

Arsenic Leaching in Mill Tailings at the AngloGold Ashanti-Obuasi Mine, Ghana: Management of Contamination in the Related Water Environment

G. Foli¹, P. M. Nude^{2*}, C. G. Amedjoe³ and L. Kyei⁴

¹ Department of Geological Engineering, College of Engineering, Kwame Nkrumah University of Science and Technology, Kumasi, Ghana

² Department of Earth Science, University of Ghana, P. O. Box LG 58, Legon-Accra, Ghana

³ Department of Earth and Environmental Science, University for Development Studies, P. O. Box 24, Navrongo Campus, Navrongo, Ghana

⁴ Department of Statistics, University for Development Studies, P. O. Box 24, Navrongo Campus, Navrongo, Ghana

*Corresponding author; Email: pmnude@ug.edu.gh

Abstract

The study investigated arsenic (As) leaching prior to stabilization in mill tailings at the AngloGold Ashanti (AGA)-Obuasi mine in Ghana, using the toxicity characterisation leaching procedure (TCLP). Total As concentrations and pH values in TCLP leachate and related water bodies, as well as the concentration mass-time for As reduction in groundwater, were monitored. The corning pH/C107 meter and the varian atomic absorption spectrometer (AAS) were used to determine pH values and total As concentrations, respectively, in TCLP leachate and water samples. Sample blanks and matrix spikes ensured quality assurance. Statistical analysis was done using the SPSS 16.0 and stata 11.0. The range of total As concentrations and pH values in TCLP leachate were 0.45–8.73 mg/l and 4.3–8.4, respectively, while the results in the water bodies ranged from 0.04–7.23 mg/l and 5.8–8.4, in stream water; 0.01–2.62 mg/l and 5.4–8.0, and also less than 0.01 and 4.2–7.0 mg/l in the unsaturated and saturated ground water zones, respectively. The unsaturated ground water zone was established as the most convenient for As leaching study. Two-tailed 80% upper confidence level (ULC) for As in TCLP leachate was 2.45 mg/l. Arsenic was mostly mobilised within pH range of 6.3–8.2 and 5.8–7.6 in stream water and ground water, respectively. The mobile As required 26, 32, and 45 months degrading to compliance values of 0.10, 0.05 and 0.01 mg/l, respectively, in the groundwater.

Introduction

Arsenic (As) contamination of water bodies in the Obuasi mining district of Ghana which has significantly affected the socio-economic life of inhabitants of most of the surrounding villages has been reported (Actionaid, 2006). The gold ores contain high sulphide minerals composed mainly of the primary As bearing mineral, arsenopyrite (Osae *et al.*, 1995). The As in the ore decomposes to release As, which is one of the dominant trace metals in mill tailings in the Obuasi environment (Foli & Nude, 2011). Arsenic in tailing is stabilized from

the As (III) ion to As (V) in the form of complexes by oxidation during the treatment of the ores (Marsden & House, 2006). In tailings impoundments, both hydrological and geochemical factors may induce desorption of the As (V) back to As (III) state (Blowes, 1997). The mobile As may then infiltrate soil pore spaces and become discharged as contaminants into underlying aquifers, shallow water sources and, or surface streams. From the various contamination water sources, As may become available for uptake by biota.

Chronic exposure of humans and animals

to high levels of As in drinking water may lead to varieties of human health effects such as skin and several internal cancers, and also cardiovascular and neurological disorders (Tondel *et al.*, 1999; Milton *et al.*, 2001; Safiuddin & Karim, 2001; Elangovan & Chalach, 2006). In the developed world, awareness of toxic impacts of As led to the enactment of stringent regulations such as the Toxicity Characteristic Leaching Procedure (TCLP) test for arsenical waste stabilisation prior to disposal (Davis, 2001; USEPA, 1989).

In the Kwabrafo stream catchment area of the Obuasi mine environment, As contamination was identified to be anthropogenic, where considerable amount of the metal was found in soils associated with tailings (Antwi-Agyei *et al.*, 2009). As a result, environmental management plans (EMPs) can be implemented to manage the impacts, if the primary sources of contamination are identified. Some of the efforts in this regard include stoppage of pyrometallurgical pre-treatment of ores, closure of the tailing dams that are sited very close to, or encroached upon by communities, implementation of land surface remediation programmes for contaminated sites, and also containing treatment plant effluent sources. The above efforts make the environment a closed system. In addition to the enactment of stringent regulations to stabilise arsenical waste prior to disposal, concerns have also made it mandatory for major mining companies to establish internationally accepted environmental standards for managing environmental issues, such as the kind due to As contamination. Common among these standards is the International

Standards Organization series on the environment (ISO 14001), for which the Obuasi mine was certified in November 2006 (Foli *et al.*, 2010).

When establishing the ISO 14001 standards environmental management system (EMS), environmental aspects such as As contamination in water are assessed for impact significance, and rated for the risk they pose to the environment (Poder, 2006; Foli *et al.*, 2010). The risk rated environmental aspects then form the basis for the design and construction of objectives and targets policy (Anthony, 2001; ISO 14001:2004, 2008) for use in EMPs. Most environmental groups of mining companies in the developing world depend on *pH* to monitor trace metals by merely ensuring near-neutral values. This approach is, however, not conclusive for managing a metal like As (Kaye, 2005; Norris, 2005) and, therefore, requires some investigations to establish more specific methods for managing the high As impacts in the environment.

In the study, a set up of laboratory scale test, using TCLP criteria, was used to determine the status of As stabilisation in tailings waste and total As concentrations and *pH* values in leachates from mill tailings. This was followed by field monitoring of total As concentrations and *pH* values in three distinct water bodies within the closed system to establish the most convenient water medium for attenuation study, determine the most effective *pH* range for arsenic mobilisation in ground water, and also evaluate the risk of As contamination in water. Finally, the study estimated As degradation patterns in the ground water, which may be used for setting environmental

objectives and targets, according to ISO 14001 standard requirements for use in environmental management plans (EMPs).

Materials and methods

Study site

Obuasi is located between latitude $5^{\circ} 35' N$ and $5^{\circ} 65' N$ and longitude $6^{\circ} 35' N$ and $6^{\circ} 90' N$, in the southern part of the Ashanti Region of Ghana (Fig. 1). Climate is of the semi-equatorial type with a double rainfall regime while mean annual rainfall ranges between 125 and 175 mm. The mean annual temperature is $25.5^{\circ} C$ and relative humidity is about 75–80% in the wet season (Obuasi Municipality, 2009). The topography is

undulating, with hills rising up to about 250 m above sea level. Fairly fast flowing streams drain the area; streams are seasonal, drying up in the dry season and leaving stream channels with probable seepage or effluent from industrial sources. Stream flow rates vary between 0 and $2,000 m^3/h$ in dry season and between 374 and $10,454 m^3/h$ in the wet season (Foli & Nude, 2011).

The Obuasi area is underlain by the Paleo-Proterozoic Birimian rocks characterised by rich gold-bearing sulphide ore deposits (Kesse, 1985). Mining operations has been going on in the area since the 1890s (AngloGold Ashanti, 2006). Both company and illegal mining activities are the

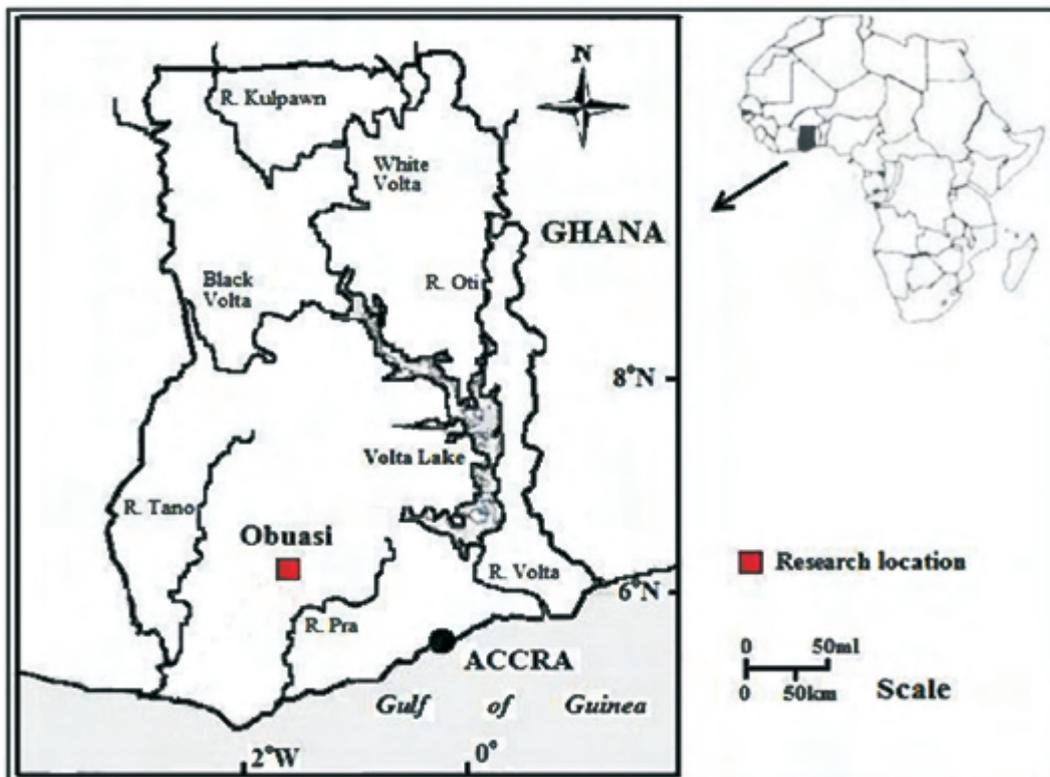


Fig. 1. A sketch map of Ghana showing the location of Obuasi

potential source of chemical contaminations in the area.

TCLP Sampling and analyses

All TCLP sampling and analytical procedures are specified in the TCLP Manual 1311 of SW-846 (1992) and the US EPA Solid Waste Sampling Guidance Manual of 1995 (US EPA, 1989). About 0.5 kg portions of tailings were scooped from tailings release points at the plant with a hand trowel. The hand trowel was washed with detergent, thoroughly flushed with distilled water and sun dried to avoid contamination. In all, 84 duplicate samples were taken on weekly basis. The paired samples were prepared by drying in an oven at 90 °C overnight, packaged into plastic bags, and labelled for analysis. The TCLP test simulates leaching by tumbling the waste in a dilute acetic acid solution for 18 h, followed by sub-micron filtration to obtain the extract for analyses; the process stabilises arsenic (III) to arsenic (V) (Baba, 2000). The pH was determined using the Corning pH/C107 meter, and the total As also determined using the Varian atomic absorption spectrometer (AAS). A sample blank and a matrix spike per 20 TCLP extractions ensured quality assurance.

For calibration purposes, the pH meter was turned on and the tip of the electrode placed in a standard buffer solution of pH 7.0 for several minutes to equilibrate at room temperature. The “Cal 2” knob was set to 100, and the “Cal 1” knob adjusted to read pH 7.0. The electrode was then removed from the buffer solution and the tip rinsed thoroughly with distilled water; excess liquid was gently blotted from the tip of the electrode with a kimwipe. The electrode was

then calibrated to pH 4 and immersed again into the buffer and the “Cal 2” knob set to display the pH 4. The electrode was then removed from the solution, rinsed with distilled water, and the tip blotted, and then used for pH determination (AGA Internal report, 1994). Statistical analysis of data was done using the SPSS 16.0 and stata 11.0.

Preliminary water monitoring

A total of 252 water samples were taken from three different sources, designated A, B and C, each of which had six representative sites; these are surface stream monitoring (SWM) sites, tailings dam monitoring boreholes (MBH) and community boreholes (CBH) (Fig. 2 and 3). The SWM source drained through series of tailings dam sites that posed site-specific local water quality problem sources in the area. The MBH sites, with depths between 30–40 m, are located at about 5–200 m from the tailings dam sites, while the CBH sites, with depths between 60–80 m, are located at about 400–800 m away from tailings dams. The sampling points were sited relative to the tailings locations and are, therefore, not regularly spaced. Sampling was done once a month, from July 2008 to August 2009. The samples were filtered through cellulose nitrate membrane filters, of pore size 0.45 µm, using vacuum filtration, and the filtrates acidified with concentrated nitric acid to pH = 2. Samples were analysed and validated as described for the TCLP data.

Concentration mass-time analysis

Based on preliminary data analysis, a follow-up sampling was done in the MBH medium for concentration-time analysis. In all, a total of 108 samples, 18 each from the

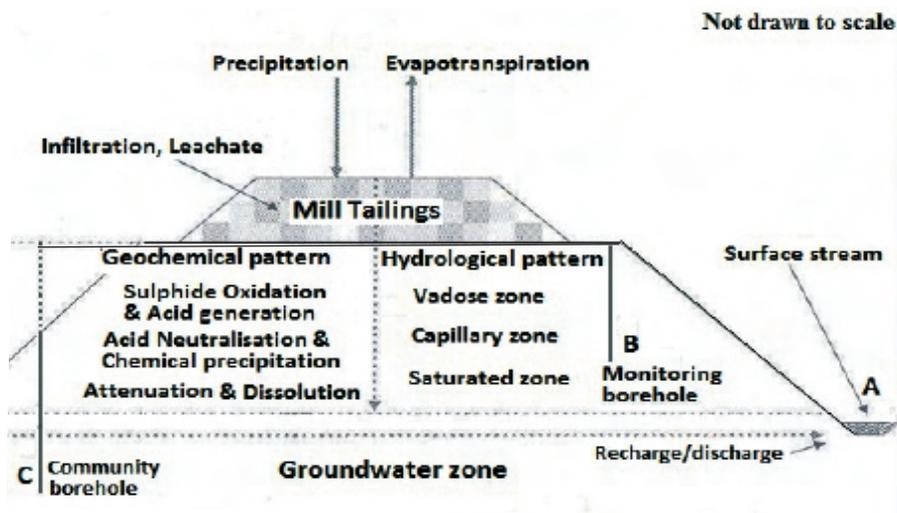


Fig. 2. Conceptualized research environment (modified after Blowes 1997; Maest *et al.* 2005)

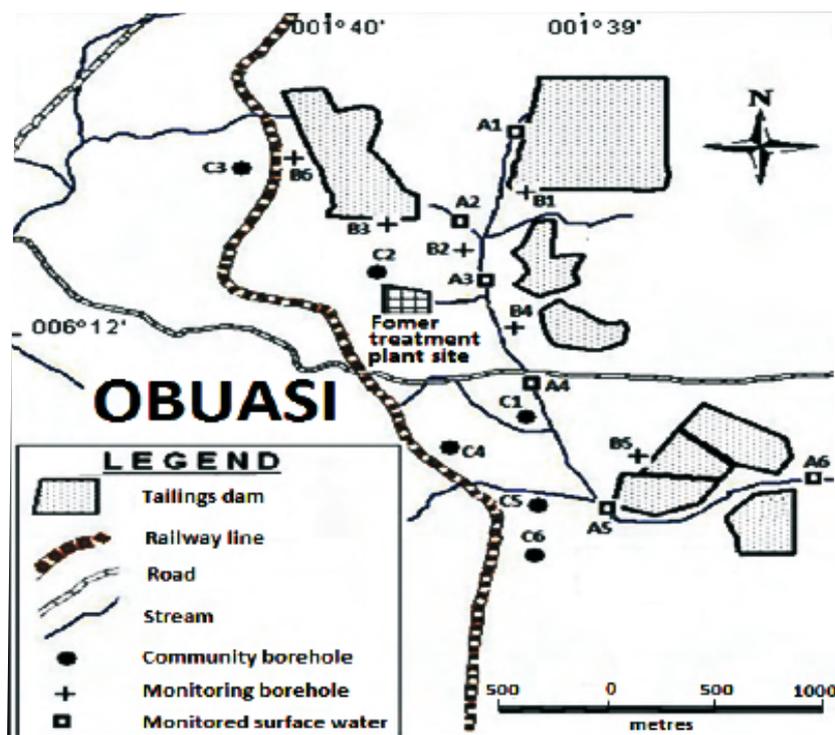


Fig. 3. Sketch map of part of Obuasi showing tailing dams, treatment plant site and sampling locations (modified after Foli & Nude, 2011)

six MBH sites, were taken over a period of 24 months. There were six missing data in the collection due to seasonal factors. Samples and statistical analyses followed the procedures for the preliminary water monitoring, while data validation followed the outline described in Azadpour-Keeley *et al.* (2001).

Risk assessment and evaluation of compliance

A modified risk assessment methodology involved a baseline review of preliminary water monitoring data to establish As contamination as a significant aspect in water. The baseline risk assessment was achieved using a four-tier criteria (AGA Internal Report, 2007). These are “can the impact due to the aspect be demonstrated; does the impact have cause of concern to interested parties; has the impact got the potential for any financial loss”; and finally, “has the impact any potential legal implication”. An affirmative response to any one of the four criteria implied that the aspect is of significance. The significant aspect is then taken through an issue-based risk assessment methodology to determine the consequence and likelihood to evaluate the degree of significance (AGA Internal Report, 2007).

The management of inorganic pollutants in the environment are based on established guideline values which are periodically evaluated for compliance. In the study, three different guideline values from different sources were quoted for As in water and used for the risk assessment. These are Ghana Environmental Protection Agency standards value of mining effluent

discharges into water bodies of 0.10 mg/l [y.] (EPA [GH], 1994); the primary maximum contamination level for drinking water of 0.05 mg/l [y.] (Wilson & Salomon, 2002) and the WHO guideline value of 0.01 mg/l [y.] for drinking water (WHO, 2010), as compliance values, were used to design consequence scenarios. Socio-economic issues, such as the availability of portable water supply for use, either directly or indirectly (Actionaid, 2006), were also used to design the likelihood scenarios. Both the consequence and likelihood scenarios were rated 1–5 for the nature of impacts (Tables 1, 2, 3) (ESIA Methodology, 2002; Broadleaf Capital International Pty Ltd, 2007). The product of consequence and likelihood determines the range of significance (Table 3).

Results and discussion

TCLP data evaluation

Relationships between pH and As, as the specific metal of concern during TCLP stabilisation of hazardous waste prior to disposal, are specified in the TCLP Manual 1311 of SW-846 (1992). The point estimates for 80% confidence interval for total As concentrations in the two batches of the TCLP samples had values (Mean \pm std err.) as 2.35 ± 0.15 mg/l and 2.15 ± 0.16 mg/l, and confidence intervals 2.15–2.55 mg/l and 1.96–2.34 mg/l, respectively. The values indicated that the deviations in As from the two batches of samples are very small, and so were pooled together (US EPA, 1989) and designated As_{TCLP} with value of 2.25 ± 0.15 mg/l and confidence interval of 2.06–2.45 mg/l. The sources of error may be due to unanticipated matrix effects, equipment malfunction, and operator error (US EPA, 1989). The pH of TCLP leachate ranged from

TABLE 1
Consequence of arsenic impacts on water resources

<i>Rank</i>	<i>Water resource usage</i>	<i>Effect on users</i>
5	Arsenic concentration higher than the EPA (GH) standards value of mining effluent discharges into water bodies of 0.10 mg/l	Permanent
4	Arsenic concentration higher than 1 ^o guideline value of 0.05 mg/l and the EPA (GH) guideline value for mine effluent discharges of 0.10 mg/l.	Long term
3	Arsenic concentration higher than WHO guideline value of 0.01 mg/l and equal to 1 ^o guideline value of 0.05 mg/l.	Short term
2	Arsenic concentration less than or equal to WHO guideline value of 0.01 m/l.	Temporary
1	No discernible Arsenic concentration in water.	No

TABLE 2
Likelihood of arsenic impacts on natural and socio-economic issues

<i>Rank</i>	<i>Definition for incidence of impact to occur upon interaction with water resource</i>
5	Necessarily
4	Very likely
3	Likely
2	Unlikely
1	Very unlikely

TABLE 3
Classification of the nature of impact on the environment

<i>Classification</i>	<i>Range</i>	<i>Definition of nature of impact</i>
High (C)	15.0-25.0	Threat to health, livelihood and total loss or degradation of social life
Medium (B)	5.1-14.9	Effects to health, disruption of livelihood pattern that may lead to high losses or degradation of social life
Low (A)	1.0-5.0	Minor effect to health and disruption of livelihood pattern that may lead to minor losses or degradation of social life

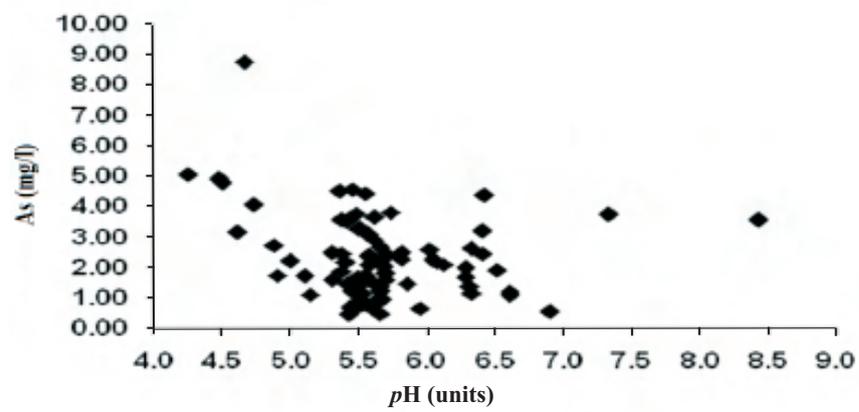


Fig. 4. Scatter plot of As-pH for TCLP leachate data

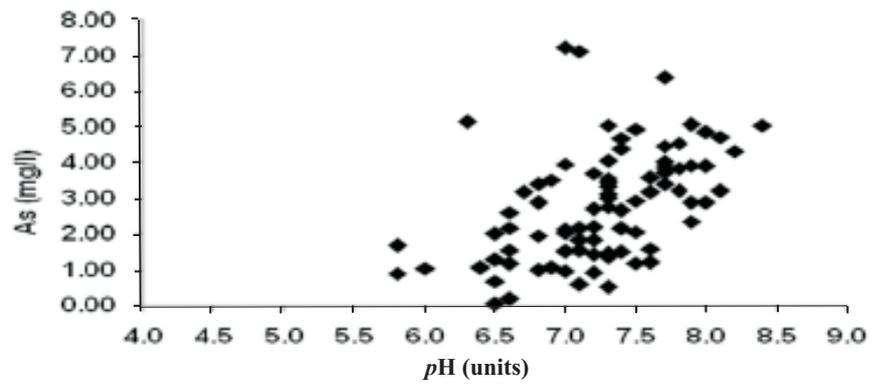


Fig. 5. Scatter plot of As-pH for MSW data

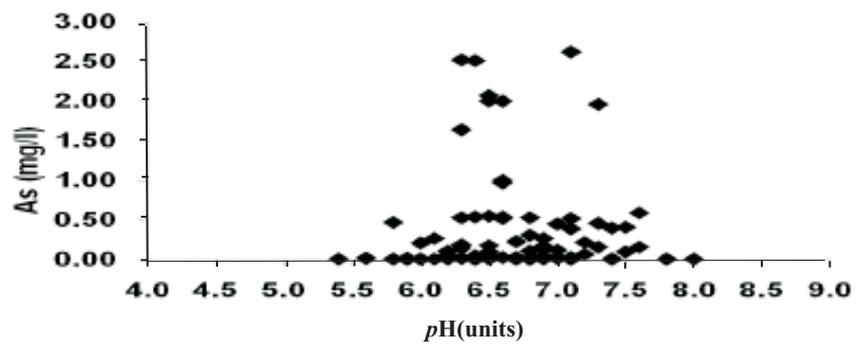


Figure 6. Scatter plot of As-pH for MBH data

4.3 to 8.4, while As_{TCLP} value and pH scatter plot is illustrated in Fig. 4. The upper confidence level of 2.45 mg/l for As_{TCLP} was below the TCLP criteria of 5.00 mg/l and, therefore, safe for disposal (Baba, 2000).

Preliminary water monitoring

From Fig. 4, there is an indication of rapid mobilisation of As into water at pH value of about 4.2 and below, while Fig. 5 and 6 clearly indicated that As was mostly mobilised within pH range of 6.3–8.2 and 5.8–7.6 in surface stream and ground water, respectively (e.g. Norris, 2005; Helm & Thorneloe, 2006). The optimal pH range for As stability in water bodies may, therefore, be inferred from about 4.2–6.3 and 4.2–5.8 in stream water and ground water, respectively (e.g. Wang & Mulligan 2006).

The range of pH values of 5.8–8.4 (mean 7.2) and 5.4–8.0 (mean 6.7) in the SWM and MBH samples, respectively, are near neutral and, therefore, justified the high mobilisation of As in both media. The near neutral pH may probably be due to buffering activity due to carbonates in the soil and tailings (Johnson, 1993; Blowes, 1997). This situation probably nullified the oxidation of sulphide minerals in tailings, which normally would have reduced pH values (Safiuddin & Karim, 2001).

The ranges of total As concentrations in SWM and MBH are 0.04–7.23 and 0.01–2.62 mg/l, both with average values of 2.82 and 0.39 mg/l, respectively, and above the primary drinking water guideline value of 0.05 mg/l. The slightly acidic pH range of 4.2–7.0 (mean, 5.7) in CBH samples probably inhibited the mobilisation of As in the CBH medium; i.e. total As concentration in CBH samples were < 0.01 mg/l. Smedley

et al. (1996) found out in the study area that groundwater sources had low total arsenic concentrations of just about 2.0 $\mu\text{g l}^{-1}$ under low ambient pH conditions of 5.4–5.8. From earlier in the text, the CBH sites, being farther away from tailings impoundments and also deeper than the MBH, probably accounted for the enhanced immobilisation or natural attenuation of As in the CBH; in line with observations by McAllister & Chiang (1994).

Test of hypotheses

T-tests were performed, at a 95% significance level of confidence, to compare the means As, with a null hypothesis that the means of As_{TCLP} is equal to the means of As_{SWM} , As_{MBH} and As_{CBH} against any of the alternatives that the means of As_{TCLP} is greater than the means of As_{SWM} , As_{MBH} and As_{CBH} . From the analysis for As_{SWM} and As_{TCLP} , the test statistic value was 0.99 compared with the table value at 0.05 significance level was -2.42. Since the table value is lower than the significance value, the null hypothesis was rejected. This implies that there is a mean difference between the two and so they are not the same. From the analysis for As_{MBH} and As_{TCLP} , the test statistic value was 0.00 compared with the table value at 0.05 significance level was 11.33. Since the table value is greater than the significance value, we fail to reject the null hypothesis. This implies that there is no difference of means among the two arsenic results. From the analysis for As_{CBH} and As_{TCLP} , the test statistic value was 0.00 compared with the table value at 0.05 significance level was 14.87. Since the table value is greater than the significance value, we fail to reject the null hypothesis that the means of As_{TCLP} is equal to the means

of As_{CBH} . This means that there is no difference of means among the two arsenic results.

From the above analyses, the TCLP leachate, MBH and CBH media are analogous. Jurjovec *et al.* (2002) showed that column experiments, involving metal attenuation processes under controlled laboratory conditions, possibly eliminate unknown influences that could affect the geochemical evolution of pore water. These unknown influences may have induced adsorption of the As(III) to As(V) state in the ground water. The ground water environment is, thus, shown to be the most convenient medium for monitoring attenuation processes of As in the study.

Risk assessment

The consequence for arsenic impacts in the environment is rated 5, based on the average values of 2.82 and 0.39 mg/l in SWM and MBH, respectively. These concentrations, therefore, will cause permanent effect on the users of water from such sources. Since sources of water are likely to be abused, there is the high likelihood that arsenic contamination will have socio-economic impacts. From Table 2, the likelihood of impact ranged between 3 and 5 rating. The nature of impact of As contamination will, therefore, be a threat to health, livelihood and degradation of social life (Table 3).

Concentration mass-time analysis

Averages of arsenic values from the data ranged from an initial value of 2.52 mg/l to the final low of 0.11 mg/l in 24 months, and with standard error 0.16, which is relatively low as compared with the individual

concentration values. Linear regression analysis indicated that the correlation coefficient between the arsenic concentration and time is 0.7967, which implied that 80% of the data demonstrated statistical significance at 95% level of confidence.

Also, for the same 95% level of significance, the spread of the data is observed to be between 0.45 and 1.135. A *t-test* conducted showed the model to be significant at *p-value* of 0.000; the null hypothesis was, therefore, not rejected, establishing that there is a relationship between arsenic concentration and the months. Missing data of arsenic concentrations in some months, also, has no effect on the validity of the sample data. The data are, thus, statistically valid. The relationship between the arsenic concentration and time indicated an exponential pattern (Fig. 7) with formula as indicated below;

$$y = 2.5203e^{-0.123x}$$

where *y* represents concentration (mg/l) and *x* represents time (months).

From the model equation above, the time frame over which the water may become usable, relative to the compliance values y_1 , y_2 and y_3 , will, thus, be;

$$x = - \frac{\ln \left(\frac{y}{2.503} \right)}{0.123}$$

Substituting y_1 , y_2 , and $y_3 = 0.10$ mg/l, 0.05 mg/l and 0.01 mg/l returned corresponding values x_1 , x_2 and $x_3 = 26.18$, 31.81 and 44.90 months, respectively. The above quantities, therefore, constitute the overall environmental goals and specific task which can be translated into detailed EMPs.

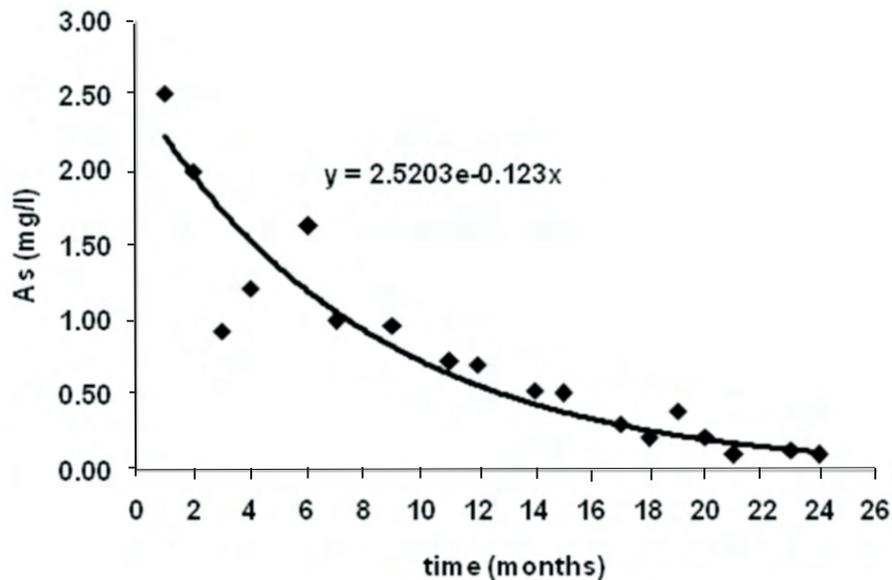


Fig. 7. Curve for mean arsenic concentration mass-time analysis

Conclusion

The point estimates for 80% confidence interval for As_{TCLP} , having the upper confidence level of 2.45 mg/l, which is below the TCLP criteria of 5.00 mg/l, indicates that the groundwater zone is analogous to the TCLP leachate medium and thus the most convenient medium for attenuation. The optimal pH zone for arsenic mobilisation is 5.8–8.2, and for a closed system, As required about 26, 32 and 45 months respectively to degrade to guideline values of 0.10 mg/l, 0.05 mg/l and 0.01 mg/l respectively in groundwater. These quantities may serve as relevant parameters and targets for the design of EMPs for the management of As impacts on water bodies.

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