Assessment of the levels of cadmium and lead in soil and vegetable samples from selected dumpsites in the Kumasi Metropolis of Ghana

Peter Twumasi*, Marina A. Tandoh, Makafui A. Borbi, Adigun R. Ajoke, Emmanuel Owusu-Tenkorang, Roseline Okoro and Rexford M. Dumevi

Department of Biochemistry and Biotechnology, College of Science, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana.

Received 15 February, 2016; Accepted 1 April, 2016

Many dumpsites in the urban communities in Ghana are used for cultivation of crops, especially vegetables. However, these dumpsites may serve as potential sources of soil heavy metals that could enter the food chain mainly through cultivated food crops with serious consequences on human health. This study investigated the levels of two heavy metals, lead (Pb) and cadmium (Cd), in the soil and tissues of vegetables grown on such dumpsites. Soil and tissue (lettuce, cabbage, spring onion, tomato and the leaves of Xanthosoma sagitifolium) samples were collected from ten locations with two of these locations used as control. The samples were acid-digested and the metal concentrations determined using atomic absorption spectrometry. Pb and Cd contents of soils from all the eight dumpsites and one of the control locations were above the guidelines recommended by FAO and WHO. The highest Cd level in the soil (13.6 mg kg⁻¹ of Cd) was found at Aketego dumpsite and the highest soil Pb (36.1 mg kg⁻¹ Pb) was recorded at Meduma dumpsite. The leafy vegetables, cabbage, lettuce and X. sagitifolium (locally called 'kontomire') recorded relatively higher amounts of Pb and Cd in the edible parts. Further studies are required to determine how much of the daily diet these vegetables contribute to the total diets of the population and special attention to calculating the overall daily doses of Cd and Pb to pregnant mothers and children <5 years of age is thus warranted.

Key words: Cadmium (Cd), dumpsites, heavy metals, lead (Pb), health risk, vegetables.

INTRODUCTION

The swelling of urban populations in Ghana reflects rural-urban migration (Black et al., 2011) and high population growth rate in the cities (Grimm et al., 2008), impacting severely on the availability of arable lands in the urban centres (Akudugu et al., 2012). In Ghana, construction of roads and buildings has been blamed for losses of otherwise agricultural lands (Kugelman, 2012).

The agricultural prowess of some of the urban residents, the high cost of living (Aguda, 2009) and the very high unemployment (Barrios et al., 2006) in most Ghanaian
cities have over the years promoted backyard farming (Appeaning, 2010). Many families living in these urban communities now depend upon backyard farming (Zeza and Tasciotti, 2010), which includes both livestock and crops (Cofie et al., 2005). Urban agriculture contributes about 20% of Ghana’s national agricultural production (Appeaning, 2010), although about 45% of the backyard farm produce is for subsistence. Unfortunately, due to the pressure on land use in the urban settings, farmers tend to use unapproved land sites for agricultural purposes (Egyir and Beinquo, 2009), including abandoned refuse dump sites (Agyarko et al., 2010). Dumpsites are commonly used for direct cultivation of vegetables and also as good source of compost to support mainland agricultural activities (Owusu-Sekeyere et al., 2013; Mwingyine, 2008).

Despite the value of plant-derived compost and soils in refuse dumps (Stefanowicz et al., 2012), many hazardous chemicals are found in these dump sites, and consequently, they pose threat to human health when they become introduced to the food chain (Bagumire et al., 2009). Because metals and other inorganic chemicals are not degraded, they can occur in elevated concentrations with time due to accumulation. Some plastics, papers, batteries, electric bulbs and bottle caps, are known to contain heavy metals (Amusan et al., 2005; Akpoveta et al., 2011; Ideriah et al., 2010; Adelekan and Alawode, 2011; Kolo et al., 2014).

There is an increasing trend of industrial waste dumping in most African countries, an activity mainly motivated by importation of cheap used manufacturing products from industrial countries by these poor African nations. This menace has been identified in Ghana with serious consequences on human health and the environment (Agyarko et al., 2010). Arsenic, cadmium, lead, chromium, nickel, cobalt and mercury are of great concern primarily because of their potential to harm soil organisms, plants, animals and ultimately humans (Singh et al., 2011a). Heavy metals are taken up alongside essential elements such as zinc, magnesium and calcium by plants (Volpe et al., 2009). Research has shown that vegetables grown on refuse dump sites have tendency to absorb these heavy metals, hyper-accumulate them in their tissues with detrimental effects on plant growth and yield (Amusan et al., 2005; Nabulo et al., 2008) as well as the health of consumers (Reilly, 2008; FAO/WHO, 2011; Järup, 2003).

This study investigated the levels of two very important heavy metals, that is, Cd and Pb, in refuse dumpsites in the Kumasi Metropolis of Ghana and their accumulations in edible tissues of vegetables commonly cultivated on these farms.

MATERIALS AND METHODS

Study site

The study sampled eight refuse dump sites (Emina, Oduom, Tafo, Aketego, Ayigya Zongo, Meduma, South Suntreso and Boadi) and two control farms (KNUST Horticulture and Ayeduase Agric Farm). The refuse dumps sites at Aketego, Ayigya Zongo and Boadi were relatively young as they were established about 5 years prior to the study according to the Assemblyman of the area - vegetable cultivation began in the last three years (Personal Interview). The study sites were grouped into two clusters (that is, Clusters 1 and 2) based on the types of vegetables cultivated. Cluster 1 consisted of six refuse dump sites and a control farm that were used for cultivation of lettuce, cabbage and spring onion. Cluster 2 was for cultivation of tomatoes and leaves of cocoyam or Xanthosoma sagittifolium (locally called ‘kontomire’). All the vegetables from these farms are regularly sold on the local markets in the Kwame Nkrumah University of Science and Technology (KNUST), surrounding communities and across Kumasi Metropolis.

Sample collection

Soil (1.0-5.0 inches deep) and vegetables (that is, lettuce, cabbage, spring onion stems, tomato fruits and cocoyam leaves) were each collected into separate transparent, air-tight polyethylene bags and kept on ice before transporting to the laboratory for the metal analysis. Five samples each of soil and the vegetables were collected for the heavy metals determination. The period of the research spanned over three years from 2012 to 2014.

Pre-digestion of samples for heavy metal analysis

In the laboratory, samples were washed using metal-free distilled water to remove soil particles on the surfaces. The washed vegetable samples were air-dried for 2 h at room temperature and immediately followed by oven-drying at 105°C for 5 h. The dried samples were ground into fine powder using mortar and pestle and stored in clean containers. Soil samples, on the other hand, were air/sun-dried for 2 h, homogenized and sieved using 2 mm nylon mesh. The powdered soil and vegetable samples were stored in clean containers.

Acid digestion of vegetable and soil samples

Triplicates of 2 g dry weight (dw) of finely ground soil and vegetable samples were each placed in 300 ml volumetric flask, 20 ml of HNO3-HClO4 di-acid mixture (9:4) added, and the contents well mixed by swirling thoroughly. The flasks with contents were slowly heated on hot plate in a fume chamber to 85°C and then rapidly to 150°C. Heating continued until the production of red NO2 fumes ceased and the contents’ volume reduced to 3 to 4 ml of colorless or yellowish slurry. The flasks and their contents were cooled and the volumes adjusted with deionized water before instrumental analysis. The resulting solutions were kept in vials and stored at 4°C pending Atomic Absorption Spectrometry (AAS) determination. The machine was calibrated using standard samples and also blanks to ensure reliability of the test results.

Statistical analysis

The experiment was a complete randomized block design with triplications of soil and vegetable samples. Each replicate consisted of three different samples of each vegetable and soil collected randomly from the farm sites. Analysis of variance (ANOVA) for the effects of refuse dump sites and vegetable type on the mean values of Pb and Cd was tested at 5% probability level using the statistical package Graphpad Prism 4 (Graphpad Software, Inc.).
RESULTS

Vegetable cultivation

Farmers cultivated specific types of vegetables per refuse dump site. Some of these vegetables thrived better on these refuse dump sites. Seven of the ten farms were involved in cultivation of lettuce, cabbage and spring onions. Tomato and cocoyam leaves or *kontomire* were produced on the remaining three farms.

Cadmium levels in agricultural soil and vegetable samples

Only three of the ten farms recorded soil Cd level below the FAO/EPAs’s recommended maximum limit of 0.270 mg kg⁻¹ (270 ppb) for food crop production (Tables 1 and 2) (Odai et al., 2008). The Cd content of soil from the Aketego dumpsite (13.623 mg kg⁻¹), one of the newly-established dumps, was exceptionally high (P<0.0001), nearly 13.5 times the maximum allowable limit for Cd in agricultural soil.

Lettuce, cabbage and spring onion from all the seven farms including the control contained <0.2 mg kg⁻¹, the FAO/WHO guideline (Table 1 and Figure 1). Although Cd in cabbage from four dump sites (Emina, Oduom, Tafo and Ayigya Zongo) was within the safe levels, the concentrations were either double than the amounts found in same vegetables from the other sites (that is, Aketego and Boadi dumpsites, and Agric Farm C1 (Table 1). On the other hand, Cd levels in *kontomire* and tomato leaves from Meduma and South Suntresos dumpsites, and KNUST Horticulture C2 (control) were above the maximum allowable limits (p<0.0001) (Table 2 and Figure 2).

The concentration ranged from 0.340 mg kg⁻¹ (340 ppb) to 0.790 mg kg⁻¹ (790 ppb), amounts in excesses of 300 to 830% the maximum level allowed for consumption (Table 2). The Cd content of the tomato fruit from the dump sites was 7 to 11 times higher than the maximum allowable level in fruiting vegetable. These measurements were comparable (P>0.05) to the control farm (KNUST Horticulture C1) which was found to be 10 times higher than the maximum level of Cd in fruiting vegetables. Clearly the concentration of Cd in the tomato leaves and fruits were different. The leaves contained about twice the amount of Cd in the fruits (Table 2).

Pb levels in agricultural soil and vegetable samples

In all the farms including the controls, significantly high amounts of Pb was recorded in the soil above the maximum limit of 0.420 mg kg⁻¹ (420 ppb) allowed for agriculture (Tables 3 and 4). The highest concentration of Pb (33.870 mg kg⁻¹) was detected in the soil from South Suntresos dump site and the lowest from the control farm (1.13 mg kg⁻¹). Lettuce, cabbage and spring onion grown on old dump sites (that is, Emina, Oduom and Tafo) had Pb levels exceeding the maximum level of Pb in leafy vegetables allowed for consumption (0.3 mg kg⁻¹). These were about 1.5 to 17 times greater than the maximum level of Pb in leafy vegetables allowed for human consumption. However, Pb levels of lettuce, cabbage and spring onion samples from newly established dump sites (Aketego, Ayigya Zongo and Boadi) and the control (Agric Farm C1) all fell below the maximum level allowed for consumption (Table 3 and Figure 3). *Kontomire* and tomato leaf samples from Meduma, South Suntresos and KNUST Horticulture C2 (control) all recorded Pb levels 8 to 13.5 times higher than the maximum level of Pb in leafy vegetables allowed for consumption.

### Table 1. Mean concentration (mg kg⁻¹) of Cd in soil and vegetable samples from refuse dump sites in the Kumasi Metropolis of Ghana.

<table>
<thead>
<tr>
<th>Soil/Vegetable</th>
<th>Emina</th>
<th>Oduom</th>
<th>Tafo</th>
<th>Aketego*</th>
<th>Ayigya Zongo*</th>
<th>Boadi*</th>
<th>Control Farm (C1)</th>
<th>FAO/WHOb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soil</td>
<td>0.453(±0.04)</td>
<td>0.243(±0.07)</td>
<td>0.543(±0.04)</td>
<td>13.623(±0.31)</td>
<td>0.363(±0.03)</td>
<td>0.280(±0.02)</td>
<td>0.253(±0.02)</td>
<td>0.27</td>
</tr>
<tr>
<td>Lettuce</td>
<td>0.017(±0.01)</td>
<td>0.067(±0.01)</td>
<td>0.017(±0.01)</td>
<td>0.037(±0.00)</td>
<td>0.003(±0.00)</td>
<td>0.053(±0.00)</td>
<td>0.010(±0.01)</td>
<td>0.2</td>
</tr>
<tr>
<td>Cabbage</td>
<td>0.200(±0.02)</td>
<td>0.117(±0.02)</td>
<td>0.107(±0.02)</td>
<td>0.073(±0.00)</td>
<td>0.143(±0.03)</td>
<td>0.013(±0.00)</td>
<td>0.023(±0.00)</td>
<td>0.05 (brassica)</td>
</tr>
<tr>
<td>Spring onion</td>
<td>0.060(±0.02)</td>
<td>0.033(±0.02)</td>
<td>0.033(±0.01)</td>
<td>0.033(±0.00)</td>
<td>0.060(±0.01)</td>
<td>0.030(±0.01)</td>
<td>0.020(±0.01)</td>
<td>0.2</td>
</tr>
</tbody>
</table>

*C1 is the control farm±SEM. *Maximum levels (MLs) of Cd in leafy and fruiting vegetables according FAO/WHO (CODEXAlimentarius Committee, 2011) are 0.2 mg kg⁻¹ and 0.05 mg kg⁻¹, respectively. *Relatively younger refuse dump sites. Averaged Cd concentration in soil is 0.270mgkg⁻¹ (WHO/FAO: from ftp://ftp.fao.org/codex/meetings/CCCF/cccf5/cf05_INF.pdf).

---

Table 1. Mean concentration (mg kg⁻¹) of Cd in soil and vegetable samples from refuse dump sites in the Kumasi Metropolis of Ghana.
Table 2. Mean concentrations (mg kg\(^{-1}\)) of Cd in soil and vegetable samples from refuse dump sites in the Kumasi Metropolis of Ghana.

<table>
<thead>
<tr>
<th>Soil/Vegetable samples</th>
<th>Refuse dump sites</th>
<th>FAO/WHO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Meduma</td>
<td>South Suntreso</td>
</tr>
<tr>
<td>Soil</td>
<td>0.317(±0.02)</td>
<td>0.617(±0.05)</td>
</tr>
<tr>
<td>Tomato leaf</td>
<td>0.873(±0.02)</td>
<td>0.753(±0.04)</td>
</tr>
<tr>
<td>Kontomire leaf</td>
<td>0.507(±0.03)</td>
<td>0.563(±0.02)</td>
</tr>
<tr>
<td>Tomato fruit</td>
<td>0.527(±0.01)</td>
<td>0.340(±0.01)</td>
</tr>
</tbody>
</table>

C2 is the control farm. Maximum levels (MLs) of Cd in leafy and fruiting vegetables according FAO/WHO (CODEX Alimentarius Committee, 2011) are 0.2 and 0.05 mg kg\(^{-1}\), respectively. Average Cd concentration in soil is 0.270 mg kg\(^{-1}\)±SEM.

Table 3. Mean concentration (mg kg\(^{-1}\)) of Pb in soil and vegetable samples from refuse dump sites in the Kumasi Metropolis of Ghana.

<table>
<thead>
<tr>
<th>Soil/Vegetable samples</th>
<th>Refuse dump sites</th>
<th>FAO/WHO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Emina</td>
<td>Oduom</td>
</tr>
<tr>
<td>Soil</td>
<td>6.040(±0.04)</td>
<td>10.650(±0.27)</td>
</tr>
<tr>
<td>Lettuce</td>
<td>3.533(±0.24)</td>
<td>7.197(±0.38)</td>
</tr>
<tr>
<td>Cabbage</td>
<td>2.433(±0.22)</td>
<td>0.700(±0.05)</td>
</tr>
<tr>
<td>Spring onion</td>
<td>0.780(±0.07)</td>
<td>0.590(±0.04)</td>
</tr>
</tbody>
</table>

C1 is the control farm. Maximum levels (MLs) of Pb in leafy and fruiting vegetables according FAO/WHO (CODEX Alimentarius Committee, 2011) are 0.3 and 0.1 mg kg\(^{-1}\) respectively. FAO Averaged Cd concentration in soil is 0.420mg kg\(^{-1}\) ±SEM.

Table 4. Mean concentrations (mg kg\(^{-1}\)) of Pb in soil and vegetable samples from refuse dump sites in the Kumasi Metropolis of Ghana.

<table>
<thead>
<tr>
<th>Soil/Vegetable samples</th>
<th>Refuse dump sites</th>
<th>FAO/WHO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Meduma</td>
<td>South Suntreso</td>
</tr>
<tr>
<td>Soil</td>
<td>36.120(±1.163)</td>
<td>33.870(±1.452)</td>
</tr>
<tr>
<td>Tomato leaf</td>
<td>4.503(±0.332)</td>
<td>4.517(±0.283)</td>
</tr>
<tr>
<td>Kontomire leaf</td>
<td>3.483(±0.070)</td>
<td>5.583(±0.321)</td>
</tr>
<tr>
<td>Tomato fruit</td>
<td>1.883(±0.082)</td>
<td>2.213(±0.160)</td>
</tr>
</tbody>
</table>

C2 is the control farm. Maximum levels (MLs) of Pb in leafy and fruiting vegetables according FAO/WHO (CODEX Alimentarius Committee, 2011) are 0.2 mg kg\(^{-1}\) and 0.05 mg kg\(^{-1}\) respectively. Averaged Cd concentration in soil is 0.420mg kg\(^{-1}\) ±SEM.

consumption. The tomato fruit on the other hand recorded about 38 to 74 times the maximum amount of Pb in fruits allowed for consumption (0.05 mg kg\(^{-1}\)) (Table 4 and Figure 4). Similar to the trends of Cd accumulation, the tomato leaves contained about twice the amount of Pb in the...
tomato fruits (Table 4).

DISCUSSION

In the past decade, Ghana’s vegetable production, local consumption and export have recorded between 20 to 25% growth annually (Sangare et al., 2012). This phenomenon is partly attributed to the increasing promotion of vegetable consumption among the local population due to its health benefits and the ever increasing horticultural export industry to countries such as those in the European Union (Thow and Priyadarshi, 2013). Despite the health benefits associated with the consumption of vegetables, a number of diseases have been linked to it (Reiss et al., 2012). Prominent among these vegetable associated diseases are those caused by microbial pathogens and toxigenic chemical compounds (Singh et al., 2011b). Dumpsite agricultural lands contribute to vegetable production especially in the urban communities where arable lands are scarce (Dubbeling and De Zeeuw, 2011). However, these dumpsite used for agriculture are important sources of dangerous heavy metals derived from components of industrial products (Fuge, 2013; Wuana and Okieimen, 2011) and thus agricultural activities on such lands provide entry route for heavy metals in the food chain.

All the eight dumpsites studied were used in the cultivation of various vegetables for human consumption. The levels of Cd and Pb, the two most injurious heavy metals to human health (Islam et al., 2007), in soil samples from the dumpsites were found to be higher than dosages approved safe for agricultural activities especially crop farming (Figures 1 to 4).

The waste composition of the dumpsites as well as the state of decomposition (or the age of the established dumpsites) affected the levels of heavy metals in the soil. Aketego dumpsite recorded 13.62 mg kg\(^{-1}\) Cd in soil, a level several times over the amounts allowed for agricultural purpose as recommended by the FAO and EPA (Bay, 2013). Pb levels in the soils from all the dumpsites were exceptionally high above the levels approved for agricultural activity (Figures 3 and 4). The results clearly show that the soils at the dumpsites may not be suitable for agricultural activity especially for growing crops such as those assessed in this study. Alternatively, these fields could be decontaminated of heavy metals through programmes involving plants with high bioremediative power for Pb and Cd.

Industrial waste is the most important sources of Pb exposure. Pb element and its compounds are components of a number of industrial products such as batteries, cables, pigments, plumbing and ceramics, gasoline, tobacco, solder and steel products, food packaging glassware and pesticides (Tangahu et al., 2011). Naturally, Cd is rare in the environment but its levels in agricultural fields have risen as a result of human activities (Cranor, 2011). Like Pb, the major source of Cd is the disposal of industrial waste containing Cd. Cd is used in the production of batteries, coatings, stabilizers and pigments (Faroon et al., 2012). These industrial wastes are dumped in Kumasi and other cities in Ghana by importers of used electronic products and machine parts from the Unites States, Europe and Asia (Schaller et al., 2009).

The leafy vegetables (lettuce, cabbage, spring onion,
Vegetables grown on refuse dump sites

Figure 2. Cadmium concentration (mg/kg$^{-1}$) in soil and vegetable samples from six refuse dump sites in the Kumasi Metropolis and a control site (KNUST Horticulture) (N = 9, bar = standard error of mean).

Vegetables grown on refuse dump sites

Figure 3. Pb concentration (mg/kg$^{-1}$) in soil and vegetable samples from refuse six dump sites and a control site (Agric Farm at Ayeduase) in the Kumasi Metropolis of Ghana (N = 9, bar = standard error of mean).

kontomire and tomato leaves) growing in soils with high levels of Cd and Pb accumulated the heavy metals at levels far above the maximum concentrations approved by the WHO and FAO for human consumption. Naturally, leafy vegetables accumulate higher levels of Cd and Pb than other crops (Mench, 1998). The uptake of Cd by plants and fungi from the soil is directly related to the concentration in the soil (WHO/FAO, 2011; Ling-Zhi et al., 2011). Generally, Cd uptake by plants from the soil is enhanced by low soil pH (Rajkumar et al., 2012), a
feature commonly associated with industrial acidic waste. It was observed in this study that vegetables from the
KNUST Horticulture, one of the control agricultural farms, produced vegetables with Pb and Cd levels significantly
above the recommended concentrations for human consumption (Tables 2 and 4). Although, no
anthropological data on the site is available, discussions with indigenes living around the university indicate that
some parts of the KNUST agricultural fields were once human settlement and thus could contribute to the high
metal contents of the soil (personal interviews).
Consumption of these vegetables cultivated on these
dumpsites in the Kumasi Metropolis contaminated with
high levels of Pb and Cd poses serious health risk to
consumers. Pb is considered a classical chronic or
cumulative poison. In humans, Pb can result in a wide
range of effects including hematological effects,
neurological and behavioral effects, renal effects,
cardiovascular effects, and effects on the reproductive
system (WHO/FAO, 2011; Abdullahi, 2013; Patra et al.,
2011; Sun et al., 2014). Children are more vulnerable to
effects of Pb than adults. Pb has been shown to be
associated with impaired neurobehavioral functioning in children and this remains the most critical effect of Pb
(Grandjean and Landrigan, 2014).
Based on dose-response analyses, it has been
established that provisional tolerable weekly intake
(PTWI) of 25000 mgkg$^{-1}$ body weight is associated with a
decrease of at least 3 IQ points in children and an
increase in systolic blood pressure of approximately 3
mmHg (0.4 kPa) in adults (WHO/FAO, 2011; Solenkova
et al., 2014; Tellez-Plaza et al., 2012). The rate of
absorption of Pb into the bloodstream ranges from 3 to
80% (WHO/FAO, 2011; Marsh and Bailey, 2013; Sabath
and Robles-Osorio, 2012). Younger persons and fasting
individuals tend to absorb higher amounts of Pb ingested
or inhaled. Half-life of Pb in blood and soft tissues is
about 28 to 36 days (Farzin et al., 2008). Cd excretion is
very slow and thus having very long half-life (decades)
one in the body (WHO/FAO, 2011; Edwards et al.,
2009). The major health risk associated with Cd exposure
in renal failure due to high accumulation in the kidney
exposure (Kazi et al., 2008).
In conclusion, soils from the eight dumpsites sampled
from the Kumasi Metropolis recorded very high
concentrations of Pb and Cd and these metals were
accumulated in the tissues of vegetables cultivated on
them. The levels of Cd and Pb in the vegetables
exceeded the maximum levels (ML) approved by WHO
for human consumption. Therefore, consumption of
vegetables cultivated on these dumpsites poses serious
health risk to the consumers.

Conflict of Interests

The authors have not declared any conflict of interests.

ACKNOWLEDGEMENTS

The authors acknowledge the Soil Research Station of
the CSIR-Ghana in Kumasi and the Faculty of Agriculture in the Kwame Nkrumah University of Science and Technology (KNUST), Kumasi, for their assistance in the atomic absorption spectrometric analysis of heavy metals. They also thank the help offered by the farmers and the Assemblymen of areas where the dumpsites were located for their permission to work and also for supply of vegetable samples for the analysis. They appreciate the technical support received from technical instructors from the laboratories of Biochemistry and Biotechnology of KNUST in Kumasi. The research was funded through Research Funds of Dr. Peter Twumasi.

REFERENCES


Singh R, Gautam N, Mishra A, Gupta R (2011a). Heavy metals and...