Assessing the Source for Arsenic in Groundwater, North Carolina Piedmont

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A—Burning pyre which is composed of lead ore with wood placed above it.
B—Workman throwing ore into another area.
C—Oven-shaped furnace.
D—Openings through which the smoke escapes.
Assessing the Source for Arsenic in Groundwater, North Carolina Piedmont

- Problem overview
- Manganese-iron coating studies – Pasour Mountain area, Gaston Co., North Carolina
- Drill core studies in the Piedmont of North Carolina
- Emerging areas of interest and future work
- Perspectives and interim conclusions
Arsenic – some background in North Carolina
Arsenic

Problem:
• Water samples from domestic water supply wells revealed the presence of arsenic (As) contaminated wells; As source unknown

Observations:
• Sulfide minerals in cores noted from areas with elevated arsenic
Arsenic

Objectives and methods:
• Help determine the arsenic source
• Major and trace element chemistry of core
• Reflected light microscope, SEM and probe
• Studies of naturally occurring manganese-iron coated ‘boulders’ and time-integrated Mn-Fe coating accumulation rates (also As and other elements on ceramic streak plates)
Arsenic

Focus:
• Collaborative investigation with NC Aquifer Protection Section at Pasour Mtn. study site, Gaston Co.

Goal:
• Improve the understanding of naturally occurring arsenic contaminated groundwater in the Piedmont
EPA Standard is 0.01 mg/L

NC health based MCL is 0.02 ug/L
Gold Prospect joined to Arsenic Sample Data

Symbols signifies concentration (mg/L) of closest arsenic sample.

- ○ 0.0000 - 0.0010
- ▲ 0.0011 - 0.0100
- ■ 0.0101 - 0.0250
- ⬤ 0.0251 - 0.0500

Arsenic Occurrence

- Non-detect
- Detect

Miles
Acidic conditions from sulphide oxidation lead to engineering problems and to geochemical releases.
Arsenic – further distribution information in North Carolina
Arsenic basics

- 20\textsuperscript{th} most abundant in the Earth’s crust
- Arsenopyrite (FeAsS), realgar and orpiment are most common mineral species (latter two uncommon in the East)
- May be trace component in more common sulphide minerals (pyrite, etc.)
Form and speciation

Understanding the form and speciation of arsenic is important as arsenic (III) (arsenite) is more toxic than As(V) to humans. In general, arsenate is more stable in oxygenated water while arsenite generally occurs in reducing water (see Eh / pH diagram right). However, the relative distribution of As (III) and As(V) is far from thermodynamic equilibrium due to microbial activity, differential absorption, presence of oxidants and reductants, and slow abiotic kinetics.
Arsenic adsorption to iron oxyhydroxides is strongly influenced by pH, redox potential, and presence of competing anions.

A number of factors are involved in the adsorption-desorption reactions. These include changes in redox potential and microbial-mediated reductive dissolution of iron hydroxides.
Gold Prospect joined to Arsenic Sample Data

Symbols signifies concentration (mg/L) of closest arsenic sample.

- ○ 0.0000 - 0.0010
- ▲ 0.0011 - 0.0100
- ■ 0.0101 - 0.0250
- ○ 0.0251 - 0.0500

Probability for Groundwater to Exceed 0.001 mg/L

- Blue: 1.00
- Light Blue: 0.75 - 0.99
- Light Green: 0.50 - 0.74
- Yellow: 0.25 - 0.49

0 25 50 100 150 Miles
Manganese – iron coating studies: Pasour Mountain area, Gaston County, North Carolina
Manganese – iron coatings

- Common features (pebbles, boulders, outcrops)
- Precipitates geochemically significant because of their concentration of Fe, Mn, Cu, Co, Ni, Ba, and other elements
- Universally present in fast flowing oxygenated streams with plentiful water
- Confined to rock exposed in flowing water and stop abruptly at the sediment-water interface
- This is an geochemical exploration tool – here applied to health-related studies and groundwater
Manganese – iron coatings (cont’d)

• Artificial substrates (streak plates) placed in streams 19 January 2004 at Pasour Mt., NC
• Coatings collected quarterly, over one year
• Natural coatings collected from same sites
Gold Prospect joined to Arsenic Sample Data

Symbols signifies concentration (mg/L) of closest arsenic sample.

○ 0.0000 - 0.0010

▲ 0.0011 - 0.0100

■ 0.0101 - 0.0250

● 0.0251 - 0.0500

Arsenic Occurrence

- Nondetect
- Detect

0 25 50 100 150 Miles
Mn-Fe coating extraction

(Left) Filtered and brought up to volume ➔ ICP analysis

Coated ‘boulder’

Nitric – hydrochloride extraction on heat plate

Coated streak plate - one month

Coated streak plate - one year

Post extraction – streak plate (left) - ‘boulder’ (above)

Process flow
<table>
<thead>
<tr>
<th>Samples #</th>
<th>Description</th>
<th>As</th>
<th>Cu</th>
<th>Fe</th>
<th>Mn</th>
<th>Pb</th>
<th>Zn</th>
<th>As accumulation rate (ppm)</th>
<th>ug/cm²</th>
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<tr>
<td></td>
<td></td>
<td>ug/L</td>
<td>ug/L</td>
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<td>10U</td>
<td>12</td>
<td>50U</td>
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<td>16</td>
<td>2,900</td>
<td>310</td>
<td>10U</td>
<td>57</td>
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<td>Tine Mine Road (TM-1)</td>
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<td>1,000</td>
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<td>1,400</td>
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<td>Tine Mine Road (TM-2)</td>
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<td>1,200,000</td>
<td>510</td>
<td>17,000</td>
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</table>

U = Indicates that the analyte was analyzed for but not detected above the reported practical quantitation limit*. The number value reported with the "U" qualifier is equal to the laboratory's practical quantitation limit*.

*PQL = The Practical Quantitation Limit (PQL) is defined and proposed as "the lowest level achievable among laboratories within specified limits during routine laboratory operation". The PQL is about three to five times the calculated Method Detection Lim
Arsenic – streak plates

Arsenic concentration increase over one year in ug/cm²
Metal Concentrations from Oxides Grown on Ceramic Tiles Location 2

Days Deployed vs Concentration (µg/cm²) for:
- As Normalized to tile area
- Cu Normalized to tile area
- Fe Normalized to tile area
- Mn Normalized to tile area
- Zn Normalized to tile area
Metal Concentrations from Oxides Grown on Ceramic Tiles Location 3

Days Deployed vs. Concentration μg/cm²

As Normalized to tile area
Cu Normalized to tile area
Fe Normalized to tile area
Mn Normalized to tile area
Zn Normalized to tile area
Metal Concentrations from Oxides Grown on Ceramic Tiles Location 4

Days Deployed

Concentration \( \text{u/cm}^2 \)

<table>
<thead>
<tr>
<th></th>
<th>As Normalized to tile area</th>
<th>Cu Normalized to tile area</th>
<th>Fe Normalized to tile area</th>
<th>Mn Normalized to tile area</th>
<th>Zn Normalized to tile area</th>
</tr>
</thead>
</table>
Boulder coatings – Interim conclusions

• Boulder coating and streak plate component of study. Arsenopyrite, a primary ore of arsenic, is present at the Long Mine area
• Arsenic occurs in significant levels in boulder coatings on the site in drainages crossing old gold mining areas at Pasour Mountain
• Associated base metals, iron and manganese are also present in high concentrations in boulder coatings
• Arsenic accumulated to measurable amounts on unglazed streak plates in about 60 days during cold, winter months
• Arsenic concentration appears greater in streams draining old gold prospects on the property
• Arsenic is present in fracture coatings in core from oxidized zones
• Arsenic is also present in significant levels in boulder coatings from the Tin Mine area, Lincoln Co., North Carolina
• Associated base metals, iron and manganese are also present in high concentrations in boulder coatings from this area
Drill core studies: North Carolina Piedmont
About the drill cores examined

• The two following geologic maps show the drill hole core examined from correlative metavolcanic units to the Lake Tillery area and to the stratigraphically underlying metarhyolite. The drill hole cores were from previous gold exploration.

• The upper map that shows drill hole core from the strike equivalents of the metavolcanic rocks in which the wells of the Lake Tillery area of arsenic concern are located. These drill holes are located some miles to the northwest as there are no other cores in this metasedimentary interval in the NCGS repository. Metavolcanic (predominantly very siliceous, rhyolitic rocks) core the antiform across Lake Tillery (also a regional arsenic stream sediment high) and arsenic problem area for domestic water wells.

• These rocks contain small amounts of gold. Pyrite is ubiquitous. Arsenopyrite, a chief host of arsenic, was reported in available drill logs. The depth of groundwater movement can be approximated by the depth of altered rock and iron oxide forming from the primary minerals, and from iron oxide coatings on fractures.
Drill hole locations shown on the Geologic Map of the Charlotte 1 x 2 Degree Quadrangle, North Carolina and South Carolina, 1988
Drill hole locations shown on the Geologic Map of the Charlotte 1 x 2 Degree Quadrangle, North Carolina and South Carolina, 1988
Cores from ‘oxidized’ zone display iron-manganese filled fractures, and a general breakdown of the framework silicate minerals (e.g., feldspar). Manganese ‘blooms’ are frequently observed. In contrast, the ‘reduced’ rocks appear competent and sulfide minerals generally are retained.
Sample locations in core hole DV-C-2-76 indicated by yellow box labeled with core hole depth in feet. Sample taken from interval immediately to the right of the yellow box. Depth of core samples increase from left to right within each divided section of the box. Core in box was subsampled from original core hole and does not represent continuous samples with depth.
Sample locations in core hole DV-C-1-76 indicated by yellow box labeled with core hole depth in feet. Sample taken from interval immediately to the right of the yellow box. Depth of core samples increase from left to right within each divided section of the box. Core in box was subsampled from original core hole and does not represent continuous samples with depth. Fe-oxides are present in the sampled intervals on exposed planar surfaces in the core.
Core Hole DV-C2-XX

Box 1  12 ft - 163 ft

Box 2  173 ft - 316 ft

Box 3  376 ft - 498 ft

Sample locations in core hole DV-C2-XX indicated by yellow box labeled with core hole depth in feet. Sample taken from interval immediately to the right of the yellow box. Reference footage labels shown with white labels. Depth of core samples increase from left to right within each divided section of the box. Note Fe-oxide staining below (to right) of 36.3-37 ft sample location. Core in box was subsampled from original core hole and does not represent continuous samples with depth.
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<thead>
<tr>
<th>NCGS repository number</th>
<th>Drill hole number</th>
<th>Rock type</th>
<th>County</th>
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<td>DV-C-1-76</td>
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<td>Davidson Co.</td>
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<td>MN-C-03-89</td>
<td>UW-89-3</td>
<td>Metarhyolite</td>
<td>Montgomery Co.</td>
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</tbody>
</table>
Cores – Interim conclusions

- Arsenic occurs in arsenopyrite; other primary arsenic minerals have not been identified
- Arsenic is present in fracture coatings in core from oxidized zones
- Whole rock arsenic chemical analyses range from 1.5 ppm to 223 ppm (ICP-based method)
- Groundwater has interacted with the metavolcanic rocks resulting in a weathering profile
- The weathering profile is characterized by generally degraded primary framework silicate minerals – many altered to clay
- Primary sulfide minerals have been destroyed resulting in iron and manganese filled fractures in which the primary sulfide minerals were located.
- Manganese and iron ‘blooms,’ recording the destruction of primary sulfide minerals, are common.
Cores – Interim conclusions (continued)

- Manganese and iron ‘blooms,’ recording the destruction of primary sulfide minerals, are common.

- While the data set is small (n=25), paired t-test of ‘extractable arsenic’ vs. arsenic by total rock chemical destruction indicates a statistical difference (P-value of .1640 for a hypothesized difference =0). This suggests arsenic mobility in the near surface environment. The unpaired t-test data, by ‘facies’ shows substantially different arsenic content, by facies, by each of the two arsenic analytical methods.

- Limited reconnaissance geochemical data suggests arsenic mobility in the oxidized ‘facies’ relative to major elements (framework mineral elements) and to transition base metals.
Outreach and education

- County health directors and interested parties – Mooresville region – summer 2004
- County health directors and interested parties in Raleigh region – September 2004
- Groundwater Professionals of North Carolina – December 2004
- Periodic news inquiries
- Aquifer Protection Section and Groundwater Planning Unit of DWQ, State Toxicologist
- National Groundwater Association – naturally occurring contaminants conference (As, U, radon, radium) – February 2005
- Interest group on coast – May 2005 (tentative)
Emerging areas for future studies: North Carolina Piedmont
Developing arsenic problem areas

Courtesy R. Bolick – Aquifer Protection Section – 2005
with contributions by Dr. Ken Rudo – 2005
Interim conclusions and perspectives for future work
Perspectives and future work

• As groundwater discharges into a stream the water moves from an oxygen poor environment to an oxygen rich environment which promotes the precipitation oxyhydroxides (i.e. the black coatings on boulders in streams or the orange red ring in the bathtub and toilet bowl).
• Arsenic is known to have an affinity for iron and manganese oxyhydroxides.
• Preliminary results indicate that arsenic is being incorporated into the formation of oxyhydroxides.
• This has implications for domestic wells and may explain part of the puzzle.
• In theory, water cascading into a domestic well after it has been pumped should behave similarly to water entering a stream. So our next step is to analyze oxyhydroxides that precipitate in well boreholes, as well as work in the Slate Belt, and Eastern Slate Belt rocks.
Geochemical maps (National Uranium Resource Evaluation – NURE database)

- Groundwater and stream sediment maps for North Carolina are on the North Carolina Geological Survey Internet site
- http://www.geology.enr.state.nc.us


Geolocated DHHS Laboratory Data through June 2004 (11,214 samples)

Focazzio, et al., 1999 (148 Samples)
Status

• Phase one completed with small grant -- $5,000 from Aquifer Protection Section (APS)
• Follow-up APS grant forthcoming for work in the Eastern Slate Belt, and likely in well studies at a Piedmont site
• Field and laboratory work on hold pending APS funding except for periodic inquiries and outreach
• Potential 319 funds for 2006 timeframe; application deadline in May 2005
• EPA previously solicited proposals in this specific area
• Geochemistry software scheduled for purchase from other source (Geochemist’s Workbench)
Elemental Ratios from Oxides Coatings formed on Stream Boulders

As/Mn ratio

Ratio

(As / Mn) + Fe ratio

As/Zn ration

As / Fe ratio
Elemental Ratios from Oxides Coatings formed on Stream Tiles

As/Mn ratio

Ratio

As / Fe ratio

(As / Mn) + Fe ratio

As/Zn ration

Graph showing elemental ratios from oxide coatings on stream tiles. The x-axis represents the As/Mn ratio, and the y-axis represents various ratios. Different markers indicate different ratios: As/Fe, (As/Mn) + Fe, and As/Zn.
Arsenic probability at the 0.01 mg/l Threshold
Arsenic probability at the 0.001 mg/l threshold.
### Arsenic analyses distribution

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<tr>
<th>Arsenic Concentration Range (mg/L)</th>
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<th>Relative %</th>
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<td>0.001 to 0.010</td>
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<td>0.141 to 0.150</td>
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<td>0.008</td>
</tr>
<tr>
<td>0.151 to 0.160</td>
<td>2</td>
<td>0.008</td>
</tr>
<tr>
<td>0.161 to 0.170</td>
<td>2</td>
<td>0.008</td>
</tr>
<tr>
<td>0.171 to 0.180</td>
<td>1</td>
<td>0.004</td>
</tr>
<tr>
<td>0.181 to 0.190</td>
<td>0</td>
<td>0.000</td>
</tr>
<tr>
<td>0.191 to 0.200</td>
<td>0</td>
<td>0.000</td>
</tr>
<tr>
<td>&gt;0.200</td>
<td>4</td>
<td>0.016</td>
</tr>
</tbody>
</table>

Total - 21,837

Non Detects (ND)
- <0.001 - 17,196
- <0.005 - 11
- <0.010 - 4,630

Frequency

Concentration Ranges
County probability to exceed 0.00001 mg/l

Number indicates total number of analyses per county. Data that did not meet selection criteria were excluded (i.e. MDL’s > threshold values).
Arsenic situation

- Arsenic public drinking water standard 10 ppb – not a health-protected standard
- Recommended groundwater standard for folks with private well 0.02 ppb – health based (State Toxicologist)
- Detection limit at lab is 1 ppb
- So any detection in private well in NC is too high for people to consume
- Studies show that As low as 5 ppb causes cancer
- Risk 1:100 etc., based 70 year exposure, 2 liters of water per day consumption
- >2,300 wells affected in NC; 1,700 in last 18 months