Radionuclides from Windscale discharges I: nonequilibrium tracer experiments in high-latitude oceanography

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ABSTRACT

The releases of soluble radionuclides from the nuclear fuel reprocessing plant at Windscale represent for some artificial radionuclides a major addition to the inventories due to nuclear test fallout on the northern hemisphere. Most of the soluble examples of the released radionuclides, especially ^{90}Sr and $^{134,137}Cs$, have passed, or will pass, through the North Sea and into the Norwegian coastal current. The releases contain, in the form of specific nuclide ratios, both event markers that should characterize specific years and elapsed time indicators. The amount of ^{90}Sr released through 1979 has exceeded the 1972 measured inventory in the North Atlantic north of 60N; and the amount of ^{137}Cs has approximated six times the inventory measured in the same area. Most of the released soluble tracers that have reached Arctic waters must still reside there. There, Windscale output in 1974 to 1979 alone would result, if mixed uniformly into the upper 150 m, in ^{137}Cs concentrations seven times and in ^{90}Sr concentrations three times those expected from fallout alone. The oceanographic potential of this tracer injection is evident, and fully justifies a feeling of urgency in proceeding to exploit it.

1. Introduction

A substantial amount of valuable oceanographic and geochemical information has been obtained from study of the distributions of the longer-lived artificial radionuclides that have been introduced to the world oceans as fallout following atmospheric tests of nuclear explosives (Broecker, 1966; Munnich and Roether, 1967; Noshkin and Bowen, 1973; Bowen *et al.*, 1974; Volchok *et al.*, 1971; Rooth and Ostlund, 1972; Ostlund and Fine, 1979; Folsom *et al.*, 1970). Interpretation of these distributions, and of their changes with time, depends critically on the assumption that the patterns and rates of introduction are well known for the various tracers. Until recently it has been safe to assume that for most nuclides and in all oceans except the Pacific (Bowen *et al.*, 1980), stratospheric fallout has been the overwhelming source, and that its patterns and rates of delivery to the

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oceans could be approximately inferred from measurements made on land (Noshkin and Bowen, 1973; Volchok, 1974). The major exception was the pulse of ^{238}Pu originating from burnup of the power source of an aborted satellite in October 1964 (Hardy *et al.*, 1973).

It is not yet widely realized that the low-level liquid waste stream released from the British Nuclear Fuels, Ltd. (BNFL), reprocessing plant at Windscale in Cumbria on the Irish Sea, has provided a quantitatively significant perturbation, in some ocean areas, of the fallout nuclide distributions. The ^{137}Cs from the French reprocessing plant, at Cap de la Hague on the Channel coast, was observed by the German Hydrographic Office in the North Sea as early as 1970 (Deutsches Hydrogr. Inst., 1971, 1972, 1980; Kautsky, 1976), both by ratios of ^{137}Cs to ^{90}Sr far above 1.45 which is typical of fallout (Bowen *et al.*, 1974), and by a steady rise in ^{137}Cs concentrations. Windscale ^{137}Cs was first reported by the German Hydrographic Office in the northwestern North Sea in 1971 (Deutsches Hydrogr. Inst., 1971). Jefferies *et al.* (1973) confirmed these latter observations and described both the use of $^{134}Cs/^{137}Cs$ ratios as elapsed time indicators and the pathway of distribution of Windscale ^{137}Cs from the Irish Sea via the Scottish coastal current into the North Sea from the northwest. At this time it was still suggested, in their Figure 3, that another important distribution path carried Windscale ^{137}Cs westward, north of Ireland into the North Atlantic. Later oceanographic studies have shown that the ^{137}Cs from Windscale is accompanied, in addition to ^{134}Cs, by measurable amounts of ^{90}Sr, $^{238,239,240}Pu$, ^{241}Am and by inference any other reasonably long-lived and reasonably soluble components of the waste stream (Livingston and Bowen, 1977). At the same time, both British (Hetherington, 1976; Mitchell, 1977a, 1977b) and German (Murray and Kautsky, 1977; Kautsky, 1977) data have accumulated giving convincing evidence that the ^{137}Cs flow around Scotland into the North Sea is in fact the major pathway for distribution of Windscale effluents leaving the Irish Sea.

The amounts of several important tracer radionuclides introduced from Windscale have been so large that, especially in view of the fact that their introduction pathway represents close to a point source located at the eastern end of the 60th parallel in the Norwegian Sea, it may be impossible to understand some recent, and most future, Arctic and North Atlantic distributions of artificial radionuclides except in the light of this perturbation. Furthermore, the Windscale waste stream carries, in the form of changing isotopic ratios, both elapsed time indicators (Jefferies *et al.*, 1973) and event markers that make this tracer introduction potentially uniquely valuable as an oceanographic tool.

History of Windscale releases. British Nuclear Fuels, Ltd. (BNFL), and the Fisheries Radiobiological Laboratory, Lowestoft (FRL), of the U. K. Ministry of Agriculture, Fisheries and Food, have provided excellent series of data concerning

the annual liquid radioactive waste releases from Windscale. More recently these sources have been supplemented by the Commission of the European Communities and others.

In Table 1 we have summarized the history, from the variety of reports shown, of the releases of ^{90}Sr, ^{134}Cs, ^{137}Cs, ^{238,239,240}Pu, ^{241}Am, and tritium from 1957 up to 1979, the latest year for which we have found data published. For comparison, at the right of the table are set out the estimated annual rates, through 1972, of fallout ^{90}Sr delivery to the Atlantic Ocean, each year increment expressed as percent of the estimated inventory at the end of the previous year (Volchok and Toonkel, 1974). After 1972, annual fallout increments represented trivial additions. The ratios, in world-wide fallout, of major nuclides like ^{3}H, ^{90}Sr, ^{137}Cs, and ^{239,240}Pu are thought to have been quite uniform and the pattern of delivery increments vs. time should apply to all of these. At the bottom of Table 1 we have also shown, from another source (Kupferman *et al.*, 1979), the ocean water-column inventories of ^{90}Sr and ^{137}Cs measured in several 10° latitude bands of the North Atlantic; we have used these inventories to calculate those of ^{239,240}Pu and ^{241}Am, using ratios reported elsewhere as referenced in Table 1.

We believe these Windscale release data are sufficiently reliable for oceanographic uses; in a number of cases there are small discrepancies between the values reported, for given nuclides in given years, by the two responsible agencies, and in those cases we have used BNFL data. It is important to express here the debt of gratitude owed by oceanographers to the British government and to the two responsible agencies for their careful documentation of the Windscale releases. No other environmental introduction of artificial radionuclides is so well described. Without this series of release data, it would have been difficult to appreciate or to use some of the most salient advantages of this tracer introduction as an oceanographic tool.

2. Discussion

Although we have included, for their considerable interest, the release data for total alpha emitters, for Pu, and for ^{241}Am, these materials must be assumed to have had only a minimal impact on the Arctic Ocean inventories. It has been emphasized by reports from FRL (Hetherington, 1976b; Hetherington *et al.*, 1976b) and confirmed by Murray and Kautsky (1977), Livingston and Bowen (1977), and Nelson and Lovett (1978) that the overwhelming preponderance of released transuranic elements from Windscale does not leave the Irish Sea, being largely immobilized locally in the sediments. The total amounts released, especially in the case of ^{241}Am, have, however, been so far in excess of those delivered to high latitudes of the North Atlantic as fallout, that one cannot completely dismiss the incremental significance of even the small fractions of Windscale transuranics that have escaped from the Irish Sea. The demonstration by Nelson and Lovett (1978),

Table 1. Windscale liquid effluent annual discharges — *Curies per year.*

Year	$^{90}Sr^{a,d,g,m}$	$^{134}Cs^{e,h,l}$	$^{137}Cs^{a,f,g,h,l}$	$Pu^{c,m}$	Total^{a,b,c,m} alpha	$^{241}Am^{b,m}$	$^{3}H^{h,m}$	Annual fallout increment (%)
1957	1,644		3,720		58			
58	2,520		6,192		62			
59	1,548		1,980		67			57.3
60	516		912		82			7.9
61	492		1,092		133			9.7
62	1,020		1,114		186			37.4
63	552		372		228			49.2
64	972		1,332		282			21.5
65	1,164		1,164		406			7.9
66	1,000		1,200		586			2.9
67	1,392		1,584	492	955		16,100	1.6
68	1,370		1,500	828	1,416	576	20,300	1.8
69	2,400	630	11,500	816	1,356	396	24,800	1.3
70	6,300	7,010	30,500	936	1,656	540	32,000	2.0
71	12,376	6,372	35,820	1,128	2,688	1,020	31,500	1.7
72	15,316	5,815	34,836	1,548	3,864	2,172	33,600	0.7
73	7,700	4,481	20,760	1,776	4,896	2,952	20,123[k]	

74	9,860	26,933	109,764	1,248	4,572	3,192	32,396[k]
75	13,140	29,211	141,360	1,200[k]	2,310	984[k]	37,952[k]
76	10,656	19,953	118,368	1,272[k]	1,600	324[k]	32,460[k]
77	11,534	16,066	121,032	981	1,241	99	24,716
78	16,160	10,909	110,483	1,567	1,837	214	28,371
1979	6,810	6,363	69,255	1,335	1,675	212	31,779
Total 1957-79:	Ci. 126,442	>145,908	825,840	>15,133	32,156	>12,675	>366,866

Atlantic Ocean
Inventory: 1972[l] - Curies:

60-66N	91,000	132,000	1,600[j]		330[j]
50-60N	347,000	503,000	6,200[j]		1,250[j]
40-50N	456,000	661,000	8,200[j]		1,640[j]

Data Sources* a) JOS-71,Table 18.
b) HET-al.-76a, Table 1.
c) HET-al.-76b.
d) WOO-73.
e) MIN-77.
f) MIT-67;-68;-69;-71a,b;-73;-75;-77a,b. HET-76b.
g) BRI-77.
h) UNS-77
i) KUP-79.
j) from ^{137}Cs inventories, using ratios from HAR-73, LIV-76.
k) HEA-78.
l) PRE-78.
m) BRI-78, BRI-79.

* References shown by initial 3 letters of first author's name, and by last 2 figures of date; in case of confusion "al." indicates more than 2 authors, and suffix a,b etc correspond to multi-publication years of authors in question.

that the soluble fraction of Windscale Pu owes this property to its stabilization in a higher valence state, suggests that once in the outflow stream, this Pu may have a quite considerable residence time in surface seawater.

In the cases of ^{90}Sr, ^{134}Cs, ^{137}Cs, or ^{3}H, no effective mechanism appears to operate to prevent their leaving the Irish Sea and, in fact, data indicate that after a delay, estimated in the neighborhood of two years (Jefferies *et al.,* 1973; Livingston and Bowen, 1977), the major amounts of these soluble waste radionuclides move northward around the Scottish coast and into the North Sea. Estimates of the time of travel from the Irish Sea (Livingston and Bowen, 1977; Livingston *et al.,* 1982a) agree that it must be comparatively short. Furthermore, both oceanographic calculations of North Sea water mass movements (Kalle, 1949; McCave, 1973; Laevastu, 1963; Dooley, 1974) and observational data (Kautsky, 1977; Livingston *et al.,* 1982a) agree that a major fraction of the water entering the North Sea between Scotland and the Shetland Islands, and carrying the Windscale tracers, moves rapidly across the northern North Sea and into the northward-flowing Norwegian coastal current. At this point we may usefully think of the Windscale effluent as representing a continuous jet of tracer radionuclides toward and finally into the Arctic Ocean (defined as including the Norwegian, Greenland, and Barents Seas).

Any point-source non-equilibrium tracer injection has great value, and British (Jefferies *et al.,* 1973; Hetherington, 1976b; Mitchell, 1977a,b) and German (Deutsches Hydrogr. Inst., 1971, 1972, 1980; Kautsky, 1976) reports have shown how much information can be wrung from little more than the changing concentrations of ^{137}Cs along the paths of distribution of Windscale effluent, or of that from Cap de la Hague. There is, however, much more to be gained by attention to the fact that the Windscale effluent contains elapsed-time indicators, such as that provided by the ratio of $^{134}Cs/^{137}Cs$ as noted by Jefferies *et al.* (1973) and Livingston and Bowen (1977), and event-markers in the form of abrupt, datable, changes in the nuclide composition of the wastes.

Brief consideration of the analytical problems involved in the oceanographic application of radionuclide tracers indicates that none of these special properties of the Windscale effluent could be of much more than very local usefulness if the amounts released had not been large in comparison to those being introduced as fallout from nuclear weapons tests. This is a critical point, and oceanography is the gainer from the fact that, indeed, Windscale has released several tracers in amounts comparable to, or exceeding, the inventories in North Atlantic areas north of 40N, produced by fallout alone.

Figure 1 shows the geographical and hydrographical setting relevant to the early stages of the dispersion of releases from Windscale and from Cap de la Hague. To illustrate the quantitative aspect of this tracer discharge, we have plotted in Figure 2 both the annual delivery of global fallout (Volchok and Toonkel, 1974), each

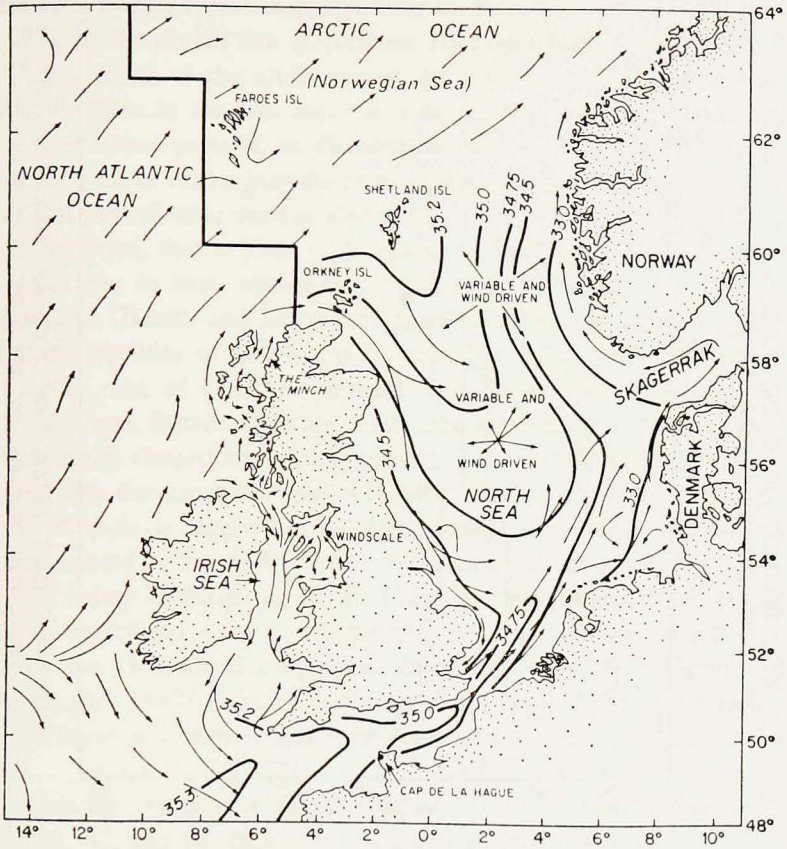

Figure 1. Mean salinities and residual surface circulation patterns in oceanic areas receiving European nuclear fuel reprocessing wastes, Atlantic Ocean boundaries from Kupferman *et al.* (1979), following Worthington and Wright (1970) and Vowinkel and Orvig (1962).

year's increment being shown as percent of the total delivered by the end of 1972, and the annual releases from Windscale of ^{137}Cs, each year's release being shown as percent of the ^{137}Cs inventory found in 1972 in the Atlantic Ocean between 60 and 66N (Kupferman *et al.*, 1979). The choice of ^{137}Cs for this illustration was dictated both by its releases having undergone great changes over the time of the Windscale operation, and by its having been the most widely used of the tracers released. Examination of the data in Table 1 and of that tabulated by Joseph *et al.* (1971) shows that comparable fluctuations, but of lesser magnitude, have characterized the releases of many other nuclides.

Changing pattern of nuclide amounts in Windscale releases. As shown in Figure 2,

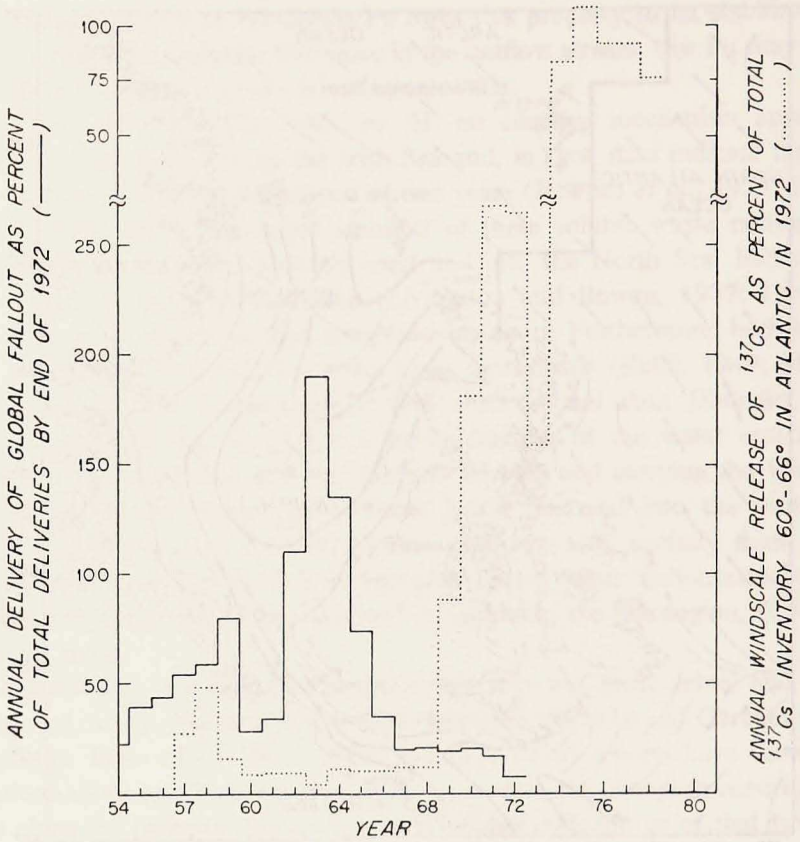

Figure 2. Relative times and magnitudes of ^{137}Cs input to high latitude North Atlantic from fallout and from Windscale releases.

in its first two years of operation Windscale released ^{137}Cs in amounts that were comparable, as fractions of the North Atlantic inventory in high latitudes, to those being delivered as a result of global fallout. During the period 1959 through 1968, however, the release rate from Windscale was kept low, and at the same time most of the 1972 inventory of fallout was being delivered. One would expect that during most of this period the Windscale tracers would not have been detectable except locally in the Irish Sea. Generally the literature bears out this expectation, with the exception of a few North Sea data discussed below.

By 1969, when fallout delivery rates had stabilized at only a couple of percent of 1972 totals, as result of the nuclear test ban treaty, the annual rate of release of ^{137}Cs from Windscale dramatically increased to over 8% of the 1972 inventory. In no year since then has the release rate fallen below 15% of the 1972 inventory, and in five years, 1974 to 1978, it ranged between 80% and 110%. Even the

1969 release increase, trivial in comparison to the later releases, was observed by March 1971 in the North Sea (Deutsches Hydrogr. Inst., 1971, 1972; Kautsky, 1976). Clearly much of the usefulness of the ^{137}Cs and other tracers from Windscale is attributable to the fact that for a decade at least, this source completely swamped, over those parts of its distribution path that have so far been studied, the amounts of tracer earlier introduced by global fallout.

During this period other sources also became active in northwestern Europe, but the largest of them, that at Cap de la Hague in France on the Channel coast, is not reported ever to have released as much as 15% of the "total beta" release from Windscale (Health and Safety Directorate, 1978) and much less, in proportion, of those nuclides of interest as tracers. The same sort of comparison also appears in the case of Dounreay (Health and Safety Directorate, 1978) on the north tip of Great Britain. Neither these, nor any others documented, represent other than trivial distortions of the massive flux of radionuclides released from Windscale, with the exception only of ^{106}Ru. During 1972 to 1977, releases of ^{106}Ru from Cap de la Hague are reported (Health and Safety Directorate, 1978) and are calculated to be 48% of the Windscale release over the same interval (Health and Safety Directorate, 1978). Like the transuranics, ^{106}Ru is believed to be largely immobilized locally in sediments near to the release point, but data reported by the German Hydrographic Office for their 1977 North Sea survey (Deut. Hydr. In., 1980) indicate that a small fraction of the ^{106}Ru released from Cap de la Hague is transported up the European coastline at least as far as the Skagerrak.

The Windscale releases of liquid effluent are not known to be substantially labelled with ^{14}C, and are deficient, compared to fallout labelling, in ^{3}H. Table 1 shows that the ratio of total tritium to ^{137}Cs released over the history of the operation is unlikely to have much exceeded 0.6, whereas the calculated 1972 ^{3}H inventory, from fallout, in the North Atlantic (6.8×10^{8} Ci, Weiss *et al.*, 1979) is close to 200 times that of ^{137}Cs (Kupferman *et al.*, 1979). Unlike most of the other nuclide constituents, tritium or ^{14}C in mixtures of Windscale effluent with seawater will usually be dominated by bomb-production sources. Their concentrations, in contrast to those of the other soluble tracers in the mixture, may be useful guides to the source and history of the seawater, and thus contribute to unravelling some of the interpretational difficulties considered below. This advantage, at least in the case of tritium, may be lost in the future. If and when the proposed expansion of the Windscale facility takes place to permit large scale reprocessing of thermal oxide fuel (the THORP plant), tritium releases will greatly increase. It has been estimated (Mummery, 1977) that tritium in the liquid effluent of the new plant could amount to 10^{6} Ci per year at full operation. These levels would ensure, for tritium, extreme future importance as a direct water tracer along Windscale effluent distribution pathways, much as ^{137}Cs is now. In this case, the $^{3}H/^{137}Cs$

ratio in the effluent stream would provide another useful elapsed-time indicator, operating over longer periods than does that of $^{134}Cs/^{137}Cs$.

Elapsed-time indicators. The use of the change in ratio of $^{134}Cs/^{137}Cs$ as an elapsed-time indicator derives from the facts 1) that these two nuclides have substantially different rates of radioactive decay (half-lives, respectively 2 y and 30 y); 2) that the annual mean discharges of the two nuclides are reported, as shown in Table 1, and their ratios have been reasonably uniform in the early 1970's though steadily decreasing from 1974 to 1979; 3) that the two nuclides, isotopes of the same element, are not discriminated by any known geochemical or biological process; and 4) because of this, that since ^{134}Cs has at present no significant sources other than in fuel-reprocessing wastes, change in its ratio to ^{137}Cs can be attributed chiefly to radioactive decay. The two salient exceptions to this are situations in which the stream of Windscale-labelled water may mix with water masses containing earlier, and therefore older, injections of the same tracer stream, or situations of significant mixing with fallout ^{137}Cs, that is free of ^{134}Cs, as discussed by Livingston and Bowen (1977). The first of these exceptions may be expected to be observable now in some special oceanographic situations, such as that characterizing the "shelf-edge current" off southern Norway (Kautsky, 1977), and eventually over much of the Arctic Ocean or high-latitude North Atlantic. One expects that careful hydrographic analysis of each tracer situation sampled may help resolve the resulting confusions. The second exception may be easier to deal with. Over a considerable portion of the pathway so far studied for Windscale ^{137}Cs (Jefferies *et al.,* 1973; Livingston and Bowen, 1977; Kautsky, 1977; Livingston *et al.,* 1982a), the fallout ^{137}Cs has been so overwhelmed that it has no significance as a diluent; further along the path it will be necessary to subtract out the contribution of ^{137}Cs from fallout, but data to support such subtraction are available as discussed below. More serious is the problem posed by the relative insensitivity of the gamma spectrometry required to distinguish the two nuclides. As decay reduces their ratio from the range about 0.2 that has characterized earlier Windscale discharges, toward tenths of a percent or less, accurate measurement requires collecting the Cs from volumes of seawater much larger than the 50-100 liters usually used. Recent developments, following the chemisorption procedures described by Folsom *et al.* (1970; 1975) and Kupferman (1971), have made easy the stripping of Cs from several hundred liter samples of seawater (Livingston *et al.,* 1982a, c; Mann and Casso, 1982).

No data have been reported that directly reveal the $^{134}Cs/^{137}Cs$ ratios that are present in Cap de la Hague releases. We believe that these ratios must be generally low, arguing from the reported British inability to find measurable ^{134}Cs in fish samples from the southern North Sea (Preston *et al.,* 1978). It cannot be claimed, however, that this is a satisfactory answer to the Cap de la Hague question.

As was originally pointed out (Jefferies *et al.,* 1973) and confirmed (Livingston

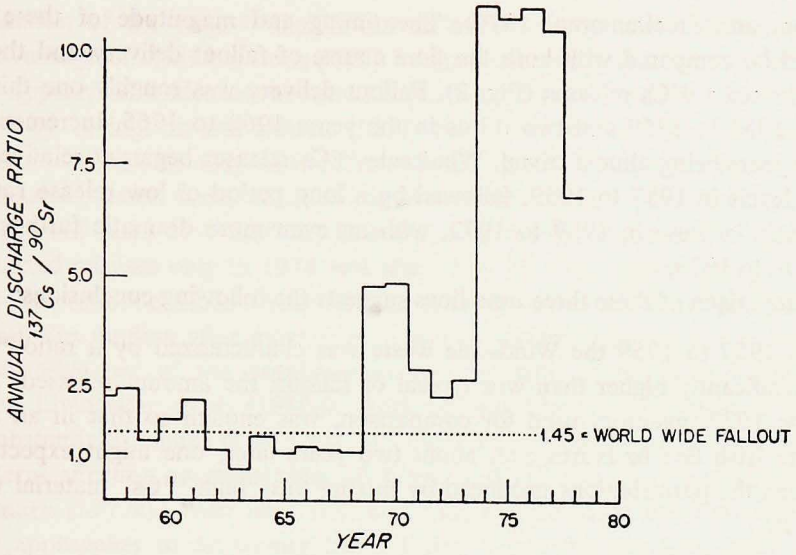

Figure 3. Annual mean ratios, $^{137}Cs/^{90}Sr$, in releases from Windscale.

and Bowen, 1977), the ^{134}Cs half-life of about 2 y is well suited to the time scale of processes within the Irish Sea. The flow around Scotland is, however, too fast to permit measurable change of the $^{134}Cs/^{137}Cs$ ratio by decay between southwest Scottish waters, the Minch, and the North Sea (Livingston and Bowen, 1977; Livingston *et al.*, 1982a). Examination of the Scottish Coastal Current for a faster-changing elapsed-time indicator promises to be very profitable.

Windscale stream certainly contains other elapsed-time indicators that would be useful, but that have not yet been exploited. For shorter-time phenomena the isotope pairs ^{89}Sr:^{90}Sr, ^{103}Ru:^{106}Ru, ^{141}Ce:^{144}Ce, or ^{242}Cm:^{244}Cm ought to be considered; for the longer term such isotope pairs as ^{241}Pu or ^{238}Pu:$^{239,240}Pu$, or ^{242m}Am:^{241}Am may be useful. A related approach would be the use of pairs of nuclides that are not isotopes, but that represent elements between which little or no fractionation would be expected; examples would be ^{147}Pm:^{155}Eu, ^{244}Cm:^{241}Am, or the like. Obtaining the basic supply-term ratios should not be difficult for any of these pairs. Clearly, more thought should be given to this aspect of the Windscale effluent as a tracer experiment.

Event markers. The ratio $^{137}Cs/^{90}Sr$ in the Windscale effluent has undergone a series of abrupt changes that illustrate well what we mean by "event-markers." Inspection of the data of Table 1 shows this, but in Figure 3 the relevant points are made clearer: we have plotted the annual mean ratios, $^{137}Cs/^{90}Sr$, that characterized Windscale releases and for comparison the line representing 1.45, the ratio believed to have characterized world-wide fallout (Bowen *et al.*, 1974, 1980;

Folsom and Sreekumaran, 1970). The timing and magnitude of these changes should be compared with both the time course of fallout delivery and the pattern of Windscale ^{137}Cs releases (Fig. 2). Fallout delivery was roughly one third in the years 1956 to 1959 and two thirds in the years 1962 to 1965, increments in the other years being almost trivial. Windscale ^{137}Cs releases began at relatively significant levels in 1957 to 1959, followed by a long period of low release rate, and a dramatic increase in 1969 to 1972, with an even more dramatic further increase in 1974 to 1979.

Comparison of these three time-lines suggests the following conclusions:

a) In 1957 to 1959 the Windscale waste was characterized by a ratio $^{137}Cs/^{90}Sr$ significantly higher than was typical of fallout; the amount released, 3-5% of the 1972 inventory used for comparison, was enough so that in an area like the Irish Sea or North Sea, about two years later, one might expect to have seen the perturbations produced by mixing this "high-^{137}Cs" material with fallout.

b) In 1959 to 1969 the Windscale waste was often characterized by ratios $^{137}Cs/^{90}Sr$ either higher (1960 to 1962) or lower (1962 to 1969) than fallout, but the annual release rates were so low that we would not expect to have seen these perturbations, except possibly in the Irish Sea.

c) In 1969 to 1971, both the ratio $^{137}Cs/^{90}Sr$ and the annual rate of release increased substantially. This should have been observable as a pulse of high-ratio tracer.

d) In 1971 to 1973, the ratio $^{137}Cs/^{90}Sr$ was reduced by almost half, but the release rate remained high. This might have been observable as a following pulse of lower-ratio tracer.

e) In 1974 to 1979, the ratio $^{137}Cs/^{90}Sr$ again increased, this time really dramatically, and the release rate simultaneously increased, also dramatically. This has been observable as a striking pulse of high ratio tracer along its distribution as far as 61N (Livingston *et al.*, 1982a) by 1978.

Clearly, the prediction of these tracer pulses depends on the assumption that both Cs and Sr are moved, in the oceans, as solute elements, with minimum discrimination. We believe this to be the case and have cited a substantial body of data that support this (Broecker, 1966; Bowen *et al.*, 1974, 1980; Livingston and Bowen, 1977; Murray and Kautsky, 1977; Kupferman and Livingston, 1979). The only demonstrated separation of these two elements in the oceans, the preferential association of ^{137}Cs with sinking particles (Noshkin and Bowen, 1973; Kupferman *et al.*, 1979) has been shown to involve fractions of fallout ^{137}Cs too small to have measurable effect on ratios $^{137}Cs/^{90}Sr$ (Kuperfman and Bowen, 1976; Kupferman and Livingston, 1979).

Application of the ratio $^{137}Cs/^{90}Sr$ as an event marker has not been widely taken advantage of. This has been partly because necessary data have been a little slow to appear, as evidenced by the comparison of our Figure 3 with the data presented by Livingston and Bowen (1977) from analyses of water and sediment collected in the Minch: they showed that based on $^{134}Cs/^{137}Cs$ ratios as elapsed time indicators, travel times of either about 2 or less than 1 year were plausible. Their observed ratio of $^{137}Cs/^{90}Sr$, however, was 6.1 in the water, a value that characterized releases only in 1974 and after (Fig. 3). This argues strongly for the about 2-year estimate, since a much higher ratio would have been expected in the 1975 Irish Sea outflow after more than a year of releases ranging higher than 10. More extensive use of the event-marker concept will appear in a forthcoming report by Livingston *et al.* (1982a). Aarkrog and Lippert have used $^{137}Cs/^{90}Sr$ ratios substantially higher than those in fallout as arguments for Windscale as a source term affecting coastal waters off Greenland (1977a), the Faeroes (1977b), or Denmark (1977c). They have not, however, attempted to use the changes in ratio as approaches to estimating travel times. There are scattered data among early German reports of dissolved radioactivity in the North Sea (Deutsches Hydrogr. Inst., 1962) showing ratios of $^{137}Cs/^{90}Sr$ that are enough higher than fallout so they could plausibly be related to the 1957 to 1959 period of high ratios in the Windscale releases (Fig. 3). At these early times, however, sampling was not sufficiently systematic for the data to be contoured and unequivocal conclusions to be drawn.

As in the case of isotopic-pair elapsed-time indicators, there is no question that the Windscale waste effluent stream has contained many event-markers in addition to those represented by changes in the $^{137}Cs/^{90}Sr$ ratio. For the period 1957 to 1967, several such changes are tabulated (Joseph *et al.*, 1971) but unfortunately comparably detailed data are not available for later years. Hetherington (1976b) used changes in the ratio $^{238}Pu/^{239,240}Pu$ as an indicator of the release time of Pu found deep in sediment cores from the Irish Sea or Ravenglass Estuary. Pentreath and Lovett (1976) showed that in the years 1974 to 1976 Windscale discharges were characterized by very large month-to-month variations in the ratio $^{241}Am/^{239,240}Pu$. These would have provided elegant event-markers, but probably of utility only within the Irish Sea. Our data (Livingston *et al.*, 1982a) as well as those from other laboratories suggest strongly that the usual two-year residence time of Windscale tracers in the Irish Sea (Jefferies *et al.*, 1973; Livingston and Bowen, 1977) must tend to obliterate, by homogenization, any really rapid fluctuations in the ratios of discharge products further along their distribution paths. There are, however, data that suggest there is relatively little mixing of discharges separated in time by several months or longer. Baxter *et al.* (1979) found amazingly good agreement, in the Clyde Sea area, of both $^{134}Cs/^{137}Cs$ ratios and the timing of peaks in ^{137}Cs concentration, with values predictable from BNFL discharge data of two

years earlier. Our interpretation of the Clyde Sea data (Baxter *et al.*, 1979) does not agree with that of the authors quoted, who feel a more complex model is indicated, but it fits excellently with our own data (Livingston *et al.*, 1982a) from similar measurements further along the distribution pathway.

Evidence of present distribution of Windscale tracer. As we have noted above, various studies (Kautsky, 1977; Livingston *et al.*, 1982a) have shown relatively rapid passage of the Windscale effluent soluble tracers from the Irish Sea, through the northern North Sea into the northward-flowing Norwegian Coastal Current. This should be delivering the tracers, by pathways that are not really well understood, largely to the Arctic Ocean (U. S. Navy Hydrographic Office, 1958; Coachman and Aagaard, 1974), (Fig. 1).

One question of considerable interest and importance is the present (1981) distribution of soluble Windscale tracers, especially of ^{137}Cs. Although the data, primarily British and German, necessary to make precise quantification of some of the distributions, undoubtedly exist and should be available in the future, it may be useful for some tracer oceanographers to think of a rough estimate derived from consideration of the discharge data pattern and of the various published surveys. A vital control on this estimate is the fact that 81% of the 826 kilocuries of ^{137}Cs discharged by the end of 1979 were released subsequent to January 1974, i.e., over only the previous six-year period. We would think that these 826 kilocuries, therefore, might now (1981) be divided about equally among:

a) The Irish Sea

b) Scottish Coastal Water and the North Sea

c) Arctic waters downstream (i.e., north) of the North Sea.

This estimate has significant uncertainties to each fraction but should be refinable as more data become available. Some of our data that contribute substantially to knowledge of these recent distribution patterns and inventories should soon appear (Livingston *et al.*, 1982a, b, c).

There are scattered pieces of evidence (Aarkrog and Lippert, 1977a, b) of the Windscale tracers in surface waters of the Atlantic near the Faeroes Islands, and in the southward-flowing East Greenland Current. The nuclide concentrations represented are, however, not sufficiently high to suggest that more than trivial leakage has yet been observed leaving the Arctic. Clearly, every effort should be made to profit by this opportunity both to establish rates, depths, and directions of movement of Windscale tracers in the Arctic, and to clarify the rates and patterns of delivery of Arctic Ocean water to the northern North Atlantic. The precision and accuracy with which these tracers can be applied would be greatly enhanced by the establishment of a closely-timed schedule, monthly at least, of monitoring the nuclide concentrations that characterize the Scottish Coastal Current, the

Norwegian Coastal Current, and the Cap de la Hague effluent flow into the southern North Sea.

Should appropriate sampling arrangements be possible, the distributions of the Windscale tracers within the Arctic Ocean should be studied in detail. These distributions, since the point of injection and the timing are well established, offer an opportunity to elucidate in a very elegant way the circulation patterns of this difficult area. The signal available is enormous as is shown by the fact that the Windscale output in the years 1974 to 1979 alone would have been enough, if mixed uniformly into the upper 150 m of the Arctic Ocean water column, to raise the concentration of ^{137}Cs to seven times that expected from stratospheric fallout, and of ^{90}Sr to three times. The combination of such a substantial increase in tracer concentrations and the presence of event and elapsed time indicators offers Arctic oceanography a remarkably powerful tool. We have ourselves, in cooperation with A. Aarkrog (Risø National Laboratory, Denmark), R. W. Moore (Dalhousie University, Canada), H. Kautsky (Deutsches Hydrogr. Inst., Germany), W. Weiss (University of Heidelberg, Germany), and D. F. Jefferies (Fisheries Radiobiological Laboratory, Lowestoft, U. K.), begun studying these distributions. The problems of sampling are, however, so severe, especially under the ice, that much more effort should be recruited.

We urge international attention to these unique opportunities.

3. Conclusions

Compilations and analyses of published data concerning the amounts, and relative nuclide contents, of the radioactive liquid effluents released by the BNFL plant at Windscale, when coupled with a variety of reports of surface seawater concentrations of radionuclides, support the following conclusions:

a) For periods, during the operating history of the plant since 1957, Windscale has been a source of ^{137}Cs, ^{90}Sr, and other nuclides, comparable to or exceeding their deposition as global fallout to adjacent ocean areas.

b) The effluent stream contains elapsed-time indicators in the form of ratios of isotope pairs like ^{134}Cs:^{137}Cs or ^{241}Pu:$^{239,240}Pu$ and of radioelement pairs like ^{147}Pm:^{155}Eu. Use of these pairs to indicate elapsed time depends on the members having markedly different half-lives and on the ratio being changed principally by radioactive decay.

c) The effluent stream also contains event-markers in the form of abrupt changes in the ratios of specific nuclides. The pair ^{137}Cs: ^{90}Sr offers a valuable example, but others must be present.

d) Although there is evidence of measurable intrusion of Windscale effluent into the North Atlantic surface circulation, most of the release appears still to be headed toward or already resident in the Arctic Ocean. Its properties and point of injection make it uniquely promising as an oceanographic and geochemical tracer of processes in the Arctic Ocean and high latitudes of the North Atlantic.

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