Geochemical Background and Methodology

1. Helium isotopes

Terrestrial ^{4}He ($^4He_{terr}$) has been successfully used as an eolian dust proxy in marine sediments [Patterson et al., 1999; Mukhopadhyay et al., 2001; Winckler et al., 2005, 2008; Marcantonio et al., 2009; Serno et al., 2014], Antarctic ice [Winckler and Fischer, 2006], and corals [Mukhopadhyay and Kreycik, 2008; Bhattacharya, 2012]. ^{4}He in continental crust material is produced by α-decay of U/Th-series elements. In source rocks, U and Th are concentrated in accessory mineral phases like zircon or uraninite, which have sufficient helium retentivity to retain radiogenic ^{4}He during weathering [e.g., Mamyrin and Tolstikhin, 1984; Martel et al., 1990]. When grains weather to typical grain sizes of long-range transported eolian dust (~2-5 μm) [Tsoar and Pye, 1987; Rea and Hovan, 1995; Serno et al., 2014], they do not accumulate more ^{4}He from α-decay since the recoil length of α-particles is ~10-30 μm [Farley, 1995; Ballentine and Burnard, 2002]. Volcanic source rocks have ^{4}He concentrations around two to three orders of magnitude lower than concentrations of typical continental crust material [Mamyrin and Tolstikhin, 1984; Patterson et al., 1999; Kurz et al., 2004; McGee, 2009].

The contrast between relatively high ^{4}He concentrations in eolian dust material and negligible concentrations in volcanic input makes ^{4}He a particularly useful proxy for eolian dust input in sediments from the SNP because this region is characterized by large lithogenic input other than eolian dust, including volcanic ash, hemipelagic material, riverine input or ice-rafted debris (IRD) [e.g., Olivarez et al., 1991; Bailey, 1993; Jones et al., 1994, 2000; McKelvey et al., 1995; Weber et al., 1996; Shigemitsu et al., 2007; Serno et al., 2014]. Because of the geological setting of the surrounding land in the SNP, non-eolian contributions to the SNP, like hemipelagic material, riverine input or IRD, have a geochemical signature similar to fine-grained volcanic ash [e.g., Jones et al., 1994; McKelvey et al., 1995; Shigemitsu et al., 2007] and are virtually ^{4}He-free. Additional confirmation for this approach has been recently provided by a spatial reconstruction of modern eolian dust input with independent geochemical

fingerprinting based on rare earth elements and ^{232}Th [Serno et al., 2014]. Therefore, we conclude that ^{4}He provides an estimate of eolian dust that is insensitive to volcanic contributions to the sediments.

Helium isotopes in marine sediments are a binary mixture of terrestrial material and extraterrestrial interplanetary dust particles (IDPs) [e.g., Patterson et al., 1999; Winckler et al., 2005]. Helium is not associated with biogenic or authigenic phases [e.g., Farley, 1995; Patterson et al., 1999]. The concentration of sedimentary $^{4}He_{terr}$ can be calculated using a two-component mixing model and the measured ^{4}He concentration ($^{4}He_{meas}$) and helium isotope ratio [$(^{3}He/^{4}He)_{meas}$] in the sample [Patterson et al., 1999]:

$$^{4}He_{terr} = {}^{4}He_{meas} \times \{[(^{3}He/^{4}He)_{meas} - (^{3}He/^{4}He)_{IDP}] / [(^{3}He/^{4}He)_{terr} - (^{3}He/^{4}He)_{IDP}]\} \quad (1)$$

with $(^{3}He/^{4}He)_{IDP}$ = helium isotope ratio of IDPs = 2.4×10^{-4} [Nier and Schlutter, 1990, 1992], and $(^{3}He/^{4}He)_{terr}$ = helium isotope ratio of terrestrial material = 3×10^{-8} [Mamyrin and Tolstikhin, 1984; Farley, 2001].

Between 70 and 230 mg of freeze-dried hand-crushed bulk sediment was wrapped in aluminum foil cups and loaded into the vacuum furnace of the gas inlet system, and analyzed for helium isotopes on a MAP 215-50 mass spectrometer at the Lamont-Doherty Earth Observatory (LDEO) following the procedure described in Winckler et al. [2005]. Calibration was performed every 4-5 samples using a known volume of a standard gas with a helium isotope ratio of $16.45R_A$ (with $R_A = {}^{3}He/^{4}He$ in air = 1.384×10^{-6}). Standard reproducibility was ~0.5% (1σ) for ^{4}He and ~1.5% (1σ) for $^{3}He/^{4}He$. Procedural blanks yielded ~0.1 ncc STP of ^{4}He (ncc STP = nano cubic centimeter at standard temperature and pressure) with atmospheric isotopic composition, and represented blank corrections of <1% ^{4}He for the samples. For 21 samples, we analyzed 2-4 replicates, with an average reproducibility of 8.7% (1σ) for ^{4}He. The contribution of $^{4}He_{terr}$ to total ^{4}He is >98% for all samples.

2. *Uranium/Thorium isotopes*

Excess ^{230}Th in the sediments ($xs^{230}Th_0$) has been established as a constant-flux proxy [e.g., Bacon, 1984; Henderson et al., 1999; Francois et al., 2004; Anderson et al., 2006]. Production of ^{230}Th occurs in the water column by α-decay of ^{234}U. Due to the short residence time of ^{230}Th in the water column as a result of efficient particle scavenging (<40 years) [e.g., Bacon and Anderson, 1982; Anderson et al., 1983; Henderson et al., 1999; Francois et al., 2004], its scavenged flux to the seafloor is approximately equal to its production rate (β_{230} = 2.67 x 10^{-5} dpm/cm^3/kyr) in the overlying water column [Francois et al., 2004]. Mass accumulation rates (MARs) estimated using this approach offer advantages over conventional stratigraphic MARs in that the ^{230}Th-normalized fluxes are insensitive to lateral sediment redistribution, are only slightly sensitive to age model uncertainties, are determined for every sample, and do not rely on the determination of dry bulk density [e.g., Henderson et al., 1999; Francois et al., 2004; Anderson et al., 2006]. The ^{230}Th-normalized MAR for a constituent i, MAR_i, with a concentration c_i in the sediment deposited at a specific water depth z was calculated following the method described in Francois et al. [2004]:

$$MAR_i = (c_i \times \beta_{230} \times z) / xs^{230}Th_0 \quad (2)$$

$xs^{230}Th_0$ (in dpm/g) is corrected for radioactive decay since its time of deposition, the fraction supported by uranium within lithogenic material (mean detrital ^{238}U/^{232}Th = 0.5 ± 0.1) [Taguchi and Narita, 1995], and the fraction of the in-situ ^{230}Th produced by decay of authigenic ^{238}U [Francois et al., 2004; Anderson et al., 2006].

For U/Th isotopes, ~200 mg of freeze-dried and hand-crushed bulk sediment was weighed into 50 ml Teflon beakers and spiked with ^{229}Th and ^{236}U prior to a complete acid digestion and anion exchange chromatography following the

method of Fleisher and Anderson [2003]. Samples were analyzed by isotope dilution using a high-resolution Element XR ICP-MS at LDEO. ^{238}U concentrations were derived using the measured $^{235}U/^{236}U$ and a $^{238}U/^{235}U$ ratio of 137.88 for the SRM129 standard [Steiger and Jäger, 1977], measured with every batch of 21 samples and one procedural blank. Blank corrections were <1.5% and average analytical reproducibility <2.2% (1σ) for ^{230}Th, ^{232}Th, and ^{238}U. Long-term reproducibility, based on multiple analyses of a surface sediment sample from the INOPEX cruise, is 2.2% (1σ) for ^{230}Th, 2.4% (1σ) for ^{232}Th, and 4.2% (1σ) for ^{238}U. Reproducibility of 2-3 replicates from 15 of the SO202-7-6 samples is 2.5% (1σ) for ^{230}Th, 2.6% (1σ) for ^{232}Th, and 2.9% (1σ) for ^{238}U.

3. Planktic foraminiferal radiocarbon dates

The planktic foraminifera *Neogloboquadrina pachyderma* (sinistral), a subsurface-dwelling species living at ~50-200 m water depth in the North Pacific [Kuroyanagi et al., 2002], was picked from the >150 μm fraction in 40 samples between mid-core depths of 130.5 and 4.5 cm for accelerator mass spectrometry (AMS) radiocarbon analyses. Each sample has a 1 cm core depth resolution. AMS radiocarbon analyses were performed on ~5-10 mg of planktic foraminifera at the National Ocean Science Accelerator Mass Spectrometry (NOSAMS) facility at the Woods Hole Oceanographic Institution in Woods Hole, Massachusetts, USA, and at the ETH accelerator facility in Zürich, Switzerland. Radiocarbon ages are reported according to the convention outlined by Stuiver and Pollach [1977] and Stuiver [1980]. For two core depths (50.5 and 56.5 cm), duplicate measurements were performed, with radiocarbon ages of 11850 ± 50 yr and 12286 ± 47 yr for 50.5 cm, and 12812 ± 50 yr and 12850 ± 50 yr for 56.5 cm core depth. The reason for the offset between the measured radiocarbon ages at 50.5 cm is uncertain. For the following conversion of radiocarbon to calendar ages, we used the average of the duplicate measurements, with the uncertainty representing the deviation of the duplicates (12068 ± 218 yr for 50.5 cm and 12831 ± 19 yr for 56.5 cm). Conversion of radiocarbon to calendar ages was performed using the downloadable version of Calib 7.0.4

(http://calib.qub.ac.uk/calib/calib.html) [Stuiver and Reimer, 1993] with the Marine13 calibration curve [Reimer et al., 2013] and a laboratory error of 0. The Marine13 calibration incorporates a global ocean reservoir correction of ~400 yr. Using the Marine13 calibration curve in Calib 7.0.4, an estimate of ΔR, the difference in reservoir age of the local region of interest and the model ocean [Reimer et al., 2013], has to be provided. All reservoir ages reported here are total reservoir ages, including the global reservoir age correction of 400 yr and ΔR. We selected the reported 2σ calibrated calendar age range of the probability distribution [Telford et al., 2004]. To generate point estimates, we used the reported median of the probability distribution, with the deviation of the upper and lower limits of the reported 2σ range from the median as an upper and lower error estimate. All calendar ages are reported in yr BP.

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