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# Metal exposures from source materials for artisanal aluminum cookware

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#### ABSTRACT

Artisanal aluminum cookware releases lead and other metals that pose significant health risk for people in low and middle-income countries. Cookware is made from recycled engine and electronic appliance parts, cans, and other materials. We obtained fourteen custom-made pots from Ghana, produced from seven different scrap aluminum sources. We sought to determine whether avoiding certain source materials could reduce leaching of metals. Cooking was simulated using dilute acetic acid and palm oil. Aluminum released from all pots exceeded recommended guidelines. Variable amounts of lead, cadmium, chromium, nickel and other metals were leached, with the most lead coming from auto radiators and mixed metals. Pots made from engine blocks did not yield detectable amounts of lead. All pots released potentially harmful concentrations of two or more metals. Selective scrap aluminum sourcing for recycled cookware does not avoid metal contamination of food, although some sources may release lower concentrations of certain metals. **ARTICLE HISTORY** 

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#### **KEYWORDS**

Lead; cadmium; aluminum; heavy metals; artisanal cookware

# 1. Introduction

Low-level toxic metal exposures pose serious health risks including neurocognitive disorders and cardiovascular disease. Chowdhury et al. (2018) concluded that arsenic, lead, cadmium and copper exposures are risk factors for cardiovascular and coronary heart disease, including stroke. Lead toxicity impacts not only cardiovascular health including high blood pressure but also causes permanent brain damage at low concentrations in children, leading to lifelong learning disability, intellectual development deficits, and behavioral problems (Braun et al. 2008; Wigle and Lanphear 2005). Imaging studies confirm lasting changes to the brain, such as decreased brain volume in adults who had low-level lead exposures in childhood (Cecil et al. 2008). Estimates place the global toll of lead exposure at 674,000 premature deaths annually (Lim et al. 2012). The economic cost of childhood lead exposure has been estimated to be 1.2% of global gross domestic product (GDP), and in Africa alone the cost was estimated to be \$135 billion or 4% of GDP (Attina and Trasande 2013).

Years after lead was removed from gasoline in most countries, blood lead levels in children remain stubbornly elevated in many low and middle-income countries (LMICs) including the Democratic Republic of Congo (Tuakuila et al. 2013), South Africa (Naicker et al. 2013), Egypt (Moawad et al. 2016), Benin (Bodeau-Livinec et al. 2016), and Uganda (Cusick et al. 2018). Numerous recent studies have sought to identify ongoing sources of lead exposure in

Supplemental data for this article can be accessed here

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these countries. Artisanal aluminum cookware is manufactured in many LMICs from scrap materials, and an initial study in Cameroon (Weidenhamer et al. 2014) found that while the overall lead content of the cookware was low, cooking with acidic foods (using dilute acetic acid as a food simulant) could result in exposures per serving as high as  $260 \mu g$ . Tests of cookware from ten LMICs, in which simulated cooking leached up to  $1426 \mu g$  of lead per serving (Weidenhamer et al. 2017) confirmed the potential risk of lead exposures from artisanal cookware. Street et al. (2020) tested twenty pots from five South African provinces, finding lead released by all samples. Jitaru et al. (2019) compared concentrations of metals prepared in stainless steel and artisanal cookware. Mean concentrations of aluminum were 6.7 times higher, and concentrations of lead were as much as 26 times higher when tomatoes were cooked in artisanal aluminum cookware.

Mixed scrap metal, including engine and electronic appliance parts, cans and other aluminum scrap are used to produce artisanal cookware as documented in previous studies (Figures 1–3; Weidenhamer et al. 2014, 2017; Mathee et al. 2020; Street et al. 2020). Artisanal production of cast aluminum pots is widespread across Africa and many LMICs (Osborn 2009). While encouraging local recycling and providing economic opportunity for producers, the industry also poses health risks. Producers and their families can be exposed to toxic metals and fine particulate matter, and consumers risk exposure to toxic metals in food cooked in these pots (Jitaru et al. 2019; Mathee et al. 2020; Shezi et al. 2020). Street et al. (2020) evaluated the migration of aluminum, lead and arsenic in comparison to European Union (EU) permissible limits. All samples tested exceeded the EU guidelines for aluminum, and allowable concentrations of lead and arsenic were exceeded by some samples. Given the many contaminants that can be found in scrap metal, the potential for multiple exposures must be considered.

Our objectives were to examine the contribution of lead and other contaminant metals from different segregated source materials used to produce artisanal cookware. The goal was to determine whether potential health risks posed by using this cookware could be reduced by excluding source materials that release greater concentrations of hazardous metals. Fourteen pots from Ghana, two each produced from seven different types of scrap aluminum, were evaluated for metal leaching by boiling dilute (4% vol/vol) acetic acid solutions. Leaching of metal during cooking with palm oil, a commonly used cooking oil in west Africa, was also examined. The total metal content of each pot was also determined.

#### 2. Methodology

## 2.1. Sample collection

In Ghana, metal recyclers collect and melt aluminum scrap into bars that are purchased by cookware producers (Figure 2(c,d)). Thus, scrap materials are generally melted twice in the production process. Source materials for the aluminum cookware used in this study were supplied by an informal metal recycler in Accra, Ghana. We requested that aluminum from the most common waste streams be segregated by recyclers and melted separately to fabricate bars. These smelters specialize in collecting and recycling aluminum. In coordination with these recyclers, we selected seven separate waste streams to process into aluminum ingots. Ingots, separated by source material, were taken to a local cookware producer who then made the finished pots.

Two pots, one larger (12 L) and one smaller (5 L), with lids, were made from each of seven source materials: blister foils, cans, condensers, engine block, mixed metal (including all scrap materials), auto radiators, and small mixed metals (without larger pieces such as engine blocks and radiators) (Figure 1). Prior to and between extractions, all pots were washed in the laboratory with soap and rinsed thoroughly with deionized water.



Figure 1. Source materials used to produce the cookware used in this study. (a) Small mixed metals. (b) Plastic-coated blister foils. (c) Aluminum cans. (d) Workers removing aluminum air conditioner fins. (e) Engine block. (f) Auto radiators.



Figure 2. Photos of aluminum recycling and artisanal cookware production. (a) Air conditioner fins being melted down. (b) Melting of plastic-coated blister foils produces noxious fumes. (c) Scrap is initially melted into ingots which are (d) sold by weight to cookware artisans.

# 2.2. Leaching tests: dilute acetic acid

There is no standardized method to replicate measurements of metals leaching from aluminum cookware. Our methods follow those used by Weidenhamer et al. (2014, 2017). A 2-hour boiling extraction with dilute acetic acid (4% vol/vol) simulated cooking with acidic foods such as tomato sauce (Inoue et al. 1988). This procedure varies slightly from the method of Street et al. (2020), which uses 3% (v/v) acetic acid and is based on EN ISO 4531:2017 for vitreous and porcelain enamels.



Figure 3. (a) Melted metal being poured into sand mold to produce pots. the coppery appearance of the molten metal is from an engine block. (b) Artisan is creating a sand mold. (c) Rough edges and other defects are removed before finishing the pots. (d) Finished pots for sale in local marketplaces.

The initial volume of 4% acetic acid added to each pot was 2.0 L (smaller pots) or 4.0 L (larger pots). Pots were brought to a gentle boil over a natural gas burner. Pot lids helped retard evaporation. Every 30 minutes, pots were refilled with acetic acid to their original volume, with a final addition 15 minutes prior to completing the boil. After cooling, solutions were transferred directly to 50 mL polyethylene centrifuge tubes and refrigerated until analysis. This procedure was repeated up to four times to assess fluctuations in metal concentrations released by corrosion during ongoing usage.

## 2.3. Leaching tests: palm oil

Zhou et al. (2017) compared amounts of lead and cadmium leached into vegetable oil from aluminum pots to amounts leached into separate test solutions of water, 15% ethanol or 4% acetic acid, and found that significantly more lead and cadmium leached into vegetable oil. For this reason, following the first two acetic acid extractions, we conducted a test of the ability of palm oil to extract lead and cadmium. Palm oil is widely used for cooking in Western Africa, and West African red palm oil (Praise Zomi \*, from Ghana) was purchased from an online vendor. Palm oil (1.0 L) was added to each of the seven smaller pots and these were heated to 110-130°C for one hour. After cooling, approximately 5 mL was transferred to a 15-mL polyethylene centrifuge tube before congealing, and refrigerated until analysis. Palm oil samples (0.25 g) were digested with 5 mL nitric

acid in a microwave digester, and diluted to 50.0 mL for analysis. Analysis of uncooked palm oil was included as a method blank, and concentrations of lead were below the instrument detection limit of 2.1  $\mu$ g/L, which corresponded to a concentration below 0.38  $\mu$ g/mL in the palm oil.

#### 2.4. Total metals analysis

As noted above, all pots were made after an initial melt of the scrap material into ingots, and subsequent re-melting of the ingots to make a large and a small pot from each source material. Pots were therefore expected to be relatively homogeneous, and the large and small pots from each material similar in composition. Samples were obtained from each pot lid, and 50-100 mg was digested in concentrated ultra high purity HCl in 15 mL acid-leached Teflon Savillex containers for 5 minutes, until effervescence ceased. To these solutions, 1 mL concentrated ultra-high purity HNO<sub>3</sub> 0.5 mL concentrated ultra-high purity HF, and 1.5 mL of 18.2 M $\Omega$ •cm water were added. After reacting 1 hour at room temperature, vessels were tightly capped and heated overnight at 120°C Samples were cooled and uncapped, returned to heat at 90°C dried halfway and brought to 12 mL volume with 6 M ultra-high purity HCl. Samples again dried halfway, and brought back to 12 mL volume with 6 M ultra-high purity HCl. Samples were then dried to approximately 1 mL volume, 10 mL of 18.2 M $\Omega$ •cm water was added to each sample, capped tightly, and heated overnight at 80°C Samples were cooled, transferred to acid-washed 50 mL tubes, and brought to a total volume of 20.0 mL with 18.2 MQ cm water. Two Standard Reference Materials which are aluminum alloys (87a and 856a, from the National Institute of Standards & Technology, Gaithersburg, Maryland USA) were analyzed to verify the results obtained with the cookware. These data are reported in Supplemental Table S3. Sample preparation and total metals analyses were carried out at the Metal Isotope Laboratory at Pennsylvania State University. Instrument detection limits for all elements are noted in Table 1.

# 2.5. Analysis of leaching solutions and palm oil digests

Leaching solutions were analyzed by inductively coupled plasma spectrometry (ICP). Palm oil digestions and all leaching solution ICP measurements were conducted by the Service Testing and Research Laboratory (Ohio Agricultural Research and Development Center – Ohio State University) on an Agilent 5110 ICP spectrometer (Agilent Technologies, Santa Clara, CA, USA). Instrument detection limits for all elements are noted in footnotes to the data tables. Blanks and spiked acetic acid samples were used to verify analytical performance. Recovery of lead from spiked acetic acid samples was 104.4% of the expected value. Stability of acetic acid extraction solutions during storage was confirmed by reanalysis of solutions and no differences were observed.

## 3. Results and discussion

Previously it was found that aluminum cookware from eleven countries released variable concentrations of lead and other metals when cooking was simulated with dilute acetic acid solutions (Weidenhamer et al. 2014, 2017). Source materials for this cookware vary from country to country and often from pot to pot, as many types of high-aluminum scrap are melted down together to make this cookware. Lead has been found to contaminate artisanal cookware, with a maximum concentration of 637 ppm and median concentration of 364 ppm by X-ray fluorescence (×RF) for pots from Cameroon (Weidenhamer et al. 2014). A subsequent study of cookware from ten additional countries included cookware items from the Ivory Coast, Kenya and Tanzania. Of 42 items tested, 32 contained less than or equal to 1000 ppm lead by XRF, and the highest amounts of lead found were in four Vietnamese pots, ranging from 3570 to 7070 ppm. The median lead concentration in that study was 615 ppm (Weidenhamer et al. 2017).

Source	Pot	•					Mn,				TI,		
Material	Size	Cd, %	Co, %	Cr, %	Cu, %	Fe, %	%	Ni, %	Pb, %	Sb, %	%	V, %	Zn, %
Blister Foils	5 L	0.0003	0.0004	0.0105	0.453	1.231	0.182	0.0084	0.0242	0.0014	BD <sup>b</sup>	0.0099	0.119
	12 L	0.0005	0.0003	0.0081	0.504	1.211	0.182	0.0072	0.0287	0.0016	BD	0.0095	0.104
	Mean	0.0004	0.0004	0.0093	0.478	1.221	0.182	0.0078	0.0265	0.0015	BD	0.0097	0.111
Cans	5 L	0.0001	0.0009	0.0236	0.223	1.403	0.595	0.0109	0.0320	0.0006	BD	0.0114	0.125
	12 L	0.0001	0.0009	0.0226	0.223	1.361	0.576	0.0106	0.0307	0.0005	BD	0.0110	0.122
	Mean	0.0001	0.0009	0.0231	0.223	1.382	0.585	0.0107	0.0314	0.0005	BD	0.0112	0.124
Condensers	5 L	0.0002	0.0017	0.0334	0.463	3.653	0.179	0.0256	0.0276	0.0013	BD	0.0086	0.314
	12 L	0.0001	0.0016	0.0342	0.386	3.758	0.185	0.0195	0.0186	0.0009	BD	0.0091	0.292
	Mean	0.0002	0.0016	0.0338	0.424	3.705	0.182	0.0225	0.0231	0.0011	BD	0.0088	0.303
Engine Block	5 L	0.0002	0.0004	0.0163	2.565	0.395	0.076	0.0189	0.0152	0.0007	BD	0.0075	0.246
	12 L	0.0002	0.0004	0.0163	2.568	0.389	0.076	0.0189	0.0153	0.0007	BD	0.0072	0.240
	Mean	0.0002	0.0004	0.0163	2.567	0.392	0.076	0.0189	0.0153	0.0007	BD	0.0073	0.243
Small Mixed	5 L	0.0003	0.0006	0.0200	0.986	2.375	0.319	0.0450	0.1735	0.0014	BD	0.0092	3.389
Metals	12 L	0.0003	0.0006	0.0209	0.880	2.368	0.334	0.0422	0.1672	0.0014	BD	0.0096	3.168
	Mean	0.0003	0.0006	0.0204	0.933	2.371	0.327	0.0436	0.1704	0.0014	BD	0.0094	3.279
Mixed Metals	5 L	0.0013	0.0008	0.0329	0.879	2.086	0.422	0.0381	0.1385	0.0041	BD	0.0101	1.574
	12 L	0.0013	0.0008	0.0332	0.879	2.090	0.420	0.0385	0.1380	0.0040	BD	0.0103	1.590
	Mean	0.0013	0.0008	0.0331	0.879	2.088	0.421	0.0383	0.1382	0.0041	BD	0.0102	1.582
Auto	5 L	0.0002	0.0018	0.0477	0.584	2.765	0.438	0.0309	0.1209	0.0025	BD	0.0093	0.793
Radiators	12 L	0.0002	0.0017	0.0480	0.550	2.789	0.444	0.0279	0.1164	0.0022	BD	0.0093	0.775
	Mean	0.0002	0.0017	0.0478	0.567	2.777	0.441	0.0294	0.1187	0.0024	BD	0.0093	0.784

Table 1. Total content<sup>a</sup> of pot materials for selected metals.

<sup>a</sup>Analysis with HCI/HNO<sub>3</sub>/HF digestion followed by ICP-AES or ICP-MS analysis as described.

<sup>b</sup>BD = Not detected; below ICP or ICP-MS detection limit. Detection limits in solution were 0.6, 0.001, 0.005, 0.01, 0.01, 0.005, 0.005, 0.005, 0.006, 0.247, 0.006 and 0.005 μg/L for Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Tl, V and Zn respectively. Based on average sample mass of 80 mg, with dilution to 20.0 mL, the IDLs for Cd, Co, Cr, Pb, Sb, Tl and V (analyzed by ICP-MS) are all below 0.0001%; IDLs for Cu and Fe (analyzed by ICP-AES) are 0.0002%, and for Mn, Ni and Zn (analyzed by ICP-AES) are 0.0001%.

The potential for lead exposures from use of artisanal cookware was conservatively estimated in previous studies based on the lead contained in a 250 mL typical serving size, which is commonly used in dietary studies (Coulibaly et al. 2009). The maximum estimated exposure was more than 1400 micrograms per serving from a Vietnamese pot (Weidenhamer et al. 2017). These results, and the fact that cookware can contribute ongoing lead exposures to entire families, suggest that inexpensive, artisanal aluminum cookware is a significant source of lead poisoning in LMICs. For this reason, we sought to determine whether these exposures could be reduced by segregating and eliminating certain source materials from the production process.

#### 3.1. Estimated lead exposures and cookware source materials

Lead concentrations of the pots, as measured by total digestions of samples from pot handles, were less than 0.031% (310 ppm) for pots made from blister foils, cans, condensers and engine blocks, while pots made from mixed material, small mixed metals and auto radiators had substantially higher lead content, up to 0.170% (1700 ppm, Table 1). These concentrations are in the range found in previous studies by XRF (Weidenhamer et al. 2014, 2017), although not as high as seen in some individual pots in those studies. Pots made from these three materials also contained the highest concentrations of antimony and zinc.

The estimated lead content per serving leached from these pots with 4% acetic acid (Table 2) varied substantially. All pots with detectable lead in the first extraction showed an average decrease of more than 90% in the second extraction. Because of this, two additional extractions were carried out for the smaller, 5 L pots following the palm oil extractions. No further tests were done on pots made from the engine blocks as no lead was detected in the initial extractions. The additional tests showed that the trend toward reduced extractable lead did not continue, with no clear patterns other than variability from one extraction to another (Table 2).

Table 2. Lead leached per 250 mL serving for pots made from various source materials, based on a 2-hour simulated cooking extraction with 4% acetic acid. Values are the results of individual simulated cooking extractions per pot. Because of high variability in initial extractions for lead, two additional extractions were carried out for the smaller pots for all source materials except the engine block, which leached undetectable amounts of lead. the overall mean is based on the four to six simulated cooking extractions were treated as zeroes. Due to the high variation between extractions, standard deviations exceed the mean values and are not reported.

		μg lead per serving						
Source Material	Pot Size	Boil 1	Boil 2	Boil 3	Boil 4	Mean		
Blister Foils	5 L	11.8	0.4	44.6	BD	14.2		
	12 L	8.3	BD	NA	NA	4.2		
				Overall Me	an	10.8		
Cans	5 L	2.5	0.2	5.3	BD	2.0		
	12 L	2.6	0.8	NA	NA	1.7		
				Overall Me	an	1.9		
Condensers	5 L	14.5	0.6	24.7	BD	10.0		
	12 L	17.6	BD	NA	NA	8.8		
				Overall Mean				
Engine Block	5 L	BD	BD	NA	NA	BD		
	12 L	BD	BD	NA	NA	BD		
				Overall Me	BD			
Small Mixed	5 L	139.4	3.0	BD	31.4	43.4		
Metals	12 L	23.0	2.8	NA	NA	12.9		
				Overall Me	33.3			
Mixed Metals	5 L	134.4	9.6	417.8	4.9	141.7		
	12 L	96.0	1.5	NA	NA	48.8		
				Overall Me	110.7			
Auto Radiators	5 L	42.9	16.2	0.8	2.05	15.5		
	12 L	89.8	3.0	NA	NA	46.4		
				Overall Me	an	25.8		

BD = Not detected; below ICP detection limit. NA = Extraction not done. The IDL (instrument detection limit) for lead was 2.1  $\mu$ g/L. Dividing the IDL by 4 will give the IDL on a per serving basis. Blanks (4% acetic acid) contained 3.6  $\mu$ g Pb/L, slightly more than the IDL, and reported values are corrected by this amount. Mean values in some cases are below the reported IDL because of undetectable concentrations in some extracts.

Estimated lead content per serving ranged from non-detectable up to  $417.8 \ \mu g$  per serving. This amount was obtained on the third extraction of the 5 L pot made from mixed metals. The estimated lead content per serving for this pot showed no clear trend, yielding 134.4, 9.6, 417.8, and 4.9  $\mu g$  on four sequential extractions. Variability likely results from uneven erosion of pot surfaces during cooking. Both scanning (Weidenhamer et al. 2014) and transmission electron microscopy (Street et al. 2020) show pronounced and uneven erosion of artisanal cookware surfaces after leaching with dilute acetic acid solutions.

Four pots (from blister foils, cans, condensers and mixed metals) yielded the most lead on the third acetic acid extraction, while two pots (from small mixed metals and auto radiators) yielded the highest amount of lead on the first extraction. Overall, mean estimated exposures per serving for pots made from different materials were 1.9  $\mu$ g for cans, 9.6  $\mu$ g for condensers, 10.8  $\mu$ g for blister foils, 25.8  $\mu$ g for auto radiators, 33.3  $\mu$ g for small mixed metals, and 110.7  $\mu$ g for mixed metals. Only pots made from engine blocks, which averaged 140 ppm in total lead content, did not yield detectable amounts of lead (Table 2).

Results from the extraction of pots while cooking with palm oil were also of concern. Size limitations of the digestion procedure used for these samples (0.25 g palm oil digested, diluted to 50.0 mL), reduced the sensitivity of this analysis compared to the acetic acid extractions. However, while no lead was detectable in pots made from four source materials (detection limit of 0.38 µg per mL palm oil), lead was detected in palm oil simmered in pots made from small mixed metals (0.48 µg/g or 0.44 µg/mL), mixed materials (0.48 µg/g or 0.44 µg/mL), and auto radiators (0.93 µg/g or 0.84 µg/mL). For auto radiators, the lead concentration in palm oil was more than nine times

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greater than in acetic acid on a per mL basis, and for small mixed metals, the lead concentration in palm oil was more than four times greater than in acetic acid on a per mL basis. For the mixed materials pot, lead concentrations in palm oil were roughly equivalent to acetic acid on a per mL basis. As noted above, pots made from these three source materials had the highest total lead content, all measuring above 0.10% (1000 ppm, Table 1). Comparisons are made here based on a per mL basis as palm oil will typically not be consumed in large servings. However, these results show that palm oil can extract as much or more lead than dilute acetic acid. Zhou et al. (2017) reported similar results, finding increased lead migration into vegetable oil compared to other food simulation solutions including dilute acetic acid.

Our objective was not to do a dietary exposure or risk assessment but rather to present data per serving and compare results to health guidance or regulatory standards. The amount of lead released by this cookware is concerning, given general agreement that there is no known threshold for lead toxicity (WHO 2011; CDC 2012; EFSA 2012). Where permissible levels for lead in food have been established, the amounts are exceedingly low (Table S1), as for the US Food and Drug Administration standard (FDA 2006) for lead in candy (0.1 ppm). Similarly, the State of California uses a Maximum Allowable Dose Level (MADL) of  $0.5 \mu g/day$  (California OEHHA 2019) to trigger consumer warnings. These levels would generally be significantly exceeded by regular consumption of food from most of the cookware tested in this study.

#### 3.2. Estimated aluminum exposures

Estimated aluminum release per serving ranged from 152–275 mg depending on source material (Table 3), higher than the mean 125 mg per serving found previously (Weidenhamer et al. 2017). Aluminum concentrations in these single servings exceed by more than seven to almost fourteen times the Joint FAO/WHO Expert Committee on Food Additives (JECFA) PTWI of 2 mg/kg body weight, which equates to 20 mg/day for a 70 kg adult (WHO 2011).

Aluminum is not generally considered hazardous (Tokar et al. 2013), and is found in many overthe-counter antacid preparations. Unlike heavy metals such as lead and cadmium, aluminum does not bioaccumulate in most individuals (WHO 2011), but can be toxic to those with kidney disease and other conditions that result in retention of aluminum in the brain and bones (Tokar et al. 2013). While a definitive link has not been established and evidence is conflicting, ongoing research continues to explore whether aluminum contributes to brain inflammation associated with neurodegenerative disease (Bondy 2016; Martinez et al. 2017). Given the magnitude of potential aluminum exposure from consuming food cooked in these pots, the long-term risks of such exposure need further study.

Although most earlier studies of metals from artisanal aluminum cookware measured metal concentrations in oil or acidic solutions, one large study assessed food contamination from this source in four countries. Jitaru et al. (2019) showed that cooked tomatoes had higher levels of aluminum and lead compared to the same foods cooked in stainless steel cookware. Another study assessed dietary exposure from these same countries and concluded that aluminum and lead exposures exceeded consensus guidance and were a public health concern (Ingenbleek et al. 2020).

#### 3.3. Exposures to other metals

As noted above, no attempt was made to conduct an exposure assessment based on dietary intake, but metal concentrations per serving were compared with applicable regulatory standards or dietary guidelines to evaluate the potential risks from using this cookware. Because there are generally no health-based standards for metals leached from cookware, we considered the most stringent regulatory standards for drinking water and in some cases upper intake levels for various metals from regulatory agencies or other scientific bodies. Metal concentrations estimated per single serving exceeded standards and guidelines for seven of the 12 metals evaluated

Table 3. Aluminum, cadmium, chromium, nickel, thallium and antimony leached per 250 mL serving for pots made from various
source materials, based on a 2-hour simulated cooking extraction with 4% acetic acid. Values are the mean of two successive
simulated cooking extractions per pot. the overall mean is based on the four simulated cooking extractions for each pot type
made from different source materials. for purposes of calculating means, undetectable concentrations were treated as zeroes.

		Al	Cd	Cr	Ni	Sb	TI
Source Material	Pot Size	mg/serving	µg/serving	µg/serving	µg/serving	µg/serving	µg/serving
Blister Foils	5 L	211	BD	20.7	3.96	2.59	1.73
	12 L	163	BD	15.7	2.96	3.80	BD
Overall Mean ± SD		187 ± 34	BD	18.2 ± 4.6	3.46 ± 0.69	3.19 ± 1.80	86 ± 1.72
Cans	5 L	218	BD	58.2	8.08	1.81	26.0
	12 L	222	BD	58.9	9.56	1.36	28.2
Overall Mean ± SD		220 ± 25	BD	58.6 ± 6.2	8.82 ± 0.95	1.59 ± .97	27.1 ± 3.9
Condensers	5 L	188	BD	50.0	10.8	1.42	BD
	12 L	173	BD	37.6	12.9	2.10	BD
Overall Mean ± SD		180 ± 14	BD	43.8 ± 12.7	11.8 ± 2.5	1.76 ± 1.19	BD
Engine Block	5 L	140	BD	21.8	1.50	1.45	BD
	12 L	163	BD	24.6	1.88	1.85	BD
Overall Mean ± SD		152 ± 20	BD	23.2 ± 5.7	1.69 ± 0.31	1.65 ± .84	BD
Small Mixed	5 L	184	1.04	26.8	14.2	2.48	3.94
Metals	12 L	207	0.68	41.7	18.8	6.02	7.20
Overall Mean ± SD		196 ± 17	0.86 ± 1.03	34.3 ± 13.3	16.5 ± 4.3	4.26 ± 2.92	5.57 ± 3.46
Mixed Metals	5 L	195	5.34	45.2	5.81	4.40	8.70
	12 L	192	5.58	43.2	5.22	6.11	9.35
Overall Mean ± SD		194 ± 22	5.46 ± 5.51	44.2 ± 15.6	5.52 ± 1.93	5.26 ± 3.22	9.03 ± 2.44
Auto Radiators	5 L	311	0.91	91.8	14.8	4.76	8.93
	12 L	239	0.64	87.0	12.3	4.16	9.81
Overall Mean $\pm$ SD		275 ± 60	$0.78 \pm 0.92$	89.4 ± 18.2	13.5 ± 2.4	4.46 ± 1.88	9.37 ± 2.50

BD = Not detected; below ICP detection limit. IDLs (instrument detection limits) were 0.69, 4.7, 4.2, 0.58, 4.6 and 8.6 µg/L for Al, Cd, Cr, Ni, Sb and TI respectively. Dividing the IDL by 4 will give the IDL on a per serving basis. Blanks (4% acetic acid) tested below the IDL for all elements except Al (76 µg/L) and Sb (5.5 µg/L), and reported values are corrected by these amounts. The correction was insignificant in the case of Al. Mean values in some cases are below the reported IDLs because of undetectable concentrations in some extracts.

(Table S1). One or more pots leached concentrations of aluminum, antimony, cadmium, chromium, lead, nickel, and/or thallium in excess of these standards and guidelines (Tables 2 and 3). Data for other metals are reported in Table S2. Total cadmium concentrations of all pots ranged from 0.0001–0.0013% (1–13 ppm, Table 1). The maximum amount of cadmium found in 4% acetic acid leachates was 5.5  $\mu$ g per serving or 22.0  $\mu$ g/L in pots made from mixed metals, while four source materials (blister foils, cans, condensers and engine blocks) did not yield detectable amounts (Table 3). Thallium was detected in leachates of all pots except those made from condensers and engine blocks. Chromium, nickel and antimony were detected in all pot leachates, with estimated exposures per serving of up to 89.4, 13.5 and 5.26  $\mu$ g, respectively (Table 3).

Our data show that recycled aluminum cookware leaches multiple metals, although in varying amounts. Interactions between metals are of concern due to common toxicological endpoints for lead, chromium and manganese, for example. However, limited information exists on these relationships among those exposed to multiple metals, but important hypotheses have been raised in the literature. Bauer et al. (2020) found evidence of an association between exposure to a mixture of metals (including manganese, lead, copper and chromium) on IQ, and recommended further research on the joint and interactive effects of these metals on neurotoxicity. Some evidence suggests that certain combinations of metals may be particularly important for predicting adverse reproductive or neurodevelopmental outcomes, and that metal mixtures may cause health effects not seen with individual metals (Henn et al. 2014). Other studies have suggested that mixed metal interactions may have different neurodevelopmental impacts at different exposure periods during pregnancy (Shah-Kulkarni et al. 2020).

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#### 3.4. Source materials and recommendations

The goal of this study was to determine whether excluding certain source materials from artisanal cookware production might reduce potential health risks posed by using this cookware. As noted above, the aluminum concentration in a single serving from all pots tested exceeds the JECFA's PTWI (2 mg/ kg bw/week) for dietary intake. Pots made from auto radiators and mixed metals (both larger and smaller mixed metals) consistently showed relatively high levels of leaching of cadmium, chromium, lead and antimony (Tables 2 and 3). This indicates that these materials should definitely be avoided in cookware manufacturing. However, other materials also yielded pots that leached amounts of metals (in addition to aluminum) that violated various regulatory standards or dietary guidelines (Table S1). Pots made from blister foils exceeded the standards/guidelines for thallium and antimony, and leached a mean 10.8  $\mu$ g lead per serving. Pots made from cans exceeded the standards/guidelines for chromium, nickel, thallium and antimony, and leached a mean 1.9  $\mu$ g lead per serving, with a maximum of 5.3  $\mu$ g lead per serving. Pots made from condensers exceeded the standards/guidelines for chromium, nickel, and antimony, and leached a mean 9.6  $\mu$ g lead per serving, with a maximum of 24.7  $\mu$ g lead per serving. Pots made from engine blocks exceeded the standards/guidelines for antimony.

#### 4. Conclusions

Our results show that artisanal aluminum cookware releases several toxic metals through simulated cooking. The findings that pots made from engine blocks leached undetectable levels of lead, and that certain source materials yielded reduced concentrations of some metals, should be regarded as tentative given limited sample sizes and variability in metal concentrations of these materials. Specifically for engine blocks, given the large size of an engine block, fewer units are needed to make a single pot and the sample cookware cannot be considered representative of all such source materials. Although contaminant metals represent only a small portion of the overall content for each material, they will leach into solution as the pots themselves slowly dissolve.

For this reason, and because all source materials leached at least one metal in addition to aluminum in potentially harmful concentrations, we conclude that selective sourcing of scrap aluminum for recycled cookware does not avoid metal contamination of food. Addressing this public health issue may require a shift to new cookware materials. Additional research to improve the safety of this cookware is urgently needed, as well as the testing of metal content in local foods cooked in artisanal cookware.

#### Abbreviations

AES: Atomic emissionspectroscopy; EU: European Union; ICP: Inductively coupled plasma spectrometry; IDL: Instrument detection limit; LMICs: Low and middle-income countries; MADL: Maximum allowable dose level; MCL: Maximum contaminant level; MS: Massspectrometry; PTWI: Provisionaltolerable weekly intake; SD: Standard deviation; UL: Tolerable upper intakelevel; XRF: X-ray fluorescence

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