THEME 4: THE EARTH'S SURFACE: Pollution, climate, anthropogenic effects

Session 4.65:

The fate of arsenic, antimony and similar elements in the environment

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Recent megascale contamination episodes such as the large-scale utilization of arsenic-contaminated water in Bangladesh, Vietnam and the United States have focused the attention of world leaders on the science of micro-pollutant fate and transport. Contamination of waters used for drinking and irrigation by As, antimony, and other trace micropollutants, is increasingly seen as a potential threat to human health and economic growth. The geo-chemical controls over micropollutant fate and transport are in many cases the critical determinant of water useability, suggesting that the international language of water utilization and health will increasingly rely on the language of geochemistry. This symposium seeks to emphasize the richness of that vocabulary by considering all aspects of fate and transport, including field studies, new analytical techniques and experimental results, global cycling measurements and models, and geochemistrybased in situ and ex situ treatment technologies. We encourage the submission of presentations that address these areas but hope for presentations that extend the boundaries of the subject as well. For this reason, we also encourage the submission of presentations that anticipate "the next wave" pollutants - such as uranium, fluoride, and perchlorate.

4.65.11

Arsenic geochemistry in reducing environments – influence of arsenicsulfide interaction on mobility

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Arsenic contamination of ambient waters can lead to significant health concerns if the water is used as a source of drinking water. Therefore, it is important to understand the processes that drive the kinetics and thermodynamics of arsenic release into waters on a molecular level. Modern speciation methods allow us to study the individual species of arsenic in solution (and to some extent in the solid phase) during such mobilization processes, and thereby help us in assessing risks, predicting fate, and developing management, prevention and remediation strategies.

Arsenic speciation in oxic waters is simple, with arsenite and arsenate making up the vast majority of the dissolved arsenic. It is commonly believed that in reducing environments, where many relevant processes leading to arsenic contamination of drinking water take place, arsenic speciation as such does not change, and that mobility is determined exclusively by the binding of arsenite and arsenate to mineral surfaces, particularly iron(oxy)-hydroxides, as well as by the reductive dissolution of such minerals. In this presentation, we will demonstrate that this assumption does not hold true in sulfidic waters or in systems containing sulfide minerals.

We have recently shown [1, 2] that in reducing waters containing free sulfide, a large fraction of the dissolved arsenic can be present in the form of soluble As-S compounds. Up to four currently unidentified As-S species are encountered, and we are in the process of determining whether these are monomeric thioarsenites or thioarsenates. The same species were also shown to play a key role in the alkaline dissolution of arsenic-sulfide minerals [3].

The four members of the homolog thioarsenate series were synthesized and characterized by mass spectrometry. The chemical properties of thioarsenates will be compared to arsenite and arsenate, and implications for environmental cycling and fate will be discussed. Model experiments were conducted to identify formation pathways and decay mechanisms for these species, and will be synthesized into a qualitative model for As speciation in sulfidic environments.

References

- [1] Wilkin R.T., Wallschläger D., and Ford R.G. (2003) *Geochem. Transact.* **4**, 1-7.
- [2] Wallschläger D., Wilkin R.T., and Ford R.G. (2004) *Environ. Sci. Technol.*, submitted.
- [3] Wallschläger D., London, J. and Mitchell, S.M. (2004), in preparation.