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Distributions of nuclear fuel-reprocessing tracers in the Arctic Ocean: Indications of Russian river influence

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ABSTRACT

Radionuclide sampling in 1986 and 1993 in the Canada Basin, and in 1993 in the Amundsen Basin and on the adjacent Laptev shelf, provides new insights into the origin, timing, pathways, and mechanisms for dispersal of non-fallout radioactive tracers in the Arctic Ocean. First, samples from the Beaufort Sea shelf, slope, and adjacent basin show a four-fold increase in ^{129}I concentrations from 1986 to 1993. Second, anthropogenic non-fallout radionuclide concentrations in the Beaufort Sea increase with proximity to slope boundary currents. Third, there is evidence for riverine contributions of anthropogenic radionuclides to surface waters of the Amundsen Basin and the Laptev continental shelf. This evidence includes high surface water burdens of ^{237}Np and ^{129}I, with the maximum in anthropogenic ^{129}I found in the least saline and most ^{18}O-depleted waters, consistent with an origin in high-latitude runoff. Additionally, the ^{237}Np/^{129}I atom ratios in the Laptev Sea and Amundsen Basin in 1993 were significantly lower than observed elsewhere in the Arctic Ocean and can be reasonably explained by ^{129}I added during transit of the Russian shelves. The ^{240}Pu/^{239}Pu ratios in the water column were mostly near 0.18, consistent both with stratospheric bomb fallout and with the discharged-weighted mean Sellafield ratio during 1966–1985. In the least saline water samples collected at the most shallow Laptev shelf station, however, the Pu ratios were lower, consistent with a non-European nuclear fuel reprocessing source. There are clear secondary maxima in ^{237}Np and ^{129}I

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near 1000 m in the Amundsen Basin, likely associated with the Barents Sea branch of Atlantic water. Finally, the ^{129}I/salinity and ^{129}I/δ^{18}O relationships in the Amundsen and Canada Basins at mid-depths are indistinguishable, suggesting effective horizontal dispersion.

1. Introduction

The transport of nuclear fuel-reprocessing byproducts, including ^{134}Cs, ^{137}Cs and ^{90}Sr, from western European sources in France (La Hague) and the U.K. (Sellafield) into Arctic waters was documented in the 1970s and early 1980s [reviewed by Livingston (1988) and Kershaw and Baxter (1995)]. Since the time of that work, the potential for contributions to the Arctic Ocean of radionuclides from Russian nuclear fuel cycle and nuclear reprocessing sources on the Ob and Yenisey rivers has also been examined [e.g. Aarkrog (1994); Vakulovsky *et al.* (1995); Baskaran *et al.* (1995); Baskaran *et al.* (1996); Beasley *et al.* (1997)]. These studies disagreed as to whether non-fallout anthropogenic radionuclides can even be detected in the mouths of the Ob and the Yenisey, illustrating the difficulty in unambiguously identifying contributions of anthropogenic radionuclides in the Arctic Ocean from Russian sources, excluding stratospheric bomb fallout. In addition to river-borne discharges, other potential sources include dumped waste in the Kara Sea, and leakage and tropospheric fallout from nuclear weapons testing on Novaya Zemlya. The large flux of radiostrontium and radiocesium entrained in northward flowing Atlantic water, including Sellafield, La Hague and Chernobyl contributions, together with the background of stratospheric bomb fallout, have all contributed to the difficulty in following the fate of non-fallout Russian sources. In recent years this has stimulated work on other Arctic tracers including ^{129}I [e.g. Cooper *et al.* (1998a)], ^{237}Np [e.g. Beasley *et al.,* (1998a)], and plutonium isotope ratios [e.g. Huh *et al.* (1997)]. In principle, absolute abundances and ratios of these and other radioisotopes can be used with discharge data from Sellafield and La Hague to constrain the magnitude of Russian sources (Beasley *et al.,* 1998a). From an oceanographic standpoint, if ratio or abundance characteristics unique to Russian sources can be identified, these tracers can for example, track Siberian river waters and distinguish western European source waters, thereby illuminating circulation and mixing within the Arctic Ocean.

The present work explores both of these interests: identifying non-fallout radionuclides of Russian origin in Arctic Ocean waters and better understanding circulation and time scales within the Arctic Ocean. We present here the results of water sampling undertaken in 1986 and 1993 in the Canada Basin of the Arctic Ocean, and in 1993 in the eastern Nansen and Amundsen basins and on the continental shelf of the Laptev Sea (Fig. 1). We also use data from the 1994 Arctic Ocean Section (Beasley *et al.,* 1998a), which crossed the Arctic Ocean.

We note that the Canada Basin is most remote from the Sellafield and La Hague nuclear fuel reprocessing sources, and the Nansen and Amundsen Basins are by contrast, much more immediately impacted. Even within the Canada Basin, however, sampling undertaken in 1985 (Livingston, 1988) demonstrated the presence of anthropogenic radionuclides of non-fallout origin, based upon ^{137}Cs/^{90}Sr ratios in excess of 1.5 at depths as great
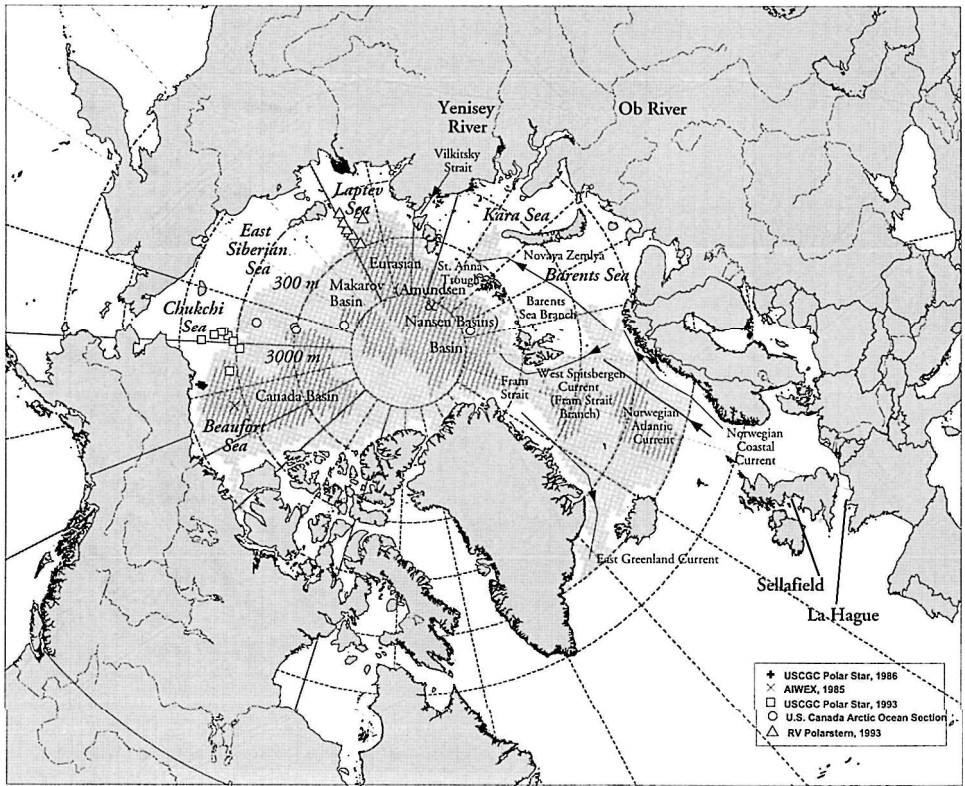
Figure 1. Sampling locations and geographic features discussed in text. Samples collected in 1985 (X) at the AIWEX ice island are from Livingston (1988); samples collected in 1994 during the U.S. Canada Arctic Ocean Section (circles) are presented in Beasley *et al.* (1998a). Other samples used as part of this study were collected in 1986 from the USCGC *Polar Star* (crosses), and in 1993 from both the USCGC *Polar Star* (squares) and the R/V *Polarstern* (triangles). Bathymetric shading is provided for depths greater than 300 m (lighter) and 3000 m (darker) for the central Arctic basin only, and is based upon data from the Environmental Working Group (1997).

as 1200 m. Livingston (1988), in his review of all Arctic radionuclide data then available, pointed out that most of the anthropogenic non-fallout radionuclides detected in the 1970s and 1980s were not concentrated in the warmest part of the Atlantic layer, suggesting a complex circulation pattern and mixing history. It is these issues that we examine here: the origin, timing, pathways, and mechanisms for dispersal of non-fallout radionuclides in the Arctic Ocean.

2. Methods

Water samples for this study were collected on four cruises (Fig. 1), in 1986 in the Beaufort Sea from the USCGC *Polar Star* (Aagaard *et al.,* 1986), in 1993 during both the ARK-IX/4 cruise of the R/V *Polarstern* (Amundsen and Nansen basins and Laptev shelf;

Fütterer, 1994), and a cruise of the USCGC *Polar Star* (Canada Basin and shelf), and in 1994 on the U.S./Canada Arctic Ocean Section (Wheeler, 1997; Beasley *et al.,* 1998a). Samples were directly obtained from bottle casts and were not filtered. Except in the case of the samples collected in 1986, which were held in storage, all other samples were poisoned with sodium azide to suppress any biological alteration prior to processing. Following arrival at the Environmental Measurements Laboratory, $\sim$4-L water samples were split for transuranic and ^{129}I measurements. Preparation for measurements of plutonium isotopes and ^{237}Np was undertaken, including yield and blank determinations, following the methods of Beasley *et al.* (1998a,b). Isotope dilution thermal ionization mass spectrometry was accomplished using an instrument at Pacific Northwest National Laboratory (Stoffels and Lagergren, 1969; Lagergren and Stoffels, 1970).

One-L samples were used in preparation of the ^{129}I targets following the detailed description available in Mann and Beasley (1994); potassium iodide with pre-bomb ^{129}I background concentrations was used as a carrier. This added iodide was equilibrated at 70°C with seawater iodide following addition of sodium hypochlorite to oxidize all iodide to iodate (IO_3^-/IO_4^-). These higher valence states were then reduced to I^- by addition of $NaHSO_3$. Following extraction into and from high purity CCl_4, iodine was precipitated as silver iodide from a dilute acid solution using an ultra-pure silver nitrate solution. After drying at 100°C, silver iodide targets were transferred to the accelerator mass spectrometry facility at the Isotrace Laboratory, University of Toronto, where measurements were undertaken, including background and blank measurements (Kilius *et al.,* 1990; 1992).

The ^{137}Cs activity was measured exclusively on the 1986 samples, which had been archived at Woods Hole Oceanographic Institution (H. Livingston, personal communication). Aliquots for ^{237}Np and ^{129}I determinations were first removed at the Environmental Measurements Laboratory and processed as described above. Following receipt at Oak Ridge National Laboratory, a known amount of an atomic absorption cesium standard was added as a yield tracer and total stable cesium was measured in processed sample fractions by inductively coupled plasma mass spectrometry (ICP-MS). Cesium was concentrated in these large volume (40 to 47-L) samples for gamma counting by acidification to pH 2, and addition of ammonium molybdophosphate. Cesium was then precipitated by addition of sodium hydroxide. Two 44 and 49-L aliquots of a water sample used at Woods Hole as an internal ^{137}C standard were also analyzed to assure data quality. Precipitates were assayed for ^{137}Cs using low-background, high-resolution, lithium-drifted germanium or high purity germanium (HPGe) detectors (Cooper *et al.,* 1998b).

Salinity data were obtained from CTD profiles compiled following each cruise. The $\delta^{18}O$ values in some samples collected in 1993 were also analyzed to establish their conservative tracer identity by standard equilibration methods (Epstein and Mayeda, 1953). Mass spectrometric determinations of $\delta^{18}O$ were made at Oak Ridge National Laboratory for samples collected on the 1993 *Polar Star* cruise. Oxygen isotope data from the 1993 *Polarstern* cruise are derived from Frank (1996). Precision of this measurement was better than $\pm0.1\%_o$ for both laboratories.

Table 1. Radiochemical and physical oceanographic data for samples collected during this study. Errors tabulated are ±1σ. In addition to data presented here, other data sources include Aagaard et al. (1986), Frank (1996), and cruise reports. Data (not tabulated) from the 1994 U.S. Canada Arctic Ocean Section are available in Beasley et al. (1998a).

Station number, coordinates	Depth (m)	Salinity	Temperature °C	$\delta^{18}O$ V-SMOW	^{129}I (10^9 atoms kg⁻¹)	^{237}Np (10^8 atoms kg⁻¹)	^{239}Pu (10^7 atoms kg⁻¹)	^{240}Pu (10^6 atoms kg⁻¹)	^{137}Cs (Bq kg⁻¹)
					USCGC *Polar Star*, August, 1986				
A8, 71.77N, 152.83W	20	31.335	—	—	0.0292 ± 0.0020	—	—	—	4.22 ± 0.25
	206	34.601	−0.23	—	0.08158 ± 0.005	—	—	—	4.03 ± 0.25
A6, 71.597N, 153.17W	20	30.836	—	—	0.0224 ± 0.00010	—	—	—	3.51 ± 0.28
A9, 71.88N, 152.47W	20	29.839	—	—	0.0328 ± 0.0040	—	—	—	4.68 ± 0.28
	30	—	—	—	—	—	—	—	1.32 ± 0.14
	111	33.092	−1.30	—	0.0265 ± 0.0010	—	—	—	5.46 ± 0.49
	201	34.594	−0.29	—	0.0740 ± 0.0031	—	—	—	3.57 ± 0.23
	301	34.795	0.36	—	0.0925 ± 0.0060	—	—	—	3.51 ± 0.18
					0.1030 ± 0.0060 (independent replicate)				
	502	34.874	0.43	—	0.0502 ± 0.0024	—	—	—	4.07 ± 0.28
					0.0470 ± 0.0032 (independent replicate)				
	706	34.893	0.25	—	0.0502 ± 0.0050	—	—	—	3.71 ± 0.26
	1003	34.902	−0.06	—	0.0327 ± 0.0031	—	—	—	4.05 ± 0.28
	1545	34.924	−0.34	—	0.0200 ± 0.0018	—	—	—	—
A10, 71.98N, 152.67W	21	29.500	—	—	0.0368 ± 0.0020	0.1987 ± 0.0040	0.0350 ± 0.0077	0.0532 ± 0.0077	6.95 ± 0.34
	131	33.372	−1.46	—	0.0194 ± 0.0013	0.2612 ± 0.0054	0.3811 ± 0.0074	0.724 ± 0.027	5.72 ± 0.44
	200	34.557	−0.34	—	0.0885 ± 0.0040	0.2063 ± 0.0044	0.3311 ± 0.0074	0.608 ± 0.027	4.60 ± 0.30
	329	34.817	0.40	—	0.0930 ± 0.0042	0.2426 ± 0.0061	1.815 ± 0.032	3.27 ± 0.81	6.42 ± 0.34
	503	34.866	0.43	—	0.0581 ± 0.0041	0.1743 ± 0.0040	0.3641 ± 0.0084	0.644 ± 0.029	4.28 ± 0.29
	700	34.893	0.22	—	0.0424 ± 0.0027	0.1656 ± 0.0042	0.883 ± 0.012	1.65 ± 0.04	3.52 ± 0.25

Table 1. (Continued)

Station number, coordinates	Depth (m)	Salinity	Temperature °C	δ¹⁸O V-SMOW	^{129}I (10⁹ atoms kg⁻¹)	^{237}Np (10⁸ atoms kg⁻¹)	^{239}Pu (10⁷ atoms kg⁻¹)	^{240}Pu (10⁶ atoms kg⁻¹)	^{137}Cs (Bq kg⁻¹)
31, 76.53N, 133.32E	1000	34.900	−0.02	—	0.0352 ± 0.0023	0.1392 ± 0.0034	0.886 ± 0.019	1.59 ± 0.06	3.03 ± 0.22
	1250	34.911	−0.20	—	0.0211 ± 0.0018	—	—	—	1.61 ± 0.12
	1573	34.927	−0.34	—	0.0202 ± 0.0016	0.0828 ± 0.0018	0.554 ± 0.008	1.03 ± 0.03	1.58 ± 0.17
RV Polarstern ARK-IX/4, August–September, 1993									
31, 76.53N, 133.32E	10	28.604	0.0262	—	1.13 ± 0.03	1.082 ± 0.041	0.1646 ± 0.0078	0.265 ± 0.034	—
	19	30.336	0.0271	—	1.20 ± 0.03	1.114 ± 0.039	0.212 ± 0.011	0.307 ± 0.035	—
	25	31.132	−0.8603	—	1.14 ± 0.03	1.189 ± 0.055	0.373 ± 0.014	0.653 ± 0.052	—
	33	32.038	−0.8465	−2.54	1.21 ± 0.03	1.176 ± 0.0411	0.441 ± 0.014	0.785 ± 0.051	—
32, 78.71N, 132.35E	12	32.769	−1.6876	−1.11	1.15 ± 0.03	0.940 ± 0.065	0.486 ± 0.073	0.92 ± 0.16	—
	98	34.188	−1.5913	−0.16	0.84 ± 0.02	0.861 ± 0.045	0.890 ± 0.049	1.53 ± 0.14	—
	200	34.823	1.557	0.20	0.42 ± 0.01	0.483 ± 0.022	1.223 ± 0.027	2.164 ± 0.075	—
	300	—	—	—	0.42 ± 0.01	0.469 ± 0.030	1.334 ± 0.034	2.462 ± 0.086	—
	500	34.871	0.7452	0.25	0.45 ± 0.01	0.480 ± 0.023	1.172 ± 0.026	2.049 ± 0.075	—
	800	34.861	−0.0396	—	0.63 ± 0.02	0.629 ± 0.027	1.245 ± 0.022	2.294 ± 0.057	—
	1002	34.881	−0.1609	—	0.59 ± 0.02	0.573 ± 0.042	1.206 ± 0.029	2.272 ± 0.082	—
	1509	34.919	−0.5269	—	0.068 ± 0.003	0.121 ± .010	1.284 ± 0.026	2.341 ± 0.073	—
33, 79.65N, 130.60E	3298	34.942	−0.7434	0.24	0.0043 ± 0.0007	—	—	—	—
35, 78.39N, 133.11E	14	32.486	−1.6419	−1.10	1.19 ± 0.03	1.223 ± 0.025	—	—	—
	75	34.166	−1.6312	—	0.91 ± 0.03	0.994 ± 0.022	—	—	—
	301	34.891	1.8488	—	0.35 ± 0.04	0.554 ± 0.015	—	—	—
	801	34.867	0.1419	0.23	0.54 ± 0.05	0.642 ± 0.015	—	—	—
40, 78.07N, 133.56E	75	34.072	−1.4776	−0.35	0.98 ± 0.03	—	—	—	—
	210	34.850	1.4443	0.27	0.42 ± 0.01	—	—	—	—
43, 77.40N, 133.60E	14	31.119	−1.4994	−2.06	1.26 ± 0.03	—	—	—	—
	48	33.093	−1.6089	—	1.27 ± 0.03	—	—	—	—
47, 77.18N, 126.25E	299	34.859	1.4232	—	0.37 ± 0.01	—	—	—	—
50, 77.71N, 125.84E	600	34.859	0.5737	—	0.56 ± 0.01	—	—	—	—
	1983	34.929	−0.8055	—	0.011 ± 0.006	—	—	—	—

Table 1. (Continued)

Station number, coordinates	Depth (m)	Salinity	Temperature °C	δ¹⁸O V-SMOW	^{129}I (10^9 atoms kg^{-1})	^{237}Np (10^8 atoms kg^{-1})	^{239}Pu (10^7 atoms kg^{-1})	^{240}Pu (10^6 atoms kg^{-1})	^{137}Cs (Bq kg^{-1})
				USCGC Polar Star, August-September, 1993					
PS026E1, 74.40N, 155.72W	2000	34.900	−0.414	0.33	0.022 ± 0.003	—	—	—	—
	3800	34.954	−0.257	0.20	0.014 ± 0.002	—	—	—	—
PS035D1, 75.40N, 163.01W	50	32.301	−1.406	−0.89	0.044 ± 0.007	—	—	—	—
	125	33.062	−1.477	−0.94	0.063 ± 0.005	—	—	—	—
PS039C5, 74.81N, 165.29W	2048	34.930	−0.305	0.38	0.019 ± 0.004	—	—	—	—
	125	32.918	−1.553	−1.44	0.065 ± 0.007	—	—	—	—
	150	33.838	−1.132	−0.72	0.126 ± 0.008	—	—	—	—
PS042C3, 74.300N, 167.10W	423	34.808	0.508	0.29	0.437 ± 0.010	—	—	—	—
	50	32.141	−1.574	−1.40	0.023 ± 0.004	—	—	—	—
	137	33.177	−1.199	−1.01	0.060 ± 0.006	—	—	—	—
PS044C1, 73.90N, 168.49W	300	34.724	0.204	0.20	0.469 ± 0.025	—	—	—	—
	10			−1.80	0.047 ± 0.005	—	—	—	—
	115			−0.76	0.122 ± 0.008	—	—	—	—
PS045B8, 73.70N, 168.22W	175			0.22	0.437 ± 0.010	—	—	—	—
	20	30.114	−1.355	−1.59	0.037 ± 0.004	—	—	—	—
	75	32.404	−1.656	−1.38	0.040 ± 0.005	—	—	—	—
PS049B4, 73.10N, 166.10W	110	33.451	−1.076	−0.61	0.147 ± 0.008	—	—	—	—
	55	32.686	−1.490	−1.21	0.045 ± 0.007	—	—	—	—
PS052B1, 72.00N, 165.35W	34	31.382	3.514	−1.19	0.021 ± 0.003	—	—	—	—

3. Results

a. Beaufort Sea and Canada Basin The maximum ^{129}I and ^{237}Np burdens in samples collected in 1986 and 1993 in the Beaufort Sea, from shelf, slope, and deep Canada Basin waters, showed an approximately four-fold increase over the seven year interval (Table 1; Fig. 2). Comparison with ^{137}Cs data collected in 1985 at the AIWEX site (Livingston, 1988), about 500 km north of Prudhoe Bay, Alaska, shows no increase in ^{137}Cs activities in Canada Basin surface waters between 1985 and 1986 (Fig. 2). Therefore, no significant atmospheric deposition of ^{137}Cs reached Canada Basin surface waters in the first five months after the Chernobyl nuclear accident of 28 April 1986. In deeper waters, however, there are significant differences in the ^{137}Cs activity distributions in our 1986 samples collected near the continental margin and the offshore AIWEX samples. The AIWEX profile shows a rapid decline in ^{137}Cs activity below the polar mixed layer and the halocline, and in fact shows lower activity everywhere above about 1000 m than the profile collected the following year (Fig. 2). Plots of ^{129}I/^{137}Cs (1986) and ^{137}Cs/^{90}Sr (1985) indicate that the nuclear fuel-reprocessing component of anthropogenic radionuclides also shows a disjunct vertical and horizontal distribution (Fig. 3). Since ^{137}Cs/^{90}Sr ratios in excess of 1.5 reflect non-fallout sources (Bowen *et al.,* 1974), and higher ^{129}I/^{137}Cs ratios also reflect non-fallout sources (Edmonds *et al.,* 1998), conservative radionuclides not originating from stratospheric bomb fallout in our 1986 samples were most prevalent at 200–300 m (Fig. 3). At this depth, the AIWEX samples, however, appear to mostly represent stratospheric bomb fallout [Fig. 3; also see Livingston (1988)]. For example, the influence of nuclear fuel-reprocessing sources on ^{129}I/^{137}Cs ratios (1986 samples) declines on a relative basis between 300 and 500 m, while the influence of nuclear fuel-reprocessing sources on ^{137}Cs/^{90}Sr ratios (1985 samples) increases between 300 and 400 m (Fig. 3). We return to this feature in the discussion section.

b. Laptev Sea continental shelf and Amundsen Basin

Both deep Amundsen Basin water samples, and those collected on the Laptev continental shelf carried relatively high surface water burdens of ^{237}Np ($>1 \times 10^8$ atoms kg^{-1}) and ^{129}I ($>1 \times 10^9$ atoms kg^{-1}; Table 1, Fig. 2). Off the shelf, these profiles also exhibited a mid-depth secondary maximum in both radionuclides, with concentrations of ^{237}Np and ^{129}I rising below 500 m and reaching secondary maxima at approximately 800–1000 m (Fig. 2). When compared to the Canada and Makarov Basin data, ^{237}Np/^{129}I atom ratios in these Amundsen Basin profiles were in general lower than observed elsewhere (Fig. 4), including within multiple basins on the 1994 Arctic Ocean Section (Beasley *et al.,* 1998a). These lower ^{237}Np/^{129}I atom ratios in Amundsen Basin samples (Fig. 4) imply either a relative excess of ^{129}I or preferential scavenging of ^{237}Np. Based upon plots of ^{237}Np relative to ^{129}I (Fig. 5), we consider it unlikely that scavenging of ^{237}Np is more important. Briefly, in our ^{237}Np/^{129}I atom ratio plot, the regression line intercepts for the Canada and Makarov Basin samples approach zero, while most others do not (Fig. 5). The 95%
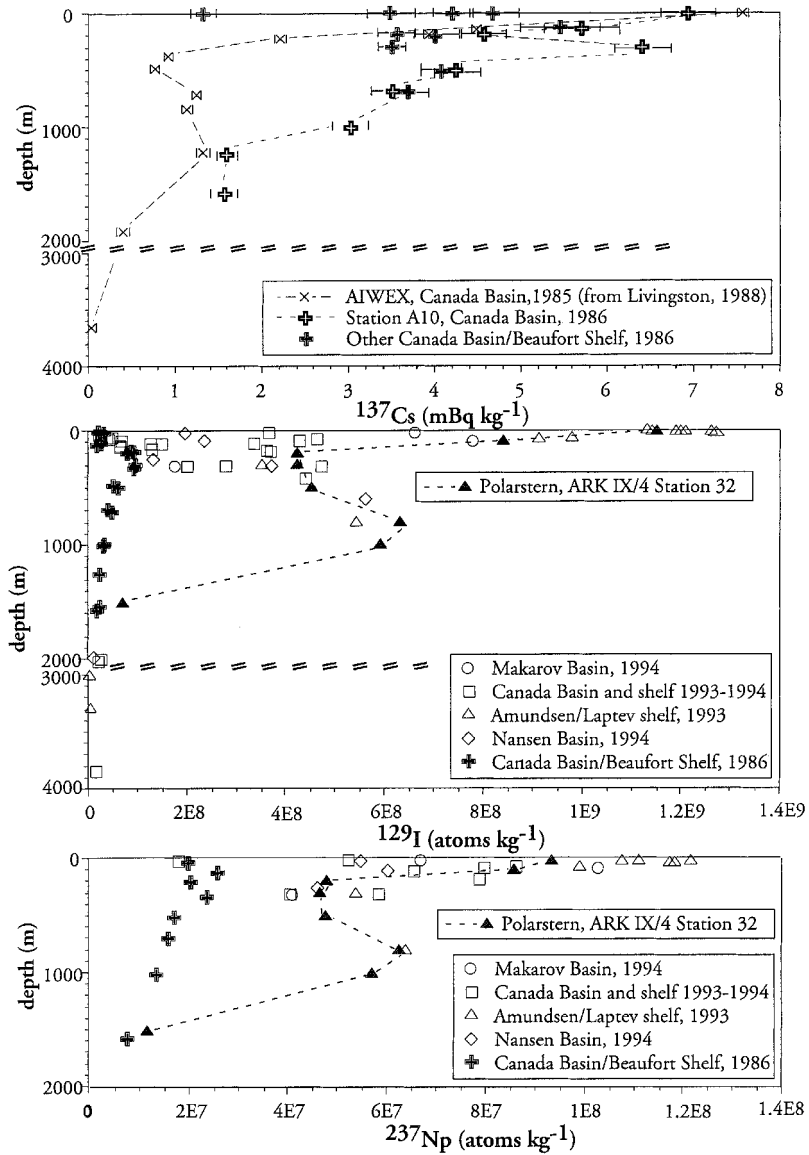
Figure 2. Depth relative to concentrations (atoms kg^{-1}) or activities (mBq kg^{-1}) for waters samples assayed during this study. ^{137}Cs activity is decay-corrected to date of collection. Error bars ($\pm 1\ \sigma$) are shown only where they are larger than symbols. Representative profiles are shown for three stations (AIWEX and Station A10, both in the Canada Basin, and *Polarstern* ARK IX/4 Station 32, in the Amundsen Basin).
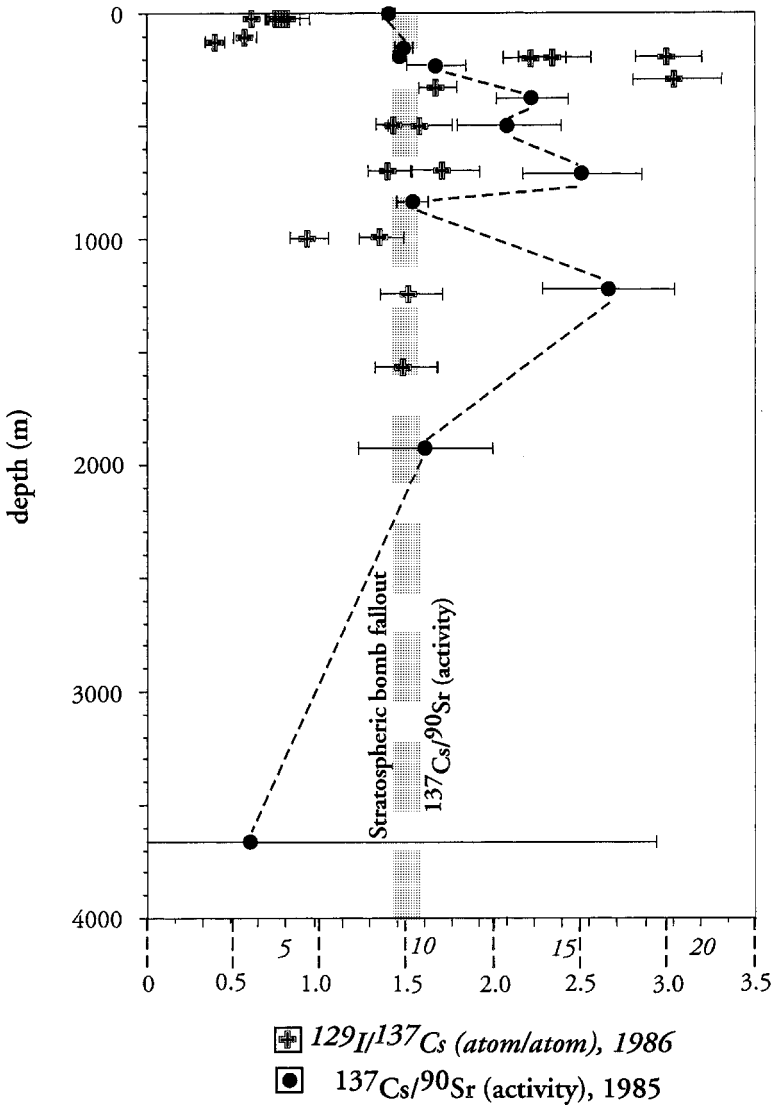
Figure 3. Ratios of $^{137}Cs/^{90}Sr$ and $^{129}I/^{137}Cs$ for samples collected in the Beaufort Sea, 1985–1986. The source of the $^{137}Cs/^{90}Sr$ data is Livingston (1988). Error bars ($\pm 1\ \sigma$) are shown only where they are larger than symbols. The dashed gray bar corresponds to the expected $^{137}Cs/^{90}Sr$ for stratospheric bomb fallout (Bowen *et al.*, 1974).

confidence intervals for the slopes of each regression line indicate that as ^{129}I concentrations reach zero, concentrations of ^{237}Np overlap or approach zero only in the case of the Canada (1986 and 1994) and Makarov Basin samples; in the Eurasian Basin samples, measurable concentrations of ^{237}Np would occur in the water column even if ^{129}I
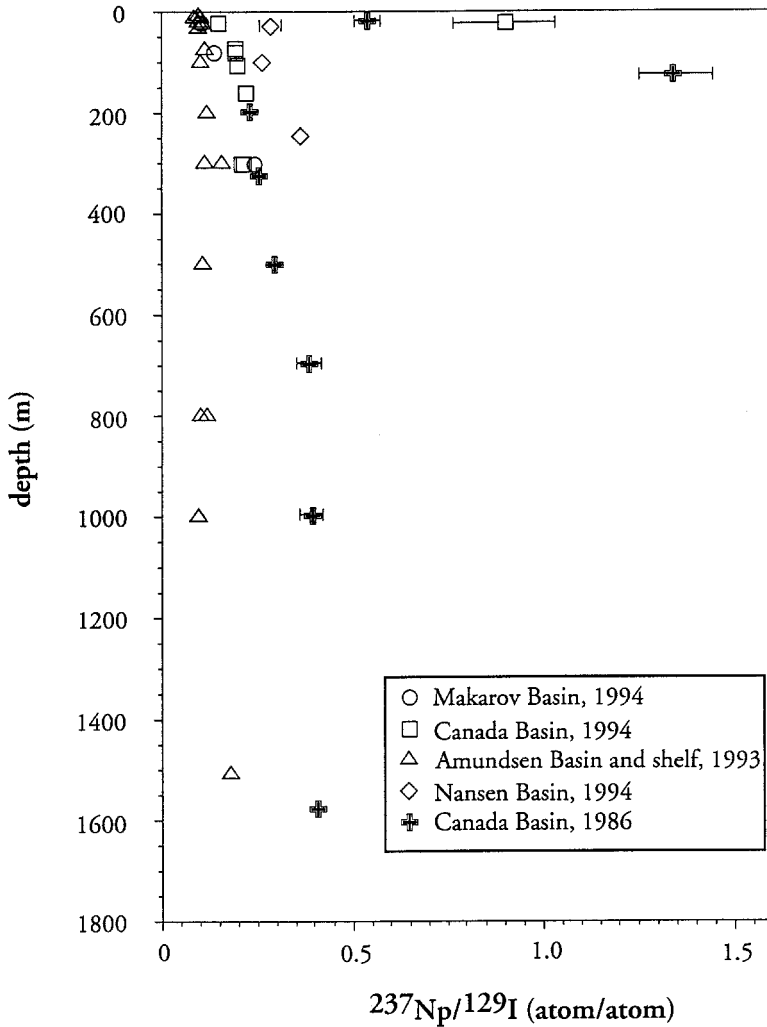
Figure 4. Ratios of ^{237}Np/^{129}I relative to depth for all currently available Arctic samples. Data from samples collected in 1994 are from Beasley *et al.* (1998a). Error bars ($\pm 1\ \sigma$) are shown only where they are larger than symbols.

concentrations were below analytical blank values. This suggests that an additional source of ^{237}Np in the samples collected within the Eurasian Basin is responsible. It also indicates that to the extent that scavenging of ^{237}Np occurs, it is not responsible for the low ^{237}Np/^{129}I atom ratios in the Amundsen Basin samples.

For the most part, the dissolved ^{240}Pu/^{239}Pu isotope ratios in these water samples were approximately 0.18 (Fig. 6), consistent with stratospheric bomb fallout (Krey *et al.*, 1976) and the discharge-weighted isotopic composition from Sellafield during 1966–1985
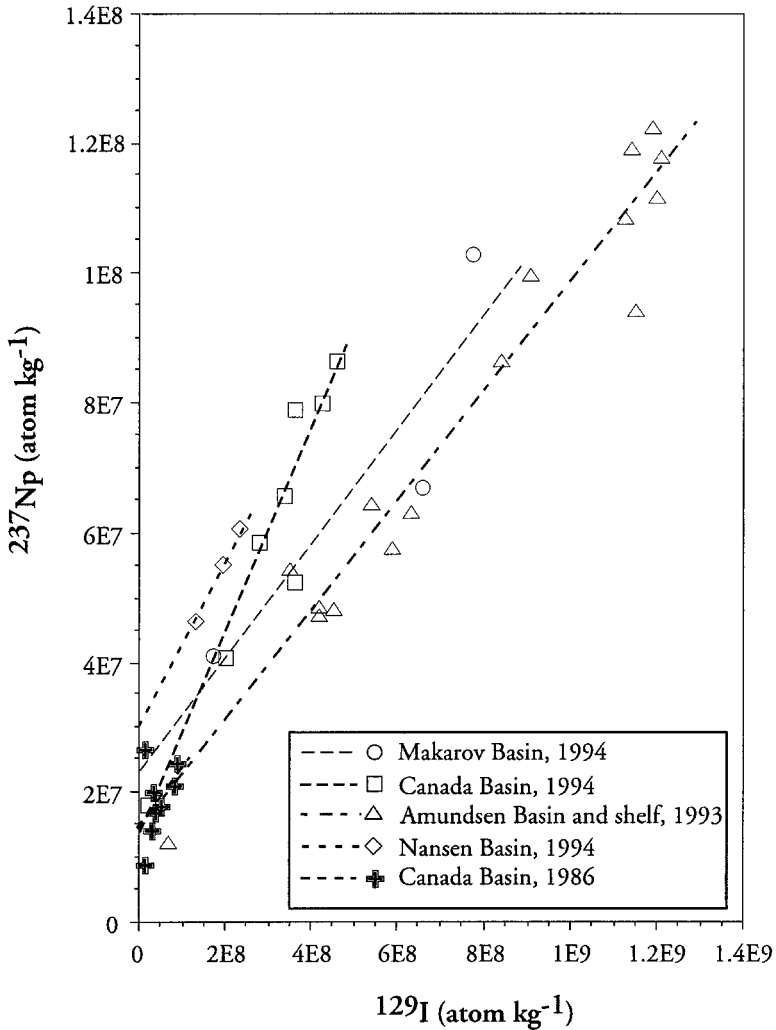
Figure 5. Concentrations of [129]I versus [237]Np for various Arctic basin and shelf samples. Data from samples collected in 1994 are from Beasley *et al.* (1998a). Errors ($\pm \sigma$) were universally smaller than symbols shown. Regression lines are the least-square fit line for each group of samples.

[Cooper *et al.* (1999), based upon Kershaw *et al.* (1990; 1995)]. The three exceptions were water samples from 10 m and 19 m at Station 31 (Table 1), the most shallow, and most inshore Laptev shelf station sampled (water depth 37 m) and in the Canada Basin sample at 21 m in 1986 at Station A10 (water depth 2100 m). These three water samples with low [240]Pu/[239]Pu ratios were also the least saline of all samples analyzed for plutonium isotope ratios (Fig. 6). As might be expected for surface waters, the total plutonium concentrations in these three water samples were also very low (Table 1), which constrains the certainty of
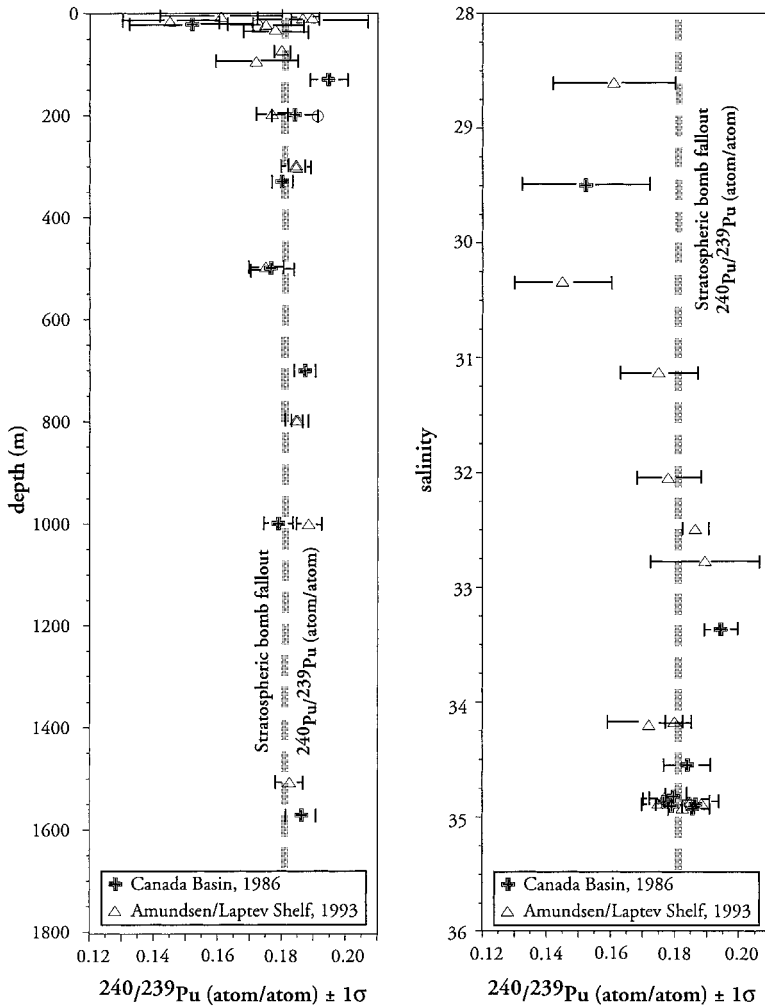
Figure 6. ^{240}Pu/^{239}Pu ratios versus water depth and salinity for samples collected on the Laptev shelf and adjacent basin waters, 1993, and for Canada Basin waters, 1986. Error bars correspond to $\pm 1\,\sigma$. The dashed gray bar corresponds to the expected ^{240}Pu/^{239}Pu ratio for stratospheric bomb fallout in the northern hemisphere (Krey *et al.,* 1976).

any provenance analysis. Nevertheless, high concentrations of anthropogenic ^{129}I were also associated with the least saline and most heavy isotope-depleted (^{18}O) waters sampled on the Laptev shelf (Fig. 7). The linear regression between salinity and δ^{18}O for the Laptev Sea samples indicates that the δ^{18}O composition of the freshwater end-member is approximately $-20\%o$ (Frank, 1996), consistent with high latitude runoff (Östlund and Hut, 1984), rather than sea ice melt. Using this δ^{18}O value for runoff and $+2.1\%o$ for melted sea ice, Frank (1996) estimated that the fraction of ice melt in surface water at Station 31 was
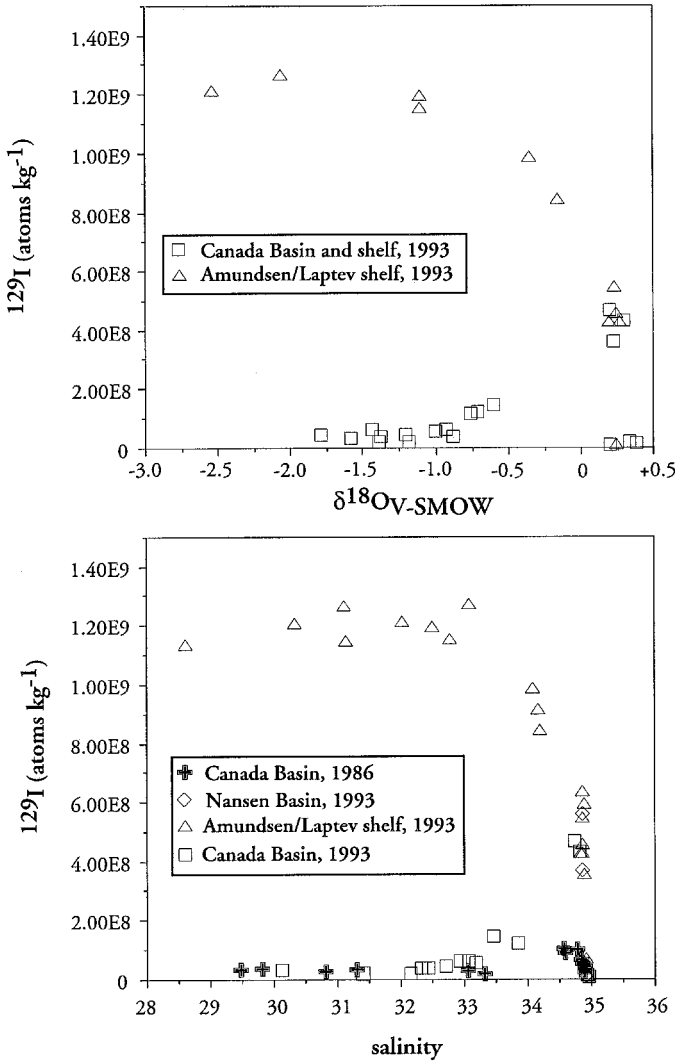
Figure 7. $\delta^{18}O$ values and salinity relative to concentrations of ^{129}I for various Arctic shelf and basin waters. Analytical errors were universally smaller than the symbols displayed. $\delta^{18}O$ values and salinity data for the Eurasian Basins and Laptev shelf samples collected in 1993 are modified from Frank (1996).

negligible, and that runoff made up 20% of surface waters. In deeper water, however, our data show that at an ^{129}I concentration of 4×10^8 atoms kg^{-1}, which is typically at a depth of $\sim$300 m (Table 1), the salinities and $\delta^{18}O$ values of Amundsen and Canada Basin samples are indistinguishable, suggesting effective horizontal dispersal near that depth (Fig. 7).

4. Discussion

a. Beaufort Sea and Canada Basin samples

The absence of an increase in surface water ^{137}Cs activities between 1985 (AIWEX) and 1986 (Fig. 2) suggests that there was no significant contribution of atmospheric ^{137}Cs activity derived from Chernobyl to surface waters of the Canada Basin in the first five months following that nuclear power plant accident. While this does not exclude the possibility for deposition of Chernobyl-derived contaminants onto ice that was later transported to the Canada Basin, independent evidence also indicates that direct Chernobyl-derived deposition in this portion of the Arctic was at most minor [e.g. Baskaran *et al.* (1991)]. These indications are in contrast to those for the Eurasian basins, where Chernobyl radiocesium was observed in Nansen Basin waters in 1987 at depths ranging from 300 m in the deep basin to 800 m on the Barents slope (Cochran *et al.,* 1995).

The well defined subsurface ^{129}I maximum at ~300 meters in 1986 (Fig. 2) coincides with the depth of the ^{129}I/^{137}Cs maximum (Fig. 3). This indicates that the nuclear fuel-reprocessing component we observe in much of the water column is concentrated in the upper Atlantic layer, below the halocline (salinities between 200–300 m were 34.6 to 34.8; Table 1). The relatively high concentrations of ^{137}Cs within this upper Atlantic layer (~4 mBq kg^{-1}) are well above activities (~1 mBq kg^{-1}) observed the previous year at the AIWEX sampling site (Fig. 2; see also Livingston, 1988). While we cannot exclude that the difference results from a rapidly moving plume of radiocesium, the far simpler and more likely explanation is that it is related to the closer proximity of the stations sampled in 1986 (Fig. 1) to the topographically steered boundary current (Aagaard, 1989) carrying a nuclear fuel-reprocessing burden into the Canada Basin. In 1986, there was a well defined subsurface peak in ^{129}I at 200–300 m, not far below the halocline, and activities of ^{137}Cs at this depth were well above those observed the previous year at AIWEX, 500 km offshore.

These tracer distributions, as influenced by boundary currents, and concentrated in Atlantic waters below the halocline, are consistent with recent measurements that have used other circulation tracers in the Arctic Ocean, including chlorofluorocarbons (e.g. Carmack *et al.,* 1997) and T/^{3}He dating (e.g. Schlosser *et al.,* 1994). In the context of recent observed change in Arctic temperature distributions (Carmack *et al.,* 1997), the changes we observed in radionuclide tracer distributions in the Canada Basin between 1986 and 1993–1994 help confirm a growing consensus that the upper Arctic Ocean is a dynamic system capable of rapidly propagating a variety of tracers, especially in the boundary current.

At deeper levels within the Canada Basin, there are few data available to differentiate transport pathways, but for waters with similar ^{129}I concentrations of ~4 $\times$ 10^8 atoms kg^{-1} observed in 1993–1994, the salinity and δ^{18}O content collected in the Nansen, Amundsen, and Canada basins were largely indistinguishable (Fig. 7), suggesting effective lateral dispersal of conservative radionuclides across much of the Arctic Ocean basins at mid-depth. The vertical distribution of radionuclides elsewhere in the water column differs significantly among the various basins (cf. below).

b. Laptev Sea continental shelf and Amundsen Basin

These water samples, collected near the continental slope and also over the continental shelf, carried relatively high surface water burdens of ^{237}Np ($>1 \times 10^8$ atoms kg^{-1}) and ^{129}I ($>1 \times 10^9$ atoms kg^{-1}), significantly higher than any samples collected on the 1994 Arctic Ocean Section (Carmack *et al.,* 1997; Beasley *et al.,* 1998a), although similar to concentrations observed in some surface samples collected in 1996 over the Lomonosov Ridge, at stations north of 85N (Josefsson, 1998; Buraglio *et al.,* 1999). The locations of those high concentrations, in conjunction with our own measurements, suggest probable export of the surface burden we observed in the Laptev Sea along the Lomonosov Ridge and thence out of the Arctic Ocean via Fram Strait. Note that the 1994 Section, in contrast to our 1986 and 1993 sampling, was centered on the deep Arctic basins and only undertook limited sampling near the continental slope and shelf, and then only in the Chukchi Sea.

Smith *et al.* (1998) reported 1993 Kara Sea surface water concentrations of ^{129}I of 1.14×10^9 atoms kg^{-1} at the north end of Novaya Zemlya, near the path of the Barents Sea branch of Atlantic water (BSBW), similar to our near-surface ^{129}I concentrations in the Laptev Sea. The salinity at the north end of Novaya Zemlya was much higher, however (32.86), suggesting a possible separate freshwater source of ^{129}I for our Laptev Sea samples. In the Laptev Sea, a mid-depth secondary maximum was also evident, with concentrations of ^{237}Np and ^{129}I increasing below 500 m and reaching secondary maxima at approximately 800 m (Fig. 2). Although the mobility of neptunium within seawater is incompletely understood [reviewed by Beasley *et al.* (1998a)], the high abundance of both radionuclides, and the high ratio of ^{129}I relative to ^{237}Np, are consistent with a nuclear fuel-reprocessing origin, rather than bomb fallout, Chernobyl, or other potential sources discussed by Beasley *et al.* (1998a).

In an earlier budget (Beasley *et al.,* 1998a) we evaluated the probable sources of neptunium to the Arctic Ocean, and took into account the likely advection of neptunium derived from direct bomb fallout upon both the Atlantic and Pacific oceans, and recent data on erosion of bomb fallout ^{237}Np from river catchments into the Arctic Ocean. We also considered discharge data from the La Hague and Sellafield nuclear fuel reprocessing facilities during 1966–1991, and the likely proportion of these discharges that would adhere to sediments close to the points of release, rather than be transported to the Arctic. We also estimated that contributions of ^{237}Np from Chernobyl were not significant relative to these other terms. We concluded that the observed concentrations of ^{237}Np in the upper Arctic Ocean (<300 m) could best be explained by an additional source, viz., reprocessing wastes from Russian riverine sources, which contributed an average of $\sim 1.5 \times 10^7$ atoms of ^{237}Np to each liter of seawater in the upper 50 m of the Arctic Ocean between 1984–1994. Based upon observations made in that study, we concluded that in the upper 300 m of the Arctic Ocean, bomb fallout ^{237}Np carried with river runoff and discharges from Russian reprocessing facilities are approximately equal in magnitude ($\sim 20\%$) but their combined total is less than the contribution ($>50\%$) from reprocessing facilities in western Europe, which are dominated by Sellafield.

Other information that is relevant to understanding the probable oceanographic origin of the radionuclides discussed here is available in Schauer *et al.* (1997) and Olsson and Anderson (1997), who sampled the same Laptev shelf and Amundsen Basin waters. According to the analyses of Schauer *et al.* (1997), these Laptev Shelf and Amundsen Basin waters sampled as part of Sections IV and V of ARK IX/4, were within the BSBW, which flows across the Barents and Kara continental shelves, and at the time of this sampling formed a 200 km-wide wedge, 200 m to 1300 m deep, which displaced the warmer Fram Strait Branch of Atlantic water (FSBW) to a more offshore position. We interpret the mid-depth maxima in ^{129}I and ^{237}Np at approximately 800 m as representing a sunken flux of anthropogenic radionuclides not derived from stratospheric bomb fallout, which was entrained within the BSBW and advected off the continental shelf, probably in or near the St. Anna Trough (Rudels *et al.,* 1994; Schlosser *et al.,* 1995; Pavlov and Pfirman, 1995; Schauer *et al.,* 1997). These radionuclides have a major source in the nuclear fuel-reprocessing facilities at Sellafield and La Hague (Kershaw and Baxter, 1995), which are eventually entrained in the northward flowing waters in the Norwegian Coastal Current. Off northern Norway this anthropogenic burden is divided, with the larger fraction continuing via the coastal current and the BSBW onto the Barents shelf, while the remainder is carried northward within the West Spitsbergen Current (Pfirman *et al.,* 1994). This smaller fraction of the western European non-fallout radionuclide signal reaches the Nansen Basin of the Arctic Ocean through Fram Strait as the FSBW (Kershaw and Baxter, 1995).

In the other study, which was undertaken simultaneously with our Laptev Sea sampling, normalized total carbonate and alkalinity data indicate that most of the Ob and Yenisey discharge had passed through Vilkitsky Strait from the Kara Sea into the Laptev Sea before leaving the shelf (Olsson and Anderson, 1997). Therefore, if there is a distinguishable non-fallout burden of anthropogenic radionuclides in the Ob and/or Yenisey discharge, it is likely to have been transported with the surface freshwater onto the Laptev shelf and continental slope, where we sampled in 1993.

Several lines of evidence suggest that our samples from the Laptev shelf and Amundsen Basin surface waters contain a detectable component of anthropogenic radionuclides derived from neither western European sources or stratospheric bomb fallout, but rather from Russian rivers.

The major evidence for a riverine origin includes:

1. An excess of ^{129}I relative to ^{237}Np in the BSBW is observed in the sector, compared with other Arctic basins. By itself, this excess in the BSBW does not rule out a Norwegian Coastal Current origin (i.e. Sellafield and La Hague). However, the presence of significant amounts of ^{237}Np as ^{129}I concentrations approach zero (Fig. 5) imply an additional source of ^{237}Np and also suggest that preferential scavenging of ^{237}Np does not dominate the observed ^{129}I/^{237}Np ratio.

2. This apparent excess of ^{237}Np, over expectations from stratospheric bomb fallout and western European nuclear fuel-reprocessing sources, is present in Arctic Ocean

water, and can best be accounted for by riverine contributions from Russian sources (Beasley *et al.,* 1998a);

3. Maxima of ^{237}Np and ^{129}I were present in the least saline and most ^{18}O-depleted waters sampled in 1993;

4. Ratios of $^{240}Pu/^{239}Pu$ lower than integrated stratospheric bomb fallout occurred at a shallow (37 m) station on the Laptev shelf; and

5. Plutonium occurs in Laptev Sea shelf sediments with $^{240}Pu/^{239}Pu$ ratios as low as 0.04 (Huh *et al.,* 1997), which are not consistent with an origin in either Sellafield or La Hague (Cooper *et al.,* 1999).

Despite these strong indications, several difficulties remain, including reconciling our Laptev Sea observations with the limited data on ^{129}I and ^{237}Np burdens available from Ob and Yenisey waters and/or sediments. Concentrations of ^{129}I on lower reaches of the Yenisey in August, 1993 ($3.57 \pm 0.75 \times 10^8$ atoms kg^{-1}, $n = 10$; Beasley *et al.,* 1997), were an order of magnitude lower than observed in our Laptev Sea samples. In the Ob catchment, concentrations of ^{129}I appear to be higher. In 1994, concentrations of 6.5– 27.1×10^8 atoms kg^{-1} were observed, with the lower concentrations observed within the estuary (Moran *et al.,* 1995). The upper limit for ^{129}I observed in the Ob (2.7×10^9 atoms kg^{-1}) is slightly higher than the mean concentrations observed in the Kara Sea ($\sim 1.4 \times 10^9$ atoms kg^{-1}; Raisbeck *et al.,* 1993; 1995), but as Moran *et al.* (1995) point out, when integrated over the annual discharge of the Ob, the contribution of ^{129}I to the Kara Sea as a whole appears to be only a small fraction of the total ^{129}I inventory of that marginal sea.

Our data by contrast, argue that the surface layer of the Laptev Sea contains a significant burden of anthropogenic radionuclides that are not of stratospheric bomb fallout origin. The $\delta^{18}O$ content of these Laptev Sea surface waters that have high ^{129}I and ^{237}Np burdens indicates that runoff contributes $\sim 20\%$ of the water in those samples, with almost no sea ice melt (Frank, 1996; Fig. 7). The collection of additional samples from the Ob and Yenisey might prove helpful, although there are difficulties with this approach also. For example, the only data on concentrations of ^{129}I in the Ob and Yenisey were collected following the transformation of the Soviet Union, which may have coincided with discharge decreases associated with declines in economic output. Furthermore, these samples were collected in late summer, well after the peak discharge in June. This is problematic since the annual freshet has the potential for mechanically and chemically releasing radionuclides stored within sediments, particularly for elements such as iodine that are preferentially stored within interstitial waters and attached to sediments (Ullman and Aller, 1980; Wong, 1991). Anoxic conditions may also be present during the winter when air exchange is limited through river ice, which would release sediment-bound forms of iodine (Wong, 1991). The limited sediment data available from the Ob does support the possibility of significant radioiodine storage in river sediments and interstitial waters. Surficial sediment concentrations of ^{129}I in the Ob are at least two orders of magnitude

higher than in overlying water collected at the same time, with typical core burdens of 3×10^{11} atoms kg^{-1} of sediments (Moran *et al.,* 1995). Also, the limited sediment core data available from the Ob floodplain (Sayles *et al.,* 1997) indicates that there have likely been episodic discharges of radionuclides not derived from stratospheric bomb fallout in the past. Consequently, based upon our observations of probable non-fallout radionuclide burdens in Laptev surface water layers, and the circumstances under which existing river data have been collected, we conclude that the potential importance of contributions of anthropogenic, non-fallout radionuclides entrained in the Ob and Yenisey may have been underappreciated.

Some of the other indicators we measured do in fact indicate that Russian river discharge significantly impacts some atomic and isotopic ratios, which are probably more sensitive indicators of provenance than concentration and dilution considerations alone (Livingston *et al.,* 1982). It is difficult, for example, to otherwise explain the presence of lower ^{240}Pu/^{239}Pu isotope ratios in the least saline and most ^{18}O-depleted waters sampled (Fig. 7), although the error associated with these low-concentration mass spectrometric measurements is high. Nevertheless, the objective measure of error still indicates that these lower ratios are consistent with significant contributions of nuclear fuel-reprocessing waste. Western European reprocessing sources are also less likely than Russian river sources because the observed ratios in Laptev surface waters are lower than the weighted ^{240}Pu/^{239}Pu of 0.18 that has been discharged from Sellafield (Kershaw *et al.,* 1990; Kershaw *et al.,* 1995; Cooper *et al.,* 1999).

It is more difficult to account for the source of the low ^{240}Pu/^{239}Pu ratio (0.15 ± 0.02) observed in the 1986 sample collected at 21 m in the Beaufort Sea (Fig. 6). We do not think it likely that surface waters with low ^{240}Pu/^{239}Pu ratios, such as we observed over the Laptev shelf, could be transferred without modification of plutonium isotope ratios to the Beaufort Sea, particularly since all other plutonium isotope ratios outside the Laptev Shelf (Fig. 6) reflect contributions consistent with stratospheric bomb fallout and/or integrated Sellafield discharges during 1966–1985. We note, however, that the absolute amounts of both ^{240}Pu (2×10^4 atoms kg^{-1}) and ^{239}Pu (3.5×10^5 atoms kg^{-1}) in this sample are an order of magnitude or more lower than concentrations in any other sample analyzed (Table 1), so the total amount of plutonium in this sample is also anomalously low. One speculative possibility is that this surface water sample had a high degree of recent sea ice melt present in it, leading to very low absolute concentrations of Pu as a result of brine rejection during ice formation. If that ice originally formed in the Laptev Sea, it is possible that the ^{240}Pu/^{239}Pu ratio reflects the ^{240}Pu/^{239}Pu ratio in the Laptev surface water as ice formed. Unfortunately, we have no ^{18}O data for the sample that would help to confirm the extent of sea ice melt content in this surface water.

These generally higher ^{240}Pu/^{239}Pu ratios in dissolved plutonium are present in almost all other deeper and more saline water samples, regardless of the concentrations in those waters of more conservative radionuclides such as ^{237}Np and ^{129}I. Since plutonium is much more particle-reactive than ^{237}Np and ^{129}I (Beasley *et al.,* 1998a), sedimentation on the

Laptev shelf may explain the lower ^{240}Pu/^{239}Pu ratios that have been reported in some Laptev shelf sediments (Huh *et al.,* 1997).

In a similar way, the distinct, relatively low and invariant ^{237}Np/^{129}I ratio observed in waters of the Laptev Sea and Amundsen Basin (Figs. 4, 5) imply an additional source of ^{129}I in the BSBW. We were unable to obtain samples from deeper than 300 m during the Arctic Ocean Section in 1994, and we are not certain whether there is a temporal cause to the difference in the ^{237}Np/^{129}I ratio in the deep water profiles between the Canada Basin (1986) and the Amundsen Basin (1993), or whether the profiles simply reflect differing sources. It also is possible that higher scavenging of ^{237}Np, relative to ^{129}I may explain some inter-basin differences, although as discussed above, we do not think that this process is particularly significant in impacting the isotopic and elemental ratios we discuss here. The water samples analyzed as part of this study were unfiltered, so discussion of processes such as colloid formation and the transport of radionuclides on such low density, "dissolved" fractions (e.g. Guo and Santschi, 1997) is beyond the scope of this work.

It is also possible that biological utilization of iodine during storage could have affected the samples collected in 1986 in the Canada Basin (see discussion by Edmonds *et al.,* 1998); all of our other samples were poisoned following collection. However, ^{237}Np/^{129}I ratios in the Canada Basin collected in 1993 were little different for samples collected at the same depths as in 1986, despite a four-fold increase in concentration in both radionuclides at their water column maxima, $\sim$300 m over that seven year period (Fig. 2). It is possible that the prevailing low biological productivity in the Canada Basin negated this possible problem of biological utilization of iodine that was inferred by Edmonds *et al.* (1998) for their stored samples, which were primarily collected from the North Atlantic. At least to the depth of 300 m, with some exceptions that probably reflect effective lateral dispersion (such as suggested by Fig. 7), waters of the Canada Basin typically had higher ^{237}Np/^{129}I ratios in 1994 than the Amundsen Basin and the Laptev shelf did in 1993. Given the indications of excess ^{237}Np in Arctic Ocean waters to depths of at least 300 m, with some originating from Russian riverine sources (Beasley *et al.,* 1998a), it seems plausible that a riverine contribution of ^{129}I (and other radionuclides) lowered ^{237}Np/^{129}I ratios on the Laptev shelf and adjacent portions of the Amundsen Basin. It is possible, however, that preferential particle scavenging of ^{237}Np may also play a role in some of the inter-basin ^{237}Np/^{129}I ratio differences.

5. Conclusions

As a result of revelations about the release of anthropogenic radionuclides from Russian sources during the Cold War era, the past decade has seen a resurgence of interest in the distributions of radioactive isotopes in the Arctic Ocean. A number of studies, including many funded as part of the United States Arctic Nuclear Waste Assessment Program, have concluded that current releases from Russian sources are insignificant in comparison to stratospheric bomb fallout and the western European discharge sources at Sellafield and La Hague. Our results are not inconsistent with these conclusions from the standpoint of the

simple dilution of [129]I originating in Sellafield and La Hague and subsequently carried in BSBW that may mix with river waters during transit of the Russian shelves. Nevertheless, analyses of isotope ratios, including [237]Np/[129]I and [240]Pu/[239]Pu, rather than the concentrations of each individual radioisotope, are likely to be more sensitive indicators of sources. Measurements of these ratios in Arctic waters influenced by runoff suggest that Russian riverine contributions are significant in determining isotopic and elemental ratios on the Laptev shelf and in the Amundsen Basin. Comparison of anthropogenic radionuclides over the Beaufort slope relative to concentrations observed at the AIWEX site in 1985 (Livingston, 1988) also suggests that part of the Sellafield/La Hague burden of radionuclides generated by western European nuclear fuel-reprocessing activities has been preferentially transported with the boundary undercurrent into the Canada Basin along the continental slope, rather than directly across the basin. Taken together, these observations indicate that anthropogenic radionuclides in the Arctic Ocean have additional significant sources beyond those previously recognized, and that the distributions of those radionuclides are conditioned by a complex circulation system. It is clear that there is a need for additional measurements of tracer ratios, including within the deeper waters of all the Arctic basins, as well as expanded studies of transport of these tracers on the Arctic continental shelves and adjacent slopes.

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