The Effectiveness of Arsenic Remediation from Groundwater in a Private Home
by Elizabeth Pratson, Avner Vengosh, Gary Dwyer, Lincoln Pratson, and Emily Klein

Abstract
Private wells are the source of drinking water for approximately 15% of households in the United States, but these wells are not regulated or monitored by government agencies. The well waters can contain arsenic, a known carcinogen that occurs in groundwater throughout the nation at concentrations that can exceed the Maximum Contaminant Level defined by the U.S. Environmental Protection Agency (10 ppb). In order to reduce arsenic exposure, homeowners can either rely on bottled water for drinking or install in-house water treatment systems for arsenic removal. Here, we document the arsenic levels associated with these options. We examined 24 different major bottled water brands and found that all have arsenic levels <1.5 parts per billion (ppb), and more than half have levels below our measurement detection limit of 0.005 ppb. For in-house treatment systems, we examined the performance of arsenic removal by point-of-use reverse osmosis filtration, and by whole-house and point-of-use filters containing granulated ferric oxide. Our results show that long-term (2 years) filtration with granulated ferric oxide reduced arsenic in well water from an initial concentration of 4 to 9 ppb down to <0.005 ppb, validating this technology as an effective form of arsenic remediation for private homes.

Introduction
Arsenic (As) toxicity is a global health problem. In many areas of the world, particularly Bangladesh and West Bengal, millions of people are exposed to high levels of As in local groundwater (Ratnaike 2003). Chronic and long-term exposure to As leads to problems such as malignancy, skin keratosis, vascular diseases, and neurological and blood effects. In addition, there is some evidence linking As exposure through drinking water to diabetes, reproductive disorders, and neurological disorders (National Research Council [NRC] 2001). As has also been linked to cancer of the bladder, lungs, skin, kidney, nasal passages, liver, and prostate (NRC 1999). The risk of contracting cancer from drinking water with an As concentration of 50 ppb is 1:100, more than a hundred times greater than any other drinking water contaminant (Smith et al. 2002).

In the United States, the Environmental Protection Agency (EPA) is responsible for regulating the concentration of As in public water supplies. Originally, this standard was set at maximum contaminant level (MCL) for As in drinking water at 50 parts per billion (ppb). In January 2001, a new standard of 10 ppb was adopted. Lower levels were considered (down to 3 ppb; NRC 1999), however, due to As’s widespread occurrence in water resources and the potentially high costs to water companies of reducing As to such low levels, this 10 ppb was adopted and became enforceable in 2006. The maximum contaminant level goal (MCLG) for As, or the level at which there are no known health effects, is 0 ppb (NRC 2001), indicating that any exposure has some, although potentially small impact on human health. A survey of 30,000 groundwater samples in the United States has shown that concentrations of naturally occurring As in groundwater vary considerably, with approximately 10% exceeding the 10 ppb MCL threshold (Welch and Stollenwerk 2003). Because private wells are exempt from the Safe Drinking Water Act (SDWA), more than 40 million people in the United States consume water through unregulated, unmonitored private wells (EPA 2004). In the southeastern United States, As levels in well water are elevated in some geological terrains. One is the Carolina terrain, also known as the Carolina slate belt, which extends from Virginia to Georgia (Pippin et al. 2005). In North Carolina, samples from hundreds of private wells in the Carolina slate belt have As levels above the MCL threshold of 10 ppb (Figure 1). Therefore, a large (but unknown) fraction of the NC population in this region is vulnerable to As exposure from drinking water.

Because the quality of private wells is not protected by SDWA, private homeowners, who may not be fully aware of the risks associated with As or the federal safety standard,
Figure 1. Map of arsenic concentrations in wells in North Carolina; arsenic concentration ≥1 ppb are marked by red circles and <1 ppb with black circles. Note that groundwater with As ≥1 ppb levels occurs in the Piedmont, which is shaded in grey. Case study home located in yellow box. Map compiled and prepared by J.L. Tootoo, Duke University.

must monitor their own wells. Testing the As level in private well is typically conducted at a laboratory administered by a state agency or a private company. Most of these laboratories report As concentrations with a detection limit of 2 to 5 ppb, so the water quality can only be established with respect to the MCL but not to the MCLG (i.e., zero As concentration). Concerned homeowners can switch to drinking bottled water and/or install one or more water filtration systems for As remediation. The level to which these two options decrease As intake down to the MGLG, however, has not been systematically analyzed.

In this paper, we evaluate the As exposure from consumer brand bottled waters and from water treatment systems in a private home. We present continuous measurements of As concentrations in a private well and treated drinking water from several in-home water filtration setups that include a whole-house or point-of-entry (POE) sediment trap, a single faucet or point-of-use (POU) reverse osmosis (RO) system, and both a POE and POU granulated ferric oxide (GFO) filters. The As detection limit reported in this study is 0.005 ppb; 3 orders of magnitude lower than generally reported by commercial and state water-testing laboratories. This paper aims to evaluate (1) the extent of As reduction upon using bottled water or installing in-house remediation and (2) how closely these options are related to the EPA MCLG level.

Study Site and Methods

This study focuses on well water from the Piedmont region of North Carolina. Figure 1 shows detectable (≥1 ppb) vs. non-detectable (≥1 ppb) levels of arsenic in groundwater from wells throughout the state. High arsenic levels are detected along the Carolina slate belt where As content in well water can reach up to 100 ppb. The Carolina slate belt is characterized by volcaniclastic rocks that have undergone low-grade metamorphism (Pippin et al. 2005).

The household whose well water was analyzed in this study is also located in the Piedmont physiographic belt containing As bearing rocks (Figure 1). Repeat measurements of the well water started in March 2003 and continued through December 2007. The only water treatment system in the house prior to sampling was a standard salt-loaded water softener installed to lower iron and manganese concentrations.

Beginning in July 2003, a sequence of additional off-the-shelf water filtration systems were installed in the house (Figure 2). This sequence began with a standard sediment filter placed inline before the water softener in order to reduce particulate matter in the groundwater (Figure 2). The filter consists of a 24 cm × 6 cm cylinder of synthetic material designed to trap particles down to 5 µm in size.

A POU-RO system was installed underneath the kitchen sink in the house in June 2005 to further cut down on As in water used for drinking and cooking (Figure 2). RO systems force water through a semipermeable membrane that filters out contaminants down to the ion level using an electric charge differential. The stronger the charge of the contaminant ions, the more they are rejected from passing through the RO membrane. This has particular relevance for filtering out As, which occurs in water as two ionic forms, As³⁺ and As⁵⁺. Because of the latter’s greater size and charge, RO systems are more effective at filtering out As³⁺. The more toxic ion, however, is As⁵⁺, which is more difficult to remove by RO filtration.

The RO system was enhanced in May 2006 by replacing one of its 6 cm × 24 cm carbon prefilter cartridges with a same-size cartridge of GFO (Figure 2). A RO system generally comes with two fine sediment filters and one carbon-based organic filter through which the water is passed before being forced through a membrane.

A POE-GFO system was then installed in July 2006 (Figure 2). The filter was set up according to guidelines proposed by New Jersey State Geologic Survey (Spayd 2005). These guidelines recommend two side-by-side 25 cm × 102 cm (or 23 cm × 259 cm) tanks, each containing at least 0.03 m³ (1 ft³) of granular ferric adsorption media if As concentrations for the well water are < 50 ppb. The filter is installed inline after the sediment filter and the water softener, and this treated water goes to all faucets (Figure 2). The lead tank is the primary working tank, and removes the majority of As. The second tank is a backup and removes any remaining As. A sampling tap between the tanks allows the homeowner to periodically test water quality and thus monitor the performance of the first tank (Figure 2). When As levels become too high, the tanks are to be switched; the
backup becomes the primary working tank and the lead tank the backup after its granular iron media has been emptied and replaced (Spayd 2005).

Throughout the sampling program, water was tested at the wellhead and kitchen faucet (Figure 2). Additional testing was done down flow of the whole-house filter, and up flow of both the POU-RO and GFO filters following their installation.

Finally, the RO system was replaced with POU-GFO filters on August 2006 (Figure 2), where the As removal applies for only the kitchen faucet.

Water samples, including those from various brands of bottled water, were analyzed using an inductively coupled plasma mass-spectrometer (ICP-MS) at Duke University. The published detection limit for this instrument, that is, the smallest concentration of a given element for which a reliable measurement can be made, is 0.002 ppb (Taylor 2001).

A test was carried out to ensure that this instrument was indeed capable of measuring As to such low concentrations (Figure 3). A series of dilutions of National Institute of Standards (NIST) water-sample standard 1643e were prepared. The As levels in these test samples ranged from the undiluted level of 60.45 ppb in the NIST standard down to 0 ppb. The results are shown in Figure 3, and suggest that under typical operating conditions the PQ3 ICP-MS detection limit for As is approximately 0.005 ppb. The detection limit was calculated using the following formula from Taylor (2001):

\[
C_{DL} = \frac{(2.262 \times 15 \text{ cps})}{7013.4 \text{ cps}} = 0.0048 \text{ ppb}
\]

(95% confidence)
where \(C_{DL}\) is the detection limit, expressed in ppb.

Figure 2. A schematic illustration of the water treatment systems in the investigated home and a time sequence of installation and monitoring. (A) Basic layout of treatment systems within the house as well as the relative location of well, pressure tank, and water softener. (B) Timeline indicating month/year when each new system was added to plumbing. Note that timeline is not to scale.

Figure 3. Resolving power of the Duke ICP-MS for measuring arsenic concentrations in water samples. Progressive dilutions of NIST water-sample standard 1643e show that the instrument can accurately measure arsenic down to levels of 0.005 ppb.
Results

Bottled Waters

We tested 24 brands of bottled water sold locally in Durham, North Carolina. These ranged from national and international brands of spring and mineral water to regional filtered waters bottled in the southeast United States. The results are shown in Figure 4A. All 24 brands are found to have As concentrations ≤1.5 ppb, with more than half of these (13) falling below the 0.005 ppb detection limit of the ICP-MS.

Figure 4B compares the As concentrations in the bottled waters to the combined concentrations of Ca, Fe, Mg, and Mn measured by the ICP-MS. The latter are some of the major rock-forming elements and occur in higher concentrations in “mineral” or “natural spring” water sources. Our data show that these mineral waters also tend to have relatively higher As concentrations, indicating that waters with higher mineral content are more likely to have higher As content.

In-Home Water Treatment Systems

Throughout the 4 years of monitoring water quality at the residence, As concentration at the wellhead fluctuated between 4 and 9 ppb (Figure 5, note the logarithmic scale along the vertical axis in order to enhance the lower values). Removal of tap water As by the water softener without the GFO addition (before July 2006) was not significant (Figure 5). In contrast, our data show that the whole-house sediment filter (prior to the softener) significantly reduced As. Its installation reduced As levels by about 2 ppb (Figure 5).
The impact of the whole-house GFO system was even greater. After it was installed in July 2006, As levels at the kitchen faucet decreased from 4.23 to 1.22 ppb (Figure 5). In fact, water sampled between the two tanks tested at As levels below the 0.005 ppb detection limit of the ICP-MS. The higher As concentration measured at the kitchen faucet appears to be due to the filtered waters reacquiring As from deposits built up in the intervening pipes. Support for this comes from repeated testing, which showed the As concentrations at the sink continued to decline over time.

Results for the RO system showed temporary initial success when installed in 2005. Following its installation, As levels at the kitchen faucet fell to below the ICP-MS detection limit. However, these levels then began to increase with time. The As level rose to 0.25 ppb after 2 weeks, to 0.83 ppb by 10 months, and over 1 ppb by 1 year (Figure 5). The pre-filters in the RO were replaced with the manufacturer’s recommended replacement filters at this point. This dropped As levels back to 0.22 ppb, but these levels then rose again even more rapidly (up to 0.84 ppb in 2 months) (Figure 5). It seems that the RO membrane was becoming clogged, diminishing its effectiveness for As rejection. Our data suggest that a sink-RO system in a private home can be an effective remediation for removing As but its performance degrades with time unless the membrane and filters are changed regularly.

The removal of As by the RO system became more effective when one of its sediment prefilters was replaced with the GFO-POU cartridge. As levels in the RO-treated water were reduced from 1 down to 0.24 ppb with the addition of this single GFO-POU cartridge (Figure 5; 6/2006).

Based on the effectiveness of the one GFO cartridge, the two remaining prefilters in the RO system were replaced with GFO cartridges and the RO membrane system was disconnected. Thus, the new kitchen POU system consisted of just three GFO cartridges, with the additional cartridges increasing the contact time between the water and GFO media. The effectiveness of this simple system was impressive. It reduced As concentrations to below detection limits.

Figure 5. Arsenic concentrations (logarithmic scale, vertical axis) over time (horizontal axis) in well water, tap water treated by POE systems, and water treated by POU systems. Dates along horizontal axis correspond to when water samples were taken and are not at regular intervals. Well water exhibits relative stability of As concentration, whereas selected treatment systems show differential remediation, with the POU-GFO filter being the most effective in removing As from well water.

Figure 6. Performance of the POU-GFO systems over a period of 15 months in several neighboring homes with similar As concentrations in their well waters. Data show that these systems are capable of reducing As level in treated water below the detection limit for more than 1 year.
(0.005 ppb) for more than 1 year, outperforming the RO system (Figure 5; 12/2007).

A simple two cartridge GFO-POU system was also tested in five neighboring homes that use private wells for drinking water with groundwater having As concentrations ranging from 2 to 8 ppb (Figure 6). Three of these homes had no whole-house water treatment system, whereas two had only a water softener. Our results show a significant As reduction; Installation of the GFO-POU system at Home D (8 ppb) As concentrations dropped in the treated water to below the ICP-MS detection limit for 10 months (Figure 6). For the home with As levels of 2.5 ppb at the wellhead, the GFO-POU system treatment kept concentrations below the detection limit for >15 months (Figure 6). This indicates that the POU-GFO works very well, even without the aid of any additional treatment systems such as a POE sediment trap and/or a POE-GFO filter.

Discussion

Homeowners with private wells have several effective options for reducing As concentrations in their drinking water by an order of magnitude or more below the EPA MCL of 10 ppb. In fact, some of these options approach attainment of the EPA MCLG of 0 ppb, yielding As concentrations that cannot be detected at even the highest measurement resolutions.

Our data show that water softeners offer little if any protection against As. They are, however, successful in filtering out other potentially unwanted elements. For example, the water softener in the residence analyzed in this study reduced both iron and manganese concentrations from 135 and 85 ppb, respectively, to below the ICP-MS detection limit. Because the GFO system requires reduced levels of Fe, Mn, and Si, a water softener may be a necessary component for home water treatment systems together with the GFO system.

Our results also show that POE sediment traps, which are relatively simple and inexpensive to install, can be effective As filters. We show that when one of these filters was placed in front of the water softener, As levels were reduced by about 2 ppb. A limitation of these filters, however, is that they eventually get clogged with sediment and have to be replaced frequently. The time between replacements depends on the turbidity of the incoming well water with the duration being longer for clearer water. A replacement frequency of 3-6 months is typical in the Piedmont area of North Carolina.

RO systems are very effective POU systems for As removal, but only for a limited period of time. Manufacturers of POU-RO systems generally recommend yearly replacement of membranes and that the prefilter should be replaced every 6 months, depending on the quality of the incoming water. We found this frequency insufficient for keeping the As concentrations ≤1 ppb. Our results suggest that with in months after installation or replacement, As concentration in POU-RO-treated water can rise above this 1 ppb concentration for incoming well waters containing ≤4 ppb of As. At higher As concentrations, the effectiveness of RO systems may decline even faster.

An additional drawback to RO filtration is that it can lower the water pH. Using a pH meter in the residence analyzed, we found that the RO unit reduced the pH of the treated water from 7.0 to 6.0. The more acidic water can enhance leaching of metals in the pipe system. Most RO units are therefore constructed of glass or plastic. The RO faucet, however, is made of metal. In order to check this effect, we found that the RO water had a higher lead content (0.8 ppb) than that from the adjacent kitchen sink faucet (0.1 ppb). This excess lead likely came from the metal RO faucet. So even when an RO system is successfully reducing As, it may be raising the concentration of other unwanted metals in the system due to the lower pH of the RO-treated water.

An alternative option for homeowners is the use of bottled waters. Our data show that brands common in North Carolina have As levels far below the EPA MCL value. Mineral water with higher dissolved minerals tends to have higher As concentrations, but only one of the brands analyzed exceeded 1 ppb. The lowest As was found in bottled waters that are distilled water, which have As levels below the detection limit.

Overall our data indicate that for in-home water treatment systems, the POE- and POU-GFO systems are the most effective for As remediation. In contrast to the POU-RO system that selectively removes charged ions (removing As\(^{3+}\) effectively, but not as effective with the more toxic As\(^{5+}\)), the POU-GFO medias high surface area of the iron oxide causes adsorption of both As\(^{3+}\) and As\(^{5+}\).

The primary advantage of the POE-GFO filter is that it treats water at the POE to the house, so As concentrations are low regardless of what faucet the water is drawn from. The advantages of the GFO-POU cartridges are that they are effective, relatively long-lived, inexpensive, and easy to install. The cartridges treat only the water coming out the POU system, but this may be sufficient if this is the only water used for drinking and cooking. The threefold POU-GFO system developed in this study works as well as the RO POU and lasts longer.

Conclusions

In combination with a whole-house sediment trap, a POU-GFO system may be the most cost-effective in-house treatment system for removing As from well water. We estimate that at 2009 costs, such a system can be commercially installed for $300 to $400, and would require an additional $80 per year for replacement of one of the filters. This is in contrast to the whole-house GFO filter, which costs $1800 to $2600 for commercial installation, and requires an additional $800 every 2 to 3 years for media replacement in one tank. POU-RO systems do not remove As as consistently as POU-GFO and require more frequent maintenance at greater cost. Commercial installation of an RO system costs between $600 and $900, with additional $100 for periodic replacement of the system’s filters and membrane.

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References


Biographical Sketches

Elizabeth Pratson, corresponding author, was a consultant and research scientist at Duke University at the time of this study. She can be reached at epratson@elon.edu.
Avner Vengosh is an associate professor at Earth and Ocean Sciences, Duke University. His expertise lies in geochemistry and water quality.
Gary Dwyer is a research scientist in Earth and Ocean Sciences at Duke University and holds a PhD from Duke University. His expertise lies in paleoclimatology and marine science.
Lincoln Pratson is a professor of energy and environment at Duke University.
Emily Klein is a professor of geology and senior associate dean and director of undergraduate studies at Duke University. Her experience lies in marine geochemistry and geology.