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Transfer of lipids through marine water columns to sediments – insights from stable and radiocarbon isotopes

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Abstract

Compound-specific ^{13}C and ^{14}C compositions of diverse lipid biomarkers (fatty acids, alkenones, hydrocarbons, sterols, and fatty alcohols) were measured in sinking particulate matter collected in sediment traps and from underlying surface sediments in the Black Sea, the Arabian Sea and the Ross Sea. The goal was to develop a multi-parameter approach to constrain relative inputs of organic carbon (OC) from marine biomass, terrigenous vascular plant, and relict sources. Marine biomass in sediment trap material from the Black Sea and Arabian Sea accounted for 66–100 % of OC, with lower terrigenous (3–8 %) and relict (4–16 %) contributions. Marine biomarkers in sediments constituted lower proportions of OC (66–90 %), with consequentially higher proportions of terrigenous and relict carbon (3–17 % and 7–13 %, respectively). Ross Sea data were insufficient to allow similar mass balance calculations. It is apparent that whereas particulate organic carbon is overwhelmingly marine in origin, there are also significant proportions of pre-aged terrigenous and relict OC that become proportionally more important in sediments. These results indicate that pre-aged OC is better preserved during vertical transport to and burial at the seafloor and/or it reaches the sediment by lateral advection rather than only by the vertical sinking/biological pump that affects the upper ocean-derived marine POC.

1 Introduction

The dynamics of sources, sinks and processes that control burial of organic carbon (OC) in marine sediments have important implications for the global carbon cycle, paleoceanographic reconstructions and understanding climate variability (Berner, 1982; Hedges and Keil, 1995; Burdige, 2007; Zonneveld et al., 2010). Assigning the provenance of sedimentary OC remains a difficult task. Most burial occurs on continental margins where terrigenous material constitutes a significant proportion of the burial flux (Hedges et al., 1997; Burdige, 2005). But even at open ocean locations remote from the continents where marine OC dominates the water column flux via the biological pump, a terrigenous component delivered by long-range aeolian transport (Zafiriou et al., 1995; Gagosian and Peltzer, 1986; Eglinton et al., 2002; Kawamura et al., 2003) is still recognizable in sediments (Prahl et al., 1989; Wakeham et al., 2002; Zonneveld et al., 2010). Myriad biogeochemical and sedimentological processes during transport through the marine water column and at the sediment-water interface affect the quantity and nature of sedimentary OC. Structural characterization of most marine organic matter is incomplete (Hedges et al., 2000; Lee et al., 2004), but it is generally thought that marine OC is more reactive than terrigenous plant and relict sediment OC (Cowie and Hedges, 1984; Wakeham et al., 1997). Even a small fraction of the most recalcitrant relict carbon is amenable to bacterial assimilation (Petsch et al., 2001, 2003; Pearson et al., 2005; Wakeham et al., 2006). Selective enrichment of terrigenous OC in marine sediments over what is observed in the water column may result from differences in the intrinsic reactivity of the organic molecules themselves, protection by degradation-resistant macro-organic matrices and mineral surfaces, or environmental conditions (oxygen availability, oscillating redox, microbial consortia present) (Hedges and Keil, 1995; Hedges et al., 2001; Aller, 1994; Wakeham and Canuel, 2006; Burdige, 2007).

A suite of geochemical tools are applied to characterize the source(s) and fate of OC in the marine water column and sediments. Elemental compositions and ratios (e.g., OC/N) are often combined with carbon isotope analyses at the bulk level. Natural abundance stable carbon isotopes (δ^{13}C) give insight into carbon source, carbon assimilation pathways and carbon flow in marine ecosystems and food webs (Hayes, 1993; Fry and Sherr, 1994; Freeman, 2001; Pearson, 2010). Natural-abundance radiocarbon analyses ($\Delta^{14}C_{OC}$ or fraction modern f_m) add the dimension of “age” to the character of organic matter and help define the residence time and redistribution of OC (Blair et al., 2003; Ingalls and Pearson, 2005; Griffith et al., 2010). Molecular analyses of biomarkers can distinguish between marine (e.g., sterols, alkenones), terrigenous (plant waxes and lignin phenols) and relict (alkanes with a carbon preference index of ~ 1 and often an unresolved complex mixture) materials in the heterogeneous mixture that is sedimentary OC. But biomarker compounds are often present at low concentrations, and extrapolations to bulk OC are formidable. Single-compound (compound-specific) isotope analyses help in this respect because they combine the source-specificity of biomarkers with $\delta^{13}C_{biomarker}$-derived information on carbon flow (Freeman et al., 1990; Hayes, 2001; Freeman, 2001) and $\Delta^{14}C_{biomarker}$-derived ages that indicate mixing of old with modern OC (Eglinton et al., 1997; McNichol and Aluwihare, 2007; Ingalls and Pearson, 2005). Early investigations in the marine water column showed that whereas sinking particulate OC comprised of fresh planktonic detritus has a young radiocarbon age, surface sedimentary OC has older ages of a "pre-aged" and/or relict terrigenous component (Druffel et al., 1996; Wang et al., 1998; Hwang et al., 2010). Compound-specific radiocarbon measurements confirm pre-aged biomarkers in sedimentary OC (review by Ingalls and Pearson, 2005), and compound-class radiocarbon analyses have subsequently shown pre-aged OC in water column particulate matter (Wang et al., 2001; review by McNichol and Aluwihare, 2007). However, to date combined biomarker/stable carbon/radiocarbon studies of marine particulate matter are few (an exception being Ingalls et al., 2006), largely due to sample size limitations.

The present investigation attempts to fill this gap. The combination of lipid biomarker composition with molecular stable- and radio-carbon isotopes offers a three-dimensional approach for investigating OC sources and transport and alteration processes in the ocean. In this study, compound-specific stable carbon and radiocarbon isotopes were measured on multiple lipid biomarkers in sinking particulate matter collected in sediment traps and from underlying surface sediments to evaluate the provenance of sedimentary organic matter. Three sets of paired sediment trap-surface sediment samples (POM – particulate organic matter; SOM – sedimentary organic matter) from the Black Sea, Arabian Sea and Ross Sea were investigated as representatives of oceanic regions characterized by widely disparate OC sources and depositional environments. Organic carbon content (% OC), atomic C/N ratios ($C/N_{(a)}$), $\delta^{13}C$ and $\Delta^{14}C$

of bulk materials and individual biomarkers (fatty acids, hydrocarbons, alkenones, alcohols, and sterols) were measured to identify major molecular and isotopic compositions, and thereby shifts in relative amounts of marine, terrigenous (pre-aged vascular plant-derived with continental residence times of decades to centuries) and relict (derived from eroded ancient sedimentary rocks and petrogenic material) carbon, in POM and underlying SOM. Throughout this discussion it is important to remember that the sediment trap samples are short snap-shots in time (up to 6 months) whereas the surface sediments may represent centuries.

2 Materials and methods

2.1 Study sites and samples

Paired sediment trap and surface sediment (~ 0–2 cm) samples from three settings (Fig. 1) were studied: Black Sea (anoxic water column; high biogenic and high terrigenous OC); Arabian Sea (oxygen minimum zone; high biogenic OC, low terrrigenous OC); Ross Sea, Antarctica (high biogenic flux, relict continental OC). The choice of these locations was predicated in part on the availability of large amounts of archived sinking particulate matter to facilitate compound-specific radiocarbon analyses.

2.1.1 Black Sea

The Black Sea site was in southwestern Black Sea (42° N, 32° E), at station BS of the joint US Turkish/German sediment trap program 50 km north of Asmara and 15 km from the base of the continental slope (Hay and Honjo, 1989; Hay et al., 1990). Anoxic conditions prevail below ~ 120–150 m water depth (Sorokin, 1983). Underlying sediments contain up to ~ 6 % OC (Ross and Degens, 1974; Premuzic et al., 1982; Calvert et al., 1991). Biogenic and lithogenic particle fluxes and sediment accumulation have been reported by Hay (1987) and Hay et al. (1990). The site is offshore of the rugged Pontic Mountains on the north-Anatolian coastline and the continental margin is particularly steep, typically 6° between 100 and 2000 m depths (Ross et al., 1974). Lithogenic material delivered by the Sarkarya Nehri, Filyos and Kocacay Rivers, usually during winter/spring, is deposited on the shelf, is frequently resuspended by storms, and is subsequently transported off-shore as either surface, mid-water (~ 150 m depth) or bottom water nepheloid layers and turbidites; a surface plume extending out to the study site is sometimes visible on LANDSAT imagery (Hay, 1987). Sedimentation of lithogenic material out of the water column is enhanced by biogenic material derived from the annual succession of blooms of the coccolithophoride *Emiliania huxleyi* (spring) and pennate diatoms *Rhizosolenia* sp. (summer), producing the characteristic light-dark laminae (varve couplets) of Black Sea sediments (Hay, 1987; Hay et al., 1991). White laminae are comprised almost exclusively of coccolithophores of *E. huxleyi*, whereas dark laminae are predominately terrigenous clay minerals.

Sediment trap material (18.5 g dry weight) from the moored BS trap at ~ 250 m depth under strongly anoxic conditions is a composite of a seven month-long time-series collection (October 1985–April 1986). Trap material was preserved with buffered formalin and stored refrigerated at 4 °C. Surface sediment (0–2 cm; 109.7 gdw) was composited from multicores collected at 2200 m water depth during the 1988 Black Sea Expedition (Murray and Izdar, 1989; Hay and Honjo, 1989) at approximately the same location as the trap mooring. Sediments were stored frozen. Radiocarbon dating of sediments near the study site gave a sedimentation rate of 26 cm kyr^{-1} (Arthur and Dean, 1998). Thus the 0–2 cm sediment interval represents approximately one century.

2.1.2 Arabian Sea

Arabian Sea samples were collected at mooring site MS-1 in the northwestern Arabian Sea (Oman Margin) during the US JGOFS Arabian Sea Process Study (ASPS) in 1994–1995 (Smith et al., 1998). MS-1 was approximately 160 km from the Oman coast off Ra’s Sharbatat (17°41′ N, 58°51″ E) at a water depth of 1445 m. High seasonal productivity during monsoon-driven upwelling enhances export into deep waters where remineralization of sinking organic matter depletes dissolved oxygen down to levels of ~ 5 µM, producing the world's largest open ocean oxygen minimum zone (OMZ) (Smith et al., 1998). Greater than 50 % of the annual particle flux in the central Arabian Sea occurs during the boreal summer southwest monsoon. Biogenic material, primarily diatom-derived, dominates over lithogenic material (Haake et al., 1996; Honjo et al., 1999), but some terrigenous material is delivered to the northwestern Arabian Sea as dust from the Horn of Africa (Somalia and Ethiopia) and from the Arabian Peninsula by strong summer monsoon winds (Ramage et al., 1972; Sirocko and Sarnthein, 1989; Dahl et al., 2005).

Sediment trap material (35.5 gdw) was a composite of material collected in time-series traps deployed in the OMZ at ~ 500 and ~ 900 m between May 1995 and January 1996, covering the southwest monsoon period (Wakeham et al., 2002). Mercuric chloride was used as a biocide. Upon recovery, trap samples were sealed and stored refrigerated at 4 °C. Surface sediments (0–2 cm) were composited (142.7 gdw) from multicores collected in 1995 at the same location as the trap deployment and stored frozen. Passier et al. (1997) have estimated the sedimentation rate for this part of the Oman Margin at 5 cm kyr^{-1}; the sediment sample thus represents about 400 years.

2.1.3 Ross Sea

The Ross Sea site was in the southwestern Ross Sea where diatoms, primarily *Nitzschia* sp., and *Phaeocystis Antarctica,* dominate the phytoplankton community and vertical flux during the austral summer bloom (Arrigo et al., 2002; Dunbar et al., 2003). Sediments are largely biogenic oozes (biogenic silica 10–30 %) with low OC (0.1–3 %, averaging 1.5 %) and negligible biogenic carbonate (Dunbar et al., 1985, 1989). Terrigeneous material, primary lithogenics with low OC content, is delivered by glaciers that drain the polar plateau and by eolian transport from the ice-free Dry Valleys and accounts for 2–25 % of the vertical flux through the water column. In the western Ross Sea, ice-rafted debris constitutes ~ 10 % of sediments (Anderson et al., 1984) and aeolian sedimentation (either through sea ice or directly onto the sea surface) could be up to 50 % of sediments in nearshore areas with limited glacial ice cover (Bentley, 1979, cited in Dunbar et al., 1989). Nonetheless, sedimentary OC is dominated by biogenic water column sources. Sediments are resuspended, mixed and redistributed within a pervasive nepheloid layer.

Ross Sea samples were collected during 1998 cruises of the ROAVERRS (Research on Ocean-Atmosphere Variability and Ecosystem Response in the Ross Sea) program (Dunbar et al., 2003). Trap material (48.2 gdw) came from Gentoo and Adelie time-series traps located at about 76° S, 172° E in the southwestern Ross Sea and deployed ~ 50 m above the sea floor in 650-m deep water (Dunbar et al., 2003). Trap material was preserved with 3 % formalin and stored at 4 °C. Surface sediments (515.1 gdw) were obtained from box cores (Ohkouchi et al., 2003) and stored frozen.

Sedimentation rate determinations for the Antarctic margin often use acid-insoluble organic carbon (AIOC) due to lack of calcareous formaninifera (Domack et al. 1989; Licht et al., 1996; Harris et al., 1996; Andrew et al., 1999). DeMaster et al. (1996) reported AIOC-derived sedimentation rates in this region of the Ross Sea at ~ 4.5 cm kyr^{-1}. AIOC-based chronology, however, is complicated by “contamination” by unknown amounts of relict OC (Sackett et al., 1974). To overcome this problem, Ohkouchi et al. (2003) applied compound-specific radiocarbon analysis of sedimentary fatty acids for cores from the Gentoo and Adelie sites and found a 1200–2000 year offset between ages of fatty acids and AIOC. The fatty acid-derived sedimentation rate was 7.5 cm kyr^{-1} vs. an AIOC-derived rate of 15 cm kyr^{-1}. The sediment sample therefore represents 130–250 years of deposition.

2.2 Elemental analysis

Freeze-dried and acidified (Hedges and Stern, 1984) trap material (bulk POM) and sediments (bulk SOM) were analyzed for organic carbon (% OC) and total nitrogen (TN) with a Fisons CHN analyzer (Model EA 1108) elemental analyzer.

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2.3 Lipid analysis

Extraction, cleanup and isolation of fatty acids, hydrocarbons, alcohols and sterols by preparative capillary gas chromatography (PCGC; Eglinton et al., 1996) are outlined in Fig. 2. All laboratory glassware and SiO_2 were precombusted at 500 °C for 8 h before use. Freeze-dried POM and SOM were Soxhlet-extracted with methylene-chloride : methanol (DCM : MeOH, 2 : 1 v/v) for 72 h. Extracts were washed with 5 % NaCl solution and solvent lipid extracts (SLEs) were partitioned into DCM, after which the DCM fraction was dried over Na_2SO_4. SLEs were saponified using 0.5 N KOH at 100 °C for 2 h, and nonsaponifiable lipids were extracted out of the alkaline mixture with hexane after which the pH was adjusted to < 2 with 6 N HCl and acids were extracted with hexane. Non-saponifiable lipids were fractionated on 5 % deactivated silica gel into a hydrocarbon fraction eluted with hexane, an alkenone fraction eluted with 10 % ethylacetate in hexane and an alcohol/sterol fraction eluted with 25 % ethylacetate in hexane. Straight-chained hydrocarbons were separated from branched and cyclic hydrocarbons by urea adduction. Alkenones were isolated by sequential silica gel, $AgNO_3$/silica gel chromatography and urea adduction after Ohkouchi et al. (2005). Alcohols and sterols were acetylated with pyridine and acetic anhydride. Acids were methylated with BF_3 : MeOH and the fatty acid methyl esters (FAMEs) were purified on columns of activated SiO_2.

2.4 Stable and radiocarbon analysis

Isolation of purified individual hydrocarbons, sterol acetates, alcohol acetates and FAMEs was by preparative capillary gas chromatography (PCGC; Eglinton et al., 1996; Wakeham et al., 2006). An HP 5980II GC equipped with an HP 7673 autoinjector, a Gerstel CIS-3 cooled injection system and a Gerstel preparative fraction collector (PFC) was fitted with RTX-1 megabore (60 m × 0.53 mm id × 0.5 µm film) capillary column. The GC temperature program was 60 °C (1 min), 20 °C min^{-1} to 160 °C, 4 °C min^{-1} to 300 °C and isothermal at 300 °C for 20 min. An effluent splitter directed 1 % of the column effluent to the FID and the remaining 99 % was sent to the zero-dead-volume splitter of the PFC. The PFC was operated at 320 °C and U-tube traps were held at room temperature. Purified fractions were checked for purity and quantified by gas chromatography-mass spectrometry (Agilent 6890 gas chromatograph, Agilent 5793 mass spectrometer, 30 m × 0.25 mm i.d. J&W DB-5 capillary column). Individual, composited compounds or operational classes were transferred to glass ampules and flame sealed for isotope analysis.

Stable and radio-carbon isotope measurements were made at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) Facility at the Woods Hole Oceanographic Institution. δ^{13}C values are reported relative to the VPDB (precision ±0.2 ‰) and Δ^{14}C values are reported according to Stuiver and Polach (1977) using the year of sample collection for age correction. Acidified POM and SOM were transferred to pre-combusted Vycor tubes containing CuO and Ag powder. Sample extracts and isolated biomarkers were transferred with solvent to pre-combusted Vycor tubes, and after evaporating the solvent, 100 mg pre-combusted CuO was added to the tube. Samples were combusted to CO_2 at 850 °C for 5 h. After purification and quantification, a split of the CO_2 was analyzed for δ^{13}C on a VG Micromass Optima isotope ratio mass spectrometer. The remaining CO_2 was reduced to filamentous graphite over either Fe or Co powder. Radiocarbon analyses of both large and small samples were performed using standard NOSAMS procedures (McNichol et al., 1994; von Reden et al., 1998; Pearson et al., 1998). Processing and combustion blanks of a hydrocarbon fraction isolated from a south Louisiana crude oil had replicate Δ^{14}C values between −980 and −998 ‰ (f_m 0.001–0.03). Contributions of added methyl carbon derived from methanol in FAMEs and acetyl carbons from acetic anhydride in alcohol and sterol esters were removed by isotopic mass balance (Pearson, 2000; Wakeham et al., 2006). The δ^{13}C and Δ^{14}C values of carbon in the BF_3 : MeOH and acetic anhydride reagents were calculated by measuring the δ^{13}C and Δ^{14}C values of palmitic acid and cholesterol standards and of methylpalmitate and cholesteryl acetate prepared using the same lots of BF_3-MeOH and acetic anhydride, respectively, and isolated by PCGC.

Several FA had unusually high Δ^{14}C values and were not used in subsequent calculations of ranges, means and standard deviations of radiocarbon data (shown in bold italics in Tables 1–3). The reasons for these enrichments are unknown (see Wakeham et al., 2006 for additional discussion). Radiotracers had never been used in the SkIO laboratory that was thoroughly checked for any radiocarbon contamination and crude oil process blanks were free of modern carbon. The most enriched FA (most often but not always 16 : 0, 18 : 1 and 18 : 0) were compounds isolated in the highest concentrations and thus for which larger amounts of carbon were analyzed by AMS rather than the less-abundant compounds. Such enrichments were not systematic, and in several cases replicate AMS analyses from splits of the same isolates were made, but with similar results indicating that any contamination must have occurred prior to or during PCGC workup. However, Levin and Kromer (1997) suggested that the average ^{14}C of atmospheric CO_2 between 1980 and present may have been ∼ 200 ‰ whereas analysis of post-bomb sediment from the Santa Monica Basin by Pearson and Eglinton (2000) indicates an average ^{14}C of atmospheric CO_2 around 1960 may have been ∼ 400 ‰. Thus while it is not possible to completely rule out incorporation of higher amounts of post-bomb ^{14}C into these biomarkers Δ^{14}C, other marine biomarkers and bulk OC are not consistent with such a scenario.

3 Results and discussion

3.1 Bulk elemental compositions

Organic carbon contents (% OC) of POM (Fig. 3a) were 8.7 % in the Black Sea (BS), 6.4 % in the Arabian Sea (AS) and 5.5 % in the Ross Sea (RS). Total nitrogen (% TN) contents were 1.1, 0.86, and 0.92 %, respectively, for the BS, AS and RS trap material. Thus atomic C/N ratios were 7.7, 7.4, and 5.9 (Fig. 3b). Sediment % OC and % TN were lower: 1.8, 2.3, and 0.54 % OC for the BS, AS, and RS, respectively; 0.20, 0.29, and 0.08 % TN for the BS, AS, and RS, respectively. $C/N_{(a)}$ ratios for sediments were somewhat higher (9.4, 7.7, 6.5 for BS, AS, and RS, respectively) than for POM. Except for the BS sediment, $C/N_{(a)}$ ratios of the other trap and sediment samples were sufficiently Redfield-like to indicate the predominance of marine OC. The higher $C/N_{(a)}$ ratio of the BS sediment suggests a somewhat higher component of terrigenous OC; alternately the higher $C/N_{(a)}$ ratio could result from preferential loss of nitrogen during degradation of OC, but this process is likely limited by the anoxic water column of the BS.

3.2 Bulk stable carbon isotopes

BS and AS POM $\delta^{13}C_{OC}$ values were typical for marine dominated OC: −22.9 ‰ and −22.4 ‰ for BS and AS, respectively (Fig. 4a). $\delta^{13}C_{OC}$ values for the corresponding SOM were both slightly more negative (BS −25.3 ‰) and more positive (AS −20.8 ‰) compared to the corresponding POM. The ^{13}C-depletion in the BS sediment could reflect a greater long-term/time-averaged terrigenous C_3-plant OC component (Collister et al., 1994; Conte and Weber, 2002; Chikaraishi et al., 2004) from the heavily wooded Anatolian coast than was present in the short-term trap sample. The relative enrichment of the AS sediment compared to the trap material could be the result of the addition of isotopically-enriched aeolian-transported OC derived from C_4-grasses in the arid Horn of Africa and Arabian Peninsula (Parker et al., 2004; Dahl et al., 2005), but again sampling time-scales for trap and sediment are different.

The Ross Sea is a very different environment. Both POM and SOM had $\delta^{13}C_{OC}$ values of −27.9 ‰, significantly isotopically depleted compared to BS and AS samples. Algal biomass at high latitudes is typically depleted in ^{13}C relative to algal OC at lower latitudes (Rau et al., 1991a, b; DeHairs et al., 1997; Freeman, 2001). Several factors may be involved in this differential photosynthetic isotope fractionation, including high algal growth rates, higher dissolved CO_2 concentrations associated with low seawater temperatures, and carbon assimilation mechanism (Rau et al., 1991a, b; DeHairs et al., 1997; Freeman, 2001). In the open southwest Ross Sea, diatom and *Phaeocystis* biomass is −28 ‰ to −27 ‰ (Villinski et al., 2000). Sea-ice algae would add ^{13}C-enriched OC (Gleitz et al., 1996; Gibson et al., 1999; Villinski et al., 2000) but is not a significant source of algal OC at the study site. Antarctic kerogen and coal have $\delta^{13}C_{OC}$ values of −22 ‰ and −24 ‰, respectively (Sackett, 1986; Burkins et al., 2000), and soils from the McMurdo Dry Valley region have a wide range of $\delta^{13}C_{OC}$ values, ~ −30 ‰ to ~ −18 ‰ (and $C/N_{(a)}$ of 11 ± 4) (Burkins et al., 2000).

Solvent lipid extracts (SLE) would help bridge the gap between bulk OC and individual biomarkers In all three sample pairs, SLE's were depleted by up to 4 ‰ in ^{13}C compared with the respective bulk OC's (Fig. 4b). $\delta^{13}C_{SLE}$ values for both traps and sediments followed the same trend among samples as $\delta^{13}C_{OC} : \delta^{13}C_{AS\text{-}SLE} > \delta^{13}C_{BS\text{-}SLE} > \delta^{13}C_{RS\text{-}SLE}$. Lipids are a relatively abundant fraction of OC in phytoplankton and zooplankton but are usually only a few percent of OC in particulate matter and sediments (Wakeham et al., 1997; Wang and Druffel, 2001). They would thus not be major contributors to particulate and sedimentary $\delta^{13}C_{OC}$ compared to more abundant proteins and carbohydrates that are ~ 4–6 ‰ enriched in ^{13}C relative to lipids. Intra-class comparisons of isotopic compositions in marine systems are few. A study of $\delta^{13}C$ of particulate and sedimentary OC in the northeast Pacific and Southern Oceans found that total hydrolysable amino acids (THAA) and total carbohydrates (TCHO) in phytoplankton, zooplankton and sediments were enriched in ^{13}C by about ~ 2 ‰ and ~ 3 ‰, respectively, relative to OC whereas lipids were depleted ~ 4 ‰ relative to OC (Wang et al., 1998; Wang and Druffel, 2001).

3.3 Bulk radiocarbon isotopes

Radiocarbon isotopic values for BS and AS trap OC ($\Delta^{14}C_{OC}$) were 21 ‰ and 14 ‰ and for BS and AS sediments were −199 ‰ and −138 ‰, respectively (Fig. 4c and Tables 1–3 which also lists radiocarbon ages and fraction (%) modern, f_m). RS POC and SOC were depleted in ^{14}C (−208 ‰ and −355 ‰, respectively) relative to the two other sites. $\Delta^{14}C_{DIC}$ values for pre-bomb and post-bomb mixed-layer DIC in the Black Sea are ~ −70 ‰ and 100 ‰, respectively (Jones and Gagnon, 1994) and in the AS are ~ −60 ‰ and ~ 70 ‰ (Stuiver and Östlund, 1983; Southon et al., 2002). Pre-bomb and post-bomb $\Delta^{14}C_{DIC}$ values in the Ross Sea are lower than elsewhere, −130 ‰ and −100 ‰, respectively (Berkman and Forman 1996; Gordon and Harkness 1992; Hall et al., 2010). In the AS and RS, upwelling of ^{14}C-depleted deep-waters and the short residence time of surface waters lead to the high surface-water reservoir ages (~ 700 yr and ~ 1100 yr for AS and RS, respectively vs. ~ 400 yr for open ocean locations, including the BS; Stuiver and Braziunas, 1993; Siani et al., 2000; Southon et al., 2002; Key et al., 2004; Hall et al., 2010). Thus trap OC displayed post-bomb signatures for their respective regions but additionally contained older carbon, especially in the Ross Sea. Sediments contained significant contributions of old carbon, and corrected $^{14}C_{OC}$-ages (Tables 1–3) are significantly older than estimated geological ages (100–400 yr, admittedly based on $^{14}C_{OC}$-derived sedimentation rates).

Trap and sediment SLE's for the BS and RS had higher $\Delta^{14}C_{SLE}$ values than corresponding $\Delta^{14}C_{OC}$ values (Fig. 4d): $\Delta^{14}C_{SLE}$ values were −96 ‰ and −150 ‰ for BS trap and sediment, respectively; −154 ‰ and −211 ‰ for RS trap and sediment. That the BS and RS trap lipid fractions had lower $\Delta^{14}C_{SLE}$ values than $\Delta^{14}C_{OC}$ values indicates that some old carbon was extractable (e.g., plant waxes and petroleum hydrocarbons). But the higher sediment $\Delta^{14}C_{SLE}$ values than sediment $\Delta^{14}C_{OC}$ suggested that residual OC remaining after solvent extraction must be, by extension, still older, such as non-extractable kerogen, especially in the RS sediment. On the other hand, $\Delta^{14}C_{SLE}$ of the AS trap material was enriched (66 ‰) compared to $\Delta^{14}C_{OC}$ value, but the sediment was depleted (−173 ‰) relative to its $\Delta^{14}C_{OC}$ value. Thus solvent extraction of the AS trap material released a greater proportion of fresh, young algal OC, as lipid, into the SLE but extraction of the sediment left behind a greater proportion of non-extractable, old OC. In the only other reports to our knowledge that measured $\Delta^{14}C_{SLE}$ (Wang et al., 1998; Wang and Druffel, 2001), plankton and sedimentary lipids in the northeastern Pacific and Southern Ocean were similar to or lower than, respectively, Δ^{14}C of total OC. Lipids, THAA and TCHO all had similar Δ^{14}C values in plankton, but in sediments lipids usually had lower Δ^{14}C signatures than THAA and THCO.

3.4 Biomarker molecular compositions

Biomarker analyses focused on fatty acids, alkenones, fatty alcohols, sterols, and hydrocarbons (Figs. 5–7). In the following discussion, the operational distinction is made between biomarkers of marine origin (hereafter termed "marine OC"), those derived from pre-aged terrestrial vascular plants ("terrigenous OC") and compounds originating from eroded ancient sediment or petrogenic sources ("relict OC"). In all POM samples, short-chain C_{14}–C_{24} *n*- and methyl branched *iso*- and *anteiso*-C_{15} and C_{17} compounds of marine biomass dominated fatty acid distributions (Volkman, 2006). Long-chain, even-carbon number predominant C_{24}–C_{30} terrestrial vascular plant *n*-fatty acids were ~ 10-fold less abundant. Sediments contained similar fatty acid distributions but with higher relative abundances (but still ~ 3–5-fold less abundant) of long-chain compounds. Long-chain C_{37}–C_{39} alkenones derived from the haptophyte, *Emiliania huxleyi* (Volkman et al., 1980), were abundant in BS (where coccoliths of *E. huxleyi* constitute the light laminae) and AS traps and sediments, but absent from the RS. Low levels of hydrocarbons, a mix of C_{15}–C_{36} *n*-alkanes and an unresolved complex mixture (UCM) in the C_{14}–C_{22} carbon number range were present in BS and AS samples at levels ~ 10-fold lower than fatty acids; none above blanks were detected in the RS. Short-chain *n*-alkanes (C_{16}–C_{22}) showed no odd-over-even carbon number predominance (CPI ~ 1) and were underlain by an unresolved complex mixture (UCM), whereas long-chain *n*-alkanes (C_{25}–C_{31}) were odd-carbon predominant (CPI > 5). C_{25}-highly branched isoprenoid (HBI) alkenes of diatom origin were the dominant hydrocarbons in AS POM but were only minor components in BS POM and sediments and AS sediments. No hydrocarbons (above blanks) were detected in RS samples. POM contained abundant *n*-hexadecanol (16 ROH, assumed derived primarily from zooplankton wax esters) and C_{27}–C_{30} – Δ^5, $\Delta^{5,22}$, $\Delta^{5,24(28)}$ sterols [e.g., cholest-5-en-3β-ol (cholesterol), abbreviated as 27Δ^5; 24-methylcholesta-5,22-dien-3β-ol, 28$\Delta^{5,22}$; 24-methylcholesta-5,24(28)-dien-3β-ol, 28$\Delta^{5,24(28)}$; and in the BS, a significant amount of 4,23,24-trimethylcholest-22-en-3β-ol (dinosterol), 30Δ^{22}], all of marine origin (Volkman, 2006). POM contained only low amounts of even-carbon number predominant C_{24}–C_{30} *n*-alcohols produced by vascular plants, but sediments contained higher abundances of terrestrial *n*-alcohols than sterols.

3.5 Biomarker isotopic compositions

PCGC isolation for compound-specific isotope analyses targeted the most abundant biomarkers of marine, terrigenous plant and relict origins. Short chain *n*-fatty acids (C_{14}–C_{24}, abbreviated as 14 : 0 FA, etc), $C_{37} + C_{38}$-alkenones, a short-chain *n*-fatty alcohol [16ROH (*n*-hexadecanol)] and sterols [cholest-5-en-3β-ol (cholesterol), abbreviated as $27\Delta^5$; 24-methylcholesta-5,22-dien-3β-ol, $28\Delta^{5,22}$; and 4,23,24-trimethylcholest-22-en-3β-ol (dinosterol), $30\Delta^{22}$], were designated as marine biomarkers. Long-chain even-carbon number *n*-acids (C_{26}–C_{30}) and *n*-alcohols (C_{24}–C_{28}) and long-chain odd-carbon numbered *n*-alkanes (C_{27}, and C_{29}) were tagged as terrigenous, vascular plant markers; in some cases the long chain fatty acids could be isolated individually but in other cases composites (e.g. $C_{24} + C_{26} + C_{28}$ fatty acids) were required. Long-chain even-carbon numbered *n*-alkanes (composited $C_{24} + C_{26} + C_{28}$) and, in the AS sediment, short-chain $C_{15} + C_{16} + C_{17} + C_{18}$ alkanes (and including some unresolved complex mixture that could not be removed) were used as relict markers since these alkanes are not abundant in marine or terrestrial biomass. As will be shown below, using long-chain *n*-alkanes as either terrigenous plant or relict markers is problematic because in the present samples there is overlap between odd vs even chain lengths and plant vs. relict sources. Thus it is possible that long-chain *n*-alcohols, if present in sufficient abundances, may represent the best “terrigenous” biomarkers since they would have neither modern marine (major alcohols in wax esters are C_{16} and C_{18}; Wakeham, 1982) nor relict sources. Highly-branched C_{25} alkenes (HBI) of diatom origin (Belt et al., 2000) were the most abundant hydrocarbons in AS trap material, less so in AS sediments, but they were underlain by a UCM that could not be removed, thus yielding unrealistically ^{14}C depleted results. In the RS POM and SOM, only fatty acids, 16 ROH, and sterols (cholest-5-en-3β-ol and 24-methylcholesta-

5,22-dien-3β-ol) could be isolated. Stable carbon and radiocarbon isotope results were weighted according to their abundance within each sample to give means for each of the three sources (marine, terrigenous, and relict) that are designated below as $\delta^{13}C_M$ and $\Delta^{14}C_M$, $\delta^{13}C_T$ and $\Delta^{14}C_T$, and $\delta^{13}C_R$ and $\Delta^{14}C_R$.

3.5.1 Black Sea

For the Black Sea POM, short-chain fatty acids, long chain alkenones, and sterols had $\delta^{13}C$ values between −26.1 ‰ to −23.2 ‰ (Fig. 8a and Table 1), generally 1–4 ‰ more ^{13}C depleted than bulk POM (−22.9 ‰). Interestingly, alkenones were the most ^{13}C depleted of this group. Overall these "marine" biomarkers had an abundance weighted average $\delta^{13}C_M$ of −25.3 ± 1.1 ‰ (Table 4), typical for marine lipids. Radiocarbon values for the marine biomarkers (excluding the highly ^{14}C-enriched 18 : 1 fatty acid outlier at 288 ‰) ranged from 65 ‰ to 146 ‰, giving $\Delta^{14}C_M$ of 92 ± 28 ‰, considerably enriched in ^{14}C relative to bulk POM ($\Delta^{14}C_{OC}$ −199 ‰) (Fig. 8b). Long-chain even-carbon number acids and alcohols and odd-carbon-number alkanes had $\delta^{13}C$ values ranging from −30.1 ‰ to −27.0 ‰, yielding a mean $\delta^{13}C_T$ – 28.7 ± 1.6 ‰, roughly 3‰ depleted in $\delta^{13}C$ compared to marine lipids. Terrigenous markers had a much greater range of $\Delta^{14}C$ values (1 ‰ for the fatty acids, −44 ‰ for the alcohols, and −181 ‰ for the alkanes) but a mean $\Delta^{14}C_T$ of −53 ± 60 ‰. It is likely that some fraction of the long chain fatty acids are zooplankton (wax ester) derived young carbon whereas the long chain alkanes may contain a relict component. The single sample of "relict" biomarkers isolated by PCGC was a composite of $C_{24} + C_{26} + C_{28}$ *n*-alkanes that had a $\delta^{13}C_R$ of −29.3 ± 0.2 ‰ and a $\Delta^{14}C_R$ of −677 ± 10‰; this $\Delta^{14}C$ value probably also reflects a mix of moderately pre-aged terrigenous ($\Delta^{14}C$ of −44 ‰ might be reasonable if the alcohols are a good representative of terrigenous OC) and radiocarbon-dead ($\Delta^{14}C_R$ of −1000 ‰) relict carbon.

Marine biomarkers in the BS sediment had $\delta^{13}C$ values ranging from −31.9 ‰ to −26.0 ‰ (mean $\delta^{13}C_M$ – 28.8 ± 1.1 ‰), about 2.5 ‰ depleted in ^{13}C relative to both BS bulk SOM (−25.3 ‰) and the marine group of BS POM (also −25.3 ‰). Radiocarbon contents of marine biomarkers ranged from −46 ‰ to 75 ‰ (mean $\Delta^{14}C_M$ 2 ± 44 ‰, excluding 16 : 0 at 214 ‰ and 18 : 0 at −374 ‰), considerably enriched in ^{14}C relative to bulk POM ($\Delta^{14}C_{OC}$ − 199 ‰) but depleted in ^{14}C compared to marine biomarkers in BS POM. Terrigenous biomarkers displayed $\delta^{13}C$ values between −31.9 ‰ to −29.7 ‰ (mean $\delta^{13}C_T$ − 30.5 ± 0.65 ‰). Thus although there was a considerable overlap in $\delta^{13}C$ values for marine and terrigenous groups, concentration weighting yielded an offset of ∼ 2 ‰ as would be expected. Plant-wax alkanes (C_{27} and C_{29}) and alcohols (C_{24} and C_{26}) were strongly depleted in $\Delta^{14}C$ relative to the marine lipids (range −231 ‰ to −100 ‰), with a mean $\Delta^{14}C_T$ of −171 ± 58 ‰. Long chain even-carbon numbered [$C_{24} + C_{26} + C_{28}$] alkanes had a $\delta^{13}C$ value of −29.3 ‰ and a $\Delta^{14}C_R$ of −609 ‰, again suggesting they are not exclusively relict.

3.5.2 Arabian Sea

The marine biomarkers in the AS trap POM had $\delta^{13}C$ values between −27.1 ‰ and −16.7 ‰ (mean $\delta^{13}C_M$ − 23.5 ± 2.5) and $\Delta^{14}C$ contents ranging from −91 ‰ to 111 ‰ (Fig. 9a and b and Table 2) (mean $\Delta^{14}C_M$ − 67 ± 40 excluding $C_{14:0}$ at 178 ‰ and HBI alkenes at −514 ‰). The negative $\Delta^{14}C$ value for the HBI alkenes is due inclusion of a UCM which could not be removed. The only vascular plant compounds in the AS trap in sufficient quantity for AMS analysis were analyzed as a composite of [$C_{27} + C_{29}$] *n*-alkanes, giving a $\delta^{13}C_T$ of −28.4 ‰ and a $\Delta^{14}C_T$ of −320 ‰. The single sample of [$C_{24} + C_{26} + C_{28}$] *n*-alkanes gave a $\delta^{13}C_R$ of −28.5±0.2 ‰ and a $\Delta^{14}C_R$ of −731±14 ‰.

The $\delta^{13}C$ values for marine biomarkers in AS sediments ranged from −26.8 ‰ to −18.2 ‰ ($\delta^{13}C_M$ mean −24.5±2.1 ‰), and $\Delta^{14}C$ ranged from 104 to −171 ‰ ($\Delta^{14}C_M$ − 59 ± 60 ‰, excluding 18 : 0 FA at 190 ‰ and HBI alkenes at −256 ‰). Vascular plant [$C_{26} + C_{28} + C_{30}$] *n*-alcohols had a $\delta^{13}C$ value of −24.1 ‰ and a $\Delta^{14}C$ value of −113 ± 13 ‰ and [$C_{27} + C_{29}$] *n*-alkanes displayed a $\delta^{13}C$ value of −27.7 ‰ and a $\Delta^{14}C$ value of

−430±11 ‰. Together, the terrigenous biomarkers had a concentration weighted $\delta^{13}C_T$ value of −27.7 ± 0.20 ‰ and a $\Delta^{14}C_T$ of −270 ± 110 ‰. Two groups of *n*-alkanes in AS sediments could be assigned to relict sources. Short chain $[C_{14} + C_{15} + C_{16} + C_{17}]$ alkanes and the UCM under this group of homologs had a $\delta^{13}C$ value of −29.4 ‰ and a $\Delta^{14}C$ value of −887 ‰. Long-chain $[C_{24} + C_{26} + C_{28}]$ *n*-alkanes had a $\delta^{13}C$ value of −27.7 ‰ and a $\Delta^{14}C$ value of −430 ‰. Thus $\delta^{13}C_R$ and $\Delta^{14}C_R$ for the AS sediments would be −28.4 ± 0.2 ‰ and −879 ± 45 ‰, respectively.

3.5.3 Ross Sea

Ross Sea POM and SOM contained only marine-derived fatty acids (here C_{26} is considered marine assuming the absence of much terrigenous FA input to the RS) and alcohols/sterols in sufficient abundance for compound-specific isotopic analysis. In keeping with a ^{13}C isotope depletion in high latitude regions, noted above, fatty acids had $\delta^{13}C$ values ranging from −35.3 to −31.7 ‰ and alcohols/sterols values ranging from −35.4 to −31.8 ‰ (Fig. 10a and Table 3), together giving a mean $\delta^{13}C_M$ of −33.3 ± 1.8 ‰. Radiocarbon contents ranged from −216 to −100 ‰, with the alcohols/sterols slightly depleted relative to the fatty acids, for a $\Delta^{14}C_M$ – 155 ± 47 ‰ (Fig. 10b).

Fatty acids and sterols/alcohols in RS sediments had $\delta^{13}C$ values ranging from −36.6 to −31.7 ‰ and −34.3 to −33.0 ‰, respectively, for a mean $\delta^{13}C_M$ of −34.3 ± 1.9 ‰. Radiocarbon contents of fatty acids from RS sediments were more variable than for RS POM, with values ranging from −302 to −83 ‰. Sterols from RS sediments were very unchanged from sterols in POM with $\Delta^{14}C$ values between −202 and −178 ‰ for sterols. Overall this gave a mean $\Delta^{14}C_M$ of −163 ± 77 ‰, also essentially the same as for $\Delta^{14}C_M$ of the trap material.

3.6 The provenance of POM and SOM in the Black Sea and Arabian Sea

Concentration-weighted isotope values (Table 4) can be used to constrain the ranges of isotopic compositions of marine, terrigenous and relict biomarkers, and by extension organic carbon, in Black Sea and Arabian Sea POM and SOM. Neither terrigenous nor relict biomarkers could be isolated from the Ross Sea in sufficient quantities for AMS analyses. Overall, $\delta^{13}C_M > \delta^{13}C_T \sim \delta^{13}C_R$ and $\Delta^{14}C_M > \Delta^{14}C_T \gg \Delta^{14}C_R$ (Fig. 11). Furthermore, SOM biomarkers were generally ^{13}C-depleted and ^{14}C-depleted relative to their corresponding POM samples, indicative of higher proportions of ^{13}C depleted but older, pre-aged OC in sediments. The range of $\delta^{13}C_R$ was relatively small compared to $\delta^{13}C_M$ and $\delta^{13}C_T$ of POM and SOM, but the span of $\Delta^{14}C_R$ values was quite large since the odd carbon number hydrocarbons constituting these groups are a mix of old but not radiocarbon "dead" terrigenous vascular plant ($\Delta^{14}C$ values similar to average $\Delta^{14}C_T$ values) and truly relict ($\Delta^{14}C = 1000$ ‰) OC.

The relative contributions of marine, pre-aged terrigenous, and relict OC (f_M, f_T, and f_R) to sediment trap material and sediments can be estimated in the Black, Arabian, and Ross Seas using an isotopic mass balance. Plots of $\delta^{13}C_{M,T,R}$ vs. $\Delta^{14}C_{M,T,R}$ in both the sediment traps and sediment (Fig. 11) show significantly different values for the radiocarbon content of the different carbon pools but relatively small differences in the stable isotopic content in the Black and Arabian Seas and in the Ross Sea sediments. The small differences in $\delta^{13}C$ coupled with significant error make it difficult to use the stable isotopic composition as a discriminating factor in determining the sources of the bulk material. However, the large differences observed in the $\Delta^{14}C$ values make it possible to use these data to constrain the relative amounts of relict, terrestrial and marine components.

For each sample, there is a $\Delta^{14}C$ value assigned to the bulk SLE, fresh marine, fresh terrestrial and a relict/terrestrial mix. Knowing that the bulk material is a mix of fresh marine and other "added" material, we can calculate the relative amounts of marine and "added" material.

$$1 = f_{\mathrm{M}} + f_{\mathrm{added}} \tag{1}$$

$$\Delta^{14}\mathrm{C}_{\mathrm{bulk}} = f_{\mathrm{M}}(\Delta^{14}\mathrm{C}_{\mathrm{mar}}) + f_{\mathrm{added}}(\Delta^{14}\mathrm{C}_{\mathrm{added}}) \tag{2}$$

Solving this equation for f_{added}

$$f_{\mathrm{added}} = \frac{\Delta^{14}\mathrm{C}_{\mathrm{bulk}} - \Delta^{14}\mathrm{C}_{\mathrm{M}}}{\Delta^{14}\mathrm{C}_{\mathrm{added}} - \Delta^{14}\mathrm{C}_{\mathrm{M}}} \tag{3}$$

The value of $\Delta^{14}\mathrm{C}_{\mathrm{added}}$ can range from that for relict material (−1000 ‰) to that of the bulk material. Using this information, we can calculate the relative amounts of marine and "added" material as a function of the Δ^{14}C of the "added" material. Further, we can combine the radiocarbon value measured on the terrestrial portion with the value of truly relict material to calculate how much of the "other" material might come from fresh terrestrial material using the equations below.

$$f_{\mathrm{added}} = f_{\mathrm{T}} + f_{\mathrm{R}} \tag{4}$$

$$\Delta^{14}\mathrm{C}_{\mathrm{added}} = f_{\mathrm{T}}(\Delta^{14}\mathrm{C}_{\mathrm{T}}) + f_{\mathrm{R}}(\Delta^{14}\mathrm{C}_{\mathrm{R}}) \tag{5}$$

Truly relict material has a Δ^{14}C value of −1000 ‰ and fresh terrigenous material has the values determined in this study ($\Delta^{14}\mathrm{C}_{\mathrm{T}}$, Table 5). If we assume that the value we measured for "relict" material (Table 5) is a good representation of $\Delta^{14}\mathrm{C}_{\mathrm{added}}$, then we have a unique solution to the mass balance. This solution is indicated by the vertical lines in Fig. 12 and the values listed in Table 5. Using this model, we force ourselves to an extreme situation where all the "added" material is either vascular or totally dead. Thus, the relative fractions of terrestrial and relict material must be considered maximum and minimum values, respectively.

For the calculations, concentration weighted $\delta^{13}\mathrm{C}_{\mathrm{M}}$, $\delta^{13}\mathrm{C}_{\mathrm{T}}$, $\delta^{13}\mathrm{C}_{\mathrm{R}}$, $\delta^{13}\mathrm{C}_{\mathrm{B}}$, $\Delta^{14}\mathrm{C}_{\mathrm{M}}$, $\Delta^{14}\mathrm{C}_{\mathrm{T}}$ and $\Delta^{14}\mathrm{C}_{\mathrm{B}}$ values listed in Table 4 were used. The relative fractions of marine, terrestrial and relict material (f_M, f_T, and f_R) as a function of $\Delta^{14}C_{other}$ are shown in Fig. 12. It is most likely that the marine component is dominant in both the trap and sediment samples, although the possible values range from > 80 to 0 %. Fresh vascular material can account for 0 up to almost 100 %. The amount of relict material is constant at a low proportion, ranging from 0 to < 20 % in both the trap and sediments. In the AS, the marine component is dominant in both the trap and sediments; the trap data indicate that there can be virtually no relict or vascular material input to this sample, while the sediment data suggest that up to 30 % of the material may come from either relict or vascular sources. In the RS sediment, marine material dominates in almost all instances, with possible values ranging from 90 to 30 %. Relict material can account for > 10 to 0 % and vascular from 0 to < 70 %.

In the Black Sea and Arabian Sea cases here, it is likely that the marine component contributes the most material because a marine biogenic flux dominates at both sites. No comparable combined biomarker, ^{13}C, and ^{14}C studies of POM at other sites exist to our knowledge. But the high amounts of marine OC in the BS and AS sediments contrasts with other environments where similar biomarker-isotopic mass balances have been made, although using a narrower range of biomarkers. In nearshore sediments on the northern California Margin off the high-energy Eel River that erodes ancient sedimentary rocks in its watershed (Blair et al., 2003), fractional contributions from marine, terrestrial and relict components were estimated at 0–10, 50–75 and 30–50 % of total OC (Drenzek et al., 2009). On the Washington Margin off the lower-energy Columbia River, marine, terrestrial and relict OC contribute 2, 89–95 and 3–9 % of bulk sedimentary OC (Feng et al., 2013). Sediments in the Beaufort Sea had f_M, f_T and f_R values of 13–27, 36–42, and 34–37 %, respectively, with the elevated f_R values consistent with the well-defined petrogenic signature of *n*-alkanes and polycyclic aromatic hydrocarbons in Mackenzie River and Beaufort Sea sediments (Yunker et al., 1993, 2002). For surface sediments in the southwest Black Sea near our study site, Kusch et al. (2010) estimate a f_R of ~ 18 % of TOC. In the Santa Monica Basin of the California Borderlands, 80–87 % of the *n*-alkanes were of terrigenous plant origin whereas up to 20 %

of the alkanes were derived from petroleum or shales (Pearson and Eglinton, 2000), but since alkanes are not abundant in most marine biota, no estimation of the marine component was made.

In the BS, the graphs for the relative contributions look similar for both the trap and sediment samples. A strict interpretation of these results suggests that there is a greater relative amount of non-marine material in the sediment trap than in the sediments themselves. Given the limitations of our data set, we believe that it is more likely that the relative amounts of material are actually very similar. This is somewhat surprising for two reasons. First, under a differential degradation/preservation case (Wakeham and Canuel, 2006), a more labile component of marine OC would be selectively degraded as POM moved between the trap depths and the surface sediments, leaving behind increased proportions of selectively preserved, more refractory terrigenous and relict components in sediments. Alternately, lateral advection of terrigenous and relict OC below the trap depths either by mid-depth or bottom currents might simply allow this material to bypass the traps to be deposited directly on the sediments. At the Black Sea site, advective transport of continental material containing terrigenous and relict carbon to the study site is likely via surface, mid-water (~ 150 m depth) or bottom water nepheloid layers and turbidites. Surface and mid-water plumes moving offshore could carry some continental material to the shallow (~ 250 m) sediment trap. But a greater amount of terrigenous and relict carbon might, after temporary storage on the narrow continental shelf, be remobilized to move laterally down the steep continental slope under the trap. This type of advective remobilization of pre-aged material is well documented at other locations for both marine derived alkenones (e.g., Ohkouchi et al., 2002; Mollenhauer et al., 2003) and terrigenous/relict OC (e.g., Aller et al., 2004; Mead and Goñi, 2006; Mollenhauer and Eglinton, 2007; Kusch et al., 2010; Hwang et al., 2010). Aeolian transport of leaf wax OC and petrogenic OC to the sediments of the central Black Sea have been reported (Wakeham 1996; Eglinton et al., 1997), but the importance of aeolian delivery to the southwestern Black Sea is unknown. Nonetheless, a small leaf wax/petrogenic signal was detected in the trap POM. Because the trap was already deployed within the anoxic zone (which starts at 120–150 m), OC degradation in the water column and surface sediments should be depressed, so that degradation might not be a significant cause for any reduction in f_M for the sediments.

In the Arabian Sea, the sediment trap material indicates the material is all marine while the sediments show a small influence of non-marine material. The Arabian Sea site was in an area of intense upwelling dominated by high export of diatomaceous material (Wakeham et al., 2002), remote from fluvial inputs but potentially affected by aeolian transport of pre-aged OC off the Arabian Peninsula and Horn of Africa during the windy monsoon periods (Dahl et al., 2005). The AS trap was deployed within the OMZ where organic matter degradation has reduced dissolved oxygen concentrations to ~ 5 µM (Smith et al., 1998), but AS sediments were collected at 1400 m water depth where bottom waters are oxygenated. Indeed within and below the OMZ, OC fluxes decreased 5–10 fold between the trap and surface sediments, even as % OC did not change as dramatically (Lee et al., 2000; see Wakeham et al., 2002 for AS lipid fluxes). Comparative studies have shown conclusively that, among other things (e.g., intrinsic reactivity of organic molecules and protection by macromolecular organic matrices and mineral surfaces), oxygen availability is a key control on OC and lipid degradation/preservation water columns and sediments (e.g., Hedges and Keil, 1995; Gong and Hollander, 1997; Wakeham and Canuel, 2006; Burdige, 2007; Mollenhauer and Eglinton, 2007). Enhanced degradation (diminished preservation) in the oxygenated AS should be greater than in the anoxic Black Sea, and if marine OC is more labile than terrigenous/relict OC, f_M in the AS POM and SOM would, as observed, be higher than in the BS. Extensive production of petroleum reserves on the Arabian Peninsula and intense tanker traffic in the Arabian Sea is an additional source of refractory and radiocarbon-dead petrogenic OC to AS particulate matter and sediments.

The RS sediment results are consistent with previous studies suggesting that marine material is the most important source of organic matter to the sediments (Arrigo et al., 2002; Dunbar et al., 2003). The solutions that suggest a relatively large input of vascular plant material seem unrealistic given these prior studies. The Ross Sea data set is very limited and in fact does not include unambiguous lipids of either terrestrial or relict origins with which to constrain the isotopic compositions of these endmembers.

4 Conclusions

This study examined the compound-specific ^{13}C and ^{14}C compositions of diverse biomarker indicators of marine biomass, terrigenous vascular plant, and relict sources of organic carbon in sediment traps and underlying surface sediments in the Black Sea, the Arabian Sea and the Ross Sea. Using an isotopic mass balance approach, it was possible to constrain relative inputs from these three sources, with marine biomass accounting for 66–100 % of extractable lipids and organic carbon in Black Sea and Arabian Sea sediment trap material. The remaining 3–8 % derives from terrigenous and 4–16 % from relict sources. Sediments contained lower proportions of marine biomarkers (66–90 %) and consequentially higher proportions of terrigenous and relict carbon (3–17 % and 7–13 %, respectively). These results suggest that although particulate organic carbon is overwhelmingly marine in origin, there are significant proportions of pre-aged terrigenous and relict OC present. Because these latter fractions become proportionally more important in sediments, it is likely that they are better preserved than the marine component, and/or they reach the sediments by lateral advection rather than only by the vertical sinking that affects the upper ocean-derived marine POC. This approach demonstrates the strengths, and limitations, of such a multi-parameter approach for studying marine OC cycling and budgeting.

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Table 1. Stable carbon and radiocarbon isotope data for Black Sea POM and SOM.

	source	ID #	δ^{13}C (‰)	Δ^{14}C (‰)	f_m	Age
			Black Sea Trap			
OC		OS-32870	−22.9	21 ± 2	1.026 ± 0.003	> Mod
SLE		OS-38316	−27.2	−96 ± 16	0.909 ± 0.016	770 ± 140
14 : 0 FA	M	OS-38328	−25.6	132 ± 13	1.064 ± 0.014	> Mod
b-15 : 0 FA	M	OS-38321	−25.3	90 ± 9	1.029 ± 0.011	> Mod
16 : 1 FA	M	OS-38327	−25.4	81 ± 11	1.025 ± 0.011	> Mod
16 : 0 FA	M	OS-38333	−24.3	***146 ± 9***	**1.086 ± 0.011**	**> Mod**
18 : 1 FA	M	OS-38318	−23.2	***288 ± 11***	**1.228 ± 0.008**	**> Mod**
18 : 0 FA	M	OS-38320	−23.9	77 ± 11	1.028 ± 0.011	> Mod
24 + 26 + 28 FA	T	OS-38331	−27.0	1 ± 16	0.967 ± 0.014	265 ± 110
alkenones	M	OS-39539	−26.6	87 ± 14	1.099 ± 0.014	> Mod
24 + 26 + 28 HC	R	OS-39911	−29.3	−677 ± 10	0.325 ± 0.009	9030 ± 210
27 + 29 HC	T	OS-39908	−30.0	−181 ± 14	0.825 ± 0.010	1550 ± 100
27Δ^{5} sterol	M	OS-53936	−26.4	79 ± 11	1.046 ± 0.011	> Mod.
28$\Delta^{5,22}$ sterol	M	OS-53934	−26.1	65 ± 9	1.034 ± 0.009	> Mod.
30Δ^{22} sterol	M	OS-53957	−25.7	69 ± 15	1.040 ± 0.015	> Mod.
24 + 26 + 28 ROH	T	OS-53956	−30.1	−44 ± 12	0.925 ± 0.012	625 ± 110
			Black Sea Sediment			
OC		OS-32871	−25.3	−199 ± 6	0.806 ± 0.004	1740 ± 35
SLE		OS-38309	−27.9	−150 ± 15	0.856 ± 0.008	1250 ± 80
14 : 0 FA	M	OS-38630	−28.6	18 ± 15	0.957 ± 0.016	350 ± 130
b-15 : 0 FA	M	OS-38632	−30.5	−27 ± 14	0.919 ± 0.019	680 ± 170
16 : 1 FA	M	OS-38628	−31.9	75 ± 14	1.019 ± 0.013	> Mod
16 : 0 FA	M	OS-38627	−29.1	***214 ± 18***	**1.151 ± 0.018**	**> Mod**
b-17 : 0 FA	M	OS-38642	−30.5	57 ± 20	1.006 ± 0.020	> Mod
18 : 1 FA	M	OS-38637	−27.0	−24 ± 20	0.999± 0.020	5 ± 100
18 : 0 FA	M	OS-38636	−29.5	***−374 ± 27***	**1.310 ± 0.023**	**> Mod**
22 : 0 FA	M	OS-38639	−28.9	−46 ± 26	0.919± 0.027	680 ± 230
24 : 0 FA	M	OS-38640	−29.2	−34 ± 26	0.934 ± 0.024	550 ± 220
26 : 0 FA	T	OS-38641	−30.5	−223 ± 31	0.754 ± 0.021	2270 ± 230
alkenones	M	OS-39536	−28.6	45 ± 13	1.069± 0.013	> Mod
24 + 26 + 28 HC	R	OS-39909	−29.2	−609 ± 11	0.393 ± 0.007	7500 ± 150
27 HC	T	OS-39907	−30.0	−231 ± 8	0.774 ± 0.011	2060 ± 110
29 HC	T	OS-39906	−31.0	−125 ± 8	0.880 ± 0.008	1020 ± 75
27Δ^{5} sterol	M	OS-53948	−27.0	−33 ± 12	0.937 ± 0012	520 ± 100
30Δ^{22} sterol	M	OS-53943	−26.0	−15 ± 9	0.942 ± 0.011	475 ± 95
24 ROH	T	OS-53951	−29.7	−176 ± 8	0.796 ± 0.011	1840 ± 110
26 ROH	T	OS-53958	−31.2	−100 ± 16	0.871 ± 0.018	1100 ± 160

M = marine; T = terrigenous; R = relict
FA = fatty acid; ROH = alcohol; HC = alkane

Table 2. Stable carbon and radiocarbon isotope data for Arabian Sea POM and SOM.

	source	ID #	δ^{13}C (‰)	Δ^{14}C (‰)	f_m	Age
			Arabian Sea Trap			
OC		OS-32868	−22.4	14 ± 13	1.020 ± 0.013	> Mod
SLE		OS- 38314	−24.7	66 ± 14	1.073 ± 0.014	> Mod
14 : 0 FA	M	OS- 37311	−25.7	***178 ± 17***	**1.107 ± 0.017**	**> Mod**
b-15 : 0 FA	M	OS-37314	−23.2	56 ± 13	0.997 ± 0.014	25 ± 110
16 : 1 FA	M	OS-37304	−24.0	55 ± 21	0.999 ± 0.014	5 ± 100
16 : 0 FA	M	OS-37298	−23.8	97 ± 14	1.039 ± 0.013	> Mod
18 : 1 FA	M	OS-37297	−23.1	38 ± 21	0.990 ± 0.013	80 ± 100
18 : 0 FA	M	OS-37302	−23.8	111 ± 15	1.060 ± 0.015	> Mod
22 : 0 FA	M	OS-37305	−23.8	51 ± 14	1.012 ± 0.014	> Mod
24 : 0 FA	M	OS-37313	−24.7	69 ± 14	1.033 ± 0.013	> Mod
26 : 0 FA	M	OS-37315	−25.2	80 ± 16	1.047 ± 0.016	> Mod
alkenones	M	OS-39910	−23.9	−6 ± 9	1.000 ± 0.011	> Mod
24+26+28 HC	R	OS-55323	−28.5	−731 ± 14	0.270 ± 0.013	10500 ± 390
HBI HC	M	OS-55248	−24.5	−514 ± 10	**0.488 ± 0.010**	5750 ± 160
27 + 29 HC	T	OS-55325	−28.4	−320 ± 24	0.684 ± 0.024	3050 ± 280
$27\Delta^{5}$ sterol	M	OS-56344	−16.7	−32 ± 12	0.939 ± 0.012	505 ± 100
$28\Delta^{5,22}$ sterol	M	OS-56348	−27.1	−86 ± 13	0.888 ± 0.013	955 ± 120
16 ROH	M	OS-56347	−22.5	−91 ± 12	0.861 ± 0.012	1200 ± 110
			Arabian Sea Sediment			
OC		OS-32869	−20.8	−138 ± 2	0.867 ± 0.003	1140 ± 30
LE		OS-38322	−25.0	−173 ± 11	0.833 ± 0.011	1470 ± 100
14 : 0 FA	M	OS-38332	−26.6	−10 ± 13	0.931 ± 0.013	575 ± 110
b-15 : 0 FA	M	OS-38324	−24.5	−70 ± 11	0.878 ± 0.011	1040 ± 100
16 : 1 FA	M	OS-38313	−26.0	−112 ± 7	0.842 ± 0.007	1380 ± 65
16 : 0 FA	M	OS- 38329	−25.0	104 ± 12	1.046 ± 0.012	> Mod
18 : 1 FA	M	OS-38334	−24.8	−171 ± 13	0.791 ± 0.013	1880 ± 140
18 : 0 FA	M	OS-38325	−24.7	***190 ± 10***	**1.135 ± 0.010**	**> Mod**
22 : 0 FA	M	OS-38326	−26.8	103 ± 12	0.864 ± 0.012	1180 ± 110
24 : 0 FA	M	OS-38317	−25.4	−91 ± 7	0.879 ± 0.006	1040 ± 55
26 : 0 FA	M	OS-38319	−24.7	−116 ± 7	0.858 ± 0.007	1230 ± 70
alkenones	M	OS-39902	−24.1	−202 ± 7	0.803 ± 0.003	1760 ± 65
24 + 26 + 28 HC	R	OS-55329	−27.6	−805 ± 9	0.197 ± 0.008	13050 ± 340
HBI HC	M	OS-56341	−18.2	***−256 ± 8***	**0.748 ± 0.008**	***2330 ± 85***
15+16+17+18 HC	R	OS-55251	−29.4	−887 ± 5	0.114 ± 0.004	17500 ± 250
27 + 29 HC	T	OS-55318	−27.7	−430 ± 11	0.573 ± 0.011	4470 ± 160
$27\Delta^{5}$ sterol	M	OS-56349	−24.0	−152 ± 13	0.822 ± 0.013	1570 ± 130
16 ROH	M	OS-56351	−23.3	−99 ± 13	0.853 ± 0.012	1270 ± 110
26 + 28 + 30 ROH	T	OS-56350	−24.1	−113 ± 14	0.861 ± 0.012	1200 ± 110

M = marine; T = terrigenous; R = relict
FA = fatty acid; ROH = alcohol; HC = alkane

Table 3. Stable carbon and radiocarbon isotope data for Ross Sea POM and SOM.

	source	ID #	δ^{13}C (‰)	Δ^{14}C (‰)	f_m	Age
Ross Sea Trap						
OC		OS-32872	−28.0	−208 ± 6	0.797 ± 0.004	1820 ± 40
SLE		OS-38330	−32.7	−154 ± 15	0.852 ± 0.015	1290 ± 140
14 : 0 FA	M	OS-38626	−34.9	−115 ± 14	0.831 ± 0.012	1490 ± 110
16 : 0 FA	M	OS-38624	−31.7	−100 ± 10	0.853 ± 0010	1270 ± 95
18 : 0 FA	M	OS-39272	−35.3	−105 ± 15	0.854 ± 0.015	1270 ± 140
22 : 0 + 24 : 0 FA	M	OS-38635	−31.8	−175 ± 19	0.796 ± 0.019	1830 ± 190
27Δ^5 sterol	M	OS-50105	−32.0	−216 ± 7	0.761 ± 0.007	2190 ± 75
28$\Delta^{5,22}$ sterol	M	OS-50107	−35.4	−180 ± 8	0.796 ± 0.007	1830 ± 70
14 + 16 ROH	M	OS-50100	−31.8	−191 ± 9	0.764 ± 0.008	2160 ± 80
Ross Sea Sediment						
OC		OS-32873	−27.9	−355 ± 3	0.649 ± 0.003	3480 ± 35
SLE		OS-38323	−30.0	−211 ± 18	0.795 ± 0.018	1850 ± 180
14 : 0 FA	M	OS-38633	−36.6	−83 ± 5	0.862 ± 0.015	1190 ± 140
br-15 : 0 FA	M	OS-38625	−32.3	−128 ± 12	0.824 ± 0.012	1560 ± 120
16 : 0 FA	M	OS-39266	−32.5	***430 ± 11***	**1.439 ± 0.011**	**> Mod**
18 : 0 FA	M	OS-38644	−31.0	***189 ± 29***	**1.196 ± 0.029**	**> Mod**
24 : 0 FA	M	OS-38634	−33.5	−208 ± 22	0.765 ± 0.023	2150 ± 240
26 : 0 FA	M	OS-38645	−30.3	−302 ± 27	0.677 ± 0.029	3130 ± 340
27Δ^5 sterol	M	OS-50108	−33.0	−178 ± 10	0.798 ± 0.010	1810 ± 95
28$\Delta^{5,22}$ sterol	M	OS-50106	−34.3	−202 ± 8	0.775 ± 0.007	2050 ± 70

FA = fatty acid; ROH = alcohol

Table 4. Abundance weighted mean table carbon and radiocarbon isotope values for composited marine, terrigenous, and relict biomarkers.

	δ^{13}C (‰) ± s.d.	Δ^{14}C (‰) ± s.d.	f_m ± s.d.	Age ± s.d.	n
BS trap					
marine	−25.3 ± 1.1	78± 9	1.110 ± 0.066	> Mod	7
terrigenous	−28.7 ± 1.6	−75 ± 94	0.930 ± 0.043	580 ± 360	3
relict	−29.1	−677 ± 9	0.325 ± 0.009	9030 ± 220	1
BS sediment					
marine	−28.8 ± 1.8	−30 ± 10	0.970 ± 0.023	240 ± 56	6
terrigenous	−30.5 ± 0.7	−171 ± 58	0.833 ± 0.066	1470 ± 615	5
relict	−29.2	−609 ± 8	0.393 ± 0.007	7500 ± 150	1
AS trap					
marine	−23.5 ± 2.5	64 ± 20	1.071 ± 0.019	> Mod	7
terrigenous	−26.8 ± 2.3	−320 ± 24	0.684 ± 0.024	3050 ± 280	2,1
relict	−28.5	−731 ± 13	0.270 ± 0.013	10 140 ± 360	1
AS sediment					
marine	−24.5 ± 2.1	−63 ± 110	0.941 ± 0.110	490 ± 1440	8
terrigenous	−27.7	−430 ± 13	0.573 ± 0.011	4470 ± 150	1
relict	−28.5 ± 1.2	−846 ± 58	0.114 ± 0.004	17 440 ± 270	1
RS trap					
marine	−33.3 ± 1.8	−155 ± 47	0.850 ± 0.045	1310 ± 420	7
terrigenous	nd*	nd*	nd*	nd*	nd*
relict	nd*	nd*	nd*	nd*	nd*
RS sediment					
marine	−33.2 ± 1.9	−105 ± 31	0.900 ± 0.030	850 ± 270	6
terrigenous	−31.8 ± 2.8	−255 ± 66	0.750 ± 0.056	2310 ± 560	2
relict	nd*	nd*	nd*	nd*	nd*

nd* not determined

Table 5. Isotopic values used in mass balance calculations. The values of f_M, f_T, and f_R are those values calculated assuming that the actual value of $\Delta^{14}C_{add}$ is that measured on the compounds defined as relict.

Sample	$\Delta^{14}C_{bulk}$ (‰)	$\Delta^{14}C_M$ (‰)	$\Delta^{14}C_T$ (‰)	$\Delta^{14}C_{add}$ (‰)	f_M	f_T	f_R
BS trap	−100	80	−75	−677	0.76	0.08	0.15
BS sed	−150	−30	−171	−609	0.79	0.1	0.11
AS trap	64	64	−320	−731	1	0	0
AS sed	−173	−63	−430	−846	0.86	0.04	0.1
RS sed	−210	−105	−255				

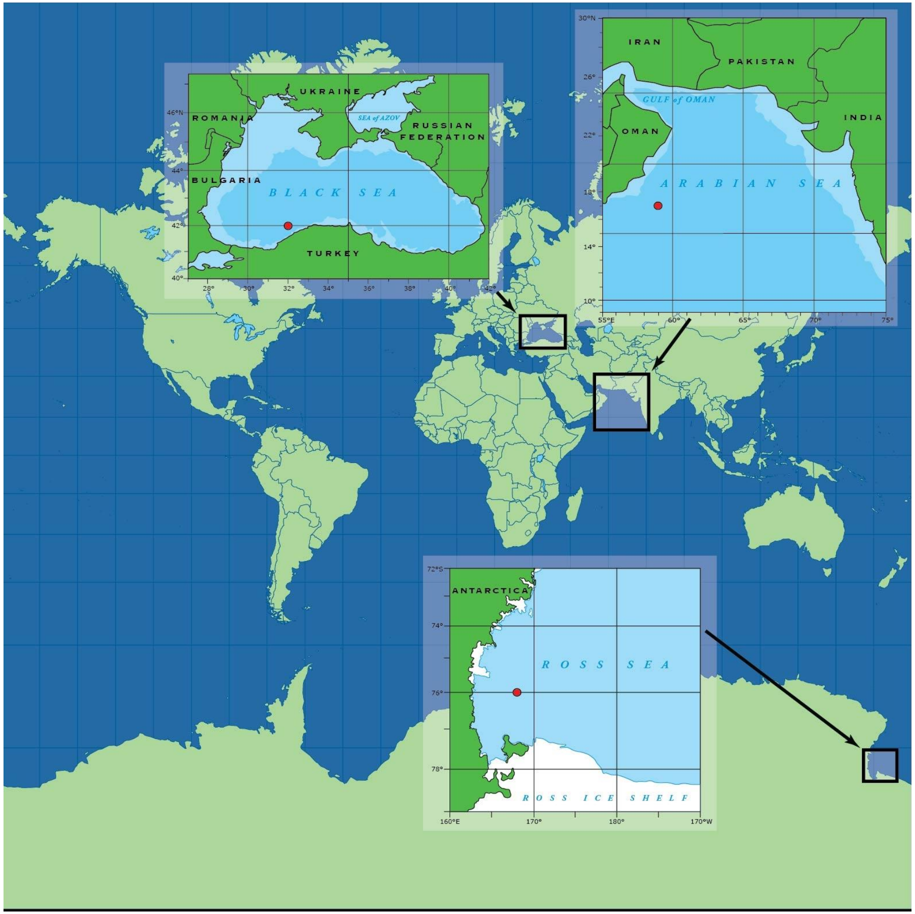

Figure 1. Sampling locations in the Black Sea, Arabian Sea and Ross Sea.

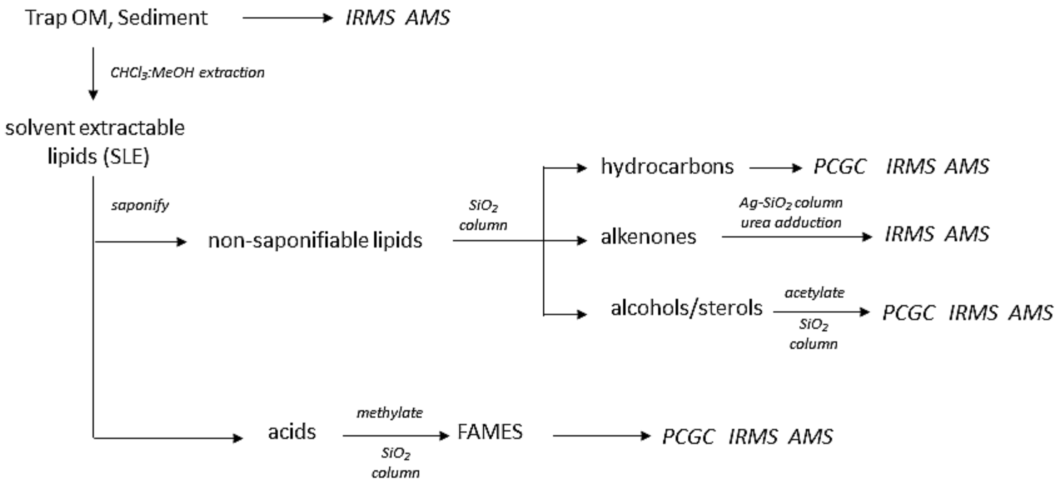

Figure 2. Scheme of extraction, isolation and analysis of biomarkers.

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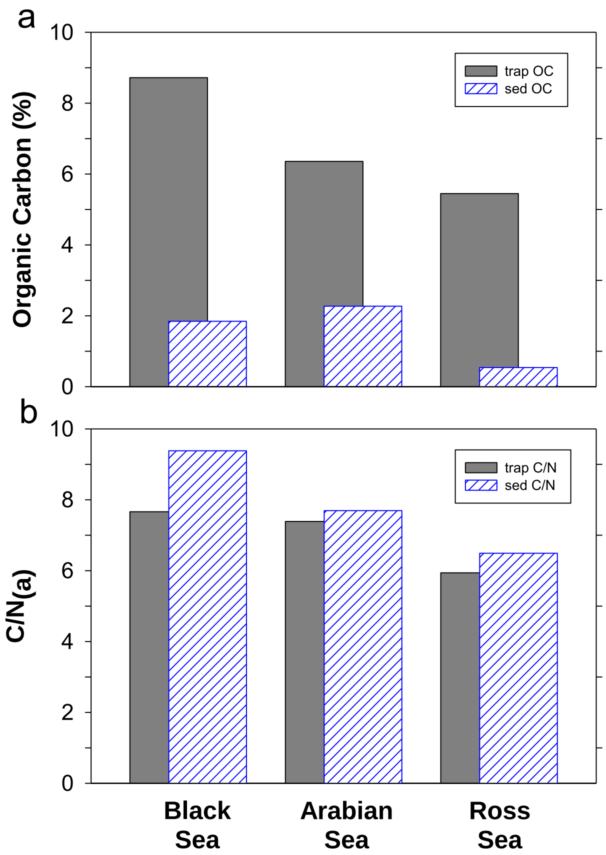

Figure 3. (a) Organic carbon (% OC) and **(b)** C/N(a) for bulk trap and sediments.

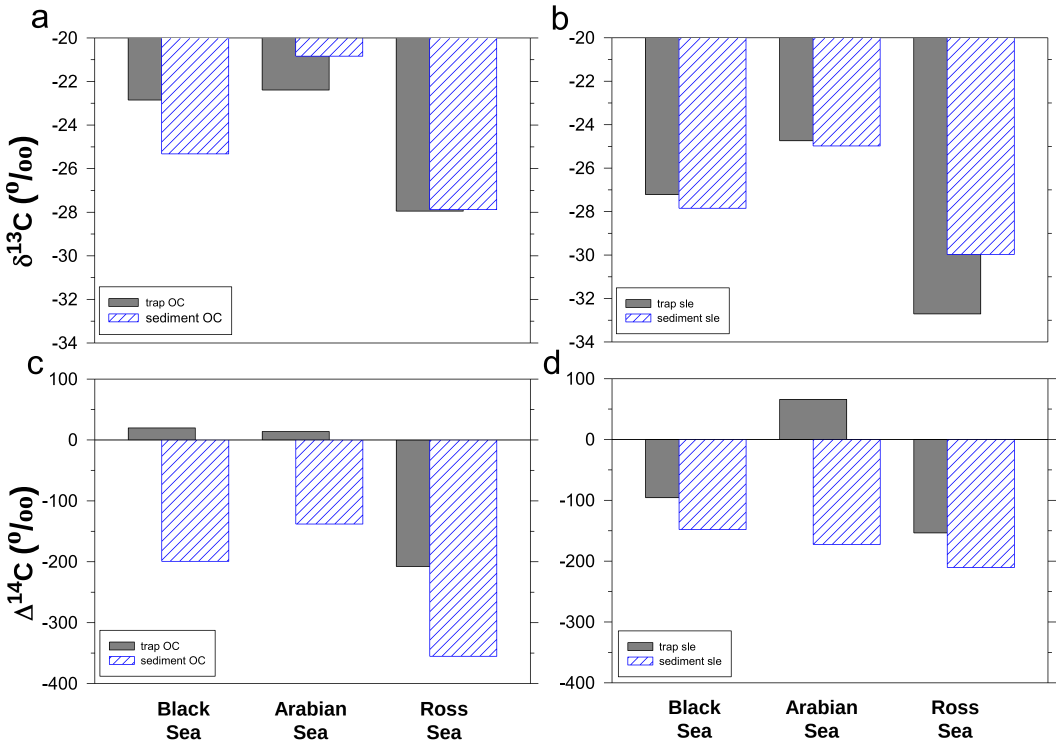

Figure 4. (a) The δ^{13}C and **(b)** Δ^{14}C values of bulk organic carbon (OC) and **(c)** δ^{13}C and **(d)** Δ^{14}C values solvent extractable lipids (SLE) for trap POM and sediments. All values have been corrected for procedural blanks and any derivative carbon.

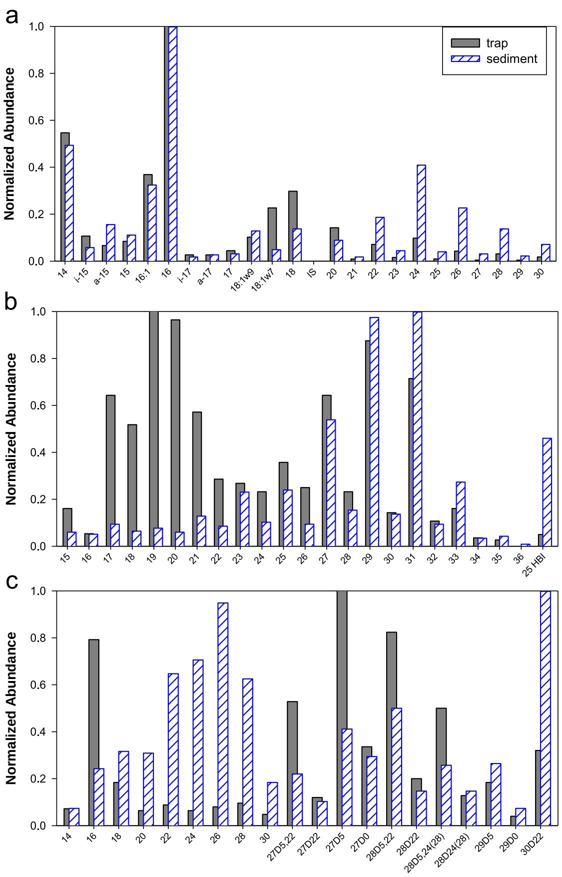

Figure 5. Histograms of relative abundances of **(a)** fatty acids, **(b)** hydrocarbons, and **(c)** sterols/alcohols POM and SOM from the Black Sea. Carbon numbers are given for fatty acids, alkanes, and alcohols; sterol abbreviations are 27D5,22 = $27\Delta^{5,22}$, etc.

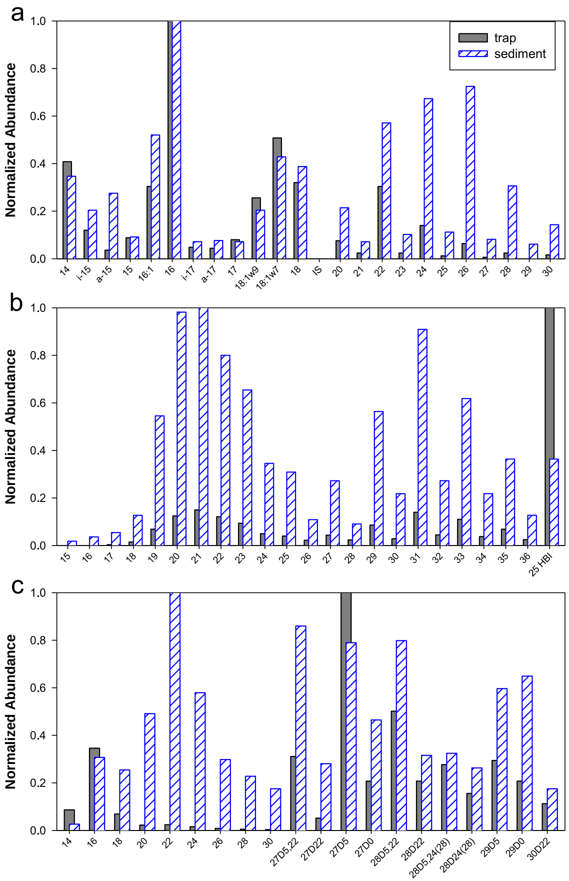

Figure 6. Histograms of relative abundances of **(a)** fatty acids, **(b)** hydrocarbons, and **(c)** sterols/alcohols in POM and SOM from the Arabian Sea. Carbon numbers are given for fatty acids, alkanes, and alcohols; sterol abbreviations are 27D5,22 = $27\Delta^{5,22}$, etc.

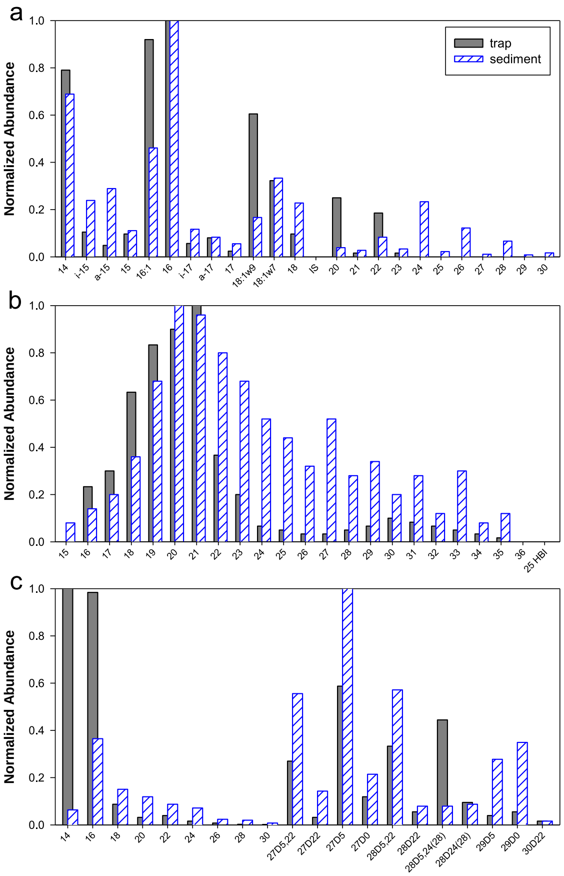

Figure 7. Histograms of relative abundances of **(a)** fatty acids, **(b)** hydrocarbons, and **(c)** sterols/alcohols in POM and SOM from the Ross Sea. Carbon numbers are given for fatty acids, alkanes, and alcohols; sterol abbreviations are 27D5,22 = $27\Delta^{5,22}$, etc.

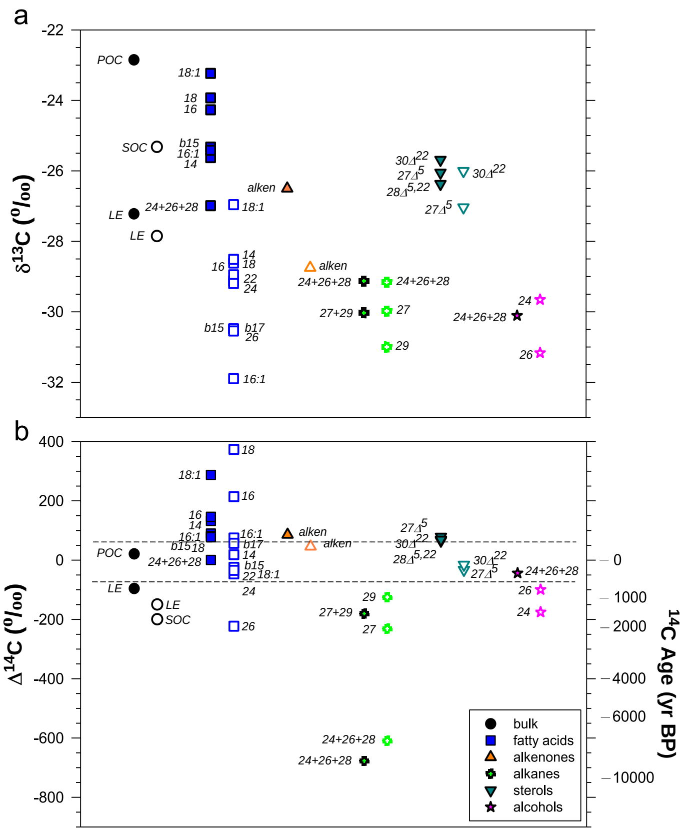

Figure 8. The **(a)** δ^{13}C and **(b)** Δ^{14}C values of bulk POC and SOC and individual biomarkers for trap and sediments from the Black Sea (see also Table 1). Filled symbols are trap biomarkers; open symbols are sediment biomarkers. All values have been corrected for procedural blanks and any derivative carbon. Dashed lines are pre-bomb (lower) and post-bomb (upper) mixed-layer $\Delta^{14}C_{DIC}$.

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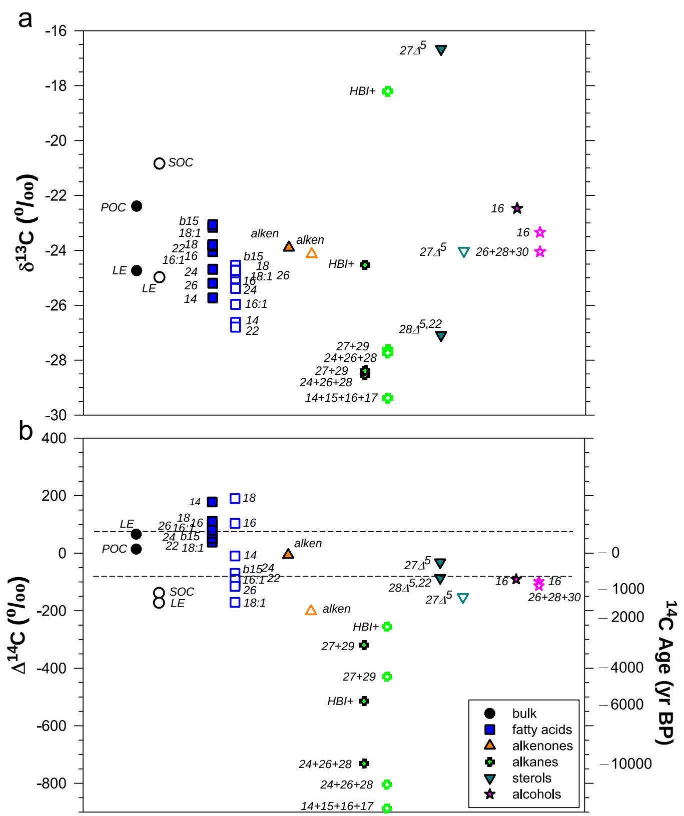

Figure 9. The **(a)** δ^{13}C and **(b)** Δ^{14}C values of bulk POC and SOC and individual biomarkers for trap and sediments from the Arabian Sea (see also Table 2). Filled symbols are trap biomarkers; open symbols are sediment biomarkers. All values have been corrected for procedural blanks and any derivative carbon. Dashed lines are pre-bomb (lower) and post-bomb (upper) mixed-layer $\Delta^{14}C_{DIC}$.

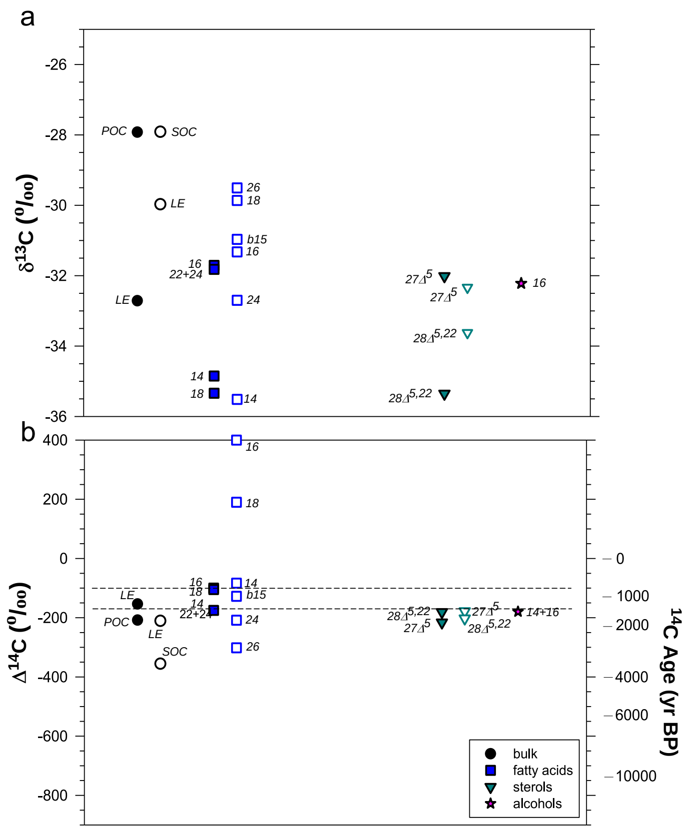

Figure 10. The **(a)** δ^{13}C and **(b)** Δ^{14}C values of bulk POC and SOC and individual biomarkers for trap and sediments from the Ross Sea (see also Table 3). Filled symbols are trap biomarkers; open symbols are sediment biomarkers. All values have been corrected for procedural blanks and any derivative carbon. Dashed lines are pre-bomb (lower) and post-bomb (upper) mixed-layer $\Delta^{14}C_{DIC}$.

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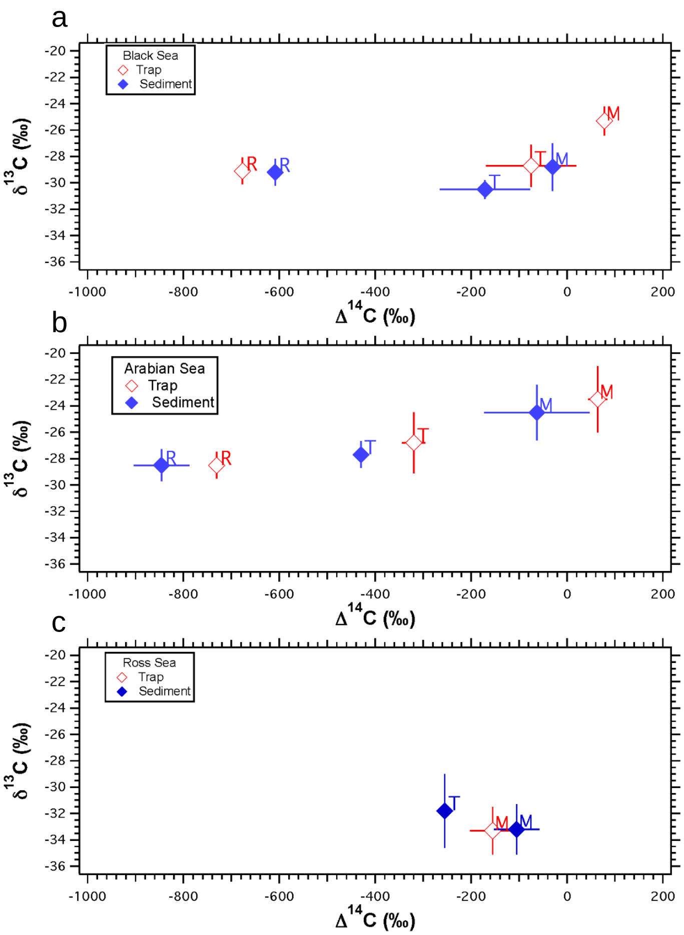

Figure 11. Concentration weighted average $\delta^{13}C$ and $\Delta^{14}C$ values of marine (M), terrigenous (T) and relict (R) lipids in **(a, b)** Black Sea, **(c, d)** Arabian Sea, and **(e)** Ross Sea traps and sediments.

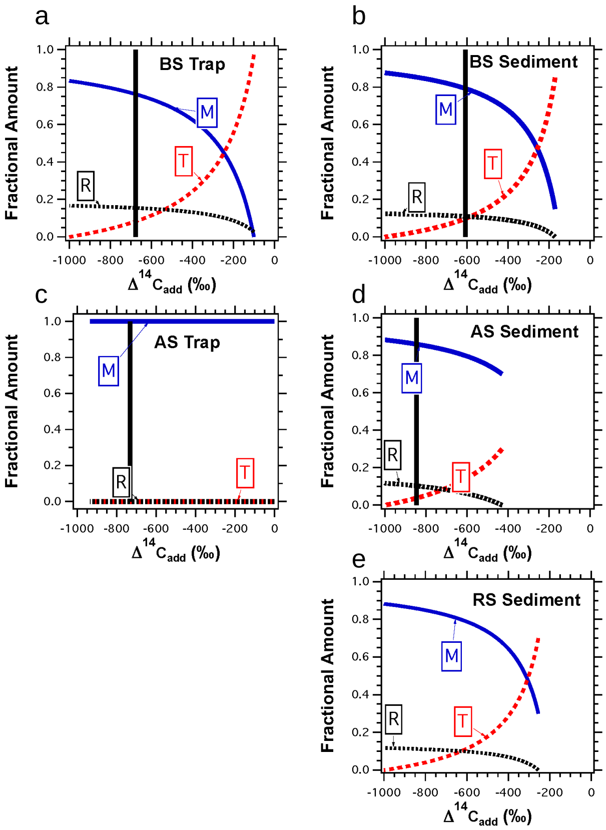

Figure 12. Relative amounts of marine (M, blue solid line), terrestrial (T, red dashed line) and relict (R, black small-dashed line) carbon as a function of $\Delta^{14}C_{add}$ in Black Sea, Arabian Sea, and Ross Sea trap (**a** and **c**, respectively) and sediment material (**b**, **d**, and **e**, respectively). Only real solutions are depicted. The solid black line indicates the solution when $\Delta^{14}C_{add}$ is equal to the value measured on compounds defined as relict. There are not enough data to construct a graph for the Ross Sea sediment trap.

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