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Arsenic in groundwater in private wells in rural North Dakota and South Dakota: Water quality assessment for an intervention trial

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Abstract

Elevated exposure to arsenic disproportionately affects populations relying on private well water in the United States (US). This includes many American Indian (AI) communities where naturally occurring arsenic is often above 10 µg/L, the current US Environmental Protection Agency safety standard. The Strong Heart Water Study is a randomized controlled trial aiming to reduce arsenic exposure to private well water users in AI communities in North Dakota and South Dakota. In preparation for this intervention, 371 households were included in a community water arsenic testing program to identify households with arsenic ≥ 10 µg/L by inductively coupled plasma mass spectrometry (ICP-MS). Arsenic ≥ 10 µg/L was found in 97/371 (26.1%) households; median water arsenic concentration was 6.3 µg/L, ranging from < 1–198 µg/L. Silica was identified as a

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Authors’ contributions

MP made substantial contributions to conception and design, collection of data, analysis and interpretation of data. JY, ET, MP, JS, MO, AB conducted and collected field data. KH, CM constructed GIS maps. PO, RC, AR, KS, LM conducted laboratory testing. DH, LGB provided guidance. CMG, ANA, MO, LGB designed SHWS study. CMG, ANA made substantial contributions to conception and design and interpretation of data. All authors read and approved the final manuscript.

Declarations/Ethics approval and consent to participate

The Indian Health Service, institutional review boards, and participating Tribal IRBs approved the study protocol. All participants provided informed consent.

Consent for publication

We have obtained consent from participating Tribal IRBs for publication of results.

Availability of data and material: All data generated or analyzed during this study are included in this published article.

Competing interests

The authors declare no financial or non-financial competing interests.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.envres.2018.09.016.

water quality parameter that could impact the efficacy of arsenic removal devices to be installed. A low-range field rapid arsenic testing kit evaluated in a small number of households was found to have low accuracy; therefore, not an option for the screening of affected households in this setting. In a pilot study of the effectiveness of a point-of-use adsorptive media water filtration device for arsenic removal, all devices installed removed arsenic below 1 µg/L at both installation and 9 months post-installation. This study identified a relatively high burden of arsenic in AI study communities as well as an effective water filtration device to reduce arsenic in these communities. The long-term efficacy of a community based arsenic mitigation program in reducing arsenic exposure and preventing arsenic related disease is being tested as part of the Strong Heart Water Study.

Keywords

Arsenic; Private well; American Indian; Drinking water; Intervention; Field test kit

1. Background

In the United States, the estimated population using domestic wells for drinking water with arsenic (As) levels above the Environmental Protection Agency (EPA) maximum contaminant level (MCL) of 10 µg/L is 2.1 million people (Ayotte et al., 2017), with an additional 384,000 people exposed through public water systems (EPA, 2017). Arsenic is a highly toxic and carcinogenic element, classified by the US Agency for Toxic Substances and Disease Registry as the top substance on its priority list of hazardous substances (ATSDR, 2015). Millions more are exposed to arsenic in drinking water below the MCL, which is of concern as the evidence suggests no safe threshold exists (Schmidt, 2014). Increasing evidence supports the role of low-moderate arsenic (< 50 µg/L) in cancer and cardiovascular disease, and potentially diabetes, neurodevelopmental toxicity, immune effects and nonmalignant respiratory disease (Council, 2013).

Private wells do not fall under the jurisdiction of the EPA's Safe Drinking Water Act, and it is the responsibility of the well owner to make sure their water is safe. Major barriers to arsenic water testing include general awareness, geographic distance to a laboratory and lack of access to water testing (Navas-Acien et al., 2009), and the high price point per test (~20 USD per sample) (Shaw et al., 2005; Flanagan et al., 2015a, 2015b). Rural and suburban families relying on ground water, including many in American Indian (AI) communities, are the most affected, especially in the Southwest, Midwest and Northeast US (Focazio et al., 2000). The concentration of naturally occurring arsenic in ground water varies regionally due to a combination of climate and geology, with greater concentrations found in certain areas of the US including the Interior Plains (Welch et al., 2016). In AIs who participated in the Strong Heart Study (SHS), an ongoing cohort study of cardiovascular disease and its risk factors among AIs, arsenic has been associated with an increased risk of cardiovascular disease, diabetes, kidney disease, and some cancers (Moon et al., 2013; Best et al., 2015; Zheng et al., 2015), highlighting the urgency of preventing arsenic exposure for those relying on private wells.

The Strong Heart Water Study (SHWS), the first randomized controlled trial of an arsenic intervention in the US, was developed to reduce arsenic exposure in AI communities in North Dakota and South Dakota. High arsenic in ground water in aquifers composed of felsic volcanic rocks is found in these areas (Welch et al., 2016). All participating households receive a point-of-use filter to remove arsenic from drinking and cooking water and are randomly assigned to receive an intensive health promotion program or the standard program. In preparation for the intervention, a water quality assessment was performed between February 2014 and January 2018 to achieve the following objectives: (1) to identify households with private wells with elevated arsenic that are eligible for the SHWS using inductive coupled plasma mass spectrometry (ICP-MS); (2) to assess water quality parameters that could interfere with the efficacy of the point-of-use adsorptive media filter and (sub-objective 1) to evaluate the accuracy of the Arsenic Econo-Quick™ (Industrial Test Systems, Inc.) test as a potential rapid screening tool for eligible households; and (3) to evaluate the efficacy of the selected point-of-use adsorptive media filter in study households.

2. Methods

2.1. Study area and private well testing

The study area consists of households using private wells for drinking water in three Tribal Nations in South Dakota and North Dakota, communities referred to as A, B, and C (as requested by the communities which prefer their names are not made public). Convenience sampling was performed by study team members to test domestic wells from 2014 to 2018 (N = 371). Strategies used to identify those homes included word of mouth, health fairs, radio, community member contacts, and a database of previously tested wells. In November 2015, a more detailed water quality assessment was performed in 29 households, chosen by convenience sampling and willingness to participate. Of those 29 households, 19 had also been tested for arsenic in February/March 2014, allowing the examination of arsenic temporality in the study area.

2.2. Water sample collection

Water samples were collected from the kitchen faucet using a sampling and analysis plan developed from EPA guidelines (U.S. EPA, 2016). Water sampling locations were determined by a handheld GPS (Global Positioning System) navigator (Garmin eTrex 10). Water samples for ICP-MS arsenic testing (Objective 1) were collected in 20 mL scintillation vials. Duplicate samples were collected from each well, however only one sample has been analyzed from each well to date. For detailed water quality analyses (Objective 2), acid-washed 125 mL plastic bottles were used to collect filtered samples for iron and phosphorus testing; non-acid-washed bottles to collect samples for alkalinity, sulfate, and silica; a non-acid-washed bottle to collect filtered samples for nitrate; a 20 mL scintillation vial to collect samples for arsenic, uranium, lead, and cadmium for ICP-MS analysis; arsenic speciation filters to collect samples for ICP-MS As(III) analysis; and a multi-parameter water quality sonde (YSI Inc./Xylem Inc.) to test for pH. To test a rapid water arsenic screening kit (Objective 3), water samples were collected in an acid-washed plastic beaker and then poured into reaction bottles (clear PVC) supplied in the arsenic testing kits.

2.3. Objective 1: ICP-MS testing of 342 households in the study area for arsenic 10 µg/L (2014–2018)

Water samples (n = 342) were transported to Mid Continent Testing Labs, Inc. (Rapid City, South Dakota) for ICP-MS arsenic analysis. The number of samples tested by Mid Continent per community was 319, 15, and 8 in communities A, B and C, respectively from 2014 to 2018. The larger number of samples in Community A is related to more extensive sampling in Community A, which is the focus of the SHWS intervention. The quality assurance program employed by the laboratory has a variety of checks to ensure accuracy of results. Prior to each day's use, the ICP-MS was run through an optimization and mass calibration, selecting the optimum instrument parameters for the day. The instrument was calibrated by testing various standards, in the case of arsenic analysis, using 2, 5, 10, 20, and 100 ppb standard solutions. The calibration was then checked on two 50 ppb stock solutions. The first is the Initial Calibration Verification (ICV), prepared from a different source from the calibration standards. The second is the Continuing Calibration Verification (CCV), prepared from the same source as the calibration standards. Both must show a value of 50 ppb, within $\pm 10\%$ of expected value. The CCV was checked at least every 10 samples tested throughout the day. A blank DI solution was also checked, the Initial Calibration Blank (ICB). Mid Continent also uses a Matrix Spike and a Matrix Spike Duplicate to ensure accuracy. At least every 10 samples, a small concentration of arsenic, 25 ppb, was added to two aliquots of the sample, and should get 90–110% recovery from the spiked samples. For any samples that are digested, the lab also carries a Blank and Fortified Blank through the process to demonstrate proper recovery and lack of sample contamination through digestion. During all calibration and sample testing, a constant flow of small amounts of internal standards, rare earth elements, were injected into the solutions being tested. These values needed to stay within a certain range and help prove there were no erroneous test results from matrix interference. All analyses were assessed to the laboratory detection limit less than or equal to 1 µg/L. Arsenic results were disseminated to study participants after ICP-MS testing was performed; participants were delivered a letter by a study team member with their signature as proof of receipt.

2.4. Objective 2: intensive water quality assessment in 29 households (2015)

In addition to arsenic, the following additional water quality parameters were tested in 29 households in 2015: arsenic speciation, nitrate, iron, sulfate, phosphorus, alkalinity, silica, pH, uranium, cadmium, and lead. The testing parameters were selected based on historic Indian Health Service data and known compounds that interfere with filtration (U.S. E, 2015). Nitrate, iron, sulfate, phosphorus, alkalinity, and silica water samples were brought back to a semi-controlled temporary laboratory and tested on a Hach DR 2800 portable spectrophotometer at the end of every sampling day. For silica samples that were over the limit of detection, a 1:100 dilution with deionized water was performed. Lead, uranium, cadmium, arsenic, and arsenic speciation water samples were stored at room temperature in a dry lab and shipped back to Johns Hopkins University Trace Metals Laboratory for ICP-MS analysis. The samples were acidified in a 1:1 dilution with 10% optima grade HNO₃ (Fisher Scientific, Columbia MD) and allowed to digest at room temperature for 48 h. The acidified samples were diluted with 2% HNO₃ and 0.5% HCl (Fisher Optima Trace Element Grade) in ultra-pure Milli-Q water and vortexed prior to analysis. A calibration curve for the

element tested (As, Pb, Cd, U) was built using an appropriate element standard solution (Multi-element Aqueous CRM, QC Standard 21. VHG Labs, Manchester, NH, US). Ge, In and Bi were added as an internal standard (CPI International, Santa Rosa, CA, US) for samples and calibration curves to control potential drifts in the elements tested signal. All elements were analyzed using ICP-MS (Agilent 7500ce Octopole ICP-MS, Agilent Technologies, Santa Clara, CA, US). For quality control purposes, a drinking water standard reference material was used (Standard B Water TMDW-B, High Purity Standards, Charleston, SC, US). The results of the QCs were within $\pm 10\%$ of the stated value. Ten-percent of reagent blanks and duplicates were also carried out for quality control. To measure the arsenic species arsenite (AsIII) and arsenate (AsV), syringe filters (Millipore PVDF Sterile Syringe Filter Unit, 0.45 μm , 33 mm Diameter) and arsenic speciation cartridges (MetalSoft Center) were used.

In sub-objective 1, arsenic rapid testing kits were chosen to be tested in 23 households in 2015 because of their potential ability to inexpensively measure arsenic in drinking water in 12 min and identify affected households. The Econo-Quick™ test brand was selected because previous studies conducted found high accuracy (Flanagan et al., 2015a; George et al., 2012, 2014; van Geen et al., 2014a; Steinmaus et al., 2006; Kinniburgh and Kosmus, 2002). Two Arsenic Econo-Quick™ test kits were evaluated: a low-range ($< 1 - > 160 \mu\text{g/L}$) (kit # 481303) and high-range ($0 - 1000 \mu\text{g/L}$) (kit # 481298). The test utilizes a modified Gutzeit method: inorganic arsenic compounds present in water samples are converted to arsine gas which reacts with mercuric bromide on the test strip to form mixed mercury halogens causing a color change from white to yellow or brown (Kinniburgh and Kosmus, 2002; Rapid Arsenic Test Kit Instruction Booklet, 2013). The test is qualitative, with the color intensity proportionally related to the arsenic concentration in the sample. The color on the test strip is visually compared against a colorimetric standard (Fig. 1) by the tester.

Even though recommended by the manufacturer, we did not use reagent #2 because of health concerns due to potassium peroxymonosulfate and potassium peroxydisulfate, which may cause irritation to skin or eyes or breathing difficulties if inhaled (Potassium persulfate; SDS. No. 228036; Potassium peroxymonosulfate; SDS. No. 379824). The practice of removing reagent #2 has shown high accuracy in comparison to ICP-MS measurements previously in Bangladesh (van Geen et al., 2014b). To manage waste in the field, our research team separated the reagents from the liquid, once tests were completed, using a coffee filter over a 5-gallon plastic bucket (aspyrx, 2017), separating out the zinc and stopping production of hydrogen and arsine gases. The used filters were stored in a plastic bag for future hazardous waste disposal. One team member ran a high-range test and one team member ran a low-range test at each household, for a total of two tests per household. Testers were blinded to the results of the other.

To assess for a potential batch effect of the rapid test kits, laboratory experiments were performed with arsenic-spiked samples of known concentrations using the same kits that were used in field experiments. Five samples of Milli-Q water were spiked with sodium arsenate dibasic heptahydrate to concentrations of: 1, 5, 10, 20, and 50 $\mu\text{g/L}$. The prepared samples were prepared under strict and well-established laboratory conditions but were not

tested by ICP-MS. One tester, blinded to the arsenic levels, tested each of the low-range and high-range tests twice ($N = 32$) with ($n = 16$) and without ($n = 16$) reagent #2.

2.5. Objective 3: efficacy of point-of-use filter in study households (2017–2018)

The Multipure® (Model CB-As-SB, Las Vegas, NV) Drinking Water System is a point-of-use adsorptive media filter tested according to NSF/ANSI Standard 53 for the reduction of arsenic. The filter utilizes block carbon to remove arsenate (As(V)) and has a relatively high flowrate. It was installed in 6 households, a convenience sample, with water $\text{As} > 10 \mu\text{g/L}$ as a pilot study in 2017. The system consists of a small filter faucet connection at the kitchen sink that is separate from the kitchen faucet that is used for washing dishes and other household tasks unrelated to drinking and cooking. The filters were monitored for nine months, with total water usage monitored and arsenic samples taken at installation and 9-month follow-up, to examine efficacy of reducing arsenic in the study setting and to determine filter life in proportion to the amount of water being used.

2.6. Statistical analysis

Objective 1: Using the fishnet tool in ArcGIS 10.5.1 (ESRI, Redlands, CA), a 5-mile by 5-mile grid of polygons was created for the three communities. The spatial join tool was used to connect the location of private wells to polygons in the grid. The averages of arsenic concentrations of wells within the same polygon was calculated for each polygon to anonymize wells. If a private well had been tested multiple times in the years 2014–2018, the highest arsenic concentration was used. For wells with arsenic concentrations below the detectable of $1 \mu\text{g/L}$, $0.5 \mu\text{g/L}$ was used for the average calculation. Polygons were shaded based on the range ($< 5 \mu\text{g/L}$; $5 - < 10 \mu\text{g/L}$; or $10 \mu\text{g/L}$) in which the average fell.

Objective 2: Descriptive statistics were shown for water parameters and stratified by community, and differences in those characteristics in water samples by arsenic concentration (< 10 and $10 \mu\text{g/L}$) using t -tests were compared. To estimate variability in arsenic levels in the subset of drinking water samples collected in 2014 and 2015, we estimated the intraclass correlation coefficient and used a paired t -test to estimate the mean difference between paired samples. We also estimated the Spearman's rank correlation coefficient for descriptive purposes. Statistical analyses were performed using Stata software, version 13.1 (StataCorp, College Station, TX, USA). Sub-objective 1: Determining the accuracy of the rapid arsenic testing kit provides an estimate of its usefulness as a screening tool, the weighted average of a test's sensitivity and specificity, where the sensitivity is weighted by prevalence and specificity is weighted by the complement of prevalence (Alberg et al., 2004). The sensitivity of the rapid test, the true positive, is defined as the proportion of wells with elevated arsenic ($10 \mu\text{g/L}$) that are correctly identified by the rapid test. The specificity of the rapid test, the true negative, is defined as the proportion of wells that do not have elevated arsenic that are correctly identified as negative by the rapid test (Gordis, 2013). Sensitivity and specificity were calculated by comparing rapid test results to ICP-MS, the reference method for arsenic assessment (Flanagan et al., 2015a; George et al., 2012; van Geen et al., 2014a).

Objective 3: Descriptive statistics were used to compare water As levels at baseline and over the follow-up.

3. Results

3.1. Objective 1: water arsenic levels

Water arsenic was $10 \mu\text{g/L}$ in 97 (26.1%) households, between $5 - < 10 \mu\text{g/L}$ in 135 (36.4%) households, and $< 5 \mu\text{g/L}$ in 139 (37.5%) households, based on ICP-MS testing (Table 1, Fig. 2). In Community A ($n = 330$), 26.7% of households had water arsenic $10 \mu\text{g/L}$, with median (interquartile range [range]) arsenic of 6.6 (4.5, 10.7 [< 1.0 , 198.0]) $\mu\text{g/L}$; in Community C ($n = 17$), 47.1% of households had water arsenic $10 \mu\text{g/L}$, with median arsenic 9.7 (3.1, 19.9 [< 1.0 , 30.1]) $\mu\text{g/L}$; and in Community B ($n = 24$), 1 household had water arsenic $10 \mu\text{g/L}$, with median arsenic below the limit of detection (range [< 1.0 , 1.3]) $\mu\text{g/L}$.

3.2. Objective 2: detailed water quality assessment in 29 households

Water arsenic was $10 \mu\text{g/L}$ in 37.9% ($n = 11$) in the 29 households included in the water quality assessment, with a median (inter-quartile range [range]) of 6.3 (< 1.0 , 15.0 [< 1.0 , 49.7]) $\mu\text{g/L}$ arsenic overall (Table 2). All parameters were similar in samples with arsenic above and below $10 \mu\text{g/L}$, except for sulfate which was borderline statistically higher in samples with lower arsenic concentrations (91.2 (lower arsenic) vs. 20.0 (higher arsenic) mg/L , $p = 0.07$). Median silica was highest in Community A (194.4 mg/L), much higher than Community B (7.96 mg/L) (Supplemental Table 1). Community A also had the highest median uranium level (6.4 $\mu\text{g/L}$) (Supplemental Table 1). Uranium was at or above the MCL (30 $\mu\text{g/L}$) in three wells in the 2015 pilot, with two located in Community A. Median uranium level was higher in water samples with arsenic 10 vs. $< 10 \mu\text{g/L}$ (uranium 4.4 vs. 2.9 $\mu\text{g/L}$, $p = 0.89$), but the difference was not statistically significant (Table 2). Nitrate, iron, and cadmium medians were all below the EPA MCL and Secondary Drinking Water Standards. While lead was detectable in all three communities (highest median concentration in Community B (0.6 $\mu\text{g/L}$)), all samples were below the lead action level (EPA Treatment Technique) of 15 $\mu\text{g/L}$. For arsenic speciation, the mean percent of total arsenic that was arsenite (As(III)) was 5.0% in Community A and 38.2% in Community C (results not shown). We found some temporal variability of arsenic in wells ($n = 19$) with ICC (95% confidence interval) of 0.77 (0.58, 0.96) and paired mean difference (95%CI) of 3.50 (0.07, 6.92) $\mu\text{g/L}$ higher arsenic in 2014 vs. 2015 (Fig. 3). The Spearman correlation coefficient between samples collected in 2014 and 2015 was $r = 0.86$ ($p = 0.05$).

In a field setting, the Arsenic Econo-Quick™ low-range test had 25.0% sensitivity and 100.0% specificity when compared to ICP-MS measurements relative to $10 \mu\text{g/L}$, and the high-range test had 100.0% true positive rate and 8.3% true negative rate (Supplemental Table 2 and Supplemental Fig. 1). In laboratory experiments with arsenic-spiked samples overall, the high-range test had moderate to high sensitivity (87.5%) and high specificity (100%); the low-range test had moderate sensitivity (62.5%) and high specificity (100%) (Supplemental Table 3). The sensitivity was higher for both the high-range (100.0%) and low-range test (75.0%) when reagent 2 was not used.

3.3. Objective 3: adsorptive filter results

The Multipure® filter effectively removed arsenic in each household, with arsenic found to be below the LOD ($< 1 \mu\text{g/L}$) in water samples collected from each filter faucet at the installation and the 9-month follow-up timepoint (Fig. 4). In the pilot households, total water usage from the filter over 9 months ranged from 80 to 1233 gallons (median 330). Silica ranged from 21.5 to 32.4 mg/L (median 22.6) in kitchen faucets pre-installation of the filter.

4. Discussion

4.1. Arsenic

Overall, a quarter of private wells tested in tribal communities in North and South Dakota had arsenic $> 10 \mu\text{g/L}$, and therefore are eligible to participate in the SHWS, which will provide arsenic-safe drinking water to affected households. This is much higher than the national average of 7% of domestic well users in the US with arsenic $> 10 \mu\text{g/L}$ (Ayotte et al., 2017). These findings are consistent with historic Indian Health Service data for the area and with urinary arsenic excretion patterns in Strong Heart Study participants (Navas-Acien et al., 2009), demonstrating that tribal communities in North Dakota and South Dakota are disproportionately exposed to arsenic in drinking water, especially in Communities A and C. Community C is undergoing the installation of a new public water system that will provide service to homes on private wells, including many of those tested in this water quality assessment. As such, the SHWS will focus on Community A, where homes often cannot be connected to community water systems, typically due to distance from the tribal water system line, and arsenic-safe drinking water is urgently needed. The Multipure® filter was found to be highly effective at lowering drinking and cooking water arsenic concentrations below $1 \mu\text{g/L}$, even after consistent usage over 9 months. As a result, this point-of-use arsenic removal device will be used to reduce arsenic exposure in the SHWS intervention.

4.2. Water quality parameters

Silica was above the manufacturer's recommended concentration for optimal filter performance ($< 30 \text{ mg/L}$) in 20 households out of 29 in the 2015 intensive water quality pilot and in 1 household out of 6 where the filter was installed (Multipure). High silica concentrations could reduce the amount of time the filter will effectively be able to remove arsenic, consequently increasing the frequency that the filter cartridge will need to be replaced (Moller et al., 2009). Our pilot study showed that the adsorptive media filter proposed for the SHWS effectively removed As for 9 months, indicating that the well with high silica in Community A did not affect the performance of the filter. Sulfate and iron can also potentially interfere with an adsorptive media water filter. However, the levels of sulfate and iron found at study communities were below the problem levels for both parameters (720 mg/L and 0.5 mg/L, respectively) (U.S. E, 2015). The majority of inorganic arsenic in the study communities occurred as As(V), which is considered less toxic than As(III) (Council, 2013). Reverse osmosis was not chosen as a treatment option, as treated water may substantially change the taste of household drinking water since during the treatment process inorganic compounds are removed, and during our formative research, community members expressed concern about certain water filters changing the taste of their drinking water. Another concern about reverse osmosis was the relatively low flow rate.

4.3. Performance of the rapid arsenic test

The Arsenic Econo-Quick™ test performed poorly in our setting and cannot be used as a screening tool at our community sites. It was difficult to meet the required test manufacturer conditions to conduct the test in the field. The manufacturer recommends water samples be warmed to 22–28 °C before testing; air temperature is recommended to be in the same range (Rapid Arsenic Test Kit Instruction Booklet, 2013). At our community site, water samples were not warmed upon collection, and tests were conducted outside in November (mean air temperature 8.3 °C) (NOAA NCDC, 2017). However, a study in Peru with similar cold weather conditions using the same kit found the high-range test to be accurate (George et al., 2014). Another potential source of interference is sulfide. When reagent #2 was removed, our ability to mitigate any interference with sulfide was also removed. Sulfide concentrations at our community site were tested in February 2017 as part of the SHWS filter pilot; all sulfide levels were < 0.050 mg/L, below the 2 mg/L sulfide threshold the manufacturer reports causes interference.

The rapid test needs to be used in a well-ventilated area due to the production of hydrogen and arsine gases, in addition to eye and hand protection (Rapid Arsenic Test Kit Instruction Booklet, 2013). Disposal of waste presented a problem for team members while conducting field work, as both liquid waste and mercuric bromide testing strips needed to be properly handled. This is an additional concern in resource limited settings where waste disposal facilities are not available.

4.4. Arsenic temporality

In the 19 households that had arsenic samples taken in both 2014 and 2015, findings suggest some temporal variability of the arsenic levels in groundwater over our one-year surveillance period, with levels being on average slightly lower in the 2nd period. Other studies have shown that the degree of temporal variability in groundwater arsenic is generally low (Zheng and Ayotte, 2015; Steinmaus et al., 2005). The arsenic concentration variability that we found could have arisen from seasonal effects, or anthropogenic reasons, like aquifer pumping (Ayotte et al., 2015).

4.5. Multipure® filter

The Multipure® filter effectively removed arsenic in each household that participated in the pilot study to below 1 µg/L at the 9-month follow-up, including one household that had elevated silica, and is an adequate water treatment intervention for the SHWS. For private well users, point-of-use filtration devices have been shown to be effective in removing arsenic from drinking water, and major challenges are consistent use of the filter and maintenance over time (Slotnick et al., 2006; George et al., 2006). The SHWS engages community promoters to deliver messages on the importance of using the filter faucet for drinking and cooking, and to explain to households how to change the filter cartridge for their arsenic removal device.

5. Conclusions

This water quality assessment and pilot of a point-of-use arsenic removal device was important for informing the next steps of the Strong Heart Water Study, the first randomized controlled trial of a community based arsenic mitigation intervention in the US. We identified that arsenic above the current arsenic MCL in drinking water was relatively common, especially in communities A and C. We also found elevated silica, which could reduce the efficacy of a point-of-use arsenic adsorptive media filter. The Arsenic Econo-Quick™ test, originally planned for use as a screening tool for recruitment of participants in the intervention, was found to have low performance in rural North Dakota and South Dakota. In our assessment of the effectiveness of the adsorptive media water filtration device for arsenic removal, all devices installed removed arsenic below 1 µg/L at both installation and 9 months post-installation. Through our water quality assessment, we identified a relatively high burden of arsenic as well as an effective water filtration device to reduce arsenic exposure in study communities. The long-term efficacy of a community based arsenic mitigation program, including these arsenic removal devices in reducing arsenic exposure and preventing arsenic related disease, is being tested as part of the ongoing intervention study.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Abbreviations:

AI	American Indian
As	Arsenic
EPA	United States Environmental Protection Agency
GIS	Geographic information systems
ICP-MS	Inductively coupled plasma mass spectrometry
MCL	Maximum contaminant level
SHS	Strong Heart Study

SHWS

Strong Heart Water Study

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Fig. 1.
Visual comparison of test strip from Arsenic Econo-Quick™ test against colorimetric standard to determine water arsenic concentration.

Fig. 2.

Water Arsenic Samples from Household Private Wells in SHWS Tribal Nations from 2014 to 2018. The total number of samples was 330 in Community A, 24 in Community B, and 17 in Community C. The mean water arsenic concentrations were estimated in a 5-mile by 5-mile grid of polygons to anonymize wells. The number of samples in each polygon ranged from 1 to 20 (median: 2).

Fig. 3.
Comparison of 2014 and 2015 ICP-MS arsenic measurements (n = 19) in SHWS study sites.

Fig. 4. Arsenic concentration at baseline and 9-month follow-up with detection limit line at 1.0 $\mu\text{g/L}$ (N = 6 households).

Table 1

Water Arsenic (µg/L) in Domestic Wells in SHWS Tribal Nations in 2014–2018 (N = 371).

	All	Community A	Community B	Community C
N	371	330	24	17
Median (IQR)	6.3 (4.0, 10.5)	6.6 (4.5, 10.7)	< 1.0* (< 1.0*, < 1.0*)	9.7 (3.1, 19.9)
Range	< 1.0*, 198.0	< 1.0*, 198.0	< 1.0*, 1.3	< 1.0*, 30.1
Number of wells:				
< 5 µg/L	139	111 (79.9%)	23 (16.5%)	5 (3.6%)
5- <10 µg/L	135	131 (97.0%)	0	4 (3.0%)
10 µg/L	97	88 (90.7%)	1 (1.0%)	8 (8.2%)

* Limit of detection

Table 2

Median Concentration (IQR) of Water Parameters Tested in 29 Households Overall and by Water Arsenic Levels Above and Below 10 µg/L.

Parameter ^c	N	Limit of detection ^a	Median (IQR)		As <10 µg/L	As 10 µg/L	P value [*]	Analytical method	Analytical location
			Overall	Median					
Arsenic (µg/L)	29	0.1	6.3 (0.3–15.0)					ICP-MS	JHU Trace Metals lab
Cadmium (µg/L)	29	0.0052	0.005 (0.005–0.005)	0.01	0.01	0.41	0.41	ICP-MS	JHU Trace Metals lab
Iron (mg/L)	29	0.2 ^b	0.1 (0.1–0.1)	0.1	0.1	0.71	0.71	Hach	Semi-controlled temp. lab
Lead (µg/L)	29	0.0029	0.49 (0.15–0.42)	0.3	0.1	0.14	0.14	ICP-MS	JHU Trace Metals Lab
Phosphorus (mg/L)	24	0.05 ^b	0.025 (0.025–0.11)	0.03	0.03	0.63	0.63	Hach	Semi-controlled temp. lab
Silica (mg/L)	27	0.01 ^{b**}	107.2 (14.4–162.4)	109.2	107.2	0.69	0.69	Hach	Semi-controlled temp. lab
Uranium (µg/L)	29	0.0015	3.42 (0.32–6.38)	2.9	4.4	0.89	0.89	ICP-MS	JHU Trace Metals lab
Nitrate (mg/L)	29	0.2 ^b	0.33 (0.1–0.79)	0.2	0.5	0.61	0.61	Hach	Semi-controlled temp. lab
Sulfate (mg/L)	29	40 ^b	41.5 (20–143)	91.2	20.0	0.07	0.07	Hach	Semi-controlled temp. lab
Alkalinity (mg/L)	29	25 ^b	214 (153–278)	214.0	230.0	0.68	0.68	Hach	field
pH	29	0 ^b	6.8 (6.5–7.1)	6.7	6.9	0.27	0.27	YSI	field

^aLimit of detection (LOD) = LOD/2 (used in analyses) for undetectable samples.

^bUpper limit: phosphorus = 1.50 mg/L; silica = 1.60 mg/L; nitrate = 30.0 mg/L; sulfate = 150 mg/L; alkalinity = 400 mg/L; iron = 6.0 mg/L; pH = 14 units.

^cUS Environmental Protection Agency's National Primary Drinking Water Regulations, maximum contaminant level (MCL): arsenic = 0.010 mg/L; cadmium = 0.005 mg/L; lead = zero; uranium = zero; nitrate = 10 mg/L.

^{*}P-value comparing difference in parameter between low and high arsenic.

^{**}For samples over the limit of detection, measurement performed by 1:100 dilution with deionized water.