Geochemistry Group
Research in Progress Meeting

GGRiP 2019

University of Portsmouth
16-17th April 2019
Dear Delegate,

Welcome to the Geochemistry Group Research in Progress Meeting (GGRiP) 2019!

The Group committee and the local organizers acknowledge our industrial partners: Sercon, OEA Labs, Metrohm, Cameca, Savillex, Teledyne, Agilent, SciMed, QMX and Thermo Scientific for their generous support of this meeting.

We hope you enjoy your visit to Portsmouth.

Craig Storey
Local organiser, on behalf of the GG committee
## MONDAY 15TH APRIL

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<tr>
<td>Public Lecture</td>
<td>Craig Storey</td>
<td>1</td>
<td>17:30</td>
<td>Parrish, R</td>
<td>University of Portsmouth</td>
<td>Keynote</td>
<td>Searching for the environmental smoking gun of Gulf War Illness</td>
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<tr>
<td>Icebreaker</td>
<td>Craig Storey</td>
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## TUESDAY 16TH APRIL

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<tr>
<td>Arrival and Reg.</td>
<td>Craig Storey</td>
<td>-</td>
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<td>Coffee</td>
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<td>Year of Carbon</td>
<td>Gordon Inglis</td>
<td>6</td>
<td>10:00</td>
<td>Badger, M.P.S.</td>
<td>The Open University</td>
<td>Keynote</td>
<td>Fixing the alkenone palaeobarometer</td>
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<td>7</td>
<td>10:30</td>
<td>Babila, T.L. &amp; Foster, G.L.</td>
<td>University of Southampton</td>
<td>-</td>
<td>The Monterey Event and the Paleocene-Eocene Thermal Maximum - two contrasting oceanic carbonate system responses to LIP emplacement and eruption</td>
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<tr>
<td>Method/Technique</td>
<td>Marie-Laure Bagard</td>
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<td>11:30</td>
<td>Mottmann, C.</td>
<td>University of Portsmouth</td>
<td>Keynote</td>
<td>Directly dating deformation with calcite U-Pb dating: the good, the bad and the ugly!</td>
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<td>10</td>
<td>12:00</td>
<td>Bevan, D.G., Coath, C.D., Lewis, J. &amp; Elliott, T.</td>
<td>University of Bristol</td>
<td>Student</td>
<td>In situ Rb-Sr dating: Triple Quad v Proteus</td>
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<td>Poster Intro</td>
<td>Susan Little</td>
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<td>12:30</td>
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<td>Lunch &amp; Posters</td>
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<tr>
<td>Investigating</td>
<td>Craig Storey</td>
<td>12</td>
<td>14:30</td>
<td>Wade, J.</td>
<td>Oxford University</td>
<td>Keynote</td>
<td>Why is Earth habitable and Mars like the solar system’s worst pub?? (*no atmosphere, no life and dry)</td>
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<td>Planetary Interiors</td>
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<td>13</td>
<td>15:00</td>
<td>Clarke, E., De Hoog, J.C.M., Kirstein, L.A., Harvey, J. &amp; Debret, B.</td>
<td>University of Edinburgh</td>
<td>Student</td>
<td>Boron isotopes in metamorphic olivine record external fluid infiltration during serpentinite dehydration in subduction zones</td>
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<td>14</td>
<td>15:15</td>
<td>Villalobos-Orchard, J., Freymuth, H., O’Driscoll, B. &amp; Willbold, M.</td>
<td>University of Manchester</td>
<td>Student</td>
<td>Tracing subduction zone fluids in Izu arc lavas using molybdenum isotopes</td>
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<td>Contamination</td>
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<td>16</td>
<td>16:15</td>
<td>Dean, J., Srivastava, P. &amp; Perkins, B.</td>
<td>Aberystwyth University</td>
<td>Student</td>
<td>The determination of the sources and cycling of Ni and associated elements in an industrialised floodplain, South Wales</td>
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<td>17</td>
<td>16:30</td>
<td>Valiteriano, C.M., Neumann, R., Alkim, A.R., Evangelista, H., Hellbron, M., Neto, C.C.A. &amp; Guerra, L.</td>
<td>University of Salzburg</td>
<td>-</td>
<td>Sm-Nd and Sr isotope fingerprinting of iron mining tailing deposits from the failed SAMARCO Fundão dam 2015 accident at Mariana, SE-Brazil</td>
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**Posters and Wine Reception**: 17:00
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<tr>
<td>Marine Biogeochemistry</td>
<td>Susan Little</td>
<td>18</td>
<td>09:30</td>
<td>Lohan, M.</td>
<td>University of Southampton</td>
<td>Keynote</td>
<td>Hydrothermal vents an important source of iron and zinc to the ocean interior</td>
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<td>19</td>
<td>10:00</td>
<td>Crocket, K.C., Baer, T., Abell, R.E., Beveridge, C., Hughes, D. &amp; Halthorne, E.</td>
<td>University of Edinburgh</td>
<td>Rare earth elements in marine biogenic carbonate as proxies for the marine carbonate system?</td>
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<td>21</td>
<td>10:30</td>
<td>Packman, H., Little, S.H., Rehkmmer, M., van De Flierdt, T., Bridgestock, L. &amp; Baker, A.</td>
<td>Imperial College London</td>
<td>Student</td>
<td>The isotope compositions of Zn and Cu in atmospheric aerosols and their role in biogeochemical cycles</td>
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<td>10:45</td>
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<td>Earth Surface Processes</td>
<td>Chris Pearce</td>
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<td>Tostevin, R.</td>
<td>Oxford University</td>
<td>Keynote (Award)</td>
<td>Microbial evolution in a ferruginous, silica-rich ocean</td>
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<td>23</td>
<td>11:45</td>
<td>Murphy, M.J., Stevenson, E.I. &amp; von Strandmann, P.A.E.</td>
<td>University College London</td>
<td>Impact of permafrost thaw on chemical weathering in the high Arctic Zackenberg River, NE Greenland</td>
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<td>24</td>
<td>12:00</td>
<td>Pogge von Strandmann, P.A.E., Jones, M.T., Schmidt, D.N. &amp; Murphy, M.J.</td>
<td>University College London</td>
<td>Lithium isotope evidence for weathering changes across the PETM</td>
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<td>AGM</td>
<td>Chris Pearce</td>
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<td>Crustal Evolution and Magmatic Processes</td>
<td>Marc-Alban Millet</td>
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<td>13:30</td>
<td>Saji, N.S.</td>
<td>Cardiff University</td>
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<td>Hadean geodynamics inferred from time-varying $^{142}$Nd/$^{144}$Nd in the early Earth rock record</td>
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<td>14:00</td>
<td>Moreira, H., Storey, C., Fowler, M., Seixas, L. &amp; Dunlop, J.</td>
<td>University of Portsmouth</td>
<td>Student</td>
<td>Latest TTG to sanukitoid transition: implications for the evolution of the continental crust during the magmatic lull</td>
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<td>Stubbs, D., Coath, C.D. &amp; Elliott, T.</td>
<td>University of Bristol</td>
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<td>Tungsten in Mariana arc basalts: evidence for mobility and isotopic fractionation during subduction</td>
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<td>Brown, R., Chalk, T., Wilson, P., Rohling, E. &amp; Foster, G.</td>
<td>University of Southampton</td>
<td>Yes</td>
<td>High Resolution CO₂ Record of the First Great Glaciation Using Boron Isotopes</td>
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<td>Chalk, T.B., Hain, M.P., Foster, G.L., Nuber, S., Rohling, E.J. &amp; Wilson, P.A.</td>
<td>University of Southampton</td>
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<td>Atmospheric CO₂ change over the past 1.5 million years</td>
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<td>Chen, L., Little, S.H., Kreissig, K., McManus, J. &amp; Severman, S.</td>
<td>Imperial College London</td>
<td>Yes</td>
<td>Isotopically light cadmium in organic-rich sediments</td>
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<td>Crocket, K.C.</td>
<td>University of Edinburgh</td>
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<td>NERC Changing Arctic Ocean: Implications for marine biology and biogeochemistry</td>
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<td>Crompton-Banks, J.G., Burke, A., Greenop, R., Rae, J.W.B. &amp; Mackenzson, A.</td>
<td>University of St Andrews</td>
<td>Yes</td>
<td>Boron isotope evidence for Southern Ocean stratification at the onset of the last glacial period</td>
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<td>Freymuth, H., Williams, H., Jenner, F. &amp; Baptiste, D.</td>
<td>University of Cambridge</td>
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<td>Copper isotope constraints on Cu enrichment in arc magmas</td>
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<td>Groome, N.T., Bucha, D.M., Fagereng, A., Wood, M., Campbell, S. &amp; Horak, J.M.</td>
<td>Cardiff University</td>
<td>Yes</td>
<td>Geochemical discrimination of metabasites from a Neoproterozoic accretionary complex in Anglesey, North Wales</td>
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<td>Harvey, J., Konig, S., Ackerman, L. &amp; Georgiev, B.</td>
<td>University of Leeds</td>
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<td>Siderophile and chalcophile element insights into the petrogenesis of pyroxenite xenoliths from Kilbourne Hole, NM: exploring a potential genetic relationship with the Potrillo Volcanic Field</td>
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<td>Holder, L., Lambelet, M., Plancherel, Y., van de Flierdt, T., McKay, R.M., De Santis, L., Kulhanek, D. &amp; the IODP Expedition 374 Scientists</td>
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<td>Yes</td>
<td>Reevaluating the neodymium isotopic signature of Ross Sea Bottom Water – Implications for paleoceanographic reconstruction</td>
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<td>Lipp, A.G., Shorttle, O. &amp; Syvret, F.</td>
<td>Imperial College London</td>
<td>Yes</td>
<td>A model for fine-grained sediment major element composition in terms of weathering and provenance</td>
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<td>Littley, E., Rae, J., Burke, A., Greenop, R., Vogel, H. &amp; Thornalley, D.</td>
<td>University of St Andrews</td>
<td>Yes</td>
<td>CO₂ perturbations during rapid climate change. What is the role of the North Atlantic?</td>
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<td>Marschalek, J., van de Flierdt, T., Carter, A., Vermeesch, P., Siegert, M., Licht, K., McKay, R.M., De Santis, L., Kulhanek, D. &amp; the Expedition 374 Scientists</td>
<td>Imperial College London</td>
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<td>A Multi-Proxy Sediment Provenance Record of Antarctic Ice Sheet Change in the Early to Middle Miocene: Preliminary Results from IODP Site U1521 (Ross Sea)</td>
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<td>Mulders, J.J.P.A. &amp; Oelkers, E.H.</td>
<td>University College London</td>
<td>Yes</td>
<td>An experimental study of sepiolite dissolution rates and mechanisms at 25°C</td>
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<td>Murphy Quinlan, M.E., Muller, T., Walker, A., Mound, J. &amp; Harvey, J.</td>
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<td>Yes</td>
<td>Deciphering planetary processes on the microscale</td>
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<td>Pickard, H., Rehkamper, M., Palk, C. &amp; Wood, B.J.</td>
<td>Imperial College London</td>
<td>Yes</td>
<td>The Origin of Earth’s Volatiles</td>
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<td>Savage, P.S., Traill, D. &amp; Moynier, F.</td>
<td>University of St Andrews</td>
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<td>Calibration of Si isotope fractionation between quartz and zircon</td>
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<td>Scharnrock, M., Freymuth, H. &amp; Williams, H.M.</td>
<td>University of Cambridge</td>
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<td>Do Molybdenum isotopes of Gorgona komatitites trace deeply subducted oceanic lithosphere?</td>
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<td>51</td>
<td>Ward, J., Freitas, S.F., Hendry, K., Arndt, S. &amp; the CHAOS Team</td>
<td>University of Bristol</td>
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<td>Atlantification of the Barents Sea and implications for Benthic-Pelagic coupling</td>
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<td>Weiss, L., Mottram, C., Parrish, R. &amp; Darling, J.</td>
<td>University of Portsmouth</td>
<td>Yes</td>
<td>Timing of brittle deformation in the Jura mountains revealed by U-Pb dating on calcite</td>
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<td>Wilson, D.J., Pogge von Stradmann, P.A.E., Tarbuck, G., White, J. &amp; Hopley, P.J.</td>
<td>University College London</td>
<td></td>
<td>Exploring lithium isotopes as a weathering tracer in natural waters</td>
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Fixing the alkenone palaeobarometer

Badger M.P.S.¹

¹School of Environment, Earth and Ecosystem Sciences, The Open University, Walton Hall, Milton Keynes. marcus.badger@open.ac.uk

Atmospheric $pCO_2$ is a critical component of the global carbon system and is considered to be the major control of Earth’s past, present and future climate. Accurate and precise reconstructions of its concentration through geological time are, therefore, crucial to our understanding of the Earth system. Ice core records document $pCO_2$ for the past 800 kys, but at no point during this interval were CO$_2$ levels higher than today. Interpretation of older $pCO_2$ has been hampered by discrepancies during some time intervals between two of the main ocean-based proxy methods used to reconstruct $pCO_2$: the carbon isotope fractionation that occurs during photosynthesis as recorded by haptophyte biomarkers (alkenones) and the boron isotope composition ($\delta^{11}B$) of foraminifer shells (see Badger et al., [1]).

Suspicions have long been held that at low levels of dissolved CO$_2$ ($[CO_2]_{aq}$) haptophytes may actively concentrate carbon from the water in which they live in order to maintain metabolic requirements. Such deviation from the assumed passive uptake of carbon could change the carbon isotopic fractionation during photosynthesis and so confound the alkenone proxy.

Here I compile existing alkenone records from the past 800 kys where overlap between proxy and ice core records allow direct comparison, and show that these records show evidence of a threshold response at low $[CO_2]_{aq}$. Consideration of this threshold allows accurate reconstruction of ice core $pCO_2$, and suggests a way to confidently reconstruct ancient CO$_2$ during the geological past.

References:
The Monterey Event and the Paleocene-Eocene Thermal Maximum - two contrasting oceanic carbonate system responses to LIP emplacement and eruption

Babila, T.L. and Foster G.L.

School of Earth and Ocean Science, University of Southampton t.babila@soton.ac.uk

The Columbia River Flood basalts and the North Atlantic Igneous Province are two of the youngest Large Igneous Provinces (LIP). This presents an excellent opportunity to study the link between the emplacement and eruption of LIPs and their associated carbon cycle and climatic responses. The emplacement of both these LIPs are associated with two well-known climate events: The Monterey Carbon Isotope Excursion (MCIE; ~17-13.5 Ma) and The Paleocene-Eocene Thermal Maximum (PETM; ~56 Ma) characterized by a positive 1‰ and negative 3-5‰ carbon isotope excursion, respectively. Both are also associated with significant global warming. However, despite these contrasting timescales and carbon isotope trends, boron-based proxy records indicate a similar oceanic carbonate system response. This is consistent with the interpretation that in both cases enhanced volcanic carbon dioxide emissions led to oceanic absorption of the carbon released and this caused a decline surface ocean pH and an increase in dissolved inorganic carbon. We conclude, that although similar underlying carbon cycle processes are at play during both events, their specific behavior is somewhat different as the magnitude, carbon emissions rate (slow vs. fast) and background climate state (icehouse vs. greenhouse) conspire to cause distinct carbon isotope expressions and variable climatic responses to each LIP emplacement.
Reconstructing the structure of Atlantic Ocean circulation during early-middle Eocene extreme climatic warmth

McIntyre, A.J.1, Sexton P.F.1 and Anand P.1

1 School of Environment, Earth and Ecosystem Sciences, The Open University, Milton Keynes MK7 6AA, UK. Correspondence to Andrew McIntyre at andrew.mcintyre@open.ac.uk

The Atlantic meridional overturning circulation (AMOC) is a key component of the modern global ocean circulation, distributing heat, salt and biologically essential nutrients globally, and exerting a fundamental influence on regional and hemispheric climates. Yet we have little understanding how unstable the AMOC will be, or its sensitivity to climate change, under acute climatic warmth such as that marking Earth’s future. The nature of any inter-hemispheric competition in deep water formation during such warmth is another unknown. These fundamental gaps in our knowledge prevent us testing the simulations of ocean models for past warm climates, and thus also diminish our confidence in the capabilities of these models to predict changes to our oceans during our future climate trajectory.

A solution is to reconstruct AMOC stability during an interval of past acute warmth. The early-middle Eocene (50-47 Myr ago) was a time of extreme warmth. The deep oceans were 10 to 14°C warmer than today [1] and atmospheric carbon dioxide levels were ~2-4 times higher [2][3]. Yet, it is currently unclear whether a well-developed AMOC existed during the early-middle Eocene, how sensitive it was to orbital forcing, and what influence it exerted on the Earth’s climate during the early-middle Eocene [4].

To reconstruct the structure of any early-middle AMOC, we use an Atlantic transect of tightly correlated sites, spanning the northern Atlantic to Southern Ocean. New bulk carbonate δ13C records for six new drill sites form a high resolution chemostratigraphy for each site, allow for detailed inter-site correlations and provide a common, high resolution age model. We use Nd isotopes to reconstruct water mass source pathways and benthic foraminifer δ13C to reconstruct water mass ventilation state across a ~400 kyr interval of magnetochron C21n to resolve orbital-scale dynamism in the structure and stability of Atlantic Ocean circulation during extreme greenhouse warmth.

References:
Directly dating deformation with calcite U-Pb dating; the good, the bad and the ugly!

Mottram, C.¹

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Deformation in the upper crust is commonly accommodated by brittle fracturing and faulting. These events are coupled with fluid flow resulting in pervasive calcite/quartz veining in these geological settings. These rapid, low temperature processes are notoriously difficult to date as the minerals usually used to record geological time (such as zircon) typically do not crystallise or record any deformation under these conditions. Emerging techniques have recently been developed for directly-dating brittle structures using calcite, a mineral ubiquitous in many brittlely deformed rocks [1-2] (e.g. Roberts and Walker, 2016, Geology; Nuriel et al., 2017, Geology). This has opened up a whole new realm of tectonic investigation in the upper crust.

This talk will explore the effectiveness of the U-Pb calcite dating technique for providing timing constraints for orogenic crustal deformation, fluid flow and mineralisation events. In similarity with other unconventional geochronometers that have low U content and incorporate common Pb into their structures (such as titanite), calcite is not always straightforward to date and results can therefore be ambiguous to interpret. We therefore present examples of the ‘good, bad and ugly’ in the world of calcite U-Pb dating. Despite these challenges, we use examples from the Main Boundary Thrust in the Himalaya, the North Anatolian Fault, and large-scale strike-slip faults in the Yukon, Canadian Cordillera to demonstrate successfully how U-Pb calcite dating can be used to reconstruct the displacement history of these various first-order structures. The results can be used to examine how orogenic stress is spatially and temporally accommodated along major seismically-active, continental-scale faults.

References:
**In situ Rb-Sr dating: Triple Quad v Proteus**

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The beta decay of \(^{87}\text{Rb}\) to \(^{87}\text{Sr}\) was one of the earliest examples of a radioactive system to be harnessed as a tool for geochronology. A characteristic and complicating feature in the isotopic analysis of beta decay systems is the inevitable isobaric overlap of the parent and daughter nuclides, making *in situ* analysis of minerals with high Rb/Sr extremely difficult.

However, the development of collision cell ICPMS (inductively coupled plasma mass spectrometer) technology, coupled with an established literature on ion gas reactions, now permit *in situ* analysis of Rb-rich mineral phases. This can be achieved using variety of reaction gases such as N\(_2\)O, SF\(_6\) and CH\(_3\)F, which are used for their reactivity with Sr\(^+\) within a collision cell to form product ions with a greater mass [1]. Crucially, Rb\(^+\) displays little or no reactivity with these gases and where it does react it does not form the same polyatomic ion product as Sr\(^+\). The contrast in behaviour of Sr\(^+\) and Rb\(^+\) with these reaction gases allow ‘chemical resolution’ of the two elements that can be achieved rapidly during analysis. This has been successfully implemented using ICP-QQQ-MS (inductively coupled plasma – triple quadrupole mass spectrometer) technology for the dating of micas [2,3].

Here we demonstrate the added advantages of using the prototype collision cell MC-ICPMS (multi collector – inductively coupled plasma mass spectrometer) ‘Proteus’ over ICP-QQQ-MS for *in situ* Rb-Sr analysis. The enhanced sensitivity relative to ICP-QQQ-MS and the simultaneous collection of all Sr isotopes using multiple faraday cups allow Proteus to achieve superior precision for radiogenic Sr isotope ratios. Another collision cell MC-ICPMS has recently been developed, but unlike Proteus this does not have a quadrupole ‘pre mass-filter’ before the collision cell, a feature unique to Proteus among collision cell MC-ICPMS. This quadrupole permits only a certain mass range to be transmitted onward to the collision cell. This is crucial for laser ablation sampling, as it vastly simplifies the variety of ion gas products formed.

Our results show that, using SF\(_6\) as a reaction gas, Sr\(^+\) can be quantitatively resolved from Rb\(^+\) by conversion to SrF\(^+\) with 80% reaction efficiency. We present the first application of laser ablation Rb-Sr dating using collision cell MC-ICPMS yielding an age and initial \(^{87}\text{Sr}/^{86}\text{Sr}\) for the Dartmoor Granite. This Rb-Sr isochron was produced using analyses of only plagioclase and alkali feldspar. This approach applies the strengths of Proteus to produce greater \(^{87}\text{Sr}/^{86}\text{Sr}\) precision and analysis of lower Rb/Sr minerals (such as alkali feldspar) than is possible with ICP-QQQ-MS. The improved constraint on the measured \(^{87}\text{Sr}/^{86}\text{Sr}\) ratios importantly results in greater age resolution than could be achieved when the same sample was analysed using ICP-QQQ-MS. The age and initial \(^{87}\text{Sr}/^{86}\text{Sr}\) measured using Proteus is in agreement with previously conducted TIMS Rb-Sr analysis of the same intrusion [4].

The effect of matrix interferences on in situ boron isotope analysis by laser ablation MC-ICP-MS

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Boron isotope analysis is an important geochemical tool for understanding Earth-based processes; with applications ranging from a palaeo-pH proxy for seawater in studies concerned with ocean acidification to a tracer for mixing processes in studies of the ocean-crust-mantle system. Laser ablation multi-collector inductively-coupled-plasma mass spectrometry (LA-MC-ICP-MS) permits the in situ collection of geochemical information at high spatial resolutions and with high sample throughput. However, despite these clear advantages over traditional methods, the adoption of laser ablation techniques to studies concerning boron isotopes has not been straightforward, with numerous authors reporting significant levels of imprecision (typically 1–2 ‰ δ11B) and inaccuracy (up to 6 ‰ Δδ11B) compared to alternate analytical techniques [1, 2].

Here we explore the role of matrix interferences on boron isotope analysis by LA-MC-ICP-MS and present a protocol for obtaining accurate δ11B data following mass bias correction with the universal NIST glass standard SRM610. Critically, our protocol enables small-scale investigations of boron isotope heterogeneities in a range of samples without the need for matrix-matched standards, which are themselves often difficult to obtain and/or characterise. Our methods will be further demonstrated through timeseries analysis of a section of a Siderastrea siderea core collected from the Southern Belize portion of the Mesoamerican Barrier Reef System.

References:
Earth and Mars both possessed ample surface water very soon after formation, yet while oceans still cover around 70% of the Earth’s surface, Mars has lost the majority of its surface water, leaving it inhospitable for the development of complex life. What happened to surface water on Mars? While the Martian water inventory was undoubtedly depleted by loss to space following the collapse of the planet’s magnetic field, the presence of hydrated clays and the widespread serpentinisation of surface basalts attests to water-rock reactions playing a critical role in its sequestration. This, then, leads to an obvious question – why has Earth remained habitable for life, and Mars became barren? This is problematic, especially given the secular evolution of the Earth, and the presence of active water transport mechanisms, such as plate tectonics, to the Earth’s essentially dry lower mantle.

One possible explanation concerns iron, the one major rock forming element whose mantle concentration varies significantly across the terrestrial bodies. We show how this subtle chemical variation, set by the differing conditions of planetary accretion and core formation, may play a disproportionate role in subsequent planetary development and habitability. The ability of a planet’s surface to retain water over times relevant to the evolution of complex life may therefore be a result of the stochastic processes that resulted in planetary core formation. In particular, it is likely that planetary habitability may not simply be a case of ‘right ingredients and distance from star’ but also the correct ‘recipe’ of planet accretion.
Boron isotopes in metamorphic olivine record external fluid infiltration during serpentinite dehydration in subduction zones
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Fluid sources and pathways at the interface between the subducting slab and overlying mantle wedge are poorly understood. Metamorphic olivines produced by serpentine dehydration at sub-arc depths contain significant concentrations of B which is a particularly powerful tracer of subduction zone fluid processes. Here we report in situ B isotope data in serpentinites from high-pressure ophiolites in the Western Alps and Spain, which allows direct comparison of δ11B in metamorphic olivine and co-existing serpentine. We find high [B] (1.7-28.6 ppm) and δ11B (+9.5 to +27.5 ‰) in metamorphic olivine as well as co-existing serpentine ([B] 1.6-50.5 ppm, δ11B -4.7 to +24.4‰). Differences between δ11Bol and δ11Bsrp (Δ11Bol-srp) are highly variable and indicate significant isotopic disequilibrium. Importantly, there is a correlation between [B]ol/srp and Δ11Bol-srp: samples with high [B]ol/srp have low Δ11Bol-srp (up to -10 ‰) whereas samples with low [B]ol/srp have high Δ11Bol-srp (up to +15‰). We suggest: B-depleted olivines represent closed-system dehydration and equilibrium fractionation of the isotopes between olivine and serpentine. B-enriched olivines have equilibrated with externally-derived fluids most likely derived from subducting sediments and crust. We conclude that open system dehydration is common in exhumed ophiolites, even in those that show little evidence based on their whole-rock B isotope signatures. We believe this provides direct evidence for large scale fluid mobility in or near the subduction interface.
Tracing subduction zone fluids in Izu arc lavas using molybdenum isotopes

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Molybdenum isotopes have gained attention as potential tracers for the recycling of crustal components in subduction zones and in the deep-mantle, due to the distinctive Mo isotope signatures (expressed as δ⁹⁸/⁹⁵Mo deviations from the NIST 3134 standard) of marine sediments, MORB and altered oceanic crust [1,2,3]. In this context, a thorough understanding of the effects of subduction zone processes on the Mo budget and on the isotope composition of the subducted slab are required. Mo isotope compositions of arc lavas heavier than MORB (δ⁹⁸/⁹⁵Mo₉⁴3ולוגiscisoNIST3134 ~ -0.20‰; [2]) have been attributed to fractional crystallisation of hydrous phases [4,5], to the addition of isotopically heavy slab components (e.g., black shales; [1]) or the involvement of subduction zone fluids [2,3]. Discriminating between these processes has so far been challenging due to the complexity of the previously studied arc settings. One important outstanding problem is that it is not clear whether the loss of Mo from the subducted slab via slab-derived fluids is linked to Mo isotope fractionation. This highlights the necessity of addressing this problem on a simple arc system, dominated by fluid transfer from the subducted slab and characterised by minimal inputs from other slab components.

Here, we present Mo isotope data for basalts and basaltic andesites from five volcanic islands of the Izu arc (Japan). Independent geochemical and isotopic evidence suggest that hydrous fluids dominate the slab-derived components added to the Izu arc magma sources [e.g. 6,7], which makes it an ideal setting to study the Mo isotope composition of slab-derived fluids. The Izu arc lavas have significantly higher δ⁹⁸/⁹⁵Mo than MORB. Indices of slab-fluid addition in the lavas, such as high Ba/Th, Ba/La, (²³⁸U/²³⁰Th) and low Ce/Pb, are associated with high δ⁹⁸/⁹⁵Mo, suggesting the addition of a slab-derived fluid with heavy Mo isotope composition to the Izu magma sources. Positive correlations between δ⁹⁸/⁹⁵Mo and ¹⁴⁳Nd/¹⁴⁴Nd and ¹⁷⁶Hf/¹⁷⁷Hf ratios exclude fractional crystallisation of hydrous phases as a cause for the observed Mo isotope variations. The latter relationships also underline the extent to which the relatively depleted Izu arc mantle has been overprinted in terms of its fluid-mobile trace element budget. The presence of well-defined trends in our data also suggests that the fluid component which infiltrated the Izu arc mantle was relatively homogeneous with respect to its δ⁹⁸/⁹⁵Mo composition. In identifying a clear link between isotopically heavy Mo and the addition of slab-derived fluids in arc lavas, our data lend support to the hypothesis that Mo isotope fractionation occurs during transport of fluids through the oceanic crust, which has been attributed to the retention of isotopically light Mo in oxide phases such as rutile during fluid flux [2,3,8]. Through geochemical modelling, we further constrain the causes and magnitudes of these Mo isotope variations and assess the consequences for the mass balance of Mo in subduction zones and the deep recycling of Mo.

References:
Arsenic remediation using TiO$_2$/Fe$_2$O$_3$ composites: investigating mixed mineral adsorption via SCM and LEIS


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The arsenic(III) rich groundwaters of Bangladesh are responsible for an estimated 30-40,000 unnecessary deaths per year. As(III)-contaminated water is difficult to treat, as As(III) is poorly removed by commercial sorbent media. A number of studies have considered bifunctional materials, whereby As(III) is first photooxidised to As(V), which is much more strongly adsorbed. Composites such as TiO$_2$/Fe$_2$O$_3$ offer one route to bifunctional behaviour. But how do we describe and model adsorption over a multicomponent material?

Surface Complexation Modelling (SCM) provides an adsorption model which, unlike Langmuir and Freundlich isotherms, can be made sensitive to changes in pH and presence of competitor adsorbates. However very few multicomponent (mixed mineral) SCMs have been developed due to difficulty in determining the contribution of each mineral phase to the overall surface area. Previous approaches have either lacked surface sensitivity (e.g. XRD) and underestimate the contribution of fine nano-precipitates, or are time consuming (e.g. fitting batch adsorption data).

In this work an atomically sensitive surface technique, Low Energy Ion Scattering (LEIS), is applied for the first time in SCM to quickly and precisely determine the composition of the meso-TiO$_2$/Fe$_2$O$_3$ atomic surface. The accuracy is verified as LEIS-results successfully predicted other properties as a linear combination of TiO$_2$ and Fe$_2$O$_3$ reference samples, such as surface charge (potentiometric titration). pH adsorption edges and isotherms (ICP-MS) are fitted using geochemical software (FITEQL, PHREEQC) and the first multicomponent SCM for arsenic adsorption developed, considering simple monodentate models with the Diffuse Double Layer electrostatic model, and more sophisticated/spectroscopically consistent bidentate Extended Triple Layer models.

Figure 1: (a) quantification of composition at the atomic surface using LEIS, and (b) LEIS data used to predict arsenic(V) Langmuir adsorption isotherm (black line) as a linear combination of reference samples (blue and green lines). (pH 7.0 ± 0.1, 0.01 M NaCl, 1 g L$^{-1}$ sorbent)
The determination of the sources and cycling of Ni and associated elements in an industrialised floodplain, South Wales.

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The present study has set out to investigate the cycling of Ni, Co, Cu, As, Se and Te at an operational metal refinery in a floodplain setting, South Wales. The refinery has produced high purity metal products since its opening ca. 1900, and has handled a range of feed material from impure sulfide matte in its beginning, to a higher purity metal-oxide today. A colourful range of processing methods have been used at the refinery throughout its history. The accumulated effect of unregulated disposal and spillage of the ensuing process by-products prior to heightened environmental awareness has resulted in poorly understood, but highly contaminated sediment and groundwater at the site. The current pump and treat system used to recover Ni and other metals from contaminated groundwater has had limited success; this project aims to develop a better understanding of sediment-water interactions, upon which a more effective remediation strategy can be developed.

Contaminated sediments at the site are highly heterogeneous. Analysis of sediments collected from around the site (n=58), digested using aqua regia, show that Ni, Cu and As are the most highly enriched metal(oids) in surface sediments with respective mean concentrations of 4700, 5000 and 5800 mg/kg. All relative standard deviations exceed 150%, highlighting the extreme heterogeneity of sediment contamination. Mean Se and Te values are 23 and 3 mg/kg, with maximum values of >1 wt% Se and 850 mg/kg Te. Water samples from 15 boreholes spread over a 55 Ha area have been collected between June 2018 and February 2019 to investigate the spatial and temporal variation in groundwater contamination. It has been found that despite the high concentration of Cu, As, Se and Te in sediment, respective groundwater concentrations rarely exceed 200, 10, 100 and 1 ug/L. In contrast, the maximum Ni concentration exceeds 20 mg/L for each sampling period, with concentrations seemingly higher in summer months. Co is geochemically similar to Ni in both groundwater and sediments (r=0.92).

The current phase of the project is concerned with delineating the interaction between sediment and the groundwater that passes through it. Work is being focused on identifying the host phase of the key metal(oids) Ni, Co, Cu, As, Se and Te. Various methodologies are being applied to address this question, including the use of sequential extraction, XRD, density separation and various digestion protocols. Geochemical modelling (Geochemists Workbench \textsuperscript{®}) has been used to support experimental and field data. Field data has been analysed using a GIS-PCA-Fuzzy Classification methodology to identify overriding spatial relationships between contaminants and other factors such as the site hydrogeology and redox zones.

The knowledge gained from field and laboratory studies has been applied to the design of a series of flow-through soil columns, where real groundwater will be used and factors such as oxygen content, pH and residence time will be tightly controlled. It is hoped that this experiment will identify major controls on the mobilisation of contaminants from sediment to groundwater, allowing for the future design of a more sophisticated remediation strategy.
Sm-Nd and Sr isotope fingerprinting of iron mining tailing deposits from the failed SAMARCO Fundão dam 2015 accident at Mariana, SE-Brazil

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This work presents the first Sm-Nd and Sr isotope data on the fine-grained fraction (< 74 µm) of iron-rich deposits related to the November 5th, 2015 Mariana accident, caused by the rupture of the Fundão dam, an iron tailings pond operated by SAMARCO Mining, a BHP Billiton and Vale joint venture. The failure of the Fundão dam produced what is considered the most serious environmental accident in Brazil, with the sudden gravitational flow of approximately 62 Mm³ of a quartz and iron oxide-rich slurry composed of water, fine sand and silt, that flowed into the Gualaxo do Norte and Carmo rivers, and eventually along the whole Doce river. At first, the mudflow caused the loss of 19 human lives and the total destruction of the Bento Rodrigues village, located ca.7 km downstream. The displacement of water created a propagating flood wave that was followed by a turbidity front that travelled for 673 km along the Rio Doce river, eventually reaching the sea 16 days later, giving rise to a large sediment plume in the adjoining seawater.

Samples were collected between 5 and 80 km downstream from the Fundão Dam, along the Santarém creek and the Gualaxo do Norte and Carmo rivers. X-Ray Diffractometry analyses show that samples are >90% composed of quartz and iron oxides (hematite, goethite and magnetite), with minor phyllosilicates (kaolinite and mica). Major and trace element contents also closely match those of the original Paleoproterozoic Cauê Formation banded iron formation ore. The five most proximal samples, collected between the Fundão Dam and Bento Rodrigues village, yielded εNd between -17.0 and -19.7, similarly to those of the samples collected more downstream along the Gualaxo River, between -17.1 and -19.7. Neodymium model ages (TDM) are between 1.94 Ga and 2.40 Ga. The most distal sample, collected at the Carmo River just downstream from the confluence with the Gualaxo River, shows more negative εNd of -23.2, and older TDM of 2.75 Ga. This contrasting value suggests a larger degree of mixing with incoming sediment derived from Archean terrain, brought by the Carmo River. Strontium 87Sr/86Sr ratios show variability between 0.7351 and 0.7729.

The reported Sm-Nd and Sr isotope compositions closely match those of the original iron ore, and contrast with those of upriver and downriver Precambrian to Ordovician bedrock units. The firstly defined isotope signature can thus be potentially used as a proxy for ongoing suspended sediment dispersion studies farther downstream along the Doce river and in adjacent marine environment.
Hydrothermal vents an important source of iron and zinc to the ocean interior
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Mid-ocean ridges are significant sources of iron to the ocean due to hydrothermal activity. This talk will use data from a recent cruise along the mid-Atlantic Ridge to highlight the large range in supply of dissolved iron from known vent sites. Comparing the vents sites TAG and Rainbow, iron isotope data reveal large differences in the isotopic signature of iron with values of -6.94‰ at Rainbow compared to -1.83‰ at TAG reflecting the differences in iron (II), dissolved iron concentrations and sulphide concentrations. Preliminary results suggest that both dissolved iron and manganese are transported away from TAG into the abyssal Atlantic Ocean. It is not only dissolved iron which is supplied from these vent sites, a large supply of excess dissolved zinc was also observed highlighting the need for hydrothermal vents to be considered in the global biogeochemical cycle of zinc.
Rare earth elements in marine biogenic carbonate as proxies for the marine carbonate system?

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The geochemical behaviour of dissolved rare earth elements (REE) in seawater shows a strong carbonate ion ($[CO_3^{2-}]$) dependence [1]. Under experimental conditions of constant salinity (35) and temperature (25°C), the REE equilibrium constants ($LnCO_3^2$ vs. $Ln(CO_3^2)_2$) span a sliding range from pH 8.47 (La) to pH 7.42 (Lu), with the negative complex dominant at higher pH [2]. If the aqueous complexation of seawater REE exerts a dominant and predictable effect on the uptake of REE during biogenic carbonate precipitation, it hints at the potential for marine calcifiers to preserve a record in their skeletons of the status of the marine carbonate system [3]. To accurately interpret REE concentrations in biogenic archives requires knowledge of seawater-to-calcite REE partitioning, together with “vital effects” (biological factors that modify the primary signal). However, the biogenic carbonate/seawater partition coefficients ($K_{D}^{REE}$) reported in the literature are variable and the mechanisms controlling them remain poorly understood (e.g. [3-6]).

To quantify the effect of the marine carbonate system on the partitioning of seawater REE into biogenic carbonate, we adopted an empirical approach. We conducted culturing experiments under three different seawater pH treatments, with constant temperature and salinity, using locally available serpulid worms (Serpula vermicularis) from the adjacent Loch Creran (West Coast Scotland). This temperate coastal species precipitates a mainly calcitic tube from which it emerges to filter feed. After 13 weeks of culturing, we harvested new tube growth and measured their REE and Ca concentrations. In tandem with the dissolved REE concentrations from seawater in the experimental tanks and from Loch Creran, we calculated the $K_{D}^{REE}$ at four pH values across the range of pH 8.1 to 7.5.

Our results show that differences between the pH treatments are expressed by lower REE concentrations in the serpulid tubes and a flattening of the $K_{D}^{REE}$ with decreasing pH, while the total dissolved REE concentrations in the experimental tanks are identical within error. We use the results to test firstly that a predictable and systematic relationship between $K_{D}^{REE}$ and pH (or other parameter of the marine carbonate system) exists. Our results conform to the hypothesis that aqueous REE complexation by carbonate ions may indeed represent the mechanism controlling the partitioning behaviour of REE into serpulid calcite. However, we cannot exclude other potential effects on REE partitioning behaviour at this stage. Secondly, we construct a calibration line using the REE concentration ratios in the serpulid tubes and parameters of the marine carbonate system to identify if this approach to reconstructing change in the marine carbonate system has potential application in fossil biogenic carbonate samples. This research is still in its very early stages, and much work remains to identify vital and other ambient seawater effects(e.g. temperature, salinity change), and to test the calibration on biogenic samples with known seawater pH or carbonate ion concentration. However, the narrow range of pH spanned by the REE equilibrium constants makes this approach an appealing option to investigate change associated with rapid variation in atmospheric CO$_2$.

The age distribution of deep-sea corals in the Pacific, Atlantic and Southern Oceans

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Deep-sea corals are distributed worldwide and live across a wide range of depths and temperatures. The skeletons of calcifying corals can be precisely dated and can offer insights into long term ecological and environmental change in the oceans. There is mounting evidence that changes in their paleo-biogeographic distributions may be associated with climatic events. In this study we use a large collection of fossil scleractinian corals from the Pacific, Atlantic and Southern Oceans. Their aragonite skeletons were dated by radio-isotopic methods (radiocarbon, uranium-thorium solution and laser) to investigate their age distributions over the last 250 thousand years. As for prior studies, we find that there were large changes in the populations over time in all locations. In addition to examining past age distributions, these fossil corals are also a useful archive for reconstructing information about their paleoenvironment because they incorporate geochemical properties of their surrounding waters during their growth. Radiocarbon isotopes can be used as a tracer of the last contact of a water mass with the atmosphere and, therefore, provide interpretations of the past oceanic circulation. Other proxies as Mg/Li, U/Ca, P/Ca, Ba/Ca, boron and neodymium isotopes can provide information about paleotemperature, carbon cycling, nutrients, pH and water masses sources, respectively. These geochemical signals can be used to investigate the causes of population changes over time and the oceanic influence on global climate.
The isotope compositions of Zn and Cu in atmospheric aerosols and their role in biogeochemical cycles
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Aerosols are a source of Zn and Cu to the oceans [1]. Stable isotope signatures of Zn and Cu can be used to trace the various external sources that contribute to these atmospheric inputs. External sources include natural mineral dust or anthropogenic activities such as factory and traffic emissions. High temperature processes for example those in Pb-Zn smelting fractionate metal isotopes [2] and may therefore influence the isotope compositions of anthropogenic aerosols.

This study presents Zn and Cu isotope data for aerosols collected during the GEOTRACES GA06 section cruise (D361, February–March 2011) in the eastern Tropical Atlantic Ocean. The aerosol \( \delta^{66}\text{Zn} \) values (relative to JMC-Lyon) fall in the range 0.05-0.41 ‰, whereby all samples but one yielded an isotope composition lighter than the measured North African dust value of ~ 0.3 ‰. These comparatively light values together with the large enrichment factors determined for Zn suggest a significant anthropogenic contribution of Zn.

The \( \delta^{65}\text{Cu} \) isotope values of the aerosols (relative to NIST SRM 976) range from -0.33 ‰ to 0.28 ‰ and hence do not differ significantly from the isotope composition of lithogenic Cu values (-0.14-0.26 ‰) [3]. Copper is less volatile than Zn and thus does not fractionate to the same extent during high temperature processes. The combination of a remote sampling location and the high boiling point results in the sources of atmospheric Cu being more ambiguous.

The atmospheric input data in this study highlights the perturbation of the natural cycles of Zn and Cu in the ocean by anthropogenic emissions whereby the extent of perturbation may vary between elements depending on their properties.

References

The deposition of Precambrian banded iron formation (BIF) is traditionally explained through upwelling of deep, anoxic waters onto shallow shelves, where iron is oxidised by microbes or through direct contact with dissolved oxygen. However, recent work has revealed that these chemical sediments are packed with tiny particles of greenalite, a ferrous-silicate mineral thought to have precipitated directly from seawater. This body of work shows that iron was originally deposited in its reduced form, and oxidation occurred later, during regional metamorphic events. How does this change our view of Archaean biogeochemistry? Iron silicates would have acted as significant sinks for nutrients such as Zn and Cu from seawater, which may have significantly impacted the evolutionary dynamics of early microbes. Metal availability in a greenalite ocean is in fact consistent with patterns of metal utilisation by early archaea and bacteria as predicted from protein structures and comparative genomics. Further, our data suggest natural greenalite may provide a novel window into ancient ocean chemistry that can be used to track nutrient availability in deep time.
Impact of permafrost thaw on chemical weathering in the high Arctic Zackenberg River, NE Greenland

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Increasing global temperatures are causing widespread changes in the Arctic. Enhanced permafrost degradation and deepening of the seasonally thawed ‘active layer’ alters subsurface water flow-paths and increases water-rock interaction. Together, these warming-induced processes promote weathering of freshly exposed, previously frozen mineral-rich horizons. Furthermore, permafrost degradation affects the weathering reactions controlling the release or removal of atmospheric carbon dioxide ($CO_2$) from high-latitude regions. This in turn alters the biogeochemical cycles of many elements into the ocean and atmosphere, however these processes remain under-constrained and largely unquantified.

Over the past two decades, summer air temperature increases of 1.5°C[1], and active layer thickness increases of 0.8-1.5 cm yr⁻¹ have been recorded in the high Arctic Zackenberg River Catchment, NE Greenland[2]. Seasonal permafrost thaw can serve as a proxy for longer-term permafrost degradation and deepening of the active layer caused by climate warming. To determine the extent of these changes, we sampled glacier- and snow-fed rivers draining continuous permafrost throughout the melt season as the permafrost ‘active layer’ thawed and refroze. The detailed time series data will be compared with historic data obtained from the Greenland Ecosystem Monitoring programme in order to enable us to better understand and predict the response of Arctic rivers in a rapidly warming climate.

Here we report riverine lithium isotopes (a proxy for silicate weathering processes), which when coupled with major and trace element geochemistry, will allow the determination of whether this high Arctic river catchment is acting as a source or a sink for $CO_2$. We show increasing intra and inter-annual Ca/Na variability, suggesting a greater proportion of carbonate relative to silicate weathering, consistent with increased active layer thaw depth. Our preliminary findings suggest that the Zackenberg River Catchment transitions from a transient sink to a source of $CO_2$ throughout the melt season. These findings provide important new insights into the weathering processes controlling $CO_2$ removal and release, as well as how these changes drive climate change in the Arctic.

References:
Lithium isotope evidence for weathering changes across the PETM

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Chemical weathering of silicate rocks is considered to be the primary mechanism for mitigating atmospheric CO₂ levels over geologic time, likely via a temperature-driven “thermostat” feedback [1]. However, the rates by which this thermostat affects or responds to climate are poorly known. The Palaeocene-Eocene Thermal Maximum (PETM) is a hyperthermal that allows us to test how weathering responds to, and controls, climate change. It is characterised by rapid warming, ocean acidification, a major biotic turnover and significant changes to the hydrological cycle, likely triggered by a carbon release event. It is of relatively short duration (~100 kyr, plus another ~100 kyr recovery phase) [2].

We investigate weathering across the PETM using lithium isotopes, a tracer of silicate weathering processes [3]. Negative isotopic excursions are observed in marine carbonates in three different IODP cores, as well as two detrital silicate sections from Denmark and Spitzbergen. In all cases the excursion size is around 3–4‰ and broadly synchronous with the carbon isotope excursion, demonstrating that the δ⁷Li excursion is global in extent.

The data imply that silicate weathering rates increased fairly dramatically across the PETM, in addition to a shift in the weathering regime to lower intensity (more congruent) weathering. They also imply that the concept that silicate weathering responses are too slow to affect carbon on PETM-length scales are likely incorrect.

References:
Hadean geodynamics inferred from time-varying $^{142}\text{Nd}/^{144}\text{Nd}$ in the early Earth rock record

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Short-lived radioactive systems such as $^{146}\text{Sm}-^{142}\text{Nd}$ provide unique insights into the earliest periods of planetary evolution for which there exists no physical evidence exists otherwise [1]. Presence of $^{142}\text{Nd}/^{144}\text{Nd}$ anomalies relative to modern accessible mantle in Archean cratons suggests differentiation of Hadean mantle to form crustal and depleted mantle reservoirs as early as $\sim 4.5$ Ga. Tracking the secular evolution of $^{142}\text{Nd}/^{144}\text{Nd}$ anomalies is important towards understanding the mode of crust-mantle dynamics in the early Earth. However, an important obstacle when it comes to its application is that the $^{142}\text{Nd}/^{144}\text{Nd}$ variations expected are at the limit of analytical precision and hence highly prone to analytical artefacts. In addition, uncertainties regarding bulk silicate Earth (BSE) composition contributes to incorrect interpretations regarding the observed $^{142}\text{Nd}/^{144}\text{Nd}$ variations [2].

New high-precision MC-ICPMS data for rocks from Isua supracrustal belt (SW Greenland) demonstrate a well-resolved temporal variability in $^{142}\text{Nd}/^{144}\text{Nd}$ signature unlike previously reported, suggesting progressive homogenization of Hadean Isua depleted mantle [3]. The secular evolution of $^{142}\text{Nd}/^{144}\text{Nd}$ anomaly provides a direct observational measure of mantle dynamics in the early Earth. Box-modelling approach that considers crustal recycling shows that the observed $^{142}\text{Nd}/^{144}\text{Nd}$ evolution is consistent with crustal residence times of $\sim 1000$-$2000$ Myr. Such long crustal residence times, suggesting preservation of mafic protocrust for periods much longer than today, is inconsistent with prevalence of modern-style plate tectonics until at least the mid-Archean. Stagnant-lid regime punctuated by episodes of mantle overturns accounts for the long life-times estimated for the Hadean proto-crust. Although the $^{142}\text{Nd}/^{144}\text{Nd}$ variations in Superior Province and Labrador are found to be related to those in SW Greenland, the global extent of these reservoirs is less clear [4]. This has implications for Earth’s accretion and Moon formation scenarios. Future work required includes expansion of the high-precision $^{142}\text{Nd}/^{144}\text{Nd}$ dataset to other Archean terranes and mass-balance models that quantify the extent of these Hadean reservoirs.

References:

Latest TTG to sanukitoid transition: implications for the evolution of the continental crust during the magmatic lull

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One of the biggest mysteries in geosciences is defining when and how the continents were formed. The evolution of Earth’s continents and plate tectonics has left geoscientists puzzled for more than 50 years trying to constrain the factors governing crust generation. Much is debated for the early stages of Earth’s evolution and what our planet would have looked like during the Hadean. Nevertheless, ideas currently are converging that plate tectonics began in the Archean as a gradual and evolving mechanism, which started at ca. 3.0 Ga and evolved to a modern style by ca. 2.5 Ga. During this period, a secular geochemical evolution is observed globally: TTGs evolve to younger magmas with sanukitoid (high Ba-Sr) composition. This is inferred to represent the onset of subduction-driven plate tectonics due to the opening of the mantle wedge angle and Earth cooling. Plutonic rocks from the Mineiro Belt, Brazil record this transition much later, during the so-called ‘magmatic lull’ between 2.4 and 2.1 Ga, when few evolved juvenile magmas were added to the continental crust globally. New data from detrital and igneous zircons in this study depict a continuous evolution of supracrustal reworking and magmatism during this period and thus a unique window into Palaeoproterozoic crustal evolution. Ultimately, the Mineiro Belt represents evidence of continental crust growth during the magmatic lull and a delayed transition from TTG to sanukitoid magmatism to shortly after the Archaean-Palaeoproterozoic boundary, compared with other cratons. The challenge is now to understand why and how this region developed and preserved this out-of-sequence magmatic record and what it means for the secular evolution of geodynamics. Our line of evidence is based on the rise of $\delta^{18}$O and decrease of $\varepsilon$Hf(t) values in zircon, which indicate a continuous process of continental crust evolution and input of sediments in the mantle wedge during the Palaeoproterozoic, similarly to the Archaean geochemical secular evolution.
What controls titanium stable isotope fractionation during magma evolution?

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Titanium (Ti) is a highly immobile, refractory lithophile element and as such it is unaffected by core formation, impact-driven volatilisation or fluid-mediated processes. Preliminary studies of its stable isotope composition in geological samples have shown that it is sensitive to oxide-melt equilibrium [1,2] This is because Ti predominantly exists in 5-fold co-ordination in silicate melts [3], in contrast to oxide minerals in which Ti occupies a 6-fold co-ordination [3]. This contrast in co-ordination serves a major driver for Ti stable isotopic fractionation in magmas during fractional crystallisation [2].

Previous work [1, 2] has demonstrated a progressive enrichment in heavy isotopes (expressed as $\delta^{49}$Ti, the deviation of the $^{49}$Ti/$^{47}$Ti ratio relative to the OL-Ti standard) correlated with increasing SiO$_2$ due to crystallisation of Fe-Ti oxides during magmatic differentiation (e.g. titanomagnetite, ilmenite), consistent with stable isotope fractionation theory. However, due to the limited data available, it is unclear if other controls also exist. In order to fill this gap, this study presents a comprehensive set of Ti stable isotope data for a range of differentiation suites from tholeiitic, calc-alkaline and alkaline magma series. All samples were processed and measured using a double spike method [1]. Isotope measurements were also bracketed by measurements of the OL-Ti standard to account for polyatomic interferences on $^{47}$Ti and $^{48}$Ti [1]. Typical precision of the double spike measurements are ca. ±0.02 ‰ at 95% confidence.

$\delta^{49}$Ti values for terrestrial differentiation suites from plume settings range from +0.005 ± 0.028‰ to +2.315 ± 0.032‰. In arcs $\delta^{49}$Ti values range from -0.010 ± 0.030‰ to +0.640 ± 0.022‰. In both settings progressive enrichment is correlated with multiple indicators of increasing magmatic differentiation i.e. increasing SiO$_2$, alkalis, Rb and Zr and decreasing TiO$_2$, MgO and FeO$_T$ contents. As first postulated by Millet et al. [2] these enrichments are the manifestation of fractional crystallisation of Fe-Ti oxides during magma evolution. However, the $\delta^{49}$Ti compositions of evolved rocks from plume settings far surpasses the typical compositions of observed in arc rocks of similar silica contents. There is also a second order contrast observed within arc settings with calc-alkaline arcs displaying a slight enrichment in heavy $\delta^{49}$Ti relative to tholeiitic arc suites. A major driver for the larger magnitude of Ti fractionation trends in plume settings could potentially be related to the elevated TiO$_2$ contents of plume magmas, combined with delayed oxide saturation as a result of lower fO$_2$ [4]. This culminates in the preponderance of ilmenite over titanomagnetite in the crystallising assemblages of plume rocks compared to arc rocks, particularly at higher SiO$_2$ contents. Such difference would hint towards the Ti isotope fractionation factor between ilmenite and melt being larger than that between magnetite and melt.

Further work will be aimed at elucidating precise isotopic fractionation factors of Ti between silicate melts and Fe-Ti oxides using both natural and experimental samples to determine the varying control of titanomagnetite vs. ilmenite during magma evolution.

References:
Tungsten in Mariana arc basalts: evidence for mobility and isotopic fractionation during subduction

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Novel application of non-traditional stable isotope systems is emerging as a powerful tool in understanding a range of low temperature and high temperature geochemical processes. Recent work on stable Mo isotope fractionation shows that arc basalts are isotopically heavier than mid ocean ridge basalts (MORB), suggesting that Mo isotopes are fractionated during subduction [1]. One mechanism for Mo isotope fractionation in these environments is by incorporation of isotopically light Mo into rutile [1]. Like Mo, W is also incorporated into rutile at typical subduction zone temperatures and pressures [2]. This process involves a coordination change from tetrahedral (4+) in melts or aqueous fluids, to octahedral within the structure of rutile (6+) [3]. The higher coordination number of W in rutile should be accompanied by preferential incorporation of light W isotopes. In addition to being highly incompatible during mantle melting, W is also fluid mobile in subduction zones [4]. Any remaining isotopically heavy W not sequestered into rutile could be mobilised by fluids at subduction zones and be added to the source of arc magmas.

The Mariana arc is an ideal locality to study because the lavas contain a fluid component only partially overprinted with a contribution from incompatible element enriched subducted sediment [5, 6]. If W isotopic fractionation occurs during subduction and is recorded by the fluid released, across-arc stable W isotopic variability would be expected.

We measured W concentrations and stable isotopic compositions of 6 islands across the Mariana arc. Mariana arc basalts are enriched in W relative to Th, an element with a similar incompatibility during mantle melting. The islands form a tightly defined trend in Ba/Th vs W/Th, confirming the fluid mobile behaviour of W in subduction zones. Samples with highest W/Th also have the highest Pb/Ce [1], suggesting a common source of W and Pb enrichment. Marianas arc basalts are isotopically heavier than the mantle as defined by ocean island basalts, and lavas that show the greatest influence from the fluid component (highest Ba/Th, W/Th, Pb/Ce) contain the isotopically heaviest W, suggesting the fluid released in subduction zones carries a heavy W isotopic composition.

Previous work on Pb isotopic compositions in Marianas arc lavas suggest the fluid component is derived from the subducted mafic oceanic crust [1]. This is consistent with stable isotope variations for both W and Mo [1]. Collectively, the data suggest the fluid component in subduction zone environments acquires its diagnostic chemical composition from deep within the subducting mafic oceanic crust. If equilibrium with rutile is the dominant mechanism for W isotopic fractionation, the magnitude of isotopic fractionation may reflect temperature conditions deep within the subducting slab.

References:
Posters
High-resolution CO$_2$ record of the First Great Glaciation using boron isotopes

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One of the most recent climate transitions was the intensification of northern hemisphere glaciation (iNHG) at 3.4-2.5 million years ago (Ma). iNHG represents the culmination of Cenozoic cooling with the development of permanent ice sheets in the northern hemisphere that periodically waxed and waned with changes in insolation [1,2]. Most recent evidence suggests that changes in atmospheric CO$_2$ are the primary driver of iNHG though the precise nature of its role is debated [3, 4]. In particular current records are not of sufficient resolution to adequately assess the relationship between changes in CO$_2$ and global climate. My PhD project, that I will introduce here, is focused on creating a high resolution (1 sample per 3 thousand years) CO$_2$ record for iNHG using the boron isotope pH proxy and using this record to evaluate the nature of the relationship between CO$_2$ and climate during this transition.

References:

Atmospheric CO₂ change over the past 1.5 million years

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During the past 1,500 thousand years (kyr) Earth’s climate has undergone major changes. Ice core-derived atmospheric CO₂ records have long been the staple of Pleistocene climate reconstruction, used to inform hypotheses and test climate model simulations of these changes, however, they are currently limited to the last 800 kyers and are unlikely to be significantly extended in a continuous record. We interrogate the future and potential of boron isotope records to provide high quality carbon cycle information for the Pleistocene. Here we present a compilation of boron isotope-derived pH-CO₂ records from low-latitude ocean drill cores which closely follow the evolution of atmospheric CO₂ over the ice core interval and data extending to 1.5 million years ago. We show that there is a mean CO₂ decline across the Mid Pleistocene Transition (MPT) which manifests as a lengthening and deepening of glacial CO₂, and confirm distinct difference in the nature of CO₂ cycles in the 41 kyr world. Through carbon cycle modelling, we attribute this decline primarily to the initiation of substantive dust-borne iron fertilization of the Southern Ocean during peak glacial stages and associated internal climate feedbacks. A change in the dynamics that govern ice sheet stability is invoked to explain an observed twofold steepening of the relationship between sea level and CO₂-related climate forcing. We argue that neither ice sheet dynamics or CO₂ change in isolation can explain the MPT.
Isotopically light cadmium in organic-rich sediments

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Cadmium (Cd) is a nutrient-like element in the ocean that has a distribution akin to phosphate (P) and has been used as a proxy for past ocean phosphate distribution. However, deviations from the global Cd/P relationship are observed under low oxygen concentrations in the modern ocean. Cadmium sulphide precipitation in oxygen minimum zones has been proposed to explain the decrease in Cd/P ratios [1]. To evaluate whether cadmium sulphides are formed within marine systems, we measured Cd and Cd isotopes in a variety of sediment samples. Our data are from continental margin sediments in suboxic-anoxic settings from the Northeast Pacific margin and provide evidence for an isotopically light sink. Preferential uptake of isotopically light Cd by phytoplankton is one means to supply isotopically light Cd to the sediment. Although, CdS particulates could also supply isotopically light Cd to the sediment, we find limited evidence for CdS precipitation occurring within the water column. Instead, a correlation between δ114Cd and total Fe suggests that scavenging of Cd by Fe oxide phases may also play a role in enriching continental margin sediments in isotopically light Cd. Further isotope fractionation through diagenetic processes in the sediment is unlikely as Cd is sequestered quantitatively in reducing sediments, possibly through precipitation of CdS. We calculate a Cd output flux to continental margin sediments of 5.2 × 10^7 mol/yr, at ~0‰. Isotopically, this estimate is in the range of the known inputs of the ocean (rivers and dust, at 0 to 0.2‰)[2][3], however, with current estimates, the Cd output flux is ~2 times larger than the inputs. This flux imbalance suggests either that current riverine/dust inputs are underestimated or the existence of another source with similar isotope composition.

References:
NERC changing Arctic Ocean: Implications for marine biology and biogeochemistry

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The Arctic is responding in unknown ways to profound changes in the physical environment as well as to multiple natural and anthropogenic events that place stress on Arctic ecosystems. The scale of the challenges facing the Arctic is immense and is further compounded by the rapid rate of change.

To address the uncertainties generated by climate change in the Arctic Ocean, NERC has invested £16 million in a research programme called “Changing Arctic Ocean: Implications for marine biology and biogeochemistry.” The over-arching goal of this 5-year (2017-2022) flagship research programme is to understand how change in the physical environment (ice and ocean) affects the large-scale ecosystem structure and biogeochemical functioning of the Arctic Ocean. The science outputs will address the potential major impacts and refine projections of change for future ecosystem services.

At the core of the programme are four large, NERC-funded projects that started in February 2017. A further 12 projects joined the programme in July 2018, co-funded by NERC and the German Federal Ministry of Education and Research. This co-funding of research projects by the UK and Germany represents a first, and brings with it benefits for the programme’s international collaboration, access to large-scale research infrastructure, and advantages of shared scientific expertise. The programme has more than 200 scientists, from 32 German and UK research institutions, working with Arctic teams in 15 other countries to meet the programme’s objectives.

The focus of the 16 projects spans many of the effects of warming on the Arctic Ocean’s ecosystem, driven principally by the cascade of impacts from the retreat and thinning of sea ice:

- release of chemical pollutants and plastics from melting sea ice;
- exposure of the ocean’s surface to the atmosphere and the release of climate-sensitive gases;
- alteration of the balance of nutrient concentrations due to ocean circulation changes and the “Atlantification” of the Arctic Ocean;
- release of soil nutrients and toxins to the Arctic Ocean from permafrost thaw;
- the impact of surface ocean changes on the seafloor, including carbon sequestration;
- changes in the timing, location and amount of plankton growth;
- migration of new species into the Arctic;
- the precarity of the Arctic ecosystem, reliant on a few dominant plankton species, and the impact on higher trophic populations.

With an extensive array and variety of approaches employed to monitor change in the Arctic, substantial datasets are being generated that cover physical, chemical and biological parameters of change in the Arctic Ocean. In this contribution, I summarise these as well as outlining the scope of the programme and the focus of the projects.
Boron isotope evidence for Southern Ocean stratification at the onset of the last glacial period

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Data derived from ice core records show us that over the past 800,000 years cold, glacial periods have been characterised by low atmospheric CO₂, reflecting a shift in carbon storage from the atmosphere to the deep ocean at these times. As the region which ventilates the majority of the deep ocean today, the Southern Ocean has been invoked to play a central role in this shift. The Southern Ocean is a high nutrient, low chlorophyll region today, meaning that there is incomplete consumption of upwelled nutrient and carbon in its surface waters. This results in a “leak” of CO₂ to the atmosphere and it has been suggested that this leak could be reduced by increased biological productivity during glacial periods. While there is evidence for increased glacial productivity north of the polar front, an additional physical mechanism in the polar Southern Ocean is needed to reduce CO₂ outgassing in this region and sequester the carbon to the deep ocean. Two such mechanisms have been proposed: increased sea-ice cover [1], which would act to impose air-sea disequilibrium on surface waters, resulting in a relatively low sea-surface pH; and enhanced surface stratification [2], which would limit exchange between carbon-rich deep waters and the atmosphere, while allowing air-sea equilibrium to continue and resulting in a high sea-surface pH relative to interglacials.

We have applied the boron isotope pH-proxy to samples of the planktic foraminifera Neogloboquadrina pachyderma from sediment core PS1506, located in the Weddell Sea. We observe an abrupt decrease in δ¹¹B (decrease in pH) across the penultimate deglaciation, consistent with a breakdown in surface stratification and the upwelling to the surface of acidic, carbon-rich deep waters. This evidence supports enhanced surface stratification as the mechanism acting to draw down atmospheric CO₂ during the onset of the last glacial cycle.

References:
The petrogenesis of pyroxenites from Kilbourne Hole, New Mexico: insights into melt-rock reactions beneath the Rio Grande rift, SW USA.

Curtis, E.¹, Granger, I.¹, Dubb, M, Privat, M.², Walker, J.¹, Stanley, T.¹, Hammond, S.J.³, Walshaw, R.D¹, Manning, C.⁴, Harvey, J¹*.

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Kilbourne Hole is a Pleistocene volcanic maar in New Mexico, USA. It is located above the southern portion of the Rio Grande Rift (RGR) which extends over 1000 km from Colorado, USA in the north to Chihuahua, Mexico in the south. The maar has been the subject of several prior studies investigating the nature of the sub-continental lithospheric mantle (SCLM) of the SW USA (e.g., [1]-[3]), where the composition of peridotite xenoliths recovered from this area were investigated.

Pyroxenite xenoliths recovered from the same locality, while only representing approximately 10% of the ultramafic material brought to the surface as accidental inclusions in alkali basalts, offer further insights into the nature of melt-rock reactions in the SCLM beneath the RGR. In addition to the interaction of alkali basalt with pre-existing peridotite at high melt-rock ratios, which formed the observed pyroxenites, a host of secondary processes recording melt-rock interaction involving compositionally diverse melts are preserved in these samples (Figure 1). Within the pyroxenites, dominated by augite, Cr-diopside and plagioclase, is preserved evidence for reactions deriving from lower melt-rock ratios, including core-and-rim textures in augite, multiples generations of veins cross-cutting all minerals present (Figure 2), plagioclase-clinopyroxene-olivine-rich melt inclusions, and trails of melt inclusions marking former grain-boundaries.

Several lines of geochemical evidence, not least the Sr-Nd isotope signatures preserved in the pyroxenites suggest that these rocks are genetically related to extrusive volcanics exposed in the Potrillo Volcanic Field [4], but unrelated to the later volcanism that brought both peridotitic and pyroxenitic xenoliths to the surface at Kilbourne Hole.

References:
Copper isotope constraints on Cu enrichment in arc magmas

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Copper ore formation is usually linked to subduction zones. Yet whether the Cu enrichment in arc magmas is caused by slab contributions or magma evolution under oxidised conditions is disputed \([1,2]\). Copper isotope ratios are a potential tracer for the Cu enrichment but have not been explored in detail in subduction zones. To explore this potential we measured Cu isotope ratios in magmas from the Izu volcanic arc in the Western Pacific ocean. In addition, we analysed serpentinites of variable metamorphic grades from the Western Alps which represent various stages of slab dehydration in a subduction setting.

The Izu arc is a global end-member where slab contributions to the magma sources are dominated by hydrous fluids whereas melts of subducted sediments are minimal. This allows a close examination of the potential for Cu mobilisation from the slab via fluids. The Izu arc Cu isotope ratios are mostly within the range previously reported for arc magmas from Kamchatka \([3]\), the Tonga rear-arc \([4]\), and the suggested composition of the depleted mantle \([3]\). Yet, Cu isotope ratios vary significantly between individual islands along the arc. These variations do not correlate with tracers for enrichment via slab components, thus suggesting that the arc magma Cu budget is dominantly mantle-derived.

Copper concentrations and Cu isotope ratios of the Alpine serpentinites are mostly within the range of unaltered peridotites \([5]\). However, \(\delta^{65}\)Cu (the permil variation in 65Cu/63Cu relative to NIST976) varies significantly within the serpentinites. This variation largely reflects processes occurring during serpentinisation. Copper concentrations in the serpentinites decrease with increased metamorphic grade but no significant correlation is observed with Cu isotope ratios, possibly indicating Cu mobilisation via fluids with mixed chloride and sulfate ligands. Our data suggest that the oceanic mantle lithosphere is not a significant contributor of Cu to arc magmas.

References:
Water in Earth’s Mantle? An Experimental and \textit{Ab-initio} study of CaSiO$_3$-walstromite

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The study of water in nominally-anhydrous mantle minerals provides fundamental knowledge of Earth’s deep-water cycle [1]. CaSiO$_3$-walstromite is a stable upper mantle mineral and the intermediate phase (4-9GPa) in the high-pressure and temperature transformation sequence to the dominant calcium bearing lower mantle phase CaSiO$_3$-perovskite [2,3]. Hydrous CaSiO$_3$–perovskite is known to be stable with a water capacity up to 0.4wt.% [4], although it may be a dry phase [5]. A minor calcium silicate intermediate phases larnite (β-Ca$_2$SiO$_4$) (9-11GPa) and a new hydrated post-hatrurite (Ca$_3$SiO$_5$) phase (17GPa) have been found to be hydrous with 4.06wt.% and 5.84 wt.% H$_2$O, respectively [6]. Given the extent of research into hydrous Ca-silicate phases there is no information for a hydrous form of the lower pressure phase CaSiO$_3$-walstromite.

Using high-pressure/temperature multi-anvil press experiments, we investigate the upper mantle phase CaSiO$_3$-walstromite and report a new high-pressure water-bearing Ca-silicate. Fourier transform infrared (FTIR) spectroscopy shows a broad hydroxyl (OH-) absorption bands at \(~3560\) cm$^{-1}$ for hydrous CaSiO$_3$-walstromite (Fig.1). In its hydrous form, hydrogen defects likely substitute Si (4H for each Si) sites rather than Ca sites (2H for each Ca). Using \textit{ab-initio} calculations, we calculate the thermodynamic stability of varies hydrous defects in at different pressure, temperature and water content condition in CaSiO$_3$-walstromite. These calculations indicate SiA position is the most favourable hydrogen substitution site in CaSiO$_3$-walstromite. We also investigate if Ti defects influence the water content and hydrogen incorporation mechanism in CaSiO$_3$-walstromite. Overall, we conclude at high-pressure calcium silicates can have water incorporated into their crystal structure. These results are essential to fully understanding the Earth’s deep-water cycle.

\textbf{Figure 1.} Fourier transform infrared (FTIR) spectra between 2800 and 3800 cm$^{-1}$ of high pressure/temperature experimental sample WA7 displaying lattice bound hydroxyl (OH) groups (indicated by black dotted lines) in CaSiO$_3$ – walstromite.

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Programme and Abstracts

Geochemical discrimination of metabasites from a Neoproterozoic accretionary complex in Anglesey, North Wales

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The Mona Complex, which extends across the Isle of Anglesey and Lleyn Peninsula, North Wales, is a Neoproterozoic-Cambrian accretionary complex formed at the margin of the Avalonian microcontinent. It hosts the Aethwy terrane, one of the world’s oldest preserved blueschist belts with a peak metamorphic age of 550-560 Ma [1]. A geodynamic model of the Mona Complex [2] suggests that the blueschist belt is a tectonic lens emplaced within the Gwna Group (or Gwna mélange), a kilometre-scale mélange ranging from sub-greenschist to greenschist facies metamorphic grades. Both the Aethwy blueschist belt and the Gwna mélange contain numerous elongate lenses of metabasites, varying in metamorphic grade and deformation extent, which locally exhibit deformed pillow lava remnants. Despite a recent geochemical study of Mona Complex greenschist basalts on the Lleyn Peninsula [3], the geochemistry and therefore the origin(s) of metabasites on Anglesey, most notably the blueschists, is poorly constrained.

A wedge of Gwna mélange found in Newborough, Anglesey, hosts numerous lenses of relatively undeformed sub-greenschist pillow basalt sequences along with dismembered units of overlying seafloor sediments. The geochemistry of these undeformed pillow basalts is similar (albeit not identical) to that of modern MORB, with variations between individual basalt lenses. Blueschists and greenschists from across Anglesey and Lleyn Peninsula have generally consistent immobile trace element contents with the Newborough pillow basalts, suggesting that they also originated from intra-oceanic magma sources. The metabasites (schists to pillows) show compositions within a MORB-OIB array, with the exception of a rare group of metabasites showing continental arc affinities. Those within the MORB-OIB array can be split into several smaller groups, including the least enriched N-MORB group that is made up predominantly of blueschists. Elemental mobility within the geochemical groups found in different parts of the Mona Complex is being assessed based on ratios of major and minor elements against Zr, which is known for its relative immobility during alteration and metamorphism [4]. This approach will benefit from the occurrence of different metamorphic grades within geochemical groups, which offers an interesting opportunity to test whether sub-greenschist, greenschist and blueschist metamorphic conditions (and more cryptic ocean floor alteration) are correlated to specific chemical changes. The preservation of hyaloclastites and peperites, for example, at Newborough also provides an insight as to how volcanic glass content, or the presence of carbonates may have affected alteration processes of the basalts.

References:

Return to the Programme
Siderophile and chalcophile element insights into the petrogenesis of pyroxenite xenoliths from Kilbourne Hole, NM: exploring a potential genetic relationship with the Potrillo Volcanic Field

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Kilbourne Hole is a volcanic maar located towards the southern end of the Rio Grande Rift (RGR) in New Mexico, USA (Figure 1). It is a site that has been previously been used to investigate sub-continental lithospheric mantle (SCLM) processes using peridotite xenoliths recently brought to the surface as accidental inclusions in alkali basalts (e.g. [1]-[3]). Less well understood is the petrogenesis of Al-augite-dominated pyroxenites found either as discrete pyroxenite xenoliths or as composite peridotite-pyroxenite xenoliths as reported in [4].

A recent study by Curtis et al (see abstract presented in this meeting) suggests that the petrogenesis of the Kilbourne Hole pyroxenites may be intimately linked to the generation and eruption of the lavas that comprise the nearby Potrillo Volcanic Field (PVF) [5]. The two suites of rocks share some major and trace element and Sr-Nd isotope characteristics, but the precise sequence of events leading to the observed pyroxenitic composition at Kilbourne Hole remains somewhat enigmatic. Moreover, using lithophile trace elements and isotope systems that are prone to resetting by metasomatic events (i.e., Sr and Nd isotopes) means that the exact relationship between the PVF and the Kilbourne Hole pyroxenites remains equivocal.

Here, we use highly siderophile elements (platinum group elements + Re) and strongly chalcophile elements (Se, Te) in addition to Re-Os isotopes to explore (i) the potential genetic relationship between the PVF basalts and Kilbourne Hole pyroxenites, (ii) establish the nature of the melt that induced the production of pyroxenites from a SCLM peridotite precursor, and (iii) explore the significance of these findings in the context of the back-arc spreading and melt generation at the RGR as a consequence of the subduction of the Farallon plate beneath the modern-day SW USA.

References:
Reevaluating the neodymium isotopic signature of Ross Sea Bottom Water – Implications for paleoceanographic reconstruction

Holder, L.1; Lambelet, M.1; Plancherel, Y.1; van de Flierdt, T.1; McKay, R. M.2; De Santis, L.3; Kulhanek, D.4 and the IODP Expedition 374 Scientists

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The $^{143}$Nd/$^{144}$Nd isotope ratio of seawater is commonly used as water mass tracer due to the ‘quasi-conservative’ behaviour of Nd away from continental margins, and a residence time shorter than the overall mixing time of the oceans. It has recently been shown that dense Antarctic Bottom Water (AABW), formed along the Antarctic margin, carries a sector specific Nd isotope fingerprint. This fingerprint may subsequently be traced along export pathways into the global ocean.

The Ross Sea is a region of interest as it produces ~25% of all AABW. So far, the only direct seawater analysis that exist to characterise the Nd isotopic composition of Ross Sea Bottom Water (RSBW) are from areas distal to the formation region in the Ross Sea. Here we present new dissolved Nd isotope results from seawater samples collected in 2018 on the GO-SHIP cruise SO4-P from sites proximal to the drainage outlets of RSBW. Our study aims to investigate for the first time, if the high salinity and low salinity variants of RSBW are isotopically distinguishable.

Our new seawater results will be used as part of a larger paleoceanographic project aiming to reconstruct variations in the export of RSBW throughout the Plio-Pleistocene. This will be achieved by firstly developing a calibration between dissolved Nd isotopes in seawater and authigenic Nd isotopes extracted from Late Holocene seafloor sediments. Secondly, we will utilise Site U1524, drilled during IODP Expedition 374 and located on the side of the Hillary Canyon which acts as a conduit for RSBW, to produce a record of RSBW outflow through time. The ultimate goal is to understand the interplay of global ocean circulation and West Antarctic Ice Sheet dynamics during Earth’s past.
A model for fine-grained sediment major element composition in terms of weathering and provenance

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The elemental composition of fine-grained sediments is principally controlled by the composition of the rocks from which they formed, ‘provenance’, and reactions with fluids transforming these source rocks into solutes and neoformed minerals in the surface environment, ‘weathering’. The challenge is how, from a given sedimentary rock, to identify the contribution of each of these factors to its formation? This challenge is compounded by the problems of working with compositional data, an unintuitive form of data due to the ‘closure constraint’ [1]. One common approach to analysing compositions is by using elemental ratios, such as the Chemical Index of Alteration (CIA) [2]. However, ratios suffer from several issues such as spurious correlations [3]. We attempt to resolve these deficiencies by developing a simple predictive and invertible model of sediment composition.

A large dataset of shale, soil, and igneous rock major element (SiO₂, Al₂O₃, Fe₂O₃, Na₂O, CaO, MgO, K₂O) compositions was compiled from the literature to calibrate our approach. To overcome the closure constraint, we utilise the centred log-ratio transformation [1]. By applying principal component analysis to the dataset we observe that most of the variation of shale compositions lies on a 2D plane defined by straight line vectors of igneous variation and weathering [4]. Comparison to soils shows that deviations from this plane are caused by diagenesis. Based on this we define a model for sediment composition as a linear sum of these two vectors with a misfit term which corresponds to diagenesis. Comparison of predicted and observed compositions indicates a 1:1 correspondence.

Our model has several benefits over other approaches. Firstly, it is able to estimate the bulk composition of the rock from which a sediment is derived. Secondly, diagenesis can be identified and quantified as deviations from a weathering-provenance composition plane. Finally, because this method uses log-ratios it has greater sensitivity to changes in weathering intensity than other indices such as the CIA. This method has the potential to improve studies of provenance and weathering in present day and ancient sediments.

References:
CO₂ perturbations during rapid climate change. What is the role of the North Atlantic?
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The northern high latitude oceans play a key role in rapid glacial climate change. Abrupt transitions of climate from cold, stadial conditions into warm Dansgaard-Oeschger (DO) events punctuate the Greenland ice core temperature records from the last glacial period. Heinrich-DO (HDO) tandems – the most extreme transitions from the coldest stadials into warm DO conditions – are linked to deep water formation and ventilation in the North Atlantic and the changing modes of Atlantic Meridional Overturning Circulation. These events also coincide with ‘pulses’ of increased atmospheric CO₂. Thus, the northern high latitude oceans could play a critical role in abrupt atmospheric CO₂ change.

Here we present a new high resolution record of surface ocean pH and pCO₂ from ODP core 980 (55°29’N, 14°42’W, 2170 m) in the North Atlantic Ocean from 41-11 kyr. Boron isotopic analysis of planktic foraminifera Neogloboquadrina pachyderma, in combination with trace element and nutrient proxy data, reveal the complex interplay of sea-ice, productivity and water mass competition on surface ocean pCO₂. We reconstruct a large decrease in pH (~2.5‰ drop of δ¹¹B) following the transition from Heinrich Stadial 4 into DO8. We attribute this transient decrease in surface pH to strengthened deep water convection following the renewal of North Atlantic Deep Water formation, causing upwelling of CO₂ rich intermediate waters. Complemented with modelling experiments conducted with the cGENIE earth system model and δ¹¹B data from another core in the North Atlantic (RAPID-17-5P, 61°29’N, 19°32’W, 2303 m) over the same event, our findings highlight the crucial role of the high latitude oceans in abrupt CO₂ change.
A Multi-Proxy Sediment Provenance Record of Antarctic Ice Sheet Change in the Early to Middle Miocene: Preliminary Results from IODP Site U1521 (Ross Sea)

J. Marschalek, T. van de Flierdt, A. Carter, P. Vermeesch, M. Siegert, K. Licht, R.M. McKay, L. De Santis, D. Kulhanek, and the Expedition 374 Scientists

An accurate record of past ice sheet change is critical for constraining models that predict future sea level contributions from Antarctica. During January to March 2018, International Ocean Discovery Program (IODP) Expedition 374 conducted a latitudinal/depth transect involving five drill sites from the outer continental shelf and rise in the eastern Ross Sea. The drilling was designed for optimal data-model integration, enabling an improved understanding of the sensitivity of Antarctic Ice Sheet mass balance during warmer-than-present climates (e.g., the early Pliocene and middle Miocene).

Site U1521 (75°41.0351'S, 179°40.3108'W, 562 m water depth) is located in the Pennell Basin on the middle-outer continental shelf, and recovered 411.5 m of Early Miocene to recent sediments. The primary objective of this site was to sample and date strata above and below Ross Sea Unconformity 4 (RSU4), previously identified in seismic stratigraphic studies.

We here analyse material from U1521 to form a low-resolution record of sediment provenance, which elucidates the erosional behaviour and extent of Miocene-to-recent ice sheets in the Ross Sea. A multi-proxy approach, using a combination of detrital zircon U-Pb dating and fine grained (<63 um) neodymium and strontium isotope ratios, was applied. Preliminary zircon U-Pb and neodymium isotope data appear to record significant shifts in provenance with two distinct end-member states. Further investigation will aim to identify the changes in ice-sheet form and flow that these data reflect.
Fluid flow in the mantle wedge of the Mariana Forearc: insights from serpentinite mud volcanoes

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Serpentinite mud volcanoes in the Mariana forearc permit sampling of fluids and xenoliths from the subducting slab and forearc mantle. IODP Expedition 366 drilled into three serpentinite mud volcanoes at different depths to slab, and other mud volcanoes were drilled in previous expeditions. The changes in major and trace element chemistry and isotopic composition (D/H, O, B and $^{87}$Sr/$^{86}$Sr) of pore fluids between seamounts are used to trace the evolution of the down going slab and water-rock interactions in the overlying mantle wedge during progressive subduction.

Pore fluids from the shallowest depths-to-slab are Ca and Sr-enriched, but otherwise solute poor, low alkalinity fluids of pH ~11. In contrast, the deeper-derived fluids have high pH (12.5) and elevated DIC, Na and Cl, as well as B and K, which are associated with the breakdown of slab sheet silicate phases, but they are depleted in Ca and Sr compared to seawater. These waters also have higher $\delta^D$ and $\delta^{11}B$ values than shallower waters ($\delta^D$ values up to +16‰; $\delta^{11}B = 14.4$‰ cf. $\delta^D < 0$‰; $\delta^{14}B = 12.2$‰). Elevated pH and DIC are likely due to liberation of slab carbonate at these depths, driving reactions that liberate CH$_4$ and OH$^-$. 
An experimental study of sepiolite dissolution rates and mechanisms at 25°C

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Two opposing models are commonly invoked to explain non-stoichiometric dissolution of multi-oxide silicates. Non-stoichiometric dissolution is either attributed to leaching of divalent metals from the mineral structure or non-stoichiometric dissolution is associated with initial congruent dissolution, followed by the precipitation of Si-rich secondary phases. This study aims to provide insights in the dissolution mechanisms of multi-oxide silicates, by assessing the dissolution rates and behaviour of sepiolite far from equilibrium under ambient conditions. Sepiolite is a Mg-rich 2:1 trioctahedral clay, characterized by its continuous inverted tetrahedral sheets and discontinuous octahedral sheets, creating channels [1]. Sepiolite is found in many important natural systems, including the CarbFix Hellisheidi CO2 storage pilot site [2]. Quantifying sepiolite dissolution rates provides us with more accurate models on Mg release, which facilitates CO2 sequestration via the precipitation of Mg-carbonates.

The steady state dissolution rates of sepiolite were determined as a function of pH from mixed flow reactor experiments based on the Si release rate. Dissolution at pH ≤5.5 and pH ≥9.25 was found to be non-stoichiometric, with relatively more Mg being released at low pH and Si being preferentially released at high pH. Steady state dissolution rates (r, in mol/cm²/s) can be described by $r = 10^{16.08}(15(a_{H^+}/a_{Mg^{2+}})^{1/6}/(1+15(a_{H^+}/a_{Mg^{2+}})^{1/6})$. Sepiolite dissolution rates at pH ≤2.7 were found to be pH independent. The Si release rate under these conditions resembles are slightly lower than those of amorphous SiO2, but higher than those measured for quartz, implicating a semi-structured solid phase. To examine the extent of non-stoichiometric dissolution and its implications on solid phase transformations, batch experiments were performed at pH 2.57, 3.40 and 4.31. From mass balance calculations, Energy-Dispersive X-ray spectroscopy (EDS) and X-Ray powder Diffraction (XRD), it can be inferred that after 688 hours, 98% of the Mg was removed from the solid structure in the pH 2.57 batch experiment, while 80% of the SiO2 was retained in an amorphous solid phase.

The rate law of sepiolite dissolution is consistent with a dissolution mechanism by which interstitial Mg is exchanged for H+, followed by the relatively slow release of Si. The stoichiometric coefficient, of 1/6, implicates that sepiolite predominantly dissolves at the channels where 6 Si atoms are bound to 1 Mg atom. The batch experiments demonstrate that non-stoichiometric steady state dissolution leads to the formation of an extensive leached layer. In concurrence with the Si-release rates at low pH, the leached layer is interpreted to be slightly more structured than amorphous SiO2. Continuous non-stoichiometric dissolution is not observed for minerals that require breakage of Si-O bonds for further divalent metal release [3]. Steady-state non-stoichiometric dissolution has been observed in wollastonite [4], for which the structure allows for Ca-release without breaking any Si-O bonds. The channel like structure of sepiolite facilitates the transport of Mg, without fully breaking the Si-tetrahedra. The preferential release of Mg from sepiolite under acidic conditions, favours precipitation of Mg-carbonates during CO2 sequestration, where acidic solutions are injected in the sub-surface.

References:
Deciphering planetary processes on the microscale

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Main Group Pallasite (MGP) meteorites originated in the earliest differentiated bodies in the Solar System, with an age of 4.5 Ga \cite{1}. They are composed primarily of an olivine network, infiltrated by iron-nickel metal \cite{2}. Both textural and chemical heterogeneity within individual samples and across the group is not fully understood. While existing oxygen isotope data suggests that MGPs all originated from the same local neighbourhood in the early Solar System, the number of parent bodies involved in their origin remains elusive due to diverse cooling rates.

Diverse formation mechanisms have been proposed, with most falling between two end-member scenarios: either MGPs represent the core-mantle boundary of a differentiated body or bodies where mantle olivine and core metal coexisted \cite{3}, or, alternatively, they are the remnants of an impact between a stripped metallic body with the olivine mantle of another planetesimal \cite{4}.

In this study, we investigate the distribution and concentration of elements across texturally varied olivine to constrain the thermal history of Pallasite meteorites. MGP olivine grains have been mapped using Energy-Dispersive X-Ray Spectroscopy (EDS) and calibrated with quantitative spot analysis using an electron microprobe, with crystal orientation obtained through Electron Backscatter Diffraction (EBSD). Combined with improved modelling of the cooling rates of small planetary bodies, these data allow new estimates for the thermal history of the parent body, which constrain the number of parent bodies required to produce the heterogeneities observed and may reveal earlier processes than those recorded by metallographic cooling rates.

References:
\begin{itemize}
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Location, location, location & proxies! Disentangling Indian Summer Monsoon dynamics across Marine Isotope Stage 5
Nilsson-Kerr, K\textsuperscript{1*}, Anand, P\textsuperscript{1}, Sexton, P. F\textsuperscript{1}, Leng, M. J\textsuperscript{2}, Hammond, S. J\textsuperscript{1}.

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Inferences on past climate are pinned on proxy understanding. Furthermore, these climate records form the basis for climate models and simulations of future climate. The proxies that have thus far been applied to reconstruct the monsoon systems have received mixed responses within the palaeoclimate community.

The Bay of Bengal represents a unique region that is subjected to some of Earth’s most dynamic interactions of ocean-atmosphere-terrestrial processes. Here we exploit two deep sea sediment cores retrieved during IODP Expedition 353 from the northern Bay of Bengal (Site U1446) and Andaman Sea (Site U1448), both situated proximal to fluvial runoff in order to reconstruct a primary signal of Indian Summer Monsoon rainfall and fluvial runoff. We present multi-proxy records of coupled Mg/Ca-derived temperatures and oxygen isotope composition ($\delta^{18}O$) in planktic foraminifera to derive the oxygen isotope composition of seawater ($\delta^{18}O_{sw-IVC}$) and bulk sediment elemental variations at a centennial-to-millennial scale across Marine Isotope Stage 5 (130-70 kyr). These records of Indian Summer Monsoon induced surface freshening and terrigenous fluxes are compared with other monsoonal records in order to assess the “global monsoon” concept across Marine Isotope Stage 5 (130-70 kyr) and explore potential discrepancies in proxy that have thus far been applied to reconstruct the monsoon.
The Origin of Earth’s Volatiles

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The origin and accretion mode of the volatile building blocks of the Earth and other terrestrial planets are major unresolved questions in planetary science, and are of particular importance because water, as a key volatile, is required for life. In particular, some posit that volatile elements were primarily delivered to Earth during main-stage accretion that was accompanied by continuous core formation. In contrast, others argue for a model in which a large late veneer was the dominant source of terrestrial volatiles.

This study utilises the double-spike technique and MC-ICP-MS to determine the mass dependent Cd and Zn isotope signatures of meteorites and terrestrial samples, with the aim of constraining the composition of the bulk Earth, before the arrival of a late veneer. This is achieved by combining estimates for the isotope composition for the bulk silicate Earth with the metal-silicate partitioning behaviour of Zn and Cd in a mass balance model. Cadmium and Zn are both volatile elements, with different chemical affinities during planetary core formation, and as such, give insights into the role played by core segregation during accretion.

Initial results from the modelling confirm that the Zn isotope composition of the bulk Earth, prior to addition of a small or large late veneer, was within the range of known carbonaceous chondrites. However, the initial Cd isotope composition of the bulk Earth must have been distinctly non-chondritic if a large late veneer is invoked. As such non-chondritic material has yet to be sampled, the result argues against a dominant role of a large late veneer in the accretion of volatiles.
Calibration of Si isotope fractionation between quartz and zircon

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Weathering and assimilation of sedimentary material into crustal melts has been explored extensively though whole-rock measurements of oxygen isotopes because supracrustal/aqueously altered rocks have fractionated O isotope compositions, typically to heavier $\delta^{18}$O values, from interaction with fluid – and this signature can be transferred to melts during anatexis of such material. Extending this observation to the mineral-scale has resulted in a clearer understanding of the relationship between crustal weathering, recycling, and isotopic composition of individual igneous phases, including zircon (e.g., [1]). Some Hadean zircons, for example, have O isotope compositions that are fractionated relative to the canonical mantle composition, implying that significant volumes of liquid water were interacting with the crust as early as 4.2 to 4.3 Ga [2]. Silicon isotopes, like oxygen, fractionate away from igneous compositions during weathering in many low-temperature water-rock interactions [3, 4] and these fractionated signatures can, as with O isotopes, be transferred to melt during anatexis [5]. However, in contrast with oxygen, silicon isotopes are unaffected during hydration of ‘fresh’ crustal rock (e.g. serpentinisation); during chemical weathering, it is only neoformation of secondary minerals that fractionate Si – providing a way of differentiating between hydrated and metasomatised (\textit{sensu lato}) material in granitoid source rocks. Recently, inferences about the silica cycle and weathering reactions have been gleaned from the analysis of both Si and O isotopes in Hadean detrital zircons, pointing to diverse supracrustal assemblages including cherts and serpentinites [6].

A significant problem when inferring source melt Si isotope composition from \textit{ex-situ} zircon is that there is limited Si isotope equilibrium fractionation data for zircon-mineral pairs. Here we apply an experimental approach to better constrain equilibrium Si isotope fractionation, as a function of temperature, between zircon and quartz by utilising both direct synthesis [7] and the 3-isotope methods [8]. Our calibration, which includes the data from both techniques to describe Si isotope fractionation between zircon and quartz, is:

$$\Delta^{30}{\text{Si}}_{(\text{Qtz-zircon})} = \frac{(0.45 \pm 0.05) \times 10^6}{T^2}$$

where $\Delta^{30}$Si is the $\delta^{30}$Si difference between quartz and zircon, and T is temperature in K. As quartz will tend to dominate the Si isotope composition of a granitoid, this fractionation factor allows us to infer the SI isotope composition of a host melt from an out of context zircon analysis given an independent estimate of T. Alternatively, this relationship may be used as a geothermometer for zircon-quartz pairs.

Do Molybdenum isotopes of Gorgona komatiites trace deeply subducted oceanic lithosphere?

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The presence of water in the Earth's mantle and its evolution are crucial for our understanding of geodynamic processes and mantle potential temperatures. Komatiites are hot, Mg-rich volcanic rocks, which result from large-degree mantle melting and hence provide valuable information on H₂O contents of the deep Earth. It is now widely accepted that komatiites are products of hydrous mantle plume melts, an idea supported by melt inclusion studies [1]. Previous works on Archaean komatiites have proposed that water and other volatile elements were entrained into mantle plumes traversing through the transition zone (~660km deep in the Earth's mantle) of the Archaean mantle [1,2]. The water was either delivered to the mantle transition zone via deep subduction, where oceanic lithosphere and sediments partly retain water in dense hydrous magnesium silicates or nominally anhydrous minerals (such as olivine), or else has been preserved in the deep mantle since the Earth's accretion.

The island of Gorgona, Columbia, is a unique place where very young (~90 Ma) komatiites have been found and studied in detail [3]. Recent research has provided evidence for a deep hydrous mantle source of the ultramafic lavas of the island [4]. Three distinctive mantle source regions (two depleted and one enriched) have been deduced based on lithophile element (Sm-Nd, Pb-Pb) isotopic heterogeneity [4]. Although this heterogeneity is consistent with entrainment of recycled oceanic lithosphere by the mantle plume, enrichment of 187Os, 186Os and 3He in some Gorgona lavas rather suggest transfer of material from the outer core to the lower mantle plume source region [5].

Molybdenum (Mo) stable isotopes offer great potential to investigate mantle heterogeneities, particularly those related to subduction processes. Mo is isotopically fractionated in sediments and altered oceanic crust producing distinct isotopically heavy or light reservoirs relative to the bulk silicate Earth. The Mo isotope signature of arc lavas can then be used as tracer of oceanic input in their mantle source [6, 7]. Ocean island and intraplate basalts often record isotopically lighter signatures compared to MORB and the primitive mantle. This difference has been related to fractionation within the subduction zone where slab fluids carry heavier Mo isotopes to the mantle wedge leaving an isotopically light slab [e.g., 6]. Recycling of slabs with light Mo isotope signatures into the deep mantle might then explain the isotopically light signatures of ocean island basalts and should also be observed in komatiites.

We will present stable Mo isotopic compositions of Gorgona komatiites, which provide information on the nature of their mantle source region. If they are isotopically lighter relative to the primitive mantle, it indeed suggests deep mantle recycling of oceanic lithosphere. This would in turn also argue for the transfer of water to the Earth's deep interior by subduction processes.

References:
Atlantification of the Barents Sea and Implications for Benthic-Pelagic Coupling

Ward, J. 1,2,3, Freitas, S.F. 1,2,3, Hendry, K. 1,3, Arndt, S 2,3, and the ChAOS Team 3

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Despite occupying just 10% of the Arctic Ocean areal extent, the Barents Sea contributes up to 40% of primary productivity and 50% of all anthropogenic carbon uptake in the Arctic Ocean, as well as the densest fraction of North Atlantic Deep Water [1,2]. At present the Barents Sea is roughly divided equally into an Arctic and Atlantic Water domain. However, recent increases in the temperature and inflow of Atlantic Water across the Barents Sea opening (termed ‘Atlantification’) has cascading, yet poorly constrained effects on ecosystem and biogeochemical dynamics, as well as potential significant implications for global biogeochemical cycles.

While the influence of Atlantification on sea-ice dynamics, primary productivity and pelagic biogeochemical cycling is receiving increasingly more attention, little is known of the potential impact on the benthic realm. This knowledge gap is critical, as in the Arctic domain up to 98% of anaerobic degradation of organic matter in the upper 12cm of the sediment column is carried out through dissimilatory iron and manganese reduction (DIR and DMR respectively), constituting a potentially significant source of bioavailable iron to shelf sediments as has been reported in the Chukchi Sea and off the West Antarctic Peninsula. Meanwhile in the southern Atlantic domain, >80% of this degradation is reported to be carried out through sulphate reduction [3,4]. This disparity is due to a number of factors, including sea-ice cover and its effect on organic matter content of the underlying sediments. Atlantification of the Barents Sea will thus most likely induce a significant shift in the dominant metabolic pathways in these shelf sediments, with the potential to stifle a significant source of bioavailable iron. This could have important implications for organic matter burial and nutrient recycling through remineralisation and thus, important implications for local and Arctic nutrient cycling and ecosystem dynamics.

Here, we use a reaction-transport model approach to explore the sensitivity of the benthic-pelagic coupling to the Atlantification of the Barents Sea. For this purpose, we run the Biogeochemical Reaction Network Simulator (BRNS) [5] over a range of model parameters, including bottom water concentrations, biological mixing parameters (bioirrigation and bioturbation coefficients), as well as organic matter concentrations and reactivities (ages) that mimic the projected changes in system dynamics associated with Atlantification. Model results indicate the potential for a curtailment in the contribution of metal reduction to organic matter degradation pathways, concurrent with a decrease in the contribution of aerobic respiration. These shifts may have important implications for nutrient, as well as alkalinity fluxes to the Arctic ocean. In addition, they show that changes in organic matter delivery and quality, as well as biological activity are the main drivers of these predicted changes.

References:
Timing of brittle deformation in the Jura mountains revealed by U-Pb dating on calcite

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The Jura fold and thrust belt (JFTB) formed as part of the Alpine orogeny as deformation propagated from thick-skinned to thin-skinned deformation during the (on-going) collision of the European and Adriatic plates. During the final stages, both the Helvetic nappes and Molasse basin were transported on a basal décollement to the north creating the Jura fold belt, where structures propagated to the north. Thereby, the Helvetic nappes include allochthonous sediments that were systematically overthrust along a basal thrust fault, while the Molasse basin formed due to erosional and sedimentary depositions consisting of marine and fluvial clastics. We will address the duration of fault propagation within the Jura and the extent to which there is along-strike temporal variation, and whether the accepted view of SE to NW propagation of structures can be validated. It was previously difficult to directly date brittle folding and thrusting events, because the minerals used for geochronology do not crystallise in low temperature conditions, and therefore the relative ages are known only indirectly from stratigraphy. These questions can now be addressed using the newly developed U-Pb calcite dating technique. Calcite formed in fractures, and as slickenfibres on fault surfaces, can be dated to provide absolute constraints for folding and faulting processes. Here, we show that calcite veins yield a spread of ages between Cretaceous and Miocene. This is indicative that the calcite preserves a longlived and complex history of a variety of processes from early diagenesis to brittle deformation. The results of this innovative study provide the first quantitative constraints on the rates and timing of deformation within the Jura mountains. This will allow refinement of structural models of the Jura of fluid-migration pathways, with wider generic implications to fold belts elsewhere.

References

Exploring lithium isotopes as a weathering tracer in natural waters

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Chemical weathering is a key process that controls Earth’s geochemical cycles and global climate, yet at present the controls on weathering are poorly understood and quantified. Lithium (Li) isotopes have emerged as a promising tracer of the modern and past weathering system [e.g. 1-3]. Unlike isotopic tracers that constrain weathering sources or fluxes [4], Li isotopes are uniquely sensitive to weathering processes because of the large isotopic fractionation that occurs during secondary clay formation [2], enabling new insights on the climate-weathering feedback.

A recent study has suggested that the Li isotopic composition of weathering fluids is recorded by speleothems [5], opening up the possibility of high resolution absolute-dated reconstructions of weathering processes in response to past climate events. However, important questions remain concerning how Li isotopes in weathering solutions are set during fluid-rock interaction, as well as the potential for seasonal variability in river and cave systems [6]. Here, I will present Li isotope data from two approaches addressing these questions.

Firstly, batch reactor experiments were carried out to replicate the weathering of shale and granite rocks in natural river waters, with monitoring of the fluid composition over hours to months as the reaction progressed. Both Li isotope and concentration data indicate that Li release from primary mineral dissolution dominates over Li removal by secondary mineral formation. These results contrast with previous experiments conducted on basaltic river sands, in which significant secondary mineral formation was inferred to have led to a net removal and fractionation of Li. Therefore, we suggest that Li isotope behaviour during weathering is sensitive to the lithology and/or prior weathering history of a sample.

In addition, to explore the magnitude and timescales of Li isotope variability in a natural system, we are analysing Li isotopes and concentrations in a series of cave drip-waters collected over the course of a year in a cave-monitoring programme in Ease Gill and White Scar caves, Yorkshire Dales. These data will allow seasonal variability in Li isotopes to be assessed, providing information on temporal variability in weathering processes and key ground-truthing for interpreting records of past changes.

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