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Radiocarbon constraint on relict organic carbon contributions to Ross Sea sediments

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[1] We estimate the relative contribution of relict organic matter to the acid-insoluble organic carbon (AIOC) fraction of surface sediments from Ross Sea, Antarctica, on the basis of ^{14}C abundance. The bulk isotopic characteristics of AIOC can largely be explained by simple two-source models of modern and relict organic carbon, when samples are grouped according to two geographical regions, namely, southwestern and south central Ross Sea. This spatial variability in relict organic carbon could be controlled by proximity to the edge of the Ross Ice Shelf and ice drainage areas. Radiocarbon abundance in the AIOC is potentially an excellent tool to estimate the contribution of relict organic carbon in the Antarctic margin sediments.

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1. Introduction

[2] In Antarctic margin sediments, radiocarbon ages of acid-insoluble organic carbon (AIOC) have been used for determining chronologies, because carbonate shells such as those from planktonic foraminifera are generally absent. However, the radiocarbon ages of the AIOC in surface sediments have frequently been found to be substantially older than the ages of dissolved inorganic carbon (DIC) in overlying surface waters. "Contamination" of relict organic matter eroded from the Antarctic Continent has thus been suspected [e.g., *Harris et al.*, 1996; *Andrews et al.*, 1999; *Domack et al.*, 1999; *Licht et al.*, 1996]. Recently, *Ohkouchi et al.* [2003] used compound-specific radiocarbon analysis [*Eglinton et al.*, 1996] to determine ages of solvent-extractable, short-chain (C_{14}, C_{16}, and C_{18}) fatty acids in Ross Sea sediments. These dates were not only substantially younger than those of AIOC, but also agreed well with post-bomb (850 yr) or pre-bomb (1300 yr) DIC reservoir ages in Ross Sea surface waters [*Berkman and Forman*, 1996; *Broecker et al.*, 1985; *Gordon and Harkness*, 1992; *Rafter*, 1968]. Ohkouchi and coworkers concluded that the difference of radiocarbon ages between sedimentary AIOC and pre-bomb DIC reflects the contribution of the relict organic matter eroded from Antarctica.

[3] In this study, we infer the source, relative abundance, and distribution of relict organic carbon in the surface sediments of southern Ross Sea mainly on the basis of ^{14}C content in sedimentary organic matter.

2. Samples and Methods

2.1. Ross Sea

[4] The Ross Sea, located in the Pacific sector of the Southern Ocean, is mostly covered by sea ice for 8 to 12 months in a year. Although the biological production in the Ross Sea is confined mostly to a short period from late austral spring to summer, the annual mean primary production was estimated to be as large as 90 to 220 gC m^{-2} yr^{-1} [*Nelson et al.*, 1996], which is among the most productive areas in the Southern Ocean. Phytoplankton blooms in the southern Ross Sea are strongly controlled by the formation of coastal polynyas (open water area surrounded by sea ice) during the austral spring to summer [e.g., *Saggimo et al.*, 1998]. Diatoms often form extensive blooms in the western Ross Sea where surface stratification is strong and mixed layer depth is shallow, whereas the colonial haptophyte *Phaeocystis antarctica* predominates in the south central Ross Sea where surface waters are weakly stratified and relatively deeply mixed [e.g., *Arrigo et al.*, 1999; *Goffart et al.*, 2000]. The cause for such a spatial variation of primary producers is still unknown. Sediment trap experiments have suggested that the sedimentation of organic matter in the deeper water column is strongly coupled with primary production in the surface water [*Dunbar et al.*, 1998; *Handa et al.*, 1992].

2.2. Samples

[5] Sediment samples were recovered using a box corer at four sites from Ross Sea, Antarctica during the ROAVERRS (Research on Ocean-Atmosphere Variability and Ecosystem Response in the Ross Sea) cruise on R/V *Nathaniel B. Palmer* in December 1998 (Table 1). The sediment cores were sub-sampled on board into 2 cm intervals and were stored frozen (−20°C) until analysis.

2.3. Methods

[6] After air drying, the sediments were acidified to remove carbonate, and combusted in quartz tube at 850°C for 5 hr according to standard procedures of the National Ocean Science Accelerator Mass Spectrometry (NOSAMS) Facility at Woods Hole [*McNichol et al.*, 1994]. Evolved CO_2 was cryogenically purified and majority was supplied for ^{14}C determination by AMS. In this study we mostly use the $\Delta^{14}C$ notation rather than radiocarbon age, since it has linear systematics that is amenable for calculating end-member contributions. The definition of $\Delta^{14}C$ notation is

$$\Delta^{14}C = (f_m \exp(1950 - x)\lambda - 1) \times 1000,$$

where λ is 1/8267 (yr^{-1}), f_m is the fraction of modern ^{14}C corrected for isotopic fractionation by use of $\delta^{13}C$ and x is the year of analysis [*Stuiver and Pollach*, 1977].

[7] A small aliquot of the CO_2 gas evolved by the combustion of AIOC was also used for ^{13}C analysis. Stable carbon isotopic compositions are reported as conventional δ notation against the PDB standard.

[8] Procedures for radiocarbon measurement in individual fatty acids were described by *Ohkouchi et al.* [2003]. A small aliquot of CO_2 produced by the combustion of purified fatty acids was used for the measurement of stable carbon isotopic composition.

[9] Analytical methods for the other 16 samples used in this study (Table 1) were described by *Andrews et al.* [1999] and *Domack et al.* [1999].

3. Results and Discussion

3.1. Isotope Mass Balance Considerations

[10] Assuming that the overestimation of surface sediment age based on AIOC is purely dependent on the magnitude of the contamination by relict organic carbon, corresponding $\Delta^{14}C$ values ($\Delta^{14}C_{AIOC}$) can mathematically be expressed as follows:

$$\Delta^{14}C_{AIOC} = f\Delta^{14}C_{relict} + (1 - f)\Delta^{14}C_{modern} \quad (0 \le f \le 1), \tag{1}$$

where $\Delta^{14}C_{modern}$ and $\Delta^{14}C_{relict}$ denote $\Delta^{14}C$ values of organic matter produced in the modern Ross Sea and those derived from relict organic matter, respectively, and f denotes a fraction of relict carbon in AIOC. On the basis of the ^{14}C measurements of DIC and organisms such as mollusca collected in known years, DIC reservoir ages of Ross Sea were estimated to be around −150‰ for the pre-bomb period (before 1957) [*Berkman and Forman*, 1996; *Gordon and Harkness*, 1992]. If assuming that the relict

Table 1. The $\delta^{13}C$ and $\Delta^{14}C$ Values of Acid-Insoluble Organic Matter in the Ross Sea Surface Sediments

Number	Sample	Depth, cm	Latitude	Longitude	Water, Depth, m	$\delta^{13}C$, ‰	$\Delta^{14}C$, ‰[a]	References[b]
				Southwestern Ross Sea (160°–170°E)				
1	95P26	0–2	76°59′S	162°53′E	800	−26.0	−241	1, 2
2	95K30	0–2	76°00s	164°35′E	752	−25.2	−418	1
3	95K34	0–1	75°10s	164°39′W	1257	−24.7	−329	1
4	Chinstrap	0–2	76°20s	165°02′E	823	−24.0	−295	this study
5	95K31	0–2	75°42′S	165°25′E	879	−23.7	−261	1, 2
6	96B15	0–1	75°02′S	166°16′E	939	−23.9	−295	1
7	95K37	0–2	74°30s	167°45′W	924	−24.4	−292	1, 2
				South Central Ross Sea (170°E–170°W)				
8	Emperor	0–2	76°59′S	172°00′E	670	−26.4	−555	this study
9	Gentoo	0–2	76°20s	172°56′E	623	−27.4	−398	this study
10	95P7	2–4	75°38′S	178°35′E	449	−25.1	−943	1
11	94P33	1–3	75°27′S	179°37′E	603	−29.4	−419	1
12	95P11	0–2	76°27′S	179°52′E	659	−27.1	−403	1, 2
13	TC16	2–4	76°56′S	179°49′W	712	−25.0	−395	1, 2
14	95P17	2–4	77°27′S	179°03′W	732	−24.1	−934	1
15	96B91	0–1	75°30s	178°20′W	451	−28.8	−360	1
16	Fairy	0–2	77°58′S	178°03′W	671	−25.7	−712	this study
17	95P12	0–2	76°47′S	177°49′W	568	−26.8	−431	1, 2
18	95T40	0–2	74°28′S	173°30′W	556	−27.9	−405	1, 2
19	95K39	0–1	74°28′S	173°28′W	557	−28.3	−324	1, 2
20	96B38	0–1	78°09′S	171°02′W	562	−26.7	−360	1

[a] $\Delta^{14}C$ values are calculated from the reported ^{14}C ages (t) based on the following equation: $\Delta^{14}C = 1000\ \{\exp(-t/8033) - 1\}$.
[b] References: 1, *Andrews et al.* [1999]; 2, *Domack et al.* [1999].

carbon is "radiocarbon dead", equation (1) can thus be simplified as

$$\Delta^{14}C_{AIOC} = -850f - 150. \qquad (2)$$

This equation implies that only 10% "contamination" of ^{14}C-dead relict organic carbon results in an overestimate the sediment age about 850 ^{14}C years, sufficiently large to influence on the climate interpretations from the sedimentary record [e.g., *Anderson et al.*, 2002; *Denton et al.*, 1989; *Ingólfsson et al.*, 1998]. Bioturbation of the surface sediments can mix organic carbon of different ages, which should be the source of errors in this formulation. However, radiocarbon ages of sedimentary fatty acids in our four sites suggested that the bioturbation would not be large enough to significantly affect above consideration [*Ohkouchi et al.*, 2003]. It is clear therefore that the degree of contribution of relict carbon in a sample should be carefully evaluated.

3.2. Two-Source Models for Acid-Insoluble Organic Carbon in the Ross Sea Surface Sediments

[11] *Andrews et al.* [1999] and *Domack et al.* [1999] reported both radiocarbon dates and stable isotopic compositions of AIOC for 16 core top or near core top sediments from the Ross Sea. Adding our results from 4 sites relatively close to the edge of the Ross Ice Shelf (Table 1) [*Ohkouchi et al.*, 2003], $\Delta^{14}C$ values of AIOC in surface sediments are plotted as a function of $\delta^{13}C$ values in Figure 1. Both $\delta^{13}C$ and $\Delta^{14}C$ values vary widely ranging from −23.7 to −29.4‰, and from −943 to −241‰, respectively. Here, we interpret the distribution of AIOC in the $\delta^{13}C - \Delta^{14}C$ diagram as a function of simple two-source models for samples from two distinct geographical regions: southwestern (160°E–170°E) and south central (170°E–170°W) Ross Sea.

[12] In the southwestern Ross Sea along the coast of Victoria Land, major primary producers in the modern environment are diatoms such as *Nitzschia* sp. or *Fragilariopsis curta* [*El-Sayed et al.*, 1983; *Wilson et al.*, 1986]. Although the $\delta^{13}C$ values of these types of phytoplankton have not been reported, *Villinski et al.* [2000] measured the $\delta^{13}C$ values of −22 ± 1‰ for suspended particulate matter (SPM) in the water column of this region during February to March. In contrast, in the south central Ross Sea, a major primary producer is prymnesiophyte *Phaeocystis antarctica* which forms extensive bloom in December to January [*DiTullio et al.*, 2000; *Dunbar et al.*, 1998]. The $\delta^{13}C$ values of SPM in this region are significantly

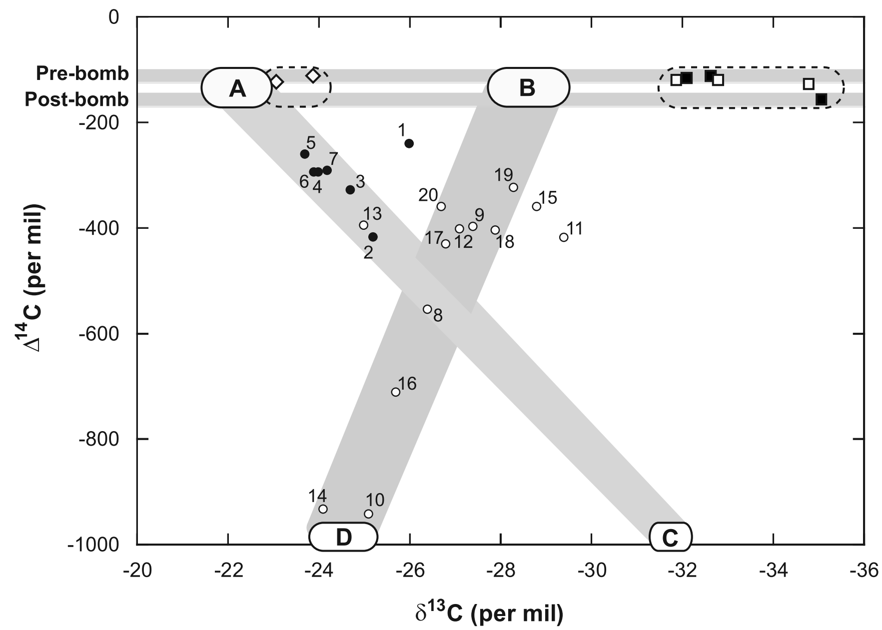

Figure 1. A cross plot of $\delta^{13}C$ versus $\Delta^{14}C$ for the acid-insoluble organic fraction in the surface sediments from Ross Sea. Samples collected from southwestern (160°–170°E) and south central (170°E–170°W) Ross Sea are represented by closed and open circles, respectively. Numbers indicate the sample number shown in Table 1. A sample from Granite Harbor is "1". "A" and "B" indicate domains of marine end-members for southwestern and south central Ross Sea, respectively, estimated from $\delta^{13}C$ values of SPM [*Villinski et al.*, 2000]. "C" and "D" indicate domains of hypothetical terrestrial end-members for southwestern and south central Ross Sea, respectively. Areas surrounded by broken lines are fields which encompass $\delta^{13}C$ and $\Delta^{14}C$ values of short-chain (C_{14}, C_{16}, and C_{18}) fatty acids in the surface sediments from Chinstrap (open star), Gentoo (open square), and Emperor sites (closed square). $\Delta^{14}C$ values of pre- and post-bomb DIC in the Ross Sea [*Berkman and Forman*, 1996] are also shown as the shaded area.

more depleted in ^{13}C than that of the southwestern Ross Sea, mostly ranging from −28 to −29‰ [*Villinski et al.*, 2000]. Such a large isotopic contrast between the phytoplankton in these two regions may be ascribed to the fact that *Nitzschia* sp., one of the major diatoms in the southwestern Ross Sea, potentially utilize C4 pathway when fixing CO_2 [*Wong and Sackett*, 1978], which has a small isotopic fractionation relative to the C3 pathway. We determined the $\delta^{13}C$ values, as well as $\Delta^{14}C$ values, for sedimentary, short-chain (C_{14}, C_{16}, and C_{18}) fatty acids [*Ohkouchi et al.*, 2003]. Since these compounds exhibit pre- or post-bomb ^{14}C ages, they must predominantly directly or indirectly reflect carbon inputs from primary producers in the overlying water column. As shown in Figure 1, the $\delta^{13}C$ values of these sedimentary fatty acids are around −23‰ in the southwestern Ross Sea and −32 to −35‰ in the south central Ross Sea. Since the fatty acids tend to be depleted in ^{13}C by ~5‰ relative to bulk tissue due to the isotopic fractionation at the time of decarboxylation of pyruvate [e.g., *DeNiro and Epstein*, 1977; *Monson and Hayes*, 1982; *Hayes*, 1993], these values are consistent with measurements of SPM from the corresponding areas [*Villinski et al.*, 2000] (Figure 1).

[13] Ice flow trajectories over the Ross Ice Shelf suggest that sea ice in the south central Ross Sea mostly originates from West Antarctic Ice Sheet (WAIS), whereas in the southwestern Ross Sea, it mainly derives from the Eastern Antarctic Ice Sheet (EAIS) through many outlet glaciers flowing over the Transantarctic Mountains [e.g., *Shabtaie and Bentley*, 1987; *Casassa et al.*, 1991]. The quality such as $\delta^{13}C$ value and quantity of reworked organic matter transported by the ice is therefore potentially different in these two regions. *Truswell and Drewry* [1984] conducted palynomorph analyses for surface sediments and found abundant (~500 grains g^{-1}) Paleozoic spores and pollen grains in the western half of the Ross Sea. Although neither $\delta^{13}C$ nor $\Delta^{14}C$ values have been measured for the organic matter in these relict fragments, we can reasonably set the $\Delta^{14}C$ value of this end-member as −1000‰ (radiocarbon dead) in both regions. As shown in Figure 1, on the basis of our mixing models, the mean $\delta^{13}C$

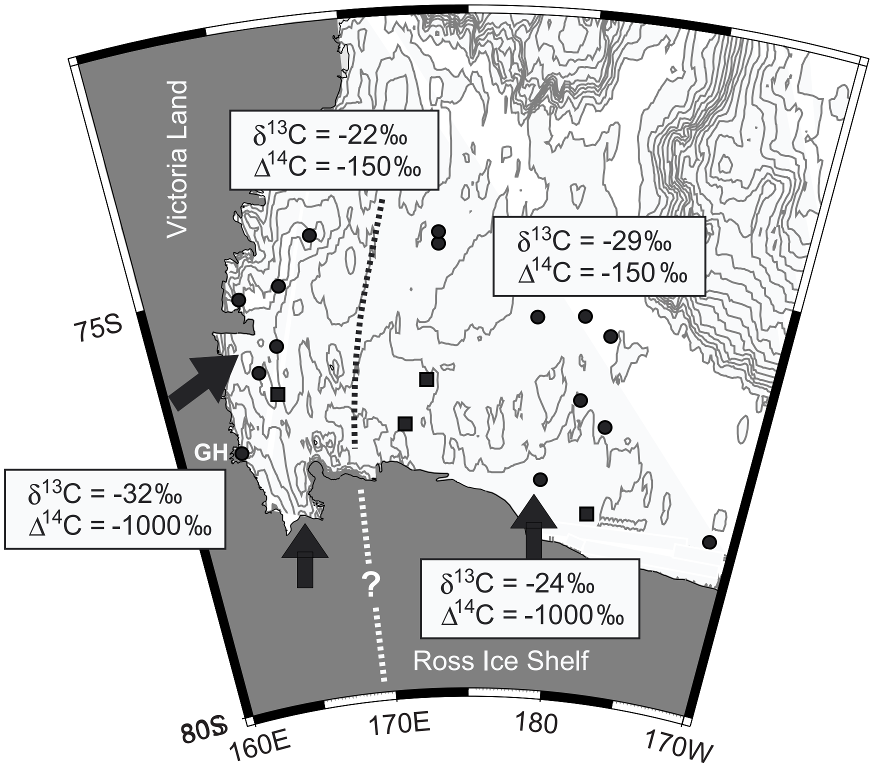

Figure 2. A proposed scheme for mixtures of relict and modern organic carbon in the Ross Sea sediments. Boundaries between two regions are indicated by broken lines. Closed squares and circles indicate our samples and samples referred from literatures, respectively. "GH" is a sample from Granite Harbor.

values of organic matter for the continental end-members are estimated to be −24 to −25‰ for the WAIS and around −32‰ for the EAIS. The isotopic composition of the continental end-member in the WAIS is consistent with those of organic carbon in sediments underlying the Ross Ice Shelf [*Sackett*, 1986]. The relict organic matter transported by WAIS may have originally been formed in marine environment, which resulted in relatively ^{13}C-enriched isotopic values of this end-member. On the basis of these considerations, we ascribe the old core top ages of Ross Sea sediments mainly to the contribution of relict organic matter, in support of the conclusions of *Andrews et al.* [1999]. The basic pattern of ^{13}C and ^{14}C distributions can primarily be explained by the scheme illustrated in Figure 2.

[14] One of the apparent exceptions to this model is the sediments from Granite Harbor in the southwestern Ross Sea, which appear to be of intermediate composition (Figure 1). Allochthonous organic matter deposited in this embayment may be influenced by the weathered materials in the Dry Valley region where modern organic matter is produced by algae and bacteria [e.g., *Maurice et al.*, 2002]. Some samples from south central Ross Sea also plot to the both sides of the mixing line (Figure 1), which might be the result of a broader distribution of $\delta^{13}C$ values for the marine end-member. Although we applied the $\delta^{13}C$ of POM [*Villinski et al.*, 2000] as the end-member, marine algal organic carbon may have a broader distribution of isotopic composition since the $\delta^{13}C$ values of fatty acids vary by more than 3‰.

[15] By applying equation (2), we estimate the relict fraction in the AIOC in the Ross Sea surface sediments (excluding the sample from Granite Harbor) (Figure 3). In the southwestern Ross Sea, relict organic carbon generally accounts for less than 20% of the bulk organic carbon, whereas in the south central Ross Sea the fraction of abundance of relict organic carbon varies widely, from 20 to 93%, but on average is higher than that of the southwestern Ross Sea. Although these values are basically similar with those based on $\delta^{13}C$ values, some samples like site 10 and 14 are significantly different. Such inconsistencies may be ascribed to the vagueness of the $\delta^{13}C$ end-members. Alternatively, the distribution pattern of reworked organic matter in the Ross Sea may be more complicated and may not well correlate with that of the phyto-

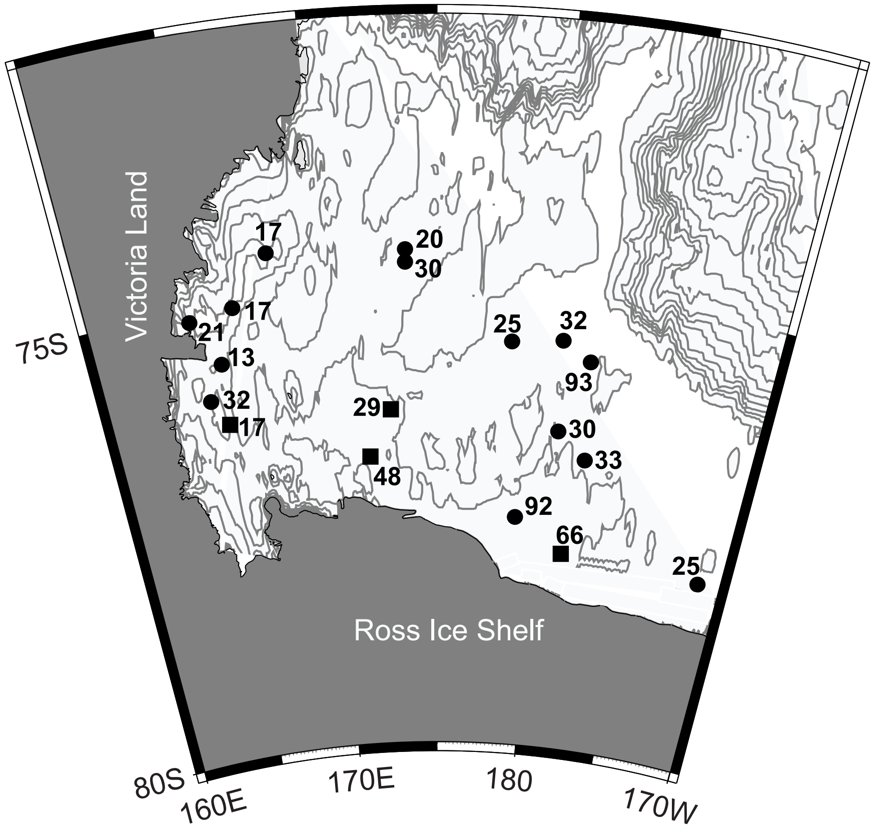

Figure 3. Relative contributions (%) of relict organic carbon estimated from ^{14}C abundance in the acid-insoluble organic fraction from surface sediments. Closed squares and circles indicate our samples and samples referred from literatures, respectively.

plankton community. Our estimates based on ^{14}C abundance are consistent with the observation by *Andrews et al.* [1999] that in the southwestern Ross Sea the reworked diatom percentage as a proxy for the contribution of relict organic matter is substantially smaller than that of the south central Ross Sea. In the south central Ross Sea, the fraction of relict organic matter is relatively high along the margin of Ross Ice Shelf. However, this is not always the case, as one site shows as high as 93% relict organic carbon near the edge of the continental shelf (Figure 3). Presently, it is difficult to estimate the mass accumulation rate of relict organic carbon in these surface sediments, mainly due to the lack of precise chronology of the sediments. *Anderson et al.* [1980] pointed out that patterns of sediment distribution on the Ross Sea continental shelf clearly reflect the different ice drainage areas. Furthermore, *Truswell and Drewry* [1984] showed that the spatial distribution of palynomorphs in the Ross Sea surface sediments is strongly controlled by ice drainage areas. Although the number of samples remains small, preliminary data indicates that the distribution of relict organic carbon in the southern Ross Sea is apparently controlled by proximity to the edge of the Ross Ice Shelf and ice drainage areas. We suggest that the radiocarbon is a valuable tool for tracing the nature of deposition of relict organic carbon in the Antarctic margin sediments.

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