Rapid appearance of Chernobyl radiocesium in the deep Norwegian Sea sediments

Chernobyl
Radiocesium
Sediments
Deep sea
Norwegian Sea

Chernobyl
Radiocésium
Sédiments
Mer profonde
Mer de Norvège

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ABSTRACT

Two deep-sea box cores, collected two months after the Chernobyl accident from the slope off mid Norway at 967 m water depth and from the Vøring Plateau (1426 m), were analysed for ^{134}Cs and ^{137}Cs and revealed a total of Chernobyl radiocesium of 220 and 330 Bq/m^2, respectively. The reactor cesium was essentielly confined to the uppermost centimeter of the cores. The depth distribution of the nuclides, which is interpreted in terms of bioturbation, gives a minimum value for the biological mixing coefficient of 1100 and 300 cm^2/ka (or 3.5×10^{-8} and 9.5×10^{-9} cm^2/s), respectively, for the two stations studied.

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RÉSUMÉ

L'apparition rapide du radiocésium de Chernobyl dans les sédiments profonds de la Mer de Norvège

Deux carottes de sédiments ont été recueillies en Mer de Norvège deux mois après l'accident de Chernobyl, l'une sur la pente norvégienne à 967 m de profondeur et l'autre sur le Vøring Plateau à 1 426 m. Les analyses de ^{134}Cs et ^{137}Cs permettent d'estimer le dépôt total de radiocésium de Chernobyl à 220 et 330 Bq/m^2, respectivement. La distribution des ^{134}Cs et ^{137}Cs, essentiellement contenus dans le premier centimètre, est interprétée en termes de bioperturbation et donne des coefficients minimaux pour la bioperturbation du sédiment de 1 100 et 300 cm^2/ka (ou $3{,}5.10^{-8}$ et $9{,}5.10^{-9}$ cm^2/s), respectivement, pour les deux localités étudiées.

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INTRODUCTION

With the disaster at Chernobyl on early April 26, 1986, a huge amount of fission nuclides was injected into the atmosphere and readily spread out to the northwest and west (Panitz *et al.*, 1986; Devell *et al.*, 1986; Smith, Clark, 1986). The radioactive fallout, which contaminated wide areas of northwest and northern Europe, was also recorded in Greenland and on Svalbard (Pourchet *et al.*, 1986) and thus can be expected to have imprinted the deep-sea sediments of the Norwegian-Greenland Sea as well. This provides a chance to study short-term effects of transport phenomena in the marine environment, particularly the particle mixing processes in the uppermost layer of the sediment. Here we present first analytical results on the radiocesium content in two cores off mid Norway.

MATERIALS AND METHOD

The two box cores (50 × 50 cm) were collected on July 4, 1986 during Meteor cruise 2, leg 2, off mid Norway (Fig. 1). Core 101 (66°59.9'N, 7°46.6'E, 967 m) was collected from an area of high silt sedimentation (Rumohr, pers. comm., 1987) ranging between about 700 and 1200 m water depth on the continental slope. Core 103 (67°39.4'N, 5°47.5'E, 1426 m) was collected further west from the central part of the Vøring Plateau, which slightly raises above a wide submarine depression towards Scandinavia and hence is protected against near-bottom supply of particulate matter from the upper continental slope. Both cores show fine-grained hemipelagic sediments with carbonates below 30% (Henrich, 1986) and organic carbon between 0.5 and 1% of dry weight.

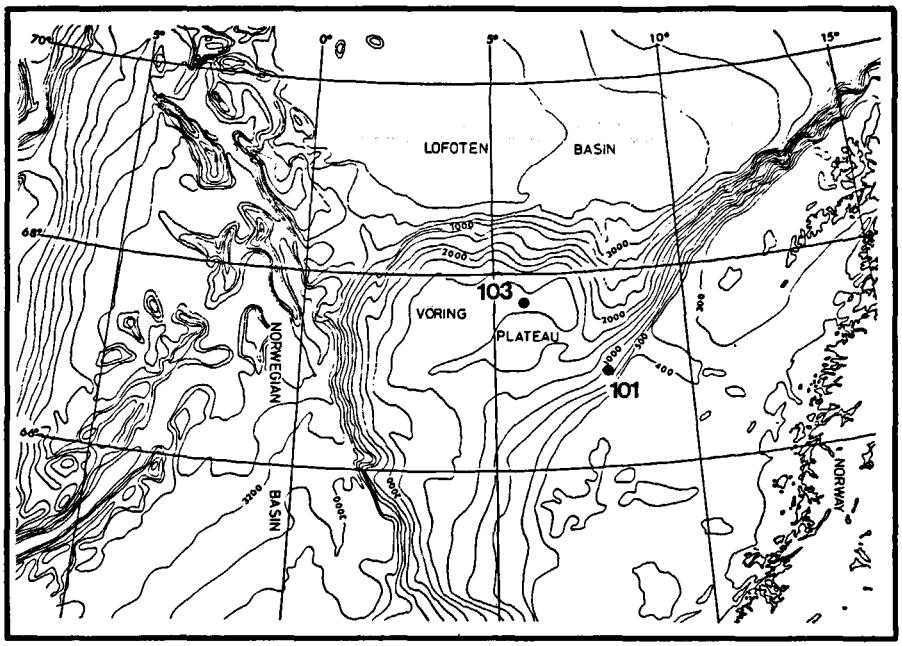

Figure 1
Coring locations; water depth in metres.
Localisation des carottages; profondeur en mètres.

A subcore of 10 cm diameter was drawn from each box core and cut. The interstitial water was expelled from the sediment slices for chemical investigation and the remainder analysed for radiocesium in February, 1987. The gamma-ray spectra of the sediments were measured on a temperature stabilized 3 × 3″ NaI scintillation spectrometer coupled to a multichannel pulseheight analyser. Samples ranged between 20 and 93 g of dry weight and were measured for about 48 hours in a well defined geometry. The spectra were corrected for background. In order to overcome spectral interference of radiocesium with the U- and Th-supported natural radioactive families, the radiocesium photopeaks were evaluated from the difference of the net spectra between the respective sample and the sediment from 3 to 5 cm depth, which appeared to be free of Chernobyl cesium. Normalizing the two net spectra prior to stripping was performed by means of the total counts in the 860 to 1200 keV energy interval. ^{134}Cs was quantified via its 796, 802 keV doublet and then was used to correct for the contribution of the 605 keV line to the 662 keV peak of ^{137}Cs. The energy dependent photopeak efficiency as well as the sample self-absorption factors were adequately taken into account in this procedure. The absolute accuracy of the radiocesium determinations shown in the Table is within 10 to 30%, or about 1 to 2 Bq/kg for the low activity values.

Due to the relatively late date of our measurements after Chernobyl, ^{134}Cs and ^{137}Cs are by far the most prominent gamma-emitters left of the Chernobyl fission nuclide assembly. Of the other longer living fission nuclides, only the ^{106}Ru/^{106}Rh pair could slightly contribute to the Cs-photopeaks by its 622 keV line (9.8% A = absolute intensity, *i.e.* gamma-rays per decay; Erdtmann, Soyka, 1979). This may have biased our ^{137}Cs values by 2.1% (calculated from the data of Sadasivan, Mishra, 1986). However, ^{106}Ru/^{106}Rh could not be identified directly, since even the stronger 512 keV line (20.6% A; interferes closely with the 511 keV line of ^{208}Tl of the natural ^{232}Th family!) did not appear in the final net pulseheight spectra as a distinct photopeak.

Table
Analytical results and biological mixing coefficients from model calculations.
Résultats des mesures et coefficients de bioperturbation, calculés par différents modèles.

	Station 101				Station 103			
	Slope 967 m				Vøring Plateau 1426 m			
Sample depth in core (cm)	Porosity	^{134}Cs (Bq/kg)[1]	^{137}Cs (Bq/kg)[1]	Water content[2]	Porosity	^{134}Cs (Bq/kg)[1]	^{137}Cs (Bq/kg)[1]	Water content[2]
0-0.5	0.788	14	36	143.0	0.861	40	95	238.2
0.5-1	0.761	5	19	122.5	0.833	6	13	191.8
1-2	0.751	0	0	116.0	0.810	0	1	164.0
2-3	0.728	0	0	102.9	0.786	2	5	141.3
Inventory (Bq/m^2)		56	161			99	236	
^{134}Cs: ^{137}Cs		0.35	1			0.42	1	
Biolog. mixing coeff. D_1, model 1								
(kg/m^2)^2/a		22.4	37.0			5.1	4.6	
cm^2/ka		646	1090	133[3]		289	321	215[3]
Multiplier of D_1 for model								
2		3.1	3.6			2.0	2.1	
3		4.6	6.2			3.4	3.3	

[1] Becquerel per kg dry sediment, decay-corrected to May 1, 1986. Analytical errors are within 10 to 30%, or 1-2 Bq/kg minimum.
[2] (weight-) percent *vs.* dry matter.
[3] Mean water content for 0-1 cm.

RESULTS AND DISCUSSION

The results are presented in the Table and Figures 2 and 3. The activities are decay-corrected and refer to May 1, 86. The radiocesium pair is found to be essentially confined to the uppermost centimeter of the sediment. Faint traces below were detected in core 103 from the Vøring Plateau. The relative rank of these traces is beyond statistical doubt.

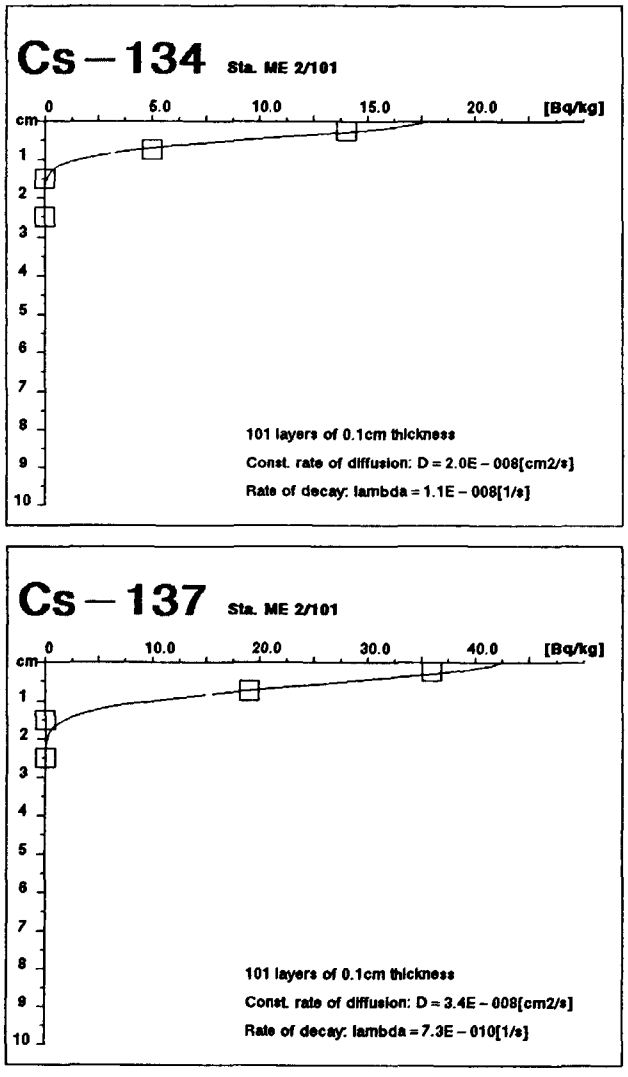

Figure 2

Core 101 (*slope*, 924 *m*): *radiocesium profiles. Activities refer to dry weight* (*D* = *mixing coefficient*, cm^2/s; *lambda* = *decay constant of radiocesium*, s^{-1}).

Carotte 101 (pente, 924 m) : concentrations en radiocésium dans le sédiment sec (D = coefficient de bioperturbation, cm^2/s; lambda = constante de désintégration du radiocésium, s^{-1}).

The $^{134}Cs/^{137}Cs$ activity ratio is, with one exception, close to 0.42. This figure is very much higher than that observed by Kautsky (1986) on the residual radiocesium waste from Sellafield Works in the waters of the Norwegian Sea, and is close to the ratio of about 0.5 reported in literature for the Chernobyl fallout (Devell *et al.*, 1986; Bondietti, Brantley, 1986; Pourchet *et al.*, 1986; Sadavisan, Mishra, 1986; Dörr, Münnich, 1987). The measured amounts of the short-lived ^{134}Cs ($t_{1/2}$ = 2.06 a) also discard the possibility that the radiocesium found might be a remnant of bomb-produced fallout from the early sixties. So there is little doubt

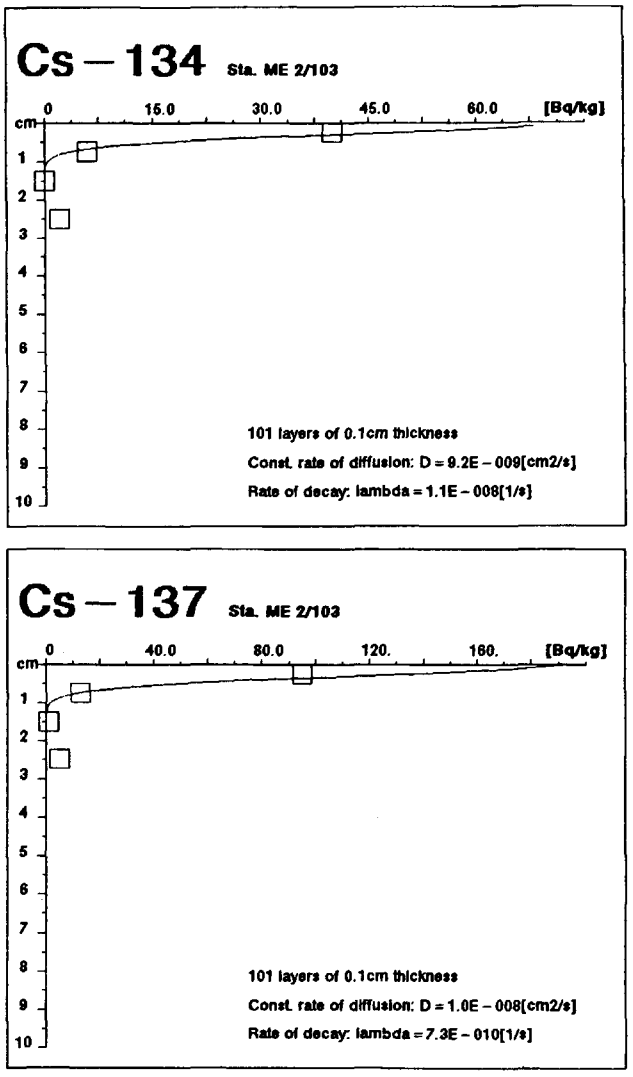

Figure 3

Core 103 (*Vøring Plateau*, 1426 *m*): *for legend see Figure 2.*

Carotte 103 (Vøring Plateau, 1426 m) : *cf.* légende de la figure 2.

as to the origin of the sedimentary radiocesium from Chernobyl. However, a small contribution of pre-Chernobyl ^{137}Cs is possible. Disregarding a possible bias from $^{106}Ru/^{106}Rh$, the $^{134}Cs/^{137}Cs$ ratios of 0.42 (measured) and 0.5 (literature) suggest a pre-Chernobyl ^{137}Cs content of 16% of the total ^{137}Cs. Curiously, however, bomb-Cs was not detected below the ^{134}Cs layers, although it should have penetrated deeper into the sediment due to the longer time bioturbation was effective, and also was missing in two comparative cores collected in 1985. This point deserves further attention.

The appearance of the radiocesium in the sediments so short a time (2 months) after its injection into the Norwegian Sea can hardly be explained by deep-convecting surface waters, as the thermohaline stratification of the water column by the time of the Chernobyl event does not favour deep water formation a long time after the winter season. Most likely the radiocesium —or a fraction of the Cs-fallout— was fixed to planktonic matter and subsequently supplied to the sediment in particulate form. This kind of transport, which partly uses fecal pellets or voluminous aggregates of plankton detritus ("fluff") as carriers, can be very rapid (*see, e.g.* Deuser *et al.*, 1981; Billet *et al.*, 1983).

The detailed mechanisms, which bring the radiocesium to the bottom, still are to be found out. Particulate radiocesium, for instance, which has been a constituent of the Chernobyl fallout (Bondietti, Brantly, 1986), is expected to be removed much faster (by scavenging) than dissolved (ionic) radiocesium and hence may partially account for the rapid appearance of ^{134}Cs and ^{137}Cs in the deep sea.

The penetration of the radiocesium isotopes into the sediment probably results from bioturbation. Redissolution, on the other hand, of the radiocesium from the settled particles at the sea bottom and subsequent downward diffusion is not thought to be of major significance, since almost 98% of the redissolving Cs would be lost into the free bottom water which allows for much faster diffusion than the sediment (the respective diffusion coefficients are 1×10^{-5} and ca. 5×10^{-9} cm^2/s; Duursma, Eisma, 1973). Very high quantities of 9900 and 15200 Bq/m^2 of ^{134}Cs and ^{137}Cs must have redissolved up to the time of coring at stations 101 and 103, respectively, if the measured inventories of 220 and 330 Bq/m^2, respectively, were built up by diffusive intrusion.

The radiocesium inventory of the deep sea bottom is slightly higher on the Vøring Plateau than on the slope. If not caused by differences in fallout, this finding may reflect a slightly lower settling efficiency at the slope station of the fine-grained matter to which the radiocesium is usually associated. The depositional environment at the site of core 101 is in favour of coarser grain (silt) rather than clay deposition (Holtedahl, 1981; Rumohr, pers. comm., 1987).

The inventory of radiocesium in the sediments studied amounts to about 200 to 300 Bq/m^2 and thus is much lower than reported for most of the continental sites. Radiocesium activities in Germany or Finnland in general are pronouncedly higher than 1000 Bq/m^2 (GSR, 1987; Dörr, Münnich, 1987). It is possible that a substantial part of the oceanic Cs-fallout either was still dissolved at the time of coring or was fixed to unsettled suspended matter in the surface water and had not yet been deposited on the sea floor. This view is suggested by the comparatively slow removal of dissolved radiocesium from the water column (Kautsky, 1985) and the very low sinking velocity of the smallest aggregates (marine snow) among the suspended particulates (Asper, 1987). Alternatively, the Norwegian Sea may have escaped the fallout from the most heavily contaminated air masses of the early days after Chernobyl on account of the prevailing meteorological situation, *i. e.* the course of the Chernobyl trajectories and the rain fall history of Northern Europe in the beginning of May (Panitz *et al.*, 1986; Smith, Clark, 1986). Heaviest fallout on Greenland and Svalbard occurred as late as May 10 to May 11, although weaker radiodeposition was recorded earlier (Pourchet *et al.*, 1986). These aspects also bear importance for models serving to describe the variation of the radionuclide input to the sediments with time.

BIOTURBATION

The radiocesium in the sediment reveals a distinct downcore distribution, which most likely reflects the mixing of sedimentary particulates by benthic organisms. Modelling this bioturbation in terms of a diffusion-like process (*see, e. g.* Christensen, Bhunia, 1986; Robbins, 1986) yields the biological mixing coefficient as a quantity reflecting the intensity of this process, which generally overprints any sedimentary record and hence gains importance as a low-pass filter acting upon the transfer of environmental signals to the sedimentary memory. Observing the long-term development of the radiocesium profiles in the sediments after the Chernobyl event in particular will later gives a chance to directly observe the variation of the mixing coefficient with depth in the sediment and relate these results to the vertical species and abundance distributions of the benthic macrofauna.

For the present cores, bioturbation was treated by a simple model based on Fick's 1st law of diffusion and the continuity equation, *i. e.* the mass conservation law. Sediment growth and radioactive decay could be neglected for the short period of time considered here.

Only the two upper samples were considered to be representative for a statistically well matured tracer profile which can only result from a sufficiently long series of stochastic sediment transport actions of the benthic individuals. The radionuclide distribution below probably is of a different character. The radiocesium traces in the deeper layers may partly represent pre-Chernobyl radiocesium or may reflect the downward diffusion of the —most likely small— fraction of the radiocesium dissolved in the interstitial waters (Duursma, Eisma, 1973; Santschi *et al.*, 1983; Sholkovitz *et al.*, 1983), or for core 103, may have been advected by deep-burrowing macrofauna such as Sipunculans, which have been observed in the sediments of the Vøring Plateau to produce a closely packed network of deep-reaching open burrows (Romero-Wetzel, 1987). These features deserve further inspection and have not been included in our model shown here.

Using the common notation for partial derivatives, the basic equations of the model, *i. e.*

Fick's law

$$j = -D c_x \qquad (1)$$

continuity equation

$$c_t = -j_x \qquad (2)$$

combine as

$$c_t = D c_{xx} \qquad (3)$$

with

x = sample depth in core (kg-dry weight/m^2);
t = time (a);
$c = c(x, t)$ = specific activity (Bq/kg-dry matter);
D = mixing coefficient, assumed to be constant with depth;

$j = j(x, t)$ = radioactivity flux (Bq $a^{-1} m^{-2}$).

The boundary conditions are

a) $c(x, 0) = 0$ at $t = 0$;

b) $c(x, t) \to 0$ for $x \to \infty$;

c) $\int_0^T j_0 \, dt = \int_0^\infty c(x, T) \, dx = A(T)$ = inventory (Bq/m^2)

at time T with j_0 = input flux density (Bq $a^{-1} m^{-2}$).

The solution of equation (3), averaged over the sample interval $x - a/2$, $x + a/2$, is

$$c(x, t) = \frac{2}{a} \int_0^t j_0(s) \left(\varphi \left(\frac{x + (a/2)}{\sqrt{2D(t-s)}} \right) - \varphi \left(\frac{x - (a/2)}{\sqrt{2D(t-s)}} \right) \right) ds \qquad (4)$$

where φ is the error integral

$$\varphi(z) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{z} e^{-(u^2/2)} \, du. \qquad (5)$$

Certainly, with only two measured values for each profile, only a 2-parameter formulation could be applied, one parameter being the mixing coefficient D and the other fitted to reproduce the input of the tracer to the sediment. Three models concerning different behaviour of the radionuclide input with time were considered.

1) $j_0 = A_0 * \delta(t)$ ($\delta(t)$ = Delta-function),

or

$$\int_0^\infty c(x, t) \, dx = A_0 = \text{constant with time},$$

i.e., a single pulse of radioactivity was input to the sediment at time $t = 0$. This model corresponds to a short-lasting, heavy radiowaste deposition, such as seen on land in early May, 1986.

2) $j_0 = j_{00}$ = constant with time, *i.e.* the radioactivity is supplied continuously at constant rate. This model considers a possible gradual runoff of radioactivity from land and/or gradual deposition by a finite sinking velocity of the Cs-loaded particulates.

3) $j_0 = b * t$, *i.e.* the input flux increases linearily with time since $t = 0$. This model is to match the possible effects of temporary depositories and entrapment sites, such as estuaries and nearcoastal zones, which are usually intercalated between the contaminated terrestrial sites and the final depositories in the deep sea.

The model parameters were calculated from equation (4). Since the radiocesium is attached to the particulate fraction rather than occurring in solution (Miettinen, Lax, 1980; Santschi *et al.*, 1983; Sholkovitz *et al.*, 1983), the depth scale of the sediment column has been expressed as dry matter weight per unit area in order to use an invariant approach with respect to porosity changes. The biological mixing coefficients (Tab.) have also been given in conventional units, using the mean water content of the upper core cm for conversion. With these latter figures given and assuming a single pulse of tracer input on May 1, a more general, numerical formulation of the differential equations, applying a finite difference scheme, has been used to prepare the graphical representation of the results (Fig. 2 and 3).

The biological mixing coefficient (Tab.) for model 1 of core 103 (Vøring Plateau) amounts to about 300 cm^2/ka or 9.5×10^{-9} cm^2/s, a reasonable value of deep-sea sediments (Schink, Guinasso, 1977). A much higher activity of bioturbation is found for core 101 from the slope. This finding nicely correlates with the richness of the benthic fauna observed at this site (Graf, pers. comm., 1987) which is well supplied with nutrient-rich matter from the shelf.

The prolonged input of radioactivity considered in model 2 and 3 results in correspondingly higher mixing coefficients by a factor 2 to 6 (Tab.). The result of model 1 hence is likely to provide a lower estimate of the intensity of bioturbation by which the sedimentary particles in the very top layers of the deep Norwegian Sea deposits are mixed up.

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REFERENCES

Asper V. L., 1987. Measuring the flux and sinking speed of marine snow aggregates, *Deep-Sea Res.*, **34**, 1-17.

Billett D. S. M., Lampitt R. S., Rice A. L., Mantoura R F. C., 1983. Seasonal sedimentation of phytoplankton to the deep-sea benthos, *Nature*, **302**, 520-522.

Bondietti E. A., Brantley J. N., 1986. Characteristics of Chernobyl radioactivity in Tennessee, *Nature*, **322**, 313-314.

Christensen E. R., Bhunia P. K., 1986. Modelling radiotracers in sediments: comparison with observations in Lake Huron and Michigan, *J. Geophys. Res.*, **91**, 8559-8571.

Deuser W. G., Ross E. H., Anderson R. F., 1981. Seasonality in the supply of sediment to the deep Sargasso Sea and implications for the rapid transfer of matter to the deep ocean, *Deep-Sea Res.*, **28**, 495-505.

Devell L., Tovedal H., Bergström U., Appelgren A., Chyssler J., Andersson L., 1986. Initial observations of fallout from the reactor accident at Chernobyl, *Nature*, **321**, 192-193.

Dörr H., Münnich K. O., 1987. Spatial distribution of soil-^{137}Cs and ^{134}Cs in West Germany after Chernobyl, *Naturwissensch.*, **74**, 249-251.

Duursma E. K., Eisma D., 1973. Theoretical, experimental and field studies concerning reactions of radioisotopes with sediments and suspended particles of the sea, Part C: Applications to field studies, *Neth. J. Sea Res.*, **6**, 265-324.

Erdtmann G., Soyka W., 1979. *The gamma rays of the radionuclides*, Verlag Chemie, New York, 862 p.

GSR Gesellschaft für Reaktorsicherheit mbH, 1987. Neuere Erkenntnisse zum Unfall im Kernkraftwerk Tschernobyl, Bericht GRS-S-40 (Feb. 87), Schwertnergasse 1, Köln 1.

Henrich R., 1987. A calcite dissolution pulse in the Norwegian-Greenland Sea during the last deglaciation, *Geol. Rundsch.*, **75**, 805-827.

Holtedahl H., 1981. Distribution and origin of surface sediments on the Norwegian continental margin between 62°N and 65°N with some remarks on the late quaternary litho- and biostratigraphy, in: *The Norwegian coastal current*, edited by R. Saetre and M. Mork, Univ. Bergen, Vol. 2, 768-792.

Kautsky H., 1985. Distribution and content of different artificial radionuclides in the waters of the North Sea during the years 1977 to 1981 (complemented with some results from 1982 to 1984), *Dtsch Hydrogr. Z.*, **38**, 193-224.

Kautsky H., 1986. Artificial radioactivity in the North Sea and the northern North Atlantic during the years 1977 to 1986, manuscript submitted to International Atomic Energy Agency, Hamburg, Nov. 1986.

Miettinen J. K., Lax M., 1980. Distribution of radionuclides in seawater and sediments, in: *Isotope marine chemistry*, edited by E. Goldberg *et al.*, Geochemical Research Association, Tokyo, 35-49.

Panitz H. J., Matzerath D., Ehrhardt J., Kusch W., Geiß H., Maßmeier K., 1986. Analyse der Ausbreitungsvorgänge nach dem Reaktorunfall von Tschernobyl. Anlage 1a, in: Bericht über den Reaktorunfall in Tschernobyl, seine Auswirkungen und die getroffenen bzw. zu treffenden Maßnahmen, Der Bundesminister für Umwelt, Naturschutz und Reaktorsicherheit, Bericht RSI1-518042-SOW/2, Bonn, 18.6.1986.

Pourchet M., Pinglot J.-F., Gascard J.-C., 1986. The northerly extent of Chernobyl contamination, *Nature*, **323**, 676.

Robbins J. A., 1986. A model for particle selective transport of tracers in sediments with conveyor belt deposit feeders, *J. Geophys. Res.*, **91**, 8542-8558.

Romero-Wetzel M., 1987. Sipunculans as inhabitants of very deep, narrow burrows in deep-sea sediments, *Mar. Biol.*, **96**, 87-91.

Sadavisan S., Mishra U.C., 1986. Radioactive fallout swipe samples from Chernobyl, *Nature*, **324**, 23-24.

Santschi P. H., Li Y.-H., Adler D. M., Amdurer M., Bell J., Nyffeler U. R., 1983. The relative mobility of natural (Th, Pb, Po) and fallout (Pu, Am, Cs) nuclides in the coastal marine environments: results from model ecosystems (MERL) and Narragansett Bay, *Geochim. Cosmochim. Acta*, **47**, 201-210.

Schink D. R., Guinasso N. L. Jr., 1977. Modelling the influence of bioturbation and other processes on calcium carbonate dissolution at the sea floor, in: *The Fate of Fossil Fuels*, edited by N. R. Andersen and A. Malahoff, Plenum Press, New York, 375-399.

Sholkovitz E. R., Cochran J. K., Carey A. E., 1983. Laboratory studies of the diagenesis and mobility of ^{239,240}Pu and ^{137}Cs in nearshore sediments, *Geochim. Cosmochim. Acta*, **47**, 1369-1379.

Smith F. B., Clark M. J., 1986. Radionuclide deposition from the Chernobyl cloud, *Nature*, **322**, 690-691.