Equatorial Pacific productivity and dust flux during the mid-Pleistocene climate transition

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Received 6 May 2005; revised 30 August 2005; accepted 19 September 2005; published 17 December 2005.

[1] We present a helium isotope record for core TT013-114PC from the central equatorial Pacific (140°W, 4°N, 4432 m water depth) spanning a period of 1 million years. We focus on the time interval from 560 to 800 kyr, largely coinciding with the mid-Pleistocene climate transition (MPT) when the dominant period of the Earth's climate variability shifted from 41 kyr to 100 kyr. The terrigenous ⁴He concentrations from our study correlate very well with published titanium concentrations in this core strongly supporting the use of terrigenous ⁴He as a monitor of continental dust. Normalizing titanium and terrigenous ⁴He concentrations to ³He suggests that the dust supply during the MPT was approximately 30% lower compared to the subsequent period (560–100 kyr). The ³He-normalized barium, aluminum and phosphorus concentrations, trace elements with a predominantly biogenic source in these sediments, are relatively constant. This is in contrast to previous studies that reported an apparent rise of titanium-normalized productivity proxies. Rather than a significant increase in productivity during the MPT, we conclude that the dust flux to the central equatorial Pacific was reduced and that the export productivity was approximately constant during this period of climate reorganization.

Citation: Winckler, G., R. F. Anderson, and P. Schlosser (2005), Equatorial Pacific productivity and dust flux during the mid-Pleistocene climate transition, *Paleoceanography*, *20*, PA4025, doi:10.1029/2005PA001177.

1. Introduction

[2] The most prominent feature of the equatorial Pacific is the upwelling of CO_2 -rich subsurface water which creates the largest natural source for the net CO_2 flux from the ocean to the atmosphere [*Takahashi et al.*, 1997, 2002]. The rich nutrients brought up by the upwelling water promote phytoplankton growth, making it one of the major sites of organic carbon export to the deep sea. Export productivity in this region represents a significant part of the global ocean carbon cycle. Thus, the equatorial Pacific has been suggested to have a major influence on global climate via feedbacks involving CO_2 [e.g., *Archer and Maier-Reimer*, 1994; *Broecker and Henderson*, 1998].

[3] One of the key questions in paleoceanography is whether biological productivity in the Pacific has changed substantially during the past. If so, have productivity changes been a direct response to climate forcing and have they provided significant feedbacks through altered air-sea partitioning of CO_2 ? Modern El Niño events disrupt the natural carbon fluxes and create significant imbalances in the ocean-atmosphere CO_2 exchange [e.g., *Feely et al.*, 1999] highlighting the importance of understanding the role of the ocean carbon cycle in climate change and its

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sensitivity to climate perturbations. In order to discern the coupling between ocean circulation, productivity and climate, many studies have attempted to estimate past ocean productivity and to develop proxies to reconstruct the history of ocean productivity from marine sediments [e.g., *Bopp et al.*, 2003; *Kohfeld et al.*, 2005].

[4] Another important variable in the global climate system is atmospherically transported mineral aerosols (dust). The concentration of dust in the atmosphere influences the climate system through a variety of processes, e.g., affecting radiative forcing (scattering and absorption of light), condensation processes and by providing nutrients (e.g., iron) to terrestrial and marine biological systems [e.g., Harrison et al., 2001]. A potential factor controlling dust delivery to the equatorial Pacific is the latitudinal position of the Intertropical Convergence Zone [Rea, 1994]. Only a few paleorecords of dust deposition in the equatorial Pacific are available. Most of them are limited to the late Pleistocene and show complex and variable patterns. Whether there is a consistent relationship between maximum dust fluxes and interglacial conditions [Rea, 1990, 1994] or glacial conditions [Anderson et al., 2005] or no consistent relationship at all [Murray et al., 1995], remains a matter of debate.

[5] The focus of this paper is on reevaluating the export productivity and dust deposition in the equatorial Pacific during the mid-Pleistocene climate transition (MPT). The MPT, i.e., the period when the dominant cyclicity of the Earth's climate evolved from \sim 41 kyr to \sim 100 kyr, is thought to represent a period of major climate reorganization [e.g., *Raymo et al.*, 1997]. There is recent evidence for

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a strong increase in export production in the central equatorial Pacific during the period from 560–800 kyr, broadly coinciding with the MPT [*Murray et al.*, 2000]. Extreme increases in export productivity were found along a transect of cores across the equator at 140°W. The maxima of the export productivity observed during the MPT are much more pronounced than its variability throughout the subsequent glacial/interglacial periods. If this interpretation is correct, then the export productivity in response to the mid-Pleistocene climate transition was greatly enhanced compared to any other time during the late Pleistocene, and represents perhaps the largest biogeochemical response to climate change in the Pacific Ocean during the Pleistocene.

[6] To further investigate the cause of this important feature, we have analyzed ³He and ⁴He concentrations over the last 1 Myr in core TT013-PC114 in the central equatorial Pacific. This core represents the site along the 140°W transect where the most pronounced relative changes in export productivity were found previously. In equatorial Pacific sediments, ³He is dominated by the extraterrestrial ³He signal from interplanetary dust particles (IDPs) and represents a tool to determine accumulation rates independent from age models [e.g., Marcantonio et al., 1995, 1996, 2001]. We apply this tool to existing records of paleoproductivity (barium, aluminum, phosphorus) and, using existing titanium data and our new ⁴He record, to reconstruct the dust flux to the central equatorial Pacific. We find relatively constant ratios of the paleoproductivity indicators to ³He throughout the record, and lower Ti/³He and ⁴He_{terr}/³He ratios during the MPT. Rather than an increase in productivity during the MPT, we conclude that there was a reduced flux of mineral aerosols to this region during this period of climate reorganization.

2. Geochemical Background

2.1. Reconstruction of Fluxes

[7] Reconstructing fluxes of sedimentary components is vitally important to many areas of paleoceanographic research. At any particular site, sediment burial rates may be influenced by the rain rate of particulate matter settling through the water column as well as by sediment redistribution. Consequently, strategies have been sought to correct for sediment redistribution and to derive past changes in rain rate unbiased by the influence of sediment focusing.

[8] For sediments deposited during the past 250-300 kyr, normalizing to decay-corrected concentrations of unsupported ²³⁰Th (²³⁰Th⁰_{ex}) is the method of choice (for a review see *Francois et al.* [2004]). For older sediments, where the 75 kyr half-life of ²³⁰Th precludes its use, there is no consensus concerning the best approach to correct for sediment redistribution.

[9] Some investigators have sought to reconstruct past changes in biological productivity by normalizing trace elements with a predominantly biogenic source (e.g., barium, phosphorus, aluminum) to titanium, the source of which is lithogenic [e.g., *Latimer and Filippelli*, 2002; *Murray et al.*, 2000]. While the elemental ratio approach circumvents the need of accumulation rates for the derivation of fluxes and thus avoids potential complications arising from sediment redistribution, it implicitly relies on

the assumption that variability in the supply of titanium has been much less than variability in export production throughout the period of study.

[10] Other investigators have suggested that the flux of interplanetary dust particles (for a review see Farley [2001]) to Earth has been relatively uniform in space and time so that the known flux of extraterrestrial ³He contained within interplanetary dust particles (IDPs) (for a review see Farley [2001]) can be used to correct for sediment redistribution [e.g., Marcantonio et al., 1995, 1996, 2001; Winckler et al., 2004]. By normalizing to 230 Th⁰_{ex}, the flux of ³He to the Earth has been shown to be constant over the past 250 kyr at a rate of $(0.8 \pm 0.24) \cdot 10^{-12}$ cm³ STP cm⁻² kyr⁻¹ [Marcantonio et al., 1996, 2001], consistent with estimates from GISP2 and Vostok ice cores $((0.62 \pm 0.27) \cdot 10^{-12} \text{ cm}^3)$ STP cm⁻² kyr⁻¹ and $(0.77 \pm 0.25) \cdot 10^{-12}$ cm³ STP cm⁻² kyr^{-1} , respectively [*Brook et al.*, 2000]). The flux of IDPs on longer timescales has not been extensively calibrated so far, but efforts to do so by normalizing ³He to ¹⁰Be, a constant flux proxy under specific conditions, are underway. In spite of the lack of a dedicated calibration data set, the accretion rate of ³He for the time period of interest to this study (250 kyr to 1 Myr) can be provisionally constrained by using records of ³He [Patterson and Farley, 1998] and ¹⁰Be (A. Mangini, personal communication, 2005) from core ODP806 in the western equatorial Pacific. Beryllium 10 data have been corrected for radioactive decay and for geomagnetically induced production rate changes following the procedure outlined by Frank et al. [2000]. The resulting ³He/¹⁰Be⁰_{corr} ratios (see Figure 1 and caption) show no trend over the time interval from 250 kyr to 1 Myr with a variability around the mean ${}^{3}\text{He}/{}^{10}\text{Be}^{0}_{\text{ corr}}$ ratio of 23% (1 σ). These findings support the use of extraterrestrial ³He as a constant flux proxy on the Myr timescale.

[11] Using the ³He approach, the accumulation rate of any sedimentary component can be calculated as

$$[F]_{\text{proxy}} = [\text{proxy}] \cdot \text{MAR}_{\text{CF}} = [\text{proxy}] \cdot \frac{f_{^{3}\text{He}}}{[^{3}\text{He}]}$$
 (1)

where MAR_{CF} is the bulk mass accumulation rate derived from the constant flux proxy, [proxy] is the concentration of the component, [³He] is the measured ³He concentration and f_{3He} is the constant flux of ³He to the sediments. As shown in equation (1), there is a linear relationship between the proxy/³He ratios and the flux itself, an essential principle that will be used in the discussion of this paper.

2.2. Barium, Aluminum and Phosphorus as Paleoproductivity Proxies

[12] The barium cycle has been shown to be strongly associated with biological activity [e.g., *Bishop*, 1988; *Goldberg and Arrhenius*, 1958; *Paytan et al.*, 1996]. Marine barite (BaSO₄) is an authigenic mineral formed within decaying organic matter. It has been shown empirically that its flux is positively correlated with increasing export production [*Dymond et al.*, 1992; *Francois et al.*, 1995]. Excess barium, defined as the fraction of barium not associated with lithogenic material, is frequently used to define barite abundance in sediments and has been extensively used as a proxy to infer paleoproductivity patterns



Figure 1. The ${}^{3}\text{He}/{}^{10}\text{Be}^{0}_{\text{corr}}$ ratios for core ODP806 in the western equatorial Pacific Ocean. ³He concentrations are from Patterson and Farley [1998] and ¹⁰Be concentrations are from A. Mangini (personal communication, 2005). The ³He concentrations were measured using aliquots of homogenized (strip) samples representing 20-30 kyr time intervals, while the ¹⁰Be analysis was generated using discrete samples. The ¹⁰Be concentrations have been corrected for radioactive decay and for variability in the ¹⁰Be production due to variable strength of the magnetic field of the Earth after Masarik and Beer [1999]. The intensity of the magnetic field was reconstructed for the past 800 kyr after Guyodo and Valet [1999] and was reconstructed for the time period before 800 kyr after Valet and Meynadier [1993]. To compensate for the different resolution of both records, ${}^{3}\text{He}{}^{10}\text{Be}{}^{0}_{\text{corr}}$ ratios were determined at the depth of the ${}^{10}\text{Be}$ sample by dividing the ${}^{3}\text{He}$ concentration in the composite sample spanning the depth of the discrete beryllium analysis by the respective ¹⁰Be⁰_{corr} concentration. The straight line gives the mean of the ³He/¹⁰Be⁰_{corr} ratio; the dotted lines indicate the standard deviation of the mean.

[e.g., Dean et al., 1997; Francois et al., 1995; Nürnberg et al., 1997; Schwarz et al., 1996].

[13] Studies in the equatorial Pacific have documented that significant amounts of aluminum are contained both in lithogenic phases and authigenic phases [e.g., *Dymond et al.*, 1997; *Murray and Leinen*, 1996; *Murray et al.*, 1993]. For these sediments, scavenged excess aluminum, the

component of aluminum that is unsupported by the lithogenic fraction, has been found to reflect the bulk biogenic particle flux through the water column and to the seafloor and is therefore used as paleoproductivity proxy [*Banakar et al.*, 1998; *Dymond et al.*, 1997; *Murray and Leinen*, 1996; *Murray et al.*, 1993, 2000].

[14] Similarly, the use of phosphorus as paleoproductivity proxy is based on observed correlations between the phosphorus distribution in sediments and biogenic fluxes through the water column to the seafloor [e.g., *Delaney*, 1998; *Froelich et al.*, 1982; *Latimer and Filippelli*, 2002].

2.3. Terrigenous ⁴He as Dust Proxy

[15] Terrigenous ⁴He has been introduced as a proxy of dust in marine sediments by *Patterson et al.* [1999]. As old continental material contains high radiogenic ⁴He concentrations (e.g., $2000 \cdot 10^{-9}$ cm³ · g⁻¹ in Asian dust [*Farley*, 1995]) compared to relatively low ⁴He contents in other terrigenous components (e.g., $5 \cdot 10^{-9}$ cm³ · g⁻¹ in volcanic ashes [*Patterson et al.*, 1999]), the terrigenous ⁴He signal tracks old continental dust and is not affected by other terrigenous components. Helium does not exist as organic complex, nor is it incorporated into biogenic phases; therefore terrigenous ⁴He can be assumed to be entirely contained within detrital material.

3. Samples and Methods

[16] Samples were analyzed from piston core TT013-PC114 (4.04°N, 139.85°W, 4432 m water depth) collected in the framework of the JGOFS (Joint Global Ocean Flux Study) program. The age model was determined by correlating the CaCO₃ concentration profile in PC114 to the CaCO₃ records of nearby cores with δ^{18} O constrained age models (cores TT013-PC18 at 2°S and TT013-PC72 at the equator, both along 140°W [*Knowlton*, 1998; *Murray et al.*, 2000]).

[17] Helium isotopes were measured on samples taken between 20 cm and 775 cm core depth, a section that represents a time interval of about 935 kyr. Individual sediment sample aliquots of 1.5 to 2.5 g were leached in 200 mL of 0.5 N acetic acid to remove carbonate material which carries no substantial amount of helium and were then washed three times in distilled water. This resulted in residues of about 200 mg which were wrapped in aluminum foil and placed in the furnace of the gas inlet system. Helium was extracted from the samples at $\sim 1300^{\circ}$ -1400°C. During extraction the furnace was kept exposed to a liquid nitrogen cooled charcoal trap in order to remove CO₂, H₂O and organic compounds. Further purification was performed by exposure to a SAES getter at room temperature. The gas was then collected on a cryogenically cooled charcoal trap held at ~ 13 K and helium was separated from neon by heating the trap to 45 K. Abundance and isotopic analyses were performed with a MAP215-50 noble gas mass spectrometer calibrated with a known volume of a Yellowstone helium standard (MM) with a ³He/⁴He ratio of 16.45 R_a (where R_a = $({}^{3}\text{He}/{}^{4}\text{He})_{air}$ = 1.384 \cdot 10⁻⁶ [*Clarke et al.*, 1976]). Typical hot blanks were 1–2 \cdot 10⁻¹⁰ cm³ STP ⁴He with approximately atmospheric ³He/⁴He ratios, and represent small corrections (<1%) to the samples. The

Table 1. Helium Isotope Data of TT013-PC114^a

| Depth, | Age, | ³ He, | ³ He/ ⁴ He | ⁴ He _{terr} | |
|------------------|------|--|----------------------------------|---|--|
| cm | kyr | $10^{-12} \text{ cm}^3 \text{ STP g}^{-1}$ | (R/R_a) | $10^{-8} \text{ cm}^3 \text{ STP g}^{-1}$ | |
| 20 ^b | 15 | 1.422 ± 0.028 | 32.2 ± 0.5 | 2.604 ± 0.051 | |
| 40 | 30 | 1.227 ± 0.026 | 35.3 ± 0.6 | 2.001 ± 0.042 | |
| 60 | 46 | 1.183 ± 0.024 | 37.9 ± 0.6 | 1.764 ± 0.036 | |
| 80^{b} | 60 | 1.7215 ± 0.034 | 30.0 ± 0.5 | 3.420 ± 0.069 | |
| 120 ^b | 113 | 5.843 ± 0.117 | 33.6 ± 0.6 | 10.11 ± 0.21 | |
| 140 | 145 | 1.392 ± 0.028 | 27.0 ± 0.4 | 3.145 ± 0.061 | |
| 150 | 153 | 1.529 ± 0.032 | 26.4 ± 0.5 | 3.542 ± 0.075 | |
| 165 | 164 | 2.997 ± 0.060 | 35.7 ± 0.6 | 4.822 ± 0.094 | |
| 185 ^b | 179 | 8.855 ± 0.186 | 27.3 ± 0.4 | 19.691 ± 0.382 | |
| 200^{b} | 219 | 4.385 ± 0.088 | 29.2 ± 0.5 | 9.034 ± 0.183 | |
| 215 | 259 | 2.084 ± 0.040 | 43.1 ± 0.7 | 2.623 ± 0.051 | |
| 225 | 265 | 3.209 ± 0.064 | 27.0 ± 0.4 | 7.264 ± 0.141 | |
| 235 ^b | 271 | 6.399 ± 0.128 | 25.8 ± 0.5 | 15.271 ± 0.322 | |
| 275 | 334 | 1.805 ± 0.036 | 28.9 ± 0.5 | 3.756 ± 0.076 | |
| 290 ^b | 358 | 5.865 ± 0.117 | 33.5 ± 0.6 | 10.226 ± 0.207 | |
| 310 | 388 | 9.047 ± 0.181 | 39.2 ± 0.6 | 12.890 ± 0.250 | |
| 325 | 400 | 2.212 ± 0.044 | 40.6 ± 0.7 | 3.012 ± 0.061 | |
| 350 | 452 | 1.444 ± 0.030 | 29.1 ± 0.5 | 2.982 ± 0.060 | |
| 380 ^b | 481 | 8.788 ± 0.176 | 46.3 ± 0.7 | 10.060 ± 0.195 | |
| 415 | 525 | 1.551 ± 0.033 | 34.9 ± 0.6 | 2.561 ± 0.054 | |
| 435 | 564 | 3.875 ± 0.078 | 31.9 ± 0.5 | 7.152 ± 0.145 | |
| 460 ^b | 583 | 2.933 ± 0.062 | 39.3 ± 0.7 | 4.158 ± 0.084 | |
| 485 | 637 | 0.999 ± 0.020 | 43.0 ± 0.7 | 1.264 ± 0.025 | |
| 500 | 659 | 0.698 ± 0.017 | 43.7 ± 1.0 | 0.863 ± 0.022 | |
| 510 | 672 | 1.191 ± 0.024 | 70.4 ± 1.1 | 0.725 ± 0.014 | |
| 525 | 677 | 0.994 ± 0.023 | 33.6 ± 0.7 | 1.720 ± 0.039 | |
| 555 ^b | 687 | 4.429 ± 0.097 | 41.7 ± 0.8 | 5.811 ± 0.128 | |
| 580 ^b | 710 | 0.953 ± 0.019 | 41.1 ± 0.7 | 1.277 ± 0.026 | |
| 595 | 724 | 1.333 ± 0.027 | 48.8 ± 0.8 | 1.418 ± 0.028 | |
| 605 | 736 | 0.869 ± 0.018 | 47.3 ± 0.9 | 0.966 ± 0.021 | |
| 630 ^b | 766 | 1.637 ± 0.033 | 37.3 ± 0.6 | 2.493 ± 0.050 | |
| 655 ^b | 795 | 3.671 ± 0.081 | 41.4 ± 0.8 | 4.916 ± 0.108 | |
| 690 ^b | 837 | 4.912 ± 0.098 | 38.4 ± 0.7 | 7.100 ± 0.144 | |
| 745 | 907 | 4.794 ± 0.096 | 41.4 ± 0.7 | 6.370 ± 0.124 | |
| 775 ^b | 935 | 3.194 ± 0.064 | 30.3 ± 0.6 | 6.278 ± 0.143 | |

^aHelium concentrations are reported in units of cm³ STP per gram of sediment. The ³He⁴He ratios are normalized to the atmospheric ratio ($R_a = 1.384 \times 10^{-6}$ [*Clarke et al.*, 1976]. Here ⁴He_{terr} represents the terrigenous fraction of ⁴He (see sections 2.3 and 4.2). Quoted errors are the analytical uncertainties of the mass spectrometric analysis.

^bMarked samples were duplicated; in these cases, average values are reported.

analytical precision of the mass spectrometric analysis is on the order of 1% for ⁴He and 2–3% for ³He. Note that the natural variability in the ³He concentration of a sample is controlled by the statistical effect of the small number of IDPs hosted in the sediments and is a greater source of uncertainty than the analytical precision. Replicates were run for samples as indicated in Table 1, and the reproducibility distribution is in good agreement with the model prediction of *Farley et al.* [1997]. Following *Farley et al.* [1997] and *Patterson and Farley* [1998], the 1 σ uncertainty is therefore estimated to be ~20% for a single and ~15% for a duplicate analysis, respectively.

4. Results and Discussion

4.1. The ³He record

[18] The observed 3 He/ 4 He ratio (see Table 1 for results) reflects the mixing of extraterrestrial and terrigenous material. Assuming typical 3 He/ 4 He ratios for these endmembers, 2 \cdot 10⁻⁸ for terrigenous helium [*Ozima and Podosek*, 1983] and 2.4 \cdot 10⁻⁴ for extraterrestrial helium

[*Nier and Schlutter*, 1992], the amount of IDP-derived ³He in the sediments can be estimated from the observed ${}^{3}\text{He}/{}^{4}\text{He}$ ratio using a simple two component mixing model

$$\frac{{}^{3}\text{He}_{\text{IDP}}}{{}^{3}\text{He}_{\text{meas}}} = \left(\frac{1 - \frac{({}^{3}\text{He}/{}^{4}\text{He})_{\text{terr}}}{({}^{3}\text{He}/{}^{4}\text{He})_{\text{meas}}}}{1 - \frac{({}^{3}\text{He}/{}^{4}\text{He})_{\text{terr}}}{({}^{3}\text{He}/{}^{4}\text{He})_{\text{ET}}}}\right)$$
(2)

where "ET" denotes the extraterrestrial and "terr" denotes the terrigenous component. Following this approach, it was determined that the non-IDP component of the ³He measured in our samples is negligible throughout the core. The lowest proportion of IDP-derived ³He (reflected by the lowest ³He/⁴He ratio) is found in a sample at 235cm (271 kyr) where it still contributes 99.95% of the total ³He concentration. Therefore we use the total ³He concentration as approximation for extraterrestrial ³He.



Figure 2. Down-core profiles of the ³He concentration and calcium carbonate content (on a reverse scale to orient the peaks with the ³He record) plotted against age of sediments for core PC114. Shaded region represents the time period between 560 and 800 kyr, identified as representing the mid-Pleistocene climate transition by *Murray et al.* [2000].



Figure 3. Property-property plot of the terrigenous fraction of ⁴He, calculated as defined in the text, versus titanium concentration for core PC114. Solid squares represent samples from the MPT; open squares represent samples from before and after the MPT. The solid line is a linear regression to the data from the MPT; the dashed line is a fit to the data representing the pre- and post-MPT period. The correlation factors for the linear fits are 0.98 and 0.95, respectively.

[19] Depth profiles of the ³He concentration and the $CaCO_3$ content are shown in Figure 2. Carbonate data as well as barium, aluminum, phosphorous, and titanium data are from *Murray et al.* [2000] and have been retrieved from World Data Center for Paleoceanography (http://www.ncdc.noaa.gov/paleo/index.html).

[20] The most prominent feature of the carbonate record is the pattern of cyclic and high-amplitude fluctuations of CaCO₃ preservation with depth as has been described by previous investigators [*Arrhenius*, 1952; *Farrell and Prell*, 1989]. The CaCO₃ preservation record exhibits a strong periodicity at about 100 kyr and is generally described by good preservation during glacial stages and poor preservation during interglacials.

[21] Helium 3 concentrations vary between $0.7 \text{ and } 9 \cdot 10^{-12} \text{ cm}^3 \text{ STP g}^{-1}$. The correlation between ³He concentration and carbonate content throughout the glacial/interglacial stages back to MIS20 is very good (Figure 2). The ³He profile apparently reflects variable dilution of the constant ³He input from IDPs by CaCO₃, the principal component of these sediments. The only sample that does not follow the tight correlation is that at 907 kyr for which the ³He concentration seems to be anomalously high. As there is no analytical reason to discard the measurement and a replicate sample was not available for analysis we decided to keep this value in the data set and flag it ("?") in Figures 3–5.

4.2. The ⁴He Record

[22] Helium 4 concentrations range from 10 to $230 \cdot 10^{-9}$ cm³ STP g⁻¹. In contrast to ³He, the ⁴He signal in the

sediment is dominated by the terrigenous component. Using a simple two-component mixing model

$$\frac{{}^{4}\text{He}_{\text{terr}}}{{}^{4}\text{He}_{\text{meas}}} = \frac{({}^{3}\text{He}/{}^{4}\text{He})_{\text{meas}} - ({}^{3}\text{He}/{}^{4}\text{He})_{\text{ET}}}{({}^{3}\text{He}/{}^{4}\text{He})_{\text{terr}} - ({}^{3}\text{He}/{}^{4}\text{He})_{\text{ET}}}$$
(3)

the amount of terrigenous ⁴He was determined (Table 1). The relative contribution of terrigenous ⁴He (⁴He_{terr}) to total ⁴He component ranges from 60 to 85%. The correlation between titanium and ⁴He_{terr} concentrations at PC114 is excellent (Figure 3) and supports the application of ⁴He_{terr} as monitor of continental dust. The strong correlation between the two proxies is inconsistent with the interpretation of Kryc et al. [2003] who recently challenged the traditional view that titanium is predominantly supplied by dust and argued that up to 80% of the titanium in biogenic sediments from the central equatorial Pacific occurs as organic complexes. Because terrigenous ⁴He does not build organic complexes, we would expect significant variability around the mean relationship between titanium and ⁴He_{terr} if, in fact, a large and variable fraction of the titanium in PC114 sediments occurred as organic complexes. Contrary to this expectation, the good correlation between ⁴He_{terr} and titanium indicates the absence of a large and variable nondetrital component of titanium. Titanium, like ⁴He_{terr}, appears to be contained predominantly within detrital mineral phases and track the dust input to equatorial Pacific sediments.

4.3. Reevaluation of the Export Productivity Record During the Mid-Pleistocene Climate Transition

[23] Figure 4 compares the barium, aluminum and phosphorus data normalized to extraterrestrial ³He to the same proxy data normalized to titanium as originally presented by *Murray et al.* [2000]. The most prominent feature of the titanium-normalized records are pronounced maxima of the elemental ratios (Ba/Ti, Al/Ti, P/Ti) during the MPT which have been interpreted as indication for a strong increase in export production [*Murray et al.*, 2000]. Normalizing to ³He eliminates the apparent peaks observed in the titanium-normalized barium, aluminum and phosphorus records during the MPT. Whereas the Ba/³He and P/³He ratios are relatively constant throughout the record, the Al/³He ratio even seems to show slightly lower values during the MPT (see discussion below).

[24] The ³He data allow us to discriminate between increased export production (increase of numerator) and reduced titanium flux (decrease of denominator) as the principal cause for increased element/Ti ratios. The long-term trend of the Ti/³He record (Figure 5b) is complementary to the trend in the Ba/Ti ratios (Figure 5a) shown as an example of the records in Figure 4. In particular, the ³He-normalized titanium values are significantly lower during the MPT than during the post-MPT period. This implies that the supply of titanium at this site was significantly lower during the MPT than during the subsequent glacial/interglacial cycles and that the maxima of the element ratios (Ba/Ti, Al/Ti, P/Ti) during the MPT is due to reduced titanium supply, rather than due to increased biological productivity. Thus, on the basis of the ³He-normalized



Figure 4. Comparison of the down-core profiles of (a) barium, (b) aluminum, and (c) phosphorus, normalized to titanium (black) and to ³He (red). Shaded region represents the time period between 560 and 800 kyr, representing the mid-Pleistocene climate transition.



Figure 5. Comparison of down-core profile of the (a) Ba/Ti ratios with the (b) ³He-normalized titanium and (c) ³He-normalized ⁴He_{terr} record. The bold lines indicate the long-term trend and represent running averages over \sim 80 kyr. This corresponds to 13-point running averages in the case of Ba/Ti and 3-point running averages in the case of the ³He-normalized values in order to compensate for the different time resolution of both records (³He, mean resolution \sim 25 kyr; Ba, mean resolution 6 kyr). Shaded region represents the time period between 560 and 800 kyr, representing the mid-Pleistocene climate transition.

Table 2. Mean Element Ratios for MPT and Post-MPT^a

| | Post-MPT | MPT | $\Delta_{\mathrm{MPT/PostMPT}}$ |
|--|----------------|-----------------|---------------------------------|
| Ti/ ³ He, $10^{13} \ \mu g \ cm^{-3}$ Ba/Ti, $g \ g^{-1}$ ⁴ He _{terr} / ³ He | 9.3 ± 0.9 | 6.5 ± 0.8 | 0.70 |
| Ba/Ti, g g^{-1} | 7.3 ± 0.7 | 10.3 ± 1.44 | 1.4 |
| ⁴ He _{terr} / ³ He | 18580 ± 2350 | 13130 ± 1690 | 0.71 |
| Al/ ³ He, $10^{15} \ \mu g \ cm^{-3}$ | 2.5 ± 0.2 | 1.92 ± 0.19 | 0.77 |

^aMid-Pleistocene climate transition (MPT) is 560-800 kyr as defined by *Murray et al.* [2000]. Post-MPT is 100-560 kyr. The time period 560-100 kyr was selected to represent the "plateaus" both in Ti³He and Ba/Ti ratios in the period following the MPT without including the drop in titanium supply and strong increase in Ba/Ti ratios toward the present. The choice of the upper limit, 100 kyr, is somewhat arbitrary; however, sensitivity tests show that the conclusions do not depend on the exact choice of the upper limit. Mean ratios were calculated for the long-term trend of the data in order to eliminate the high glacial/interglacial variability in the time period 560-100 kyr. Quoted errors are standard deviations of the mean ratios.

records (Figure 4) we infer that productivity during the climate reorganization of the MPT was comparable to the productivity levels in the late Pleistocene.

4.4. Dust Input During the Mid-Pleistocene Climate Transition

[25] In the central equatorial Pacific, the predominant source of titanium is aeolian dust [e.g., *Chuey et al.*, 1987; *Murray and Leinen*, 1993]. The drop in titanium supply during the MPT could reflect either a substantially reduced dust input to this site or, alternatively, a major change in dust provenance, from a dust source with higher titanium concentrations to a dust source low in titanium.

[26] The good correlation between ⁴He_{terr} and titanium argues against a significant change in provenance. As shown in Figure 3, the correlation between the two proxies is almost identical for the samples during the MPT and before and after. A change in provenance of the dust, without an accompanying change in the Ti/⁴He_{terr} ratio of the source region, appears to be highly unlikely. Therefore we conclude that the drop in titanium supply reflects a reduced dust input during the MPT compared to late Pleistocene levels. Because the supply of dust to the ocean surface is affected by climate and is potentially variable in time [e.g., *Harrison et al.*, 2001; *Kohfeld and Harrison*, 2001], a significant change in dust flux appears to be a likely response to a major climate reorganization.

[27] Latitudinal variations in the position of the Intertropical Convergence Zone (ITCZ) represent a potential control on the dust delivery to the equatorial Pacific [Rea, 1994]. Given the proximity of core PC114 (4°N) to the modern position of the ITCZ, it is an important question whether the variability of the titanium flux recorded at PC114 could have been influenced by a meridional shift in the mean position of the ITCZ or is rather representative of changes in the dust supply to the entire central equatorial Pacific. Evidence against a significant influence by an ITCZ shift comes from the study of *Murray et al.* [2000] who reported alike patterns of variability of Ba/Ti profiles at station PC114 and station PC32 at 5°S along the meridional transect at 140°W, particularly a similar amplitude of the strong MPT maximum. If there was a shift in the position of the ITCZ during the MPT, we would expect the Ba/Ti ratios at PC114 to be more affected than those at PC32 at 5°S. On

the basis of the similarity between the Ba/Ti records at 4° N and 5° S we therefore conclude that the drop in titanium supply recorded at PC114 most likely represents a reduction in dust supply throughout the region during the MPT rather than a relocation of the ITCZ.

4.5. Quantifying the Variability of the Dust Input

[28] The long-term trend of the dust flux, as inferred from the ⁴He_{terr}/³He ratios (Figure 5c), shows the same basic features as the Ti/³He record (Figure 5b). Following the relatively low dust input levels during the MPT, there is an increase to higher values in the period 550–250 kyr and a subsequent decrease during the past ~250 kyr to values comparable to the MPT. While the Ti/³He ratios are relatively stable during the MPT, there is considerable variability in the ratios during the period since 560 kyr.

[29] We quantitatively evaluated the extent to which dust fluxes increased following the MPT by comparing the mean values for the MPT time slice and the subsequent period (560–100 kyr, Table 2). The mean Ti/³He ratio during the MPT is about 70% of the Ti/³He ratio during the post MPT period, indistinguishable from the corresponding changes in the ⁴He_{terr}/³He ratio. Both independent dust proxies, titanium and ⁴He_{terr}, indicate a consistent reduction of ~30% in aeolian dust flux to this site during the mid-Pleistocene climate reorganization compared to average late Pleistocene levels.

[30] The information in Table 2 can also be used to check if the reduced dust level can quantitatively explain the apparent productivity peak. Scaling the Ba/Ti ratio of the post-MPT period with this reduced titanium supply yields

$$\left(\frac{\text{Ba}}{\text{Ti}}\right)_{\text{MPT}} = \frac{\left(\frac{\text{Ba}}{\text{Ti}}\right)_{\text{Post-MPT}}}{\Delta_{\text{Ti}}} = \frac{7.3}{0.7} = 10.4 \qquad (4)$$

and matches well the observed value (10.3, Table 2). Accordingly, the 30% decrease of the titanium supply fully accounts for the observed mean increase of the Ba/Ti ratio during the MPT without invoking any change of the barium flux, and thus paleoproductivity.

4.6. Cross-Check With Aluminum Data

[31] The ³He-normalized aluminum record (Figure 4b) provides a sensitive internal consistency check of the hypothesis of a reduced dust flux during the MPT. Particularly, it provides an independent approach to evaluate whether reduced dust flux or an increased IDP and thus ³He flux is responsible for the patterns shown in Figure 5. Since only part of the total aluminum is contained in dust (section 2.2), its flux should be less affected by a reduced dust input than that of the "pure" dust tracers. Accordingly, if the flux of mineral aerosols were lower during the MPT, then we would expect the amplitude of the change in $Al/^{3}He$ to be less than that for $Ti/{}^{3}He$ or ${}^{4}He_{terr}/{}^{3}He$, respectively. On the other hand, if the flux of IDPs was greater during the MPT (and responsible for the features of Figure 4), then we would expect the MPT reductions in Al/³He, Ti/³He and ⁴He_{terr}/³He to be of similar amplitude. We observe a Al/³He reduction that is smaller, 23% versus \sim 30% from Ti/³He and ${}^{4}\text{He}_{\text{terr}}/{}^{3}\text{He}$ (Table 2), consistent with the observation of



Figure 6. (a) Comparison of the down-core profile of barium, normalized to titanium (black) and 230 Th $_{ex}^{0}$ (red). (b) The 230 Th $_{ex}^{0}$ -normalized titanium record for the past 150 kyr. (Thorium data are available from US JGOFS database at http://usjgofs.whoi.edu.)

slightly higher (by about 12%) Al/Ti ratios during the MPT. This observation strongly supports the view that the flux of IDPs remained relatively constant while the flux of dust was diminished during the MPT.

4.7. Glacial/Interglacial Variability

[32] Apart from the long-term trend discussed so far, the dust and productivity records show considerable variability on shorter timescales. However, owing to the limited resolution of the post-MPT ³He record the pattern of the late Pleistocene glacial/interglacial cycles cannot be resolved.

[33] A high-resolution ${}^{230}\text{Th}^{0}_{ex}$ record is available for PC114 for the past 150 kyr [Anderson et al., 2005] (raw data archived in the U.S. JGOFS database, http://usjgofs. whoi.edu) and can be used to evaluate the variability of the productivity and the dust signal on a glacial-interglacial timescale. As an example, the Ba/Ti ratios show significant variability over the past 150 kyr (Figure 6a), including a strong increase at about 65 kyr to relatively high values during the past 50 kyr. When the barium concentrations are normalized to 230 Th_{ex}, the variability is eliminated, and the $Ba/^{230}Th_{ex}^{0}$ ratios are relatively constant over the past 150 kyr. The $Ti/^{230}Th_{ex}^{0}$ record is complementary to the Ba/Ti values indicating that the temporal variability of the Ba/Ti ratios is dominated by changes in the dust (Ti) supply and is not due to changes in export production. The $^{230}\text{Th}_{ex}^{0}$ -based results are analogous to our observations for the long-term trend, based on the ³He normalization, in showing that the variability of Ba/Ti ratios reflects changes in the supply of titanium rather than changes in the flux of barium. Productivity at this location has been relatively constant over the last glacial cycle, as it was through the MPT.

5. Conclusions

[34] Reconstruction of paleofluxes on longer timescales provides a challenge to paleoceanography. Extraterrestrial ³He is shown to be a valuable tool to normalize fluxes on a Myr timescale. Our study also confirms terrigenous ⁴He as a reliable tracer of continental dust in the sedimentary record that provides independent and complementary information to other dust proxies such as titanium.

[35] Normalizing titanium and terrigenous ⁴He to ³He reveals dust fluxes to the central equatorial Pacific during the MPT that were lower by approximately 30% compared to the subsequent time period. Relatively constant $Ti/^{4}He_{terr}$ ratios rule out a change in provenance of the dust to be responsible for the reduced titanium supply during the MPT.

[36] The ³He normalization of barium, aluminum, and phosphorus implies that export productivity was relatively constant throughout the entire record. In particular, the mid-Pleistocene climate transition does not appear to be characterized by elevated productivity levels. This implies that the increase in dust levels from the MPT to the post-MPT phase did not have a significant impact on the export productivity.

[37] Our record implies that mean dust levels recorded in the central equatorial Pacific core site were higher during the late Pleistocene (<560 kyr) than during the previous period (>560 kyr). If this trend was extrapolated toward the early Pleistocene, then it would imply that the 100 kyr world was on average dustier than the 41 kyr world, consistent with the change in amplitude of the global ice volume. Alternatively, if the dust levels were higher during the period before ~ 900 kyr one could speculate about modulation of the dust input with a low frequency. With its limitation to \sim 930 kvr. our present record is definitely too short to check these hypotheses, and future work is needed toward answering this question. This will also help to understand whether the reduced dust flux during the MPT is a local signal or reflects a regional or global climate pattern.

[38] Comparing the dust flux records (Figure 5) with the calcium carbonate record (Figure 2) reveals a strong similarity. The lower dust levels during the MPT correlate with a period of high calcium carbonate contents (Figure 2) indicative of exceptionally good preservation. Also, both records show a relatively stable signal over the period of the MPT, in comparison to the considerably higher variability during the subsequent glacial/interglacial cycles. Assuming that carbonate preservation is driven by changes in the paleochemistry of the deep Pacific Ocean, i.e., less corrosive waters because of better ventilation of the bottom waters, the systematic correlation between both records indicates a large-scale coordination between the Southern Ocean, where the deep Pacific Ocean is ventilated, and the lowlatitude wind fields of the Northern Hemisphere, the probable source region of the dust. This potential long-term teleconnection merits further investigation.

[39] Acknowledgments. Joerg Schaefer and Sidney Hemming provided valuable comments on an earlier draft of this paper. We thank Martin Stute for his collaboration and continued support of the lab. We are grateful to A. Mangini, B. Schwarz, C. Strobl, and M. Christl from the "Radiometric Dating Group" at the Academy of Sciences Heidelberg who generously provided us with the ¹⁰Be data from ODP806 and to Martin Frank for help with the magnetic field corrections for the ¹⁰Be data. We thank Marty Fleisher and Yong Lao for the ²³⁰Th data. Reviews by Ralf Tiedemann and an anonymous referee are much appreciated. Funding for this research was provided by NSF grant OCE-97-11870. G.W. acknowledges support of the Leopoldina Fellowship from Deutsche Akademie der Naturforscher Leopoldina as well as support from the Comer Science and Education Foundation. Samples were generously provided by the curator at the University of Rhode Island core repository (OCE-9102410). This is LDEO contribution 6805.

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