PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

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Phil. Trans. R. Soc. Lond. A 1993 **343**, 519-529
doi: 10.1098/rsta.1993.0063

Seasonal fluxes across the sediment–water interface, and processes within sediments

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Measurements of oxygen uptake across the sediment–water interface suggested that between 17–45% of the net primary production in the southern North Sea was degraded in the bottom sediments. Similar measurements of nutrient exchange fluxes showed that the sediments were significant sources of nutrients transferred to the water column. The sediments are, therefore, important sites of organic matter degradation and nutrient recycling, and must be included in any models for the North Sea. The sediments are also accumulators of radionuclides, particularly associated with the silt/clay fraction. At one site in the more central area of the North Sea where the water column stratifies during summer, vertical profiles of radionuclides (^{137}Cs, ^{239,240}Pu, ^{210}Pb) suggested a deposition rate of sediment of 0.3–0.6 cm a^{-1}, but at other sites vertical sediment profiles were unsuitable to measure deposition.

1. Introduction

In coastal marine environments fluxes of both inorganic and organic materials occur between the water column and bottom sediments. Such materials are derived both from primary production within the water column and from terrigenous sources. Particulate inorganic material settles to the bottom and may contribute to gradually accumulating bottom sediments, while settling particulates may also carry adsorbed or complexed trace chemicals with them. Exchange between sediments and overlying sea water will therefore lead to the trapping of trace chemicals in bottom sediments. As a sediment slowly accumulates, therefore, vertical profiles of sedimentary characteristics (below a surface mixed layer) reflect the history of exchange between the sediment and sea water. While the chemical and physical characteristics of the overlying water change in response to tidal, seasonal and interannual variations, the sedimentary characteristics provide a time-integrated eulerian record of the exchange processes which have occurred in the past between the sediment and the seawater.

Organic matter is decomposed within the water column, but in shallow coastal waters a large proportion of the organic input may survive settlement through the water column, and therefore the bottom sediments are important sites of organic matter degradation. The importance of the bottom sediments to organic matter decomposition is inversely related to water depth (Jørgensen 1980, 1983; Nedwell

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Phil. Trans. R. Soc. Lond. A (1993) **343**, 519–529
Printed in Great Britain

1984) and the southern half of the North Sea is a shallow water area with water depths less than 80 m. However, there is little information on the quantitative importance of the bottom sediments to the ecology of this region.

Aquatic sediments are largely anaerobic beneath a shallow oxic layer which is usually only a few millimetres deep (Revsbech *et al.* 1980). Decomposition of organic matter in marine sediments is therefore a function of aerobic metabolism in this narrow surface layer together with anaerobic decomposition which proceeds below the oxic layer. Anaerobic decomposition relies upon the interactions of a complex community of anaerobic bacteria, each of which brings about partial oxidation of the organic substrates leading to simpler organic molecules, of which acetate is the most important. In the terminal stages the sulphate respiring bacteria are responsible for the ultimate conversion of organic carbon to carbon dioxide (Abram & Nedwell 1978; Nedwell 1984). The reason for their importance is that seawater contains approximately 20 mM sulphate, providing abundant electron acceptors for this group of bacteria. It has been demonstrated that in coastal marine sediments approximately half of the organic matter mineralization is driven by sulphate respiration with the balance due to aerobic metabolism (Jørgensen 1980; Parkes & Buckingham 1986). Therefore, in marine sediments measurement of the rates of utilization of these two predominant respiratory electron acceptors, oxygen and sulphate, will permit the estimation of the stoichiometrically equivalent rates of organic carbon degradation.

Along with carbon, organic matter contains other elements (N, P, S etc.), which in coastal diatom communities are typically in the ratios $C_{106}N_{16}P_1$ (Redfield *et al.* 1963; Stefansson & Richards 1963). Therefore, decomposition of organic detritus will result also in the mineralization of these elements to inorganic forms. If bottom sediments are important sites of organic matter degradation they will also be sites of accumulation of mineral nutrients which may be transported from the sediment back to the water column where they will be available to sustain further production in the water column. Not all deposited organic matter will be recycled, but a proportion may be refractory and will be progressively buried in the sediment at an accumulating site. In general, the North Sea has negligible net sedimentation in its southern area (Eisma 1987; Rutgers van der Loeff 1980) with sandy bottom sediments. However, in the more central areas some deposition occurs and the sediments have a higher silt/clay content.

The present work was undertaken, as part of the NERC North Sea Community Research Programme (NSP), to establish the significance of the bottom sediments in organic matter degradation, and to measure the fluxes of inorganic nutrients across the sediment–water interface. In addition, natural and artificial radionuclides in North Sea sediments were investigated, both to elucidate their distribution and to obtain information on the depositional history of the sediments.

2. Study sites

During the initial survey cruise in April–May 1988, 41 stations were sampled along the NSP survey track and sediment cores taken with a multicorer (Barnett *et al.* 1984). Samples of surface sediment (0–5 cm layer) were taken for measurement of the sedimentary organic carbon content determined as per cent ash-free dry weight (%AFDW); and of particle size analysis, particularly for the silt/clay fraction (percentage < 63 μm) with which radionuclides are associated. Figure 1 illustrates

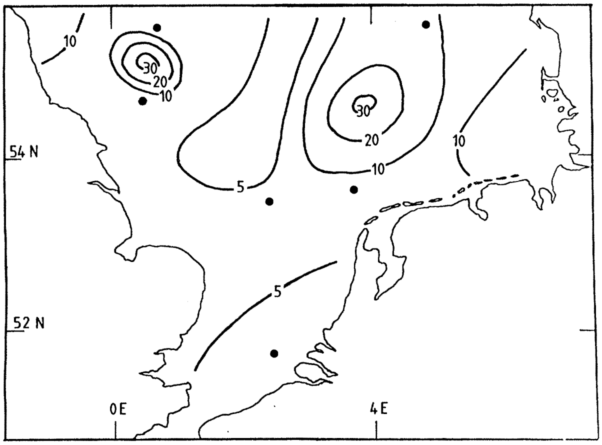

Figure 1. Distribution of the silt/clay fraction in sediments of the southern North measured during the initial survey cruise. Isolines show the percentage of sediment with grain size < 63 μm.

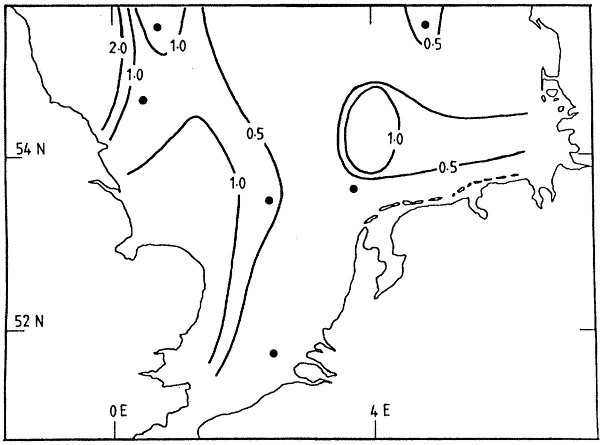

Figure 2. Organic content in sediments of the southern North Sea, measured during the initial survey cruise. Isolines show the %AFDW in the sediments.

the distributions of the silt/clay fraction. The more northerly stations which stratified during summer had generally finer sediments than the more southerly stations near the Channel mouth in the region of mixed water which did not stratify.

Low organic matter was present in the more coarse, sandy sediments in the south (figure 2), and higher organic contents in the finer, more northerly sediments. Figure 3 shows the distribution of the five types of bottom sediment which were significant within the study area: sand 23%; mud/sand 21.3%; sand/gravel 38.2%; mud/sand/gravel 10.2%; rock/sand/gravel 6.9% (*Oceanographic atlas of the north Atlantic Ocean*, p. 71. Washington, D.C.: U.S. Naval Hydrographic Office (1965); Veenstra 1971; D. J. Hydes, personal communication).

From the initial survey cruise, six stations, renumbered 1–6, were selected (figure 3) to cover the range of bottom types, and to encompass the more northerly stratified area, the frontal central area and the most southerly mixed area. These stations were visited on eight subsequent occasions, at approximately two-monthly intervals between September 1988, and August 1989. Stations 3 and 4 had the finest particle sediments, and the highest organic contents which decreased with depth in the

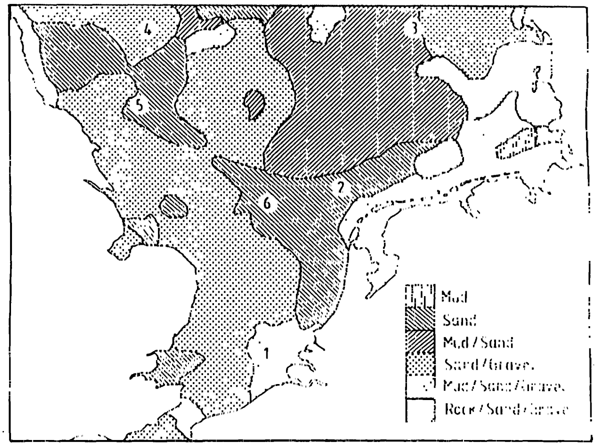

Figure 3. Distribution of different sediment types in the southern North Sea.

sediment. At the most southerly stations 1 and 2 the sediments were coarse sand with very little silt/clay, and low organic content which increased with depth in the sediment. Stations 5 and 6 were of intermediate characteristics.

3. Organic carbon decomposition in the bottom sediments

(a) *Rates of exchange of oxygen and nutrients between sediment and water column*

On each visit cores of sediment were taken at each of the six stations with the multicorer and were used to measure benthic oxygen uptake and benthic sulphate reduction rates. (Further details of methodology are given by Upton *et al.* 1993.) Cores were held on board ship in a water bath, immersed in sea water taken from near the sea bed at that station, for 6 h to permit the sediment cores to re-equilibrate after coring. The temperature of the water bath was maintained at that of the *in situ* bottom sediment with a thermocirculator. After re-equilibration, samples of seawater were removed, filtered through glass fibre filter papers, and frozen until analysed colorimetrically later for dissolved nutrients (nitrate, ammonium, phosphate and silicate). A further water sample was taken from within each core tube and analysed for its dissolved oxygen concentration by an automated amperometric back-titration method (Talling 1973). Three core tubes were then capped, enclosing approximately 300 ml of seawater above the sediment, and incubated in the water bath for up to 6 h. Further samples of water for analyses of dissolved oxygen and of nutrients were removed at the midpoint of the incubation period and at the end. Changes in the concentrations of dissolved oxygen and nutrients in the water samples during the incubation period permitted the calculation of the rates of exchange between the sediment and the overlying water column of these solutes (Parkes & Buckingham 1986).

(b) *Sulphate reduction rates in bottom sediments*

After the re-equilibration period, three further cores were removed and sampled in 5 cm deep sections under oxygen-free nitrogen. Triplicate subsamples from each 5 cm deep horizon were taken with 5 ml polypropylene hypodermic syringes, with their distal ends removed, and then sealed with butyl rubber seals to exclude air. Each subsample of sediment was injected with 2 μCi (74 MBq) of [^{35}S]sulphate solution and incubated for 24 h at the *in situ* temperature. The samples were then frozen at

Table 1. *Mineralization of organic matter by aerobic and sulphate respiration in bottom sediments at each site in the North Sea*

	organic degradation ($mmolC\ m^{-2}\ a^{-1}$)		percentage due to	
station	O_2 uptake	SO_4 reduction	O_2	SO_4
1	3236.3	346.6	89.3	10.7
2	4583.9	1398.4	69.5	30.5
3	4392.2	1112.6	74.7	25.3
4	3771.7	699.6	81.4	18.6
5	2836.1	410.8	85.5	14.5
6	4451.5	2365.0	46.9	53.1

−20 °C until later analysis in the laboratory. The samples were subsequently analysed for [^{35}S] in acid-volatile sulphide and pyrite+elemental sulphur (Parkes & Buckingham 1986). Sulphate reduction rates were calculated according to the method of Jørgensen (1978).

(*c*) *Organic matter mineralisation*

Oxygen uptake rates during winter were between 5–10 mmol $O_2\ m^{-2}\ day^{-1}$ at all sites, but during summer were more variable. At sites 4 and 5, where the water column stratified during summer and where there was very little seasonal change of bottom water temperature, there was little increase in O_2 uptake rate during the summer. In contrast, at sites 2 and 3 there were summer increases to 15–28 mmol $O_2\ m^{-2}\ day^{-1}$, and at sites 1 and 6 increases to 7–17 mmol $O_2\ m^{-2}\ day^{-1}$. These values corresponded well with others reported in the literature for oxygen uptake by North Sea sediments (van Raaphorst *et al.* 1990). The measured daily rates of O_2 uptake were integrated with time by a simple trapezium method to estimate the annual O_2 uptake by the sediment at each of the six stations (table 1). By comparison, van Raaphorst *et al.* (1992) estimated similar values for annual oxygen uptake of 3 and 10 mol $O_2\ m^{-2}\ a^{-1}$ at sites in the Broadfourteens and the Friesian Front respectively.

Similarly, measured sulphate reduction rates were integrated with depth (0–20 cm) to yield for each station a daily rate of sulphate reduction on each sampling date. These were integrated with time to give an estimate of the annual sulphate reduction at each station. In table 1 the amounts of organic carbon which were estimated to be mineralized are shown. There were no statistically significant correlations ($P > 0.05$) between the organic mineralization rates and the organic content of the sediment at each station, probably reflecting differences in the rates of turnover, and hence differences in the availability of the organic matter. Overall, there were statistically significant correlations ($P < 0.05$) between temperature and daily oxygen uptake rate, and between temperature and sulphate reduction rate, although the correlation may be because of either direction temperature effects on these processes, or indirect effects via seasonal changes of net primary production (NPP) and organic input to the sediments.

Sulphate reduction accounted for 10.7–53.1 % of the total organic matter mineralization. The smallest proportion was at the most southerly station 1 where a high proportion of coarse sand presumably facilitated O_2 transport into the sediment and hence minimized the contribution of anaerobic organic matter degradation.

Estimates of the total organic matter mineralization in the sediments can be derived in two ways. First, an average of the annual benthic organic matter degradation for all six stations (4.06 ± 0.8 mol C m^{-2}) can be multiplied by the total area of the southern North Sea (2.4×10^{11} m^2 between 55° 30′ N and the entrance of the Dover Straits) to yield 9.74×10^{11} mol C a^{-1} (1.17×10^7 tonnes C a^{-1}). Secondly, the areas of different types of bottom can be used to obtain an alternative estimate. Stations 2, 5 and 6 occurred on sand; station 3 on mud/sand; station 4 on sand/gravel; and station 1 was taken as characteristic of both mud/sand/gravel and rock/sand/gravel. The annual organic matter mineralization in each sediment bottom type was calculated and summed. This gave a total annual organic matter mineralization of 1.16×10^7 tonnes C a^{-1}, which was identical to the previous estimate.

What does this estimate of benthic mineralization represent as a proportion of the organic input in the southern North Sea? Postma (1973) suggested that the major part of organic material in offshore waters was not allochthonous, but derived *in situ*, and estimates of net primary production (NPP) are available. Aletsee & Rick (1988, cited in Kempe & Pegler 1991) estimated the NPP in the 517000 km^2 of the whole North Sea as 15×10^7 tonnes C a^{-1}; or, proportionately, 6.96×10^7 tonnes C a^{-1} in the area of the southern North Sea. Reid *et al.* (1990) indicated that NPP in the southern North Sea was about 200 gC m^{-2} a^{-1}, equal to 4×10^7 tonnes C a^{-1} total. Finally, as a component of the NSP, Joint & Pomroy (1992) measured the NPP in the six southerly ICES boxes. For equivalent areas, their estimate of annual NPP for the southern North Sea corresponded to about 2.6×10^7 tonnes C a^{-1}. Thus, estimates of NPP in the southern North Sea range from 2.6–6.96×10^7 tonnes C a^{-1}. Our calculation of benthic organic matter mineralization represents between 17–45% of the annual NPP in the southern North Sea, and emphasizes that the bottom sediments are significant sites of organic matter degradation.

4. Nutrient exchange between sediment and water column

When organic matter is degraded and mineralized, apart from carbon, elements such as nitrogen, phosphorus, silicon, etc., bound in the organic matter, will be liberated as inorganic forms. Measurements of nitrate, nitrite, ammonium, phosphate and silicate in the porewaters at each of the six stations revealed that their concentrations were usually greater than in the overlying seawater. Whether or not exchange of these solutes between the sediment and the overlying water will occur is controlled by the redox conditions near the sediment/water interface, and the microbial processes which occur there. For example, nitrification in the aerobic surface layer may prevent efflux of ammonium to the water (Nedwell *et al.* 1983); while release of phosphate from the sediment may be prevented by the presence of a surface oxidized layer (Mortimer 1941; van Raaphorst *et al.* 1988). Our measurements of the exchange fluxes of mineral nutrients between the sediment and water column were measurements of the resultant of the physical, chemical and biological processes.

The nutrient fluxes at the six stations were measured on eight occasions throughout the year. Maximum rates of efflux of nutrients were seen during the summer: for nitrate during August–September 1989, with rates of *ca.* 800 and 700 μmol m^{-2} day^{-1} at stations 1 and 2 respectively. The lowest summer rate of nitrate output was *ca.* 200 μmol m^{-2} day^{-1} at station 4. Greatest efflux of ammonium

Table 2. *Net annual fluxes of nutrients between the sediment and overlying water at six stations in the southern North Sea, measured between September 1989, and September 1990*

(Positive values indicate flux into the sediment; negative values indicate flux out of sediment.)

	net annual flux of nutrient (mmol m^{-2} a^{-1})				
station	nitrate	nitrite	ammonium	phosphate	silicate
1	−88.3	54.6	−83.4	−36.1	−64.0
2	26.1	33.7	−160.3	6.8	−294.6
3	−97.9	13.7	−116.9	−7.6	−353.6
4	−15.4	26.5	−39.4	1.0	−396.4
5	−56.1	25.0	36.1	−3.6	−231.8
6	−73.3	0.9	−377.7	−17.9	−383.2

was 2400 and 2050 µmol m^{-2} day^{-1} at stations 6 and 2 respectively. Station 5 was always a sink for ammonium, in contrast to the other stations. Greatest rates of silicate output were at sites 2 and 3 (3200 and 2500 µmol m^{-2} day^{-1} respectively). These rates of nutrient exchange are similar to others reported in the literature for the region. van Raaphorst *et al.* (1990) investigated 12 stations over the Dogger Bank during summer (July–August) and reported the following average outward fluxes of nutrients (µmol m^{-2} day^{-1}): Si 333; NH$_4^+$ 167; NO$_3^-$ 161; P 32. At stations in the Broad Fourteens and the Friesian Front (near our station 2) maximal rates of ammonium release during summer were 500 and 1700 µmol m^{-2} day^{-1} respectively (van Raaphorst *et al.* 1992). We will restrict ourselves to consideration of the net annual fluxes at each station, which were derived by integrating daily flux rates with respect to time for the whole year (table 2).

It can be seen that on annual basis the sediments were significant exporters of nitrate, ammonium and silicate to the water column. In contrast, the sediments were sinks for nitrite. Overall, they were always net exporters of nitrogen when nitrate, nitrite and ammonium were considered together. There were only small net fluxes of phosphate on an annual basis, probably because of the low solubility of iron phosphates in the oxidized environment near the sediment/water interface. The annual net fluxes of these mineral nutrients were converted to total annual fluxes for the southern bight using the areas of different bottom types described previously for the benthic organic matter mineralization (table 3).

From the estimates of NPP for the southern North Sea of 2.6–6.96 × 10^7 tonnes C a^{-1}, and elemental ratios for a coastal diatom population of C:N:P:Si 106:16:1:22 (Stefansson & Richards 1963), the total annual outputs of each element from the sediments (table 3) can be calculated as a percentage of that element assimilated during NPP as: nitrogen 4–10%; phosphorus 10–26%, and silicon 14–38%. On an annual basis, the sediments are therefore important sources of nutrients used during NPP in the water column, particularly of P and Si. The significance of the sedimentary source of nutrients would have been greater during the period in summer when the water column was most deficient in available nutrients.

5. Radionuclides

Subsections of surface sediment (0–1 cm depth) from 24 of the stations during the initial survey cruise were subsequently analysed for the presence of natural (Pb, Th, U isotopes) and artificial (Cs, Pu isotopes) radionuclides (Anderson & Fleer 1982;

Table 3. *Total annual fluxes of nutrients between sediment and water column in the southern half of the North Sea, estimated from the proportions of each bottom type present*

bottom type	percentage of total area	flux across each bottom type ($\times 10^9$ mol a^{-1}) nitrate	nitrite	ammonium	phosphate	silicate
sand	23.0	-1.90	1.10	-9.25	-0.27	-16.77
mud/sand	21.3	-5.00	0.70	-5.99	-0.39	-18.10
sand/gravel	38.2	-1.42	2.44	-3.62	-0.09	-36.47
mud/sand/gravel	10.2	-2.16	1.34	-2.04	-0.88	-1.57
rock/sand/gravel	6.9	-1.47	0.91	-1.38	-0.60	-1.06
total flux (mol a^{-1})		-1.20×10^{10}	6.48×10^9	-2.23×10^{10}	-2.05×10^9	-7.40×10^{10}

Table 4. *Radionuclide measurements for subsections of a core from station 4*

(Errors are $\pm 1\sigma$ counting errors. Errors for U and Th isotopes are < 10 %.)

depth/cm	percentage particles < 63 µm	radionuclide activity (Bq kg^{-1}) ^{137}Cs	^{239,240}Pu	^{210}Pb	^{234}U	^{238}U	^{228}Th	^{230}Th	^{232}Th
0–1	16.1	7.2 ± 1.3	0.14 ± 0.03	20.2 ± 0.7	6.9	6.1	8.4	8.2	7.2
2–3	14.3	11.1 ± 1.9	0.16 ± 0.03	19.1 ± 0.8	6.2	5.9	8.2	8.3	7.9
4–5	19.7	15.3 ± 1.9	0.22 ± 0.04	18.8 ± 1.0	8.3	6.9	8.5	6.3	6.3
6–7	11.9	9.3 ± 1.5	0.41 ± 0.10	18.7 ± 0.50	7.9	7.4	7.1	6.4	7.5
8–9	13.5	8.9 ± 0.9	0.21 ± 0.04	18.2 ± 0.9	5.2	5.1	6.4	6.3	6.9
10–11	12.8	4.7 ± 1.4	0.16 ± 0.03	16.3 ± 1.1	8.9	9.9	9.2	9.1	9.0
12–13	11.1	4.1 ± 0.7	0.15 ± 0.03	14.3 ± 1.2	11.1	10.1	9.8	6.9	6.2
14–15	12.0	4.8 ± 0.7	0.17 ± 0.03	12.8 ± 1.4	11.4	10.4	10.8	14.8	9.9
16–17	13.7	4.0 ± 0.8	0.14 ± 0.03	10.6 ± 0.7	10.5	9.6	8.4	7.7	6.3
18–19	13.0	3.5 ± 0.8	0.14 ± 0.03	7.8 ± 0.4	11.3	10.6	7.3	7.4	7.3

Assinder 1982) to assess the horizontal variability of radionuclide distribution. Particular attention was paid to stations 3 and 4, in sites of accumulation, where there was sufficiently fine-grained sediment for study of sediment-bound pollutant accumulation.

There were significant correlations ($P < 0.05$) between the percentage of the silt/clay fraction in the sediments and the concentrations of ^{137}Cs, ^{239,240}Pu, ^{234}U, ^{238}U, ^{228}Th, ^{230}Th, and ^{232}Th, emphasising the significance of this sediment fraction in sequestering these radionuclides (Assinder 1983). The correlation of the silt/clay fraction and ^{210}Pb was not significant, however, possibly because of run-off of ^{210}Pb in coastal areas from coal mining, and fertilizer production and use. These give rise to higher levels of ^{210}Pb due to uranium in coals and natural phosphates (McCartney *et al.* 1990).

The ^{137}Cs detected over the study area ranged between 0.8–9.0 Bq kg^{-1} (dry weight), and can be compared with measurements of 8.5 Bq kg^{-1} in coastal sand near the Hartlepool nuclear establishment (Hunt 1990). It may be derived either from Sellafield, from Cap de la Hague, or from nuclear fallout. As there was no detectable ^{134}Cs (a major product of Chernobyl fallout), and discharges from Sellafield have always been greater than from Cap de la Hague, it may be deduced that the majority of the ^{137}Cs was derived from Sellafield, via known transport pathways around Scotland to the North Sea (Prandle 1984; Hunt 1990).

(*a*) *Radionuclide profiles and sediment deposition*

Vertical profiles of radionuclides in a core from station 4 are given in table 4. A pronounced peak of ^{137}Cs occurs at 5 cm depth, and an absence of ^{134}Cs suggests that this is related to high discharges from Sellafield in the mid-1970s which have been preserved in the sediment profile. Mixing and remobilization processes have not been sufficient to obscure the trend of the discharge record at this site, therefore. A sedimentation rate of *ca.* 0.6 cm a^{-1} can be deduced from this peak, assuming a five year lag in transport from the Sellafield source (Kautsky 1979). The same core showed a peak of ^{239,240}Pu at about the same depth, and if this reflected the peak Sellafield discharge in 1973 it also suggested a tentative sedimentation rate of *ca.* 0.6 cm a^{-1}, assuming the same five year lag from discharge.

The activity of ^{210}Pb in all cores showed a decrease consistent with the classic use of excess ^{210}Pb decay to determine sedimentation rate. To calculate excess ^{210}Pb in the simplest way requires a knowledge of the supported ^{210}Pb levels, usually obtained by determining the activity at which total ^{210}Pb levels off at depth in the core. This point was not reached in any core. However, using a value of 5 Bq kg^{-1} for supported ^{210}Pb allowed tentative sedimentation rates of 0.3–0.5 cm a^{-1} to be estimated for site 4. This is likely to be unreliable as the quantity of excess ^{210}Pb is small to the relatively coarse samples collected, but the estimate is similar to that calculated from ^{137}Cs and ^{239,240}Pu. The data seem to suggest that for site 4 sedimentation rates were less than 1 cm a^{-1}. At other stations sediment profiles were unsuitable to calculate sedimentation rates due to mixing.

Uranium activities increased with depth down the cores, and the ^{234}U/^{238}U ratios also increased with depth. Both of these features support the theory of post-depositional removal of authigenic U to these deeper layers, as noted in other shelf sediments (Aller & Cochran 1976; Carpenter *et al.* 1984). Activity of benthic organisms introduces overlying sea water with about 40 mBq dm^{-3} of dissolved ^{238}U and ^{234}U with a ^{234}U/^{238}U ratio of 1.15 (but little dissolved Th) several centimetres down into the sediment. The uranium is then deposited under the reducing conditions present in the deeper sediment (Cochran 1982). Thorium profiles show no identifiable trends with sediment depth. Since essentially all ^{232}Th in marine sediments is detrital in origin (Cochran 1982) constant ^{232}Th activity with depth in the North Sea sediments indicates that there has been little change in the composition of detritus deposited throughout the cores.

In conclusion, the bottom sediments were important sites of both organic matter degradation and of recycling of nutrients back to the water column. They were particularly significant in the more northerly stations in the stratified area of the North Sea. Both natural and artificial radionuclides accumulated in the sediments, their concentrations correlated with the silt/clay fraction. Over most of the southern North Sea there was no net deposition of sediment, but at one site in the stratified region a deposition rate of 0.3–0.6 cm a^{-1} was measured.

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