

Recommended Sampling Intervals for Arsenic in Private Wells

by Brian J. Mailloux¹, Nicholas A. Procopio³, Mark Bakker⁴, Therese Chen^{2,5},
Kazi Matin Ahmed⁵, M. Rajib H. Mozumder⁶, Tyler Ellis⁶, Steve Chillrud⁶, and Alexander van Geen⁶

Abstract

Geogenic arsenic in drinking water is a worldwide problem. For private well owners, testing (e.g., private or government laboratory) is the main method to determine arsenic concentration. However, the temporal variability of arsenic concentrations is not well characterized and it is not clear how often private wells should be tested. To answer this question, three datasets, two new and one publicly available, with temporal arsenic data were utilized: 6370 private wells from New Jersey tested at least twice since 2002, 2174 wells from the USGS NAWQA database, and 391 private wells sampled 14 years apart from Bangladesh. Two arsenic drinking water standards are used for the analysis: 10 µg/L, the WHO guideline and EPA standard or maximum contaminant level (MCL) and 5 µg/L, the New Jersey MCL. A rate of change was determined for each well and these rates were used to predict the temporal change in arsenic for a range of initial arsenic concentrations below an MCL. For each MCL and initial concentration, the probability of exceeding an MCL over time was predicted. Results show that to limit a person to below a 5% chance of drinking water above an MCL, wells that are $\frac{1}{2}$ an MCL and above should be tested every year and wells below $\frac{1}{2}$ an MCL should be tested every 5 years. These results indicate that one test result below an MCL is inadequate to ensure long-term compliance. Future recommendations should account for temporal variability when creating drinking water standards and guidance for private well owners.

Introduction

Consumption of groundwater contaminated with trace levels of geogenic arsenic is a worldwide health problem (Smith et al. 2000). This problem is hardest to assess in areas where privately owned wells are used for drinking water and the prevalence of testing can vary along with outreach campaigns and laws (Zheng and Flanagan 2017). Testing programs are implemented by both governmental and non-governmental agencies worldwide. However, even with the large number of spatially based sampling

programs that exist, little data are available to elucidate the temporal variability of arsenic in wells over time and to recommend a protective sampling interval.

Spatial surveys have been carried out at the national (Dhar et al. 1997; Berg et al. 2001; BGS and DPHE 2001; Focazio et al. 2006; Ayotte et al. 2017), regional (Berg et al. 2001; Erickson and Barnes 2005; Ayotte et al. 2006), and local (van Geen et al. 2003; Yang et al. 2012) scale to determine the extent of the arsenic problem. Building on the spatial surveys, statistical modeling has been used to better understand the controls on arsenic contamination and to predict other potential hotspots (e.g., Amini et al. 2008; Winkel et al. 2008; Rodríguez-Lado et al. 2013; Ayotte et al. 2017). However, fewer studies examine temporal variability (e.g., Cheng et al. 2005; Erickson and Barnes 2006; Savarimuthu et al. 2006; McArthur et al. 2010; Ayotte et al. 2015; Levitt et al. 2019).

The controls and the amount of temporal variability of arsenic in groundwater are poorly constrained. Hourly (Erickson and Barnes 2006), seasonal (Schaefer et al. 2016; Levitt et al. 2019; Degnan et al. 2020), and longer (McArthur et al. 2010) trends have been observed but many wells are also fairly stable (Ayotte et al. 2003; Cheng et al. 2005; Thundiyil et al. 2007; van Geen et al. 2007; Dhar et al. 2008). The trends are usually thought to be caused by changes in redox

¹Corresponding author: Environmental Science Department, Barnard College, NY, New York 10027; bmaillou@barnard.edu

²Environmental Science Department, Barnard College, NY, New York, 10027

³New Jersey Department of Environmental Protection, Division of Science and Research, Trenton, NJ, 08064

⁴Water Management Department, Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft, Netherlands

⁵Department of Geology, University of Dhaka, Dhaka, Bangladesh

⁶Lamont-Doherty Earth Observatory, Columbia University, Palisades, NY, 10964

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status or change of groundwater flow patterns (Schreiber et al. 2000; Gotkowitz et al. 2004). In a study of 1245 public and private wells sampled twice, 87% of samples were within 4 $\mu\text{g/L}$; the remaining 13% showed larger variability, with the variability potentially related to seasonal variations in water levels or changes in the redox status of the groundwater (Ayotte et al. 2015). During well installation, more oxidized water can enter an aquifer, which can become more reducing over time and arsenic levels can increase (Erickson et al. 2019). In production wells, when the pumps are off, Fe oxides form that can sorb arsenic and lower concentrations which rebound during pumping (Erickson and Barnes 2006). Changing flow paths associated with groundwater pumping and urbanization can also impact arsenic concentrations (McArthur et al. 2010; van Geen et al. 2013). In summary, knowledge about the physical and chemical processes that cause temporal variation of arsenic is still limited.

Within public utilities, regular monitoring is required; in the United States, this is under the federal Safe Drinking Water Act (U.S. Environmental Protection Agency [U.S. EPA] 1974). In the United States, however, there are approximately 44 million people drinking water from privately owned wells (Johnson and Belitz 2017; Dieter et al. 2018). These private wells are not covered by this federal statute and face very little regulation by the states. Only five states require testing for arsenic, and only at specific occasions like new well construction or during real estate transactions (Zheng and Flanagan 2017). In New Jersey, a state with one of the most stringent laws, testing has been mandated during all residential housing transactions and rental leases since 2002 (State of New Jersey N.J.S.A. 58:12A-26 et seq 2002); this required testing has only reached about 25% of estimated private wells. In an arsenic impacted region of Maine, 78% of survey respondents had completed any test of their well water, but only 59% believed that it included arsenic as a parameter and only about 26% could recall the arsenic result (Flanagan et al. 2015a, 2015b). Similar findings were found in New Jersey (Flanagan et al. 2016). Outside of the United States, in one region of Bangladesh where multiple blanket surveys have occurred, only 48% of respondents knew the status of arsenic in their well in relation to the country's drinking water standard (van Geen et al. 2014). Considering the lack of knowledge about arsenic concentrations in private wells, a clear data-driven message about how often to sample a private well is needed.

Current arsenic standards or maximum contaminant levels (MCLs) around the world vary: 50 $\mu\text{g/L}$ is the MCL in Bangladesh and in India for private groundwater in absence of another drinking water source (Bureau of Indian Standards 2012), 10 $\mu\text{g/L}$ is the EPA MCL (U.S. EPA 2001) and WHO guideline (World Health Organization 2017) and is used in most of the world, whereas 5 $\mu\text{g/L}$ is the New Jersey MCL (New Jersey Department of Environmental Protection 2004) and is the new MCL for New Hampshire that will take effect by 2021 (New Hampshire Department of Environmental Services 2018) and may be a future target for other jurisdictions. In this

paper we are using the term MCL generically to indicate a standard or cutoff in arsenic concentrations that should be met by a homeowner with a private well. When these MCLs were formulated it was based on multiple criteria: health outcomes, the ability to measure arsenic and treat contaminated water as well as cost concerns (Schmidt 2014). Water below an MCL still poses a health risk (e.g., Moon et al. 2013). A small change in concentration when crossing an MCL may not drastically change health risks even if it has important public policy implications. The 50 $\mu\text{g/L}$ MCL will not be considered in this study as it poses a larger health risk and is not recommended by the WHO. This study considers the 10 and 5 $\mu\text{g/L}$ MCLs which are used for guidance for private well owners by the state agencies in the United States. Other laws govern the testing of public supply wells.

This work uses three large datasets where arsenic was tested multiple times at the same well in order to determine changes in groundwater composition and provide a recommended sampling frequency to homeowners which includes a level of confidence that the concentration will remain below a recommended level or MCL. Datasets are from New Jersey, across the United States from the U.S. Geological Survey (USGS), and Bangladesh. The rates of arsenic change from each dataset are used to independently predict the change in arsenic in well water over time. The calculated rates of change represent both measurement error and actual changes in arsenic concentrations over time. However, for this analysis, separating measurement error from trends is not required as both factors contribute to the calculated rates. The goal of this paper is to provide recommended protective resampling intervals for private well owners based on the probability of changing arsenic levels, which is an approach that could be used for generating recommended sampling frequency guidelines for other contaminants, especially ones with geogenic sources.

Methods

Arsenic Data

Datasets from New Jersey, the USGS, and Bangladesh were used to assess the rates of change of arsenic. The New Jersey and Bangladesh datasets are previously unpublished whereas the USGS data were acquired via open access.

Beginning in 2002, as part of the New Jersey PWTA, homes in the northern region of New Jersey with a private well are required to have their raw well water tested before a real-estate transaction or every 5 years for rental properties (State of New Jersey N.J.S.A. 58:12A-26 et seq 2002). From September 2002 through March 2014, 42,994 arsenic tests were conducted. During this time, 6273 unique wells were tested multiple times with the tests at least 7 days apart (Table 1). State-certified commercial laboratories were used to test for arsenic. The detection limits for arsenic varied with time and lab. The detection limits have ranged from 0.01 to 15 $\mu\text{g/L}$

and the mean annual detection limit has decreased from 4.4 $\mu\text{g/L}$ in 2002 to 2.7 $\mu\text{g/L}$ in 2014. These data were evaluated using a unique well identifier with no connection to the specific location to ensure confidentiality of the data and the well owner throughout the analysis.

Arsenic data were downloaded from the USGS National Water-Quality Assessment Project (NAWQA) via the Water Quality Portal (<https://www.waterqualitydata.us/portal/>). When downloading the data, the Project ID was NAWQA NWIS and the characteristic was arsenic with sample results chosen as the download option. This resulted in 2174 wells with multiple data points. Wells that only consisted of field duplicates were removed from the dataset (two samples from 1 day). Sampling years spanned from 1970 to 2018. The detection limits ranged from 0.022 to 13.5 $\mu\text{g/L}$. The mean annual detection limit varied from 1.0 to 1.26 $\mu\text{g/L}$ from 1970 to 2000 and decreased to 0.03 to 0.45 $\mu\text{g/L}$ after 2000.

In Bangladesh, 6000 wells in a 25 km² area were tested in 2001 (van Geen et al. 2003). The area was revisited in 2014 and 391 of the original wells were plausibly reidentified and resampled (Mozumder 2019). The wells were verified by looking for well tags along with comparing the latitude, longitude, depth, and year of installation between the two datasets. The original archived samples were reanalyzed by ICP-MS along with the newly collected samples using published methods (Cheng et al. 2004). All samples were above the detection limit of 0.1 $\mu\text{g/L}$.

Statistical Analysis

The measured arsenic concentrations were used to estimate the probability that the arsenic concentration will exceed a MCL in X years if the current value is Y $\mu\text{g/L}$. For example, what is the probability that an MCL of 5 $\mu\text{g/L}$ is exceeded in 3 years ($X = 3$) if the current concentration is 2 $\mu\text{g/L}$ ($Y = 2$). Probabilities were estimated for up to 10 years ($X = 1, 2, \dots, 10$). Probabilities were estimated for nine initial concentrations for each MCL, $Y = 0.5, 1, \dots, 4.5$ $\mu\text{g/L}$ for the 5 $\mu\text{g/L}$ MCL, and $Y = 1, 2, \dots, 9$ $\mu\text{g/L}$ for the 10 $\mu\text{g/L}$ MCL. For each initial concentration Y , the critical rate was computed for exceeding an MCL in X years. The critical rate is the rate necessary to reach an MCL over a given time period. For example, for an initial concentration of $Y = 2$ $\mu\text{g/L}$, the critical rate to reach an MCL of 5 $\mu\text{g/L}$ in $X = 3$ years is 1 $\mu\text{g/L/yr}$. The following procedure was followed for each dataset to estimate the probabilities.

Step 1: The data were organized by well and time. Only wells with two or more samples were utilized and all others were discarded, as no rate of change can be estimated.

Step 2: The rate of change in arsenic concentration was computed for each well. For a well with only two measurements, the rate of change was the difference between the measured arsenic concentrations divided by the time interval between the measurements. For

a well with more than two measurements, the rate of change was obtained from simple linear regression using Pearson's correlation. For wells where a sample was below the detection limit, the value was set to $1/2$ the detection limit. For a well with all samples below the detection limit the rate of change was set to zero. The method for utilizing nondetect data can have large implications for the results, especially for the New Jersey data which has a large number of non-detects. The implications of setting the values to $1/2$ the detection limit and the rates of change to zero are further elaborated in the discussion section.

Step 3: For each measured concentration Y of interest, the data were subsampled to only use rates from wells with a mean arsenic concentration near the concentration of interest. This was accomplished by filtering the observed rate data using the mean arsenic concentration for each well and a window of plus or minus 2.5 $\mu\text{g/L}$ around the mean measured arsenic concentration. If the window intersected zero, it was not adjusted. For example, if the concentration of interest was $Y = 9$ $\mu\text{g/L}$ then rates from wells with a mean arsenic concentration from 6.5 to 11.5 $\mu\text{g/L}$ were utilized. If the concentration of interest was $Y = 2$ $\mu\text{g/L}$ then rates from wells with a mean arsenic concentration from 0 to 4.5 $\mu\text{g/L}$ were utilized. This window method was utilized because the rates of change increase with arsenic concentration and thus using all rates would overestimate change at lower concentrations.

Step 4: The probability of a well with concentration Y $\mu\text{g/L}$ exceeding an MCL in X years was estimated from the cumulative distribution of the rate of change of the wells selected in step 3. The probability that the critical rate is exceeded is estimated as the fraction of the selected wells with a rate larger than the critical rate for concentration Y and time X . For example, consider again the concentration of $Y = 2$ $\mu\text{g/L}$ with a critical rate of 1 $\mu\text{g/L/yr}$ to exceed the 5 $\mu\text{g/L}$ MCL in $X = 3$ years. If 10% of the measured rates of change of all wells with a median concentration between 0 and 4.5 $\mu\text{g/L}$ are above the critical rate of 1 $\mu\text{g/L/yr}$, then the probability that a well with a measured concentration of 2 $\mu\text{g/L}$ exceeds an MCL within 3 years is estimated as 10%.

Results

Arsenic Data Summary

In New Jersey 6273 unique wells where multiple samples were collected were evaluated with 5343 having two samples from the same well, 803 having three samples, 116 having four samples, 10 having five samples, and 1 having six samples (Table 1). The elapsed time (time between first and last sample) was calculated for each well and was then averaged for wells with the same number of samples and ranged from 3.8 years

Table 1
Summary of Wells from the Three Datasets

Number of Times Sampled	New Jersey		NAWQA		Bangladesh	
	Number of Wells Sampled	Average Elapsed Time (Years)	Number of Wells Sampled	Average Elapsed Time (Years)	Number of Wells Sampled	Average Elapsed Time (Years)
2	5343	3.8	1270	9.9	391	14
3	803	5.8	361	17.1		
4	116	6.8	99	11.4		
5	10	5.7	92	9.6		
>5	1	8.0	352	13.4		
Total	6273	4.1	2174	11.7	391	14

The number of times each individual well was sampled and the average elapsed time for wells that were sampled multiple times. The elapsed time is defined as the length of time between earliest and latest sample for each well. For the New Jersey dataset only wells with an elapsed time over 7 days were utilized.

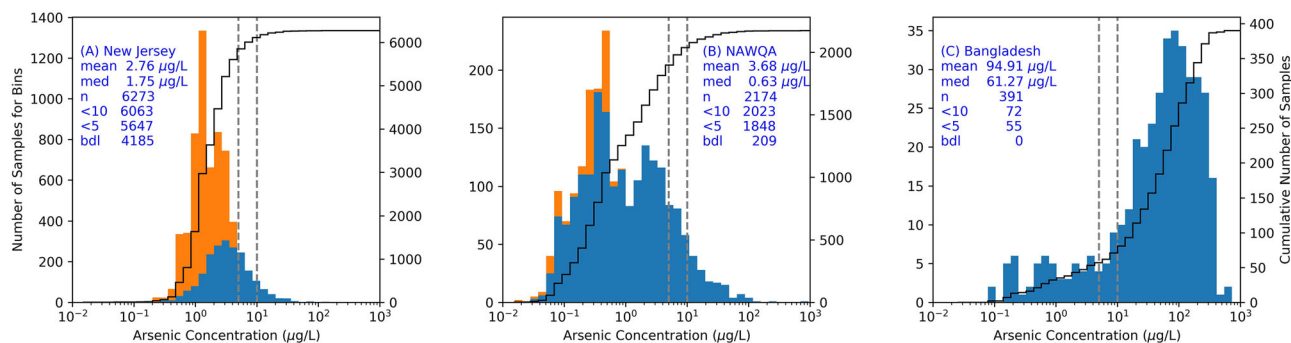


Figure 1. The distribution of mean arsenic concentrations for each well (e.g., if a well has two samples we used the two values to calculate the mean) and cumulative number of samples from three datasets; (A) New Jersey, (B) USGS NAWQA, and (C) Bangladesh. The orange bars represent the mean values when all data are below the detection limit is included. The blue bars are means of arsenic concentrations when at least one sample was above the detection limit. The solid line is the cumulative number of samples. All nondetect samples were taken as $1/2$ the detection limit. The vertical dashed lines are drawn at 10 and 5 $\mu\text{g/L}$, the WHO/EPA standard and the New Jersey standard respectively. Note that the number of samples are different between datasets and are shown on each graph along with the number of samples below each standard and the number of samples below the detection limit (bdl).

for wells sampled twice to 8.0 years for wells sampled more than five times (Table 1). Any well with an elapsed time of less than 7 days was removed. The majority ($n = 6063$, 96.7%) of wells had a mean arsenic concentration below 10 $\mu\text{g/L}$ and 5647 (90.0%) were below 5 $\mu\text{g/L}$ with 4185 (66.7%) below the variable detection limits of each lab (blue in Figure 1 are samples above the detection limit and orange below). The overall mean arsenic concentration (including non-detects at $1/2$ the detection limit) was 2.76 $\mu\text{g/L}$ and the median was 1.75 $\mu\text{g/L}$ (Figure 1). The one-half detection limits were clustered between 1 and 5 $\mu\text{g/L}$ consistent with levels from commercial labs (Figure 1a, orange histogram). The one-half detection limits and not a smoothed interpretation (e.g., maximum likelihood estimation) (Helsel 2005) are shown in order to represent what is reported to private well owners (Figure 1). For each well the number of samples (both detect and non-detect) were determined and then summed and grouped by the number of times a well was sampled (Table 2). Except for the one well sampled six times, the highest percentage of wells had all samples below the detection limit regardless of the

number of times sampled (zero results above detection) (Table 2). For wells with both detectable and non-detectable samples the distribution was relatively uniform (Table 2). Samples in the dataset that were collected less than 7 days apart were discarded, as they likely represent retesting after, for example, sample collection error, resulting in unrealistically high rates of change. Even still, the rates of change vary from -1294 to $383 \mu\text{g/L/yr}$. The few remaining higher magnitude rates are geochemically unrealistic and likely represent outliers, but they are part of the dataset and are representative of future misclassifications and thus were kept for the analyses. The rates of change of 2 standard deviations around the median ranged from -1.67 to $1.95 \mu\text{g/L/yr}$ (dotted blue vertical line in Figure 2).

The NAWQA dataset includes 2174 sampled wells with 917 having two samples, 240 having three samples, 72 having four samples, 90 having five samples, and 233 having more than five samples (Table 1). The mean elapsed time ranged from 9.6 to 17.1 years (Table 1). The mean arsenic concentration was 3.68 $\mu\text{g/L}$ and the median was 0.63 $\mu\text{g/L}$. The majority of the wells, 2023 (93.1%),

Table 2
Distribution of Samples in Regard to the Detection Limit from the New Jersey Dataset

Number of Times Sampled	Number of Wells Sampled	Number of Samples Above the Detection Limit						
		0	1	2	3	4	5	6
2	5343	3615 (68%)	821 (15%)	907 (17%)				
3	803	496 (62%)	121 (15%)	81 (10%)	105 (13%)			
4	116	70 (60%)	10 (9%)	14 (12%)	11 (9%)	11 (9%)		
5	10	4 (40%)	1 (10%)	2 (20%)	1 (10%)	0 (0%)	2 (20%)	
6	1	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	1 (100%)

For each well the number of samples that were above the detection limit were determined and then summed and grouped by number of times a well was sampled. The count is presented with the percentage in parentheses. Zero indicates all samples were non-detect whereas the rightmost column in each row indicates all samples were above the detection limit.

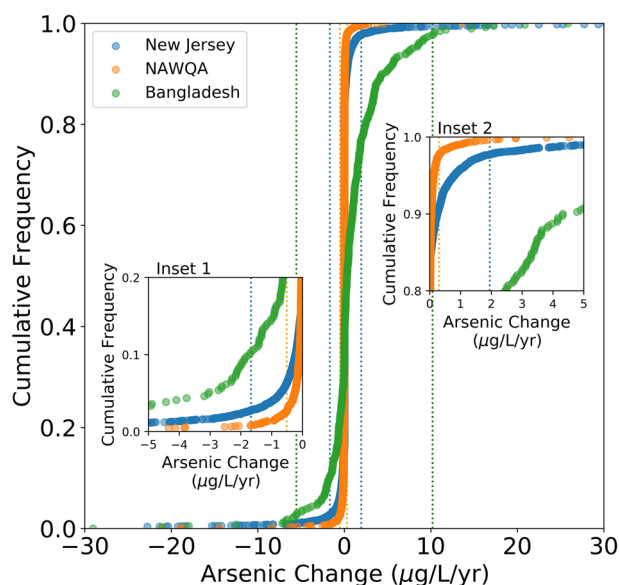


Figure 2. Cumulative distribution of rates of change in arsenic concentrations. The axes were limited to -30 to $30 \mu\text{g/L/yr}$ which does not show the larger rates for the New Jersey dataset that are associated with samples taken close in time. Inset 1 shows the distribution of rates for arsenic change ranging from -5 to $0 \mu\text{g/L/yr}$. Inset 2 shows the distribution of rates for arsenic change ranging from 0 to $5 \mu\text{g/L/yr}$. The dotted vertical lines show two standard deviations around the median for each dataset and match the color of the symbols. The orange line for NAWQA is barely visible as it is on the edge of the main data points.

had a mean concentration below $10 \mu\text{g/L}$ and 1848 (85.0%) were below $5 \mu\text{g/L}$ with 209 below the variable detection limit (Figure 1). The rates of change ranged from -126 to $13.6 \mu\text{g/L/yr}$ with the rates of change of 2 standard deviations around the median ranging from -0.51 to $0.30 \mu\text{g/L/yr}$ (orange dotted vertical line in Figure 2).

In the Araihasar region of Bangladesh, 391 private household wells were sampled twice approximately 14 years apart (Table 1). The mean arsenic concentration was $94.9 \mu\text{g/L}$ and the median was $61.3 \mu\text{g/L}$ with 72 locations (18.4%) below $10 \mu\text{g/L}$ and 55 (14.1%) below $5 \mu\text{g/L}$ with no samples below the detection limit

(Figure 1). The rates of change ranged from -29.1 to $25.9 \mu\text{g/L/yr}$ with the rates of change of 2 standard deviations around the median ranging from -5.55 to $10.2 \mu\text{g/L/yr}$ (green dotted vertical line in Figure 2).

Probability of Exceeding a Standard

For each dataset we determined the probability that the arsenic concentration exceeds the 5 and $10 \mu\text{g/L}$ MCLs in X years as estimated using the described methodology. The probability that a well with arsenic below $10 \mu\text{g/L}$ will exceed $10 \mu\text{g/L}$ generally increases with time (Figure 3). This probability is the same as the calculated fraction of wells that crosses an MCL. In all three datasets, a well with an initial concentration of $9 \mu\text{g/L}$ is estimated to have a greater than 5% chance of exceeding $10 \mu\text{g/L}$ within 2 years. When the initial concentration is lower, the probability that a well will exceed $10 \mu\text{g/L}$ decreases but probabilities still almost always increase over time. Repeating the analysis with the $5 \mu\text{g/L}$ MCL shows the same pattern (Figure 3). The probabilities flatline for the Bangladesh dataset and do not always increase over time. This occurs because of the smaller number of wells; the change in rates between the ordered wells is larger and thus as time increases more wells do not cross a threshold rate at each interval and the probability remains constant for multiple years. Given an initial sample concentration and time since sampling, the probability of exceeding an MCL can be determined directly from the graphs (Figure 3).

Utilizing the change in probability over time, it is possible to determine the predicted time for a well to have a 5% chance of exceeding an MCL given a starting concentration (Figure 4). In the New Jersey dataset, a well that starts with $5 \mu\text{g/L}$ arsenic has a 5% chance of exceeding $10 \mu\text{g/L}$ within 4 years and a well that starts with $3 \mu\text{g/L}$ arsenic remains below a 5% chance of exceeding $10 \mu\text{g/L}$ within 10 years (Figure 4A). With the $5 \mu\text{g/L}$ MCL, a well that starts with $3 \mu\text{g/L}$ has a 5% chance of exceeding an MCL within 5 years (Figure 4B). Only a well that begins with $1.5 \mu\text{g/L}$ or lower, has less than a 5% chance of exceeding the $5 \mu\text{g/L}$ MCL within 10 years (Figure 4B).

In the NAWQA dataset, a well that starts with $8 \mu\text{g/L}$ arsenic has a 5% chance of exceeding $10 \mu\text{g/L}$ within

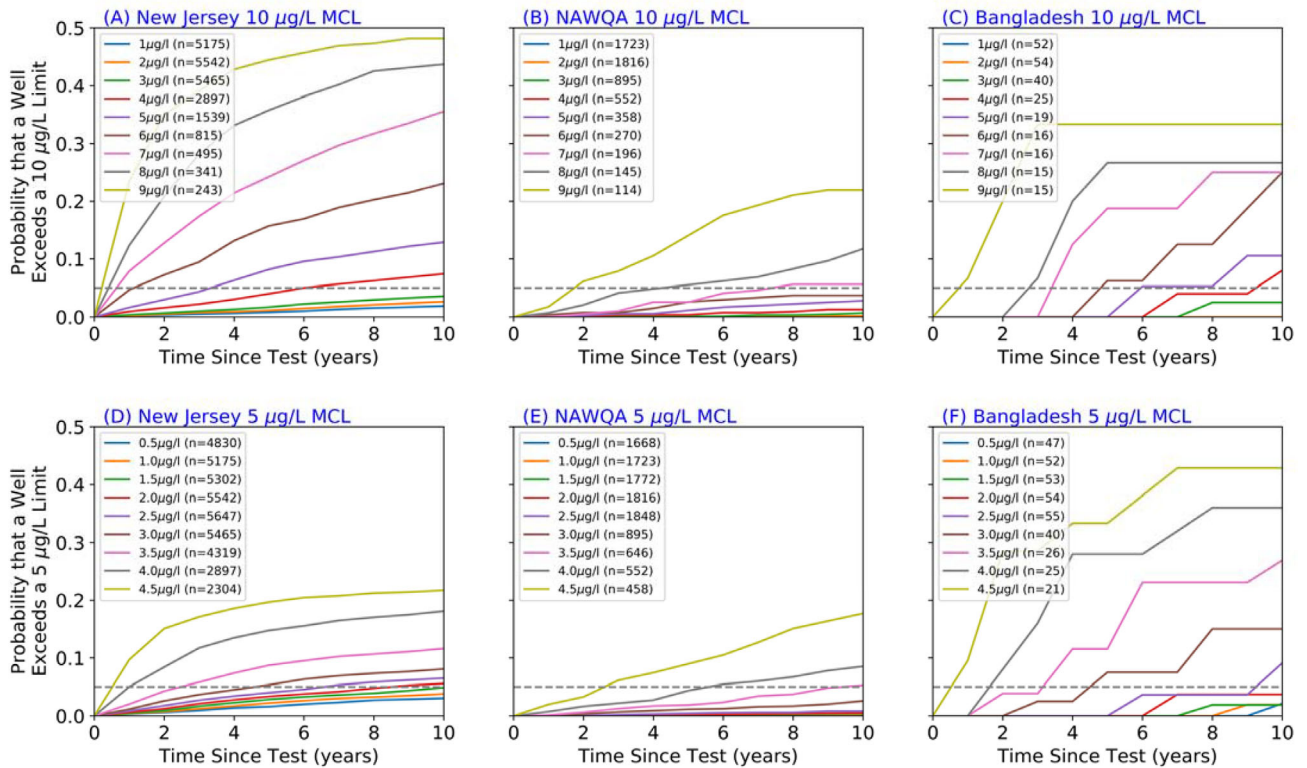


Figure 3. The probability that a well exceeds a 5 or 10 $\mu\text{g/L}$ maximum contaminant level (MCL) given a measured concentration and the number of years since sampling. Results are shown at a 10 $\mu\text{g/L}$ MCL for (A) New Jersey, (B) USGS NAWQA, (C) Bangladesh data, and a 5 $\mu\text{g/L}$ MCL for (D) New Jersey, (E) USGS NAWQA, and (F) Bangladesh. The number of wells used to estimate the probabilities at each concentration are listed in the legend. The wells chosen for each starting concentration were chosen using a window of plus or minus 2.5 $\mu\text{g/L}$ around the mean measured arsenic concentration (see step 3 in methods).

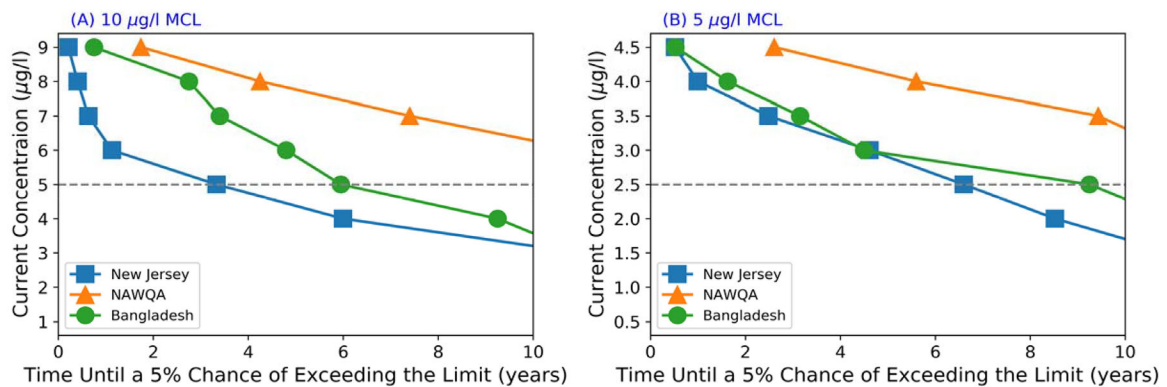


Figure 4. The length of time for a sample with current arsenic concentration (vertical axis) to have a 5% chance of exceeding (A) 10 $\mu\text{g/L}$ and (B) 5 $\mu\text{g/L}$. The time was determined using the data presented in Figure 3 by linearly interpolating between the two closest data points. The x-axis is truncated at 10 years to show the timeframe of interest. The horizontal line shows one-half the MCL for reference when developing the recommendations.

4 years and a well that starts with 7 $\mu\text{g/L}$ arsenic has a 5% chance of exceeding 10 $\mu\text{g/L}$ within 8 years (Figure 4A). Using a 5 $\mu\text{g/L}$ MCL, a well that starts with 4 $\mu\text{g/L}$ has a 5% chance of exceeding the standard within 6 years (Figure 4B). All samples that start with 3.5 $\mu\text{g/L}$ or less have lower than a 5% chance of exceeding the 5 $\mu\text{g/L}$ standard within 10 years (Figure 4B).

In the Bangladesh dataset, a well that starts with 6 $\mu\text{g/L}$ arsenic has a 5% chance of exceeding

10 $\mu\text{g/L}$ within 5 years and a sample that starts with 4 $\mu\text{g/L}$ arsenic has a 5% chance of exceeding 10 $\mu\text{g/L}$ within 10 years (Figure 4A). Using a 5 $\mu\text{g/L}$ MCL, a well that starts with 3.0 $\mu\text{g/L}$ has a 5% chance of exceeding the standard within 5 years (Figure 4B). Only a well with less than 2 $\mu\text{g/L}$ has less than a 5% chance of exceeding 5 $\mu\text{g/L}$ within 10 years (Figures 4B).

Discussion

Through time, arsenic concentrations in wells increased, decreased, or remained constant. Fortunately, the majority of wells have rates of change close to zero, but enough are changing that this has to be accounted for when estimating sampling intervals. Rates varied across datasets (Figure 2). The Bangladesh dataset consisted of the smallest number of wells and had the highest mean arsenic concentrations. This can lead to less precision in the estimates of the probabilities at concentrations near an MCL. The NAWQA data have the smallest magnitude rates of change (Figure 2) but these wells are mainly utilized for research purposes, and were analyzed in a government lab using standard protocols and thus might be less variable than private wells analyzed at various commercial labs. The New Jersey dataset is from private wells in an arsenic impacted region and the samples were analyzed at various state certified commercial labs.

The three datasets represent a diverse set of arsenic values collected using different methods from different parts of the world in different aquifer units with different geochemistries and well depths. These differences were not further investigated as the goal is to use a wide range of wells sampled multiple times for arsenic. The temporal nature of the data enables estimations of sampling frequencies. The goal of sampling a private well is to determine if the well is below an MCL for drinking water and to provide guidance to the well owner about the status of the well. It is important to be able to communicate to a well owner what the probability is that they are drinking water with arsenic concentrations exceeding the local, federal, or WHO drinking water MCL and to suggest a reasonable period of time in which they should retest the groundwater source.

In each of the datasets the observed temporal variability in the arsenic could be due to temporal changes in the actual arsenic concentrations or due to variability (errors) in the laboratory measurements. Temporal variability in arsenic has been observed in wells before and has been linked to changing hydrological and geochemical conditions (Erickson and Barnes 2006; Savarimuthu et al. 2006; McArthur et al. 2010; van Geen et al. 2013; Ayotte et al. 2015; Levitt et al. 2019). Laboratory errors, especially at locations with only two samples, will appear identical to temporal variability. However, for this analysis, separating measurement error from trends is not required because the goal is to determine how often a well-owner should sample and both factors contribute to the probability of a well being reported from going below an MCL to above it.

The New Jersey dataset has the largest fraction of non-detects and the methods used to analyze this portion of the data could impact recommendations (Figure 1). Methods exist for estimating slopes when some of the samples are below a detection limit, for example using the Akritas-Theil-Sen slope estimate, but proper usage of such methods is limited by sample size and the percentage of wells above detection levels (Helsel 2005). In the New Jersey dataset, 67% of the wells have all samples below

the detection level limiting the ability to calculate the rate of change for these wells. A value could be estimated for each sample using a method such as the maximum likelihood estimation or regression on order statistics (ROS) methods (Helsel 2005) and then calculating a rate using the estimated values. This approach is inappropriate for this assessment, as rates would be calculated with individually imputed or estimated values. Using only wells above the detection limit would skew results toward higher rates and ignore a large amount of valuable information. Setting the rates to zero for wells with all samples below the detection limit also skews the results as it assumes there is no change when concentrations could be trending toward or away from an MCL. Because of the high number of non-detects, there is no ideal method to estimate the rates in the New Jersey dataset. Setting the rates to zero for wells was chosen as the preferred approach for the New Jersey data since it retains the majority of data and does not rely on artificial imputed values for data below the detection limit. The results for New Jersey proved to be similar to the USGS and Bangladesh data. In addition, after determining a recommendation, it is possible to compare and validate the recommendation using the original data (see below).

A recommendation for a sampling interval for private well owners is proposed to ensure that there is less than a 5% chance that the water from a well exceeds an arsenic MCL (Table 3). This 5% probability was chosen to balance the frequency of sampling with risk, but a different value can be selected to evaluate different levels of risk tolerance. The recommendation is based on Figures 3 and 4 and is as follows (see summary in Table 3). At the 10 $\mu\text{g/L}$ MCL, wells with an arsenic concentration of 5 to 10 $\mu\text{g/L}$ should be tested every year and wells with less than 5 $\mu\text{g/L}$ should be tested every 5 years (Table 3). At the 5 $\mu\text{g/L}$ MCL, wells with an arsenic concentration of 2.5 to 5 $\mu\text{g/L}$ should be tested every year and wells with less than 2.5 $\mu\text{g/L}$ should be tested every 5 years (Table 3, panel B). Combining the recommendations, if the arsenic concentration is above one-half an MCL, a well owner should test every year, and if the concentration is below one-half an MCL a well-owner should test every 5 years. The recommendation, which is independent of the chosen MCL, will ensure that homeowners have less than a 5% chance of inadvertently consuming water with arsenic above an MCL.

The wells from the New Jersey dataset with two samples were used to check the recommendations. Wells with two samples were chosen as these are the most numerous, the time between the two samples is known, and it is easy to determine if a well goes from below to above an MCL. The recommendation was verified at two time intervals, 0 to 5 years and 4 to 6 years. There are 3642 wells that were sampled twice within a 5-year period. Twenty-two wells (0.60%) went from $<5 \mu\text{g/L}$ to exceeding the 10 $\mu\text{g/L}$ MCL within five years and 40 (1.10%) went from $<2.5 \mu\text{g/L}$ to exceeding the 5 $\mu\text{g/L}$ MCL within five years. There are 994 wells sampled

Table 3
Sampling Recommendations

A. Recommendation for sampling interval for a 10 µg/L Arsenic MCL.	
Initial Sample Concentration	Recommendation
5 to 10 µg/L	Test every year
<5 µg/L	Test every 5 years
B. Recommendation for sampling interval for a 5 µg/L Arsenic MCL.	
Initial Sample Concentration	Recommendation
2.5 to 5 µg/L	Test every year
<2.5 µg/L	Test every 5 years

twice between 4 and 6 years apart. Eight wells (0.80%) went from <5 µg/L to exceeding the 10 µg/L MCL and 9 (0.91%) went from <2.5 µg/L to exceeding the 5 µg/L MC. In both cases, the percentage of wells going from less than 1/2 an MCL to above an MCL was lower than 5%. Hence, it is concluded that for the New Jersey dataset, the recommendation indeed results in less than a 5% chance of inadvertently exceeding an MCL.

The results provide a clear data-driven message on sampling frequency for arsenic in private wells. The use of three different datasets that produce similar results indicates that the results are broadly applicable and independent of country and geology. It is not clear if the results would be the same for wells analyzed with field kits which are a popular low-cost method (George et al. 2012; van Geen et al. 2019) but this can be tested when datasets become available. Furthermore, as more datasets become available and the New Jersey dataset continues to grow the recommendation can be continually verified and, if needed, updated.

Conclusions

A single analytical sample of arsenic below a MCL is not enough to ensure that drinking water from private wells remains low because of measurement error and because arsenic concentrations change over time. Rates of arsenic change were analyzed from three diverse datasets, New Jersey Private wells, the USGS NAWQA database, and Bangladesh private wells. In each dataset, arsenic concentrations varied with time and a subset went from below to above a MCL between sampling events. The rates of change in arsenic concentration from each well were utilized to estimate the probability that a well will exceed an MCL in the future and to recommend how often a private well owner should test a well for arsenic. The goal was to develop a clear data-driven message about how often to sample private drinking water wells to ensure with a high probability that well water is below an MCL. Based on the analysis it is recommended that private wells

are tested for arsenic every 5 years but yearly if the well exceeds 1/2 the MCL. This recommendation holds for both the 5 and 10 µg/L MCL. This probability-based approach for determining recommendations for testing frequency may be useful for other contaminants as well as being considered when setting or revising MCLs.

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