PALEOCEANOGRAPHY, VOL. 16, NO. 5, PAGES 468–478, OCTOBER 2001

Evidence from authigenic uranium for increased productivity of the glacial Subantarctic Ocean

Zanna Chase[1] and Robert F. Anderson

Lamont-Doherty Earth Observatory, Columbia University, Palisades, New York, USA

Department of Earth and Environmental Science, Columbia University, New York, New York, USA

Martin Q. Fleisher

Lamont-Doherty Earth Observatory, Columbia University, Palisades, New York, USA

Abstract. Authigenic uranium is precipitated in reducing sediments and therefore responds both to changes in particulate organic carbon flux to the sediment and to changes in the oxygen concentration of bottom waters. By examining a large number of cores over a wide latitudinal and depth range in the Atlantic sector of the Southern Ocean, we hope to distinguish between a predominantly productivity-driven authigenic U signal and a circulation or sediment-focusing signal. We find little to no authigenic U in Holocene sediments throughout the South Atlantic Ocean. Glacial sediments north of ~40°S lack authigenic U, whereas sediments from the Subantarctic Zone show substantial enrichments in authigenic U relative to the Holocene (up to ~5.3 ppm). The widespread distribution of glacial U enrichment, even in cores with no glacial-interglacial change in mass accumulation rate, implies that U deposition was not caused by an increased supply of organic carbon via sediment focusing. Authigenic U and organic carbon in a shallow core (~1000 m) from the Subantarctic region show the same glacial increase found in the deep cores. Because this site was well ventilated during the Last Glacial Maximum, its record provides further evidence that increased glacial productivity of the Subantarctic region contributed to the high concentrations of authigenic U found in the glacial sections of deep cores.

1. Introduction

In the modern Southern Ocean (SO), upwelling supplies abundant nutrients to surface waters yet phytoplankton productivity is relatively low, so nutrients are returned to the deep ocean largely unused. This inhibition of phytoplankton growth has been attributed to intense grazing [*Hart*, 1942], light limitation [e.g., *Mitchell et al.*, 1991], trace metal limitation [*de Baar et al.*, 1995; *Martin et al.*, 1990], and combinations of these factors. During glacial periods the physical setting of the SO must have been very different from today, with possible changes in wind strength [*Petit et al.*, 1981] and dust inputs [*De Angelis et al.*, 1987; *Petit et al.*, 1999], sea ice and iceberg distributions [*Cooke and Hays*, 1982; *Crosta et al.*, 1998], meltwater stabilization [*Labeyrie et al.*, 1986], frontal positions [*Prell et al.*, 1979], and upwelling intensity [*Francois et al.*, 1997]. Given their role in determining modern primary productivity in the SO, these environmental changes are likely to have affected productivity during the last glacial period. Reconstructing this biological response is important, as it has been suggested that more complete utilization of nutrients in the SO would be a significant perturbation to the global carbon cycle and might help explain the observed decrease in atmospheric CO_2 during glacial periods [e.g., *Knox and McElroy*, 1984; *Sarmiento and Toggweiler*, 1984] (but see *Archer et al.* [2000]).

Past changes in biological productivity can be reconstructed either by estimating past nutrient utilization in surface waters or by reconstructing particle export. Given a constant supply of nutrients via upwelling, increased nutrient utilization produces a decrease in surface water nutrient concentrations and an increase in particle export. Attempts to reconstruct past changes in SO surface water nutrient concentrations using various tracers including $\delta^{13}C$, Cd/Ca, and $\delta^{15}N$ have yielded inconsistent results and generally fail to produce clear evidence of the glacial drawdown expected from increased phytoplankton productivity. Research directed toward unraveling these apparent conflicts continues [e.g., *Anderson et al.*, 2001; *Kohfeld et al.*, 2000; *Rickaby and Elderfield*, 1999], but at present there is no consistent interpretation of glacial nutrient concentrations based on multiple proxies.

Perhaps the most obvious tracer of past biogenic particle export is the concentration of organic carbon in the sediment. However, because most organic carbon is remineralized before reaching the sediments, the signal is small (<0.5% of sediment mass). Furthermore, there is considerable debate over whether the sediment concentration of organic carbon reflects primarily differences in supply (i.e., export production) or preservation efficiency [e.g., *Calvert and Pedersen*, 1992; *Hartnett et al.*, 1998]. Biogenic opal is another obvious candidate, and productivity reconstructions based on the accumulation and abundance of biogenic opal suggest either a decrease in total SO productivity during the Last Glacial Maximum (LGM) or no change [*Charles et al.*, 1991; *Mortlock et al.*, 1991]. However, opal is (1) subject to variable dissolution, both in the water and sediment column e.g., *Nelson and Gordon*, 1982; *Van Bennekom et al.*, 1988], (2) only an indicator of export production by diatoms, and (3) not necessarily produced by diatoms in constant proportion to carbon [*Hutchins et al.*, 1998; *Takeda*, 1998].

In an effort to avoid some of the ambiguities associated with these previously used tracers of export production, a number of new proxies have been developed and applied to the SO, including ^{231}Pa:^{230}Th and ^{10}Be:^{230}Th ratios, biogenic barium, and authigenic U. The radionuclide flux proxies, ^{231}Pa:^{230}Th and ^{10}Be:^{230}Th, are

[1]Now at Monterey Bay Aquarium Research Institute, Moss Landing, California, USA.

Paper number 2000PA000542.
0883-8305/01/2000PA000542$12.00

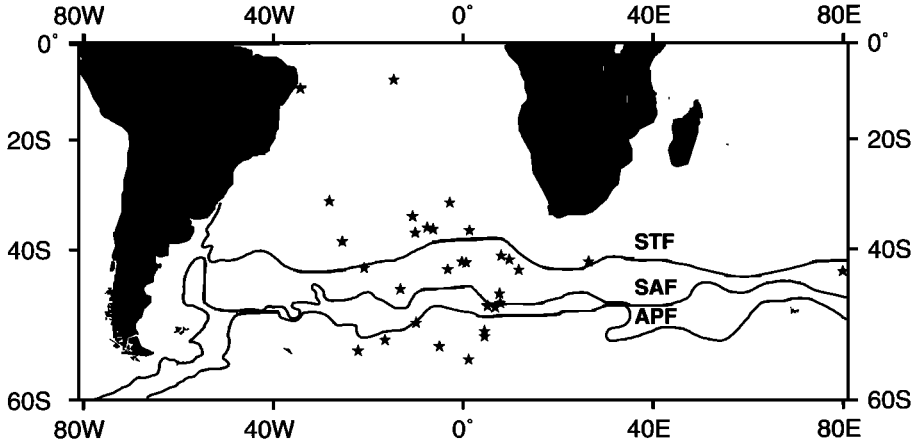

Figure 1. Geographic distribution of the 30 cores examined in this study. Also shown are the present locations of the Subtropical Front (STF), the Subantarctic Front (SAF), and the Antarctic Polar Front (APF) [after *Orsi et al.*, 1995]. Core names are given in Table 1.

based on the slightly longer residence times of Pa and Be with respect to scavenging compared to the residence time of Th [*Anderson et al.*, 1983, 1990; *Bacon*, 1988; *Kumar et al.*, 1993]. These proxies argue for a glacial increase in the export of biogenic particles from the Atlantic and Indian sectors, north of the modern Antarctic Polar Front (APF) [*Francois et al.*, 1993; *Kumar et al.*, 1995, 1993]. However, recent evidence suggests that high ^{231}Pa:^{230}Th ratios in the SO are associated with a high opal content of sinking particles and not necessarily with high particle flux [*Chase*, 2001; *Walter et al.*, 1997].

The association between the flux of nondetrital barium (excess barium) and particulate organic carbon (POC) in sediment traps [*Dymond and Collier*, 1996; *Francois et al.*, 1995] suggests that excess barium in sediments may serve as a proxy for POC rain rate. Barium accumulation rates during the LGM were generally lower than those today in sediments south of the APF and exhibit little glacial-interglacial change in sediments to the north of the APF [*Kumar*, 1994; *Nurnberg et al.*, 1997]. Although the evidence from biogenic barium conflicts with the evidence from radionuclide ratios, an important caveat in the use of barium as a productivity proxy is its poor preservation in reducing sediments [*McManus et al.*, 1998].

In this paper we consider authigenic uranium as a proxy for POC rain to the seafloor. Under suboxic conditions, pore water uranium (U(VI)) is reduced to the insoluble species, U(IV), and precipitated. This lowers the pore water U concentration, and dissolved U from the overlying bottom waters then diffuses into the sediment across this concentration gradient and precipitation continues, leading to an enrichment of so-called authigenic U in the sediment [*Anderson*, 1982; *Barnes and Cochran*, 1990; *Klinkhammer and Palmer*, 1991]. For a given bottom water oxygen concentration, sedimentary authigenic U increases (non-linearly) with increasing organic carbon flux [*Kumar et al.*, 1995]. Thus, all else being equal, downcore profiles of authigenic U should reflect past changes in the redox state of oceanic sediments and hence the rain of organic carbon supplied by export production.

Previous workers found elevated concentrations of authigenic U in glacial-aged sediments of the Atlantic [*Anderson et al.*, 1998; *Kumar et al.*, 1995] and Indian [*Bareille et al.*, 1998; *Francois et al.*, 1993; *Rosenthal et al.*, 1997] sectors of the SO, suggesting a net increase in the rain of organic carbon during glacial times. Very large enrichments of authigenic U in the polar frontal region of the Atlantic sector, during the LGM, were interpreted as reflecting productivity that was equivalent to that found today in productive ocean margin regions [*Kumar et al.*, 1995]. As with the other paleoflux proxies, there are alternative interpretations of the authigenic U record. Interpreting the authigenic U record is difficult because the reducing conditions that favor its precipitation are a product of the supply rate of organic carbon from surface productivity, the supply of organic carbon from sediment focusing, and the oxygen concentration of the overlying bottom water. To the extent that there were glacial-interglacial changes in sediment focusing and deepwater ventilation, this complicates the interpretation of authigenic U as a paleoproductivity record. Here we present authigenic U records from 30 cores in the South Atlantic Ocean. By considering a large number of cores with broad geographic and depth coverage, we hope to clarify the factors controlling authigenic U precipitation in sediments of the SO and thereby further constrain glacial-interglacial changes in primary productivity and deepwater circulation.

2. Site Description and Methods

The cores presented in this paper cover a latitudinal range from 7°S to 55°S: from well north of the Subtropical Front (STF) to south of the Antarctic Polar Front (APF) (Figure 1). The region between the STF and the Subantarctic Front (SAF) is known as the Subantarctic Zone, and the region between the SAF and the APF is the Polar Frontal Zone (PFZ). In this paper we will combine the PFZ with the Subantarctic Zone, since our records show similar behavior across the two zones. Today, ventilation of the deep core sites is from the north, by North Atlantic Deep Water (NADW), and from the south, by Antarctic Bottom Water (AABW). Two of the cores, VM22-109 and TN057-20, were recovered from the Discovery Seamount. These sites lie within Antarctic Intermediate Water (AAIW). Many of the cores presented in this paper have been studied previously. Table 1 describes core location, depth, age control, and data sources. A number of the subtropical cores have chronologies constrained only by *CLIMAP Project Members* [1981] (hereinafter referred to as *CLIMAP* [1981]) estimates of the LGM. This is sufficient age control for our purposes, because authigenic U is generally absent throughout these cores. In all of the cores the LGM is associated with a peak in the detrital content of the sediment, which is reflected in a peak in the ^{232}Th concen-

Table 1. Depth, Location, Age Control, and Radionuclide Data Source for the Cores Examined in This Study[a]

Core	Depth, m	Lat., °S	Lon., °W	Age Control	^{232}Th and ^{238}U Data	Chronology
VM22-177	3290	7.75	14.62	C, D	this study	*CLIMAP* [1981]; *Mix* [1986]
VM22-38	3797	9.55	34.25	B, C, D	this study	*CLIMAP* [1981]; *Shackleton* [1977]
VM24-240	4327	31.73	28.2	C, D	this study	*CLIMAP* [1981]
VM24-221	4204	32.03	2.82	C, D	this study	*CLIMAP* [1981]
VM24-229	4202	34.45	10.6	C, D	this study	*CLIMAP* [1981]
VM27-196	1106	36.33	7.57	D	this study	this study
VM27-197	4089	36.68	6.27	D	this study	this study; C. Charles, unpublished data, 1994
RC13-243	4790	36.9	−1.33	B, C, D	this study	*Lozano and Hays* [1976]; *Shackleton* [1977]
RC12-294	3308	37.26	10.1	A, B, C, D	this study	*Shackleton* [1977]
RC12-267	4144	38.68	25.45	B, C, D	*Anderson et al.* [1998]	*Lozano and Hays* [1976]
VM14-65	4825	41	−8	D	this study	this study
RC11-83	4718	41.6	−9.72	A, B	this study	*Charles et al.* [1996]
VM34-157	3636	41.95	−26.41	B, D	*Anderson et al.* [1998]	*Verardo* [1995]
VM22-109	733	41.97	0.25	A	this study	this study
TN057-20	1312	42.1	−0.6	A, B	this study	this study; *Ninnemann and Charles* [1997]
RC15-94	3762	42.9	20.85	B	*Kumar et al.* [1995]	*Charles et al.* [1991]
VM22-108	4171	43.18	3.25	B	*Kumar et al.* [1995]	*Charles et al.* [1991]
PS2082	4610	43.21	−11.73	B, C, D	*Frank* [1996]	*Frank* [1996]
RC11-120	3193	43.5	−79.86	B, D	*Anderson et al.* [1998]	*Martinson et al.* [1987]
RC15-93	2714	46.1	13.21	A, B	*Anderson et al.* [1998]	*Charles et al* [1991]
PS1754-1	2471	46.77	−7.61	B, C, D	*Frank* [1996]	*Frank* [1996]
RC13-254	3636	48.56	−5.11	A, B	*Kumar et al.* [1995]	*Charles et al.* [1991]
PS1756	3787	48.73	−6.71	B, C, D	*Frank* [1996]	*Frank* [1996]
RC11-78	3115	50.86	9.86	C, D	this study	P. Froelich and R. Mortlock, unpublished data, 1994
RC13-271	3634	51.98	−4.51	A, B	*Kumar et al.* [1995]	*Charles et al.* [1991]
PS1768	3270	52.6	−4.48	A, B, C, D	*Frank* [1996]	*Frank* [1996]
RC11-77	4098	53.05	16.45	C, D	*Anderson et al.* [1998]	P. Froelich and R. Mortlock, unpublished data, 1994
RC13-259	2677	53.88	4.93	B	*Kumar et al* [1995]	*Charles et al.* [1991]
RC11-76	5229	54.38	22.13	C, D	*Anderson et al.* [1998]	P. Froelich and R. Mortlock, unpublished data, 1994
PS1772	4135	55.46	−1.166	B, C, D	*Frank* [1996]	*Frank* [1996]

[a] Age control symbols are as follows: A, ^{14}C; B, oxygen isotope stratigraphy; C, biostratigraphy; D, lithic composition.

tration. Thus, for cores VM27-196 and VM27-197, where no other age control was available, we identified the LGM as the depth of the peak in ^{232}Th concentration. Core VM14-65 was dated by correlating its $CaCO_3$ record with that of its well-dated pair, RC11-83. In time-slice comparisons, reported Holocene and LGM values were calculated as averages between 0–10 and 17–24 kyr, respectively.

Uranium (^{238}U) and thorium (^{232}Th) concentrations were determined after total dissolution (HNO_3, HF, and $HClO_4$ treatment) by isotope dilution inductively coupled plasma-mass spectrometry. Uranium 236 was used as an internal standard for both ^{238}U and ^{232}Th in all the cores except TN057-20, VM22-109, and RC11-83, where ^{230}Th was used as a yield monitor for ^{232}Th. Replicate digests agreed to within 1–2%, and an internal standard was stable over 9 months to within 3%. Authigenic uranium (U_A) is the difference between total and detrital uranium:

$$^{238}U_A(\text{ppm}) = {}^{238}U_T - (^{238}U/^{232}Th)_D(^{232}Th_T),$$

where the subscript T denotes total nuclide concentration (ppm) and $(^{238}U/^{232}Th)_D$(=0.197) is the average $^{238}U/^{232}Th$ ratio (by weight) in detrital material from the South Atlantic Ocean [*Anderson et al.*, 1994; R. F. Anderson, unpublished data, 1996]. Note that $(^{238}U/^{232}Th)_D$ in the Southern Ocean is low relative to that from other ocean basins with the highest observed $(^{238}U/^{232}Th)_D$ being ~0.23 (R. F. Anderson, unpublished data, 1996). Although the detrital U/Th ratio may have changed by up to ~10% in response to changing detrital sources, for example, during the LGM, the authigenic component is typically >60% of the total U, so this correction is not large. Temporal and spatial deviations from the average detrital ratio are the largest source of uncertainty in determining U_A. For concentrations greater than 1 ppm the overall uncertainty is generally less than 10%.

Organic carbon was measured in decarbonated samples using a Carlo Erba NA 1500 elemental analyzer, following a modification of the method of *Verardo et al.* [1990]. Dry bulk density was used in the calculation of the mass accumulation rate (MAR) and was estimated from $\%CaCO_3$ using the relationship derived by *Froelich* [1991] for South Atlantic sediments.

3. Results

Throughout most of the South Atlantic Ocean we find little to no authigenic U in Holocene sediments, although there is a small but systematic enrichment around the APF (Figure 2a). During the LGM, sediments south of 40°S, particularly north of the APF, contain high concentrations (up to 5.3 ppm) of authigenic U. We find no authigenic U in glacial sediments north of ~38°S (Figure 2b). Authigenic Cd, which like U, is precipitated in suboxic sediments, was determined by inductively coupled plasma-mass spectrometry (ICP-MS) in a limited number of cores, primarily from the subtropical zone (cores VM22-177, VM22-38, RC12-294, and VM14-65). Authigenic Cd concentrations were generally <0.3 ppm and displayed little downcore variability. The highest concentrations (~1 ppm) were found in the Holocene section of core VM22-177. In core VM34-157, from the Subantarctic, authigenic Cd decreased from 0.6 ppm during the LGM to 0.1 ppm during the Holocene.

The downcore record of authigenic U and organic carbon in core VM22-108 is typical of deep cores from the Subantarctic Zone.

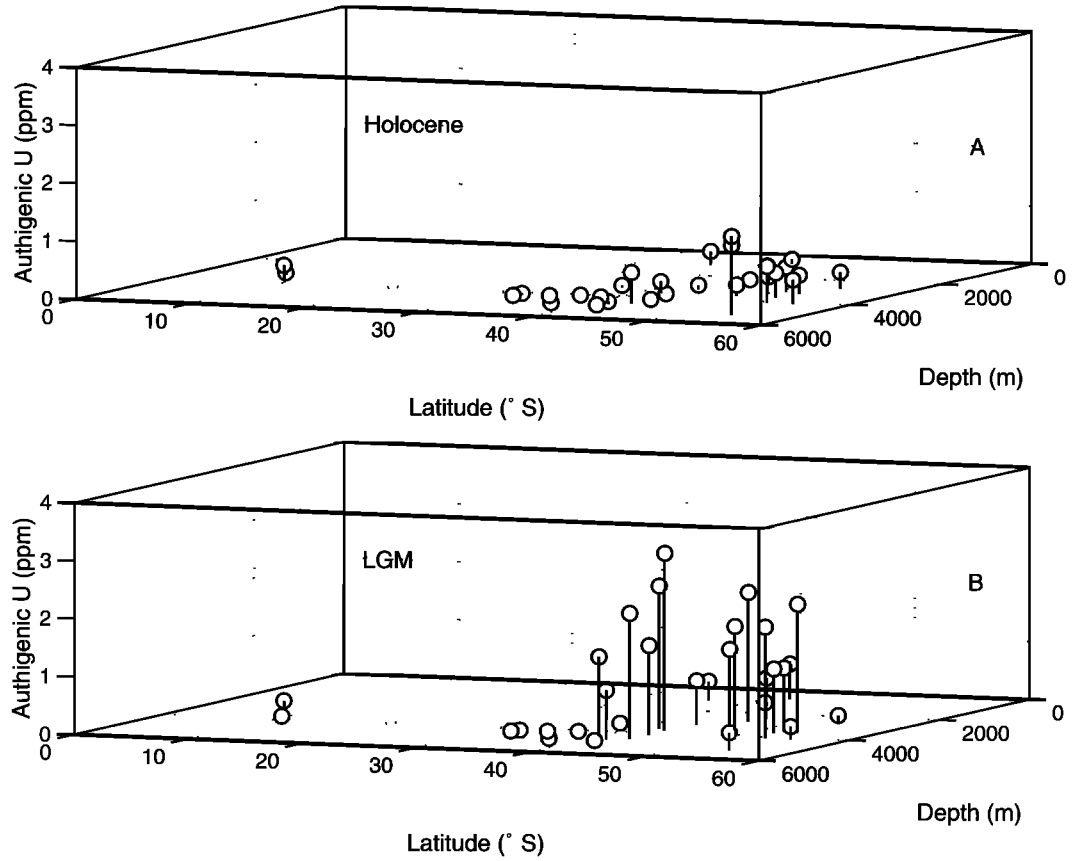

Figure 2. Latitudinal and depth dependence of the concentration of authigenic uranium in sediments from the South Atlantic Ocean during (a) the Holocene and (b) the Last Glacial Maximum (LGM).

Both tracers show substantial enrichment during glacial periods (Figures 3a and 3b), accompanied by an increased vertical flux of lithogenic material (Figure 3c).

3.1. Shallow Cores

Two cores recovered from the Discovery Seamount (cores TN057-20 and VM22-109) provide records of authigenic U and organic carbon that should be unaffected by changes in deepwater ventilation. Age models were constructed by linear interpolation between bulk carbonate ^{14}C ages ($n = 7$ in core VM22-109 and $n = 4$ in core TN057-20), converted to calendar age using *Bard et al.*'s [1990] calibration. Note that the age scale for core VM22-109 was simply extrapolated beyond the corrected ^{14}C ages and therefore involves greater uncertainty beyond the limits of ^{14}C dating. Core VM22-109 has a peak in organic carbon during the LGM and a slightly deeper (~20 cm) peak in authigenic U (Figure 4). Throughout the length of the core there are similar paired peaks, with maximum authigenic U lying below maximum organic C concentrations. This pattern is very similar to that observed in core VM22-108, a deep core located near core VM22-109 (Figure 3). However, the sampling interval in core VM22-108 cannot resolve potential offsets between organic carbon and authigenic U peaks. In core TN057-20, organic carbon and authigenic U covary in a similar manner, and both constituents show a general increase downcore (Figure 4). However, there is no clear peak in either proxy, and we believe that this core has been affected by post-depositional oxygenation. That is, a sudden change in the depth of oxygen penetration in this core may have led to organic carbon oxidation together with solubilization of previously deposited authigenic U. This may also explain the offset between organic carbon and authigenic U peaks observed in core VM22-109; oxygenation may have attenuated the organic carbon signal and caused mobilization of the authigenic U and reprecipitation further downcore [e.g., *Colley and Thomson*, 1985; *Crusius and Thomson*, 2000; *Thomson et al.*, 1990].

3.2. Lithogenic Inputs

The average thorium content of the upper continental crust is 10.7 ppm [*Taylor and McLennan*, 1985], and our work throughout the global ocean suggests that lithogenic sediments have a uniform Th content of ~10 ppm. Because ^{232}Th has a purely detrital source [*Brewer et al.*, 1980], it serves as a convenient proxy for lithogenic phases. During the LGM, we find elevated concentrations of ^{232}Th throughout the South Atlantic region (Figure 5). On average, LGM concentrations are 2 times greater than Holocene concentrations, with some sites recording a sixfold glacial-interglacial decrease in lithogenic content. This material must have been delivered either as ice-rafted debris, aeolian dust or eroded fine-grained sediment, because riverine inputs of detrital material are unlikely to contribute to these open-ocean sites.

4. Discussion

Interpreted strictly as a proxy of particulate organic carbon (POC) rain to the seafloor, the glacial enrichment of authigenic U in South Atlantic sediments suggests that between the APF and the STF, surface productivity was substantially greater during the LGM than it is today. The large enrichments suggest that the increased POC flux was more than can be explained by a northward migration of the productive polar frontal region, and more than can be explained by increased mineral ballasting of organic matter (R. Francois, personal communication, 2001). However, factors other than POC

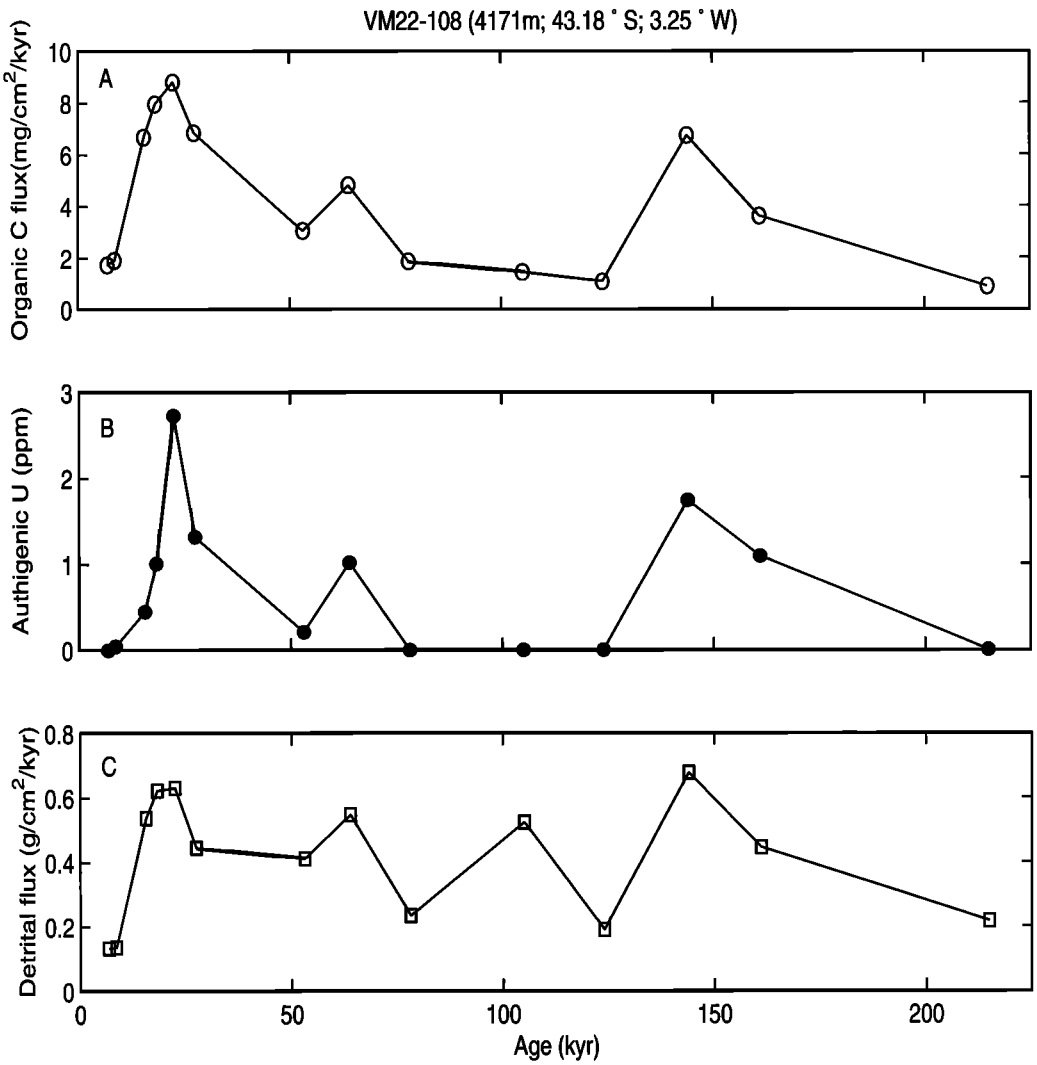

Figure 3. Downcore profiles in core VM22-108 of (a) organic carbon flux, (b) authigenic uranium concentration, and (c) detrital flux. Detrital content was derived from ^{232}Th concentration: detritus (%) = 10 × ^{232}Th (ppm). Organic carbon and detrital flux were corrected for sediment focusing using the ^{230}Th normalization method [*Francois et al.*, 1990]. The data are from *Anderson et al.* [1998] (Figure 3a), *Kumar et al.* [1995] (Figure 3b), and *Kumar* [1994] (Figure 3c).

flux can affect the sedimentary authigenic U record, and we must address these before identifying a paleoproductivity signal from the data. First, changes in sediment focusing can influence authigenic U burial. Here we use mass accumulation rates (MARs) to help identify those cores where variable sediment focusing may have influenced authigenic U burial. Second, the chemically reducing conditions required to form authigenic U can be induced by some combination of high POC rain and low bottom water oxygen (BWO) content. Authigenic U records from cores from the depth of Antarctic Intermediate Water argue for a significant contribution from increased POC flux.

4.1. Changes in Sediment Focusing

The redox conditions that control authigenic U precipitation are dictated by the total flux of organic carbon to the sediment, whether it is supplied vertically, through biological production, or horizontally via sediment focusing. By supplying POC laterally, changes in sediment focusing might therefore drive changes in authigenic U precipitation independent of any changes in export production. In order to identify cores that may have been affected by variable sediment focusing, we have, where possible, used downcore MARs to estimate changes in sediment focusing. We were able to estimate MARs for all but the subtropical cores. Of these 20 cores, half show either no glacial-interglacial change in MAR or greater MAR during the Holocene (Figure 6 and Table 2). The remaining 10 cores, for which glacial MARs are greater than Holocene MARs, are distributed evenly throughout the study area. Thus, while some of the cores with high glacial authigenic U also have higher glacial MARs, sediment focusing cannot explain all of the observations of high glacial authigenic U (Figure 6). Indeed, in all but four of the cores showing authigenic U enrichment during the LGM, glacial MARs were not more than 3 g cm^{-2} kyr^{-1} greater than Holocene MARs (Figure 6). For example, core VM22-108, with no glacial-interglacial change in MAR (Figure 6), has a pronounced enrichment of both authigenic U and organic carbon during the LGM (Figure 3). Cores RC15-94 and VM34-157 also show substantial LGM enrichment of authigenic U without significant change in MAR. We conclude that the observed glacial-interglacial pattern of authigenic U

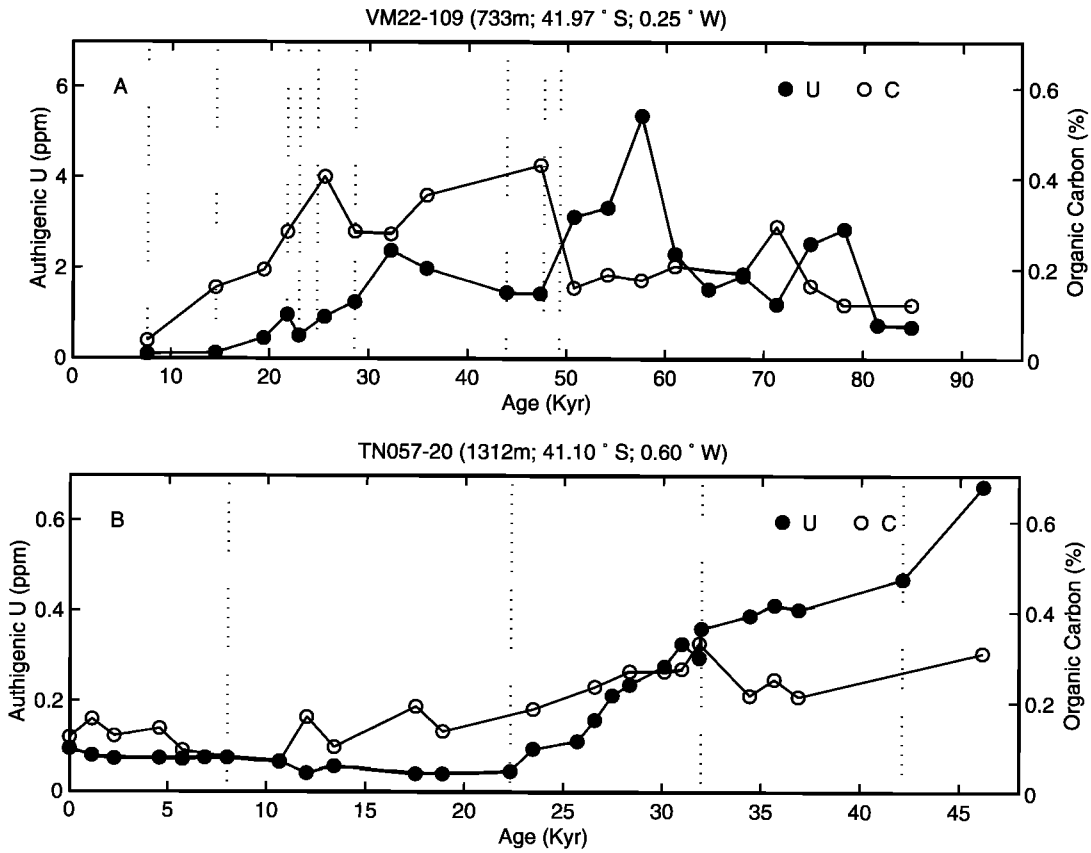

Figure 4. Downcore profiles of authigenic uranium (solid circles) and organic carbon (open circles) in shallow cores (a) VM22-109 and (b) TN057-20. Note the factor of 10 difference in the authigenic U scale. Dotted vertical lines indicate the depth of ^{14}C bulk carbonate analyses used to derive timescales.

enrichment cannot simply reflect changes in MAR or sediment focusing.

4.2. Changes in Bottom Water Oxygen

Both organic carbon supply and bottom water oxygen concentration determine sediment redox conditions. Thus a decrease in bottom water oxygen could potentially cause authigenic U enrichment independent of any increase in organic carbon supply. Indeed, a decrease in the oxygen concentration of bottom waters has been invoked to explain the presence of authigenic U in the glacial section of two cores from the Indian sector of the Southern Ocean [*Francois et al.*, 1997].

4.2.1. Altered deepwater circulation. Changes in bottom water oxygen (BWO) concentration could arise from altered deepwater circulation during the LGM. For example, a shift in the location of deepwater formation to near 40°S, well to the north

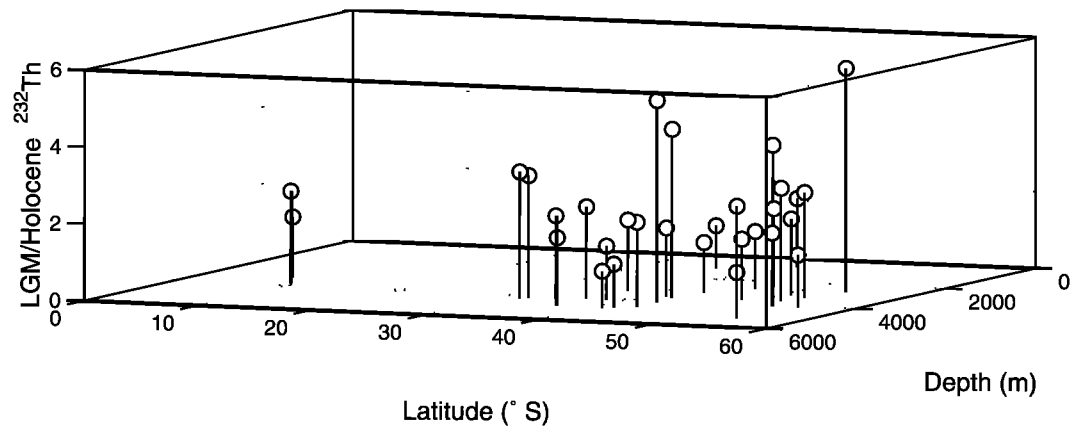

Figure 5. Latitudinal and depth dependence of the ratio of Last Glacial Maximum (LGM) to Holocene ^{232}Th concentrations in sediments from the South Atlantic Ocean. Thorium 232 has a purely detrital source [*Brewer et al.*, 1980] and is thus an indicator of lithogenic inputs.

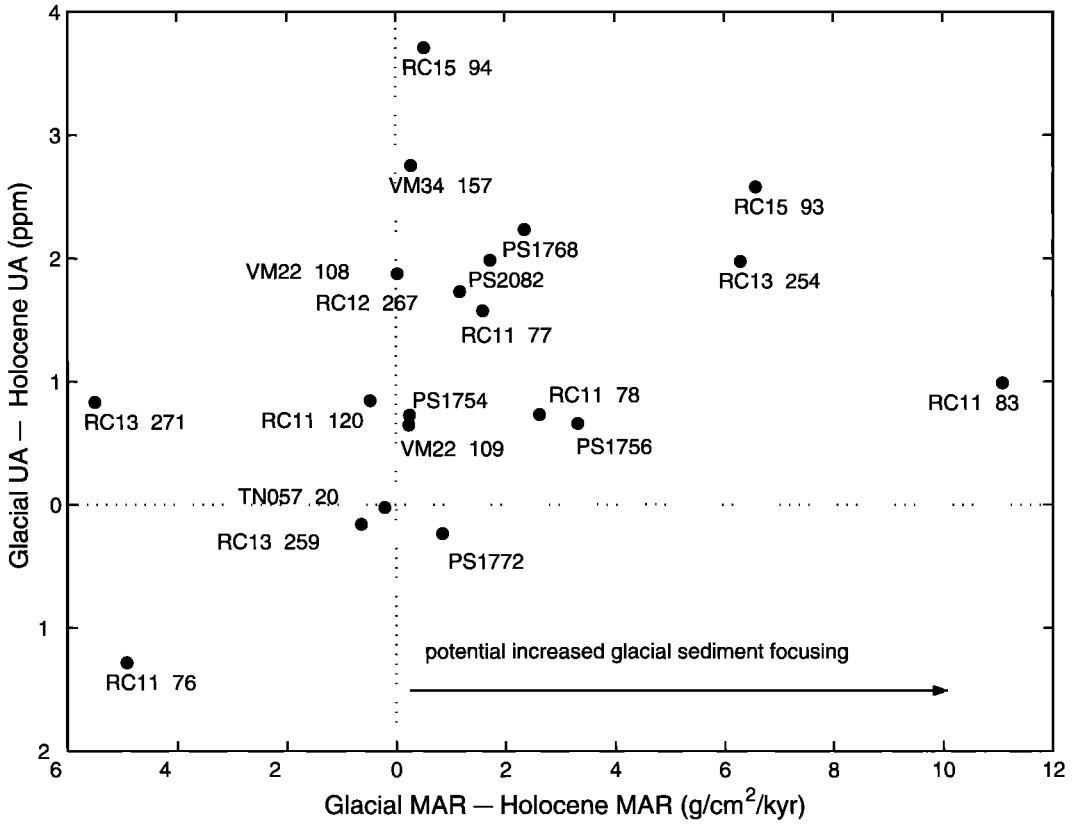

Figure 6. Glacial-interglacial changes in mass accumulation rate (MAR) and authigenic uranium (U_A) in cores from the South Atlantic Ocean.

of current sites [e.g., *Michel et al.*, 1995], would have produced lower oxygen concentrations in the nonventilated bottom waters to the south. Sediments could potentially have been reducing enough to precipitate authigenic U south of the STF while ventilation north of 40°S would have prevented authigenic U precipitation in the subtropical zone. Although this mode of deepwater formation is consistent with the latitudinal distribution of U during the LGM, it is not consistent with the relatively warm winter sea surface temperature (SST) reconstructions for the LGM near 40°S (~5°C [*CLIMAP*, 1981]). Even allowing for an error of as much as 2°–3°C in the CLIMAP winter SST values, winter surface temperatures in this region would have been too warm to permit convection to great depth, as proposed by *Michel et al.* [1995].

Recent work suggests that there was a shoaling or reduction in North Atlantic Deep Water (NADW) during the LGM [*Marchitto et al.*, 1998; *Oppo and Lehman*, 1995]. Assuming no change in Antarctic Bottom Water (AABW) production, if NADW flux was indeed much weaker during the LGM, then the impact on BWO would be greater north of the STF than to the south. This is because sites to the south would continue to be ventilated by AABW. It is conceivable that reduced NADW flux may have produced low oxygen concentrations beneath the productive APF and thereby lowered the threshold POC flux required to produce conditions reducing enough to precipitate U, with the result being enhanced deposition of U within the Subantarctic Zone. However, the evidence for reduced NADW flux during the LGM is still equivocal [e.g., *Matsumoto*, 2000; *Yu et al.*, 1996].

In the modern South Atlantic Ocean the intrusion of Circumpolar Deep Water (CDW) from the Pacific contributes to an oxygen minimum zone roughly coincident with the region of high glacial authigenic U (Figure 7) [*Mantyla and Reid*, 1983]. Although the CDW oxygen minimum zone is a subtle feature in the modern ocean, it is possible that extremely low oxygen in the glacial CDW may have led to the observed glacial authigenic U enrichment between 40°S and 50°S. That is, a latitudinally uniform POC rain rate, below the threshold for authigenic U precipitation under modern conditions, may have combined with near-anoxic CDW to produce significant authigenic U precipitation between 40°S and 50°S. Two processes determine the oxygen content of modern CDW entering the Atlantic through the Drake Passage. First, mixing occurs with low O_2 deep waters of the Pacific and Indian Oceans, and second, northern source waters lose O_2 during their circum-Antarctic transit before reentering the Atlantic [*Callahan*, 1972; *Reid*, 1989]. The transit time of CDW around the SO and hence the opportunity for O_2 consumption were probably not greater during the LGM, as modeling work suggests that the velocity of the Antarctic Circumpolar Current (ACC) actually increased in response to increased wind speeds during the glacial period [*Klinck and Smith*, 1993]. The second possibility, an exaggeration of the CDW oxygen minimum through mixing, would seem to require extremely low oxygen concentrations throughout the glacial deep Pacific.

An important constraint on the contribution of bottom water oxygen to authigenic U deposition is the observation that glacial-aged and modern sediments from the western Pacific sector of the Southern Ocean (at 170°W) lack authigenic U [*Chase*, 2001]. In today's ocean, primary production along 170°W is comparable to the productivity of the Atlantic sector [*Moore et al.*, 2000]. Unless the Pacific sector experienced dramatically lower levels of productivity during the LGM, relative to today, the absence of authigenic U in glacial sediments from the SW Pacific strongly suggests that BWO levels in that sector were not much lower during the LGM than today. We conclude, therefore, that during the LGM the deep Atlantic was not supplied by the Pacific with CDW

Table 2. Average Authigenic U (U_A) Concentration, ^{232}Th Concentration, and Mass Accumulation Rates for the Holocene and Last Glacial Maximum in the Cores Used in This Study[a]

Core	Holocene ^{232}Th, ppm	LGM ^{232}Th, ppm	Holocene U_A, ppm	LGM U_A, ppm	Holocene MAR, g cm^{-2} kyr^{-1}	LGM MAR, g cm^{-2} kyr^{-1}	Holocene *n*	LGM *n*
VM22-177	0.71	1.12	0.05	0.21			1	1
VM22-38	5.07	12.11	0.33	0.00			1	1
VM24-240	4.11	13.66	0.00	0.00			1	1
VM24-221	0.85	2.71	0.03	0.00			1	1
VM24-229	1.27	2.72	0.00	0.00			1	1
VM27-196	1.14	1.26	0.29	0.40			1	1
VM27-197	1.48	3.54	0.00	0.00			1	1
RC13-243	4.76	8.39	0.00	0.00			1	1
RC12-294	1.56	2.91	0.05	0.00			1	1
RC12-267	4.65	6.50	0.00	1.59	2.06	3.23	1	3
VM14-65	6.18	5.90	0.00	0.00			1	1
RC11-83	5.79	6.46	0.05	1.04	7.79	18.88	6	35
VM34-157	1.13	2.01	0.25	3.00	3.56	3.84	7	5
VM22-109	0.25	0.44	0.09	0.73	2.23	2.46	1	2
TN057-20	0.24	0.71	0.08	0.06	1.70	1.50	8	4
RC15-94	1.56	6.84	0.02	3.72	3.01	3.52	1	1
VM22-108	1.27	6.66	0.00	1.87	4.10	4.13	2	2
PS2082	3.31	7.26	0.66	2.64	3.31	5.04	3	8
RC11-120	2.11	2.75	0.09	0.93	2.46	1.98	7	7
RC15-93	2.44	2.84	0.41	3.15	3.04	9.62	2	2
PS1754-2	0.77	1.57	0.14	0.27	1.41	1.66	2	2
RC13-254	1.45	1.96	0.25	3.27	2.32	8.62	6	8
PS1756	2.88	2.14	1.10	1.76	1.04	4.37	2	15
RC11-78	1.23	2.44	0.51	1.24	2.77	5.40	2	5
RC13-271	0.82	2.42	0.52	1.35	20.00	14.50	2	3
PS1768	1.03	2.82	0.40	2.63	6.20	8.56	2	4
RC11-77	2.01	3.83	0.76	2.33	2.15	3.75	1	4
RC13-259	0.45	2.63	0.34	0.18	3.39	2.75	2	3
RC11-76	1.29	3.76	1.65	0.37	8.92	4.00	4	3
PS1772	3.86	5.29	0.51	0.27	0.64	1.49	2	3

[a]Note that some of these data have been published elsewhere (see Table 1). The *n* refers to the number of samples averaged for each time period. Previously unpublished data are available electronically from the World Data Center-A for Paleoclimatology, NOAA/NGDC, 325 Broadway, Boulder, Colorado (paleo@mail.ngdc.noaa.gov; URL: http://www.ngdc.noaa.gov/paleo). MAR, mass accumulation rates; LGM, Last Glacial Maximum.

bearing an oxygen content much lower than that which exists today. Consequently, the widespread enrichment of authigenic uranium in glacial Atlantic sediments must indicate that productivity in the Atlantic Subantarctic Zone was significantly greater than it is today.

4.2.2. Shallow depth authigenic U record. The most compelling evidence that the observed glacial increase in authigenic U was largely driven by increased POC rain throughout the Subantarctic Atlantic Ocean, and not simply reduced bottom water oxygen, comes from core VM22-109 on the Discovery Seamount. Because this is a shallow site, one might expect greater POC rain at all times, relative to deeper sites [*Suess*, 1980] as well as a greater preservation of particulate, nonlithogenic uranium associated with sinking particles [*Zheng et al.*, 2001]. Yet the authigenic U record shows a real change through time, from Holocene sediments which lack authigenic U to a clear enrichment (1–2 ppm) into the LGM. It is difficult to invoke changes in bottom water oxygen concentration, as this site lies within

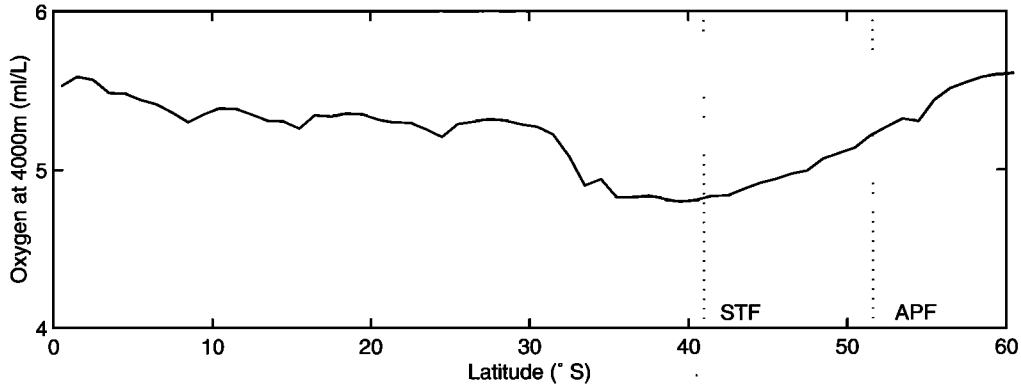

Figure 7. Zonally averaged oxygen concentration at 4000 m in the South Atlantic Ocean. Data are from *Levitus and Boyer* [1994].

Antarctic Intermediate Water and was therefore probably better oxygenated during the LGM than today [*Lynch-Stieglitz et al.*, 1994]. Indeed, measurements of $\delta^{13}C$ in benthic foraminifera from core TN057-20, an adjacent core from the Discovery Seamount, show a glacial-interglacial change of only −0.2‰ [*Ninnemann and Charles*, 1997]. After applying a whole-ocean change of −0.32‰ [*Duplessy et al.*, 1988], this suggests lower nutrient concentration and thus higher oxygen concentration at this intermediate water site during the LGM [*Ninnemann and Charles*, 1997]. The large enrichments of authigenic U observed in the glacial section of core VM22-109 were therefore most likely caused by a high rain rate of organic carbon.

Naturally, if the POC rain in the Subantarctic Atlantic Ocean was indeed much greater than today during the LGM, then the oxygen concentration of bottom waters may very well have been lower than that in the modern ocean [*Francois et al.*, 1997], as respiration couples oxygen consumption to POC rain. However, the absence of authigenic U in glacial Pacific sediments together with the enrichment of authigenic U in glacial sediments from the Discovery Seamount indicate that reduced deepwater oxygen concentrations are unlikely to have acted alone in producing widespread deposition of authigenic U in glacial sediments of the Atlantic sector of the Southern Ocean.

4.3. Postdepositional Oxygenation

Another important issue is the extent to which the authigenic U records have been affected by postdepositional oxygenation. The relatively low sedimentation rates in the Southern Ocean make this region particularly susceptible to such "burn-down" [*King et al.*, 2000; *Rosenthal et al.*, 1995]. Indeed, our shallow cores do show some evidence for burn-down (see section 3.1). Note that burn-down could either eliminate a U signal or accentuate a peak and cause it to be displaced downward. Any postdepositional oxygenation would therefore tend to produce, on average, lower concentrations of authigenic U in glacial sediments. This means that our conclusions regarding increased POC fluxes during the LGM are, if anything, conservative, since authigenic U concentrations during the LGM may have been underestimated. It is possible, for example, that U was precipitated north of the STF during the LGM but has since been oxidized away. While the systematic absence of U throughout the length of these records (a total of 11 cores north of the STF) argues against this possibility, unequivocal demonstration of the absence of authigenic U deposition during the LGM in this region awaits the study of a core with a sediment accumulation rate sufficiently high to assure that any authigenic U formed during the LGM would have been buried below the depth of plausible burn-down.

5. Conclusions

We have found glacial enrichment of authigenic uranium in sediments from the Atlantic sector of the Southern Ocean between the Antarctic Polar Front and the Subtropical Front, which we interpret as reflecting primarily increased particulate organic carbon (POC) rain in this region during the LGM. This interpretation is based on three observations: (1) Lateral supply of POC via sediment focusing has been ruled out as a major contributor to glacial authigenic U by identifying cores in the region with little change in mass accumulation rate between the LGM and the Holocene that still show substantial glacial authigenic U enrichment; (2) the absence of authigenic U in glacial sediments from the western Pacific sector of the SO, where modern productivity is comparable to that found in the Atlantic sector, suggests that the authigenic U signal in the Atlantic cannot be due entirely to a reduction in bottom water oxygen concentration, because this mechanism would have led to emplacement of authigenic U in the Pacific sediments as well; and (3) a higher glacial POC flux is required to account for the glacial authigenic U in a shallow core, VM22-109, since a benthic $\delta^{13}C$ record from a nearby core suggests that sediments at this site experienced greater oxygen concentrations during the LGM than during the Holocene, when authigenic U is absent.

Without further constraints on bottom water oxygen concentrations during the LGM, we cannot say whether the large glacial enrichment of authigenic U in the Subantarctic reflects a POC flux that was substantially greater than today (e.g., similar to ocean margins [*Kumar et al.*, 1995]), or rather, a flux that was moderately greater than today but with deposition of authigenic U enhanced by the presence of bottom water oxygen concentrations somewhat lower than those occurring today [e.g., *Francois et al.*, 1997]. Our data suggest that there must have been some increase in POC to sediments of the Atlantic sector of the Southern Ocean during the LGM, and our data are consistent with a very substantial increase in productivity just north of the modern position of the APF. The higher POC fluxes inferred from our records of authigenic U during the LGM are thus consistent with the suggestion [*Martin*, 1990] that enhanced dust flux to the Southern Ocean during glacial periods delivered iron to nutrient-rich surface waters and stimulated phytoplankton growth. These sites lie directly downwind from Patagonia, the principal source responsible for the large increases in dust reaching Antarctic ice cores during glacial times [*Basile et al.*, 1997]. This region is thus ideally positioned to record a glacial release from iron limitation driven by increased dust fluxes during the LGM. Quantifying the net effect on atmospheric CO_2 concentrations will require better data coverage from all three sectors of the Southern Ocean and better constraints on the supply of CO_2 to surface waters via upwelling.

Acknowledgments. We thank Anitra Ingalls and Yong Lao for laboratory assistance. John Crusius and two anonymous referees provided helpful comments on an earlier draft of this paper. This work was supported by National Science Foundation grants OCE93-14634 and OPP95-30379 and a fellowship (awarded to Z. C.) from the Fonds pour la Formation de Chercheurs et l'Aide á la Recherche (Québec). This is LDEO contribution 6183.

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R. F. Anderson and M. Q. Fleisher, Lamont-Doherty Earth Observatory, Columbia University, Palisades, NY 10964-8000, USA.

Z. Chase, Monterey Bay Aquarium Research Institute, 7700 Sandholdt Road, Moss Landing, CA 95039, USA. (zanna@mbari.org)

(Received May 8, 2000;
revised April 20, 2001;
accepted May 25, 2001.)