Effects of CO_2 leakage on benthic biogeochemistry – Results from a large scale field experiment

ANNA LICHTSCHLAG[1*], DOUG CONNELLY[1], HENRIK STAHL[2], PETE TAYLOR[2] AND RACHAEL H. JAMES[1]

[1]National Oceanography Centre, University of Southampton Waterfront Campus, European Way, Southampton, SO14 3ZH, UK,
correspondence: alic@noc.ac.uk
[2] Scottish Association for Marine Sciences, Oban, Argyll PA37 1QA, UK

Over the last few decades subseafloor Carbon Capture and Storage (CCS) has been recognized as having great potential as an effective and safe method to reduce anthropogenic CO_2 emissions. However, the impact of any CO_2 seepage from a storage site on the marine environment is poorly understood. Released CO_2 will potentially dissolve into sediment pore waters, increasing the concentration of total dissolved carbonate and decreasing pH. As a consequence, metals may be released into pore waters and potentially into the water column. The effects of these changes to pore water chemistry on benthic ecosystems needs to be assessed.

To this end, we have analysed the chemical composition of pore waters, sediments, and the water column during a CO_2 release experiment that we carried out in Ardmucknish Bay, Scotland, as part of the 'Quantifying and Monitoring Potential Ecosystem Impacts of Geological Carbon Storage' (QICS) project. CO_2 gas was injected into the sediment 12m below seafloor at a known flow rate through a borehole drilled through the underlying bedrock. Sediment cores and water column samples were taken before, during and after the CO_2 release from 3 sites, at varying distances from the injection point.

Our data demonstrate that injected CO_2 was detected in the pore waters within 30 cm of the sediment-seawater interface within 33 days after the start of the CO_2 injection, but only at the site located closest to the injection point. As the CO_2 dissolved, concentrations of dissolved inorganic carbon (DIC) and total alkalinity (TA) increased and the $\delta^{13}C$ of the DIC decreased, reflecting the low $\delta^{13}C$ value of the injected CO_2 gas ($\delta^{13}C$= -26.6 ‰). Highest DIC concentrations (up to 30 mmol L^{-1}, compared with background levels of ~ 2.2 mmol L^{-1}) were observed 5 days after the injection had stopped. Pore waters with highest DIC concentrations also had enhanced concentrations of cations (e.g. iron, calcium) and nutrients. However, concentrations of all pore water consituents had returned to background values 18 days after the CO_2 injection was stopped.

Paired Sr Isotope ($^{87}Sr/^{86}Sr$, $\delta^{88/86}Sr$) Systematic of Sulfates and Pore Waters: New Perspectives in Marine Weathering, Seepage Signatures and Fractionation Processes

V. LIEBETRAU[1*], M. HAECKEL[1], A. EISENHAUER[1], H. VOLLSTAEDT[1], L. HAFFERT[1] AND C. HENSEN[1]

[1]Helmholtz Centre for Ocean Research Kiel (GEOMAR), Germany (*vliebetrau@geomar.de)

The simultaneous but independent determination of the conventional normalized radiogenic ($^{87}Sr/^{86}Sr$) and the fractionation reflecting stable ($\delta^{88/86}Sr$) Sr isotope ratio on pore waters, sediments and precipitates (e.g. carbonates and sulfates) opens a new perspective in the field of marine weathering and Sr contribution to the ocean chemistry.

As established tool in provenance studies on silicates and SIS-geochronology (Sr-Isotope-Stratigrapghy) of marine carbonates the radiogenic $^{87}Sr/^{86}Sr$ signature is in our approach directly combined with the isochemical stable Sr ($\delta^{88/86}Sr$) fingerprint of isotope fractionation, mainly due to water/rock-interaction and secondary precipitation processes.

One of our initial case studies, includes gypsum bearing sediments of active mud volcanoes (MV) and mounds from the Gulf of Cadiz (GoC). The Mercator MV is characterised by high $\delta^{88/86}Sr$ ratios of 0.72‰ for small ideomorphic (mm scale, most probably authigenic) and up to 0.92 ‰ for large (cm scale) heterogeneous gypsum crystals. The related pore water profile reflects also elevated signatures from around 0.7 ‰ at the crystals to 0.52 ‰ close to the sediment surface at $^{87}Sr/^{86}Sr$ ratios around 0.7105. Referred to NIST-SRM-987 the IAPSO seawater (SW) standard of this study plots on 0.39 ‰ (±0.03, 2SD) in $\delta^{88/86}Sr$ and on 0.70917(1) in $^{87}Sr/^{86}Sr$.

Potential explanation for the observed trend and PW signatures heavier than SW are (a) strong fractionation processes enriching light isotopes in secondary precipitates and remineralisation products and/or (b) preferential dissolution of mineral phases high in $\delta^{88/86}Sr$.

In order to develop a better understanding of isotope fractionation processes we conducted experiments providing contrary Sr and Ca isotope systematics for the precipitation of gypsum.

Whereas the precipitate is enriched in ^{88}Sr by more than 0.1 ‰ when compared to the initial fluid source, the $\delta^{44/40}Ca$ isotope signature is depleted as known for carbonates.

In contrast, Sr of biogene $SrSO_4$ dominated sulfates of acantharia follows the Ca fractionation systematic with a strong depletion of 0.3‰ in $\delta^{88/86}Sr$ compared to seawater.

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