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INVENTORIES AND DISTRIBUTION OF RADIOCAESIUM IN ARCTIC MARINE SEDIMENTS: INFLUENCE OF BIOLOGICAL AND PHYSICAL PROCESSES

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Extensive surveys of sediment burdens of radiocaesium, specifically ¹³⁷Cs, and other radioactive contaminants in the Arctic during the 1990's, indicate that almost all anthropogenic radionuclides buried on continental shelves adjacent to Alaska are derived from global bomb fallout. The ¹³⁷Cs (half-life: 30.2 y) activities observed in surface (0–4 cm) marine sediments however, vary widely, albeit much less than the expected current inventory resulting from bomb fallout at this latitude (~100 mBq cm⁻²). This observed geographical variation provided the opportunity to evaluate physical and biological mechanisms that may affect caesium biogeochemistry on Arctic continental shelves. We investigated whether high biological productivity in portions of the Bering and Chukchi Seas is effective in removing dissolved radiocaesium from the water column, and whether biological production in overlying water affects total radiocaesium inventories in sediments. Based upon C/N ratios in the organic fraction of shallow sediments, we found no evidence that higher inventories or surface activities of radiocaesium are present in areas with higher deposition of particulate organic matter. Based upon stable carbon isotope ratios of organic matter in sediments, we found no evidence that terrestrial runoff contributes proportionally to higher surface activities, although terrestrial runoff may affect total inventories of the radionuclide. Radiocaesium content of surface sediments was significantly correlated with total organic carbon content of sediments and the proportion of sediments in the finest sediment fractions. Because high current flow can also be expected to influence distributions of those sedimentary parameters, we conclude that re-distribution of

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radiocaesium in sediments on near-Alaska continental shelves is mostly governed by physical processes. Data from other Arctic continental shelves are discussed in the context of these findings.

Keywords: Radiocaesium; Arctic; contaminants; radionuclides; Arctic marine sediments; Arctic seas; biological physical processes

INTRODUCTION

Caesium is the rarest alkali metal with at least one stable isotope, both in the earth's crust (1 part in 2.6×10^6) and in sea water (one-half a part per billion, or approximately $1.5 \times 10^{-4} \text{ mol m}^{-3}$; Cox, 1989). Typical mid-latitude Northern hemisphere surface sea water concentrations of the most common anthropogenic radioisotope of the element, ^{137}Cs , at the present time ($\sim 5 \text{ Bq m}^{-3}$, or $7 \times 10^9 \text{ atoms m}^{-3}$), are ten orders of magnitude smaller than the concentrations of the only stable isotope, ^{133}Cs . In the ocean, the element is predominantly in a dissolved form, and its residence time is estimated to be 10^6 years (Cox, 1989). Given the 30.2 year half-life of ^{137}Cs , it is clear that only a minute proportion of the anthropogenic radiocaesium that has been introduced directly into the ocean from atmospheric fallout and direct ocean discharge over the past 40 years will be incorporated into particulate organic matter and affect radiological exposures in marine food webs. Although this statement is true both in the open ocean, as well as on continental shelves, at least two other factors need to be considered with respect to caesium biogeochemistry on continental shelves.

First, terrestrial runoff introduces significant amounts of radiocaesium into coastal areas, although in most cases the radioisotope is tightly bound to clay mineral surfaces (Olsen *et al.*, 1993). Secondly, higher biological productivity on continental shelves relative to most of the open ocean has the potential for removing some dissolved radiocaesium from the water column, through biological uptake and binding to particles associated with biological productivity. Over shorter term periods, there is little evidence that this is a significant process. For example, in a month-long period following the Chernobyl accident, only a small fraction of deposited radiocaesium ($\sim 0.2\%$) was removed from Mediterranean surface waters by biological activity

(Fowler *et al.*, 1987). These observations are also consistent with short term experiments (*e.g.*, Fisher, 1985) that show that volume to volume concentration factors for caesium in marine plankton are not significantly different than zero. Despite these shorter term indications that caesium is not actively removed from the water column by biological activity, over the 30 year period since the cessation of nuclear weapon testing, it is possible that biological activity might be cumulatively more significant in influencing sedimentation.

The recent revelations of large-scale nuclear waste dumping in the Russian Arctic (Yablokov *et al.*, 1993) have led to widespread radiological surveys of the Arctic Ocean, where the world's largest continental shelves are located. For the most part, these surveys (*e.g.*, Cooper *et al.*, 1995; Baskaran *et al.*, 1996) have found that radiocaesium burdens in Arctic continental shelf sediments are consistent with contributions from atmospheric bomb fallout only. Because atmospheric bomb fallout is almost certainly the only significant source of radiocaesium on the continental shelf regions of the Arctic adjacent to Alaska, the analyses of these data may therefore provide useful insight into the processes that have affected the re-distribution of radiocaesium in continental shelf sediments since the peak of nuclear weapons testing in the mid-1960's, namely the significance of biologically mediated removal from the water column, terrestrial runoff, and re-distribution by currents.

One of the major goals of the United States Arctic Nuclear Waste Assessment Program has been assessing the radiological risk to subsistence users of marine food resources in Alaskan coastal communities. This goal has been fortuitous from the standpoint of facilitating the collection of an extensive set of radiocaesium data ($n > 500$) for marine Arctic sediments obtained between 1990 and 1995 in the Bering, Chukchi, Beaufort, and East Siberian Seas. These data include the total inventories of radiocaesium buried in marine sediments relative to known variations in overlying biological production and current flow, and the depth of radiocaesium burial in the sediments relative to benthic bioturbation.

The western portions of the Bering and Chukchi Seas have the highest biological productivity of any Arctic continental shelves (Grebmeier *et al.*, 1995). Moreover, the eastern portions of these

two seas are characterized by sharply lower biological productivity and these differences in biological productivity are reflected in the benthic biological communities and sediment characteristics underlying the differing water masses (Walsh *et al.*, 1989; Grebmeier and Barry, 1991). As a result, these sediment surveys for radiocaesium and other radionuclides may be useful for comparing the importance of autochthonous removal of dissolved radiocaesium from the water column in relation to allochthonous, terrestrially derived, sedimentation of radiocaesium.

We chose as an indicator of overlying biological productivity the carbon:nitrogen (weight to weight) ratio in the organic fraction of surface sediments. In this region, this indicator has been shown to be relatively low ($\sim 5-7$) under the productive continental shelf waters of the Bering Shelf and the Anadyr Current, and relatively higher (up to 10) under unproductive Alaska Coastal Waters (Walsh *et al.*, 1989; Grebmeier and McRoy, 1989; Grebmeier and Barry, 1991).

The stable carbon isotope composition of the organic carbon in a subset of our samples was measured to evaluate the importance of terrestrially derived radiocaesium contributions to marine sediments. Terrestrially derived organic carbon is more depleted in ^{13}C than marine derived organic carbon (Craig, 1953), and thus can be used as a tracer of terrestrial runoff in marine settings (Parker, 1964; reviewed by Fry and Sherr, 1984). We limited our carbon isotope determinations to regions of the East Siberian and Chukchi Seas sampled in 1995 because carbon isotope data for the organic fraction of sediments in much of the rest of our study area had already been compiled (Naidu *et al.*, 1993).

Other factors that were evaluated during our analyses included a comparison of the regional variation in total inventories of radiocaesium buried in marine sediments. The analyses of several other sediment parameters, including grain size, and the organic matter contents, permits evaluating the potential physical mechanisms for radiocaesium re-distribution within Arctic marine sediments. Finally, comparison of the surface radiocaesium activities in the Kara and Laptev Seas observed in the early 1990's by other investigators (*cf.* Cooper *et al.*, 1998) also permits an evaluation of the causes of variation in radiocaesium activities in continental shelf sediments on a more pan-Arctic basis.

METHODS

The sediment sample data we present here were collected on a number of research cruises between 1990 and 1995 in the Bering, Chukchi, Beaufort, and East Siberian Seas (Tab. I). Sediments were collected using several devices, including a HAPS benthic corer with removable Plexiglas[®] insert sleeves (133 cm² surface area; design derived from Kannevorff and Nicolaisen (1973)) which under optimal conditions, recovers 15–25 cm deep cores with a very low degree of apparent disturbance. We assume low sediment disturbance based upon the presence of clear water at the sediment–water interface, the presence of flocculent materials such as faecal pellets at the base of benthic burrows at the sediment surface, continued feeding activity by

TABLE I Radioactivity in surface benthic sediments collected during this study. All radioactivity data are corrected to January 1, 1996. Data from Kara Sea are derived from Baskaran *et al.*, 1996; data from Laptev Sea were provided by Martha Scott, Texas A and M University, as reported in Cooper *et al.* (in press, 1998)

<i>Mean</i> ¹³⁷ Cs (Bq kg ⁻¹) ± S. D.	<i>Area of collection</i>	<i>Year</i>	<i>Ship platform or cruise</i>	<i>Sediment sampling technique</i>	<i>n</i>
1.6 ± 1.00	Bering Sea	1990	Alpha Helix Cruise 139	HAPS core van Veen grab	21
4.2 ± 1.7	Kasaguluk Lagoon (Chukchi Sea)	1991	small boat	Ponar grab	8
4.0 ± 3.0	Bering, Beaufort and Chukchi Seas	1992	Alpha Helix Cruise 165	HAPS core, van Veen grab	12
2.0 ± 0.9	Bering and Chukchi Seas	1993	Alpha Helix Cruise 171	HAPS core, van Veen grab	50
1.5 ± 1.4	Bering and Chukchi Seas	1993	Okean (U.S. – Russia BERPAC programme)	HAPS core, van Veen grab	64
4.4 ± 3.5	Chukchi and Beaufort Seas	1993	USCGC Polar Star	HAPS core, bottom grabs	33
14.2 ± 15.6	Kara Sea	1993, 1994	Texas A and M	box core	107
1.6 ± 1.0	Bering Sea	1994	Alpha Helix Cruise 177	HAPS core, van Veen grab	140
2.1 ± 2.1	East Siberian Sea	1995	Alpha Helix Cruise 189	HAPS core, van Veen grab	122
12.6 ± 5.9	Laptev Sea	1995	Texas A and M	box core	11

macrobenthic filter feeders, and the presence of atmospherically derived, short-lived radionuclides such as ^7Be (half-life 53 d) only in the surface layer of sediments (Grebmeier and Cooper, 1995). For determination of total radiocaesium inventories, a 7 cm diameter weighted Benthos[®] gravity core was utilized. Both HAPS and gravity cores were sectioned on ship board as follows: in 1 cm intervals to 4 cm, then 2 cm intervals to 20 cm depth, then 4 cm depth to the base of the core. In many instances in which acceptable cores could not be recovered, due to either water depth or sediment characteristics, a 0.1 m² van Veen grab, weighted with 32 kg of lead to facilitate bottom penetration, was used to obtain surface sediments, in which top sediments were removed before the grab was opened. Since in the van Veen grabs, the sediment stratigraphy was more compromised than in the cores, we have assigned nominal surface depths of 0–4 cm in our data presentations, although in the case of most of the HAPS core collections, the sediments counted were in the 0–1 cm interval.

Sediments collected using the HAPS core were packed wet in 90 cm³ aluminum cans; sediments from the van Veen grab were packed wet in 500 cm³ Marinelli beakers. All samples were returned to Oak Ridge National Laboratory, and analyzed for gamma emitters using low-background, high-resolution, lithium-drifted germanium or high purity germanium (HPGe) detectors equipped with a Canberra Genie personal computer system programmed to record gamma spectra in 4096 channels. Calibration of the detectors was performed using standards with traceability to the U.S. National Institute of Standards and Technology (NIST).

Background corrections were performed and appropriate control samples were analyzed to verify detector performance. Data are presented as either surface (0–4 cm) activities, with units of Bq kg⁻¹, or total inventories (Bq cm⁻²). Inventories were calculated as the sum of total activity detected in each sediment interval in an individual core, taking into account the area of the core (*e.g.*, 133 cm² for the HAPS core), as well as the volume of sediment in each core increment that was counted. For example, in HAPS cores, sediments were packed in 90 cm³ containers, so for the core increments sectioned at each cm, total activity in the 90 cm³ can was divided by 90 cm² to provide an inventory per cm². Adjustments were made for cores sectioned at greater than one cm intervals. Total inventories are only reported for

those cores where ^{137}Cs declined to non-detectable levels; typically we count at least several cm below the last sediment interval where ^{137}Cs is detected to ensure that all ^{137}Cs recovered in the core has been accounted for.

Additional sediment characteristics that were determined as part of this study included grain size, carbon : nitrogen weight/weight ratios, carbon isotope analysis, and the total organic carbon (TOC) fraction. The proportions of specific sediment grain sizes were determined by dry sieving, using standard geological sieves (0–4 phi sizes) following the methods of Grebmeier *et al.* (1989). For the TOC, carbon : nitrogen weight/weight ratios, and carbon isotope analysis, one g sub-samples of sediments were acidified with 2 ml of 1 N hydrochloric and dried at 105°C overnight to obtain carbonate-free sediments, followed by homogenization. The TOC content and the carbon : nitrogen ratio of the organic fraction of these sediments was measured on a Model 240XA CHN analyzer (Control Equipment Corp., Lowell, Massachusetts) located at the University of California, Santa Barbara. For those carbonate-free sediments that were also analyzed for stable carbon isotope composition, sample preparation was undertaken by combustion in vacuo at 875°C for two hours in 9 mm O.D. Vycor[®] glass tubing, in the presence of 1 g cupric oxide, 1 g copper, and silver foil. A programmable furnace with slow cool-down was used to prevent the production of oxides of nitrogen (Lajtha and Michener, 1994). Following cryogenic separation of evolved gases, stable carbon isotope ratios were determined using a VG SIRA Series II dual inlet stable isotope mass spectrometer at the Environmental Sciences Division, Oak Ridge National Laboratory. Precision, based upon measurements of three replicates of one of the sediment samples, was $\pm 0.18\%$ (standard deviation). Calibration of the internal standard used was assured through simultaneous batch processing of a sample of the NBS-22 oil standard.

RESULTS

Cross-comparisons of Samples Collected Using Different Methods

Since several devices were used to collect bottom sediments, an initial comparison of the radiocaesium activities of surface sediments

obtained using different methods was undertaken to see if surface radiocaesium activities were dependent upon the equipment used. In most cases, we expected that the radiocaesium activities of surface benthic sediments collected with the HAPS core and the van Veen grab would be similar. We based this expectation on the high biological activity in the benthos of the Bering and Chukchi Seas, which tends to mix radiocaesium homogeneously within the surface sediments (Cooper *et al.*, 1995; Baskaran and Naidu, 1995). Marine cores exhibit penetration of ^{137}Cs much deeper than expected, based upon reasonable sedimentation rates for this region (Grebmeier, 1993), and do not show the subsurface peaks often observed in laminated settings that would be associated with the peak of atmospheric bomb testing in the mid-1960's (Cooper *et al.*, 1995). The hypothesis that there were no significant differences in radiocaesium activities between surface sediments collected using the van Veen grab and the less disturbed surface sediments collected using the HAPS core was tested directly by comparing radiocaesium activities at the same stations where both sediment collection devices had been used. This comparison was undertaken on two separate cruises, Alpha Helix 177, May–June, 1994, in the Bering Sea, and Alpha Helix 189, August–September, 1995, in the Chukchi and East Siberian Seas. On both cruises, no significant differences were observed in surface radiocaesium activities for samples collected at the same stations using both the HAPS and van Veen grab (Alpha Helix 171: Wilcoxon signed-rank test; $p = 0.40$; $n = 25$; Alpha Helix 189: Wilcoxon signed-rank test; $p = 0.36$; $n = 24$).

Relationship to Pelagic Biological Productivity

Radiocaesium activities in surface sediments showed significant relationships ($p < 0.001$) to total organic carbon in the sediments (Fig. 1) and the proportion of the finest sediment fraction (Fig. 2). However, there were not significant relationships ($p > 0.05$) between radiocaesium activity in surface sediments and either the carbon:nitrogen ratio of the organic fraction of those sediments or the carbon isotope composition of the organic fraction (Figs. 3 and 4). Mean surface radiocaesium activities for sediment samples collected on each cruise and from two published sources are provided in Table I.

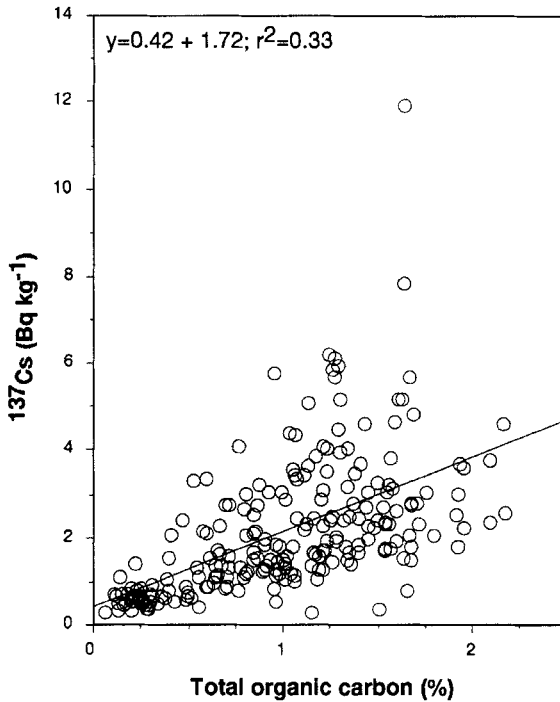


FIGURE 1 Total organic carbon in surface sediments as a fraction of total sediment dry weight relative to radiocaesium activity, decay-corrected to January 1, 1996.

Total Inventories of ¹³⁷Cs in Sediments

The total inventory of radiocaesium present in cores is dependent on water depth in an asymptotic manner (Fig. 5). Higher inventories are only found in shallow water, but some shallow water cores have relatively low total inventories. The highest inventory (69.5 mBq cm^{-2}) and the deepest radiocaesium penetration into the sediments (36 cm) was observed in a gravity core from the East Siberian Sea, at a station ($70^{\circ}01.59 \text{ N}$, $167^{\circ}41'.06 \text{ E}$) influenced by freshwater discharge from the Kolyma River. The lowest radiocaesium inventory, with $< 1 \text{ mBq cm}^{-2}$, present only in the surface layer, was observed in two cores collected from 3800 m depth in the Canada Basin (Fig. 6). During two crossings of the Canada Basin continental slope on the 1993 Polar Star cruise, the sharp decline in total radiocaesium inventories with increasing water depth is readily apparent (Fig. 6).

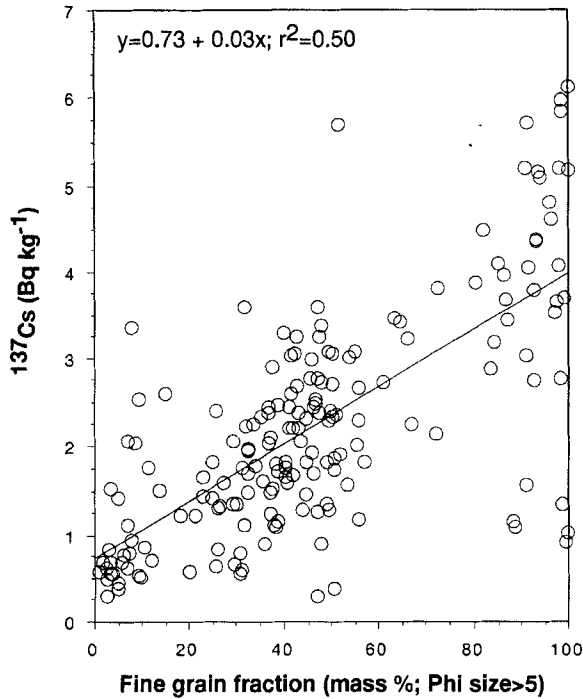


FIGURE 2 Percentage of finer grained sediments (>5 phi) as a fraction of total sediment dry weight relative to radiocaesium activity, decay-corrected to January 1, 1996.

DISCUSSION

Potential for Radiocaesium Removal by Biological Productivity

The Bering and Chukchi continental shelves are characterized by highly variable biological productivity, with unproductive Alaska Coastal Water to the east ($\sim 10 \text{ g C m}^{-2} \text{ year}^{-1}$) and much more productive Anadyr Water to the west (locally as high as $800 \text{ g C m}^{-2} \text{ year}^{-1}$), and a mixed Bering Shelf water in between (Walsh *et al.*, 1989; Springer *et al.*, 1996). Because of the shallow shelf, much of this variation in pelagic productivity is transmitted to the underlying sediments, with lower C/N ratios, higher benthic biomass and higher sediment oxygen demand underneath the more productive Anadyr and Bering Shelf waters (Grebmeier and McRoy, 1989; Grebmeier and

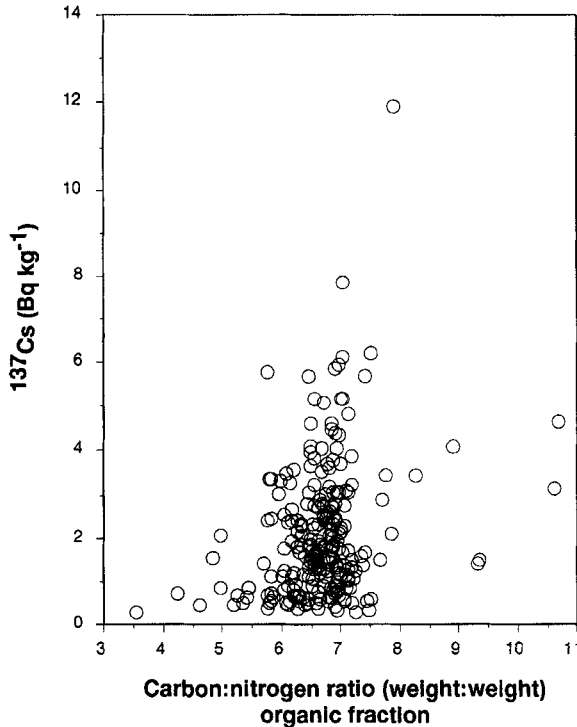


FIGURE 3 Carbon:nitrogen (weight:weight) ratios of organic fraction of surface sediments relative to radiocaesium activities, decay-corrected to January 1, 1996.

Barry, 1991). Despite this difference in productivity, and the direct coupling of this pelagic productivity to the shallow benthos, there is no evidence that the higher productivity on the western, Asian side of the continental shelf results in the accumulation of greater amounts of radiocaesium in the underlying surface sediments. We base this conclusion on the lack of any significant relationship between C/N ratios and surface radiocaesium activities in surface sediments (Fig. 3). Although the lack of an observed correlation indicates that high biological production has no significant impact upon radiocaesium removal from the water column, it might be argued that biological processes that remove radiocaesium from the dissolved ionic form do not remove caesium preferentially relative to other dissolved sea water constituents that are incorporated into marine particulates. If the ratio

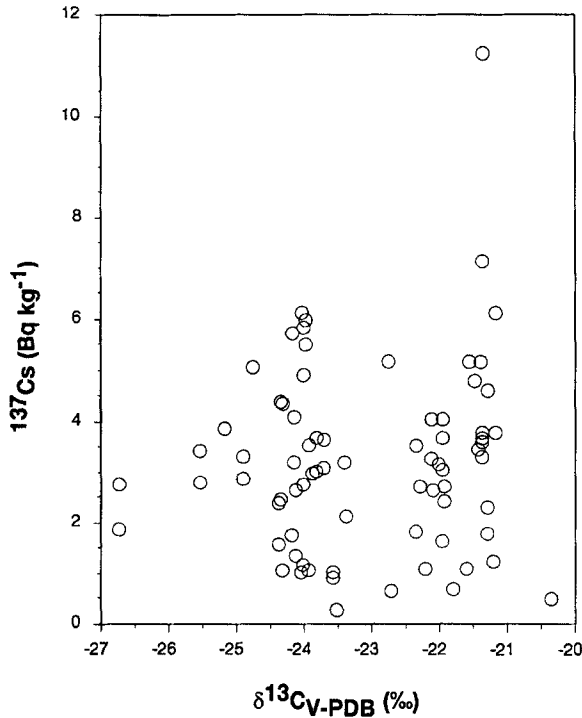


FIGURE 4 Carbon isotope composition of organic fraction of surface sediments relative to radiocaesium activities, decay-corrected to January 1, 1996. Data are limited to collections made on Alpha Helix cruise 189, Chukchi and East Siberian Seas, August–September, 1995.

of radiocaesium incorporated into settling particulate organic matter relative to other stable components of organic matter (*e.g.*, carbon, nitrogen, phosphate, silica), is a constant regardless of the rate of biological productivity, we would expect similar surface radiocaesium activities in unproductive and productive waters, which is what is observed (Fig. 3). It is also possible that radiocaesium removed by biological activity is not bound irreversibly upon reaching the sediments, and is subject to being released back into the water column. For example, radiocaesium removed by biological activity might migrate within the sediments during diagenesis or be released into interstitial waters.

Taking this possible argument one step further, if biological removal of radiocaesium from the water column is at all significant, total core

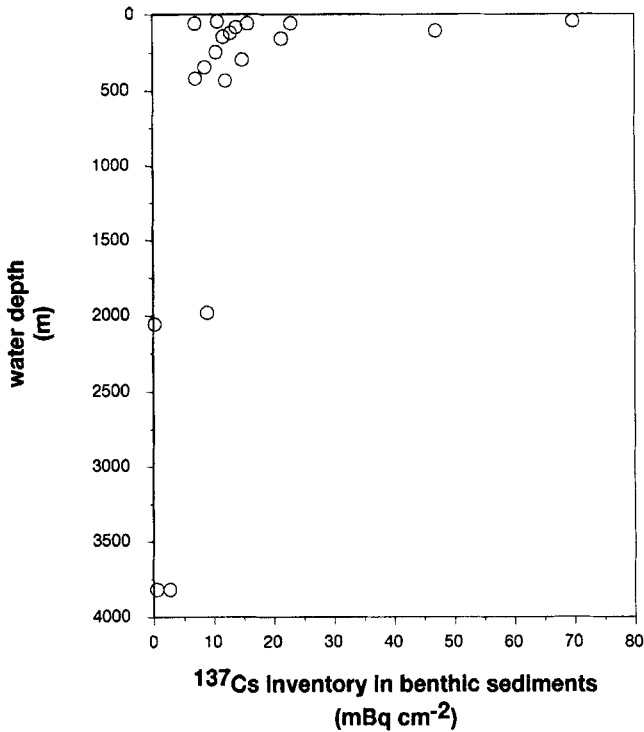


FIGURE 5 Total radiocaesium inventories in sediments relative to water depth from which the core was collected. Cores were collected in 1993–1995, and are decay-corrected to date of collection. Profiles of ^{137}Cs are shown for 10 of these 19 cores on Figure 6.

inventories of radiocaesium in the sediments should be higher underlying productive waters, when direct deposition to the underlying sediments is prevalent, as in this study. Although our numbers of complete cores and calculated inventories are far from as extensive as our data on surface radiocaesium activity, there is no clear evidence that total radiocaesium inventories are higher under more productive waters of the Bering and Chukchi continental shelves. The highest single inventory was observed in the relatively unproductive East Siberian Sea, and the Chukchi Sea continental shelf cores collected on the USCGC Polar Sea (Fig. 6) were also collected in unproductive areas influenced by Alaska Coastal Water.

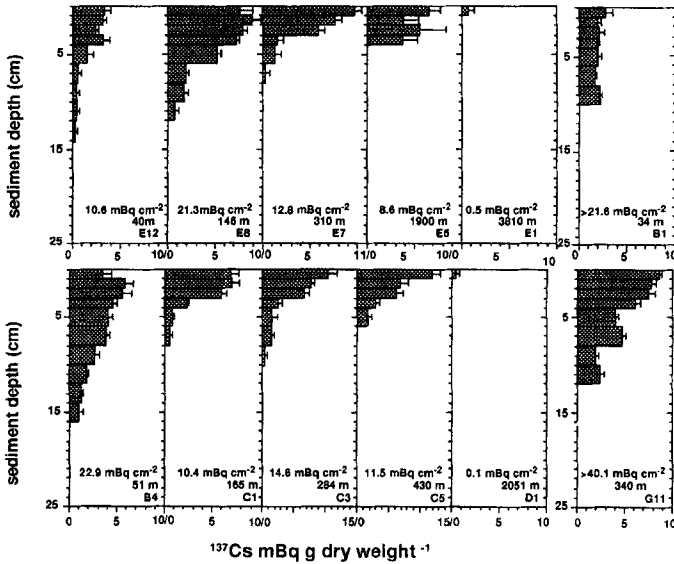
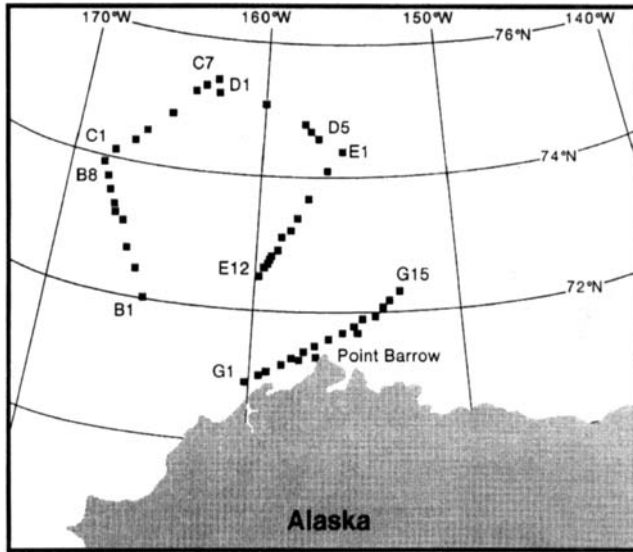


FIGURE 6 Radiocaesium inventories and distributions in 12 sediment cores collected from the USCGC Polar Star, August, 1993. Activities correspond to date of collection. Two cores (B1 and G11) still contained detectable ^{137}Cs at the bottom of the recovered cores, and are therefore not considered complete inventories. Error bars shown are $\pm 1\sigma$.

Riverine Inputs of Radiocaesium

The presence of the relatively high inventory in the one East Siberian Sea core, close to the mouth of the Kolyma River, implicates river transport of radiocaesium from terrestrial sources (reviewed by Olsen *et al.*, 1993), as being more crucial in determining total inventories than any deposition of formerly dissolved radiocaesium that is removed from the water column by biological activity. However, as with biological productivity, the proximity to terrestrial runoff does not result in higher radiocaesium surface activities, even if total inventories may be higher in river out-flow areas. No correlation was observed between radiocaesium activities and the stable carbon isotope composition of organic carbon in associated carbonate-free sediments (Fig. 4). If terrestrial sediment runoff had an inherently higher radiocaesium activity than marine sediments, we should have observed higher surface activities where terrestrial contributions are greater. The lack of a significant correlation (Fig. 4) indicates that terrestrial sedimentary runoff does not have higher radiocaesium activity (in Bq g^{-1}) than that found in predominantly marine sediments, even if terrestrial runoff does result in higher total inventories (in Bq cm^{-2}). This result is somewhat surprising because total radiocaesium inventories in terrestrial tundra in this region are generally higher (Cooper *et al.*, 1995; Cooper *et al.*, 1996) than we report here for marine cores. Also, while the atmospheric flux of radiocaesium at specific latitudes would have been relatively even over both ocean and land, most radiocaesium in the ocean remains in a dissolved form, and is not recorded in sediment inventories, unlike on land, where most is now bound to mineral surfaces (Zucker *et al.*, 1984; Avery, 1996). Since river runoff includes both terrestrial sediments uncontaminated by nuclear weapons testing as well as terrestrial sediments contaminated by radiocaesium, it is possible that the annual proportion of radiocaesium that is being lost from land surfaces in the Kolyma basin is relatively small compared to sedimentation derived from terrestrial sediments uncontaminated by nuclear weapons testing.

It is uncertain if this pattern is valid for other Arctic rivers. Surface radiocaesium activities are significantly higher in sediments at the mouth of the Yenisey River (up to 70 Bq g^{-1}) (Vakulovsky *et al.*, 1995;

Baskaran *et al.*, 1996). It is not clear if these higher activities are in part due to releases from the nuclear fuel cycle facilities at Krasnoyarsk, or simply reflect a higher degree of terrestrial runoff of fine particles with sorbed bomb fallout relative to the order of magnitude lower activities we observe at the mouths of more pristine rivers such as the Kolyma or the Yukon (Cooper *et al.*, 1995).

Based upon the physical chemical association of caesium with fine clay minerals including illite, which predominates in Arctic sediments (Darby *et al.*, 1989) and has a high capacity to exchange with caesium (Tamura and Jacobs, 1960), it is possible that the proportions of illite (and smectite) play a role in determining areas of radiocaesium accumulation on Arctic continental shelves. Nevertheless, we recently found no significant correlation between proportions of any specific clay mineral in Arctic sea ice-entrained sediments and radiocaesium activities (Cooper *et al.*, 1998).

Significant positive correlations were observed between surface radiocaesium activities and sediment total organic carbon (Fig. 1) and the proportion of fine grained sediments (Fig. 2). It seems most reasonable that the causative association is between fine grain proportions, as controlled by current flow, rather than any association between radiocaesium and sediment organic content. In previous work (Cooper *et al.*, 1995), we pointed out that the patterns of surface radiocaesium activities in the Bering and Chukchi Sea bear a striking relationship to current flow patterns. In regions such as Bering and Anadyr Straits, where there is high current flow, radiocaesium activities in surface sediments are low. This pattern is also observed south of St. Lawrence Island, where a winter polynya apparently facilitates brine-induced currents that transport fine grained sediments southward and away from the island (Grebmeier and Cooper, 1995).

Potential Non-fallout Sources of Radiocaesium

The spatial distribution of radiocaesium in Bering, Chukchi, East Siberian and Beaufort Sea sediments reported here are significantly different from sediment activities reported for some other Arctic continental shelves. For example, mean radiocaesium sediment

activities in surface sediments of the Laptev and Kara Seas are significantly higher (Tab. I; Baskaran *et al.* (1996); also cited from Cooper *et al.*, in-press (1998)). These significant differences are based upon unpaired *t*-test comparisons among each of the individual cruises reported here and each set of data from the Laptev and Kara Seas. In all cases, the probability of the null hypothesis being correct is less than 0.05, in most cases < 0.001, that there is no significant difference among our measured radiocaesium activities, and the reported Laptev and Kara Sea sediment radiocaesium activities (Tab. I).

There is a considerable evidence that radionuclides, including ^{137}Cs , that are discharged as waste from Sellafield (United Kingdom) and La Hague (France) nuclear fuel reprocessing plants, are transported northward to waters of the Arctic Ocean via the Norwegian Coastal Current (*e.g.*, Aarkrog, 1994; Dahlgaard, 1994). Water column concentrations of ^{137}Cs in coastal waters of the Barents and Kara Seas, were for example 3 to 20 Bq m^{-3} in 1992 (Strand *et al.*, 1994) compared to $\sim 1-3 \text{ Bq m}^{-3}$ observed in shallow waters of the Bering and Chukchi Seas in 1988 (Medinets *et al.*, 1992). The magnitude of the differences in water column concentrations of the radionuclide between the European and North American Arctic shelves are similar to the differences observed in mean surface sediment burdens (Tab. I). The possible incorporation of at least some western European derived radiocaesium into Russian Arctic continental shelf sediments suggests that biological and or physical processes are effective in removing a measurable portion of the dissolved water column radiocaesium burden to the surface sediments. One major difference between the Laptev and Kara Sea shelves and much of our study area, however, is the markedly lower benthic biomass on the west and central Russian Arctic continental shelves. The probable lower degrees of bioturbation present on those shelves may be the basis of the observed higher surface radiocaesium activities. In the Bering and Chukchi Seas, burial of anthropogenic radiocaesium well below the surface is prevalent (Fig. 6; see also Cooper *et al.*, 1995), but the lower biological activity on the Russian Arctic shelves may keep radiocaesium activity much more concentrated within the top several layers of marine sediments, and thus bias the mean surface activities comparison (Tab. I). Total radiocaesium inventory data presented by Baskaran *et al.* (1996) are consistent with this hypothesis. In five cores collected within the Kara

Sea, total radiocaesium inventories ranged from 15.7 mBq cm^{-2} to 160 mBq cm^{-2} . Only the highest inventory observed is significantly higher than total inventories reported here. Despite the higher apparent surface radiocaesium activity in surface sediments of the Eurasian Arctic, these higher surface activities may be misleading in the absence of total core inventory data. Additional information on bioturbation rates and radiocaesium penetration depths in Barents and Kara Sea sediments, and the collection and reporting of additional total core inventories from these less productive Arctic shelves, will be required prior to establishing the cause of the apparent difference in surface radiocaesium activities between the American and Eurasian Arctic marginal seas.

In the interim, however, we conclude that atmospheric bomb fallout is the only significant source of radionuclide contamination in sediments of the Bering, Chukchi, Beaufort and East Siberian Seas. Physical resuspension of fine sediments appears to be the major determinant of surface sediment radioactivities. Finally, even in the most productive portions of these continental shelf seas, biological removal of dissolved radiocaesium from the water column appears to be, at most, of minor importance.

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