

Anthropogenic radiotracer evolution in the Central Greenland Sea

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ABSTRACT

The distribution patterns of several anthropogenic radiotracers in the Central Greenland Sea are presented together with a discussion of the significance of the changes in these patterns observed during the period 1972 to 1981. The radiotracers are the radionuclides ^3H , ^{137}Cs , ^{90}Sr , and $^{239,240}\text{Pu}$, introduced to the region primarily as fallout from atmospheric nuclear weapons testing. Some ^{137}Cs is also derived from recent advective input of traces of radioactive waste discharge to European coastal waters.

The observed patterns and their changes with time are discussed in the context of the source input history of each tracer, the physical properties of deep ventilation in the Central Greenland Sea gyre and the chemical reactivity of the tracer. As conservative water tracers, the distributions with time of ^3H , ^{137}Cs and ^{90}Sr can be explained in terms of their input histories and the resultant evolution of each tracer distribution by the purely physical mixing processes operating in the Greenland Sea. On the other hand, although physical mixing processes represent the major controls, the evolution of the $^{239,240}\text{Pu}$ distribution between 1972 and 1979 must be explained in terms of some Pu particle association and vertical scavenging by the net flux of particle populations sinking through the water column.

INTRODUCTION

The Greenland Sea sits in one of the two major basins which connect the Arctic Ocean with the northern North Atlantic. The Norwegian Sea in the other major basin, and the Iceland Sea in the small basin south of the Greenland Sea, complete the major connections between the Atlantic and Arctic Oceans. The Greenland Sea has long been recognized as an important region of deep water formation — both in terms of intermediate water masses which feed into the dense overflows entering the deep North Atlantic between Scotland and Greenland and of deep water masses which enter the deep water circulation in the Greenland and Norwegian Seas and the Eurasian Basin of the Arctic Ocean. Aa-

gaard et al. (1985) have recently described their present understanding of the circulation in these regions which they collectively describe, following earlier authors, as the Arctic Mediterranean Seas. They draw attention to the likelihood that deep ventilation of the basins of the Arctic Mediterranean does not originate uniquely in Greenland Sea winter surface waters but must draw on input from other shallow regions — the wide Arctic shelves in particular. Nevertheless, the nature of the production of deep water in the Greenland Sea and its contribution to deep circulation in the Arctic Mediterranean is critical to an understanding of the regional circulation processes.

As in other oceanic regions, anthropogenic

tracers have been added to the suite of natural tracer tools, both traditional and new, which have been used to unscramble details of the nature of circulation in the Arctic Mediterranean. New anthropogenic tracers include radiotracers from nuclear weapons tests and radioactive waste releases as well as non-radioactive tracers. It is beyond the scope of this paper to review this wide range of tools and their application in the Arctic and sub-Arctic. The purpose of this paper is to describe the evolution in the Central Greenland Sea of the anthropogenic radiotracers ^3H , ^{137}Cs , ^{90}Sr and $^{239, 240}\text{Pu}$ during the period 1972–1981. Since these tracers have been and will be used in the Arctic Mediterranean to address several questions or test various hypotheses connected to the regional circulation (e.g. Östlund 1982; Östlund et al. 1982, 1984; Livingston et al. 1984; Swift et al. 1983; Kautsky 1980; Aarkrog et al. 1983), it is essential that their temporal evolution be characterized in such a major circulation component as the Greenland Sea.

With the exception of Pu, these tracers have been demonstrated to behave essentially conservatively in the open ocean and have been widely used as transients which trace the advective and diffusional components of oceanic circulation. In contrast, Pu has been shown to exhibit significant association with particulates in ocean water columns (e.g. Livingston and Anderson 1983) and consequently is being scavenged and removed to ocean sediments — albeit at rates somewhat slower than were initially predicted. All four tracers have been introduced to the oceans as a result of nuclear age activities but may be viewed as originating ultimately from two very different sources. The first source, usually of major quantitative significance, is the world-wide fallout of radioisotopes from atmospheric nuclear weapons tests. This input was primarily to the northern hemisphere and heaviest at mid-latitudes. Apart from minor

subsequent additions, the time of the input can be thought of as a small pulse in the late 1950's and a considerably larger one in the early 1960's. Subsequently, this primary source has been modified by advective redistribution in oceanic current systems and augmented in areas which are significantly affected by runoff of terrestrial fallout from the continents. The second source, a relatively more recent input, derives from the release of waste radioactivity from nuclear fuel reprocessing operations to points in the shallow European continental seas. For practical purposes this input has been dominated by the discharges to the eastern Irish Sea from the U. K. plant in Cumbria (Sellafield — formerly Windscale). Quantitatively less significant releases have derived from the French plant at Cap de la Hague on the Normandy coast. The time-history and composition of the U. K. releases for the nuclides discussed in this paper have been described (Livingston et al. 1982a) in the context of their use as high northern latitude tracers. The releases of ^{137}Cs and, to a lesser extent, ^{90}Sr increased sharply in the decade beginning in 1970: by two orders of magnitude for ^{137}Cs and by one for ^{90}Sr . The rate of the ^{137}Cs releases was sufficiently high to produce surface water concentrations in the Arctic inflow areas around Spitsbergen which are many times that from fallout derived sources (Holm et al. 1983; Casso and Livingston 1984). The estimate of about 5 years for the transit time for a release to the Irish Sea to reach the Barents Sea (Livingston et al. 1984) effectively means that prior to 1979, artificial radioisotope concentrations in the Greenland Sea derived mostly from fallout sources; but around and after that time, additional input from nuclear fuel reprocessing sources began to make significant additions. An account of the evolution of ^{137}Cs , ^{90}Sr and ^3H in the source waters supplying the Norwegian and Greenland Seas appears elsewhere (Livingston et al. 1985a).

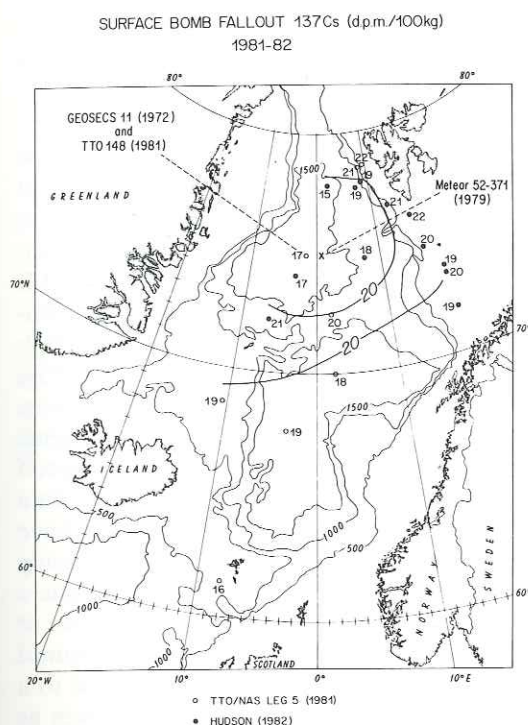


Fig. 1. a) Central Greenland Sea station locations 1972–1981 — positions of GEOSECS 11, METEOR 52-371 and TTO 148. b) Distributions of fallout ^{137}Cs concentrations estimated for surface water in 1981–1982.

METHODS: SAMPLE COLLECTION AND ANALYSIS

The radiotracer data in this paper were from water samples collected in the Greenland Sea in 1972, 1979 and 1981. In August 1972 and again in July 1981, a station in the Central Greenland Sea was occupied by R/V KNORR, as part of the Geochemical Ocean Sections (GEOSECS) programme and Transient Tracers in Oceanography-North Atlantic Study (TTO-NAS) respectively. The position of this station is noted in Figure 1. Also shown in Figure 1 is the position of a station occupied by R/V METEOR in August 1979. On each occasion, large volume water samples were collected for radiochemical analyses — using

Gerard water samplers. Smaller volume samples for tritium analyses were also collected at these stations, although not always on the same lowerings as the Gerard bottles. Standard hydrographic data were collected at each station by conventional methods.

The water samples from the different cruises were analyzed for the various radionuclides in shore based laboratories. ^{137}Cs , ^{90}Sr and $^{239, 240}\text{Pu}$ concentrations were determined at the Woods Hole Oceanographic Institution by radiochemical methods which have been described in detail elsewhere (Bowen et al. 1980). ^3H concentrations were determined at the Tritium Laboratory, University of Miami, also by methods previously reported (Östlund et al. 1974).

RESULTS

The changing distribution of the various radiotracers discussed in this paper need to be set in the context of the water masses which they characterize at any given point in time. Coachman and Aagaard (1974) have presented a review of Arctic and sub-Arctic physical oceanography which sets Greenland Sea water masses in the context of their relationships with other regional water masses. A more recent paper focuses on the complexity of the water mass formation processes which take place in the Greenland Sea as they respond to annual and seasonal forcing (Swift and Aagaard 1981). The major feature of the Central Greenland Sea water masses is the remarkable homogeneity of deep water properties. In summer, the variations in temperature or salinity are quite small over perhaps the lower 80% of the water column. In winter, water masses with these nearly homogeneous water properties are found at depths much closer to the surface. Although the region has been described as being characterized by a spectrum of water

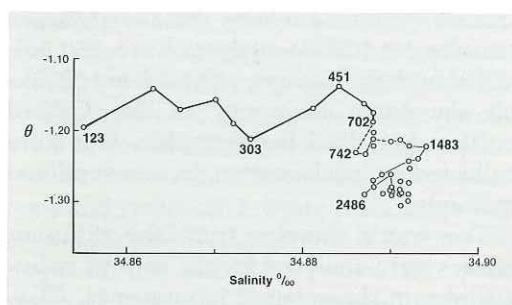


Figure 2. Θ -S diagram for TTO Station 148.

- Arctic Intermediate Water
- Upper Deep Water
- Lower Deep Water

masses in its upper layers (Swift and Aagaard 1981), for the purposes of the present discussion it is perhaps sufficient to divide the upper layers into two categories, as defined by Swift and Aagaard (1981). These are, firstly, the Arctic Surface Water which, in summer, might be typically restricted to the upper 100 m; and secondly, Arctic Intermediate Water, which might be considered to extend down to the top of the deep water. The Θ -S correlation observed at TTO/NAS station 148 in the Central Greenland Sea may be used to illustrate a water mass categorization (Fig. 2). The Arctic Surface water is not shown, being both warmer and fresher than the deeper layers. The Arctic Intermediate Water shows some slight temperature maxima around 200 and 450 m which presumably originate from Atlantic layers. The deep water, if defined as being water colder than -1.2° and saltier than 34.887, is found at depths greater than 700–800 m. For purposes of comparing the change in tracer distribution patterns with time, it seemed useful to subdivide the deep water into two layers, Upper and Lower, the depth of the break being around 1400 m. This is principally because it will be seen later that strong tracer gradients are frequently observed in the Upper layer, in contrast to the more usual nearly constant pattern found in the

Lower layer. In addition, the Upper layer is slightly warmer than the Lower layer although at essentially the same salinity (Fig. 2).

A considerable amount of the data of radionuclide concentrations measured at the three stations in 1972, 1979 and 1981 has been reported in detail elsewhere. The tritium data for GEOSECS station 17 and TTO station 148 are taken from University of Miami reports (Östlund and Brescher 1982; Östlund 1983). The ^{137}Cs , ^{90}Sr and $^{239, 240}\text{Pu}$ data for the GEOSECS and TTO stations appear in two Woods Hole Oceanographic Institution reports (Casso and Livingston 1984; Livingston et al. 1985b). The data from METEOR station 371 in 1979 have not appeared separately and therefore only appear as plotted in Figure 3. To facilitate a comparison of the distribution patterns of the various radiotracers, the measured concentrations have all been corrected to a single reference date, arbitrarily chosen as 1/1/81. Also indicated in Figure 3 are the water mass divisions discussed above.

In order to quantify the differences in distributions of the radiotracers shown in Figure 3, both by water mass and by time, the concentrations of each radionuclide at a given station were converted into integrated radionuclide inventories per unit area, over depths selected to correspond to water mass divisions. For this integration, the concentration of each radionuclide at any given depth was taken to represent the average concentration over the depth interval bounded by the midpoints of the distance to the sample depth above and below any given sample depth. The results of these calculations are listed in Table 1 as a function of water mass, year collected and radionuclide. The water mass at the shallowest depth, although labelled as Arctic Intermediate Water, was integrated to the surface and, hence, includes the very shallow Arctic Surface Water layer.

As has been noted elsewhere (Livingston

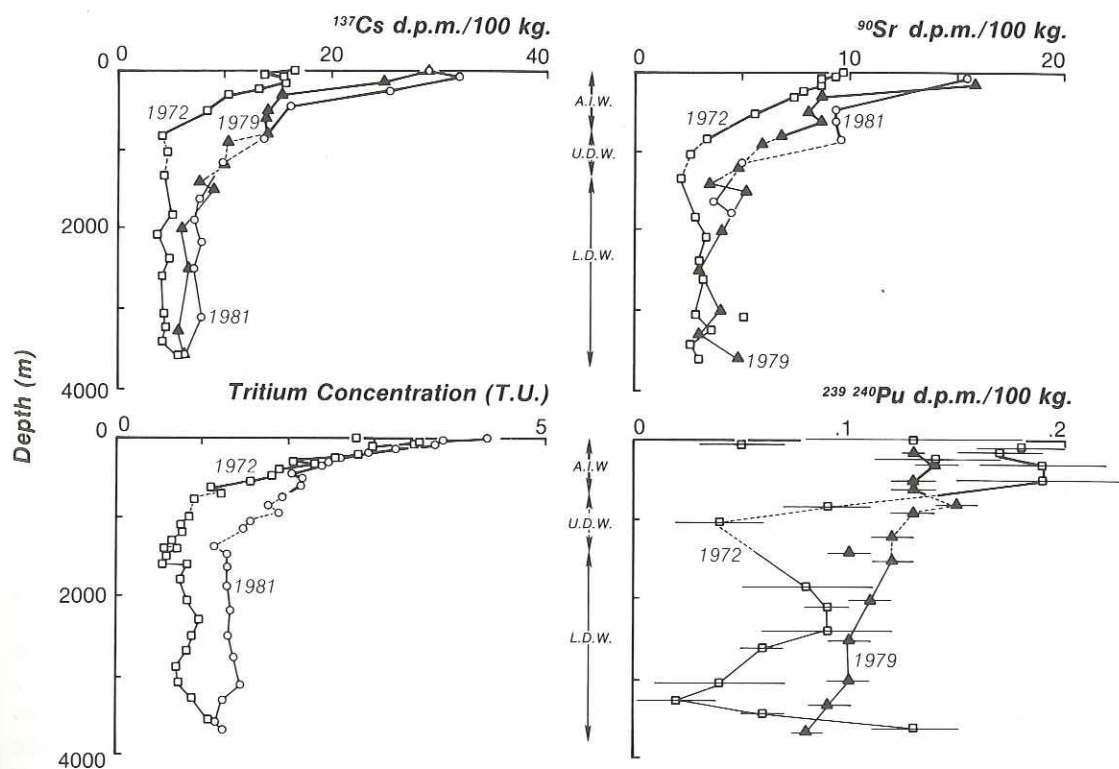


Fig. 3. Depth profiles of ^{137}Cs , ^{90}Sr , ^3H and $^{239, 240}\text{Pu}$ concentrations in the Central Greenland Sea 1972–1981. All data decay corrected to 1 January 1981. \square GEOSECS Station 11, 1972; \blacktriangle METEOR 52, Station 371, 1979; \circ TTO Station 148, 1981; — Arctic Intermediate Water (A. T. W.); - - - - Upper Deep Water (U. D. W.); — Lower Deep Water (L. D. W.).

et al. 1985a), there has been a supply of ^{137}Cs and ^{90}Sr to the Greenland and Norwegian Seas from European nuclear fuel reprocessing sources. Livingston et al. (1985a) also note that tritium from this source is insignificant at such distances from the discharge point. In the case of Pu, although the amounts discharged are relatively large, most of this is deposited in sediments close to the point of release and the amounts which move towards the Arctic are insufficiently high to add significantly to fallout Pu in the sub-Arctic seas (Livingston et al. 1982b). Because of the different input history of ^{137}Cs and ^{90}Sr from the two sources, both in terms of the time-history and route of entry, it is desirable to resolve

the measured concentrations shown in Figure 3 into their fallout and nuclear fuel reprocessing components. This has been attempted. No non-fallout component is assumed to contribute to the concentrations measured in 1972 at GEOSECS station 17, since the earlier releases which could have moved to the Central Greenland Sea by that year were insignificant relative to fallout sources (Livingston et al. 1982a). The resolution of fallout-derived ^{137}Cs and ^{90}Sr in the upper 1400 m of the water column was made through use of the measured tritium concentrations (representing a pure fallout source) after conversion by use of an appropriate ratio of fallout tritium to each of ^{137}Cs and ^{90}Sr . The appropriate ratios were

taken to be those observed in North Atlantic water flowing into the Norwegian Sea, prior to any influence from the European coastal water source. The relevant ratios used, following Livingston et al. (1985a), were 0.253 for the ratio $^3\text{H}/^{137}\text{Cs}$ and 0.380 for the ratio $^3\text{H}/^{90}\text{Sr}$ (being ratios observed in the North Atlantic inflow in 1981). The same approach was used to derive the surface distribution of fallout ^{137}Cs in the Norwegian/Greenland Seas in 1981–1982. The tritium data used to derive the fallout ^{137}Cs distribution were from measurements made on samples collected on TTO/NAS Leg 5 in the region (Östlund 1983, 1984). The resultant pattern of fallout ^{137}Cs concentrations in surface waters is shown in Figure 1.

A somewhat different approach was made to the resolution of the two components in samples collected in the Lower Deep Water. This was in consequence of the fact that the proportion of fallout tritium to ^{137}Cs or ^{90}Sr has been slowly increasing with time in the North Atlantic inflow to the Norwegian Sea (Livingston et al. 1985a). As Greenland Sea Deep Water can be expected to have a composition more influenced by "older" inflowing Atlantic water, it did not seem appropriate to use recent ratios, nor is it clear how to derive those which would be correct averages of earlier ratios. As an alternative approach, use was made of the uniformity of the fallout ratio $^{137}\text{Cs}/^{90}\text{Sr}$ of 1.5 in open ocean sea water (Bowen et al. 1974). This ratio is taken to characterize deeper water which mixes with Atlantic water containing ^{137}Cs and ^{90}Sr from southern European coastal water sources. Since the West Spitsbergen Current (WSC) is the likeliest candidate to represent this "contaminated" Atlantic water, calculations were made of the fallout-derived deep water concentrations of ^{137}Cs and ^{90}Sr . These were calculated by solving the following two simultaneous equations:

$$^{137}\text{Cs}_{\text{Fallout}} = ^{137}\text{Cs}_{\text{Measured}} - ^{137}\text{Cs}_{\text{WSC}}$$

$$^{90}\text{Sr}_{\text{Fallout}} = ^{90}\text{Sr}_{\text{Measured}} - ^{90}\text{Sr}_{\text{WSC}}$$

— by substitution of a fallout $^{137}\text{Cs}/^{90}\text{Sr}$ ratio of 1.5 and a recently observed ratio for the West Spitsbergen Current (WSC). Little difference was found for the deep water fallout ^{137}Cs and ^{90}Sr concentrations calculated using values for the West Spitsbergen Current $^{137}\text{Cs}/^{90}\text{Sr}$ ratios in the range 3–4, representative of recent observed values (Casso and Livingston 1984), so that a representative value of 3.5 was used. This method was applied to data from TTO station 148 to resolve the observed Lower Deep Water concentrations into "fallout" and "nuclear fuel reprocessing" components, the latter being the West Spitsbergen Current component.

The derived "fallout" concentrations were then integrated over the same water mass layers as in Table 1 to produce "fallout" ^{137}Cs and ^{90}Sr inventories. These were then subtracted from the observed inventories in Table 1 to derive inventories of both nuclides from the nuclear waste labeled coastal current. Since no tritium data are available for the 1979 station, it was not possible to take the same approach to resolution of the data from this METEOR station into fallout and non-fallout components. Since the fallout tracers have been present in the region for about two decades, it is unlikely that substantial differences in their distribution in the Central Greenland Sea between 1979 and 1981 would have occurred. On the other hand, this is the time period essentially right at the beginning of the injection into the region of the new sources of ^{137}Cs and ^{90}Sr (the magnitude of the new ^{137}Cs source being substantially larger than that of the new ^{90}Sr source). An assumption was made that the fallout ^{137}Cs and ^{90}Sr inventories in 1979 were the same as those listed in Table 2 for 1981. These were then subtracted from the observed 1979 in-

TABLE 1

Radionuclide Inventories in the Central Greenland Sea 1972–1981.

Water Mass	Year	$^3\text{H}^{1)}$	$^{137}\text{Cs}^{1)}$	$^{90}\text{Sr}^{1)}$	$^{239, 240}\text{Pu}^{1)}$
		Ci/km ²	mCi/km ²		
Arctic Intermediate Water 0–700 m	1972	4.91	34.5	22.1	0.53
	1979	—	57.5	34.2	0.42
	1981	5.85	68.9	36.5	—
Upper Deep Water 700–1400 m	1972	1.85	13.9	8.5	0.18
	1979	—	33.6	16.9	0.40
	1981	3.56	36.0	21.7	—
Lower Deep Water 1400–Bottom	1972	6.17	47.9	32.2	0.76
	1979	—	69.4	41.7	1.08
	1981	9.67	78.1	38.2	—
Total Water Column	1972	12.93	96.3	62.8	1.46
	1979	—	161	94.0	1.90
	1981	19.1	183	99.6	—

¹⁾ All activities as of 1/1/81; 1 Ci/km² = 617 MBq/km² (Ci = Curie; MBq = Megabecquerel).

ventories to derive estimates of the distribution of nuclear waste tracers throughout the water column at that time.

Finally, the percentage inventory change between 1972 and 1979/1981 was calculated for each water mass, fallout-derived radionuclide and year, and is listed in Table 3. This allows a contrast to be made between the decadal scale changes in the fallout tracers and the recent perturbation from the arrival of the new nuclear waste input.

DISCUSSION

The surface distribution of ^{137}Cs in 1981–1982 in the Norwegian and Greenland Seas shows a pattern of ^{137}Cs concentrations dramatically elevated from the fallout ^{137}Cs pattern presented in Figure 1. The total ^{137}Cs distribution has been assembled from data presented elsewhere (Casso and Livingston 1984) but is reproduced for convenience here as Figure 4. It is very evident from a comparison of the patterns in Figure 1 and Figure 4 that the region has received a

substantial input of ^{137}Cs from the new nuclear waste source which appeared in the late 1970's in the shallow continental seas along the eastern margin of the region. Obviously no understanding of the tracer evolution pattern of ^{137}Cs (and to a lesser extent ^{90}Sr) in the Central Greenland Sea can be reached without taking into account both the fallout and the nuclear waste source. This becomes immediately obvious from a comparison of the changes in the profiles in Figure 3, or in the inventories in Table 1, of the total radionuclide concentrations between 1972 and 1981 throughout the water column. Although the concentrations and inventories of all radiotracers increased over this time interval, the largest percentage increase is for ^{137}Cs (90%). This ^{137}Cs increase and the ^{90}Sr increase (59%) both exceed the ^3H increase (48%). However, when the fallout derived ^{137}Cs and ^{90}Sr are resolved from the total observed concentrations, this pattern is reversed. Fallout ^{137}Cs and ^{90}Sr increase by only an average of 5% (Table 3) as compared with the 48%

TABLE 2

Estimate of Fallout-Derived and Nuclear Fuel Reprocessing Derived ^{137}Cs and ^{90}Sr in Central Greenland Sea in 1979 and 1981.

Water Mass	Year	Fallout		Nuclear Fuel Reprocessing	
		$^{137}\text{Cs}^{1)}$ mCi/km	$^{90}\text{Sr}^{1)}$ mCi/km	$^{137}\text{Cs}^{1)}$ mCi/km ²	$^{90}\text{Sr}^{1)}$ mCi/km ²
Arctic Intermediate Water					
0-700 m	{ 1979	—	—	25.1	12.6
	{ 1981	32.4	21.6	36.5	14.9
Upper Deep Water					
700-1400 m	{ 1979	—	—	13.7	3.6
	{ 1981	19.9	13.3	16.1	8.4
Lower Deep Water					
1400-Bottom	{ 1979	—	—	21.4	9.7
	{ 1981	48.8	32.0	30.1	6.2
Total Water Column	{ 1979	—	—	61	27.1
	{ 1981	100	66.9	83	32.7

¹⁾ All activities decay corrected to 1/1/81.

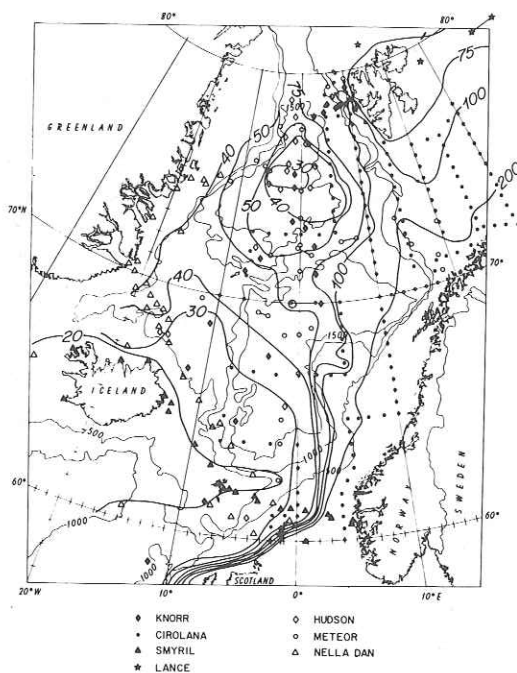


Fig. 4. Surface distribution of ^{137}Cs (d.p.m./100 Kg) in the Norwegian/Greenland Seas in 1981-1982. Station positions in 1981 were occupied by R/V KNORR (◆), R/V CIROLANA (●), M/V SMYRIL (▲), R/V LANCE (★), and in 1982 by R/V HUDSON (◇), R/V METEOR (○) and R/V NELLA DAN (△).

increase in ^3H . The change in $^{239,240}\text{Pu}$ between 1972 and 1979 is seen to be the smallest change in all four radiotracers (+30%), but this represents a larger increase than that of fallout ^{137}Cs and ^{90}Sr ; this is a meaningful difference in view of the fact that, as noted earlier, only $^{239,240}\text{Pu}$ from fallout is believed to contribute to present distributions in the Greenland Sea. This difference and the very different water mass distribution pattern found for $^{239,240}\text{Pu}$ will be discussed later.

Fallout Radionuclide Evolution in the Central Greenland Sea

^3H , ^{137}Cs and ^{90}Sr . As noted above, fallout tritium has increased much more sharply than has fallout ^{137}Cs or ^{90}Sr throughout the Central Greenland Sea water column. This difference is maintained when the changes within the major water mass layer are compared. The relatively greater increase in ^3H is maintained in the intermediate and deep water layers (Table 3), but the change is more marked in the deep waters. In fact, fallout ^{137}Cs and ^{90}Sr inventories in the Lower Deep Water are almost unchanged be-

TABLE 3

Change in Central Greenland Sea Inventories of Fallout Derived Radioisotopes Between 1972 and 1981.

Water Mass	Year	% Inventory Change from 1972			
		^3H	^{137}Cs	^{90}Sr	$^{239, 240}\text{Pu}$
Arctic Intermediate Water					
0-700 m	{ 1979	—	—	—	-21
	{ 1981	+19	-6	-3	—
Upper Deep Water					
700-1400 m	{ 1979	—	—	—	+126
	{ 1981	+92	+43	+56	—
Lower Deep Water					
1400-Bottom	{ 1979	—	—	—	+43
	{ 1981	+57	0	-1	—
Total Water Column	{ 1979	—	—	—	+30
	{ 1981	+48	+4	+6	—

tween 1972 and 1981, whereas ^3H has increased by 57%. An even more marked increase (92%) is seen for ^3H in the Upper Deep Water, which is partially followed by an average increase in ^{137}Cs and ^{90}Sr of 50%. In the Arctic Intermediate Water, ^{137}Cs and ^{90}Sr actually decline, in contrast to the 19% increase in ^3H .

The contrast between the changes in time and in water mass of the ^3H distribution with those of fallout ^{137}Cs and ^{90}Sr , seem consistent with the relative change in their concentrations in surface water flowing into the region over time. ^{137}Cs and ^{90}Sr concentrations in surface Atlantic water flowing into the Norwegian Sea have declined more rapidly between 1967 and 1981 than have those of ^3H (Livingston et al. 1985a). The reason for the lesser decline in ^3H concentrations is believed to reflect its larger continuing input from the troposphere due to hydrological recycling, including ^3H derived from re-evaporation of terrestrial freshwater sources. Between 1967 and 1981 there has been an increase in the ratio of ^3H to ^{137}Cs or ^{90}Sr , in Norwegian Sea inflow water, of 55% (Livingston et al. 1985a). The Greenland Sea water column is then simply

responding to this changing input pattern with time. In 1981 the deep water is relatively more strongly enriched in tritium since the concentrations and inventories then reflect the more recently enriched surface ^3H concentrations flowing into the region. In 1972, in contrast, the deep waters were influenced by the previous several years input of ^3H in lesser amounts relative to ^{137}Cs and ^{90}Sr . The intermediate water shows a less dramatic change in ^3H relative to ^{137}Cs and ^{90}Sr , a 19% increase as opposed to a decline in fallout ^{137}Cs or ^{90}Sr of 4-5%. This is consistent with its rapid response to surface properties, and hence it reflects a later period of surface input than that seen by the deep water. Since this later period corresponds to a less pronounced relative increase in ^3H (to ^{137}Cs or ^{90}Sr) in inflowing waters, the relative changes in concentrations and inventories were correspondingly not as different as in the deep waters.

Plutonium. As noted earlier, Pu transport in the oceans has a component related to its particle association. Although only a small fraction of Pu present in open water columns is associated with particulates (Bowen et al. 1980), the movement of this

fraction towards the sea floor is geochemically important as a process affecting the distribution of Pu. The data in this paper on the changing distributions of Pu in the Central Greenland Sea between 1972 and 1979 provide further indirect evidence of Pu transport with sinking particles.

Both the increase in the inventory of Pu in the whole water column between 1972 and 1979 and the changes in the Pu distribution within the various water masses show evidence of Pu vertical transport. During this period the only significant new input of Pu came from the horizontal advection of North Atlantic water into the upper water column. Any accelerated downward transfer of Pu from this input would tend to lead to an accumulation of Pu in the region, because of this transfer to deep water with longer residence times. Therefore the 30% increase in fallout Pu in the whole water column between 1972 and 1979 would (Table 3) be an indication of this process, if no corresponding change in soluble tracers occurred simultaneously. Only fallout ^{137}Cs and ^{90}Sr data are appropriate to address this question as ^3H changes are complicated by the additional input from hydrological cycles discussed earlier. By 1981, fallout ^{137}Cs and ^{90}Sr showed only an average increase of 5% in the Central Greenland Sea so that the 30% Pu increase in 1979 is interpretable by a mechanism of accumulation through transfer to deeper water via particle sinking (and subsequent release), especially when viewed in the even stronger evidence about to be discussed on the changing distribution within the individual water masses.

When the Pu concentration profiles in 1972 and 1979 (Fig. 3) are compared, it is clear that concentrations have declined in the upper water column and increased in the lower water column during this period. The Pu inventory in the Arctic Intermediate Water has declined by 21% (Table 3) while the Upper and Lower Deep Water in-

ventories increased by 126% and 43% respectively during the same period. With the exception of the Upper Deep Water (which showed an increase of about 50%), fallout ^{137}Cs and ^{90}Sr inventories changed little from 1972 to 1981, and presumably were similar in 1979. The decline in the Pu content of the Intermediate Water during this interval is not surprising in view of the significant contribution of Atlantic water input to this water mass. Pu is relatively rapidly removed from the surface ocean by scavenging in the euphotic zone (Bowen et al. 1980). Hence Pu concentrations in the shallow North Atlantic inflow to the Norwegian Sea would be expected to decline much more rapidly than those of fallout ^{137}Cs or ^{90}Sr (Livingston et al. 1985). The resultant effect in the Central Greenland Sea is the net loss of Pu which is observed. On the other hand, such Pu as is advected into the region from the south is subject to vertical transport by ongoing active scavenging processes. The observed increases in Pu concentrations and inventories in regional deep waters in 1979 are markedly higher than the corresponding changes in fallout ^{137}Cs or ^{90}Sr . In fact, the 43% Pu increase in the Lower Deep Water even exceeds the increase in total ^{90}Sr (30%) between 1972 and 1979, so it is reasonably clear that processes involving Pu association with sinking particles and subsequent release at depth, in addition to pure physical water movements, determine the evolution of fallout Pu in the region.

CONCLUSIONS

The changing patterns with time of the radiotracers ^3H , ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ are consistent with rapid ventilation of the Greenland Sea. The tracer composition throughout the water seems to be very sensitive to changes in the tracer composition of the surface water inflow to the region. The fallout ^3H and the fallout derived fraction of measured ^{137}Cs and ^{90}Sr concentra-

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tions have distributions reflecting broadly similar input histories — but modified by the slower decline in ^3H concentrations in the regional surface inflow as compared to ^{137}Cs and ^{90}Sr concentrations changes. Increased concentrations of fallout Pu in the deep water of the Greenland Sea draw attention to the transfer of this tracer by processes involving its association with sinking particles.

At least as early as 1979, levels of ^{137}Cs had started to rise throughout the whole water column of the Central Greenland Sea in response to the input of this radionuclide (and ^{90}Sr) from the new radioactive waste source in Norwegian Coastal Waters. Deep water concentrations of ^{137}Cs from this new source were already about 20% of near surface concentrations of "new" ^{137}Cs . Given that the new source had just begun to reach the Greenland Sea, its rapid penetration throughout the water column illustrates very dramatically the short time scale of the ventilation processes taking place.

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