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Arsenic Concentrations in Ground and Surface Waters across Arizona Including Native Lands

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Abstract: Parts of the Southwestern United States report arsenic levels in water resources that are above the United States Environmental Protection Agency's current drinking water limits. Prolonged exposure to arsenic through food and drinking water can contribute to significant health problems including cancer, developmental effects, cardiovascular disease, neurotoxicity, and diabetes. In order to understand exposure risks, water sampling and testing have been conducted throughout Arizona. This information is available to the public through often non-overlapping databases that are difficult to access and in impracticable formats. The current study utilized a systemic compilation of online databases to compile a spreadsheet containing over 33,000 water samples. The reported arsenic concentrations from these databases were collected from 1990-2017. Using ArcGIS software, these data were converted into a map shapefile and overlaid onto a map of Arizona. This visual representation shows that arsenic levels in surface and ground water exceed the United States Environmental Protection Agency's drinking water limits for many sites in several counties in Arizona, and there is an underrepresentation of sampling in several tribal jurisdictions. This information is useful for water managers and private well owners throughout the State for determining safe drinking water sources and limiting exposure to arsenic.

Keywords: *arsenic, groundwater, surface water, Arizona, monitoring map, water quality*

Arsenic contamination represents a growing public health concern in numerous countries across the globe (Mukherjee et al. 2006; Uddin and Huda 2011; Alarcón-Herrera et al. 2013; Huang et al. 2015; Ayotte et al. 2017; Hsu et al. 2017; Malloch et al. 2017; Saint-Jacques et al. 2018; Zeng et al. 2018). It has been responsible for some of the most devastating natural mass poisoning incidents in recent times, according to the World Health Organization (WHO) (Flanagan et al. 2012), and represents a looming threat as concerns about water security and water shortages increase (IPCC 2013, 2014). Its potency for damage to health prompted the WHO in 1999 to lower maximum contaminant levels (MCL) from 50 μg/L or parts per billion (ppb) to 10 ppb and recommend emergency corrective measures be taken in waters that exceed 50 ppb (Smith et al. 2000). Following this policy change, most governments adopted similar regulations globally (Shankar et al. 2014; Nigra et al. 2017).

Arsenic exists in two common forms, organic and inorganic, and this characteristic determines its toxicological potential (Dani and Walter 2018). Organic forms of arsenic, such as monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA), are commonly found in aquatic fish and other consumable sea products and are generally viewed as relatively non-toxic (Husain et al. 2017), although this view is under debate (Moe et al. 2016; Wei et al. 2017). This non-toxic designation is attributed to the fact that MMA and DMA are both metabolic products produced by the liver during natural arsenic metabolism, and ingestion of these products typically results in normal excretion by the organism (Vahter and Concha 2001). Inorganic arsenic, in relation to contamination and toxic health effects, is broken into its pentavalent, arsenate, and trivalent, arsenite, forms (Thomas et al. 2001). Both arsenate and arsenite contaminate ground and surface drinking water sources and can be taken up by plants, such as rice (Hughes et al. 2011; Chung et al. 2014). Inorganic arsenic can be metabolized by organisms; however, it bioaccumulates in various organs such as the liver, kidneys, heart, and lungs causing progressive damage with chronic exposure (Arslan et al. 2016). Arsenic is recognized as a potent carcinogen and is associated with vascular damage, which can lead to congestive heart failure (Martinez et al. 2011; Moon et al. 2012). Additionally, studies have demonstrated that arsenic acts as a potentiating agent with other toxins exacerbating their detrimental effects (Singh et al. 2011; Tyler and Allan 2014).

While arsenic's previous use in industry poses a possible legacy contaminant in parts of the globe, its application has been diminishing and its natural occurrence represents the primary environmental source to contaminate air, food, and water resources (Ribeiro et al. 2000; Mandal and Suzuki 2002). For example, tube wells drilled in Bangladesh in the 1960s and 1970s by the United Nation's Children Fund (UNICEF) went deeper than previously drilled wells to access cleaner water sources not contaminated by microbial organisms (Sen and Biswas 2013); deep wells have provided arsenic laden water to parts of Vietnam for more than a hundred years (Winkel et al. 2011). The aquifers in these deeper wells have a different surrounding geologic structure, and the mineralite matrix was associated with heavy arsenic concentrations, which led to subsequent contamination of these new wells (Hoque et al. 2017; Rahman et al. 2018). A report by the United States Geological Survey (USGS) in the early 2000s revealed similar geologic conditions which could lead to contamination of water by naturally occurring arsenic across many parts of the United States (Welch et al. 2000). A combination of rich iron-sulfur bearing rocks, agricultural backgrounds, and extensive mining history increases the potential for arsenic contamination in the Southwestern United States. Mining operations in the Southwest result in increased risk of contamination by disturbing underlying bedrock and iron-sulfur rocks; as ore is brought to the surface, this increases the surface area of these rocks, which may increase arsenic mobilization into the environment (Focazio et al. 2000; Etschmann et al. 2017). Waste and tailing piles represent an added source for contamination, increasing the potential for concentration of arsenic contaminants (Lim et al. 2009; Larios et al. 2012; Laird et al. 2014).

Because of water treatment to meet drinking water standards, arsenic in Arizona does not pose a significant concern for urban centers; however, much of the state is designated as rural or frontier regions (Gordon 1987; U.S. Census Bureau 2010). Citizens in these regions still rely on private wells or water hauling practices, which are unregulated and unmonitored, and are vulnerable to unchecked contamination by arsenic. Both the Verde Valley and the Hopi Tribe have faced litigation and legal ramifications for exceedances in their water from arsenic (Foust et al. 2004; Wildeman 2016). For these reasons, numerous separate studies and databases are publicly searchable, and show arsenic levels across the state (see references- National Water Quality Monitoring Council (NWQMC); U.S. Environmental Protection Agency (USEPA)). These databases are now combined within the NWQMC site, and the USEPA site is no longer available. However, a recent study investigating regional water quality on the Navajo Nation in Arizona demonstrates that the use of these databases, combined with new sampling, can provide information regarding water quality such as arsenic concentrations above the regulatory drinking water limit across a landscape (Hoover et al. 2017, 2018; Jones et al. 2020). The Navajo Nation represents the largest contiguous Native American reservation in the United States, has an extensive history of environmental injustice and environmental contamination issues associated with uranium mining, and is primarily rural or frontier in designation (Lewis et al. 2017). The purpose of this paper is to combine water quality and arsenic concentration data on the water in

Arizona from various databases and scientists into a single location. This information is important for identifying which populations and communities are at risk of consuming arsenic contaminated drinking water, especially for those reliant on unregulated sources.

Methods

Retrieval of Datasets

Datasets were collected through the methods summarized in Figure 1. Data were downloaded in May 2017 from https://ofmpub.epa.gov/storpubl/dw_pages.querycriteria, which is no longer available through the USEPA online sites, and https://www.waterqualitydata.us/portal/, where the USEPA data may have now migrated. Search criteria for the geographic location was entered as Arizona and then more specifically broken down by county and or tribal lands. Data from all Organization Types and Station Types were used. The Date Range was set as January 1, 1990-May 1, 2017. Water was chosen as the "Activity Medium" and all "Intents and Communities" were used. "Arsenic" and all synonyms were chosen as the "Characteristic" and all "Warehouse Data Sources" were used. Once the results were generated, they were downloaded and converted into Microsoft Excel files.

The location information and corresponding arsenic levels had to be combined into a single file for results downloaded from https://www.waterqualitydata.us/portal/. To do this conversion, the Monitoring Location Identifier was matched on each of the documents. A copy of the Results file was saved in order to avoid corrupting the original information. The sample type (groundwater or surface water), latitude, and longitude were then added to the copy of the Results file for each individual county. The date of the sample was also changed to a recognizable date format using the formula =DATAVALUE.

Condensation by County

After combining the Water Quality Database site location and arsenic level information into single files for each county, all of the information for each county from both websites was merged into a single Excel document that was used to create the shapefiles in ArcGIS. In order to ensure quality was maintained, the background color of six to ten line items was changed per county. Random spot checking was also conducted by comparing the merged file to the originally downloaded data.

Organization and Formatting

When the county samples were merged into a single Excel document, the website containing the original information was included for each sample site. Additionally, the location information for each sample was included, specifically the county where the sample was taken, latitude, longitude, and whether the sample was taken from groundwater, surface water, or unspecified. The date the sample was taken, the original numerical value of the concentration of arsenic, the original units, and the converted value and units were also included. Each sample was converted to ppb. After all of this information was entered, the file was saved in comma-separated value (CSV, comma delimited) format.

Conversion into Shapefiles

ArcGIS version 10.4 software was used to make the shapefiles. The CSV file was converted into an XY table with Longitude as X and Latitude as Y (this information comes from ESRI technical support: How to import XY data tables). The XY table is then uploaded as a shapefile to the map of Arizona. The map of Arizona was loaded from the ESRI online database and tribal reservations were delineated (Figure 2; CAPGISadmin 2017; Central_Arizona_Project 2019; Esri 2020; MPD_GIS 2020). Once loaded as a shapefile, it was converted to a layer. The symbols were identified by quantity and broken up into the following three categories based on concentration in ppb: 0.0-10.0, 10.1-100.0, and greater than 100.0. The color became darker and the size larger as the value increased.

Upon completion of the map of the entire state of Arizona, there were 33,099 samples represented. The previously listed information (see above) per sample is viewable in the attribute table for the newly created layer in ArcGIS (http://www.arcgis.com/home/item.html?id=191c7abbce0445409a190522ccb3db2c).

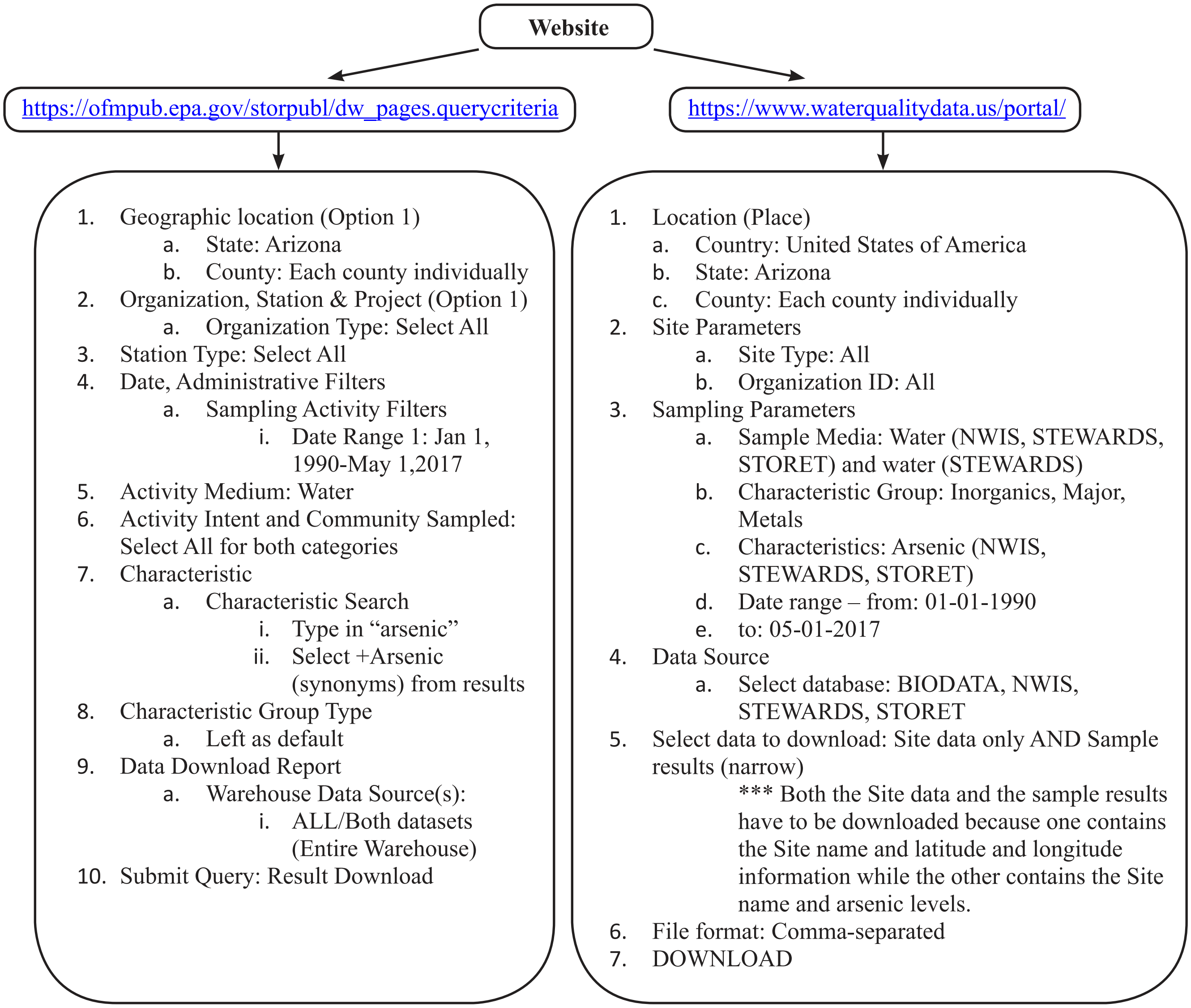

Figure 1. Flowchart demonstrating the procedures used for querying each website for downloading data regarding arsenic measurements in ground and surface water in Arizona.

Data Analysis and Reporting

In order to determine the number of samples per county with a measured arsenic concentration over 10.0 ppb, the 33,099 samples were separated by county and broken up further by concentration level. The number of samples with an arsenic concentration of 10.1 ppb or above was divided by the total number of samples reported for that county in order to get the percentage of reported samples over 10 ppb using the formula below where *P%* is percentage, *X* the portion of total samples in the concentration category, and *Y* the total number of samples in the category (equation 1).

$$\frac{P\%}{100} = \frac{X}{Y} \tag{1}$$

Similar analysis was also conducted for arsenic levels on tribal lands. These analyses are reported in further detail in the Results section.

Results

The compilation of data from 33,099 ground and surface water samples provides a clear picture of the extent of known arsenic levels throughout the state of Arizona (Figures 3-6; surface water, groundwater, and sites where the water source was not specified, respectively). Sixty-four %

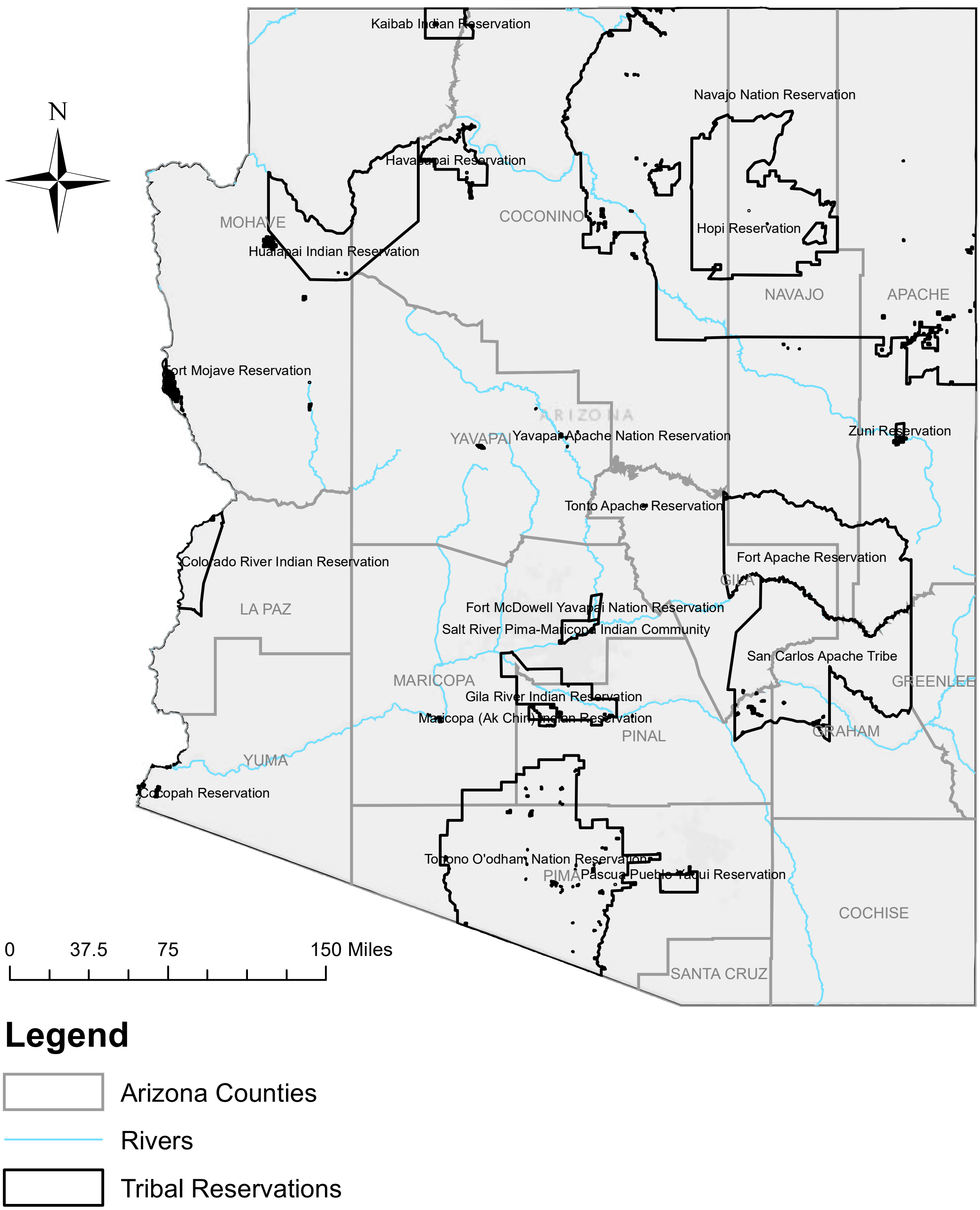

Figure 2. Map of Arizona with tribal names and jurisdictions outlined.

(21,194 samples) of the total samples taken did not specify where they were from, while 6.4% were from groundwater and 29.6% were from surface water. Many samples came from repeated site sampling at the same location over the course of the timeframe evaluated; therefore, the number of sites represented on the map appears to be fewer than the total sampled, especially for the surface water sites. For the sites where the type of sample was “unspecified,” the information may have been recorded when the sample was taken, but has not been made publicly available. For several of these unspecified sites, samples were taken from areas with little or no surface water, so it may be possible to assume the sites were sampled from groundwater resources. Any area with no indication of arsenic sampling on the maps is a result of there being no data indicated in the searched databases for GIS coordinates in that region. Regions where there are no shapes on the maps either have not been sampled or samples were not provided to the queried databases suggesting that further sampling may be useful especially for the evaluation of groundwater.

Across counties, many ground and surface water samples demonstrated arsenic levels above the regulatory safe drinking water limit of 10 ppb as put forth by the USEPA. The results indicate that 20.7% of all the samples taken throughout the state measured over 10 ppb for arsenic in the water (Table 1). More than 40% of samples from Pinal and Yavapai Counties have arsenic concentrations over 10 ppb (Table 1). The county with the overall lowest concentrations is Greenlee. Several of the tribal jurisdictions also had samples that exceeded 10.0 ppb, especially Fort McDowell. For the most part, sampling on tribal lands is limited and in some cases is either non-existent or unavailable through the searched public databases (Table 2).

Discussion

The geologic profile and climate of Arizona lend itself to naturally occurring valuable mineral deposits and fertile agricultural lands. Unfortunately, both of these factors plus the large and primarily rural nature of Arizona contribute to issues securing clean water resources that are safe for human consumption (Cordy et al. 2000). The arsenic water quality information represented in the USEPA and National Water Quality Monitoring Council databases demonstrated water resources that exceeded the USEPA MCL of 10 ppb were widely distributed across the state, with most exceedances located in the central and southern regions (Figures 3-6). The frequency of contaminated wells in Arizona at 20.7% exceeded the national average of 12%; however, several states including Illinois, Maine, Minnesota, and Nevada have a high percentage of exceedances similar to the levels found in Arizona (Uhlman et al. 2009; Ayotte et al. 2011, 2017). The USGS points to the geologic substrate across Arizona as the explanation for an elevated background concentration of arsenic in water resources, which explains the statewide contamination (Ryker 2001). Areas that have experienced previous mining or significant ground disturbances, including deep water exploration, demonstrate clusters of elevated arsenic concentrations exceeding drinking water regulatory limits, such as those seen in Yavapai and Pinal Counties (Anning et al. 2012).

All of the maps demonstrate a lack of information regarding sampling of arsenic levels in water resources within most tribal jurisdictions. This lack of representation in these databases does not demonstrate an absence of arsenic in water resources across these regions, but rather an absence of either sampling by federal agencies and/or a lack of centralized information being publicly available. These Native American Nations are sovereign entities recognized and separate from the federal government that maintain their own utility services, including water monitoring programs (U.S. Department of the Interior 2006; Washington and van Hover 2011). The most sampling has occurred on the Navajo and Hopi lands, where water quality issues, especially related to widespread arsenic and uranium contamination occur (Brugge and Gobble 2002; Hoover et al. 2017, 2018). Unfortunately, while water quality information exists for some of these Nations (TerraSpectra Geomatics et al. 2000; Orescanin et al. 2011; Hoover et al. 2017, 2018), for many, if the data exist, it requires strict approval by the various tribal governments to publish them in a public location (Kickingbird and Rhoades 2000). An added barrier to such publication is the

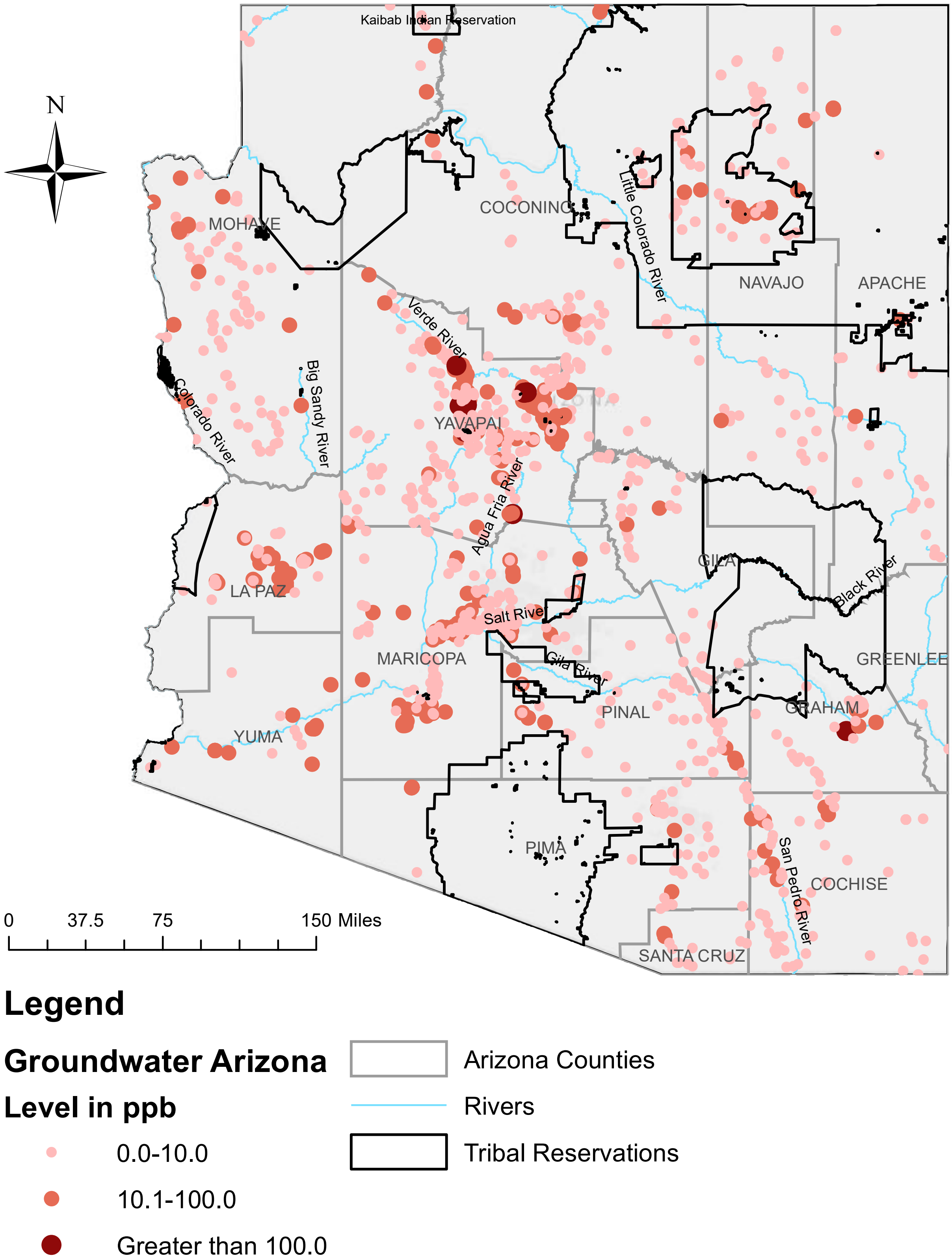

Figure 3. Map of Arizona representing arsenic levels in ppb for samples taken from groundwater resources between 1990 and 2017. Increasing circle size indicates higher arsenic concentrations.

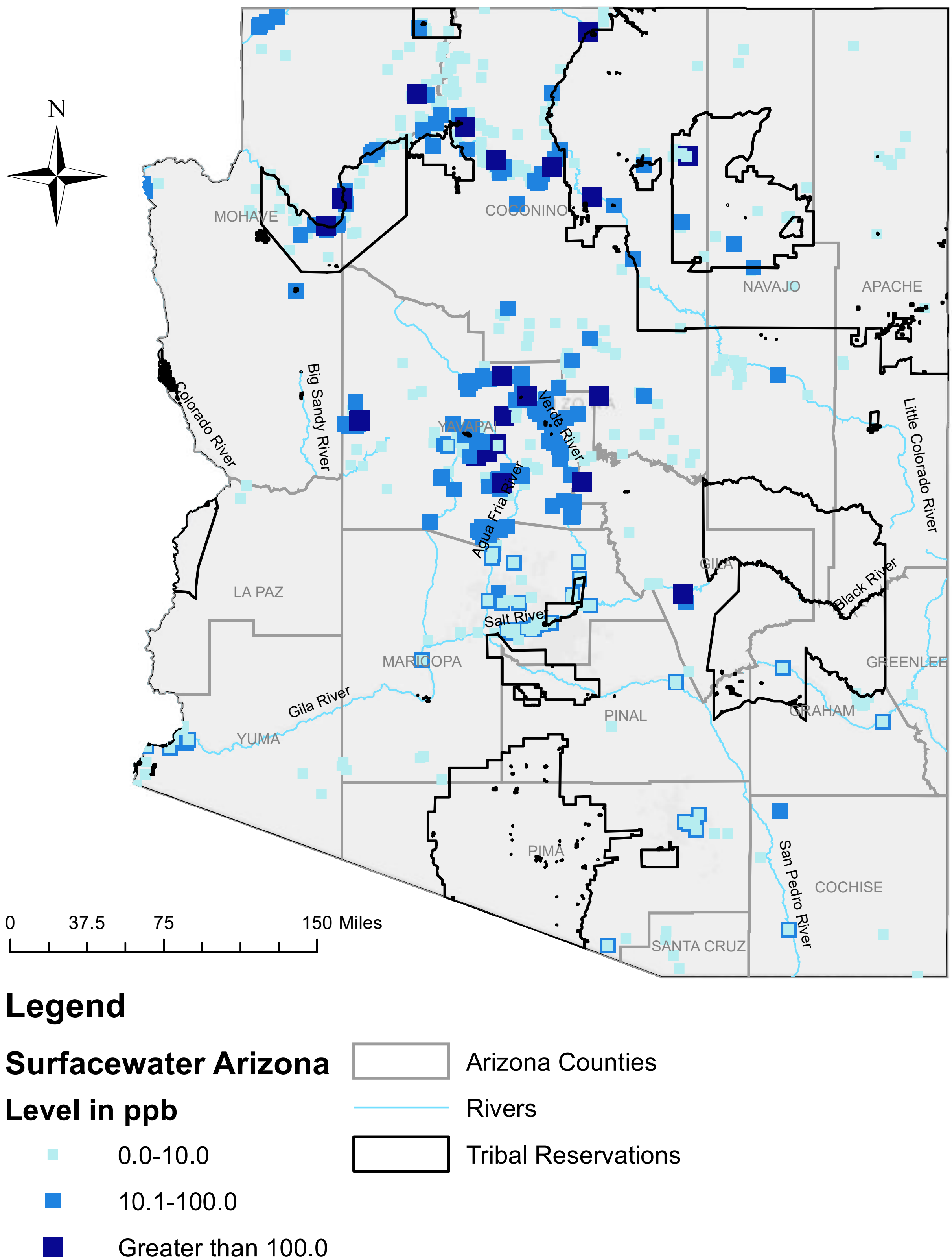

Figure 4. Map of Arizona representing arsenic levels in ppb for samples taken from surface water resources between 1990 and 2017. Increasing square size indicates higher arsenic concentrations.

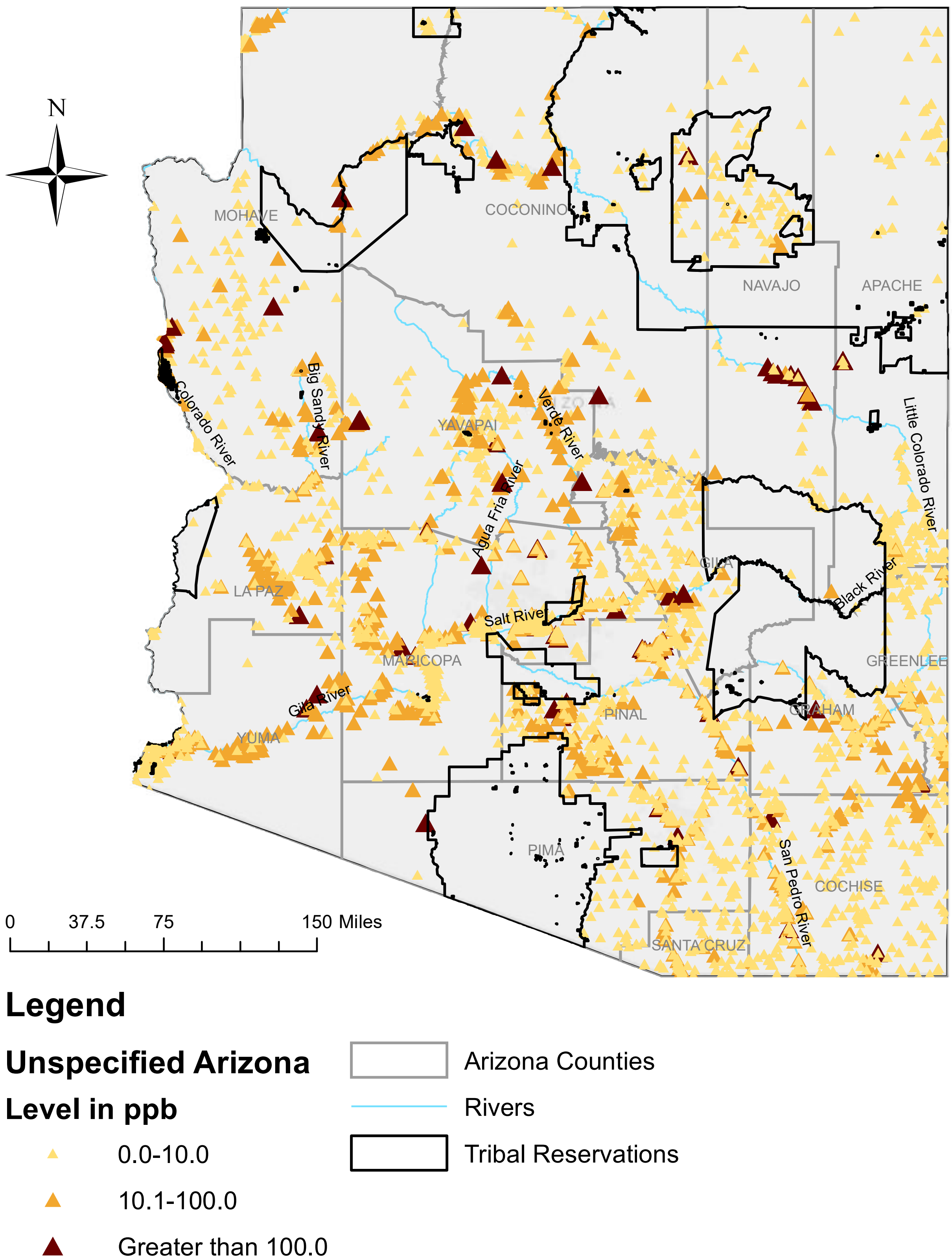

Figure 5. Map of Arizona representing arsenic levels in ppb for samples that did not have the type of water resource provided and were taken between 1990 and 2017. Increasing triangle size indicates higher arsenic concentrations.

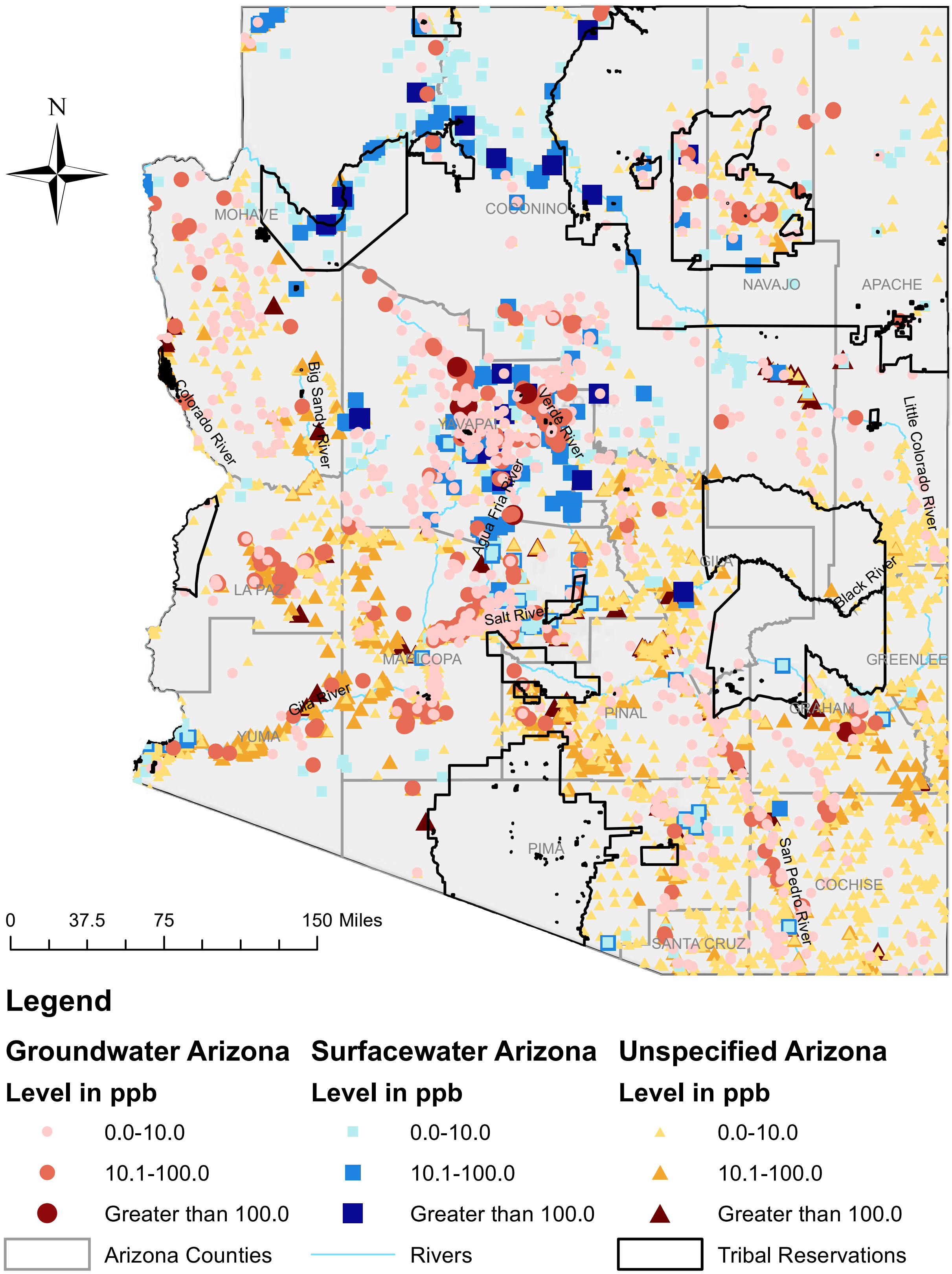

Figure 6. Map of Arizona representing arsenic levels in ppb for all samples taken between 1990 and 2017. Marked places on the map are the same as for Figures 3-5.

Table 1. The distribution of samples across concentration ranges of arsenic and the % of samples that are above the USEPA drinking water limit (10.0 ppb) for each Arizona County. Percentages highlighted in red represent those counties with the highest % of samples above USEPA drinking water limits.

	Apache	Cochise	Coconino	Gila	Graham	Greenlee	La Paz	Maricopa	Mohave	Navajo	Pima	Pinal	Santa Cruz	Yavapai	Yuma	Total for Arizona
Square Miles	11,218	6,219	18,661	4796	4641	1848	4513	9224	13,470	9959	9189	5374	1238	8128	5519	113,997
Total Number of Samples	1407	1511	3712	2870	1162	1164	794	6774	1622	782	1411	1801	720	5705	1664	33,099
# of Samples 0.0-5.0 ppb	169	235	1618	555	559	114	207	1246	437	259	230	182	66	463	883	7,223
# of Samples 5.1-10.0 ppb	25	43	151	286	159	16	278	1954	233	49	312	240	103	432	304	4,585
# of Samples 10.1-50.0 ppb	20	78	267	226	90	33	137	1393	339	74	78	718	41	2227	183	5,904
# of Samples 50.1-100.0 ppb	11	17	38	31	7	1	9	113	20	26	50	92	3	127	3	548
# of Samples > 100.0 ppb	32	4	26	52	3	0	2	26	22	36	3	56	0	136	4	402
% of Samples > 10.0 ppb	**4.5**	**6.6**	**8.9**	**10.8**	**8.6**	**2.9**	**18.6**	**22.6**	**23.5**	**17.4**	**9.3**	**48.1**	**6.1**	**43.6**	**11.4**	**20.7**

Table 2. The distribution of samples across concentration ranges of arsenic and the total number of samples for each Arizona tribal jurisdiction.

	Cocopah Reservation	Colorado River Reservation	Fort Apache Nation Reservation	Fort McDowell Yavapai Nation Reservation	Fort Mojave Reservation	Fort Yuma Indian Reservation	Gila River Indian Reservation	Havasupai Indian Reservation	Hopi Reservation	Hualapai Indian Reservation	Kaibab Indian Reservation
Square Kilometers	26.1	1202.1	6814.8	100.9	136.6	182.1	1514.4	714.4	6560.8	4155.9	491.5
# of Samples 0.0-10.0 ppb	4	1	12	71	12	0	75	4	284	75	35
# of Samples 10.1-100.0 ppb	2	0	1	86	2	0	3	2	55	61	2
# of Samples > 100.0 ppb	0	0	0	0	0	0	0	0	2	11	0
Total Number of Samples	**6**	**1**	**13**	**157**	**14**	**0**	**78**	**6**	**341**	**147**	**37**

	Maricopa Ak Chin Indian Reservation	Navajo Nation Reservation	Pascua Pueblo Yaqui Reservation	Salt River Pima Maricopa Reservation	San Carlos Apache Reservation	Tohono O'odhan Nation Reservation	Tonto Apache Reservation	Yavapai-Apache Nation Reservation	Yavapai-Prescott Reservation	Zuni Reservation
Square Kilometers	84.9	62563.9	5.7	221.2	7580.7	11535.4	0.3	2.6	5.7	1879.9
# of Samples 0.0-10.0 ppb	33	1246	0	35	132	1	0	0	69	0
# of Samples 10.1-100.0 ppb	1	38	0	20	19	0	0	4	11	0
# of Samples > 100.0 ppb	0	1	0	0	0	0	0	0	1	0
Total Number of Samples	**34**	**1285**	**0**	**55**	**151**	**1**	**0**	**4**	**81**	**0**

hesitation of some tribal governments to associate a location identification with any specific problem, which may lead to social or local stigmatization for continued monitoring (Sharp and Foster 2002; Manson et al. 2004). Last, resources for testing may be limited.

The information presented in this study strictly focused on the extent of arsenic contamination in water supplies across Arizona from readily available databases. The lack of information regarding water resource characteristics, sampling practices, and other hydrogeochemical information were not presented and therefore do not allow comment on how these factors could influence arsenic contamination or mobilization. Though these limitations do not paint a complete hydrogeochemical profile of water in Arizona, they provide a collected map that displays arsenic contamination and details counties where arsenic is prevalent. The map additionally demonstrates whether water resources are likely to be contained in surface or groundwater, which allows regulatory and governmental agencies to take steps to locate and possibly mitigate input into these resources.

As Arizona, and much of the Southwestern U.S., prepares for another year of limited precipitation and drought conditions, the question of clean and safe consumable water is important. A recent study from Sonora, Mexico demonstrates a clear link between arsenic levels in wells used for drinking water, urinary arsenic levels in children, and hazard risk for negative health outcomes (García-Rico et al. 2019). Such studies demonstrate the importance of understanding the potential risk of arsenic exposure especially for those populations who may not always have access to municipal water resources.

Spurred by worsening drought conditions, in 2017 the Governor of Arizona authorized the Arizona Department of Water Resources to conduct studies that detail the extent of water security and purity (MacEachern 2017). Tribal governments, such as the Navajo Nation, have already adopted similar policies and contingency plans to address this growing concern (Navajo Nation Department of Water Resources 2003). Decreased precipitation and snowmelt recharge in combination with increased water consumption from growing population centers and resource extraction represent a significant stressor to water resources (Maupin et al. 2014; Eden et al. 2015). These factors could act to concentrate arsenic as the amount of water in these systems drop and present another looming concern for public health and safety. The collective maps in this study provide another resource for legislators, regulators, and community members to face the challenge of providing safe drinking water to Arizona and limit public health risks.

Conclusions

This study demonstrates that many ground and surface water resources in Arizona have levels of arsenic above the current drinking water limits. The data also demonstrate a lack of data available for many of the Native American jurisdictions throughout the state. Many populations in rural areas throughout the state rely on well water and do not have access to the water treatment available to municipal customers. These maps may provide information for local water resource managers to evaluate both the need for more arsenic sampling and for providing information to water users regarding their water quality. This need is especially important for many of the tribal regions throughout Arizona.

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Dr. Catherine R. Propper (corresponding author) has been a Professor of Biological Sciences at Northern Arizona University (NAU) since 1991 where she has been dedicated to supporting students from underrepresented backgrounds. She is the Program Director for the National Institutes of Health RISE for Native American Students and the Minority Health International Research Training program, Native Americans Exploring Global Health Disparities. Dr. Propper is co-Lead for the Southwest Health Equities Research Collaborative's Research Infrastructure Core. Her research focuses on how environmental contaminants affect development, reproduction and behavior, and she has published more than 60 peer-reviewed journal articles. Locally and statewide, she has served on the City of Flagstaff's Contaminants of Emerging Concern Panel and the Arizona Department of Environmental Quality's Advisory Panel on Emerging Contaminants. Nationally, she has participated in several U.S. Environmental Protection Agency Scientific Advisory Panels. She may be contacted at Catherine.Propper@nau.edu or via mail at 617 S. Beaver Street, Box 5640, Northern Arizona University, Flagstaff, AZ 86011.

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