sweden (Becker et al. 2011), and Pakistan (Parveen et al. 2003). But Cu levels were lower in tomato than those reported by Cherfi et al. (2014) and Rodriguez-Iruretagoiena et al. (2015). It should be noticed that these authors (Rodriguez-Iruretagoiena et al. 2015) did their study in a relatively polluted area of the Basque country. Those relatively high contaminations of some vegetables by Cu might be closely related to the pollutants in irrigation water. Indeed, the Poitou-Charentes region is subjected to chronic pollution by

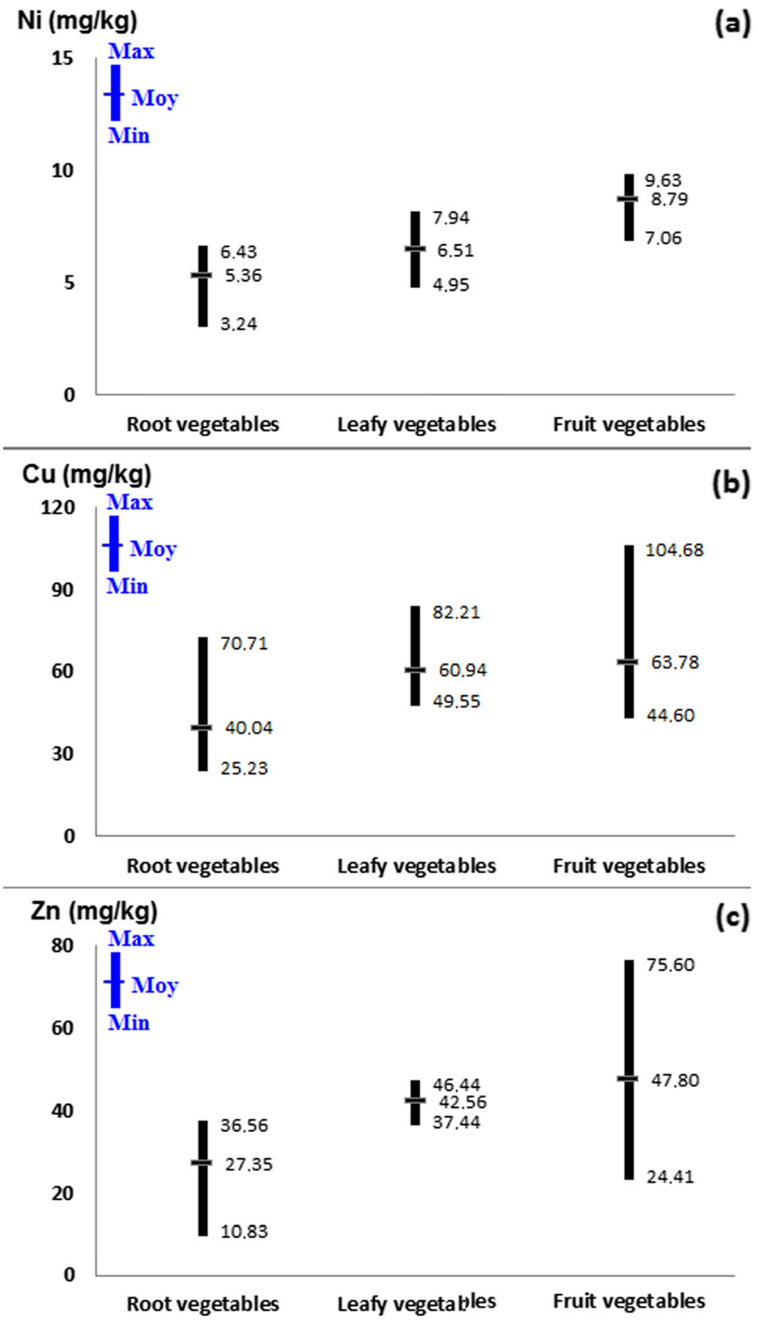

Fig. 1 Distribution of heavy metals by vegetable category: fruit vegetables, root vegetables, and leafy vegetables

chemical micro-pollutants, especially by three metals: Hg (mercury), Cu, and Zn (ORE Poitou-Charentes 2007). For Cu, three types of sources have been reported: the effluents of industrial farming, grapes processing, and replacement of tributyltin by Cu in antifouling paints on ships less than 254 m (ORE Poitou-Charentes 2007). However, considering the average consumption of the studied vegetables given in Table 3 and the relatively low toxicity of Cu, these levels do not seem to put a real threat to human health. In fact, the estimated daily intakes of the studied heavy metals caused by ingestion of vegetables and presented in Table 3 show that the contribution of vegetables to the total intake of Cu, Zn, and Ni ranged, respectively, between (0.04–4.41), (0.03–1.89) and (0.07–6.84) μg/kg body wt. The highest contribution for all intakes of metals was

Table 4 Individual, combined and total target hazard quotients (*THQ*$_i$, *CTHQ*, *TTHQ*) of heavy metals contents in vegetables

Categories	Foodstuffs	Target hazard quotient (THQ_i)			CTHQ[a]	
		Cu	Zn	Ni		
Root vegetables	Potato	0.1101	0.0063	0.3422	0.4587	0.7115
	Carrot	0.0287	0.0045	0.1009	0.1341	
	Onion	0.0248	0.0016	0.0379	0.0643	
	Leek	0.0103	0.0011	0.0354	0.0469	
	Turnip	0.0017	0.0002	0.0058	0.0076	
Fruit vegetables	Tomato	0.0614	0.0040	0.2570	0.3224	0.4654
	Zucchinis	0.0115	0.0021	0.0880	0.1016	
	Green peppers	0.0044	0.0005	0.0214	0.0262	
	Eggplant	0.0020	0.0001	0.0130	0.0151	
Leafy vegetables	Salad	0.0178	0.0013	0.1031	0.1223	0.1640
	Cabbage	0.0089	0.0010	0.0271	0.0371	
	Fennel	0.0010	0.0001	0.0035	0.0046	
TTHQ[b]		0.2826	0.0229	1.0353	1.3408	

[a] Combined (multi-metal) target hazard quotient

[b] Total target hazard quotient

noticed for potato while the lowest was noticed for fennel. Furthermore, the total dietary intake of Cu and Zn (11.30 and 6.86 μg/day kg body wt., respectively) are largely below the Provisional Tolerable Daily Intake values (*PTDI*) of FAO/WHO (500–1000 μg/day kg body wt. for Cu and Zn, respectively). On the other hand, the total dietary intake of Ni (20.71 μg/day kg body wt.) is four times the threshold PTDI of FAO/WHO (5 μg/day kg body wt.) and is far above the tolerable limit. As a consequence, adverse effects related to Ni on resident's health could be expected from consumption of vegetables. From results in Table 3, it can also be concluded that the higher contribution to the total multi-metal intake was caused by consumption of root vegetables: 21.56 from 38.88 μg/day kg body wt., which represents 55.46 %. The contribution of fruit vegetables was 32.88 % while that of leafy vegetables was 11.66 %.

Based on these insights, we concluded that although root vegetables were the less contaminated among the selected vegetables, the risks associated with their consumption are the higher because they were the most consumed vegetables. We concluded also that Ni is the major component contributing to the potential health risk via consumption of vegetables by the local inhabitants.

Health risks assessment, based on the target hazard quotient (*THQ*)

Oral reference doses (R_fD) were based on 40, 300, and 20 (μg/day kg body wt.) for Cu, Zn, and Ni, respectively (US EPA 2000a; 2003; 2007a; 2007b). THQ_i values obtained from Eq. (3) are presented in Table 4 along with total and combined (multi-metal) target hazard

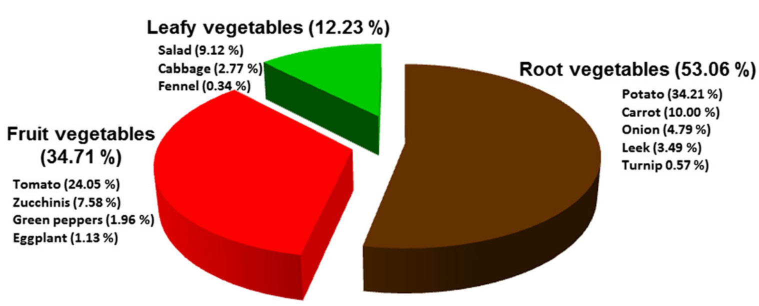

Fig. 2 Percentage of contribution of each vegetable and vegetable category in the total multi-metal THQ

quotients (*TTHQ* and *CTHQ*) calculated from Eqs. (4) and (5), respectively.

Results in Table 4 show that the THQ_i of Cu, Zn, or Ni is generally less than 1 for all vegetables, suggesting that it is not risky for the citizens to consume these elements separately. But, some attention should be paid for the Ni content in potato, tomato, and carrot because their THQ_i values are not far below the threshold value of 1. This suggests that the ingestion of Ni may pose risk for inhabitant's health via consumption of these vegetables together. Indeed, among the three selected metals, the Ni's *THQ* is largely the highest whatever the vegetable. From Table 4, we can also conclude that the average contribution of Ni in the total multi-metal *THQ* exceeds 77 %, followed by Cu (21.08 %) and Zn (1.71 %). We notice then that Ni is the most dangerous metal among those studied. It is known as a carcinogenic metal whose continuous intake has been connected with increased risk of lung cancer, cardiovascular disease, neurological deficits, developmental deficits in childhood, and high blood pressure (Chervona et al. 2012). Hence, consumption of vegetables may have a relatively high potential health risk, for the local inhabitants. On the other hand, the potential health risk of Zn is the lowest, which may be ascribed to its higher R_fD.

The results in Table 4 show also that among the 12 studied vegetables, the *CTHQ* associated with the consumption of potato, the most consumed among the studied vegetables, is the highest (0.459), followed by tomato (0.322), carrot (0.134), salad (0.122), and zucchinis (0.102). This ranking is best shown in Fig. 2 in terms of percentage of contribution for each vegetable and vegetable category in the total multi-metal *THQ*: Potato (34.21 %), tomato (24.05 %), carrot (10 %), salad (9.12 %), zucchinis (7.58 %), etc. According to Fig. 2, it can also be concluded that root vegetables take the lead with a percentage of contribution in the total multi-metal *THQ* exceeding 53 %, followed by fruit vegetables (34.71 %) and leafy vegetables (12.23 %).

Finally, the results in Table 4 show a *TTHQ* higher than 1 for Ni and a *CTHQ* which can exceed 1 by combination of more than four foodstuffs, suggesting that consumers may experience major health risks related to this metal. All the measurements highlight the necessity to take action in order to overcome some dangerous consequences consumers undergo in the short future with regard to health issues.

Conclusions

The present study generated additional useful data of heavy metal contents in vegetables sold in La Rochelle city (France) and the health risk of the local inhabitants, based on the estimated daily intake (*EDI*) and the target hazard quotient (*THQ*). The study showed that some consumed vegetables were highly contaminated with some heavy metals and exceeded their standards established by FAO/WHO. The bioaccumulation of metals seems to be favored in fruit vegetables, followed by leafy vegetables and finally root vegetables. The evaluation of *EDI* and *THQ* has indicated that health risks involving a single heavy metal are not significant for Cu and Zn. However, the total *THQ* of Ni was higher than 1 which signifies an obvious hazardous exposure over a life time to this toxic metal through consumption of vegetables. This study suggests that much more attention should be given particularly to the Ni levels to assure food safety.

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References

Alexander, P. D., Alloway, B. J., & Dourado, A. M. (2006). Genotypic variations in the accumulation of Cd, Cu, Pb and Zn exhibited by six commonly grown vegetables. *Environmental Pollution, 144*(3), 736–745.

Andrade Korn, M. D. G., da Boa Morte, E. S., Batista dos Santos, D. C. M., Castro, J. T., Barbosa, J. T. P., Teixeira, A. P., et al. (2008). Sample preparation for the determination of metals in food samples using spectroanalytical methods—a review. *Applied Spectroscopy Reviews, 43*(2), 67–92.

Becker, W., Jorhem, L., Sundström, B., & Grawé, K. P. (2011). Contents of mineral elements in Swedish market basket diets. *Journal of Food Composition and Analysis, 24*(2), 279–287.

Bellamy, V., & Beaumel, C. (2013). Bilan démographique 2012: La population croît, mais plus modérément. Insee Première n° 1429. Available from http://www.insee.fr/fr/ffc/ipweb/ip1429/ip1429.pdf.

Brewer, G. J. (2010). Copper toxicity in the general population. *Clinical Neurophysiology, 121*, 459–460.

Cherfi, A., Abdoun, S., & Gaci, O. (2014). Food survey: levels and potential health risks of chromium, lead, zinc and copper content in fruits and vegetables consumed in Algeria. *Food and Chemical Toxicology, 70*, 48–53.

Cherfi, A., Achour, M., Cherfi, M., Otmani, S., & Morsli, A. (2015). Health risk assessment of heavy metals through consumption of vegetables irrigated with reclaimed urban wastewater in Algeria. *Process Safety and Environmental Protection, 98*, 245–252.

Chervona, Y., Arita, A., & Costa, M. (2012). Carcinogenic metals and the epigenome: understanding the effect of nickel, arsenic, and chromium. *Metallomics, 4*(7), 619–627.

Chien, L. C., Hung, T. C., Choang, K. Y., Yeh, C. Y., Meng, P. J., Shieh, M. J., & Han, B. C. (2002). Daily intake of TBT, Cu, Zn, Cd and as for fishermen in Taiwan. *Science of the Total Environment, 285*, 177–85.

De Brouwere, K., Buekers, J., Cornelis, C., Schlekat, C. E., & Oller, A. R. (2012). Assessment of indirect human exposure to environmental sources of nickel: oral exposure and risk characterization for systemic effects. *Science of the Total Environment, 419*, 25–36.

De Saint Pol, T. (2007). L'obésité en France: les écarts entre catégories sociales s'accroissent. Institut national de la statistique et des études économiques. Insee Première n° 1123. Available from http://www.insee.fr/fr/ffc/ipweb/ip1123/ip1123.pdf.

Finster, M. E., Gray, K. A., & Binns, H. J. (2004). Lead levels of edibles grown in contaminated residential soils: a field survey. *Science of the Total Environment, 320*(2), 245–257.

Haswell, S.J. (1991). *Atomic absorption spectrometry: theory, design and applications*. Elsevier Science Publishers BV

Hu, J., Wu, F., Wu, S., Cao, Z., Lin, X., & Wong, M. H. (2013). Bioaccessibility, dietary exposure and human risk assessment of heavy metals from market vegetables in Hong Kong revealed with an in vitro gastrointestinal model. *Chemosphere, 91*, 455–461.

Jarup, L. (2003). Hazards of heavy metal contamination. *British Medical Bulletin, 68*, 167–182.

FAO/WHO. (2001). Joint FAO/WHO food standards programme, Codex alimentarius commission, Twenty-fourth session, Geneva, switzerland, 2–7 July 2001.

Khan, A., Khan, S., Khan, M. A., Qamar, Z., & Waqas, M. (2015a). The uptake and bioaccumulation of heavy metals by food plants, their effects on plants nutrients, and associated health risk: a review. *Environmental Science and Pollution Research, 22*(18), 13772–13799.

Khan, S., Cao, Q., Zheng, Y. M., Huang, Y. Z., & Zhu, Y. G. (2008). Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. *Environmental Pollution, 152*(3), 686–692.

Khan, S., Rehman, S., Khan, A. Z., Khan, M. A., & Shah, M. T. (2010). Soil and vegetables enrichment with heavy metals from geological sources in Gilgit, northern Pakistan. *Ecotoxicology and Environmental Safety, 73*(7), 1820–1827.

Khan, S., Reid, B. J., Li, G., & Zhu, Y. G. (2014). Application of biochar to soil reduces cancer risk via rice consumption: a case study in Miaoqian village, Longyan, China. *Environment International, 68*, 154–161.

Khan, S., Waqas, M., Ding, F., Shamshad, I., Arp, H. P. H., & Li, G. (2015b). The influence of various biochars on the bioaccessibility and bioaccumulation of PAHs and potentially toxic elements to turnips (*Brassica rapa L.*). *Journal of Hazardous Materials, 300*, 243–253.

Khillare, P. S., Jyethi, D. S., & Sarkar, S. (2012). Health risk assessment of polycyclic aromatic hydrocarbons and heavy metals via dietary intake of vegetables grown in the vicinity of thermal power plants. *Food and Chemical Toxicology, 50*, 1642–1652.

Li, Z., Ma, Z., Jan van der Kuijp, T., Yuan, Z., & Huang, L. (2014). A review of soil heavy metal pollution from mines in China: pollution and health risk assessment. *Science of the Total Environment, 468–469*, 843–853.

Liao, Y. P., Wang, Z. X., Yang, Z. H., Chai, L. Y., Chen, J. Q., & Yuan, P. F. (2011). Migration and transfer of chromium in soil-vegetable system and associated health risks in vicinity of ferro-alloy manufactory. *Transactions of Nonferrous Metals Society of China, 21*, 2520–2527.

Minkina, T. M., Motuzova, G. V., Mandzhieva, S. S., & Nazarenko, O. G. (2012). Ecological resistance of the soil–plant system to contamination by heavy metals. *Journal of Geochemical Exploration, 123*, 33–40.

ORE Poitou-Charentes. (2007). Bilan des polluants. http://www.environnement-poitou-charentes.org/Bilan-des-polluants-version.html. Accessed 26 Mars 2015.

Parveen, Z., Khuhro, M. I., & Rafiq, N. (2003). Market basket survey for lead, cadmium, copper, chromium, nickel, and zinc in fruits and vegetables. *Bulletin of Environmental Contamination and Toxicology, 71*(6), 1260–1264.

Pennington, J. A. T., & Young, B. (1990). Iron, zinc, copper, manganese, selenium, and iodine in foods from the United States total diet study. *Journal of Food Composition and Analysis, 3*(2), 166–184.

Rodriguez-Iruretagoiena, A., Trebolazabala, J., Martinez-Arkarazo, I., de Diego, A., & Madariaga, J. M. (2015). Metals and metalloids in fruits of tomatoes (*Solanum lycopersicum*) and their cultivation soils in the Basque country: concentrations and accumulation trends. *Food Chemistry, 173*, 1083–1089.

Roychowdhury, T., Tokunaga, H., & Ando, M. (2003). Survey of arsenic and other heavy metals in food composites and drinking water and estimation of dietary intake by the villagers from an arsenic-affected area of West Bengal, India. *Science of the Total Environment, 308*(1), 15–35.

Säumel, I., Kotsyuk, I., Hölscher, M., Lenkereit, C., Weber, F., & Kowarik, I. (2012). How healthy is urban horticulture in high traffic areas? Trace metal concentrations in vegetable crops from plantings within inner city neighbourhoods in Berlin, Germany. *Environmental Pollution, 165*, 124–132.

Song, B., Lei, M., Chen, T., Zheng, Y., Xie, Y., Li, X., & Gao, D. (2009). Assessing the health risk of heavy metals in vegetables to the general population in Beijing, China. *Journal of Environmental Sciences, 21*(12), 1702–1709.

Tuzen, M., Sesli, E., & Soylak, M. (2007). Trace element levels of mushroom species from east black Sea Region of Turkey. *Food Control, 18*, 806–810.

US EPA. (2000). *Risk-based concentration table*. Philadelphia PA: United States Environmental Protection Agency, Washington DC.

US EPA. (2000b). *Handbook for non-cancer health effects evaluation*. Washington (DC)7 U.S. Environmental Protection Agency.

USEPA. (2003). Integrated Risk Information System Database (IRIS). Available from http://www.epa.gov/IRIS.

USEPA. (2007a). EPA Region 3 Risk-based Concentration Table. Available from http://www.epa.gov/reg3hwmd/risk/human/rbc/RBCapr07.pdf.

USEPA. (2007b). Risk-based Concentration Table, May 2007. Available from http://www.epa.gov/reg3hwmd/risk/human/index.htm.

Volatier, J. L. (2000). Enquête nationale sur les consommations alimentaires (INCA). *Tec&Doc. CREDOC-AFSSA-DGAL*

Wang, X., Sato, T., Xing, B., & Tao, S. (2005). Health risks of heavy metals to the general public in Tianjin, China via consumption of vegetables and fish. *Science of the Total Environment, 350*, 28–37.

Waqas, M., Khan, S., Qing, H., Reid, B. J., & Chao, C. (2014). The effects of sewage sludge and sewage sludge biochar on PAHs and potentially toxic element bioaccumulation in Cucumis sativa L. *Chemosphere, 105*, 53–61.

Yang, Q. W., Xu, Y., Liu, S. J., He, J. F., & Long, F. Y. (2011). Concentration and potential health risk of heavy metals in market vegetables in Chongqing, China. *Ecotoxicology and Environmental Safety, 74*, 1664–1669.