Lead exposure from aluminum cookware in Cameroon

Jeffrey D. Weidenhamer a, Peter A. Kobunski a, Gilbert Kuepouo b, Rebecca W. Corbin a, Perry Gottesfeld c,⁎

a Department of Chemistry, Geology & Physics, 401 College Ave., Ashland University, Ashland, OH 44805, USA
b Research and Education Centre for Development (CREPD), Yaounde, Cameroon
c Occupational Knowledge International, San Francisco, CA, USA

HIGHLIGHTS

• Cookware is manufactured in Cameroon from scrap aluminum including car parts.
• Twenty-nine cookware samples were evaluated for their potential to leach lead.
• Boiling extractions to simulate the effects of cooking released significant lead.
• Potential lead exposures per serving are estimated as high as 260 μg.
• Artisanal aluminum cookware may be a major contributor to lead poisoning.

GRAPHICAL ABSTRACT

ABSTRACT

Blood lead levels have decreased following the removal of lead from gasoline in most of the world. However, numerous recent studies provide evidence that elevated blood lead levels persist in many low and middle-income countries around the world at much higher prevalence than in the more developed countries. One potential source of lead exposure that has not been widely investigated is the leaching of lead from artisanal aluminum cookware, which is commonly used in the developing world. Twenty-nine samples of aluminum cookware and utensils manufactured by local artisans in Cameroon were collected and analyzed for their potential to release lead during cooking. Source materials for this cookware included scrap metal such as engine parts, radiators, cans, and construction materials. The lead content of this cookware is relatively low (<1000 ppm by X-ray fluorescence), however significant amounts of lead, as well as aluminum and cadmium were released from many of the samples using dilute acetic acid extractions at boiling and ambient temperatures. Potential exposures to lead per serving were estimated to be as high as 260 μg, indicating that such cookware can pose a serious health hazard. We conclude that lead, aluminum and cadmium can migrate from this aluminum cookware during cooking and enter food at levels exceeding recommended public health guidelines. Our results support the need to regulate lead content of materials used to manufacture these pots. Artisanal aluminum cookware may be a major contributor to lead poisoning throughout the developing world. Testing of aluminum cookware in other developing countries is warranted.

© 2014 Elsevier B.V. All rights reserved.

Abbreviations: GFAAS, graphite furnace atomic absorption spectrometry; ICP, inductively coupled plasma spectrometry; MADL, maximum allowable dose level; PTIIL, provisional tolerable total intake level; PTWI, provisional tolerable weekly intake; SEM, scanning electron microscopy; XRF, X-ray fluorescence.

⁎ Corresponding author. Tel.: +1 415 221 8900; fax: +1 415 221 8903. E-mail address: pgottesfeld@okinternational.org (P. Gottesfeld).
1. Introduction

In recent decades lead has been removed from gasoline in all but a few countries resulting in a substantial decrease in blood lead levels. However, evidence suggests that elevated blood lead levels persist in many low and middle-income countries around the world at much higher prevalence than in the more developed countries. Recent research from India (Kalra et al., 2013), China (Li et al., 2014; Xie et al., 2013), South Africa (Naicker et al., 2013), the Democratic Republic of Congo (Tuakuila et al., 2013), Thailand (Swaddiwudhipong et al., 2013) and Saudi Arabia (El-Desoky et al., 2013) indicate that blood lead levels among young children are many times higher than levels reported in the U.S. or EU despite progress from earlier decades.

Lead accounts for 674,000 deaths annually (Lim et al., 2012) and is a risk factor for attention-related behaviors, learning disabilities, and criminal behavior. Lead toxicity in low- and middle-income countries costs $977 billion annually (Attina and Trasande, 2013). There is no identified threshold for toxicity (Lanphear et al., 2005; Wigle and Lanphear, 2005). The continued prevalence of high blood lead levels in children after the phase-out of leaded gasoline is therefore of great concern from a public health perspective.

Evidence suggests that it may be increasingly difficult to identify a single source of exposure to account for the current exposure scenario and to provide relief to millions of exposed individuals. To address this next phase of the global lead poisoning epidemic it is necessary to identify significant exposure sources that contribute to the patterns observed in recent studies from emerging market countries so that appropriate responses can be developed. One known source repeatedly cited has been contaminated food (Bergkvist et al., 2010; Xie et al., 2013), but little direct evidence is available to explain how lead has entered the food supply. Although some reports link lead contamination of soil to these exposures, many other sources may also play a contributing role (Bergkvist et al., 2010; Ritchie and Gerstenberger, 2013).

In addition to some more prominent exposure sources including industrial emissions, lead in paint, and contaminated water supplies, many consumer products have been shown to contain lead in significant concentrations (Weidenhamer and Clement, 2007; Weidenhamer, 2009). While conducting education and outreach programs in Cameroon around the need to control lead levels in new paints (Gottesfeld et al., 2013), several participants raised concerns about the lead content of locally available cooking pots.

An investigation was subsequently launched to determine whether lead may be entering the manufacturing process of locally made aluminum pots. In visiting a number of artisanal cookware manufacturing facilities, we discovered that the raw material used is scrap metal including used car and motor bike engine parts, waste aluminum and computer components (Fig. 1). These inexpensive pots, which are not anodized, are widely used throughout Cameroon. Lead introduced through aluminum cookware may be a significant source of contamination relative to other environmental exposures. Recently other investigators have noted a possible link between the use of inexpensive aluminum cookware and lead absorption (Bergkvist et al., 2010; Swaddiwudhipong et al., 2013).

This investigation was designed to help fill the gap in explaining the potential lead exposures from aluminum cookware. We investigated the levels of lead and other heavy metals that may be released into foods cooked in finished pots obtained from a number of local sources in four cities in Cameroon where artisanal cookware production is prevalent. The results of our investigation are outlined along with recommendations to regulate the lead content of aluminum cookware.

2. Methodology

2.1. Sample collection

Cookware samples were collected from four towns located in four of the ten regions in Cameroon where cookware production is concentrated: Kumba (4 samples), Douala (6 samples), Ngaoundéré (5 samples) and Yaoundé (11 samples). Sample collection information including potential scrap metal sources and used or new condition of the pot (Figs. 1 and 2) was documented, and all samples were numbered as collected. In addition to the cookware samples, three cooking ladles (Fig. 2, inset) were also collected. To facilitate international shipment of this large number of samples to the analytical laboratory, the ladles and one pot from each location with its lid were sent intact, while 4 × 4 cm pieces were cut from all remaining samples and sent for analysis.

![Fig. 1. Some source materials used in the manufacture of artisanal aluminum cookware in Cameroon. (A) Computer hard drive cover; (B) mixed scrap including computer parts; (C) motor bike engine part; and (D) molten scrap being poured into molds for pots.](image-url)
2.2. X-ray fluorescence (XRF) analysis

XRF screening of samples was conducted using a Niton XL3t GOLDD XRF spectrometer mounted in a test stand (Thermo Fisher Scientific, Billerica, MA). Samples were analyzed in “Test all” mode for 60 s.

2.3. Leaching tests

There are no standard methods for the measurement of leaching of heavy metals from aluminum cookware. However, investigations of the concentration of aluminum in foods prepared from aluminum cookware have typically used either (a) dilute acidic liquids such as tomato juice, citric acid, or acetic acid (4% vol/vol) to simulate slightly acidic foods (Al Zubaidy et al., 2011; Inoue et al., 1988; Mohammad et al., 2011), or (b) soup broths to examine the effect of added salt and various meat and vegetable extracts (Al Juhaiman, 2010, 2012; Al Zubaidy et al., 2011; Mohammad et al., 2011). In addition, a 24 h extraction with dilute acetic acid at ambient temperature is typically used to evaluate the release of lead and cadmium from ceramic cookware (Cheng, 2010). Here, we used dilute acetic acid in two separate extractions: (a) a 24 h ambient temperature extraction; and (b) a 2 h boiling extraction. Both extractions are relevant to the actual use of this cookware. Cooking times using this heavy aluminum cookware in Cameroon can range from 0.5 to 4 h depending on food type and energy supply (cooking gas or wood), and food is often stored in the pots for 1–2 days with reheating before additional meals.

Volumetric flasks and all other glassware used in these experiments were rigorously acid-washed with concentrated nitric acid prior to analysis. All cookware was washed with soap and water prior to undertaking these experiments. The cookware pieces (4 × 4 cm) were cut into smaller 2 × 2 cm pieces to allow multiple measurements with different leaching times on individual pieces. The dimensions of each piece (length, width, and thickness) were determined with a micrometer so that the surface area of each sample was known. Each cookware piece was accurately weighed prior to the experiment. Intact cookware items were analyzed as received.

Samples were leached with 4% acetic acid (vol/vol) for 24 h at ambient temperature, or for 2 h in boiling solutions. (a) The 24 h measurements were made by placing individual pieces in sealed plastic cups with 25 mL of 4% acetic acid. For whole cookware pieces, the pot was filled with 4% acetic acid, and ladies were placed in 4% acetic acid in a sealed beaker. (b) For the 2 h boiling measurements, 25 mL portions of 4% acetic acid were put into 100 mL beakers which were then covered with watch glasses. Ladies were placed in large beakers covered with a watch glass and a volume of acetic acid sufficient to immerse the ladle to a depth of approximately 10 cm. For whole cookware, vinegar was boiled in the pot, and ladles were placed in 4% acetic acid in large beakers for this extraction. Except for the whole pots where solutions were brought to a boil within the pot, all solutions were brought to a gentle boil and the samples were added. The samples were then boiled for 2 h, during which time 4% acetic acid was added as needed to maintain solution volumes. After cooling, solutions were transferred with multiple rinses to volumetric flasks to bring to constant volume. Following the leaching experiments, all samples were dried and weighed.

2.4. GFAAS and ICP methods

Leaching solutions were analyzed by both graphite furnace atomic absorption spectrometry (GFAAS) and by inductively coupled plasma spectrometry (ICP). GFAAS measurements were carried out on a SpectrAA 220 FS instrument (Varian, Walnut Creek, CA, USA). Lead absorbance was measured at 283.3 nm using background correction and standard addition. Char and atomization temperatures were set at 800 °C and 1800 °C respectively. A 1% wt/vol ammonium dihydrogen phosphate matrix modifier solution was used. Fresh working standard (30 μg/L) was prepared daily from certified reference material lead standard for AA (Fluka Analytical, St. Louis, MO, USA; product no. 16595) solution (1000 μg Pb/mL). All standards were prepared in 4% acetic acid. Blanks and spiked samples were used to verify analytical performance.

ICP measurements were carried out by the Service Testing and Research Laboratory (Ohio Agricultural Research and Development Center — Ohio State University) on a Prodigy Dual View ICP spectrometer (Teledyne Leeman Labs, Hudson, NH, USA). Blanks and spiked samples were used to verify analytical performance.

2.5. Scanning electron microscopy (SEM)

Scanning electron micrographs of cookware samples at 350, 1000, and 2500× magnification were obtained using a Hitachi S-3500N Scanning Electron Microscope under high vacuum by the Molecular and Cellular Imaging Center (Ohio Agricultural Research and Development Center, Ohio State University). For SEM imaging, three 2 × 2 cm samples were cut from the rim of three intact pots. Because the outside of the pots had not been exposed to cooking liquids, this provided a point of comparison to determine the effect of boiling on the pot surface. SEM images were obtained before and after boiling in 4% acetic acid for 2 h.

3. Results

3.1. X-ray fluorescence analysis

Cookware samples contained as much as 850 ppm lead when analyzed by XRF (Tables 1–3). The median lead content of the cookware and utensils was 355 ppm. With one exception, aluminum content for all samples exceeded 940,000 ppm (94.0%), with an average concentration of 97.0% and median of 97.3% (data not shown). This is consistent with our field observations that the artisans manufacturing these items are using scrap aluminum in the production process.

3.2. Leaching of lead from cookware samples

Despite the relatively low lead content of the cookware, many of the samples released significant quantities of lead in dilute acetic acid. Lead content of the leachates for all samples is shown in Tables 1–3. Boiling for 2 h leached considerably higher concentrations of lead from the cookware pieces than the ambient 24 h extraction.

Lead content as measured by XRF was weakly correlated to the concentrations released by the 2 h boiling ($r^2 = 0.39$) and 24 h ambient
temperature \( (r^2 = 0.32) \) extractions. Lead concentrations determined by GFAAS and ICP were strongly correlated \( (r^2 = 0.83 \text{ for the 2 h leaching data, and } 0.97 \text{ for the 24 h leaching data}) \), although GFAAS data were on average 15\% (2 h) to 45\% (24 h) higher than those for ICP \( (\text{data not shown}) \). Here, we have reported the ICP data because quality control results for GFAAS slightly exceeded FDA quality control limits of \( \pm 10\% \) for check solutions \( (\text{Cheng, 2010}) \). However, GFAAS results were used to identify which solutions would be further characterized by ICP. Samples containing less than 4 \( \mu \text{g Pb/L} \) were not analyzed by ICP.

The three ladles tested were much more resistant to corrosion than the cookware samples \( (\text{Table 2}) \). Except for the 24 h ambient leaching of ladle 3Y, none of the ladles released significant amounts of lead. Only three whole pots were tested, as one used pot was found to leak and was unable to be tested. One of the pots tested \( (\text{sample 5 N, Table 3}) \) contained a very low lead content by XRF, and no detectable lead in the 2 h boiling extraction. Pieces cut from the rim of this pot also yielded no detectable lead. For reasons that are unclear, the second extraction as well as extraction of pieces cut from the rim yielded significantly more lead than the initial extraction.

### 3.3. SEM shows significant surface erosion of pots with leaching

All samples showed significant loss of aluminum in dilute acetic acid \( (\text{Tables 4 and 5}) \). Total corrosion rates based on measurements of cookware mass before and after extraction were as high as 4.58 mg/cm\(^2\) over the 2 h boiling extraction, and the average corrosion rate was 1.64 mg/cm\(^2\) over 2 h \( (\text{data not shown}) \). SEM micrographs confirm the tremendous corrosive effect of boiling for 2 h in dilute acetic acid \( (\text{Fig. 3}) \). Significant pitting of the surface was obvious on all samples, and provides further evidence that trace metals present in the pot alloy \( (\text{e.g. lead}) \) will be released into foods with regular use.

### 3.4. Estimating exposures from cookware

Lead concentrations in the leaching solutions, which resulted from soaking or boiling cookware pieces in specified volumes of 4% acetic acid, cannot be directly related to food contamination without first adjusting for the volume to surface area ratio of typical cookware. For this reason, the results obtained were first converted to corrosion rates \( (\mu \text{g lead/cm}^2\text{-h}) \) by the following calculation:

\[
\text{Corrosion rate} (\mu \text{g lead/cm}^2\text{-h}) = \frac{\text{Total lead in extraction solution}, \mu \text{g Pb}}{(\text{sample area, cm}^2) \times \text{h})}
\]

Exposures to lead per typical serving size were then estimated with the following calculation:

\[
\text{Estimated exposure} = \text{Corrosion rate} \times 2\text{ h cooking time} \times \text{estimated pot surface area} (865 \text{ cm}^2, \text{based on average of four pots}) / \text{Pot volume (1700 mL, based on average of four pots)} = \mu \text{g lead per mL} \times 250 \text{ mL per serving} = \mu \text{g lead from consumption of one serving of liquid cooked in pot.}
\]

The 250 mL volume of a metric cup has been used in other dietary studies in Africa \( (\text{Coulibaly et al., 2009}) \). The lead exposures estimated...
from cooking in these pots based on these calculations range up to 260μg per serving (Tables 1 and 3). For the 22 pieces of cookware reported in Table 1, the median estimated exposure was 97μg lead per serving. Analyses for other metals (Tables 4 and 5) suggest that depending on the source materials used, other trace metals in the cookware (e.g. cadmium, manganese, vanadium) may be elevated in pot leachates. Leaching was again higher when samples were extracted by boiling for 2 h as compared to the ambient 24 h soak. For vanadium a maximum concentration of 1.34 mg/L was observed, and for manganese, concentrations were as high as 3.78 mg/L. Cadmium concentrations were lower, at a concentration of 58μg/L. Using the calculations above, maximum estimated exposures per serving were 34.0μg vanadium, 937μg manganese and 15.6μg cadmium respectively (Table 5).

4. Discussion

Artisanal aluminum cookware has the dominant market share in Cameroon and most African countries as it has replaced cast iron. These pots are inexpensive, readily available, and often made locally in small facilities with five to ten workers. Although several studies have examined the potential for aluminum to leach from cookware (Al Juhaiman, 2010, 2012; Al Zubaidy et al., 2011; Inoue et al., 1988), very little effort has been made to assess the health risks of potential lead and other heavy metal exposures that may result from regular use of aluminum pots and utensils.

XRF analysis indicated that the pots and utensils tested contain a moderate amount of lead as a contaminant ranging from 15 to 850 ppm. These results are consistent with another recent study examining aluminum cookware manufactured in China, India, Saudi Arabia and Syria, which reported lead levels ranging from 150 ppm to 700 ppm (Al Juhaiman, 2012). To better assess the health risk from lead content in this range, we conducted a further investigation to estimate the concentration of lead that may be released into food from cooking and the use of utensils.

Although the U.S. Food and Drug Administration (Cheng, 2010) and the European Commission (2005) have prescribed test methods for determining whether harmful concentrations of lead leach from ceramic Table 3
Summary of XRF analyses of whole pots and ICP analyses of extraction solutions. Corrosion rates and exposures per serving are calculated as described in the text.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Condition</th>
<th>XRF, ppm Pb</th>
<th>2 h boil, μg Pb/L</th>
<th>μg Pb/cm²-h</th>
<th>μg Pb/serving</th>
</tr>
</thead>
<tbody>
<tr>
<td>4D whole pot</td>
<td>New</td>
<td>850</td>
<td>68</td>
<td>0.070</td>
<td>18</td>
</tr>
<tr>
<td>Repeat</td>
<td>371</td>
<td>0.380</td>
<td>96</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4Da (piece)</td>
<td>830</td>
<td>1.023</td>
<td>260</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4Db (piece)</td>
<td>869</td>
<td>0.961</td>
<td>244</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5N whole pot</td>
<td>New</td>
<td>&lt;15</td>
<td>&lt;4.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Repeat</td>
<td>&lt;4.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5Na (piece)</td>
<td>&lt;4.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5Nb (piece)</td>
<td>&lt;4.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1Y whole pot</td>
<td>New</td>
<td>336</td>
<td>NT d</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1Ya (piece)</td>
<td>769</td>
<td>0.756</td>
<td>192</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1Yb (piece)</td>
<td>899</td>
<td>0.887</td>
<td>226</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Median</td>
<td>336</td>
<td>68</td>
<td>0.822</td>
<td>209</td>
<td></td>
</tr>
</tbody>
</table>

a Letter indicates cookware location source; D = Douala, K = Kumba, N = Ngaoundéré, and Y = Yaoundé.
b Calculation method described in text.
c NT = Not tested; samples containing less than 4 μg L⁻¹ of lead by GFAAS were not analyzed by ICP.
d Rounded shape of pot bottom prevented effective boiling.

Table 4
Concentrations of aluminum, cadmium, manganese and vanadium measured by ICP in 24 h (ambient temperature) and 2 h (boiling) extractions of cookware pieces.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Al, mg/L</th>
<th>Al, mg/L</th>
<th>Cd, μg/L</th>
<th>Cd, μg/L</th>
<th>Mn, μg/L</th>
<th>Mn, μg/L</th>
<th>V, μg/L</th>
<th>V, μg/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>1D</td>
<td>4.51</td>
<td>239</td>
<td>1.4</td>
<td>3.3</td>
<td>28.4</td>
<td>690</td>
<td>&lt;3</td>
<td>20</td>
</tr>
<tr>
<td>2D</td>
<td>5.10</td>
<td>686</td>
<td>0.4</td>
<td>0.4</td>
<td>23.6</td>
<td>808</td>
<td>&lt;3</td>
<td>65.8</td>
</tr>
<tr>
<td>3D</td>
<td>2.72</td>
<td>703</td>
<td>3.8</td>
<td>4.3</td>
<td>33.6</td>
<td>942</td>
<td>4.2</td>
<td>69.3</td>
</tr>
<tr>
<td>5D</td>
<td>6.00</td>
<td>608</td>
<td>2.4</td>
<td>7.1</td>
<td>66.2</td>
<td>1563</td>
<td>4.1</td>
<td>50.1</td>
</tr>
<tr>
<td>6D</td>
<td>5.33</td>
<td>594</td>
<td>6.3</td>
<td>6.0</td>
<td>35.1</td>
<td>927</td>
<td>&lt;3</td>
<td>44.4</td>
</tr>
<tr>
<td>1K</td>
<td>4.79</td>
<td>547</td>
<td>0.600</td>
<td>1.3</td>
<td>36.1</td>
<td>256</td>
<td>6</td>
<td>39.5</td>
</tr>
<tr>
<td>3K</td>
<td>5.33</td>
<td>794</td>
<td>&lt;0.4</td>
<td>0.4</td>
<td>5.7</td>
<td>290</td>
<td>4.1</td>
<td>44.8</td>
</tr>
<tr>
<td>4K</td>
<td>4.18</td>
<td>752</td>
<td>0.600</td>
<td>8.9</td>
<td>25.7</td>
<td>953</td>
<td>4.5</td>
<td>62.9</td>
</tr>
<tr>
<td>1N</td>
<td>6.37</td>
<td>893</td>
<td>1.1</td>
<td>0.9</td>
<td>75.9</td>
<td>1727</td>
<td>4.9</td>
<td>86.6</td>
</tr>
<tr>
<td>2N</td>
<td>7.03</td>
<td>423</td>
<td>2.1</td>
<td>1.1</td>
<td>50.4</td>
<td>619</td>
<td>&lt;3</td>
<td>35.3</td>
</tr>
<tr>
<td>3N</td>
<td>4.2</td>
<td>596</td>
<td>2.1</td>
<td>1.3</td>
<td>10.6</td>
<td>362</td>
<td>&lt;3</td>
<td>28.4</td>
</tr>
<tr>
<td>4N</td>
<td>8.03</td>
<td>818</td>
<td>2.6</td>
<td>8.1</td>
<td>60.0</td>
<td>1427</td>
<td>8.1</td>
<td>74.5</td>
</tr>
<tr>
<td>2Y</td>
<td>62.25</td>
<td>1853</td>
<td>13.0</td>
<td>21.6</td>
<td>472.4</td>
<td>3785</td>
<td>7.4</td>
<td>130.1</td>
</tr>
<tr>
<td>3Y</td>
<td>2.80</td>
<td>749</td>
<td>2.5</td>
<td>16.1</td>
<td>79.7</td>
<td>1419</td>
<td>&lt;3</td>
<td>56.6</td>
</tr>
<tr>
<td>4Y</td>
<td>4.92</td>
<td>801</td>
<td>4.4</td>
<td>7</td>
<td>51.6</td>
<td>1347</td>
<td>&lt;3</td>
<td>60.4</td>
</tr>
<tr>
<td>5Y</td>
<td>5.75</td>
<td>955</td>
<td>8.5</td>
<td>25</td>
<td>49.5</td>
<td>1175</td>
<td>&lt;3</td>
<td>44.9</td>
</tr>
<tr>
<td>6Y</td>
<td>3.51</td>
<td>531</td>
<td>0.8</td>
<td>27.2</td>
<td>1.7</td>
<td>113</td>
<td>&lt;3</td>
<td>51.5</td>
</tr>
<tr>
<td>7Y</td>
<td>3.15</td>
<td>582</td>
<td>0.6</td>
<td>&lt;0.4</td>
<td>14.9</td>
<td>550</td>
<td>&lt;3</td>
<td>41.7</td>
</tr>
<tr>
<td>8Y</td>
<td>4.88</td>
<td>262</td>
<td>0.6</td>
<td>0.8</td>
<td>6.2</td>
<td>186</td>
<td>&lt;3</td>
<td>19.9</td>
</tr>
<tr>
<td>9Y</td>
<td>NT a</td>
<td>368</td>
<td>NT a</td>
<td>&lt;0.4</td>
<td>NT a</td>
<td>133</td>
<td>NT a</td>
<td>26.9</td>
</tr>
<tr>
<td>10Y</td>
<td>7.39</td>
<td>287</td>
<td>97.6</td>
<td>58.3</td>
<td>71.4</td>
<td>1290</td>
<td>&lt;3</td>
<td>33.3</td>
</tr>
<tr>
<td>11Y</td>
<td>6.20</td>
<td>251</td>
<td>4.7</td>
<td>8.8</td>
<td>39.3</td>
<td>3352</td>
<td>4.8</td>
<td>38.8</td>
</tr>
<tr>
<td>Median</td>
<td>5.01</td>
<td>602</td>
<td>2.1</td>
<td>5.2</td>
<td>36.1</td>
<td>924</td>
<td>&lt;3</td>
<td>44.8</td>
</tr>
</tbody>
</table>

a NT = Not tested; samples containing less than 4 μg L⁻¹ of lead by GFAAS were not analyzed by ICP.
cookware, no standard methods exist for metal cookware. In this study, methods similar to those used with ceramic cookware were adopted to simulate the exposures that may result from the consumption of cooked food made with these pots.

Our results indicate that lead can migrate from these pots and enter food at unacceptable levels that could significantly contribute to a child’s or adult’s body burden of this metal. The lack of refrigeration also necessitates the storage of food in pots sometimes for a number of days. The repeated re-heating of food also increases the potential for a lead build-up in the food. Further work needs to be done to examine the impact of salt content of broths and the lead leaching rates of this cookware over time, but the potential for dangerous exposures is clear.

4.1. Estimated lead exposures in comparison to relevant standards

Lead serves no useful function in the body and therefore the joint Food and Agriculture (FAO) and World Health Organization (WHO) Expert Committee on Food Additives (WHO, 2011) recommends identifying contributing sources and limiting exposures to the extent possible. The committee withdrew its provisional tolerable weekly intake (PTWI) and noted that given the lack of a threshold, it would be impossible to set a health protective level for lead exposure from dietary sources.

In 1993 the U.S. Food and Drug Administration (FDA, 1993) established a provisional total tolerable intake level (PTTIL) for lead for small children of 6 micrograms per day (FDA, 1993). This guidance was based on the U.S. Centers for Disease Control and Prevention (CDC) “blood lead level of concern” of 10 μg/dL that was adopted in 1991. The agency has subsequently determined that there is no level of exposure without deleterious effects in children, so this terminology and action level has been rescinded, although the FDA has not yet revised its guidance (CDC, 2012). However, the European Food Safety Authority (EFSA) rejected a Provisional Tolerable Weekly Intake (PTWI) of 25 μg/kg BW as inappropriate because of the lack of evidence for a threshold for lead toxicity (EFSA, 2010, 2012).

Comparing our results to the 1993 PTTIL indicates that the median lead content of even a single serving of food from this cookware exceeds this obsolete guidance level by more than 15-fold. For the 22 samples in Table 1, the median estimated lead content per serving based on the two hour boiling extraction is 97.0 μg. An adult consuming a single serving from many of the cookware samples would also exceed the PTTIL for adults of 75 micrograms per day. One serving of food from this cookware would exceed California’s Maximum Allowable Dose Level (MADL) of 0.5 μg day⁻¹ for lead (known as the safe harbor level) under Proposition 65 by up to almost 500-fold (Sample 11Y, Table 5).
Results from the leachate tests indicate that a single 250 mL serving estimated range of ing was 174 mg, which exceeds the World Health Organization’s reported in Table 5, the median potential exposure to aluminum per serving was 174 mg, which exceeds the World Health Organization’s estimated range of weekly consumption of aluminum through the diet of 11–136 mg/person (WHO, 2011).

Cadmium is a by-product of zinc and is also used as a coating on iron and steel. It is a carcinogen and is linked to cardiovascular, reproductive, neurological, gastrointestinal and respiratory symptoms (OSHA, 2011). Results from the leachate tests indicate that a single 250 mL serving from this cookware may contain 15.6 μg of cadmium (see Table 5). By contrast California set a MADL of 4.1 μg day⁻¹ for oral ingestion (California OEHHA (Office of Environmental Health Hazard Assessment), 2001). Five (23%) of the cookware pieces tested (Table 5) exceed this amount based on the estimated cadmium per serving, indicating that cadmium contamination is another concern for some of this cookware.

Levels of other two metals detected in cookware leachates, manganese and vanadium, do not appear to be harmful. A single 250 mL serving from this cookware may contain up to 988 μg manganese (Table 5). For the samples reported in Table 5, the median manganese content per serving is 248 μg. Studies in children suggest that manganese may impact brain development impacting behavior and the ability to learn (ASTDR, 2012a). The interim Tolerable Upper Intake Level established by the Institute of Medicine (IOM) for 70 kg adults is 11,000 μg/manganese per day (ASTDR, 2012a). Thus it does not appear that levels of manganese leaching from this cookware pose a significant hazard. There also appears to be little hazard posed by the levels of vanadium that were measured. The IOM Upper Tolerable Limit for vanadium is 1,800 μg/day (ASTDR, 2012b). In contrast, the maximum concentration of vanadium leached under our testing procedure was 34 μg/serving (Table 5).

4.3. Exposure assessment

We note that our assessment of lead exposures per serving may underestimate actual conditions in Cameroon. A recent study suggests that lead contamination levels in typical foods consumed as part of the local diet in Cameroon can also be a significant contributor to lead ingestion (Gimou et al., 2014).

Although our investigation made no attempt to directly quantify the contribution of lead or other metals from cookware to body burdens in the exposed population, one recent study from Thailand investigated the possible contribution from aluminum cooking pots to blood lead levels (Swaddiwudhipong et al., 2013). The study was conducted in a rural area where few other sources of lead were present, and found that among 254 children, those in households with inexpensive non-certified aluminum cooking pots had higher blood lead levels in contrast to those who used certified pots. No data was available on the total or soluble lead levels from these pots. One additional study investigated the possible contribution to urinary lead levels from cookware commonly used in rural Bangladesh. Mean lead levels in the leachate of six new aluminum pots extracted at ambient temperature for 24 h with 4% acetic acid averaged 113 μg/L. Pots released a maximum of 381 μg lead/L, or 95 μg lead per 250 mL serving (Bergkvist et al., 2010).

In addition, our investigation uncovered significant concerns of occupational exposures to workers in the manufacturing process and those later involved in finishing the pots. The latter work is conducted by filing and scraping the metal to smooth out rough surfaces and shine the interior and exterior surfaces. During this process significant dust is released. (see online video in Appendix A. Supplementary data)

We are unaware of any blood lead testing having been conducted among any group in Cameroon. No laboratories with blood lead testing capacity have been identified in the country. However based on these results, it is highly recommended that public health authorities conduct an investigation of blood lead levels and assess possible exposure sources particularly among vulnerable populations including children and pregnant women.

4.4. Regulatory approaches and future research directions

These results also point to an immediate need to regulate the use of source materials in aluminum cookware. Aluminum pots by themselves may not necessarily pose a significant hazard if they are manufactured with strict controls to prohibit or limit the sources and use of scrap metal contaminated with significant amounts of lead. Given the large number of small artisanal manufacturing businesses operating in Cameroon, it is unlikely that manufacturing processes can be significantly improved in the short-term. However, more controls can be placed on limiting the use of scrap metal to restrict the content of lead and other heavy metals in such pots. Enforcement of such measures can possibly be rapidly determined with portable XRF testing equipment.

Lead does not serve any useful function in aluminum products. Industry guidelines call for limiting the use of alloying elements to silicon, magnesium, zinc, titanium, chromium, and manganese depending upon the application (Cookware Manufacturers Association (CMA), 2012). Our results underline the immediate need to regulate the metal content of aluminum pots and other cookware produced around the world. Although there are government standards in the U.S. and U.K. for the amount of lead allowed to leach from ceramic ware, there are no comparable standards for metallic cookware. Such legal frameworks are needed to specify the maximum concentration of lead and other harmful metals that can be used in these products.

Given the expense in testing such cookware and the lack of capacity among most governments, any regulatory solution should include a provision to require third party certification of cookware against a health protective standard for the concentration of lead and other metals permitted in a leachate test. Certification programs may also benefit from the use of XRF testing to screen a large sample of aluminum cookware in order to select a subsample for leachate testing. A manufacturer then wishing to sell their wares would have to first obtain certification and display the certification mark before they could market their products. Such efforts could be successfully implemented even in countries with very limited enforcement resources.

One third-party certification standard for cookware that includes criteria for lead and other heavy metals is NSF Protocol 390 for stovetop cookware for home use (NSF International, 2011). This standard, or other equivalent standards, can be referenced in a mandatory certification program to control the contamination level of metals in food from aluminum cookware.

4.5. Future research

All of the cookware tested was made at small manufacturing facilities that lack the infrastructure to anodize cookware. Anodization provides a more resilient coating on aluminum that also reduces the leaching of metals from cookware (Sekheta et al., 2010). Therefore, we do not know if anodized finishing of these surfaces would offer sufficient protection to reduce or eliminate the high concentration of metals that were observed in the leachate from the pots tested. Future
investigations should include samples from anodized aluminum to better gauge the potential reduction of lead and other metals migrating into food. It seems unlikely that the amount of aluminum corrosion observed by SEM (Fig. 3) and confirmed by our analyses (Table 5) will be reduced without changing the manufacturing process to provide a more corrosion-resistant finish.

Given the widespread use of inexpensive aluminum cookware throughout the developing world very little research has been conducted on the potential exposures to lead and other heavy metals from this source. Additional research is needed to quantify the extent of this problem throughout Africa, the Mideast, and Asia where earlier research has suggested that a similar situation may be present. It will be important to identify source materials that introduce harmful levels of lead and other metals.

Given the ubiquitous use of non-anodized aluminum cookware in developing nations, and the documented presence of lead in similar concentrations in products from six other countries (Al Juhaiman, 2012; Al Zubaidy et al., 2011; Bergkvist et al., 2010), our results may foretell an important yet largely overlooked source of lead poisoning around the world. The results from our leachate testing indicate that food cooked in such cookware may be readily contaminated with lead at harmful levels that can impact the health of children and adults.

At the same time, our investigation strongly suggests that the source of lead in these products is a contaminant of the recycled aluminum used in their manufacture. Given the relatively small concentration of other metals used in these pots, the removal of scrap of questionable quality from the production of aluminum cookware should not be cost prohibitive.

4.6. Implications for public health

Even low-level lead exposures can result in significant health impacts. Unlike some other sources of lead contamination, lead poisoning from cookware can impact entire families over a life-time. Many of the pots tested are capable of leaching enough lead into food that could greatly increase exposure levels particularly among the most vulnerable populations. This problem is magnified by the poor nutritional status of many people in Cameroon that will generally facilitate greater lead absorption.

Although adults will generally absorb less of the lead that they ingest than children, these results suggest that lead contamination of food from aluminum pots may be a significant contributor to the increased risk of cardiovascular disease that we are witnessing in developing countries. Pregnant women also absorb a greater proportion of the lead they ingest and are susceptible to higher rates of miscarriages, stillbirths, and pre-term deliveries. Their exposures are also passed on to the developing fetus. Children are generally the most susceptible as lead exposures that are likely from this source will contribute to neurological deficits, hearing loss, reduced growth, and a range of other health effects. The costs to society in terms of lost educational opportunity, reduced IQ, and potentially increased crime rates greatly outweigh any additional costs that may be incurred by better controls in the manufacturing of cookware to eliminate the use of lead and other heavy metals (Attina and Trasande, 2013).

5. Conclusions

This paper presents the findings of an unusual investigation initiated after individuals at outreach programs on lead hazard awareness that the authors conducted in Cameroon suggested that aluminum pots may be a source of lead exposure. Based on the findings from our investigation this previously unrecognized exposure source has the potential to be of much greater public health significance than lead paint or other well-known sources that are common in Cameroon. Given the widespread use of this type of inexpensive aluminum cookware around the world, and limited evidence from additional studies with similar results, these types of pots may be one of the most significant contributors to lead poisoning throughout the developing world.

Typical single serving size portions of foods cooked in these pots can greatly exceed daily intake guidelines established by public health authorities for lead, aluminum and cadmium. As a result, further investigation should be prioritized to better assess the extent of this hazard in Cameroon and in other countries where similar production methods are commonly used for aluminum pots.

Competing interests

The authors declare no actual or competing financial interests.

Acknowledgements

Samples were collected by Yves Nzitat, Assistant program officer of the Research and Education Centre for Development (CREPD) in Cameroon. The National Science Foundation (DUE 9952552) provided financial support for acquisition of the Varian 220 AA spectrometer used in these studies.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2014.07.016.

References


Weidenhamer JD, Clement ML. Widespread lead contamination of imported low-cost jewelry in the US. Chemosphere 2007;67:561–5.

