PLOS WATER

RESEARCH ARTICLE

Geologic predictors of drinking water well contamination in North Carolina

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OPEN ACCESS

Citation: Alvarado TR, Austin RE, Bradley PJ, Eaves LA, Fry RC, George A, et al. (2024) Geologic predictors of drinking water well contamination in North Carolina. PLOS Water 3(1): e0000194. https://doi.org/10.1371/journal.pwat.0000194

Editor: Daniel Reddythota, Faculty of Water Supply & Environmental Engineering, ArbaMinch Water Technology Institute (AWTI), ETHIOPIA

Received: August 1, 2023

Accepted: December 4, 2023

Published: January 11, 2024

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Data Availability Statement: Census-tract and county -level datasets constructed from the NCWELL database are available online at https://doi.org/10.15139/S3/BDQG90 [13,15]. The base map of North Carolina is available from https://www.nconemap.gov/. All other data associated with this paper are included in the supporting information.

Funding: This research was funded in part by a UNC Superfund Program grant from the National Institute of Environmental Health Sciences

Abstract

More than 200 million people worldwide, including 11 million in the US, are estimated to consume water containing arsenic (As) concentrations that exceed World Health Organization and US EPA standards. In most cases, the As found in drinking water wells results from interactions between groundwater and geologic materials (geogenic contamination). To that end, we used the NCWELL database, which contains chemical information for 117,960 private drinking wells across North Carolina, to determine the spatial distribution of wells containing As contaminated water within geologic units. Specific geologic units had large percentages (up to 1 in 3) of wells with water exceeding the EPA As maximum contaminant level (MCL, 10 μg/L), both revealing significant variation within areas that have been previously associated with As contamination and identifying as yet unidentified problematic geologic units. For the 19 geologic units that have >5% of wells that contain water with As concentrations in exceedance of 10 μg/L, 12 (63%) are lithogenically related to the Albemarle arc, remnants of an ancient volcanic island, indicating the importance of volcanogenic materials, as well as recycled (eroded and deposited) and metamorphosed volcanogenic material. Within geologic units, wells that have As concentrations exceeding 10 μg/L tended to have pH values greater than wells with As concentrations less than 10 μg/L, emphasizing the importance of the extent of interaction between groundwater and geologic materials. Using census information with the geologic-based exceedance percentages revealed the importance of regional geology on estimates of population at risk compared to estimates based on county boundaries. Results illustrate that relating As contamination to geologic units not only helps explain sources of geogenic contamination but sharpens the

(P42ES031007). This work was supported by the USDA National Institute of Food and Agriculture, Hatch projects NC02713 and NC02951. This work was made possible by a US Department of Defense SMART Fellowship Program awarded to T.R.A. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

Competing interests: The authors have declared that no competing interests exist.

identification of communities at risk for exposure and further illuminates problematic areas through geologic interpretation.

1. Introduction

Consumption of arsenic (As) contaminated drinking water is associated with increased risk of cancer, diabetes, and kidney disease [1]. Worldwide, >200 million people are estimated to drink water containing As concentrations that exceed the United States (US) Environmental Protection Agency (EPA) and World Health Organization (WHO) standards (EPA maximum contaminant level (MCL) and WHO provisional guideline = 10 μg/L) [2, 3]. In the US, where roughly 15% of the population utilize private wells for domestic water [4], nearly 11 million people are exposed to As concentrations exceeding 10 μg/L in well water [5]. Most commonly, As contamination of well water is geogenic [5], resulting from the solubilization of naturally occurring As from geologic materials. Generally, private well users are responsible for monitoring and remediating their wells and not subject to regulatory standards such as the MCL. It is worth noting that the EPA Maximum Contaminant Level Goal (MCLG), the level at which a contaminant is deemed to pose no hazard to human health, for arsenic is 0, although this is a non-enforceable standard. Individual decisions about private well water are hindered by socioeconomic barriers, such as lack of access to testing, cost of well testing, cost of treatment, and lack of awareness of exposure risk [6–8].

North Carolina has been proposed as model area for well water quality in the US [9, 10] because it: (1) ranks in the top five states in population using private domestic wells as their drinking water supply [4, 11]; (2) has an estimated 40,000–120,000 of people consuming As contaminated drinking water [12–14]; (3) has mandatory new well testing and a large water chemistry data set from private wells [13, 15]; (4) exhibits significant socioeconomic and racial disparities in access to safe drinking water [10, 16]; and (5) presents a diverse aquifer lithology, including volcanic, intrusive, sedimentary, and metamorphic rocks thought to contribute variable amounts of As to groundwater [17]. North Carolina's geology, consisting of Blue Ridge and Piedmont crystalline rock terranes, Coastal Plain sedimentary formations, and minor interior Mesozoic sedimentary basins, is broadly typical of other Eastern US states from Alabama to New York [18].

Previous studies have documented statewide spatial variation in As concentrations in water from in private wells at the county level [13, 14]. However, the distribution of groundwater As concentrations within a broad geographic area (larger than the scale of the local flowpath) likely result from geogenic occurrence at the scale of a map-scale geologic unit [19–21]. Consequently, studies have examined As in drinking water from private wells at the scale of single county or region or a few counties [22–26]. In addition, Coyte and Vengosh [21] related the occurrence of As and other elements in drinking water wells to regional-scale geologic belts or terranes, finding that As occurs most frequently in the Carolina Slate belt. Although these previous studies greatly inform our understanding of As contamination in NC private wells, they have either been limited in the extent of geographic coverage of well water sampling data, resulting in a lack of statistically meaningful sample sizes across many rock types, or have analyzed the data at a county level and were thus not informed by geology. Thus, a comprehensive statewide analysis that relates As contamination of water in private drinking wells, as well as estimates of population served by them, to individual fine-scale geologic units is currently lacking [19–21].

Identifying and characterizing map-scale geologic units that pose significant risk of geogenic contamination, the objective of this study, will improve protection of private well users from As exposure by focusing testing, information, and treatment efforts on areas with the highest probability of As occurrence. Here we relate the prevalence of As occurrence greater than 10 μg/L in water from private wells from a large, recently-compiled statewide dataset with >100,000 As analyses [13] to a new reclassification of map-scale geologic units in North Carolina (NC). Using this new, comprehensive geospatial analysis of As occurrence, we estimate the potential population exposure to As in drinking water based on the populations of people drinking well water from within each geologic unit, suggesting that population-scale As exposure is both concentrated in a volcanogenic geologic units and dispersed in several other regionally extensive rock types where As occurrence >10 μg/L frequently occurs.

2. Methods and materials

2.1 NCWELL database

The well water database (NCWELL [13, 15]) that was used for this analysis was constructed from NC Department of Health and Human Services State Laboratory for Public Health well water tests from October 19, 1998, to May 20, 2019, resulting in a dataset of 117,960 total well tests. This dataset was used in our geospatial analysis to determine the influence of geology on the occurrence of As concentrations >10 μg/L in private well water. This dataset represents all samples submitted by well owners to the State Laboratory of Public Health, most often via County Health Departments, during the time period noted above. Further details about well testing and analytical methods, geocoding, data cleaning, and temporal trends are presented in Eaves et al. [13]. In brief, approximately 80% of tests from NCWELL are unique well locations and the remaining are likely duplicate tests. Temporal variability in As in NCWELL was not noted in previous analyses and thus averaging over sampling time period is justified [13]. Based on an estimated 1.6 million domestic wells in NC [4], the NCWELL database represents approximately 5.9% of known private domestic wells in the state, and on average represents ~0.67 tested wells per km^2.

A subset of well tests with As included pH measurements taken in the same sampling event as the water sample for As analysis ($n = 66064$, representing 56% of the entries in the NCWELL database). This subset was used in an analysis to determine relationships between pH and dissolved As concentrations >10 μg/L. We note that the NCWELL dataset includes only private wells and does not include any public supply wells (i.e., wells supplying at least 25 people year round, which would constitute a community water system and be subject to the Safe Drinking Water Act).

2.2 Geologic units

The digitized and georeferenced version of the Geologic Map of North Carolina [17] provides the spatial foundation for our analysis of naturally occurring As. The map, as published, contains the boundaries and attributes for 19 regional scale geologic belts (e.g., regionally extensive rock complexes with internally consistent tectonic histories and age ranges. It is worth noting here that, the term “belt”, used in this study in order to remain consistent with the North Carolina geological map, has been abandoned and “terrane” has been adopted by the geologic academic community. On a finer scale, the state map contains 134 “geocodes”, which are abbreviations used to denote areas on the map that contain a specific rock type. A limitation of the “geocodes” is that one geocode may be used to denote rocks of the same type in multiple geologic belts (that is, genetically unrelated rocks with similar map-scale lithological or stratigraphic characteristics, e.g., “mafic volcanic rocks” could receive a single geocode).

Another example of the limitations of the previous geocodes, is that some geocodes refer to rock texture (e.g., gneiss) without regard to the protolith (that is, the rock that originally formed, such as a volcanic rock, before its eventual metamorphism). Therefore, geocodes could group together rocks of distinct origins and therefore also, group together rocks with dissimilar As content. For these reasons, we modified the geocodes, as published, into a belt-geocode classification, referred to as "geologic units". Geologic units within each belt are designated as distinct from similarly geocoded rocks in other belts. This approach results in grouping spatially disparate areas bearing the same geocode (e.g., rock type) within a geologic belt (belts are assumed to be internally genetically related) while dividing similar geocodes from different geologic belts into separate geologic units. Dividing 134 geocodes resulted in 201 unique geologic units (S1 Fig and S1 Table). Where our geologic units coincide with formally named Groups, Formations, or Members, we apply these accepted names to our operationally defined geologic units.

2.3 Statistical analysis

The US EPA maximum contaminant level (MCL, 10 μg/L) was used as the threshold concentration for this study, although private wells are not regulated by the MCL. Although laboratory reporting limits for As were not consistent within the NCWELL dataset, all reporting limits were $\leq$10 μg/L, and thus differences in reporting limits did not affect our analysis. To estimate the frequency of As $>$10 μg/L (referred to herein as the As exceedance percentage), the count, Y_i, of well containing water with As $>$10 μg/L, was divided by the number of arsenic analyses in the NCWELL database, n_i, in each geologic unit, i. Approximate 95% confidence intervals [27] for the theoretical relative frequency of exceedance, π_i, were constructed under the model which takes $\{Y_i\}$ to be binomial random variables with exceedance frequencies $\{\pi_i\}$. This method accommodates the zeros ($y_i = 0$) observed in the data, where no wells were observed to be in exceedance for a particular geologic unit combination i. The intervals were computed using SAS PROC FREQ [28]. Visualizations of these frequencies was conducted using ArcGIS Pro version 2.292.

To compare observed pH of water in wells within geologic units in the NCWELL database, normal theory t-tests were performed along with non-parametric Mann-Whitney tests. Within each geologic unit samples were divided into two populations, those above $>$10 μg/L As, and those below. T-tests were used to determine significance between population means when distributions of pH levels were assumed normal and variances unrestricted. Mann-Whitney tests were run to determine significance on non-normal distributions. To assess the assumption of normally distributed pH levels, residuals were computed by subtracting the average pH corresponding to samples in the population with As concentrations greater than or less than the 10 μg/L As threshold. Tests for the goodness-of-fit of a normal distribution were conducted (Anderson-Darling) after forming a single sample of all of the residuals for each formation. The geologic units with significance differences in mean pH between those above and below the 10 μg/L As threshold, and under assumptions of both normality and no-normality, were reported as significant. Formations with less than five samples were excluded from mean difference tests for statistical power; however, results for all formations with an exceedance probability greater than 5% are included in the figures for comparison.

2.4 Estimations of population at risk to As exposure

To help understand the influence of underlying geology on human exposure within counties, an estimate of the population that may be served by private wells containing As concentrations $>$10 μg/L was calculated. The calculation utilized the exceedance percentages within each geologic unit multiplied by an estimate of individuals consuming water from private wells underlain by that unit.

Within a county, the number of people consuming water from private wells was obtained from the US Geological Survey database for 2015 [42]. County population totals were captured from the US Census Bureau Population Estimates for 2018 through the tidycensus R package [43]. To determine the percent of the population drinking from private wells within a county, the number of people estimated to consume water from private wells was divided by the total population. To transfer the percentage of the county drinking from private wells to the percentage in geologic units, an overlay operation was performed within a Geographic Information System with the assumption that the population using private drinking wells is uniformly distributed across each county. This distribution was calculated by dividing the population using private drinking wells by the total area, yielding the population per area on private wells. County boundaries were then intersected with the geologic unit boundaries to create new features representing the unique geologic units within the county. The area for each unique geologic unit was calculated and multiplied by the population per unit area, resulting in the population on private wells within the geologic unit. The geologic units were then reassembled across county boundaries providing an estimate of the population using private wells. The total number of people within the geologic unit consuming private well water was multiplied by the As exceedance percentage within each geologic unit to estimate the exposed population.

3. Results and discussion

3.1 Arsenic in well water in North Carolina

Of the 117,960 total As results in the NCWELL database, 2493 exceeded 10 μg/L (2.1%). Among geologic belts (Fig 1A and Table 1), the Carolina Slate belt (equivalent to the Carolina Terrane) had the greatest As exceedance percentage (5.3%, with 1584 of 28156 wells with water containing As concentrations that exceeded 10 μg/L). This observation is consistent with findings of Coyte and Vengosh [21], who found the Carolina Slate belt to have the most frequent occurrence of elevated As concentrations in groundwater, and Pippin et al [24], who focused their study primarily within the Carolina Slate belt. The Grandfather Mountain Window (3.1%; 24 of 755 wells containing water with As concentrations greater than 10 μg/L) and the Inner Piedmont (2.9%; 217 of 7271 wells containing water with As concentrations greater than 10 μg/L) were the second and third belts in terms of percentage of wells containing water with As exceedance of 10 μg/L, respectively. It is worth noting that the Carolina Slate belt and the Inner Piedmont cover large geographic areas [17] and thus the probability of As exceedance may vary with location within belts.

3.2 Analysis of arsenic in well water at the geologic unit scale

In finer scale analyses, 19 of the 201 geologic units have As exceedance percentages above 5% (Fig 2A and Table 2). Notably, these geologic units span nine of the 19 major geologic belts (S1 Fig and S1 Table), showing the potential for As contamination from rocks with varying compositions and geologic histories. This analysis also highlights the variability within a geologic belt. For example, geologic units within the Carolina State belt, the geologic belt with greatest (5.3%) exceedance percentage (5.3%), have well test exceedance percentages that vary from effectively 0 to as large as 33.8%. It is worth noting that some units (Zbg, Ashe Formation, 2nd greatest exceedance percentage and Ccl, Lower Chilhowee Group, 5th greatest exceedance percentage) contain small datasets ($n \leq 6$), resulting in very large confidence intervals. These geologic units thus are potential targets for future testing to determine if they represent areas of increased exposure risk.

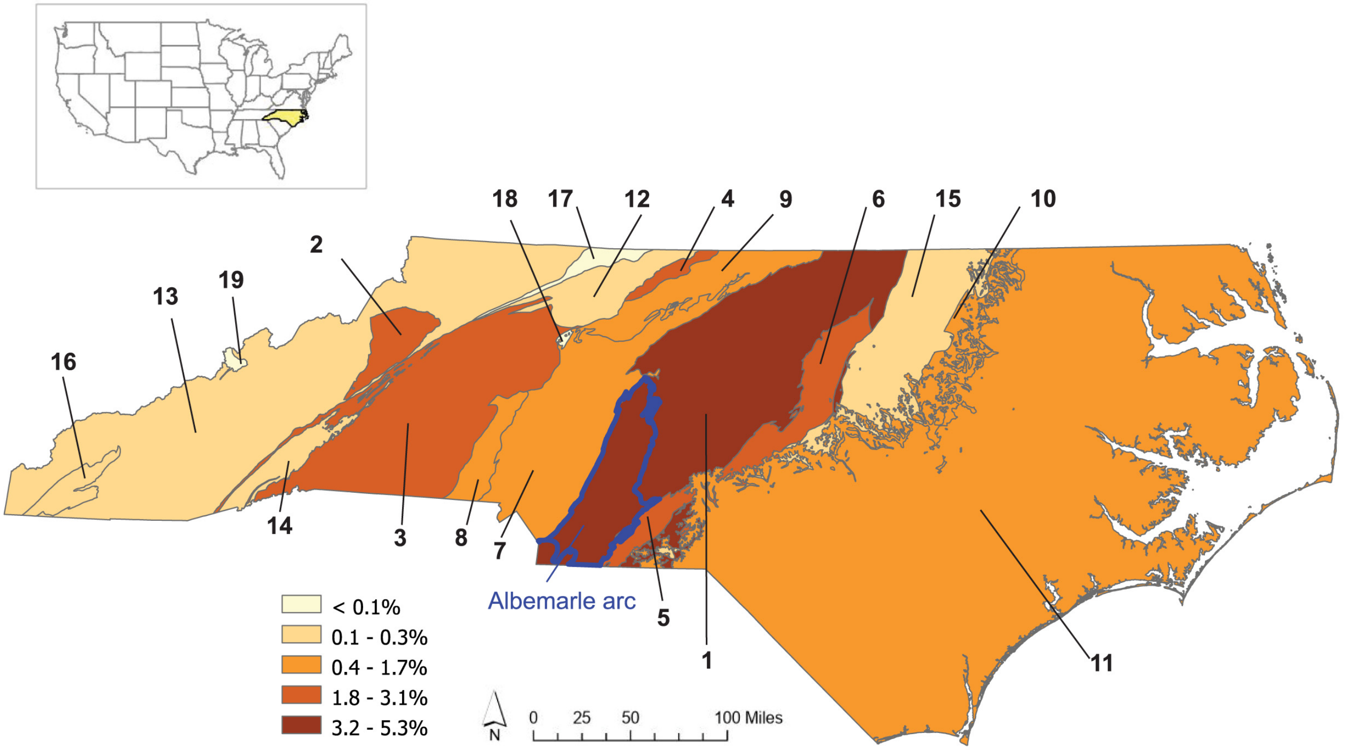

Fig 1. Percentage of tested private wells containing water with as concentrations exceeding 10 μg/L within geologic belts. Numeric labels correspond to rows in Table 1. The Albemarle arc, an area of interest within the Carolina Slate belt, is outlined in blue. The map and license information can be found at: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03.

https://doi.org/10.1371/journal.pwat.0000194.g001

A cluster of geologic units with large As exceedance percentages occurs in the youngest portion of the Carolina Slate belt—the Albemarle arc (Fig 2B)—that lies in a geographic region previous noted as having elevated As concentration in well water [13, 14, 24]. The Albemarle arc along with the rest of the Carolina Slate belt was a zone of suprasubduction that accreted to North America [29, 30]. Other geologic events in these regions included intense metamorphism, folding, faulting, and igneous intrusive activity, resulting from collisions and rifting events associated that contribute to the complex geology of the Albemarle arc [31, 32]. Solid phase As concentration (~8–21 mg As/kg C horizon material) in subsurface material taken from within the Albemarle Arc is greater than in the surrounding area (<5 mg As/kg C horizon material) [33], and that volcanogenic sediment has been identified as a source of As in groundwater in many parts of the world [19, 34–36].

Across all geologic units, the greatest percentage of wells containing water exceeding 10 μg/L As (Fig 2A, 2B) is associated with mudstones within the Cid Formation (Table 2, geocode: CZmd2), which has approximately 1/3 of wells containing water with As concentrations in exceedance of 10 μg/L and was previously been identified as a formation with large As exceedance percentage [21, 24, 37]. Generally, the metasedimentary Cid mudstone is thought to represent the sediment eroded from volcanic formations that formed ~545 Ma. Related geologic units in the metavolcanic-metasedimentary Albemarle Arc also have large exceedance percentages, including the metasedimentary Tillery Formation (geocode: CZmd1, 3rd greatest exceedance percentages, 20%), the Cid Formation metasedimentary (CZmd2, mudstone, greatest exceedance, 33.8%) and metavolcanic rocks (CZfv2, felsic metavolcanics, 7th greatest

Table 1. Occurrence of As in private drinking water wells for major geologic belts in North Carolina. Percent exceedance indicates the percentage of values greater than 10 μg/L. LCL = lower confidence interval; UCL = upper confidence interval.

Rank	Geologic belt	Arsenic analyses (n)	Results above >10 μg/L (n)	LCL (95%)	Percent Exceedance	UCL (95%)
1	Carolina Slate belt	28156	1584	5.1	5.3	5.6
2	Grandfather Mountain Window	755	24	2.0	3.1	4.6
3	Inner Piedmont	7271	217	2.5	2.9	3.3
4	Dan River Basin	255	7	1.1	2.7	5.4
5	Wadesboro Subbasin	89	2	0.27	2.2	7.7
6	Durham-Sanford Subbasins	1548	33	1.4	2.1	2.9
7	Charlotte belt	14112	236	1.4	1.6	1.9
8	Kings Mountain belt	2900	47	1.2	1.6	2.1
9	Milton Belt	2173	31	0.96	1.4	2.0
10	Eastern Slate belt	1909	26	0.88	1.4	2.0
11	Coastal Plain	22234	232	0.90	1.0	1.2
12	Sauratown Mountains Anticlinorium	1398	4	0.08	0.29	0.73
13	Blue Ridge	21663	36	0.12	0.17	0.23
14	Chauga belt	1944	3	0.03	0.15	0.45
15	Raleigh belt	5679	8	0.06	0.14	0.28
16	Murphy belt	2397	3	0.03	0.13	0.36
17	Smith River Allochthon	441	0	0	0	0.83
18	Davie Basin	34	0	0	0	10.3
19	Hot Springs Window	33	0	0	0	10.6

https://doi.org/10.1371/journal.pwat.0000194.t001

exceedance, 11.4%; CZmv1, mafic metavolcanic rocks, 8th greatest exceedance, 10.0%), the Floyd Church Formation metasedimetary rocks (CZmd3, 11th greatest exceedance, 8.3%), and the Yadkin Formation metasedimentary greywacke (CZy, 13th greatest exceedance, 6.9% exceedance) [31, 32].

Additional rocks surrounding the Albemarle arc and influenced by it contain As exceedance percentages that are >5%. The Gold Hill Shear zone (CZph, 4th greatest exceedance, 19.1%), a zone of deformation that marks the contact between the Carolina Slate belt and Charlotte belt, is hosted in rocks associated with the Cid and Tillery Formations [29]. Additionally, recent mapping efforts [38] have indicated that a geologic unit composed of undifferentiated Albemarle arc and Hyco arc sediments (CZmd, 14th greatest exceedance, 6.8%) in the eastern part of the Carolina Slate belt is also likely related to the Cid Formation.

As described above, eight of the 19 formations that have >5% exceedance are part of or directly linked to the Albemarle arc. Formations surrounding the Albemarle arc, both within and outside the Slate belt, exhibit lower exceedance percentages. The northern (and older) part of the Carolina Slate belt, the Hyco arc, has significantly lesser exceedance percentages for As (<1.9%; S1 Table), supporting the geologic influence and spatial variability within the Carolina Slate belt [24]. With the exception of the Gold Hill shear zone, the geological units in the Charlotte belt, with its distinct geologic history from the Caroline Slate belt [29], have comparatively small exceedance percentages (<2.8%; S1 Table).

3.3 Relationships between geologic units with large exceedance probabilities

The influence of the Albemarle arc and its associated geologic units span beyond the Carolina Slate belt. The rocks of the Cat Square terrane (CZms, 16th greatest exceedance, 6.4%), part of the Inner Piedmont, are thought to have originated as sediment sources from the Albemarle

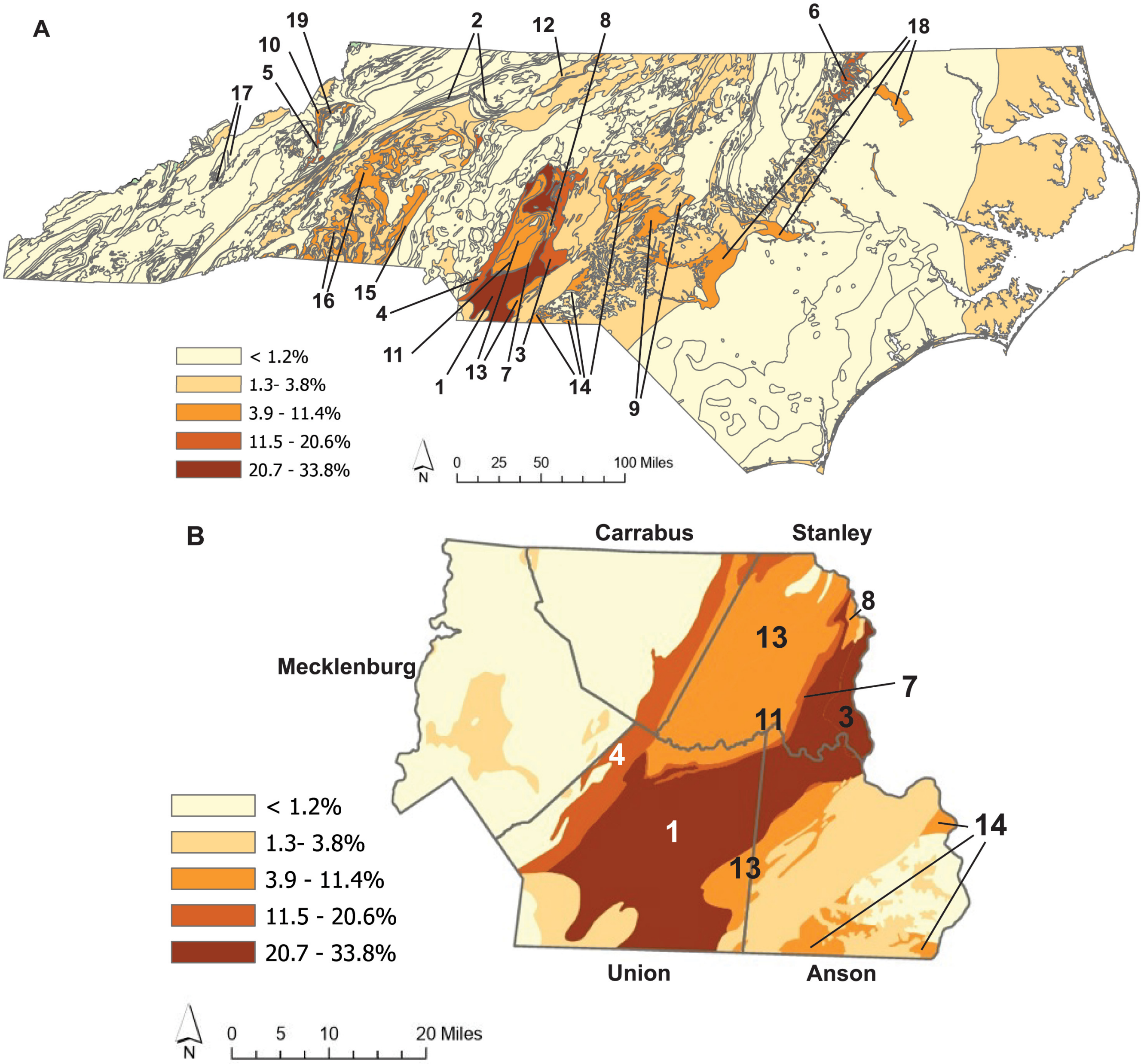

Fig 2. Percentage of tested wells with As greater than 10 μg/L within (a) geologic units. Exceedance probabilities vary significantly within and across county boundaries with elevated risk associated with units related to the Albemarle arc (b). Numeric labels correspond to rows in Table 2. The map and license information can be found at: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03.

https://doi.org/10.1371/journal.pwat.0000194.g002

arc as the paleo-continent Carolinia accreted to ancient North America [39], implying that these geologic units share a similar As source despite the Cat Square terrane being located ~60–70 km from the Albemarle arc today (Fig 2A). Among sedimentary aquifers, the Sanford sub-basin of the Deep River Triassic basin (TRcs, 9th greatest exceedance, 9.6%), likely received sediments from erosion of rocks in the Albemarle arc, which may explain the As source within the Triassic sedimentary formations. Furthermore, the Blacksburg Formation, consisting of metasedimentary rocks (CZms, 15th greatest exceedance, 6.5%), and the Eastern Slate belt metamorphosed felsic metavolcanics (PzZq, 6th greatest exceedance, 12.9%) are both similarly aged volcanic island arcs that are genetically related to the Albemarle arc [29]. Due to the similar age of volcanic activity within these geologic units, and presence of felsic volcanogenic rocks, we infer that these have a similar As source as the Albemarle Arc rocks.

Thus, of the 19 units with greater than 5% exceedance, 12 can be linked genetically to the Albemarle arc. This highlights the value of using geologic history as tool to identify possible regions that may pose a risk for geogenic contamination. Furthermore, we again note that Ashe Formation and Lower Chilhowee Group contain small datasets ($n \leq 6$), and that confidence intervals on their exceedance percentage are large and approach zero. Exceedances in the Grandfather Mountain Window (Zgma, Grandfather Mountain meta-arkose, 10th greatest exceedance percentage, 8.5% and Zgms, Grandfather Mountain metasiltstone, 18th greatest exceedance percentage, 5.3%) are thought to have volcanogenic origins analogous to the Albemarle arc, but from older events. The Grandfather Mountain Window consists of metavolcanic and associated metasedimentary rocks developed during the breakup of an older supercontinent, Rodinia [40]. As with the younger Albemarle arc, the presence of volcanic rocks and sediments derived from the erosion of those volcanic rocks seems to control groundwater As in multiple terranes and settings, including distant from the well-documented Cid formation mudstone in the Carolina Slate belt. Therefore, we see the consistent impact of volcanogenic material on groundwater As that requires detailed understanding of the genetic history of rocks in the region. Common terminology for some rock units (e.g., schist, mudstone) does not communicate the proto-lithologic origin, that is critical for predicting As in groundwater (e.g., felsic metavolcanic rocks or volcanic-source sediment in mudstone).

In general, the presence of naturally occurring As in the regional groundwater is connected to ancient volcanic activity, the erosion and deposition of volcanic source rock and the recycling of those As prone rocks into sedimentary basins (i.e., Cat Square terrane and Sanford sub-basin of the Triassic basin). Our results generally agree with previous analysis of Pippin et al. [24] who specifically noted frequent detection (>1 μg/L) in metavolcanic and associated metasedimentary rocks within, primarily within the Slate Belt and Inner Piedmont geologic belts. However, we note that nine of the geologic units with $>5\%$ of wells containing water with As concentration exceeding 10 μg/L (Table 2) were not identified by Pippin et al. [24], likely due to the larger data set (13,337 vs. 117,960 As well tests) used in our analysis.

3.4 Indicators of arsenic contamination within geologic units

Within geologic units that have large exceedance percentages ($>5\%$), the location of wells containing water with As concentrations exceeding 10 μg/L were spatially independent (average Moran's $I = 0.2$, $p = 0.37$), with occurrence of exceedance and non-exceedance of As varying over short (<100 m) distances. This spatial variation occurs at a smaller scale than the mapped geological units, at the scale of a local groundwater flow path. While this study has identified the distributions of As exceedance in map-scale rock types, prediction of As in an individual well could depend significantly on local factors, including water flowpath and small-scale subsurface heterogeneity.

Although map-scale geology is a primary control of groundwater As, and all drinking water wells are recommended for As testing [41], identifying indicators for hydrogeochemical processes that mobilize As into groundwater within geologic units may further improve our ability to predict wells at risk and set priorities for public health interventions, such as providing treatment or alternative water sources. Fig 3 depicts pH distributions from wells testing above and below the As MCL for the geologic units with As exceedance percentages $>5\%$. The median pH of groundwater with As concentrations that exceed 10 μg/L tends to be higher

Table 2. Occurrence of As in drinking water wells for 19 geologic units that have percent exceedance greater than 5%. Percent exceedance indicates the percentage of values greater that EPA primary drinking water standard (10 μg/L). LCL = lower confidence interval; UCL = upper confidence interval.

Rank	Geologic Units	GeoCode	As analyses (n)	As above 10 μg/L (n)	LCL (95%)	Percent Exceedance	UCL (95%)	Estimated well users above 10 μg/L
1	Cid Formation -metamudstone	CZmd2	2720	919	32.0	33.8	35.6	10086
2	Ashe Formation	CZbg	3	1	0.84	33.3	90.6	104
3	Tillery Formation	CZmd1	936	193	18.1	20.6	23.4	2099
4	Gold hill shear zone	CZph	961	184	16.7	19.2	21.8	1701
5	Lower Chilhowee group	Ccl	6	1	0.42	16.7	64.1	42
6	Eastern Slate Belt metamorphosed felsic metavolcanics	PzZq	54	7	5.4	13.0	24.9	510
7	Cid Formation—felsic metavolcanics	CZfv2	289	33	8.0	11.4	15.7	416
8	Cid Formation—mafic metavolcanics	CZmv1	30	3	2.1	10	26.5	88
9	Sanford sub-basin	TRcs	218	21	6.1	9.6	14.4	658
10	Grandfather Mountain Window meta-arkose	Zgma	165	14	4.7	8.5	13.8	151
11	Floyd Church Formation	CZmd3	948	79	6.7	8.3	10.3	1518
12	Cow Branch Formation	TRdc	24	2	1.0	8.3	27.0	112
13	Yadkin Formation	CZy	318	22	4.4	6.9	10.3	488
14	Uwharrie and Hyco mudstone	CZmd	815	55	5.1	6.8	8.7	852
15	Blacksburg Formation	CZbl	573	37	4.6	6.5	8.8	548
16	Cat Square Terrane	CZms	1863	119	5.3	6.4	7.6	4115
17	Earlies Gap Formation	Ybam	16	1	0.16	6.3	30.2	5
18	Cape Fear Formation	Kc	810	47	4.3	5.8	7.6	1724
19	Grandfather Mountain Window metasiltstone	Zgms	150	8	2.3	5.3	10.2	65

https://doi.org/10.1371/journal.pwat.0000194.t002

than that of wells containing water with As concentrations less than 10 μg/L. For nine geologic units, the pH distributions are significantly different at the 95% (*) confidence level; the lack of significance is largely found in geologic units that have a small number of wells containing water exceeding 10 μg/L As, including six geologic units that do not have adequate sample size to support the test.

Alkaline pH has previously been suggested to indicate increased extents of interaction with aquifer materials because weathering and reduction reactions consume protons [20, 42]. In the case of geogenic contamination, As in groundwater is thought to result from solubilizing interaction between water and soil, saprolite, and aquifer materials, so increased extent of reaction within the same geologic unit may result in higher dissolved As concentrations [12, 23, 25, 36]. Greater pH could record: (1) proton consumption from weathering reactions, which would promote desorption of oxyanionic forms of As(V) species; and/or (2) proton consumption from As, manganese (Mn), and iron (Fe) reduction, all of which consume protons and may result in As solubilization through formation of more soluble As(III) and concomitant As dissolution from the reduction of Mn and Fe (oxyhydr)oxide minerals. Through either pathway, increasing pH occurs as part of the evolution of groundwater along flowpaths undergoing acid-base and/or redox reactions [34, 42]. Additional studies on the relationship between pH and other geogenic metals are needed to better understand contaminant distributions.

3.5 Human exposure to arsenic

Due to the complex and spatially distinct geology in the Piedmont region and the presence of geologic units with large and small As exceedance percentages in the same county, assignment

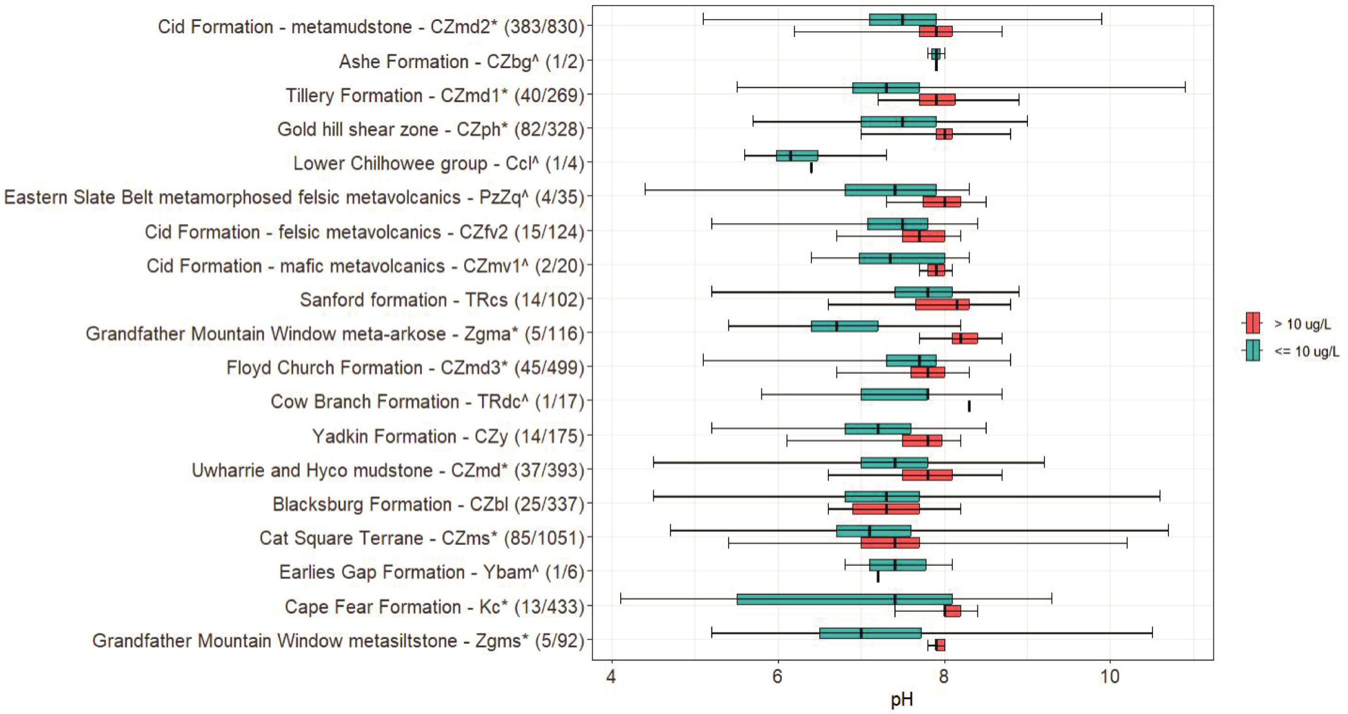

Fig 3. Distribution of pH in drinking well water from wells testing above (red) or below (blue) the 10 μg/L for the 19 geologic units with exceedance probabilities >5%. Significant differences at the 95% confidence level denoted with an asterisk (*) whereas those denoted with a caret (^) do not support the analysis (<5 data points that exceed 10 μg/L).

https://doi.org/10.1371/journal.pwat.0000194.g003

of exceedance percentages for geologic units provides finer resolution for assessing exposure than county-level analyses. For example, Union County was previously identified as having a large percentage of wells containing water that exceed 10 μg/L As (19.5% [13, 14]). Much of this county is underlain by geologic units that contain a large percentage of wells containing water with As concentrations in exceedance of 10 μg/L, such as the Cid mudstone (CZmd2, 33.8%) and the Gold Hill Shear zone (CZph, 19.1%). However, volcanic tuffs in the western part of the county and younger intrusive rocks in the southwestern corner of the county have a small percentage of wells with water in exceedance of 10 μg/L As (0.54% and 0.30%, respectively). Therefore, expressing As exceedance percentages by county rather than by geologic unit can obscure exposed populations by averaging geologic units with large As exceedance percentages with adjacent units containing lesser As exceedance percentages. For example, the estimated percentage of wells with water exceeding 10 μg/L, a proxy for the risk of exposure, within geological units in Union County may vary by as much as 110-fold (Fig 2B).

The spatial distribution of geologic units also impacts estimates of human exposure to As through well water consumption. By estimating the population on wells living on a geologic unit, the exposed population can be estimated. We recognize that this approach has limitations: (1) estimates of exceedance probabilities are based on one time measurements over a 21 year period [13]; (2) estimates of population on wells are derived from 1990 census data with assumptions about the percentage of households on private wells; and (3) population distributions are not evenly distributed within counties and thus their projections onto geologic units may not reflect local patterns of housing development. Nonetheless, we present an estimate of population exposed to As to compare to county based estimates [13] and other geology based estimates [12], and to provide an additional framework to help identify population with the potential of elevated risk of As exposure.

Based on the estimate that 2.4 million people drink from private wells in NC [43], and census information about well use projected at a county-scale, we calculated a population exposed to As concentrations greater than 10 μg/L within geologic units. We estimate that 1.8% (~42,500 people) may consume water from wells containing As concentrations greater than 10 μg/L (S1 Table). However, exposure risk is highly regional; within the Carolina Slate belt, we estimated 407,000 people using private well water, 5.3% of which are estimated to consume water with >10 μg/L As. This distribution results in >50% of the estimated population with wells containing water exceeding 10 μg/L As (22,300 of the total 42,500 people) being located in the Carolina Slate belt. However, geology within the Carolina Slate belt is also critical to understanding exposure and further concentrates As exposure within a few high-As metasedimentary geologic units. Of the 22 geologic units within the Carolina Slate belt, metamudstones comprise 20% of the land area (3365/15635 km^2), but contain an estimated 58% (13,026/22,254 people) of the population with wells with >10 μg/L. Thus, around 25% of statewide As exposure through private wells is associated with metamudstones, again highlighting strong geologic control on As exposure.

Within the Carolina Slate belt, where approximately 200,000 residents consume private well water live within the boundaries of geologic units with greater than 5% exceedance percentages (S1 Table), ~13% (27,600/198,000 people) use a well that tested about >10 μg/L. Furthermore, 53% (106,210/198,011 people) of the population in the Carolina Slate belt live within the footprint of eight geologic units related to Albemarle arc, where 20% of the ~100,000 people (21,400/106,200 people) are estimated to consume water from wells >10 μg/L. Here it must be emphasized that while we estimate that more than half of As exposure from NC private well water use is concentrated in the Carolina Slate belt, which has previously been identified as a region of concern for As occurrence [14, 21, 22, 24], nearly half of As exposure is dispersed around North Carolina in various geological units, and which may be more difficult to predict or locate with targeted testing.

An additional 33% (66,200/198,000 people) of the population live over four geologic units with suspected origins similar to the Albemarle arc (recycled volcanogenic material) with 6% exposed to As >10 μg/L. The most significant of these four geologic units (3,199/4,457 people) is the Cat Square terrane, which comprises both 70% of the area within those four geologic units and 70% of the population with wells above the EPA limit. The Cat Square terrane illustrates a geologic unit with only moderate exceedance percentages, but relatively large exposure population due to the large geographic area and population living on the geologic unit. It is also a region that has received less statewide attention than the known As-bearing geologic units of the Carolina Slate belt [25].

The estimated NC population using domestic wells containing water with As in exceedance 10 μg/L is approximately 1/3 of the value (~120,000 people) estimated by Ayotte et al. [12] We speculate that this difference may be related to the scale of the analyses, assumptions about the population using well water, and distribution of data used in the analysis. Namely, Ayotte et al. [12] estimates the number of private well users in NC to be 43% greater (3.3 vs. 2.3 million) than the estimated used in this paper. Additionally, the geospatial distribution of the data underpinning their analysis may also lead to differences in our estimations (cf. Fig 1 of Ayotte et al. [12]). Specially, a greater proportion of their data is from regions that typically have greater exceedance probabilities as compared to the NCWELL dataset (i.e., more samples from the Carolina Slate than from the Coastal Plain), which could contribute to a greater population estimate.

In contrast, our analysis provides a similar estimate to an earlier analysis using the NCWELL database [13] (~39,000 people). In our analysis, the median As exceedance percentages are weighted by geology and population to estimate exposed populations in specific counties, and thus our results differ from previous county-by-county estimates derived from the NCWELL database because of the more concentrated distribution of As wells containing water with As greater than 10 μg/L within specific geologic units (S2 Table). For example, when comparing the county-based estimates of people consuming water with greater than 10μg/L AS, the geologic-based estimates of the top 10 at-risk counties (S2 Table) vary by as much as 100%. The largest discrepancies are found in Union, Anson, and Stanly Counties (Fig 2B), previously identified as hot spots of As contamination [13, 14, 24] where there is a mix of underlying geologic units with varied As exceedance percentages. In counties such as Union, where almost 60% of the county is underlain by the Cid Formation, the population at risk is estimated 16% greater than the county-level estimate of Eaves et al. [13]. In Anson and Stanly Counties, where only 12% of the area is over the Cid Formation, the geologic-based estimates are on average 50% smaller. In the context of public health, the geologic-based estimates provide greater specificity in counties with highly localized As contamination.

4. Conclusions

Our analysis leverages a leverages a large private well chemistry database to [13, 15] highlight potential advantages of using the geologic history of a region to identify areas that may contain aquifers at risk of geogenic contamination, and thus populations at risk of exposure to drinking water with As concentrations that exceed 10 μg/L. Our results indicate the majority of the population at risk can be determined by identifying problematic volcanogenic geologic units (e.g., the Albemarle Arc); however, additional exposed populations may be determined by identifying related (metamorphosed or deposited) geologic units (such as the Dan River Basin or Cat Square terrane), as well as other units with similar (e.g., Eastern Slate Belt metamorphosed felsic metavolcanics and Blacksburg formation) or analogous (e.g., within the Grandfather Mountain Window) geologic origins. While the highest exceedances statewide are concentrated in a few volcanic and volcanogenic sedimentary rock units (e.g., Cid Formation mudstone), problematic geologic units are more dispersed around North Carolina than previously inferred using smaller arsenic data sets and at the terrane-belt scale. Within these geologic units, finer-scale factors such as the local flow system, depth, chemical weathering profiles, and/or human land use influence groundwater geochemistry such as pH, redox state, and competing ion concentrations may exert important secondary controls on natural contaminant concentrations [42]. However, the extent of interaction with geologic material impacts dissolved As concentrations, and thus pH may be valuable measurement for understanding and predicting geospatial distributions of As contamination.

Our results also suggest that analysis by geologic unit may result in a different estimate of the geographic distribution of exposed population compared to county-level analyses. Most notably, the analysis demonstrates differences in the exceedance percentage within counties as well as differences in the estimate of exposed populations at the county level. Without incorporating geology into analyses, counties with a relatively small proportion of people on private wells but a highly regionalized As problem are less likely to identify as at-risk when compared to the geologic-based estimates. However, challenges remain when trying to identify and predict As occurrences that are dispersed across a large population with a small exceedance percentage. Nearly one-half of NC private wells with water that tested As >10 μg/L occur in such a pattern. Where As occurs in a more concentrated pattern, by including the geologic controls in the assessment of the at-risk populations, public health officials may be able to better identify communities that may otherwise be unnoticed or overlooked by public outreach or intervention efforts.

Supporting information

S1 Fig. **S1A Fig**. Map showing geologic units within the Carolina Slate belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03. **S1B Fig**. Map showing geologic units within the Grandfather Mountain Window. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03. **S1C Fig**. Map showing geologic units within the Inner Piedmont. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1D Fig**. Map showing geologic units within the Dan River Basin. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1E Fig**. Map showing geologic units within the Wadesboro Subbasin. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1F Fig**. Map showing geologic units within the Durham-Sanford Subbasin. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1G Fig**. Map showing geologic units within the Charlotte belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1H Fig**. Map showing geologic units within the Kings Mountain belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1I Fig**. Map showing geologic units within the Milton belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1J Fig**. Map showing geologic units within the Eastern Slate belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1K Fig**. Map showing geologic units within the Coastal Plain. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1L Fig**. Map showing geologic units within the Sauratown Mountains Anticlinorium. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1M Fig**. Map showing geologic units within the Blue Ridge. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1N Fig**. Map showing geologic units within the Chauga belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1O Fig**. Map showing geologic units within the Raleigh belt. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03**. S1P Fig**. Map showing geologic units within the Murphy belt. County boundaries are shown to help orient the reader. Map and license information: https://www.

nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03. **S1Q Fig**. Map showing geologic units within the Smith River Allochthon. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03. **S1R Fig**. Map showing geologic units within the Davie Basin. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03. **S1S Fig**. Map showing geologic units within the Hot Springs Window. County boundaries are shown to help orient the reader. Map and license information: https://www.nconemap.gov/datasets/ncdenr::bedrock-geology-1/explore?location=35.144009%2C-79.920510%2C8.03.
(PDF)

S1 Table. Exceedance percentages and estimates of population with wells that contain with concentrations greater than 10 μg/L in geologic units. A geocode is an abbreviation used to denote a map area associated with a rock type on the geologic map.
(XLSX)

S2 Table. Geology based estimates of populations with private wells that contain water with As concentrations greater than 10 μg/L. County-based estimates are from Eaves et al. [13].
(XLSX)

Acknowledgments

We thank Matthew Polizzotto, Cass Miller, Hannah Peel, Sarah Yelton, and Megan Lane for valuable discussion.

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