

## Sediment-water exchange on the Georgia mid-shelf

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In permeable sediments, exchange of near-interface porewater constituents with the overlying water is driven primarily by advective movement of water through the sand matrix. As part of BOTTOMS-UP, we have developed novel independent and non-invasive methods for time series sampling of porewater constituents and for estimation of time- and depth-dependent fluid exchange rates within sediments (Figures 1). We have combined these data within advection-diffusion models to make realistic estimates of *in situ* respiration and methanogenesis rates in permeable shelf sands.

Advection rates are derived from modelling propagation of temperature changes in the overlying water into the sediment. Porewater concentrations of metabolically active gases (O<sub>2</sub>, CO<sub>2</sub>, N<sub>2</sub>, and CH<sub>4</sub>) are obtained by repeatedly profiling the sediment with a newly developed sampling probe for an underwater Membrane Inlet Mass Spectrometer uMIMS [1].

Oxygen consumption rate (-17.7 mmol m<sup>-2</sup> day<sup>-1</sup> obtained by this method agree closely with core incubation data [2]. Advective exchange between the sediments and water column on the shelf is deep and rapid, even in the absence of significant topography. Forcing of exchange appears to be driven primarily by tides. Summertime cooling of the interface due to large-scale intrusion of cold bottom water onto the shelf may lead to enhanced flushing of porewater due to unstable thermal convection.

[1] Bell *et al.* (2007) *Environ. Sci. Technol.* **41**, 8123–8128.

[2] Jahnke *et al.* (2005) *Cont. Shelf. Res.* **25**, 433–452.

## CO<sub>2</sub> dissolution kinetics studied by Raman spectroscopy

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Numerical models predict that anthropogenic CO<sub>2</sub> emissions will increase global temperatures and acidify the ocean to levels that will impact human activities by the end of the 21<sup>st</sup> century. One strategy to mitigate future CO<sub>2</sub>-related climate change is the sequestration of CO<sub>2</sub> in marine sediments. At typical oceanic conditions, CO<sub>2</sub> is denser than seawater at water depths >3000 m. Here, the liquid CO<sub>2</sub> forms gravitationally stable pools either on the ocean floor or in the sediment pore space. Assessing the suitability of these two regimes of anthropogenic CO<sub>2</sub> disposal requires accurate knowledge of the *in situ* physicochemical processes triggered by the injection of liquefied CO<sub>2</sub> in sediments.

We carried out high pressure laboratory experiments to investigate the kinetics of dissolution and transport of CO<sub>2</sub> into a) pure water and b) water-saturated sediment. A capillary filled with water or water-saturated sediment was used to mimic natural conditions. The capillary was exposed to liquefied CO<sub>2</sub> in a pressure cell. The transient concentration of CO<sub>2</sub> dissolved in water was monitored by non-invasive Raman Spectroscopy. The rates of dissolution and transport were constrained with a transport-reaction model which considers both the kinetics of dissolution of CO<sub>2</sub> from a pure CO<sub>2</sub> phase into water and the transport of dissolved CO<sub>2</sub> away from the liquid CO<sub>2</sub>-water interface.

We observed two distinct behaviours with regards to the transport mechanism of dissolved CO<sub>2</sub> in water: in the case of the water-saturated sediment, the model reproduced the CO<sub>2</sub> Raman data by considering molecular diffusion as the sole transport mechanism; in the pure water set-up, however, the model highlights an enhanced rate of CO<sub>2</sub> transport in water, implying that another transport process is present in addition to molecular diffusion. We propose that the presence of sand in the capillary inhibits the small-scale convective processes that enhance transport of CO<sub>2</sub> in the absence of sand.

Enhanced transport was modelled numerically by adding an eddy diffusion term to the differential equation describing the transport of CO<sub>2</sub> in water. The eddy diffusion coefficient we constrained in the set-up with water-saturated sediment is 30 times larger than the molecular diffusion coefficient of CO<sub>2</sub> in pure water. This result highlights the importance of understanding the process of CO<sub>2</sub> dissolution and transport in the context of marine CO<sub>2</sub> sequestration.