Hydrothermal iron flux variability following rapid sea-level changes
Jennifer L. Middleton¹², Charles H. Langmuir³, Sujoy Mukhopadhyay⁴, Jerry F. McManus³⁴, Jerry X. Mitrovica¹

¹Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA, ²Department of Earth and Planetary Sciences, University of California Davis, Davis, California, USA, ³Lamont-Doherty Earth Observatory, Palisades, New York, USA, ⁴Department of Earth and Environmental Sciences, Columbia University, New York, New York, USA

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Introduction

This supporting information includes additional methodical details regarding the extraterrestrial helium-3 constant flux proxy (Text S1, Figure S1), the non-hydrothermal Fe and Cu fluxes for KN207-2-GGC3 (Figure S2), a comparative figure demonstrating the similarity between the down-core Fe contents of KN207-2-GGC3 and 4 previously analyzed neighboring sediment cores (Figure S3), and two plots illustrating the low sensitivity of the relative sea-level curve presented in the main text to ice sheet model and mantle viscosity profile (Figure S4).
Text S1: Determination of Extraterrestrial Helium-3

Natural variability in the distribution of rare, helium-rich IDPs within marine sediments can generate uncertainties when using limited sample sizes to determine representative sediment \(^{3}\text{He}_{\text{ET}}\). Such uncertainties are a function of the area-time product of the sediment aliquots analyzed, determined by dividing the sample mass (in g) by the mass accumulation rate (in g m\(^{-2}\) a\(^{-1}\)), and are reduced by increasing sample masses and analyzing replicate samples [Farley et al., 1997]. A typical area-time product of 0.25 m\(^2\) a\(^{-1}\) yields a 1σ uncertainty in sediment \(^{3}\text{He}_{\text{ET}}\) of \(\approx 20\%\), much larger than the 1-2% analytical uncertainties for each sample [Farley et al., 1997; Patterson and Farley, 1998]. In contrast, sediment concentrations of terrigenous \(^{4}\text{He}\) are well constrained, with 1σ values reflecting the analytical uncertainties of 1-2% (Table S2).

Sample aliquots of 1.5 g and the bulk sedimentation rates of GGC3 sediments lead to an areal-time product of 0.15 m\(^2\) a\(^{-1}\). To constrain the \(^{3}\text{He}_{\text{ET}}\) uncertainties associated with such an areal-time product, replicate helium analyses were performed on 108 samples from GGC3 and an additional Mid-Atlantic core, KN207-2-GGC6 (29.21°N, 43.23°W, 3018 m water depth), with a similar mass accumulation rate (Table S2). A Gaussian fit to the distribution of fractional difference in measured \(^{3}\text{He}_{\text{ET}}\) within the replicated sample population yields estimated 1σ uncertainties of 28% for single analyses of sediment \(^{3}\text{He}_{\text{ET}}\). Applying the 28% uncertainty to the average \(^{3}\text{He}_{\text{ET}}\) of the replicated sample population yields a 1σ uncertainty of 0.66 pcc STP \(^{3}\text{He}_{\text{ET}}\) g\(^{-1}\).

The 28% 1σ uncertainty in sediment \(^{3}\text{He}_{\text{ET}}\) is reduced for replicated samples by \(n^{1/2}\), where \(n\) is the number of replicated analyses for a given sample. To further reduce uncertainties associated with sampling rare He-rich particles, sediment fluxes reported in the main text represent a 3-point running mean through the \(^{3}\text{He}_{\text{ET}}\)-derived sediment rain rates. Propagation of additional uncertainties in the IDP \(^{3}\text{He}_{\text{ET}}\) flux from space (7.7 ± 1.7 x 10\(^{13}\) cm\(^3\) STP cm\(^{-2}\) ka\(^{-1}\); [Higgins, 2001]) leads to typical 1σ values for \(^{3}\text{He}_{\text{ET}}\)-derived sediment rain rates of 30% for each sampled depth and 18% for the 3-point running mean.

Computation of \(^{3}\text{He}_{\text{ET}}\) concentrations requires negligible helium contributions from mantle sources (\(^{4}\text{He}/^{3}\text{He} \sim 8\) R\(_{\text{a}}\) for average mid-ocean ridge basalts (MORB) [Graham, 2002]) such as gas-bearing basalt fragments or vent fluid-bearing sulfides. Mantle-like \(^{4}\text{He}/^{3}\text{He}\) ratios have been observed in near-vent deposits [Jean-Baptiste and Fouquet, 1996; Stuart et al., 1994], but there is no correlation between high concentrations of hydrothermal Fe and \(^{3}\text{He}_{\text{ET}}\) (Figure S1) in the GGC3 sediment core.

Some basalt fragments occur in GGC3 sediments and the upper limit of MORB \(^{4}\text{He}\) contamination from these fragments was constrained as follows. Basalt fractions \(f_{\text{basalt}}\) in the carbonate-free sediments were calculated using Mg/Rb ratios assuming a two component mixture of MORB (Mg/Rb = 2.54 wt.%/ppm; [Gale et al., 2013; Table S1]) and a hypothetical zero-Mg endmember. Resulting \(f_{\text{basalt}}\) values range from 1 to 3%. Calculations of \(f_{\text{basalt}}\) made using the MORB Mg/Th ratio of 18.7 [Gale et al., 2013] also ranged from 1 to 3%. These values overestimate basalt concentrations within He analyzed aliquots as non-basalt sediment is not completely Mg-free and ICP aliquots were not sieved at the 64 μm level, which systematically removed coarse basalt chips relative to the finer terrigenous sediments.
Representative He concentrations and isotope ratios for the fine basalt fragments were determined from the average values of 3 MORB glass samples (KN207-2 D4-1, D42-4, and D57-1; Table S3) dredged from the TAG and Broken Spur segments of the Mid-Atlantic Ridge on the same cruise as the GGC3 and GGC6 sediment core retrievals. Sample D42-4 is known to be gas rich due to its popping behavior when recovered from the seafloor. MORB samples were crushed and sieved to the <200 μm level and He was extracted by heating under vacuum at 1285°C following the same procedure as the sediment samples (yielding a $^3\text{He}/^4\text{He}$ ratio of 8.19 $R_a$ and a $^{3}\text{He}_{\text{MORB}}$ concentration of $2.85 \times 10^{-11}$ cc STP g$_{\text{basalt}}^{-1}$).

$^{3}\text{He}_{\text{ET}}$ concentrations were then recalculated, using the same two component assumption as above, after subtracting maximum $^{3}\text{He}_{\text{MORB}}$ and $^{4}\text{He}_{\text{MORB}}$ contributions proscribed for each sample based on the Mg/Rb-derived f$_{\text{basalt}}$ values. Resulting basalt-corrected $^{3}\text{He}_{\text{ET}}$ concentrations were only 1 to 7% lower than original $^{3}\text{He}_{\text{ET}}$ estimates. Thus, the fractional $^{3}\text{He}_{\text{ET}}$ correction due to basalt contamination is lower than natural uncertainty of the uncorrected $^{3}\text{He}_{\text{ET}}$ measurements and does not affect interpretation of $^{3}\text{He}_{\text{ET}}$-derived sediment rain rates. We conclude that a two-component mixture of IDP and terrigenous helium remains a valid assumption for the GGC3 sediment core.
Figure S1. Mir zone hydrothermal Fe vs. $^3$He$_{ET}$ for GGC3 carbonate-free sediments (a) and bulk sediments (b). Note that there is no correlation between high Fe contents (indicative of increased hydrothermal contribution) and high $^3$He$_{ET}$, suggesting no significant vent-fluid $^3$He contamination of the $^3$He$_{ET}$ signal.
**Figure S2.** Mir zone (GGC3) non-hydrothermal fluxes. 3-point running mean fluxes (points) of non-hydrothermal Cu (a) and Fe (b) within 1σ uncertainty envelope (solid lines). Gray bar highlights the interval of the LGM-hydrothermal peak.
Figure S3. a, Location of KN207-2-GGC3 (yellow star) and neighboring Mirzone sediment cores of Cherkashev [1995] (numbered circles). b, Carbonate-free Fe contents of GGC3 and the Cherkashev cores are plotted as a function of depth, due to the lack of age control on the Cherkashev cores. Similarities in Fe deposition between cores from all sides of the Mirzone suggest that the hydrothermal Fe peaks were caused by net increases in hydrothermal plume flux, rather than variations in plume directionality.
Figure S4. Relative sea-level prediction sensitivity. a, Predictions of relative sea-level (RSL) change and b, its rate above the TAG vent field on the Mid-Atlantic Ridge since 20 ka. The solid black lines are computed using the ICE-5G ice history and the VM2 mantle viscosity model [Peltier, 2004], as in the bottom frames of main text Figure 3. Blue lines, ICE-6G ice history and the VM5a viscosity model [Peltier et al., 2015]. Red lines, ICE-5G ice history and a viscosity model (LM) within the class of models favored in a suite of studies [e.g., Nakada and Lambeck, 1989; Mitrovica and Forte, 2004]. The LM model is characterized by an elastic lithosphere of thickness 95 km, an upper mantle viscosity of $5 \times 10^{20}$ Pa-s and a lower mantle viscosity of $5 \times 10^{21}$ Pa-s. The dotted black line is the same as the solid black line, except that 3-D variations in mantle viscosity and lithospheric thickness, including plate boundaries, are included in the calculation [e.g., Kendall et al., 2006]. This line is indistinguishable from the solid black line in b. The figure demonstrates that the main features of the relative sea-level prediction in Figure 3 are insensitive to the details of ice history and Earth structure.
Table S1. Compiled Isotopic Data and ICP-MS Results.

Table S2. Sediment Helium Analyses.

Table S3. MORB Helium Analyses.

Table S4. Benthic Oxygen Isotope Analyses.

Table S5. Radiocarbon Measurements.

References


Stuart, F., G. Turner, R. C. Duckworth, and A. Fallick (1994), Helium isotopes as tracers of trapped hydrothermal fluids in ocean-floor sulfides, Geology, 22(9), 823-826.