

## Sediment response to persistently low oxygen levels in bottom waters: The Lower St. Lawrence Estuary

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Reports of hypoxia ( $O_2$  concentrations  $< 62.5 \mu\text{mol L}^{-1}$ ) in the bottom waters of the coastal ocean, and the extent of the areas that are affected by hypoxia, have increased at an alarming rate. Most studies of hypoxic environments focused on the impacts on pelagic and benthic populations, but the impact of hypoxia on sediment chemistry is poorly documented. We compared the chemical composition of sediment and porewater in cores recovered between 1982 and 2007 at two sites in the Lower St. Lawrence Estuary (LSLE), where the bottom water has been persistently hypoxic for more than 25 years. The concentrations and the vertical distributions of total Fe and As in the cores have not changed since the 1980s, but the speciation of solid phase Fe and As have changed significantly. The proportions of reactive Fe and As components have increased while the degree of pyritization of Fe and As has decreased by 50% and 75%, respectively. In addition, the concentrations of porewater Fe and As have increased since 1982. We propose that in marine, iron-rich environments, such as the LSLE, the hypoxia interferes with pyritization, which normally would immobilize elements such as As by incorporating them into stable authigenic pyrite phases.

## Fate of As upon microbial Fe(III) reduction of As-bearing biogenic Fe minerals

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In arsenic-contaminated groundwater and soil, aqueous arsenic enters the human food chain directly via drinking water or indirectly via plants and animals, potentially leading to devastating health effects on people. Hence, research has focused on the geochemical and biogeochemical processes leading to the mobilization (release) and immobilization (removal) of arsenic from aquifers and soil. Former studies by Hohmann *et al.* [1] demonstrated that Fe(II)-oxidizing bacteria can efficiently immobilize arsenic by forming biogenic Fe(III) hydroxides. In the presence of arsenic these microorganisms form co-precipitates of Fe(III) (hydr)oxides and arsenic and additionally arsenic is sorbed to the mineral surfaces.

However, the co-existence of Fe(II)-oxidizing with Fe(III)-reducing bacteria in the environment could result in Fe cycling by the reduction of biogenic Fe(III) (hydr)oxides. This could lead either to a dissolution of the Fe(III) minerals causing a release of the bound arsenic or alternatively to the formation of secondary Fe(II/III) mineral phases [2] and an immobilization of arsenic.

In this study, we followed the reduction of biogenic arsenic-bearing Fe(III) minerals by the Fe(III)-reducer *Shewanella oneidensis* MR-1. We then compared the As (im-)mobilization during the reduction of these biogenic Fe(III) hydroxides to the As (im-)mobilization from chemically-synthesized Fe(III) minerals including the poorly-crystalline mineral ferrihydrite and the highly crystalline mineral goethite.

The resulting iron minerals were identified and characterized by X-ray diffraction, Mössbauer spectroscopy and electron microscopy. First results show that arsenic is only partially remobilized during microbial reduction of biogenic Fe-As-co-precipitates and that the remaining arsenic is bound either to the non-reduced goethite or to the newly formed Fe (II) carbonate and phosphate mineral phases.

[1] Hohmann *et al.* (2010) *ES&T* **44**, 94-101. [2] Tufano and Fendorf (2008) *ES&T* **42**, 4777-4783.