Conceptual Model for Arsenic Distribution and Its Relationship with Groundwater Depth

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Abstract: In this study, arsenic distribution in groundwater and its relationship with the depth of groundwater was studied. During the study, water samples were taken from wells and community boreholes in the Obuasi Municipality. The samples were analyzed, and models were developed to provide a better understanding of arsenic distribution and groundwater depth. The study concludes that, at greater depth in community boreholes (CBH > 50 m), there is a positive correlation of depth with total arsenic indicating total arsenic mobilization whereas in shallower depth (wells and CBH < 50 m) the correlation is not very clear suggesting total arsenic may be mobilized or demobilized depending on prevailing conditions. There is contribution of microbial activity in the mobilization of arsenic in the groundwater. Arsenite mobilizes with increasing borehole depth and demobilizes with decreasing borehole depth. In wells, there appear to be two arsenite groups all negatively correlated with depth. There is positive correlation between carbonate and silicate weathering and arsenic distribution in the order Wells > shallow MBH > deep MBH > shallow CBH > deep CBH. Shallow monitoring boreholes (MBH) appear to mobilize arsenic through silicate weathering and deep MBHs and CBHs generally mobilize arsenic through carbonate weathering, whilst shallow CBHs and wells appear to mobilize arsenic through a combination of carbonate and silicate weathering. There is therefore the need to identify areas with suitable deep aquifers where there is adequate knowledge of the recharge mechanism and the soil profile to determine protected aquifers for location of new boreholes.

Keywords: Arsenic Groundwater Depth Conceptual Model

I. INTRODUCTION

rsenic can be found in over 200 minerals in the form of arsenate, arsenites, arsenides, sulphides and oxides (Smedley et al., 2002). The concentration of this metalloid in surface water and groundwater is increasing through among others the use of arsenical pesticides, mining, processing, oxidation-reduction of soil minerals and burning of fossil fuels (Ferguson et al., 1972 and Igboamalu et al., 2018). The greatest concentrations of the element tend to occur in sulphide minerals such as arsenopyrite (FeAsS) (Smedley, 1996). Groundwater pollution by arsenic is a serious threat to mankind all over the world (Igboamalu et al., 2017 and Smedley et al., 2007). Studies have linked long-term exposure to arsenic in drinking water to cancer of the bladder, lungs, skin, kidney, liver, and prostate (Anon., 2001). Buruli ulcer is linked to arsenic exposure in Ghana (Duker, 2004; Gyasi et al., 2011, Gyasi et al., 2012a; Gyasi et al., 2012b; Gyasi et al., 2012c; Gyasi et al., 2012d; and Gyasi et al., 2014). Baseline concentrations of arsenic in surface water range between 0.1 μ g/l and 0.8 μ g/L, but can increase up to 30 μ g/l depending on the composition of the surface recharge, the contribution from base-flow and the bedrock lithology (Smedley et al., 2002). Arsenic has been found in concentrations above 50 µg/l in many large aquifers in the world. The following are areas are noted to have significant concentration environments: Bangladesh (up to 1000 µg/l), West Bengal of India (up to 500 μ g/l), Taiwan (10~ 1000 μ g/l), Vietnam (1~3050 μ g/l), Romania (up to 150 µg/l), and Hungary (up to 150 µg/l) (Smedley et al., 2002). In the arid oxidizing environments such as Argentina (up to 225 μ g/l), Chile (up to 500 μ g/l), China (up to 175 µg/l), Mexico (up to 1400 µg/l), and mixed oxidizing and reducing environments such as many parts of USA (up to 50 µg/l), particularly in the southwest regions (Planer-Friedrich et al., 2008; Smedley et al., 2008; and Xie et al., 2008). In the United States of America, wells that had >50 µg/l arsenic were abandoned as they did not meet the 50 µg/l minimum compliance limit (Planer-Friedrich et al., 2008). Ghana has a legacy of metalliferous mining, and Obuasi is a historical mining town that has seen continuous mining operations since the 1890's (Akabzaa et al., 2005; Anon., 2006; and Bowell, 1993). Intensive exploitation of metalliferous ore deposits, combined with the natural geochemical dispersion from arsenic mineralization, has resulted in the creation of areas of arsenic contaminated water, plants, and soils at Obuasi (Ahmad et al., 2000). Amonoo-Neizer et al. (1980) found high concentrations of arsenic in soils, plants and fish in the Obuasi area. Arsenic can transform through oxidation-reduction reactions, ligand exchange, and bio-transformations (Anon., 2000; Foli et al., 2011; Mayo, 2006; and Welch et al., 2000). The type of arsenic species present, Eh, pH, Iron and sulphide concentrations, salinity, temperature, aerobic or anaerobic state, dissolved organic content and presence of microbiological organisms in the groundwater are factors likely to determine the fate and transport processes of arsenic in groundwater (Anon., 2000; Igboamalu et al., 2020; Roberston, 1989; and Welch et al., 2000). The predominant form of arsenic is usually arsenate (As^{5+}) , although arsenite (As^{3+}) may be present under some conditions (Welch et al., 2000). However, it has been noted that arsenite might be more prevalent than anticipated (Anon., 2001).

A. Groundwater Characteristics of Obuasi

Groundwater occurrence in the Obuasi Municipality is from fissures or fractured zones and within the thick weathered saprolitic zone overlying the bedrock (Amanor *et al.*, 1988). There is high rainfall and resultant short residence times of many of the groundwater bodies in the aquifer (Ewusi *et al.* 2013). The depths of boreholes in the rock vary between 30 m and 100 m with an average depth of 42 m. The aquifer horizons are intercepted at depths between 17 m and 42 m (Asamoah, 2004). Water sources for domestic use in the study area fall into the following:

- Hand dug wells with depth of 10 m and above with hand pumps.
- Hand dug wells with depth between 5 m and 10 m without hand pumps.
- Open pits recharged from groundwater (with or without) surface water inflows.
- Direct use of streams and rivers; and
- Groundwater boreholes with depth between 30 m and 100 m.

The availability of water in the study area tends to be seasonal (Ewusi *et al.*, 2013). During the dry season, many of the shallow wells and some surface streams dry out and the productivity of the various wells and boreholes decrease (Ewusi *et al.*, 2013). Typical water uses are for drinking, cooking, spiritual cleansing, washing, and bathing with other general uses including swimming and fishing. The degradation of the local water supplies is caused by a variety of causes including poor disposal of garbage, poor sanitation, potential release of faecal and coliform bacteria from domestic animals within villages, erosion/siltation, oxidation reactions with exposed soils as water levels drop, the activities of artisanal miners and the Obuasi Mine (Banson *et al.* 2016a).

B. Groundwater Geology of Obuasi

The host geology of Obuasi Municipality is predominantly Birimian metasediments (Kesse, 1985). Fractures, quartz veins and stringers, along with shear zones and stockworks are the main determining factors of the permeability of the study area (Kesse, 1985). The extent of weathering of the lithology and the nature, length, thickness, density, infilling of the fractures and the interconnection of the above anomalies largely define the development of groundwater aquifers in the lithology (Amanor *et al.*, 1988). For significant volumes of groundwater to be retained and transmitted in the lithology, there is the need for numerous and well interconnected fractures to generate enough porosity and permeability (Bowell, 1991). The groundwater of the study area can therefore be linked to the extent of weathering of the lithology and the nature, length, thickness, density, infilling of the fractures and the interconnection of the rocks (Amanor et al., 1988).

C. Groundwater Flow Direction

In the study area, the flow of groundwater in aquifers tend to occur from secondary permeabilities through fractures, joints, quartz veins and other discontinuities instead of through interstitial flows. (Amanor et al., 1988 and Banson et al., 2018b). There are many barriers to continuous groundwater flow laterally as well as with depth (Ewusi et al., 2013). As a result of the different rock types within the lithology, there is weathering at different depths within the lithology. The nature and degree of weathering also varies spatially (Banson et al., 2016b). It is noted that there are areas with unfractured lithology which often pose as lateral barriers to groundwater flow (Gyapong and Amanor, 2003). The determination of groundwater flow direction in any geologic formation is very complex and depends upon a variety of factors (Banson et al., 2018c; and Gyapong and Amanor, 2003). The presence of geological structures such as faulting, folding and jointing constitute important controlling factors. AngloGold Ashanti Limited identified the direction of groundwater flow towards the Offin River to the South West (Banson et al., 2018b and Anon., 2012b).

D. Recharge Areas and Rate of Recharge

Recharge of an unconfined aquifer occurs at the ground level directly above the aquifer (Banson et al., 2018a and Gyapong and Amanor, 2003). Infiltration of rainwater and subsequent percolation into the groundwater system as base-flow depends on the aperture, density, and degree of interconnection of the fractures and the permeability of the weathered rock (Ewusi et al., 2013). Topographic highs are normally considered as areas of groundwater recharge and topographic lows as areas of groundwater discharge, especially in a terrain in which the occurrence of groundwater is controlled by the geomorphology, geology, and tectonic setting (Asamoah, 2004). Other critical inputs to groundwater recharge include the amount of precipitation, the extent of evapo-transpiration, rock type, topography, permeability of aquifer, and surface runoff (Anon., 2011). The rate of recharge to the aquifers in the study area will require a continuous data gathered over a period of years to be estimated. Generally, about 15% of the total precipitation ends up as groundwater, but this varies locally and regionally from 1 to 20% (Anon., 2011). The recharge for the study area based on studies carried out by Jay Minerals Services around Obuasi and Dunkwa community is estimated at 3 to 5% of the total annual rainfall (Anon., 2011). Adequate and dependable groundwater is known to occur within the study area. Borehole yields vary from 62 to 259 m^{3}/day with an average of 124 m^{3}/day and the recorded static water levels vary from 3.1 m to 15 m with an average of 7.6 m compared to an average borehole depth of 42 m (Asamoah, 2004).

II. MATERIALS AND METHODS

The main methods used in the study were field assessment, collection and analysis of water samples to identify the main arsenic release indicators in the study area. Statistical analysis was conducted on identified arsenic release indicators in the study area.

Sampling locations are shown in Figure 1. Water depth sampler was used for sampling wells. Groundwater samples from existing monitoring boreholes were taken with bailers after purging when possible. Groundwater samples were collected from 108 community boreholes (CBH), 19 monitoring boreholes (MBH), 11 hand-dug wells and 5 mechanized borehole systems in 96 settlements in the study area.

Some samples were analyzed directly in the field for pH, Eh and Dissolved Oxygen (DO), whilst others were sent to the Perseus Mining Edikan Environmental laboratory and University of Mines and Technology Environmental Laboratory for analysis of arsenite, total arsenic, Fe, Ca, Mg, Na, K, $SO_4^{2^-}$, HCO_3^- and dissolved organic carbon (DOC). A depth sampler was used to measure the depth of every well whilst the depth of the boreholes was obtained from the Anglogold Ashanti Obuasi Mine Monitoring Department.



Figure 1: Sampling Locations with Satellite Image at Background (Source: Modified after Anon., 2010 and Anon., 2012b)

III. RESULTS AND DISCUSSION

A. Effect of Groundwater Depth on Arsenic Distribution in Obuasi Municipality

The mean DO content of the groundwater in the rainy season is 4.79 mg/l (0.84 to 5.17 mg/L) while the mean DO for dry season is 3.99 mg/l (0.78 to 4.76 mg/L). This indicates the suitability of groundwater for hypoxic and anaerobic action. The dissolved oxygen in CBH's decreases with depth whereas dissolved oxygen for wells (shallower depth) increases with depth (Figure 2).



A plot of total arsenic concentration against CBH and well depth (Figure 3) shows that at greater depth in the CBH (> 50 m), there is a positive correlation of depth with total arsenic indicating total arsenic mobilization whereas in shallower depth (wells and CBH < 50 m) the correlation is not very clear suggesting total arsenic may be mobilized or demobilized



Figure 3: Plots of Total Arsenic vs. Depth of CBH and Wells

Figure 4 shows the presence of two groups of total arsenic in CBH and wells with the higher total arsenic group having a positive trend with DOC and the lower total arsenic group having no clear trend with DOC. This indicates total arsenic is mobilized with increasing DOC, but total arsenic demobilization is not dependent on DOC content of the water. This suggests the contribution of microbial activity in the mobilization of arsenic in the groundwater of the study area.



Figure 4: Plot of Total Arsenic vs. DOC in Wells and CBH

Figures 5 and Figure 6 shows two groups of arsenite in CBH and wells respectively. The higher arsenite in CBH group has a positive trend with DOC and the lower arsenite in CBH group has a negative trend with DOC. This suggests arsenite demobilization with increasing DOC (low arsenite concentration) and arsenite mobilization with increasing DOC. In wells (Figure 5.91), there are two arsenite groups all positively correlated with DOC. However, the arsenite mobilization trend is less pronounced with increasing DOC.



Figure 5: Plot of DOC vs. Log of Arsenite in CBH



Figure 6: Plot of DOC vs. Log of Arsenite in Wells

Figures 7 and Figure 8 show two groups of arsenite in CBH and wells respectively. The higher arsenite CBH group has a positive trend with borehole depth and the lower CBH arsenite group has a negative trend with borehole depth (Figure 7). This indicates arsenite mobilization with increasing borehole

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depth and arsenite demobilization with decreasing borehole depth. In wells, there appear to be two arsenite groups all negatively correlated with depth (Figure 8).







Figure 8: Plot of Log of Arsenite vs. Well Depth

Figure 9 shows two groups of deep and shallow CBH and one group of shallow wells. The shallow CBH have a positive trend with Fe whilst the deeper CBH (> 60 m) have a negative trend with Fe. The wells are all positively correlated with depth (Figure 10). This suggests Fe is mobilized by oxidation of pyrite and iron-based minerals until depth of 60 m and above where the Fe begins to demobilize with the possible precipitation of pyrite or siderite in groundwater of the study area.



Figure 9: Plot of Log of Fe against CBH Depth



Figure 10: Plot of Log of Fe against Well Depth

This suggests microbial mediated aerobic activity may be responsible for dissolution of organic matter and oxidation of pyrite with a consequent mobilization of total arsenic in shallower groundwater (wells and CBH < 60 m) from the host environment. Once the arsenic is mobilized, changing conditions in the shallow wells or CBH is likely to cause the arsenic to be demobilized. It also suggests microbial anaerobic activity may be responsible for utilization of DOC and other nutrients for the demobilization of Fe through the formation of secondary minerals of Fe and mobilization of total arsenic as well as arsenite in deeper CBHs.

B. Arsenic Distribution and Stoichiometric Relations

Community boreholes and wells undergo agitations and aerations which aid the introduction of oxygen into solution. In these groundwater types the reaction will continue till all the sulphide is oxidised into solution mostly as sulphate or till the iron oxidises as ferric oxide and coats the surface of the mineral. It appears from the relationship between HCO_3^{-1} $/(Na^{+}+K^{+})$ ratio and arsenic concentration (Figure 11) that there is stronger positive correlation between silicate weathering and arsenic concentration in the order Wells>Low MBH>High MBH>CBH. The weathering of sulphides is therefore likely to generate an acidic environment for subsequent weathering of carbonate. This is confirmed by the relationship between sulphate concentration and HCO3- $/(Na^{+}+K^{+})$ in Figure 12. There is a positive linear correlation which appears to have a decreasing R^2 (Well> Low MBH > High MBH) with deeper boreholes.



Figure 11: $HCO3^{-}/(Na^{+}+K^{+})$ Ratio vs. Total Arsenic Concentration in Groundwater

The $HCO_3^{-/}(Na^++K^+)$ ratio of <1 in Figure 11 suggests shallow MBHs mobilize arsenic through silicate weathering whilst deeper MBHs and CBHs >1 suggest they are generally mobilizing arsenic through carbonate weathering. Figure 11 further suggests that shallow CBHs and wells are mobilizing arsenic through a combination of carbonate and silicate weathering. This indicates that shallow wells and CBHs are within the zone of carbonate and silicate mineralization whereas deep MBH are within the zone of silicate mineralization. This therefore proposes that the deeper the groundwater, the less silicate weathering takes place and the less the arsenic mobilization potential of the groundwater.



Figure 12: HCO3'/(Na'+K') Ratio vs. Sulphate Concentration in Wells and MBH

Figure 13 shows that generally shallow MBH, shallow CBH and deep MBH are within the $SO_4^{2-}/Ca^{2+}+Mg^{2+} < 1$ region, whilst the wells and deep CBH are in both regions. It indicates sulphate reduction is generally occurring in the MBH through the weathering of silicates and it is likely to mobilize arsenic. However, generally in deep CBH and wells, a combination of both silicate and carbonate weathering is likely to expose sulphides to be oxidized to mobilize arsenic.



Figure 13: $SO_4^{2-}/(Ca^{2+}+Mg^{2+})$ Ratio vs. Total Arsenic in Groundwater

C. Relationship between Hole Depth, Eh and Total Arsenic Most of the CBHs of the study area were weakly to strongly oxidizing with Eh values ranging between 123 mV and 511 mV. A plot of Eh against depth of CBHs (Figure 14) appear to show two main groups of reducing and oxidizing relatively deep CBHs (depth between 51 m and 100 m) with the reducing deep CBH positively correlating with the borehole depth. The oxidizing deep CBH appears to have no clear trend but split into two groups of high and medium depth. However, shallow CBHs with depth ranging between 18 m and 50 m appear to be in two groups of reducing shallow CBH correlating positively with borehole depth. Oxidizing shallow CBH correlates negatively with borehole depth (Figure 15). Although leakages from the underlying aquifer may be responsible for the high arsenic in the deeper CBHs, it is also possible that the relatively more reducing environment of the deep CBH will cause the mobilization of arsenic from arsenic sinks in the groundwater. The sealed nature of the CBH made it impossible to explore relationships between CBH water level and other parameters. Figure 14 also shows that wells with depth above 10 m had relatively lower Eh values ranging between 123 mV and 452 mV. There appears to be three groups of wells. However, the trend between Eh, well depth and total arsenic is not consistent for the wells in the study area (Figures 14 and Figure 15).





Figure 15: Plot of Total as against Hole Depth

Figure 16 to Figure 18 present plots of water level against Eh, total arsenic and HCO_3^- of the wells in the study area which appear to show two groups of wells (shallow and relatively deeper water levels). One group of wells with low water level had water level negatively correlating with Eh and HCO₃ concentration but positively correlating with total arsenic, whiles the other group with high water level positively correlated with Eh and HCO₃ but negatively with total arsenic. This is an indication that the depth of wells is not a critical factor in distinguishing between the types of wells in the study area but rather the water level should be investigated. It also suggests that at water levels of up to 3 m, aerobic microbial mediated leaching of the sulphides decrease pH gradually causing a decrease in the concentration of bicarbonates to carbonate. The oxidation process can be aided by agitation because of anthropogenic activity. It suggests the process is mobilizing arsenic from the sulphide matrix. It also suggests that at water levels of above 4 m, the process is reversed because of relatively lesser contact with the surface soils which contains more organic matter, oxygen and aerobes for microbial mediated oxidation. Anaerobic activity in these wells is therefore elevated with the possible precipitation of pyrite or siderite for demobilization of arsenic.



Figure 17: Plot of Well Water Level vs. Total Arsenic Concentration



Plot of Well Water Level vs. HCO3⁻ Concentration Figure 18:

Figure 19 appears to show three groups of wells with two groups of low and high HCO₃⁻ having a positive correlation with Eh and the third group of HCO₃⁻ having a negative correlation with Eh. Figures 20 and Figure 21 also appear to show three groups of wells with two groups of low and high total arsenic having a negative correlation with Eh and HCO₃⁻ and the third group of total arsenic having a positive correlation with Eh and HCO3. This suggests presence of aerobic and anaerobic microbial mediated activities either oxidizing or reducing the wells to decrease or increase HCO_3^{-1} content of wells respectively. The oxidation process mobilizes total arsenic whilst the reduction process demobilizes total arsenic in the wells.



Figure 20:









There is positive correlation between carbonate and silicate weathering and arsenic concentration in the order Wells > shallow MBH > deep MBH > shallow CBH > deep CBH. Shallow MBHs appear to mobilize arsenic through silicate weathering and deep MBHs and CBHs generally mobilize arsenic through carbonate weathering, whilst shallow CBHs and wells appear to mobilize arsenic through a combination of carbonate and silicate weathering. The positive influence of depth on Fe in shallow CBH and the negative influence of depth on Fe in deep CBH suggest Fe is oxidized by aerobic microbial activity of pyrite and iron based minerals in shallow groundwater until depth of 51 m and above where the Fe begins to demobilize by anaerobic activity with the possible precipitation of pyrite or siderite in groundwater of the study area. The generally weakly to strongly oxidizing Eh of CBH confirms arsenate will be predominant in most CBH of the study area. The indication of shallow CBHs with depth ranging between 18 m and 50 m showing two low total arsenic (0.011 mg/l) CBH groups of reducing and oxidizing CBH which relate positively and negatively with CBH depth respectively and a reverse of the process for deep CBH (depth between 51 m and 100 m) with relatively higher total arsenic values up to 0.166 mg/l suggests leakages into the underlying aquifer may be responsible for the high total arsenic in some deep CBHs, it is also possible that the relatively more reducing environment of the deep CBH and the demobilization of Fe will cause the mobilization of arsenic from arsenic sinks (Fe oxyhydroxides) into the CBH. The sealed nature of the CBH made it impossible to explore relationships between CBH water level and other potential factors. Figure 22 shows a conceptual model of arsenic distribution and groundwater depth in the study area.



Figure 22: Conceptual Model of Borehole Depth and Arsenic Distribution

E. Influence of Well Level on Arsenic Distribution

There is generally poor relationship between total arsenic in wells, their depth and Eh. The indication of water levels as the critical factor as opposed to depth of well in identifying wells with high arsenic concentrations arises from the suggestion that at well water levels of up to 3 m, aerobic microbial mediated leaching of available sulphide mineral occurs to decrease pH and simultaneously reduces the water in the relatively shallow wells, thereby causing a decrease in the concentration of bicarbonates to carbonate and microbial mediated oxidation with arsenic mobilization. There is another suggestion that at water levels of above 4m, the process is reversed because of relatively lesser contact with the surface soils which contains relatively more organic matter, oxygen and aerobes for microbial mediated oxidation. Anaerobic activity in these wells is therefore elevated and reduction is likely to occur with the possible precipitation of pyrite or siderite for demobilization of arsenic. Figure 23 shows a conceptual model of arsenic distribution and well water level in the study area.



Figure 23: Conceptual Model of Well Water Level and Arsenic Distribution

IV. CONCLUSION

The study concludes that, at greater depth in community boreholes (CBH > 50 m), there is a positive correlation of

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depth with total arsenic indicating total arsenic mobilization whereas in shallower depth (wells and CBH < 50 m) the correlation is not very clear suggesting total arsenic may be mobilized or demobilized depending on the best prevailing conditions. There is contribution of microbial activity in the mobilization of arsenic in the groundwater of the study area. Arsenite mobilizes with increasing borehole depth and demobilizes with decreasing borehole depth. In wells, there appear to be two arsenite groups all negatively correlated with depth. There is positive correlation between carbonate and silicate weathering and arsenic distribution in the order Wells > shallow MBH > deep MBH > shallow CBH > deep CBH. Shallow monitoring boreholes (MBH) appear to mobilize arsenic through silicate weathering and deep MBHs and CBHs generally mobilize arsenic through carbonate weathering, whilst shallow CBHs and wells appear to mobilize arsenic through a combination of carbonate and silicate weathering. There is also high probability of high concentrations of arsenic in surface water around active mining areas with inadequate layers of impermeable lithology to reduce water penetration to the groundwater in the study area. Arsenic concentrations in groundwater of these areas are likely to increase overtime because of mixing of relatively fresh water with water from the leakages and spills from the arsenic storage structures. In upstream areas with deep aquifer which is recharged by vertical infiltration of rainfall through coarse laterite, bauxite and/or carbonate media before horizontal movement of the water when it hits the impermeable barrier, the aquifer is more likely to have low concentrations of arsenic. Boreholes sited in such areas are likely to have low levels of arsenic. However, such boreholes require sealing of the annular space around their PVC pipes with clay and bentonite or cement to protect the boreholes and deep aquifer from the contaminated top layer. There is therefore the need to identify areas with suitable deep aquifers where there is adequate knowledge of the recharge mechanism and the soil profile to determine protected aquifers and decide location of new boreholes.

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