Incidence of waterborne lead in private drinking water systems in Virginia

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ABSTRACT

Although recent studies suggest contamination by bacteria and nitrate in private drinking water systems is of increasing concern, data describing contaminants associated with the corrosion of onsite plumbing are scarce. This study reports on the analysis of 2,146 samples submitted by private system homeowners. Almost 20% of first draw samples submitted contained lead concentrations above the United States Environmental Protection Agency action level of 15 μg/L, suggesting that corrosion may be a significant public health problem. Correlations between lead, copper, and zinc suggested brass components as a likely lead source, and dug/bored wells had significantly higher lead concentrations as compared to drilled wells. A random subset of samples selected to quantify particulate lead indicated that, on average, 47% of lead in the first draws was in the particulate form, although the occurrence was highly variable. While flushing the tap reduced lead below 15 μg/L for most systems, some systems experienced an increase, perhaps attributable to particulate lead or lead-bearing components upstream of the faucet (e.g., valves, pumps). Results suggest that without including a focus on private as well as municipal systems it will be very difficult to meet the existing national public health goal to eliminate elevated blood lead levels in children.

Key words | corrosion, drinking water, lead, private drinking water systems, well water

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INTRODUCTION

The United States' Centers for Disease Control and Prevention and the World Health Organization classify lead poisoning as a preventable disease as exposure can be prevented through the direct removal of sources of lead and/or implementation of measures aimed at controlling lead releases. Since the 1970s, various federal regulations, including the phase-out of lead in gasoline and the ban of lead-based paint, have significantly reduced blood lead levels in the United States (Pirkle *et al.* 1994; Jones *et al.* 2009). However, recent reports suggest that exposures from secondary sources (i.e., non-paint sources) have likely been underestimated (Levin *et al.* 2008), and that these exposures can result in elevated blood lead levels in children and perhaps even fetal death (Edwards *et al.* 2009; Brown *et al.* 2011; Edwards 2014). The United States Environmental Protection Agency (USEPA) has estimated that waterborne lead may contribute up to 20% of an adult's total lead exposure and up to 60% of total lead exposure for an infant consuming formula (US Environmental Protection Agency 1993); however, as other sources of exposure are removed from the environment, the relative percentage of exposure related to water is likely rising (Levin *et al.* 2008).

In 1991, the USEPA promulgated the Lead and Copper Rule (LCR) to minimize waterborne lead exposure through the identification and control of corrosion, which is the primary mechanism for lead release in municipal drinking water systems. The LCR states that if more than 10% of high-risk households sampled at the point of use (POU; i.e., consumers' taps) have lead concentrations greater than the 'action level' of 15 μg/L, the utility must take

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actions to control corrosion, and educate the public regarding risks and water-lead avoidance strategies (US Environmental Protection Agency 1993).

Approximately 10–15% of US households are dependent on private drinking water systems (i.e., systems that serve an average of less than 25 individuals for at least 60 days per year and have less than 15 service connections), which by definition are not regulated by the USEPA under the Safe Drinking Water Act (SDWA) or subject to LCR requirements (US Environmental Protection Agency 1993, 2012, 2013). Although some states have regulations related to the initial siting of private wells, these regulations do not address long-term water quality monitoring to ensure safe drinking water. As the monitoring and maintenance of private systems are solely the responsibility of the homeowner, water quality testing is often inconsistent and systematic reporting of water quality at the community or state level is uncommon (Francis *et al.* 1982; Knobeloch *et al.* 2013; Swistock *et al.* 2013). Past studies report that 20–50% of private systems exceed at least one SDWA health-based standard, most often for coliform bacteria (DeSimone 2009; Knobeloch *et al.* 2013; Swistock *et al.* 2013). Contamination due to the corrosion of internal plumbing is rarely assessed, but results from the few studies that measured lead concentrations in 'first draw' samples (i.e., sample collected from the POU following a minimum of 6 hours of stagnation) suggest that corrosion may be a concern in private systems.

The first comprehensive national survey of rural water supplies in the United States (e.g., individual wells and small community systems) conducted in the late 1970s and early 1980s reported that 9.2% of the 2,654 household samples contained lead concentrations in excess of 50 μg/L (Francis *et al.* 1982). During a 1985 investigation of 55 private wells in rural Nova Scotia, 20% of first draws exceeded 50 μg/L (Maessen *et al.* 1985). In 1988, 34% of samples from 605 private systems in North Carolina had lead concentrations greater than 10 μg/L (Maas & Patch 1990); the authors noted that private system homeowners were at greater risk of lead exposure than homeowners connected to municipal systems. A study of over 1,500 private systems in Pennsylvania conducted between 1989 and 1992 reported that 19% of household samples exceeded 15 μg/L lead, with concentrations as high as 2,800 μg/L (Swistock *et al.* 1993). Roughly a decade later, the same group in Pennsylvania observed lead concentrations greater than 15 μg/L in 12% of 251 submitted samples (Swistock *et al.* 2013). Although these studies are limited in scope, they support the need for further, more detailed, investigation of corrosion in private systems.

To more effectively prevent lead release within plumbing networks, recent studies of municipal systems have distinguished and quantified the physical states of lead (i.e., soluble versus particulate lead). Soluble lead is operationally defined as the concentration passing through a 0.45 μm pore size filter, and was previously the sole target of most sampling efforts, often resulting in systematic underestimates of actual lead levels and the potential for human exposure (McNeill & Edwards 2004; Triantafyllidou *et al.* 2007). The incidence of particulate lead (i.e., lead retained by a 0.45 μm filter) during sample collection is considered highly variable and is often associated with higher flow rates and/or hydraulic disturbances (Schock 1990; Triantafyllidou *et al.* 2007; Deshommes *et al.* 2010; Triantafyllidou *et al.* 2013). Studies consistently emphasize that the characterization of particulate lead levels is critical when estimating human exposure and designing appropriate remediation efforts (Triantafyllidou *et al.* 2007; Deshommes *et al.* 2012; Triantafyllidou *et al.* 2013; Clark *et al.* 2014). To date, only one identified study has evaluated particulate lead in private drinking water systems (Swistock *et al.* 1993). The authors concluded that solely quantifying soluble lead may have underestimated lead concentrations by 6–18 μg/L in their 126 samples, emphasizing the importance of designing appropriate sampling strategies to ensure accurate measurement.

Although regulatory mechanisms such as the SDWA and LCR ensure the provision of safe drinking water to homes connected to municipal systems, recent studies have highlighted the need to more deeply examine private drinking water systems where exposure to waterborne contaminants of human health concern are increasingly recognized (Wallender *et al.* 2014). In particular, without characterizing waterborne lead exposure in these systems, achieving the federal goal to eliminate elevated blood lead levels in children by 2020 will be challenging (US Department of Health and Human Services 2014). To better characterize the incidence of lead in samples from these

systems, this study: (1) documents the occurrence of lead in water samples collected from the POU of households dependent on private drinking water systems; (2) quantifies the relative amounts of dissolved and particulate lead in these samples; (3) identifies major system and environmental characteristics associated with high lead concentrations; and (4) evaluates associations between homeowner perception of water quality and the presence of high lead concentrations.

METHODS

Samples were collected via a collaboration with the Virginia Household Water Quality Program (VAHWQP; www.wellwater.vt.edu), a long-standing state-sponsored Cooperative Extension program based at Virginia Tech. Although private water supply systems provide water for several domestic purposes, this research focused on drinking water quality (e.g., samples at the POU) and will refer to these systems as 'private drinking water systems'. As lead was only added to those target contaminants included in the VAHWQP testing package in January of 2012, this partnership represents the first identified investigation of lead concentrations in private drinking water systems in the Commonwealth of Virginia.

Sample collection

Through VAHWQP's county-based drinking water clinics, 2,146 water samples were collected between March 2012 and November 2013. Participation in the drinking water clinics was wholly voluntary and participants were therefore self-selected. Homeowners who wished to participate purchased a water sampling kit that included: (i) sampling instructions on how to properly collect each sample; (ii) a questionnaire about system characteristics, perception of water quality, and household demographics; and (iii) sampling bottles pre-prepared per standard methods (e.g., autoclaved, acid washed) for the collection of water for different analyses. Per the instructions, homeowners collected samples from a non-swivel faucet on a predetermined morning. After a minimum of 6 hours of stagnation, homeowners removed the aerator and collected 250 mL of water at a pencil-thin flow ('first draw'). The system was then flushed for a minimum of 5 minutes, and three additional samples (two 250 mL and one 100 mL) were collected at a pencil-thin flow ('flushed samples'). Two bottles were used to evaluate bacteria and physico-chemical parameters, while the third bottle (250 mL) was evaluated for flushed metal concentrations. After collection, participants brought the samples to a designated location for transportation on ice to Virginia Tech. Samples were processed within 8–12 hours of collection.

Water quality analyses

Samples were processed in the Virginia Tech Biological Systems Engineering Water Quality Laboratory for pH, conductivity (proxy for total dissolved solids), nitrate-N, sulfate, and fluoride per methods 4500-H^+, 2510, and 4110 C (American Public Health Association American Water Works Association & Water Environment Federation 1998). Bacteria (i.e., total coliforms and *Escherichia coli*) were quantified using the IDEXX Colilert 2000 method (www.idexx.com, Westbrook, MN, USA). Metals (aluminum, arsenic, cadmium, calcium, chloride, chromium, copper, iron, lead, magnesium, manganese, nickel, silver, sodium, tin, and zinc) were analyzed in the Virginia Tech Environmental and Water Resources Engineering Water Quality Laboratory using inductively coupled plasma-mass spectrometry (ICP-MS) per method 3125 B (American Public Health Association American Water Works Association & Water Environment Federation 1998). Hardness was calculated based on the concentrations of calcium and magnesium. For data quality assurance and quality control (QA/QC), blanks and spikes of known concentrations were measured every 10–15 samples for all analyses, except in the case of Colilert, where a positive and negative control were included for each clinic's sample set.

Acidification for metals analysis

During the 2012 clinics, metal concentrations were quantified via 10 mL aliquots of the first draws and flushed samples; this practice of sample transfer was previously used by extension to quantify metals and has been used by several municipalities to quantify lead (Freud 2012; Triantafyllidou *et al.* 2013; Etchevers *et al.* 2015). The first draw

and flushed aliquots were acidified with 2% nitric acid and digested for a minimum of 16 hours. Owing to the unexpectedly high levels of lead observed during the first year of sampling, the 2013 sampling protocol was modified to permit acidification in the original sampling bottle in keeping with the USEPA LCR protocol.

Evaluating systematic error and quantifying particulate lead

To evaluate the systematic error associated with acidifying a sample aliquot (2012 method) rather than the full bottle (2013 method), and to quantify particulate lead, 20% of the first draws and flushed samples submitted during 2013 were selected for additional analyses. Samples with visible particles, discoloration, and/or odor were preselected for this analysis, with a remaining number of samples randomly selected to reach a total of 20% of samples submitted. Two 10 mL aliquots were pipetted from each sample: the first aliquot was used to evaluate the 2012 method, and the other aliquot was filtered through a 0.45 μm pore size Whatman nylon filter to quantify dissolved lead. Both aliquots and the remaining sample (~230 mL) were acidified with 2% nitric acid and digested for a minimum of 16 hours prior to ICP-MS analysis. Note that total lead concentrations were used exclusively throughout this study, except for the 'Occurrence of particulate lead' section, which discusses both particulate and total lead concentrations.

Statistical analysis

All statistical analyses were conducted in R version 3.0.2 (R Development Core Team 2012) assuming an alpha of 0.05 as an indication of significance. Owing to the non-normal distribution of the lead data (Shapiro-Wilk; $p < 0.05$), nonparametric statistics were used throughout this study. Potential associations between lead concentrations and the other water quality parameters (e.g., copper, zinc, and tin) were evaluated using the Spearman's rank correlation (ρ), while the Wilcoxon signed-rank test and Kruskal–Wallis test compared lead concentrations based on categorical household characteristics (e.g., income, education, piping material) and homeowner perception of water quality (e.g., taste, odor). Odds ratios (OR) were calculated to determine the odds of having elevated lead concentrations based on homeowners' perceptions of water quality.

RESULTS AND DISCUSSION

Overview of participating systems

Between March 2012 and November 2013, 2,146 water samples from 61 of the 95 counties in Virginia were submitted for analysis through the VAHWQP Cooperative Extension program. As participation in clinics was dependent on local interest and awareness, the number of samples submitted per county varied substantially. Of the samples submitted, 94.5% were collected from a private well ($n = 2{,}029$), 3.6% from a spring ($n = 78$), less than 0.1% from a cistern ($n = 2$), and the remaining 1.7% of samples were either from a municipal system or the source was not identified ($n = 37$). Of the 2,029 wells, 79% were 'drilled', 12% were 'dug/bored', and the remaining 9% were 'unknown'. Response rates regarding the year the well was constructed and the depth of the well were somewhat low (69% and 59% responding, respectively). The average year of construction reported was 1988, although responses ranged from 1850 to 2013. Reported well depths varied from 1.2 m to 381 m, with a mean and median depth of 77.7 m and 67.0 m, respectively. Roughly half (54%) of homeowners indicated that their system included some sort of treatment, with the most common type of treatment being a water softener (23%) or a sediment filter (29%).

Although previous studies report infrequent rates of water quality testing (Francis *et al.* 1982; Swistock *et al.* 1993), 55% of homeowners submitting samples to this program indicated that they had previously tested their water. Reported household demographics suggested that homeowners who participated in the VAHWQP drinking water clinics were largely older, more affluent, 'White or Caucasian', and had a college or graduate-level education. Based on self-reported household income ($n = 1{,}796$), 51% of homeowners reported an annual household income greater than $65,000, and only 8% reported an income in the lowest category (i.e., below $24,000). With respect to education

($n = 1{,}983$), 69% of homeowners received a college degree with 37% having obtained a post-college degree (i.e., masters or doctoral). The population sampled also tended to be older ($n = 2{,}024$) as 81% were older than 50 and 59% of homeowners were older than 60. During the 2013 clinics, self-reported race was added to the questionnaire ($n = 1{,}115$), and 89% of homeowners self-identified as being 'White or Caucasian'. In addition to evaluating the population participating in this extension water quality testing, these results are important in the context of lead exposure as they highlight the low rate of participation in this water quality testing among young adults (<50 years old) most likely to have children under the age of six, who are at greatest risk for detrimental health effects following lead exposure.

Overall prevalence of contamination

Using the SDWA standards for municipal systems as guidelines, 58% of submitted samples ($n = 1{,}250$) exceeded at least one Maximum Contaminant Level (MCL) (Table 1). Bacterial contamination was the most common issue, with 46% of systems testing positive for total coliforms. Using the action level for lead and copper as a threshold, 19% of systems had elevated lead concentrations (≥15 μg/L) and 12% had elevated copper concentrations (≥1.3 mg/L) in the first draw. Twenty-six percent of households ($n = 553$) had a pH value outside the recommended 6.5–8.5 range, and 89% of these pH values ($n = 491$) were below 6.5. Values exceeding other water quality constituents targeted by VAHWQP were relatively rare.

Table 1 | Summary statistics of water quality parameters in samples collected from private drinking water systems

Target water quality constituent	Standard		Number of observations	Mean	Median	90th percentile	Percent exceeding
[a]Arsenic, in mg/L	MCL	0.010 mg/L	2,146	BDL	BDL	BDL	0.1%
[b]Cadmium, in mg/L		0.005 mg/L	2,144	BDL	BDL	BDL	0.6%
[a]Chromium, in mg/L		0.1 mg/L	2,144	BDL	BDL	0.001	0.0%
[a]Fluoride, in mg/L		4.0 mg/L	2,146	BDL	BDL	BDL	0.4%
[a]Nitrate, mg/L		10 mg/L	2,146	1.5	BDL	4.2	1.3%
[a]Total coliforms, in cfu		ABSENT	2,143	–	–	–	46%
[a]*E. coli*, in cfu		ABSENT	2,143	–	–	–	10%
[b]Copper, in mg/L	Action	1.3 mg/L	2,144	0.677	0.153	1.727	12%
[b]Lead, in mg/L	Level	0.015 mg/L	2,144	0.022	0.004	0.027	19%
[b]Aluminum, in mg/L	SMCL	0.05–0.2 mg/L	2,144	0.047	0.003	0.061	3.8% (12%*)
[a]Chloride, in mg/L		250 mg/L	1,378	14.3	5.4	31.4	0.2%
[a]Copper, in mg/L		1.0 mg/L	2,144	0.677	0.153	1.727	15%
[a]Fluoride, in mg/L		2.0 mg/L	2,146	BDL	BDL	BDL	2.7%
[a]Iron, in mg/L		0.3 mg/L	2,146	0.142	0.008	0.234	8%
[a]Manganese, in mg/L		0.05 mg/L	2,146	0.032	0.002	0.049	10%
[a]pH, in standard units		6.5–8.5	2,146	7.1	7.2	8.0 (6.0[c])	26% (23%**)
[b]Silver, in mg/L		0.1 mg/L	2,144	BDL	BDL	BDL	0.0%
[a]Sulfate, in mg/L		250 mg/L	2,146	45.1	8.0	52.3	2.4%
[a]TDS, in mg/L		500 mg/L	2,146	237.1	179.6	501.1	10%
[b]Zinc, in mg/L		5 mg/L	2,144	0.868	0.231	2.247	3.1%

MCL: Maximum Contaminant Level – associated with risk to human health; SMCL: Secondary Maximum Contaminant Level – associated with aesthetic considerations; BDL: below detection limit; cfu: colony forming units.

[a]Total concentration measured in the flushed sample.

[b]Total concentration measured in the first draw.

[c]10th percentile.

*Percent above 0.05 mg/L.

**Percent below pH of 6.5.

Lead concentrations

Compliance with the LCR requires that no more than 10% of samples exceed the action level. As each private system is an independent distribution system and only one first draw sample was collected during this study, for comparison purposes, the action level was used as a threshold. Lead concentrations in the first draws ranged from below detection (<1 μg/L) to 24,740 μg/L (Figure 1(a)). Eighty percent of households had detectable lead concentrations (≥1 μg/L) and 19% of samples exceeded the lead action level of 15 μg/L.

Flushing the system for 5 minutes appeared to reduce lead concentrations to the recommended concentration (i.e., below 15 μg/L) for most households in this study; less than 1% of flushed samples exceeded 15 μg/L, and 74% of samples had non-detectable (<1 μg/L) concentrations (Figure 1(b)). However, 2% of the households ($n = 46$) had higher lead concentrations in the flushed sample relative to the first draw (Figure 2; i.e., above the 1:1 line). For most of these 46 households, flushed concentrations increased by a relatively small amount (mean and median increase of 5.2 μg/L and 1.0 μg/L, respectively), but in eight homes increases were >5 μg/L (max observed increase = 130 μg/L). In households with higher flushed concentrations, there may be components upstream of the faucet that are responsible for lead release, such as brass valves (Sandvig *et al.* 2008) or submersible pumps (Maas *et al.* 1998), and/or particulate lead may have been mobilized (Schock 1990).

Occurrence of particulate lead

To better understand patterns and mechanisms of lead release, 20% of first draw and corresponding flushed samples submitted to the 2013 drinking water clinics ($n = 253$) were filtered through a 0.45 μm filter to quantify dissolved lead. As previously mentioned, samples with noticeable characteristics (e.g., odor, flakes; $n = 69$) were preselected for this analysis, which therefore may represent a worst-case scenario for estimation of particulate lead. The remaining quantity ($n = 184$) were randomly selected. Four samples were removed from this analysis as these samples did not meet associated QA/QC checks. Particulate lead was calculated as the difference between total lead and dissolved lead.

On average, for the preselected first draws with detectable total lead concentrations (≥1 μg/L, n = 55), 75% of the total lead was in the particulate form, although some samples were entirely composed of particulate lead (Figure 3(a); i.e., >99%). For the randomly selected first draws with detectable total lead concentrations (≥1 μg/L, $n = 143$), 47% of the total lead was in the particulate form, and again, some samples were composed entirely of particulate lead (i.e., >99%). The median lead concentration for preselected first draws ($n = 67$,

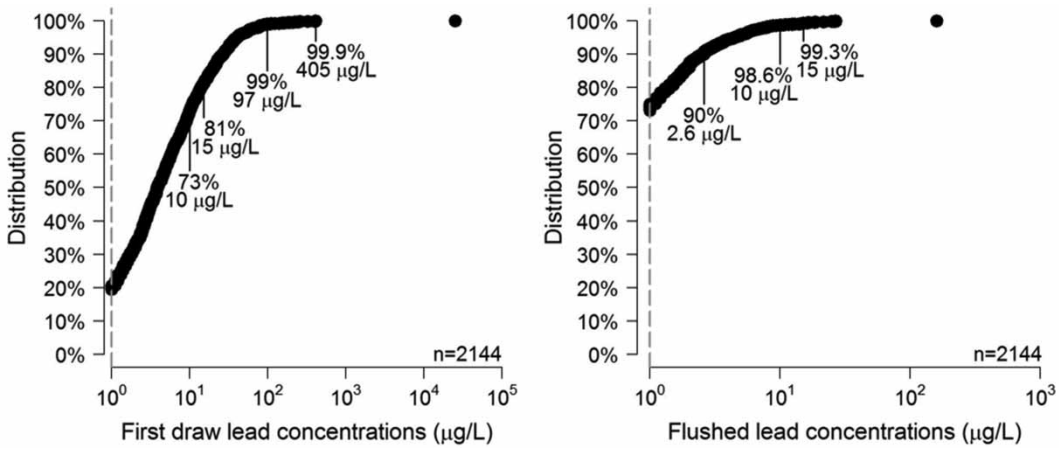

Figure 1 | Lead concentrations in (a) first draws and (b) flushed samples collected during the 2012 and 2013 VAHWQP drinking water clinics. Dashed lines represent the detection limit (1 μg/L).

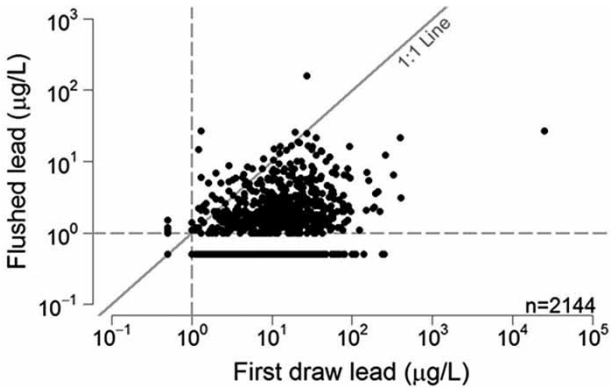

Figure 2 | First draw lead concentrations compared to lead concentrations after 5 minutes of flushing. Dashed lines represent the detection limit (1 µg/L). Samples below the detection limit were set to 0.5 µg/L.

5.2 µg/L) was significantly higher (Wilcoxon signed-rank tests, $p < 0.05$) than randomly selected first draws ($n = 183$, 1.1 µg/L). Only 30% of the corresponding flushed samples ($n = 76$) measured detectable lead concentrations, but 62% of the total lead was in the particulate form (Figure 3(b)).

Overall, the fraction of particulate lead in the first draw did not increase with higher total lead concentrations. Households with elevated total lead concentrations had varying fractions of particulate lead ranging from non-detectable to 99% composition (Figure 3(a)). This is consistent with prior work indicating that the occurrence of particulate lead is variable and unpredictable in public water systems, even when sampling is rigorously controlled (Triantafyllidou *et al.* 2007; Sandvig *et al.* 2008; Triantafyllidou *et al.* 2013; Clark *et al.* 2014).

When considering the systematic error associated with acidifying a sample aliquot (2012 method) rather than acidification in the bottle (2013 method), it is important to note that differences between subsample aliquots and total lead values were less than 1 µg/L for 64% of first draws and 92% of flushed samples. The first draw subsamples that under- or overestimated total lead ($n = 92$) were primarily composed of particulate lead (mean 70% particulate form), and had a mean absolute error of 11.0 µg/L (max under- and overestimate of 96 µg/L and 39 µg/L, respectively). Flushed subsamples that under- or overestimated total lead ($n = 20$) were also primarily composed of particulate lead (mean 75% particulate form), and had a mean absolute error of 3.6 µg/L. Given the unpredictability of particulate lead release and the potential high associated error noted in both this study and previous studies of municipal water (Freud 2012; Triantafyllidou *et al.* 2013), acidification in the bottle is highly recommended to ensure an accurate measure of lead concentration.

Potential factors predictive of elevated lead concentrations in first draw samples

System characteristics

The median lead concentration for dug/bored wells ($n = 248$, 9.4 µg/L) was significantly higher (Kruskal–Wallis test, $p < 0.05$) than drilled wells ($n = 1{,}607$, 3.6 µg/L) and springs ($n = 77$, 3.5 µg/L). While 32% of samples from

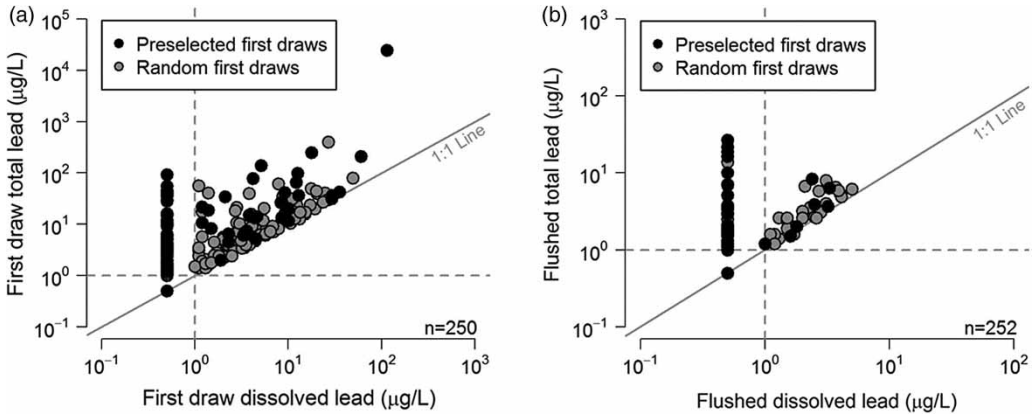

Figure 3 | Presence of particulate lead in (a) first draws and (b) flushed samples. The presence of samples along the 1:1 line indicated that 100% of the lead was in the dissolved form (i.e., total and dissolved concentrations were equal). Dashed lines represent the detection limit (1 µg/L). Samples below the detection limit were set to 0.5 µg/L.

dug/bored wells exceeded the 15 μg/L action level, only 17% and 14% of drilled wells and springs exceeded this value, respectively. Swistock *et al.* (1993) observed higher lead concentrations in springs and dug wells compared to drilled wells, which the authors attributed to the acidity of shallow groundwater. This study did not observe a correlation between self-reported well depths and lead concentrations ($n = 1{,}204$, $\rho = -0.25$), but lead concentrations were negatively correlated with pH values ($\rho = -0.42$, $p < 0.05$). pH varied significantly (Kruskal–Wallis test, $p < 0.05$) based on system type. The median pH for dug/bored wells (pH 6.2) was significantly lower than for springs (pH 6.9), which was significantly lower than for drilled wells (pH 7.2). Not surprisingly, 63% of dug/bored wells, 29% of springs, and 17% of drilled wells had a pH below 6.5. There were no correlations between lead concentrations and fluoride, hardness, nitrate, sodium, sulfate, or total dissolved solids (TDS).

Participants who indicated the use of a water treatment device did not have significantly lower median lead concentrations (Wilcoxon signed-rank tests, $p = 0.84$). Consistent with the literature, most systems that included some form of water treatment had a sediment filter (29%) or water softener (23%). Reverse osmosis units and acid neutralizers are recommended treatments for households with corrosion, but only 2.6% and 4.8% of homeowners, respectively, indicated having installed these treatment devices. As this study did not independently verify or evaluate the maintenance of these treatment units, determining the influence of specific treatment types on lead concentrations was not possible. Although past studies have observed that bacterial contamination in private systems is more prevalent in lower income and education households (Francis *et al.* 1982; Smith *et al.* 2014), in the present study there was no significant difference in lead concentrations based on household income ($p = 0.57$), education ($p = 0.55$), race ($p = 0.21$), or age ($p = 0.23$).

Plumbing components

There was no significant difference (Wilcoxon signed-rank tests, $p = 0.21$) in lead concentrations between households that identified only having copper ($n = 514$) versus plastic plumbing ($n = 915$). Given that several studies estimate that the volume of water contained within the faucet and the connective piping is approximately 125 mL (Sandvig *et al.* 2008), and the first draw in this study was 250 mL, high concentrations in first draws may reflect leaching from brass within the faucet. Brass is an alloy composed mainly of copper (60–80%), zinc (4–32%), and lead (2–8%), but can contain other metals such as tin, nickel, aluminum, iron, or cadmium (Lytle & Schock 1996). In keeping with this hypothesis, lead concentrations were strongly correlated ($p < 0.05$) with zinc ($\rho = 0.67$), copper ($\rho = 0.61$), and nickel ($\rho = 0.56$), and weakly correlated ($p < 0.05$) with aluminum ($\rho = 0.27$). Based on these findings, the interior piping material (e.g., copper, plastic) may not be as important in predicting lead concentrations as the type of fittings used in the plumbing network (e.g., brass).

Households constructed pre-1988 had a significantly higher ($p < 0.05$) median lead concentration (5.4 μg/L, $n = 600$) compared to households constructed post-1988 (3.3 μg/L, $n = 805$). This is in keeping with Swistock *et al.* (2013), who observed that the majority of households with elevated lead concentrations had copper plumbing installed before 1991. This may reflect the 1986 Lead Ban, which required the use of 'lead-free' plumbing components in the installation or repair of any municipal system or residential and non-residential buildings connected to a municipal system after June 1988 (US Environmental Protection Agency 1993). While the Lead Ban was not directly aimed at private systems, these 'lead-free' plumbing components are being installed in private systems and reducing the risk of lead release. It is important to note that as of January 2014, the Reduction of Lead in Drinking Water Act lowered the allowable lead content in 'lead-free' plumbing components, which should reduce lead concentrations in newly constructed or repaired private drinking water systems (US Environmental Protection Agency 2014).

Perception

Although lead cannot be directly identified by smell or taste in drinking water (US Environmental Protection Agency 1993), previous studies suggest lead is associated with parameters (e.g., pH) that have discernable physical characteristics (e.g., metallic taste, blue-green stains, and obvious signs of corrosion) (Swistock *et al.* 1993). OR

describing relationships between homeowner-reported perceptions of water quality and observed lead concentration in the first draws analyzed in this study are illustrated in Figure 4.

Homeowners who identified obvious signs of corrosion (OR = 1.72), blue-green staining on plumbing fixtures (OR = 2.78), and/or described the taste of water as metallic (OR = 2.29) were 1.7–2.8 times more likely to have elevated lead concentrations compared to homeowners who did not identify these characteristics. Median lead concentrations for homeowners who identified these characteristics were also significantly higher (Wilcoxon signed-rank tests, $p < 0.05$). As lead was correlated with pH and copper (most likely leaching from brass), these indicators are most likely linked to copper corrosion and lower pH values. Homeowners who noted that their water had an odor (OR = 0.62), a sulfur odor (OR = 0.49), a sulfur taste (OR = 0.42), identified white/chalk staining on plumbing fixtures (OR = 0.56), and/or noticed white flakes in the water (OR = 0.40) were 1.6–2.5 times less likely to have elevated lead concentrations compared to homeowners who did not identify these characteristics. Median lead concentrations for homeowners who identified these characteristics were also significantly lower (Wilcoxon signed-rank tests, $p < 0.05$), except for white/chalk staining ($p = 0.06$). As median lead concentrations were not significantly different for white/chalk staining on plumbing fixtures, this characteristic may not be a preferred indicator. These characteristics are most likely associated with the formation of passivating films or scales, which could inhibit corrosion (Oliphant 1983; Sandvig *et al.* 2008).

Given that previous studies have linked perception of water quality issues with increased stewardship, these observations may be useful in designing public health messaging (Imgrund *et al.* 2011); although it is important to note that an individual's perception of drinking water quality is highly variable and can be influenced by perceived risk, familiarity, chemical composition, and demographics (Cuppett *et al.* 2006; Doria *et al.* 2009; Doria 2010; Mirlohi *et al.* 2011).

LIMITATIONS

As previously discussed, this work represents a collaborative effort with an ongoing extension effort. Homeowners volunteered to participate and therefore, had both the knowledge and resources to procure water quality testing, which may have introduced sample bias. It is unknown if the rates of contamination were elevated as concerned homeowners sought testing, or reduced as a specific demographic was primarily targeted. The sampling protocol was designed to quantify contamination commonly found in private drinking

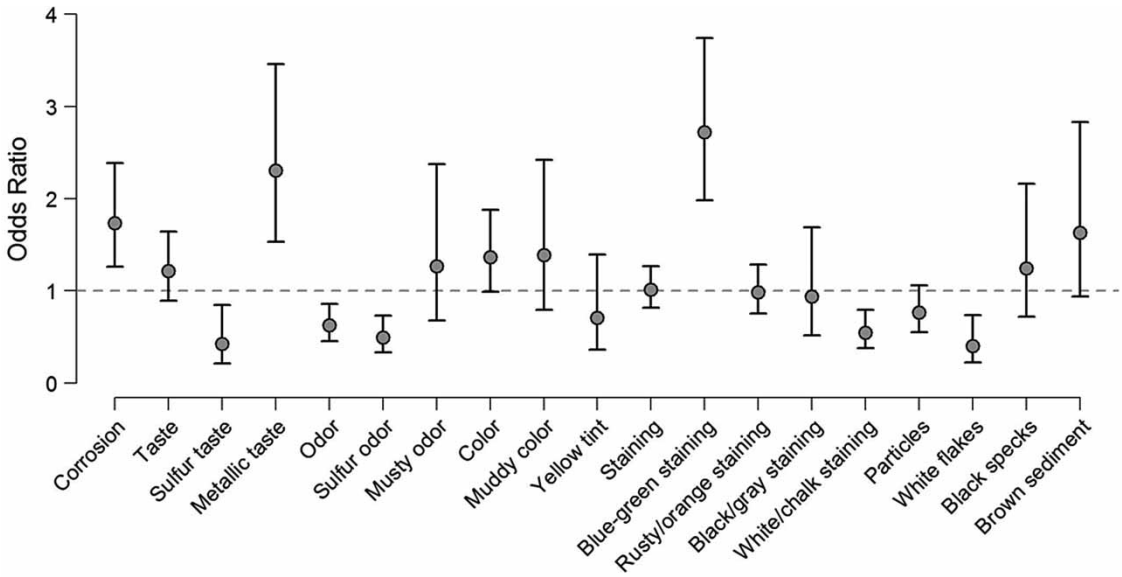

Figure 4 | Odds ratio for physical characteristics identified by homeowners.

water systems, with bacteria as a primary interest. Homeowners were instructed to collect samples from a non-swivel faucet with the aerator removed as these characteristics are believed to increase the probability of bacterial contamination. Without an aerator attached, homeowners were also instructed to collect samples at 'low pencil-thin flow' to minimize the amount of water missed during collection; collecting at 'low flow' may have resulted in an underestimation in particulate lead. The origin of the sample collected after 5 minutes of flushing was unknown and depended greatly on the characteristics of the system (e.g., presence of treatment, length of piping). With lead-bearing components upstream of the faucet and within the well, lead concentrations are highly dependent on location. Questionnaire information was voluntarily provided by the homeowner, and has not been verified. When quantifying particulate lead, samples were preselected based on physical characteristics, which may have introduced bias and resulted in the worst-case estimation of particulate lead, as discussed previously.

CONCLUSIONS

Almost 60% of samples submitted during this study exceeded at least one USEPA health-based standard for municipal systems. In keeping with past studies targeting private systems, bacterial contamination was primarily responsible for these violations, although metal leaching due to corrosion was not uncommon. Samples provided by 19% of households had lead concentrations above the USEPA action level of 15 μg/L. Average particulate lead composition was 47% for the randomly selected subsample set, and 75% for the preselected first draws. Particulate lead, on average, composed 62% of the total lead in corresponding flushed samples. The occurrence of particulate lead was highly variable, and future studies are highly encouraged to quantify all forms of lead in sampling and analytical procedures in order to ensure an accurate measure of potential human exposure.

Perhaps not surprisingly given the 1986 Lead Ban, households constructed before 1988 had higher lead concentrations; however, it is important to note that elevated lead concentrations were still observed in residences constructed after 1988. Lead appeared most likely to be leaching from the brass within faucets and/or fittings as lead concentrations were highly correlated with copper and zinc. There was no significant difference in lead concentrations based on identified plumbing materials (i.e., copper and plastic), which further supports that lead is leaching from brass fittings and fixtures. However, with the Reduction of Lead in Drinking Water Act, lead release from brass components should be reduced. Although elevated concentrations of lead were observed in samples from all types of systems, samples from dug/bored wells had significantly higher lead concentrations. This finding is likely due to the acidity of groundwater associated with shallow depths as pH values were also significantly lower in these wells. The influence of geology was not assessed during this study, but may be an important variable in understanding lead release as a large percentage of private systems do not use treatment that adjusts the acidity of the source water. Lead cannot be identified by smell or taste in drinking water, but increased concentrations were associated with obvious signs of corrosion such as blue-green staining and metallic taste, which may be useful in communicating the potential risks of corrosion and promoting private system maintenance. The demographic data highlighted the primary population sampled, which did not encompass a large portion of young adults (<50 years old). As children are at greatest risk of lead poisoning, future efforts to prevent waterborne lead exposure should focus on engaging this population.

For most of the private systems sampled in this study, flushing for 5 minutes reduced lead concentrations below 15 μg/L, although 2% of households experienced an increase in lead concentrations with flushing. This finding suggests that there may be other components within the plumbing network that release lead and/or particulate lead may have been mobilized. To develop effective remediation efforts and sampling protocols, an increased understanding of the mechanisms of lead release is essential.

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