

Transport of ^{137}Cs and $^{239,240}\text{Pu}$ with Ice-rafted Debris in the Arctic Ocean

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ABSTRACT. Ice rafting is the dominant mechanism responsible for the transport of fine-grained sediments from coastal zones to the deep Arctic Basin. Therefore, the drift of ice-rafted debris (IRD) could be a significant transport mechanism from the shelf to the deep basin for radionuclides originating from nuclear fuel cycle activities and released to coastal Arctic regions of the former Soviet Union.

In this study, 28 samples of IRD collected from the Arctic ice pack during expeditions in 1989–95 were analyzed for ^{137}Cs by gamma spectrometry and for ^{239}Pu and ^{240}Pu by thermal ionization mass spectrometry. ^{137}Cs concentrations in the IRD ranged from less than 0.2 to 78 $\text{Bq}\cdot\text{kg}^{-1}$ (dry weight basis). The two samples with the highest ^{137}Cs concentrations were collected in the vicinity of Franz Josef Land, and their backward trajectories suggest origins in the Kara Sea. Among the lowest ^{137}Cs values are seven measured on sediments entrained on the North American shelf in 1989 and 1995, and sampled on the shelf less than six months later.

Concentrations of $^{239}\text{Pu} + ^{240}\text{Pu}$ ranged from about 0.02 to 1.8 $\text{Bq}\cdot\text{kg}^{-1}$. The two highest values came from samples collected in the central Canada Basin and near Spitsbergen; calculated backward trajectories suggest at least 14 years of circulation in the Canada Basin in the former case, and an origin near Severnaya Zemlya (at the Kara Sea/Laptev Sea boundary) in the latter case. While most of the IRD samples showed $^{240}\text{Pu}/^{239}\text{Pu}$ ratios near the mean global fallout value of 0.185, five of the samples had lower ratios, in the 0.119 to 0.166 range, indicative of mixtures of Pu from fallout and from the reprocessing of weapons-grade Pu. The backward trajectories of these five samples suggest origins in the Kara Sea or near Severnaya Zemlya.

Key words: sea ice, ice-rafted debris, radionuclides, cesium-137, plutonium

RÉSUMÉ. Le transport glaciaire constitue le principal mécanisme responsable du transport des sédiments à grain fin depuis les zones côtières jusqu'à la fosse du bassin Arctique. La dérive des débris du transport glaciaire pourrait constituer un important mécanisme de transport, depuis la plate-forme continentale jusqu'à la fosse marine, pour des radionucléides provenant d'activités connexes au cycle du combustible nucléaire, radionucléides qui sont éliminés vers les zones côtières arctiques de l'ancienne Union Soviétique.

Dans cette étude, on a analysé 28 échantillons de débris de transport glaciaire recueillis dans la glace arctique au cours d'expéditions effectuées de 1989 à 1995, en vue d'y déceler du ^{137}Cs par spectrométrie gamma ainsi que du ^{239}Pu et du ^{240}Pu par spectrométrie de masse réalisée par thermo-ionisation. Les concentrations de ^{137}Cs dans les débris de transport glaciaire allaient de moins de 0,2 à 78 $\text{Bq}\cdot\text{kg}^{-1}$ (poids sec). Les deux échantillons ayant les concentrations en ^{137}Cs les plus élevées ont été recueillis à proximité de l'archipel François-Joseph, et leurs trajectoires régressives suggèrent qu'ils proviennent de la mer de Kara. Parmi les plus faibles valeurs de ^{137}Cs , sept ont été mesurées sur des sédiments arrivés sur la plate-forme continentale nord-américaine en 1989 et 1995 et prélevés sur celle-ci moins de six mois plus tard.

Les concentrations en ^{239}Pu et ^{240}Pu allaient d'environ 0,02 à 1,8 $\text{Bq}\cdot\text{kg}^{-1}$. Les deux valeurs les plus élevées venaient d'échantillons recueillis au centre du bassin Canada et près du Spitzberg; le calcul des trajectoires régressives suggère que le ^{239}Pu est resté au moins 14 ans en circulation dans le bassin Canada et que le ^{240}Pu tire son origine des environs de Severnaïa Zemlia (à la frontière de la mer de Kara et de la mer des Laptev). Tandis que la plupart des échantillons de débris de transport glaciaire révélaient des rapports $^{240}\text{Pu}/^{239}\text{Pu}$ proches de la valeur moyenne (0,185) des retombées radioactives mondiales, cinq des échantillons affichaient des rapports inférieurs, allant de 0,119 à 0,166. Cette fourchette est caractéristique de mélanges de Pu provenant de retombées radioactives et du retraitement du Pu pouvant être utilisé à des fins militaires. Les trajectoires régressives de ces cinq échantillons suggèrent qu'ils proviennent de la mer de Kara ou des environs de Severnaïa Zemlia.

Mots clés: glace de mer, débris de transport glaciaire, radionucléides, césium 137, plutonium

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INTRODUCTION

Sea ice in the Arctic Basin is produced by large-scale divergences within the ice pack, by the fall freezing of the seasonally ice-free waters, and by wind-driven ice production in flaw leads (i.e., the open water zones between the pack ice and the coastal, fast ice) during the winter. The ice cover of the Arctic Ocean consists largely of sea ice formed on surrounding continental shelves (Colony and Thorndike, 1985; Reimnitz et al., 1992). Rigor and Colony (1997) estimated that 256 000 km² of sea ice is produced in the shallow waters of the Laptev Sea. The predominant wind patterns in the Laptev Sea are conducive to flaw lead production of sea ice and to northward advection of this new ice into the Arctic Basin. In the Kara Sea, an estimated 209 000 km² of sea ice is produced; sea ice production in the Laptev and Kara Seas accounts for at least 20% of the total area of ice flux through the Fram Strait (I. Rigor and R. Colony, pers. comm. 1997).

In cold regions, such as the coastal zones bordering the Arctic Ocean, ice represents an important medium for the transport of sediments. "Frazil ice" consists of fine ice crystals that form in suspension in supercooled, turbulent water. Such ice has been shown to scavenge fine-grained sediment efficiently from the water column while rising to the surface. Thus, on a shallow coastal shelf, where storm-induced turbulence has produced an initial suspension of fine-grained bottom sediments, "suspension freezing" and the formation of a seawater-slush layer of frazil ice can entrain high concentrations of fine-grained sediment. This slush congeals into a solid ice pack, which can move seaward, away from the zone of entrainment, under the influence of wind and currents. Frazil ice that attaches to bottom sediments in such shelf regions forms "anchor ice," which can later break loose and carry coarser-grained bottom sediments into the forming coastal pack ice. These two processes yield turbid ice with sediment concentrations much higher than those of the ambient seawater (Kempema et al., 1986; Clayton et al., 1990; Reimnitz et al., 1992, 1993a, b).

Because they are so shallow and wide, the Eurasian shelf regions (Fig. 1) are much more important for sediment entrainment into sea ice than those of North America (Pfirman et al., 1989, 1990; Reimnitz et al., 1993a). Ice rafting appears to be the predominant mechanism responsible for the transport of fine-grained sediments from coastal zones to the deep Arctic Basin (Barnes et al., 1982; Kempema et al., 1989). Recent reports (Yablokov et al., 1993; Office of Technology Assessment, 1995) documenting past radioactive waste disposal practices in the former Soviet Union have raised concerns regarding the release of radioactive materials into the environment of coastal Arctic regions and into major rivers, such as the Ob and Yenisey, which drain into the Arctic Ocean. Therefore, ice-rafted debris (IRD) sampled in a variety of locations in the Arctic Ocean may give valuable information regarding the flux of sediment-bound, anthropogenic radionuclides from zones of release to other parts of the ocean basin and borders.

MATERIALS AND METHODS

Most of the IRD samples available for this study were collected in different years (1989–95) from various ice-breakers wherever opportunities arose. The samples tend to cluster into four groups (Fig. 1). The *Polar Star* 1989 samples cluster over the continental shelf and slope east of Prudhoe Bay, Alaska. To this cluster can be added the samples shown in Figure 1 as numbers 27 and 28, because all represent entrainment off North America, as discussed below; these latter two samples, from the shelf a little farther to the west, were collected by helicopter in a land-based expedition in March 1995 (referred to hereafter as "Alaska 1995"). The *Polar Star* 1993 samples represent a cluster along a deep water transect extending north from Barrow, Alaska. The *Polar Star* 1991 samples and part of the *Polarstern* 1993 samples came from the vicinity of Svalbard and Franz Josef Land. The remaining *Polarstern* 1993 samples came from the Laptev Sea (Table 1; Fig. 1).

IRD samples were collected from the surface of ice floes, where they are concentrated mainly by summer melting from underlying, sediment-rich "dirty ice." During summer, this sediment commonly occurs at the bottom of meltwater depressions of various sizes. The water with associated sediment often was drawn up with a turkey baster into a container. For some samples, where the surface and upper 3 cm of ice contained the highest sediment concentration, the sample was collected by scraping with a sharpened spatula, spoon, or shovel; after mid-August, the sediment-rich surface ice was sometimes blanketed by clean new snow, which could not be totally eliminated from samples collected. After these samples were melted aboard ship, the containers held turbid water, to which a small amount of table salt was added (to ~10–15 ppt NaCl) to speed flocculation. As this concentration is about one-third to one-half of the total dissolved solids concentration of seawater (~34 ppt), desorption of radionuclides from the IRD beyond that which would have already occurred from prior contact with seawater is assumed to be minimal. The clear water then was siphoned off, and the remaining mud placed into small containers and stored under refrigeration. Cuts of 10 to 100 g of sediment (dry weight basis) were used for radionuclide analyses. These cuts were dried at 54°C and lightly crushed with a mortar and pestle. The remaining sample material was used for sedimentological, mineralogical, and micropaleontological studies.

Grain size distributions in the clay–silt range were determined by pipette procedures (Galehouse, 1971). Sand fractions were analyzed using a Rapid Sediment Analyzer (RSA) modified after Gibbs (1974), measuring the settling rates of different grain sizes in a sample, and relating them to known settling velocities of different sphere diameters. Textural comparisons were made using parameters for sorting, skewness, and mean size (Folk and Ward, 1957). The total carbon and inorganic carbon analyses of IRD samples were done using a CO₂ coulometer with an induction furnace and

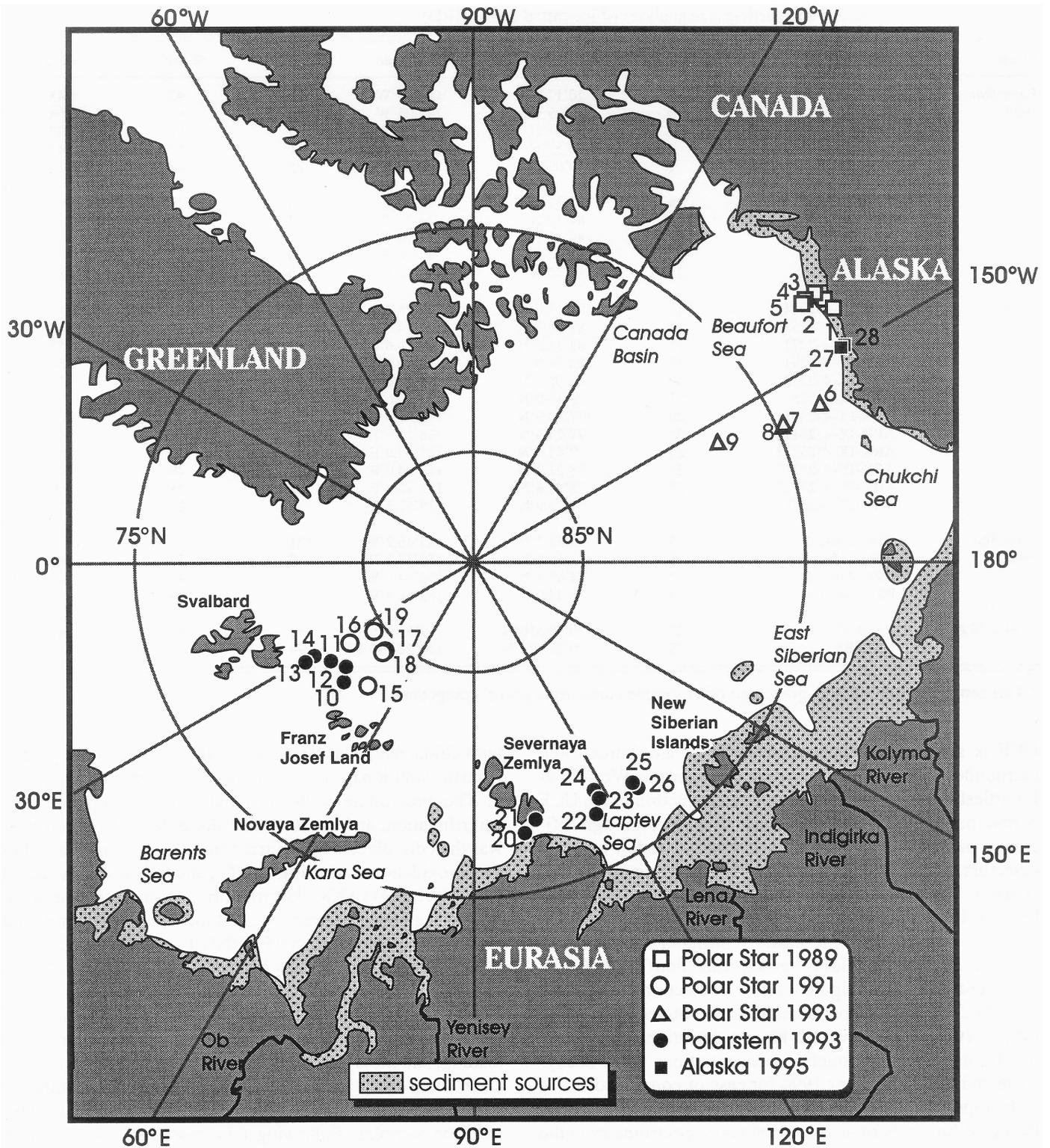


FIG. 1. Sampling locations in the Arctic Ocean. Shelf regions shallower than 30 m are shown as "sediment sources" (Reimnitz et al., 1992). Much of the stippled area is protected by fast ice most of the year, so that it is not likely to be involved in sediment entrainment in ice that escapes the shelves and survives to be included in the central Arctic drift.

acid digester (Huffman, 1977). The percentages of organic carbon were determined by subtracting inorganic carbon percentages from the total carbon.

Gamma-emitting radionuclides were determined by high-resolution gamma spectrometry using high-purity germa-

nium (HPGe) detectors. The dried sediment was placed in a standard geometry container and counted for 16 hours or more. These samples were counted on a 90% efficient HPGe detector located in the Underground Counting Facility (UCF) at the U.S. Department of Energy Savannah River Site. The

TABLE 1. Sample locations and particle size analyses of ice-rafted debris (IRD).

Cruise	Station	Map Location #	Latitude	Longitude	% Sand	% Silt	% Clay
<i>Polar Star</i> 1989	PS89-12	2	70°17'N	143°34'W	15	42	43
	PS89-15	5	71°09'N	142°05'W	0	42	58
	PS89-18	3	70°31'N	142°08'W	9	59	32
	PS89-19	4	70°53'N	141°52'W	1	52	47
	PS89-23	1	70°09'N	144°59'W	25	44	31
<i>Polar Star</i> 1991	PS91-229-1-11	16	83°21'N	32°51'E	0	20	80
	PS91-236-1-11	19	84°30'N	34°32'E	84	5	11
	PS91-241-1-06	18	84°19'N	45°10'E	1	5	94
	PS91-241-2-02	17	84°14'N	44°48'E	0	12	88
	PS91-245-1-11	15	82°40'N	49°14'E	0	20	80
<i>Polarstern</i> 1993	ARK-IX-4-2241	13	81°13.0'N	30°26.7'E	0	46	54
	ARK-IX-4-2251	14	81°39.2'N	30°14.3'E	100	0	0
	ARK-IX-4-2271	11	82°11.5'N	34°29.6'E	2	41	57
	ARK-IX-4-2291	12	82°34.0'N	39°19.0'E	1	45	54
	ARK-IX-4-2332	10	82°05.0'N	42°28.0'E	0	24	76
	ARK-IX-4-23811	21	78°10.0'N	103°05.0'E	0	43	57
	ARK-IX-4-24011	20	77°40.0'N	100°31.9'E	0	35	65
	ARK-IX-4-25122	26	77°24.3'N	126°12.9'E	2	52	46
	ARK-IX-4-25313	25	77°41.4'N	125°54.9'E	2	45	53
	ARK-IX-4-25812	24	78°21.2'N	117°54.0'E	7	48	45
	ARK-IX-4-25822	23	78°02.4'N	118°12.7'E	3	38	59
	ARK-IX-4-2621	22	77°25.9'N	115°57.5'E	1	27	72
<i>Polar Star</i> 1993	PS93-230-1-1	6	72°52.7'N	155°46.2'W	*21	11	4
	PS93-235-1-2	8	74°50.7'N	156°47.9'W	0	52	48
	PS93-236-1-2	7	74°49.4'N	156°44.6'W	10	47	43
	PS93-241-1-4	9	77°44.0'N	153°58.4'W	1	43	56
Alaska, 1995	I195AR3	27	70°46.516'N	149°51.296'W	13	46	41
	I195AR8	28	70°38.978'N	149°52.800'W	99	1	0

* This sample contained 64% gravel; no other sample contained a gravel component.

UCF is shielded by more than 12 m of overburden and surrounded by 1.2 m of specular hematite and pre-World War II battleship steel. Further background reduction in the UCF is obtained by removing particles in the air with high-efficiency particulate air filters, by displacing radon near the detectors, and by preventing contamination using clean room controls. The data were analyzed using customized software for low-level counting (Winn, 1987). The 90% efficient HPGe is situated within a plastic scintillator shield, which detects and rejects cosmic background radiation (Winn, 1991). These controls and equipment allow a detection limit of less than 0.02 Bq of ^{137}Cs per sample to be obtained routinely; thus, a 100 g sample would have a detection limit below 0.2 Bq·kg⁻¹. All radionuclide concentrations were decay-corrected to 1 September 1994 for ease of comparison.

Isotopes of plutonium (^{239}Pu , ^{240}Pu) were determined by isotope dilution thermal ionization mass spectrometry in the Environmental Technology Section Mass Spectrometry Facility (Buesseler and Halverson, 1987; Beals et al., 1995). After completing the gamma spectrometry analysis, a 10 g sample of the sediment (only 5 g of sample PS91-245-1-11) was spiked with a ^{242}Pu tracer (10 pg). The sample was leached with hot 8M nitric acid and hydrogen peroxide for several hours. The slurry was transferred to a centrifuge tube and the undigested residue separated from the supernate. The supernate was taken to near dryness and then diluted

with deionized water. Hydroxides of iron were precipitated from the solution by adding ammonium hydroxide to a pH of 8. The precipitate with entrained Pu was separated by centrifugation, and the supernate discarded. The precipitate was then dissolved in 8M nitric acid. The Pu was reduced to the 4+ oxidation state using NaNO_2 , and the Pu was extracted onto BioRad AG 1X8, chloride form, anion exchange resin. Interfering elements were washed through the column, and Pu was finally eluted with a $\text{HCl}/\text{NH}_4\text{I}$ solution. The purified Pu was loaded onto a resin bead, which was mounted on a filament for mass spectrometric analysis. The isotopic abundances were measured on a single sector mass spectrometer (30.5 cm, 90° deflection). All the above purification, bead loading, and mass spectrometric analyses were conducted in clean room facilities, class 10 000 or better. All results are corrected for a process blank which was run concurrently with the samples. Following this procedure the detection limits for a 10 g sediment sample are: ^{239}Pu , 0.91 mBq·kg⁻¹; ^{240}Pu , 3.35 mBq·kg⁻¹.

Backward trajectories have been calculated for sea ice sampled to help identify source areas. Pfirman et al. (1997a) have analyzed monthly fields of ice motion in the Arctic Basin using optimal interpolation (Gandin, 1963; Thorndike, 1986) to combine buoy motions and observed winds from the International Arctic Buoy Program (Rigor and Heiberg, 1995). These fields take advantage of the reduced error in

the measurement of monthly ice velocities by buoys ($e_u = 0.02 \text{ cm s}^{-1}$), the longer correlation length scale of 1400 km between monthly observations of ice motion, and, for areas where the buoy data is sparse, the intrinsic relationship between ice motion and the geostrophic winds (Pfirman et al., 1997a). Using these monthly fields of ice motion, Pfirman et al. (1997a) have shown that clay minerals taken from sea ice sampled in the Arctic Basin can be tracked back to the shelf source areas by matching mineralogies. These same fields of ice motion have been used here to backtrack radionuclide samples. Given a sampling position and time, the back trajectories are estimated by integrating the positions backwards in time to the near-shore/fast-ice zones, their most likely points of origin.

RESULTS AND DISCUSSION

Most of the samples were silt-clay mixtures. However, four samples contained more than 20% sand and coarser material; one of these (PS93-230-1-1) was predominantly a gravel (Table 1). These coarse materials probably reflect shelf materials entrained by anchor ice, while the predominantly fine-grained samples are thought to result from suspension freezing. Preliminary studies of benthic foraminifera and ostracodes, when present in the samples, indicate that they were entrained on shelf surfaces mainly shallower than 30 m, as indicated in Figure 1. Analyses of these microfossils so far do not reveal whether they lived off North America or off Siberia. Mineralogical investigations are in progress to help delineate what part of the circum-Arctic shelf region is the sediment source area.

The ^{137}Cs and Pu-isotope concentrations in the IRD are shown in Tables 2 and 3, respectively. The linear correlation coefficient (LCC) between the ^{137}Cs and total Pu concentrations is 0.78. The coarse-textured samples tend to be low in both ^{137}Cs and Pu. When looking at the complete sample set, the LCCs for % clay content vs ^{137}Cs and Pu concentrations are 0.69 and 0.62 respectively; for % organic carbon content vs ^{137}Cs and Pu concentrations, the LCCs are 0.52 and 0.61 respectively. The ^{137}Cs correlation with clay content (Fig. 2) was greater than that observed by Meese et al. (1997) for IRD sampled during the 1994 Arctic Ocean Section (AOS-94) transect from the Chukchi Sea to the North Pole [LCC = 0.11]; however, it must be pointed out that the samples in our study show a larger particle size range than the uniformly fine-grained AOS-94 samples. The ^{137}Cs correlation with organic carbon content for the AOS-94 samples was also poor.

Other gamma-emitting radionuclides detected were ^{60}Co , ^{134}Cs , and ^{152}Eu , as shown in Table 2. Only one sample (ARK-IX-4-2291, collected near Franz Josef Land) shows detectable levels of the short half-life (2 y) ^{134}Cs (Table 2). The $^{134}\text{Cs}/^{137}\text{Cs}$ ratio is consistent with that of worldwide fallout, including Chernobyl effects; however, some of the radiocesium may also be from reactor sources. ^{60}Co is a neutron activation product often observed in the discharge water of reactor coolant lines. ^{152}Eu is a fairly long-lived (half

TABLE 2. Concentrations (in $\text{Bq}\cdot\text{kg}^{-1} \pm 1 \sigma$) of ^{137}Cs and other gamma-emitting nuclides in IRD, dry weight basis (errors are 1 σ counting error; systematic errors $\sim 5\%$)

Cruise/Station	Map Location #	^{60}Co	^{134}Cs	^{137}Cs	^{152}Eu
<i>Polar Star</i> 1989					
PS89-12	2			5.4 ± 0.3	
PS89-15	5			7.9 ± 0.3	
PS89-18	3			4.0 ± 0.3	
PS89-19	4			4.7 ± 0.2	
PS89-23	1			3.4 ± 0.3	
<i>Polar Star</i> 1991					
PS91-229-1-11	16			31.0 ± 0.7	
PS91-236-1-11	19			8.9 ± 0.4	
PS91-241-1-06	18	1.9 ± 0.2		71.5 ± 1.1	3.4 ± 0.6
PS91-241-2-02	17	2.9 ± 0.3		78.1 ± 1.1	4.3 ± 0.9
PS91-245-1-11	15			17.6 ± 0.8	
<i>Polarstern</i> 1993					
ARK-IX-4-2241	13	0.5 ± 0.1		48.8 ± 0.7	
ARK-IX-4-2251	14			0.4 ± 0.1	
ARK-IX-4-2271	11			29.8 ± 0.6	
ARK-IX-4-2291	12		0.6 ± 0.1	37.0 ± 0.6	
ARK-IX-4-2332	10	1.0 ± 0.2		64.4 ± 0.7	
ARK-IX-4-23811	21	0.8 ± 0.2		54.8 ± 1.1	
ARK-IX-4-24011	20			29.1 ± 0.6	
ARK-IX-4-25122	26			24.6 ± 0.5	
ARK-IX-4-25313	25			19.5 ± 0.4	
ARK-IX-4-25812	24			23.3 ± 0.4	
ARK-IX-4-25822	23			33.3 ± 0.6	
ARK-IX-4-2621	22			35.5 ± 0.5	
<i>Polar Star</i> 1993					
PS93-230-1-1	6			1.7 ± 0.2	
PS93-235-1-2	8			18.7 ± 0.4	
PS93-236-1-2	7			58.8 ± 0.7	
PS93-241-1-4	9			67.3 ± 0.7	
Alaska, 1995					
I195AR3	27			3.2 ± 0.2	
I195AR8	28			< 0.2	

life = 12.7 y) neutron-activated fission product; it is probably from reactor sources, but its production in power reactors is much lower than that of ^{154}Eu and ^{155}Eu (Benedict et al., 1981), which were not observed. However, ^{152}Eu has been observed to dominate the activity of Eu isotopes in effluents of research reactors (Hooper et al., 1994).

The following screening criteria were applied to the data in Tables 2 and 3 to identify samples with elevated radionuclide contents and anomalous Pu isotope ratios: 1) ^{137}Cs concentration greater than $50 \text{ Bq}\cdot\text{kg}^{-1}$; 2) $^{239}\text{Pu} + ^{240}\text{Pu}$ concentration greater than $1000 \text{ mBq}\cdot\text{kg}^{-1}$; 3) $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio which differs by more than three standard deviations from the mean global fallout (above 55°N latitude) ratio of 0.1853 ± 0.0053 (1σ). This value represents the mean of isotopic ratios measured by Krey et al. (1976) for soils collected above 55°N latitude (Beasley et al., 1996). Table 4 shows the subset of samples meeting one or more of these criteria.

For comparative purposes, concentrations of $^{239}\text{Pu} + ^{240}\text{Pu}$ and ^{137}Cs in sediments from various marine and terrestrial environments have been tabulated from the literature (Tables 5 and 6). The mass spectrometric Pu concentrations measured for the IRD samples have been converted to radioactivity units (Table 3) for ease of comparison with the alpha

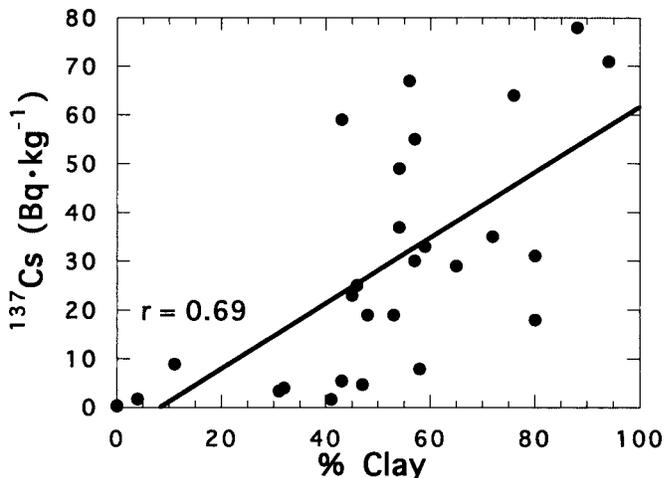
TABLE 3. Plutonium concentrations and isotopic composition of IRD, dry weight basis (not determined for Alaska, 1995 samples).

Cruise	Station	Map Location #	Total Pu pg·g ⁻¹	²³⁹ Pu + ²⁴⁰ Pu mBq·kg ⁻¹	²³⁹ Pu atom % ± 1 σ	²⁴⁰ Pu atom % ± 1 σ	²⁴⁰ Pu/ ²³⁹ Pu atom ratio ± 3 σ
<i>Polar Star</i> 1989	PS89-12	2	0.110	364	82.66 ± 0.20	16.61 ± 0.09	0.201 ± 0.004
	PS89-15	5	0.119	392	83.68 ± 0.15	16.22 ± 0.07	0.194 ± 0.003
	PS89-18	3	0.066	219	83.56 ± 0.15	16.30 ± 0.07	0.195 ± 0.003
	PS89-19	4	0.084	275	83.87 ± 0.14	16.13 ± 0.06	0.192 ± 0.002
	PS89-23	1	0.061	199	83.69 ± 0.15	16.31 ± 0.07	0.195 ± 0.003
<i>Polar Star</i> 1991	PS91-229-1-11	16	0.210	674	84.62 ± 0.30	15.00 ± 0.13	0.177 ± 0.005
	PS91-236-1-11	19	0.105	335	85.39 ± 0.15	14.52 ± 0.07	0.170 ± 0.002
	PS91-241-1-06	18	0.300	903	87.99 ± 0.25	11.64 ± 0.10	0.132 ± 0.003
	PS91-241-2-02	17	0.186	548	89.20 ± 0.13	10.58 ± 0.05	0.119 ± 0.002
	PS91-245-1-11	15	0.174	554	85.23 ± 0.16	14.32 ± 0.07	0.168 ± 0.003
<i>Polarstern</i> 1993	ARK-IX-4-2241	13	0.396	1255	85.55 ± 0.11	14.21 ± 0.05	0.166 ± 0.002
	ARK-IX-4-2251	14	0.005	18	84.44 ± 0.92	15.56 ± 0.44	0.184 ± 0.017
	ARK-IX-4-2271	11	0.201	643	85.13 ± 0.15	14.68 ± 0.06	0.172 ± 0.002
	ARK-IX-4-2291	12	0.225	728	84.46 ± 0.14	15.36 ± 0.06	0.182 ± 0.002
	ARK-IX-4-2332	10	0.457	1410	86.91 ± 0.12	12.94 ± 0.05	0.149 ± 0.002
	ARK-IX-4-23811	21	0.326	1017	86.41 ± 0.08	13.42 ± 0.03	0.155 ± 0.001
	ARK-IX-4-24011	20	0.166	533	84.80 ± 0.18	15.11 ± 0.05	0.178 ± 0.003
	ARK-IX-4-25122	26	0.152	496	84.01 ± 0.13	15.93 ± 0.06	0.190 ± 0.002
	ARK-IX-4-25313	25	0.139	446	84.96 ± 0.12	15.00 ± 0.05	0.177 ± 0.002
	ARK-IX-4-25812	24	0.138	447	84.59 ± 0.14	15.28 ± 0.06	0.181 ± 0.002
	ARK-IX-4-25822	23	0.166	534	84.72 ± 0.09	15.21 ± 0.04	0.179 ± 0.001
	ARK-IX-4-2621	22	0.214	690	84.65 ± 0.07	15.23 ± 0.03	0.180 ± 0.001
<i>Polar Star</i> 1993	PS93-230-1-1	6	0.033	108	83.80 ± 0.24	16.20 ± 0.11	0.193 ± 0.004
	PS93-235-1-2	8	0.134	425	85.54 ± 0.12	14.35 ± 0.05	0.168 ± 0.002
	Replicate Analysis	8	0.133	424	85.31 ± 0.13	14.63 ± 0.06	0.171 ± 0.002
	PS93-236-1-2	7	0.367	1186	84.47 ± 0.07	15.32 ± 0.03	0.181 ± 0.001
	PS93-241-1-4	9	0.563	1824	84.36 ± 0.07	15.35 ± 0.03	0.182 ± 0.001

spectrometric determinations of ²³⁹Pu + ²⁴⁰Pu found most often in the literature. The Pu concentrations in the IRD samples are generally higher than those in ocean bottom sediments in potential source areas and other regions, sampled by other investigators and summarized in Table 5. The ¹³⁷Cs concentrations in the IRD are likewise generally elevated in relation to those found in bottom sediments of the Bering, Chukchi, Greenland, and Barents Seas (Table 6). The elevated radionuclide concentrations in the IRD may reflect unidentified source areas with elevated radionuclide concentrations or the selective entrainment of fine materials of higher-than-ambient radionuclide concentration by the suspension freezing process.

²⁴⁰Pu/²³⁹Pu

A low ²⁴⁰Pu/²³⁹Pu ratio suggests a mixture of recent mean global fallout with low-yield weapons test fallout from early (pre-1959) atmospheric testing or material associated with the reprocessing of weapons-grade plutonium. As an example of the former in the Barents Sea region, Forman et al. (1996) showed that sediments from Chernaya Bay, on the southwestern coast of Novaya Zemlya (a former Soviet nuclear weapons testing site), had a ²⁴⁰Pu/²³⁹Pu ratio of 0.03. They attribute this low ratio to either low yields of ²⁴⁰Pu in relatively inefficient bomb tests, or the use of low-irradiation plutonium in weapon construction. As an example of the latter, Livingston et al. (1996) and Cochran et al. (1996) have shown low ratios in suspended and bottom sediments in

FIG. 2. Plot of ¹³⁷Cs concentration vs. clay content of IRD.

reaches of the Ob River downstream of nuclear weapons production facilities at Tomsk and Chelyabinsk. The backward trajectories of these five low-ratio samples in our study suggest origins in the Kara Sea or near Severnaya Zemlya (Fig. 3). The five low-ratio samples all exhibit elevated ¹³⁷Cs concentrations; three of these samples also exhibit a high total Pu concentration.

The April 1993 explosion at the reprocessing facility at Tomsk-7 (in the Ob River drainage basin) represents a potential source for the low ²⁴⁰Pu/²³⁹Pu ratio material and for the ¹³⁴Cs seen in some of the 1993 samples. The prevailing wind

TABLE 4. Subset of samples with elevated radionuclide concentrations and/or anomalous Pu-isotope ratios.

Cruise	Station	Map Location #	^{137}Cs ($\text{Bq}\cdot\text{kg}^{-1}$)	$^{239}\text{Pu} + ^{240}\text{Pu}$ ($\text{mBq}\cdot\text{kg}^{-1}$)	$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio
<i>Polar Star</i> 1991	PS91-241-1-06	18	71	—	0.132
	PS91-241-2-02	17	78	—	0.119
<i>Polarstern</i> 1993	ARK-IX-4-2241	13	49*	1255	0.166
	ARK-IX-4-2332	10	64	1410	0.149
	ARK-IX-4-23811	21	55	1017	0.155
<i>Polar Star</i> 1993	PS93-236-1-2	7	59	1186	—
	PS93-241-1-4	9	67	1824	—

— indicates a measured value that did not meet or exceed the screening criteria: (1) ^{137}Cs concentration greater than $50 \text{ Bq}\cdot\text{kg}^{-1}$; (2) $^{239}\text{Pu} + ^{240}\text{Pu}$ concentration greater than $1000 \text{ mBq}\cdot\text{kg}^{-1}$; (3) $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio which differs by more than 3 standard deviations from the mean global fallout (above 55°N latitude) ratio of 0.185.

* included, as within 1σ of criterion value.

TABLE 5. Published values of $^{239,240}\text{Pu}$ concentrations (dry weight basis) in surficial ocean and river sediment. Unless otherwise noted, sampling locations are remote from reported nuclear facilities or disposal sites.

Reference	Type sample; date collected	$^{239} + ^{240}\text{Pu}$ concentration [range or single value] ($\text{mBq}\cdot\text{kg}^{-1}$)
Holm et al. (1983)	ocean bottom sediment; 0–4 or 5 cm; Greenland and Barents Seas; 1980	33 – 1010
Beasley et al. (1996)	ocean bottom sediment; 0–2; Canada Basin; 1992	51 – 800
Pisias et al. (1995)	ocean bottom sediment; 0 to 1, 2, or 3 cm; Laptev Sea, 1963; Chukchi Sea, 1985	91 328–363
Buesseler (1986)	ocean bottom sediment; approx. 0–2 cm or less; N. Atlantic; 1983–85	289–1913
Miettinen (1975)	ocean bottom sediment; Gulf of Finland	660
Fowler et al. (1990)	ocean bottom sediment; 0–1 or 2 cm; Mediterranean Sea; 1983–85	44–677
Scott et al. (1983)	Mississippi River suspended sediment; Gulf of Mexico shelf sediment; 0–1 cm; 1978	225 105 – 295
Baskaran et al. (1995)	Ob and Yenisey River estuaries, and Kara Sea bottom sediment; 0–3 cm; 1993 (downstream of nuclear fuel facilities; dumped reactors in Kara Sea)	9.4 – 677 [mean = 250]
Smith et al. (1995)	Pechora Sea (>100 km from Chernaya Bay) off Novaya Zemlya nuclear weapons test site; bottom sediment; 0–2 cm; 1992	70 – 1650
Delfanti et al. (1995)	ocean bottom sediment (remote from power plant); 0–1 cm; Mediterranean Sea	~ 210 – 1000

at the time of the accident was to the northeast. Air sampling within Russia of the plume from that explosion showed ^{106}Ru to be a major component (Vakulovski et al., 1994). ^{106}Ru was detected in high-volume air samples in the United States (Winn, 1997). With its 368 day half-life, and the gamma emissions of its short-lived daughter product, ^{106}Rh , one would expect to see ^{106}Ru in the IRD if it had been contaminated by the Tomsk-7 explosion. The fact that this nuclide was not detected argues against the Tomsk accident as the source of the ^{134}Cs , the elevated ^{137}Cs concentrations, or the low $^{240}\text{Pu}/^{239}\text{Pu}$ ratios observed.

Ocean circulation modelling studies by Preller and co-workers suggest that during the 1980s, radionuclide

releases to the Irish Sea from the Sellafield fuel reprocessing facility may have represented a contribution to the Kara Sea of magnitude comparable to the river inputs (Ruth Preller, U.S. Naval Research Laboratory, Stennis Space Center, Mississippi; pers. comm. 1997). The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios of Sellafield effluents, while not reported, have been reconstructed on the basis of measurements made on an Irish Sea sediment core (Kershaw et al., 1995). The ratios increased steadily from about 0.06 in the early 1960s to a peak of about 0.24 in the early 1980s, reflecting the reactor practice of increased burn-up of fuel prior to reprocessing that occurred in later years; in the mid-1980s, the Sellafield ratios fell to near global fallout values. Thus Sellafield-

TABLE 6. Published values of ^{137}Cs concentrations (dry weight basis) in surficial soils and freshwater or marine sediments. Unless otherwise noted, sampling locations are remote from reported nuclear facilities or disposal sites.

Reference	type sample; date collected or *1 year before publication if not stated	^{137}Cs concentration [range or single value] ($\text{Bq}\cdot\text{kg}^{-1}$)	decay-corrected to mid 1994
Holm et al. (1983)	ocean bottom sediment; 0 – 4 or 5 cm; Greenland and Barents Seas; 1980	1.1 – 18	0.8 – 13
Meese et al. (1997)	ocean bottom sediment; 0 – 4 cm; Bering and Chukchi Seas; 1992–93	~0.5 – 12.9	~0.5 – 12.2
Smith et al. (1995)	Pechora Sea (>100 km from Chernaya Bay) off Novaya Zemlya nuclear weapons test site; bottom sediment; 0 – 2 cm; 1992	0.7 – 11.6	0.7 – 11.1
D'Anglejan (1980)	surficial bottom sediments; James Bay, Ontario; 1976–77	regional background ~26 max ~57	~17.2 ~37.7
Brooks et al. (1996)	Ob and Yenisey River estuaries, and Kara Sea bottom sediment; 0 – 3 cm; 1993–94 (downstream of nuclear fuel facilities; dumped reactors in Kara Sea)	<10 – >50	<9.8 – >49
Callender and Robbins (1993)	surficial (0 – 1 or 2 cm) bottom sediment, in Missouri River reservoir, South Dakota; 1986–88	~5.8 – 15	~4.9 – 12.8
Bryant et al. (1993)	lake sediment; 0 – 1 cm; Scotland; 1992*	407 – 1322	389 – 1263
Joshi et al. (1989)	surficial bottom sediment; Lake Athabasca, Saskatchewan; 1983	~10 – 100	~7.8 – 77.6
Kachanoski and De Jong (1984)	soils near Saskatoon, Saskatchewan; 0 – 10 cm; 1981	1.9 – 17	1.4 – 13
Zach et al. (1989)	forest soil; 0 – 21.5 cm; Manitoba; 1978–79	30 – 210	21 – 145
Olsen et al. (1994)	tundra soil; ~0 – 4 cm; Fairbanks, Alaska; 1985	~200 – 450	~163 – 366

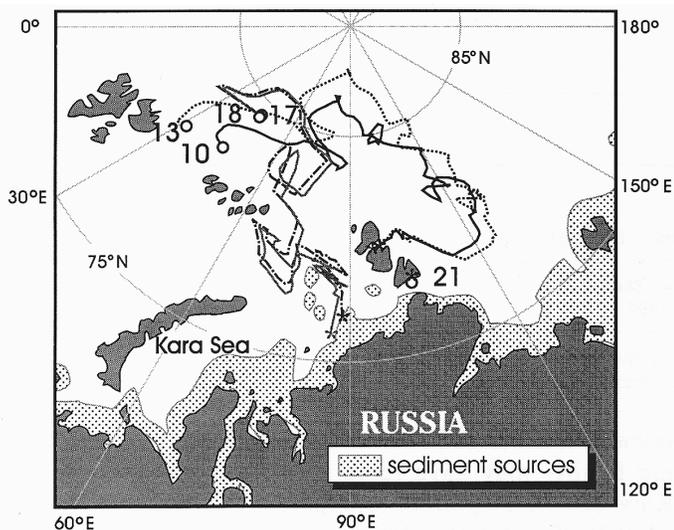


FIG. 3. Backward trajectories of IRD samples with anomalous $^{240}\text{Pu}/^{239}\text{Pu}$ ratios. The open circle and number indicate the IRD sampling location. The * at the other end of the back trajectory trace is the intercept with the 30 m isobath.

derived Pu transported to Kara Sea shelf sediments by ocean currents may yield IRD with Pu-ratio signatures that can vary greatly with time. As sediment entrained in sea ice does not form in the temperate latitudes of the nuclear facilities at Sellafield or La Hague, those sites cannot be a

direct source for the contaminant-bearing sediment observed in our sampled Arctic Ocean ice floes.

While low-value deviations from mean global fallout $^{240}\text{Pu}/^{239}\text{Pu}$ ratios suggest reprocessing sources (from weapons-grade plutonium production or low burn-up commercial fuel), these results must be interpreted with caution. They may also reflect fallout signals associated with low-yield nuclear weapons tests, such as those that occurred at the Nevada Test Site (Hicks and Barr, 1984) and on Novaya Zemlya (Forman et al., 1996). Even larger deviations from the 0.185 ratio than those seen in the present study have been observed in seafloor cores from areas far from reprocessing facilities, i.e., as low as 0.069 in the Gulf of Mexico (Scott et al., 1983) and as low as 0.11 in the central North Atlantic (Noshkin et al., 1974). These low ratios may reflect mixed sources and diagenetic processes after deposition (Scott et al., 1983). Thus, there are several possible sources of low-ratio Pu to the Kara Sea: the fuel reprocessing plants on the Ob and Yenisey Rivers, the reprocessing facility at Sellafield, and close-in fallout from Novaya Zemlya.

^{137}Cs

Among the lowest ^{137}Cs concentrations measured were in the IRD sampled in the Beaufort Sea shelf region during

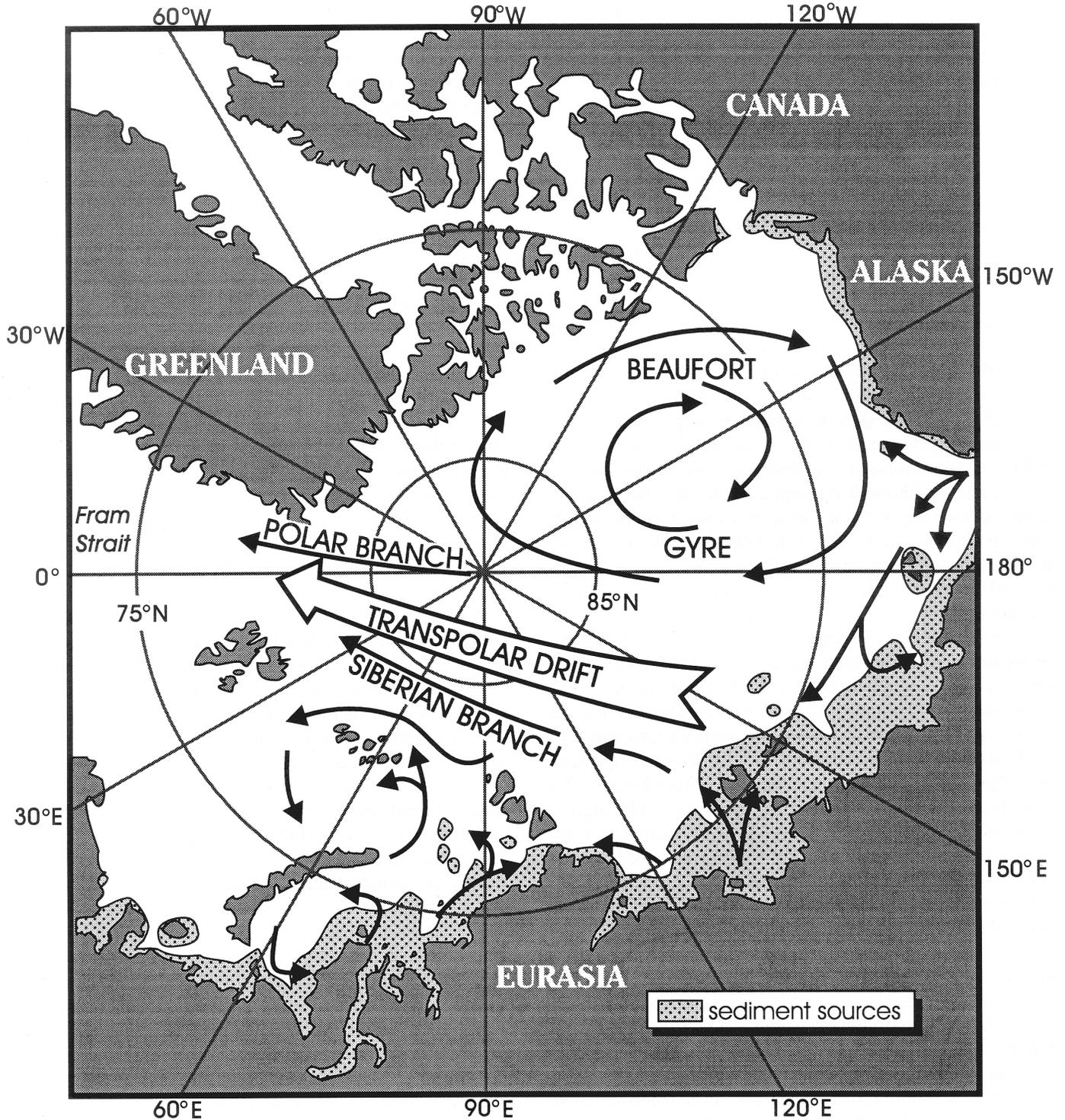


FIG. 4. Ice drift in the Arctic ocean, generalized after Gordienko and Laktionov (1969).

the *Polar Star* 1989 cruise and the Alaska 1995 expedition. The granular nature of the dirty ice sampled from the *Polar Star* in the summer of 1989 indicated that this was first-year ice that could not have originated from far away (Reimnitz et al., 1993a). Delineating possible sediment entrainment areas from satellite images and ice charts of the previous winter, and speculating on possible drift during and shortly after entrainment, Reimnitz et al. (1993a)

concluded that the sediment originated in the western Beaufort or the Chukchi Sea during late winter, 1989. The two samples from 1995 are from the zone of newly formed grounded pressure ridges near the 20 m isobath, and most likely were entrained nearby during the previous fall (Peter Barnes, USGS, pers. comm. 1995). Extensive surface sediment sampling of the northern Alaska shelves (Bering, Beaufort, and Chukchi Seas) by Cooper et al. (in

press) showed ^{137}Cs concentrations generally less than $10 \text{ Bq}\cdot\text{kg}^{-1}$. Thus, the North American shelf would not appear to be a likely source area for IRD with elevated ^{137}Cs concentrations.

IRD Transport

The generalized Arctic Ocean ice circulation pattern is shown in Figure 4. Most of the ice with sediment entrained on the shallow (< 30 m) Eurasian shelves (dotted area of Fig. 1) is transported out of the basin through the Fram Strait by the Siberian Branch of the Transpolar Drift. Figure 4 shows that ice from the Kara Sea, Laptev Sea, and East Siberian Sea can be transported to the vicinity of Svalbard and Franz Josef Land, where most of the anomalous samples were collected. Indeed the backward trajectories of all 16 of the samples from the *Polar Star* 1991 and *Polarstern* 1993 cruises (Table 1) show origins in the Kara Sea or near Severnaya Zemlya. (The backward trajectories of all the cruise samples are available at the following web site: <http://iabp.apl.washington.edu:80/ERK/>.) The likelihood that samples collected in the Beaufort Gyre had their origin on the Eurasian shelves is more remote. The samples from the *Polar Star* 1989 and 1993 cruises all show origins on the Beaufort shelf or, in the case of sample 9 (PS93-241-1-4), at least 14 years of circulation within the Canada Basin.

Both sample 9 (PS93-241-1-4) from the *Polar Star* 1993 cruise and a nearby one from the same cruise (PS93-236-1-2), with ^{137}Cs concentrations of 67 and 59 $\text{Bq}\cdot\text{kg}^{-1}$ respectively, are at the high end of the range seen in this study (less than 0.2 to 78 $\text{Bq}\cdot\text{kg}^{-1}$) (Table 2). These two samples also had high total Pu concentrations (Table 3). The AOS-94 sampling of IRD (Meese et al., 1997; Cooper et al., in press) showed ^{137}Cs concentrations ranging from 5 to 73 $\text{Bq}\cdot\text{kg}^{-1}$. The highest ^{137}Cs concentration seen in the AOS-94 IRD samples came from a floe in this region ($75^{\circ}57'\text{N}$, $171^{\circ}57'\text{W}$). The temporal (1993–94) and spatial cluster of three high- ^{137}Cs IRD samples off the northern coast of Alaska is of note, but neither in the two *Polar Star* 1993 samples studied here, nor in the AOS-94 sample (Meese et al., 1997) do back trajectories show positions outside of the Beaufort shelf/Canada Basin. The $^{240}\text{Pu}/^{239}\text{Pu}$ ratios of these three samples (0.182 and 0.181 for the two *Polar Star* samples and about 0.172 for the AOS-94 sample) were all close to that of mean global fallout (Cooper et al., in press; Table 3).

Ice-motion simulations by Pfirman et al. (1997a) show that whereas ice from the East Siberian Sea can potentially cross the Transpolar Drift stream, get caught in the Beaufort Gyre, and be transported to the northern Canadian Arctic Archipelago and the Alaskan coast, this route is unlikely in most years for ice from the Kara and Laptev Seas, most of which is exported through Fram Strait and the Barents Sea. Transport of significant quantities of ice and IRD from the Kara Sea, through the Vilkitsky and Sannikov Straits to the East Siberian Sea and from there to the Beaufort Gyre also seems unlikely, as the straits and coastal pathways are blocked by fast ice for much of the year (S. Pfirman, Barnard College,

Columbia University, pers. comm. 1997). However, recent studies by Pfirman et al. (1997b) demonstrate interannual variations in ice pathways, and show that in some years, Kara Sea ice is advected quite far to the east, to the region north of the East Siberian Sea. From this position, it may perhaps cross the Transpolar Drift stream and get caught in the Beaufort Gyre.

Recent analysis of trajectories of sea ice floes and the mineralogical composition of their entrained sediments has shown that the ice with the highest sediment loads entering the Arctic Ocean originates in the region to the north and east of the New Siberian Islands and the Central Kara Plateau (Pfirman et al., 1997a). Natural driftwood logs found in pack ice and on Arctic beaches may eventually reveal their place of origin. So far we can say with certainty only that most of the driftwood found on beaches in the Canadian Arctic Archipelago originated in Siberia, not in North America (Dyke et al., 1997). This conclusion is strongly supported by observations of locally abundant driftwood in the Siberian Branch of the Transpolar Drift, and the lack of such observations in the Beaufort Gyre.

CONCLUSIONS

An assessment of the risk associated with the transport of radionuclides in ice-rafted debris is beyond the scope of this paper. However, we note that the highest concentrations of ^{137}Cs found (Table 2) were lower than those found in some surficial soils in pristine environments (Table 6). On the other hand, the potential for ice transport of radionuclides associated with sediment does exist, and such material would be in a surface environment accessible to humans and other animals and plants in the Arctic Basin. The possibility that such contaminants move by IRD from the Siberian shelf to the mid-Arctic basin has been demonstrated in this study, although we cannot identify sources conclusively. IRD might be a significant dispersal mechanism in the event of a catastrophic release of radioactivity, for example, from a dam failure at a radioactive waste disposal reservoir such as those known to exist in the Ob River watershed. The predominant flux of contaminants associated with IRD from Siberian shelf source areas would be carried by the Transpolar Drift out of the Arctic Ocean, through Fram Strait, and into the North Atlantic (Fig. 4), where melting would release the contaminants from the ice (Weeks, 1994; Pfirman et al., 1995). Additional studies would be needed to assess the flux of such contaminants under various radionuclide release scenarios, and their environmental impact.

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