The occurrence and geochemistry of arsenic in groundwaters of the Newark basin of Pennsylvania

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Abstract

Elevated As concentrations in groundwater in the eastern United States have been recognized predominantly in the accretionary geologic terranes of northern New England. A retrospective examination of more than 18,000 existing groundwater samples from the Pennsylvania Department of Environmental Protection (PA DEP) Drinking Water and Sampling Information System database indicates that elevated groundwater As concentrations occur throughout the northern half of the Piedmont Province of Pennsylvania. Chemical analyses of 53 samples collected in 2005 from drinking water wells in this area all had detectable As, and 23% of these samples contained elevated (>133 nmol/L or >10 μg/L) concentrations of As.

Elevated concentrations of As in the groundwater samples were most common in the Mesozoic sedimentary strata composed of sandstone and red mudstone with interbedded gray shale, and gray to black siltstone and shale. Arsenic was typically not elevated in groundwater of diabase intrusions of the Newark Basin or in crystalline and calcareous aquifers to the north of the Newark Basin. Geochemical parameters such as pH and oxidation–reduction potential can indicate mobility mechanisms of As in some regions. In this area, measured groundwater conditions were predominantly oxidizing (Eh > +50 mV), and more than 85% of samples contained arsenate as the dominant As species. Variations in pH were strongly correlated to the As concentration, with highest As concentrations observed at pH values greater than 6.4. The original source of As is most likely the black and gray shales that contain some arsenian pyrite with groundwater concentrations likely to be controlled by adsorption/desorption reactions with Fe oxides in the red mudstone aquifer materials.

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1. Introduction

1.1. Arsenic geochemistry

Arsenic is a ubiquitous trace element found throughout the environment. Arsenic concentrations in groundwater vary greatly due to the heterogeneous distribution of source materials and subsequent geochemical controls on aqueous As mobility in aquifers (Cullen and Reimer, 1989). The causes of elevated As concentrations in groundwater, including the complex interactions between water, geologic substrate and biological processes, are not yet completely understood. Recent work around the globe, in such locations as West Bengal (Nickson et al., 2000), Bangladesh (Ahmed et al.,...
Elevated As in groundwater has been identified within the United States in the West and SW (Savage et al., 2000; Welch and Lico, 1998), upper Midwest (Schreiber et al., 2000), and New England (Lipfert et al., 2006; Peters and Blum, 2003; Peters et al., 1999). Dissolution and desorption of As from naturally occurring As-containing minerals, geothermal water, and mining activity appear to be the key contributors to high-As groundwater provinces within the United States.

Arsenic is present in the environment in both inorganic and methylated forms, though the inorganic species are considered to be the most prevalent in groundwater (Cullen and Reimer, 1989; Welch et al., 2000). Inorganic forms are common in As-containing minerals (O’Day, 2003; Thornburg and Sahai, 2004), sorbed on amorphous Fe(III) oxyhydroxides (Raven et al., 1998; Wilkie and Hering, 1996), sorbed on crystalline Fe oxide phases (Manning et al., 1998), as surface precipitates on sulfides or pyrite (Bostick and Fendorf, 2003), and as discrete nanoparticulate phases (Utsunomiya et al., 2003). Widespread occurrences of elevated As concentrations can be derived from oxidation of sulfide minerals, particularly trace-substituted pyrite, arsenian pyrite, and arsenopyrite, and also from desorption of As from mineral surfaces, or dissolution of the minerals with adsorbed As (Welch et al., 2000). In the case of As desorption from Fe oxide minerals, redox conditions, pH, solid-to-solution ratios, specific surface area of minerals, and competing ions such as phosphate may affect As mobility and thus As concentrations in surrounding waters e.g., (Dixit and Hering, 2003; Peters et al., 2006).

Typical geochemical parameters that may indicate desorption from mineral surfaces include elevated Eh (>100 mV), dissolved O2 (0.1–0.2 mmol/L and greater), pH (>8), and alkalinity (5–8 mmol/L and greater), and possibly high dissolved organic C (>1 mmol C/L) (Smedley and Kinniburgh, 2002).

The harmful health effects related to the consumption of elevated As in drinking water are well documented in groundwater provinces with As concentrations in excess of 667 nmol/L (50 µg/L) (Chowdhury et al., 2000; Morales et al., 2000; Smith et al., 2000). Health effects that may occur at lower concentrations have prompted the evaluation of large regions where groundwater As concentrations do not appear to be as significant, though are exacerbated when combined with other high risk activities, such as smoking (Karagas, 2002; Karagas et al., 2004).

1.2. Local geologic setting

The state of Pennsylvania, USA, consists of five physiographic provinces, the Central Lowlands, Appalachian Plateaus, Ridge and Valley, the New England Province, and the Piedmont (dashed lines, Fig. 1). The detailed field study area is located in the Newark Basin within the Piedmont physiographic province. This physiographic province is composed primarily of Upper Triassic and Lower Jurassic units that filled a half graben coincident with Mesozoic rifting. This portion of the failed rift zone is locally called the Newark Basin and the rocks primarily consist of red sandstone, shale, siltstone and conglomerate with igneous diabase intrusions (Berg et al., 1986). Local lithologic units include playa-lacustrine deposits named the Passaic formation that laterally transition into lacustrine deposits called the Lockatong formation. Although they do not outcrop in the study area, fluvial deposits of the Stockton Formation interfinger with and are overlain by Lockatong deposits to the south of the study area.

The Passaic Formation consists of quartzose sandstone grading into red mudstone, with some interbedded gray shale and argillite whereas Lockatong deposits are dark siltstones, shales and argillites (Low et al., 2000). The diabase sheets are quartz normative continental tholeiites of the York Haven type (Lyttle and Epstein, 1987; Smith et al., 1975). Units of gray mudstone, argillite, red siltstone, and red mudstone are metamorphosed to hornfels near the diabase intrusions (Froelich and Gottfried, 1999). Cambrian limestone conglomerate, quartzite conglomerate and Precambrian gneiss occur to the north of the rift basin, but still within the field area.
The hydrologic properties of the sedimentary rocks of the Newark Basin create thin aquifers separated by thicker aquitards, while the crystalline bedrock aquifers are considerably more complex, anisotropic and heterogeneous (Low et al., 2000). Water-bearing zones in the crystalline rocks tend to be located in the weathered zones near the ground surface and through fractures and joints at depth (Sloto and Schreffler, 1994). Previous studies of the aqueous geochemistry in this area indicate that waters are neutral to slightly basic in the Lockatong Formation whereas water is neutral to slightly acidic in the Passaic Formation and diabase, and elevated concentrations of Fe, Mn and SO₄ occur but are not common throughout the aquifers (Low et al., 2000).

A comprehensive analysis of the extent, levels, and causes of naturally occurring As enrichment has not been conducted in Pennsylvania, USA. The present research occurs at two scales, and with two intents. At the regional scale, a GIS analysis was conducted using existing groundwater quality data from state and federal databases (PA DEP, 2004; USGS, 2001) to highlight areas within the state that are more likely to have elevated As. Based on these data, a field study was then conducted at a local scale to focus on As concentration, speciation and potential mobilization mechanisms. The two primary intents of this paper are to highlight the importance of using existing data to guide research efforts and to evaluate field evidence for experimentally observed As geochemical phenomena. This study, which uses southeastern Pennsylvania as a natural laboratory, has specific applicability locally, but also potentially within other Mesozoic rift basins along the Atlantic Ocean margin.

2. Methods

2.1. Statewide database and GIS analysis

A GIS database was prepared to analyze the relationship between As occurrence, geography, and bedrock geology in Pennsylvania to determine an
appropriate field study location (Fig. 1). The main criteria used to determine an appropriate location were evidence of both low (<133 nmol/L, 10 µg/L) and elevated (>133 nmol/L) As concentrations in the groundwater within a single region. Test results for As concentration in drinking water samples were obtained from the Pennsylvania Department of Environmental Protection (PA DEP) Drinking Water and Sampling Information System for 1994 through 2004 (PA DEP, 2004), and the USGS National Water Information System (USGS, 2001). Data entry errors in the PA DEP database required extensive manual review and correction before analysis of the geochemical data.

2.2. Field methods

Fifty-three water samples were collected (July–October, 2005) in southeastern Pennsylvania (Fig. 2) from 49 private homes, 2 schools and one municipal government building using groundwater wells as their source of drinking water, and one hillside spring used by local residents who fill their own containers. Groundwater wells were screened in the following lithostratigraphic units: Passaic Formation, Lockatong Formation, diabase, Leithsville Formation, felsic to mafic gneiss, and hornblende gneiss. Well and spring locations were collected with a Garmin GPSMAP 76S handheld GPS unit. Temperature, pH (Corning 3-in-1 Combination IP67) and Eh (Hanna HI 98201) were determined in the field using a continuous flow through cell. The probes were calibrated daily with 4.0, 7.0 and +468 and +220 mV standards for the pH probe and Eh probe.

Groundwater samples were collected from wells using outside taps after thoroughly flushing the tap for at least 15 min or until temperature measurements indicated that all onsite storage was purged and water originated from the well bore or aquifer (~11–14 °C). All sampling containers were rinsed three times with sample water before collecting the final sample using a syringe and a 0.45 µm polypropylene filter (Whatman PP Puradisc). Samples for total As, major anion and cation analysis were collected in acid washed LDPE bottles. Samples for major cation analysis were fixed to pH 2 with double-distilled ultrapure HNO₃. Samples for As speciation analysis were collected in 40 mL cleaned glass vials with Teflon-lined caps with no headspace. All samples were kept on ice and in the dark until analyses were complete.

2.3. Analytical methods

Within 24 h, groundwater samples were analyzed for As speciation using continuous-flow coupled ion chromatography (IC, Dionex IONPAC AG4A Guard and AS4A Analytical Columns), hydride generation (HG, in-house design) with ultraviolet oxidation and inductively coupled plasma-mass spectrometry (ICP-MS, Thermo X-Series CCT) generally following the method of Klaue and Blum (1999). Arsenic species were separated on a strong anion exchange column using the standard eluent of 1.8 mM Na₂CO₃ and 1.9 mM NaHCO₃ at a flow rate of 1 mL/min and ~75 bar pressure. The eluent containing separated As species was mixed online with a solution of 1 M HNO₃ and allowed to react for ~10 s before mixing with a solution of 1% NaBH₄ (w/v) stabilized in 0.1 M NaOH (Klaue and Blum, 1999). Cross calibrations using single species and total As standards were used to verify the column separation and the performance of the entire analytical apparatus. Samples were also analyzed for total As using hydride generation with ultraviolet oxidation coupled to ICP-MS. The analytical detection limit was approximately 0.1 nmol/L total As concentration, which is lower than all sample concentrations. Arsenic concentrations are generally reported as nmol/L, and where applicable, breaks between categories correspond to US regulatory concentrations (e.g. 133 nmol/L = 10 µg/L). Major cations and other trace elements were determined using ICP-MS with pneumatic nebulization and anions were determined using IC with suppressed conductivity detection.

3. Results

3.1. Statewide survey

Groundwater samples (n = 18,277) submitted to state testing agencies were compiled, entered into a GIS, and plotted in Fig. 1. Concentrations range from below detection to 1.1 µmol/L with a median value that was below detection. All the PA DEP drinking water sample results from within a given geologic formation were averaged and assigned to that formation wherever it occurs (Fig. 2). Areas with no shading have As concentrations less than 66 nmol/L. Dark gray and black regions have average As concentrations greater than 66 nmol/L, and in some cases greater than 133 nmol/L. This map
Fig. 2. Map of geologic units coded by arsenic concentration (upper). Field area is indicated by the box. Map of geologic units in field study area, including drinking water arsenic concentrations (lower).
<table>
<thead>
<tr>
<th>#</th>
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suggests the occurrence of elevated concentrations of As in three geographic areas: sandstone aquifers in both the NE and east-central regions of Pennsylvania, and in mudstone and shale aquifers in the SE region of the state. The main criteria used to determine an appropriate location for a detailed field investigation were evidence of both low (<133 nmol/L) and elevated (>133 nmol/L) As concentrations in the groundwater within a single region. From these three regions, the SE corner was chosen as the study area because it met the criteria for the presence of elevated concentrations of As and was also located close to the Lehigh University geochemical laboratory where time-sensitive chemical analyses could be conducted.

### 3.2. Field survey

Arsenic was detected in all 53 samples from the study area, and concentrations ranged from 0.16 nmol/L up to 864 nmol/L with a median value of 38 nmol/L (Tables 1 and 2). Approximately 23% of all groundwater samples contained elevated (>133 nmol/L) concentrations of total As. When examined in the context of aquifer host material, 33% of groundwater samples from wells within two sedimentary formations, the Lockatong and the Passaic, of the Newark Basin contained elevated concentrations of total As. The average and maximum As concentration in water samples from wells within the Lockatong Formation is greater than surrounding formations, followed by the Passaic Formation (Table 2). Samples from wells located in the Precambrian gneisses and Cambrian conglomerate rocks in the northern part of the study area did not contain elevated As. Arsenic was also not elevated in the diabase aquifers except for one sample (ID# 48) located in the northeastern part of the study area near the contact between the diabase and the Passaic Formation.

The statistical distribution of As concentrations in the detailed study area (Table 2) compares favorably with summary data from the same geologic units (Table 3) within the Pennsylvania Drinking Water Information System database drinking water dataset (PA DEP, 2004). In both datasets, samples from the Passaic and Lockatong formations contained the highest concentrations of As. In both datasets, the percentage of samples with elevated As concentrations are similar for all formations except for the Lockatong Formation, which is due to a large number of nondetect results in the PA DEP dataset.
3.3. Geochemical indicators of arsenic release

In this study, elevated total As concentrations occur generally at circumneutral to high pH (Fig. 3a). All samples less than pH 6.4 contained less than 27 nmol/L total As, while samples above pH 6.4 contained up to 894 nmol/L. This transitional zone is noted on Fig. 3a by a dashed line. None of the samples collected as part of this study had a measured pH greater than 7.5. Measured Eh values of the groundwater samples ranged from $\pm 232$ mV to $+456$ mV with most of the samples having values in the $+25$ mV to $+300$ mV range. The behavior of Fe in this system may help explain some of the As data due to the close association between Fe and As in mineral form, and the ability of Fe-oxides to adsorb As (Smedley and Kinniburgh, 2002). Iron concentrations range from below detection (<0.02 µmol/L) up to 10.5 µmol/L (Fig. 3b). The lowest concentrations of Fe occur throughout the range of measured pH values. The highest concentrations of Fe occur at pH $\sim 5.8$ and decrease with increasing pH. The pH dependence of dissolved Fe concentrations and oxidation/precipitation reactions has been well characterized (Stumm and Morgan, 1996). In this system, the stability of Fe minerals can be represented by an activity diagram of dissolved Fe vs. pH at varying Eh values (Fig. 4). According to this diagram, samples from this area plot near the boundary of Fe oxyhydroxides, suggesting precipitation is likely to occur if pH values increase or O$_2$ becomes more available. The precipitation of Fe-oxyhydroxides may affect As concentrations by providing substrate for As adsorption (Pichler et al., 1999). Dissolved Fe concentrations do not appear to be directly correlated to As, and the highest concentrations of As occur at a wide range of Fe concentrations (Fig. 3c). Interestingly, the sample containing the highest concentration of Fe contains very low As.

Sulfate concentrations range from 43 to 5400 µmol/L, with a median value of 260 µmol/L. Iron and SO$_4$ in water samples do not appear to have a direct dissolution relationship (Fig. 5a), suggesting either Fe sulfide dissolution followed by Fe oxide precipitation or non-Fe sources of SO$_4$, such as gypsum. Gypsum has not been reported in the field area, and is unlikely to explain the excess of the SO$_4^{2-}$.
Total As appears inversely correlated with both NO$_3$ and Cl, with high values of As occurring at low values of these two ions (Fig. 5b and c). Relationships did not exist between total As concentration and F, SO$_4$, Mg, Ca, Na or K ($R^2$ range of 0.001–0.1431, not shown). Nitrite was not detected in enough samples to determine a relationship, though the few samples that did contain detectable NO$_2$ did not have different ranges of As concentrations than those that did not ($p > 0.20$). Phosphate was not detected in any samples (detection limit of 5 μmol/L). Manganese was only detected in 8 samples; however, 4 of the 8 samples also contained the 4 highest concentrations of As.

### 3.4. Arsenic speciation

Approximately 70% of groundwater samples from the study region contained mostly (>80%) the oxidized As species arsenate. Slightly more than 3% of the samples contained mostly arsenite, and the remaining samples had an intermediate mix of 20–80% of both species. Of the 12 samples with elevated concentrations of total As, 10 are mostly arsenate while the remaining two samples were mostly arsenite. Arsenate-dominated samples occur throughout the pH range of the samples while the arsenite-dominated samples occur within a smaller pH range (pH 5.8–7.2). Not surprisingly, the proportion of arsenate is generally positively related to Eh ($R^2 = 0.383$), with arsenate-dominated samples generally occurring at high Eh. An Eh-pH diagram was constructed of the Fe–As–SO$_4$–H$_2$O system using the software Geochemist’s Workbench to evaluate the aqueous and mineral thermodynamic stabilities of the system (Fig. 6). This study’s data were plotted in the context of the model according to measured Eh and pH. Samples dominated by arsenate plot within the arsenate stability fields indicating that the As species in these samples are at or close to equilibrium. Samples with intermediate compositions of each species (20–80%) plot within both arsenate and arsenite stability fields, but occur close to the boundary between these species. A few samples plot in stability fields opposite to their measured speciation and indicate that either they are not in equilibrium with their environment, or that the measurement technique for Eh did not accurately record the redox conditions. The sample that plots within the orpiment stability field is likely due to the inaccurate measurement of Eh that is not surprising when using a probe-type system.

### 4. Discussion

The prevalence of As in this groundwater system is generally consistent with a natural source of As whose concentrations are moderated by exchange reactions between the aqueous and solid phases. In the following paragraphs, the relationship between this study and other examinations of As behavior in both the laboratory and field environments are discussed.

In regions where it is unclear whether As is derived from natural or anthropogenic sources, geochemical indicators may help identify potential anthropogenic sources. For example, high
concentrations of Cl might indicate landfill leachate or road salt (Christensen et al., 1994; Fritz et al., 1994; Panno et al., 2006) and NO₃ could be due to agricultural fertilizer infiltration (Hamilton and Helser, 1995). It would be expected that if water samples contained high concentrations of these two compounds (or other indicator compounds), that the As in the groundwater may be related to the processes that contribute these indicator elements. Arsenic concentrations in groundwater from this site show either inverse or no correlation with either of these parameters (Fig. 5b and c), suggesting that As is derived from natural sources. Theoretically, if As were due to mineral dissolution and possibly adsorption/desorption reactions, then evidence of these processes should be present in the water chemistry. Arsenic concentrations appear dependent on a threshold value of pH, above which elevated As is present in groundwater, and below which As concentrations remain low. The moderately oxidizing conditions demonstrated by the dominance of arsenate and measured Eh values greater than 100–200 mV along with the observed pH dependence suggest that the controlling mechanism is likely desorption from mineral surfaces (e.g., Peters and Blum, 2003). At low concentrations of aqueous Fe²⁺ in solution, the adsorption and desorption of As anions from Fe-oxyhydroxide minerals is strongly controlled by pH-controlled changes in surface affinity of available mineral substrate as observed by others (Smedley et al., 2005). In some areas where a few results indicate weakly oxidizing to strongly reducing conditions, reductive desorption or primary sulfide dissolution may be possible as localized mechanisms. Some support for reducing mobilization triggers exists in water samples that have intermediate proportions of each As species, and those that plot in opposing stability fields suggest an ongoing redox transformation or possibly mixing with waters of different redox status (Peters and Blum, 2003).

Work conducted by Serfes (2005) in the same geologic formations in the New Jersey portion of the Newark Basin is consistent with desorption pro-
cesses leading to the presence of aqueous As in waters of the Passaic Formation; clay minerals coated with early hematite were determined to likely be the source of the As via desorption (Serfes, 2005; Serfes et al., 2005). Sulfide dissolution mechanisms were predicted in the black shales of the Lockatong Formation (Serfes, 2005; Serfes et al., 2005). Although not common, reducing conditions were found with elevated concentrations of As in some samples from this study. Samples with As concentrations dominated by arsenite were generally found in both the Passaic and Lockatong formations. Although oxidizing to mildly reducing conditions present in all the Lockatong samples (Table 1; Eh range 17–112 mV) may support oxidative dissolution of pyrite, other geochemical factors measured in this study are less supportive. For example, SO$_4$ is far more abundant in solution relative to Fe. Molar ratios close to 2:1 to 1:1 for SO$_4$ and Fe would be expected from pyrite or arsenopyrite oxidation and subsequent congruent dissolution in the mildly reducing conditions, especially at neutral to low pH. Actual ratios range from a minimum SO$_4$:Fe of 6:1 to a maximum of more than 3600:1 and median of 120:1. Sulfate and Ca in solution do not correlate and therefore gypsum dissolution is unlikely to explain the elevated SO$_4$. Excess SO$_4$ likely remains in solution from the primary dissolution of pyrite during the evolution of groundwater whereas the Fe is precipitated as amorphous and crystalline forms of Fe-oxyhydroxides. A considerable amount of Fe could easily have precipitated after initial sulfide dissolution, but there is no conclusive evidence that pyrite dissolution delivered the initial As to these groundwaters, and it would be difficult to conceive a study that would deliver definitive proof. Comparing the concentrations of SO$_4$ and As to the possible source materials can test the feasibility of As being derived from the dissolution of arsenian pyrite. For the sample containing the highest As (865 nmol/L) and lowest SO$_4$ (260 μmol/L) concentrations, arsenian pyrite of approximately 0.3% As (mol/mol) is needed. This calculation assumes congruent dissolution and conservative transport, but illustrates that very little arsenian pyrite is needed to explain the observed concentrations of As in groundwater.

The strength of arsenate adsorption does not differ between more crystalline forms of Fe-oxyhydroxide minerals such as goethite and magnetite to less crystalline forms such as hydrous ferric oxides (HFO); however, more crystalline forms exhibit an overall decrease in specific surface area and therefore site density (Dixit and Hering, 2003). Therefore, a conversion from less crystalline forms to more crystalline forms could result in decreased As adsorption and increased As mobility. In the case of precipitation of aqueous Fe to amorphous forms of Fe-oxyhydroxides, the newly formed Fe minerals may adsorb more As from solution relative to the surrounding crystalline Fe minerals. Over time, as these amorphous phases mature into more crystalline phases, As may be slowly released as adsorption site density decreases.

A tradeoff exists at higher pH values between an increase of the precipitation rate of Fe oxyhydroxides and the decrease in the affinity of the
precipitated substrate for As. At low pH Fe-oxyhydroxides are stable in a solid state and have a positively-charged surface that readily attracts As anions. As the pH increases through the circumneutral range, the net Fe precipitation rate increases but the surface charge of the mineral substrate becomes less positive thereby decreasing the total affinity of the Fe-oxyhydroxide surface for As anions. The presence of As in solution at both circumneutral and high pH values may be related to a combination of the availability of precipitated Fe substrates at circumneutral pH and possibly the adsorption properties of Fe minerals for As at higher pH values. This study’s results were plotted in the context of thermodynamic modeling of Fe activity under different redox conditions using Geochemist’s Workbench (Peters and Blum, 2003), forms of Fe present (Raven et al., 1998), surface structure attributes of Fe (Goldberg and Johnston, 2001; Sherman and Randall, 2003), and speciated forms of As (Dixit and Hering, 2003) simultaneously affect As mobility in the groundwater.

5. Conclusions

Geographic analysis of an existing drinking water quality database has proven to be a useful tool to

Fig. 6. Eh–pH diagram of the Fe–As–SO₄–H₂O system. Stability fields modeled using Geochemist’s Workbench (Bethke, 2000) and the LLNL thermodynamic dataset V8 R6. As concentration fixed at 133 nmol/L for modeling purposes. Fe concentration fixed at 180 nmol/L. SO₄ concentration fixed at 312 µmol/L. Temperature fixed at 13 °C. Dashed line indicates lower bound of water stability. This study’s data are plotted by measured Eh and pH according to different percentages of arsenate (As(V)) measured in solution Open circles contain less than 20% As(V). Open squares contain between 20% and 80% As(V). Solid triangles contain more than 80% As(V). n = 45.
identify a region for further study with elevated concentrations of As in the groundwater. While developing a research program utilizing this approach, researchers should be aware of the sensitive nature of drinking water source locations, and the presence of erroneous results due to data entry errors.

The presence of elevated As in groundwaters of the Newark Basin, Pennsylvania, USA, is primarily due to desorption from Fe oxide minerals under oxidizing conditions at pH values greater than 6.4. The original source of the As may be from As-bearing Fe sulfide minerals in the shales of the Lockatong Formation and interbedded shales of the Passaic Formation (Serfes, 2005). The specific geologic observations from this study may be useful in directing efforts to understand As mobility in other Mesozoic rift basins along the Atlantic Coast.

Considerable emphasis has been placed on studying reducing conditions in Quaternary river delta environments (e.g. Bangladesh) because of the gravity and magnitude of human health impacts; however, aquifers located in older and more crystalline geologic environments can also have elevated As concentrations in groundwater, although the range of concentrations is considerably less than concentrations reported from Bangladesh and geologically similar areas. With the implementation of more stringent As standards in drinking water in the United States, there is also a need to examine As mobility in groundwater environments previously considered to be low risk.

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