

Report as of FY2006 for 2006AR125B: "Metal Mobilization, Especially Arsenic, Related to Ground-water Level Fluctuation in the Alluvial Aquifer"

Publications

- Conference Proceedings:
 - Sharif, M.U., B. Kim, R.K. Davis, K.F. Steele, T.M. Kresse, J.A. Fazio, 2006, Lithologic and recharge controls on the spatial variability of arsenic in the Mississippi River Valley Alluvial aquifer, southeastern, Arkansas. Proceedings of the Geological Society of America, Annual Meeting, Philadelphia, PA, October 2006 (poster).
 - Kim, B., M.U. Sharif, T.M. Kresse, J.A. Fazio, J. Cothren, R.K. Davis, K.F. Steele, 2006, Comparison of potentiometric maps produced by densely spaced measurement points and USGS 7.5' manual elevation picks for the alluvial aquifer of eastern Arkansas, Proceedings of the Geological Society of America Annual Meeting, Philadelphia, PA, October 2006 (poster).
- Other Publications:
 - Sharif, M.U., R.K. Davis, K.F. Steele, B. Kim, T.M. Kresse, J.A. Fazio, 2007, Distribution and variability of redox zones controlling spatial variability of arsenic in the Mississippi River Valley alluvial aquifer, southeastern Arkansas, Arkansas Water Resources Center Annual Conference., April 24-25, 2007 (oral presentation)[CD].
- Articles in Refereed Scientific Journals:
 - Sharif, M.U., R.K. Davis, K.F. Steele, B. Kim, T.M. Kresse and J.A. Fazio, 2007, Distribution and variability of redox zones as control of spatial variability of arsenic in the Mississippi River Valley alluvial aquifer, southeastern Arkansas, USA (submitted to Contaminant Hydrology).
 - Sharif, M.U., R.K. Davis, K.F. Steele, B. Kim, T.M. Kresse and J.A. Fazio, 2007, Surface complexation modeling using PHREEQC for predicting solid-phase arsenic concentrations in the sediments of the Mississippi River Valley alluvial aquifer, Arkansas, USA(submitted to Aqueous Geochemistry)
 - Sharif, M.U., R.K. Davis, K.F. Steele, B. Kim, T.M. Kresse and J.A. Fazio, 2007, Inverse geochemical modeling of groundwater evolution with emphasis on arsenic in the Mississippi River Valley alluvial aquifer, Arkansas (submitted to Journal of Hydrology)
- Dissertations:
 - Kim, B. In Preparation (expected completion December 2007) Hydrogeochemical Evolution of Ground Water in an Intensively Pumped Alluvial Aquifer. Ph.D Dissertation, University of Arkansas, Fayetteville
 - Sharif, M.U., In Preparation (expected completion December 2007) Flow and Hydrogeochemical Evolution of Arsenic in Groundwater: Tracking Sources and Sinks in the Mississippi River Alluvial Aquifer Southeastern Arkansas, USA. Ph.D. Dissertation, University of Arkansas

Report Follows

Title: Metal Mobilization, Especially Arsenic, Related to Ground-water Level Fluctuation in the Alluvial Aquifer

Statement of Regional or State Water Problem

Kresse and Fazio (2002, 2003) reported that about 18 % of wells in the Bayou Bartholomew watershed in eastern Arkansas region (Figure 1) exceed the EPA drinking water maximum contamination level (MCL) of 10 ppb arsenic. Chronic exposure to low levels of arsenic can affect the skin, liver, kidney, circulatory systems, gastrointestinal tract, nervous system, and heart (NRC, 1999). Twenty one out of 118 irrigation water wells completed in shallow Quaternary alluvial deposits (alluvial aquifer) with depths of 25-30 m had arsenic concentrations exceeding 10 $\mu\text{g/L}$. These wells also had high iron concentrations (median 10 mg/L and maximum of 43 mg/L). Thus, 18 % of wells should not be used anymore as a drinking water source without treatment and it is highly unlikely that private domestic wells will obtain treatment. Arsenic data from two wells that were bored with a rotary drill and drilling mud indicate variable concentrations in the sediment. In one well arsenic concentrations of about 1000-2000 $\mu\text{g/kg}$ occur below 12 m of the surface but concentrations of only 75 $\mu\text{g/kg}$ are encountered in the production zone (coarse sand and gravel 40-43 m). Another well produced values about 300 - 500 $\mu\text{g/kg}$ (including the production zone at 30 m) except at the 12 m depth where the concentration was about 150 $\mu\text{g/kg}$. Although the drilling method is not appropriate for obtaining complete cores, the results nonetheless indicate significant and variable arsenic concentrations in the aquifer sediments.

In addition, since 1975, rapid increases of ground-water withdrawal for irrigation has occurred in eastern Arkansas. The total withdrawals of ground water in the State of Arkansas were about 2596 Mgal/day in 1975 (Bryant et al., 1985), but it increased about 6952.25 Mgal/day in 2000 (Holland, 2004). Most ground-water withdrawals of Arkansas State are in eastern Arkansas and for agricultural and industrial usage (Bryant et al., 1985; Holland, 2004; Joseph, 1999). It is reported that the ground-water level has declined more than 10 ft since the 1960s in some parts of the Mississippi River Valley region including Cross, Jackson, Jefferson, Lee, Lonoke, Monroe, Prairie, and Woodruff counties (Joseph, 1999). Some farmers' interviews reveal that in the growing season, daily ground-water level fluctuation as well as annual fluctuation was significant level due to the intensive ground-water pumping in eastern Arkansas, i.e. in the growing season.

The investigation of the relationship between reduction-oxidation condition in ground water and metal mobilization (Stumm and Morgan, 1996) indicate heavy metals precipitate under oxic conditions or are sorbed on hydroxyoxides that can be leached or dissolved under reducing condition (Pandey et al., 2002). Considering the high iron concentration in the wells with high arsenic values, the probable process in eastern Arkansas is "oxyhydroxides reduction" (Pandey et al., 2002). Fiedler and Sommer (2004) state that ground-water level variation caused oxidation-reduction potential energy (E_h) change. The linear correlation between E_h and ground-water level indicated increased iron mobilization with increasing duration of saturation in the soil. These research results indicate that

ground-water fluctuation in eastern Arkansas can accelerate metal mobilization from the sediment as a result of the accompanied changes in pH and Eh. In addition ground-water level fluctuation can change ground-water flow direction. These mobilized metals ultimately can be transported to surface water resources. In summary, the arsenic problem of eastern Arkansas alluvial aquifer appears be related to the oxidation-reduction conditions of the aquifer and its sediment characteristics. However there is little research on the water quality and/or geochemical processes associated with intensive ground-water withdrawals causing significant ground-water level fluctuation which can decrease ground-water quality. This proposed project will provide useful information on the influence of ground-water level fluctuation to the water quality in the alluvial aquifer. The mechanism for releasing the metals from the sediment is not easy to investigate in the field. Appropriate lab-scale column experiments can provide fundamental information on the relationship between the sediment and water chemistry with ground-water level fluctuation. Characteristics of sediment determined by analyses of acid-extractions and hydrogeologic parameters can be input into physico-chemical models. The results from these models can be used to estimate potential amount of heavy metals mobilized from the sediment. This will allow us to manage water resources properly and secure the best drinking water resources.

Statement of Results or Benefits

Lab-scale column experiment can be easily handled and allows us the ability to examine more diverse conditions than is possible in the field. The results can provide very important information in understanding ground-water quality and the relationship between ground-water level fluctuations caused by intensive ground-water usage and metal mobilization causing ground-water quality problems. Moreover, the differences from the comparison of chemical speciation modeling and actual measured data by the column experiment in the laboratory will provide fundamental data to setup an integrated capacity of metal mobilization index. The mobilization index must include critical characteristics of sediments in the region, as well as chemical constituents which can be modeled. In addition the index can be used to calibrate chemical speciation modeling to estimate ground water heavy metal concentrations.

Monitoring ground-water level and hydrochemical parameters can provide important information to investigate the relationship between ground-water level fluctuation caused by intensive ground-water withdrawals and metal mobilization mechanism from the aquifer sediment associated with decrease of ground-water quality including arsenic mobilization. Also, understanding the distribution of arsenic and other potentially harmful elements (e.g. Pb and Cd) in geologic materials, i.e. sediment, is critical to understanding their influence on surface water and ground water. This knowledge is also important for defining geochemical baselines for potentially harmful elements so that any future perturbations caused by human influence or natural processes may be recognized and measured.

This proposed project will be the first detailed study of the influence of ground-water level fluctuation in a major alluvial aquifer with relatively low arsenic concentrations and no obvious contamination source. Because of these attributes, the results of this study

should have wide application to large areas of the Mississippi River Valley alluvial aquifer. Information about sources of metals i.e. arsenic and geochemical processes in the alluvial aquifer should be considered important in developing a safe and sustainable water supply, and ultimately, a technically feasible and cost effective water resources development and management system. Also, although the emphasis of the project is arsenic mobilization, geochemical evolution of the aquifer sediment along the flow path associated with ground-water quality will also provide useful information on the behavior of both major and other trace ions (e.g., Fe, Mn, Cu, Pb, Cd, Zn) in alluvial aquifers.

The results of the project will be applicable to the remainder of the Mississippi River Valley alluvial aquifer, especially in Louisiana, Mississippi, Tennessee, and Missouri. The area comprises the Mississippi Delta Region of the central U.S. The Delta has been identified by both the Clinton and Bush administrations as being the most impoverished, least developed region of the Nation with a tremendous need for growth that will be dependent upon region resources, as such the Delta can little bear development of problematic ground-water level decrease and fluctuation potentially causing acceleration arsenic mobilization from the sediment and contaminated water without a clear understanding of the problem and how to best manage and make use of regional ground-water resources. Much of the data will also be of value in the study of other alluvial aquifers around the world. A better understanding of the influence of intensive ground-water usage associated with metal mobilization mechanism from the aquifer sediments to ground water and geochemical evolution of the aquifer. It can be used to assess the potential risk of ground-water over-usage related to the use of ground water for a drinking water source.

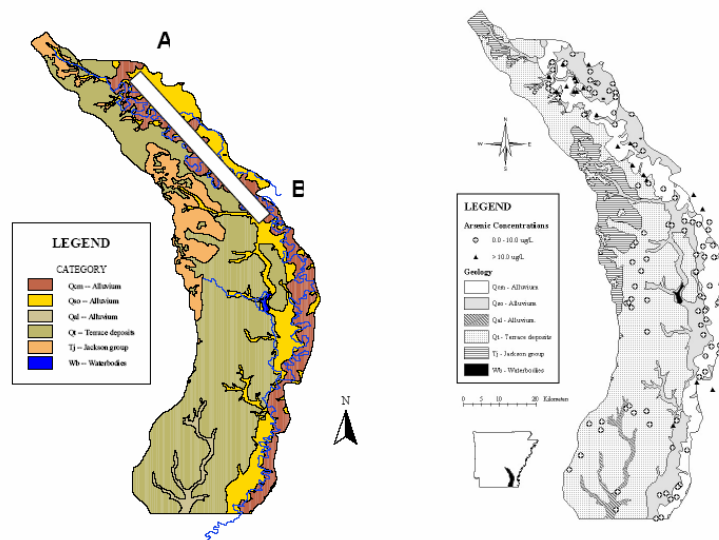


Figure 1. Geological map of the study area (left) and plot of As concentrations from alluvial aquifer (right) wells. Inset of Arkansas shows the location of Bayou Bartholomew in the state. Line A-B is the proposed transect for the monitoring wells.

Nature, Scope, and Objectives of the Project (Timeline)

This proposed project will study the influence of ground-water level fluctuation in hydrogeochemistry and hydrogeology i.e. heavy metal mobilization mechanism from the alluvial aquifer sediments. The decrease in ground-water quality due to heavy metal mobilization will be investigated also. Through the lab-scale column experiment with the alluvial aquifer sediments under various ground-water level and hydrologic condition will be conducted with focusing heavy metals, i.e. arsenic. Sediment acid-extraction analyses and physico-chemical modeling will be conducted for studying mobilization along the flow path in both aerial and vertical dimensions within the alluvial aquifer in southeastern Arkansas with the following objectives.

Objective 1. Investigate the relation between ground-water level fluctuation and metal mobilization mechanism, i.e. arsenic in an alluvial aquifer hydrogeochemical environments (soils, sediments, ground water, and their interaction). This investigation includes lab-scale column experiment to study the influence of ground-water level fluctuation on metal mobilization mechanism from the alluvial aquifer sediments under the various oxidation-reduction conditions.

Objective 2. Determine the ground-water flow pattern and its relationship to hydrochemical evolution of the ground-water chemistry, fate of arsenic in the ground water, and potential amount of heavy metals mobilized from sediments by using physico-chemical model. Steele et al. (2004, 2003a) have demonstrated that significant chemical variation can occur in the alluvial aquifer and Kresse and Fazio. (2003, 2002) have shown that some mineral precipitation (e.g., calcite) occurs. A goal then is to construct a mobilization index to assist the prediction of heavy metal concentrations in the alluvial aquifer.

Also, this study will evaluate three conceptual models of arsenic mobilization and release mechanisms, which are a sulfur oxidation model (Acharyya, 1997; Pearse, 1995; Sinha Roy, 1997), iron oxyhydroxide reduction model (Matisoff, 1982; Korte, 1991; BGS, 1999; Nickson et al. 1997, 1998, 2000), and competitive ion exchange model (Acharyya et al., 2000) by interpreting chemical analyses of sediment and ground water, and results of model simulations, and column tests.

Ultimately, real-time ground water level monitoring and associated water quality will be needed. Unfortunately only preliminary data will be possible for this project due to the limited funding. Samples and water levels for the three monitoring wells will be collected during a “dry” period and during a “wet” period.

Project Schedule

Table 1. Activity Time Line

Research Activity (Task)	'06. 3	4	5	6	7	8	9	10	11	12	'07. 1	2
Identify Monitoring site, field data collecting & sampling sediment												
Lab analysis of sample including sequential acid extraction												
Column test and field monitoring data collecting												
Data compilation and interpretation												
Physical and chemical modeling												
Preparation of final report												

Methods, Procedures and Facilities

For the current project, the direction of ground-water flow in the aquifer in the Bayou Bartholomew watershed is dominantly southward with the slope of the land within the watershed (Kresse and Fazio, 2002). So, the transect of the research area (drilling locations) will be oriented in a general NW-SE direction to ensure that the wells will be oriented on the same flow path. Sediments from one of these wells will be used for the lab-column experiments.

Parameters to be studied are ground-water chemistry including major and minor cations and anions and other physico-chemical parameters including dissolved oxygen (DO), redox potential (Eh), pH, temperature (T). Sequential acid extractions for investigating sediment characteristics and potential metal mobilization capacity will be conducted. The extractions for chemical forms and leaching agents will be as a) acid soluble form (mainly fixed in carbonates), which will be leached using 0.1 M acetic acid; b) reducible form (fixed in Fe- and/or Mn-oxides), which will be leached using 0.1 M hydroxylamine hydrochloride solution, adjusting pH to 2 using HNO₃ acid; c) organic form, which will be first decomposed by 0.1 M sodium pyrophosphate solution heated on a hot plate and dissolved into 1 M ammonium acetate solution; and d) insoluble form (fixed mainly in sulfide and rarely in silicate minerals), which will be dissolved by a mixture of concentrated HNO₃ and HClO₄ acid if appropriate ventilation equipment is available to work with the HClO₄. Some hydrogeologic parameters including porosity, hydraulic conductivity and isotopic analysis will be conducted to characterize alluvial aquifer hydrogeology.

Drilling and Sampling Operations

The wells for this project will be shared with the ongoing 104 B project. The wells are drilled along a flow path of ground water, which is based on highest arsenic concentration reported by Kresse and Fazio (2002). Hollow-stem augers equipped with PVC core liners or other drilling methods that do not require drilling fluids will be used to drill this wells. Optimum depth of the wells will be to the base of the

alluvial aquifer (about 46 m). Sediment samples will be collected at a preset regular interval to cover the range of aquifer heterogeneity from the drilling process. The collected sediment will be used for lab-scale column experiment and acid-extraction.

Quality assurance and quality-control measures will be followed at every step to ensure data quality. Methods, sampling, and analytical protocols of USGS's National Water-Quality Assessment (NAWQA) programs will be followed (<http://water.usgs.gov/nawqa/>).

Column Test

The most important interactions to control ground-water quality are sediment-water interaction in the subsurface environment (Kim et al., 2004; Steele et al., 2004). To understand the mechanism of metal mobilization by changing oxidation-reduction condition in the aquifer caused by ground-water level fluctuation and to identify controlling factors of sediment-water and surface-ground water interactions, column test with field collected sediments will be conducted. The experimental equipment is illustrated at figure 2.

The parameters to be measured in the column experiment will be pH, dissolved oxygen (DO), oxidation-reduction potential (ORP), alkalinity, total organic carbon (TOC), dissolved organic carbon (DOC), total dissolved solids (TDS), conductance, Ca, Mg, Na, K, HCO₃, Cl, SO₄, Fe, Mn, NH₃, NO₃, PO₄, As, Cr, and temperature under various water level and flow conditions.

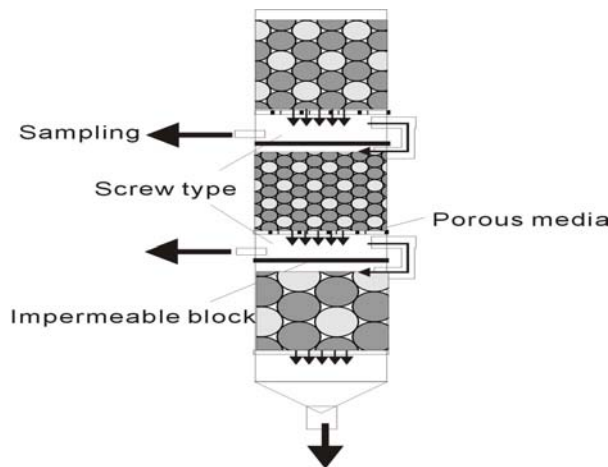


Figure 2. Diagram of column experiment equipment. Acrylic tubing will be used to make the column. The inner diameter of column is about 5 in, and it has 1 in diameter sampling port at each stage, and 1.5 in diameter transport tube which is linked with next stage. Each stage has a porous media block to prevent sediment loss, and impermeable layer to separate water from each sediment layer. The location of transport port should be higher than sampling port to give sufficient gradient to hold water sample at the sampling area. Each column stage can be assembled by screw type linking system to give flexibility of experimental multi-stage and diverse condition design.

Physico-Chemical Modeling and Data Analysis

A three-dimensional finite-difference ground-water flow model (MODFLOW) (McDonald and Harbough, 1996) and/or integrated hydrologic modeling of surface water and ground water will be used to simulate flow path and ground-water level change in growing season. The modeling results will provide information on mobilized metal i.e. arsenic transport and potential risk of water resource development. The program PHREEQC (Parkhurst, 1995) has been extensively used to model evolution of ground water chemistry along a flow path. Results of chemical analyses of water and sediment collected from the monitoring wells, as well as the results of the column experiments, will be used as input into the speciation module of the PHREEQC program. The model will provide speciation and solubilities of mineral phases and their relation to arsenic mobility in the aquifer.

Related Research

Fiedler and Sommer (2004) investigated the relationship between ground-water level and redox potential (Eh) and its influence in iron mobilization in a wetland. The result showed that ground-water level and Eh were linearly correlated ($r^2=0.88$), and indicated increased iron mobilization with increasing duration of saturation. The research was conducted in order to study wetland soil redoximorphic features, so they did not consider water quality and metal mobilization mechanism. However, the result clearly show that ground-water level fluctuation affected redox potential causing metal mobilization conditions.

Davis et al. (1994) used speciation and adsorption modeling (MINTEQA2) using data from an historical tanning and rendering site at Woburn, Massachusetts. At this site, ground water was contaminated by arsenic and chromium. They defined three distinct redox zones with a reduced core of the plume close to the hide piles, with dissolved sulfide, high dissolved iron and ammonium. An intermediate zone of occurrence further down gradient with sporadic sulfide, high dissolved iron and intermediate concentrations of ammonium. Finally a peripheral zone of the plume was oxidized, with detectable concentration of oxygen and precipitation of iron hydroxide. Modeling was used to determine saturation indices, which indicated undersaturation of ground water with respect to all arsenic mineral phases. This result was comparable with electron microprobe analysis indicating no arsenic precipitated in the solid phase. The adsorption model used the calculated amount of iron hydroxide as a quantity of adsorbent in a diffuse double layer model to quantify arsenic adsorption, which was based on the same principle followed by PHREEQC. The modeling results indicated very efficient removal of arsenic from solution. The results indicated soil water interaction and chemical modeling usage for investigating metal mobilization mechanism under various hydrogeochemical conditions. However, solute transport with reflecting hydrogeologic factors and ground-water level fluctuation affecting redox potential were not considered.

My current 104 B project is also focusing on arsenic mobilization mechanism in the ground water but there are limited sediment-water interaction data. This proposed project is focusing on the interaction between sediment and ground-water level fluctuation and the resulting oxidation reduction condition variation by using lab-scale column test. The results from this study will provide better data for the PHREEQC geochemical model. The improved data will allow the construction of a mobilization index for the heavy metals in the aquifer.

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Training Potential

The project will directly involve one Ph.D. student in ENDY Ph.D program. Data, field methods, and laboratory techniques used in this project will be incorporated into course content for graduate level courses in Hydrogeology, Geochemistry, Hydrochemical Method and Hydrologic Modeling, which are currently taught by the principal investigator and other Geoscience faculties.

Investigator Qualifications

Dr. Ken Steele has had wide experience in water studies in Arkansas and has conducted several projects on the alluvial and Sparta aquifers. He also has extensive experience in supervising graduate research assistants. See attached resume.

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Professional Preparation

University of North Carolina, Chapel Hill, Chemistry B.S. 1966
University of North Carolina, Chapel Hill, Geology Ph.D. 1971

Appointments

1983 – Present: Professor, University of Arkansas
1988 – 2001: Director of Arkansas Water Resources Center, University of Arkansas
1979 – 1981: Coordinator of Advising, College of Arts and Sciences, Uni. of Arkansas
1970 – 1983: Instructor, Assistant Professor, Associate Professor,

Selected Publications—closely related

- Kim, Burmshik and Kenneth F. **Steele**, 2005, Application of geologic information for ground-water quality management, Proceedings of the XXXI International Hydrologic Resources Congress, Seoul, Korea, p.5577-5585.
- Kim, Burmshik, Kenneth F. **Steele**, and Todd Fugitt, submitted and invited, Comparison of dissolved and acid-extractable metal concentrations of ground water, eastern Arkansas, U.S.A., Journal of Environmental Informatics.
- Kim, B., K. **Steele** and T. Fugitt, 2004. Comparison of Dissolved and Acid-Extractable Metal Concentrations of Ground Water, Eastern Arkansas, USA, Environmental Informatics Archives, Vol. 2, p. 272-284.
- Kresse, T.M., Fazio, J.A., Hays, P.D., **Steele, K.F.**, Davis, R.K. and J.V. Brahana, submitted, Source and Release Mechanisms for Arsenic in the Alluvial Aquifer, Southeastern Arkansas, Applied Geochemistry.
- Kim, B., **K. Steele** and T. Fugitt, 2004. Comparison of Dissolved and Acid-Extractable Metal Concentrations of Ground Water, Eastern Arkansas, USA, Environmental Informatics Archives, Vol. 2, p. 272-284.
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Publications—significant

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Synergistic Activities

This project will allow me to continue exploration of the geochemical regimes in the alluvial aquifer and the factors controlling element mobility, especially As. The proposed research will compliment my teaching (undergraduate and graduate courses) where I lecture on the major aquifers in Arkansas and assign hydrochemical projects. The knowledge gained from the project will enhance the quality of my service as reviewer for journals, e.g. *Environmental Quality and Ground Water*, and reviewer and panelist for National Science Foundation, U.S. Department of Agriculture, U.S. EPA and Natural Environment Research Council (United Kingdom). Expansion of collaborative research with the Arkansas Department of Environmental Quality and the Arkansas Soil and Water Conservation Commission will additional out comes from this project.