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Lead exposure from aluminum cookware in Cameroon

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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

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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.

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

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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.

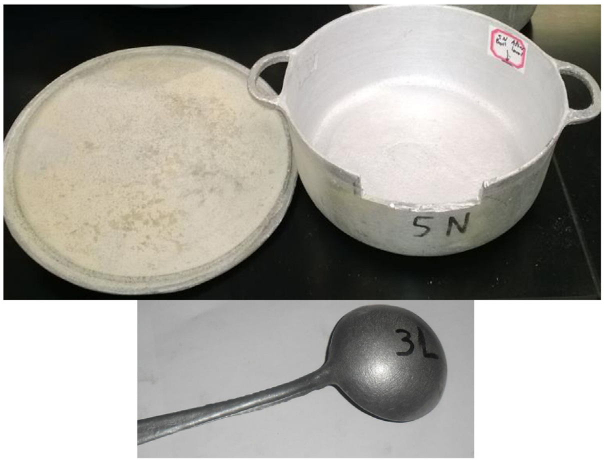

Fig. 2. Examples of cookware evaluated in this study. Above, one of the whole pots after testing and removal of pieces from the rim for further testing and microscopy. Below, one of three ladles tested.

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 firewood), 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 ladles 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. Ladles 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

Table 1
Summary of XRF analyses of cookware pieces and ICP analyses of extraction solutions. Corrosion rates and exposures per serving are calculated as described in the text.

Sample[a]	Condition	XRF, ppm Pb	24 h ambient, μg Pb/L	2 h boil, μg Pb/L	μg Pb/cm²-h[b] (based on 2 h boiling extraction)	μg Pb/serving[b]
1D	New	446	183	229	0.259	66
2D	New	240	27	122	0.124	32
3D	New	547	88	478	0.565	144
5D	New	268	138	449	0.403	103
6D	New	409	139	354	0.413	105
1K	New	111	27	69	0.076	19
3K	New	99	50	88	0.083	21
4K	New	412	149	433	0.438	112
1N	New	213	67	358	0.417	106
2N	New	342	45	128	0.195	50
3N	New	389	138	423	0.442	112
4N	New	458	212	675	0.775	197
2Y	Used	476	402	734	0.753	192
3Y	New	546	240	561	0.631	161
4Y	New	460	148	422	0.450	115
5Y	New	208	74	184	0.154	39
6Y	New	<21	18	18	0.022	6
7Y	New	225	76	189	0.221	56
8Y	Used	374	13	91	0.086	22
9Y	Used	143	NT[c]	71	0.081	21
10Y	New	637	131	339	0.356	91
11Y	New	355	294	866	0.974	248
	Median	364	110	346	0.380	97

[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.

temperature ($r^2 = 0.32$) extractions. Lead concentrations determined by GFAAS and ICP were strongly correlated ($r^2 = 0.83$ for the 2 h leaching data, and 0.97 for the 24 h leaching data), although GFAAS data were on average 15% (2 h) to 45% (24 h) higher than those for ICP (data not shown). Here, we have reported the ICP data because quality control results for GFAAS slightly exceeded FDA quality control limits of ±10% for check solutions (Cheng, 2010). However, GFAAS results were used to identify which solutions would be further characterized by ICP. Samples containing less than 4 μg lead L^{-1} by GFAAS were not analyzed by ICP.

The three ladles tested were much more resistant to corrosion than the cookware samples (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 (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 extractable lead. A second pot (sample 1Y, Table 3) also showed no detectable lead in the 2 h boiling extraction, but pieces cut from the rim did yield significant amounts of lead when tested. Due to the rounded shape of the pot bottom, it was difficult to maintain this pot at a constant boil, which may have resulted in less than maximal extraction in the 2 h test. A third pot (sample 4D, Table 3) was tested twice by boiling

Table 2
Summary of XRF analyses of ladles, and ICP analyses of lead in the 24 h (ambient temperature) and 2 h (boiling) extraction solutions.

Sample[a]	Condition	XRF, ppm Pb	24 h ambient, μg Pb/L	2 h boil, μg Pb/L
Ladles				
1Y	Used	430	NT[b]	NT
2Y	Used	332	NT	NT
3Y	Used	431	19	NT
	Median	430		

[a] Letter indicates cookware location source; D = Douala, K = Kumba, N = Ngaoundéré, and Y = Yaoundé.
[b] NT = Not tested; samples containing less than 4 μg/L of lead by GFAAS were not analyzed by ICP.

extraction. 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 (Tables 4 and 5). Total corrosion rates based on measurements of cookware mass before and after extraction were as high as 4.58 mg/cm² over the 2 h boiling extraction, and the average corrosion rate was 1.64 mg/cm² over 2 h (data not shown). SEM micrographs confirm the tremendous corrosive effect of boiling for 2 h in dilute acetic acid (Fig. 3). Significant pitting of the surface was obvious on all samples, and provides further evidence that trace metals present in the pot alloy (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 (μg lead/cm²-h) by the following calculation:

Corrosion rate (μg lead/cm²-h) = (Total lead in extraction solution, μg) / ((sample area, cm²) × (h)).

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

Estimated exposure = Corrosion rate × 2 h cooking time ×estimated pot surface area (865 cm², based on average of four pots) / Pot volume (1700 mL, based on average of four pots) =μg lead per mL × 250 mL per serving = μg 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 (Coulibaly et al., 2009). The lead exposures estimated

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.

Sample[a]	Condition	XRF, ppm Pb	2 h boil, μg Pb/L	μg Pb/cm^2-h[b]	μg Pb/serving[b]
				(based on 2 h boiling extraction)	
4D whole pot	New	850	68	0.070	18
Repeat			371	0.380	96
4Da (piece)			830	1.023	260
4Db (piece)			869	0.961	244
5N whole pot	New	<15	<4.0		
Repeat			<4.0		
5Na (piece)			<4.0		
5Nb (piece)			<4.0		
1Y whole pot	New	336	NT[c,d]		
1Ya (piece)			769	0.756	192
1Yb (piece)			899	0.887	226
	Median	336	68	0.822	209

[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.

from cooking in these pots based on these calculations range up to 260 μg per serving (Tables 1 and 3). For the 22 pieces of cookware reported in Table 1, the median estimated exposure was 97 μg 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 58 μg/L. Using the calculations above, maximum estimated exposures per serving were 34.0 μg vanadium, 937 μg manganese and 15.6 μg 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 4
Concentrations of aluminum, cadmium, manganese and vanadium measured by ICP in 24 h (ambient temperature) and 2 h (boiling) extractions of cookware pieces.

Sample	Al, mg/L	Al, mg/L	Cd, μg/L	Cd, μg/L	Mn, μg/L	Mn, μg/L	V, μg/L	V, μg/L
	24 h	2 h	24 h	2 h	24 h	2 h	24 h	2 h
1D	4.51	239	1.4	3.3	28.4	690	<3	20
2D	5.10	686	0.4	0.4	23.6	808	<3	65.8
3D	2.72	703	3.8	4.3	33.6	942	4.2	69.3
5D	6.00	608	2.4	7.1	66.2	1563	4.1	50.1
6D	5.33	594	6.3	6.0	35.1	927	<3	44.4
1K	4.79	547	0.600	1.3	36.1	256	6	39.5
3K	5.33	794	<0.4	0.4	5.7	290	4.1	44.8
4K	4.18	752	0.600	8.9	25.7	953	4.5	62.5
1N	6.37	893	1.1	0.9	75.9	1727	4.9	86.6
2N	7.03	423	2.1	1.1	50.4	619	<3	35.3
3N	4.2	596	2.1	1.3	10.6	362	<3	28.4
4N	8.03	818	2.6	8.1	66.0	1427	8.1	74.5
2Y	62.25	1853	13.0	21.6	472.4	3785	7.4	130.1
3Y	2.80	749	2.5	16.1	79.7	1419	<3	56.6
4Y	4.92	801	4.4	7	51.6	1347	<3	60.4
5Y	5.75	955	8.5	25	49.5	1175	<3	44.9
6Y	3.51	531	0.8	27.2	1.7	113	<3	51.5
7Y	3.15	582	0.6	<0.4	14.9	550	<3	41.7
8Y	4.88	262	0.6	0.8	6.2	186	<3	19.9
9Y	NT[a]	368	NT[a]	<0.4	NT[a]	133	NT[a]	26.9
10Y	7.39	287	97.6	58.3	71.4	1290	<3	33.3
11Y	6.20	251	4.7	8.8	39.3	3352	4.8	38.8
Median	5.01	602	2.1	5.2	36.1	934	<3	44.8

[a] NT = Not tested; samples containing less than 4 μg L^{-1} of lead by GFAAS were not analyzed by ICP.

Table 5
Potential exposures serving for aluminum, cadmium, manganese and vanadium based on data in Table 4.

Sample	Al, mg	Cd, μg	Mn, μg	V, μg
1D	69	1.0	199	5.8
2D	178	0.1	209	17.0
3D	211	1.3	283	20.8
5D	139	1.6	357	11.4
6D	176	1.8	275	13.2
1K	153	0.4	72	11.1
3K	191	0.1	70	10.8
4K	194	2.3	246	16.1
1N	265	0.3	512	25.6
2N	164	0.4	240	13.7
3N	158	0.3	96	7.5
4N	239	2.4	417	21.8
2Y	484	5.6	988	34.0
3Y	214	4.6	406	16.2
4Y	218	1.9	366	16.4
5Y	203	5.3	250	9.6
6Y	162	8.3	34	15.8
7Y	173	<0.1	164	12.4
8Y	63	0.2	45	4.8
9Y	107	<0.1	39	7.8
10Y	77	15.6	345	8.9
11Y	72	2.5	959	11.1
Median	174	1.4	248	12.8
Range	63–484	<0.1–15.6	34–988	4.8–34.0

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. In addition, 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,

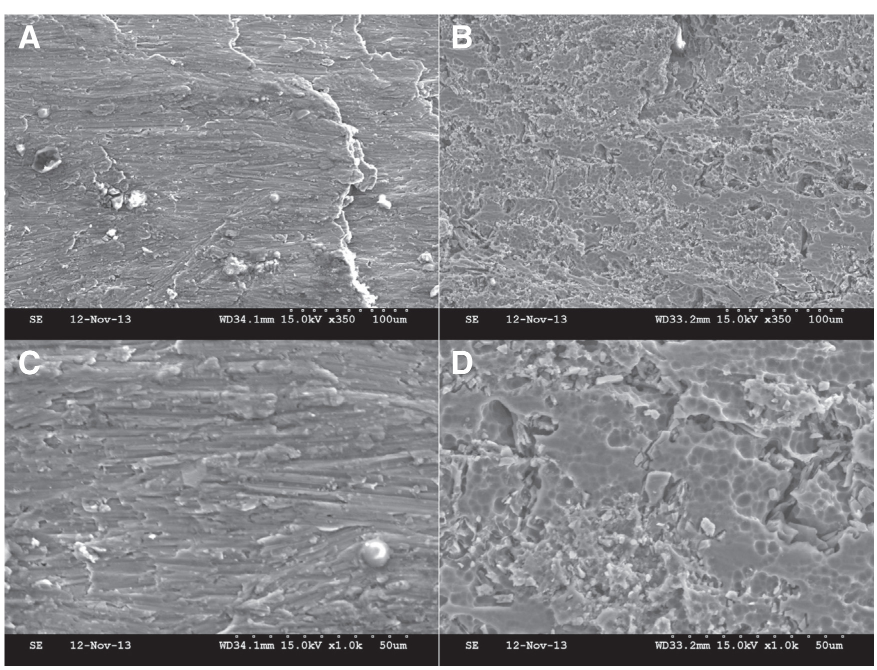

Fig. 3. Scanning electron micrograph images of aluminum cookware surfaces before and after extraction with boiling 4% acetic acid for 2 h. (A and C) Prior to extraction at magnifications of 350× (A) and 1000× (C). (B and D) Following extraction surface erosion is obvious at magnifications of 350× (B) and 1000× (D).

Table 1) (California OEHHA (Office of Environmental Health Hazard Assessment), 2013).

4.2. Estimated exposures to other metals

Other metals were found in the leachates of these pots at potentially harmful levels. The PTWI for aluminum of 140 mg/person/week or 20 mg/person/day (based on a typical 70 kg adult) would be exceeded with a single serving from all of the pots tested in this study (Table 5) (WHO, 2011). Similarly, for children a single serving from all of the pots tested would exceed the PTWI of 60 mg/child per week or 8.6 mg/child per day (based on a 30 kg child). For the 22 samples 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 two other 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.

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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

Al Juhaiman LA. Estimating aluminum leaching from aluminum cook wares in different meat extracts and milk. J Saudi Chem Soc 2010;14:131–7.

Al Juhaiman LA. Estimating aluminum leaching from aluminum cookware in different vegetable extracts. Int J Electrochem Sci 2012;7:7283–94.

Al Zubaidy EA, Mohammad F, Bassioni G. Effect of pH, salinity and temperature on aluminum cookware leaching during food preparation. Int J Electrochem Sci 2011;6:6424–41.

ATSDR (U.S. Agency for Toxic Substances, Disease Registry). Toxicological profile for manganese. [Cited 24.4.2014]. Available from http://www.atsdr.cdc.gov/toxprofiles/tp151.pdf, 2012.

ATSDR (U.S. Agency for Toxic Substances, Disease Registry). Toxicological profile for vanadium. [Cited 24.4.2014]. Available from http://www.atsdr.cdc.gov/toxprofiles/tp58.pdf, 2012.

Attina TM, Trasande L. Economic costs of childhood lead exposure in low- and middle-income countries. Environ Health Perspect 2013;121:1097–102.

Bergkvist C, Kippler M, Hamadani JD, Grander M, Tofail F, Berglund M, et al. Assessment of early-life lead exposure in rural Bangladesh. Environ Res 2010;110:718–24.

California OEHHA (Office of Environmental Health Hazard Assessment). Proposition 65 maximum allowable daily level (MADL) for reproductive toxicity for cadmium (oral route). [Cited 24.4.2014]. Available from http://www.oehha.org/prop65/law/pdf_zip/cadmium%20MADL.pdf, 2001.

California OEHHA (Office of Environmental Health Hazard Assessment). Proposition 65 no significant risk levels (NSRLs) for carcinogens and maximum allowable dose levels (MADLs) for chemicals causing reproductive toxicity. [Cited 24.4.2014]. Available from http://www.oehha.org/prop65/pdf/safeharbor081513.pdf, 2013.

CDC (Centers for Disease Control, Prevention). Low level lead exposure harms children: a renewed call for primary prevention. [Cited 24.4.2014]. Available from http://www.cdc.gov/nceh/lead/ACCLPP/Final_Document_030712.pdf, 2012.

Cheng J. U.S. Food and Drug Administration elemental analysis manual: Section 4.6. Inductively coupled plasma-atomic emission spectrometric determination of cadmium and lead extracted from ceramic foodware. [Cited 21.3.2014]. Available from http://www.fda.gov/Food/FoodScienceResearch/LaboratoryMethods/ucm221685.htm, 2010.

Cookware Manufacturers Association (CMA). Technical engineering standards for cookware and bakeware. Mountain Brook: AL:CMA; 2012.

Coulibaly A, O'Brien H, Galibois I. Development of a Malian food exchange system based on local foods and dishes for the assessment of nutrient and food intake in type 2 diabetic subjects. S Afr J Clin Nutr 2009;22:31–5.

EFSA. Lead dietary exposure in the European population. EFSA J 2012;10(7):2831. [Cited 24.4.2014]. Available from http://www.efsa.europa.eu/en/efsajournal/doc/2831.pdf].

EFSA (European Food Safety Authority). EFSA panel on contaminants in the food chain (CONTAM); scientific opinion on lead in food. EFSA J 2010;8(4):1570. [Cited 24.4.2014]. Available from http://www.efsa.europa.eu/en/search/doc/1570.pdf].

El-Desoky GE, Aboul-Soud MA, Al-Othman ZA, Habila M, Giesy JP. Seasonal concentrations of lead in outdoor and indoor dust and blood of children in Riyadh, Saudi Arabia. Environ Geochem Health 2013;36:583–93.

European Commission. Official journal of the European communities: ED 84/500/EEC. Council Directive of 15 Oct. 1984 on the approximation of the laws of the Member States relating to ceramic articles intended to come into contact with foodstuffs (84/500/EEC) amended by Commission Directive 2005/31/EC of 29 April 2005;

2005. [Cited 24.4.2014]. Available from http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri= OJ:L:2005:110:0036:0039:en:PDF].
FDA (U.S. Food, Drug Administration). Lead-soldered food cans. Fed Regist 1993;58(17):33860–71.
Gimou M-M, Pouillot R, Charrondiere UR, Noël L, Guérin T, Leblanc J-C. Dietary exposure and health risk assessment for 14 toxic and essential trace elements in Yaoundé: the Cameroonian total diet study. Food Addit Contam A 2014. http://dx.doi.org/10.1080/19440049.2014.909953.
Gottesfeld P, Kuepouo G, Tetsopgang S, Durand K. Lead concentrations and labeling of new paint in Cameroon. J Occup Environ Hyg 2013;10:243–9.
Inoue T, Ishiwata H, Yoshihira K. Aluminum levels in food-simulating solvents and various foods cooked in aluminum pans. J Agric Food Chem 1988;36:599–601.
Kalra V, Sahu JK, Bedi P, Pandey RM. Blood lead levels among school children after phasing-out of leaded petrol in Delhi, India. Indian J Pediatr 2013;80:636–40.
Lanphear BP, Hornung R, Khoury J, Yolton K, Baghurstl P, Bellinger DC, et al. Low-level environmental lead exposure and children's intellectual function: an international pooled analysis. Environ Health Perspect 2005;113:894–9.
Li T, Dai YH, Xie XH, Tan ZW, Zhang SM, Zhu ZH. Surveillance of childhood blood lead levels in 11 cities of China. World J Pediatr 2014;10:29–37.
Lim SS, Vos T, Flaxman AD, Danaei G, Shibuya K, Adair-Rohani H, et al. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990–2010: a systematic analysis for the global burden of disease study 2010. Lancet 2012;380:2224–60.
Mohammad F, Al Zubaidy E, Bassioni G. Effect of aluminum leaching process of cooking wares on food. Int J Electrochem Sci 2011;6:222–30.
Naicker N, Mathee A, Barnes B. A follow-up cross-sectional study of environmental lead exposure in early childhood in urban South Africa. S Afr Med J 2013;103:935–8.
NSF International. NSF protocol P390–2011: stovetop cookware for home use. Ann Arbor, MI: NSF International; 2011.
OSHA (U.S. Occupational Safety, Health Administration). Cadmium. [Cited 24.4.2014]. Available from https://www.osha.gov/SLTC/cadmium/index.html, 2011.
Ritchie JAB, Gerstenberger SL. An evaluation of lead concentrations in imported hot sauces. J Environ Sci Health B 2013;48:530–8.
Sekheta MAF, Sahtout AH, Ahmad HF, Sekheta AHF, Sharabi RO, Airoud KA. The group of hidden hazards in enhanced HACCP and ISO-22000 based quality systems. Internet J Food Saf 2010;12:146–57.
Swaddiwudhipong W, Tontiwattanasap W, Khunyotying W, Sanreun C. Blood lead levels among rural Thai children exposed to lead-acid batteries from solar energy conversion systems. Southeast Asian J Trop Med 2013;44:1079–87.
Tuakuila J, Kabamba M, Mata H, Mata G. Blood lead levels in children after phase-out of leaded gasoline in Kinshasa, the capital of Democratic Republic of Congo (DRC). Arch Public Health 2013;71:5.
Weidenhamer JD. Lead contamination of inexpensive seasonal and holiday products. Sci Total Environ 2009;407:2447–50.
Weidenhamer JD, Clement ML. Widespread lead contamination of imported low-cost jewelry in the US. Chemosphere 2007;67:961–5.
WHO (World Health Organization). Evaluation of certain food additives and contaminants: seventy-fourth report of the Joint FAO/WHO Expert Committee on Food Additives. (WHO technical report series; no. 966). [Cited 24.4.2014]. Available from http://whqlibdoc.who.int/trs/WHO_TRS_966_eng.pdf, 2011.
Wigle DT, Lanphear BP. Human health risks from low-level environmental exposures: no apparent safety thresholds. PLoS Med 2005;2:1232–4.
Xie XH, Tan ZW, Jia N, Fan ZY, Zhang SM, Lu YY, et al. Blood lead levels among children aged 0 to 6 years in 16 cities of China, 2004–2008. Chinese Med J Peking 2013;126:2291–5.