Radioactive tracer studies in the surface waters of the northern North Atlantic including the Greenland, Norwegian and Barents Seas

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ABSTRACT

Discharges of radiocesium, strontium-90 and plutonium from the UK reprocessing plant Sellafield have been used for radioactive tracer studies in the northern North Atlantic including the Greenland, Norwegian, and Barents Seas. Transfer factors for ^{90}Sr, ^{134}Cs and ^{137}Cs from the discharge to various water masses have been calculated.

INTRODUCTION

Since 1974 discharges of radiocesium to the Irish Sea from the British Nuclear Fuels Ltd, Sellafield (formerly Windscale) installation in Cumbria have increased the concentrations of ^{137}Cs and ^{134}Cs in the northern North Atlantic (Jefferies et al. (1982; McKinley et al. 1981; Kautsky and Murray 1981; Livingston et al. 1981; Livingston et al. 1982; Casso and Livingston 1984; Holm et al. 1983; Aarkrog et al. 1983, 1984; Dahlgaard et al. 1984; Aarkrog et al. 1963–1984). Other radionuclides such as ^{90}Sr, ^{99}Tc, Pu-isotopes and ^{241}Am are also present in the effluent (BNFL 1978–1982; Cambray 1982).

The various radionuclides may be used as tracers for waterborne pollution entering the coastal West European waters. In particular, we have dealt with the situation in the Norwegian, Barents, and Greenland Seas. Our studies began in 1980 with the Swedish Ymer expedition to the Barents and Greenland Seas and continued in 1981 with a sampling from Norway to the Faroe Islands and around Iceland. In 1982 samples were collected around the northern UK, between Norway and East Greenland and along the West and East Greenland coasts. In 1983 a cruise along the Norwegian coast and between Spitsbergen and East Greenland was carried out on board the German icebreaker Polarstern. Furthermore, samples were obtained that year from Ireland and along the west coast of Greenland. This paper will give emphasis to the results obtained from the cruise with the Polarstern, in particular those from the Fram Strait.

MATERIALS AND METHODS

Sea water was usually collected with a pump from the ship. The radiocesium was precipitated with ammonium molybdophos-

phate (AMP, Dutton 1970) on board from 200 or 1800 l samples; 100 g AMP was used to precipitate the radiocesium. The average yields were 91% and 62%, respectively. The AMP samples with very low ^{134}Cs levels were dissolved in NaOH and reprecipitated as Cs_2PtCl_6. As explained earlier (Aarkrog et al. 1983) this procedure improved our limit of detection to 5–10 mBq ^{134}Cs m^{-3}. The radiocesium was measured by γ-spectroscopy on 100 cm^3 Ge(Li) detectors. Strontium-90 was determined in the 50 l samples by the fuming nitric acid procedure (Harley 1972) after an initial precipitation of the activity as carbonate. Finally, plutonium and americium were assayed on board the ship on 200 l or 1800 l samples with an initial precipitation as hydroxide with yield tracers of ^{242}Pu and ^{243}Am, followed by the radiochemical analysis developed by Talvitie (1971) and Holm et al. (1979), respectively.

RESULTS

The detailed data of our North Atlantic sea water samplings since 1980 are published elsewhere (Aarkrog et al. 1963–1984). The ^{137}Cs concentrations in North Atlantic surface water have shown nearly the same distribution from year to year during the period of observations (1980–1983). In the North Sea the mean concentration was 95 ± 7 (N = 55, ± 1SE) Bq ^{137}Cs m^{-3}; in the Norwegian Coastal Current we found 84 ± 6 (N = 34, ± 1SE) Bq m^{-3}. Between Norway and Spitsbergen (Svalbard) in the Barents Sea the mean concentration was 19 ± 3 (N = 14, ± 1SE) Bq m^{-3}. In the Fram Strait between Spitsbergen and East Greenland we found 14 ± 2 (N = 25, ± 1SE) Bq m^{-3}. In West Greenland coastal water the mean level was 5.4 ± 0.1 (N = 52 ± 1SE) Bq m^{-3}, and around Iceland and the Faroe Islands close to the North Atlantic fallout background of 3 Bq ^{137}Cs m^{-3}. The pattern of ^{134}Cs is similar to that of ^{137}Cs, but the local variations are more pronounced for ^{134}Cs. If we correct for radioactive decay, the ^{134}Cs concentrations for a given location have been nearly constant during the period, similar to that seen for the ^{137}Cs levels. In 1983 the concentrations of ^{134}Cs were 2–3% of those of ^{137}Cs.

With regard to other radionuclide analyses the material is less complete. The ^{90}Sr concentrations followed those of ^{137}Cs but the local variations were less pronounced, and the levels were as expected lower. The plutonium content of North Atlantic surface sea water has been nearly constant since 1980 at about 13 m Bq m^{-3}. In general the local variations have been small. Only in the North Sea and the British waters the plutonium levels have been enhanced significantly. Relatively high $^{238}Pu/^{239,240}Pu$ ratios in the Norwegian Coastal Current, however, suggest that some non-fallout plutonium may be present here too (Dahlgaard et al. 1984).

DISCUSSION

During the eighties the decay-corrected (to 1983) concentration patterns of ^{90}Sr, ^{134}Cs, and ^{137}Cs have been rather constant in the North Atlantic region. The annual discharges from Sellafield (BNFL 1978–1982; Cambray 1982) have also been approximately constant during the period 1974–1981:

3.278 ± 0.758 PBq ^{137}Cs a^{-1}
(N = 8, rel SD: 23%)

0.0725 ± 0.0128 PBq ^{134}Cs a^{-1}
(N = 8, rel SD: 18%)

0.343 ± 0.091 PBq ^{90}Sr a^{-1}
(N = 8, rel SD: 27%)

It would therefore seem obvious to postulate that we have attained an equilibrium between discharges from Sellafield and surface water concentrations of the three above-mentioned radionuclides in the North Atlantic. If so, we may use the data to calculate transfer factors from Sellafield dischar-

ges to various water masses in the North Atlantic.

A transfer factor to sea water from a source discharging radioactivity to the sea may e.g. be given as Bq m^{-3} per annual discharge in PBq.

In the North Atlantic surface sea water the fallout backgrounds are 3 Bq ^{137}Cs m^{-3}, 0 Bq ^{134}Cs m^{-3} and 2 Bq ^{90}Sr m^{-3}. These figures have to be subtracted from the measured concentrations before any transfer factors can be calculated.

In the surface waters of the Arctic Ocean the fallout backgrounds of ^{90}Sr and ^{137}Cs are higher than those in the Atlantic, as inflow of Pacific Ocean water and runoff from the Siberian rivers to the Arctic Ocean carry relatively higher fallout levels than the Atlantic Ocean water.

In the Fram Strait the boundary between Polar and Atlantic water in 1983 nearly followed the 0° longitude (Fig. 1). Temperatures, salinities and the position of the ice edge

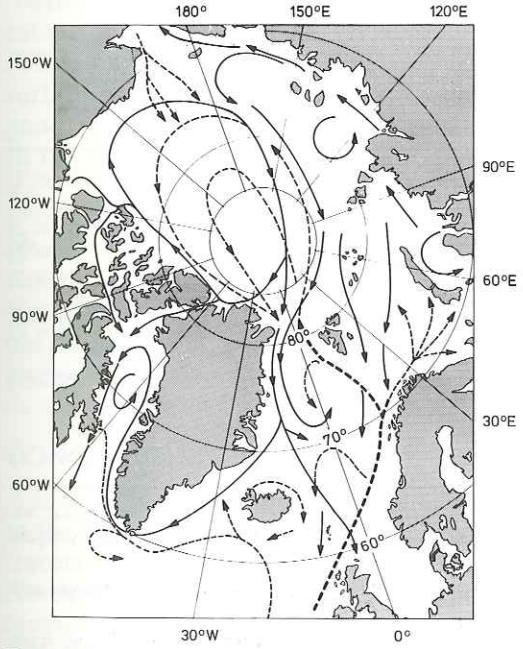

Fig. 1. Main ocean currents in the northern North Atlantic and the Arctic Ocean (dotted lines: warm; solid lines: cold). After Rey (1982).

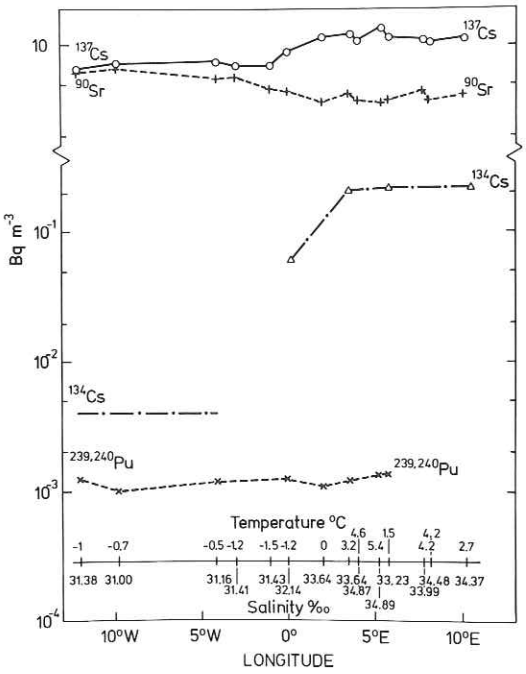

Fig. 2. Radionuclide concentrations in surface water (8 m depth) collected around 80°N in the Fram Strait in July 1983.

showed that this was the case. Thus the stations west of 0° were in the East Greenland Current and those east of 0° in Atlantic water.

Figure 2 shows the concentrations of ^{90}Sr, ^{134}Cs, ^{137}Cs and $^{239,240}Pu$ in the surface water collected inside and outside the East Greenland Current in the Fram Strait. The mean calculated concentrations in the Polar and Atlantic surface waters in the Fram Strait are given in Table 1.

In the Atlantic water (east of 0° in the Fram Strait) the fallout concentrations are 3 Bq ^{137}Cs m^{-3} and 2 Bq ^{90}Sr m^{-3}. Hence the contributions from Sellafield become $11.38 - 3 = 8.38$ Bq ^{137}Cs m^{-3} and $4.00 - 2 = 2.00$ Bq ^{90}Sr m^{-3}. Caesium-134 comes from Sellafield alone and the ratio of Sellafield activity in Atlantic to polar water thus becomes $0.186/0.004 = 46.5$. Hence the contributions from Sellafield to polar water

become 8.38/46.5 = 0.18 Bq ^{137}Cs m^{-3} and 2/46.5 = 0.043 Bq ^{90}Sr m^{-3}, and consequently the fallout concentrations in the polar water are 7.04 − 0.18 = 6.86 Bq ^{137}Cs m^{-3} and 5.76 − 0.043 = 5.72 Bq ^{90}Sr m^{-3}.

In Table 2 are summarized the transfer factors from Sellafield to surface seawater collected in various parts of the North Atlantic. From the North Sea to the eastern part of the Fram Strait the activities in the water are diluted by a factor of 10 and in the polar water the dilution is 300−500 times. At Jan Mayen the dilution (relative to the North Sea water) was 10−20 times. According to Casso and Livingston (1984) other in-

TABLE 1

Radionuclide mean concentrations (± 1SD) in surface sea water at 79°–80°N collected in July 1983 in the Fram Strait.

	Length	^{90}Sr		^{134}Cs		^{137}Cs		$^{239,\ 240}Pu$	
		Bq m^{-3}	*N*	*Bq m^{-3}*	*N*	*Bq m^{-3}*	*N*	*mBq m^{-3}*	*N*
"Polar Stations"	12°05′W−1°09′W	5.76 ± 0.78	5	0.004[1]	5	7.04 ± 0.33	5	11.4 ± 1.4	3
"Atlantic" Stations	0°05′E−10°25′E	4.00 ± 0.30	9	0.186 ± 0.049[2]	9	11.38 ± 1.24	9	12.3 ± 1.0	5

[1] 3 samples of 1800 litres combined to one before analysis; counting error 43%.

[2] The $^{134}/^{137}Cs$ ratio at 0°05′E was 0.0067. The ratio of the other eight "Atlantic" stations was assumed to be equal to the mean ratio (0.0173) measured at three of these stations. The ^{134}Cs mean concentration at the nine "Atlantic" stations was calculated from these ratios and the actual 4 measurements of ^{134}Cs.

TABLE 2

Transfer factors[1] (Bq m^{-3} per PBq a^{-1}) for ^{90}Sr, ^{134}Cs and ^{137}Cs discharged from Sellafield in surface water samples collected in the North Atlantic in 1983.

Sea area	*Latitude or longitude range of samples*	[2] ^{90}Sr ± *1SD*			^{134}Cs ± *1SD*			^{137}Cs ± *1SD*		
North Sea	57°54′N−60°33′N	42	±2	(N = 2)	33	±8	(N = 2)	17	±2	(N = 2)
Norwegian Coastal Current	63°21′N−65°33′N	20		(N = 1)	22		(N = 1)	13	±	(N = 2)
— —	68°30′N−70°36′N	27		(N = 1)	18	±2	(N = 2)	15	±	(N = 2)
Barents Sea	71°32′N−73°00′N	13		(N = 1)	12		(N = 1)	10	±0.4	(N = 2)
— —	73°44′N−77°07′N	6.2	±0.3	(N = 5)	3.1	±0.5	(N = 2)	3.3	±0.7	(N = 5)
Arctic Ocean ...	81°13′N−81°32′N	5.3	±0.7	(N = 2)	2.6		(N = 1)	2.3	±0.5	(N = 2)
Fram Strait (East)	0°05′E−10°25′E	4.3	±0.3	(N = 9)	2.6	±0.7	(N = 4)	2.6	±0.3	(N = 9)
Fram Strait (West)	1°09′W−12°05′W	0.13	±0.07[3]		0.06	±0.03[3]		0.03	±0.015[3]	
Jan Mayen[4]	71°N 8°W	—			—			1.3		(N = 1)

[1] In the calculation of the transfer factors there has been corrected for radioactive decay of the radionuclides, i.e. we have assumed no radioactive decay of any radionuclide. Hence ^{134}Cs and ^{137}Cs should in theory display equal transfer factors.

[2] It has been assumed that 83% of the non-fallout ^{90}Sr come from Sellafield (17% are from Cap de la Hague and Dounreay). (Luykx and Fraser 1983).

[3] As the calculations depend upon the ^{134}Cs determination (cf. the text) which has a counting error of 43%, we have estimated the SD as 50%. The errors from the discharge figures are not included in the error terms in this table.

[4] The fallout background was estimated from the concentrations found in the East Greenland Current between Danmarkshavn and Angmagssalik, i.e. 4.87 Bq ^{137}Cs m^3 in 1983.

vestigators (Jefferies et al. 1982; Kautsky and Murray 1981 have found ^{137}Cs surface water concentrations of 8 Bq m^{-3} around Jan Mayen. This is compatible with our single observation of 9 Bq m^{-3}. It seems thus possible that transfer of Sellafield activity to the East Greenland Current may occur at the lattitudes around Jan Mayen. This probably happens by entrainment of eddies due to the strong fronts between polar water and arctic domain water (Swift and Aagard 1981).

Table 2 furthermore shows that the transfer of ^{90}Sr is approximately two times that of radiocesium. This is due to a greater sedimentation of ^{137}Cs (and ^{134}Cs) than of ^{90}Sr, particularly in coastal, low salinity waters. Similar observations have been made in the Danish Straits (Aarkrog et al. 1984). In other words ^{90}Sr behaves more conservatively in coastal waters than radiocesium and may thus be a better tracer for the water masses.

At the "Polar" stations the $^{137}Cs/^{90}Sr$ ratio from fallout was 1.20. Livingston et al. (1984) found a ratio of 1.23 in 1979 in the surface mixed layer of the Arctic Ocean, in good agreement with our calculations. In the same water masses Livingston found a $^{239,\,240}Pu/^{90}Sr$ ratio of 0.0025 and a $^{239,\,240}Pu/^{137}Cs$ ratio of 0.0020. At the "Polar" stations we found 0.0020 and 0.0016, respectively, i.e. 80% of the Livingston figures. This may be due to a preferential sinking of particulate Pu to deeper water layers from 1979 to 1983.

CONCLUSION

It is shown that radionuclides discharged to the Irish Sea can be used as long-distance tracers for water masses in the northern North Atlantic region. A waterborne pollutant in the North Sea is diluted by a factor of 300–500 when it enters the East Greenland Current in the Fram Strait. But higher concentrations may be seen southward in the Current due to a short-circuit in the Greenland Sea. Although the radioactive contamination from Sellafield is easily measurable in the North Atlantic, its radiological impact outside the Irish Sea is extremely low. The individual annual doses to the populations in north-western Europe from consumption of fish containing radionuclides from Sellafield are on the order of 10^{-3} to 10^{-4} times the doses received from the naturally occuring background radiation.

ACKNOWLEDGEMENTS

The present project was supported by the CEC Radiation Protection Programme. The Alfred Wegener Institute for Polar Research, Bremerhaven is acknowledged for its kind invitation to participate in the 1983 MIZEX- and Fram Strait cruise on board the R. V. "Polarstern". We are indebted to Erik Buch of the Greenland Fisheries and Environmental Research Institute, for providing us with water samples from West Greenland.

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