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SHORT REPORT

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A pilot study to assess lead exposure from routine consumption of coffee and tea from ceramic mugs: comparison to California Safe Harbor Levels

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Abstract

Background: Lead (Pb) is a pervasive metal that can be found in, and potentially leached from, ceramics, particularly into acidic foods and beverages. The purpose of this study was to investigate potential lead exposure from coffee and tea consumption, given that both are acidic and routinely consumed from ceramic mugs. We measured the concentration of lead in coffee and tea at two different time points brewed in five readily available mugs known to contain lead. Results were compared to EPA's action level for drinking water and FDA's allowable level for bottled water. The measured concentrations, along with consumption patterns, were also used to calculate potential daily lead doses, which were compared to California's Safe Harbor Levels under Proposition 65. Additionally, we estimated changes in adult and fetal blood lead levels using EPA's Adult Lead Methodology model.

Findings: The results of this pilot study suggest that lead in ceramic mugs can leach into coffee and tea. The measured lead concentrations ranged from 0.2 to 8.6 μg/L in coffee, and from <0.2 to 1.6 μg/L in tea. No statistical differences were found between the measured concentrations in coffee, tea, or water within each cup, or in the measured concentrations between retention times within each cup. However, a statistically significant difference was observed in the lead concentrations measured between cups, indicating that the lead concentrations were dependent on the cup used, rather than on the beverage or retention time. The estimated daily dose of lead exceeded the California Maximum Allowable Dose Level of 0.5 μg per day for one of the five mugs tested. Blood lead levels did not increase above regulatory or guidance values.

Conclusions: This preliminary investigation provides data on potential lead exposures from daily beverage consumption among typical consumers, relevant to a substantial portion of the population, with particular implications for pregnant women.

Keywords: Lead, Coffee, Tea, Ceramics, Proposition 65, Leaching

Introduction

Lead (Pb) is a naturally occurring metal pervasive in the environment that can cause well-known adverse health effects in humans upon sufficient exposure (Brown and Margolis 2012; ATSDR 2007b). The primary target of lead toxicity in both children and adults is the nervous system, although children are more sensitive to lead's neurotoxic effects. Moreover, children generally absorb more ingested lead into their blood than do adults. Children absorb approximately 50% of ingested lead, while adults absorb approximately 10% (ATSDR 2007b, ATSDR 2007a; Philip and Gerson 1994). Lead exposure may begin *in utero*, as it can cross the placenta (Mason et al. 2014; Brown and Margolis 2012). Decreasing cognitive function has been observed with increasing lead exposure in both children and adults (Mason et al. 2014; Brown and Margolis 2012). IQ deficits of one to five points have been associated with blood lead level increases of 10 μg/dL or less in children (ATSDR 2007a). At high levels of exposure, lead can cause fatal damage to the brain and kidney

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in both adults and children (ATSDR 2007b). Moreover, the International Agency for Research on Cancer (IARC), the U.S. Department of Health and Human Services (DHHS), and the U.S. Environmental Protection Agency (EPA) have all considered lead or lead compounds as probably carcinogenic in humans (ATSDR 2007b).

Historically, lead was used in gasoline, paint, plumbing, and various industrial processes (Levin et al. 2008). Because of concerns over lead's human health effects, many of these uses have been limited or banned under regulatory actions in the U.S. For example, lead was phased out of gasoline beginning in 1973, residential lead-based paint was banned in 1978, and lead solder in food cans was banned in 1995 (Brown and Margolis 2012). While these efforts over the past several decades have significantly reduced the potential for lead exposure in the U.S., lead poisoning remains an appreciable health concern, especially for children.

In January 2012 *Consumer Reports* published an article indicating that 25% of 88 juices tested exceeded the Food and Drug Administration (FDA) bottled water standard for lead of 5 μg/L (CR 2012; USFDA 1995). More recently, the lead-contaminated drinking water in Flint, Michigan, has drawn the public eye to lead poisoning. Based on elevated blood lead levels, the President declared a state of emergency in Flint in early 2016 (The White House 2016). The Centers for Disease Control and Prevention (CDC) estimates that at least four million U.S. households contain children exposed to "high levels of lead" (CDC 2016b). In 2012, the CDC lowered the blood lead level at which it recommends public health action from 10 to 5 μg/dL for children. Though average blood lead levels have significantly declined in U.S. children since the 1970s, the CDC estimates that approximately half a million U.S. children between the ages of one and five have blood lead levels above 5 μg/dL (CDC 2016b; CDC 1997).

The most common route of lead exposure is through ingestion, and multiple regulatory and guidance values exist to limit lead ingestion in the U.S. (ATSDR 2007a). The Environmental Protection Agency (EPA) has set a lead action level of 15 ppb in drinking water, for example, and the FDA has set an allowable level for lead in bottled water of 5 ppb (EPA 1991; USFDA 1995). The World Health Organization (WHO) formerly set a provisional tolerable weekly intake (PTWI) of 25 μg/kg/week for lead from food and water, but withdrew it in 2010. The WHO stated that it did not issue an updated PTWI because it was "not possible" to establish a value that would be "health protective" (WHO 2010). In California, lead is listed as a chemical known to cause cancer and reproductive toxicity under the Safe Drinking Water and Toxic Enforcement Act (Proposition 65). Accordingly, the California Office of Environmental Health Hazard Assessment (OEHHA) has set Safe Harbor Levels for lead, consisting of a Maximum Allowable Dose Level (MADL) of 0.5 μg/day, based on reproductive toxicity, and a No Significant Risk Level (NSRL) of 15 μg/day based on lead's potential carcinogenicity (OEHHA 2016b). There is currently a petition to lower the MADL to 0.2 μg/day (Cal/EPA 2015a; Cal/EPA 2015b).

Ceramics with lead-containing paint or glaze are one potential source of lead exposure (ATSDR 2007b). Several studies have suggested that lead may leach from such ceramics, particularly in acidic environments (Sheets 1997; Mohamed et al. 1995; Levin et al. 2008; Markowitz 2000; Gonzalez de Mejia and Craigmill 1996; Feldman et al. 1999). In fact, the FDA limits the amount of lead in cups and mugs to that which results in no more than 0.5 μg/mL lead in an acidic leaching solution (USFDA 2015). The purpose of the current pilot study, then, was to investigate potential lead exposure from coffee and tea consumption, given that both beverages are acidic and routinely consumed from ceramic mugs in the U.S. Specifically, we measured the concentration of lead in coffee and tea at two time points brewed in five commercial mugs known to contain lead.

Materials and methods

The five mugs chosen for this study were selected because they were found to contain lead in a screening-level assessment. Specifically, 24 mugs from the authors' office were tested using an Olympus Innov-X Delta handheld X-Ray fluorescent (XRF) analyzer. Each mug was measured once with the XRF gun at its highest sensitivity setting, which required the tester to hold the analyzer over the mug for 45 s. The three mugs with the highest resulting lead concentrations (1,223 to 7,034 mg/kg) were selected for the present study. These mugs each had decorative elements and will be referred to by their predominant colors: Green Decorative, Yellow Decorative, and Red Decorative. In addition, two representative mugs were selected from the batch of office mugs baring the authors' company's logo. These will be referred to as Black Logo1 and Black Logo2. All five mugs selected were in active use in the authors' San Francisco, California, office environment, and were typically washed daily in an automatic dish washer. Four of the five mugs were purchased in the U.S., and one was purchased in Europe (Red Decorative). The mugs all appeared to be in good condition, with no obvious signs of damage or wear.

Five beverage-making events were performed in the five mugs, as well as in a glass cup, in duplicate, allowing the collection of 60 total samples. The five mugs and one glass cup will hereafter be collectively referred to as 'cups'. The sampling protocol is described in greater detail below. The five beverages were hot water, coffee 10 and 60 min post-brewing, and tea 10 and 60 min post-brewing. Tap water was used for each scenario; the first tap water collection occurred at approximately 9:30 in the morning, after the tap had been used intermittently for several hours. Between each event, the cups and all utilized equipment were washed with dish soap and water and dried.

In each sampling situation, the "end temperature" of the beverage was measured immediately before pouring approximately 250 mL of the beverage into a plastic sampling container. Sampling containers are depicted in Fig.1a. Four control samples of tap water were also collected. All containers were shipped on ice to a laboratory accredited by the Environmental Laboratory Accreditation Program (ELAP), where they were analyzed for the presence of lead using a Perkin Elmer inductively coupled plasma mass spectrometer (IC-PMS), according to EPA Method 200.8. As specified in Method 200.8, with each set of samples, the laboratory ran method calibration blanks and multiple laboratory control samples and duplicates to verify the instrument performance and determine instrument precision. A matrix spike of 50 μg/L lead was utilized as a calibration procedure.

Water

Water was boiled in an electric kettle, and approximately 300 mL of water was poured into each of the six cups. After 30 min had elapsed, the water was stirred. After 60 min had elapsed, the water was stirred again, the temperature of the water was recorded, and the water was poured into the sample containers. The scenario was repeated for each of the six cups such that a total of 12 samples were collected.

Coffee

Coffee was brewed utilizing a single cup pour-over cone with a paper filter (Fig.1b, c). Approximately 300 mL boiling water was poured over three teaspoons of a nationally available, freshly-ground, medium roast, 100% Arabica coffee for each of the six cups. Two scenarios were performed in duplicate: in the first, the coffee remained in the cup for 10 min, and in the second, the coffee remained in the cup for 60 min before being poured into the sample container. After 5 min had elapsed, the pour-over cone was removed, and the coffee was stirred in both scenarios. The 60-min samples were also stirred after 30 min, and all samples were stirred prior to transfer to the sampling containers.

Tea

Tea was prepared by pouring boiling water over one nationally available, 2 g black tea bag in each cup (Fig.1d). Water was boiled in an electric tea kettle, and 300 mL of water was poured into each cup. Two scenarios were performed in duplicate: in the first, the tea remained in the cup for 10 min, and in the second, the tea remained in the cup for 60 min before being poured into the sample container. The tea bag was dunked in the water several times within 5 min of steeping in both scenarios. After 5 min of steeping, the tea bag was removed, and the tea was stirred. The 60-min samples were also stirred after 30 min, and all samples were stirred prior to transfer to the sampling containers.

Tap water controls

On a separate day from the beverage making scenarios, two samples of tap water that had been boiled and retained in the electric tea kettle for 60 min were collected. After boiling was reached, the water was allowed

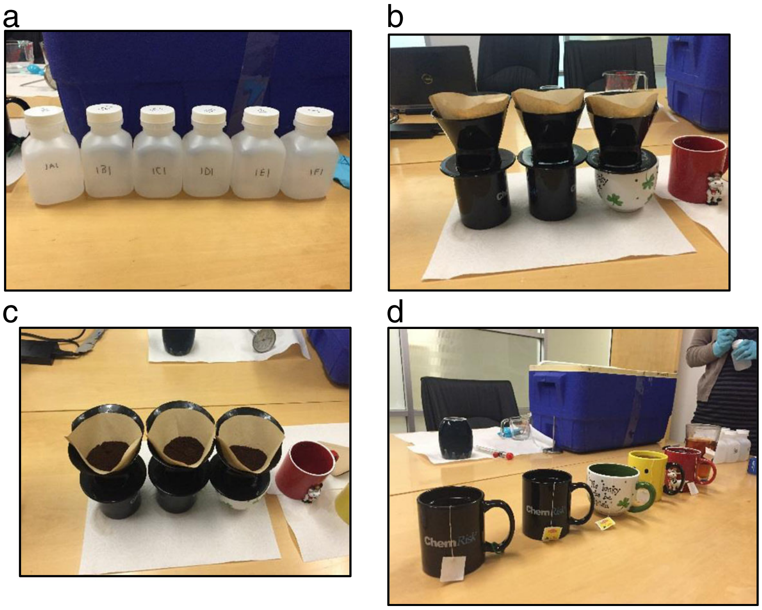

Fig. 1 Brewing and Sampling Equipment. **a** Sampling containers. **b**, **c** Coffee brewing. **d** Tea brewing

to cool, was stirred inside the kettle at the 30 and 60-min marks, and then was poured into the sampling containers. Two additional samples were collected directly from the tap.

Risk assessment: comparison to regulatory limits

For simple comparison purposes, and to put our results in the context of regulatory limits, our measured concentrations were compared to the EPA's lead action level of 15 ppb in drinking water and the FDA's allowable level for lead in bottled water of 5 ppb.

The sampling results were also utilized along with general assumptions to determine the level of lead exposure expected from daily consumption of coffee and tea from the cups. These estimated daily doses were compared to the current Safe Harbor Levels under Proposition 65 in California (MADL: 0.5 μg/day, NSRL: 15 μg/day). The EPA currently has no Reference Dose for lead.

The daily dose of lead was calculated simply as the amount of lead in a serving of the beverage from the study cups multiplied by the number of servings typically consumed in one day, or:

$$\frac{\text{Amount of Lead }(\mu\text{g})}{\text{Beverage Serving }(\text{L})} \times \frac{\text{Beverage Servings}(\text{L})}{\text{Day}} = \text{Amount of Lead Ingested per Day }(\mu\text{g/day})$$

Basing these calculations on typical coffee and tea consumption is appropriate, since Proposition 65 dictates that the MADL be calculated "using the reasonably anticipated rate of intake or exposure for average users of the consumer product" (OEHHA 2016a). Note that our estimate accounts only for lead exposure from drinking beverages from the subject cups, and ignores any other potential sources or routes of lead exposure.

Coffee

A 2009 survey by the National Coffee Association reported that the average consumption among coffee drinkers in the U.S. aged 18 and older was 3.3 eight-ounce cups per day (USFDA 2012). We thus assumed a daily consumption of 26.4 fluid ounces, or 0.78 L, of coffee per day in our risk assessment.

Tea

According to the most recent available data (2011–2012) from the National Health and Nutrition Examination Survey (NHANES), median tea consumption among U.S. tea drinkers age 18 to 80 is 355.2 g per day, or 12.01 fluid ounces per day (CDC 2014). We thus assumed a daily consumption of 12.01 fluid ounces, or 0.36 L, of tea per day in our risk assessment.

Estimation of blood lead levels following exposure to lead from mugs

As another point of comparison, we utilized our measured concentration data to estimate changes in adult blood lead levels (BLLs) using the EPA's Adult Lead Methodology (ALM) model. The ALM model has previously been used to estimate BLLs as a result of exposure to lead from beverages and consumer products (Monnot et al. 2015). The average baseline BLL for adults was assumed to be 1.0 μg/dL (ALM default value); however, we also ran the model assuming a baseline of 0 μg/dL in order to determine the contribution of lead from the beverages ingested in the lead-containing ceramic mugs. The gastrointestinal absorption for lead was assumed to be 12% in adults, the default for the model. Any ingestion from soil and dust was assumed to be zero because of their irrelevance to beverage exposure. The average consumption of coffee per day (in g/day) was used because average coffee consumption is higher than average tea consumption. We assumed an exposure frequency of 365 days/year, assuming that a person would drink the same amount of the beverage every day. The BLLs estimated from the model were compared to regulatory and guidance values for BLLs set by the CDC and EPA. Additionally, the model was used to estimate fetal blood lead concentrations in women exposed to lead from beverages contaminated by ceramic mugs.

Findings

Lead was measured at levels at or above the limit of detection (0.2 μg/L) in 56 out of 60 samples (93.33%) (Table 1, Fig. 2). The percent recovery from quality control samples ranged from 95.6 to 102%. The percent recovery from matrix spikes ranged from 98.3 to 102%. Out of the four samples for which the lead concentration was below the limit of detection, three results were from 60 min water samples and one was from a 10 min tea sample. The total range of lead levels measured was <0.2 μg/L to 8.6 μg/L. The highest concentration of 8.6 μg/L came from a 10 min coffee sample in the Green Decorative mug. The other lead concentrations measured from the Green Decorative mug, though consistently higher than the measures for all other cups, did not exceed 1.8 μg/L. All four tap water control samples resulted in measurements below the limit of detection (<0.2 μg/L) (Results not shown).

Statistical tests were performed in order to determine what factors contributed to the concentrations of lead measured in beverages from each cup. Non-parametric methods were used, as the data were found to be non-normally distributed. Additionally, a value of 0.1 μg/L (the limit of detection divided by the square root of 2) was substituted for the four samples that resulted in concentration measures below the limit of detection.

Table 1 Measured lead concentrations by cup, beverage, and time

Cup	Medium	Time in cup (min)	Result (μg/L)	End temperature (°C)
Black Logo1	Coffee	10	0.2	55
Black Logo1	Coffee	10	0.2	55
Black Logo1	Coffee	60	0.2	34
Black Logo1	Coffee	60	0.2	36
Black Logo1	Tea	10	<0.2	35
Black Logo1	Tea	10	0.2	35
Black Logo1	Tea	60	0.2	59
Black Logo1	Tea	60	0.2	60
Black Logo1	Water	60	0.2	36
Black Logo1	Water	60	<0.2	37
Black Logo2	Coffee	10	0.2	55
Black Logo2	Coffee	10	0.2	57
Black Logo2	Coffee	60	0.2	34
Black Logo2	Coffee	60	0.2	34
Black Logo2	Tea	10	0.2	35
Black Logo2	Tea	10	0.2	35
Black Logo2	Tea	60	0.2	60
Black Logo2	Tea	60	0.2	60
Black Logo2	Water	60	0.2	37
Black Logo2	Water	60	0.2	37
Green Decorative	Coffee	10	8.6	55
Green Decorative	Coffee	10	1.7	56
Green Decorative	Coffee	60	1.8	31
Green Decorative	Coffee	60	1.3	33
Green Decorative	Tea	10	1.6	33
Green Decorative	Tea	10	1.2	33
Green Decorative	Tea	60	1.0	57
Green Decorative	Tea	60	0.9	58
Green Decorative	Water	60	0.8	34
Green Decorative	Water	60	1.6	34
Yellow Decorative	Coffee	10	0.4	55
Yellow Decorative	Coffee	10	0.2	56
Yellow Decorative	Coffee	60	0.2	35
Yellow Decorative	Coffee	60	0.2	36
Yellow Decorative	Tea	10	0.2	36
Yellow Decorative	Tea	10	0.2	37
Yellow Decorative	Tea	60	0.2	60
Yellow Decorative	Tea	60	0.2	60
Yellow Decorative	Water	60	0.3	35
Yellow Decorative	Water	60	0.2	35
Red Decorative	Coffee	10	0.2	58
Red Decorative	Coffee	10	0.2	56
Red Decorative	Coffee	60	0.2	33
Red Decorative	Coffee	60	0.2	36
Red Decorative	Tea	10	0.2	35
Red Decorative	Tea	10	0.2	35
Red Decorative	Tea	60	0.3	60
Red Decorative	Tea	60	0.2	60
Red Decorative	Water	60	0.3	35
Red Decorative	Water	60	<0.2	37
Glass	Coffee	10	0.3	59
Glass	Coffee	10	0.2	58
Glass	Coffee	60	0.2	32
Glass	Coffee	60	0.2	35
Glass	Tea	10	0.2	35
Glass	Tea	10	0.2	35
Glass	Tea	60	0.2	57
Glass	Tea	60	0.2	60
Glass	Water	60	0.2	36
Glass	Water	60	<0.2	35

The Mann-Whitney Wilcoxon test was performed between the coffee samples retained in the cups for 10 and 60 min, and between the tea samples retained in the cups for 10 and 60 min, respectively. No statistical significance was found for either test, indicating that there was no difference in lead concentrations measured depending on the amount of time the beverages were retained in the cups (coffee samples: $p = 0.43$; tea samples: $p = 0.60$). As a result of this test, the 10 and 60 min samples for coffee and tea, respectively, were pooled from each cup to perform the Kruskal-Wallis test. This test was performed to determine if there was a difference in lead measured depending on the beverage in the cups. No statistical significance was found ($p = 0.64$). Finally, the Kruskal-Wallis test was performed on the samples from each cup to determine if there was a difference in lead concentrations measured depending on which cup was used. The Nemenyi multiple comparison test was utilized to determine statistical differences among the cups, and it found that samples from the Green Decorative mug were statistically significantly higher than samples from all other cups ($p = 0.0001–0.0125$).

Risk assessment: comparison to regulatory limits

Measured lead concentrations in the beverage samples from the six cups in this study ranged from <0.2 to 8.6 μg/L. Mean lead concentrations across all beverage scenarios were 0.18 μg/L in the Black Logo1 mug, 0.2 μg/L in the Black Logo2 mug, 2.05 μg/L in the Green Decorative mug, 0.23 μg/L in the Yellow Decorative

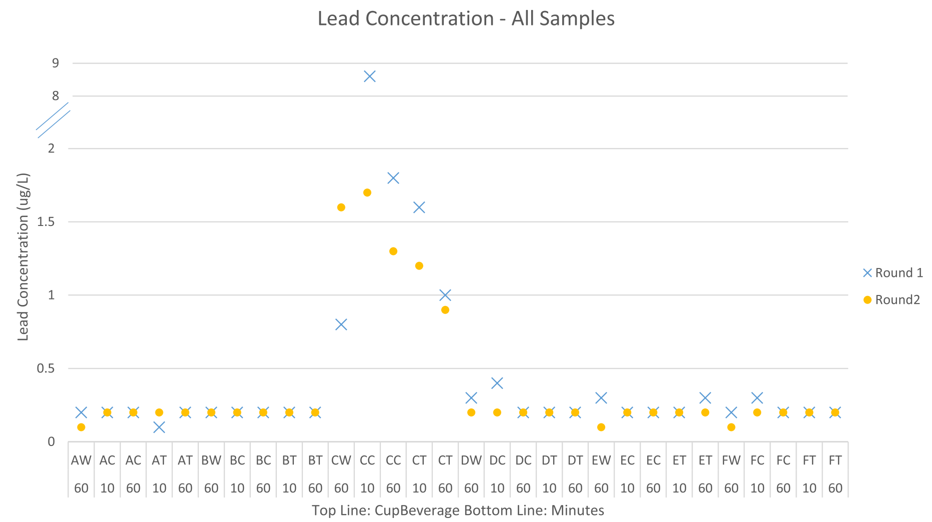

Fig. 2 Lead Concentration – All Samples. Measured lead concentrations by mug/cup, beverage and time. Cup A = Black Logo1 mug; Cup B = Black Logo2 mug; Cup C = Green Decorative mug; Cup D = Yellow Decorative mug; Cup E = Red Decorative mug; Cup F = Glass Cup. Beverage W = Water. Beverage C = Coffee. Beverage T = Tea. Samples below the limit of detection (<0.2 μg/L) graphed at the limit of detection divided by the square root of two (0.1 μg/L)

mug, 0.21 μg/L in the Red Decorative mug, and 0.2 μg/L in the glass cup (Table 2).

All measured lead concentrations were below the EPA's lead action level of 15 ppb in drinking water. However, the highest concentration measured from the Green Decorative mug (8.6 μg/L or 8.6 ppb) exceeded the FDA's allowable level for lead in bottled water of 5 ppb. This simple comparison, although interesting, does not have clear health implications, since these EPA and FDA health - based values use water consumption rates, which are much higher than coffee or tea rates of consumption.

For comparison the to the California Proposition 65 Safe Harbor Levels, given that the samples from the Green Decorative mug were statistically different from samples from all other cups, we calculated daily lead intake based on three values: 1) the mean lead concentration from all cups excluding the Green Decorative mug (0.2 μg/L); 2) the mean lead concentration from the Green Decorative mug (2.05 μg/L); and 3) the maximum concentration measured from the Green Decorative mug (8.6 μg/L). These three values incorporate all beverage scenarios because, as described above, there was no statistical difference between beverages or retention time in the cups in this study.

Table 2 Range of measured lead concentrations by cup

Cup Name	Lead Concentration (μg/L)			
	Min	Max	Mean	Median
Black Logo1	<0.2	0.2	0.18	0.2
Black Logo2	0.2	0.2	0.2	0.2
Green Decorative	0.8	8.6	2.05	1.45
Yellow Decorative	0.2	0.4	0.23	0.2
Red Decorative	<0.2	0.3	0.21	0.2
Glass	<0.2	0.3	0.2	0.2

Concentrations reported to be below the limit of detection (<0.2 μg/L) were assumed to have a value of the limit of detection divided by the square root of two (0.1 μg/L) in statistical analyses

Coffee

Using the mean lead concentration of 0.2 μg/L from all cups except the Green Decorative mug, and the average daily coffee intake among coffee drinkers of 0.78 L, the daily dose of lead from coffee consumption would be 0.156 μg. This dose is below both the current Proposition 65 NSRL of 15 μg/day and the MADL of 0.5 μg/day. Using the mean lead concentration of 2.05 μg/L from the Green Decorative mug, the daily dose of lead for the average coffee drinker would be 1.60 μg, which exceeds the current MADL by over three fold. Using the maximum measured lead concentration of 8.6 μg/L and the same average daily coffee intake of 0.78 L, the daily dose of lead from coffee consumption is estimated to be 6.71 μg/day, which is 13.4 times the current MADL. Even consuming one eight-ounce serving of coffee with this concentration of lead would exceed the MADL four-fold, resulting in a daily dose of lead of 2.03 μg.

Tea

Using the mean lead concentration of 0.2 μg/L from all cups except the Green Decorative mug and the average daily tea intake among tea drinkers of 0.36 L, the daily dose of lead from tea consumption would be 0.071 μg, which is below both the current Proposition 65 NSRL of 15 µg/day and the MADL of 0.5 µg/day. Using the mean lead concentration of 2.05 µg/L from the Green Decorative mug, the daily dose of lead for the average tea drinker would be 0.728 µg, which exceeds the current MADL. Using the maximum lead concentration of 8.6 µg/L and the same average daily tea intake of 0.36 L, the daily dose of lead from tea consumption would be 3.05 µg/day, or approximately six times the current MADL.

BLLs resulting from exposure scenario

Table 3 represents the predicted BLLs for adults and fetuses based on the measured lead concentrations in this study. Two lead concentrations were used to estimate BLLs; one scenario used the maximum concentration of lead measured from all beverages (8.6 µg/L), and the other used the mean concentration of lead measured from all beverages, excluding results from the Green Decorative mug (0.2 µg/L). Based on the average consumption of coffee (0.78 L per day, or approximately 780 g assuming coffee has the same density as water), the predicted BLLs for adults ingesting coffee daily ranged from 0.0 to 1.3 µg/dL and from 0.0 to 1.2 µg/dL in fetuses, assuming baseline BLLs of 0 and 1 µg/dL, respectively. The contribution of lead, therefore, from coffee from the cups was estimated to increase BLLs above background by a maximum of 0.3 µg/dL in adults and by 0.2 µg/dL in fetuses. All of the estimated BLLs were below the BLL of concern of 5 µg/dL set by the CDC and 10 µg/dL set by EPA, and did not raise BLLs by more than +1.0 µg/dL, which is California's proposed benchmark for risk assessment (Carlisle and Dowling 2007; CDC 2016a; EPA 2016).

Discussion

Lead was detected in over 90% of our beverage samples; most samples resulted in concentrations similar to the analytical limit of detection (0.2 µg/L). However, in one of the mugs tested (Green Decorative), the results ranged from 0.8 to 8.6 µg/L. This finding indicates that under the conditions of this study, lead may leach from mugs into hot beverages such as coffee and tea, or even hot water, and result in individual lead exposures well above maximum allowable dose levels set by the State of California. Although this finding only pertained to one of the five mugs tested, it was unexpected, given that these mugs were randomly selected from an office environment, and that all but one were purchased in the U.S. From the small sample size of this study no conclusion can be drawn about the prevalence of mugs with leachable lead in the U.S. market; however, the findings do indicate a need for further research with greater sample sizes (and thereby more robust statistical analyses) in this area. Regarding blood lead levels, we found that background lead exposure in the models primarily contributed to the BLLs for both adults and fetal exposures. As shown in Table 3, lead ingestion from coffee using the highest concentration measured in this study increased the BLL estimated in adults by 0.3 µg/dL. Given the limitations of this study, these results should be considered a screening-level assessment.

The U.S. FDA regulates lead content in ceramics used with foods, and in mugs specifically. In 1970, the FDA

Table 3 Estimated blood lead levels following exposure to lead from mugs

	Exposure Scenario			
Background (baseline BLL)	0.0 µg/dL[a]		1.0 µg/dL[b]	
Coffee Ingestion (consumption; concentration)	780.0 g/day; 0.00022 ppm[c]	780.0 g/day; 0.0086 ppm[d]	780.0 g/day; 0.00022 ppm[c]	780.0 g/day; 0.0086 ppm[d]
Maternal Blood Lead levels (µg/dL)	0.0	0.3	1	1.3
Fetal Blood Lead levels (µg/dL)	0	0.3	0.9	1.2

Key

"Low" Exposure	"High" Exposure

[a] Chosen value for background

[b] Default value from the ALM model

[c] Maximum Pb concentration in beverages from results

[d] Mean Pb concentration excluding green decorative mug

conducted a survey of imported pottery and found "high levels" of lead leaching from the products (USFDA 1979a, p. 51237). The FDA instituted a compliance program for domestic and international pottery in 1971, limiting the amount of lead that leached from pottery into a leaching solution to 7 μg/L (USFDA 1979a). In 1979, the FDA revised the guidelines for lead leached from ceramic foodware based on a recommended tolerable total lead intake value of 100 μg/day for infants up to 6 months of age and of 150 μg/day for children from 6 months to 2 years of age, based on the endpoint of altered heme synthesis (USFDA 1979b; USFDA 1989). In response to new data and updated international reference values, in 1989 the FDA adopted a range of 6 to 18 μg/day as the provisional tolerable lead intake from food for a 10 kg child, and proposed that the guidelines for ceramic foodware again be lowered (USFDA 1989). The agency noted at that time that it was not possible to establish a threshold for lead toxicity (USFDA 1989).

In 1992, the lead release guidelines were amended to include levels specific to cups and mugs "because these articles are frequently used under conditions that may enhance lead leaching" (USFDA 1992, p. 29734). In particular, the FDA noted that cups and mugs are "generally used to hold acidic beverages, such as…coffee or tea" (USFDA 1992, p. 29735). The FDA reported that the acidity in conjunction with the higher temperatures of these beverages enhances the lead leaching rate (USFDA 1992). According to the 1992 guidance, the FDA "may take enforcement action" when cups or mugs exceed a lead level of 0.5 μg/mL in a 4% acetic acid leaching solution in any of six mug or cup units examined (USFDA 1992, p. 29735). This limit currently is still in place for cups and mugs.

The FDA reported that based on its request for information and subsequent review, the amount of lead that will leach into a leaching solution is approximately 2.5 to 5 times the amount that will leach into "hot coffee during 15 to 30 min" (USFDA 1992, p. 29735). Applying this estimation to our results, we can estimate that 2 to 43 μg/L lead would leach from the Green Decorative mug into leaching solution, which is 0.002 to 0.043 μg/mL, and well within the FDA's limit of 0.5 μg/mL. Our results, then, suggest that mugs in compliance with federal regulatory limits for lead may still well-exceed California's Safe Harbor Levels.

Furthermore, the State of California recently proposed reducing the lead MADL to 0.2 μg/day based on modeling of exposure levels that would result in maximum blood lead levels of 15 μg/dL (Cal/EPA 2015a; Cal/EPA 2015b). In 2015, the State clarified that the MADL is intended to be a daily exposure dose, but that alternative exposure doses (increased doses) are plausible within the law, if exposures can be determined not to occur daily (Cal/EPA 2015a). Nonetheless, given the proposed MADL, and assuming mugs are used daily, coffee consumption from the four mugs with lower associated lead levels in this study (mean of 0.2 μg/L, or daily intake of 0.156 μg) nearly results in exposures above the proposed MADL. The lower MADL would also further widen the discrepancy between California and federal compliance levels.

Given the existing low MADL, and the fact that the proposed MADL for lead is less than half of the current MADL, understanding the implications for product testing is important. Many analytical methods are not sensitive enough to detect the presence of lead at meaningful concentrations in terms of exposure levels in compliance with the Safe Harbor Level. Inherent variability in measurements at such low levels of analytical detection also exists, which must be characterized and understood properly in order to rely on them for regulatory compliance. For example, the two 10 min coffee samples in the Green Decorative mug were 1.7 and 8.6 μg/L, a seemingly wide range. Also, coffee that had been in the glass cup for 10 min had a detected lead concentration of 0.3 μg/L, but the same coffee after 60 min in the glass resulted in a detected lead concentration of only 0.2 μg/L. These ranges and "reductions" highlight the uncertainty and normal variations in the measurements.

In addition, definitively identifying and segregating the relative contributions of lead from different sources in this study is not possible. Lead is ubiquitous in our environment, and its presence in glass, equipment, tea, coffee, and drinking water cannot be ruled out. Several studies, for example, have reported concentrations of lead in solid coffee beans or solid residues of coffee infusions ranging from 0.053 to 1.239 μg/g (Nędzarek et al. 2013; Onianwa et al. 1999; Federal Republic of Germany and Federal Länder unknown; Santos et al. 2004; Othman 2010). One study reported lead concentrations in liquid coffee of 2.37 and 2.57 μg/L (Ong 2014), and another study reported lead concentrations below the limit of detection of 1.5 μg/L (Ashu and Chandravanshi 2011). Additionally, studies have reported lead in tea leaves or residues from tea infusions of 0.046 to 15.479 μg/g (Li et al. 2015; Shekoohiyan et al. 2012; Shokrzadeh et al. 2008; Onianwa et al. 1999; Othman 2010; Al-Othman et al. 2012; Zheng et al. 2014). Although we did not detect any lead in our direct tap water or boiled tap water control samples, the presence of lead in drinking water is a known concern, as shown by the recent state of emergency issued in Flint, Michigan, because of its drinking water lead content. The U.S. EPA reported that 1,831 (8%) of 22,808 residential water samples collected in Flint between September 2015 and June 2016 were above the action level of 15 ppb (State of Michigan 2016). Samples reported above the action level ranged from 16 to 22,905 ppb.

These other highly variable potential sources of lead exposure must be considered when assessing total lead intake; in this study, though, the beverage lead content appeared to be most strongly determined by the mug.

To our knowledge, ours is one of only a handful of studies in the peer-reviewed literature to evaluate lead leaching into coffee or tea from lead-containing ceramics. In 1985, Wallace et al. tested Italian-originating coffee mugs found in a U.S. household. The authors reported that approximately 4000 μg of lead were leached into a 250 mL serving of coffee (16,000 μg/L) over 15 min at a temperature of 65 °C and a pH of 5.1 (Wallace et al. 1985). Wallace et al. stated that the mugs were "badly degraded," and that a similar new cup released only 200 μg of lead (800 μg/L) in the same conditions (Wallace et al. 1985, p. 290). Ajmal et al. (1997) investigated lead leaching into tea from ceramic mugs in India. They reported that measured lead concentrations in the tea were below the limit of detection (Ajmal et al. 1997). To our knowledge, then, the current study is the first to evaluate lead leaching into coffee and tea from ceramic mugs purchased in the 21st century in the U.S. (with the exception of the Red Decorative mug, which was purchased in Europe).

A greater number of studies have investigated lead leaching from ceramics associated with various other foods and beverages (Sheets 1997; Mohamed et al. 1995; Levin et al. 2008; Markowitz 2000; Gonzalez de Mejia and Craigmill 1996; Feldman et al. 1999; Belgaied 2003; Hight 1996; Valadez-Vega et al. 2011). In studies that measured lead concentrations leached from a variety of ceramics using 4% acetic acid (the same leaching solution used by the FDA to evaluate ceramics), reported values reached up to 2004.7 ppm (2,004,700 μg/L), significantly higher than values we obtained in this study (Gonzalez de Mejia and Craigmill 1996). Many of these studies also evaluated the amount of lead that leached into various acidic and non acidic foods, such as salsa, beans, tamarind juice, pickle juice, wine, and milk products, with results reaching up to 244 ppm (244,000 μg/L). The highest value was associated with salsa, a highly acidic food (Gonzalez de Mejia and Craigmill 1996). The study, however, noted that there was a mean background level of lead of 0.93 ± 0.13 ppm in the salsa.

Overall, the potential for lead ingestion from contaminated ceramic mugs is minimal when compared to other sources, such as food. The Agency for Toxic Substances and Disease Registry (ATSDR) reported that the average daily intake of lead from food sources in the general population is approximately 56.5 μg/day (ATSDR 2007a). In comparison, the maximum daily lead intake from drinking 3.3 eight-ounce cups of coffee based on the data collected in our study resulted in a dose of 6.71 μg, over eight times less than the average daily lead intake from food. Nonetheless, exposure to lead should be minimized to the extent possible. The U.S. EPA does not publish a safe dose for lead because it felt it was "inappropriate to develop a reference dose (RfD) for inorganic lead (and lead compounds) because some of the health effects associated with exposure to lead occur at blood lead levels as low as to be essentially without a threshold" (ATSDR 2007a, p. 403). Similarly, WHO withdrew its provisional tolerable weekly intake (PTWI) for lead in 2010 because it did not believe establishing a value that would be "health protective" would be possible (WHO 2010).

This study is limited in that a small number of mugs were randomly selected from the authors' work environment, and were not purchased for the purpose of evaluating the full range of lead contamination in ceramic mugs. We also were not able to test multiple mugs from the same manufacturer or origin, with the exception of the black logo mugs. Overall, then, this study can be considered a pilot study, and the results should be considered as such until additional research can be conducted, and more samples collected. This preliminary investigation, however, provides data on potential lead exposures from daily beverage consumption among typical consumers, with particular implications for pregnant or breastfeeding women. This potential source of lead exposure is less well-characterized than are some other lead exposure sources (e.g., paint, gasoline), yet is relevant to a substantial portion of the U.S. population.

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Authors' contributions

All named authors contributed significantly to the development and writing of this manuscript. Generally, GA took the lead in designing the study; LG took the lead in estimating blood lead levels; MF took the lead in the statistical analysis; and SG assisted with the design of the study and took the lead in the comparison to regulatory limits. All authors were involved in executing the study. All authors read and approved the final manuscript.

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Competing interest

The authors declare that they have no competing interest, and that no external funding was provided for writing this manuscript.

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