

Glacial to Interglacial changes in the carbonate ion signature of deep and intermediate water masses in the Southern Ocean

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The Southern Ocean (SO) currently acts as a sink for CO₂, which is transported to intermediate [1] and deep waters [2] and consequently alters the inorganic carbon chemistry there. With regard to the rise in atmospheric pCO₂ since the Last Glacial Maximum (LGM), we aim to reconstruct fluctuations in [CO₃²⁻] in the Circumpolar Deep Water (CPDW) and the Antarctic Intermediate Water (AAIW) in the Australian and Pacific Sector of the SO. For this purpose, B/Ca ratios in benthic foraminiferal shells (*C. wuellerstorfi*, *C. mundulus* and *Uvigerina spp.*) are obtained. Deep water samples from the South Tasman Rise show a clear decrease in average B/Ca ratios from the LGM (43 μmol/mol) towards the Holocene (29 μmol/mol), from which we reconstruct average LGM and Holocene carbonate ion contents of 130 μmol/kg and 81 μmol/kg, respectively. Holocene values are corroborated by data from nearby GLODAP sites and the observed drop in seawater- [CO₃²⁻] is consistent with an increase in atmospheric pCO₂ since the LGM. Reconstructed values from Challenger Plateau sediments (bathed by AAIW) follow the trend observed in deep water cores, showing a decrease in average [CO₃²⁻] from 93 μmol/kg (LGM) to 81 μmol/kg (Holocene). Results from the shallowest core (958 m water depth) however reveal rising [CO₃²⁻], which might reflect location- and depth-related differences in CO₂ uptake and release. To investigate this further, we are currently analysing sediments from the SE of New Zealand, which stem from AAIW and CPDW depths. Moreover, a continuous [CO₃²⁻]- record for the last 20 ka will hopefully enable us to further constrain the temporal relationship between changes in SO carbonate chemistry and the step-wise release of CO₂ into the atmosphere.

[1] Sabine *et al.* (2004) *Science* **305**, 267–371. [2] Sandrini *et al.* (2007) *Antarctic Sci.* **19**, 395–407.

Plutonium transport: Identifying the biogeochemical mechanisms controlling its behavior

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A major challenge in predicting the mobility and transport of actinides is determining the dominant biogeochemical processes that control its behavior in the subsurface. Plutonium (Pu) is of particular concern due to its large worldwide production inventory, high toxicity to human health and long half-life. There is more than 2200 metric tons of Pu throughout the world (ISIS website). The long half-life of Pu (²³⁹Pu ~ 2.4 x 10⁴ years) together with existing inventories guarantees that significant quantities will remain in our environment for a very long time. Effectively managing, controlling, and disposing of these materials in order to stop inadvertent release and transport through the geosphere is a serious scientific challenge. Currently, scientists cannot reliably predict how or how much Pu will move once deposited in the subsurface preventing accurate assessment of risk to human health. The behavior of Pu is complex because the reaction chemistry of Pu (i.e. aqueous speciation, solubility, sorptivity, redox chemistry, and affinity for colloidal particles, both abiotic and microbially-mediated) is particularly complicated. Its migration is known to be oxidation-state dependent and facilitated by transport on particulate matter (i.e. colloidal particles). Yet very little is known about how and under what geochemical conditions colloids facilitate the transport of Pu.

Despite the gaps in our understanding, recent field and laboratory experiments are helping to shape our conceptual understanding. I will summarize our current understanding of the biogeochemical processes controlling Pu transport by discussing recent field and laboratory studies. Field studies include weapons facilities where contamination of large quantities of Pu have been deposited and migrated in the subsurface. These studies demonstrate that colloids can and do play an important role in the transport of low-solubility radionuclides; yet, colloids are not always responsible for transport. It is becoming clear that the depositional history of the contamination, as well as the site-specific hydrogeology is critical to assessing the dominant biogeochemical processes controlling the migration of Pu. In addition, the dominant biogeochemical processes controlling migration of Pu can change along the flow path from the source to the far-field environment.