Combination of Hydrous Iron Oxide Precipitation with Zeolite Filtration to Remove Arsenic from Contaminated Water

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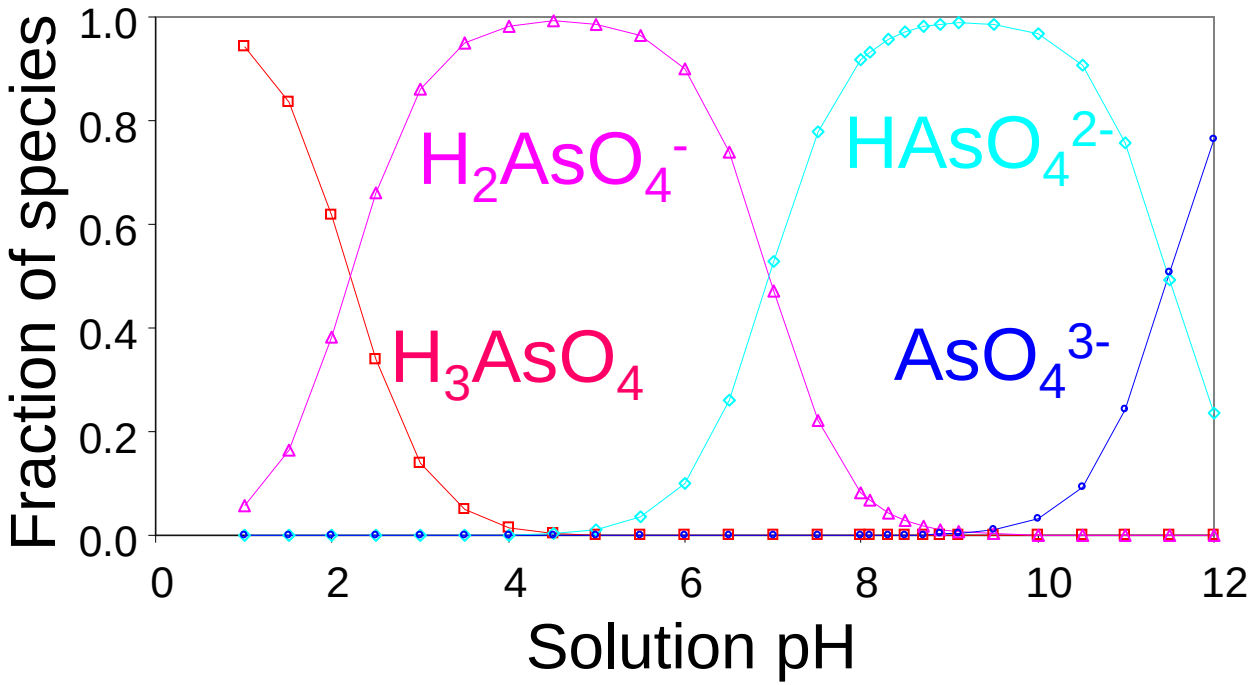
Topic of discussion

1. Why combination of zeolite + Fe?
2. Materials and methods
3. Results
4. Instrumental characterization
5. Conclusions
6. Extension of the project

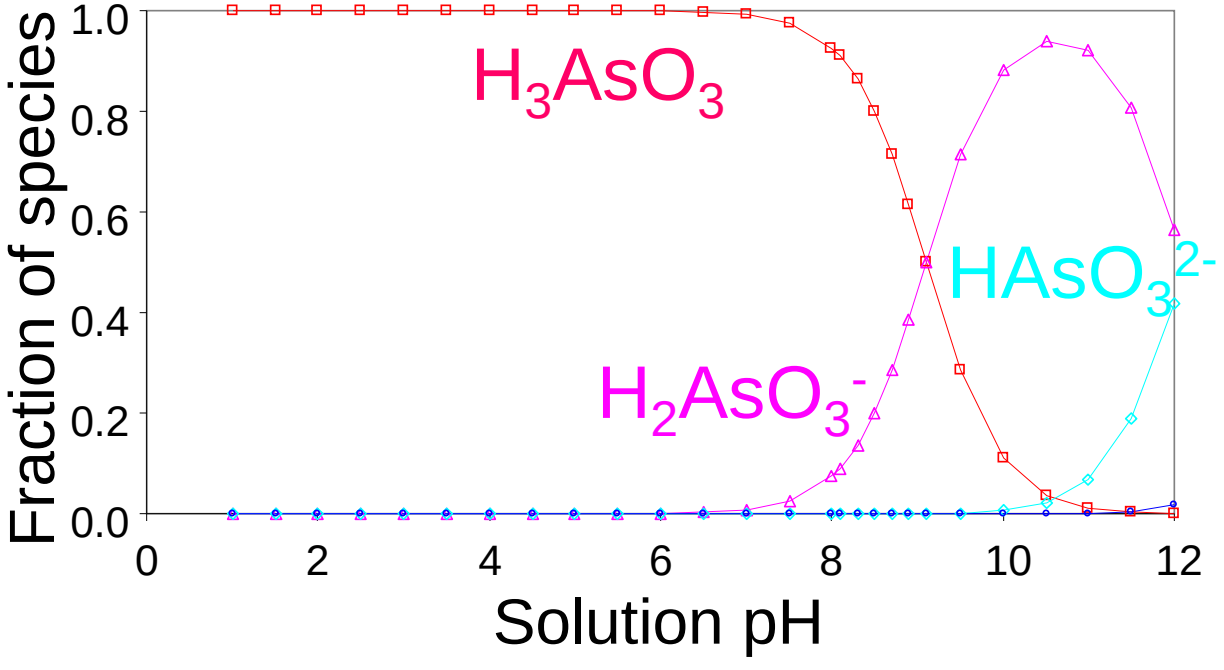
1. Why combination of zeolite + Fe?

- $Fe(OH)_3$ have strong affinity for As.
- Precipitation of $Fe(OH)_3$ from water could remove most of dissolved As.
- Filtration of As-laden $Fe(OH)_3$ could be achieved using sand packs.
- Zeolite has higher cation exchange capacity, thus used as cation exchanger for water treatment.
- Propose to use zeolite as filtration media

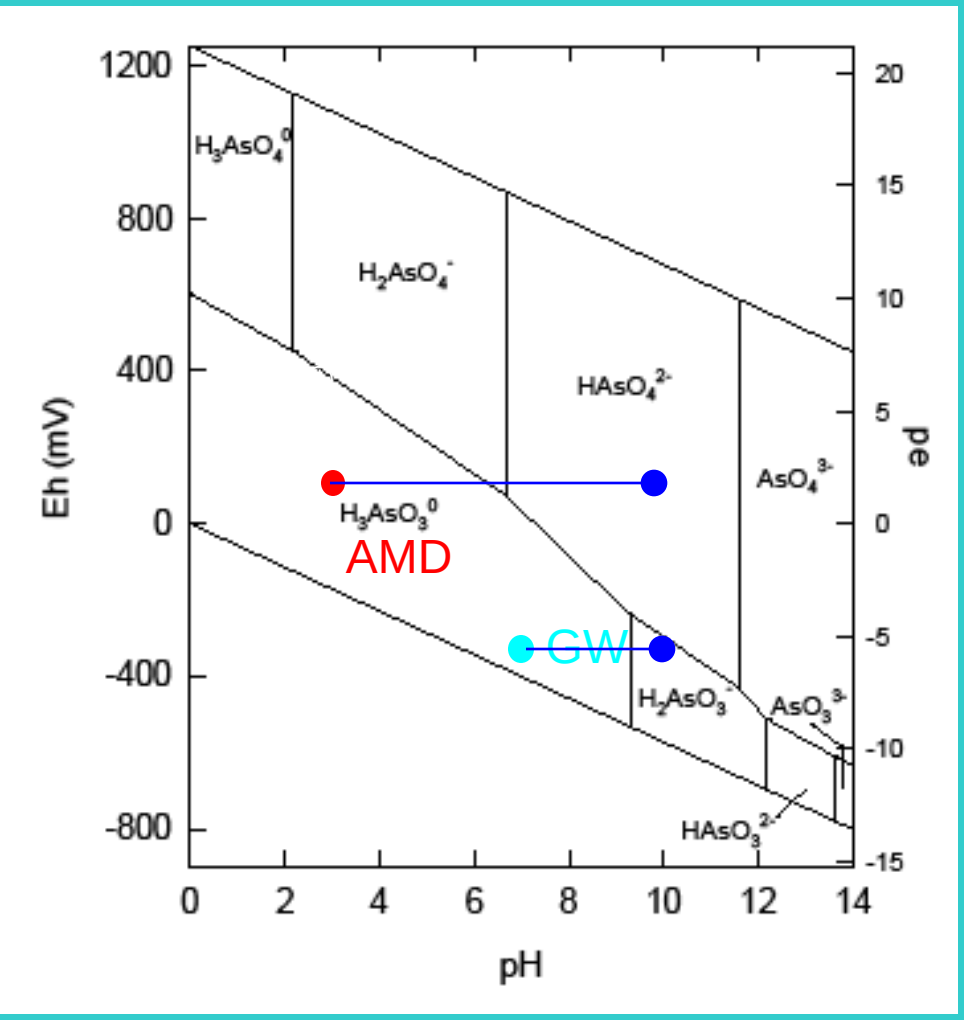


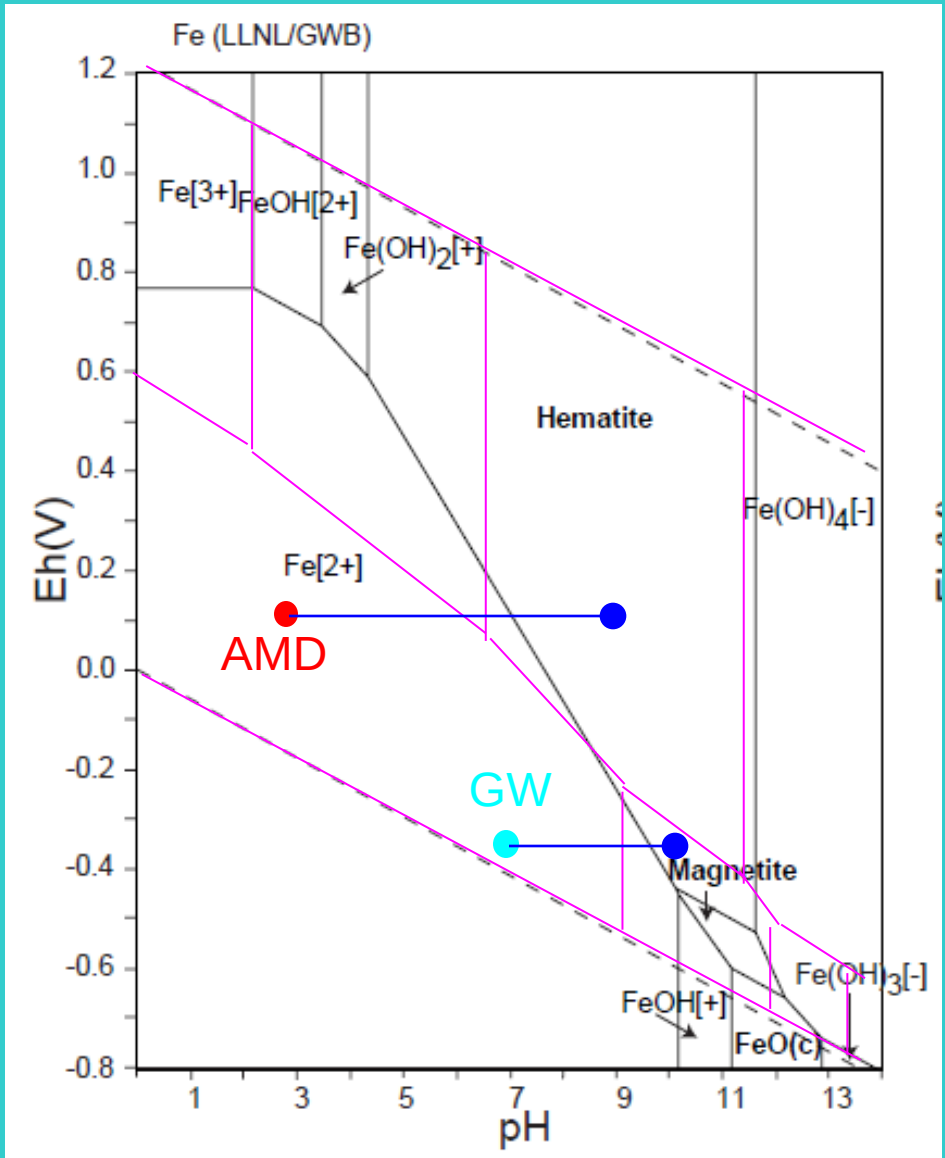
Arsenate speciation As(V)

Arsenite speciation As(III)



pH-Eh diagram of As and Fe

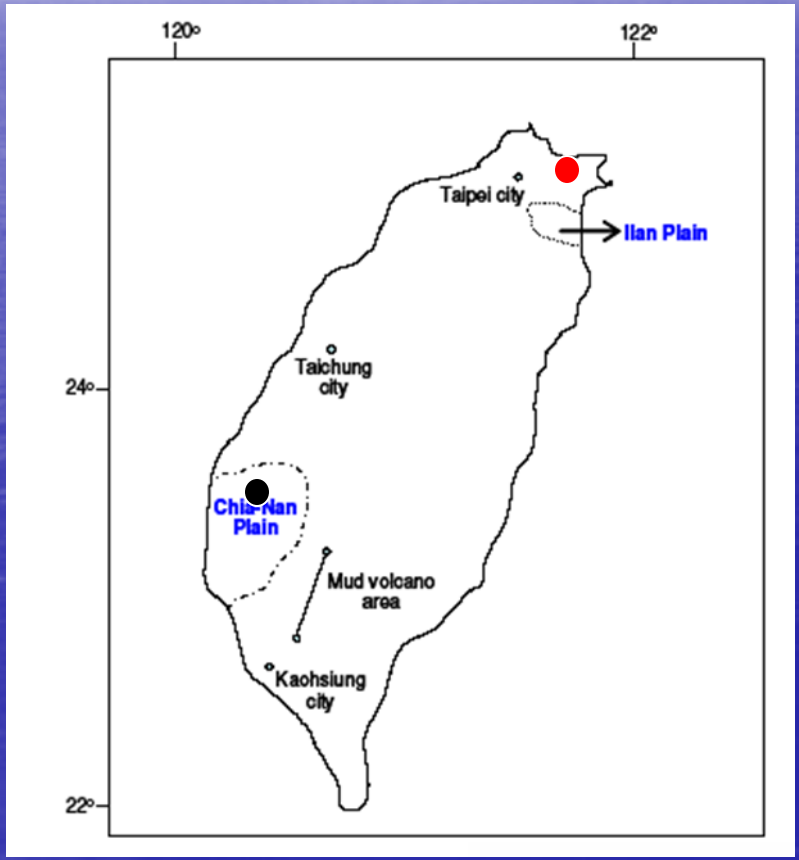




2. Materials and methods

- The zeolite obtained from the St. Cloud mine (Winston, NM) with a particle size range of 8-14 mesh (1.4 to 2.4 mm).
- Real groundwater collected from Chia-Nan Plain aquifer (SW Taiwan): Humic acid = 34 to 468 QSU (mean = 128±126.8 QSU) (quinine standard unit); ORP = -179 mV; As = 511 μg/L; neutral pH
- Acid mine drainage (AMD) water collected from (Chinkuashih, N Taiwan): pH = 2.9; ORP = 374 mV; Fe^{TOT} >100 mg/L; As = 147 μg/L

2. Materials and methods



Batch studies

- Varying amounts of stock solution of As (100 mg/L) was added to DI water (178–196 mL)
- Varying amounts of 10 mM $FeCl_3 \cdot 6H_2O$ added
- Solution pH raised to 7–9 by adding 1 M NaOH
- Samples were analyzed for equilibrium As and Fe concentrations

Small column studies

- Each 60 mL syringe sleeve was filled with 50 mL with zeolite (about 50 g as the bulk density of zeolite is about 1 g/cm^3)
- To 1 L of 100 or 1000 µg/L As solution, 1, 3, or 10 mL of 10 mM Fe stock was added to reach a final Fe concentration of 10, 30 and 100 µM
- The solution pH was raised to 7–9 by adding 1 M NaOH drop-wise before being passed through the column

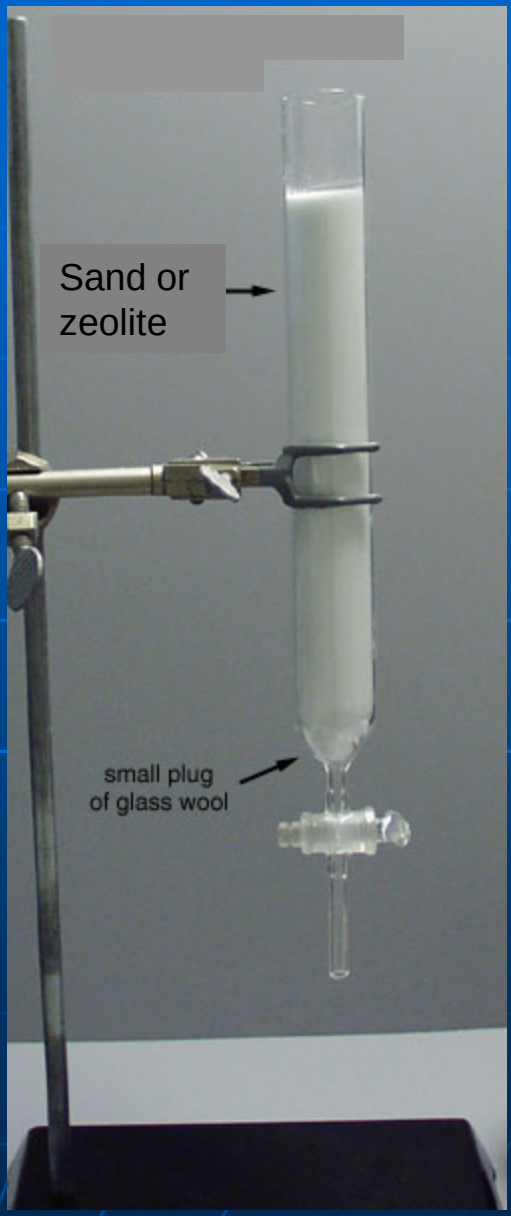
Large column studies

- 4.5 cm in diameter and 70 cm in length
- 400 g of zeolite packed to a height of 28 cm
- 10 L of 100 µg/L As (V) synthetic arsenic containing water
- 270 mg of $FeCl_3 \cdot 6H_2O$ added to make a final Fe concentration of 5.6 mg/L
- 3.1 mL of 1 M NaOH added to raise the pH to 9.1
- The mixture stirred vigorously to induce precipitation
- Water passed through the column at a flow rate about 300 mL/min

Large column studies

- Samples taken every liter
- Real groundwater collected from Chia-Nan Plain aquifer (SW Taiwan)
- Added Fe(III) equivalent to 0.2 mM, or 11.2 mg/L
- Acid mine drainage (AMD) water collected from (Chinkuashih, N Taiwan)
- No Fe(III) added, as it contained 101 mg/L of dissolved Fe already
- Only pH was raised by adding appropriate amounts of NaOH.
- The flow rate between 125 and 150 mL/min

Small column studies



Large column studies

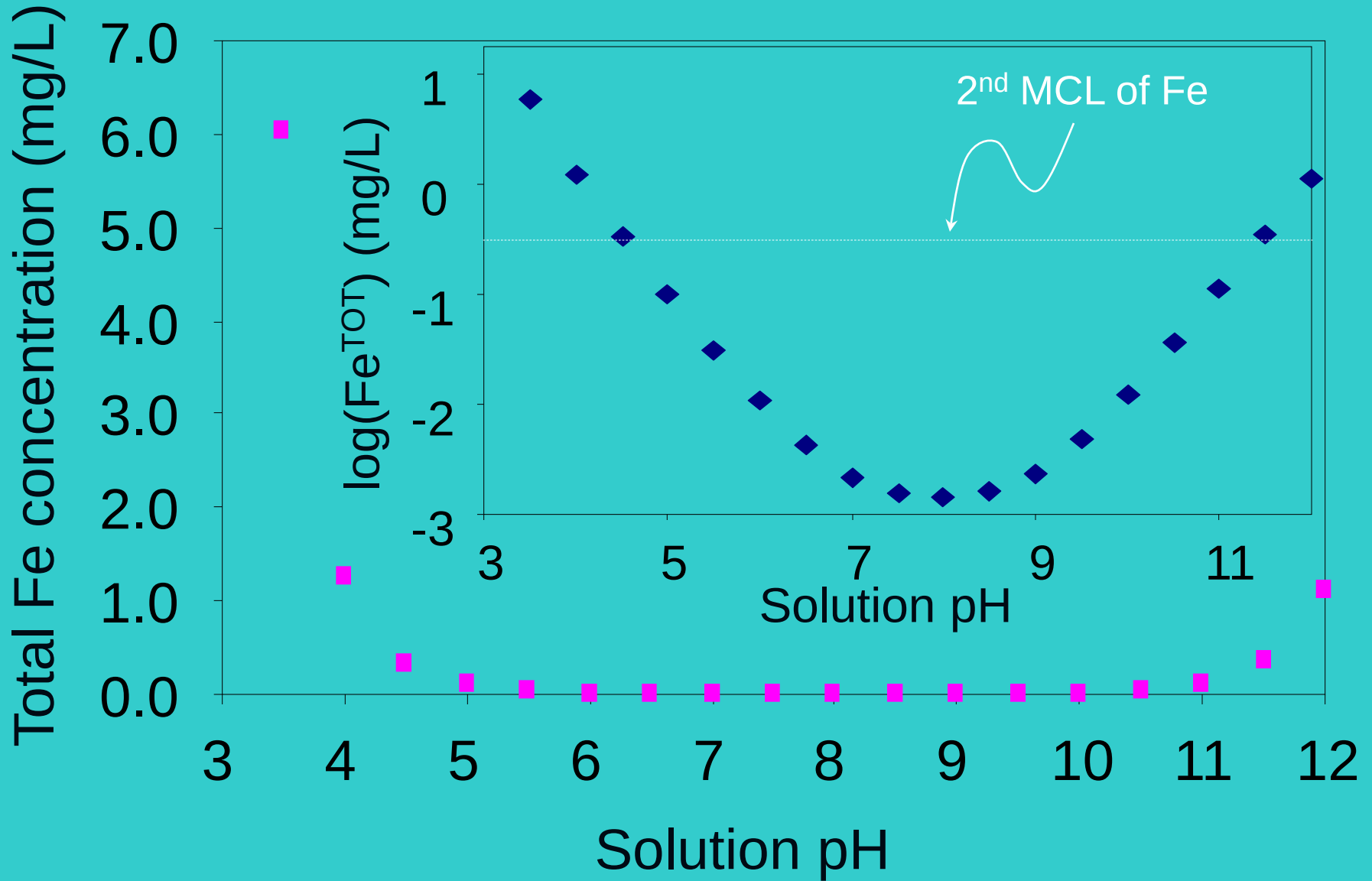
Chemical Analyses

- Fe^{TOT} was determined using Loviband MutiDirect Photometric System (The Tintometer Ltd., Dortmund, Germany) with an analytical range of 0.02 to 1.0 mg/L
- As was analyzed on either PE Optima 7000 DV ICP-OES with a detection limit of 1 µg/L or PSA Millennium System Excalibur (PS Analytical Ltd., UK) with a detection limit of 0.1 µg/L

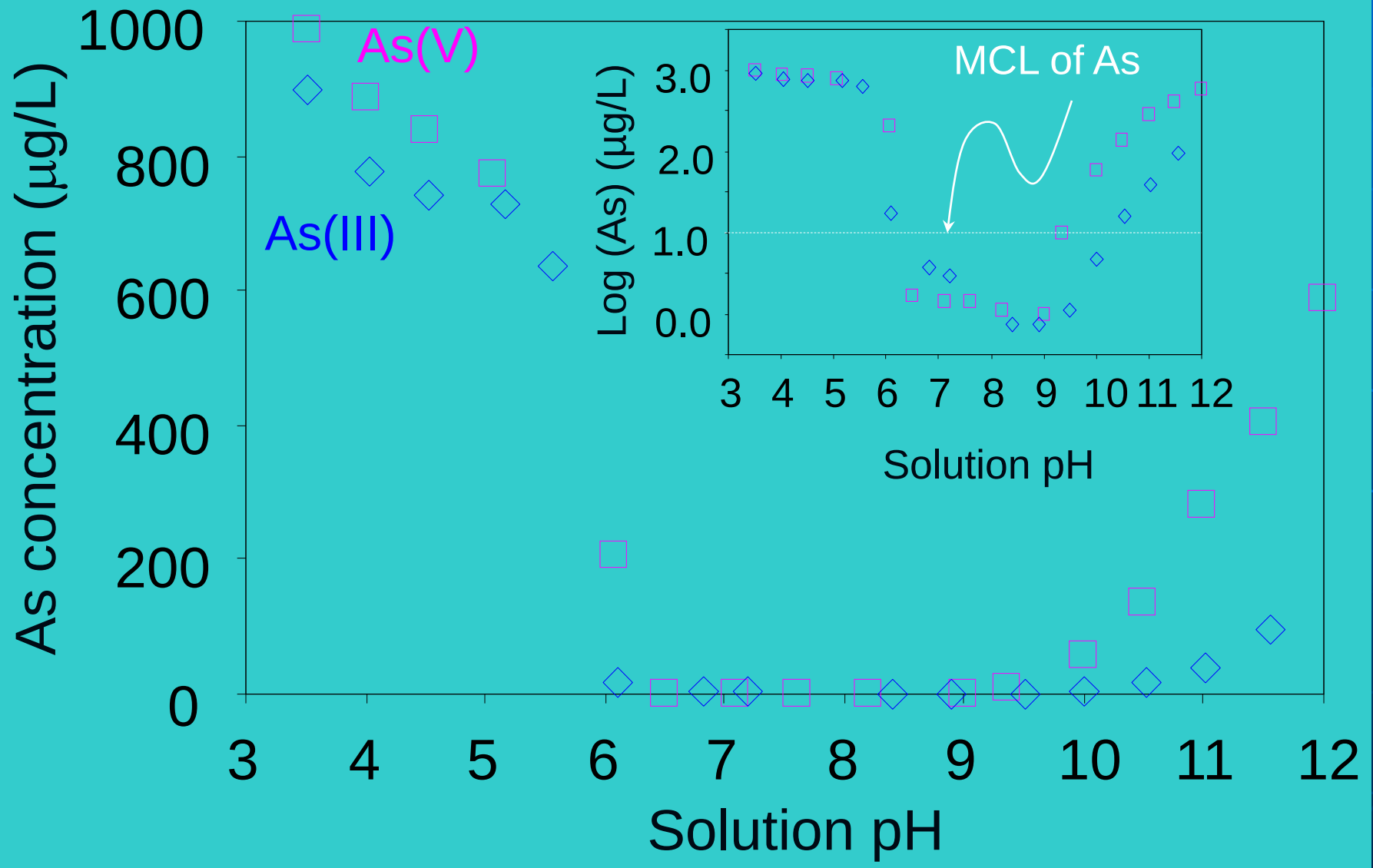
3. Results

- pH effect on Fe concentration
- pH effect on As concentration
- As adsorption on Fe precipitates
- Influence of Fe input on As removal
- Small column tests
- Tests with real water

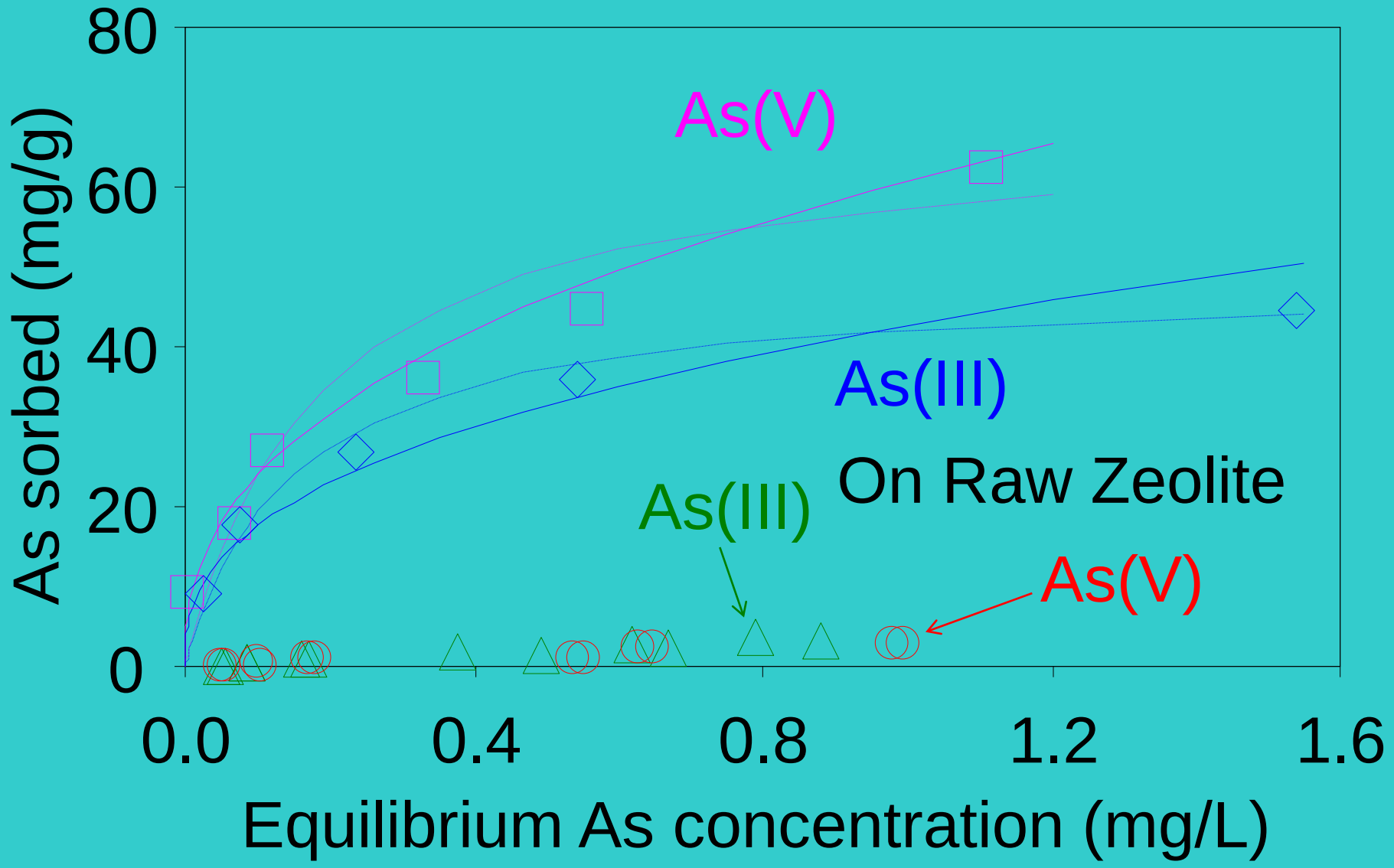
Fe concentration in solution



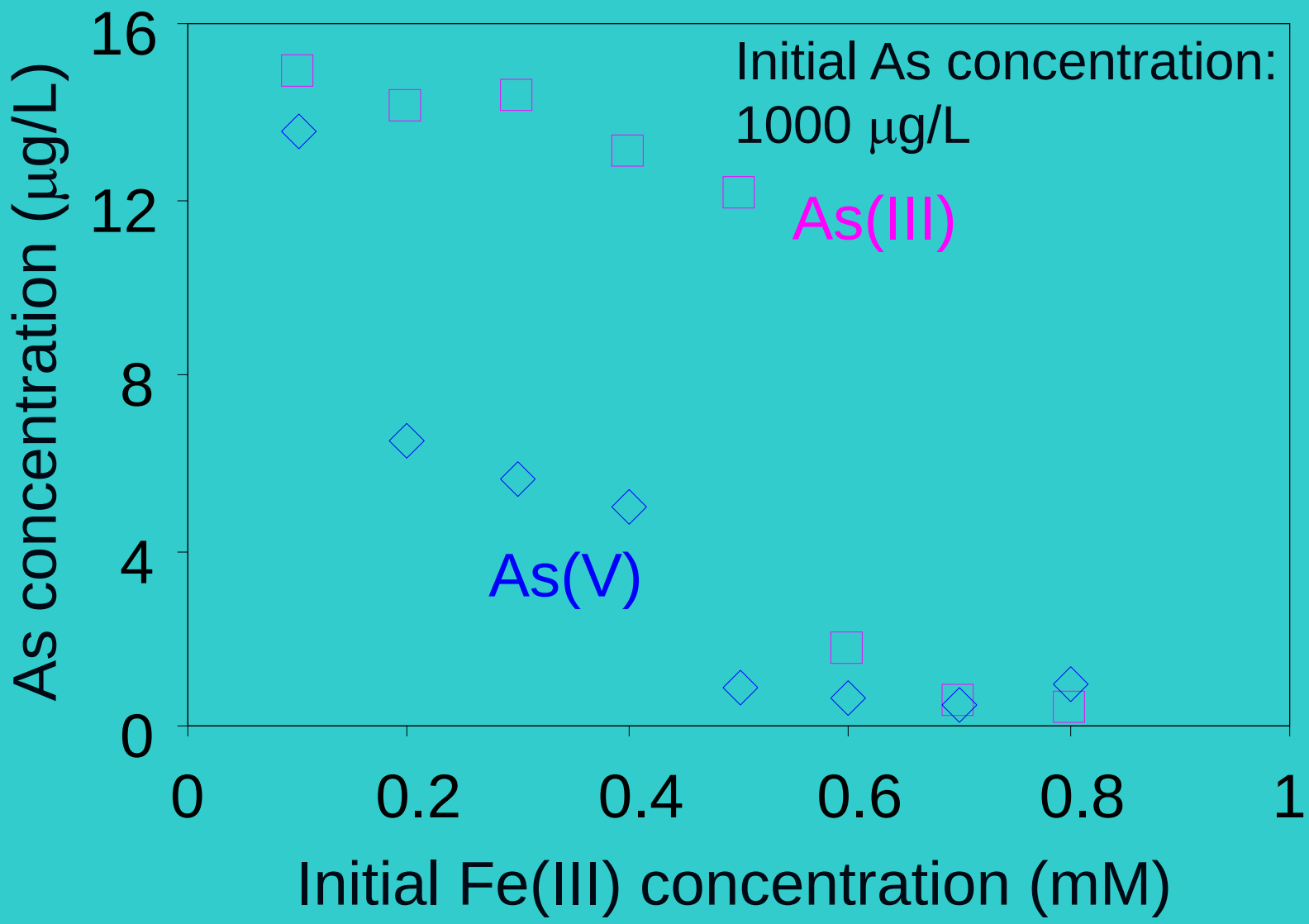
As concentration in solution



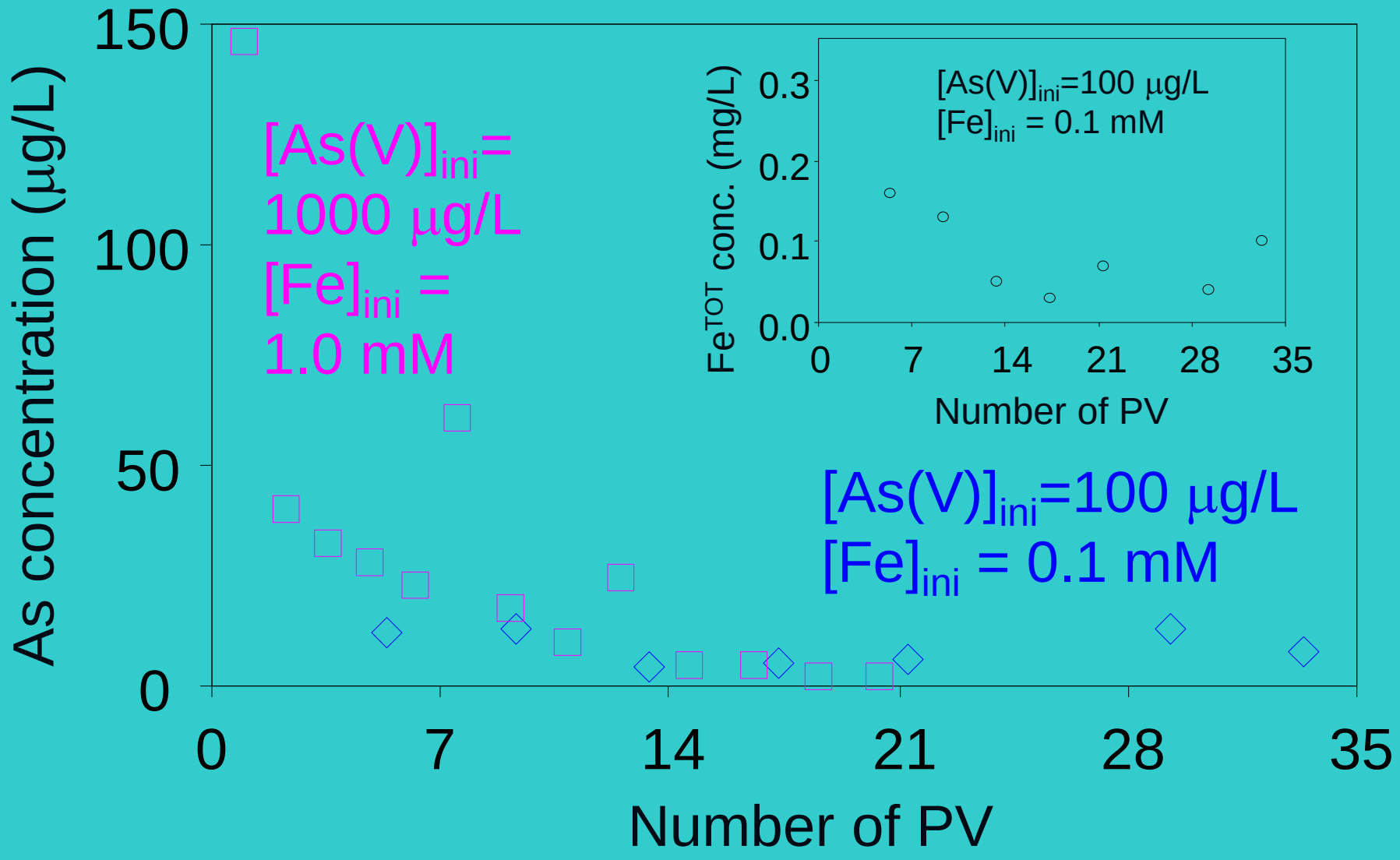
As sorption on Fe(III) precipitates



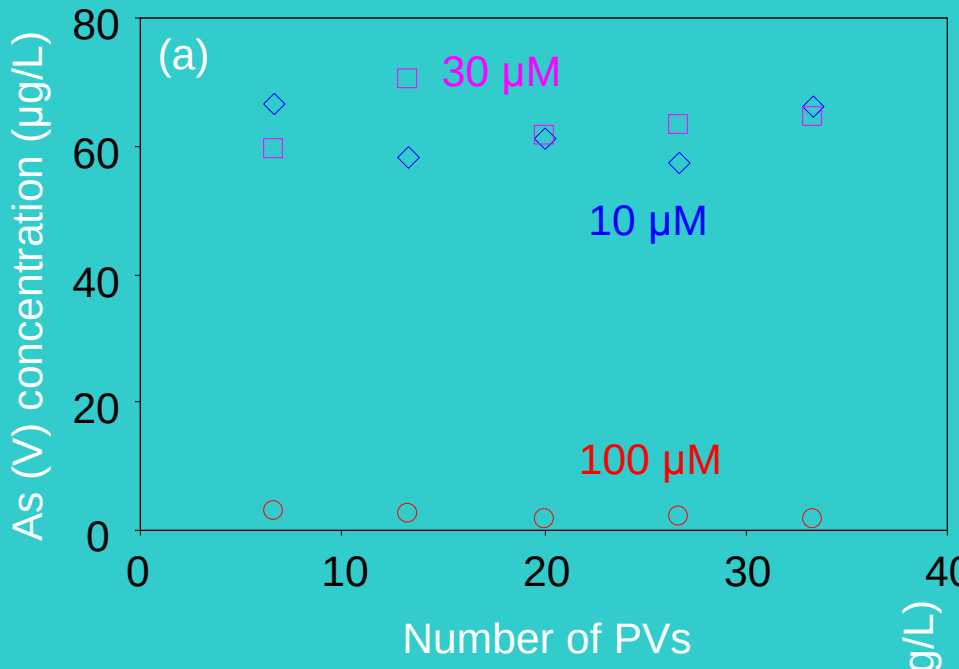
Equilibrium As concentration

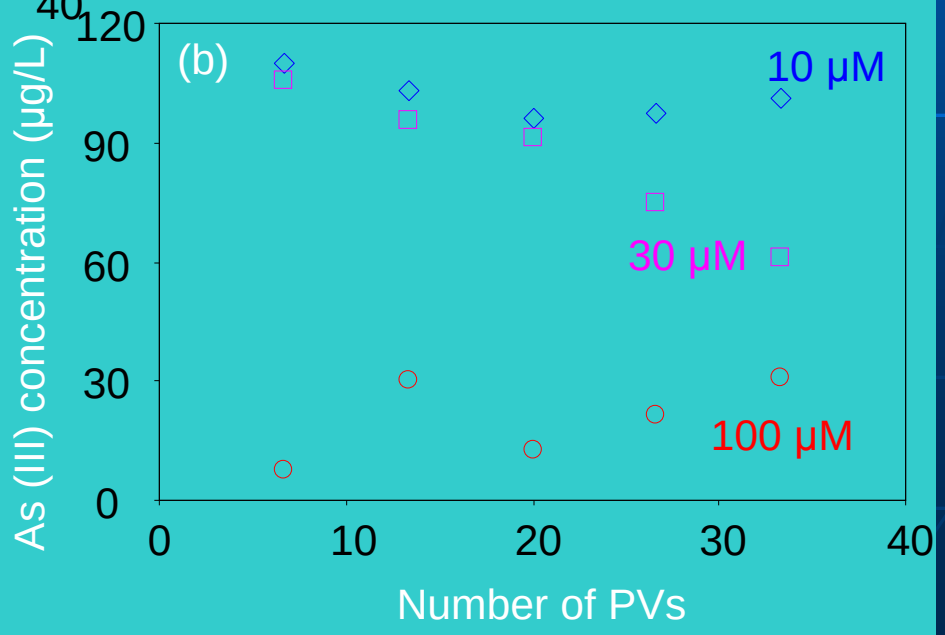


Effluent As concentration



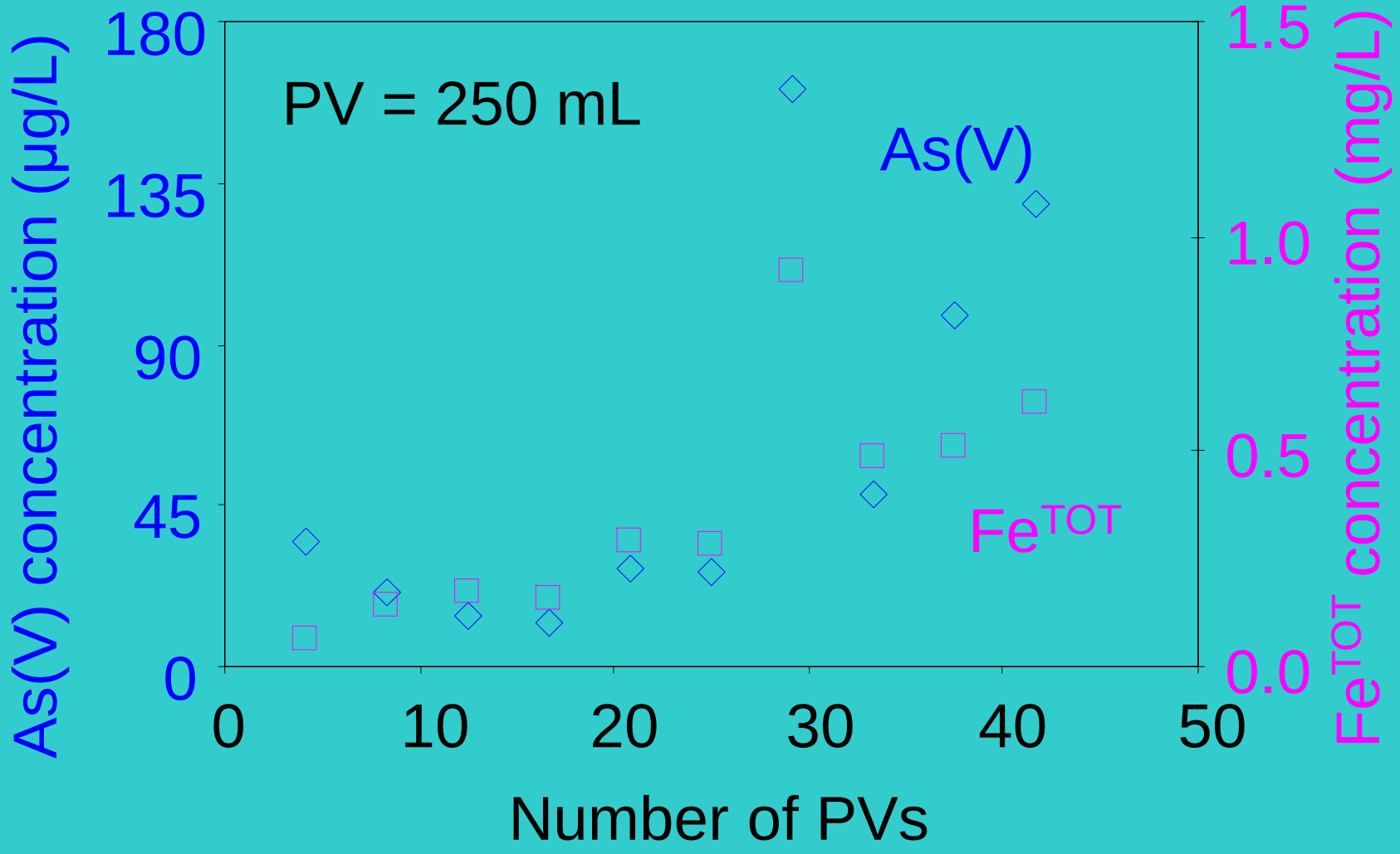
Amount of Fe Usage on As removal



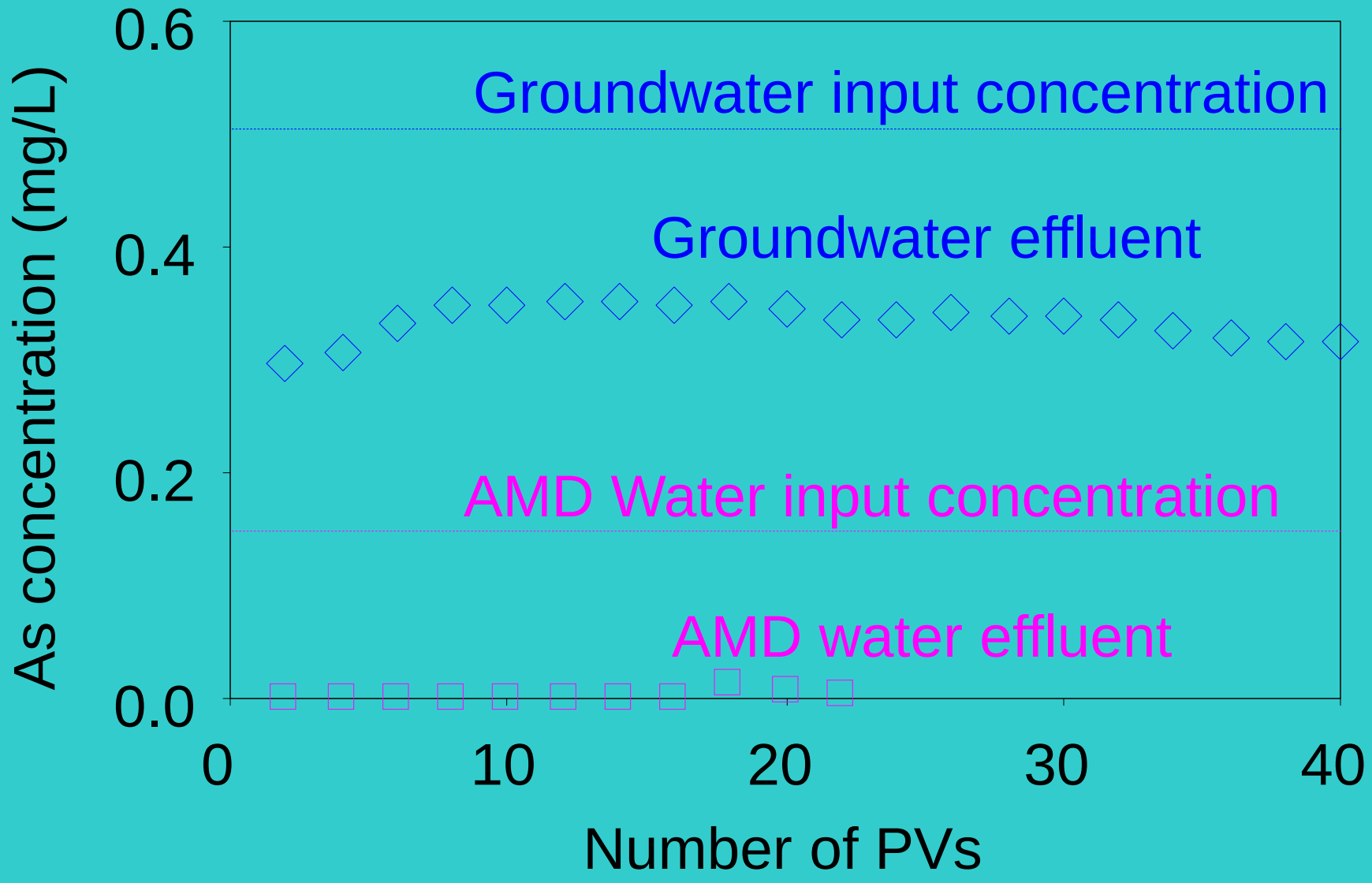


As and Fe concentrations

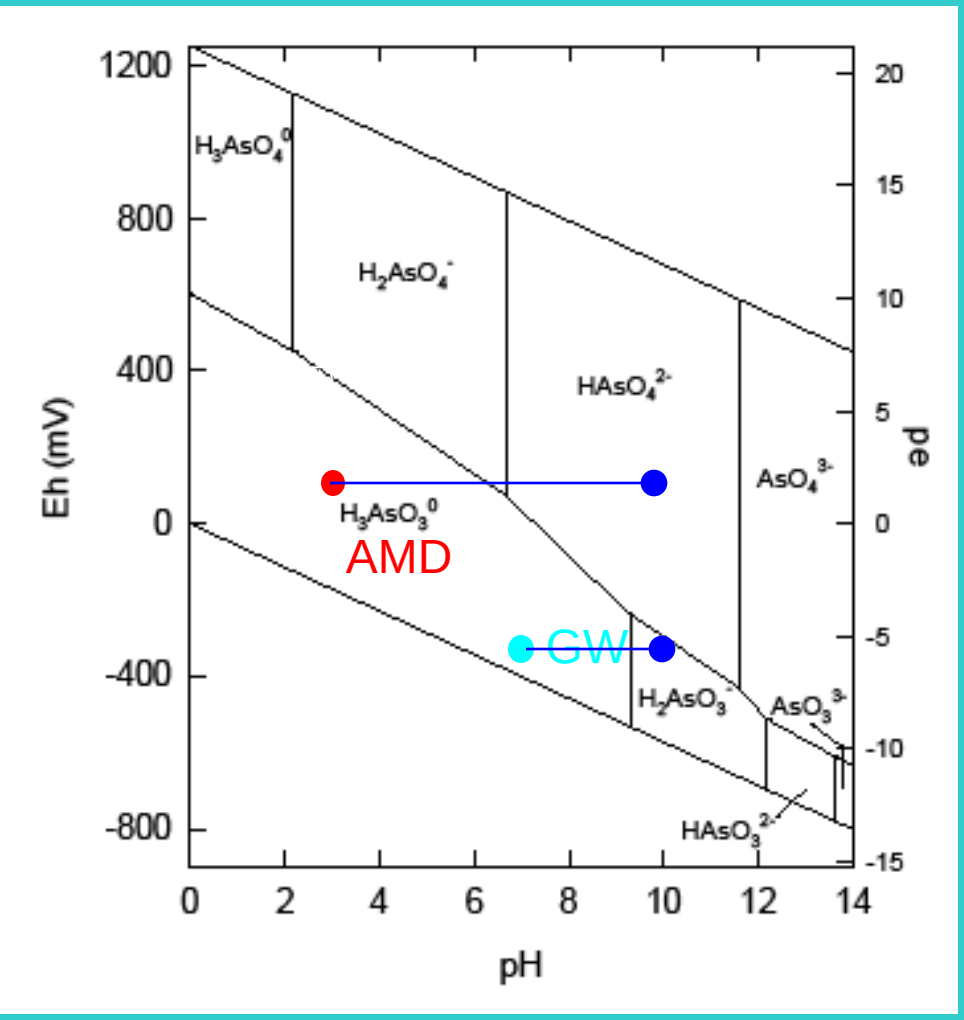
Large column: 28 cm long, 4.5 cm ID, 400 g zeolite

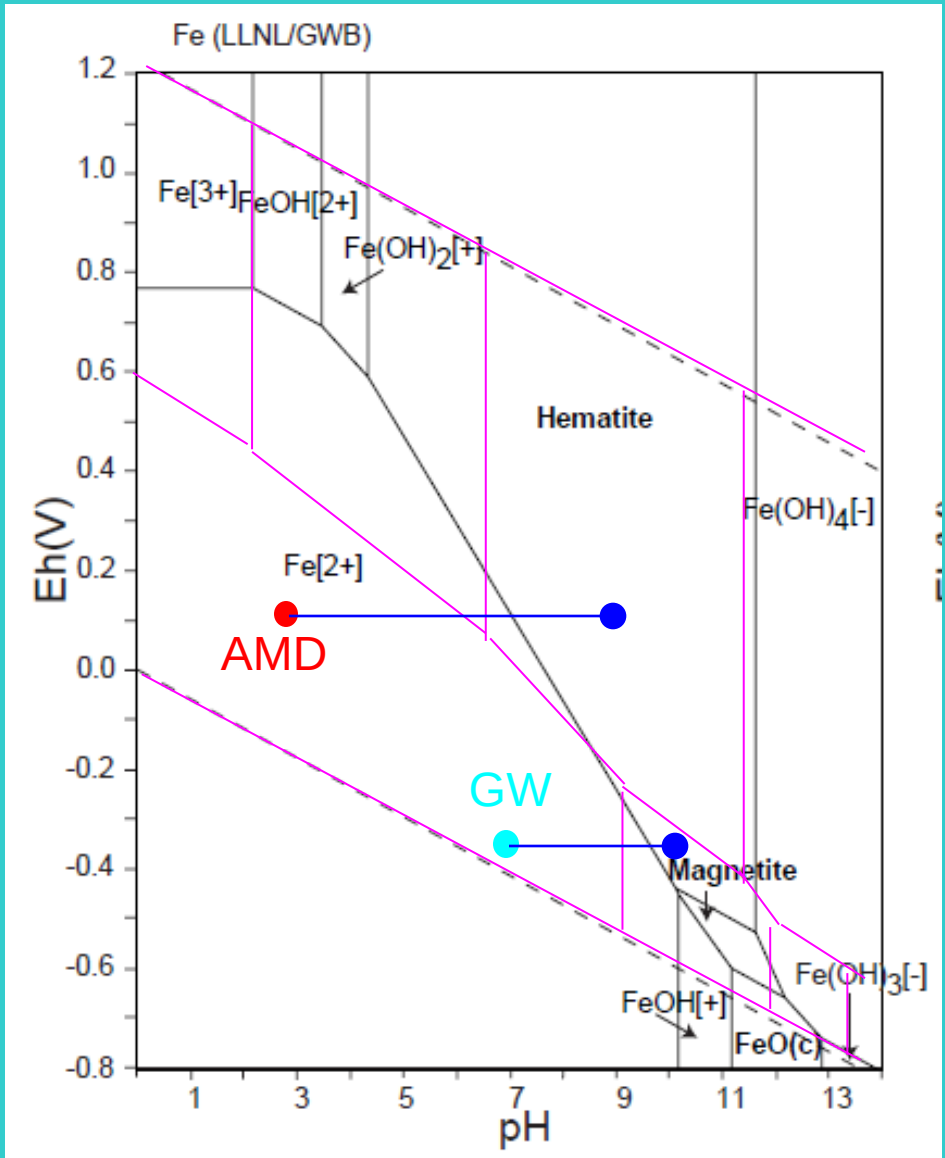


Tests on real water



pH-Eh diagram of As and Fe

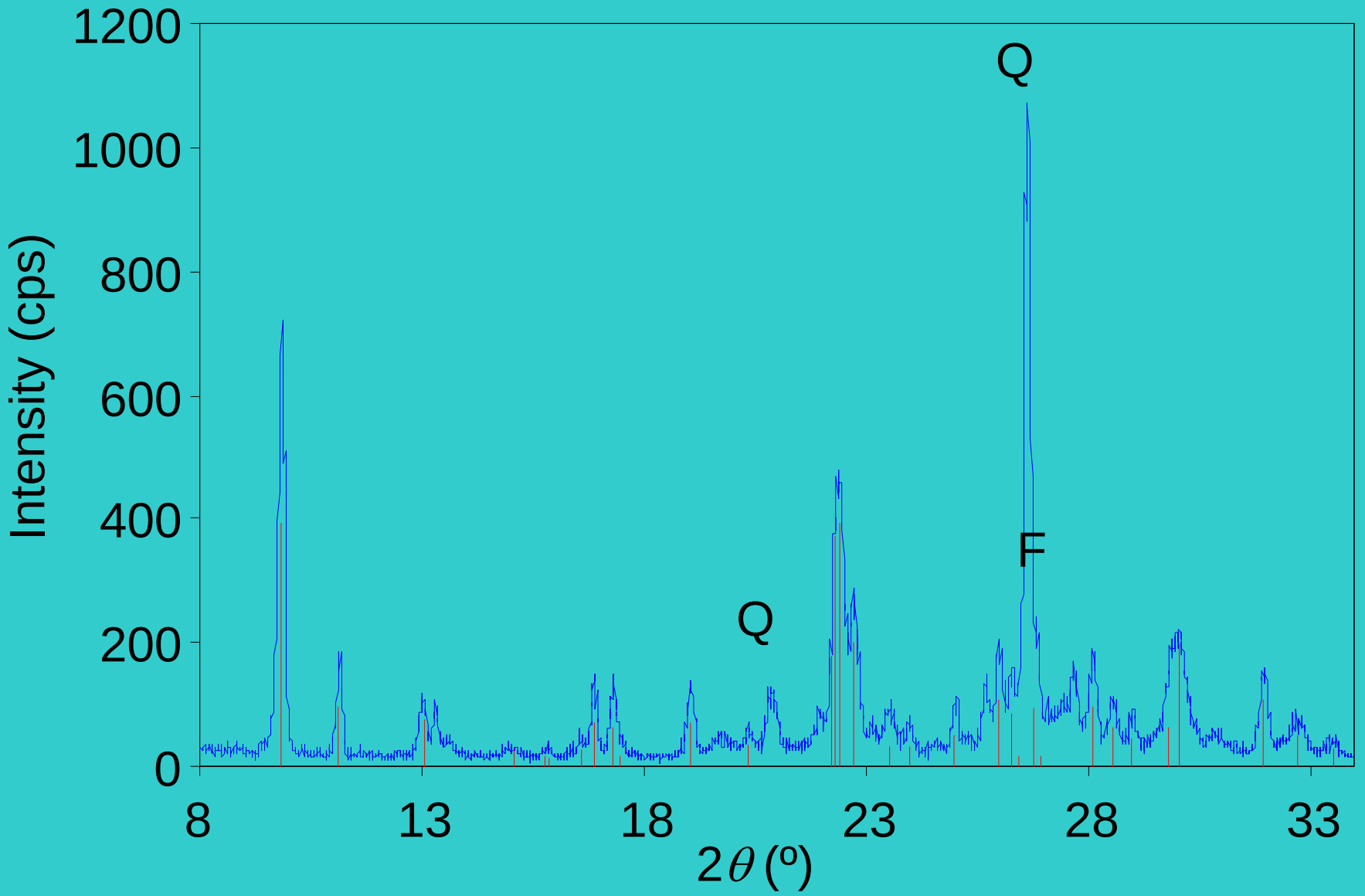




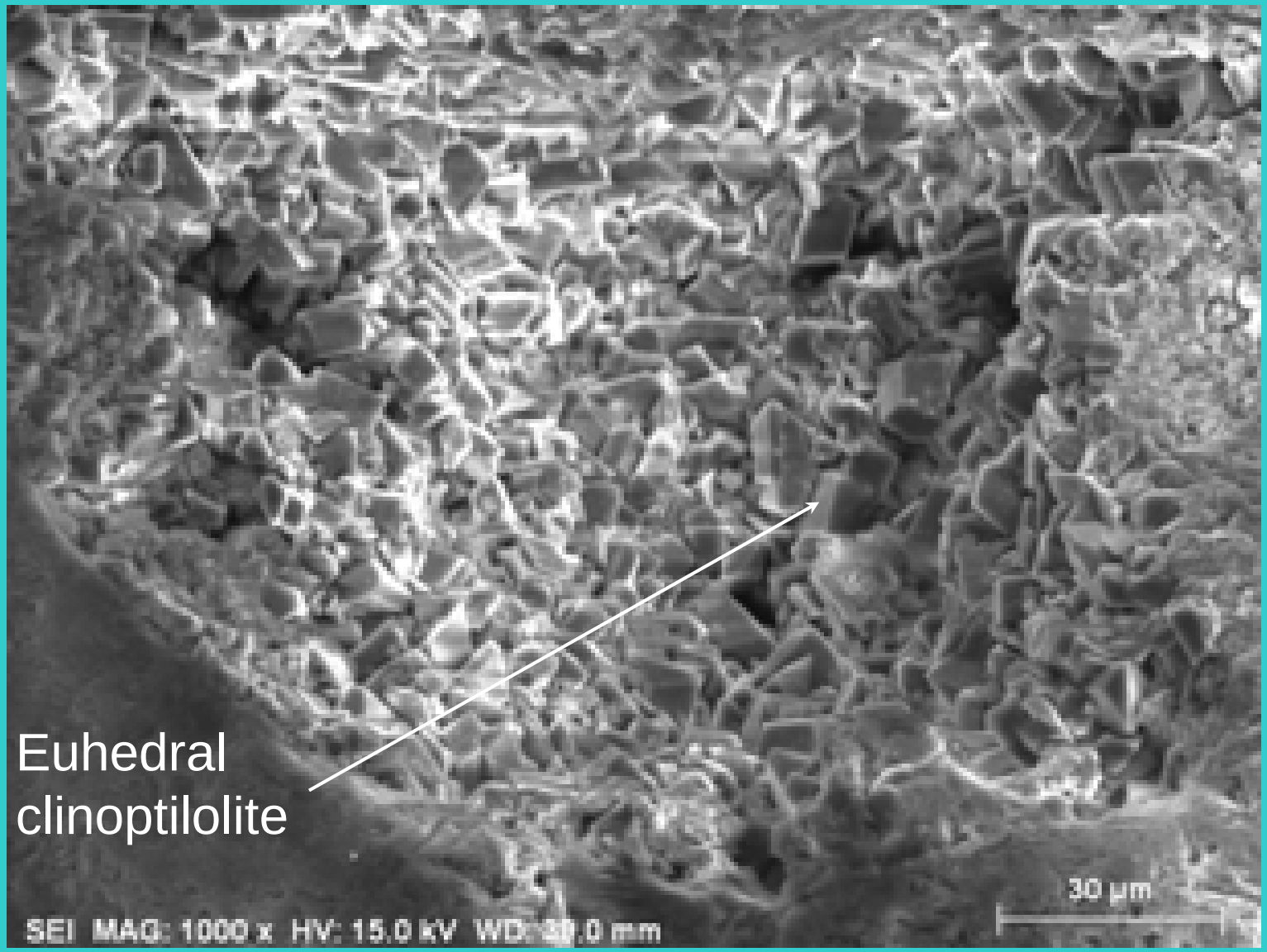
4. Instrumental characterization

- XRD analyses to determine the mineral composition of raw zeolite and the crystalline phases of Fe precipitates
- SEM observation of the morphology of raw zeolite and the Fe precipitates and energy dispersion spectrum determination of chemical composition for Fe and As
- Mossbauer spectrum to determine the state of Fe precipitates

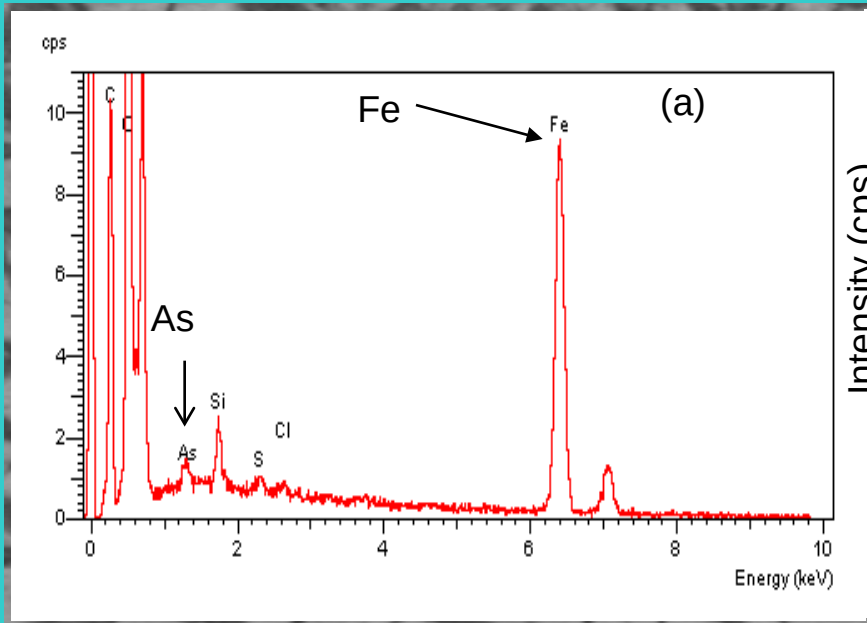
XRD of raw zeolite

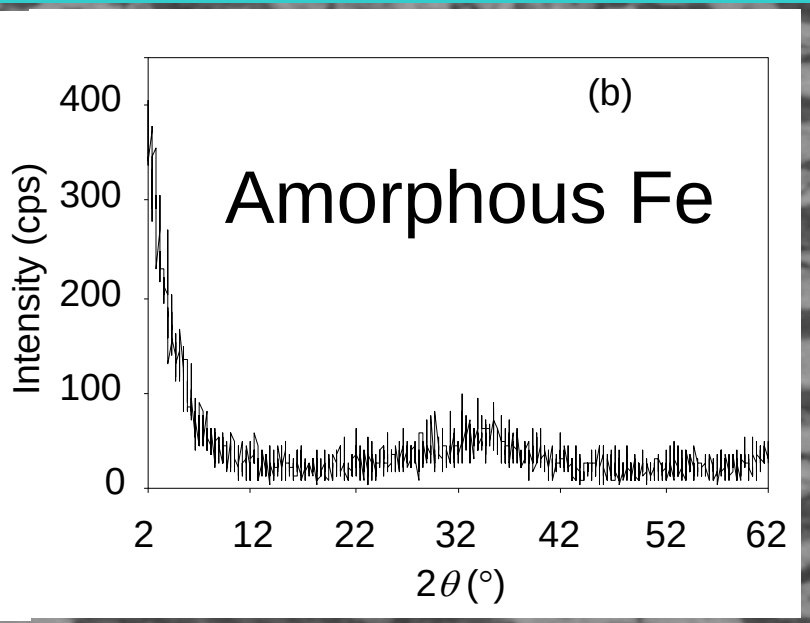


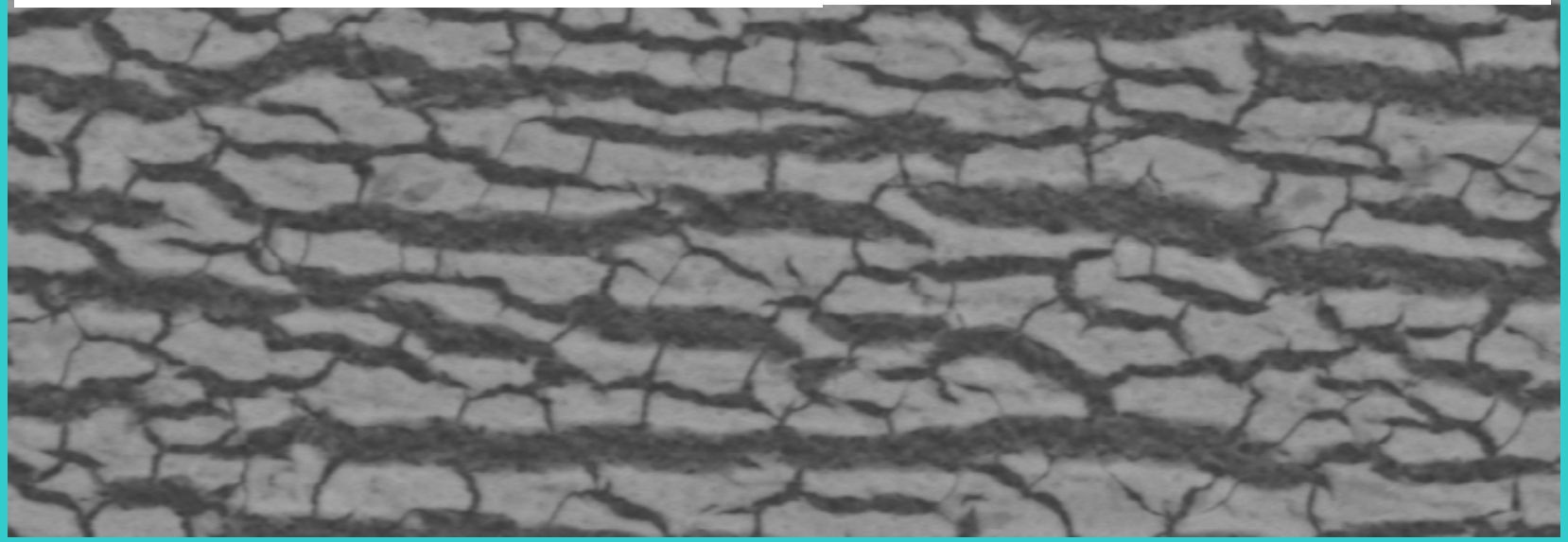
SEM observation



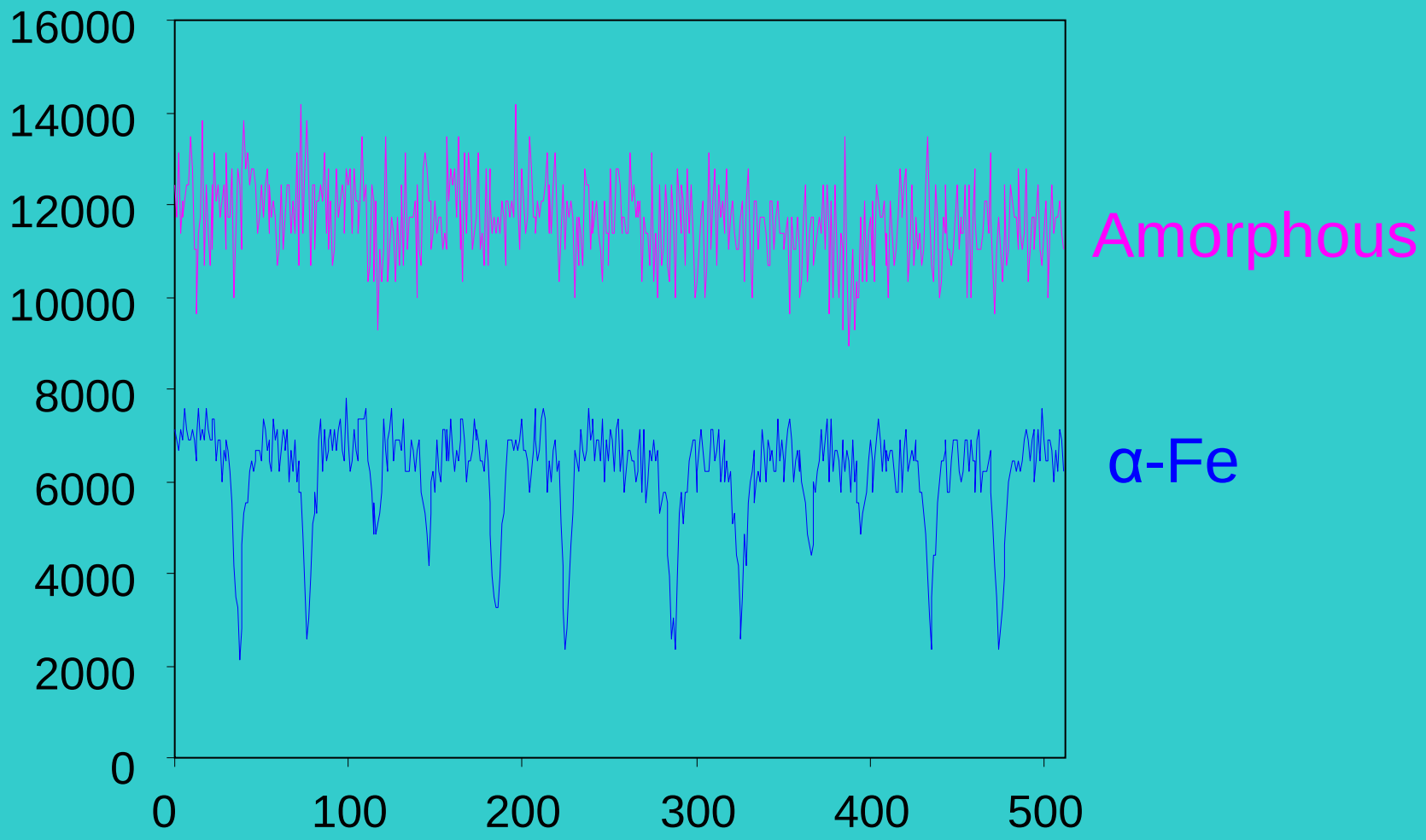
Material Characterization







Mössbauer spectrum



5. Conclusions

- Removal of As from water can be achieved relatively effectively using zeolite as filtration media after HFO precipitation and As sorption onto precipitated HFO
- The As adsorption capacities were 48 and 68 mg/g for As(III) and As(V) adsorption on precipitated HFO, respectively
- The optimal pH for maximum As removal was in neutral range between 6.5 to 9
- Depending on the initial As concentration and redox conditions, the minimal Fe(III) need to reduce the As concentration below standard would be around 10 mg/L
- The method works better for waters under oxygenated condition rather than under reduced conditions

6. Extension of the project

- Use of Fe-exchanged zeolite for the removal of As from water.
- Use of Fe-exchanged zeolite for the removal of Cr from water

Publications

1. Du, G., Li, Z., Hong, H., Ackley*, Leick*, S., Fenske*, N., Demarco, N. (2012) Removal of Cr(VI) from water using Fe(II)-exchanged zeolite. In preparation.
2. Du, G., Li, Z., Liao, L., Hanson*, R., MacDonald*, R., Hoeppner*, N. (2012) Influence of present Fe(III) on Cr(VI) retention and transport through natural zeolite. *J. Hazard. Mater.*, in review.
3. Li, Z., Jiang, W.-T., Jean, J.-S., Hong, H., Liao, L., Lv, G. (2011) Combination of hydrous iron oxide precipitation with zeolite filtration to remove arsenic from contaminated water, *Desalination*, **280**, 203-207.
4. Li, Z., Jean, J.-S., Jiang, W.-T., Chang, P.-H., Chen, C.-J., Liao, L. (2011) Removal of arsenic from water using Fe-exchanged zeolite, *J. Hazard. Mater.*, **187**, 318-323.

Acknowledgment

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Questions?