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Radionuclides from Windscale discharges II: Their dispersion in Scottish and Norwegian coastal circulation

by Hugh D. Livingston[1], Vaughan T. Bowen[1,3] and Stuart L. Kupferman[1,2]

ABSTRACT

Measurement of the concentrations of ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{238}Pu, and $^{239,240}Pu$ in Scottish and Norwegian coastal waters in 1976 and 1978 provides information on dispersal pathways, transport times, and dilution in these waters of radioactive waste discharged to the Irish Sea from the Windscale nuclear fuel reprocessing plant in Cumbria, U. K. Consideration of such nuclide ratios as $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ both in Windscale discharges and in these coastal waters is shown to be a more powerful tool for transport rate derivation than are the changing concentrations in space and time of individual radionuclides. The soluble radionuclides in the discharges are held in the Irish Sea for about two years, moved relatively rapidly to the North Sea and discharged from there to the Norwegian coastal current, subject in hydrographic regions of the North Sea to residence times which increase from north to south from a few months to less than two years. Dilution in the main Windscale plume between the Irish Sea outflow and North Sea inflow, though variable over the short-term is estimated to average a factor of three. Certainly by 1978, if not a little earlier, the North Sea outflow in the Norwegian coastal current had begun to be labelled with the increased concentrations of ^{134}Cs and ^{137}Cs resulting from the sharp increase which began in 1974 in their discharge to the Irish Sea.

1. Introduction

The marginal seas of the European continental shelf have been used during recent years to dilute and disperse radionuclides from low-level liquid waste streams generated in connection with the reprocessing of spent nuclear fuel. The principal sources for these discharges are the British Nuclear Fuels Ltd. (BNFL) reprocessing plant at Windscale on the northeast coast of the Irish Sea and the French plant at Cap de la Hague on the Normandy coast. The Cap de la Hague discharges, not as well documented as are those of Windscale, do not appear to have exceeded 10-15% of the latter (discussed in detail later), in respect to those nuclides with which we are concerned. A third source, from the experimental facility at Dounreay, Scotland, is not reported ever to have approached even 1% of the Windscale amounts (Luykx and Fraser, 1978). Livingston *et al.* (1982) have drawn attention to the

1. Woods Hole Oceanographic Institution, Woods Hole, Massachusetts, 02543, U.S.A.
2. Present Address: Sandia Laboratories, Division 4536, Albuquerque, New Mexico, 87185, U.S.A.
3. Present Address: P.O. Box 504, Wayne, Pennsylvania, 19087, U.S.A.

contribution that releases from Windscale from 1957 to 1979 have made to high latitude North Atlantic distributions and inventories of artificial radionuclides and to the enormous potential use of these releases for North Atlantic and Arctic tracer oceanography.

For the various radionuclides released in these discharges, it is recognized that knowledge of their subsequent fates is essential for determination of the capacities of the various marine compartments which receive them. The biogeochemistry of a given radionuclide determines the compartment principally involved in its fate after release. Radionuclides such as ^{90}Sr and $^{134,137}Cs$ are believed to behave predominantly as solute and are transported, diluted, and dispersed by water mass movement (Deutsches Hydrographisches Institut, 1971, 1972; Kautsky, 1977; Jefferies *et al.*, 1973; Hetherington, 1976a; Mitchell, 1977a,b; Livingston and Bowen, 1977; Baxter *et al.*, 1979; McKinley *et al.*, 1981a,b). Other nuclides, such as the transuranic and lanthanide elements, are mostly immobilized locally in sediments near the points of discharge (Hetherington *et al.*, 1976; Hetherington, 1976b; Nelson and Lovett, 1978).

British (Hetherington, 1976a; Mitchell, 1977a,b) and German (Deutsches Hydrographisches Institut, undated; Kautsky *et al.*, 1980) studies of ^{137}Cs distributions from Windscale releases into U. K. coastal waters reveal details of its (and by inference other soluble radionuclide components of the waste stream) dispersion pathways within and outside the Irish Sea. These data convincingly demonstrate that the ^{137}Cs flow around Scotland into the North Sea is the major pathway for dispersion of nuclide-labelled water leaving the Irish Sea. Other studies (Kautsky, 1976, 1977; Murray and Kautsky, 1977; Kautsky *et al.*, 1980) have demonstrated that the major flow of soluble nuclides released by the Cap de la Hague plant proceeds northward through the English Channel, along the continental coastline to the Skagerrak then northward along the Norwegian coast and out of the North Sea.

Despite the fact that such nuclides as the transuranics are, as noted above, mostly immobilized in sediments near to the discharge point, several studies have indicated that small but significant concentrations of Pu and Am move with the soluble nuclides dispersed in the water circulation. This has been seen from the Windscale releases within the Irish Sea (Hetherington, 1976b; Nelson and Lovett, 1978) and in Scottish coastal water (Livingston and Bowen, 1977; Murray *et al.*, 1978), and from Cap de la Hague in the southern North Sea (Murray and Kautsky, 1977).

Knowledge of the rates at which soluble radionuclides are dispersed throughout the whole coastal circulation system is highly relevant to the establishment of levels of the various radionuclides safely dischargeable over time. Kautsky (1977) concluded, from comparisons of ^{137}Cs concentrations in surface water, that a patch of water labelled with 3 or 4×10^3 Ci of ^{137}Cs released from Cap de la Hague in early 1971, took two years to travel to the Skagerrak. Jefferies *et al.* (1973) used the comparison of the ratio ^{134}Cs to ^{137}Cs in seawater to that in the Windscale dis-

charges to conclude that the "transit time"[4] for water to be moved from the Windscale area to the North Channel (northern exit of the Irish Sea) was between 1.1 and 1.8 years. Later work with these nuclides led them to conclude that the residence half-times of water (a) between the Isle of Man and the Cumbrian coast and (b) between the Isle of Man and the Irish coast are 150 and 100 days respectively (Ministry of Agriculture, Fisheries and Food, 1977). Livingston and Bowen (1977) concluded that the "transit time"[3] for water to move from the Windscale area to the Minch (off N.W. Scotland) was two years—also based on a comparison of measured seawater ratios of Cs radionuclides with those believed to characterize the Windscale discharges.

One of the principal purposes of this paper is to demonstrate that, in developing a good time framework for viewing the discharge transport, measurements of radionuclide *ratio* changes with time are more informative than *concentration* changes. This conclusion is based on data obtained from samples collected in Scottish and Norwegian coastal waters in 1976 and 1978. These data should be compared to, and viewed in the framework of, the extensive ^{137}Cs surveys made in 1976 by the British (Mitchell, 1977b; McKinley *et al.*, 1981a) and Germans (Deutsches Hydrographisches Institut, undated; Kautsky *et al.*, 1980), in 1977 by the British (McKinley *et al.*, 1981a) and in 1978 by the British and Germans, when data from their joint survey become available. Beside our examination of nuclide ratios amongst the conventionally viewed soluble nuclides, we compare with the discharge data the changing concentrations of Pu in coastal seawater and their ratios to ^{90}Sr concentrations—in the context of their indication of the significance of secondary remobilization phenomena in the transport of reactive radionuclides such as Pu.

2. Sample collection, analyses, and results

The water samples for which data are presented in this paper were collected on three cruises during 1976 and four in 1978. As part of the Flex Program (Fladen Ground Experiment—part of Jonsdap '76 International program on the interplay of environment and a plankton bloom) a series of 50-1 water samples and 21 cm diameter sediment cores was collected for radionuclide analysis from the North Sea and northern European coastal waters by Woods Hole Oceanographic Institution (WHOI) on cruise number 54 of R.V. *Knorr*, May and June 1976. Positions, dates, and salinities for the Woods Hole samples are included in Table 1. As part of the continuing program of Danish measurements of radionuclides in the North Sea, R.V. *Dana* collected two series of surface samples in 1976—in February and

4. As this term has recently been defined (Otto, 1981) in the context of time constants which apply to the exchange between reservoirs of specific sea areas, it may be more appropriate to substitute the term "average time of transport." We have adopted this terminology throughout the paper and we use the term "turnover time" (equivalent to mean residence time) for the concept of how long a body of water spends in a given reservoir.

Table 1. 1976 European coastal water concentrations of radionuclides from nuclear fuel reprocessing releases.

Ship and cruise no., station no., sample depth m, and date	Latitude/ Longitude	Salinity ‰	^{134}Cs pCi/Kg	^{137}Cs pCi/Kg	$^{134}Cs/^{137}Cs$	^{90}Sr pCi/Kg	$^{137}Cs/^{90}Sr$	$^{239,240}Pu$ fCi/Kg	^{238}Pu fCi/Kg	$^{238}Pu/^{239,240}Pu$
Western Scotland coastal water										
Knorr 54/6, 84, surface	58°15′N	34.698	0.79	6.13	0.128	0.998	6.15	2.6	0.28	0.11
5/27/76	5°50′W		±0.04	±0.03	±0.007	±0.009	±0.07	±0.2	±0.09	±0.04
" , 30 m	"	34.712	0.82	6.52	0.126	1.00	6.51	2.6	0.41	0.16
			±0.04	±0.05	±0.005	±0.02	±0.13	±0.1	±0.05	±0.02
" , 60 m	"	34.751	0.77	5.84	0.132	0.923	6.32	2.6	0.46	0.18
			±0.02	±0.04	±0.004	±0.009	±0.08	±0.2	±0.05	±0.02
" , 90 m	"	34.871	0.59	4.66	0.126	0.793	5.87	2.2	0.28	0.13
			±0.01	±0.04	±0.003	±0.023	±0.18	±0.1	±0.05	±0.02
Northern North Sea										
Knorr 54/6, 24, surface	58°26′N	34.953	0.27	2.18	0.122	0.53	4.14	1.2	0.27	0.22
5/12/76	1°45′E		±0.02	±0.01	±0.006	±0.01	±0.04	±0.1	±0.05	±0.04
" , 30 m	"	34.987	0.30	2.44	0.124	0.57	4.27	1.4	0.27	0.19
			±0.02	±0.12	±0.006	±0.01	±0.06	±0.2	±0.09	±0.07
" , 60 m	"	35.016	0.30	2.00	0.14	0.51	3.97	1.7	0.27	0.16
			±0.09	±0.06	±0.04	±0.01	±0.12	±0.2	±0.14	±0.09
" , 90 m	"	35.039	0.16	1.70	0.10	0.43	3.99	1.4	0.27	0.19
			±0.05	±0.03	±0.04	±0.01	±0.08	±0.1	±0.05	±0.04
Knorr 54/6, 19, surface	58°36′N	35.010	0.31	2.55	0.124	0.46	5.56	1.3	0.27	0.21
5/11/76	0°03′E		±0.02	±0.01	±0.005	±0.01	±0.17	±0.1	±0.05	±0.04
Knorr 54/6, 19, 80 m	58°26′N	35.100	0.23	1.93	0.122	0.44	4.36	0.81	0.23	0.28
5/11/76	0°03′E		±0.01	±0.01	±0.006	±0.01	±0.1	±0.14	±0.05	±0.08
Knorr 54/6, 14, surface	59°20′N	35.055	0.26	2.16	0.122	0.37	5.78	1.1	0.16	0.15
5/11/76	0°02′E		±0.03	±0.02	±0.013	±0.003	±0.07	±0.1	±0.03	±0.03

Knorr 54/6, 32, surface 5/14/76	58°51′N 0°42′E	35.125	0.19 ±0.03	1.59 ±0.02	0.116 ±0.017	0.31 ±0.002	5.13 ±0.07	1.0 ±0.1	0.12 ±0.03	0.12 ±0.03
" , surface duplicate	"	35.125	n. m.	1.73 ±0.01	—	0.32 ±0.01	5.41 ±0.17	0.90 ±0.09	0.14 ±0.04	0.16 ±0.05
" , 40 m	"	35.133	n. m.	1.65 ±0.01	—	0.31 ±0.01	5.32 ±0.17	0.95 ±0.09	0.05 ±0.05	0.05 ±0.06
" , 40 m duplicate	"	35.133	n. m.	1.65 ±0.01	—	0.31 ±0.01	5.32 ±0.17	0.63 ±0.09	0.1 ±0.5	0.16 ±0.80
" , 80 m	"	35.144	n. m.	1.77 ±0.01	—	0.39 ±0.01	4.54 ±0.12	0.95 ±0.09	0.14 ±0.05	0.15 ±0.05
" , 120 m	"	35.174	n. m.	1.27 ±0.01	—	0.29 ±0.01	4.38 ±0.15	0.86 ±0.05	0.09 ±0.05	0.10 ±0.06
Knorr 54/6, 9, surface 5/10/76	59°20′N 1°46′E	35.215	0.10 ±0.03	0.66 ±0.01	0.15 ±0.04	0.17 ±0.01	3.88 ±0.23	0.86 ±0.09	0.03 ±0.02	0.035 ±0.024
Knorr 54/6, 68, surface 5/21/76	61°49′N 0°50′E	35.215	n. m.	0.154 ±0.002	—	0.073 ±0.001	2.11 ±0.04	1.2 ±0.1	0.01 ±0.1	0.01 ±0.01
Knorr 54/6, 48, surface 5/16/76	60°22′N 1°58′E	35.207	n. m.	0.287 ±0.002	—	0.091 ±0.003	3.15 ±0.11	0.63 ±0.09	0.03 ±0.02	0.05 ±0.03
Norwegian coastal current										
Knorr 54/6, 30A, surface 5/13/76	58°55′N 4°10′E	32.995	0.068 ±0.010	0.88 ±0.03	0.078 ±0.012	0.44 ±0.01	2.00 ±0.08	0.45 ±0.05	<0.05	<0.11
" , 60 m	"	34.951	n. m.	0.636 ±0.005	—	0.266 ±0.002	2.39 ±0.03	0.39 ±0.08	0.01 ±0.06	0.03 ±0.16
" , 120 m	"	35.109	0.044 ±0.014	0.459 ±0.014	0.096 ±0.031	0.197 ±0.001	2.33 ±0.07	0.63 ±0.05	0.06 ±0.02	0.10 ±0.03
" , 180 m	"	35.211	n. m.	0.377 ±0.002	—	0.135 ±0.002	2.79 ±0.04	0.39 ±0.08	0.02 ±0.05	0.03 ±0.10
" , 240 m	"	35.223	n. m.	0.427 ±0.003	—	0.104 ±0.001	4.11 ±0.05	0.56 ±0.11	0.05 ±0.05	0.06 ±0.06

Table 1 (continued)

Ship and cruise no., station no., sample depth m, and date	Latitude/ Longitude	Salinity ‰	^{134}Cs pCi/Kg	^{137}Cs pCi/Kg	$^{134}Cs/^{137}Cs$	^{90}Sr pCi/Kg	$^{137}Cs/^{90}Sr$	$^{239,240}Pu$ fCi/Kg	^{238}Pu fCi/Kg	$^{238}Pu/^{239,240}Pu$
Knorr 54/6, 50, surface 5/17/76	61°29'N 3°26'E	34.282	0.032 ±0.003	0.527 ±0.013	0.060 ±0.005	0.277 ±0.001	1.90 ±0.05	0.72 ±0.09	0.09 ±0.03	0.13 ±0.04
" , 50 m	"	34.929	n. m.	0.518 ±0.005	—	0.235 ±0.001	2.20 ±0.02	0.60 ±0.07	0.02 ±0.02	0.04 ±0.04
" , 100 m	"	35.101	0.035 ±0.014	0.345 ±0.012	0.10 ±0.04	0.146 ±0.001	2.36 ±0.08	0.90 ±0.09	0.05 ±0.02	0.06 ±0.03
" , 150 m	"	35.179	n. m.	0.186 ±0.002	—	0.109 ±0.005	1.71 ±0.08	0.67 ±0.08	0.01 ±0.03	0.01 ±0.05
" , 200 m	"	35.150	n. m.	0.253 ±0.016	—	0.108 ±0.003	2.34 ±0.16	0.68 ±0.09	0.05 ±0.02	0.07 ±0.03
" , 300 m	"	35.208	n. m.	0.286 ±0.002	—	0.130 ±0.003	2.20 ±0.05	1.0 ±0.1	0.01 ±0.02	0.01 ±0.02
Norwegian Sea										
Knorr 54/6, 70, surface 5/22/76	63°50'N 0°54'E	35.123	n. m.	0.127 ±0.002	—	0.080 ±0.001	1.59 ±0.03	0.77 ±0.09	0.03 ±0.02	0.04 ±0.03
" , 100 m	"	35.109	n. m.	0.103 ±0.003	—	0.058 ±0.003	1.76 ±0.10	0.77 ±0.09	−0.05 ±0.05	−0.06 ±0.06
" , 300 m	"	34.904	n. m.	0.095 ±0.003	—	0.061 ±0.005	1.56 ±0.14	0.68 ±0.09	0 ±0.05	<0.07
" , 500 m	"	34.917	n. m.	0.092 ±0.003	—	0.067 ±0.007	1.37 ±0.15	0.81 ±0.09	0.01 ±0.02	0.01 ±0.02
" , 1250 m	"	34.912	n. m.	0.028 ±0.002	—	0.018 ±0.001	1.56 ±0.14	0.32 ±0.05	−0.03 ±0.02	−0.1 ±0.1
" , 2150 m	"	34.911	n. m.	0.014 ±0.003	—	0.011 ±0.002	1.27 ±0.34	0.22 ±0.04	−0.02 ±0.02	−0.1 ±0.1

n. m. = Not measured

June (Aarkrog *et al.*, 1977). In July and August of 1978, R.V. *Atlantis II* (WHOI) and R.V. *Endeavour* (University of Rhode Island) collected a series of 50-1 surface water samples along tracks from Glasgow, Scotland, or Bergen, Norway, to the site of the international JASIN (Joint Air Sea Interaction Program) experiment between Scotland and Iceland. R.V. *Explorer* of the Marine Laboratory of the Department of Agriculture and Fisheries for Scotland, in Aberdeen, collected samples in the central and northern North Sea and in the coastal stream around North Scotland on two cruises in April and June 1978.

Analytical methods used for analysis of the samples collected on these cruises (all of which, except those of R.V. *Dana*, were analyzed at WHOI) were referenced in an earlier publication describing radionuclide measurements of water and sediments from station 84 in the Minch (Livingston and Bowen, 1977) of R.V. *Knorr* cruise 54. Those used in analysis of the R.V. *Dana* samples have been referenced by Aarkrog *et al.* (1977). The quality of the analytical data was assured through the analysis of samples of duplicates, environmental blanks (Bowen, 1978), and reagent blanks within the regular analytical sample series. In addition, we receive confirmation of the reliability of our analytical data through regular participation in intercomparison exercises such as are organized by IAEA, NBS, EPA or the Office of Health & Environmental Research of the U.S. Department of Energy. A fuller account of our quality control procedures has been referenced in a recent report (Bowen *et al.*, 1980).

The analytical data for the Woods Hole samples are presented in Tables 1 and 2. The positions of the samples collected in 1978 are shown in Figure 1, and of the 1976 samples in Figure 2 (by plots of the ratio $^{137}Cs/^{90}Sr$ measured in samples from each location—including those from the Danish cruises). For convenience in discussing the data, four regions A, B, C, and D are identified in Figures 1 and 2; these correspond respectively to West Scottish coastal water, North Scottish coastal water, northern North Sea water and the Norwegian coastal current. The boundary between regions C and D was defined on the basis of salinities measured at the various stations; stations with salinities less than 34‰ were taken to be in the Norwegian coastal current (region D) while those greater were assigned to region C.

3. Discussion

a. Surface circulation

The fates of radionuclides moved by water transport and the complexities of their passage to and through the North Sea will be more easily visualized if we first summarize the residual (i.e., nontidal) surface circulation and water budget of the area. These and other North Sea properties were recently reviewed (Lee, 1980; Becker, 1981). Figure 3 shows the residual surface currents after a recently pub-

Table 2. 1978 European surface coastal water concentrations of radionuclides from nuclear fuel reprocessing releases.

	Ship, (cruise), station no., and date	Latitude/ Longitude	Salinity ‰	^{134}Cs pCi/Kg	^{137}Cs pCi/Kg	$^{134}Cs/^{137}Cs$	^{90}Sr pCi/Kg	$^{137}Cs/^{90}Sr$	$^{239,240}Pu$ fCi/Kg	^{238}Pu fCi/Kg	$^{238}Pu/^{239,240}Pu$
Western Scotland coastal water*											
9+	*Atlantis II*, (102)	55°31′N	34.618	1.16	12.4	0.10	1.71	7.26	6.22	1.7	0.28
	No. 9, 8/15/78	7°30′W		±0.17	±0.2	±0.02	±0.01	±0.12	±0.59	±0.3	±0.06
8	*Atlantis II*, (102)	55°30′N	35.157	0.37	3.37	0.11	n. m.	—	n. m.	n. m.	—
	No. 8, 8/15/78	8°08′W		±0.13	±0.14	±0.04					
7	*Atlantis II*, (102)	55°48′N	35.239	0.06	0.75	0.08	n. m.	—	n. m.	n. m.	—
	No. 7, 8/15/78	8°45′W		±0.02	±0.03	±0.03					
2	*Atlantis II*, (102)	56°00′N	34.283	1.82	20.6	0.09	3.38	6.09	9.14	2.2	0.24
	No. 2, 7/26/78	7°02′W		±0.20	±0.2	±0.01	±0.01	±0.06	±0.68	±0.3	±0.04
3	*Atlantis II*, (102)	56°29′N	34.709	0.79	8.02	0.10	0.93	8.62	3.1	0.47	0.15
	No. 3, 7/26/78	8°00′W		±0.21	±0.28	±0.03	±0.03	±0.41	±0.5	±0.18	±0.06
11	*Atlantis II*, (102)	56°30′N	35.177	0.06	0.51	0.12	n. m.	—	n. m.	n. m.	—
	No. 11, 8/22/78	9°00′W		±0.02	±0.17	±0.05					
6	*Atlantis II*, (102)	56°30′N	35.349	<0.03	0.23	—	n. m.	—	n. m.	n. m.	—
	No. 6, 8/15/78	10°00′W			±0.01						
42	*Explorer*	56°49′N	34.725	1.90	19.4	0.098	2.90	6.69	8.5	1.9	0.22
	No. 11, 4/13/78	6°55′W		±0.08	±0.2	±0.004	±0.01	±0.07	±0.3	±0.1	±0.01
4	*Atlantis II*, (102)	57°30′N	34.738	0.98	10.9	0.090	1.65	6.62	2.57	0.49	0.19
	No. 4, 7/26/78	9°00′W		±0.03	±0.1	±0.003	±0.02	±0.10	±0.14	±0.05	±0.02
12	*Atlantis II*, (102)	57°31′N	35.087	0.05	0.60	0.090	n. m.	—	n. m.	n. m.	—
	No. 12, 8/22/78	10°04′W		±0.03	±0.03	±0.04					
13	*Atlantis II*, (102)	58°00′N	35.091	<0.02	0.088	<0.2	0.060	1.5	0.50	0.009	0.02
	No. 13, 9/7/78	14°00′W			±0.003		±0.002	±0.2	±0.09	±0.009	±0.02
5	*Atlantis II*, (102)	58°30′N	35.178	0.13	1.47	0.086	n. m.	—	n. m.	n. m.	—
	No. 5, 7/26/78	10°00′W		±0.01	±0.02	±0.008					
1	*Atlantis II*, (102)	55°29′N	n. m.	1.45	15.4	0.094	2.68	5.75	2.3	0.54	0.23
	No. 1, 7/26/78	6°29′W		±0.20	±0.2	±0.013	±0.04	±0.11	±0.2	±0.14	±0.06

10	*Atlantis II*, (102)	55°30′N	n. m.	n. m.	n. m.	—	n. m.	—	n. m.	n. m.	
	No. 10, 8/15/78	7°00′W									
North Scotland coastal water**											
41+	*Explorer*	58°39.8′N	34.364	1.21	13.2	0.091	1.93	6.84	6.4	1.4	0.22
	No. 17, 4/7/78	5°15.5′W		±0.4	±0.1	±0.004	±0.01	±0.06	±0.1	±0.1	±0.02
40	*Explorer*	58°42′N	34.413	1.22	13.9	0.087	1.85	7.51	5.0	1.2	0.24
	No. 7, 4/7/78	4°43′W		±0.04	±0.1	±0.003	±0.01	±0.07	±0.1	±0.1	±0.02
39	*Explorer*	58°45′N	34.743	0.64	7.00	0.090	0.970	7.21	3.41	0.62	0.18
	No. 22, 4/7/78	3°30′W		±0.01	±0.07	±0.010	±0.026	±0.21	±0.15	±0.06	±0.02
14	*Endeavour*	59°30′N	35.254	0.03	0.15	0.16	n. m.	—	n. m.	n. m.	—
	No. 16, 8/27/78	7°30′W		±0.02	±0.02	±0.12					
15	*Endeavour*	59°30′N	35.136	0.21	2.72	0.081	n. m.	—	n. m.	n. m.	—
	No. 15, 8/27/78	5°30′W		±0.01	±0.02	±0.004					
16	*Endeavour*	59°30′N	35.182	0.15	1.68	0.090	n. m.	—	n. m.	n. m.	—
	No. 14, 8/26/78	4°30′W		±0.03	±0.05	±0.020					
17	*Endeavour*	59°30′N	35.011	0.38	4.35	0.090	n. m.	—	n. m.	n. m.	—
	No. 13, 8/26/78	4°00′W		±0.04	±0.05	±0.010					
18	*Endeavour*	59°30′N	35.050	0.64	7.38	0.090	0.997	7.40	2.54	0.53	0.21
	No. 12, 8/26/78	3°30′W		±0.05	±0.08	±0.010	±0.005	±0.09	±0.11	±0.05	±0.02
19	*Endeavour*	59°30′N	34.725	0.95	10.2	0.093	1.25	8.13	3.54	0.74	0.21
	No. 11, 8/26/78	2°30′W		±0.07	±0.1	±0.007	±0.01	±0.10	±0.13	±0.05	±0.02
20	*Endeavour*	59°20′N	34.707	0.82	11.2	0.073	1.40	8.00	3.81	0.84	0.22
	No. 10, 8/26/78	2°20′W		±0.04	±0.1	±0.004	±0.01	±0.09	±0.14	±0.06	±0.02
21	*Endeavour*	59°30′N	34.925	0.53	6.48	0.081	0.913	7.10	2.59	0.55	0.21
	No. 9, 8/26/78	2°00′W		±0.04	±0.05	±0.005	±0.004	±0.06	±0.12	±0.05	±0.02
22	*Endeavour*	59°45′N	35.085	0.13	1.61	0.075	n. m.	—	n. m.	n. m.	—
	No. 8, 8/28/78	1°30′W		±0.03	±0.03	±0.014					
23	*Endeavour*	60°00′N	34.675	0.10	0.88	0.12	n. m.	—	n. m.	n. m.	—
	No. 7, 8/26/78	0°30′W		±0.08	±0.05	±0.11					

* = Region A of Figure 1
** = Region B of Figure 1
+ = Sample identification number (Fig. 1)
n. m. = Not measured

Table 2 (continued)

Ship, (cruise), station no., and date	Latitude/ Longitude	Salinity ‰	^{134}Cs pCi/Kg	^{137}Cs	$^{134}Cs/^{137}Cs$	^{90}Sr pCi/Kg	$^{137}Cs/^{90}Sr$	$^{239,240}Pu$ fCi/Kg	^{238}Pu	$^{238}Pu/^{239,240}Pu$
Northern North Sea***										
38+ *Explorer*	57°53′N	34.650	0.55	6.18	0.090	0.736	8.40	2.49	0.50	0.20
No. 1, 4/6/78	1°55′W		±0.05	±0.09	±0.09	±0.021	±0.27	±0.14	±0.06	±0.03
36 *Explorer*	57°15′N	34.501	0.64	6.92	0.091	1.06	6.53	2.5	0.34	0.13
No. 16, 4/26/78	1°21′W		±0.03	±0.06	±0.004	±0.01	±0.08	±0.1	±0.05	±0.02
37 *Explorer*	57°10′N	32.314	0.77	9.14	0.084	1.16	7.88	2.39	0.35	0.15
No. 13, 4/6/78	1°59′W		±0.10	±0.13	±0.011	±0.01	±0.13	±0.18	±0.06	±0.03
35 *Explorer*	57°27′N	33.841	0.63	6.73	0.090	0.851	7.91	1.43	0.32	0.13
No. 14, 4/18/78	0°37′E		±0.05	±0.19	±0.009	±0.005	±0.23	±0.20	±0.06	±0.03
34 *Explorer*	57°45.5′N	35.014	0.40	4.52	0.089	0.584	7.74	1.83	0.35	0.19
No. 2, 4/25/78	1°35.5′E		±0.03	±0.03	±0.005	±0.003	±0.06	±0.11	±0.05	±0.03
Norwegian coastal current****										
33+ *Explorer*	57°45′N	33.023	0.29	2.49	0.12	0.580	4.29	0.88	0.04	0.04
No. 4, 6/30/78	3°00′E		±0.04	±0.05	±0.02	±0.003	±0.09	±0.08	±0.01	±0.01
33 Duplicate			0.22	2.82	0.079	n. m.	—	n. m.	n. m.	—
			±0.03	±0.04	±0.021					

32	*Explorer*	58°45′N	32.277	0.09	1.41	0.06	0.429	3.29	0.51	0.03	0.06
	No. 3, 6/29/78	2°31′E		±0.02	±0.03	±0.02	±0.002	±0.07	±0.05	±0.02	±0.04
31	*Explorer*	59°46′N	31.736	0.17	1.55	0.11	0.384	4.04	0.64	0.03	0.05
	No. 12, 6/28/78	3°35′E		±0.03	±0.05	±0.02	±0.014	±0.20	±0.03	±0.02	±0.03
30	*Explorer*	60°46′N	31.071	0.10	1.41	0.07	0.327	4.31	0.77	0.08	0.10
	No. 5, 6/27/78	3°25′E		±0.03	±0.04	±0.03	±0.001	±0.12	±0.09	±0.04	±0.06
29	*Endeavour*	60°00′N	31.814	0.15	1.88	0.08	0.436	4.31	0.45	0.05	0.11
	No. 1, 8/25/78	5°00′E		±0.04	±0.05	±0.02	±0.004	±0.12	±0.09	±0.03	±0.07
28	*Endeavour*	60°00′N	32.079	0.15	1.93	0.08	0.462	4.18	0.50	0.05	0.10
	No. 2, 8/25/78	4°30′E		±0.02	±0.03	±0.01	±0.002	±0.07	±0.09	±0.03	±0.07
27	*Endeavour*	60°00′N	32.940	0.07	1.23	0.05	0.260	4.73	0.49	0.02	0.04
	No. 3, 8/25/78	4°00′E		±0.02	±0.03	±0.02	±0.003	±0.13	±0.05	±0.02	±0.04
26	*Endeavour*	60°00′N	33.498	0.13	1.08	0.12	0.317	3.41	0.40	0.01	0.03
	No. 4, 8/25/78	3°30′E		±0.03	±0.04	±0.03	±0.009	±0.16	±0.05	±0.01	±0.03
25	*Endeavour*	60°00′N	33.813	0.08	0.95	0.08	0.234	4.06	0.32	0.02	0.06
	No. 5, 8/26/78	3°00′E		±0.03	±0.04	±0.03	±0.003	±0.18	±0.05	±0.02	±0.06
24	*Endeavour*	60°00′N	33.696	0.06	1.21	0.05	0.273	4.43	0.40	0.03	0.08
	No. 6, 8/26/78	2°00′E		±0.01	±0.01	±0.01	±0.003	±0.06	±0.05	±0.01	±0.03

*** = Region C of Figure 1
**** = Region D of Figure 1
⁺ = Sample identification number (Fig. 1)
n. m. = Not measured

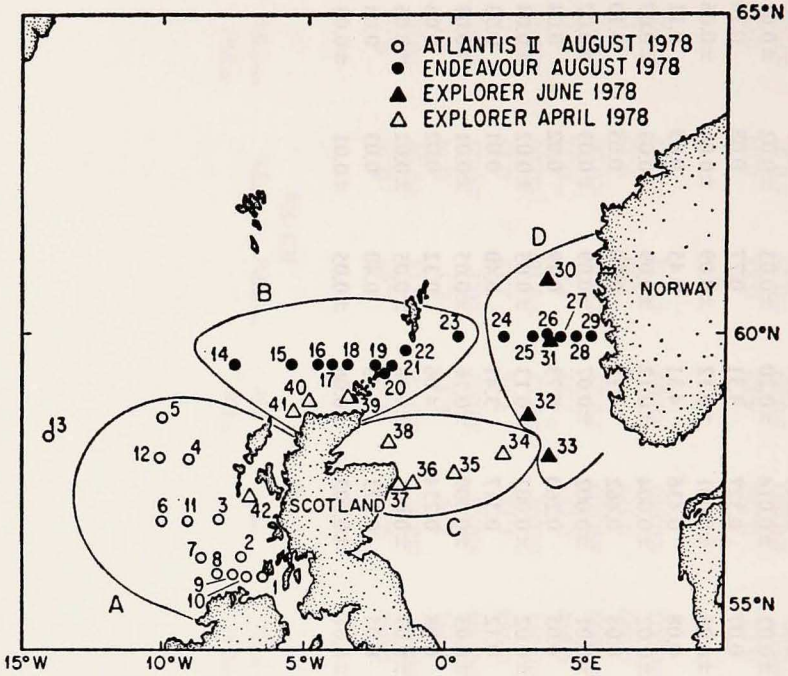

Figure 1. Surface seawater sampling positions—1978 (cf. Table 2).

lished chart (Lee and Ramster, 1981). These current patterns were inferred through the use of standard physical oceanographic techniques independently of any radioisotope results. Superimposed are isohalines showing the mean annual surface salinity distribution (ICES, 1933). The figure also shows the locations of Windscale and Cap de la Hague, the major points of radionuclide releases. The large-scale movement of water in the area is northward up the European shelf, driven by the prevailing winds.

The geographical setting of the land-masses around the Irish Sea leads to its being flushed almost wholly by Atlantic Ocean water that enters from the south through the St. George's Channel. This inflow is balanced by outflow principally northward through the North Channel. There is a counterclockwise gyral circulation within the Irish Sea, a very small and probably sporadic return flow southward out of the St. George's Channel, and some variability in the distribution of the outflow water into the Scottish coastal current. Nevertheless, the mean circulation patterns of the Irish Sea are relatively simple and consistent.

The North Sea is a much more difficult proposition. The largest inflow, estimated at 23,000 km^3/yr (Kalle, 1949), consists of relatively high salinity water from the N. Atlantic entering from the northwest between the Shetland Isles and Norway.

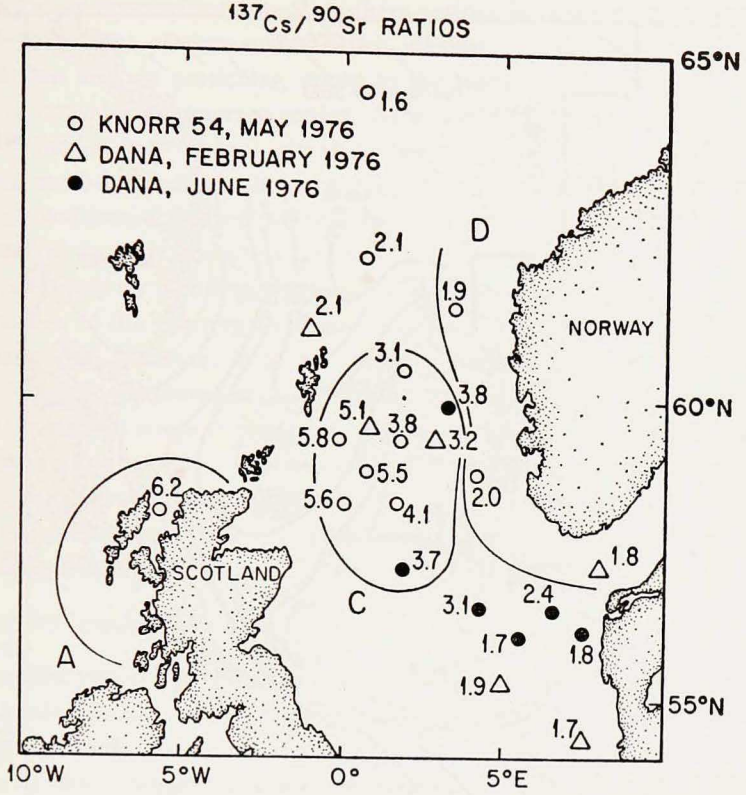

Figure 2. $^{137}Cs/^{90}Sr$ ratios in surface seawater—1976 (cf. Table 1).

Dooley (1974) has concluded the N. Atlantic water inflow to the North Sea north of Shetland is 35,000 km³/year.

Coastal water enters the North Sea around the north coast of Scotland, both as lower salinity Scottish coastal water nearer shore and as a higher salinity mixed water mass flowing between the Orkneys and the Shetlands. The latter stream firstly flows southeastward and subsequently eastward along about 57N, then leaves the North Sea to the north, west of the Norwegian coastal current. The former passes southward along the east coast of Britain and mixes counterclockwise at various latitudes through the central and southern regions of the North Sea.

The southern North Sea receives water of salinity somewhat below oceanic values from the eastern English Channel. The resulting current continues northward along the continental coast becoming less saline as it receives precipitation and river input. After following the coastline into the Skagerrak, this stream exits, with the addition of a relatively small amount (compared to the North Atlantic inflow) of fresher water from the Baltic, and continues northward along the coast of Norway

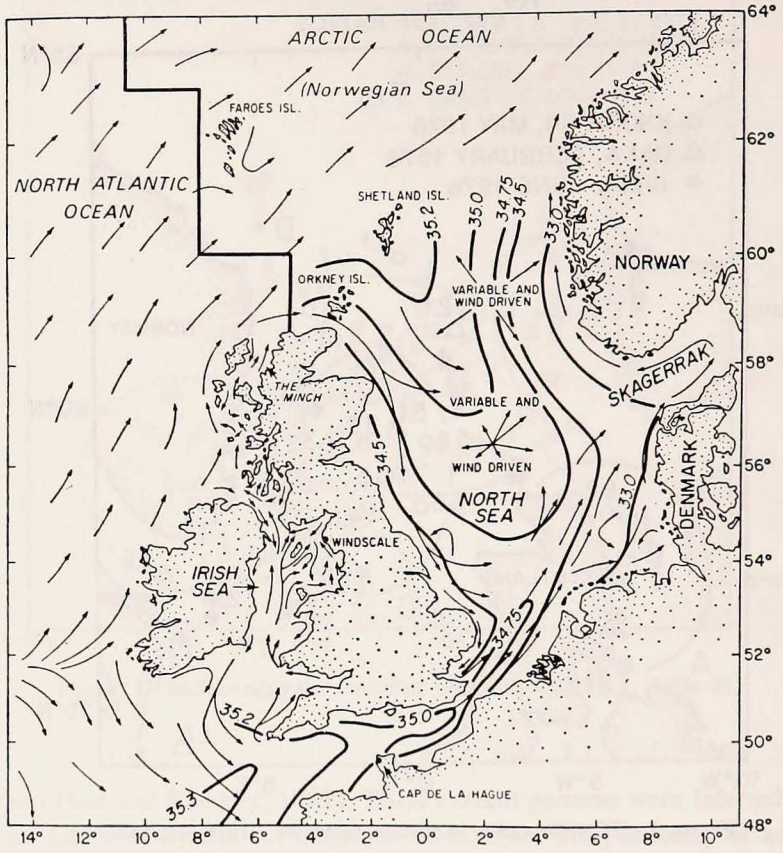

Figure 3. Mean salinities and residual surface circulation patterns in oceanic areas receiving European nuclear fuel reprocessing wastes, cf. Livingston *et al.* (1982).

as the Norwegian coastal current. North Sea near-coastal salinities are lower than those in the north and central regions due to the addition of 145 km³/year of river runoff (Kalle, 1949).

On the basis of information available to Kalle (Kalle, 1949), the total input of water to the North Sea is 25,000 km³/year or just over 45% of its volume. McCave (1973), relying on more recent estimates of inflow through the English Channel, determined an annual input of 31,000 km³/year. Both these estimates imply a turnover time of about two years for the water in the North Sea, the inflows balanced by the northward outflow along the coast of Norway (Fig. 3 and Laevastu, 1963). A still more recent review of the North Sea water budget (Lee, 1980) suggests that the turnover time for the North Sea could be as short as about one year, primarily because of the 35,000 km³/year North Atlantic inflow estimate (Dooley, 1974). These overall turnover times combine means of northern regions having

shorter turnover times with those of southern regions having longer turnover times.

The development of time-scales which characterize the detailed circulation of the North Sea and, in particular, apply to the transit of Windscale-derived radionuclides, is considerably more complex. As noted above, the inflowing water in the northwest divides into several streams, one short-circuiting across the northern North Sea, the others moving into the central and southern sections before turning east to join northward flowing water eventually leaving the North Sea in the Norwegian coastal current. The short-circuiting stream, being of relatively high salinity, could be expected to travel to the west of, perhaps sinking underneath, the lower salinity waters of the Norwegian coastal current. Turnover times are of very little relevance in this situation; it is our primary purpose to show how, with these detailed circulation patterns as a framework for the analysis, available radiotracer data can be used to confirm particular pathways of circulation, and to support the estimation of rates of flow. The conclusion that turnover times for the North Sea are of little relevance to considerations of North Sea flushing has been reached by others (Otto, 1981) who also have suggested that the estimates of turnover times in the various North Sea compartments may be more relevant.

b. Uses of Windscale tracer data

In an earlier report (Livingston *et al.*, 1982) we have summarized the history of the Windscale nuclear waste outflow into the Irish Sea, calling attention to the properties that make this waste stream uniquely valuable as a tracer experiment. It may be as well to supplement that discussion with a brief exposition, here, of the uses we are to make of Windscale tracer data.

1. Surface water data as tracers for whole water column. Most of the available data concerning tracer concentrations and ratios in the Irish Sea-North Sea system derive from measurements of surface water samples. We use these data to infer circulation patterns and their time scales, as well as the fate of the Windscale tracers leaving the North Sea. It is important first, however, to inquire how far these surface water data represent the water columns under investigation. Kautsky (1973) and Kautsky *et al.* (1980) reported that ^{137}Cs distributions in the North Sea were generally homogeneous in the vertical; he observed inhomogeneity only in summer stations about the central and northern North Sea, but at all stations in the Skagerrak. The vertical distribution in the southern North Sea was found to be largely homogeneous throughout the year—in contrast to the Norwegian Deep where permanent stratification of nuclide concentrations was noted. In May 1976, we noted reasonable homogeneity for several nuclides, or for their ratios, at four stations off N. Scotland (stations 19, 24, 32 and 84 in Table 1). At these stations the surface concentrations of ^{137}Cs were within a factor of 1.3 of the maximum or minimum concentrations found at any depth, while $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ ratios showed

Table 3. Annual $^{137}Cs/^{90}Sr$ and $^{134}Cs/^{137}Cs$ Ratios in Discharges from Windscale*.

Year	$^{137}Cs/^{90}Sr$	$^{134}Cs/^{137}Cs$
1957-1968	1.6 (Mean)	N. A.
1969	4.8	0.06
1970	4.8	0.23
1971	2.9	0.18
1972	2.3	0.17
1973	2.7	0.22
1974	11.1	0.25
1975	10.8	0.21
1976	11.1	0.17
1977	10.5	0.13
1978	6.8	0.099
1979	10.2	0.092

* = After Livingston *et al.*, 1982
N. A. = Not available

substantially less vertical variation. At stations 30A and 50, in the Norwegian Channel, the subsurface samples down to depths of 300 m had salinities which seem to indicate influence of the southward flowing shelf-edge current described by Dooley (1974), in which salty oceanic water flows toward the Skagerrak under the northward moving brackish coastal water. The surface ^{137}Cs concentrations at these stations were nevertheless no more than about twice those found down to 200 m. The ratios of $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ reveal trends increasing with depth by factors up to 2.1, which is clearly indicative of the vertical structure in those water columns, with surface and deep water carrying nuclide ratios characteristic of labelling from releases of differing ages. The distributions of water column tracer properties are sufficiently similar as far "downstream" from the Irish Sea as the North Sea outflow to persuade us that surface water data are useful for tracing the movement of a substantial portion of the water column at least this far.

2. *Rates of movement from tracer ratios.* Comparison of the ratios ^{137}Cs to ^{90}Sr, and ^{134}Cs to ^{137}Cs, in Windscale effluent with those measured in waters flowing into, within, and out of the North Sea might be expected (Livingston and Bowen, 1977) to provide insight into the rates at which labelled Irish Sea water moves through this region. As noted by Livingston *et al.* (1982) the ratio of ^{137}Cs to ^{90}Sr in Windscale effluent increased radically in 1969 and again, even more radically, in 1974; since the two nuclides decay with similar half lives, these changes in their ratios should serve as good event markers. The ratio ^{134}Cs to ^{137}Cs in the discharges was relatively uniform until 1973 (Table 3) but since has been declining steadily from a maximum of 0.25 in 1973 to 0.092 in 1979. The half life of ^{134}Cs is only about one-fifteenth that of ^{137}Cs, so this ratio can provide a useful elapsed time indicator

(Livingston *et al.*, 1982). Calculations of time passage from such radioisotope ratios are very sensitive to estimates of the holdup time and mixing within the Irish Sea. As mentioned previously, the average transport time from the Windscale pipeline to entry into the North Sea appears to be about two years.

3. Nuclide ratio changes during dispersion. The ratio ^{137}Cs to ^{90}Sr in these samples is relatively insensitive to factors other than source term variation. As will become clear from later discussion, mixing during transit from Windscale to waters north of Scotland (Region B of Fig. 1) only reduced the 1976 discharge ratio of between 10 and 11 to the observed average of 7.5 in 1978. For samples with ^{90}Sr concentrations less than 0.25 pCi/1, a value roughly four times that expected from fallout alone, the effect of the fallout component (Tables 1 and 2) was to cause these ratios to be less than would have been observed in the absence of fallout. The fraction of water column Cs radionuclides transferred to coastal sediments (estimated at $< 3.5\%$ for the Windscale stream, 5-6% for fallout derived ^{137}Cs (Livingston and Bowen, 1977)) is insufficient to lower the discharged $^{137}Cs/^{90}Sr$ ratio substantially in the course of the dispersion of the discharges in the coastal circulation. As discussed by Livingston *et al.* (1982) change in the ratio $^{134}Cs/^{137}Cs$ occurs chiefly as a result of radioactive decay of ^{134}Cs, which has a half life (2.06 years) short compared with that of ^{137}Cs (30 years). Fallout-derived ^{137}Cs, essentially free of ^{134}Cs, is certainly at sufficiently low concentrations (estimated at < 0.1 pCi/kg) in these waters not to affect the Windscale-derived ratio to a significant extent.

The question of nuclide ratio changes resulting from mixing of older labelled water with younger water, of differing ratios, is not easily answered. It is our view that the patterns of nuclide ratios in the discharges and in the dispersal stream, by location and by time, are more consistent with an interpretation which suggests that *extensive* horizontal mixing is not an important process over many of the areas traversed by the waste stream. We would expect that the record in the dispersal plume of short-term fluctuations in release rates taking place over a few months would be erased or sharply damped by short-term mixing by tidal, wind-driven, and density currents. But features in release rate variations that persist over longer time-periods appear to be observable all along the advecting plume.

4. Use of salinity normalized data. West and North Scottish coastal waters (regions A and B in Fig. 1) connect the Irish and North Seas. They may be viewed as a system wherein northerly flowing Irish Sea water is mixing with higher salinity North Atlantic water. In a steady state situation the salinity in the mixture should correlate inversely with the concentration of the radionuclide being diluted. In August 1978 there are good indications that steady state was being approximated. Figure 4 shows the correlation between salinity and ^{137}Cs concentration in coastal

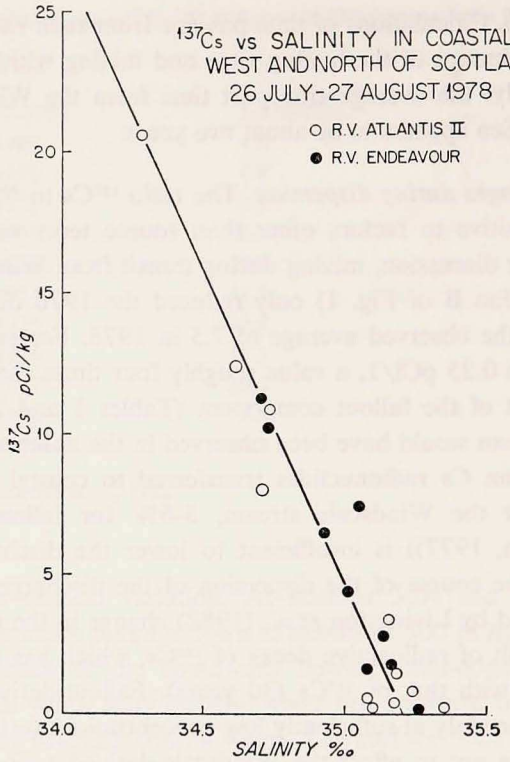

Figure 4. Dilution of ^{137}Cs labelled Irish seawater in Scottish coastal water by North Atlantic water.

water west and north of Scotland during that month. Similarly strong ^{137}Cs and salinity inverse correlations can be seen in data (McKinley *et al.*, 1981a) from measurements in the sea of the Hebrides in 1976 through 1977. These correlations encouraged us to construct a salinity-normalized history of the changes with time in the concentrations of ^{137}Cs and ^{90}Sr in these Scottish coastal waters and to compare this history with data for these radionuclides as released from Windscale. The upper half of Figure 5 shows the annual amounts and ratios of ^{137}Cs and ^{90}Sr discharged in the years 1966 through 1978 (Livingston *et al.*, 1982), while the lower half shows the corresponding response in these Scottish coastal waters (normalized to a salinity of 34.5‰—an intermediate value between Irish Sea outflow water salinities and the higher values of the offshore North Atlantic water). In addition to data from Tables 1 and 2, this figure uses data from the Danish surveys of 1972, 1973, and 1975 (Aarkrog *et al.*, 1973, 1974, 1976). Salinity normalized data will be used later to address questions of rates of water movement along certain parts of the dispersal pathway.

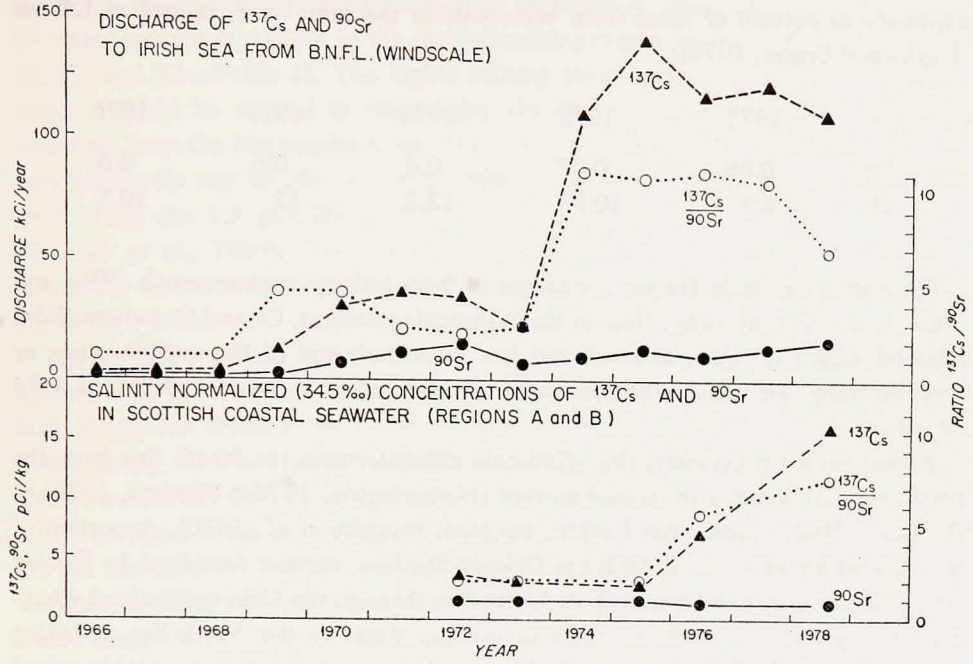

Figure 5. ^{137}Cs, ^{90}Sr, and their ratio in annual releases to Irish Sea from Windscale compared with resultant concentrations and ratios of these nuclides in Scottish coastal seawater. (The lines connecting the points in the lower half of the figure are meant to draw the reader's eye to the data and are not meant to give an indication of *continuous* temporal trends. For example, ^{137}Cs concentrations, as referenced in the text, passed through a maximum in early 1977.)

c. Pattern of tracer movement to and through the North Sea

The pattern of movement of soluble radioisotopes to and through the North Sea may be inferred from ^{137}Cs concentrations measured from the British and German surveys in the area (Hetherington, 1976a; Mitchell, 1977a,b; Kautsky, 1973, 1976, 1977; Kautsky *et al.*, 1980; Deutsches Hydrographisches Institut, undated; Murray and Kautsky, 1977). Some uncertainty in the interpretation of such data is produced by the consistent inadequacy of documentation of the discharges from Cap de la Hague. As we noted earlier, the evidence available indicates the latter source to be negligible, except very locally in the Channel and southern North Sea, compared to Windscale. In 1972 and 1973, the earliest years for which data have been published (UNSCEAR, 1977), Cap de la Hague discharged 1,200 and 2,000 curies, respectively, of ^{137}Cs, at ratios of $^{137}Cs/^{90}Sr$ of 1.38 and 1.96. In 1974, 1975 and 1976, the ^{137}Cs discharges were 1,513, 931 and 939 curies respectively (Ancellin and Bovard, 1980). Gross alpha and gross beta releases from Cap de la Hague,

expressed as percent of those from Windscale in the same year, ranged as follows (Luykx and Fraser, 1978):

	1972	1973	1974	1975	1976
α	0.08	0.07	0.6	0.6	0.6
β	8.3	10.8	12.2	13	10.5

Since the Cap de la Hague discharges of beta activity contain much ^{106}Ru and ^{144}Ce (relatively both richer than in the Windscale releases), Cs and Sr radionuclides released must have been at levels too low to perturb any of the nuclide ratios or concentrations on which our discussion of circulation is based, except as mild diluents.

As mentioned previously, the Windscale effluent enters the North Sea from the northwest via the Scottish coastal current (Hetherington, 1976a; Mitchell, 1977a,b; Deutsches Hydrographisches Institut, undated; Kautsky *et al.*, 1980). A portion of this current appears to mix with the Orkney-Shetland current described by Dooley (1974) as entering the northwestern North Sea through the Orkney-Shetland Channel. Dooley also describes the entry of oceanic water to the North Sea as taking place to the north of the Shetland Is. If the entering waters were thoroughly mixed throughout the volume of the North Sea, their turnover time would be that inferred above from the total water inflow, less than two years. The patterns of circulation shown by the surveys of ^{137}Cs concentration in the North Sea during 1973-1976 (Kautsky *et al.*, 1980), especially the German surveys of 1973 (Kautsky, 1973; Deutsches Hydrographisches Institut, 1974), and the German and the British surveys of 1976 (Deutsches Hydrographisches Institut, undated; Mitchell, 1977b) strongly suggest that the system is partially short-circuited in respect to the Orkney-Shetland current inflow since the Windscale ^{137}Cs in it appears never to mix throughout the North Sea (Mauchline, 1980). Instead most of it appears to join the northward moving outflow after only a brief residence largely north of 56N and (along this parallel) west of about 3E (although this boundary changes with summer/winter movement of surface salinity contours). This is also Kautsky's conclusion (Kautsky, 1977, Fig. 2; Kautsky *et al.*, 1980) and is in general agreement with the pattern of residual surface currents shown in Figure 3. It should be noted, furthermore, that the short-circuiting through the northern North Sea apparently requires that the residence time of water in the southern half of the North Sea must be considerably in excess of the total North Sea turnover time of two years.

Kautsky *et al.* (1980) noted that the fate of this water mass was not completely clear. We would argue that the pattern of increasing ^{137}Cs/^{90}Sr ratios with increasing depth and salinity found in May 1976 at Station 30A in the Norwegian Channel (Table 1) gives some clues to the fate of this body of water. The high salinity and

$^{137}Cs/^{90}Sr$ ratio found at depth seem to be relatable to those found over much of the water column at stations in the short-circuiting region to the southwest (Stations 19, 24, and 32—Table 1). The higher salinity water mass from the area of these stations would be argued to ride under the less dense water masses extending westward from the Norwegian coast. The locations at which this takes place should vary seasonally and be affected by storms, but may have been defined in summer 1976 along the 1.8 pCi/liter ^{137}Cs contour of the German survey of that year (Kautsky *et al.*, 1980). This water would then be transported by the southerly subsurface flow towards the Skagerrak (Lee, 1980) prior to its joining the North Sea outflow from there.

The time framework implied by the various isotope ratio data for Windscale nuclide-labelled water moving to and through the North Sea is also consistent with both North Sea flushing times and patterns of short-circuiting referred to above. Conceptually it would seem helpful to view the dispersal of Windscale-derived soluble radionuclides in this system in four distinct regions, defined in terms of residence times of the Windscale stream in each region. It should be noted that they are not the same regions defined in Figures 1 and 2 and that it was not considered worthwhile in this context to subdivide these regions further as has been done elsewhere (Lee, 1980; Clark and Webb, 1981). They are: first, the Irish Sea—where a turnover time of something less than two years would seem to be indicated, second, coastal water from southwest around to northeast Scotland—for which relatively rapid transport times of the order of months seem indicated, third, the North Sea—which, as indicated above, may have an overall turnover time of one to two years, but with considerable short-circuiting in the northern section, and fourth, the Norwegian coastal current—which is fed by the outflow from the North Sea and in which one would expect transport to be relatively rapid.

d. Average time of transport from Windscale to Scottish coastal water

1. From $^{137}Cs/^{90}Sr$ data. In respect of the discharge history, it should be noted from Figure 5 that the rate of ^{90}Sr discharge has remained relatively steady since the early 1970's while that of ^{137}Cs peaked in 1970 through 1972 and again (much larger) in 1974 through 1978. The $^{137}Cs/^{90}Sr$ ratios characterizing the releases did not exceed 5 until 1974, then rose to and stayed around 11 during 1974 through 1977. In these Scottish coastal waters at salinity about 34.5‰, ^{90}Sr concentrations can be seen to have remained relatively constant during 1972 through 1978 whereas corresponding data for ^{137}Cs, relatively constant during 1972 through 1975 (although we believe the higher ^{137}Cs reported for the Pentland Firth in October 1974 to be a real departure from this constancy (Kautsky *et al.*, 1980)), show sharp increases in 1976 and higher still by 1978. The ratio $^{137}Cs/^{90}Sr$ did not exceed 5 in Scottish coastal seawater until 1976. These data seem to confirm the arguments (Livingston and Bowen, 1977) that pointed to an average time of transport of about

Table 4. $^{137}Cs/^{90}Sr$ and $^{134}Cs/^{137}Cs$ ratios in European coastal water masses in 1976 and 1978+.

	$^{137}Cs/^{90}Sr$*		$^{134}Cs/^{137}Cs$*	
Water mass	1976	1978	1976	1978
West Scotland	6.2	5.8 – 8.6	0.13	0.08 – 0.10
(region A)	6.2	6.8 ± 1.0	0.13	0.093 ± 0.008
	(1)	(6)	(1)	(10)
North Scotland	—	6.8 – 8.1	—	0.073 – 0.098
(region B)		7.5 ± 0.5		0.085 ± 0.007
		(7)		(11)
N. North Sea	3.1 – 5.8	6.5 – 8.4	0.12 – 0.15	0.084 – 0.091
(region C)	4.3 ± 1.0	7.7 ± 0.7	0.13 ± 0.02	0.089 ± 0.003
	(10)	(5)	(5)	(5)
Norwegian coastal	1.8 – 2.0	3.3 – 4.7	0.06 – 0.08	0.05 – 0.12
current	1.9 ± 0.1	4.1 ± 0.4	0.07 ± 0.01	0.079 ± 0.021
(region D)	(3)	(10)	(2)	(9)
				0.056 – 0.066**
				0.064 ± 0.005
				(4)

+ Regions as in Figures 1 and 2
* Range
Mean ± 1σ
(Number of samples)
** Cs collected on copper ferrocyanide absorbers

two years for water in the Windscale area to reach N.W. Scottish coastal areas, with most of the hold-up being, as Jefferies suggested (Jefferies *et al.*, 1973; Jefferies, 1977), in the Irish Sea. From comparisons of mean annual ^{137}Cs releases from Windscale with Aberdeen coastal water concentrations of ^{137}Cs, Mauchline (1980) has also concluded that the Windscale effluent takes two years to travel to the North Sea region off Aberdeen on the Scottish east coast.

2. *From $^{134}Cs/^{137}Cs$ data.* This transport time is also derivable from comparisons of the $^{134}Cs/^{137}Cs$ ratios in these coastal waters in 1976 and 1978 with their ratios in the releases from Windscale. Table 4 lists for 1976 and 1978 the ranges, means, standard deviations and number of samples represented, of surface water $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ ratios in the regions around Scotland and in the Norwegian coastal current relative to sampling positions in 1976 and 1978 depicted in Figures 1 and 2. The ratios of $^{134}Cs/^{137}Cs$ in the discharges from 1974 through 1976 declined significantly, namely 0.25, 0.21, and 0.17 (Table 3). After two years these ratios would be reduced by decay to 0.13, 0.11, and 0.087, respectively. Paralleling this, but offset by two years, the mean ratio in Scottish coastal water of regions A, B, and C, declined from 0.13 in 1976 to 0.089 in 1978. This change fits exactly

that which would result from decay in a little less than two years during transport from the discharge area to these Scottish coastal waters; i.e., the 1974 discharge ratio of 0.25 being reduced by decay during transit to the value of 0.13 found in 1976 in Scottish coastal water and, likewise, the 1976 discharge ratio of 0.17 being similarly reduced to 0.089 in 1978. The $^{134}Cs/^{137}Cs$ ratios in Scottish coastal water in 1976 are very close to those measured in the same year in the sea area of the River Clyde estuary viz. 0.14 (Baxter *et al.*, 1979). Likewise, the ratios reported there in 1977 (Baxter *et al.*, 1979), averaging 0.11 can be viewed as resulting from about two years decay of the 1975 discharge ratio of 0.21. Baxter *et al.* concluded, on the basis of a box-model treatment of their data, that water transit times from the Windscale area to the Clyde are in the range 8 ± 3 months. In a subsequent report (McKinley *et al.*, 1981b), considerable mixing in the Irish Sea of Windscale releases is suggested and water movement to the North Channel from Windscale is modelled as being characterized by a residence half-time of 12 months and a transit time of six months. This model approach was adopted in place of earlier approaches to transport which did not involve large area mixing and which were strongly advective in nature. The need for the model arose from the difficulties in matching monthly ^{137}Cs releases from Windscale during 1972-77 with North Channel and Clyde Sea area ^{137}Cs concentrations during the period 1974-77. The monthly data can, for the following reasons, be deceptive in making a comparison:

a) McKinley *et al.* (1981b) note, for example, that there is a "decreasing trend in output during 1974 to 1977." The monthly output data appear to follow such a trend but the annual releases were in fact 108, 141, 118, and 121 kilocuries respectively for that period—which follow a fairly uniform annual release (excepting the 1975 maximum). These data are referenced in our earlier paper (Livingston *et al.*, 1982) or are derivable (in relative units) from McKinley *et al.* (1981b).

b) The time series of ^{137}Cs concentrations in North Channel or Clyde Sea area waters are subject to annual and seasonal fluctuations due to processes which affect the degree of dilution during transport. These include, for example, in the North Channel, the proportion of Atlantic water which is mixed with exiting lower salinity Irish Sea water at any given time.

The processes of mixing and transport of Windscale tracers during their movement out of the Irish Sea are no doubt more complex and variable than are described by either a single or multi-box model approach or a purely advective approach with only localized mixing during transport. In any approach we believe that an addition can profitably be made to the direct matching of release rates with downstream isotope concentrations—even when appropriate concentration normalization techniques have been applied. The addition of isotope ratio comparison, such as $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$, between releases and along the dispersion path-

way, largely eliminates variable dilution effects which bring substantial changes in isotope concentrations.

In the context of the difficulties in matching release rate change patterns with downstream isotope concentration change patterns, it has become evident that some modification is required to earlier remarks (Livingston *et al.*, 1982) which were made in regard to the matching of Windscale ^{137}Cs release data with concentrations of this isotope in Clyde Sea waters reported by Baxter *et al.* (1979). We accept that the *intensity* of the maximum they observed in spring 1977 was as a consequence of the unusual low salinity water outflow from the Irish Sea at that time (which they later described (McKinley *et al.*, 1981a)). However, annualized averages of their data, even excluding the anomalous perturbation affect of the low salinity outflow, still show a maximum which we hold to be highly influenced by the Windscale ^{137}Cs release maximum of 141 kilocuries in 1975. This was larger than those of 1974 and 1976 by 31% and 19%, respectively.

Comparison of the $^{134}Cs/^{137}Cs$ ratios measured in both 1976 and 1978 around the coast of Scotland (Regions A-C of Table 4) allows conclusions to be made about the rate of water transport along this pathway. The constancy of $^{134}Cs/^{137}Cs$ ratios from West Scotland around to the northern North Sea in both years could be used to argue that travel around Scotland into the North Sea is relatively fast. However, the pattern of change in ratio $^{134}Cs/^{137}Cs$ in Clyde Sea area water between 1975 and 1978 (Baxter *et al.*, 1979) is very close to ratio half-life change through decay of 2.2 years. Accordingly, this is a more substantial factor creating constancy in the ratio at any point in time around the Scottish coast.

e. Windscale stream dilution in Scottish coastal water

The various transport times which apply to water movement through the system are critical with respect to estimates of the extent of discharge dilution at the various points downstream from the Irish Sea. Preston *et al.* (1978), presumably using the results of 1976 surveys, estimated that mixing between the North Channel and Cape Wrath (region A of Fig. 1 essentially) reduced surface water ^{137}Cs concentrations by factors of about 5 to 10. It is clear from Figure 5 and from other data (Baxter *et al.*, 1979; Kautsky *et al.*, 1980) that ^{137}Cs concentrations in this water mass were rising sharply in 1976 in response to the large increase in release rate that had occurred two years previously. The concentration gradients which resulted along the circulation pathway complicate any estimation of dilution factors as the system was far from steady state with respect to ^{137}Cs release and dispersion. As we point out above, in 1978 the coastal water around Scotland was closer to steady state because the annual releases from 1974 through 1976 were more uniform than before and, by 1978, created a more uniform supply of ^{137}Cs to Scottish coastal water. Comparison of ^{137}Cs concentrations (Table 2) at positions close to

the northern exit of the Irish Sea, with those off the north coast of Scotland, would support substantially lower estimates of dilution factors between these areas, probably only half those Preston *et al.* (1978) suggested. Similarly, comparison of ^{90}Sr concentrations between these two areas would indicate dilution of this nuclide, one discharged in amounts varying much less annually than those of ^{137}Cs, by around a factor of 3. This degree of dilution agrees with the estimate of 2-5 reported by Mauchline (1980). The further dilution by a factor of 2 which Mauchline suggests occurs during transport from Cape Wrath to the Pentland Firth is in contrast to the little variation of ^{137}Cs and ^{90}Sr concentrations which Kautsky reported in this area from his 1978 survey (Deutsches Hydrographisches Institut, 1981). Dilution effects are almost certain to fluctuate locally with seasonal and weather dependent factors.

f. North Sea circulation rates from inflow and outflow isotope ratio changes

Comparisons of the $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ ratios measured in the Norwegian coastal current in 1976 and 1978 with those in North Sea inflow water in these years (Table 4) and with those measured by the Danes in the southern North Sea in 1977 and 1978 (Aarkrog *et al.*, 1978, 1979) provide some information on the passage of Windscale-labelled waters through the central and southern North Sea. The uniform and high $^{137}Cs/^{90}Sr$ ratios (averaging 4.1) which characterized the Norwegian coastal current in 1978, especially as found in those samples collected by R.V. *Endeavour* on the E-W section along 60N (Fig. 1 and Table 2), are persuasive that, by that time, high Cs isotope discharges from Windscale dominated and controlled the composition of the North Sea outflow. Even one year earlier, surface water measurements of ^{137}Cs and ^{90}Sr in the outflow and the Skagerrak (Deutsches Hydrographisches Institut, 1980) were still indicating $^{137}Cs/^{90}Sr$ ratios about 2. The lesser impact of the Cap de la Hague releases is suggested by the comparative discharge history of la Hague and Windscale (Luykx and Fraser, 1978), the low rates of ^{137}Cs released by Cap de la Hague from 1972 to 1976 referred to earlier (UNSCEAR, 1977; Ancellin and Bovard, 1980) and by the 1975 and 1976 surveys of ^{137}Cs concentrations in the English Channel and southern North Sea (Mitchell, 1977b; Murray and Kautsky, 1977; Kautsky *et al.*, 1980). Also, $^{137}Cs/^{90}Sr$ ratios measured in water collected off the German North Sea coast by the German lightship F.S. "P8" during 1974 through 1978 did not exceed 2 (Deutsches Hydrographisches Institut, 1981). As this location, 54°16′06″N, 7°11′24″E, would be traversed by water which would, at least in part, have come through the English Channel bearing la Hague-derived radionuclides, Cs radionuclides released by la Hague can be argued to be no more than twice the published ^{90}Sr releases (Luykx and Fraser, 1978) during this time interval. Accordingly, the contrast between the considerable difference found in the $^{134}Cs/^{137}Cs$ ratios of the North Sea inflow water and Norwegian coastal current in 1976 (0.13 and 0.07 and the closer values

of 0.09 and 0.064 found in 1978) is easier to understand. (It is worth noting, parenthetically, that our 1978 Norwegian coastal current ratios, derived from measurements made of Cs isotopes collected by copper ferrocyanide chemisorption from several hundred liters of water, are the most precise from that region (Table 4).) As stated above, the $^{134}Cs/^{137}Cs$ discharge ratio from Windscale in 1974 through 1976 declined from 0.25 to 0.17. Accordingly, the "younger" (in terms of time of labelling with Windscale radioactivity) water in the North Sea inflow was being characterized by increasingly lower ratios $^{134}Cs/^{137}Cs$. The outflow water in the Norwegian coastal current, representing water which has been held up in the North Sea circulation, was substantially "older." So the outflow water ratio in 1978 still reflected considerably earlier, e.g., 1976, inflow ratios, reduced by decay in the North Sea circulation. It is tempting to relate the inflow ratios in 1976 (0.13) to the outflow ratios in 1978 (0.072) and attribute the difference to decay by just under two years within the North Sea. This really cannot result from longer decay of water labelled with higher $^{134}Cs/^{137}Cs$ ratio because the 1976 inflow water was reflecting the 1974 peak (0.25) in the discharge ratio. As longer times of circulation of water traversing the central and southern North Sea would be necessary to compensate for the short-circuiting in the northern section and yet produce the older estimate of two years for the turnover time for the whole North Sea, these data appear to support the more recent (Lee, 1980) estimate of about one year. Certainly the $^{137}Cs/^{90}Sr$ and $^{134}Cs/^{137}Cs$ ratios measured in the central and southern North Sea by the Danes in 1977 and 1978 (Aarkrog *et al.*, 1978, 1979) do not provide evidence of sluggish circulation and longer turnover times.

A similar North Sea counterclockwise circulation transport time of two years can be inferred from measurements of ^{137}Cs concentrations in inner Danish waters from 1972-1980 (Aarkrog *et al.*, 1981). Their conclusion is that the peak ^{137}Cs concentration found in 1979 corresponds to the peak release in 1975, a transport time of four years. If, as argued earlier, transport to the inflow to the North Sea takes two years, their arguments imply counterclockwise transport around the perimeter of the North Sea of a further two years.

g. Plutonium from Windscale in Scottish coastal waters

1. Transport with soluble nuclides. Although the coastal circulation system around the U. K. has been studied more intensively in regard to its transport of those radionuclides which remain primarily in true solution, it is also the vehicle which transports and disperses reactive elements whose form after release, or after undergoing biogeochemical change subsequent to discharge, is such as to maintain them in solution to some extent. Such an element is Pu which is largely immobilized in sediments, mostly near to the point of discharge (Hetherington, 1976b; Hetherington *et al.*, 1976), but which has been shown to be moving with the soluble

radionuclides in the coastal circulation to a limited extent (Livingston and Bowen, 1977; Hetherington, 1976b; Murray *et al.*, 1978). Nelson and Lovett (1978 and 1981) proposed that such transport is mostly of Pu in a higher oxidation state, ^{V}Pu and/or ^{VI}Pu. Although the data are presently somewhat limited, it seemed worthwhile to examine the changes with time of Pu from Windscale which has been transported with the soluble nuclides outside of the Irish Sea. Windscale-derived Pu is readily distinguishable from weapons-test fallout Pu by its high ratio ^{238}Pu/^{239,240}Pu (Livingston and Bowen, 1977).

2. Pu concentration normalized in Scottish coastal water. As was demonstrated earlier for comparing ^{137}Cs and ^{90}Sr concentration changes with time in Scottish coastal water, a normalization technique helps to factor out mixing and dilution effects.

In Scottish coastal waters sampled during the years 1975 through 1978, there have been a substantial number of measurements of ^{239,240}Pu, both as shown in Tables 1 and 2 in this work and as reported by Murray *et al.* (1977, 1978) and by the Deutsches Hydrogr. Inst. (Deutsches Hydrogr. Inst., 1980, 1981). It was not possible to combine these data using normalization to salinity as no salinity data were given by Murray *et al.* or the Deutsches Hydrogr. Inst. Normalization to ^{137}Cs was not useful as ^{137}Cs concentrations were changing dramatically during this period (Fig. 5). Strontium-90 concentrations were, however, relatively constant when salinity normalized (Fig. 5). Accordingly, ^{239,240}Pu concentrations were normalized to those of ^{90}Sr from data in Tables 1 and 2 and, likewise, from Murray *et al.* (1977, 1978) and the Deutsches Hydrogr. Inst. (1980, 1981). It was necessary to convert the data of Murray *et al.* from a ^{137}Cs-normalized base, as they did not report ^{90}Sr but did report ^{137}Cs concentrations. Conversion was made using ^{137}Cs/^{90}Sr ratios from Table 4 or from the Danish report for 1975 (Aarkrog *et al.*, 1976) as appropriate to the time and place of sampling. The ratio for the single 1975 (February-March cruise) measurement reported from the data of Murray *et al.* (1977) does not correspond to the published data. It is, however, clear from that paper that the reported ratio of ^{239,240}Pu/^{137}Cs of 0.20% disagrees with the value of 2% derived from the actual ^{239,240}Pu and ^{137}Cs concentrations reported. We assume that the ratio was incorrectly reported; i.e., that the reported concentrations were correct.

The results of this comparison are shown in Table 5. It is hard to discern a distinct pattern of ratio change with time, either seasonally or annually. In fact, during 1976 through 1978, the ratio was remarkably constant between 2×10^{-3} and 3×10^{-3}. The 1975 value, 5×10^{-3}, seems significantly higher than those of 1976 through 1978 and although it is the only ratio measurement reported for that year, similar Pu concentrations were found later that year further south along the Scottish East Coast on a subsequent cruise. No ^{137}Cs data were reported so normalization to ^{137}Cs or ^{90}Sr was not possible.

Table 5. Variation in the ratio $^{239,240}Pu/^{90}Sr$ in Scottish coastal water*—1975-1978.

Month, year	Ratio $^{239,240}Pu/^{90}Sr$+	Reference
Feb., March 1975	5.0×10^{-3} (1)	Murray, Kautsky, 1977
May 1976	$2.7 \times 10^{-3} \pm 0.1 \times 10^{-3}$ (4)	Livingston, Bowen, 1977
July-Sept. 1976	$1.9 \times 10^{-3} \pm 0.3 \times 10^{-3}$ (6)	Murray *et al.*, 1978
March, April 1977	$2.0 \times 10^{-3} \pm 0.7 \times 10^{-3}$ (6)	Deutsches Hydrographisches Institut, 1980
February 1978	$2.1 \times 10^{-3} \pm 0.5 \times 10^{-3}$ (11)	Deutsches Hydrographisches Institut, 1980
April 1978	$2.9 \times 10^{-3} \pm 0.3 \times 10^{-3}$ (4)	This work
August 1978	$2.5 \times 10^{-3} \pm 0.8 \times 10^{-3}$ (8)	This work

* Coastal water flowing into the North Sea — Regions A and B of Figure 1
+ Mean $\pm$ 1σ (Number of samples)

We agree with the suggestion (Mauchline, 1980) that these elevated concentrations may have reflected the 1973 maximum in Windscale release, following about a two-year transport time. The uniformity of salinity-normalized ^{90}Sr concentrations in these waters during 1972 through 1978 has been noted earlier. Accordingly, the very uniform ratios $^{239,240}Pu/^{90}Sr$ shown in Table 5 for Scottish coastal waters during 1976 through 1978 can be viewed as reflecting the uniformity of their annual release rates from 1974 through 1976. Pu releases, for example were not outside the range 1,200-1,272 curies/year (Livingston *et al.*, 1982). Although some of the plutonium travelling in the Windscale plume has been transferred to underlying sediments (Livingston and Bowen, 1977) the plutonium concentration of the water column would appear to be principally controlled by the release pattern to the Irish Sea. Conversely, Scottish coastal water Pu concentrations are not controlled at the present time by remobilization of Windscale plutonium accumulating in Irish Sea sediments. If the discharges are substantially reduced following the commencement of new reprocessing technology in the 1980's, it will be of considerable interest geochemically to follow the water column concentration changes that ensue.

4. Conclusions

1) The concentrations of Windscale-derived radionuclides measured in surface water flowing to and through the North Sea in 1976 and 1978 support the patterns of movement of these tracers in this region inferred from other published ^{137}Cs surveys. The chief features in this pattern are the passage of some of the Windscale tracers through the northern North Sea and the counterclockwise passage of another fraction down the U. K. east coast and through the southern North Sea.

2) Within defined regions, use of salinity-normalized concentrations of Windscale radionuclides can factor out dilution and mixing effects and provide a better basis for comparison of concentration changes as a function of time.

3) Transport times of Windscale tracers from the Irish Sea to the North Sea have been derived from comparison of the measured ratios $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ in 1976 and 1978 surface Scottish coastal and North Sea water with those reported in their releases. Windscale tracers appear to enter the Scottish coastal circulation slightly less than two years after their release, but are moved thereafter quite rapidly (on the order of a few months) into the North Sea.

4) Dilution of Windscale nuclides in the main dispersal plume between the Irish Sea outflow and the North Sea inflow, though liable to fluctuate over short periods of time, averages about a factor of 3.

5) Consideration of 1976 and 1978 ratios of $^{134}Cs/^{137}Cs$ and $^{137}Cs/^{90}Sr$ in surface seawater in the North Sea, inflowing Scottish coastal water and outflowing Norwegian coastal water suggest that Windscale tracers traverse the North Sea by different pathways and at different rates. One stream appears to short-circuit the Northern regions spending only a few months there. Another stream moving southward down the East coast of Britain takes about two years to pass through the North Sea to the Norwegian coastal current.

6) The high $^{137}Cs/^{90}Sr$ ratios found in the Norwegian coastal current in 1978 provide evidence that the sharp increases of ^{134}Cs and ^{137}Cs release to the Irish Sea in 1974 and subsequent years had started to control the Cs isotope composition of the North Sea outflow in the surface waters of the Norwegian coastal current in the time interval 1977-78.

7) Changes in the ratios $^{239,240}Pu/^{90}Sr$ measured in Scottish coastal water in the years 1975-78 were compared with changes in the release from Windscale of these radionuclides. The small fraction of the Pu release travelling in Scottish waters reflected its discharge pattern by time. These data show no indications that Scottish coastal water Pu concentrations are increasing as a result of remobilization of Windscale Pu from Irish Sea sediments as the major fraction of released transuranics accumulates there with time.

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