^{239,240}Pu transport into the Arctic Ocean from underwater nuclear tests in Chernaya Bay, Novaya Zemlya

J.N. Smith[a,*], K.M. Ellis[a,1], L. Polyak[b], G. Ivanov[c], S.L. Forman[d], S.B. Moran[e]

[a]*Bedford Institute of Oceanography, P.O. Box 1006, Fisheries and Oceans Canada, Dartmouth, NS, Canada B2Y 4A2*
[b]*Byrd Polar Research Center, Ohio State University, Columbus, OH, USA*
[c]*Okeangeologia, St. Petersburg, Russia*
[d]*Department of Earth and Environmental Sciences, University of Illinois, Chicago, IL, USA*
[e]*Graduate School of Oceanography, University of Rhode Island, Narragansett, RI, USA*

Abstract

Radionuclide measurements have been conducted on sediment, seawater and biota samples collected in Chernaya Bay, on the southern coast of Novaya Zemlya, the site of two underwater nuclear tests conducted in the 1950s. ^{239,240}Pu levels in sediments from the central region of Chernaya Bay exceed concentrations of 15,000 Bq/kg, and are among the highest ever reported for the marine environment. It is estimated that approximately 11 TBq of ^{239,240}Pu from the tests has been retained in the sediments of Chernaya Bay. Plutonium from Chernaya Bay is distinguished by ^{240}Pu/^{239}Pu atom ratios of 0.03 that are much lower than ratios of 0.18 typical of global fallout. High levels of ^{137}Cs (Bq/kg) and ^{60}Co (Bq/kg) were also measured in surface sediments in the central regions of Chernaya Bay near the presumed epicentre of the explosions. Applications of a biodiffusion model to excess ^{210}Pb sediment depth profiles indicate that the distribution of ^{239,240}Pu is governed mainly by sediment mixing in this low sedimentation rate (< 0.1 cm/yr) regime and, as a result, most of the ^{239,240}Pu has been retained in the upper 20 cm of the sediment column. Elevated levels of ^{239,240}Pu measured in *Macoma* (104 Bq/kg), *Fucus* (15 Bq/kg) and polychaete (1292 Bq/kg) from Chernaya Bay, indicate that ^{239,240}Pu levels in the benthos are comparatively high and that significant uptake has occurred in the food chain. Although levels of ^{239,240}Pu in bottom water from Chernaya Bay are high (4.2 Bq/m^3), restricted exchange over the fjord sill limits the present rates of ^{239,240}Pu transport from

---

* Corresponding author. Tel.: + 1-902-426-3865; fax: + 1-902-426-6695.

E-mail address: smithjn@mar.dfo-mpo.gc.ca (J.N. Smith)

[1] Deceased, 23 July 1999.

contaminated sites in Chernaya Bay into the eastern Barents Sea. However, low ^{240}Pu/^{239}Pu atom ratios measured in sediment cores collected throughout the eastern Barents Sea indicate that significant offshore transport of plutonium from Chernaya Bay has occurred in the past, probably at the time of the original nuclear tests. The large difference in end member ^{240}Pu/^{239}Pu atom ratios for Chernaya Bay fallout (0.03) and atmospheric fallout (0.18) has been exploited to estimate that 2 TBq of ^{239,240}Pu in Barents Sea sediments was originally derived from Chernaya Bay. Further, a plume of low ^{240}Pu/^{239}Pu ratio plutonium, distributed in a northwestward direction, is evident in sediments along the southern coastline of Novaya Zemlya, indicating that an additional quantity of Chernaya Bay plutonium may have been transported into the Arctic Ocean. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Environmental concerns about radioactivity contamination of the Russian Arctic Ocean precipitated by the Yablokov Report (Yablokov et al., 1993) have led to numerous investigations, including recent studies of radioactive waste dumpsites in the Kara Sea (Salbu et al., 1997), downstream transport of radioactivity from nuclear facilities on the Ob and Yenesey Rivers (Sayles et al., 1997), long-range transport from European nuclear fuel reprocessing plants (Kershaw and Baxter, 1995) and offshore transport of radioactivity into the Barents Sea from nuclear test sites on the southern coastline of the island of Novaya Zemlya (Smith et al., 1995). This latter investigation revealed that some of the highest levels of ^{239,240}Pu ever measured in the marine environment can be found in the sediments of Chernaya Bay, a small (1–6 km wide, 15 km long) fjord on the southwestern coast of Novaya Zemlya. Chernaya Bay is a former Soviet Union nuclear weapons test site where at least two underwater nuclear tests were conducted in 1955 and 1957. The high level of ^{239,240}Pu (> 8000 Bq/kg) measured on a single surface sediment sample collected from Chernaya Bay in 1992 indicated that a large inventory of ^{239,240}Pu from the nuclear tests may have been sequestered in bottom sediments (Smith et al., 1995). Chernaya Bay plutonium was also shown to be distinguished by a low ^{240}Pu/^{239}Pu isotope atom ratio of 0.03 (compared to typical fallout ratios of 0.18) that is typical of low yield nuclear tests. In addition, elevated levels of ^{239,240}Pu measured in box cores collected within 50 km of the approaches to Chernaya Bay indicated that ^{239,240}Pu transport had occurred from this embayment into the eastern Barents Sea (also known as the Pechora Sea) and could represent a potential radiological threat to its important commercial fishery.

More extensive oceanographic investigations were undertaken aboard the R/V *Geolog Fersman* in 1993 and 1996 to collect seawater, sediment and biota samples in Chernaya Bay and its surrounding approaches in the eastern Barents Sea. An important element in these studies was to trace the transport of plutonium from Chernaya Bay using its characteristic Pu isotopic signature. The specific questions to

be addressed in this study were:

1. What are the magnitude, isotopic composition and bio-geochemical partitioning of the radionuclide inventory in Chernaya Bay?
2. Has there been significant biological uptake of ^{239,240}Pu in the food chain?
3. What is the magnitude of the ^{239,240}Pu flux from Chernaya Bay into the eastern Barents Sea and central Arctic Ocean?

These questions bear upon both the bio-geochemical mobility of fallout ^{239,240}Pu in arctic marine sediments and other environmental issues associated with the only recorded detonation of nuclear weapons in an Arctic Ocean fjord. They also address an important issue underlying many of the radionuclide studies in the Arctic Ocean; namely, the extent to which the Russian continental shelves represent a significant source of contamination, both for North American shelves and for the central basins of the Arctic Ocean.

2. Methods

Sediment and large volume seawater samples were collected during 1993 and 1996 cruises of the RV *Geolog Fersman* to the eastern Barents Sea (Fig. 1) and Chernaya Bay (Fig. 2). Supplementary data from a 1992 *Geolog Fersman* cruise and 1992 Dalnie
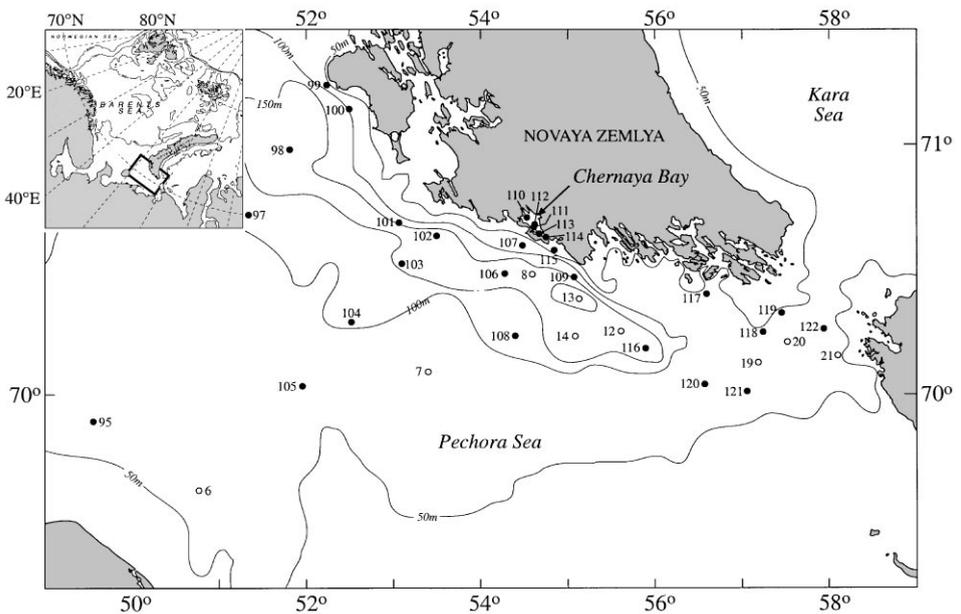

Fig. 1. Sediment grab and core samples were collected at stations occupied by the RV Dalnie Zelentsy in 1992 (open circles) and the RV *Geolog Fersman* (closed circles) in 1993 in the Barents Sea.
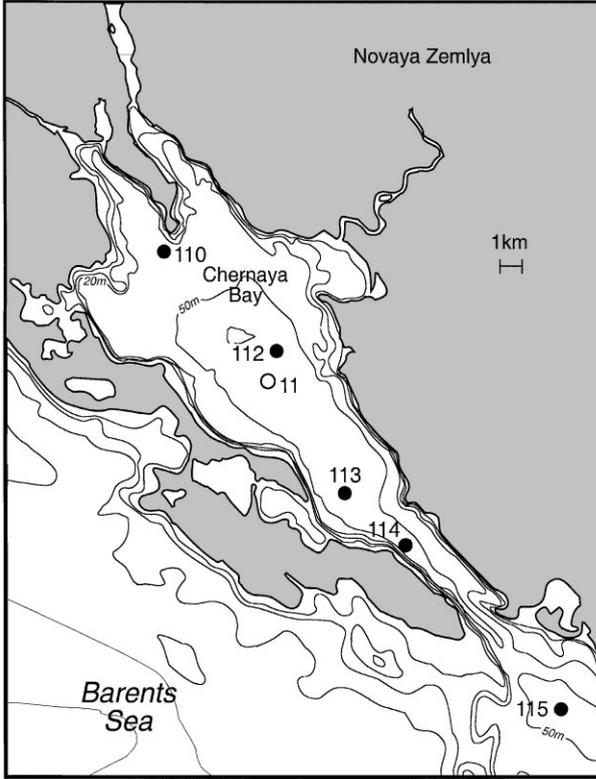
Fig. 2. Sediment box cores, large volume seawater samples and biota were collected in 1993 at the five stations (closed circles) located within and near the entrance to Chernaya Bay. Station 11 (open circle) marks the location of a sediment grab sample collected aboard the RV *Dalnie Zelentsy* in 1992.

Zelentsy cruise (Smith et al., 1995) to the eastern Barents Sea are also reported in this paper. In 1993, sediment grab samples were collected at Stations 95–119 using a Van Veen sampler. The grab samples collected in these cohesive, clayey sediment regimes were sufficiently consolidated that they were subsequently subsampled on the ship at 2 cm intervals (using a polypropylene subcore cylinder) to a depth of 8 cm. Five sediment cores were collected at Stations 110–115 in Chernaya Bay using a large box corer having dimensions, 0.5 m × 0.5 m × 2 m. The box cores were subsampled using a 50 cm PVC cylinder with a vacuum pump attached to the top core cap to prevent sediment compression during insertion of the subcore. The subcores were sampled at 1 cm intervals aboard the ship, stored in plastic containers having air tight lids and returned to the Bedford Institute of Oceanography (BIO) for geochemical and radionuclide analyses. Two meter long, PVC Lehigh gravity cores (10 cm ID) were also collected at some locations. ^{210}Pb was measured on sediment samples using alpha particle spectroscopy and the ^{226}Ra supported contribution to the total ^{210}Pb activity was determined using a radon gas emanation technique (Smith and Walton,

1980). Pu and Am were radiochemically separated from acid-digested sediment samples using ion-exchange techniques, electro-deposited onto stainless steel disks and measured by alpha particle spectroscopy (Smith et al., 1995). Gamma emitting radionuclides were measured using a 25% Ge hyperpure well detector. Metal analyses were performed by ICP-MS (Loring et al., 1995). Particle size measurements were conducted by standard sieving/pipetting techniques (size fraction < 63 μm is termed "percentage clay $+$ silt") and total organic carbon was measured using a CHN analyser. Biota samples were removed from the sediments using screens, rinsed of particulate material, freeze dried and analysed for radionuclides using techniques similar to those referenced above. ^{14}C analyses were conducted on dried sediment samples by accelerator mass spectrometry (Polyak et al., 1995).

Plutonium isotopic concentrations were measured using a Finnigan MAT ELE-MENT high-resolution ICP-MS at the Graduate School of Oceanography, University of Rhode Island. Plutonium was de-plated in a clean-room from the stainless steel disks used for alpha counting by leaching for 24 h in 1.5 m HNO_3 in a Teflon vial and analysed by HR-ICP-MS equipped with a MCN 6000 desolvator. Successive leachings and analyses of test disks indicated that the plutonium recoveries were generally $> 98\%$. Instrument sensitivity was ~ 2 Mcps per ppb U. The low resolution mode was employed to analyse the isotopes ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. Each isotope was scanned 100 times to reduce counting uncertainties in ^{240}Pu/^{239}Pu ratios to $< 4\%$ RSD. Mass fractionation of the Pu isotope ratios was calibrated using NIST reference standard material U050, which has a certified ratio of ^{235}U/^{238}U $= 0.05278$ and a mass fractionation of 0.18% per AMU.

Large volume seawater samples were collected with a 100 l GoFlo Niskin bottle, passed through KCFC resin cartridges to extract ^{137}Cs (Smith and Ellis, 1995) and subsequently analysed using gamma spectroscopy. A lower limit of extraction of ^{137}Cs from solution of 96% was estimated from measurements conducted on second KCFC resin columns placed in series (Smith and Ellis, 1995). ^{242}Pu tracer was added to acidified, unfiltered 60 l seawater samples on board the vessel and Pu isotopes were co-precipitated using ferric hydroxide, with the precipitates subsequently being analysed by techniques referenced above.

3. Results and discussion

3.1. Environmental setting

Chernaya Bay is a shallow (92 m deep) embayment on the southern coast of Novaya Zemlya bordered by low elevation (< 50 m) accretionary spits formed by long-shore drift from the southeast (Fig. 2). It is 14 km long, 1–6 km wide and has a 46 m sill at its seaward entrance. The dominant sources of sediments in this region of the eastern Barents Sea are coastal erosion and inputs of suspended matter from the Pechora River, which drains into the southern part of the sea, in addition to long shore drift of suspended sediments from the Kara Sea (Pfirman et al., 1995). Bottom sediments have a comparatively high percentage of fine grained clays ($>80\%$) and
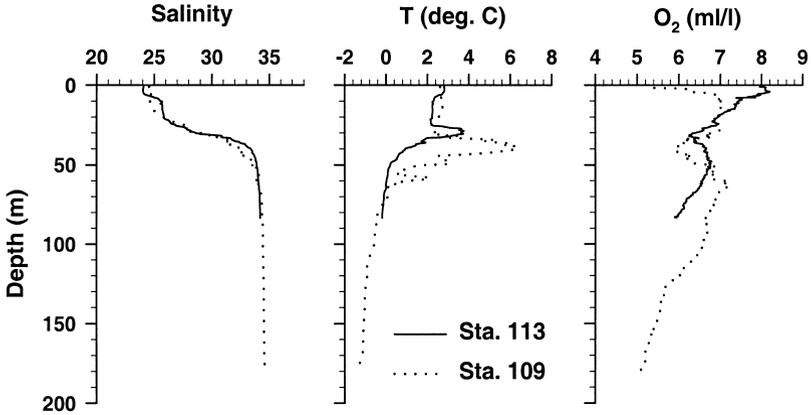
Fig. 3. Salinity, temperature and dissolved oxygen depth profiles for Station 113 in Chernaya Bay and Station 109, 15 km offshore from Chernaya Bay in the Barents Sea.

organic matter (1.5–2.0% total organic carbon; Smith et al., 1995). ^{14}C ages (T_{C-14}) were measured on sediment samples collected at Station 113 in Chernaya Bay using a Lehigh gravity corer. The 2 m core was subsampled at depths of 30 cm ($T_{C-14} = 0.665$ kyr), 60 cm (1.51 kyr), 90 cm (1.54 kyr), 150 cm (2.43 kyr) and 190 cm (2.94 kyr). The ^{14}C sedimentation rate between 30 and 60 cm is 0.036 cm/yr and the mean sedimentation rate over the entire depth range (30–190 cm) is 0.073 cm/yr. These results are consistent with Holocene-averaged sedimentation rates of 0.05 cm/yr generally measured in depositional basins in the southeastern Barents Sea proximal to Novaya Zemlya (Polyak et al., 1995).

Freshwater discharge to the fjord occurs via a small river at the head of Chernaya Bay. The hydrography of the fjord is characterised by a relatively fresh (salinity = 24–26) surface layer down to a depth of 20 m and a steep halocline at the 30 m level (Fig. 3), with maximum salinities of 34.85 measured in bottom water. The intrusion of Atlantic water into the fjord is characterised by a temperature maximum (>3°C) located several metres above the halocline with temperatures decreasing to 0°C in bottom water. Although the bottom water is relatively well-oxygenated (oxygen > 70% saturation levels at 87 m; Fig. 3), there is an O_2 deficit extending from the sill depth of 50 m to the bottom of the inner basin indicating that exchange between the inner basin and the Barents Sea is partially restricted.

Since the 1950s, Chernaya Bay has been the site for a series of atmospheric, underground and underwater nuclear weapons tests (Boyarsky, 1993). Three underwater tests having yields of less than 20 kilotons were reportedly conducted in Chernaya Bay in 1955, 1957 and 1961 Matuschenko et al., 1994), although the latter test is also reported as having taken place off the southern coastline of Novaya Zemlya (Nilsen and Bohmer, 1994). The 1955 test was the first nuclear explosion carried out on Novaya Zemlya and was apparently conducted at a water depth of 12 m using a nuclear tipped torpedo (Matuschenko et al., 1994), but little information is available on either this or subsequent tests.
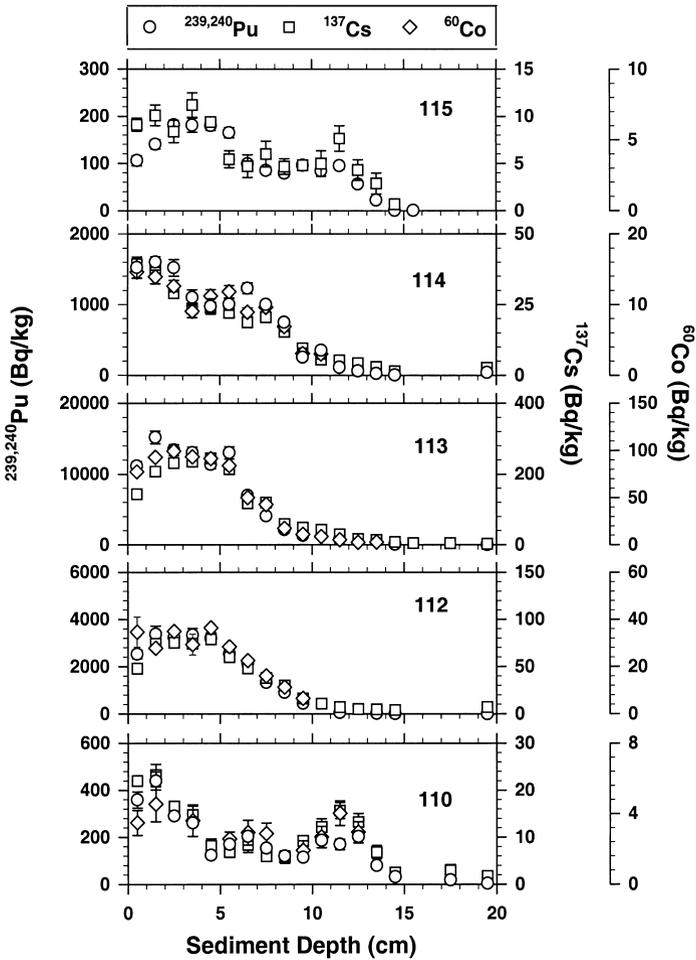
Fig. 4. ^{137}Cs, ^{60}Co and ^{239,240}Pu concentration profiles in Chernaya Bay sediments reveal the highest levels in the upper 10 cm of core 113 from the central basin where ^{239,240}Pu levels exceed values of 15,000 Bq/kg.

3.2. Radionuclide sediment-depth profiles in Chernaya Bay

The sediment-depth profiles for ^{137}Cs, ^{60}Co and ^{239,240}Pu for the four box cores from Chernaya Bay and the core from Station 115 located immediately outside its entrance are illustrated in Fig. 4. The highest levels of radioactivity were measured in cores from Stations 112 and 113 in the deep (60–80 m), central part of the basin. Maximum levels of ^{239,240}Pu ($> 15,000$ Bq/kg), ^{137}Cs (> 300 Bq/kg) and ^{60}Co (> 100 Bq/kg) are orders of magnitude greater than fallout levels (eg. < 1 Bq/kg for ^{239,240}Pu) typically measured in fine-grained depositional regimes in the Barents and Kara Seas (Smith et al., 1995). These radionuclide levels are similar to those measured

in 1972 in surface sediments at Enewetak (1000–15,000 Bq/kg for ^{239,240}Pu; 100–500 Bq/kg for ^{137}Cs; 20–200 Bq/kg for ^{60}Co decay corrected to 1992), a coral atoll in the Pacific Ocean that was the site of several US underwater nuclear tests conducted during the 1950s (Nelson and Noshkin, 1973). The only comparable radionuclide levels measured in an arctic environment are in sediments in Bylot Sound near Thule, Greenland, where a B-52 aircraft carrying four nuclear weapons crashed in 1968 resulting in ^{239,240}Pu contamination of local sediments at levels as high as 10,000 Bq/kg (Holm et al., 1988; Smith et al., 1994). These results support the hypothesis, based entirely on radionuclide analyses of one surface sediment sample collected in 1992 at Station 11 (Fig. 2), that underwater nuclear tests conducted in the 1950s have produced widespread ^{239,240}Pu contamination in Chernaya Bay sediments (Smith et al., 1995).

The sediment-depth profiles (Fig. 4) for each of the three isotopes ^{239,240}Pu, ^{137}Cs and ^{60}Co are similar for each core (110, 112, 113 and 114) within Chernaya Bay. Least-squares regressions between the concentrations of the three radionuclides over the entire depth range of each core all have regression coefficients, r^2, greater than 0.95. The high correlation between concentrations of radionuclides having very different half-lives is consistent with delivery from a single input event. Multiple sources (having similar isotopic yields) separated by periods of more than a few years would result in higher relative values of the longer-lived plutonium isotopes compared to ^{60}Co ($t_{1/2} = 5.4$ yr) at deeper depths in the sediment column. This result supports the validity of the assumption of a single pulsed input event that was used to calculate a date of 1957 for the delivery of Pu to Chernaya Bay sediments based on their ^{241}Pu/^{239}Pu atom and ^{241}Am/^{240,239}Pu activity ratios (Smith et al., 1995).

The highest levels of Pu (> 3000 Bq/kg) were measured in the 0–6 cm levels of cores collected in the central region (Sta. 112 and 113) of the fjord (Fig. 5) while lower ^{239,240}Pu levels were measured in the shallower (29 m) surface sediments of Station 110 at the northern extremity of the fjord and at Station 114, near the 50 m sill. ^{239,240}Pu levels decrease further to values of the order of 100 Bq/kg in sediments at Station 115 immediately outside the fjord, indicating that Chernaya Bay has been relatively effective in retaining the original inventory of bomb-produced, particle-reactive radionuclides. Maximum values of ^{137}Cs and ^{60}Co (250 Bq/kg and 100 Bq/kg, respectively), measured in surface sediments at Sta. 113, were much lower than those of ^{239,240}Pu. The radionuclide concentration gradient along the axis of the fjord (Fig. 5) suggests that the epi-centre of the detonation was located between Stations 112 and 113. The limited depth of sediment penetration of radioactivity indicates that minimal disturbance of the sediments occurred as a result of the underwater tests.

3.3. Sediment biodiffusion model

The inventory of artificial radionuclides in Chernaya Bay sediments is almost entirely retained in the upper 20 cm of the sediment column. Sediment-depth distributions of ^{239,240}Pu, ^{137}Cs and ^{60}Co usually exhibit a plateau or sub-surface maximum in the upper 5 cm and then decrease to background levels by the 15–20 cm level. Levels of excess ^{210}Pb (ie. ^{210}Pb$_{ex}$; equal to total ^{210}Pb minus ^{226}Ra-supported
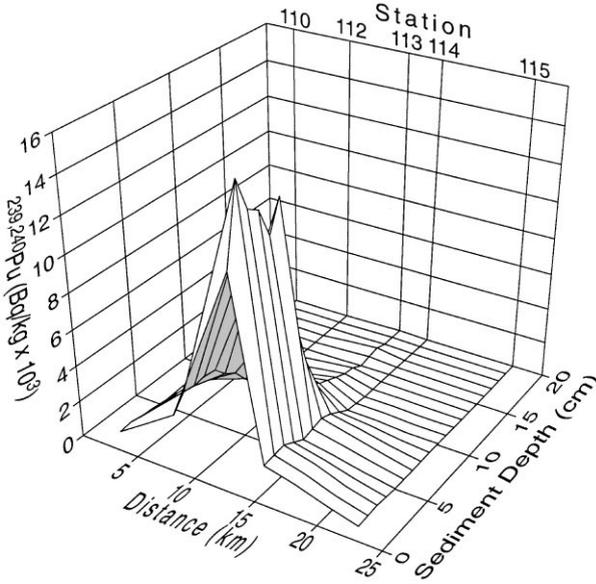
Fig. 5. ^{239,240}Pu sediment profiles shown as a function of distance from the head of the fjord illustrate the high degree of spatial focusing in the vicinity of Stations 112 and 113, close to the assumed epi-centre of the nuclear test.

^{210}Pb) in Chernaya Bay sediments (Fig. 6) are typical of those normally measured in fine-grained, marine sediments and this naturally occurring radionuclide can be used to constrain the sedimentation and biodiffusion (bioturbation) rates governing the artificial radionuclide distributions (Smith et al., 1995).

A two layer, biodiffusion model has been used to simulate the ^{210}Pb$_{ex}$ and ^{239,240}Pu sediment-depth profiles for Chernaya Bay sediments assuming that the tracer distributions are subject to continuous sedimentation and particle mixing and can be described by

$$\frac{\partial A}{\partial t} = \frac{\partial (D_b \partial A/\partial z)}{\partial z} - \omega \frac{\partial A}{\partial z} - \lambda A \qquad (1)$$

where A is the tracer activity, D_b (cm^2/yr) is the biodiffusion coefficient (represented by D_b^0 in the upper surface mixed layer and D_b^∞ in the deep layer), ω (cm/yr) is the sedimentation rate and z (cm) is the sediment depth (Guinasso and Schink, 1975; Smith et al., 1995). The relatively small gradient in the solid volume fraction ($< 15\%$) in the upper 20 cm of the sediment cores validates the assumption that sediment-depth compaction is approximately constant (Boudreau, 1986a; Mulsow et al., 1998). The flux of ^{210}Pb across the sediment–water interface was assumed to be constant while an impulse model was employed for ^{239,240}Pu (Smith et al., 1994). The pulsed input of ^{239,240}Pu was assumed to have occurred in 1957 as previously determined from plutonium and americium isotope ratios (Smith et al., 1995). In all of the cores,
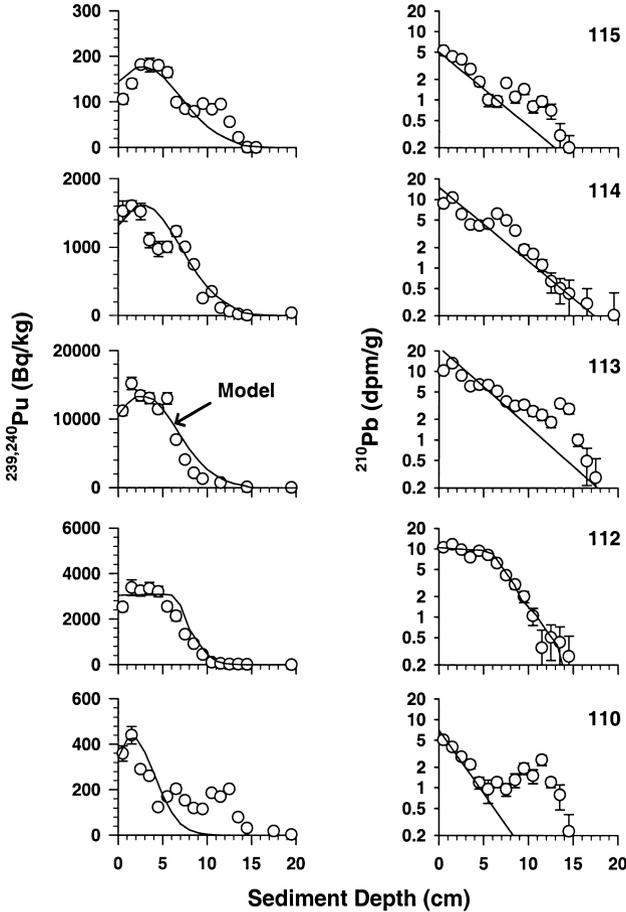
Fig. 6. Measured excess ^{210}Pb and [239,240]Pu sediment-depth profiles have been simulated using a two layer biodiffusion model and the following parameter values: Core 115 ($L = 20$ cm, $D_b^\circ = 0.3$ cm^2/yr, $D_b^\infty = 0$); Core 114 ($L = 20$ cm, $D_b^\circ = 0.3$ cm^2/yr, $D_b^\infty = 0$); core 113 ($L = 20$ cm, $D_b^\circ = 0.25$ cm^2/yr, $D_b^\infty = 0$); core 112 ($L = 5$ cm, $D_b^\circ = 10$ cm^2/yr, $D_b^\infty = 0.05$ cm^2/yr); core 110 ($L = 20$ cm, $D_b^\circ = 0.1$ cm^2/yr, $D_b^\infty = 0$). The sedimentation rate, ω, was assumed to be 0.05 cm/yr for each core. Steady-state and pulsed input functions have been used for [210]Pb$_{ex}$ and [239,240]Pu, respectively. [239,240]Pu model data have been normalised to peak values in each measured profile.

the artificial radionuclides were measurable at the base of the [210]Pb$_{ex}$ distribution, which was usually located between 15 and 20 cm. This result suggests that sediment mixing in each core has occurred at least to the base of the excess ^{210}Pb distributions. Under these conditions, separately resolving the individual sedimentation and mixing rates for each core is impractical. Therefore, the simplifying assumptions were made that (1) mixing occurred at least to 20 cm; (2) the sedimentation rate (based on the ^{14}C results) is 0.05 cm/yr for each core.

The bio-diffusion model, with mixing restricted to the upper 20 cm of the sediments (i.e. $D_b^\infty = 0$; see Fig. 6 caption) results in reasonable agreement between modelled and measured profiles for cores 110, 113, 114 and 115 for values of the biodiffusion coefficient, D_b°, in the range of 0.1–0.3 cm^2/yr. However, the ^{210}Pb$_{ex}$ distribution in core 112 exhibits a plateau near the sediment-water interface that is indicative of more rapid mixing in the upper 5 cm of the core (e.g., $D_b^\circ \cong 10$ cm^2/yr), while some degree of deeper mixing ($D_b^\infty = 0.05$ cm^2/yr) is required to adequately simulate the entire ^{210}Pb$_{ex}$ and ^{239,240}Pu profiles for this core. Several cores (eg. 110 and 115) exhibit anomalous sub-surface maxima in ^{239,240}Pu and ^{210}Pb$_{ex}$ activities. These sub-surface maxima are also correlated with elevated worm tube densities observed in *x*-radiographs of the cores. No attempt was made to model these features, because they are probably non-local mixing artefacts associated with the activities of benthic infauna (Boudreau, 1986b). Simulation of these features would require site-specific, mixing parameters (Smith et al., 1986) having limited general relevance to the Chernaya Bay sediment regime. Despite the simplicity of the biodiffusion model, it does permit a prediction of future ^{239,240}Pu sediment distributions in Chernaya Bay. For example, it can be shown that it will require approximately 35 yr for ^{239,240}Pu concentrations in surface sediments to decrease to 50% of their present values, at which time most of the plutonium inventory will still be retained in the upper 20 cm of the sediments.

3.4. Radionuclide sediment inventories

Radionuclide inventories have been estimated for each core in Chernaya Bay (Table 1) from the sediment porosity and radionuclide distributions. The highest inventories for each radionuclide were measured at the deepest station (Sta. 113) in the central basin and the lowest were measured at Sta. 115 near the entrance to Chernaya Bay. Least-squares regressions show high correlations between the ^{239,240}Pu and ^{137}Cs inventory ($r^2 = 0.997$, $n = 5$) and the ^{239,240}Pu and ^{60}Co inventory ($r^2 = 0.993$, $n = 5$). This result confirms that there was minimal biogeochemical fractionation among the three radionuclides following the nuclear detonation. A value of 11.2 TBq (Table 1) has been calculated for the total ^{239,240}Pu inventory in Chernaya Bay by assuming that the ^{239,240}Pu inventory in each sediment core is representative of bottom sediments bounded by the 50 m isobath (that marks the sill depth) on the sides of the fjord and the midpoints between the positions of each of the cores. Clearly, the lack of knowledge of axial gradients introduces considerable uncertainty into any estimate of total inventories. It is therefore concluded that 11.2 TBq represents a maximum value for the ^{239,240}Pu inventory, because the sampling stations are located along the axis of the fjord where the deposition rates of the fine grained sediments (that most efficiently scavenge ^{239,240}Pu) are probably highest. This inventory of ^{239,240}Pu is greater than the values of 1 and 8.5 TBq estimated to reside in the sediments of Bylot Sound, Thule and Enewetak Lagoon, respectively, and in the marine environment is exceeded only by the 519 TBq of plutonium discharged from the Sellafield nuclear fuel reprocessing plant, currently sequestered in the sediments of the Irish Sea (Kershaw et al., 1995).

Table 1
Total radionuclide inventories (bottom line of table) have been estimated from the sum of individual radionuclide inventories for each station (from box cores) times the sediment area below the 50 m isobath representative of each station

Station	Water depth (m)	Area under 50 m isobath (m$^2 \times 10^6$)	^{239,240}Pu inventory (Bq/m$^2 \times 10^4$)	^{137}Cs inventory (Bq/m$^2 \times 10^4$)	^{60}Co inventory (Bq/m$^2 \times 10^4$)
110	31	—	1.91	0.116	0.028
112	42	27.8	14.0	0.373	0.150
113	76	12.3	56.3	1.07	0.408
114	68	5.4	7.37	0.167	0.0711
115	69	—	0.576	0.0591	0.0046
Total inventory in Chemaya Bay (TBq)			11.2 TBq	0.244 TBq	0.096 TBq

3.5. ^{240}Pu/^{239}Pu and ^{241}Am/^{239,240}Pu isotope ratios

The plutonium in a surface sediment sample collected from Chernaya Bay in 1992 was reported by Smith et al. (1995) to have a ^{240}Pu/^{239}Pu atom ratio of 0.0304 ± 0.0003 as measured by thermal ionisation mass spectrometry (TIMS). This ratio is much lower than the value of 0.18 that is characteristic of soils in northern latitudes contaminated by nuclear fallout. This result labelled this plutonium as low irradiation material typical of debris from the low yield nuclear tests conducted by the US at the Nevada Test Site between 1951 and 1955 (Hicks and Barr, 1984; Smith et al., 1995). This single previous result is confirmed by the present ICP-MS measurements (Table 2) of material from the 1993 expedition which give a mean ^{240}Pu/^{239}Pu atom ratio of 0.0324 ± 0.0003 for four surface sediment samples collected at locations both inside and immediately outside the entrance to Chernaya Bay. These ratios are only slightly greater than ^{240}Pu/^{239}Pu atom ratios of 0.0283 reported by Beasley et al. (1998) for soils contaminated by the 1957 accident at the Mayak nuclear complex in the southern Ural Mountains. The agreement between the ^{240}Pu/^{239}Pu atom ratios in material from Chernaya Bay and Mayak is consistent with the obvious inference that a former USSR production centre is the source of the weapons grade nuclear material detonated in Chernaya Bay, since it is known that only relatively small isotopic changes occur in Pu subjected to the low thermal neutron fluxes typical of these early nuclear weapons devices.

Plutonium in Chernaya Bay is characterised by a ^{241}Pu^{239}Pu atom ratio of 0.00123 ± 0.00006, decay corrected to April 1957 (Smith et al., 1995). This latter date was estimated to be the time of the detonation (or mean time for two or more detonations), based on the present measured concentration ratio of ^{241}Am/^{241}Pu and assuming that all of the ^{241}Am ($t_{1/2} = 433$ yr) in Chernaya Bay sediments was produced by radioactive decay from ^{241}Pu ($t_{1/2} = 14.3$ yr). Again, this value is in close agreement with the value of 0.00138 measured by Beasley et al. (1998) on Mayak soil samples, decay corrected to the time of the accident in 1957. These ^{241}Pu/^{239}Pu atom ratios are an order of magnitude lower than values of 0.015 typical of global

Table 2

Radionuclide inventories (calculated from core profiles), ^{240}Pu/^{239}Pu atom ratios (surface sediments), ^{241}Am/^{239,240}Pu inventory ratios, % of ^{239,240}Pu transported from Chernaya Bay ($f_{CB} \times 100$; from Eq. (2) and ^{239,240}Pu inventory in Barents Sea sediments transported from Chernaya Bay[a]

Station	^{239,240}Pu Inventory (Bq/m^2)	^{137}Cs Inventory (Bq/m^2)	^{240}Pu/^{239}Pu (atom ratio)	^{241}Am/^{239,240}Pu (activity ratio)	% ^{239,240}Pu: Cher. Bay ($f_{CB} \times 100$)	^{239,240}Pu Inventory: Cher. Bay (Bq/m^2)
6	67.8 ± 4.2	470 ± 20	0.1601 ± 0.0035	0.292 ± 0.030	13	9
7	26.4 ± 1.6	101 ± 12	0.1473 ± 0.0046	0.298 ± 0.054	22	6
8	864 ± 20	1092 ± 30	0.0566 ± 0.0004	0.102 ± 0.011	84	725
12	298 ± 10	1128 ± 21	0.0971 ± 0.0027	0.198 ± 0.030	56	167
13	781 ± 24	1644 ± 48	0.0816 ± 0.0006	0.149 ± 0.012	67	523
14	247 ± 8	864 ± 36	0.1057 ± 0.0014	0.158 ± 0.019	51	124
19	45.1 ± 3.6	198 ± 17	0.1199 ± 0.0041	0.267 ± 0.045	41	19
20	99 ± 6	325 ± 26	0.1053 ± 0.0031	0.179 ± 0.028	51	50
21	19.0 ± 2.0	214 ± 15	—	0.394 ± 0.078	0	0
95	23.6 ± 1.8	166 ± 23	—	0.338 ± 0.043	4	1
97	57.3 ± 3.6	196 ± 25	—	0.358 ± 0.035	0	0
98	253 ± 14	672 ± 69	0.0948 ± 0.0018	0.157 ± 0.019	58	146
99	447 ± 20	654 ± 66	0.0607 ± 0.0013	0.104 ± 0.009	81	362
100	300 ± 10	504 ± 39	—	0.082 ± 0.007	89	268
101	320 ± 16	642 ± 63	0.056 ± 0.0013	0.097 ± 0.010	84	270
102	450 ± 17	810 ± 69	0.066 ± 0.0010	0.109 ± 0.010	80	361
103	198 ± 13	894 ± 105	0.1123 ± 0.0023	0.216 ± 0.023	46	91
104	150 ± 10	642 ± 75	0.1459 ± 0.0034	0.291 ± 0.041	23	35
105	61.2 ± 1.8	353 ± 99	0.1391 ± 0.0015	0.294 ± 0.031	28	19
106	652 ± 23	1374 ± 144	—	0.119 ± 0.007	77	502
107	1000 ± 30	936 ± 147	—	0.072 ± 0.004	93	926
108	178 ± 10	990 ± 126	0.1204 ± 0.0023	0.226 ± 0.018	41	72
109	761 ± 29	1572 ± 150	0.0684 ± 0.0009	0.124 ± 0.008	76	578
110	$1.91 \pm 0.08 \times 10^4$	1160 ± 120	0.0334 ± 0.0001	0.053 ± 0.004	100	1.91×10^4
112	$14.0 \pm 0.6 \times 10^4$	3730 ± 180	0.0316 ± 0.0001	0.048 ± 0.005	100	14.0×10^4
113	$56.3 \pm 3.1 \times 10^4$	10700 ± 540	—	0.049 ± 0.004	100	56.3×10^4
114	$7.37 \pm 0.42 \times 10^4$	1670 ± 82	0.0331 ± 0.0002	0.051 ± 0.006	100	7.37×10^4
115	$5.76 \pm 0.24 \times 10^3$	591 ± 25	0.0315 ± 0.0001	0.050 ± 0.005	100	5.76×10^3
116	218 ± 14	1074 ± 102	—	0.231 ± 0.021	39	86
117	55.8 ± 5.4	270 ± 33	0.0898 ± 0.0034	0.129 ± 0.013	61	34
118	52.8 ± 6.6	348 ± 45	—	0.173 ± 0.024	59	31
119	99.0 ± 5.4	495 ± 92	—	0.224 ± 0.021	42	42
120	48.6 ± 3.9	426 ± 42	—	0.173 ± 0.050	59	29
121	38.4 ± 3.6	300 ± 39	—	0.234 ± 0.025	39	24
122	45.6 ± 4.5	384 ± 39	—	0.197 ± 0.026	51	23

[a]*Note*: Uncertainties represent 1 σ counting errors.

fallout (Smith et al., 1995). The low yield of ^{241}Pu has also resulted in a mean ^{241}Am/^{239,240}Pu activity ratio of 0.050 in Chernaya Bay sediments (Table 2) which is much lower than values of 0.3–0.4 that are typical of modern sediments contaminated by atmospheric fallout (Smith et al., 1987).
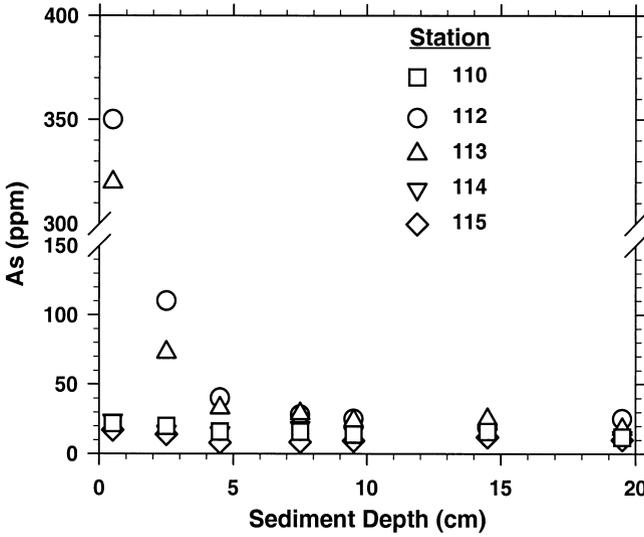
Fig. 7. Arsenic sediment-depth profiles in Chernaya Bay exhibit a diffusional shape, but are most elevated at stations 112 and 113 having the highest ^{239,240}Pu concentrations.

3.6. Arsenic concentrations in Chernaya Bay

Trace metal concentration profiles in Chernaya Bay sediments are characterised by unusually high levels of As which co-vary with ^{239,240}Pu levels, a feature previously reported on a broader scale for Barents Sea sediments (Loring et al., 1995). In fact, the highest levels of As (in excess of 300 ppm) reported for the Barents Sea are those measured near the sediment–water interface of cores 112 and 113 (Fig. 7), the cores having the highest ^{239,240}Pu activities. A linear regression of As concentration on ^{239,240}Pu activity for surface sediments in the Barents Sea (Loring et al., 1995) has a regression coefficient, $r^2 = 0.792$ ($n = 15$). In contrast, As levels measured in sediments from the Kara Sea exhibit lower correlations with artificial radionuclide concentrations (Loring et al., 1998).

Arsenic has a sediment-depth profile characterised by maximum concentrations near the sediment surface, while ^{239,240}Pu exhibits a sub-surface maximum in most cores. The surface enrichment of As is probably caused by the initial burial of particulate As compounds and subsequent reduction, remobilisation and upwards migration in pore waters to the surface oxidised zone (Loring et al., 1998). In the latter region, As undergoes oxidation and precipitation/adsorption onto solid phases in association with Fe and Mn. Although the covariance of As and ^{239,240}Pu in Barents Sea sediments may reflect contamination from the Chernaya Bay nuclear tests, it is surprising that other heavy metals such as Fe, Mn, Cr, Sn and Pb, that might be present in military/industrial contaminants, exhibit minimal enrichment at the sediment-water interface. One possible explanation is that As may have been deliberately incorporated into the nuclear device in order to produce

short-lived, neutron activation products of As as environmental tracers for that specific nuclear test.

3.7. Radionuclide levels in seawater

Radionuclide concentrations measured in unfiltered seawater samples collected in Chernaya Bay are given in Table 3. ^{137}Cs levels of 5–8 Bq/m^3 are similar to those measured in other regions of the Barents and Kara Seas and represent combined inputs from fallout, Chernobyl and European reprocessing plants (Kershaw et al., 1997; Smith et al., 1998). In contrast, ^{239,240}Pu surface water concentrations of 94.9 ± 9.7 mBq/m^3 and 123 ± 12 mBq/m^3 measured at Stations 110 and 113 are an order of magnitude greater than a level of 7.5 mBq/m^3 reported for surface waters of the southern Barents Sea (Strand et al., 1994). ^{239,240}Pu levels in the Barents Sea were higher (> 17 mBq/m^3) in the 1980s owing to long-range transport from the Sellafield nuclear fuel reprocessing plant in the UK (Kershaw and Baxter, 1995). These plutonium levels were distinguished by the elevated ^{238}Pu/^{239,240}Pu ratios (> 0.05), characteristic of a reprocessing plant source, compared to ratios ($\cong 0.03$) typical of fallout (Holm et al., 1986; Kershaw et al., 1995). The ^{238}Pu/^{239,240}Pu ratios measured in seawater in Chernaya Bay lie in a range (0.02–0.04; Table 3) which is consistent with a fallout source and indicate that contributions from long-range transport of plutonium from reprocessing plants are minimal. Higher ^{239,240}Pu levels of 209 ± 21 and 4200 ± 67 mBq/m^3 were measured in unfiltered bottom water samples from Stations 110 and 113, respectively. A similar range of ^{239,240}Pu levels has been measured at highly contaminated sites in Enewetak Lagoon (Nelson and Noshkin, 1973) and at locations near the Sellafield discharge outfall in the Irish Sea (Kershaw et al., 1995). The high ^{239,240}Pu levels in Chernaya Bay bottom water may have resulted from the release of dissolved ^{239,240}Pu from bottom sediments or from the resuspension of plutonium-enriched, surficial sediments.

A simple box model can be used to estimate water column ^{239,240}Pu inventories and residence times in Chernaya Bay. It will be assumed that the Chernaya Bay water column is composed of halocline water, extending from the surface to the sill depth of 50 m, and a deep layer, having ^{239,240}Pu concentrations of 100 and 4200 mBq/m^3, respectively. The ^{239,240}Pu inventory in the water column is then estimated to be 2.1×10^{-3} TBq, 90% of which is in the deep layer. It is further assumed that a steady-state O$_2$ deficit of approximately 0.7 ml/l (estimated from O$_2$ profiles given in Fig. 3), is maintained in the deep layer owing to O$_2$ uptake by benthic infauna. For an O$_2$ respiration rate of 2 ml m^{-2} hr^{-1} for Arctic marine sediments (Anderson et al., 1988), a deep layer residence time of 2 yr would be required to maintain this O$_2$ deficit. A 2 yr residence time for bottom water in Chernaya Bay would result in an annual turnover of 0.5×10^9 m^3 of seawater and a yearly discharge of approximately 10^{-3} TBq to the Barents Sea or approximately 0.01% of the total Chernaya Bay inventory on an annual basis. Although this is a crude calculation it should provide an estimate to the correct order of magnitude of the residence time and present-day ^{239,240}Pu flux from Chernaya Bay.

3.8. Radionuclide uptake in biota in Chernaya Bay

Benthic organisms occupy a particularly important position in the marine ecosystem of the Barents Sea as they provide the main food source for many birds and much of the commercial fishery. During the 1993 RV *Fersman* cruise, 30 benthic organism samples were collected from the five sediment sampling sites in and near Chernaya Bay (Pogrebov et al., 1995). Samples were also collected from benthic locations at stations near the mouth of Chernaya Bay having similar water depths and sediment types. The total biomass of the benthic community measured in Chernaya Bay in 1993 was 246 g/m^2. The benthos is dominated by deposit feeders, with the two species, *Golfingia sp.* and *Maldane sarsi* composing 60% of the total biomass. In contrast, open ocean regimes outside the fjord have a greater proportion of suspension feeders, mainly bivalves and barnacles. The open ocean locations were also distinguished by greater species diversity and an average biomass six times greater than that observed in Chernaya Bay. However, there was no evidence for differences in species mass size or morphology between the macrobenthos in Chernaya Bay compared to open ocean regimes (Pogrebov et al., 1995).

Radionuclide levels measured in benthic invertebrate samples from Chernaya Bay are given in Table 3. Concentration factors, CF (=*contaminant concentration in organism/contaminant concentration in sediment*), in the range of 0.5–10 are commonly measured for heavy metals in deposit feeders in highly contaminated sediment regimes. The concentration of ^{137}Cs in polychaete (27.5 Bq/kg) at Station 110 reflects a concentration factor of only 1.6 (Table 3), although these levels are still significantly greater than the highest ^{137}Cs values (17 Bq/kg) in polychaete measured elsewhere in the Barents Sea in 1993–1994 (Matishov et al., 1995). ^{60}C is more highly concentrated in polychaete with levels of 43.5 Bq/kg in this organism reflecting a concentration factor of about 12. ^{239,240}Pu levels in polychaete (1292 Bq/kg) are also much higher than levels of 295 Bq/kg measured in proximal sediments (CF = 4.4) and many orders of magnitude greater than levels (<1 Bq/kg) typically measured in benthic regimes contaminated solely by fallout. In contrast, ^{239,240}Pu levels in the suspension feeding, bivalve, *Macoma calcarea* (104 Bq/kg) collected at Sta. 114 were lower than levels (1390 Bq/kg) in the proximal sediments. A similar CF of 0.1 was measured in *Macoma* in Bylot Sound, Thule, where the biomass of the *Macoma* community (100 g/m^2) was close to that measured in Chernaya Bay (Aarkrog et al., 1987). Using a *Macoma* biomass of 148 g/m^2 (Pogrebov et al., 1995), a CF of 0.07 and surface (0–1 cm) ^{239,240}Pu sediment concentrations for the stations in Chernaya Bay, the inventory of ^{239,240}Pu in the *Macoma* community is estimated at 2.4×10^{-3} TBq. This is an order of magnitude greater than the ^{239,240}Pu inventory of 1.9×10^{-4} TBq estimated for the *Macoma* community in Bylot Sound (Aarkrog et al., 1987).

Plutonium is efficiently accumulated by benthic algae and, as a result, ^{239,240}Pu levels are comparatively high in *Laminaria* (5.1 Bq/kg) and *Fucus* (14.7 Bq/kg) samples collected at Station 110 in Chernaya Bay (Table 3). These ^{239,240}Pu levels are almost two orders of magnitude greater than those in algae collected from other locations in the Barents Sea in 1993 and 1994 (Ikaheimonen et al., 1997) where contamination is mainly from fallout. ^{239,240}Pu levels in both algae and surface water at Station 110

Table 3
Radionuclide concentrations in environmental samples from Chernaya Bay

| Station	Sample matrix	Units	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{241}Am/^{239,240}Pu	^{137}Cs	^{60}Co
110	Sediment (0–5 cm)	Bq/kg (dry wt.)	295 ± 10	0.0340 ± 0.0024	0.0531 ± 0.0036	17.0 ± 2.7	3.5 ± 1.2
	Polychaete (0–5 cm) (*Spiochaetoperus*)	Bq/kg (dry wt.)	1292 ± 106	0.0345 ± 0.0070	0.0533 ± 0.0050	27.5 ± 3.8	43.5 ± 5.1
	Algae (*Laminaria sp.*)	Bq/kg (dry wt.)	5.12 ± 0.92	0.029 ± 0.010	0.0254 ± 0.0073	13.3 ± 2.5	< 4.7
	Algae (*Fucus sp.*)	Bq/kg (dry wt.)	14.7 ± 1.2	0.032 ± 0.011	0.0442 ± 0.0072	5.2 ± 2.7	< 3.8
	Sea urchin (*Strongglocentrotus*)	Bq/kg (dry wt.)	22.2 ± 1.9	0.039 ± 0.012	0.0541 ± 0.0049	2.9 ± 1.4	< 3.6
	Seawater (2 m)	Bq/m³	0.095 ± 0.010	0.059 ± 0.022	—	5.48 ± 0.20	—
	Seawater (25 m)	Bq/m³	0.209 ± 0.021	0.023 ± 0.012	—	5.81 ± 0.22	—
113	Seawater (1 m)	Bq/m³	0.123 ± 0.012	0.040 ± 0.017	—	5.64 ± 0.25	—
	Seawater (71 m)	Bq/m³	4.200 ± 0.067	0.0267 ± 0.0043	—	7.56 ± 0.14	—
114	Sediment (0–5 cm)	Bq/kg (dry wt.)	1390 ± 28	0.0273 ± 0.0019	0.0511 ± 0.0061	31.0 ± 2.1	12.7 ± 2.1
	Mollusc (0–5 cm) *Macoma*	Bq/kg (dry wt.)	104 ± 10	0.024 ± 0.011	0.0647 ± 0.0071	1.25 ± 0.50	1.24 ± 0.66

are an order of magnitude greater than [239,240]Pu levels in algae and seawater at the weapons accident site near Thule, Greenland (Aarkrog et al., 1984). However, the [239,240]Pu concentration ratios, CR (= *Bq/kg dry wt. in plant/Bq l in seawater*) are approximately the same ($1–2 \times 10^4$) at both locations indicating that this parameter is a robust indicator of the [239,240]Pu partitioning between algae and seawater.

Ten species of *ciliophora* were also collected from the five sampling locations, Stations 110–115 (Pogrebov et al., 1995). Samples collected at Sta. 115 were observed to be of similar composition (1–3 species per sample) and morphology to those collected at open ocean control sites. However, the samples collected at Station 110 were distinguished by changes in morphology and movement character compared to samples from control sites (Pogrebov et al., 1995). Further, sediments at Stations 112, 113 and 114, having the highest levels of [239,240]Pu were found to be totally barren of infusorians. These results indicate that, although elevated levels of plutonium in Chernaya Bay sediments have had little effect on the macrobenthos, there may have been some deleterious impact on the meiobenthos (Pogrebov et al., 1995).

3.9. [239,240]Pu transport from Chernaya Bay into the Arctic Ocean

Radionuclide activities were measured on sediment subsamples (2 cm intervals) in the upper 6 cm of grab samples collected in the eastern Barents Sea in 1993 (Fig. 1). [137]Cs, [239,240]Pu and [241]Am inventories were calculated assuming that the upper 6 cm of the sediment column retained 55% of the radionuclide inventory, as was generally observed in box cores collected during the 1992, 1993 and 1996 Geolog Fersman cruises in the Barents Sea (Smith et al., 1995). During the 1992 Dalnie Zelentsy cruise, the upper 2 cm of sediment was subsampled from grab samples collected at stations (open circles) indicated in Fig. 1 (Smith et al., 1995). In these cases, the radionuclide inventory was estimated assuming that the upper 2 cm represented 15% of the total inventory in the core (Table 2). The ^{137}Cs inventories calculated by this method are contoured in Fig. 8(a) for the eastern Barents Sea. The ^{137}Cs inventories are highly correlated with fine particle concentrations (percentage clay + silt; $r^2 = 0.850$; $P \leqslant 0.001$) and total organic carbon ($r^2 = 0.872; P \leqslant 0.001$) and as a result, the ^{137}Cs inventories tend to conform to the bathymetry. In contrast [239,240]Pu inventories (Fig. 8(b)) have a lower correlation with percentage clay + silt ($r^2 = 0.504; P \leqslant 0.01$) and total organic carbon ($r^2 = 0.626, P \leqslant 0.01$) and tend to be more concentrated closer to the mouth of Chernaya Bay. This latter observation is supported by a higher inverse correlation measured between radionuclide inventory and distance from Chernaya Bay for [239,240]Pu ($r^2 = 0.776$, $n = 30$) compared to [137]Cs ($r^2 = 0.616$,

Fig. 8. (a) Contours of ^{137}Cs inventories (Bq/m^2), estimated for grab and box core samples, tend to conform to the bathymetry of the eastern Barents Sea with highest values associated with finer-grained, basin sediments. (b) [239,240]Pu inventory (Bq/m^2) contours tend to be more centred around the entrance to Chernaya Bay. (c) Contours of the [239,240]Pu inventory derived exclusively from transport from Chernaya Bay illustrate the northwestward transport of [239,240]Pu carried by the Litke Current. (d) Contours illustrating the percentage of the total [239,240]Pu inventory derived from Chernaya Bay have been calculated from the [240]Pu/[239]Pu and [241]Am/[239,240]Pu ratio data for each core.
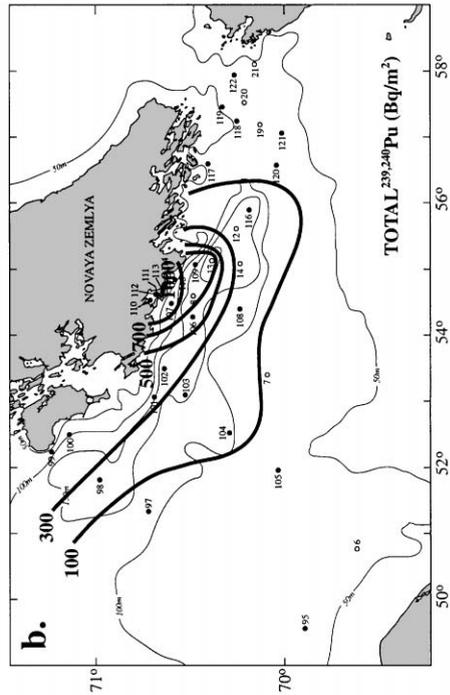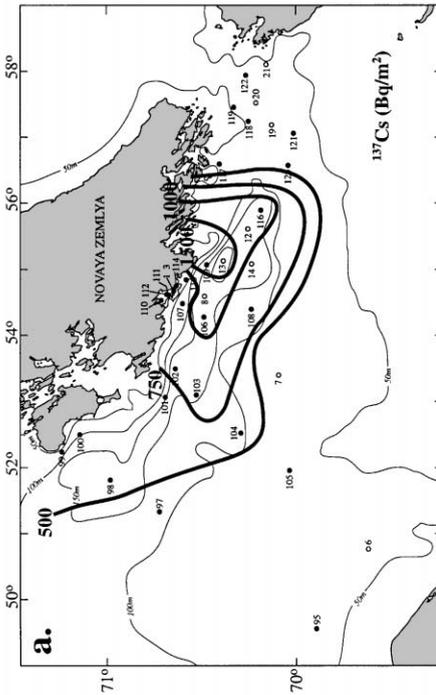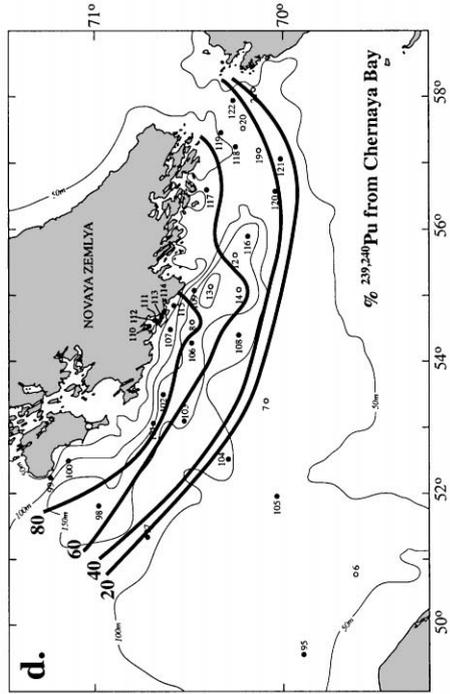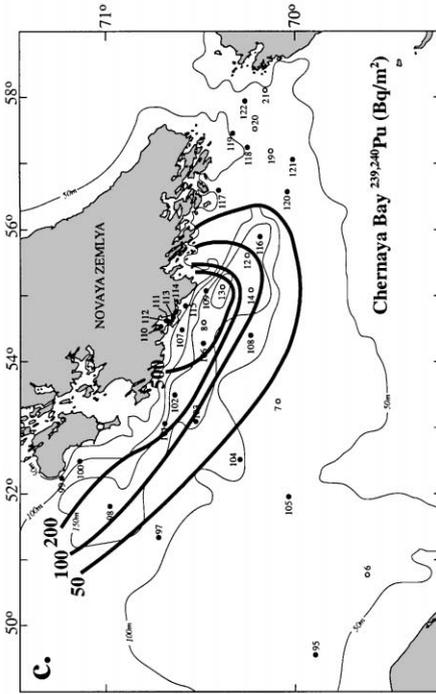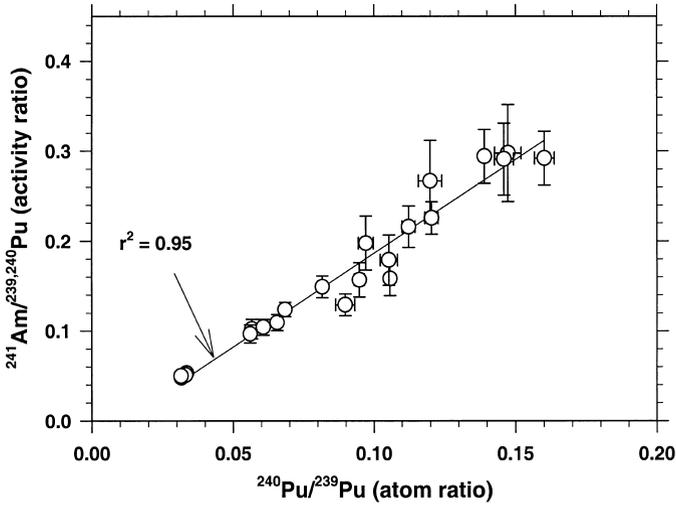
Fig. 9. Linear regression of total $^{241}Am/^{239,240}Pu$ activity ratios on $^{240}Pu/^{239}Pu$ atom ratios in Barents Sea sediments has a relatively high regression coefficient, r^2, of 0.95 ($P < 0.001$) that is consisent with the plutonium inventory being an end member mixture of fallout and Chernaya Bay components.

$n = 30$). These results support the hypothesis that the principle sources of ^{137}Cs in the Barents Sea are non-localised inputs from fallout and Sellafield while the sources of $^{239,240}Pu$ are mainly atmospheric fallout and suspended sediments transported from Chernaya Bay. These results are in contrast to those reported by Baskaran et al. (1996) who concluded that plutonium inventories in Kara Sea sediments were entirely associated with atmospheric fallout.

The hypothesis that the plutonium inventory in eastern Barents Sea sediments consists of a mixture of Chernaya Bay and fallout plutonium can be further tested by comparisons of the $^{240}Pu/^{239}Pu$ and $^{241}Am/^{239,240}Pu$ isotope ratios. $^{240}Pu/^{239}Pu$ atom ratios in Chernaya Bay sediments and atmospheric fallout are 0.033 and 0.18 (Krey et al., 1976), respectively, while the $^{241}Am/^{239,240}Pu$ activity ratios for Chernaya Bay and atmospheric fallout are approximately 0.05 and 0.35, respectively (Table 2). A linear regression of $^{241}Am/^{239,240}Pu$ activity on $^{240}Pu/^{239}Pu$ atom ratios, shown for samples collected throughout the eastern Barents Sea in Fig. 9, has a regression coefficient, r^2, of 0.95 ($n = 22$), indicating that a linear relationship exists between the two pairs of isotope ratios. This concordance confirms that the plutonium inventories can be considered to be end member mixtures of Chernaya Bay and fallout plutonium. Under these conditions, the fraction, f_{CB}, of Chernaya Bay $^{239,240}Pu$ in a sediment sample having a $^{240}Pu/^{239}Pu$ ratio, R_s, is given by Krey et al. (1976) as

$$f_{CB} = (R_f - R_s)/(R_f - R_{CB}) \tag{2}$$

where R_{CB} and R_f are the $^{240}Pu/^{239}Pu$ atom ratios of Chernaya Bay and fallout plutonium, respectively. Assuming that there is minimal fractionation between ^{241}Am and its parent, ^{241}Pu subsequent to ^{241}Am ingrowth, then Eq. (2) can also be applied

to measured ^{241}Am/^{239,240}Pu ratios (R_s) in sediment samples, using end member ^{241}Am/^{239,240}Pu ratios for Chernaya Bay (R_{CB}) and fallout (R_f) material. This is a less robust technique for determining relative contributions from low and high irradiation nuclear sources compared to the use of ^{240}Pu/^{239}Pu ratios, because it has higher analytical uncertainties, but it also has the advantage of using alpha spectroscopy rather than ICP-MS or thermal ionisation mass spectrometry (TIMS).

Eq. (2) has been applied to ^{240}Pu/^{239}Pu results from surface sediments and ^{241}Am/^{239,240}Pu results for sediment inventories for the eastern Barents Sea and the results are given in Table 2. Values of f_{CB} given in Table 2 were calculated using the ^{240}Pu/^{239}Pu ratio data except when this data was unavailable, in which case the results were calculated using the ^{241}Am/^{239,240}Pu data. The inventory of ^{239,240}Pu derived from Chernaya Bay is contoured in Fig. 8(c) while the percent of the total ^{239,240}Pu inventory derived from Chernaya Bay is contoured in Fig. 8(d). The influence of Chernaya Bay discharges decreases rapidly with distance seaward into the eastern Barents Sea, but extends for much greater distances (> 100 km) northwestward along the Novaya Zemlya coastline. This is consistent with circulation patterns which indicate that longshore sediment transport occurs northward along the southern Novaya Zemlya coastline, mainly as a result of the Litke Current flowing westward through the Strait of Karskiye Vorota or Kara Gate (Pfirman et al., 1995, 1997). Although the actual magnitude of plutonium transport southeastward into the shallower, coarser-grained sediments of the Barents Sea is relatively small, Chernaya Bay ^{239,240}Pu still represents more than 40% of the total ^{239,240}Pu inventory (Fig. 8(d)) in much of the region off the southern coast of Novaya Zemlya. The inventory of Chernaya Bay plutonium in the eastern Barents Sea, estimated by linear interpolation between adjacent stations from the contour plots in Fig. 8(c), is 2.0 TBq. This can be compared to the 11.2 TBq of ^{239,240}Pu still retained in the sediments of Chernaya Bay. However, the decreasing ^{239,240}Pu inventory gradient, northwestward along the Novaya Zemlya coastline, is relatively small, suggesting that a significant additional quantity of ^{239,240}Pu could have been transported farther along the Novaya Zemlya coastline by the Litke Current.

In Section 3.4 it was estimated from a water residence time calculation that the present day flux of ^{239,240}Pu to the Barents Sea is approximately 10^{-3} TBq/yr, a rate that would correspond to a cumulative discharge of only 0.04 TBq over the past 40 yr. This cumulative discharge is less than 2% of the ^{239,240}Pu inventory derived from Chernaya Bay that is presently observed in sediments in the eastern Barents Sea. This low current rate of ^{239,240}Pu discharge from Chernaya Bay suggests that most of the measured 2 TBq of ^{239,240}Pu from Chernaya Bay was transported into the Barents Sea at the time of the initial nuclear detonations.

Recently, Huh et al. (1997) measured ^{239,240}Pu inventories in sediments collected during the 1994 Arctic Ocean Section (AOS) icebreaker cruise across the central Arctic Ocean. They reported relatively low ^{240}Pu/^{239}Pu atom ratios (0.09–0.16) in surface sediments, indicating that 20–60% of the plutonium in the central Arctic Ocean is derived from low yield, weapons grade sources. The results of Huh et al. (1997) also indicate that much of the low ^{240}Pu/^{239}Pu plutonium has been deposited in the deeper, high latitude basins of the central Arctic Ocean rather than over the

large, polar continental shelves. If their limited data set for AOS is assumed to be representative of the entire Arctic Ocean then it can be estimated that the total inventories of low yield, weapons grade plutonium are 0.18 and 0.06 TBq in the Eurasian and Makarov Basins, respectively.

Chernaya Bay is one of the few documented sources of low $^{240}Pu/^{239}Pu$ ratio plutonium known to have been discharged directly into the Arctic Ocean. Although low $^{240}Pu/^{239}Pu$ ratio plutonium was also released from Sellafield in the early 1960s, the largest plutonium releases from Sellafield occurred during the 1970s when $^{240}Pu/^{239}Pu$ ratios were greater than 0.15 (Kershaw et al., 1995). It was noted above that a significant quantity of low $^{240}Pu/^{239}Pu$ ratio plutonium appears to have been transported northwestward along the Novaya Zemlya coastline. Plutonium transported along the western coastline of Novaya Zemlya via the Litke and Western Novaya Zemlya Currents (Pfirman et al., 1997) could have been carried into the Eurasian Basin through the Santa Anna Trough (Fig. 1), either in colloidal or dissolved form or in very fine particulate phases, and subsequently transported through the Arctic Ocean in the Barents Sea branch of the Atlantic water flow (Rudels et al., 1994; Frank et al., 1998). It would only require an additional 0.2–0.3 TBq of Chernaya Bay plutonium, or approximately 10–15% of the inventory already measured in the eastern Barents Sea, to account for the low $^{240}Pu/^{239}Pu$ ratio, plutonium inventories recently measured in the Eurasian and Makarov Basins. This hypothesis will be addressed in future work that will examine the distribution of plutonium in sediments from the western coastline of Novaya Zemlya to determine if there is a plume of low $^{240}Pu/^{239}Pu$ plutonium delineating the transport pathway from Chernaya Bay into the central basins of the Arctic Ocean.

4. Conclusions

1. Elevated levels of artificial radionuclides, as high as 15,000 Bq/kg for $^{239,240}Pu$, 250 Bq/kg for ^{137}Cs and 100 Bq/kg for ^{60}Co, were measured in sediment cores collected in Chernaya Bay, Novaya Zemlya which have been contaminated by at least two underwater nuclear weapons tests conducted in the 1950s. The $^{239,240}Pu$ inventory in Chernaya Bay sediments estimated from sediment-depth concentration profiles in four box cores is approximately 11 TBq.
2. Sediment-depth distributions of $^{239,240}Pu$ and other artificial radionuclides are consistent with the results of a biodiffusion model, whose parameters are constrained by ^{210}Pb sediment-depth distributions. The results are consistent with low sedimentation rates ($\cong 0.05$ cm/yr) in Chernaya Bay and indicate that downward transport of radioactive contaminants is governed mainly by bioturbation with the entire inventory being retained in the upper 20 cm of the sediments.
3. Arsenic levels are elevated (350 μg/g) in the most contaminated regions of Chernaya Bay and exhibit a high correlation with $^{239,240}Pu$ throughout the eastern Barents Sea for reasons that remain unknown.
4. Elevated levels of $^{239,240}Pu$ in surface (123 mBq/m^3) and bottom (4200 mBq/m^3) water at Station 113 in the central basin of Chernaya Bay are maintained as a result

of the release of dissolved ^{239,240}Pu from surface sediments or the resuspension of plutonium-enriched, bottom material. However, only limited exchange of high ^{239,240}Pu, bottom water appears to occur across the fjord sill with Barents Sea water.

5. Significant uptake of ^{239,240}Pu has occurred in Chernaya Bay from contaminated sediments into benthic invertebrates (1292 and 104 Bq/kg in polychaete and *Macoma*, respectively) and from seawater into brown algae (5.1 and 14.7 Bq/kg in *Laminaria* and *Fucus*, respectively). The latter measurements are consistent with concentration ratios ($\cong 10^4$) similar to those measured in other highly contaminated marine environments.

6. The low ^{240}Pu/^{239}Pu atom and ^{241}Am/^{239,240}Pu activity ratios (0.03 and 0.05, respectively) which characterise Chernaya Bay plutonium (compared to fallout values of 0.18 and 0.35, respectively) have been used to track this signal over distances greater than 100 km into the Barents Sea. At least 2 TBq of ^{239,240}Pu has been transported from Chernaya Bay into offshore depositional basins and north-westward along the Novaya Zemlya coastline. It is possible that low ^{240}Pu/^{239}Pu plutonium from Chernaya Bay has also caused some contamination of the central basins of the Arctic Ocean.

Acknowledgements

References

Anderson, L.G., Jones, E.P., Lindegren, R., Rudels, B., Sehlstedt, P.l., 1988. Nutrient regeneration in cold, high salinity bottom water of the Arctic shelves. Continental Shelf Research 8 (12), 1345–1355.

Aarkrog, A., Dahlgaard, H., Nilsson, K., Holm, E., 1984. Further studies of plutonium and americium at Thule Greenland. Health Physics 46, 29–44.

Aarkrog, A., Boelskifte, S., Dahlgaard, H., Duniec, S., Holm, E., Smith, J.N., 1987. Studies of transuranics in an arctic marine environment. Journal of Radioanalytical and Nuclear Chemistry 115, 39–50.

Baskaran, M., Asbill, S., Santschi, P., Brooks, J., Champ, M., Adkinson, D., Colmer, M., Makeyev, V., 1996. Pu, ^{137}Cs and excess ^{210}Pb in Russian Arctic sediments. Earth and Planetary Science Letters 140, 243–257.

Beasley, T.M., Kelley, J.M., Orlandini, K.A., Bond, L.A., Aarkrog, A., Trapeznikov, A.P., Pozolotina, V.N., 1998. Isotopic Pu, U and Np signatures in soils from Semipalatinsk-21, Kazakh Republic and southern Urals, Russia. Journal of Environmental Radioactivity 39, 215–230.

Boudreau, B.P., 1986a. Mathematics of tracer mixing in sediments. 1. Spatially dependent, diffusive mixing. American Journal of Science 286, 161–198.

Boudreau, B.P., 1986b. Mathematics of tracer mixing in sediments: 11. Non-local mixing and biological conveyor-belt phenomena. American Journal of Science 286, 199–238.

Boyarsky, P.V., 1993. In: Boyarsky, P.V. (Ed.), The Integrated Marine Arctic Expedition Proceeding. Novaya Zemlya, Vol. 1, Russian Federation Ministry of Culture, Moscow, Russia, pp. 96–97.

Frank, M., Smethie Jr., W.M., Bayer, R., 1998. Investigation of subsurface water flow along the continental margin of the Eurasian Basin using the transient tracers tritium, ^{3}He and CFCs. Journal of Geophysical Research 103 (C13), 30773–30792.

Guinasso Jr., L., Schink, D.R., 1975. Quantitative estimates of biological mixing rates in abyssal sediments. Journal of Geophysical Research 80, 3032–3043.

Hicks, H.G., Barr, D.W., 1984. Nevada Test Site Fallout Atom Ratios: ^{240}Pu/^{239}Pu and ^{241}Pu^{239}Pu. UCRL-53499/1. Lawrence Livermore National Laboratory, 4 p.

Holm, E., Aarkrog, A., Ballestra, S., Dahlgaard, H., 1986. Origin and isotopic ratios of plutonium in the Barents and Greenland Seas. Earth and Planetary Science Letters 79, 27–32.

Holm, E., Gastaud, J., Oregioni, B., Aarkrog, A., Dahlgaard, H., Smith, J.N., 1988. Chemical partitioning of plutonium and americium in sediments from the Thule, Greenland region. In: Guary, J.C., Guegueniat, P., Pentreath, R.J. (Eds.), Radionuclides: A Tool for Oceanography. Elsevier Science, London, pp. 351–362.

Huh, C.-A., Pisias, N.G., Kelley, J.M., Maiti, T.C., Grantz, A., 1997. Natural radionuclides and plutonium in sediments from the Western Arctic Ocean: sedimentation rates and pathways of radionuclides. Deep-Sea Research II 44 (8), 1725–1745.

lkaheimonen, T.K., Rissanen, K., Matishov, D.G., Matishov, G.G., 1997. Plutonium in fish, algae and sediments in the Barents, Petshora and Kara Seas. Science of the Total Environment 202, 79–87.

Kershaw, P.J., Baxter, A.J., 1995. The transfer of reprocessing wastes from NW Europe to the Arctic. Deep-Sea Research II 42 (6), 1413–1448.

Kershaw, P.J., Gurbutt, P., Woodhead, D., Leonard, K., Rees, J., 1997. Estimates of fluxes of ^{137}Cs in northern waters from recent measurements. Science of the Total Environment 202, 211–223.

Kershaw, P.J., Woodhead, D.S., Lovett, M.B., Leonard, K.S., 1995. Plutonium from European reprocessing operations-its behaviour in the marine environment. Applied Radiation Isotopes 46 (11), 1121–1134.

Krey, P.W., Hardy, E.P., Pachucki, C., Rourke, F., Coluzza, J., Benson, W.L., 1976. Mass isotopic composition of global fallout in plutonium in soil. In: Transuranium Nuclides in the Environment. IAEA-SM-199/39, IAEA, Vienna, pp. 671–678.

Loring, D.H., Naes, K., Dahle, S., Matishov, G.G., 1995. lllin, G., 1995. Arsenic, trace metals and organic micro contaminants in sediments from the Pechora Sea. Russia. Marine Geology 128, 153–167.

Loring, D.H., Dahle, S., Naes, K., Santos, J.Dos, Skei, J.M., Matishov, G.G., 1998. Arsenic and other trace metals in sediments from the Kara Sea and the Ob and Yenisey estuaries, Russia. Aquatic Geochemistry 4, 233–252.

Matishov, D.G., Matishov, G.G., Rissanen, K., 1995. Peculiarities of radionuclide accumulation in benthic organisms and fish of the Barents and Kara Seas. In: Strand, P., Cooke, A. (Eds.), International Conference on Environmental Radioactivity in the Arctic, Oslo, Norway, August 21–25, 1995. Symposium Proceedings, pp. 233–237.

Matuschenko, A.M., Kaurov, G.A., Krasilov, G.A., Kharitonov, K.V., 1994. Nuclear testing ground unclassified (dates and events). In: Boyarsky, P.V., (Ed.), Integrated Marine Arctic Expedition, Novaya Zemlya, Vol. 3. pp. 54–67 (in Russian).

Mulsow, S., Boudreau, B.P., Smith, J.N., 1998. Bioturbation and porosity gradients. Limnology and Oceanography 43 (1), 1–9.

Nilsen, T., Bohmer, N., 1994. Sources to (sic) radioactive contamination in Murman'sk and Arkhangel'sk counties. In: Bellona Report, Vol. 1. The Bellona Foundation, Oslo, Norway.

Nelson, V., Noshkin, V.E., 1973. Enewetak Survey Report. USAEC Nevada Operations Office. Las Vegas, Nevada. NVO-140, pp. 131–224.

Pfirman, S.L., Kogeler, J., Anselme, B., 1995. Coastal environments of the western Kara and eastern Barents Seas. Deep-Sea Research II 42, 1391–1412.

Pfirman, S.L., Kogeler, J.W., Rigor, I., 1997. Potential for rapid transport of contaminants from the Kara Sea. Science of the Total Environment 202, 111–122.

Polyak, L., Lehman, S.J., Gataullin, V., Jull, A.J.T., 1995. Two-step deglaciation of the southeastern Barents Sea. Geology 23, 567–571.

Pogrebov, V.B., Fokin S.I., Kiyko, O.A., 1995. Benthos communities of Gouba Chernaya (Novaya Zemlya Test Site) status 30 years after underwater explosions. unpublished manuscript.

Rudels, B., Jones, E.P., Anderson, L.G., 1994. On the intermediate depth waters of the Arctic Ocean. In: Johannessen, O.M., Muench, R.D., Overland, J.E. (Eds.), The Polar Oceans and Their Role in Shaping the Global Environment: The Nansen Centennial Volume, Geophysical Monograph Series 85. AGU, Washington, DC., pp. 33–46.

Salbu, B., Nikitin, A.I., Strand, P., Christensen, G.C., Chumichev, V.B., Lind, B., Fjelldal, H., Bergan, T.D., Rudjord, A.L., Sickel, M., Valetova, N.K., Foyn, L., 1997. Radioactive contamination from dumped nuclear waste in the Kara Sea – results from the joint Russian–Norwegian expeditions in 1992–1994. Science of the Total Environment 202, 185–198.

Sayles, F.L., Livingston, H.D., Panteleyev, G.P., 1997. The history and source of particulate ^{137}Cs and ^{239,240}Pu deposition in sediments of the Ob River Delta, Siberia. Science of the Total Environment 202, 25–41.

Smith, J.N., Walton, A., 1980. Sediment accumulation rates and geochronologies measured in the Saguenay Fjord using the ^{210}Pb dating method. Geochimica et Cosmochimica Acta 44, 225–240.

Smith, J.N., Boudreau, B.P., Noshkin, V., 1986. Plutonium and ^{210}Pb distributions in northeast Atlantic sediments: subsurface anomalies caused by non-local mixing. Earth and Planetary Science Letters 81, 15–28.

Smith, J.N., Ellis, K.M., 1995. Radionuclide tracer profiles at the CESAR Ice Station and Canadian Ice Island in the Western Arctic Ocean. Deep-Sea Research II 42 (6), 1447–1470.

Smith, J.N., Ellis, K.M., Aarkrog, A., Dahlgaard, H., Holm, E., 1994. Sediment mixing and burial of the Pu-239,240 pulse from the 1968 Thule, Greenland nuclear weapons accident. Journal of Environmental Radioactivity 25, 135–159.

Smith, J.N., Ellis, K.M., Kilius, L.R., 1998. ^{129}I and ^{137}Cs tracer measurements in the Arctic Ocean. Deep-Sea Research I 45, 959–984.

Smith, J.N., Ellis, K.M., Naes, K., Dahle, S., Matishov, D., 1995. Sedimentation and mixing rates of fallout radionuclides in Barents Sea sediments off Novaya Zemlya. Deep-Sea Research II 42 (6), 1471–1493.

Smith, J.N., Ellis, K.M., Nelson, D.M., 1987. Time-dependent modelling of fallout radionuclide transport through a drainage basin; significance of "slow" erosional and "fast" hydrological components. Chemical Geology 63, 157–180.

Strand, P., Nikitin, A., Rudjord, A.L., Salbu, B., Christensen, G., Foyn, L., Kryshev, I.I., Chumichev, V.B., Dahlgaard, H., Holm, E., 1994. Survey of artificial radionuclides in the Barents Sea and the Kara Sea. Journal of Environmental Radioactivity 25, 99–113.

Yablokov, A.V., Karasev, V.K., Rumyantsev, V.M., Kokeyev, M., Petrov, O.I., Lystov, V.N., Melyanenkov, A. F., 1993. Russian Federation State Report On Sea Burial Of Radioactive Wastes. Admin. Pres. Russ. Fed., Moscow, Russia, 108 pp.