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Examining the utility of barium isotopes as a tracer of large-scale seafloor methane venting

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Global warming has the potential to release large quantities of methane (CH_4) from marine sediments, representing a positive carbon cycle-climate feedback [1]. Unambiguous evidence of this feedback in the geological record will improve understanding of the potential risk it poses for exacerbating anthropogenic global warming. For example, climate driven sedimentary CH_4 release is one of the hypothesized mechanisms for the onset of the Paleocene-Eocene Thermal Maximum (PETM) [2]. Increased sedimentary barium (Ba) burial rates have been interpreted as evidence of this mechanism [2,3], but these records are also sensitive to other processes [4]. Stable Ba isotope variations are a new geochemical tool that may improve interpretations of such records, potentially leading to clearer geological insights into the significance of this carbon cycle-climate feedback.

This study aims to determine (1) the flux and isotope composition of Ba across the sediment-water interface associated with seafloor CH_4 venting and (2) the significance of these fluxes for the marine Ba inventory. To achieve this, Ba concentration and isotope data is presented for seawater samples at different altitudes above the seafloor (1m to 60m) collected across the Regab pockmark, a methane cold seep offshore Congo. Samples were collected with a remotely operated vehicle, providing a high resolution of sample collection within the benthic boundary layer, spanning areas of varying CH_4 venting fluxes.

The measured Ba isotope values from all sites possess $\delta^{138/134}$Ba values +0.20 to +0.40 ‰ and [Ba] values 80.0 – 90.6 nmol kg^{-1}, which are typical of ambient seawater from this depth range. Furthermore, no difference in dissolved Ba isotopes or Ba concentrations with altitude at each location is observed and there is no significant difference in seawater [Ba] and Ba isotope composition between locations featuring different dissolved seawater CH_4 concentrations.

These results are interpreted to show that there is no resolvable difference in the [Ba] vs. $\delta^{138/134}$Ba relationship over the pockmark, and any Ba fluxes are too small to resolve in a circulating water column. This likely reflects the quantitative removal of pore water Ba by barite

precipitation within the upper sediments, preventing significant Ba release to the water column.

The findings indicate CH_4 seeps do not seem to significantly impact either the dissolved Ba concentration or isotope composition of the ocean, and consequently makes the use of sedimentary Ba concentration and isotope records as a tracer of past CH_4 release events questionable. These insights suggest caution should be held when developing Ba isotopes as a novel tracer of past large-scale seafloor methane release i.e. PETM sediments. The study also provides insights on the influence of methane seep environments on Ba isotopes and the factors governing the stable isotope distribution of Ba in both modern and ancient sediments and oceans.

Reference:

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[3] Frieling et al., (2019), *Palaeoceanography and paleoclimatology,* 34*,* 546-566

[4] Bridgestock et al., (2019), *Earth and Planetary Science Letters*, 510, 53-63