Spatiotemporal Variation of Groundwater Arsenic in Pampanga, Philippines

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Abstract: Several confirmed cases of arsenic (As) poisoning have been reported in Central Luzon, the Philippines, in recent years. There is a growing interest in As research in the Philippines due to the reported As poisoning cases. However, an extensive spatiotemporal As study has not been conducted. In this work, As concentration measurements were conducted in 101 wells in Guagua, Pampanga, in Central Luzon, the Philippines, from November 2018 to November 2019. The wells included 86 public hand pumps, 10 pumping stations, and 5 private, jet-powered pumps. Using hydride generation—inductively coupled plasma—optical emission spectroscopy (HG-ICP-OES), analysis of the wells in 12 barangays in Guagua revealed that 38.7% had average As concentrations beyond the 10 ppb limit with some wells having high Mn (4.0 ppm) and Fe (2.0 ppm) content as well. The high pH and reducing conditions in the wells in Guagua may have contributed to the persistence of As in the groundwater. The mean difference in wet season versus dry season As measurements were −4.4 (As < 10 ppb), −13.2 (10 to 50 ppb As), and −27.4 (As > 50 ppb). Eighty-three wells (82.2%) had higher As concentrations in the dry season, 8 wells (7.92%) had higher As concentrations in the wet season, 7 wells (6.93%) had no significant difference between the wet and dry season, and 3 wells had been decommissioned. These results indicate that there is a significant difference in As concentrations in the wet and dry seasons, and this could have implications in water treatment technology and policy implementation. The work resulted in the first year-long characterization of groundwater As in the Philippines.

Keywords: arsenic; groundwater; mapping; seasonal variation

1. Introduction

Arsenic (As) is a known pollutant present in groundwater aquifers. Chronic exposure to As-containing compounds promotes genetic mutation resulting in an increased risk of cancer, skin diseases, and various damage to other organ systems [1]. Groundwater As contamination is reported worldwide and in Asia, there is an estimated total of 150–240 million people affected by high levels of As in countries such as India, Bangladesh, Cambodia, and Vietnam [2]. The Philippines shares many similar geological features with these countries and is heavily reliant on groundwater for consumption. Arsenic poisoning, or arsenicosis, is an emerging health issue in several regions in the Philippines. The species of As in the groundwater contribute to the exposure risk, and the order of decreasing toxicity is $R_3As > H_2AsO_3 > H_3AsO_4 > R_4As^+ > As^0$, where R is a proton or an alkyl
group [3,4]. Several As removal technologies have been tested and implemented, such as adsorption (iron fillings, lignite, manganese ore), electrocoagulation [5], and filtration (modified ultrafiltration and nanofiltration) [6]. There is still much to cover when it comes to As pollution mapping, As remediation technology implementation, and research-driven policy-making in the country [7].

A couple of studies conducted in various parts of the Philippines have reported elevated concentrations of As in river sediments, surface water, and groundwater. The majority of stream sediment samples ($N = 39$) collected from the Zambales Mountain Range in Central Luzon contained 0.05–1.0 ppm As, while the highest measured was 114.8 ppm [8]. A study on rice paddy soil samples revealed As values beyond the soil background value of 9.0 ppm in some parts of Laguna (15.5 ppm) and Camarines Sur (9.2 ppm) in Luzon and Negros Occidental (11.4 ppm) in Visayas [9]. Groundwater samples from Central Luzon were found to contain As beyond the 2017 Philippine National Standards for Drinking Water (PNSDW) limit of 10 ppb, with values as high as 300 ppb [7]. A study on the Marbel–Matingao river catchment near the Mt. Apo geothermal plant in South-Central Mindanao showed that the natural hot springs feeding into the river system contributes significantly to the elevated As levels (260 ppb) at the lower reaches of the catchment [10]. The Dutch-led mission report in 2015 noted 214 cases of suspected arsenicosis from Central Luzon between 2010 and 2014 [7]. Other cases of arsenical keratosis were reported in Southern Luzon [11–13] and Southern Mindanao [13,14].

The natural presence of As in Philippine groundwater could be attributed to geothermal activity characterized by the leaching out of As from As-rich rocks in the geothermal reservoir and the subsequent mixing of geothermal water with shallow groundwater and even with surface water [15]. There is also evidence presented that clay layer compaction typical of ground subsidence events contribute to an increase in As content in groundwater. This is due to the introduction of As-rich porewater in the clay layer to the aquifer [16]. Studies have also been conducted to characterize the seasonal variation of As content in groundwater and these yielded mixed results; work done by Shrestha et al. (2014) in Kathmandu Valley, Nepal observed an increase in As levels during or immediately following the dry season [17], while Savarimuthu et al. (2006) observed higher values during or after the wet season in West Bengal, India [18]. Still, there are studies that show no significant difference in mean As values between the wet and dry seasons [19,20]. It is suggested that precipitation could affect As values by way of dilution via recharge of low As rainwater, pumping rates, and variation in redox conditions [21].

The majority of the Philippine public is unaware of the emerging As problem in drinking water. There are only a handful of studies that map the As concentration in areas with reported arsenicosis, and published works on long-term As monitoring are non-existent. It is, therefore, important to work with local government units (LGU) to monitor As and implement sustainable solutions. In this study, the As concentration of groundwater in Central Luzon was measured over a 12-month period to capture the temporal variation during the wet and dry season. The results of the monitoring will then be used to help the LGU decide on interventions and perform an information dissemination campaign to raise the awareness of the public. In addition, the data will be used to select a specific area in one of the municipalities for the installment of a community-managed water-treatment facility using electrocoagulation, which is a technology that has seen success in other parts of Asia [22].

2. Materials and Methods

The municipality of Guagua occupies 48.67 km$^2$ of land in Pampanga, Central Luzon, has a population density of 2400/km$^2$ according to a 2015 census, and is composed of four districts: Poblacion, Loción, Betis, and Pangulo. These are further sub-divided into 31 smaller political units called barangays [23]. The study site is shown in Figure 1. The municipality is geographically flat, and in some places, below sea level. Floods are a common occurrence during the rainy season. The study site is 36.2 km from Mt. Pinatubo; when it erupted in 1991, parts of Guagua were completely covered in several meters of pyroclastic material. The climate in Guagua is the same as most parts of the Philippines; the wet season is from June to November, and the dry season is from December to May [24].
The districts of Pangulo, Locion, and Betis are predominantly agricultural areas, and the rivers coursing through Locion and Betis make these two districts ideal for freshwater farming.

Water samples were extracted from public and private-owned handpumps and faucets, to be collectively referred to as wells, in Guagua. Well identification and initial sample testing from 2 wells per barangay were conducted from September to October 2018. Convenience sampling was performed from November 2018 to November 2019, except in June. Additional wells were included in the study after three months (November 2018–January 2019) for barangays with As values > 10 ppb. Subsequent sampling from wells located in remote areas, which recorded As values < 10 ppb for three months (November 2018–January 2019), were done quarterly. Two to five more wells from Barangay San Isidro and San Antonio were included in the study at the final quarter (September 2019–November 2019) of the sampling period. There were 3 wells that were decommissioned during the sampling period due to a major earthquake, while some well locations became inaccessible during the rainy season due to flooding. Sampling of wells located in the surrounding municipalities of Guagua (N = 8) was conducted in January, April, July, and October 2019. Samples were also obtained in January, May, September, and November 2019 from wells (N = 14) in Castillejos, Zambales, which lies 70 km from Guagua and is in close proximity to Mt. Pinatubo. This step was performed to compare As values between two municipalities that have similar characteristics, such as abundance in pyroclastic deposits and rainfall. There was a total of 101 wells tested; 35 were monitored monthly, while the rest were checked quarterly, were decommissioned, or were recent additions. A total of 493 measurements were conducted throughout the study period.

Global positioning data was obtained using the Garmin eTrex 20X GPS. Short interviews with public officials and people living around the wells were conducted to gather other information, such as well depth, well age, perception of groundwater, and groundwater use. Global imaging system coordinates were projected to GCS Luzon 1911 UTM Z51N, and maps were generated using ArcGIS 10.5. An administrative boundary shapefile was provided by the National Mapping and Resource Information Authority (NAMRIA). Central Luzon water quality assessment reports from 2010 to 2015, provided by the Local Water Utilities Authority (LWUA) and National Water Resources Board (NWRB), were reviewed for baseline water quality parameter values. Rainfall data from November 2018 to November 2019 was provided by the Municipal Environment and Natural Resources Office (MENRO) of Guagua.
Qualitative assessment, such as visual and odor inspection, of water samples was performed in-situ. Groundwater samples were collected in acid-washed 250 mL High density polyethylene (HDPE) bottles and transported to the laboratory for analysis. The pH of the samples was lowered to 3 using concentrated HNO\textsubscript{3}. Approximately 10 mL of sample was passed through a 0.4 \textmu m syringe filter and stored in 15 mL polypropylene centrifuge tubes for analysis. Samples over 3 months old were discarded to make room for additional samples. The total As, total manganese (Mn) and total iron (Fe) levels of the water samples were measured using the Teledyne-Leeman Prodigy7 hydride-generation-inductively coupled plasma-optical emission spectroscopy (HG-ICP-OES), which runs on Salsa software version 5.0. Approximately 10 mL of filtered water samples were acidified with conc. HCl in a 1:10 v/v ratio and 250 mL of fresh 0.13 M NaBH\textsubscript{4} solution was prepared before ICP analysis for As hydride generation. Other heavy metals were analyzed without using the hydride generation system. An initial calibration method and continuous calibration method were performed throughout the analysis, while a quality assurance check at 50 ppb ± 10% was performed every 15 samples. The method detection limit for total As was 5.5 ppb. The pH, conductivity, turbidity, ORP, and amount of dissolved oxygen of the water samples were measured using the Hanna HI 9829 Multiparameter Water Quality Meter. A Department of Health (DOH) accredited third-party laboratory performed the alkalinity test using titrimetric analysis and determined total phosphate using SnCl\textsubscript{2} analysis, total sodium using Flame AAS, total chloride using Argentometric titration, total silica as silicates using a molybdosilicate test, and total sulfate using turbidimetric analysis.

3. Results

3.1. Sample Well Profile

Analysis of the well profiles reveal that of the 101 wells, 86 are publicly available hand pumps (85.1%), 5 are private-owned but publicly available, machine-operated pumps (4.95%), and 10 are from municipal groundwater distribution pumps (9.90%). The location of the wells is indicated in Figure 1. There are 27 wells identified as sources of drinking water (26.7%). However, only residents living within the immediate vicinity of the wells were interviewed with regard to their water use. Based on another study on Philippine groundwater consumption, approximately 60% of groundwater use in the Philippines is without a permit [25]. The interviews conducted were limited to locals living within 10 m of the wells and did not include those that might use the wells but were living further out. These factors lead the researchers to believe that the actual number of wells used for drinking is much higher. The community also uses water for cooking, laundry, and other domestic functions. Most households have access to piped water, but some residents complain of a strong scent of chlorine and prefer the taste and familiarity of hand-pumped water. Only 33 wells have depth data, which range from 6 m to 90 m; many of the shallow wells (<15 m) are located in Pangulo District. Most of the wells included in this study are located in low-lying areas (<20 m above sea level), as indicated in Figure S1. These areas are densely populated and are submerged in floodwater at varying depths during the wet season.

3.2. Arsenic and Other Water Quality Parameters

Average arsenic values and the number of samples measured per barangay are indicated in Table 1. Of the 441 samples obtained from Guagua, 32.9% were obtained from Locion district, 29.9% were from Poblacion district, 21.1% from Betis district, and 16.1% from Pangulo district. Five barangays in Poblacion district had average values >10 ppb, with the highest measured in San Nicolas 1st (95 ppb). All six barangays in the Locion district had average As values exceeding the 10 ppb limit with the highest measured in San Isidro (82 ppb). Locion and Poblacion districts are 9 m below sea level to 5 m above sea level. Only San Juan Nepomuceno, which is below sea level, in Betis district had an average As value exceeding 10 ppb. The wells from Pangulo district, which is located 3-20 m above sea level, had average As concentrations below 10 ppb.
Table 1. Groundwater arsenic concentration in the 31 barangays of Guagua, Pampanga. MDL = minimum detection limit.

| District  | Barangay            | No. of Samples | Average As Conc. in ppb | Max. As Conc. in ppb | Min. As Conc. in ppb |
|-----------|---------------------|----------------|-------------------------|----------------------|----------------------|
| Poblacion | Sta Filomena        | 27             | 22.4 ± 1.8              | 54                   | 10                   |
|           | Padre Burgos        | 18             | 7.8 ± 2.5               | 31                   | MDL                  |
|           | Sto Cristo          | 7              | 8.9 ± 4.2               | 21                   | MDL                  |
|           | Sto Nino            | 12             | 10.1 ± 2.8              | 26                   | MDL                  |
|           | Bancal              | 15             | 15.0 ± 2.1              | 29                   | MDL                  |
|           | San Rafael          | 11             | 32.1 ± 2.3              | 66                   | 16                   |
|           | San Pedro           | 18             | 29.0 ± 2.0              | 65                   | 10                   |
|           | San Nicolas 1st     | 16             | 32.8 ± 2.2              | 95                   | MDL                  |
|           | San Roque           | 8              | 4.8 ± 4.8               | 11                   | MDL                  |
| Locion    | San Pablo           | 9              | 39.6 ± 4.2              | 71                   | 26                   |
|           | San Juan 1st        | 20             | 12.7 ± 2.8              | 55                   | MDL                  |
|           | San Jose            | 26             | 19.9 ± 2.6              | 42                   | MDL                  |
|           | San Matias          | 25             | 19.0 ± 3.9              | 44                   | MDL                  |
|           | San Isidro          | 26             | 26.7 ± 4.4              | 82                   | MDL                  |
|           | San Antonio         | 39             | 32.5 ± 3.7              | 75                   | MDL                  |
| Betis     | San Juan Bautista   | 16             | 2.8 ± 5.5               | 25                   | MDL                  |
|           | San Juan Nepomuceno | 19             | 15.5 ± 3.4              | 55                   | MDL                  |
|           | San Miguel          | 16             | 2.5 ± 3.3               | 6                    | MDL                  |
|           | San Nicolas 2nd     | 8              | 5.0 ± 6.0               | 11                   | MDL                  |
|           | Sta Ines            | 9              | 3.1 ± 6.0               | 7                    | MDL                  |
|           | Sta Ursula          | 10             | 8.9 ± 5.8               | 35                   | MDL                  |
|           | San Agustin         | 15             | 2.0 ± 4.1               | 7                    | MDL                  |
| Pangulo   | Natividad           | 5              | 1.4 ± 7.9               | MDL                  | MDL                  |
|           | San Vicente Ebus    | 5              | 0.9 ± 8.4               | MDL                  | MDL                  |
|           | Lambac              | 5              | 2.0 ± 7.1               | 8                    | MDL                  |
|           | Pulungmasle         | 24             | 0.5 ± 2.9               | 7                    | MDL                  |
|           | Rizal               | 7              | 2.0 ± 5.9               | 8                    | MDL                  |
|           | Magsaysay           | 8              | 1.7 ± 6.3               | 8                    | MDL                  |
|           | Maquiapo            | 9              | 3.2 ± 6.8               | 15                   | MDL                  |
|           | Ascomo              | 6              | 0.1 ± 8.3               | MDL                  | MDL                  |
|           | Jose Abad           | 2              | 4.3 ± 9.6               | 6                    | MDL                  |

Water quality assessment of groundwater samples was performed, and the results are summarized in Table S1. The pH of groundwater samples from Guagua was between 7.3 and 8.1, which is slightly basic, and the oxidation-reduction potential (ORP) value was between $-142$ and $-158$ mV. At higher pH, oxyanions become less sorbed because the active sites of adsorbents, such as minerals in the aquifer, tend to be occupied by the OH molecule. Dipole–dipole repulsion occurs between the oxyanions and the OH molecule; hence, the oxyanions stay in the aqueous phase [26]. As oxyanions are more mobile than other oxyanions, such as selenate, Cr (VI) oxyanion, and molybdate, in reducing conditions. The predominant As oxyanion in high pH and reducing conditions is the neutral-charged $\text{H}_3\text{AsO}_3$ [27]. Negative ORP readings of the sample water indicate reducing conditions typical of a deep aquifer, which was observed by Shrestha et al. (2015) [19], however reducing conditions, high Mn, Fe, and As content was also measured in shallow aquifers [28]. Manganese (Mn) levels beyond the 500 ppb PNSDW limit for drinking water were recorded in 11 barangays in Locion, Betis, and Pangulo districts. Manganese and iron-containing minerals were abundant in clay layers of aquifers. The reduced form of manganese is more soluble than its oxidized form; hence, high levels of Mn is typical of anoxic and reducing conditions in groundwater. Iron (Fe) levels beyond the 1 ppm PNSDW limit were measured in 4 barangays in the Betis and Pangulo districts. Groundwater samples ranged from being colorless and odorless to orange–brown, having a rusty smell, and were turbid. These observations correspond
well with accounts from locals who source their water from these wells. The locals attributed these undesirable conditions to earthquakes and flooding of nearby rivers.

3.3. Seasonal Variation in Arsenic Levels

Dry season samples \((N = 306)\) were collected from December 2018–May 2019, while wet season samples \((N = 187)\) were collected in November 2018 and from July 2019–November 2019. The distribution of As measurements per 10 ppb is shown in Figure 2. Arsenic measurements were categorized into three main levels to reflect the previous and current PNSDW for As; low (As value < 10 ppb), moderate (10 to 50 ppb), and high (As value > 50 ppb). Water arsenic levels were low in 246 samples (49.9%), moderate in 221 samples (44.8%), and 26 samples contained high As level (5.3%). At least 12 of the 27 (44.0%) wells used for drinking exceeded the 10 ppb limit. Statistical analysis (2-sample t-test) was used to evaluate the difference in mean As levels between the wet and dry season at a 5% level of significance. The summary of results is indicated in Table 2. For samples with low As levels, comparison of the mean As value during the dry season (5.70 ppb) versus the wet season (1.30 ppb) resulted in a \(p\)-value of \(1.37 \times 10^{-4}\), which suggests that there was a significant difference between the two means. For samples with moderate As levels, comparison of the mean As value during the dry season (28.9 ppb) versus the wet season (15.6 ppb) resulted in a \(p\)-value of \(5.67 \times 10^{-3}\), which suggests that there was a significant difference. In the high As level category, the mean value during the dry season (56.6 ppb) and the wet season (29.2 ppb) were found to be significantly different with a \(p\)-value of 0.02. The 2-sample t-test of the low, moderate, and high As groups indicate a significant difference in As measurements per group during the dry season and the wet season. A chi-square test was performed to prove that season affects the increase in As values among the groups. The low and moderate groups were further divided into As < 5 ppb and 5 < As < 10 ppb, and 10 ≤ As < 25 ppb and 25 ≤ As < 50 ppb, respectively, to capture the variation among data points within the two large groups. The resulting \(p\)-value was <0.001, which indicates a significant difference among the As groups relative to the seasons. A plot of the average As values for the wet (x-axis) and dry (y-axis) season in 35 wells in Guagua is shown in Figure 3. This figure shows that generally higher As values observed during the dry season from the same wells, as shown by the location of most points above the 1:1 line.

![Figure 2. Distribution of samples per As level. Measurements were taken from November 2018 to November 2019 in Central Luzon (N = 493).](image-url)
Table 2. The number of wells in which the arsenic level is higher or lower in the dry season compared to the wet season.

| Arsenic Concentrations (ppb) | Number of Wells | Mean Seasonal Difference in As (ppb) | p-Value (t-Test at 5% Significance) |
|--------------------------------|-----------------|---------------------------------------|-------------------------------------|
| Total                          |                 |                                       |                                     |
| Wet > Dry                      | 60              | 7 46                                  | −4.4 1.37 × 10^{−4}                |
| Dry > Wet                      | 10              | 34                                    | −13.2 5.67 × 10^{−3}               |
| <10 (low)                      |                 |                                       |                                     |
| 10-50 (moderate)               | 35              | 34                                    | −27.4 0.02                         |
| >50 (high)                     |                 |                                       |                                     |

Figure 3. Comparison of average groundwater As values in 35 wells in Guagua, the Philippines, shows most points above the 1:1 line indicating higher values during the dry season.

4. Discussion

The arsenic heat map of Guagua for the wet season and dry season is shown in Figure 4. Moderate (10–50 ppb) to high levels of As (>50 ppb) were measured in the Locion and Poblacion districts for both seasons; more intense readings were observed during the dry season. As shown in the map, some areas in Betis district recorded mean As values above 10 ppb during the dry season. The commercial district, as well as pockets of residential spaces, is located in these hotspots. Very low As levels were recorded in other areas of Guagua, which are mostly allotted to agricultural purposes.

Rainfall data for Guagua was plotted, along with the average monthly As measurements, as shown in Figure 5, to describe the differences observed in As values during the wet and dry season. The lowest average precipitation was recorded in February at 10 mm. On average, 55 mm of rainfall was recorded in the dry season, which is approximately six times less than the recorded 317 mm average in the wet season. Peak precipitation at 522 mm was recorded in August. There were generally higher monthly As concentrations in the dry season with the peak average recorded in December. The figure shows that the mean As level had a decreasing trend going to the wet season and then steadily rose, leading to the dry season. The increase in rainfall coincided with a general dip in monthly As values. This result shows that rainfall could have a dilution effect most likely felt in the shallow wells. The importance of rainfall pattern to the occurrence of As is evident in spatiotemporal studies conducted in tropical
However, the lowest recorded average for the wet season in July (17.0 ppb) was insignificantly different \((p\text{-value} = 0.47)\) from the lowest recorded during the dry season in April (17.3 ppb) at 5% significance interval. This observation implies that other factors could affect the As levels aside from rainfall. Other studies have looked into ground subsidence \([16]\) as a likely factor that influences As content in groundwater. The compaction of clay layers in the aquifer causes As-rich porewater to be introduced to the aqueous layer. This increases groundwater arsenic concentration. Guagua and some parts of Pampanga are located in the delta region north of Manila bay. A study on the compaction rates in this region revealed that Guagua is generally sinking at a rate of 1.5–2 cm \(\text{y}^{-1}\) due to natural compaction, compounded by groundwater over-extraction \([29]\). The impact of ground subsidence on the measured As levels is still being investigated.

**Figure 4.** The wet season and dry season As heat maps of Guagua. Inset map shows several barangays of the Locion and Poblacion districts with high levels of As.

**Figure 5.** Monthly rainfall and monthly average As data in Guagua, the Philippines, from December 2018 to November 2019.
Water quality records of water districts in Central Luzon reported no As values beyond 10 ppb, which is contradictory to the findings of this and other studies conducted in the region [7,30]. The Guagua water district office submitted independent water quality reports to the NWRB, which showed elevated As values in the groundwater being distributed to households (December 2018–May 2019). These reports support the findings of this study. There are still more areas to cover in Central Luzon when it comes to arsenic monitoring based on the records available.

5. Conclusions

The municipality of Guagua and other places in Central Luzon were found to have groundwater As concentration beyond the 10 ppb limit using ICP-OES analyses. Arsenic test results from wells used for drinking confirmed that some residents have been exposed to As beyond the drinking limit. These areas are located where large amounts of pyroclastic material from nearby Mt. Pinatubo have been deposited, are undergoing ground subsidence, are reliant on groundwater for domestic and commercial use, and are predominantly agricultural. There was a significant difference in As concentrations between the wet and dry season. High amounts of precipitation during the wet season may induce a dilution effect in some wells. The results of the spatiotemporal study could aid authorities in regulating the installation of new hand pumps in barangays with consistently high As groundwater content and to close down existing hand pumps in heavily affected areas. The results may be used to encourage the local water district to perform additional treatment steps before releasing groundwater to households. In addition, the results of the water quality monitoring are being used for the site-selection study. A 600 L community-managed, electrochemical arsenic removal treatment plant will be deployed in the selected barangay.

Supplementary Materials: The following are available online at http://www.mdpi.com/2073-4441/12/9/2366/s1, Table S1. Water quality parameters of groundwater samples from Guagua, Philippines, Figure S1: Elevation map of Guagua, Figure S2. Well location in Guagua, Pampanga and corresponding well depth; larger radius indicate deeper wells, Figure S3. Mean As level in each barangay in Guagua, Pampanga, Figure S4. The total number of samples measured per barangay in Guagua, Pampanga from November 2018 to November 2019.

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