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Contamination of market garden soils by metals (Hg, Sn, Pb) and risk for vegetable consumers of Ngaoundéré (Cameroon)

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Abstract The objective of this work is to study the contamination risks of market garden soils by metals using combustion debris (CD) as fertilizers. Therefore, two parcels of land usually used by farmer for market gardening have been rented on two sites. Soils were plowed, amended with CD and three plants species were grown. Samples of non-amended soils, amended soils and leaves of three species plants such as *Amaranthus hybridus*, *Corchorus olitorius* and *Lactuca sativa* were treated and then metals were analyzed by ICP-MS. The results obtained revealed the presence of several metallic elements such as Hg, Sn and Pb in soils with maximum concentrations of 15.03 ± 0.52, 6.16 ± 0.07 and 307.20 ± 16.30 mg/kg, respectively. The highest concentrations were found on amended soils. This means that a significant amount of metals found in amended soils would come from either direct or indirect CD. The same metals were identified in the leaves of three plants species, with concentrations which in some cases exceeded the recommended limit values. The *L. sativa* species is also one that has the most accumulation of Hg and Pb with average concentrations of 3.12 ± 1.18 and 3.1 ± 0.57 mg/kg, respectively, on the two sites. The results of the risk index also indicate the existence of a real risk of ingesting metals by consumers of vegetables grown on these soils.

Keywords Market garden soil · Contamination · Risk · Hg · Pb · Sn

Introduction

Urban agriculture is practiced all over the world for direct food and merchandising support in a total economy. Generally, in Africa and particularly in Cameroon, this activity is a source of employment, food and financial support for a portion of the population. Unfortunately, urban and sub-urban farming is still tarnished by poor agricultural practices (Allison et al. 1998; Hoffman et al. 2001; Pasquini 2006). In the town of Ngaoundéré, to amend the soil and promote the growth of vegetables, some farmers use leachates and debris from urban waste combustion originating from landfills. The farmers go to the urban landfills which are near the fields, burn the garbage and recover combustion debris (CD) to enrich the soil prior to growing the vegetables. Therefore, the farmed soils and

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vegetables harvested are directly contaminated with metallic trace elements (MTE) such as Cr, Cu, Ni, Zn, Co, Cd and Pb (Pasquini and Alexander 2004; Adjia et al. 2008, 2010; Odai et al. 2008).

The amendment of soils with municipal wastes is a common practice. Some studies conducted in France showed that the annual amount of mercury (Hg) introduced into cultivated soils for fertilization was evaluated to be between 0.05 and 0.3 g/ha/year. These studies also revealed that the contamination of market garden soils by Hg was always linked with specific and very intense cultural practices, such as using household waste products, spreading of sludge from wastewater treatment plants and using little or not treated urban wastewater (Baize et al. 2001). A model study conducted in the region of Aquitaine (France) has shown that soil amendment with sludge from wastewater treatment plants could also promote tin (Sn) contamination of the soil. This generates a potential risk of transfer of this metal in the lettuces that are grown on this soil (Lespes et al. 2009). Similarly, measurements of trace metals in soils performed on market garden soils treated with the spreading of wastewaters into the full-Pierrelaye Bessancourt (Val d'Oise) and in Montesson (Yvelines), for example, showed average levels of Hg 2.11 and 1.35 mg/kg, respectively (DDAF du Val d'Oise 1999; Baize et al. 2001). Sequential extraction studies on contaminated Hg soil samples have shown that low mercury concentrations occur in the water-soluble fractions, indicating a significant Hg availability after weathering processes (Bernaus et al. 2006). This highlights the possible availability of trace elements via contaminated amendments to the plant on these soils.

Mercury root uptake by *Lavandula stoechas* and the distribution of this metal through the plant under greenhouse conditions along three consecutive seasons was studied by Sierra et al. (2009). They noticed that Hg concentrations in the useful parts of the plant was low (0.03–0.55 mg/kg). Likewise, the essential oil, toilet water and tea obtained from these plants presented very low mercury levels, below the detection limit of the equipment used (<0.5 μg/kg). To our knowledge, no research on the contamination of soils by mercury (Hg) and tin (Sn) on agricultural soils has still been conducted in Africa's southern Sahel region.

There is, however, a strong interest on this topic and these elements, since these two metals are routinely handled daily under many forms in many household products. Further, like many African countries, and more specifically Cameroon, these countries suffer from a real problem in the management of plastic wastes. In Cameroon, for example, due to the disrespect for waste management principles, bulbs (low power) containing Hg that are widely used in households are routinely found in the garbage when they are no longer of use. Plastics that contain Sn (mostly as organotins usually used in the plastics are catalyzers) represent a 12% average of the component of urban garbage dumps in Ngaoundéré City. Both these sources are direct important inputs of Hg and Sn that are added to the amendments of the soils. It is therefore very important to follow these two metals (Hg and Sn) in the present agricultural context.

In this study, we have evaluated the potential risk of contamination of vegetable grown on soils which are consistently amended with the landfill leachates and the combustion debris. The work presented here consists in the study of metals in market garden soils in Ngaoundéré, the impact of the fertilization of soil by combustion debris (CD), the potential contamination of leaves of three plant species by metals such as Hg, Sn and Pb and their ingestion risk by consumers of these plants.

Materials and methods

Experimental sites

In this study, we have reproduced the same procedures for the preparation of farmed soils as routinely practiced in situ by farmers using CD to amend market garden soils. This cultural practice is a tradition that has been operated for the last 10 years. Two of the five sites commonly exploited for urban and suburban agriculture in the town of Ngaoundéré were chosen on the basis of previous studies (Adjia et al. 2008), which have already made a preliminary assessment of metal accumulation and transfer via this practice. Our study was conducted on the most contaminated sites based on the previous studies. They are located in Bali quarter (7°19′01,22″N; 13°35′01,42″E; altitude, 1114 m) and in Sabongari railroad station quarter (7°19′55,93″N; 13°35′44,55″E; altitude, 1096 m) close to a river (in the town of Ngaoundéré). These locations have already been identified to be seriously contaminated by Pb. The organization of these farmed lots was similar to the ones traditionally used. A typical lot of farmland per site was rented and organized into ridges of 3 m length and 0.5 m width (Figure 1 on supplementary materiel). The initial specific amendment was done once at the beginning of the study by bringing 20 L of CD from the nearby urban rubbish dumps. They were collected and spread on the ridges, and the amended ridges were irrigated for 3 weeks twice a day from a nearby river. We must specify that the soil of both sites were treated in the same way. So there is not witness site, but there are rather two types of non-amended soil samples which play witness soil role. *Amaranthus hybridus*, *Corchorus olitorius* and *Lactuca sativa* species, shown in Fig. 1, were sown on the soil amended by CD. Sowings were made during the transition
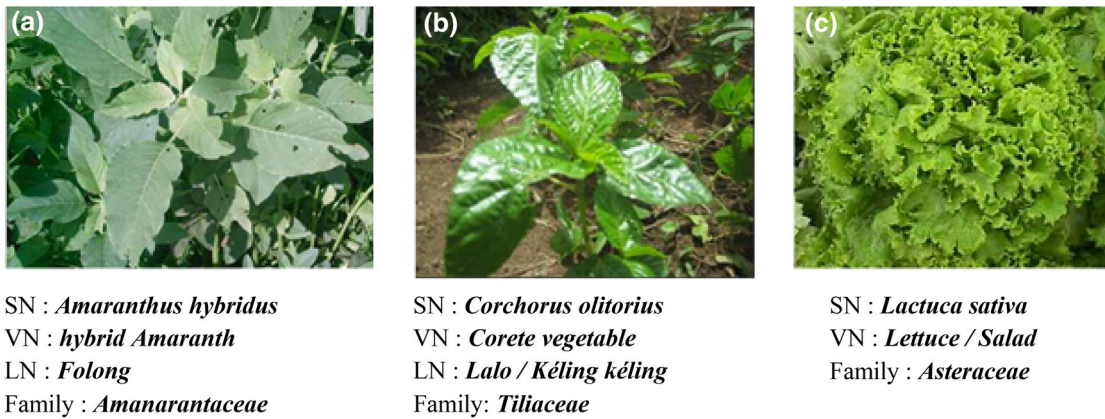
SN : *Amaranthus hybridus*
VN : *hybrid Amaranth*
LN : *Folong*
Family : *Amanarantaceae*

SN : *Corchorus olitorius*
VN : *Corete vegetable*
LN : *Lalo / Kéling kéling*
Family: *Tiliaceae*

SN : *Lactuca sativa*
VN : *Lettuce / Salad*
Family : *Asteraceae*

Scientific Name (SN); Vernacular Name (VN); Local Name (LN)

Fig. 1 Studies of the three plant species. *A. hybridus* (**a**), *C. olitorius* (**b**), *L. sativa* (**c**)

period between the dry season and the rainy season, more precisely in mid-April. Crops were watered every day with water from a nearby river.

Samples

Before spreading the combustion debris (CD) on the soils, initial reference soil samples not amended with CD were collected at 20 cm depth, which represents the maximum depth reached by the roots of the vegetable plants on sites 1 and 2, respectively (SnCDS1 and SnCDS2). Another sample of soil amended with CD was taken (SaCDS1 and SaCDS2) at a depth of 20 cm, after 3 weeks of irrigation with water from River Soum-soum which runs through the two experimental sites. (The representation of sampling points on the ridges for a representative soil sample is shown in Figure 2 of supplementary material). A further three samples of soils were taken from each site at 1 m depth (SS1 and SS2 at 1 m), to evaluate the natural geochemical background levels of Pb, Sn and Hg. The primary sources of amendment (combustion debris (CD)) were directly sampled to evaluate the influence of the amendment on the contamination of the soils by metallic elements. These CD are a mixture of soil and ash that is the product of combustion from household waste. All samples were at first characterized with general soil parameters such as acidity, conductivity, cationic exchange capacity and organic matter content.

Only the leaves of plants, which are the edible part of our three vegetable species, were harvested. They were harvested at their maturity stage, 2 months after having been planted. They were washed and rinsed with demineralized water, dried in an oven at 40 °C, crushed and conditioned at 4 °C in sachets.

Reagents and reference material

Reagents used for the extractions are of ultrapure analytical quality, like instar hydrochloric acid (JT B**A**KER), instar nitric acid (JT B**A**KER) and ultrapure water of resistivity 18.2 MΩ.cm (Millipore). CCS-6 element solution was used as the external standard for Pb, Hg and Sn. Rhodium (Rh) was used as the internal standard. Quality control of the metal analysis on the soils was performed using a soil-certified reference material (CRM) BCR-320. Another certified reference material such as channel sediment from the International Atomic Energy Agency (IAIE-433) was also used. For plants, CRM NIST-1573a tomato leaves and NIST-1570a spinach leaves were used. All of them were purchased from the European Institute for Reference Materials and Measurements (IRMM).

Physicochemical parameters of soil

Residual moisture was evaluated according to the NF ISO 11465 (AFNOR 1994a). pH and electrical conductivity were measured using the methods proposed by the standards NF ISO 10390 and 11265 (AFNOR 1994b, 1995a), respectively, and the CEC was found following the standard method AFNOR (1993). Organic matter was determined by the oxidation method using $K_2Cr_2O_7$, developed by the Centre of Expertise in Environmental Analysis of Quebec (2006).

Total element extractions and analyses

All soil samples were dried, crushed and sieved to obtain a mean grain size diameter less than 2 mm. The vegetables were powdered dry of leaves of each vegetables species that were used. In a Teflon reactor, 0.2 g of

each sample was mixed with 5 mL HCl and 2 mL HNO$_3$ Ultrex. After 12 h of pre-digestion, the mixture was heated at 90 °C for 3 h using a DigiPREP hot block platform. After cooling, the residue was diluted with MilliQ water to a final volume of 50 mL. The dilute solution was shaken for 5 min and left to rest throughout the night. Total extraction of metal elements such as Zn, Fe, Co, Mn, Ni, Ti, Cu and Cd was performed on all samples. The soil total extraction of Hg, Pb and Sn was conducted simultaneously following a combination of modified methods detailed by Zagury et al. (2009), Li et al. (2011) and Wragg and Cave (2012). Two milliliters of the extract, 100 μL of Rh (for a final concentration of 500 ppb) and 100 μL of Y (1 ppm) were used as internal standards and mixed and diluted to a total volume of 10 mL. The analyses were performed on an ICP-MS (DRC ELAN 600) (using the parameters displayed in Table 1 on supplementary materiel). The data obtained were processed using Microsoft Excel software and analyses of variance were conducted by the Stat-graphics software.

Results

The general results for global soil characterization of all samples collected are displayed in Table 1. It appears that the soils of both sites are globally acidic, despite the influence of soil amendment on the pH. We have noted indeed that the addition of CD on the soil causes a significant decrease of its acidity in site 1. This is logically explained by the basic character of CD (pH = 8.51). On the contrary to the logical phenomenon observed at site 1, we unfortunately cannot explain the slight increase of acidity induced by soil amendment in site 2. These results also show that soil amendment with CD induces a significant increase of electrical conductivity (ECond), residual

moisture (Mr), cation exchange capacity (CEC) and organic matter (OM) in the topsoil. The strong cationic charge contained in the CD (as we can see in results displayed in Tables 2 and 4) explains the increase of ECond in soils amended compared to their initial state. The frequency of irrigation by water performed on the soil after amendment with CD, as described in "Materials and methods", explains the significant increase of Mr on soils amended compared to their initial state. This increase of Mr creates favorable conditions to microbial proliferation in soil. Microorganisms, developed in the soil, transform organic soil residues including the CD to form humus. This causes a progressive increase of OM in soil and justifies the influence of soil amendment. Otherwise, the soil amendment with CD, by favoring the increase of OM through the formation of humus, results in increased complex soil adsorbent (clay–humus) and increased electronegativity. This induces therefore the increase of the adsorption sites of cations and thus an increase of the CEC of the soil.

Total extraction in soil

The concentrations of the other few metallic elements in the soil are presented in Table 2, and the concentrations of total mercury, tin and lead in the soil are also presented in Table 4.

Metallic elements

The global concentrations of some elements in soil are presented in Table 2. The iron is the most abundant metal in the soil, as much in the first 20 cm depth as in 1 m depth. It is followed by manganese (Mn) and zinc (Zn), of which their maximum concentrations are 267.67 and 175.49 mg/kg at 20 cm depth, respectively, and observed at site 2 contrary to Fe (Table 2). Other metals of less significant

Table 1 Physical parameters of soils and CD

Samples	pH	E Cond (μS/cm)	MR (%)	CEC (cmol$_c$/kg)	OM (%)
SnCDS1	5.13 ± 0.06[Aa]	2333 ± 84[Ba]	11.65 ± 1.02[Ba]	52.78 ± 2.54[Aa]	3.88 ± 0.44[Aa]
SaCDS1	5.82 ± 0.07[Ab]	7900 ± 227[Bb]	15.67 ± 0.29[Ab]	66.67 ± 4.25[Ab]	5.32 ± 0.07[Ab]
SnCDS2	6.33 ± 0.05[Ba]	900 ± 1[Aa]	4.67 ± 0.27[Aa]	86.11 ± 3.92[Ba]	5.15 ± 0.84[Ba]
SaCDS2	6.14 ± 0.01[Ba]	6900 ± 233[Ab]	20.89 ± 0.35[Bb]	91.67 ± 4.65[Bb]	6.11 ± 0.08[Bb]
S1 at 1 m depth	5.09 ± 0.14[A]	323 ± 1[A]	26.12 ± 1.25[A]	–	2.81 ± 0.42[A]
S2 at 1 m depth	5.03 ± 0.01[A]	526 ± 1[B]	35.24 ± 2.53[B]	–	2.51 ± 0.60[A]
CD (amendment)	8.51 ± 0.02	1176 ± 2	19.38 ± 0.81	–	7.58 ± 6.33

A, B; a, b: for each column, values with the same capital letter (for sites as between SnDCS1 and SnDCS2 or SaDCS1 and SaDCS2) or small letter (for amended soil or non-amended) are not significantly different ($P < 0.05$)

SnCDS1 soil non-amended with combustion debris, site 1; *SnCDS2* soil non-amended with combustion debris, site 2; *SaCDS1* soil amended with combustion debris, site 1; *SaCDS2* soil amended with combustion debris, site 2; *CD* combustion debris; *SS1 at 1 m depth* soil at 1 m depth, site 1; *SS2 at1 m depth* soil at 1 m depth, site 2; *OM* organic matter; *Mr* residual moisture; *E. Cond* electric conductivity; *CEC* cationic exchange capacity

Table 2 Other metallic elements in the soil

Samples	Most abundant metallic elements					Least abundant metallic elements		
	Cu (mg/kg)	Zn (mg/kg)	Mn (mg/kg)	Fe (mg/kg)	Ni (mg/kg)	Co (mg/kg)	Ti (mg/kg)	Cd (mg/kg)
SnCDS1	10.30 ± 0.28^{AaI}	30.60 ± 3.54^{AaI}	191.38 ± 2.01^{AaI}	$28{,}604 \pm 908^{BaII}$	24.59 ± 0.01^{AaI}	4.93 ± 0.17^{AaII}	0.26 ± 0.00^{AaII}	nd
SaCDS1	23.26 ± 1.37^{AbI}	74.61 ± 1.68^{AbI}	223.86 ± 6.71^{AbI}	$30{,}593 \pm 303^{BbII}$	25.96 ± 1.49^{AaI}	5.12 ± 0.11^{AaII}	0.46 ± 0.04^{AaII}	0.03 ± 0.02^{AbII}
SnCDS2	19.66 ± 1.23^{BaI}	164.73 ± 8.71^{BaI}	223.65 ± 12.29^{BaI}	$25{,}789 \pm 613^{AaII}$	26.30 ± 1.30^{AaI}	6.00 ± 0.21^{BaII}	0.86 ± 0.03^{BbII}	0.02 ± 0.00^{BaII}
SaCDS2	22.11 ± 0.42^{AbI}	175.49 ± 1.94^{ABI}	267.67 ± 5.45^{BbI}	$25{,}353 \pm 508^{AaII}$	25.20 ± 1.22^{AaI}	5.55 ± 0.20^{BaII}	0.42 ± 0.01^{AaII}	0.05 ± 0.00^{BbII}
SS1 at 1 m depth	8.45 ± 1.18^{AI}	28.03 ± 5.25^{AI}	206.72 ± 66.95^{BI}	$22{,}793 \pm 1008^{AII}$	19.22 ± 0.76^{AI}	4.58 ± 0.76^{AII}	0.39 ± 0.00^{BII}	nd
SS2 at 1 m depth	11.02 ± 0.55^{AI}	20.43 ± 2.06^{AI}	167.25 ± 61.32^{AI}	$26{,}033 \pm 248^{BII}$	20.07 ± 1.02^{AI}	6.86 ± 0.57^{BII}	0.27 ± 0.00^{AII}	nd
CD	24.66 ± 0.22^{I}	669.48 ± 1.93^{I}	212.49 ± 11.29^{I}	$17{,}387 \pm 1560^{II}$	18.87 ± 0.83^{I}	2.99 ± 0.23^{II}	0.38 ± 0.02^{II}	61.13 ± 7.08^{I}

A, B; a, b: for each column, values with the same capital letter (for sites as between SnDCS1 and SnDCS2 or SaDCS1 and SaDCS2) or small letter (for amended soil or non-amended) are not significantly different ($P < 0.05$)

I, II: for each line, values with the same Roman numeral are not significantly different ($P < 0.05$)

SnCDS1 Soil non-amended with combustion debris, site 1, *SnCDS2* soil non-amended with combustion debris, site 2, *SaCDS1* soil amended with combustion debris, site 1, *SaCDS2* soil amended with combustion debris, site 2, *CD* combustion debris, *SS1 at 1 m depth* soil at 1 m depth, site 1, *SS2at 1 m depth* soil at 1 m depth, site 2

abundance were also identified: nickel (Ni) with a maximum content of 26.30 mg/kg, copper (Cu, 23.26 mg/kg) and cobalt (Co, 6.86 mg/kg) of concentrations that did not significantly vary between 20 cm and 1 m depth. Titanium (Ti) and cadmium (Cd) are less abundant metals. Their maximum values are observed on site 2 with values of 0.86 and 0.05 mg/kg, respectively, at 20 cm depth.

Total mercury (Hg)

Mercury has been identified at 1 m depth soil with an average concentration on both of the sites of 0.25 ± 0.09 mg/kg. These values can be considered to be close to the natural geochemical background and are good reference values to estimate the increase brought by the amendment. The Hg concentration in the first 20 cm depth of soil ranged from 0.59 to 15.03 mg/kg. It is also observed that soil amendment with the DC multiplies by at least 23 the Hg enrichment factor of non-amended soil (Table 4). Among the least abundant metals identified into our soils, Hg represents the metal that the use of DC will have introduced into the topsoil (Tables 3 and 4).

Total tin (Sn)

On the contrary to the results observed for Hg, Sn in general does not display significant enrichments between the bottom and surface samples. Tin has been identified at 1 m depth soil with an average concentration on both sites of 3.59 ± 0.14 mg/kg. The results of ANOVA reveals that there is no significant difference at the 95.0% confidence level between the amount of Sn at 20 cm depth non-amended soil and the one at 1 m depth (P value $= 0.1120$). It was also found in the first 20 cm depth of cultivated soils at concentrations ranging from 3.59 to 6.16 mg/kg, showing little enrichments due to the influence of soil amendment on tin contamination. However, we cannot observe also significant differences between the two sites at all levels.

Total lead (Pb)

Compared to the other elements (Hg and Sn), lead shows much larger enrichments both between the sites and the depths with a significant effect in the amendments procedure. Lead has been identified at 1 m depth soil with an average concentration on both sites measured around 20 mg/kg. In the surface samples, the concentration increases significantly by a factor of 10 in the amended samples. We can clearly see that the surface-amended soils present elevated levels of Pb around 200–300 mg/kg when amended. It is remarkable that the level of Pb detected in the CD for the amendments show much lower values. This

Table 3 Enrichment factor (EF) of metallic element in the soil

Samples	Most abundant metallic elements					Least abundant metallic elements		
	Cu	Zn	Mn	Fe	Ni	Co	Ti	Cd
SnCDS1	1.058	1.263	1.024	1.172	1.252	0.860	0.781	0
SaCDS1	2.389	3.079	1.197	1.253	1.321	0.895	1.405	–
SnCDS2	2.020	6.800	1.196	1.056	1.339	1.048	2.602	–
SaCDS2	2.271	7.243	1.432	1.039	1.283	0.970	1.279	–

Table 4 Total concentrations of Hg, Pb and Sn in non-amended and amended soils

Samples	Hg		Sn		Pb	
	mg/kg	EF	mg/kg	EF	mg/kg	EF
SnDC S1	0.64 ± 0.04^{AaI}	2.6	3.59 ± 0.01^{AaI}	0.99	17.51 ± 0.10^{AaII}	0.8
SaDC S1	11.58 ± 0.42^{AbI}	47.3	5.70 ± 0.25^{AbI}	1.87	307.20 ± 16.30^{BbII}	14.0
SnDC S2	0.59 ± 0.03^{AaI}	2.4	3.85 ± 0.10^{AaI}	1.07	53.37 ± 6.00^{BaII}	2.4
SaDC S2	15.03 ± 0.53^{BbI}	61.3	6.16 ± 0.07^{BbI}	1.71	190.90 ± 17.40^{AbII}	9.1
SS1 at 1 m depth	0.31 ± 0.06^{AI}	–	3.69 ± 0.06^{AI}	–	27.59 ± 0.18^{BII}	–
SS2 at 1 m depth	0.18 ± 0.02^{AI}	–	3.50 ± 0.34^{AI}	–	16.22 ± 0.22^{AII}	–
DC	60.01 ± 3.84^{I}	–	6.14 ± 1.54^{I}	–	49.80 ± 1.50^{II}	–

A, B; a, b: for each column, values with the same capital letter (for sites as between SnDCS1 and SnDCS2 or SaDCS1 and SaDCS2) or small letter (for amended soil or non-amended) are not significantly different ($P < 0.05$)

I, II: for each line, values with the same Roman numeral are not significantly different ($P < 0.05$)

SnCDS1 soil non-amended with combustion debris, site 1, *SnCDS2* soil non-amended with combustion debris, site 2, *SaCDS1* soil amended with combustion debris, site 1, *SaCDS2* soil amended with combustion debris, site 2, *CD* combustion debris, SS1 at 1 m depth soil at 1 m depth, site 1, *SS2 at 1 m depth* = soil at 1 m depth, site 2, *EF* enrichment factor

suggests that either the CD are heterogeneous, which is a realistic hypothesis, or the sources of Pb brought to the surface soils amended are different.

Total extraction in leaves of plants

The concentrations of the metallic elements in the leaves of plants are shown in Table 5. In this table, we notice the ability of the three plant species to accumulate metallic elements such as Cu (1.6–9.3 mg/kg), Zn (13.6–53.4 mg/kg), Mn (8.6–62.9 mg/kg), Fe (89.6–2723 mg/kg), Ni (2.3–7.7 mg/kg), Ti (0.07–0.25 mg/kg), Co (0.06–0.38 mg/kg) and Cd (0.04–0.2 mg/kg) in their leaves. Fe and Zn are the most abundant metals in leaves with higher average concentrations in *L. sativa* species.

The results of the total extraction of Hg, Sn and Pb are compiled in Table 6 with varying concentrations of 0.1–3.9, 0.04–0.77 and 0.1–4.9 mg/kg, respectively. Tin is least accumulated by plants. *L. sativa* accumulates the greatest amount of Hg and Pb with 3.1 mg/kg and 3.1 mg/kg average concentrations on both sites, respectively. It is followed by *C. olitorius* and *A. hybridus* with average concentrations of 2.3 and 1.4 mg/kg for Hg, and 2.6 and 2.1 mg/kg for Pb, respectively. The results of Table 6 globally reveal a site effect on the contamination of leaves

by the three metals, because the concentration of metals is significantly higher in leaves harvested from one site compared to the other. In the present case, Hg, Pb and Sn are globally most accumulated in the leaves of the three plant species harvested on site 2, compared to the leaves of plants harvested on site 1.

Discussion

Total extraction in soil

Metallic elements

In general, according to the reports of CEMAC/FAO (2008), Ngaoundéré soils are ferruginous. This certainly justifies the high levels of iron (Fe) in soils as shown in Tables 2 and 3. The enrichment factors observed in general for all other elements are mainly significant for Zn where a maximum enrichment of 7.2 can be observed and to a lesser extent for Cu, Fe and Ni where only a factor of 1.5 on average was observed. We assume that these levels are close to the natural geochemical background levels. The concentrations of these elements in the CD are displayed in Table 2 and are in general higher by a factor of 2 compared

Table 5 Concentration of metallic elements in leaves (dry weight) of the three studied plants species and reference maximum values

Samples	Cu (mg/kg)	Zn (mg/kg)	Mn (mg/kg)	Fe (mg/kg)	Ti (mg/kg)	Co (mg/kg)	Ni (mg/kg)	Cd (mg/kg)
Site 1								
A. hybridus	1.64 ± 0.46[Aal]	34.78 ± 21.96[Aal]	14.40 ± 4.33[Aal]	89.57 ± 37,82[AaII]	0.14 ± 0.08[Aal]	0.09 ± 0.00[Aal]	4.604 ± 0.470[AbI]	0.04 ± 0.01[Aal]
C. olitorius	5.70 ± 1.05[AbI]	29.13 ± 8.53[Aal]	8.58 ± 7.59[Aal]	90.91 ± 33,35[AaII]	0.07 ± 0.01[Aal]	0.06 ± 0.02[Aal]	3.432 ± 0.311[Aal]	0.06 ± 0.01[Aal]
L. sativa	7.89 ± 1.12[AabI]	53.41 ± 2.06[Aal]	21.90 ± 7.19[Aal]	1853 ± 32[AaII]	0.14 ± 0.01[Aal]	0.06 ± 0.02[Aal]	3.818 ± 0.411[Aal]	0.09 ± 0.00[Aal]
Site 2								
A. hybridus	5.56 ± 3.70[Aal]	39.66 ± 3.63[Aal]	62.87 ± 18.63[BaI]	1876 ± 50[BaII]	0.23 ± 0.12[BaI]	0.32 ± 0.02[BaI]	7.680 ± 0.672[AbI]	0.20 ± 0.14[BaI]
C. olitorius	9.35 ± 1.18[AbI]	38.05 ± 8.06[Aal]	41.12 ± 6.41[BaI]	2723 ± 29[BaII]	0.25 ± 0.18[BaI]	0.38 ± 0.04[BaI]	5.354 ± 0.870[Aal]	0.16 ± 0.01[BaI]
L. sativa	2.25 ± 1.25[AabI]	13.65 ± 7.10[Aal]	20.54 ± 6.65[BaI]	2248 ± 30[BaII]	0.23 ± 0.12[BaI]	0.09 ± 0.02[BI]	2.273 ± 0.623[Aal]	0.05 ± 0.01[BaI]
Standard	20***	100***		450*			10***	<0.2**

* Standards FAO/OMS, 1984; ** Standards (CE) n°1881/2006; *** Standards of State Environmental Protection Administration (SEPA), 2005

to the bottom samples (1 m depth), except for Zn where high levels are observed. This shows that the use of CD has no significant effect on the soil concentration of several metals such as Cu, Mn, Fe, Ni, Co and Ti. However, it is worth mentioning that Cd levels at 1 m depths were extremely low and could not be detected under our analytical conditions in all samples. Its presence in such samples in the 50 ng/g range still means that it is brought about by the amendment procedure. Its presence is however very significant in the CD where it reaches up to 60 mg/kg, illustrating certainly the heterogeneity of the combustion debris. The concentrations obtained here are similar if not lower than the average concentrations obtained on soils worldwide (20 mg/kg for Cu, 10–300 mg/kg for Zn, 40 mg/kg for Ni and 0.06 mg/kg for Cd) (He et al. 2005). They are also similar to those obtained by Anikwe and Nwobodo (2002) and Oyedele et al. (2008) on the soil sites of municipal waste landfills operated in Nigeria for agricultural production. However, some of the results (Ni and Cu) obtained here are lower than those obtained by Oluyemi et al. (2008) on the soils of municipal waste landfills operated in Nigeria for the production of plants for food purposes. Kouassi et al. (2008) also obtained levels larger than the ones obtained here for Zn (101.2–3448.3 mg/kg), Ni (11.5–986.2 mg/g) and Cu (17.4–115.8 mg/kg) in market garden soils amended with poultry manure in Abidjan. Compared to the normal values established by the European Union in 2000, it is determined that the contents of Cu and Cd are acceptable contrary to those of Zn and Ni which are greater.

Total mercury

There is no significant difference between the amount of mercury obtained in the first 20 cm of soil and those obtained at 1 m depth, meaning that a significant amount of mercury present in these soils is more natural than from anthropogenic origin. The treatment effect on the soil due to the increased concentration of Hg in cultivated soil with the addition of CD means that a large amount of soil contamination of Hg is from the combustion products of CD, which is used for organic amendment. Indeed, in addition to the fact that about 60 mg/kg Hg was found in CD (Table 4), it was established that the mercury in the CD was from household wastes such as plastics, batteries and bulbs (low consumption). The studies carried out by ADEME (1998) revealed that 82% of Hg in the household wastes was from battery, 7% from putrescible wastes (food and green wastes), 2% from papers/cardboard and 5% from plastics. It is true that the Hg contained in household waste would volatilize during combustion, but ADEME (1998) reported that after incineration 25% (3 mg/kg) of Hg was in

Table 6 Total concentrations of Hg, Sn and Pb in leaves (dry weight) of the three studied plant species and reference maximum values

Samples	Hg (mg/kg)	Sn (mg/kg)	Pb (mg/kg)
Site 1			
A. hybridus	0.103 ± 0.001[AaII]	0.043 ± 0.004[AaI]	0.134 ± 0.005[AaII]
C. olitorius	0.624 ± 0.001[AaII]	0.043 ± 0.004[AaI]	0.283 ± 0.005[AaII]
L. sativa	2.287 ± 0.001[AaII]	0.085 ± 0.004[AaI]	3.503 ± 0.005[AaII]
Site 2			
A. hybridus	2.682 ± 0.001[BaII]	0.770 ± 0.004[BaI]	4.148 ± 0.005[BaII]
C. olitorius	3.947 ± 0.001[BaII]	0.760 ± 0.004[BaI]	4.980 ± 0.005[BaII]
L. sativa	3.953 ± 0.001[BaII]	0.202 ± 0.004[BaI]	2.697 ± 0.005[BaII]
Standards (CE) n°1881/2006)			
Vegetables	0.1	<0.2	0.1

A, B; a, b: for each column, values with the same capital letter (for sites) or small letter (for plants species) are not significantly different ($P < 0.05$); I, II: for each line, values with the same Roman numeral are not significantly different ($P < 0.05$)

the clinker, 11% (23.5 mg/kg) in the ashes and 64% (0.42 mg/Nm^3) in combustion gases.

Total tin

Table 4 shows the effect of soil amendment by CD on its contamination with tin. So, soil amendment with CD increases the content of tin in the soil, meaning that CD contains a significant amount of Sn (6.14 mg/kg) as seen in Table 4. It is worth mentioning that CD is the product of combustion of garbage from urban landfills. The dustbins are the drop points of several wastes, resulting in the presence of tin in CD. This is example of the white-iron, which is found in metal packaging such as boxes tinned food are frequently thrown in the trash cans, and old electronics device containing alloys (metal) tin deserted in landfills (Vignes et al. 2013). Besides these mentioned above, there are waste plastics containing dialkyltins and silicone rubbers containing dibutyltin dilaurate [(nC_4-H$_9$)$_2$Sn(OOCC$_{11}$H$_{23}$)$_2$] that are still found very frequently in the population bins. Furthermore, an amount of Sn present in the non-amended soil is the result of the degradation of residual plastic matter found in the soil by negligence or insouciance of man.

Total lead

It was observed that the soil at site 1 was more contaminated with Pb than that at site 2, which had already been shown by Adjia et al. (2008) at the time of assessment of the market garden soil contamination with heavy metals. The presence of this metal is more abundant in the first 20 cm of soil than at 1 m. This logically justifies the origins of more than 50% of the exogenous metal in these cultivated soils and confirm the observation made by Noubissie et al. (2011) when studying the mobility of Pb in agricultural soils of the same sites.

The CD treatment effect on the contamination of soil by Pb is in accordance with the presence of this metal in the combustion products as shown in Table 4, where approximately 50 mg/kg of Pb was identified in the CD. It is believed that a great amount of Pb come from household waste, especially from products widely used in households such as non-rechargeable batteries (for remote control) and phone batteries. In 1993, ADEME (Agence de l'Environnement et de la Maîtrise de l'Energie) determined the quantity of Pb to be 795 mg per 1 kg of dry household waste, particularly from metal Pb and glass. Regarding the origin of Pb in CD, it is established internationally that the range of Pb content in bottom ash from household waste is between 98 and 13700 mg/kg (Chandler et al. 1997). It would be logical in this context considering the poor habit of throwing battery in trash cans in addition to the origins of Pb cited above.

The Pb concentrations are higher than the average concentrations of Pb obtained in soils worldwide ranging from 10 to 150 mg/kg (He et al. 2005). They are also higher than those obtained by Anikwe and Nwobodo (2002) and Oyedele et al. (2008) on soils of municipal waste landfills sites operated in Nigeria for agricultural production. These are almost similar to those obtained by Adjia et al. (2008) on the same soil sites. But they are less than those mentioned by Kouassi et al. (2008) who obtained Pb concentrations ranging from 31.3 to 3544.6 mg/kg in market garden soil amended with poultry manure in Abidjan. However, these concentrations are above the normal value set by the European Union, which is 70 mg/kg at pH <7.

Metallic elements in the leaves of plants

Several essential (Fe, Zn, Cu, Mn, Ni and Co) and non-essential elements (Cd, Ti, Hg, Pb and Sn) were analyzed

Table 7 Bioconcentration factor (BF = leaves/soil) of metals obtained with average of metal concentration on SaDCS1 and SaDCS2

Sites	Samples	Cu	Zn	Mn	Fe	Ti	Co	Ni	Cd	Hg	Sn	Pb
Site 1	*A. hybridus*	0.07	0.47	0.06	ns	0.30	0.02	0.18	1.33	0.01	0.01	ns
	C. olitorius	0.25	0.39	0.04	ns	0.15	0.01	0.13	2.00	0.05	0.01	ns
	L. lactuca	0.34	0.72	0.10	0.06	0.30	0.01	0.15	3.00	0.20	0.01	0.01
Site 2	*A. hybridus*	0.25	0.23	0.23	0.07	0.55	0.06	0.30	4.00	0.18	0.13	0.02
	C. olitorius	0.42	0.22	0.15	0.11	0.60	0.07	0.21	3.20	0.26	0.12	0.03
	L. lactuca	0.10	0.08	0.08	0.09	0.55	0.02	0.09	1.00	0.26	0.03	0.01

ns Non-significant

in the selected plants. Bioconcentration factors (BF) of metals in the leaves of vegetables (generally <1) show that almost all metals identified in the leaves come from the soil, as shown by the results presented in Table 7. However, we note that the average BF of Cd on the two sites is ≥2. It is very curious that the Cd is metal which has no physiological use for the plant. We believe that this observed phenomenon highlights the Cd hyperaccumulating ability of these three plant species. This phenomenon also reveals the phytoavailable character of Cd in soil. This has besides been demonstrated by Noubissie et al. (2011). Indeed, by carrying out sequential extractions of Cd with distilled water, CaCl$_2$ and EDTA on these same soils, they showed that Cd was found on average to be more than 50% under the mobilizable form and hence bioavailable. This availability of Cd is often favored by acid soil pH (Kirkham 2006; Waisberg et al. 2004; Yanai et al. 2006) and soil richness in OM (Sauvé et al. 2003). The coupling of hyperaccumulating capacity of the three plant species for Cd and bioavailable character of the latter in the soil that is favored by acid pH and the significant presence of OM in the soil could explain this FB ≥2.

The presence of Hg, Pb and Sn in the leaves primarily originate from the contamination of the soil on which these three species were grown. Actually, the root barriers of these plants have certainly allowed these metals to cross, and once inside the cells of the roots migrated to the crude sap up to the leaves through translocation (Arazi et al. 1999; Greger 1999; Sunkar et al. 2000). Based on these results, the site effect observed on contamination of leaves by Hg and Sn is logical; there is indeed a positive correlation between the concentration of a metal in the soil and the concentration of the same metal in the plant grown on this soil. In other words, when there is more of Hg, Pb or Sn in the soil, the plant that grows in such a soil will accumulate more from it. Due to the fact that the soil at site 2 was more contaminated by Hg and Sn than the one at site 1 (Table 4), it is logical that plants harvested on site 2 had more accumulated metals in their leaves compared to those harvested on site 1. This means that the site effect of the Pb contamination, observed in *L. sativa* species, is logical. Hence, as seen previously, the leaves of *L. sativa* harvested

on site 1 are more contaminated by Pb than those harvested on site 2, as the soil of site 1 is more contaminated by Pb compared to the one of site 2. In this case, the site effect on Pb contamination on the leaves for site 2, observed in *A. hybridus* and *C. olitorius* species (Tables 4 and 5), was probably influenced by the high availability of Pb in soil. Indeed, the results of the study of mobility of Pb in this market garden soils conducted by Noubissie et al. (2011) showed that Pb was more available in the soil of site 2 than in site 1. On this basis, it is logical that two out of three plant species accumulate more Pb on site 2 than on site 1.

In Africa, there are still no standards for limits of heavy metal concentrations in foodstuffs intended for human consumption. However, the maximum permitted levels for foodstuffs intended for human consumption have been defined by FAO and WHO (2001). Our results show that Fe concentrations in the leaves of the three vegetables are significantly above the limit value permitted. The obtained results (Table 5) confirm the ability of these three plant species to accumulate Pb in their leaves. The concentrations obtained in the current work are lower than those obtained by Adjia et al. (2010), Because they had analyzed lead in the leaves of vegetables grown on the same sites and under the same conditions and mostly reported concentrations greater than 4 mg/kg. The ability of these three plant species to accumulate mercury, tin and lead highlights the "phyto-available" character of these metals. This ability also shows that these plants can be used in a phytoremediation context of contaminated soils by mercury, tin and lead. With respect to food safety, except for Sn, the concentrations of Pb and Hg found in these edible leaves were higher than the maximum concentration set at 0.1 mg/kg according to regulation no. 1881/2006 by the Committee of European Communities.

Risk of exposure to the total metals in plants

These three plant species were chosen in this study because their leaves are highly consumed in Africa and Cameroon, in particular, through the recent years. The results of daily exposure doses (DED) of total Hg, Sn and Pb by ingestion of the leaves of the three vegetable plants are shown in

Table 8 Daily exposure doses of metals by ingestion of the leaves of three vegetables species (ng/g bw/day)

Pollutants	Vegetable species consumed			Limit	
Metals	*A. hybridus*	*C. olitorius*	*L. sativa*	ADI (man)	LD50 (rata)
Hg	2.00 ± 0.40	2.29 ± 0.33	4.46 ± 0.45	0.71[a]/2[b]	10–40[c]
Sn	0.58 ± 0.12	0.40 ± 0.06	0.21 ± 0.02	2000[d]	–
Pb	3.06 ± 0.61	2.63 ± 0.38	4.43 ± 0.44	3.57[b]	<500[e]

ADI acceptable daily intake (ng/g bw/day) from Ricoux and Gasztowtt (2005)[a]; [b]INERIS (2009); JECFA (1982)[c]

LD50 median lethal dose (μg/g bw) from INRS (1997)[d]; power for a word of application (2004) Material safety data sheet. March, 25th 2004. Rev. 12[e]

Table 9 Risk index for consumer of the leaves of three vegetables species (DED/ADI)

Pollutants	Vegetables species consumed			Limit
Metals	*A. hybridus*	*C. olitorius*	*L. sativa*	ADI (man)
Hg	1	1.15	2.23	2[a]
Sn	ns	ns	ns	2000[b]
Pb	0.86	0.74	1.24	3.57[a]

ns non-significant

[a] INERIS (2009)

[b] JECFA (1982)

Table 8 for the total metal content. We have not found references on Sn toxicity. In Table 9, we present the risk index (RI) for the three plants consumed. Table 8 shows that Hg risk index (RI) is greater than or equal to 1 (RI[Hg] ≥ 1), while it is globally less than or equal to 1 for Pb (RI[Pb] ≤ 1). This suggests that a real risk for the consumer exists in terms of Hg exposure, and that the risk is lower for Pb exposure. With an index of RI >1, there is a significant risk for the consumer of *L. sativa* species for the exposure of any of the metals considered. The lead DED by ingestion of *L. sativa* obtained in this work was significantly lower than that obtained by Adjia et al. (2010) and varied between 8 and 19 ng/g bw/day on the same cultural sites (Ngaoundéré—Cameroon). With Hg and Pb DED widely below LD50, we can generally conclude that the observed risk related with the accumulation of Hg or Pb in the leaves does not present a direct threat to the consumer if eaten randomly.

Conclusion

Ngaoundere soils naturally contain some metallic elements that are derived from the mother rock, like most of the world's soils. With the exception of tin, the presence of the other two metals studied in these agricultural soils is mainly the result of a strong exogenous influence. This strong exogenous influence of Pb and Hg is due in the present work to the use of amendments which are made from substrates rich in these metals. Indeed, when this practice is repeated over several years, the excess metal which has not been undercover (vertical or horizontal) in the soil remains on the surface where they accumulate and make themselves available for plants cultivated there. This obviously causes a health risk for consumers of plants grown on these soils. If the results of this work show a risk index predominantly <1 for Pb and Sn, we also note that there is a real risk of exposure to mercury for consumers in the town of Ngaoundere. So the cropping practice of amending the soil with CD is very dangerous for consumers of products grown there. Given the results of this work, gardeners of Ngaoundere and all those who exploited this crop practices elsewhere should change it immediately. For example, if they do not have money to buy fertilizer, they may use a biological practice such as co-culture, associating leguminous plants (which enrich the soil in nitrogen and phosphorus) with the species of plants they want to grow. Despite the disappointing findings observed in this study on the health plan, the available character of these metals for plants allows us to think that, the gardeners who operate these contaminated soils must already thinking about their cleaning up. For example on this view, we could through phytoremediation technique use varieties (edible or not) of these three plant species to assess their effectiveness in the remediation of contaminated agricultural soils by metals, because through this work, these three plant species have shown their ability to accumulate metals including Hg, Sn and Pb. If not, the danger may be more serious in the future. When these metals are stored in the soil, they can mutate into organometallic forms under the influence of certain physical and chemical factors, similar to the nature of the waste constituting the substrate used to make the CD could contain organometallic compounds that are the most toxic forms of the metals. It is therefore important for the result of this work to study organometallic compounds of these three metals in the market garden soils.

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Compliance with ethical standards

Conflict of interest We are the authors of the manuscript entitled "Contamination of market garden soils by metals (Hg, Sn, Pb) and risk for vegetable consumers of Ngaoundéré (Cameroon)", and we declare that no conflict of interest exists.

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