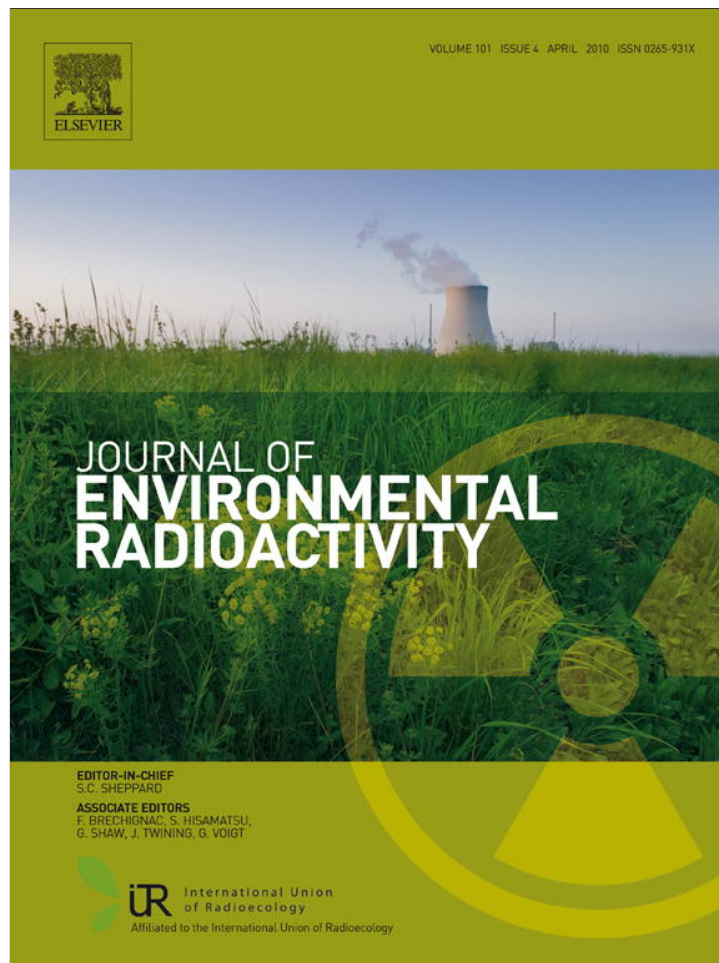


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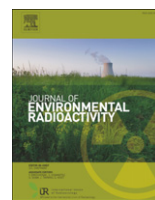
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journal homepage: www.elsevier.com/locate/jenvradSources and distributions of ^{137}Cs , ^{238}Pu , $^{239,240}\text{Pu}$ radionuclides in the north-western Barents SeaAgata Zaborska^{a,*}, Jerzy Wojciech Mietelski^b, JoLynn Carroll^c, Carlo Papucci^d, Janusz Pempkowiak^a^a Institute of Oceanology, Polish Academy of Sciences, Sopot, Poland^b Institute of Nuclear Physics, Polish Academy of Sciences, Kraków, Poland^c Akvaplan-niva, Polar Environmental Center, Tromsø, Norway^d The Italian National Agency for New Technologies, Energy and the Environment, La Spezia, Italy

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ABSTRACT

Sediment deposits are the ultimate sink for anthropogenic radionuclides entering the marine environment. The major sources of anthropogenic radionuclides to the Barents Sea are fallout from nuclear weapons tests, long range transport from other seas, and river and non-point freshwater supplies. In this study we investigated activity concentrations, ratios, and inventories of the anthropogenic radionuclides, ^{137}Cs , ^{238}Pu , $^{239,240}\text{Pu}$ in dated sediment cores collected along a north-south transect in the northwestern Barents Sea. The data were used to evaluate the influence of different sources on the derived spatial and temporal patterns of anthropogenic radionuclides in seafloor sediment deposits. Activity concentrations of ^{137}Cs ranged from <0.1 Bq/kg to 10.5 Bq/kg while $^{239,240}\text{Pu}$ ranged from <0.01 Bq/kg to 2.74 Bq/kg and ^{238}Pu activity concentrations ranged from <0.01 Bq/kg to 0.22 Bq/kg. Total inventories of ^{137}Cs ranged from 29.5 ± 1.5 Bq/m² to 152.7 ± 5.6 Bq/m² and for $^{239,240}\text{Pu}$ inventories (6 sediment layers only) ranged from 9.5 ± 0.3 Bq/m² to 29.7 ± 0.4 Bq/m². Source contributions varied among stations and between the investigated radionuclides. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios up to 0.18 indicate discharges from nuclear fuel reprocessing plants as a main contributor of plutonium. Based on $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio, it was calculated that up to 19–27% of plutonium is supplied from sources other than atmospheric global fallout. Taking into account Atlantic current flow trajectories and that both activity concentrations and inventories of plutonium negatively correlate with latitude, Sellafield is a major source for the Barents Sea. Concentrations and inventories of ^{137}Cs correlate positively with latitude and negatively with distance from the Svalbard archipelago. The ^{137}Cs concentrations are highest in an area of intensive melting of sea ice formed along the Siberian coast. Thus, sea ice and supplies from Svalbard may be important source of ^{137}Cs to the Barents Sea seafloor.

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1. Introduction

Caesium-137 (half life = 30.2 years), $^{239,240}\text{Pu}$ (half life = 24.1×10^3 and 6.5×10^3 years) and ^{238}Pu (half life = 88 years) are ubiquitous in the environment primarily as a result of nuclear bomb testing that began in 1945 and peaked in the late 1950's and early 1960's. Discharges from nuclear reprocessing plants (Sellafield, La Hague) and accidental releases (e.g. Chernobyl accident) are additional sources to the marine environment. The major sources of plutonium and ^{137}Cs to the Arctic are believed to be global fallout and discharges from European reprocessing plants respectively (Matishov et al., 1999).

In the ocean, plutonium radionuclides are readily scavenged from the water column onto sinking particles. The ^{137}Cs is less particle reactive and, in marine environments, it is mainly derived from terrestrial sources (already attached to soil particles) (Baskaran et al., 1996). The $^{239,240}\text{Pu}$ and ^{137}Cs are often used as time markers in geochronology investigations; Carroll and Lerche, 2003; Zaborska et al., 2008) and as bioturbation tracers (Cochran, 1985; Smith et al., 1995; Roberts et al., 1997; Carroll et al., 2008). Sediment radionuclide activity concentrations and activity ratios ($^{239,240}\text{Pu}/^{137}\text{Cs}$, $^{238}\text{Pu}/^{239,240}\text{Pu}$), have also been extensively used to identify sources of radionuclides to the marine environment (Smith et al., 1995, 2000; Baskaran et al., 1996; Efurud et al., 2005). If global fallout is the only source of these radionuclides, the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio, in 1983 was 0.028 (Beck and Krey, 1983) and today is 0.044 after correcting for radioactive decay. This ratio however, cannot be used for definitive determination of radionuclide sources, due to

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different geochemical behaviour (fractionation) of caesium and plutonium. More often, the $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio is used to establish sources of plutonium to the marine environment. Ratios characteristic for global fallout should be about 0.028 (Smith et al., 2000), higher $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios of ~ 0.3 are characteristic for discharges from reprocessing plants (e.g. Sellafield) (MacKenzie et al., 1998; Vintrò et al., 2000) and accidental releases of plutonium isotopes (e.g. Chernobyl) (Mietelski and Wąs, 1995; Ketterer et al., 2004).

In the present investigation, we examined the distribution of ^{137}Cs and plutonium in the Barents Sea. The Barents Sea is a highly productive Arctic shelf sea, accounting for $\sim 50\%$ of the total pan-Arctic primary production of the Arctic coastal shelf seas (Sakshaug, 2004). It exhibits highly dynamic hydrographic features due to changing supplies and mixing of Atlantic and Arctic water masses (Ingvaldsen et al., 2002; Smolyar and Adrov, 2003) (Fig. 1). The seafloor topography is complex, consisting of platform areas, basins and troughs. Average depths are 230 m and rarely exceed 300 m (Fig. 1). Material flux in the summer is large, as a result of intense primary production, as well as riverine and ice transport (Wassmann et al., 2006). In the winter, the material flux is small, but intense mixing processes, together with formation of dense water layers leads to resuspension and redeposition of bottom sediments in some areas, particularly on shallow platforms such as Hopen Bank (Sternberg et al., 2001).

Plutonium and ^{137}Cs activity concentrations in sediments are often studied in the Arctic, but the majority of data concerns surface samples collected by sediment grabs. Several papers reported ^{137}Cs activity concentrations of 0.5 Bq/kg to 9.3 Bq/kg for surface sediments of southern and central Barents Sea (Føyn and Sværen, 1997; Nies et al., 1999; Heldal et al., 2002). Smith et al. (1995) studied down-core ^{137}Cs activity concentrations in the eastern Barents Sea. They reported ^{137}Cs inventories of 251–1065 Bq/m² in the vicinity of Novaya Zemlya. Heldal et al. (2002) studied $^{239,240}\text{Pu}$ in sediments collected south of Svalbard and reported activity concentrations of 0.10–2.37 Bq/kg. These data are similar to surface sediment activity concentrations (0.8–1.6 Bq/kg) reported by Matishov et al. (1999) for the southern Barents Sea, Kola Bay region. However, Baskaran et al. (1996) and Smith et al. (1995) reported

a factor 2 lower activity concentrations in sediments collected in the Barents and Kara Seas. The $^{239,240}\text{Pu}$ activity concentrations measured in the vicinity of Novaya Zemlya reached as high as 15 000 Bq/kg (Smith et al., 2000).

The aim of the present investigation was to compare data on activity concentrations and inventories of anthropogenic radionuclides: ^{238}Pu , $^{239,240}\text{Pu}$ and ^{137}Cs in context with information on the radionuclide depositional history based on ^{210}Pb geochronology. The ^{210}Pb sediment accumulation rates and mixing coefficients were reported previously by Zaborska et al. (2008) and Carroll et al. (2008). Maximum sediment accumulation rates are on the order of 0.3 mm/year (station IV and XI) to 1.3 mm/year (station XVIII). Sediment mixed depth range from 0 cm (stations III, XVII, XVIII) to 2 cm (station X). Mixing rates based on ^{234}Th are from 1 cm²/year (station XII) to 12 cm²/year (station XVIII). Based on ^{210}Pb are from 0.1 cm²/year (station XI) to 1 cm²/year (station VIII and XII); no mixing was found at stations III, XVII and XVIII. The assembled data were used to evaluate sources and transport processes of radionuclides detected in sediments of the Barents Sea.

2. Materials and methods

2.1. Field activities

Sampling was carried out on board *R/V Jan Mayen* as part of the large multi-disciplinary research project CABANERA (Wassmann et al., 2008). Sediment cores were collected along a north–south transect in the north-western Barents Sea. Four stations were sampled in 2003 (stations I, II, III, IV), four stations were sampled in 2004 (stations VIII, X, XI, XII) and three stations were sampled in 2005 (stations XVI, XVII, XVIII) (Fig. 1).

Undisturbed sediment cores were collected using a multicorer with four 50-cm-long, 10-cm diameter core tubes. Three sediment cores from each multicore cast were used for gamma measurements (sliced every 5 mm, the same depth interval combined) while the fourth core was dedicated for measurement of alpha emitting radionuclides (sliced every 10 mm), sediment porosity, sediment density and grain size analyses. Sediment cores were described after collection (length, colour, grain size and compaction, presence of animals or stones). The outer 1 cm of each sediment slice was discarded to avoid contamination from surface sediment smearing down the core tube walls during sampling. Sediment slices were dried at 60 °C and ground into fine particles. Sediment sub-samples were removed during core slicing, and placed into pre-calibrated (weight and volume) vials for sediment porosity and density determinations. Sediment sub-samples were also retained for analyses of grain size and organic content.

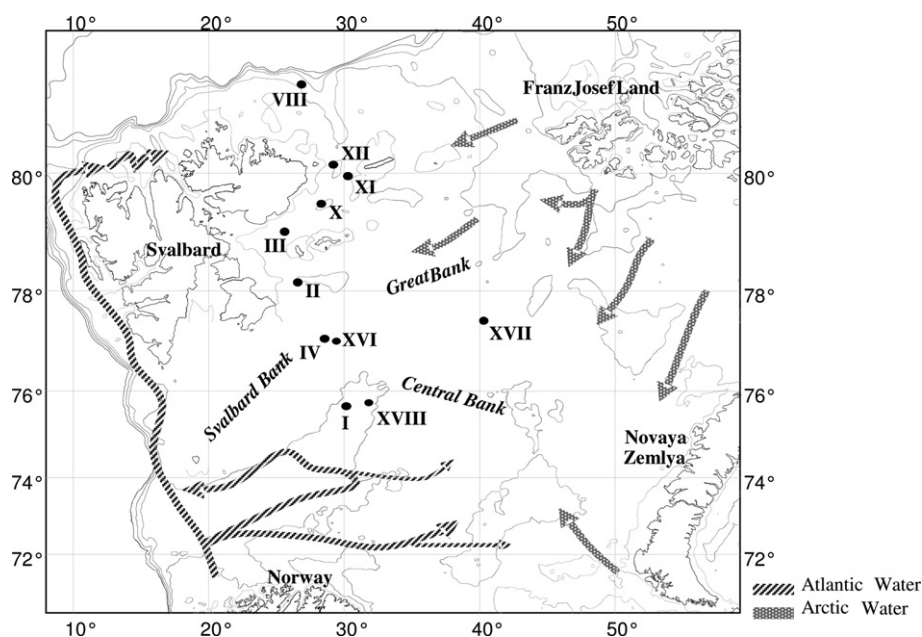


Fig. 1. Study area and location of sampling stations in the north-western Barents Sea (Atlantic and Arctic water masses circulation based on Sundfjord et al. (2007)).

2.2. Laboratory measurements

Caesium-137 was measured simultaneously with ^{210}Pb and other gamma emitting radionuclides in ORTEC, high purity, germanium detectors at the Italian National Agency for New Technologies, Energy and the Environment (ENEA) in La Spezia, Italy. Twenty grams of dried sediment were placed in counting vials with calibrated geometry. Detector efficiencies were calibrated using several sources, and confirmed using IAEA standard material (IAEA-300). Blanks were equal to natural background as assessed over counting periods of several days in an empty detector. Sediment samples were counted for 1–2 days. In the Results section, ^{137}Cs activity concentrations as measured are presented; while for the Discussion ^{137}Cs activity concentrations were decay corrected for sediment layer age (based on ^{210}Pb dating).

Plutonium analyses were carried out at the Institute of Nuclear Physics PAS in selected sediment core layers. Sediment layers: 0–1 cm, 1–2 cm, 3–4 cm, 5–6 cm, 10–12 cm and 18–20 cm of cores collected at six stations: I, III, IV, VIII, XI and XVII were selected for analysis. Approximately 10 g of sediment were combusted at 600 °C for six hours. After cooling, a wet digestion procedure was performed according to the procedure developed by LaRosa et al. (1992) and modified by Mietelski and Was (1995). Plutonium was co-precipitated with NdF_3 (LaRosa et al., 1992). The precipitate was filtered through a 25-mm-diameter (100 nm pore size) membrane filter. A filter with the precipitate was washed with ethanol and deposited on a stainless steel disc. Samples were placed in a Canberra alpha-spectrometer, counting efficiency (with chemical recovery included) was of ~36%. The ^{242}Pu was added to every sample before the plutonium separation as an internal standard. The spectrometer efficiency was confirmed with IAEA-375 standard reference material. Each sample was counted for 2 days.

Radionuclide inventories represent the cumulative sum of radionuclide activity concentrations (Bq/kg) multiplied by the cumulative mass of each sediment layer (g/cm^2), and expressed in Bq/m^2 . Caesium-137 inventories were already published for brief comparison of studied stations in Zaborska et al. (2008); profiles of radionuclide activity concentrations were not presented however.

Radionuclide uncertainties include measurement error only. Statistical calculations were performed using the computer program, STATISTICA. Regression analyses were performed to assess relationships between a given radionuclide activity concentration and sediment properties (e.g. grain size, organic carbon concentration etc.). ANOVA tests were performed to assess the statistical significance of differences in sediment properties among stations. The data did not fulfill the test assumptions (normality of distribution and homogeneity of variance), and therefore we applied the non-parametric Kruskal–Wallis test (later called K–W test). Post hoc testing (Pair-wise Mann–Whitney U-tests) were also performed on stations pairs. Probability value, $p > 0.05$ was used for all data if not stated otherwise. When performing calculations (subtracting, ratio counting) on radionuclide activity concentrations, the relative uncertainty was calculated. The uncertainty is the square root of the sum of the squares of the relative errors of individual activity concentrations. Original data on radionuclide activity concentrations is presented in supplementary tables: S1 and S2.

3. Results

Caesium-137 was measured at 11 stations. The highest activity concentrations ($>7 \text{ Bq}/\text{kg}$) were measured at stations III, VIII, X, XI and XII (Table S1). At stations XVI and XVIII, measured ^{137}Cs activity concentrations were lower, with a maximum value of 4 Bq/kg. The ^{137}Cs activities differed significantly among stations (K–W test, $H = 73.2$, $P < 0.0001$), even those within a relatively close vicinity of one another e.g. IV and XVI, I and XVIII, XI and XII (post hoc tests).

The $^{239,240}\text{Pu}$ activity concentrations were measured at selected layers of six stations. The measured activity concentrations ranged from 0.26 to 2.74 Bq/kg at station I, from 0.01 to 0.75 Bq/kg at station III, from <0.01 to 1.1 Bq/kg at station IV, from 0.54 to 0.63 Bq/kg at station VIII, from <0.01 to 0.71 Bq/kg at station XI and from 0.05 to 0.29 Bq/kg at station XVII (Tab. S2). Relatively uniform plutonium activity concentrations were detected at station VIII, a station where sediment mixing was identified (Carroll et al., 2008). For this limited number of samples measured, $^{239,240}\text{Pu}$ activity concentrations did not correlate with organic matter or organic carbon content (for $p > 0.05$).

The ^{238}Pu activity concentrations were very low at stations III, IV, XI, VIII and XVII ranging from <0.01 to 0.07 Bq/kg (Table S2). Slightly higher activity concentrations were measured at station I ranging from <0.01 to 0.22 Bq/kg.

4. Discussion

4.1. Radionuclide activity concentrations and inventories

4.1.1. ^{137}Cs activity concentration

Surface sediment (0–2 cm) ^{137}Cs activity concentrations (1.2–8.8 Bq/kg) presented in this study compare well with activity concentrations published by other authors. Nies et al. (1999) report ^{137}Cs activity concentrations in the surface sediments of the southern and central Barents Sea ranging from 1.5 Bq/kg to 5.8 Bq/kg, while Føyn and Sværen (1997) report ^{137}Cs activity concentrations in the same region ranging from 1.0 Bq/kg to 8.6 Bq/kg. Heldal et al. (2002) report ^{137}Cs activity concentrations ranging from 0.5 Bq/kg to 9.3 Bq/kg. However, sample collection in these studies was performed by sediment grab with no control for differences in sediment ages. This is critical when studying anthropogenic radionuclides with varying source function as is the case for ^{137}Cs and plutonium.

There were significant ($p > 0.05$) differences in ^{137}Cs activity concentrations even between stations located in close proximity to one another. However, we found no statistically significant correlation between the pelite (silt and clay) fraction and ^{137}Cs ($R^2 = 0.19$, $n = 65$) nor between ^{137}Cs and organic carbon content ($R^2 = 0.03$, $n = 65$). Thus sedimentary ^{137}Cs activity concentrations are more likely a reflection of the variable influence of different radionuclide sources or variation in individual source terms with time and not particle composition.

The ^{137}Cs inventories varied among stations, ranging from $29.5 \pm 1.5 \text{ Bq}/\text{m}^2$ to $152.7 \pm 5.6 \text{ Bq}/\text{m}^2$ (Fig. 2; Zaborska et al., 2008) with southern stations (I, XVIII, IV, XVI, XVII, II) exhibiting lower inventories than northern stations. There was a positive correlation of ^{137}Cs inventory with latitude ($R^2 = 0.66$, $n = 11$). The highest ^{137}Cs inventories were calculated for stations XI, VIII and III ($153 \pm 6 \text{ Bq}/\text{m}^2$, $142 \pm 2 \text{ Bq}/\text{m}^2$ and $122 \pm 3 \text{ Bq}/\text{m}^2$ respectively), located in Erik Erikssentret between Nordauslandet and Kong Karls Land (Zaborska et al., 2008). Possible processes leading to higher total inventories at these stations are intense melting of sea ice originating from Siberian coastal areas (Pfirman et al., 1997; Dethleff et al., 2000). The other explanation could be fact observed by Kershaw et al. (1997) that the central Arctic is itself acting as a source of reprocessing ^{137}Cs to the Barents Sea through recirculation of polar water contaminated with earlier discharges. Since the ^{137}Cs inventories correlated negatively ($R^2 = 0.61$, $n = 11$) with distance from Svalbard, terrigenous supplies may be an additional ^{137}Cs source. Not surprisingly,

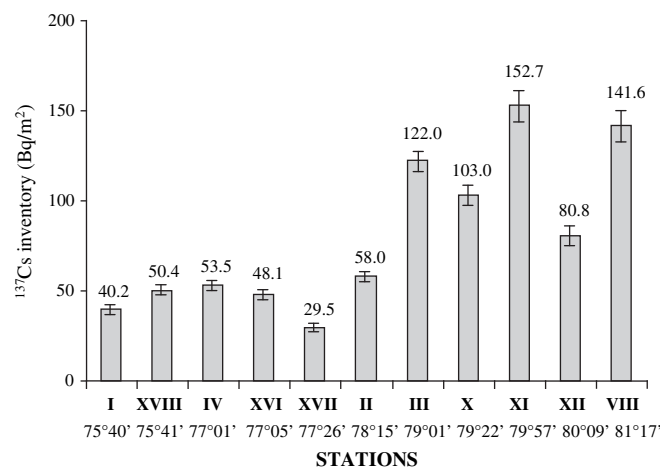


Fig. 2. Distribution of ^{137}Cs (Bq/m^2) inventories in the northern Barents Sea (Zaborska et al., 2008).

^{137}Cs inventories did not correlate with the pelite sediment fraction ($R^2 = 0.06$, $n = 11$) or organic carbon content ($R^2 = 0.17$, $n = 11$). These ^{137}Cs inventories in the northwestern Barents Sea are lower than inventories for the Barents Sea near Novaya Zemlya ($251\text{--}1065\text{ Bq/m}^2$) as reported by Smith et al. (1995) and an order of magnitude lower than inventories for the Ob river delta ($1000\text{--}5000\text{ Bq/m}^2$) (Sayles et al., 1997).

High ^{137}Cs activity concentrations and inventories are most likely influenced by ice transport of anthropogenic radionuclides in the Arctic (Emery et al., 1997; Messe et al., 1997; Pfirman et al., 1997; Landa et al., 1998; Hölemann et al., 1999; Pavlov et al., 2004; Eicken et al., 2005). Sea ice sediments containing radioactive pollutants from Novaya Zemlya were tracked by Emery et al. (1997). The ice moved north and west and after 9 months melted in the central Barents Sea. Landa et al. (1998) report very high activity concentrations of ^{137}Cs (up to 78 Bq/kg) and $^{239,240}\text{Pu}$ (up to 1.9 Bq/kg) in sea ice sediments collected in the northern Barents Sea. This confirms that the central and northern regions of the Barents Sea are highly influenced by sedimentary material containing radionuclides from melting ice cover. Ice drift trajectories for the study area during the cruises were modelled from satellite images by Hop and Pavlova (2008). Their study confirms conclusions made by others that ice on the Barents Sea is generally transported from the Kara Sea (e.g. Pfirman et al., 1997; Pavlov et al., 2004). We conclude that high ^{137}Cs inventory at the central and northern stations (III–VIII) may be partly explained by intense ice cover melting.

The radionuclide concentrations may be also enhanced by sediment focusing. Carroll et al. (2008) reports sediment focusing, due to the input of non-local supplies of organic material, in trench stations VIII and XII. On the contrary, low content of pelite fraction at stations IV and XVI (Zaborska et al., 2008) may indicate high energy conditions and possibility of erosion processes. This might partly explain lower ^{137}Cs inventories at both stations. High primary production on ice edges may enhance radionuclide scavenging. Føyen and Sværen (1997) conclude that higher ^{137}Cs inventories in Svalbard vicinity are caused by increasing scavenging of ^{137}Cs due to increased primary production.

Other, but less important, transport pathways are long range particle transport by aerosols (Pacyna, 1995; Macdonald et al., 2000; Quinn et al., 2002; Treffeisen et al., 2004) and fluvial material transport to the Barents Sea sediments.

4.1.2. The $^{239,240}\text{Pu}$ activity concentration

The $^{239,240}\text{Pu}$ activity concentrations obtained in this study ($0.01\text{ Bq/kg} \pm 0.003\text{ Bq/kg}$ to $2.74\text{ Bq/kg} \pm 0.06\text{ Bq/kg}$) are in agreement with activity concentrations ($0.10\text{--}2.37\text{ Bq/kg}$) reported by Heldal et al. (2002) for sediments collected south of Svalbard and by Matishov et al. (1999) for the southern Barents Sea, Kola Bay region ($0.8\text{--}1.6\text{ Bq/kg}$). However, Baskaran et al. (1996) and Smith et al. (1995) report levels in sediments collected in the Barents and Kara Seas that are a factor of 2 lower. On the other hand, $^{239,240}\text{Pu}$ activity concentrations measured in the vicinity of Novaya Zemlya were as high as 15000 Bq/kg , as a consequence of the underwater nuclear tests performed in Chernaya Bay (Smith et al., 2000). These differences further exemplify that proximity to source is the primary factor controlling radionuclide content in sediments of the Barents Sea. In contrast to ^{137}Cs , $^{239,240}\text{Pu}$ activity concentrations decreased northward, correlating significantly ($p > 0.05$) with latitude ($R^2 = 0.75$, $n = 5$). However, the number of stations is limited, necessitating further studies to confirm this trend.

Like others (e.g. Smith et al., 1995), our $^{239,240}\text{Pu}$ inventories were poorly resolved because $^{239,240}\text{Pu}$ measurements were made only for selected sediment layers (7 layers per core: 0–1 cm, 1–2 cm, 2–3 cm, 3–4 cm, 5–6 cm, 10–12 cm, 18–20 cm). Having measured $^{239,240}\text{Pu}$ in similar layers in each core, we report $^{239,240}\text{Pu}$

inventories as the cumulative sum of all measured $^{239,240}\text{Pu}$ activity concentrations (Bq/m^2), providing a relative measure of inventories among the investigated stations. The $^{239,240}\text{Pu}$ inventories among the six investigated stations ranged from $1.8 \pm 0.1\text{ Bq/m}^2$ at station XVII to $29.7 \pm 0.4\text{ Bq/m}^2$ at station I (Fig. 3). Others assume that the surface interval (2 cm) of sediments represents 15 % of the total $^{239,240}\text{Pu}$ inventory (Smith et al., 1995). This assumes that $^{239,240}\text{Pu}$ activity concentration profiles follow the atmospheric fallout controlled depositional trend. Given the influence of multiple sources to the investigated region, this assumption may not be correct for the present investigation. However, for comparison purposes, inventories derived based on the assumption of 15 % ranged from $5.4 \pm 0.1\text{ Bq/m}^2$ at station XVII to $77.7 \pm 0.7\text{ Bq/m}^2$ at station I. Inventories derived in this way are higher by a factor of 2–3 than inventories based on measured levels alone. Still, there is a distinct pattern of higher inventories in the south compared to the north. Compared to sediment inventories for the Pechora Sea (Novaya Zemlya vicinity) which ranged from 26 to 150 Bq/m^2 and from the Barents Sea off Novaya Zemlya ($13\text{--}155\text{ Bq/m}^2$) (Smith et al., 2000), our inventories are relatively low.

Activity concentrations of anthropogenic radionuclides in sediments are controlled by many processes, including local inputs, time dependent source functions, sediment accumulation rates, pelite and organic matter (carbon) content and lateral transport of water and particles (Ab Razak et al., 1996; Baskaran et al., 1996). Caesium-137 is sorbed most efficiently on clay minerals (Duursma and Carroll, 1996). The ^{137}Cs activity concentrations in sediments generally increase with increasing pelite fraction content (Meili, 1994). However, there were no correlations observed between ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations, inventories, or sediment properties (pelite fraction content, organic matter and organic carbon content) in the present investigation. Different sediment sources (rivers, ice transport) enriched in ^{137}Cs and plutonium is likely the cause of the major differences in down core radionuclide profiles. The correlation was not observed between radionuclide activities and pelite fraction content as has been found in other studies (Matishov et al., 1999; Smith et al., 2000; Heldal et al., 2002).

Based on ^{90}Sr global fallout within the $70\text{--}80^\circ\text{N}$ latitude band (UNSCEAR, 1982), and assuming $^{239,240}\text{Pu}$ fallout is proportional to ^{90}Sr fallout, the integral deposition for global fallout $^{239,240}\text{Pu}$ is approximately 12 Bq/m^2 . Data obtained during this study showed higher activity inventories, suggesting again additional plutonium

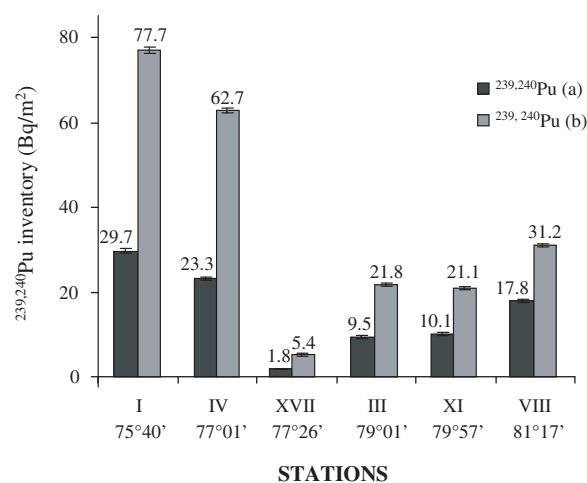


Fig. 3. Distribution of $^{239,240}\text{Pu}$ (Bq/m^2) inventories in the northern Barents Sea. Two calculation methods are used; (a) light grey coloured bars – $^{239,240}\text{Pu}$ inventory is the sum of activity concentrations in six sediment layers, (b) dark grey colored bars – $^{239,240}\text{Pu}$ inventory is calculated from the assumption that surface sediment layers (0–2 cm) contain 15% of the total $^{239,240}\text{Pu}$ inventory.

sources. Using a similar approach for ^{137}Cs inventories (decay corrected), the integral deposition for ^{137}Cs global fallout is approximately 550 Bq/m^2 . The measured inventories of both ^{137}Cs and $^{239,240}\text{Pu}$ in the north-western Barents Sea were higher; a further indication of the relative importance of sources other than global fallout to this region.

4.2. Radionuclide deposition history

The ^{137}Cs activity concentrations corrected for decay (Fig. 4) and $^{239,240}\text{Pu}$ activity concentrations (Fig. 5) were plotted against deposition year based on sediment accumulation rates reported in Zaborska et al. (2008).

For most stations, the depth of ^{137}Cs penetration rather agrees with sediment layer age (we expect ^{137}Cs presence from 1950), when considering mixing of the surface sediment as reported by Carroll et al. (2008). Visual interpretation of profiles leads to the conclusion that sediments at all stations were mixed, since none of the profiles tracked the history of ^{137}Cs deposition (i.e. introduction in 1950, global fallout maximum 1960–1970). At most profiles (stations: I, II, IV, X, XI, XII), recent deposition of ^{137}Cs was evident. Since very little ^{137}Cs is now introduced to the environment, recent radionuclide deposition may be attributed to reintroduction of ^{137}Cs from highly contaminated Irish Sea sediments (Hallstadius et al., 1986; MacKenzie et al., 1998). The ^{137}Cs activity concentrations at several stations were relatively low, resulting in significant measurement error (20–30%). Moreover, the combination of low sediment accumulation rates at some stations, evidence of mixing,

and the likelihood of sediment resuspension and transport processes, limits the value of this radionuclide as a time marker in sediment profiles.

None of the $^{239,240}\text{Pu}$ profiles exhibit activity concentration peaks (Fig. 5) as would be expected from global fallout. At all stations recent deposition of $^{239,240}\text{Pu}$ was apparent. Moreover at stations: I, III, VIII, and XI the highest $^{239,240}\text{Pu}$ activity concentration was measured at the surface-most sediment layer. Plutonium is no longer introduced to the environment, the recent discharges from reprocessing plants are negligible. This interesting finding may be related to plutonium reintroduction from the Irish Sea sediments as shown by Hallstadius et al. (1986) and MacKenzie et al. (1998). The $^{239,240}\text{Pu}$ penetrated to depths dated to 1850, well before plutonium was introduced to the environment. However, with sediment accumulation rates of, for example, 0.3 mm/year (station IV), time resolution is relatively poor for cores sliced at 10 mm intervals for $^{239,240}\text{Pu}$ analysis. Consider that a 10 mm layer of the core represents 33 years. If plutonium is present in only the surface 1 mm sediment layer (representing 3 years sedimentation), this activity will contribute to the reported average activity for the entire interval. This also leads to a diminished appearance of peaks in radionuclide activity concentrations within profiles of $^{239,240}\text{Pu}$ versus sediment depth.

Mixing further contributes to a loss of resolution in depth profiles of $^{239,240}\text{Pu}$. The ^{210}Pb based sediment mixed depths at stations I, IV and VIII were at least 1–2 cm (Carroll et al., 2008). If these layers are added (to the penetration depth obtained by sediment accumulation rate) then penetration of $^{239,240}\text{Pu}$ to deeper layers is reasonable. While Carroll et al. (2008) reported no

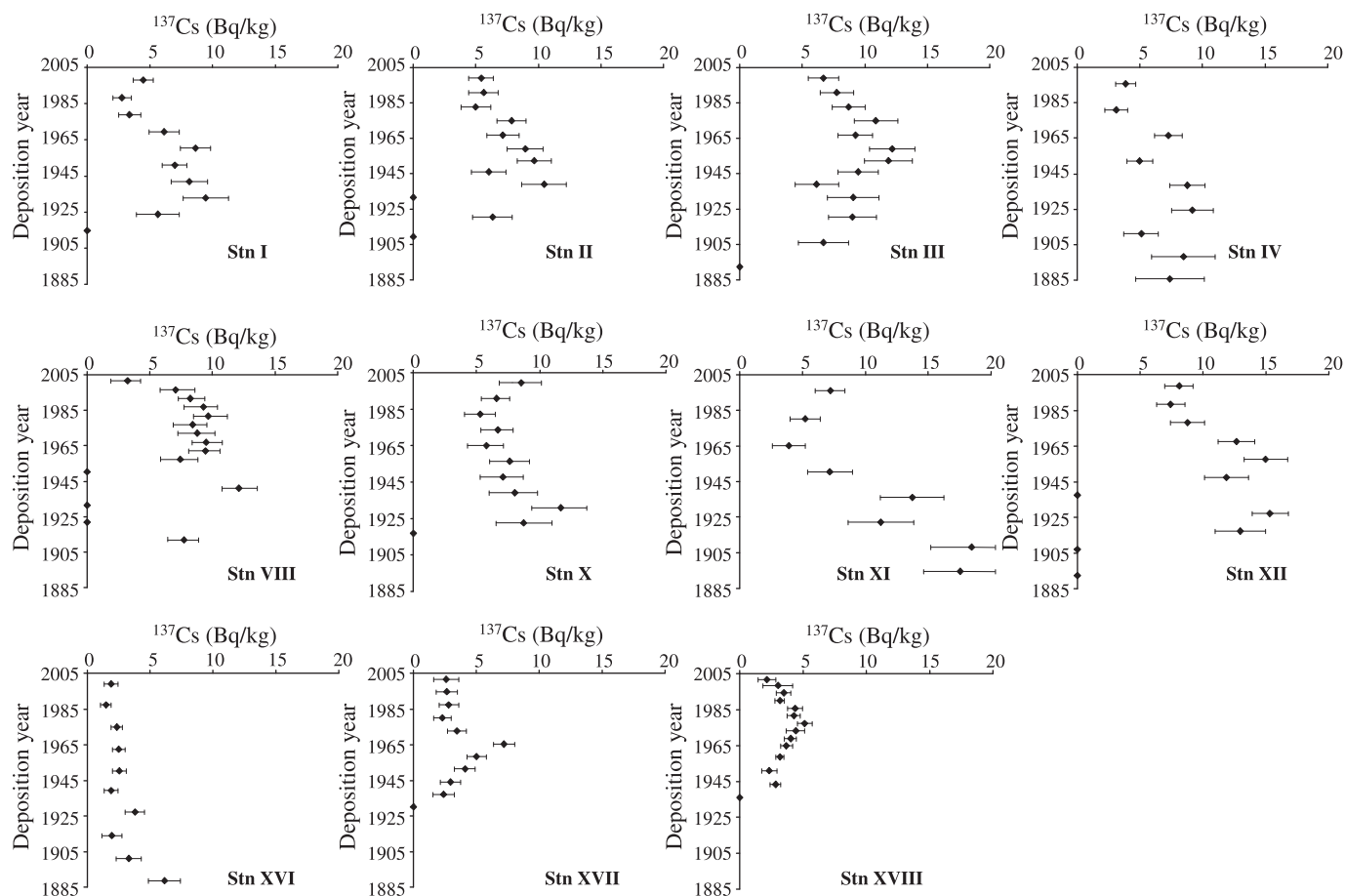


Fig. 4. ^{137}Cs activity concentrations (Bq/kg) versus year of deposition of sediment layers (determined by ^{210}Pb method) in 11 sediment cores collected from the north-western Barents Sea.

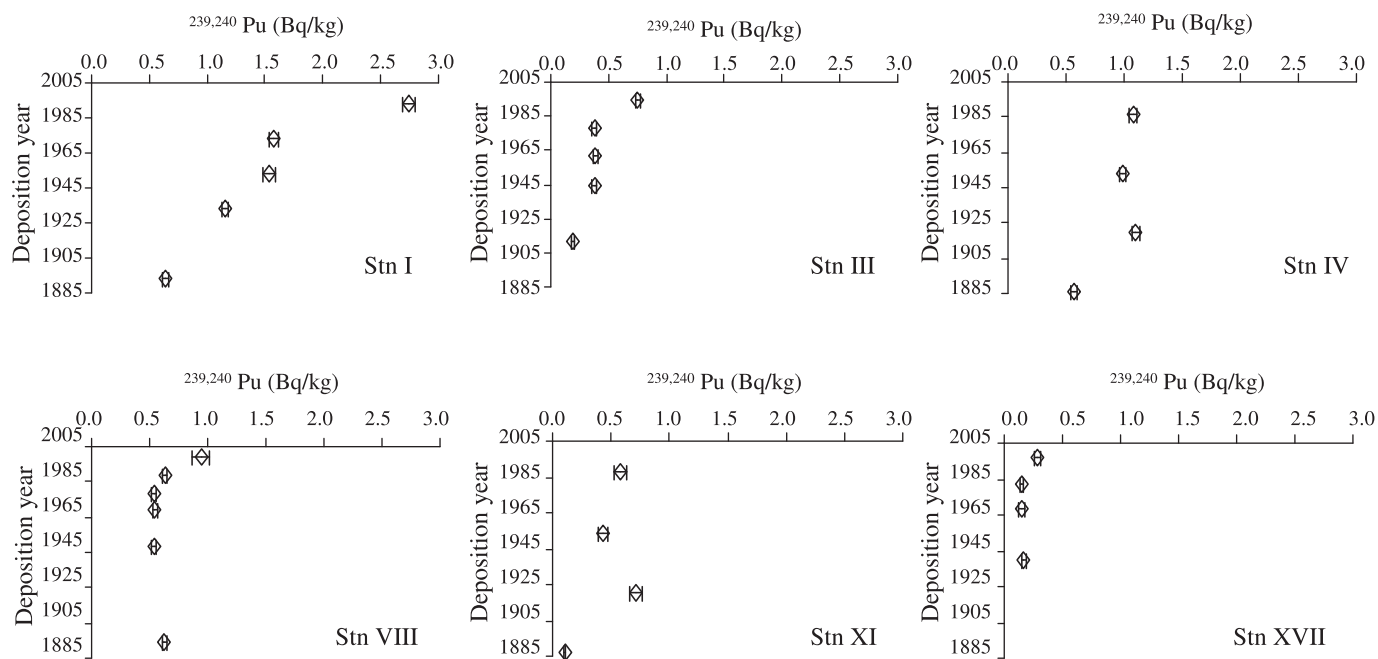


Fig. 5. ^{239,240}Pu activity concentrations (Bq/kg) versus deposition year (obtained by ²¹⁰Pb method) in 6 cores collected from the north-western Barents Sea.

clear mixed depth at station III, if mixing is relatively constant, the obtained ²¹⁰Pb profile will still exhibit an exponential decrease in activity concentrations with depth (e.g. Carroll and Lerche, 2003). Thus, the ^{239,240}Pu data may indicate mixing at station III as well or constant input of this radionuclide to the area.

Plutonium isotopes may also diffuse to deeper layers through pore water exchange. This process has been discussed by Buesseler and Sholkovitz (1987), who found low rates of ^{239,240}Pu diffusion with pore water in the North Atlantic shelf sediments. This was later confirmed by Smith et al. (1995); however, diffusion is negligible compare to the influence of sediment mixing.

Others have also encountered problems with ^{239,240}Pu penetration profiles of sediment collected in the Arctic Ocean. Huh et al. (1997) attributed the presence of ^{239,240}Pu below the predicted depth of penetration to contamination during sampling or sediment mixing. Moreover they found the highest ^{239,240}Pu activity concentrations at the sediment surface (0–1 cm) and not in the layers representing years of highest global input. Lateral processes such as boundary scavenging of ^{239,240}Pu from other areas was suggested as an explanation for their results. Excluding contamination, these are reasonable explanations for the Barents Sea data as well. To avoid problem with contamination caused by smearing sediments along the core tube walls, we removed 1 cm of sediment from the outer edge of each core slice after sediment collection.

Where peaks were observed in both ¹³⁷Cs and ^{239,240}Pu activity concentrations, they occurred at different sediment depths. This is to be expected when both the radionuclide behaviour and sources to the marine environment differ, as in the present case. Generally, plutonium is delivered to the sea in dissolved form, but being particle reactive it is rapidly scavenged from the water column to particles which then are deposited on the seafloor. Particle transport via sediment resuspension, sea ice and the atmosphere redistribute plutonium in the sea, while sedimentary ¹³⁷Cs in shelf areas is derived mainly from terrestrial sources, instead of being scavenged from the water column (Baskaran et al., 1996). Additionally, both radionuclides may be reintroduced from contaminated Irish Sea sediments (MacKenzie et al., 1998). Transit of radionuclides to the Barents Sea may occur in different times and

rates that would be related to their different behaviours. Generally we see the need to include sediment dating tools when interpreting sediment profiles of pollutants in order to assess contributions from variable source inputs and to evaluate temporal trends.

4.3. Radionuclide sources

The ^{239,240}Pu activity concentrations have been applied as tracers of marine environmental behaviour (e.g. fractionation) and radionuclide sources (Sayles et al., 1997; Smith et al., 2000). A strong correlation ($R^2 > 0.95$) between radionuclides characterised by different half lives would mean delivery from a single source while multiple sources separated in time would result in elevated values of plutonium as compared to ¹³⁷Cs. To use these hypotheses as a basis for interpreting ^{239,240}Pu and ¹³⁷Cs activity concentrations in the present study, we first decay corrected our reported ¹³⁷Cs activity concentrations based on the sediment accumulation rates of Zaborska et al. (2008). The derived ^{239,240}Pu/¹³⁷Cs ratios ranged from 0.01 to 0.75 (Table S2). This large range of values for this ratio further indicated that multiple sources control the distribution of these anthropogenic radionuclides in the northwestern Barents Sea. If global fallout alone controlled the decay-corrected ^{239,240}Pu/¹³⁷Cs, the value of this ratio would be approximately 0.044. The ^{239,240}Pu/¹³⁷Cs ratio is not the most relevant tool for radionuclide source studies since behaviour of both radionuclides in the marine environment differs. However, it is often used to assess contributions from variable source (Baskaran et al., 1996; Heldal et al., 2002).

Baskaran et al. (1996) reported ratios up to 0.08 for Kara Sea sediments. Ratios of 0.02–0.12 were calculated by Matishov et al. (1999) for the southern Barents Sea (Kola Bay region). Using ¹³⁷Cs and ^{239,240}Pu activity concentrations presented by Heldal et al. (2002) for the Barents Sea (south Svalbard region), the derived ratios range from 0.10–0.41. Similarly, data reported by Smith et al. (1995) for the Barents Sea (Novaya Zemlya region) resulted in ^{239,240}Pu/¹³⁷Cs ratios ranging from 0.03 to 0.78. Thus our results confirm earlier observations from different areas of the Barents Sea.

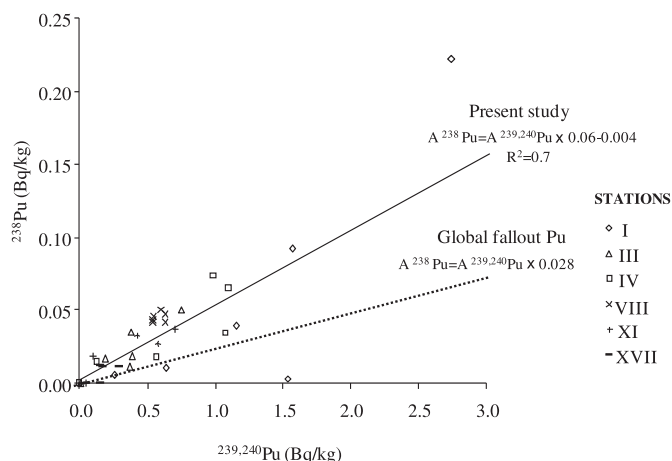


Fig. 6. ^{238}Pu activity concentrations (Bq/kg) versus $^{239,240}\text{Pu}$ activity concentrations (Bq/kg) in sediments from stations I, III, IV, VIII, XI and XVII. The figure shows that $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio is higher (mean ratio 0.06, solid line) compare to only global fallout source (expected ratio 0.03, dashed line).

More often, the $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio is used to establish sources of plutonium to the marine environment. Ratios characteristic for global fallout should be about 0.028 (Smith et al., 2000), with similarly low ratios connected to nuclear tests (Ikaheimonen et al., 1997; Beasley et al., 1998; Efurud et al., 2005). High $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios of about 0.3 are characteristic of discharges from reprocessing plants (eg. Sellafield) (MacKenzie et al., 1998; Vintrò et al., 2000) and accidental input of plutonium isotopes (eg. Chernobyl) (Mietelski

and Was, 1995; Ketterer et al., 2004). The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio for Sellafield varied in time (1950–2000) from 0.03 to 0.36, but the time integrated mean value is reported to be 0.18 (MacKenzie et al., 1998; Kershaw et al., 2001). Radionuclide ratio in dumped reactors in the Kara Sea ranges from 0.26 to 0.49 (Kershaw et al., 2001).

Ratios in the investigated samples range from 0.02 at station I (layer 6–7 cm) to 0.12 at station IV (layer 5–6 cm), but there was one sample with a ratio of 0.19 (station XI, layer 3–4 cm). The average $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio of 0.06 (solid line) is about 2 times higher than the global fallout ratio (dashed line) (Fig. 6). Generally these ratios indicate that sedimentary plutonium is derived from sources in addition to global fallout.

For comparison, $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios reported by Heldal et al. (2002) south of Spitsbergen ranged from 0.03–0.04 but ratios for the Greenland Sea are as high as 0.12. Ratios reported for the Barents Sea (Novaya Zemlya region) ranged from 0.01 to 0.18 (Smith et al., 1995) and in the Kara Sea ratios were from 0.01 to 0.07 (Baskaran et al., 1996).

According to Baskaran et al. (1996), the percentage of global fallout versus local sources (e.g. discharges from contaminated Novaya Zemlya testing sites or discharges from Sellafield) may be estimated assuming that there are two main sources of plutonium in the Barents Sea: global fallout with a $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio = 0.028 and non-global fallout with $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio = 0.26:

$$f_G = (A_R - A_M) / (A_R - A_G)$$

where f_G is the fraction of plutonium isotopes from global fallout sources (%). A_R is $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio of non-global sources (0.26). A_M is the measured $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio and A_G is $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio of global fallout (0.028).

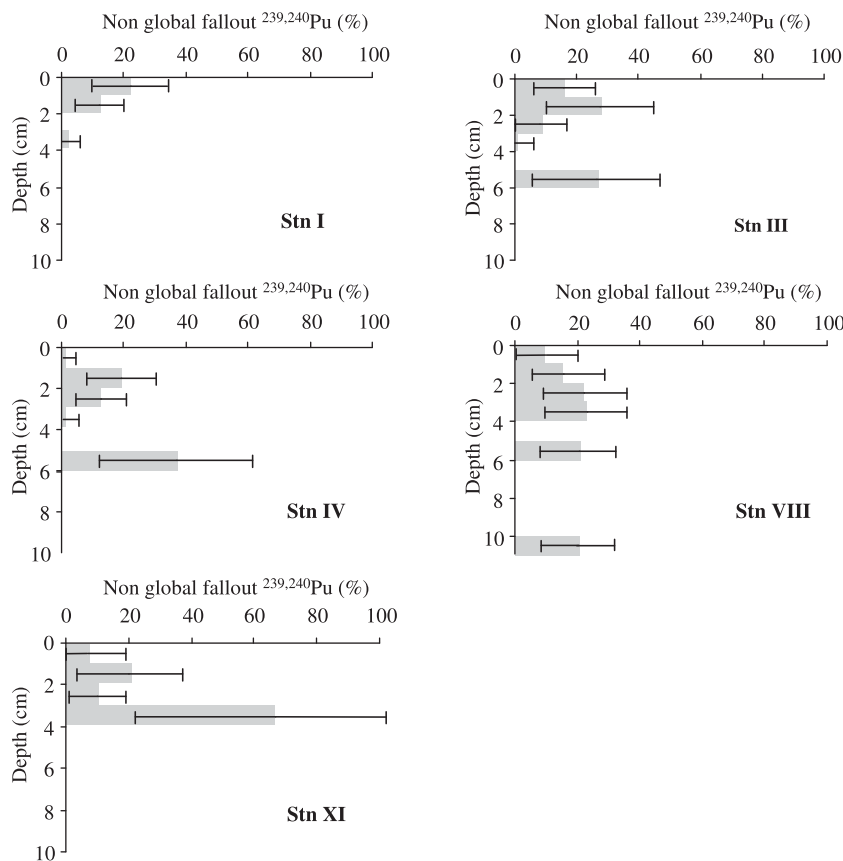


Fig. 7. The percentage of non-global fallout plutonium in sediment cores collected at five stations (at station XVII, ^{238}Pu was below the detection limit).

This analysis was performed for all stations except XVII (Fig. 7). The ^{238}Pu activity concentrations at station XVII were below the detection limit. For the formula, the 0.26 as a ratio of non-global fallout was used but some authors report that the average $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio for Sellafield is actually 0.18 (Kershaw et al., 2001). Since both isotope ratios from Sellafield discharges changed in time, it is problematic to establish this ratio. Some deal with this problem in the Arctic by using 0.2 for “historical sediments” and 0.3 for “recent sediments” (Gwynn et al., 2004). If a ratio <0.26 is applied, the determined contribution of non global fallout plutonium will obviously increase.

At station I, up to $22 \pm 11\%$ of plutonium originated from other sources than global fallout and a similar percentage of non-global fallout sources was derived for station III, up to $27 \pm 15\%$. For station VIII, the fraction was relatively constant with depth with highest contribution of $23 \pm 13\%$, a further indication of sediment mixing at this station. Generally similar proportion of non-global fallout plutonium was derived for stations IV ($19 \pm 11\%$) and station XI ($20 \pm 16\%$). The exception for station IV was one sediment layer (5–6 cm) with non-global fallout contribution of $37 \pm 20\%$ and for station XI one sediment layer (3–4 cm) with fraction of even $66 \pm 40\%$. This phenomena could be related to other source of plutonium. Both sediment layers were deposited before plutonium introduction to the environment. The complexity of plutonium behaviour in sediments makes these results difficult to interpret. There are little results to compare, in Kara Sea, 2–14 % of sedimentary plutonium derives from non-global fallout sources (local fallout or submerged reactors) (Baskaran et al., 1996).

As discussed above, higher $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios were caused by discharges from reprocessing plants. Taking into account the strength of the Atlantic current flow into the Barents Sea (Sundfjord et al., 2007) and that $^{239,240}\text{Pu}$ activity concentrations negatively correlate with latitude; we confirm that Sellafield as an important source of plutonium to the Barents Sea. This is in accordance with Herrmann et al. (1998) who reported that plutonium derived from Sellafield was traced in deep waters (1000–1500 m) of the Norwegian Sea, Greenland Sea and Barents Sea. Although it is believed that 99% of plutonium discharged to sea from nuclear plants is deposited near the plants (Vintrø et al., 2000); studies of many authors (Hallstadius et al., 1986; MacKenzie et al., 1998) underline the importance of plutonium reintroduction from the Irish Sea sediments.

5. Conclusions

Anthropogenic radionuclide ($^{239,240}\text{Pu}$, ^{238}Pu , ^{137}Cs) activity concentrations in seafloor sediment deposits in the north-western Barents Sea were studied. The concentrations of these anthropogenic radionuclides varied among stations, but there were clear latitudinal trends. Activity concentrations and inventories of ^{137}Cs correlated positively with latitude and negatively with distance from Svalbard archipelago. The region of highest ^{137}Cs activity concentrations was an area of melting of sea ice originating mainly along the Siberian coast. Thus our data provide further support that the main ^{137}Cs sources for the Barents Sea sediments are sea ice transport and supplies from Svalbard. The highest $^{239,240}\text{Pu}$ activity concentrations were measured at southern stations influenced by Atlantic Current. Predominant transport pathways for plutonium and ^{137}Cs appear to be sea current transport, ice transport and coastal erosion. No correlation was found between ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations or inventories with sediment properties (e.g. pelite or organic matter content). However, through several lines of evidence derived through the interpretation of ^{137}Cs and $^{239,240}\text{Pu}$ activity concentrations, inventories, ratios and depth profiles, ^{137}Cs and $^{239,240}\text{Pu}$ sedimentary patterns predominantly

reflected changing spatial and temporal influences of multiple anthropogenic sources. The changing influence of different source terms in turn diminishes any underlying relationships between sediment properties and radionuclide activity concentrations. In particular, we have shown that $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios indicated that generally 19–27 % of plutonium was not derived from atmospheric global fallout - but rather was primarily derived from nuclear fuel reprocessing plants, most likely from Sellafield. Caesium at most cases corresponded with the depositional history derived from ^{210}Pb geochronology, while plutonium profiles did not. This is mainly attributed to the influence of sediment mixing and low data resolution.

The reported data highlight the value of using dated sediment cores to monitor and evaluate changes in the levels and distributions of anthropogenic radionuclides in marine systems exhibiting complex depositional patterns such as the Barents Sea.

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Appendix. Supplementary data

Supplementary data associated with this article can be found in online version at doi: [10.1016/j.jenvrad.2010.01.006](https://doi.org/10.1016/j.jenvrad.2010.01.006).

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