

Arsenic and Geochemical Characteristics of Groundwater from Domestic Wells in Greater Augusta, Maine

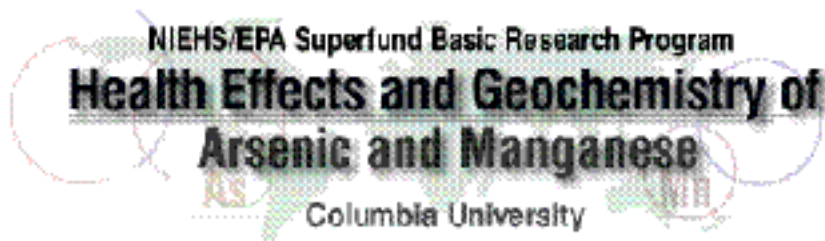
Yan Zheng^{1,2}

Co-authors: Hun Bok Jung¹, Qiang Yang¹, Charles W. Culbertson³, Robert G. Marvinnney⁴, Marc C. Loiselle⁴, Daniel Locke⁴, Heidi Cheek⁴, Hilary Thibodeau⁴,

Collaborators on Rn: C. T. Hess⁵, and P. Smitherman⁵

¹Queens College and Graduate Center, City University of New York, Flushing, NY 11367 ²Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY 10964

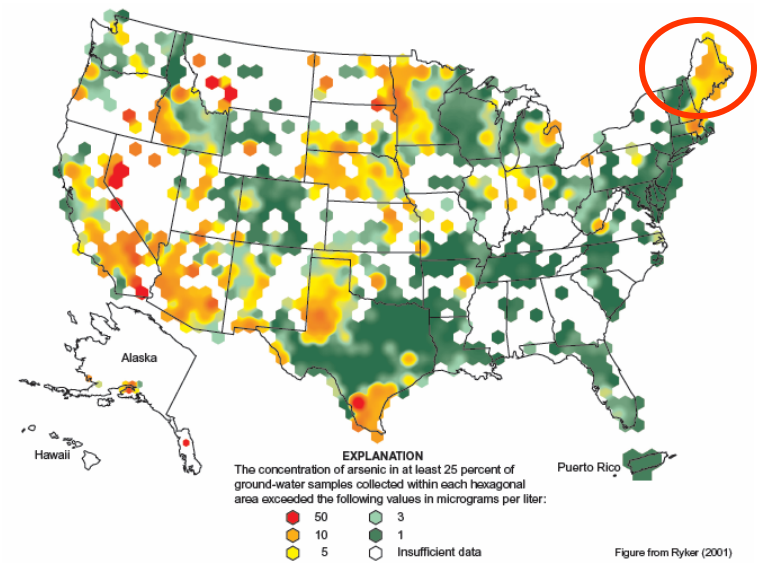
³USGS, Maine Water Science Center, 196 Whitten Rd., Augusta, ME 04330 ⁴Maine Geological Survey, 22 State House Station, Augusta, ME 04333 ⁵University of Maine, Dept. of Physics, 120 Bennet Hall, Orono, ME 04469



World-wide problem of elevated groundwater arsenic



Smedley and Kinniburgh, 2002, Appl Geochem

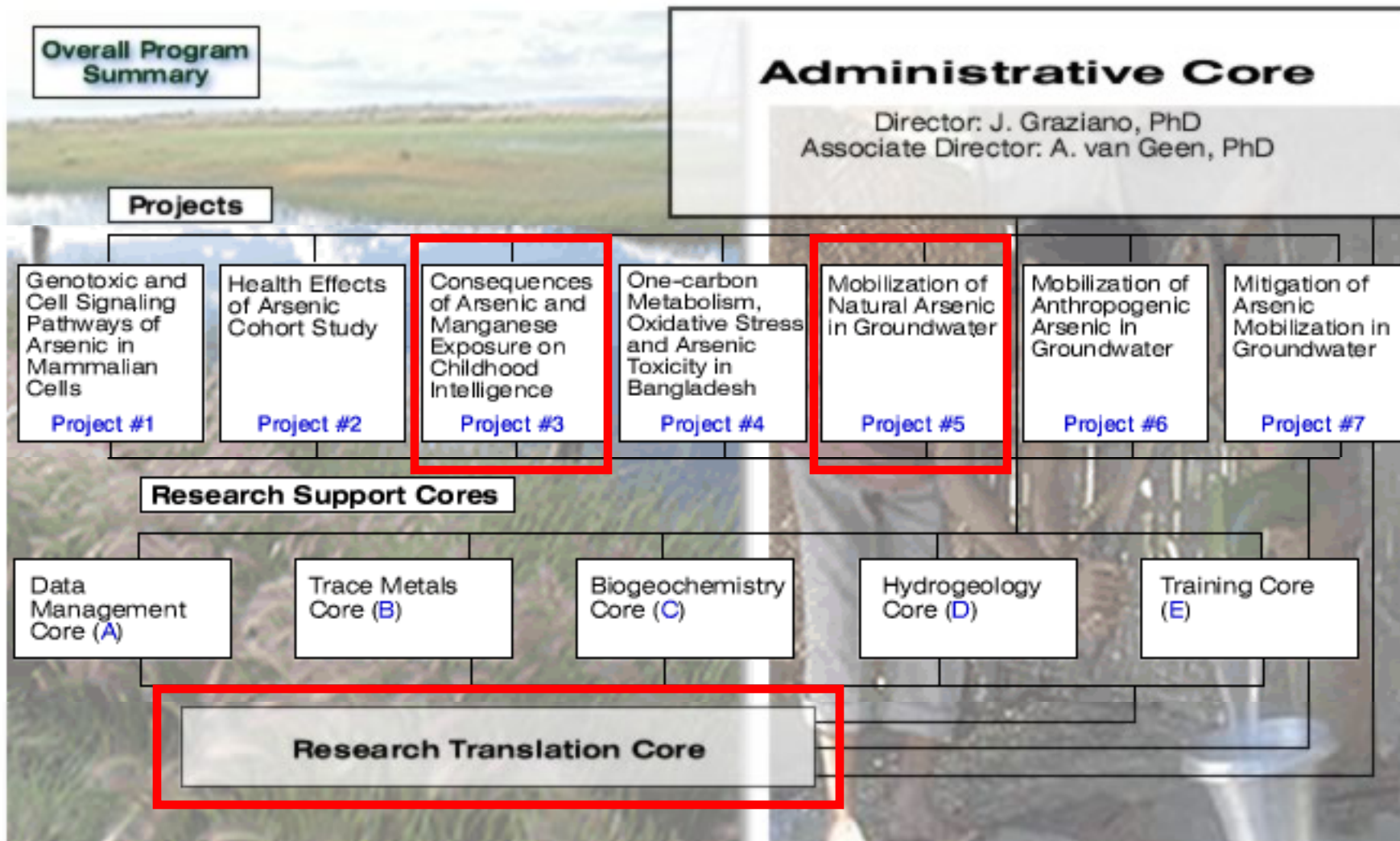


USGS

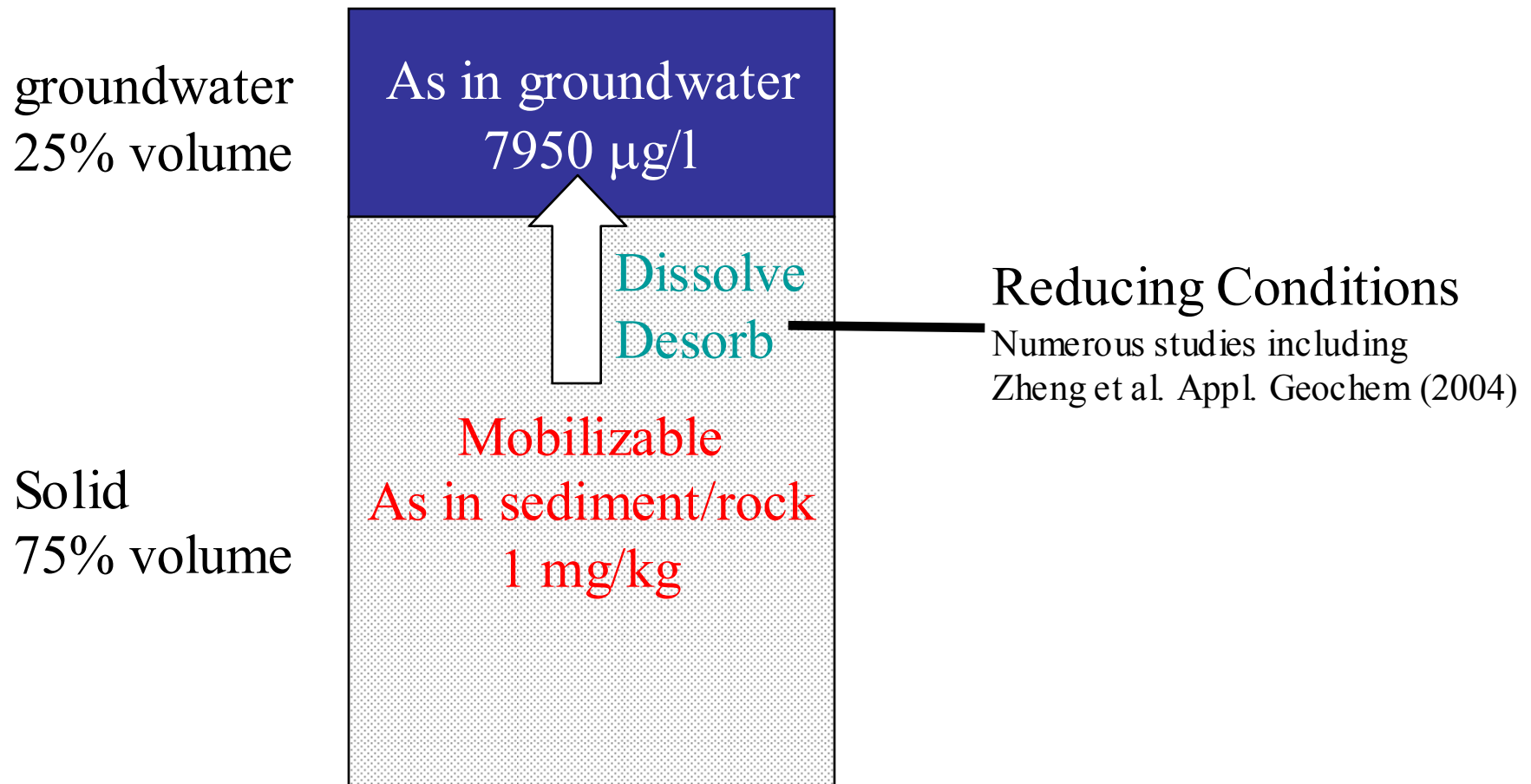


NIEHS/EPA Superfund Basic Research Program
**Health Effects and Geochemistry of
Arsenic and Manganese**

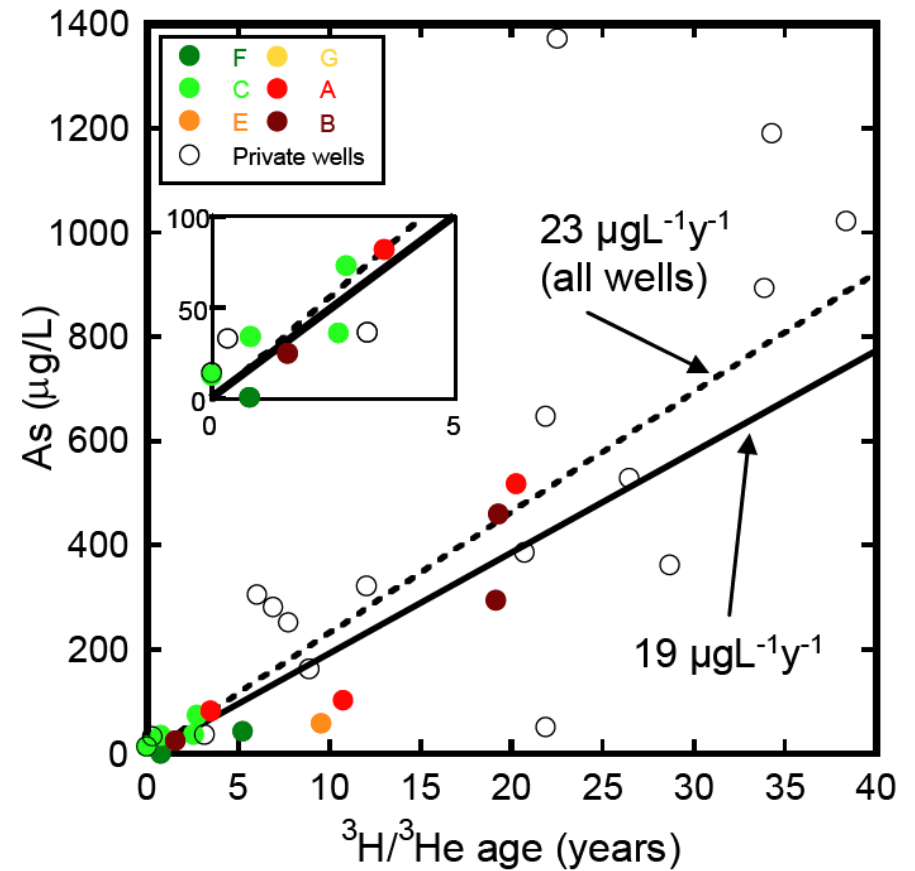
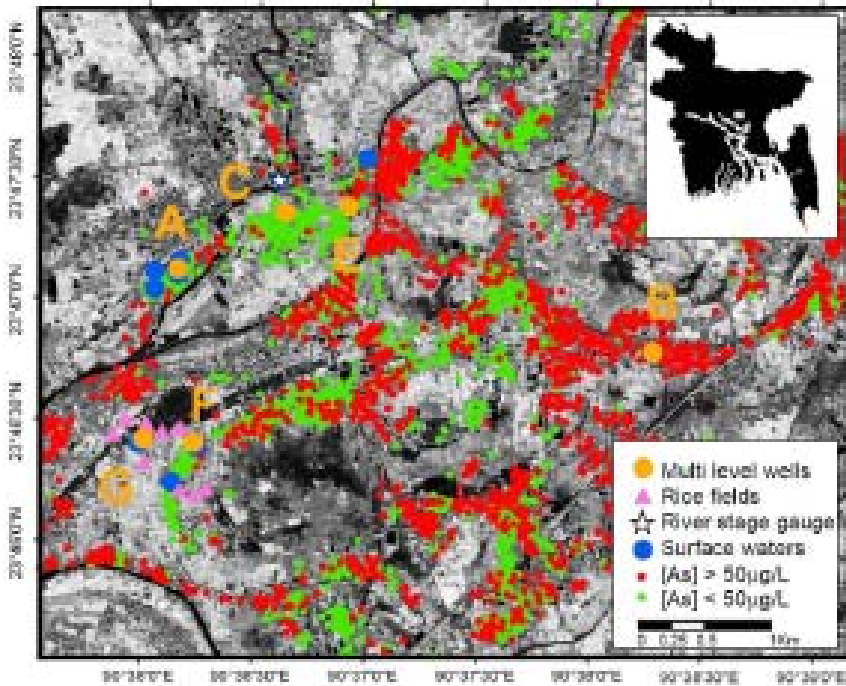
Columbia University



Arsenic of natural origin is sufficient to increase water As to > 10 $\mu\text{g/L}$



Motivation



Stute et al., 2007, Water Resources Research,
in press

Arsenic is released at a rate of ~ 20 $\mu\text{g/L}$ per year

Sampling of groundwater

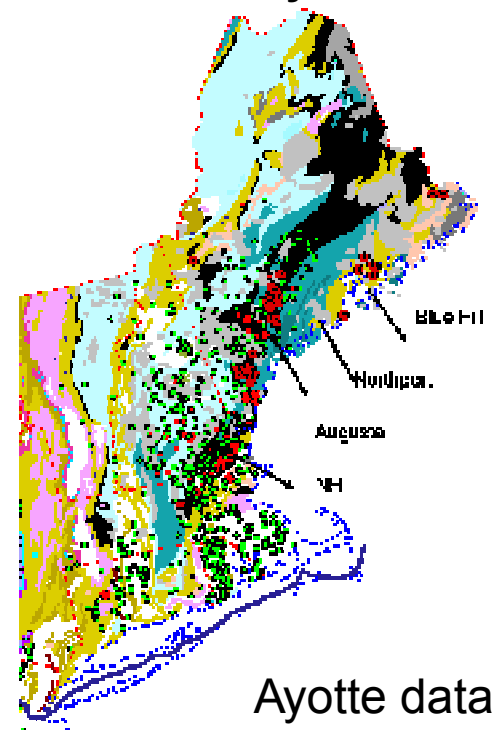
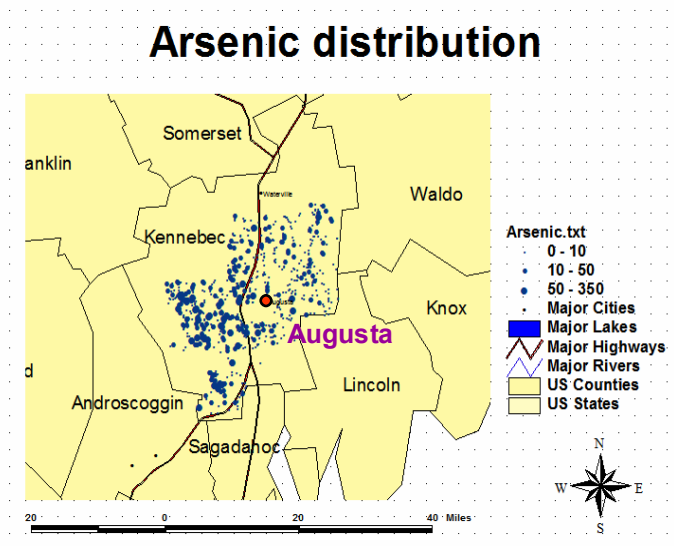
786 samples were collected from private bedrock wells of 12 towns in Kennebec county, Maine from May 25 to Oct 18, 2006 by Maine Geological Survey/USGS, and analyzed for 27 elements and 6 anions by HR ICP-MS at Lamont and IC at Queens College.

Temperature, conductivity, pH, and DO were measured in the field. Radon was determined immediately after returning from field at U of Maine.

Statistics of 12 towns in Kennebec County, Maine

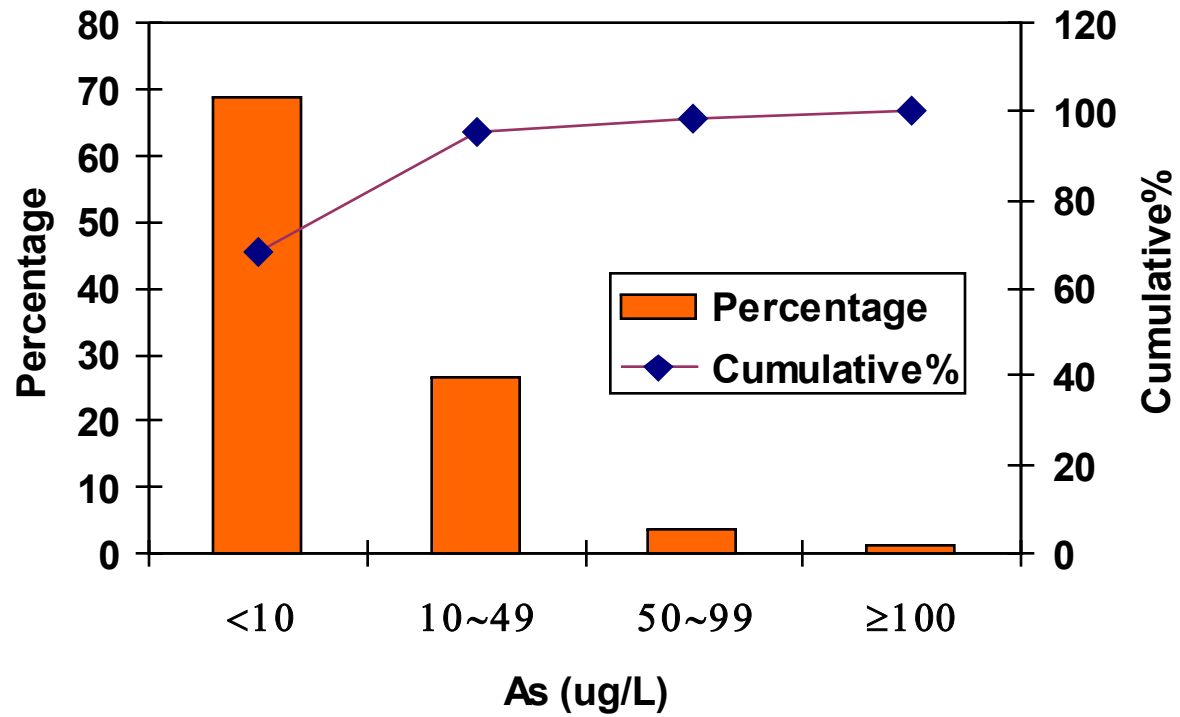
Total population: 54,428, total household: 22,658; Total area: 1,069 km² (US census 2000)
About 42% of Maine's citizens use a private groundwater well for their household water supply: Individual drilled well: 32.5%, Individual dug well: 9.4%. (US census 1990)

Sampling density: an average of 1 sample was taken for every ~9 households with private well, and every ~1.4 km²



Comparison of our study scheme with the previous studies on groundwater As in Maine and New England

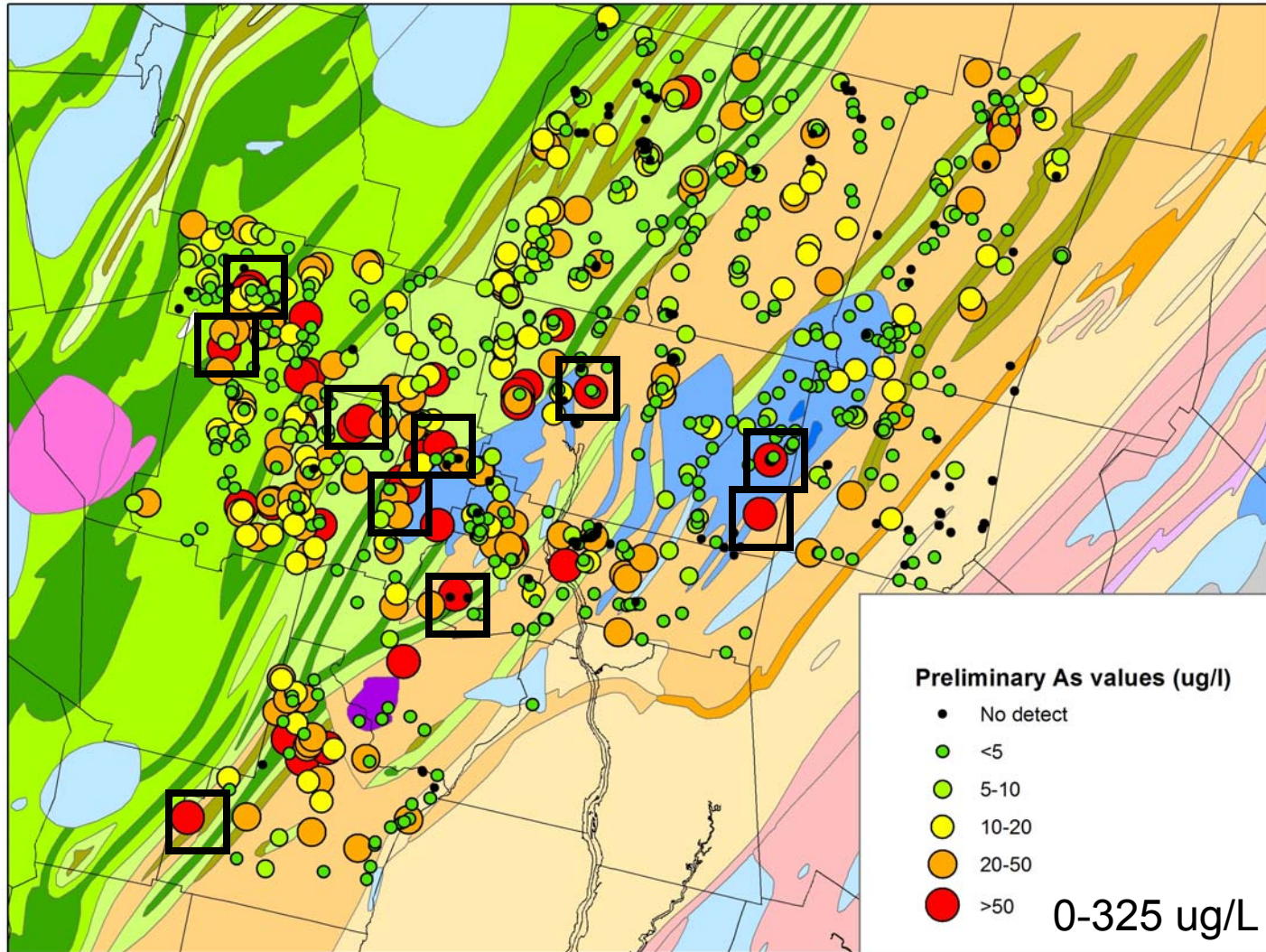
Researcher (year)	Study area	Number of Sampling	Sampling density	Percentage of elevated As
Ayotte et al (1999)	New England	800 public wells + 190 private wells	1 sample every 183 km ²	23% > 5 ug/L 10-15% > 50 ug/L
Peters et al (1999)	New Hampshire	992 drinking water from households	1 sample every 24 km ²	15% of domestic bedrock well > 10 ug/L 3% of domestic bedrock well > 50 ug/L
Loiselle et al (2002)	Maine (Towns of Buxton and Hollis)	1200 private domestic wells	1 sample every 151 km ²	12-13% > 10 ug/L 1-3% > 50 ug/L
Ayotte et al (2003)	New England	30 unconsolidated aquifer 28 calcareous metamorphic bedrock 30 undifferentiated metamorphosed marine sediment and felsic igneous rock	1 sample every 2061 km ²	3% > 10 ug/L 29% > 10 ug/L 7% > 10 ug/L
Peters et al (2003)	South central New Hampshire	127 bedrock wells	1 sample every 1.8 km ²	62% > 10 ug/L
Lipfert et al (2006)	Maine (Northport)	35 bedrock wells 7 drift wells	1 sample every 0.05 km ²	55% > 10 ug/L
Present study	Maine (Kinnebec county)	786 domestic bedrock wells	1 sample every 1.4 km ²	31% > 10 ug/L 5% > 50 ug/L



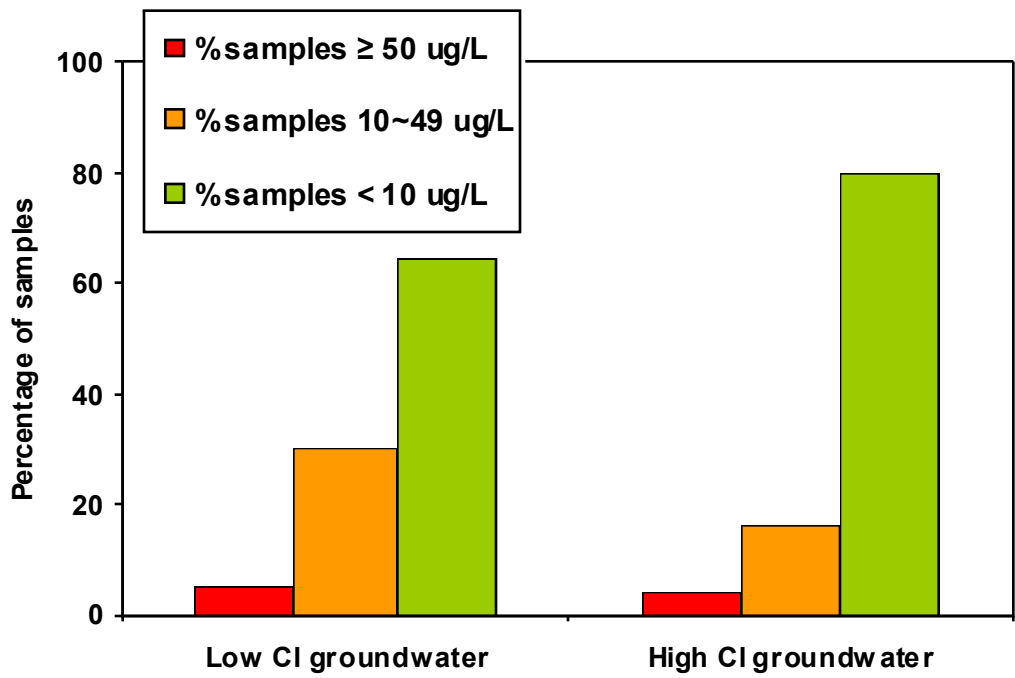
As (ug/L)	Frequency	Percentage	Cumulative%
<10	540	68.6	68.6
10~49	209	26.6	95.2
50~99	27	3.4	98.6
>100	11	1.4	100.0

Maximum As: 325 ug/L

Arsenic distribution

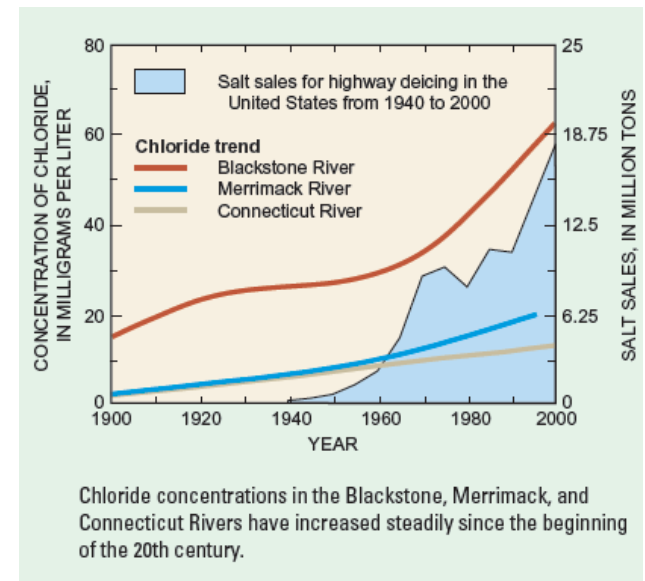


Wells with low Cl (< 30 mg/L) have higher percentage of >10 ug/L As



Both older or younger than 50 years?

Younger than 50 years?



Presumably aged groundwater
(High pH + Low DO + Low Cl)

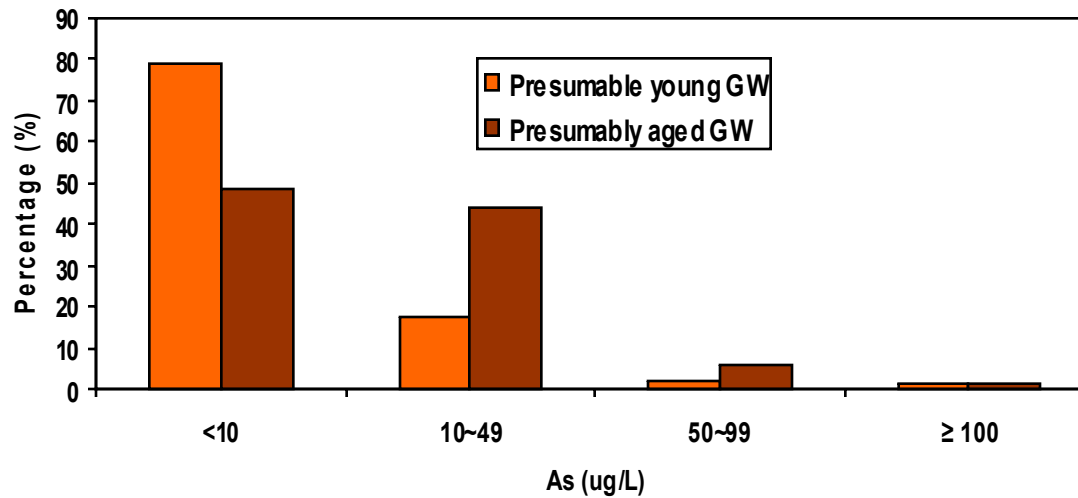
Presumably young groundwater

As (ug/L)	Frequency	Percentage	Cumulative%
<10	131	48.5	48.5
10~49	118	43.7	92.2
50~99	17	6.3	98.5
≥100	4	1.5	100.0

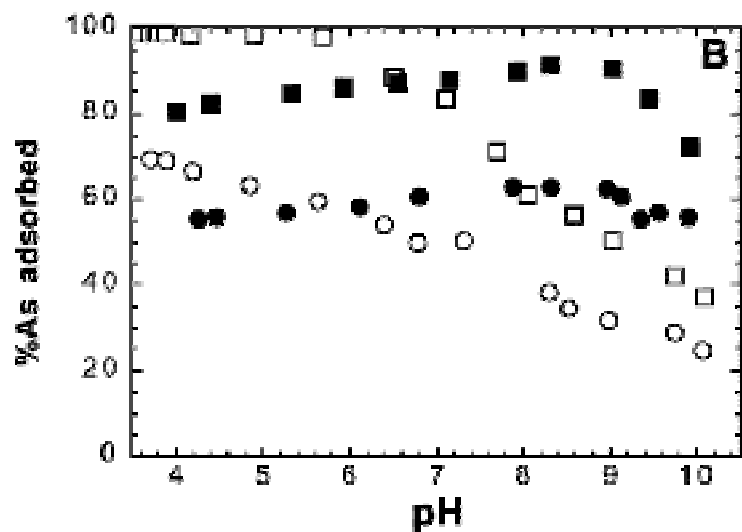
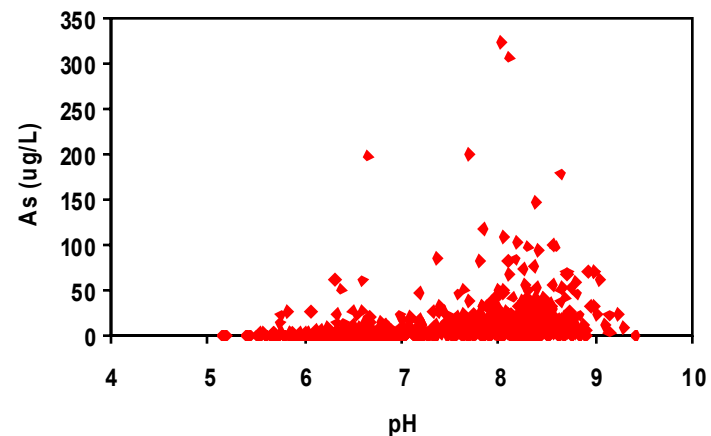
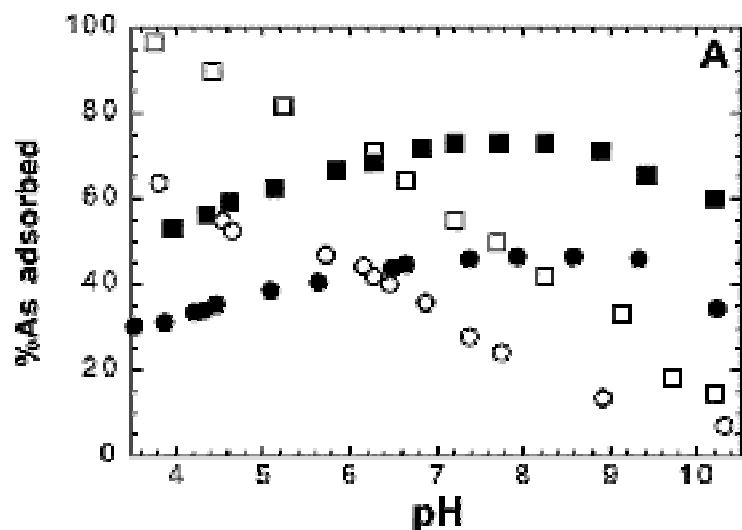
As (ug/L)	Frequency	Percentage	Cumulative%
<10	409	79.1	79.1
10~49	91	17.6	96.7
50~99	10	1.9	98.6
≥100	7	1.4	100.0

Number of samples: 270
Median: 10.42 ug/L
Average: 18.92 ug/L

Number of samples: 516
Median: 2.14 ug/L
Average: 8.81 ug/L



1. Desorption of As(V) from Iron oxyhydroxides due to increase of pH



Comparison of As(V) and As(III) sorption edges on (a) HFO and (b) goethite. The total arsenic concentrations shown are 100 M (circles) and 50 M (squares). Open symbols represent As(V) and closed symbols As(III). Experimental conditions: 0.01 M NaClO₄; 0.03 g L⁻¹ HFO or 0.5 g L⁻¹ goethite. Data are taken from Figures 1 and 2. (Dixit and Hering, 2003, EST)

Test: Determine AsIII and AsV in high As high pH wells

2. Reductive dissolution of Iron oxyhydroxides

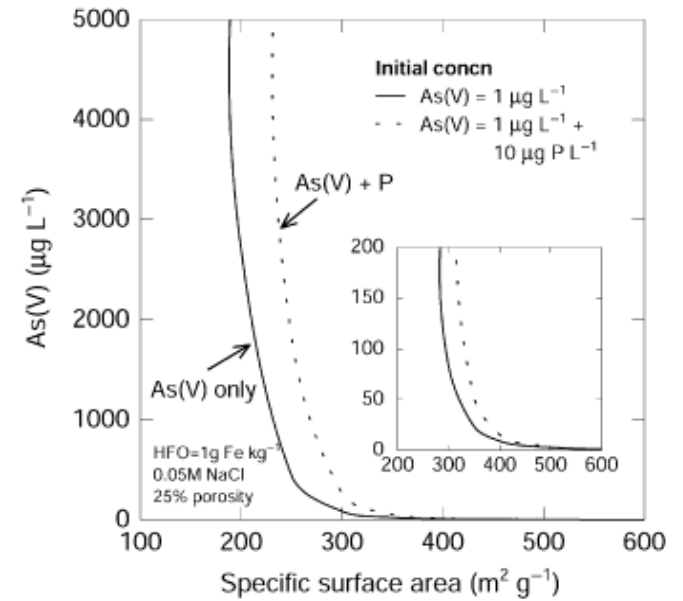
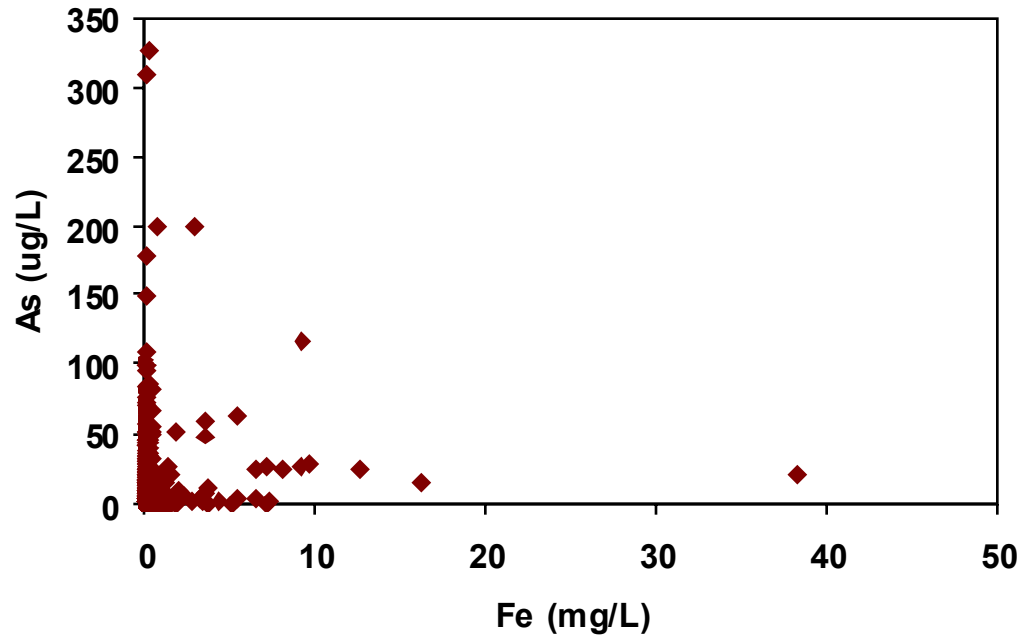
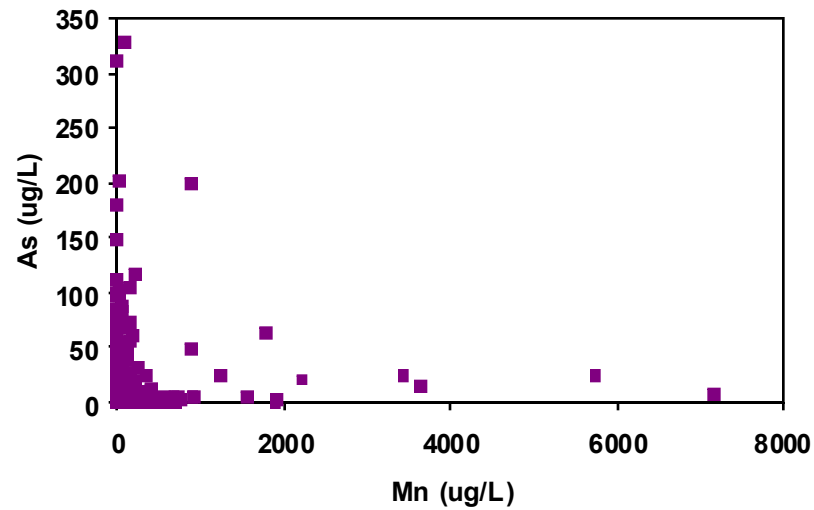
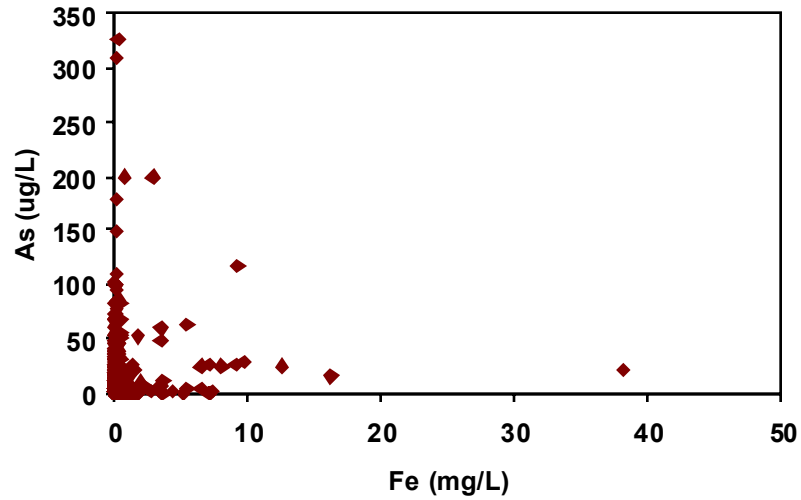


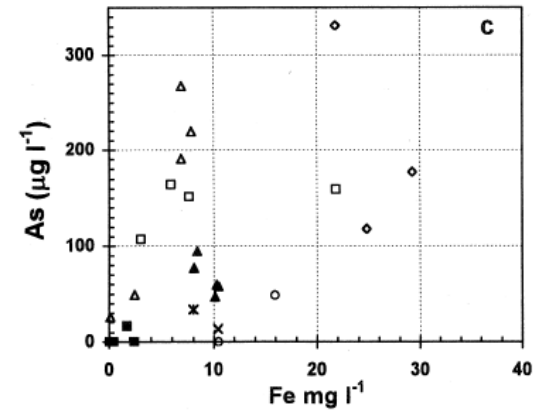
Fig. 6. Calculated increase in As concentration when the specific surface area of HFO in a sediment containing 1 g kg^{-1} Fe as HFO is reduced from its initial value of $600 \text{ m}^2 \text{ g}^{-1}$ under closed-system conditions.

Smedley and Kinniburgh, 2002, Appl Geochem

Inverse relationship
between As and Fe, Mn



Decoupling between As and Fe
In Bangladesh groundwater



Nickson et al, 2000, Appl Geochem

3. Oxidation of sulfide mineral

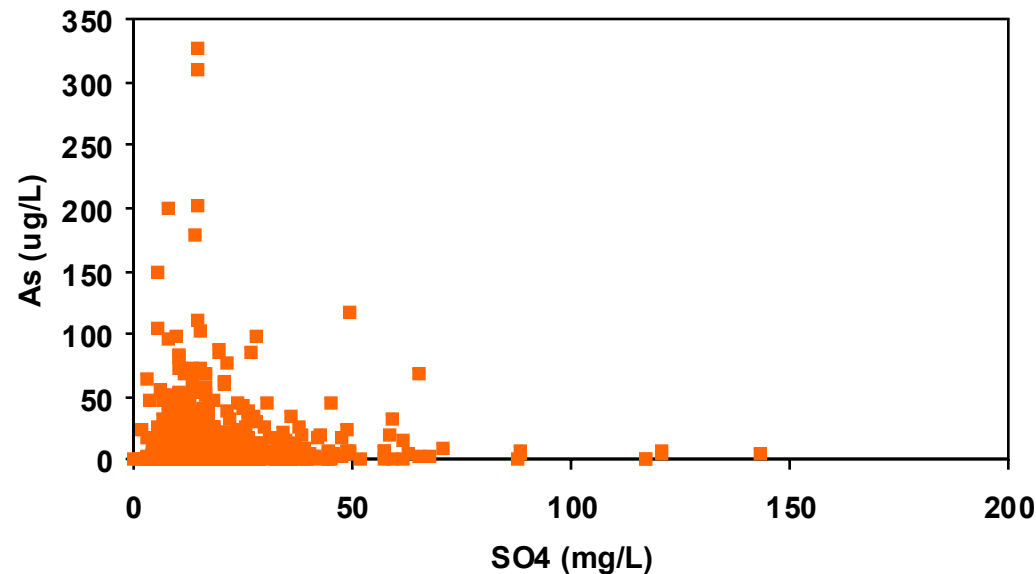
(1) Groundwater samples containing high As frequently have low DO (≤ 1 mg/L)

(2) The pH is weakly alkaline (mean pH : 7.6)

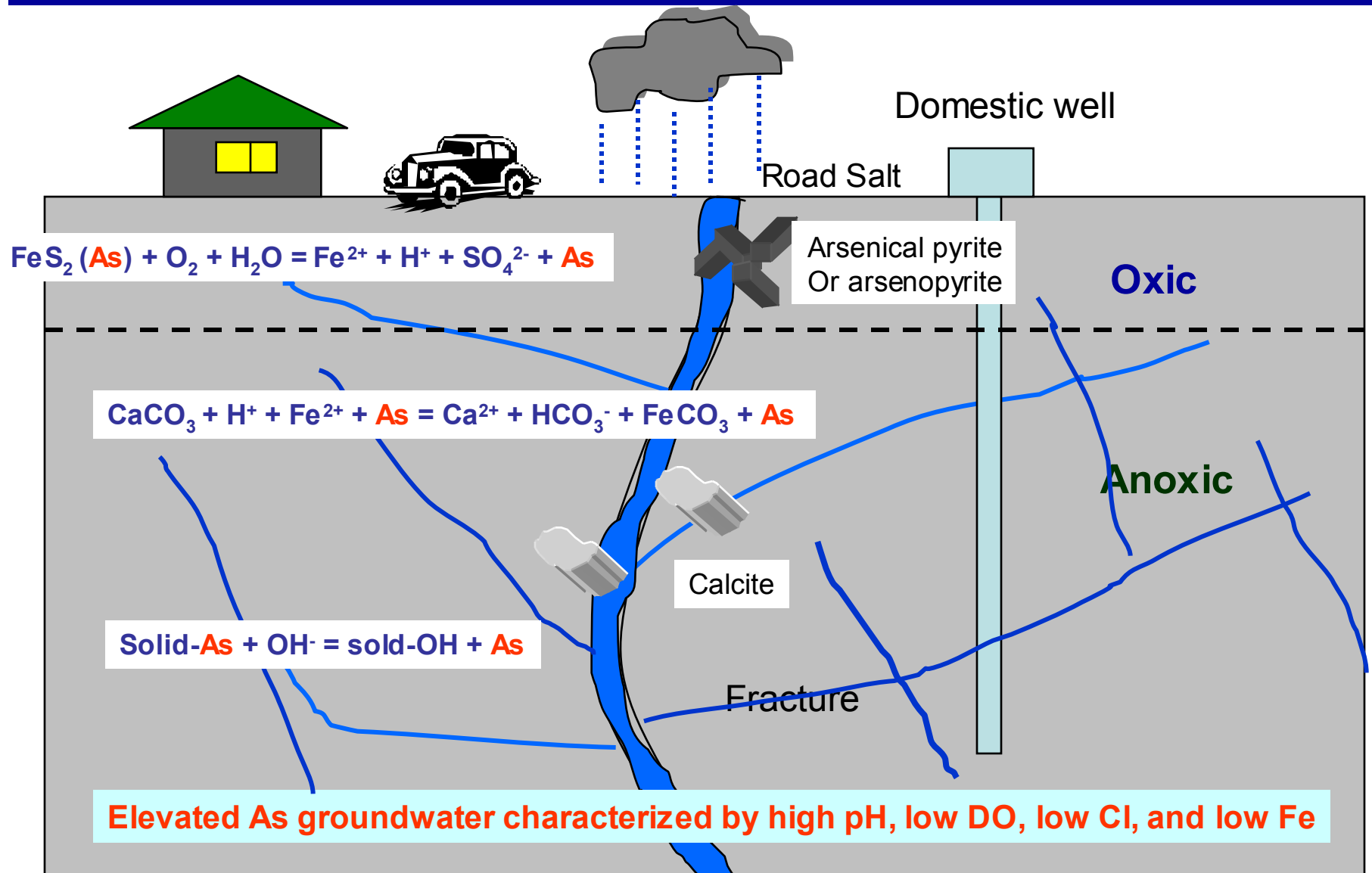
If sulfide oxidation is the direct As mobilization Mechanism, pH is likely to be acidic.

(3) Sulfate concentration in groundwater samples is relatively low
(Mean SO₄ = 15 mg/L)

(4) Sulfide minerals, especially pyrrhotite (FeS-FeS₂) are abundant in the graphitic sulfide-rich schists (Ferry, 1981)



Proposed combined mechanism of As mobilization in Maine bedrock aquifer



Conclusions

- A significant percentage (31%) of domestic well waters from Greater Augusta area contained > 10 ug/L As, with a maximum As level of 325 ug/L.
If same occurrence rate is assumed for the entire Kennebec county, Maine, then $\sim 14,782$ households are exposed to As greater than EPA-MCL of 10 ug/L.
- Elevated As (> 10 ug/L) is distributed more frequently in the wells tapping Silurian calcareous metamorphic rock (see Jung et al. poster).
- Groundwater samples containing high pH (≥ 7), low DO (≤ 1 mg/L), and low Cl (< 30 mg/L) showed higher percentage of elevated arsenic, which suggests that aged and evolved groundwater tends to contain higher As.
- Elevated As in Maine groundwater seems to result from combined mechanisms including sulfide oxidation and neutralization by calcite along the groundwater flow path (see Yang et al. poster).
- 29 % of groundwater samples were found to contain elevated Rn and frequently distributed in the wells tapping undifferentiated metamorphic rock or felsic igneous rock (see Smitherman et al. poster).

Future Studies

1. To evaluate the percentage of domestic wells with elevated As in different bedrock geologic unit based on Augusta regional (2006) and cluster (2007) sampling.
2. To estimate population exposed to elevated As in New England based on this work and previous high density sampling.
3. Hydrogeological investigations of a high As cluster area in Greater Augusta, following established USGS Mirror Lake fractured bedrock site strategy.
4. Blue Hill –volunteer program in 2007

Bedrock represents an older sedimentary rock unit deposited during the Ordovician, which is similar to the younger Silurian calcareous metamorphic rock and is composed mostly of inter-bedded pelites and sandstone

