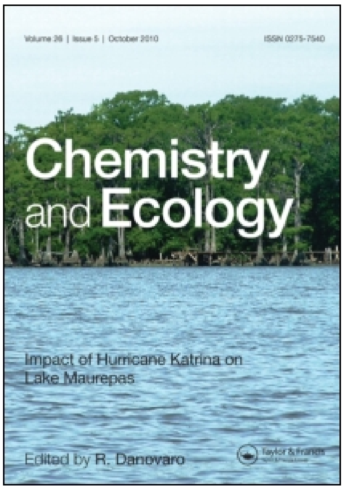

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ORGANIC MATTER COMPOSITION IN SEDIMENTS OF THE OMAN MARGIN

RENGASAMY ALAGARSAMY*

Chemical Oceanography Division, National Institute of Oceanography,
Donapaula, Goa-403 004, India

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The quantity and biochemical composition of organic matter (OM) in the sediments underlying and below oxygen minimum zone (OMZ) in the Arabian Sea were studied to provide information on the diagenetic processes of organic carbon under different environmental conditions. Concentrations of total organic carbon (TOC), total nitrogen (TN) and total hydrolysable amino acids (THAA) were significantly higher in sediments within rather than below OMZ, while those of total carbohydrates (TCHO) were slightly lower in the latter, suggesting the presence of a larger supply of labile compounds into the sea bottom at the shallower site (*i.e.* within OMZ). Hydrolysable amino acid and carbohydrate contribution to TOC were even lower (about 10% in surficial sediments) than the values obtained from the abyssal oligotrophic North Pacific, suggesting that OM food availability in the Oman Margin sediments within OMZ was lower than that observed at abyssal depths. The presence of the highest THAA and TCHO concentrations in the top 40 mm of the sediment core at both sites reflected the presence of bioturbation processes. In contrast with the general view of the deep sea as a stable and constant system, below OMZ in the Arabian Sea sediments, some differences were observed in the two investigated cores indicating the presence of a certain spatial variability in OM content and diagenesis.

Keywords: Sediments; Oxygen minimum zone; Carbon; Nitrogen; Amino acids; Carbohydrates

1 INTRODUCTION

The Arabian Sea is one of the most productive areas of world oceans and has attracted a number of physical, chemical and biological studies because of seasonal fluctuations in primary productivity due to monsoon dynamics. The seasonal reversal of both Southwest (SW) and northeast (NE) winds induces upwelling of nutrient-rich deep waters along the narrow continental shelf, resulting in enhanced surface productivity and particle flux export from the euphotic zone (Qasim, 1982; Sen Gupta and Naqvi, 1984; Nair *et al.*, 1989). A strong regional and temporal variability, related to the varying intensity of monsoons, can be observed in the vertical fluxes of particulate organic carbon towards the deep ocean (Haake *et al.*, 1993; Rixen *et al.*, 1996). Primary productivity and particulate fluxes into the deep Arabian Sea are largely controlled by the monsoon system (Witte and Pfannkuche, 2000). The high primary organic matter (OM) production in the surface water layers of the Arabian Sea and the limited water renewal may result in an over consumption of dissolved oxygen, followed by the development

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* Fax: 91-832-2450602/03; E-mail: alagar@darya.nio.org

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of an intense and unusual deep oxygen minimum zone (OMZ). Such phenomenon depresses OM recycling in mid waters, allowing a large amount of detrital material sinking into deeper depths largely undegraded, and contributing to the enhancement of the settlement rates of more labile organic material into the deep sea floor.

Several factors may control OM composition and diagenesis in marine sediments (Pedersen *et al.*, 1992). As the sedimentary record may provide the key to understanding carbon cycling in the whole Arabian Sea, some studies have been carried out in this region in order to assess factors controlling organic carbon preservation. However, the nature, biochemical composition and degradation rates of sediment OM in this area are still far from being completely assessed (Hedges and Keil, 1995). The OM origin, composition and processes controlling its degradation patterns may provide fundamental information dealing with the actual quantity of OM preserved in deep-sea sediments.

Currently, there is a strong debate over the processes that are potentially able to control the preservation and burial of organic carbon in marine sediments, especially in oceanic areas characterized by high OM inputs, such as the Arabian Sea. Among these, primary productivity, vertical fluxes, sedimentation and degradation rates of particulate organic carbon and bottom-water oxygen concentrations have been invoked as key factors controlling organic carbon preservation and burial in deep-sea sediments (Hartnett *et al.*, 1998). In the Oman Margin, apparently, oxygen does not play a key role in organic carbon burial (Pedersen *et al.*, 1992) and, despite the high amount of deposited OM, rates of sulphate reduction in the sediments underlying the OMZ are very low. Such discrepancy may reflect the presence of a relatively low amount of labile material and suggests that OM arriving at the sediment–water interface is mostly composed of refractory compounds (Pedersen and Shimmield, 1991).

Organic matter in the marine environment consists of labile and refractory compounds. The labile fraction of OM is composed of simple monomers and/or biopolymers such as proteins, carbohydrates and lipids, which may be rapidly mineralised. The refractory fraction of OM consists of structural compounds such as humic and fulvic acids, which are more slowly broken down (Henrichs, 1992; Fabiano and Danovaro, 1994). Only the labile fraction of OM undergoes early diagenesis whereas the refractory one accumulates in the sediments (Hedges and Keil, 1995).

The objectives of the present study were to investigate the quantity and biochemical composition of OM in the sediments underlying and below the OMZ in the Arabian Sea, in order to provide a proxy of diagenetic processes of organic carbon under different environmental conditions.

2 MATERIALS AND METHODS

2.1 Environmental Characteristics of the Study Area

Bottom-water oxygen concentrations in the deep Arabian Sea region under investigation generally range from ~0.13 ml l^{-1} at 400 m (see Levin *et al.*, 2000) to ~2.99 ml l^{-1} at 3400 m depth (Gage, 1995; Smith *et al.*, 2000). In these sediments, oxygen depletes rapidly, reaching values close to ~0.13 ml l^{-1} within the first millimeters of the sediments, which is entirely reduced below this depth. Sediments within the OMZ were muddy (mean particle diameter 29 μm) and displayed a mixing layer at 20–40 mm, as indicated by the vertical profiles of ^{210}Pb contents (Smith *et al.*, 2000). In spite of the low-oxygen concentrations at the sediment–water interface, macrofaunal abundance is surprisingly high there (12,362 ind m^{-2}, largely dominated by spionid and cirratulids polychaetes), suggesting that the mixing of surficial sediments might be ascribed to bioturbation (Smith *et al.*, 2000).

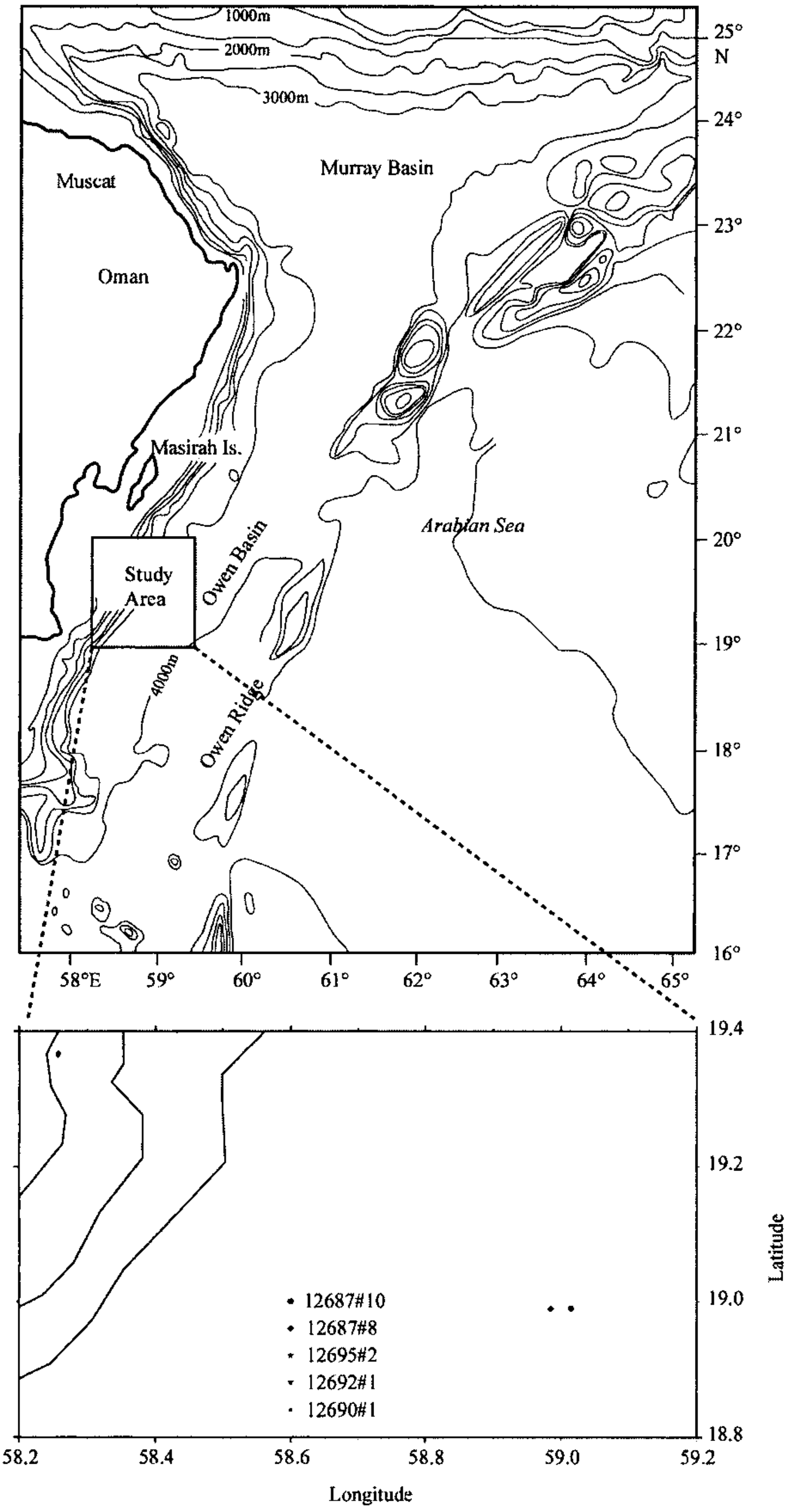

FIGURE 1 Map of sampling sites during *Discovery Cruise 211* in the Arabian Sea.

TABLE I Sampling Stations Location, Sediment Type, Depth, Core Length and Date of Collection.

Station	*Location*	*Sediment type*	*Water depth (m)*	*Core length (mm)*	*Date*
12690#1	19°22.00′ N 58°15.43′ E	Within OMZ	377	130–150	22/10/94
12692#1	19°21.83′ N 58°15.42′ E	Within OMZ	372	130–150	22/10/94
12695#2	19°22.07′ N 58°15.43′ E	Within OMZ	382	130–150	22/10/94
12687#8	18°59.33′ N 58°59.09′ E	Below OMZ	3372	270–290	20/10/94
12687#10	18°59.42′ N 59°00.92′ E	Below OMZ	3385	310–330	20/10/94

2.2 Sampling

Undisturbed sediment samples were collected using a multi-corer from separate multiple-corer deployments (Barnett *et al.*, 1984) within and below the OMZ during *RRS Discovery cruise 211/94* in the Arabian Sea (Fig. 1). Station coordinates are reported in Table I. Sediment samples were immediately frozen (−20 °C) on board, and then freeze-dried in the laboratory. The dried samples were stored at −20 °C until analysis. Frozen cores collected within the OMZ area (12690#1, 12692#1 and 12695#2) were sliced at 5 mm intervals down to 10 mm depth; and at 10 mm intervals between 10 and 70 mm depth; and 20 mm intervals between 70 and 150 mm collected within OMZ site. The sediment cores collected below the OMZ area (cores 12687#8 and 12687#10; Tab. I) were sliced at 10 mm intervals down to 70 mm; and at 20 mm intervals between 70 and 290–330 mm depth, respectively.

2.3 Elemental and Biochemical Analyses

Total organic carbon (TOC) and total nitrogen (TN) contents were determined by a CHN analyser (Carlo Erba 1106) according to Hedges and Stern (1984). The determinations of total hydrolysable amino acids (THAA) and total hydrolysable carbohydrates (TCHO) were carried out according to Patience *et al.* (1990). Coefficient of variation of replicate analyses ($n = 4$) of a homogenised sample was ±6% for TOC and TN; ±2% for THAA ($n = 10$) and ±8% for TCHO ($n = 6$). The contribution of THAA and TCHO carbon (THAA-C and TCHO-C) to TOC were calculated according to Cowie and Hedges (1994).

2.4 Statistical Analyses

Vertical and spatial changes in organic compounds sediment content were investigated separately by means of a one-way analysis of variance (ANOVA).

3 RESULTS

3.1 Total Organic Carbon and Nitrogen

Organic matter concentrations in the sediments of the Oman Margin are reported in Table II. In core 12690#1, TOC concentrations increased slightly with depth in the sediment down to ~50 mm (from ~5 to 7% in the top sediment layer and at 50 mm depth, respectively) then

TABLE II Total Organic Carbon (TOC), Total Nitrogen (TN), Total Hydrolysable Amino Acids (THAA) and Total Carbohydrates (TCHO) in the Cores Collected from OMZ and Below the OMZ in the Oman Margin of the Arabian Sea.

Core	*Depth (mm)*	*TOC (%)*	*TN (%)*	*C:N (w/w)*	*THAA* $(mg\,g^{-1})$	*TCHO* $(mg\,g^{-1})$
12690#1	0–5	4.71	0.59	8.0	17.3	2.4
	0–10	6.02	0.78	7.8	10.0	3.4
	10–20	6.64	0.79	8.4	12.1	4.9
	20–30	6.85	0.52	13.2	18.5	5.9
	30–40	6.92	0.81	8.5	14.0	5.4
	40–50	7.49	0.90	8.3	12.6	6.7
	50–60	5.30	0.62	8.6	13.4	4.6
	60–70	5.30	0.62	8.6	11.1	3.0
	70–90	4.25	0.50	8.5	11.7	5.7
	90–110	5.89	0.69	8.5	16.2	4.6
	110–130	6.13	0.71	8.7	14.6	4.4
	130–150	4.88	0.57	8.6	11.2	3.9
12692#1	0–5	6.49	0.82	7.9	13.4	–
	5–10	4.94	0.63	7.8	17.8	–
	10–20	7.25	0.89	8.2	12.6	–
	20–30	7.00	0.83	8.4	19.5	–
	30–40	5.83	0.67	8.7	13.5	–
	40–50	6.52	0.75	8.7	13.1	–
	50–60	7.32	0.99	7.3	12.4	–
	60–70	7.29	0.88	8.3	10.0	–
	70–90	7.10	0.81	8.8	14.6	–
	90–110	6.61	0.79	8.4	10.7	–
	110–130	5.45	0.67	8.2	9.2	–
	130–150	6.57	0.53	12.5	15.1	–
12695#1	0–5	4.96	0.64	7.8	13.8	–
	5–10	6.14	0.81	7.6	10.7	–
	10–20	4.35	0.55	7.9	11.9	–
	20–30	6.00	0.69	8.7	18.4	–
	30–40	5.57	0.67	8.3	10.0	–
	40–50	4.86	0.55	8.8	11.1	–
	50–60	4.68	0.49	9.5	9.0	–
	60–70	6.17	0.70	8.8	10.0	–
	70–90	4.54	0.58	7.9	11.1	–
	90–110	5.75	0.68	8.4	13.8	–
	110–130	6.33	0.74	8.6	9.2	–
	130–150	4.40	0.47	9.4	9.6	–
12687#8	0–10	2.16	0.27	7.9	7.5	4.4
	10–20	1.95	0.23	8.6	3.2	4.6
	20–30	2.24	0.28	8.1	5.6	5.1
	30–40	2.22	0.27	8.2	6.5	5.0
	40–50	2.27	0.27	8.5	6.7	4.7
	50–60	2.54	0.30	8.5	4.4	4.9
	60–70	2.33	0.27	8.5	3.8	3.5
	70–90	3.11	0.35	8.9	5.7	4.9
	90–110	3.34	0.35	9.5	4.8	4.5
	110–130	0.19	0.02	11.9	0.5	0.4
	130–150	0.16	0.01	14.7	0.3	0.5
	150–170	0.34	0.03	12.2	0.4	0.7
	170–190	0.36	0.03	10.8	0.3	0.6
	190–210	1.78	0.22	8.2	1.3	3.2
	210–230	1.61	0.19	8.5	1.1	2.7
	230–250	1.58	0.19	8.4	1.8	2.7
	250–270	1.87	0.23	8.1	1.4	2.1
	270–290	1.56	0.17	9.2	0.7	1.5

(*Continued*)

TABLE II *Continued.*

Core	*Depth (mm)*	*TOC (%)*	*TN (%)*	*C:N (w/w)*	*THAA ($mg\,g^{-1}$)*	*TCHO ($mg\,g^{-1}$)*
12687#10	0–10	2.68	0.35	7.8	1.8	2.1
	10–20	2.40	0.29	8.2	8.3	5.2
	20–30	2.33	0.28	8.4	5.3	5.2
	30–40	2.96	0.34	8.7	3.7	5.1
	40–50	2.71	0.30	8.9	6.5	4.3
	50–60	2.99	0.33	8.9	4.7	5.0
	60–70	2.95	0.33	8.9	3.9	6.0
	70–90	3.89	0.42	9.2	6.6	5.8
	90–110	4.42	0.45	9.8	5.0	5.9
	110–130	1.83	0.17	10.6	3.4	3.4
	130–150	0.18	0.02	9.3	0.7	0.7
	150–170	0.38	0.04	10.9	0.8	0.5
	170–190	0.20	0.02	10.1	0.4	0.5
	190–210	0.72	0.05	10.1	1.2	0.6
	210–230	0.98	0.11	8.7	2.3	1.4
	230–250	1.83	0.22	8.4	3.5	2.1
	250–270	1.59	0.22	8.2	2.3	1.9
	270–290	1.41	0.17	8.4	2.8	1.9
	290–310	1.54	0.18	8.7	2.3	2.2
	310–330	1.59	0.18	9.1	3.3	2.1

roughly decreased down-core (~4% in 70–90 mm layer). The average TOC concentrations below 100 mm (~5%) were slightly lower (Fig. 2) than those above 50 mm (~6%). Total nitrogen concentrations generally increased with depth in the sediment down to ~50 mm (from ~0.6 to 0.9% in the top sediment layer and at 50 mm depth, respectively) then slightly fluctuated down-core. The C/N values (core 12690#1) were rather constant (~8), except a maximum (~13) in the 20–30 mm layer. In core 12692#1, the average TOC concentration below 100 mm (~7%) was significantly (ANOVA, $p < 0.001$) higher than above 50 mm (~6%), whereas in core 12695#2, the TOC concentrations were rather constant along the entire core. The C/N values showed similar vertical patterns as TOC in both cores. Total nitrogen concentrations generally decreased with depth in the sediment (12692#1 and 12695#2).

The TOC contents of deeper cores (12678#8 and 12687#10) varied from <1 to 4% and showed significant changes with depth in the sediment (ANOVA, $p < 0.001$) (Fig. 2). Values of the C/N ratio in both cores showed a significant (ANOVA, $p < 0.05$) increase (from ~8 to 10) from the top layer down to 50 mm depth (Fig. 2). Highest values of the C/N ratios occurred at ~150 mm depth (~10–12, Tab. II). The TN content of deeper cores (12678#8 and 12687#10) displayed a similar sediment depth-related pattern as TOC (Fig. 2).

3.2 Hydrolysable Amino Acids and Total Carbohydrates

In core 12690#1 (within OMZ), TCHO concentration were significantly (ANOVA, $p < 0.001$) lower than those of THAA (Tab. II). The THAA concentrations displayed changes with depth in the sediment in all investigated cores (ANOVA, $p < 0.001$). Extremely high TCHO concentrations (up to 19 $mg\,g^{-1}$ at 20–30 mm) were observed in all the investigated cores. Concentrations of THAA were similar (at all depth, $p < 0.001$) in both cores collected below OMZ (Tab. II) and showed vertical patterns with depth in the sediment similar to those observed for TOC. In these sediments, the highest concentrations of THAA were found in

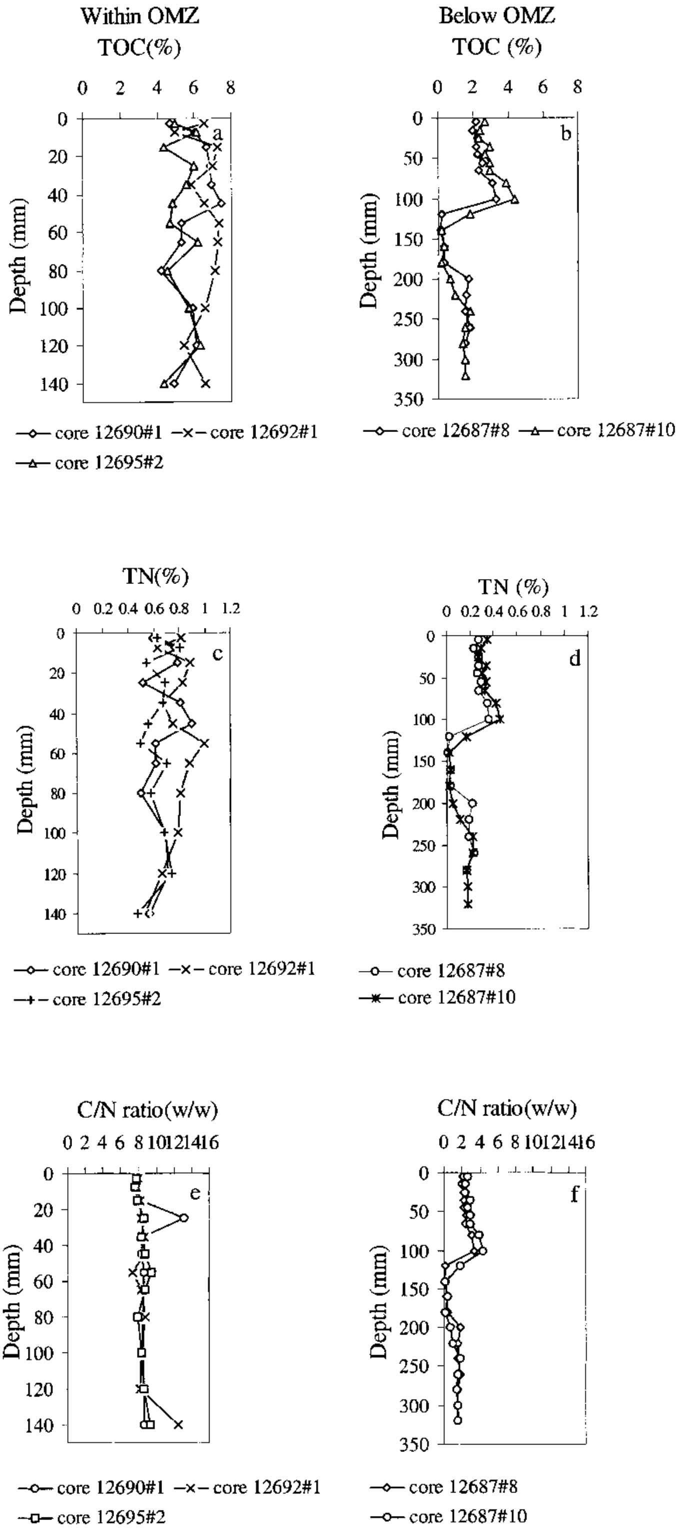

FIGURE 2 Down core variation in TOC (a, b), TN (c, d) and C/N ratio values (e, f) in the sediments within and below the OMZ in the Arabian Sea.

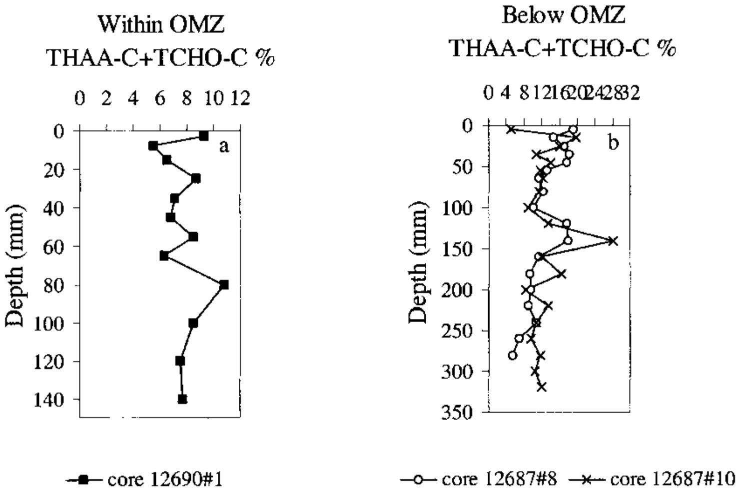

FIGURE 3 Down core variation in the contribution of total hydrolysable amino acids plus total carbohydrates (THAA-C + TCHO-C) to TOC within (core 12690#1, a) and below (cores 12687#8 and 12687#10, b) OMZ in the Arabian Sea.

0–10 mm and 90–110 mm (7.5 and 4.8 mg g^{-1}, respectively) and the lowest in 170–190 mm and 110–130 mm (0.3–0.5 mg g^{-1}, respectively).

The analysis of the vertical patterns of the contribution of THAA plus TCHO carbon equivalents to TOC% indicate that there was down-core variability in this parameter in both sediments within (0–5 mm, ~9%) and below (0–10 mm, ~19%) OMZ, with a general decreasing trend with depth in the sediment (Fig. 3). The TCHO concentration both within and below OMZ sediments changed significantly with depth in the sediment (ANOVA, $p < 0.001$) when compared to THAA contents. Total hydrolysable amino acids displayed the highest values in 0–10 mm, 7.5 mg g^{-1} and 10–20 mm, 8.3 mg g^{-1} in core 12687#8 and 12687#10, respectively.

4 DISCUSSION

4.1 Origin, Composition and Digenetic Patterns of Organic Matter Within OMZ

Concentrations of TOC, TN and THAA were higher in sediments within rather than below OMZ, while those of TCHO were slightly lower in the latter (Tab. II), suggesting the presence of a larger supply of labile compounds to the sediments at the shallower site (*i.e.* within OMZ).

While TOC mean concentrations in sediments within OMZ was similar in all the sampled cores, in only one of the three cores within OMZ (*i.e.* 12690#1) there was a down-core increase in TOC content from 5% at the sediment surface to 7% at 40–50 mm depth. In sediments within OMZ, the TN concentrations ranged from ~0.5 to 0.9% (Tab. II) and, generally, correlated with the TOC content (core 12690#1, $r = 0.86$; core 12692#1, $r = 0.85$; core 12695#1, $r = 0.95$; ANOVA, $p < 0.001$). Within OMZ, sediments were also characterized by C/N values (~8) typical of relatively 'fresh' marine-derived OM (*e.g.* Jasper and Gagosian, 1990; Macdonald *et al.*, 1991; Cowie and Hedges, 1994; Smallwood and Wolff, 2000; Suthhof *et al.*, 2000), but only two of the three cores (12692#1 and 12695#2) showed an increasing pattern of C/N ratio values with depth in the sediment (Fig. 2). These results suggest that although OM in sediments within OMZ apparently had the same origin (as the C/N ratio values were similar in the three cores), the three cores exhibited different diagenetic patterns of OM, especially of the nitrogen fraction (*e.g.* Degens and Mopper, 1976; Bender *et al.*, 1989). These differences are surprising, but reasons for such variability in OM composition in a relatively small area underlying the same environmental conditions (in terms of oxygen concentration) are difficult to explain. In organic-rich sediments of the Peru upwelling region, a rapid loss of organic N has been reported to occur with increasing depth of the sediment (Patience *et al.*, 1992; Lewis and Rowland, 1993). This did not occur in the present study. A possible explanation for the apparently weak diagenesis of OM in sediments within the OMZ in the Arabian Sea may lie in the nature of the OM. For instance, while OM in Peru Margin sediments is mostly labile, it is likely that in the Oman Margin sediments, most of the organic material reaching the sea bottom was already degraded (Emeis *et al.*, 1991; Smallwood and Wolff, 2000) due to the long residing time of sinking particles in the water column (Rao and Lamboy, 1995). Such a hypothesis is supported by data of hydrolysable amino acids and carbohydrates, whose contribution to TOC is rather low (about 10% in surficial sediments, Fig. 3). These values are closer to those reported by Cowie and Hedges (1994) for oxidised turbidites from the Madeira Abyssal Plain, but lower than those from the abyssal oligotrophic north Pacific (4990 m water depth, Lallier-Vergès and Albèric, 1990). This comparison supports the hypothesis that OM in the Oman Margin sediments within OMZ was mostly of refractory composition (Suthhof *et al.*, 2000). The significant variation (ANOVA, $p < 0.001$) in concentrations of both THAA and TCHO was observed in the down-core with higher values within the top 40 mm of the sediment. Such a pattern is in good agreement with ^{210}Pb data reflecting the presence of bioturbation processes down to that depth in the sediment (Smith, 2000; Horsfall and Wolff, 1997).

4.2 Origin, Composition and Diagenetic Patterns of Organic Matter Below OMZ

Total nitrogen concentrations in below OMZ sediments ranged from ~0.01 to ~0.4% (Tab. II) and were well correlated with the organic carbon contents (core 12687#8, $r = 1$; core 12687#10, $r = 0.99$; $p < 0.001$). Both sampled cores displayed the same decreasing patterns of both TOC and TN concentrations with depth in the sediment (Fig. 2). Again, C/N values (~8) were typical of relatively 'fresh' marine-derived OM (Jasper and Gagosian, 1990) as also observed in sediments within OMZ and sinking particles collected in the same area (about 9.5, Haake *et al.*, 1992). These results indicate that these two cores were characterized by OM of similar origin, also subjected to similar diagenetic patterns. The C/N ratio values in the sediments below OMZ were, however, characterized by a slight increasing pattern with depth in the sediment, suggesting that surface sediments were recently deposited and labile in nature, whereas deeper sediments were characterized by older and possibly allochtonous OM inputs (*e.g.* aeolian input; Smallwood and Wolff, 2000). This hypothesis is partially supported by the sediment particle size, whose changes follow those in OM content and elemental composition. Sandy sediments in the central portion of both cores (from below 100 mm down to 170–190 mm in core 12687#8; 190–210 mm in core 12687#10) were indeed accompanied by the lowest concentrations of OM content, as compared with finer-grained sediments in the surficial and deepest sediment layers, where OM contents were accordingly higher. It is likely that such shift in sediment grain size and OM content could be due to larger proportions of terrigenous material in intermediate deposits. An alternative hypothesis is that with relatively higher C/N ratio values in intermediate deposits, OM degradation (in particular the nitrogen fraction) in that layer was significantly faster than in the other two horizons. Such a hypothesis is strengthened by the vertical patterns of the THAA-C plus TCHO-C contribution to TOC (Fig. 3), which showed values in surficial sediments (up to ~20%) higher than those deeper in the sediment. These values are rather high and even similar to those reported for oxygenated coastal sediments (Cowie and Hedges, 1994). Also in below OMZ sediments, the presence of sub-surface spikes in labile organic compounds concentration reflects the presence of bioturbation down to 25 mm depth (Horsfall and Wolff, 1997), as revealed by the depth of ^{210}Pb mixed-layer and supported by the extremely high macrofaunal abundance (2485–3190 ind m^{-2}) (Smith *et al.*, 2000).

Deeper in the sediments below OMZ, the contribution of labile compounds to TOC decreased significantly (ANOVA, $p < 0.001$) although some differences were observed in the two investigated cores. Although inferentially, this discrepancy allows us to suspect that there is also a certain spatial variability in OM content and diagenesis in deep-sea sediments of the Arabian Sea.

The contribution of labile compounds to TOC in the intermediate sandy section of the below OMZ sediments was surprisingly high. Despite this, both THAA and TCHO concentrations as well as TN content in that section were very low. There are no compelling explanations for such discrepancies, but experimental errors associated with measurement of THAA and TCHO as well with TN determination may well have led to spurious values of the contribution of labile compounds to TOC, so these data should be considered with caution.

Although the down-core patterns of THAA and TCHO concentrations in sediments below OMZ were similar to those of TN and TOC (Fig. 2), THAA concentrations exhibited several sub-surface 'spikes' (*e.g.* at 30–50 mm in 12687#8). These peaks have been already reported in both oxic and anoxic sediments and have been ascribed to varying supply of THAA through time (Henrichs *et al.*, 1984; Balzer *et al.*, 1987) or to bioturbation (Horsfall and Wolff, 1997).

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