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Metal exposures from aluminum cookware: An unrecognized public health risk in developing countries

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HIGHLIGHTS

- Inexpensive aluminum cookware is widely used throughout the developing world.
- Cookware from ten developing countries was tested for the leaching of toxic metals.
- Simulated cooking leached up to 1426 micrograms of lead per 250 mL serving.
- Al, As and Cd were present in some leachates at potentially harmful levels.
- Exposure to metals by corrosion of cookware may pose significant public health risks.

GRAPHICAL ABSTRACT

Cookware made from recycled aluminum is widely used in the developing world.

Lead and other metals leach from cookware during simulated cooking

- Al, As and Cd were released at potentially harmful levels.
- Lead levels ranged from 1-1426 micrograms per 250 mL serving in 15 items.
- Metal exposures may pose significant health risks.

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ABSTRACT

Removing lead from gasoline has resulted in decreases in blood lead levels in most of the world, but blood lead levels remain elevated in low and middle-income countries compared to more developed countries. Several reasons for this difference have been investigated, but few studies have examined the potential contribution from locally-made aluminum cookware. In a previous study of cookware from a single African country, Cameroon, artisanal aluminum cookware that is made from scrap metal released significant quantities of lead. In this study, 42 intact aluminum cookware items from ten developing countries were tested for their potential to release lead and other metals during cooking. Fifteen items released ≥1 microgram of lead per serving (250 mL) when tested by boiling with dilute acetic acid for 2 h. One pot, from Viet Nam, released 33, 1126 and 1426 micrograms per serving in successive tests. Ten samples released >1 microgram of cadmium per serving, and fifteen items released >1 microgram of arsenic per serving. The mean exposure estimate for aluminum was 125 mg per serving, more than six times the World Health Organization's Provisional Tolerable Weekly Intake of 20 mg/day for a 70 kg adult, and 40 of 42 items tested exceeded this level. We conducted preliminary assessments of three

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Abbreviations: ICP, Inductively coupled plasma spectrometry; MADL, Maximum allowable dose level; PTTIL, Provisional tolerable total intake level; PTWI, Provisional tolerable weekly intake; UL, Tolerable upper intake level; XRF, X-ray fluorescence.

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Cookware
Developing countries

potential methods to reduce metal leaching from this cookware. Coating the cookware reduced aluminum exposure per serving by >98%, and similar reductions were seen for other metals as well. Potential exposure to metals by corrosion during cooking may pose a significant and largely unrecognized public health risk which deserves urgent attention.

1. Introduction

A wide variety of aluminum cookware and utensils are used throughout the world. Much of this cookware is locally made, uncoated and not anodized. Previous investigations indicated that in Cameroon, some of this cookware is made in informal shops by casting liquid aluminum melted from a collection of scrap metal (Osborn, 2009; Weidenhamer et al., 2014). Other studies have reported on the potential leaching of metals from aluminum cookware in India, Egypt, China, Saudi Arabia, Syria, and Bangladesh (Al Juhaiman, 2012; Bergkvist et al., 2010; Mohammad et al., 2011). In this study, we evaluate the potential for aluminum cookware from additional countries to contribute to metal exposures through normal cooking.

Despite the removal of lead additives from gasoline more than a decade ago in all but a small number of countries, numerous recent reports document the widespread persistence of elevated blood lead levels in low and middle-income countries (El-Desoky et al., 2013; Kalra et al., 2013; Kapitsinou et al., 2015; Li et al., 2014; Naicker et al., 2013; Swaddiwudhipong et al., 2013; Tuakuila et al., 2013; Xie et al., 2013). There is no safe level of lead exposure (CDC, 2012; Lanphear et al., 2005; Wigle and Lanphear, 2005). The toxic effects of lead are well known. Lead exposures are linked to learning disabilities, attention-related behaviors, deficits in intellectual development, high blood pressure and cardiovascular disease. The estimated global toll from lead poisoning is 674,000 premature deaths annually (Lim et al., 2012) and economic costs approaching $1 trillion (Attina and Trasande, 2013). The persistence of elevated blood lead levels in much of the world is therefore of great concern for public health and economic development.

Lead consumption is growing rapidly around the world primarily for the production of lead batteries. Emissions from the manufacturing and recycling of these batteries have been documented to contribute to lead exposures in populations surrounding these plants (Gottesfeld and Pokhrel, 2011). In addition, a wide variety of consumer products contain lead additives including lead paint that is commonly used in at least 40 countries around the world (Clark et al., 2009, 2015; Gottesfeld et al., 2013, 2014; Kumar and Gottesfeld, 2008; Occupational Knowledge International, 2016). Other consumer products that contain lead and are often unregulated include plastics, lipsticks, jewelry, solder, brass, and ceramic glazes (e.g. Gilmore et al., 2013; Weidenhamer and Clement, 2007; Zhao et al., 2016).

Locally-made aluminum cookware is a potential source of lead exposure that has largely been overlooked. This cookware is widely used throughout the developing world (Al Juhaiman, 2012; Al Zubaidy et al., 2011; Bergkvist et al., 2010; Osborn, 2009). The potential for metals to leach from this type of cookware has been studied previously, but typically with a focus on potential hazards of aluminum (e.g. Al Juhaiman, 2010, 2012, 2016; Inoue et al., 1988; Karbouj et al., 2009; Mohammad et al., 2011). Previously we reported (Weidenhamer et al., 2014) that cookware items manufactured by local artisans in Cameroon pose a significant risk from the leaching of multiple metals during cooking. Investigations in that country revealed that scrap metal was the primary source material for this cookware including waste engine parts, vehicle radiators, lead batteries, computer parts, and other materials. Potential lead exposures from cooking were estimated to be as high as 260 μg per serving, indicating a serious potential health hazard. In addition, simulated cooking in the laboratory released significant concentrations of other metals. Up to 15.6 μg Cd was released per serving, and all items tested released aluminum in amounts per serving which exceeded the provisional tolerable total intake level (PTWI) for aluminum of 140 mg/person/week or 20 mg/person/day for a 70 kg adult (WHO, 2011b). Because of the widespread use of inexpensive aluminum cookware in many countries (e.g. Osborn, 2009), determining the extent of metal exposures from this source and evaluating potential solutions is an urgent need.

Our objective in this investigation was to explore how widespread the potential health risks posed by aluminum cookware may be in the developing world. We obtained and tested 42 intact aluminum cookware items from ten developing countries in Asia, Africa and Central America for their potential to release lead and other metals during cooking. We also conducted preliminary studies on possible means to reduce corrosion of the cookware and thereby reduce metal exposures.

2. Methodology

2.1. Sample collection

Forty-two cookware samples were collected from ten countries (Bangladesh, Guatemala, India, Indonesia, Ivory Coast, Kenya, Nepal, the Philippines, Tanzania and Viet Nam). The majority were new items, and varied in appearance (Fig. 1). All were locally manufactured and available for purchase in the country. Half (21 of 42 samples) from five countries (Bangladesh, India, Indonesia, the Philippines, and Viet Nam) indicated brands on labels or commercial logos imprinted on the pots when received for analysis.

2.2. X-ray fluorescence (XRF) analysis

XRF screening of cookware samples was conducted using a Niton XL3t GOLDD XRF spectrometer (Thermo Fisher Scientific, Billerica, MA). Prior to analysis, an internal system calibration was performed. Samples were analyzed for metals in “general metals” mode using an aluminum alloy (grade 6061) reference material.

2.3. Leaching tests

There is no standardized method to replicate normal cooking for the measurement of metals leaching from aluminum cookware. Here, we used a 2 h boiling extraction with dilute acetic acid (4% vol/vol), an extraction which simulates cooking with weakly acidic foods such as tomato sauce (Al Zubaidy et al., 2011; Inoue et al., 1988; Mohammad et al., 2011; Weidenhamer et al., 2014).

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.

Samples were leached with 4% acetic acid (vol/vol) for 2 h in boiling solutions. Cookware was filled to within 1 cm of the rim, and brought to a gentle boil on a hotplate. Pots that had curved rather than flat bottoms were heated over natural gas burners. For pots which did not have lids, glass plates were placed on top of the pots to retard evaporation. 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 directly to 50 mL polyethylene test tubes and stored under refrigeration until analysis.

The test procedure was repeated for fifteen of the pots to determine the consistency of metal concentrations released by corrosion during

Fig. 1. Photos of representative cookware items (clockwise from upper left) from (a) Ivory Coast; (b) Philippines (with inset showing pot after boiling with dilute acetic acid); (c) Viet Nam; and (d) Bangladesh.

ongoing usage. Pots for these tests were selected based on the concentrations of metals, especially Pb, released during the first extraction. Ten of the pots selected to retest released an estimated 5 or more μg Pb per serving in the initial extraction, while five pots released no detectable Pb initially (Table 1).

2.4. Fluoropolymer coating

Four cookware items which released arsenic, cadmium and/or lead in initial testing were chosen for coating at a commercial cookware manufacturer with a fluoropolymer finish (Xylan®) to determine the impact of this treatment on the concentration of metals leaching from the pots. A cabinet blaster and dust collector was used to mechanically clean the substrate. This removed surface oils and roughened the surface for increased coat adhesion. The cookware coating was applied with a compressed air pressurized spray gun, followed by oven curing at 800 °C. The curing process hardened and smoothed the coating.

2.5. Evaluation of potential corrosion inhibitor treatments

Karbouj et al. (2009) reported that boiling water in aluminum cookware prior to cooking can reduce leaching of aluminum substantially. Recently, Al Juhaiman (2016) reported that curcumin, a component of the spice turmeric, could reduce leaching of aluminum from cookware into various vegetable and meat solutions by 60 to 80%. These treatments were evaluated on four pots which had released >90 mg Al per serving in previous tests. Pots were held at 95 °C in deionized water for 5 h (Karbouj et al., 2009) prior to extraction two subsequent times with boiling 4% acetic acid as described above. Following these extractions, the pots were extracted a third time with boiling 4% acetic acid which contained 100 mg/L curcumin (Al Juhaiman, 2016).

2.6. ICP methods

Leaching solutions were analyzed by inductively coupled plasma spectrometry (ICP). 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. In addition, stability of extraction solutions during storage was confirmed by reanalysis of solutions and no differences were observed.

3. Results

3.1. Leaching of aluminum, arsenic, cadmium and lead from cookware

Estimated exposures for all metals were based on the 250 mL typical serving size, which has been used in other dietary studies in developing countries (Coulibaly and Galibois, 2009) and was used in our previous study of Cameroonian cookware.

3.2. Aluminum

The estimated mean exposure from the cookware (averaged for all items and extractions) is 125 mg per serving, more than six times greater than the World Health Organization (WHO) PTWI of 20 mg day^{-1} for a 70 kg adult (WHO, 2011b). Forty (95%) of the forty-two items tested exceeded this level (Table 1). In one case (Viet Nam C), the aluminum released was only 7 mg per serving during a first extraction but, increased significantly with repeated testing to 71 and 111 mg per serving on subsequent extractions. For the fifteen pots tested by repeat boiling, ten of fifteen pots had higher aluminum concentrations on the third boil. The mean difference from first to third boil was a net increase of 42 mg per serving. Only two cookware items (Indonesia B and Kenya D) had <94% aluminum content as measured by XRF (Table 2).

3.3. Arsenic

Twenty-three (55%) items yielded detectable levels of arsenic, with a maximum level of 10 μg per serving in one pot from the Philippines. All four cookware items from Bangladesh, a country with serious arsenic poisoning issues due to contaminated groundwater (Uddin and Huda, 2011; WHO, 2011a), were found to release arsenic up to a maximum of 6 μg per serving (Table 1).

3.4. Cadmium

Thirteen samples (31%) released >1 μg per serving in one or more extractions, with a maximum estimated exposure of 7.5 μg per serving from a pot from Bangladesh. A total of twenty-four cookware items yielded detectable levels of cadmium. For the pots tested by repeat boiling, cadmium concentrations generally decreased (e.g. Indonesia B, Ivory Coast A and B; Table 1), but there were exceptions (e.g. Nepal D and Philippines E).

Table 1
Aluminum, arsenic, cadmium and lead leached per 250 mL serving based on a 2 h simulated cooking extraction with 4% acetic acid.

Country of origin	Item	Boil	Al mg/serving	As μg/serving	Cd μg/serving	Pb μg/serving
Bangladesh	A	1	9	6	ND	2.0
	B	1	219	5	7.5	ND
		2	343	4	4.5	ND
		3	216	4	5.7	ND
	C	1	218	5	ND	ND
		2	177	3	ND	ND
		3	247	6	ND	ND
	D	1	154	3	ND	ND
		2	172	3	ND	ND
		3	198	3	ND	ND
Guatemala	A	1	175	3	1.1	1.3
	B	1	128	ND	0.9	ND
	C	1	86	ND	1.1	1.7
	D	1	103	ND	0.9	ND
India	A	1	46	ND	ND	ND
	B	1	58	5	0.7	ND
	C	1	54	ND	ND	ND
Indonesia	A	1	102	8	0.3	ND
	B	1	159	ND	4.7	177
		2	151	6	1.4	19
		3	170	6	1.0	ND
	C	1	92	ND	ND	ND
Ivory Coast	A	1	131	5	7.5	19
		2	117	ND	ND	ND
		3	240	9	0.3	29
	B	1	96	ND	2.5	ND
		2	138	ND	ND	ND
		3	148	3	ND	ND
	C	1	87	3	1.8	ND
	D	1	76	ND	1.5	ND
Kenya	A	1	54	ND	0.2	ND
	B	1	38	ND	ND	ND
	C	1	37	ND	ND	ND
	D	1	113	ND	ND	ND
Nepal	A	1	70	ND	ND	ND
	B	1	67	ND	ND	ND
	C	1	69	ND	ND	ND
	D	1	71	5	0.6	11
		2	82	ND	ND	ND
		3	54	7	1.3	ND
	E	1	97	ND	ND	ND
Philippines	A	1	104	ND	0.4	15
		2	328	3	ND	32
		3	62	6	ND	ND
	B	1	127	ND	0.2	ND
	C	1	172	ND	ND	ND
	D	1	233	10	ND	19
		2	249	4	ND	ND
		3	212	7	0.5	ND
	E	1	213	7	0.1	13
		2	211	3	ND	ND
		3	126	9	2.5	406
		4	302	4	ND	ND
		5	325	5	ND	ND
Tanzania	A	1	34	ND	1.5	5.3
		2	53	ND	0.2	ND
		3	220	8	0.8	4.8
	B	1	45	ND	3.0	ND
		2	89	ND	ND	ND
		3	81	ND	ND	ND
	C	1	58	ND	1.5	ND
Viet Nam	A	1	ND	5	0.1	1.0
	B	1	45	5	ND	ND
	C	1	7	5	ND	32
		2	71	ND	ND	18
		3	111	ND	ND	ND
	D	1	33	6	0.1	33
		2	161	3	0.3	1126
		3	225	4	0.3	1426
	E	1	56	8	0.4	20
		2	74	ND	ND	5.9
		3	95	3	ND	14
	F	1	29	4	ND	1.5
	G	1	60	6	ND	ND

ND = Not detected; below ICP detection limit. Method quantitation limits (MQLs) were 30, 9, 0.4 and 4.0 μg/L for Al, As, Cd and Pb respectively.

3.5. Lead

Fifteen (36%) samples released >1 μg per serving on the first boil (Table 1), with a maximum estimated exposure of 177 μg per serving from a pot from Indonesia. For pots tested by repeated boiling, results were quite variable. Lead release from Indonesia B decreased to undetectable levels by the third extraction. For Ivory Coast A, lead levels decreased from 19 μg per serving to undetectable levels on the second extraction, before yielding 29 μg per serving on the third repeated boil. Additional pots showed similar erratic results for lead, as with Philippines E, which had undetectable levels for the second, fourth and fifth extractions, but released 13 μg per serving on the first extraction and 406 μg per serving on the third extraction. Of serious concern were the results for Viet Nam D, which yielded 33 μg per serving on the first extraction, increasing to 1126 and 1426 μg per serving on subsequent extractions.

Thirty-six (86%) of the cookware items showed detectable lead by X-ray fluorescence (Table 2) and eleven (26%) items showed lead content >0.100% (1000 ppm). The highest concentrations were found in Viet Nam items B and D, at 0.707% (7070 ppm) and 0.700% (7000 ppm) respectively. As noted above, item Viet Nam D released the highest amount of lead per serving observing in this or our previous study (Weidenhamer et al., 2014), but item Viet Nam B, which was extracted

Table 2
Summary of XRF analyses of cookware samples.

Country of origin	Item	Al, %	Pb, %
Bangladesh	A	98.6	0.028
	B	95.4	0.108
	C	94.8	0.157
	D	94.8	0.132
Guatemala	A	94.2	0.067
	B	94.7	0.046
	C	95.0	0.086
	D	94.0	0.100
India	A	99.3	<LOD
	B	96.8	0.262
	C	97.8	0.063
Indonesia	A	99.0	<LOD
	B	89.8	0.269
	C	98.5	<LOD
Ivory Coast	A	96.5	0.062
	B	95.9	0.024
	C	97.8	0.085
	D	98.2	0.057
Kenya	A	97.9	0.057
	B	98.7	0.038
	C	99.7	<LOD
	D	85.5	0.061
Nepal	A	98.3	0.053
	B	98.6	0.034
	C	97.9	0.086
	D	98.3	0.037
	E	97.3	0.302
Philippines	A	97.0	0.086
	B	97.2	0.027
	C	97.2	0.041
	D	96.6	0.066
	E	95.9	0.072
Tanzania	A	99.2	<LOD
	B	99.4	<LOD
	C	98.3	0.062
Viet Nam	A	97.9	0.052
	B	96.2	0.707
	C	99.3	<LOD
	D	96.7	0.700
	E	97.2	0.357
	F	96.4	0.475
	G	99.4	<LOD

<LOD = Below limit of detection. The MQL for aluminum was <0.5%, and for lead below 0.02%.

only once, did not release detectable amounts of lead. However, it seems likely that any lead present in these items will be released as corrosion continues to occur through cooking.

3.6. Other metals

Concentrations of other metals were also variable from extraction to extraction but these were generally below health standards where those have been identified (Supplemental Table 1). For copper, which is both an essential micronutrient and toxic at high concentrations, a Tolerable Upper Intake Level (UL) has been proposed, ranging from 1 mg day^{-1} for children 1–3 years in age to 10 mg day^{-1} for adults (Institute of Medicine, 2001). In two cases (Philippines A, second boil, and Philippines D, fifth boil) the estimated copper exposures of 1267 μg per serving and 1032 μg per serving, respectively exceeded the 1 mg day^{-1} limit for young children.

3.7. Effect of fluoropolymer coating

We evaluated the potential of fluoropolymer coatings to reduce corrosion and metal exposures on a small sample of four cookware items. The average reduction in aluminum exposure per serving was >98%, from 198 mg per serving to 3 mg per serving, and substantial reductions were seen for other metals as well (Table 3). Arsenic and cadmium were not detected in extractions of any of the coated pots. Lead levels following the coating ranged from non-detectable to 19 μg per serving. Although the highest lead concentration was detected in item Viet Nam D following coating, this represented a reduction of >98% from the final extraction prior to coating the pot.

3.8. Effect of potential corrosion inhibitor treatments

Based on a limited sample of four pots, heating cookware for 5 h in near-boiling water as proposed by Karbouj et al. (2009) resulted in a 76% reduction of estimated Al exposure from the amount leached immediately prior to treatment. However, the effect diminished following a second leaching with acetic acid, yielding a mean 31% reduction of estimated Al exposure. The effect diminished further in a final leaching with acetic acid despite the addition of curcumin, as estimated Al exposures showed only a 5% reduction from the amounts leached prior to treatment with near-boiling water. The impact of these treatments on As, Cd and Pb are much less clear as levels variably rose and fell with subsequent testing. For all four pots, the estimated Al exposure on the final extraction with curcumin was more than double the 20 mg daily limit corresponding to the PTWI for a 70 kg adult established by the WHO (2011b).

Table 3
Effect of Xylan® coating on aluminum, arsenic, cadmium and lead corrosion during simulated cooking with 4% acetic acid.

Country of origin and item	Coating	Boil	Al mg/serving	As μg/serving	Cd μg/serving	Pb μg/serving
Bangladesh B	No	1	219	5	8	ND
		2	343	4	4	ND
		3	216	4	6	ND
	Yes	1	2	ND	ND	1
		2	2	ND	ND	ND
Ivory Coast A	No	1	131	5	8	19
		2	117	ND	ND	ND
		3	240	9	0.3	29
	Yes	1	1	ND	ND	3
		2	1	ND	ND	ND
Philippines D	No	1	233	10	ND	19
		2	249	4	ND	ND
		3	212	7	0.5	ND
	Yes	1	8	ND	ND	ND
		2	10	ND	ND	ND
Viet Nam D	No	1	33	6	0.1	33
		2	161	3	0.3	1126
		3	225	4	0.3	1426
	Yes	1	1	ND	ND	19
		2	2	ND	ND	8

ND = Not detected; below ICP detection limit. MQLs were 30, 9, 0.4 and 4.0 μg/L for Al, As, Cd and Pb respectively.

4. Discussion

Previously we demonstrated that artisanal aluminum cookware from one West African nation, Cameroon, released as much as 260 μg Pb per serving in extractions designed to simulate cooking with mildly acidic solutions (Weidenhamer et al., 2014). While lead was present as a minor component in the cookware (<1000 ppm by XRF), rapid corrosion of the aluminum in dilute acetic acid solutions liberated lead and other metals present in the aluminum alloy. Inexpensive aluminum cookware, often locally made, is widely used throughout the developing world. Other studies have documented the presence of lead in similar concentrations in cookware from four other countries in addition to items from Bangladesh and India, which are also examined in this study (Al Juhaiman, 2012; Bergkvist et al., 2010; Mohammad et al., 2011). On this basis, we suggested that our results might indicate a much larger global problem with inexpensive, non-anodized aluminum cookware as a heretofore unrecognized source of lead poisoning. The present study supports this hypothesis, and suggests that exposure to metals including lead through corrosion of such cookware is a major public health problem.

One limitation of our study design is that we estimated exposures based on cooking with a slightly acidic liquid. We have made no attempt to estimate the leaching potential during cooking of solid food, or less acidic or salty vegetable or meat broths. It is possible that solid foods and less acidic cooking broths may leach lower concentrations of metals from this type of cookware during a similar cooking duration. In addition, in many countries cookware is also used to store food cooked in batches that may be reheated to serve multiple meals over several days. We have not evaluated the potential leaching from longer-term contact with this cookware. Our tests were of intact, new cookware items, so it must be recognized that the corrosion of damaged or older cookware may show different patterns. Solid fuels used for cooking in many countries may influence cooking temperatures and thereby change rates of corrosion. In addition, the actual absorption of metals present will depend on the amount of food consumed, the efficiency of uptake of the various metals in the gastrointestinal tract, and the acidity of the cooking solution.

4.1. Estimated heavy metal exposures and health implications

We previously found that significant levels of cadmium and lead were extracted along with aluminum from a number of the cookware items (Weidenhamer et al., 2014). In our previous study, 22 of 22 cookware pieces tested released 6 to as much as 248 μg lead per serving, with a median of 97.0 μg per serving. Here, fifteen of the 42 items tested released ≥1 μg lead per serving, but maximum exposures were as high as 1426 μg lead per serving (Viet Nam D, Table 1). Hazards were not confined to one geographic region, as at least one cookware item from eight of the ten countries sampled released >1 μg lead per serving (Table 1).

As it is generally accepted that there is no known threshold for lead toxicity few regulatory authorities have attempted to set maximum levels of intake (CDC, 2012; EFSA, 2012; WHO, 2011b). Where dose-based levels have been articulated, they are very low. California had set a Maximum Allowable Dose Level (MADL) for lead of 0.5 μg day^{-1} (California OEHHA, 2016). Our results confirm that inexpensive aluminum cookware can be a significant source of lead exposures for those who use it.

The toxic effects of cadmium are also well known. The bones (Järup and Alfven, 2004; Satarug et al., 2010) and kidneys (Noonan et al., 2002; Satarug et al., 2005, 2010) are the primary targets of chronic exposure, and it is classified as carcinogenic (Group 1) by the International Agency for Research on Cancer (IARC, 2016; Joseph, 2009). Certain subpopulations such as diabetics may be more susceptible to toxic effects of cadmium based on epidemiologic studies (Nordberg et al., 2009). Food and tobacco smoke are generally regarded as the major sources of cadmium exposure (Satarug and Moore, 2004), and additional sources of exposure are of concern due to the fact that cadmium bioaccumulates in the body. In our previous study of cookware from Cameroon, we found that 23% of the items tested exceeded the MADL of 4.1 μg day^{-1} set by the state of California (California OEHHA, 2016), with a maximum estimated exposure of 15.6 μg (Weidenhamer et al., 2014). Here, 24 of the 42 cookware items tested (57%) yielded detectable levels of cadmium, though only three (7%; Bangladesh A, Indonesia B, and Ivory Coast A) exceeded an exposure of 4.1 μg on one or more extractions.

The toxic effects of arsenic include skin lesions, cancer and neurotoxicity (WHO, 2011a). In the United States, the standard for arsenic in drinking water was reduced to 10 μg L^{-1} in 2001. More than half of the cookware items tested (23 or 42 items, or 55%) released detectable levels of arsenic, ranging from 3 to 10 μg per 250 mL serving. On a per liter basis, these values exceed this drinking water standard, indicating potentially hazardous exposures. As noted above, all four cookware items from Bangladesh, a country where many wells produce water containing >50 μg L^{-1} of arsenic (Smith et al., 2000; Uddin and Huda, 2011), released detectable levels of arsenic which would add to exposures from drinking water.

NSF International (2011) has a third-party certification standard (NSF Protocol 390) for cookware manufacturers, with extraction testing limits for metals including arsenic (1 μg L^{-1}), cadmium (0.2 μg L^{-1}), chromium (1 μg L^{-1}), and lead (1 μg L^{-1}). All of these criteria are exceeded by many of the cookware items tested, in some cases by 2–3 orders of magnitude (Table 1 and Supplemental Table 1).

In addition to toxic effects of individual metals, the potential for harm from chronic exposures to multiple toxic metals, combined with exposures from other sources, should not be overlooked. The variable composition of this cookware, and the multiple metals identified from the leaching procedure, indicate the high probability of multiple metal exposures in significant concentrations from this cookware. Occasional samples were found to release toxic metals other than those reported, such as thallium, which was released by two pots in one or more extractions in amounts of 14 to 71 μg per serving (Indonesia B and Philippines E, data not shown). The health impacts of multiple metal exposures are a subject of ongoing research, but studies have identified mixtures of metals that appear to act synergistically (Cedergreen, 2014), or are additive in their effects (Vijver et al., 2011).

Our results indicate that cookware from every country tested yielded hazardous exposures to one or more metals. Based on the low number of samples from individual countries, it is not possible to conclude that specific hazards such as lead contamination are absent from cookware from those countries. In addition, most of the cookware tested was acquired in large urban cities and may not be representative of cookware from smaller cities and more rural areas. It seems reasonable to conclude that the risks of toxic metal exposure from this inexpensive aluminum cookware is geographically widespread.

There were no consistent differences in metal release by branded compared to unbranded pots in this study. For aluminum, all three of the pots which released >300 mg aluminum per serving (Bangladesh B, Philippines A and E) were branded items. Arsenic was detected in leachates of 16 of 21 branded pots (76%), while only 7 of 21 unbranded pots (33%) yielded detectable amounts. Cadmium was more frequently detected in unbranded (52%) than branded pots (9.5%), and lead exposures exceeded 1 microgram per serving for 9 of 21 branded pots (43%) and 6 of 21 unbranded pots (29%). It seems likely that the quality of the source material used by both artisans and larger manufacturers is the determining factor in the amount of toxic metals present and/or released from cookware.

The amounts of metals released by sequential extractions of the cookware were highly variable. For example, as previously noted, sample Viet Nam D released 33, 1126 and 1426 micrograms of lead per serving on three subsequent extractions. Sample Philippines D, which was extracted five times, released 13 micrograms of lead per serving on the first extraction, undetectable amounts on the second, fourth and fifth extractions, and 406 micrograms of lead per serving on the third extraction. Examination of the data (Table 1 and Supplemental Table 1) show that this variability is seen for other metals (e.g. Cr, Cu and Mn) and samples. These data imply that the cookware is not homogeneous in its metal composition.

When aluminum is extracted from mined ore, the product is typically >99% pure (Pohl, 2011). Because the cookware examined in this study has an average concentration of 96.5% aluminum, it is likely derived from recycled materials. The major source of the other metals in these pots may come from discarded items such as electronic components that are collected and used for this purpose. Aluminum itself melts at the relatively low temperature of 660 °C. Given the different melting points (MP) of all of these metals, Al-alloys, and other alloys found in waste (e.g. $\mathrm{MP_{Pb}} = 163$ °C, $\mathrm{MP_{Pb\text{-}Ti\ Alloy}} = 725$ °C, $\mathrm{MP_{Cu}} = 1083$ °C, $\mathrm{MP_{Brass\ (Cu\text{-}Zn)\ Alloy}} = 930$ °C; $\mathrm{MP_{Cr}} = 1860$ °C, $\mathrm{MP_{Mn}} = 1244$ °C), as well as varying densities of these metals and alloys, it is not surprising that the resulting pots are heterogeneous in their composition since some of the metals and alloys probably never melted or completely mixed under conditions needed to melt the aluminum (American Elements, 2016). Thus, as the pots corroded, portions of the pots that were higher in particular metals or alloys were exposed so that the amounts extracted were not constant.

4.2. Estimated aluminum exposures and health implications

Aluminum ingestion is usually regarded as nonhazardous, based on typical food-borne exposure levels (Tokar et al., 2013; Stahl et al., 2011). Ingestion of food is the primary route of exposure to aluminum for most people (WHO, 1997; Stahl et al., 2011), and absorbed aluminum is rapidly distributed in the body via circulation. It passes through the blood brain barrier at all ages and can reach the fetus via maternal circulation. It is primarily eliminated via urine. The absorption of aluminum in the gastrointestinal tract is low, 0.2–1% (Priest et al., 1998; WHO, 1997), and it does not bioaccumulate in most people (WHO, 1997). Kidney disease and other conditions have led to brain and skeletal retention of aluminum, leading to toxicity (Tokar et al., 2013).

High ingested levels of aluminum have resulted in acute toxicity, interfering with nutrient absorption (e.g., calcium, iron) and can cause bone deficiencies, osteomalacia and aluminum osteodystrophy (Tokar et al., 2013). Symptoms of acute aluminum toxicity may include multiple fractures, damage the hematopoietic system (Medscape, 2016). In rare cases resulting from high level exposure it is neurotoxic, causing neurodegenerative diseases (Chin-Chan et al., 2015) that may manifest as speech disorders followed by dementia, convulsions and myoclonus (Tokar et al., 2013). While evidence is not consistent and has been the subject of much debate, aluminum is suspected of contributing to neurodegenerative diseases including Alzheimer's disease and experimental evidence suggests that aluminum can contribute to brain inflammation (Bondy, 2016; Chin-Chan et al., 2015; Shen et al., 2014).

Our results show that 40 of 42 cookware items released amounts of aluminum exceeding the WHO PTWI of 20 mg/day for a 70 kg adult, with a mean estimated exposure of 125 mg/serving and maximum exposure of as much as 343 mg/serving (Table 1). This suggests that the potential for harm from chronic aluminum exposures from this type of cookware should not be overlooked. Vulnerable individuals with kidney disease would be at particular risk from the chronic exposures likely from the regular use of such cookware.

4.3. Prevention of corrosion as a strategy to reduce exposures to lead and other metals

One way to minimize harmful metal exposures from the use of inexpensive aluminum cookware would be to regulate or screen the source materials used in its production. Efforts to prohibit the use of scrap metal for this purpose would be difficult to enforce and may result in shifting smaller manufacturers to operate in more clandestine locations. Alternatively, scrap metal could be field tested with XRF analyzers to screen out objects with high levels of contaminant metals. These approaches could potentially reduce exposure to heavy metals such as lead and cadmium, but would not address potential health issues related to aluminum exposure.

Three other potential approaches to reducing overall corrosion include: (a) boiling or near-boiling water pretreatment as proposed by Karbouj et al. (2009); (b) addition of curcumin, a natural product found in the spice turmeric, as a corrosion inhibitor as proposed by Al Juhaiman, 2016; and (c) coating of cookware with a fluoropolymer finish. The first two have been tested with a focus on aluminum in limited studies and we conducted the latter test for the first time that we are aware.

Based on the four samples of cookware coated with the Xylan fluoropolymer finish, extracted aluminum concentrations decreased by approximately 98% along with levels of lead and other heavy metals (Table 3). Treatment of cookware that had previously been extracted with boiling acetic acid solutions with near boiling water appeared to reduce aluminum corrosion on the first extraction following treatment, but the effect decreased with the second extraction following treatment (Table 4). Addition of curcumin to serve as a corrosion inhibitor for the third extraction following treatment did not reduce corrosion, as the amount of aluminum released was higher for each of the four pots in the curcumin extraction (Table 4).

The results from the fluoropolymer treatment of artisanal aluminum cookware suggest that coating these inexpensive aluminum cookware may be effective at substantially reducing the hazardous levels of metals observed leaching into the solution and exposures from normal cooking practices. In addition to Xylan coating, other fluoropolymers, enamel coatings, or anodization treatments could be effective in minimizing the leaching of heavy metals from cookware. However, all of these approaches necessitate an increase in the capital equipment used by small producers and would increase the cost to consumers for this cookware. There are also likely implications on sales, as well as cultural concerns with switching cooking surfaces and product appearance. In many cultures, selection of cookware is closely linked to cooking practices that may not be readily transferred to coated or anodized aluminum cookware. In West Africa for example, artisanal aluminum cookware is ubiquitous in homes, food stalls, and institutional applications (Osborn, 2009). The lack of cultural acceptance to alter the product in this way may preclude a simple fix.

Table 4
Impact of pre-treatment in near boiling water and curcumin on corrosion during simulated cooking with 4% acetic acid (Boil 0 = last previous boil reported in Table 1; AB1 and AB2 = first and second boils after pre-treatment in near boiling water; C3 = final boil with addition of 100 mg/L curcumin).

Country of origin and item	Boil	Al mg/serving	As μg/serving	Cd μg/serving	Pb μg/serving
Indonesia C	0	92	ND	ND	ND
	AB1	10	ND	ND	ND
	AB2	36	ND	ND	ND
	C3	72	ND	ND	ND
Ivory Coast B	0	148	3.0	ND	ND
	AB1	75	ND	ND	ND
	AB2	141	8.6	ND	ND
	C3	167	ND	0.5	ND
Philippines C	0	172	ND	ND	ND
	AB1	31	ND	ND	94
	AB2	151	11	ND	ND
	C3	206	ND	0.2	31
Viet Nam C	0	111	ND	ND	ND
	AB1	11	ND	0.6	49
	AB2	34	12	ND	27
	C3	53	15	ND	111
Mean, Al corrosion	0	131			
	AB1	32			
	AB2	90			
	C3	124			

ND = Not detected; below ICP detection limit. MQLs were 30, 9, 0.4 and 4.0 μg/L for Al, As, Cd and Pb respectively.

It is estimated that three pots can be coated with Xylan for a material cost of about $1.00 (U.S.). Although the material cost of the Xylan or other coating may be reasonably absorbed into the cost of the finished product, the equipment to appropriately clean and spray apply this type of material is estimated to be approximately $10,000–20,000 (U.S.). Therefore it would be impractical for most individual manufacturing shops to perform this task but instead would require developing a new specialty business to handle a large volume of cookware from multiple artisanal producers to provide the necessary scale to reduce costs. In large cities where these producers are concentrated, it may be feasible to establish a cooperative or central facility to apply a coating for a fixed fee to new cookware from a large number of producers.

Even if these barriers could be overcome, there are additional concerns about the safety of fluoropolymer based coatings to workers applying these materials as well as to end users. In the course of spray applying Xylan, workers are potentially exposed to significant levels of *N*-methyl-2-pyrrolidone, a solvent that is readily absorbed and associated with teratogenic effects and reduced fetal weight based on multiple animal studies (European Commission, 2011). These and other concerns resulted in a conclusion that cosmetic products containing a maximum 5% concentration of *N*-methyl-2-pyrrolidone were not safe for consumers (European Commission, 2011). The application of fluoropolymers has also been associated with a respiratory illness with flu-like symptoms similar to polymer fume fever (Hays and Spiller, 2014). Significant costs would be necessary to properly ventilate areas and the equipment used for the spray application.

The environmental and health risks of perfluorochemicals remain an area of active and ongoing research (Agency for Toxic Substances and Disease Registry, ATSDR, 2015a; Begley et al., 2005; Blum et al., 2015; Lindstrom et al., 2011; Shoeib et al., 2016; Trudel et al., 2008). Generally as a class fluoropolymers are considered safe for end users although they are known to persist in the human body and in the environment (Kelly et al., 2009). Direct exposures to perfluorochemicals from cookware are typically considered to be low in comparison to exposures from drinking water and contaminated food (ATSDR, 2015b). Some of this class of chemicals are suspect carcinogens but evidence is sparse despite their widespread use.

Our study made no attempt to evaluate the potential for fluoropolymers to be released from Xylan coatings during normal cooking, and we are unaware of any studies that have specifically examined the release of perfluorinated chemicals from Xylan-coated cookware. Studies of the extractability of perfluorinated compounds from nonstick cookware have reported variable results (Powley et al., 2005; Washburn et al., 2005; Sinclair et al., 2007). Pentadecafluorooctanoic acid (PFOA) has been a particular focus of both public health and regulatory concern, due to accumulating evidence of serious health risks (ATSDR, 2015a; EPA, 2016). The Xylan coating used in this study is a PFOA-free material.

Beyond the specific issues with PFOA, there are growing environmental and safety concerns with the use of other poly- and perfluoroalkyl compounds. >200 scientists have signed the 2015 Madrid Statement on poly- and perfluoroalkyl substances calling for the development of safer alternatives and to stop their use when these alternatives exist (Blum et al., 2015). In the case of aluminum cookware the use of any coating material must be weighed against the health protection benefits that may be provided by reducing exposures to these metals. Additional risk-benefit analysis to assess environmental and health trade-offs should be considered in evaluating proposed solutions to respond to the concerns posed by cookware made with recycled metal. Due to the small sample size in this preliminary evaluation, it is not possible to conclude that Xylan or any other coating would reduce metal exposures from this type of cookware over time. Additional studies on the potential for metals to be released over multiple boiling procedures and other types of cooking simulation would be warranted. In addition, further study is needed to determine the ability of fluoropolymer compounds to leach during normal cooking from this type of coating material used for this application.

Anodization of aluminum cookware can also provide a surface that is far more resistant to corrosion and leaching (Sekheta et al., 2010). However, the infrastructure needed to treat locally made aluminum pots would also have substantial cost implications. It is unclear whether the production quality and the significant concentrations of contaminant metals in cookware from most of the artisanal producers would be appropriate for undergoing the anodization treatment. Given the serious risks of heavy metal poisoning posed by this cookware, it is imperative that the feasibility and safety of corrosion-resistant coatings be urgently investigated.

5. Conclusions

This paper presents the findings of an investigation of the leaching characteristics of inexpensive aluminum cookware from ten countries in Africa, Asia and Central America. This investigation demonstrates that artisanal aluminum cookware is a significant and previously unrecognized source of exposure to lead and other metals. Here, we report that simulated cooking leached 1 to 1426 micrograms of lead per serving from fifteen items purchased in eight countries. In addition, arsenic and cadmium were present in some leachates at potentially harmful levels, and estimated aluminum exposures per serving exceed the recommended maximum intake for 40 of the 42 items tested. We conclude that exposure to metals by the corrosion of inexpensive, aluminum cookware may pose significant public health risks throughout the developing world.

Our results suggest that corrosion-resistant coatings may be effective in reducing metal leaching from this type of cookware. There are a number of possible alternative coatings that require additional investigation including fluoropolymer materials as well as anodization and enamel coatings. Improving the corrosion resistance of this cookware with a post-production coating treatment appears to be a more feasible option than to regulate or screen material inputs used by small producers. However, it would be useful to consider piloting various approaches in different markets to better assess the economic feasibility, cultural acceptance and practical reach of these alternative strategies. Research is urgently needed to identify safe and effective corrosion-resistant coatings that could improve the safety of this cookware.

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Competing interests

The authors declare no actual or competing financial interests.

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