

Article

Arsenic in Groundwater Sources from Selected Communities Surrounding Taal Volcano, Philippines: An Exploratory Study

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Abstract: Arsenic (As) is a highly toxic, carcinogenic trace metal that can potentially contaminate groundwater sources in volcanic regions. This study provides the first comparative documentation of As concentrations in groundwater in a volcano-sedimentary region in the Philippines. Matched, repeated As measurements and physico-chemical analyses were performed in 26 individual wells from 11 municipalities and city in Batangas province from July 2020 to November 2021. Using the electrothermal atomic absorption spectrometric method, analysis of the wells revealed that in 2020, 23 out of 26 (88.46%) had As levels above the WHO limit of >10 ppb while 20 out of 26 wells (76.92%) had persistently high As levels a year later. Using a Wilcoxon signed-rank test, levels of As were found to be statistically elevated compared to the national safe limit of 10 ppb in the 26 matched sampling sites in both 2020 (p -value < 0.001) and 2021 (p -value = 0.013). Additionally, a two-paired Wilcoxon signed-rank test revealed that As levels were statistically higher in 2020 than in 2021 (p -value = 0.003), suggesting that As levels may be higher in years when there is more volcanic activity; however, this remains to be further elucidated with suitable longitudinal data, as this study is still in its preliminary stages. The data was also analyzed using a bivariable regression, which showed no evidence of a significant relationship between As levels and distance from the danger zone (Taal volcano crater); however, results showed an inverse but statistically insignificant relationship between As levels and elevation. Due to the toxic profile and persistence of As in groundwater in Batangas Province, continuous groundwater As monitoring, timely public health risk communication, and the provision of alternative water sources to affected populations are recommended.

Keywords: Taal volcano; arsenic; groundwater; aquifer



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1. Introduction

Arsenic (As) is a naturally-occurring trace element that is well distributed in the earth's crust, and as such, may be present in the soil, water, air, and even in living organisms at variable concentrations [1–3]. Humans may be exposed to high levels of As through several pathways: through contaminated air, food, water, and soil. However, drinking water from contaminated groundwater sources is the most common cause for As poisoning and is therefore a cause of greater public health concern compared to other exposure pathways [4].

The acceptable limit of As in drinking water is established as 0.010 mg/L (10 ppb) [5]. As-contaminated water, even at high concentrations, shows no apparent change in taste, odor, or appearance and may go undetected without chemical tests and analysis [4]. This poses a major public health concern, as the failure to conduct routine screening or extensive monitoring of groundwater sources in regions at risk for high levels of As can lead to long-term consumption, unbeknownst to the population [6]. Chronic exposure to the metalloid, for months or even years, is needed to build up in the body before symptoms manifest. Health impacts may vary amongst individuals depending on the amount ingested, the

duration of exposure, immunity, and genetics, but manifestations may show signs of acute or chronic toxicity, ranging from the development of dark spots on the skin (arsenicosis) from as early as six months, and over time, to cancer in the skin and other internal organs secondary to prolonged exposure [1,3,7].

Cases of As poisoning from contaminated groundwater sources are widely reported worldwide, including in the Philippines [1,2,8]. One of the largest incidences of As poisoning from contaminated groundwater was documented in Bangladesh [9], affecting more than 70 million people. In the Philippines, an index case of a patient admitted for chronic As exposure in Lubao, Pampanga in 2014 prompted immediate water quality assessments, revealing groundwater samples containing As levels of as much as 300 ppb due to both industrial and geologic contaminants [10]. Data validation from five barangays in the same city showed 215 cases of suspected arsenicosis documented from 2010 to 2014, and further interviews revealed 69 respiratory, 47 neurological, and 98 dermatologic incidences of symptoms in 123 interviewed residents from the same and neighboring municipalities [10]. Similar case studies of dermatologic symptoms and squamous cell carcinoma due to As toxicity were also reported in the Southern Luzon and Southern Mindanao regions of the country [11–13].

As contamination of groundwater sources is the result of a complex combination of natural geochemical processes exacerbated by manmade pollution [3,7]. Water contamination may occur as a result of anthropogenic processes such as mining, improper waste disposal, pesticides, fertilizer use, industrial processes such as smelting, and the burning of fossil fuels, and their environmental impacts may be felt for decades [1,8,10]. In fact, a study in Hungary demonstrated that improperly disposed liquid waste can continue to accumulate and contaminate groundwater even several decades after the elimination of pollution sources [14]. It is, however, suggested that the extensive, regional extent of As in groundwater in arid to semi-arid countries such as the Philippines is likely to be largely accounted for by the mobilization of geological sources [7].

Volcanism, in particular, is postulated to be one of the major drivers of As mobilization into groundwater sources [9,15]. Toxic levels of As have been found in groundwater near volcanic regions all over the world, including Central Italy [16,17], wells fed by the Tahlab aquifer in Iran [18], groundwater from thermal springs all around Latin America [6], and the deep groundwater of a volcano-sedimentary region in north-central Mexico [19]. Deteriorating conditions in these waters, along with optimal pH, temperature, and solution compositions, favor the solubilization of geogenic As into water [1,8].

On the 12 January 2020, Taal Volcano, located in Southwest Luzon in the Philippines, erupted for the first time since 1977, greatly impacting the livelihood and health of the people in Batangas Province and nearby regions [20]. This index volcanic event, which was followed by continuous volcanic activity throughout 2020, prompted an investigation into the quality of groundwater sources in communities surrounding the volcano. To date, there are very few studies on levels of As in groundwater near volcanic areas in the Philippines. This study provides the first comparative documentation of As concentrations in groundwater in a volcano-sedimentary region in the Philippines and whether there is a significant change in As levels after a considerable amount of time. It also aims to investigate the relation of As levels with elevation and distance from Taal Volcano. Through the use of both quantitative and qualitative measures, this study contributes to the growing literature on the extent of known public health risks in volcanic regions in arid-to-semi-arid countries and provides information on water quality and safety in the Philippines.

2. Materials and Methods

2.1. Study Area

Batangas Province, located in Southwest Luzon in the Philippines, has a total land area of 3119.75 km² and is subdivided into six administrative districts. Its 30 municipalities and 4 cities are further subdivided into 1078 smaller political units called barangays. As

of 2020, the province has a reported population of 2,908,494 and a population density of 934 persons/km² [21].

Geographically, the province is a combination of plains and mountains, notably Taal Volcano, with an elevation of 600 m (2000 ft). Taal Volcano is one of the most active volcanoes on earth. It is surrounded by Taal lake and sits on the Macolod Corridor, an active fault system and volcanic area [22]. A magnetotelluric survey previously characterized an active large-scale hydrothermal system located beneath the volcano [23]. The main crater, with a diameter of 2 km, is filled with volcanic fluids and meteoric water from its hydrothermal system, as well as seawater from the South China Sea [22,24]. There is the presence of seawater, volcanic water, and Taal Lake water in the main crater lake and spring discharges in the province suggest a relationship or connection between the volcanic hydrothermal system, the lake, and the groundwater aquifer in Batangas Province [22,25,26].

Batangas province sits on a regional, deep, and confined groundwater aquifer; pockets of unconfined and shallow groundwater aquifers also abound inland and some even extend toward the province's coastal borders. The recharging of the aquifer largely relies on rainfall, which collects into watersheds and then percolates into the soil [27,28]. The provincial groundwater aquifer almost exclusively feeds the majority of the point sources of drinking water in the province (Figure 1). The top point sources include own-use faucets (54.9%), bottled water (15.4%), and shared-use faucets (8.86%)—all of which are supplied by local water districts and village waterworks (Level 3 water sources) that pump groundwater from the main provincial aquifer [21]. Recent groundwater assessments by the local government in 2019 also showed that point source (Level 1) deep (>25 m) and shallow wells (<25 m)—also fed by the provincial aquifer—likewise remain a significant source of drinking water for households, private business owners, and public establishments in Batangas province [27,29]. Though groundwater supply is currently sufficient for the demand, a continued reliance on groundwater extraction may prove to be unsustainable in the long run due to rising urban development and the destruction of watersheds [27].

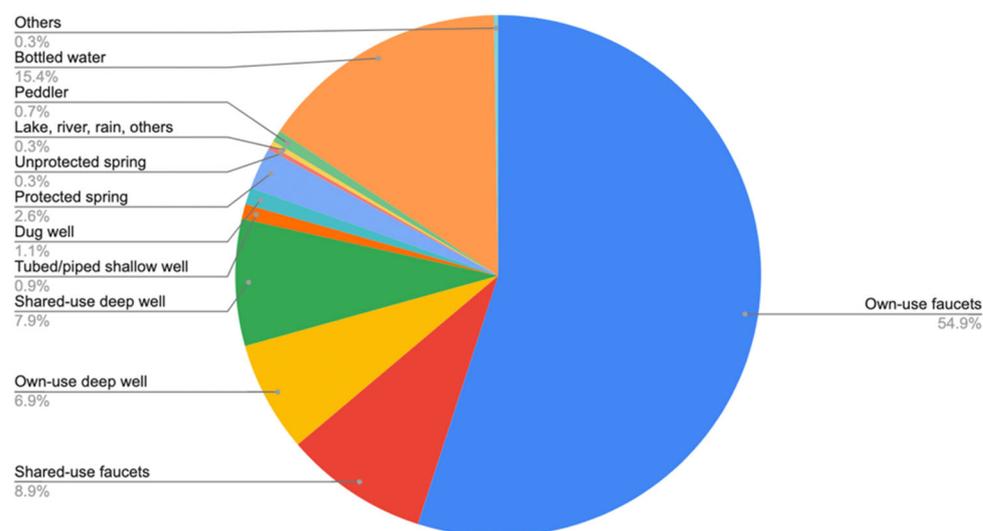


Figure 1. Point sources of drinking water supply in Batangas Province.

2.2. Profile of Sample Wells

In this study, 26 individual wells were sampled in 2020 and 2021. The majority (69.23%) of the wells were classified as Level 1 or point water sources, which includes deep wells ($n = 13$, 50%), shallow wells ($n = 4$, 15.38%), and one protected spring (3.85%). The rest of the samples ($n = 8$, 30.77%) were collected from wells classified as Level 3 waterworks systems, operated by municipal and barangay water districts, which supply multiple households. Limited interviews with residents living within 20 m of the wells included in the study showed that the households tend to use water from these sources for domestic purposes, i.e., for cooking, washing food and non-food items, bathing, laundry, and other domestic

functions. Fourteen wells (53.85%) were identified as sources of drinking water based on the limited interviews conducted. Several wells ($n = 6$, 23.08%) were in low-lying areas (<20 m above sea level) and the majority of the wells are rarely submerged in floodwaters during the rainy season.

2.3. Water Sampling Method

Water samples were extracted from both public and privately-owned handpumps and faucets, herein referred to as wells. Due to mobility restrictions brought on by the COVID-19 pandemic and upon the recommendation and approval of local leaders and stakeholders, convenience sampling was performed in order to identify the wells to be tested across 11 municipalities and 1 city in Batangas province within the 7 km, 10 km, and 14 km danger zones from the crater of Taal Volcano [30–33]. These danger zone classifications are used by the Philippine Institute for Volcanology and Seismology (PHIVOLCS) and the United Nations Office for the Coordination of Human Affairs (UN-OCHA) to guide risk management initiatives and emergency responses during volcanic eruptions. Among the 26 wells sampled, 5, 8, and 7 wells belonged to the 7 km, 10 km, and 14 km danger zones, respectively. The rest ($n = 6$) were found beyond 14 km from the Taal Volcano crater. The first set of measurements were conducted from July to August 2020, and repeated measurements were conducted in November 2021.

The operating standards of the Philippine National Standards for Drinking Water 2017 were followed during water sampling collection and analysis. Proper personal protective equipment (i.e., face masks and gloves) were used to prevent the contamination of samples. Prior to sampling, faucet taps, and pumps were visually inspected for rust, dirt, or other contaminants. The outlets were then turned on or pumped for 2–3 min to flush out service lines. Disinfection was then performed by flaming the solution of sodium hypochlorite that was applied to the faucet/outlet, which was then turned on for another 2–3 min. Afterward, water samples were collected in clean PET bottles, labeled accordingly, and placed in an icebox to preserve the samples for same-day transport and analysis at the National Reference Laboratory in Metro Manila. Approximately 10 mL of sample was passed through a 0.4 μm syringe filter and stored in 15 mL polypropylene centrifuge tubes for analysis.

The total levels of As in the water samples were measured via the electrothermal atomic absorption spectrometric method using the GTA120 Graphite Tube Atomizer from Agilent Technologies [34]. Analysis was achieved by dispensing a small volume of sample into a graphite tube, which was then heated until the sample was dried, charred, and atomized. The vapor produced absorbs the monochromatic radiation from a light source, and a photoelectric detector measures the intensity of transmitted radiation. Data from the graphite furnace AAS was analyzed using the SpectrAA software [34]. Both an initial calibration method and a continuous calibration method were performed throughout the analysis, while a quality assurance check at 50 ppb \pm 10% was performed every 15 samples. The method detection limit for total As was 0.9 ppb. The electrometric method was used to measure pH, the visual comparison method to measure color, and the Nephelometric method to measure turbidity.

The geographic coordinates of the sampling points were also recorded and then mapped using the R statistical package “ggmap”. Figure 2, below, shows the 26 matched, geocoded sampling sites in 2020 and 2021, classified according to the 7 km, 10 km, and 14 km danger zones set by the PHIVOLCS and the UN-OCHA. Most of the locations are located within these volcanic site danger zones, with a few of them being more distant. Estimated elevations ranged from 8.4–178 m above sea level, with a median elevation of 53.2 m.

were found to have As levels > 10 ppb, the highest being 46 ppb and from the same well in the same municipality. There were 20 wells in 2021 that had persistently high As levels above 10 ppb in between the two sampling periods (Figure 4). Two wells with previously elevated As levels in 2020 had levels below 10 ppb in 2021.

Table 1. Profile and As levels of study wells in selected municipalities surrounding Taal volcano, 2020 vs. 2021.

Code	Location of Sampled Well *	Dist. from Mt. Taal (In km)	Est. Elevation (In m)	Type of Source	Used for Drinking?	2020 As Levels (In ppb) **	2021 As Levels (In ppb) **
A	Banyaga, Agoncillo	5	38.4	Waterworks	Yes	4.7	1.2
B	Bilibinwang Agoncillo	5	60.7	Deep well	Yes	1.2	0.27
C	Buso-buso, Laurel	6	113.3	Shallow well	No	14	8.4
D	Bugaan East, Laurel	7	11.5	Shallow well	No	11	2.6
E	San Sebastian, Balete	7	23.9	Deep well	No	77	20
F	San Sebastian, Balete	8	23.9	Waterworks	Yes	20	46
G	Looc, Balete	10	34.9	Waterworks	Yes	27	16
H	Sampaloc, Talisay	10	38.5	Prot. spring	No	3.2	2.2
I	Abelo, San Nicolas	10	21.3	Waterworks	Yes	39	28
J	Poblacion, San Nicolas	10	17.9	Deep well	No	27	28
K	Abelo, San Nicolas	10	21.3	Deep well	No	44	33
L	Balukbaluk, Sn Nicolas	10	23.5	Waterworks	Yes	21	20
M	Balete, San Nicolas	11	23.8	Waterworks	Yes	16	13
N	Kinalaglagan, MnK	10	53.2	Shallow well	No	16	13
O	Lumang Lipa, MnK	11	160	Deep well	Yes	13	10
P	Poblacion W, Alitagtag	14	178.8	Deep well	No	16	11
Q	Burol, Sta. Teresita	13	84	Waterworks	Yes	21	15
R	Cahilan, Lemery	14	36.9	Deep well	Yes	12	12
S	Sinisian, Lemery	18	16.7	Deep well	No	12	16
T	Butong, Taal	18	10.6	Deep well	Yes	12	12
U	Butong, Taal	18	10.6	Deep well	No	43	44
V	Bolbok, Taal	27	122.5	Waterworks	Yes	14	11
W	Ambulong, Tanauan	12	8.6	Deep well	No	28	14
X	Sulpoc, Tanauan	16	267	Deep well	Yes	18	11
Y	Pagaspas, Tanauan	19	140.4	Deep well	Yes	15	15
Z	Balete, Tanuan	12	137.1	Deep well	No	10	6.6

* Exact locations of sampling sites were anonymized in compliance with the Philippine Data Privacy Act of 2012.

** Wells with As levels 10 ppb and higher are highlighted in red.

In terms of other physico-chemical parameters, water from the sample wells was found to be clear, colorless, and odorless overall. The median pH of groundwater samples was determined to be 7.48 (range: 7.04–7.99) in 2020 and 7.475 (range: 7.02–7.97) in 2021, thus slightly basic, though still within the national standard range of 6.5 to 8.5. At higher pH, the active adsorption or binding sites for As compounds, such as the minerals in the aquifer, tend to be occupied more by the hydroxyl (OH⁻) molecule from water, neutralizing these sites. This favors the mobilization of As oxyanions into the aqueous phase, increasing the concentration of As in groundwater [36]. Dry, arid conditions in the region may also further increase As concentrations (through evaporation and/or desorption due to the relatively high pH of groundwater sources found under these conditions). Lastly, the increasing compaction of clay layers into the aquifer, either by natural means or as a result of groundwater over-extraction, may also have caused As-rich porewater to be introduced to the aqueous layer [37,38].

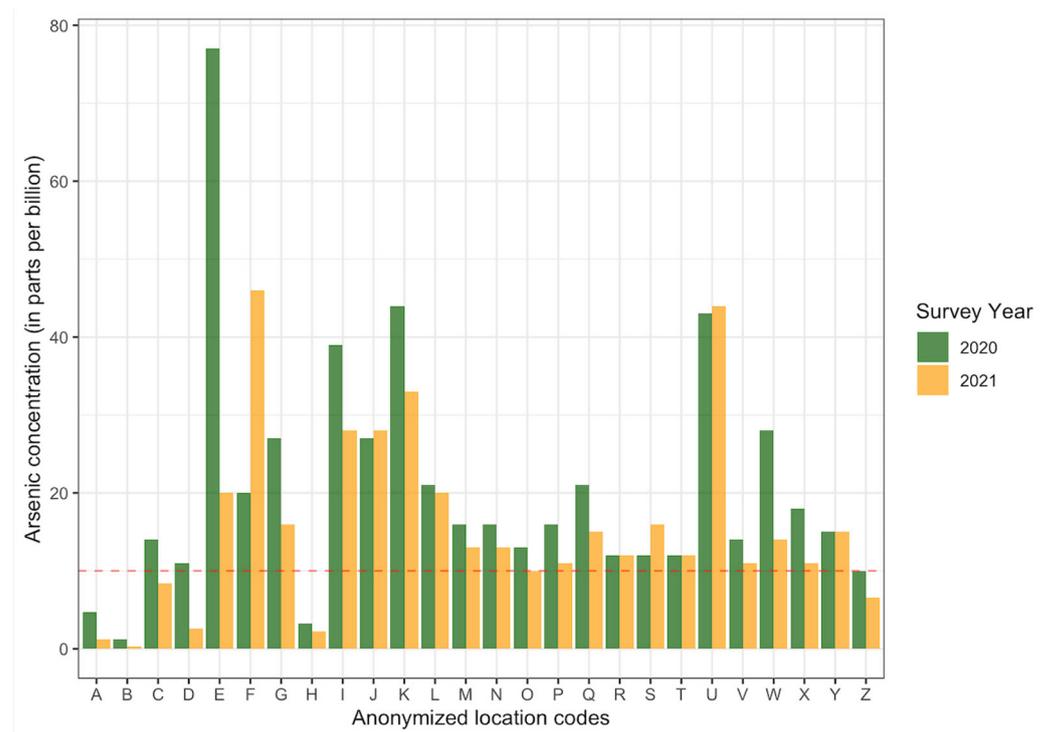


Figure 3. Comparative graph of As levels in study well surrounding Taal volcano, 2020 vs. 2021.

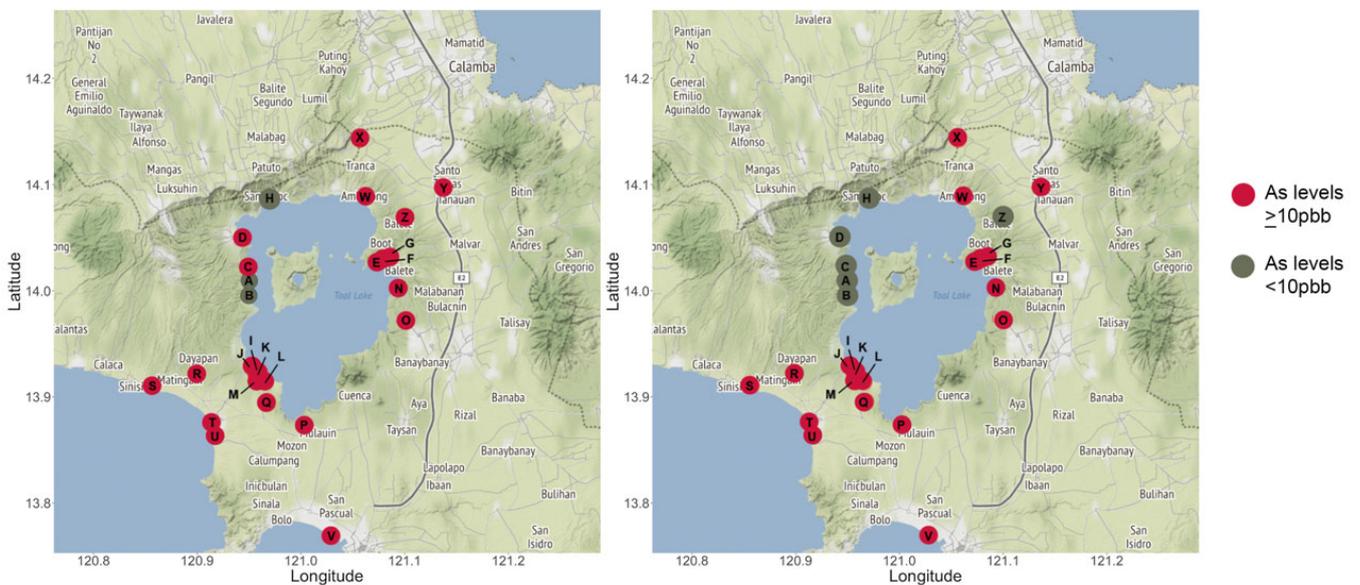


Figure 4. Geocoded map of study wells with persistently high As levels in 2020 and 2021.

3.2. Exploratory Analyses

In the succeeding exploratory analyses, we examined whether (1) current As levels (in either or both years) were significantly higher than the national and global As standard for drinking water (10 ppb), and whether (2) the distance (in km) as well as elevation (in m) of the wells in relation to Taal volcano is associated with increased As levels.

As shown in Table 2, the median value of As levels in 2020 was 16, whereas in 2021 it was 13 ppb. We observed a wider variability in As levels in 2021 (with an interquartile range (IQR) of 13.5) compared to in 2020 (IQR = 9.75). In 2020, As levels across the 26 locations were statistically higher (p -value < 0.001) than the 10 ppb limit set by the World Health Organization (WHO) and the Philippine National Standards for Drinking Water (PNSDW).

We also observed statistically higher As levels (compared to the 10 ppb limit) in 2021 (p -value = 0.013).

Table 2. Descriptive summary statistics for As levels in selected wells surrounding Taal volcano as compared to the national standard, and comparing 2020 vs. 2021.

	2020	2021
Median	16	13
Interquartile range (IQR)	13.5	9.75
One-sample Wilcoxon signed rank test	$p < 0.001$ *	$p = 0.013$ *
Paired, two-sample Wilcoxon signed rank test	$p = 0.003$ *	

* p -value < 0.05.

In the paired, two-sample Wilcoxon signed-rank test (Figure 5), we observed a statistically significant difference (p -value = 0.003) between the sampling years. In particular, we noted that the 2020 levels were statistically higher than the 2021 levels. The increased activity of the Taal volcano in 2020 potentially explains the significantly higher than usual As levels in the wells sampled that year. In 2020, PHIVOLCS reported one Taal volcano eruption in January that reached Alert Level 4 and increased seismic activity (1529 reported earthquakes, $n = 1529$) in Batangas province, 820 of which happened in January 2020 ($n = 820$, 53.63%). In comparison, a phreatomagmatic eruption occurred in July 2021, followed by multiple, smaller phreatomagmatic bursts over the following days, reaching Alert Level 3 [39]. Seismic activity in Batangas province was also reported to be significantly lower in 2021 ($n = 872$), with July having the most instances ($n = 170$, 19.50%). The statistics on seismic activity were aggregated from the PHIVOLCS Earthquake Information Database [40]. Ground movements, as a result of volcanic activity, may cause a greater interaction between groundwater and As-rich rocks and magma, causing the As to concentrate to increase. Previous studies have shown that As concentrations are highest where active hydrothermal circulation takes place [41]. High temperatures and lower pH favor mineral solubility during rock leaching and the mobilization of As through chemical processes after prolonged water-rock interactions at the reservoir [1,8,17,41]. However, this remains to be further elucidated with suitable longitudinal data, as this study is still in its preliminary stages.

We observed no evidence of increased As levels in proximal sampling sites in both years, as shown in Table 3. In a modeling study, Saunders et al. [42] argued that geochemical and geomicrobiologic processes working in concert may lead to the long-distance transportation of As, which may be partially related to the lack of a difference in proximal and distal As levels in the study. In Madrid, Spain, Gomez-Gonzalez et al. [43] also noted a long-distance dispersion of As, but this was mainly associated with mobile colloids, which is in contrast to several studies noting that proximate locations measure substantially higher As concentrations. A study in Central Italy [16] found that groundwater As levels were higher in areas with more deeply faulted zones. In the Dalsung Cu–W mine, in Korea, As in stream sediments sampled at various distances decreased with the distance from the mine [44]. Similarly, in Iran, Hajalilou et al. [45] observed decreasing As levels with increasing distances from the source.

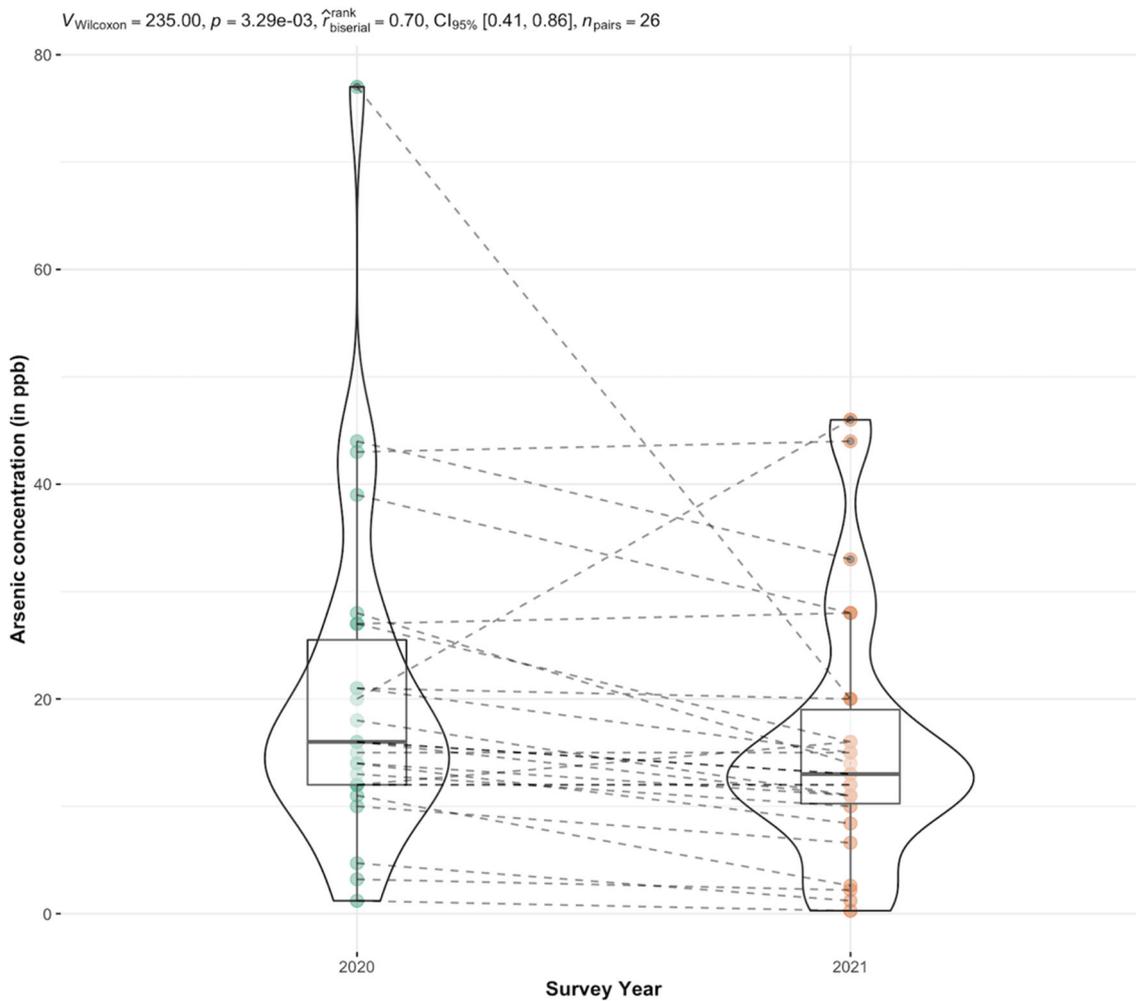


Figure 5. Graphical representation of the paired, two-sample Wilcoxon signed-rank test comparing As levels in selected wells, 2020 vs. 2021.

Table 3. Bivariable regression.

Sampling Year	Adjusted Covariates	Estimate * (95% CI)	p-Value
2020	distance	1.020 (0.959, 1.085)	0.537
	elevation	0.998 (0.993, 1.003)	0.500
2021	distance	1.066 (0.984, 1.154)	0.128
	elevation	0.998 (0.991, 1.004)	0.584

* The beta coefficient derived from the linear regression was back-transformed by exponentiating the beta coefficient. CI: Confidence Intervals.

Similarly, we did not observe any association between elevation (of water source) with As levels. This contrasts with studies noting either a positive or negative (correlational) association between water source depth and As levels. In California, Fujii and Swain [46] observed increasing groundwater As concentrations with an increase in well depth. Similarly, in Xinjiang Province, China, Wang and Huang [47] noted that As concentrations in artesian groundwater were found to increase with depth. In Bangladesh, however, a negative correlation (correlation coefficient (r) = -0.0999765) between aquifer depths and As concentrations was observed [48]. The current study’s results, albeit statistically not significant, indicate an inverse association of As level with elevation. Several plausible physio-chemical mechanisms may potentially be related to this directional association [49]. This is, however, beyond the scope of this study, and thus subsequent examination is warranted.

3.3. Limitations

The current exploratory study, which is still in its preliminary stages, has several limitations. First, only 26 sampling sites were continuously observed in both years. The limited sample size makes it to rule out substantial explanations for the observed patterns in the measurements and the subsequent statistical significance. It is thus recommended to have an increased number of sampling sites to be monitored consistently across sampling years. Second, the lack of possible control locations poses a challenge to the internal validity of the results only observed in areas affected by the increased As levels. A case-control study, to be empirically performed subject to the availability of suitable data, may address this limitation.

4. Conclusions

Through the use of a Wilcoxon signed-rank test, this study found significantly elevated As levels in 23 out of 26 (88.46%) individual wells in unique locations in Batangas Province in 2020 (p -value < 0.001), which is as high as seven times more than the 10 ppb safe limit. Of these, 20 out of the 26 wells (76.92%) had persistently elevated As levels through to 2021 (p -value = 0.013). A two-paired Wilcoxon signed-rank test revealed that As levels were statistically higher in 2020 than 2021 (p -value = 0.003). The reduction of As levels in some of the wells may be explained by higher volcanic activity in 2020 compared to in 2021, but this must be further confirmed by more extensive longitudinal data. Increasing the number of sampling sites and the frequency and duration of the investigation in future studies may mitigate this limitation. Bivariable regression showed no evidence of an association between As levels and distance (p -value 2020, 2021 = 0.537, 0.128) or elevation in relation to the source (p -value 2020, 2021 = 0.500, 0.584), which supports the notion of As groundwater contamination through volcanic hydrothermal systems. We recommend geomapping of the hydrothermal system that interacts with the groundwater aquifer in Batangas Province and the inclusion of sampling sites at the perimeter of the aquifer in order to more accurately determine the extent of the affected local inhabitants' exposure to As. Continuing and province-wide water quality assessment (blanket testing) of drinking water will also help to identify and predict areas at risk of future groundwater contamination and locate safe and unsafe drinking water sources. Furthermore, we recommend investigations on the relationship of As levels with other physico-chemical parameters such as pH, oxidation-reduction potential, and sulfide levels, as well as geo-climatic parameters such as precipitation and terrain, among others. Doing so may be helpful in assessing future options for groundwater resource management. Due to the persistence and innate nature of As, population bioexposure assessments and screening for arsenicosis is recommended for the affected communities. In terms of risk mitigation, the rapid conduct of risk communication efforts for the affected populations, alongside the provision of safe drinking water alternatives in the short-term, as well as the identification of cost-effective treatment technologies and/or the exploration of uncontaminated groundwater sources are critical steps in minimizing public health risks and protecting the population's health moving forward.

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