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A mass balance for ^{137}Cs and ^{90}Sr in the North Atlantic Ocean

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ABSTRACT

The total inventory of ^{137}Cs (3272 kCi) and ^{90}Sr (2257 kCi) in the North Atlantic Ocean in 1972, as well as the subinventories in the 0 to 1000 m, 1000 to 2000 m, 2000 m to bottom layers, continental shelf waters and bottom sediments, have been estimated. We have been careful to provide reliable estimates of uncertainty for each of these values. We have also estimated the inputs of ^{137}Cs to the Atlantic Ocean as direct fallout (2065 kCi), or as ocean current transport (240 kCi) since the start of large scale nuclear testing. The inputs are 20 to 30% less than the total inventory. We believe that the input has been underestimated by use of the traditional assumption that the mean fallout rates in 10° latitude bands are the same over the ocean as measured on land by the fallout in precipitation network. We have estimated the uncertainties inherent in using land data and find that the difference is not irreconcilable.

25% of the ^{137}Cs in the North Atlantic is found beneath the 1000 m level. If it is assumed that water, once it sinks below the 1000 m level, is not returned within decades to the surface, then based on estimates of the ^{137}Cs concentration in northern near surface waters between 1952 and 1972, the mean sinking rate of near surface water below the 1000 m level for the 20 years prior to 1972 must have been close to $14 \times 10^6 \text{m}^3/\text{s}$.

1. Introduction

The soluble, long lived fission products ^{90}Sr ($T_{1/2} = 29.0 \text{ y}$) and ^{137}Cs ($T_{1/2} = 30.2 \text{ y}$) have entered the oceans through the air sea interface, primarily as fallout from atmospheric testing of nuclear weapons. They have been found to be good water tracers (Bowen and Sugihara, 1965; Broecker, 1966; Folsom and Sreekumar, 1970; Bowen and Roether, 1973; Kupferman and Bowen, 1976; Kupferman and Livingston, 1979).

These radioisotopes have a particularly great potential usefulness in the Atlantic Ocean because their collection and measurement in subsurface water samples along with the simultaneous collection of hydrographic data have been carried out continuously since 1957. The bulk of this sample collection has been carried out by the

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Woods Hole Oceanographic Institution (WHOI), while analyses have been carried out by WHOI or associated contractor laboratories. During this same period, U.S. government agencies have maintained extensive global fallout precipitation and soil monitoring networks. Thus the rate of delivery of these tracers to the land surface is comparatively well documented. ^{90}Sr and ^{137}Cs thus offer significant advantages over the more widely used tritium and ^{14}C , since for these there exists neither so extensive a time series of data in the Atlantic Ocean, nor so substantial a basis for estimating the input function. During the period 1958 through 1963, for instance, almost no subsurface samples collected in the Atlantic Ocean were analyzed for either ^3H or ^{14}C .

Unfortunately the use of many of the earlier data concerning ^{90}Sr or ^{137}Cs , especially in the deep ocean, was inhibited by uncertainty as to the reliability of reported values; this was especially a problem because earlier evaluations of these data led to conclusions widely at variance with then-accepted models of deep ocean circulation. This situation was well summarized in Volchok *et al.* (1971, p. 68-69). Recently it has been shown that the early suspicion that much of the deep-water ^{137}Cs was attached to particles was ill-founded (Kupferman and Bowen, 1976), and a new approach has permitted the estimation of the magnitude, year-to-year variations and uncertainty of the overall analytical "blank" that characterized the work of each participating laboratory (Kupferman and Livingston, 1979). As reported there, this approach has resolved most of the disturbing conflicts among the various data sets, and it has substantially eliminated, as we discuss below, those historical tracer measurements that were utterly unreconcilable with conventional ideas about the deep circulation.

In the present paper we present a quasi-synoptic set of data concerning ^{90}Sr and ^{137}Cs distributions through the North Atlantic Ocean during 1972; all of the radiochemical data have been corrected as described by Kupferman and Livingston (1979). We attempt to balance the total quantity of ^{137}Cs and ^{90}Sr found in the North Atlantic Ocean in 1972 against estimates of the amounts introduced as direct fallout, as fallout advected from elsewhere or from other sources. It is important to have a clear understanding of the degree of consistency in these quantities before we attempt to reconstruct the most probable pathways by which these isotopes have entered the subsurface ocean.

We first estimate the total inventory of ^{90}Sr and ^{137}Cs in the North Atlantic Ocean using samples from vertical profiles collected during the 14 month period centered around November 1972. This is done by estimating the inventory in each Marsden Square (squares each of 10° latitude by 10° longitude, systematically numbered to indicate position) in three depth intervals: from the surface to 1000 m, 1000 to 2000 m, and 2000 m to the bottom. The Marsden Square format is widely used in organizing hydrographic data and the number of Marsden Squares is roughly equal to the number of profiles in our data set. This approach also serves to mini-

mize sampling error as will be discussed in subsection 2i3. The inventory in the deep ocean sediments and in the waters and sediments of the continental shelves is estimated also. We then estimate the total amount of these fission products that have been introduced directly by fallout, or by current transport or by discharge of low level wastes from nuclear facilities.

While it is relatively straightforward to make these estimates given a reasonable amount of data, it is at least as important, and far more difficult, to provide reliable estimates of uncertainty. We have tried carefully to provide this information as well.

2. Methods and calculation procedures

a. Data set. The basic data set for estimating the total inventory of ^{137}Cs and ^{90}Sr in the North Atlantic consists of 46 profiles, collected in a 14-month period in 1972-73, 25 collected as part of the GEOSECS program and by WHOI and 21 by the University of Delaware (UD). These data are supplemented by nine profiles taken outside of the base period but providing additional information in regions we felt were otherwise inadequately represented.

The locations of the profiles are shown in Figure 1. The number assigned to each profile is its serial number in the WHOI-UD combined data set. Relevant cruise, station numbers and dates are given in Appendix 1. The letters show the locations of WHOI profiles not yet in the WHOI-UD data set but used in estimating transport of ^{137}Cs and ^{90}Sr by ocean currents. Relevant information about these profiles is also given in Appendix 1. The base period chosen was one during which relatively intense sampling in the North Atlantic was carried out on GEOSECS as well as on several other cruises.

b. Collection and analysis of ^{137}Cs and ^{90}Sr from seawater. Relevant procedures for collecting and measuring ^{137}Cs and ^{90}Sr concentrations in seawater have been discussed and referenced by Kupferman and Bowen (1976). Briefly they may be summarized as follows: WHOI seawater values are obtained by laboratory extraction of bulk water samples of approximately 60 l. These samples were collected by WHOI scientists and staff or by the GEOSECS Operations Group. They were analyzed at WHOI or at commercial laboratories under contract. Comparison of WHOI and contractor analyses has been discussed by Bowen *et al.* (1974). A given water sample may be analyzed for ^{90}Sr , for ^{137}Cs or for both, as well as for transuranic elements. For use in our present calculations contractor and WHOI values for ^{90}Sr and ^{137}Cs and their uncertainties have been corrected using the Deep Ocean Blank (DOB) procedure (Kupferman and Livingston, 1979). Uncertainties so corrected are called sample uncertainties. Although it was necessary to apply the DOB procedure to WHOI values only from before the base period, since the corrections during and after the base period were so small, we refer to all the WHOI values' uncertainties also as sample uncertainties.

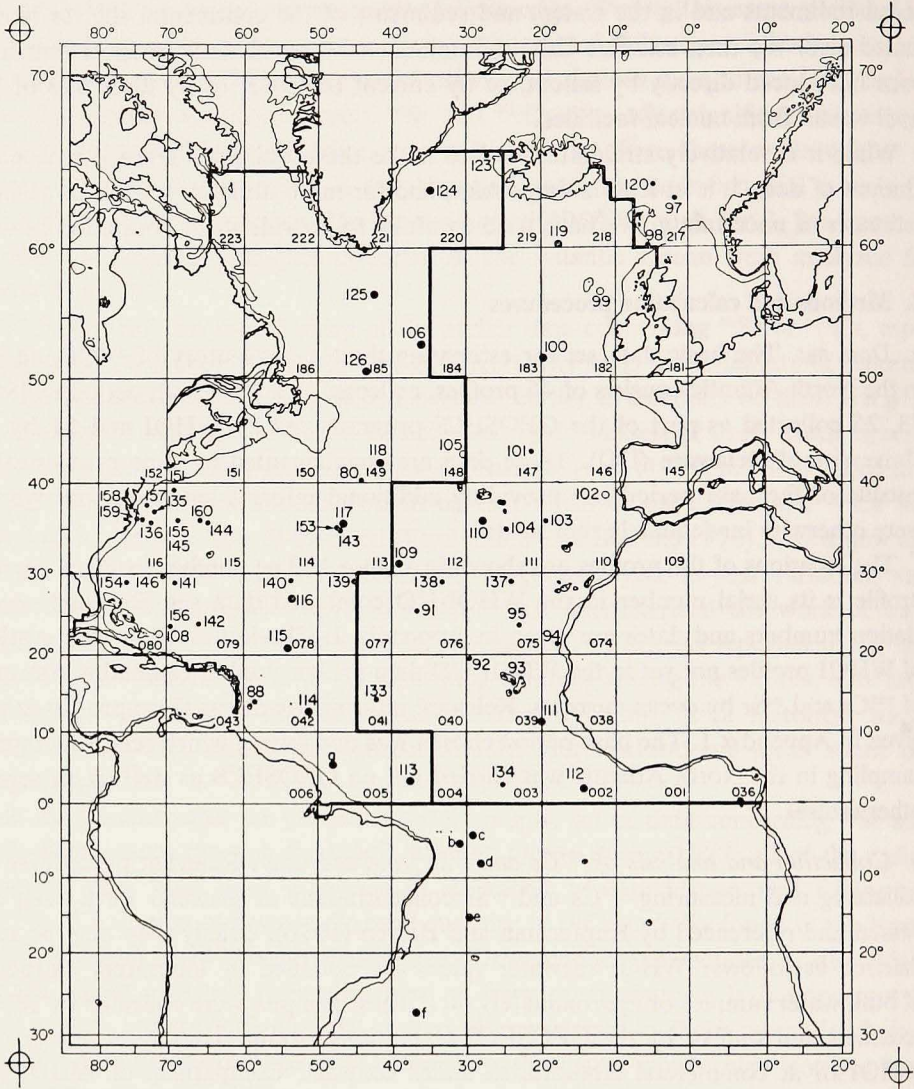


Figure 1. Chart of Atlantic Ocean. 100 fathom curve is indicated. Numbers of the Marsden Squares in the North Atlantic in lower right-hand corners. Heavy lines show the boundaries of the North Atlantic and simplified from Wright and Worthington (1970) the approximate axis of the mid-Atlantic ridge. Filled circles represent profiles used to determine inventories in all three layers; open circles, upper two layers; dots, upper layer. Letters indicate supplementary profiles used to estimate ^{137}Cs transports by currents. Cruise and station numbers for profiles are listed in Appendix 1.

The ratio of ^{137}Cs to ^{90}Sr in debris from nuclear explosions has been computed by Harley *et al.* (1965) to be 1.45. This is in excellent agreement with mean values of the ratio found in open ocean seawater (Bowen *et al.*, 1974). Therefore ^{90}Sr values (and uncertainties) were multiplied by 1.45 to make them directly comparable with ^{137}Cs values. Where both ^{137}Cs and ^{90}Sr values were available for a water sample, the $^{90}\text{Sr} \times 1.45$ and ^{137}Cs value were treated as duplicate analyses. The uncertainty in these combined values was taken to be either one half the absolute value of the difference between the two, or the propagated error (Bevington, 1969) of the sample uncertainties, whichever was larger. For a group of samples, the mean uncertainty estimated by either of the two methods is comparable (Kupferman and Bowen, 1976). *In the remainder of this paper, ^{137}Cs , $^{90}\text{Sr} \times 1.45$ and combined values will all be referred to as ^{137}Cs values, unless otherwise noted.*

University of Delaware ^{137}Cs profiles consist of samples obtained by absorption of cesium in situ (Folsom and Sreekumaran, 1970; Kupferman, 1971). The samplers are open and collect cesium in passing to and from the sampling depths as well as while held at these depths. The precision with which analytical data can be corrected for collection during passage decreases with sampler depth. The in situ profiles did not, therefore, extend substantially below 1000 m and an applicable DOB could not be determined. However, concentrations measured by UD and by WHOI have been shown to be in excellent agreement (Kupferman and Bowen, 1976) as discussed later. Uncertainties in UD values which are based primarily on counting variations are referred to as sample uncertainties also.

The units used for ^{137}Cs concentration are dpm/100(STP) liters, without correction for the compressibility of seawater. This correction, a function of sample depth and typically 1.5% with a maximum of about 3%, is negligible compared to other uncertainties, especially when the relatively small amount of ^{137}Cs in the deeper layers is considered.

c. Collection and analysis of ^{137}Cs and ^{90}Sr from sediments. Sediment concentrations and inventories of ^{137}Cs were derived from analyses of a large series of recent sediment cores from most parts of the Atlantic Ocean. These cores were collected using a 21 cm diameter gravity corer (Burke, 1968) designed for sediment retrieval for fallout radionuclide measurement. The cores were sectioned into horizontal slices either on the ship or, after freezing, in the laboratory. Portions of sections were dried and analyzed for various artificial radionuclides, e.g. ^{137}Cs , ^{90}Sr , ^{55}Fe , $^{239,240}\text{Pu}$ and ^{241}Am , using an acid leach prior to radiochemical separation and radioactivity measurement. The inventory in the sediment, for each isotope measured, was obtained by integrating the sectional activities under a unit area. ^{137}Cs concentrations in sediments are typically four times ^{90}Sr concentrations (Noshkin and Bowen, 1973; Livingston and Bowen, to be published). In estimating the amount of ^{137}Cs in the sediments we have used only ^{137}Cs values.

d. *Calculation of surface areas and volumes.* Volumes of Marsden Squares were determined by summing volumes of 1° squares whose depths were taken from a chart provided by Worthington (unpublished data). The procedure used to determine these depths is outlined in Wright and Worthington (1970). Our boundaries (Fig. 1) for the North Atlantic Ocean are the same as those used by Wright and Worthington. Volumes were also determined by Marsden Squares for regions whose depths were less than 100 m. These regions were taken to be the shelf regions. Their total area is 4% of the total area of the North Atlantic. This is in reasonable agreement with 5.6% of the area of the entire Atlantic Ocean (excluding adjacent seas) in the 0 to 200 m depth interval (Sverdrup *et al.*, 1942, p. 21).

In the boundary squares S of 40N, we apportioned the deeper volume region (beyond the 100 m isobath) between the different depth layers as if the lateral boundaries were vertical. Since the continental slope is steep in this region, this procedure did not introduce significant (i.e., greater than 1 or 2%) errors in the volumes assigned to the different layers.

A different procedure was used in apportioning volumes further north, where the ocean becomes shallower. For the regions north of 50N, we calculated the volume below 2000 m, for each Marsden Square, from the depths of 1° squares. In the 40 to 50N band, we calculated this volume from the depth of 5° squares. For all squares N of 40N, at the ocean edges, the volume in the two upper layers was apportioned assuming that the square was bounded on one side by a plane surface of constant slope, the other sides forming three sides of a rectangular parallelepiped. With this assumption, and with the volume below 2000 m, and the total volume and surface area beyond the 100 m isobath known, the volumes in the 0 to 1000 m and 1000 m to 2000 m layers were uniquely specified. For squares not near the boundaries the depth was almost everywhere greater than 1000 m. With the total volume and the volume below 2000 m known the volume in the 1000 to 2000 m layer was uniquely specified. In Appendix 2 are listed areas, volumes and depths of North Atlantic Marsden Squares, calculated in this way.

e. *Estimation of the ^{137}Cs inventory and its uncertainty, 0 to 1000 m.* (1) *Profile inventories.* The ^{137}Cs inventory at each profile, in millicuries/km² was calculated assuming that the concentration (in dpm/100 l) varied linearly between sample points. The bias introduced by using this straight line interpolation procedure was estimated in the following manner for the 0 to 1000 m layer. For each 10° latitude band we determined the profile with the greatest number and that with the least number of sample depths as well as the mean number for all profiles in the band. The ^{137}Cs inventory was calculated for the profile with the greatest number of samples in three ways: method A using all the samples; method B using only samples at those depths closest to the depths of the samples in the profile with the smallest number of sample depths; method C using a number of samples equal to the largest integer

Table 1. Effect of number of samples in profile on calculated ^{137}Cs inventory, 0 to 1000 m (inventories in mCi/km²).

Latitude Band (°N)	Mean \pm SD, Number of Samples Per Profile, 0 to 1000 m ^a	Inventory, Profile with Largest Number of Samples		
		All Samples ^b	Reduced Number ^b	Number Equal to Largest Integer Less than Mean ^{b,c}
0-10	9.7 \pm 2.0 (3)	14.0 \pm 1.8 (12)	17.2 \pm 1.8 (7) ^γ	16.5 \pm 1.7 (9)
10-20	8.2 \pm 1.1 (6)	26.1 \pm 2.3 (9)	28.7 \pm 2.4 (5) ^γ	26.7 \pm 2.6 (8)
20-30	9.9 \pm 2.3 (15)	82.1 \pm 1.9 (14)	93.5 \pm 3.6 (6) ^δ	81.1 \pm 2.7 (9)
30-40	9.6 \pm 2.3 (18)	101.4 \pm 1.7 (13)	113.7 \pm 2.9 (4) ^δ	101.9 \pm 2.1 (9)
40-50	8.3 \pm 1.9 (4)	92.7 \pm 1.8 (11)	85.9 \pm 3.2 (7) ^δ	88.9 \pm 2.4 (8)
50-60	7.4 \pm 1.5 (5)	77.0 \pm 3.2 (9)	75.6 \pm 4.1 (6) ^δ	76.7 \pm 4.0 (7)
60-66	8.8 \pm 1.0 (4)	87.1 \pm 2.2 (10)	88.6 \pm 2.8 (5) ^γ	89.2 \pm 2.2 (8)

^a Number of profiles in latitude band shown in parentheses.

^b Uncertainty is propagated sample uncertainty, number of samples shown in parentheses.

^γ Retain sample closest to surface and to 1000 m, for remainder, exclude alternate samples.

^δ Number of samples equal to number in profile with fewest samples in latitude band. Depths of samples retained chosen to correspond most closely with depths of these samples.

^ε Depths of these samples chosen to correspond approximately to mean depth distribution of samples for profiles in this latitude band.

less than the mean in that latitude band. The depths of the samples in C were chosen to correspond, roughly, to the mean distribution with depth of the samples of the profiles in the latitude band. In those 10° bands where all the profiles had about the same number of samples, we replaced method B by a procedure where alternate samples in the profile were excluded keeping the samples closest to the surface and to 1000 m.

The results of these profile inventory calculations are shown in Table 1. The uncertainty shown for each ^{137}Cs inventory is the propagated error for the calculation, based on the sample uncertainties. As can be seen, the values calculated using method A and those calculated using method B are in three of seven latitude bands somewhat (but not greatly) different. The largest difference is 14% (inventory by method B minus inventory by method A, divided by inventory by method A) and the mean difference for the three cases is 6%. For all the other possibilities tested, there is no real difference between values obtained using the maximum and less than the maximum number of samples. This suggests that the set of profiles contained sufficient samples to allow calculation of profile inventories by the linear interpolation procedure without the introduction of a significant bias. Thus the total uncertainty in a profile inventory value is reasonably well approximated by propagating the sample uncertainties.

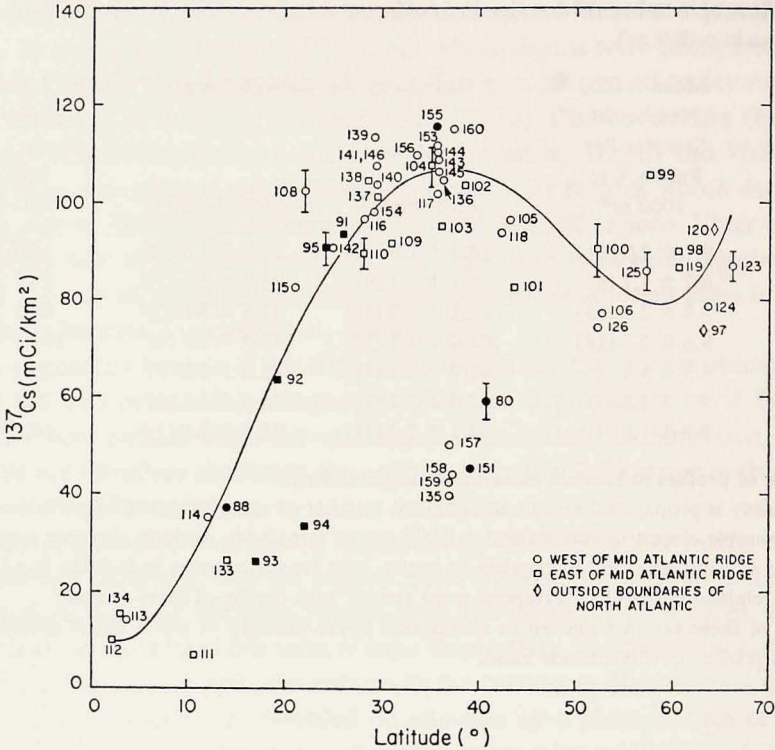


Figure 2. ^{137}Cs inventories for profiles, 0 to 1000 m. Uncertainties calculated by propagation of sample uncertainties. These are only shown when greater than ± 3.0 mCi/km² (mean uncertainty and its standard deviation for all profiles shown in figure is 2.1 ± 0.9 mCi/km²). Solid line is fourth degree least squares polynomial fit to all points except those from upwelling areas (94, 111) or northeastern U.S. slope water (80, 135, 151, 157-59). Filled symbols for profiles outside base period.

(2) *Lateral variation in profile inventories, 0 to 1000 m.* In Figure 2 the ^{137}Cs profile inventories in the 0 to 1000 m layer are plotted as a function of latitude. The filled symbols represent supplementary profiles. Since they are in most cases within one or two years of the center of the base period, we have not, in view of the near 30 year half lives of the isotopes, felt it necessary to correct the sample values for decay to the center of the base interval. They are, as are the sample values of the other profiles, corrected for decay to the time of collection (UD) or corrected for decay to the time of collection if their activity was measured more than one year after the date of collection (WHOI).

Circles represent profiles from west of the mid Atlantic Ridge; squares, profiles from east of the ridge. We made this distinction because we found significant differences in ^{137}Cs concentration in the upper thousand meters, on surfaces of constant σ_τ (approximately corresponding to surfaces of constant density), on the eastern

and western sides of the ocean. It is also known that the temperature/salinity characteristics and the lateral circulation on the two sides of the mid Atlantic ridge are different (Wright and Worthington, 1970; Worthington, 1976).

With the exception of profiles from areas affected by upwelling (94 and 111) or from the slope water off the Northeastern United States (80, 135, 151, 157-59), it is apparent that the principal variation in the ^{137}Cs profile inventories is with latitude. There are two latitude regions with significant longitudinal differences. First, profiles north of 50° and east of the ridge in the Atlantic tend to contain about 10% more ^{137}Cs than those west of the ridge. Profile 99 360 km west of Scotland contains an anomalously high ^{137}Cs inventory. It is possible that this high value reveals the effect of the discharge of low level liquid wastes by the nuclear fuel reprocessing plant at Windscale on the Cumbrian Coast of Great Britain. We return to this point in subsection 3c. Secondly, in the 10° - 20° band, profiles west of the ridge contain about 25% more ^{137}Cs than eastern profiles. A possible reason for this is that the Gulf Stream Gyre (Worthington, 1976) tends to recirculate water of higher latitude origin west of the ridge. This could also explain the anomalously high inventory in profile 108. This profile contains only six samples in the upper layer with none between 450 and 1000 m so the estimate is more likely to be in error due to non-representative sampling (however, the profile inventory is also anomalously high in the 1000 to 2000 m layer).

The solid line in Figure 2 is a 4th degree least squares polynomial fit to the data, excluding the slope water and upwelling area profiles. The minimum at 58N is probably an artifact. Comparing the spread of the data points about this curve shows clearly that the principal variation of profile inventory is with latitude. The correlation coefficient of the polynomial fit was 0.93, the standard error of the estimate ± 11.0 mCi/km².

If we knew the joint probability density function of profile inventory, latitude and longitude we could use it to find the regression surface of profile inventory on latitude and longitude (for example see Hays and Winkler, 1971, section 10.7); in practice the regression surface would be estimated from the available profile inventories using a least squares criterion. However, our ultimate interest is in the relationship between the ^{137}Cs distribution and the hydrographic structure. For this reason we prefer to focus on the values of the individual profile inventories rather than on the shape of an estimated regression surface. We have therefore chosen to estimate the layer inventory by Marsden Squares as if there were no lateral variation within a square. The lateral variation thus becomes a statistical component of the uncertainty in the square inventory which would otherwise be determined only by profile propagated sample uncertainties. We have done this by determining the interprofile variations when more than one profile was in a Marsden Square and using these as an index of uncertainty. Since the principal lateral variation in profile inventory is with latitude, we have grouped interprofile variations by latitude.

Table 2. Estimate of sampling variation in a Marsden Square, 0 to 1000 m (inventories in mCi/km²).

Latitude Range (°N)	Marsden Square Number	Number of Profiles	Mean Inventory ± SD	Mean Absolute Deviation ^a	Mean SD	Mean, Mean Absolute Deviation	Remarks
0-10	— ^β	3			3.0		
10-30	039	2	39.9 ± 19.8	14.0			Variation of inventory with latitude is large
	042	2	36.2 ± 2.1	1.5			
	075	3	91.9 ± 8.4	6.3			
	076	2	98.3 ± 7.7	5.5			
	078	3	93.7 ± 10.7	7.7			
	079	2	98.9 ± 12.8	9.1			
	080	3	102.3 ± 4.7	3.2			
30-40					9.5	6.8	Variation of inventory with latitude is small
	110	2	99.0 ± 6.3	4.5	9.2 ^γ	6.5 ^γ	
	113	3	106.7 ± 4.8	3.6			
	115	5	110.7 ± 3.9	3.2			
	116	5 ^δ	44.2 ± 4.0	2.8			
40-50					4.8	3.5	Means calculated in 10° squares, (not restricted to Marsden Squares) to minimize effect of anomalously high profile 99 in square #182.
					4.4 ^γ	3.3 ^γ	
					7.8		
N of 50°	182,218	3	94.1 ± 9.7	7.4			
	184,185	3	78.9 ± 6.2	4.6			
	182,217	2	97.5 ± 10.8	7.7			
	219,220	2	82.5 ± 5.6	4.0			
					8.1	5.9	
					8.1 ^γ	5.9 ^γ	

^a Mean absolute deviation, sometimes called average deviation $\equiv \frac{\sum |x - \bar{x}|}{N}$.

^β No more than one profile in any Marsden Square. "Mean SD" value is SD of all inventories in this latitude range.

^γ Mean of values weighted by number of profiles per Marsden Square.

^δ These are all slope water profiles.

In the 30-40° band, the variation with latitude was small, both for the slope water and non-slope water groups. In this band for each Marsden Square which contained more than one profile, we calculated the mean value, the standard deviation and the mean absolute deviation (Square #115 contained one slope water profile. This profile was included with the slope water profiles in square #116). Table 2 shows these statistics for each of the Marsden Squares with multiple profiles in this latitude band. Also shown are the number of profiles per square, the mean standard deviation and mean, mean absolute deviation, both unweighted and weighted by the numbers of profiles in the squares. In this latitude band, a single profile will on the average be within 3.5 mCi/km² of the mean profile inventory obtained by more intense sampling of the Marsden Square. In the Marsden Squares between 10 and 30N, where the variation with latitude was the most rapid, a single profile will on the average be within 7.0 mCi/km² of the mean for a square. In the region where the variation with latitude was intermediate, north of 50°, a single profile will on the average be within 5.9 mCi/km². In obtaining this last value, we have not restricted ourselves to profiles within the same Marsden Square but considered multiple profiles from within any 10° square. This was done to minimize the effect of profile 99 which had an anomalously high ^{137}Cs inventory.

In the 0-10° and 40-50° latitude bands none of the Marsden Squares contained more than one profile. In Figure 2 it can be seen that the three profiles in the 0-10° band did not show much variation in profile inventory. We have taken the standard deviation of these three values (± 3.0 mCi/km²) as the mean standard deviation for this band. We have followed the same procedure with the profiles in the 40-50° band. In this band the standard deviation was ± 7.8 mCi/km².

We have seen in subsection e(1) that the total uncertainty in a profile inventory value is reasonably well approximated by the propagated sample uncertainties. The mean of the propagated uncertainty for all profiles used in determining the amount of ^{137}Cs in the 0 to 1000 m layer is ± 2.1 mCi/km². The mean standard deviations in Table 2 are all larger than this value, most of them substantially so, indicating that the variability among profiles within Marsden Squares is larger than the uncertainties in individual profile inventories. Therefore in estimating the ^{137}Cs inventory in a Marsden Square from a single profile in that square we determine the uncertainty in this estimate from the mean weighted standard deviation for the latitude band, shown in Table 2, rather than from the propagated sample uncertainty of the profile.

(3) *Procedures for determining the ^{137}Cs inventory and its uncertainty, 0 to 1000 m.* The ^{137}Cs inventory in the 0 to 1000 m layer was calculated by 10° latitude bands in the following manner. In Marsden Squares containing more than one profile, the mean profile inventory and the standard error $\left(\frac{SD}{\sqrt{N}}\right)$ were used to determine the

total amount of ^{137}Cs and its uncertainty. For squares with one profile, the amount of ^{137}Cs in the square was determined from this profile. The uncertainty in this estimate was the weighted mean standard deviation for the latitude band from Table 2 as discussed at the end of subsection e(2). For squares that did not contain profiles, we averaged the mean profile inventories in the closest east and west squares that contained at least one profile. The uncertainties were taken to be one half the difference in the mean profile inventories of the adjacent squares. For squares near the meridional boundaries of the ocean that did not contain profiles, the inventory was determined using the mean profile inventory in the closest square in the latitude band which contained at least one profile. In these cases the uncertainty was estimated by applying the mean weighted standard deviation from Table 2 to the entire volume whose inventory was estimated using the mean profile inventory.

Nineteen out of 53 Marsden Squares did not contain profiles. Many of these were partial squares from boundary areas. Their combined volume was 24% of the total volume of the 0 to 1000 m layer.

In regions believed to contain upwelled or slope water, profiles thought to represent these areas were used to estimate inventories. Upwelling was assumed to be confined to square #038, the inventory determined by profile 111. Based on our understanding of the mean position of the Gulf Stream (U.S. Naval Oceanographic Office Chart No. 131, 1972), one third of the area of square #116 and all of squares #150 and #151 were assumed to be in slope water. Total ^{137}Cs in these areas was calculated using the appropriate slope water profiles. Uncertainties were determined in the same way as in the other regions.

The uncertainty in the estimate of the total amount of ^{137}Cs in a 10° latitude band was found by propagation of the uncertainties in the inventories for each Marsden Square or group of squares. The uncertainty for the entire 0 to 1000 m layer was found by propagation of the uncertainties for the 10° bands. These values, along with their uncertainties are shown in the second column of Table 3.

f. Estimation of the ^{137}Cs inventory and its uncertainty, 1000 to 2000 m and 2000 m to bottom. In Figures 3 and 4 mean ^{137}Cs concentrations are plotted as a function of latitude in the 1000 to 2000 m and 2000 to bottom layers respectively. These mean values were not calculated by simple averaging of ^{137}Cs concentrations but by dividing the profile inventories by the layer thickness and then converting to dpm/100 l. For this reason we will call these the weighted mean ^{137}Cs concentrations. The error bars correspond to the propagated sample uncertainties for the profiles treated in the same way. We have plotted the values in Figures 3 and 4 as concentrations rather than as profile inventories because it is more convenient to compare concentrations with sample and other uncertainties. Also, we have found that north of 50N in the 2000 m to bottom layer the profile inventory tends to be proportional to the depth, whereas weighted mean ^{137}Cs concentration is relatively independent of depth and may show more clearly the variation with latitude and longitude.

Table 3. ^{137}Cs inventory by 10° latitude bands (in kCi).

Latitude Band	0 to 1000 m	1000 to 2000 m	2000 m to Bottom	Shelf Water Column	Total in Water Column	In Sediments	Total Water and Sediments
0-10	78 ± 11	11 ± 11	12 ± 13	1.4 ± 0.3	102 ± 20	6 ± 1	108 ± 20
10-20	174 ± 22	29 ± 12	-2 ± 26	α	201 ± 36	9 ± 6	210 ± 37
20-30	668 ± 19	53 ± 15	43 ± 32	$0.1 \pm \alpha$	764 ± 40	5 ± 4	769 ± 40
30-40	704 ± 11	154 ± 23	18 ± 33	0.5 ± 0.1	877 ± 42	12 ± 4	889 ± 42
40-50	382 ± 20	144 ± 24	112 ± 13	4.1 ± 0.8	642 ± 31	19 ± 17	661 ± 36
50-60	280 ± 13	126 ± 12	90 ± 8	1.9 ± 0.4	498 ± 17	5 ± 2	503 ± 18
60-66	88 ± 6	26 ± 3	14 ± 2	0.5 ± 0.1	129 ± 5	3 ± 2	132 ± 6
Total, North Atlantic Ocean	2374 ± 39	543 ± 41	287 ± 57	8.5 ± 2	3213 ± 81	59 ± 20	$3272 \pm 84^{\beta}$ $(-260)^{\gamma}$ $\left(\begin{array}{c} +84 \\ -344 \end{array} \right)^{\delta}$

^a Less than 0.05 kCi.^b Statistical uncertainty in total inventory.^c Systematic uncertainty in total inventory.^d Combined statistical and systematic uncertainty in total inventory.

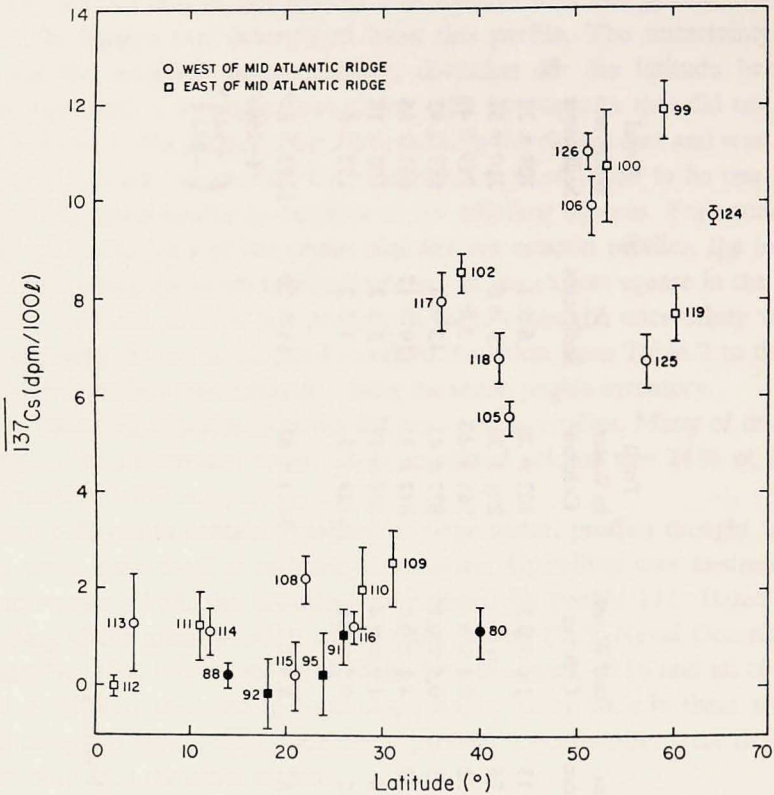


Figure 3. Weighted mean ^{137}Cs concentrations 1000 to 2000 m layer. Filled symbols for profiles before base period.

If we examine Figure 3, for the 1000 to 2000 m layer, there is, (with the exception of profile 108, which had an anomalously high value in the 0 to 1000 m layer also) no statistically significant trend in concentration with either latitude or longitude south of 30N except for a rapid increase with latitude that begins near 30N. For this reason, we do not feel that the profiles from outside of the base period, most of which lie to the south of 30N, are necessary for calculating the total burden of this, or the deeper layer which also had relatively uniform values south of 30N.

(1) *Procedures for determining the ^{137}Cs inventory and its uncertainty, 1000 to 2000 m.* The total ^{137}Cs content of the 1000 to 2000 m layer and its uncertainty were calculated in a manner similar to that used in the 0 to 1000 m layer with some slight differences in procedure, as follows.

a) Since fewer profiles were available, the Marsden Square volumes in a 10° latitude band were each assigned to the closest profile (in some cases to pairs of profiles) in that band.

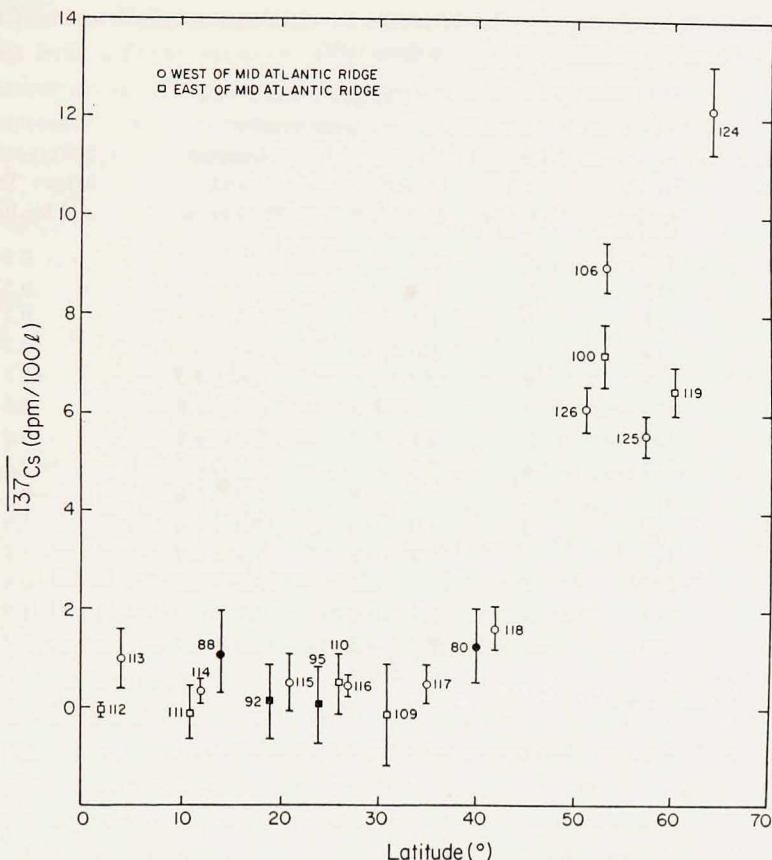


Figure 4. Weighted mean ^{137}Cs concentrations 2000 m to bottom. Filled symbols for profiles before base period.

b) For the 30 to 40° latitude band, we assumed that the mean cesium concentration varied linearly with latitude as determined by a least squares fit to the weighted mean ^{137}Cs concentrations for profiles 116, 110, 109, 117, 102, 118 and 105. The uncertainty in the total ^{137}Cs in this latitude band was determined from the standard error of the mean (i.e., standard error of the estimate divided by \sqrt{N}) of the least squares fit. The correlation coefficient was 0.81, standard error of the mean ± 0.7 dpm/100 l.

c) For Marsden Squares #221 and #222 we used the average of the weighted mean ^{137}Cs concentration of profiles 124 and 125 since 124 was in the appropriate latitude band and 125 should represent the Labrador Sea. This was done in the deeper layer also.

d) In this and the deeper layer there was no special treatment of the upwelling areas since profile 111 in an upwelling region did not appear to contain concentra-

Table 4. Effect of number of samples in profile on calculated weighted mean ^{137}Cs concentration, 1000 to 2000 m (concentrations in dpm/100l).

Latitude Band (°N)	Profile Number	Number of Samples	Weighted Mean ^{137}Cs Concentration		Concentration Difference "All" Minus "Deepest and Shallowest"
			All Samples ^a	Deepest and Shallowest ^a	
0-10	112	4	0.0 ± 0.2	-0.6 ± 0.3	0.6
	113	5	1.3 ± 1.1	0.8 ± 1.1	0.5
10-20	111	3	1.2 ± 0.7	1.0 ± 0.9	0.2
	114	3	1.2 ± 0.5	1.0 ± 0.7	0.2
20-30	115	4	0.2 ± 0.7	2.6 ± 1.5	-2.4
	116	3	1.2 ± 0.3	0.6 ± 0.6	0.6
30-40	109	3	2.6 ± 0.8	2.6 ± 0.8	0.0
	117	4	7.9 ± 0.6	8.7 ± 0.7	-0.8
40-50	118	6	6.8 ± 0.5	6.9 ± 0.6	-0.1
50-60	99	3	11.9 ± 0.6	12.9 ± 0.6	-1.0
	125	3	6.7 ± 0.5	7.8 ± 0.7	-1.1
	126	3	11.0 ± 0.4	10.5 ± 0.4	0.5
60-66	119	6	7.7 ± 0.6	6.5 ± 0.8	1.2
	124	6	9.7 ± 0.2	10.5 ± 0.4	-1.2
Mean uncertainty			± 0.6	± 0.7	
Mean ±SD of differences					-0.2 ± 1.0

^a Uncertainties calculated from propagated sample uncertainties.

tions of ^{137}Cs significantly different than the other profiles near its latitude. There was no special treatment of the slope water region either. A 1969 profile (Figure 3, 80) indicates that this may lead to a slight overestimate of the inventory in slope water. Since the volume involved is only 13% of the volume in the 30 to 40° latitude band, we feel that any overestimate will not be a serious one.

The uncertainties in the inventory calculation for the 1000 to 2000 m layer were estimated in a manner similar to that used for the 0 to 1000 m layer. The weighted mean ^{137}Cs concentration for each profile was, as in the upper layer, calculated assuming that the concentration varied linearly between sample depths.

In Table 4 the weighted mean ^{137}Cs concentrations of all the profiles (14) with more than two samples in the 1000 to 2000 m layer are compared with the weighted mean ^{137}Cs concentrations calculated using only the deepest and shallowest samples in each profile. The mean difference and its standard deviation are -0.2 ± 1.0 dpm/100 l. The standard deviation is close to what might be expected from the mean propagated sample uncertainties which are also shown in the table. This suggests that the numbers of samples in these profiles were adequate to provide un-

biased estimates of the mean ^{137}Cs concentrations since it would appear that any deviations from a linear variation with depth are, on the average, smaller than the sample uncertainties. Since the variation of ^{137}Cs concentration with depth appears to be very nearly linear, weighted mean ^{137}Cs concentrations calculated for the six profiles which had only two samples each in this layer are also likely to be unbiased estimates of the mean ^{137}Cs concentration.

We estimate the uncertainty resulting from the lateral variation in the weighted mean ^{137}Cs concentrations in the 1000 to 2000 m layer separately for different latitude regimes. The uncertainty for the 0° - 30N regime is estimated from the standard deviation of the weighted mean ^{137}Cs concentrations of all base period profiles south of 30N , with the exception of the anomalous profile 108. In doing this we are not saying that the weighted mean ^{137}Cs concentration in this region is uniform. We are assuming that the standard deviation obtained by treating these profiles as if they were drawn from a parent population with no geographical variation gives a reasonable estimate of the upper limit of the lateral variability in this region. The standard deviation of the weighted mean ^{137}Cs concentrations of these profiles is ± 0.6 dpm/100 l. The mean propagated sample uncertainty for these profiles is also ± 0.6 dpm/100 l. We therefore conclude that the total uncertainty in our estimate of the ^{137}Cs in this region can be accounted for simply by using the propagated sample uncertainty for each profile.

We have estimated in the same fashion the uncertainty resulting from lateral variation for the stations north of 35N although here it is obvious that there is a systematic variation with latitude in the weighted mean ^{137}Cs concentration values and that the assumption of a parent population with no geographical variation is not a valid one. However, as noted above this assumption simply results in an estimate of the upper limit of the uncertainty preferable, we feel, to treating the variation with latitude in a more elaborate fashion at this time. The uncertainty, estimated from the 11 relevant weighted mean ^{137}Cs concentrations north of 35° , i.e. their standard deviation, is ± 2.1 dpm/100 l. This value was used in place of the profile propagated sample uncertainties to estimate the total uncertainty in the ^{137}Cs inventory in this layer, north of 40N . For the 30 to 40° latitude band the uncertainty in the total amount of ^{137}Cs in the layer was determined from the standard error of the mean of a least squares fit as mentioned earlier. The layer inventory and uncertainty values are shown in Table 3.

(2) *Procedures for determining the ^{137}Cs inventory and its uncertainty, 2000 m to bottom.* For the deepest layer, 2000 m to bottom, our procedure for calculating the total ^{137}Cs in the layer and for assigning uncertainties was basically similar to that used in the 1000 to 2000 m layer.

In Figure 4, if the weighted mean ^{137}Cs values from the profiles in the base period are examined, there do not appear to be appreciable concentrations of ^{137}Cs south

Table 5. Effect of number of samples in profile on calculated weighted mean ^{137}Cs concentration, 2000 m to bottom (concentrations in dpm/100l).

Latitude Band (°N)	Profile Number	Number of Samples	Total Depth (m)	Weighted Mean ^{137}Cs Concentration		Concentration Difference "All" Minus "Deepest and Shallowest"
				All Samples ^a	Deepest and Shallowest ^a	
0-10	112	11	5178	-0.1 ± 0.2	0.6 ± 1.1	-0.7
	113	6	4824	1.0 ± 0.6	1.5 ± 1.0	-0.5
10-20	111	4	4954	-0.2 ± 0.7	0.2 ± 0.8	-0.4
	114	5	5073	0.3 ± 0.3	0.5 ± 0.4	-0.2
20-30	110	4	5313	0.5 ± 0.7	1.8 ± 1.0	-1.3
	115	7	5241	0.5 ± 0.6	0.4 ± 1.3	0.1
	116	9	6055	0.4 ± 0.2	0.4 ± 0.5	0.0
30-40	109	3	3646	-0.2 ± 1.0	-0.4 ± 0.9	0.2
	117	5	5021	0.5 ± 0.5	1.4 ± 1.0	-0.9
40-50	118	6	4899	1.7 ± 0.4	3.9 ± 0.9	-2.2
50-60	126	5	4310	6.1 ± 0.4	8.6 ± 0.7	-2.5
	125	5	3388	5.7 ± 0.4	6.8 ± 0.7	-1.1
60-66	119	3	2534	6.5 ± 0.5	7.6 ± 0.7	-1.1
	124	3	2444	12.2 ± 0.9	12.1 ± 0.9	0.1
Mean uncertainty				± 0.5	± 0.9	
Mean \pm SD of differences						-0.8 ± 0.8

^a Uncertainties calculated from propagated sample uncertainties.

of 40°N. However, there appears to be an east-west asymmetry with generally higher values on the west side of the mid Atlantic ridge. The profiles from which these values were obtained were all collected on GEOSECS, the western ones in September and October 1972 and the eastern ones in March 1973. 1972 Deep Ocean Blanks were used for the contractor analyses in all of these profiles, but a slight secular change (0.5 dpm/100 l) in some of the DOB values could account for the difference. This possibility has been discussed in an earlier publication (Kupferman and Livingston, 1979). Because of the difference we will treat the stations east and west of the ridge as separate populations for the purpose of calculating the total ^{137}Cs in this layer.

Figure 1 shows a simplified version of the path of the axis of the mid Atlantic ridge (simplified from Wright and Worthington, 1970). This path has been chosen to follow the boundaries of the Marsden Squares or to divide them in half. This line serves to separate the eastern and western volumes of the deep layer when assigning volumes to profiles.

Table 5, which is analogous to Table 4, provides the information required to

judge the adequacy of the numbers of samples in the profiles in the 2000 m to bottom layer to provide unbiased estimates of mean ^{137}Cs concentrations. The mean difference between using all the samples and just the top and bottom samples in the layer is -0.8 ± 0.8 dpm/100 l. The absolute value of the difference is larger than was found in the 1000 to 2000 m layer. The negative sign indicates the influence of the more northern profiles where ^{137}Cs concentrations increase with proximity to the bottom. Similar behavior may be seen in GEOSECS tritium profiles (Östlund et al., 1976) and is most probably the result of dense Norwegian Sea overflow together with entrained North Atlantic water in deeper samples. This mixture transports water with high tracer content to the proximity of the bottom in this region. The overflows are considered further in the discussion section.

We believe that the mean number of samples in the profiles we used is adequate to make a reasonably precise estimate of the total inventory. The profiles south of 40N, which have a mean ^{137}Cs concentration and standard deviation of 0.3 ± 0.4 dpm/100 l can hardly be said to seriously overestimate the ^{137}Cs concentration in the deep layer. The standard deviation represents mostly the uncertainty resulting from the lateral variation in the weighted mean ^{137}Cs concentrations in the region south of 40N. The mean of the propagated sample uncertainties for these profiles is 0.5 dpm/100 l. In view of this, it does not seem unreasonable to use the propagated sample uncertainty of each profile to calculate the total uncertainty in the deep layer south of 40N.

For the four profiles in Table 5 which are north of 50° , where the increase in ^{137}Cs concentration near the bottom is most pronounced, the mean difference between "all" minus "deepest and shallowest" is -1.2 dpm/100 l. The mean number of samples per profile in this region is four. We have represented the ^{137}Cs concentration in this region by a function which decreases exponentially with distance above the bottom, approaching a constant value and have selected the scale height of the exponential, and the bottom and constant concentrations to roughly approximate the four northern profiles. The weighted mean ^{137}Cs concentration obtained by a linear approximation to this function using four equally spaced samples is only 0.3 dpm/100 l larger than that obtained by integrating the function exactly. While we do not consider this method of estimating the bias introduced by using the linear interpolation procedure to be entirely satisfactory, it does indicate the size and sense of the expected differences. They are about the same size as the propagated sample uncertainties for these profiles and less than the uncertainty resulting from the lateral variation in the weighted mean ^{137}Cs concentration to be estimated below.

The uncertainty resulting from lateral variation in the 50-60N band is estimated from the standard deviation of the weighted mean ^{137}Cs concentrations of the profiles in this band, including 119 which is just north of 60° and 100 and 106 which with two samples each were not included in Table 5. The standard deviation, ± 1.3 dpm/100 l, will be used in estimating uncertainty in the 50-60N band and for those

volumes assigned to profile 119. We did not include profile 124 in calculating the standard deviation since this profile, at 64N is clearly dominated, in the deep layer, by the Denmark Straits overflow water and cannot be considered even approximately a member of the same parent population as the others. However, to simplify computations, an uncertainty of ± 1.3 dpm/100 l was assigned to this profile for purposes of estimating the total uncertainty of the volumes assigned to it. Since these volumes are small, this arbitrary assignment results in no serious error in the estimate of the total uncertainty in this layer.

The total ^{137}Cs in the deep layer in the 40-50N band was determined from a linear least squares fit to the mean weighted ^{137}Cs concentrations for profiles 109, 117, 118 and the mean of the profiles in the 50-60N band, taken to be located at 55N. The total uncertainty for the 10° band was determined using the standard error of the mean of the least squares fit (all the profiles in the 50-60° band being counted as one point). The correlation coefficient was 0.98, standard error of the mean ± 0.4 dpm/100 l. The layer inventory and uncertainty values are shown in Table 3.

g. Estimation in shelf waters of the ^{137}Cs inventory and its uncertainty. The total ^{137}Cs in shelf areas ($D \leq 100$ m) was calculated by Marsden Squares assuming that these areas were well mixed vertically and horizontally and that their mean ^{137}Cs concentration was equal to the surface ^{137}Cs concentration of the profiles closest to the shelf region. This procedure was found to estimate ^{137}Cs concentrations within 20% of mean values in the Middle Atlantic Bight shelf (Garfield, 1978, Table 5). Shelf concentrations estimated in this way are sufficiently precise in view of the small relative contribution of the shelves to total inventory. We have arbitrarily used 20% from our experience with Middle Atlantic Bight values as the uncertainty for each shelf square, for the total shelf inventory in each 10° latitude band and for the total shelf contribution to the North Atlantic inventory. These values are also shown in Table 3.

h. Estimation in bottom sediments of the ^{137}Cs inventory and its uncertainty. Since the total ^{137}Cs concentration in the sediments is only about 2% of the amount in the water column we have adopted the following procedure for estimating the amount in the sediments, and the uncertainty in this value.

We have averaged the total ^{137}Cs inventory from cores beyond the shelf collected in 10° latitude bands after 1969. These average values and their standard deviations were multiplied by the areas of the ocean deeper than the 100 m isobath to obtain the deep ^{137}Cs sediment inventory and its uncertainty.

We have averaged the total ^{137}Cs inventory from all cores on the continental shelf in 10° latitude bands also, except for those contaminated by local discharge of radioactive wastes. These mean values and standard deviations were multiplied by the shelf area to obtain the total ^{137}Cs inventory in shelf sediment and its uncertainty.

The deep and shelf values were then combined and are shown in Table 3. Data from 41 cores were used in making these estimates (Noshkin and Bowen, 1973; Livingston and Bowen, 1979; Livingston, Bowen and others, to be published).

i. Other sources of uncertainty. Systematic uncertainties are attributable to differences in instrument calibration and analytical procedures in the laboratories analyzing the samples and to problems in estimating the DOB's for the different laboratories. Uncertainty is also produced by the nonrandom distribution of profiles in relation to the systematic variation in profile inventory with latitude and longitude. These sources of uncertainty will be considered in the following subsections.

(1) *Uncertainty resulting from differences in calibration and procedure.* The WHOI laboratory has participated in a number of international intercalibration studies. In these studies (ILMR, 1976a; ILMR, 1976b; also see results reported in Kupferman and Bowen, 1976) which included surface Sargasso Sea water, water collected in the vicinity of a nuclear installation and seawater contaminated with fuel reprocessing wastes, WHOI values were typically within 3% of "consensus values." We believe that "consensus values" which are simply statistical means of those data which are not ruled out by Chauvenet's test are often appreciably biased and that Woods Hole values are likely to be closer to the true values. This confidence is reinforced by general very close correspondence between values reported by WHOI and such laboratories as those at Monaco, HASL, and Lawrence Livermore. The correspondence between WHOI and UD results will be discussed below.

An interlaboratory comparison between WHOI and the principal contractor laboratory producing data for this study (Bowen *et al.*, 1974), using Woods Hole Harbor water, indicated that this laboratory's DOB corrected ^{90}Sr values were 9.0% higher and ^{137}Cs values 2.2% lower than WHOI values. This laboratory analyzed approximately twice as many samples as WHOI and produced ^{90}Sr analyses for all, and ^{137}Cs analyses for half of these samples. Therefore, it might be expected that the total bulk water analysis data set, with ^{90}Sr converted ($\times 1.45$) to equivalent ^{137}Cs would have a mean ^{137}Cs value 3% higher than if WHOI had performed all of the analyses. UD in situ analyses were typically 3.2% smaller than WHOI bulk analyses in a recent comparison of seawater data (Kupferman and Bowen, 1976). Since that comparison, the UD gamma ray spectrometer was recalibrated using ^{137}Cs standard *HASL 16 August 1973* (obtained from H. L. Volchok, Department of Energy, Environmental Measurements Laboratory, 376 Hudson Street, New York, NY 10014). The new calibration indicated that UD ^{137}Cs values be increased by 5.6%. This would make the altered UD values used here 2.4% larger than WHOI values.

This brief discussion of interlaboratory comparison indicates that systematic differences between the laboratories in this study resulted in a data set whose mean

^{137}Cs value is slightly less than 3% larger than if all the data used had been obtained from WHOI bulk water analyses.

(2) *Uncertainty resulting from the use of the DOB procedure.* The systematic uncertainty resulting from applying the DOB procedure to the contractor data is estimated in the following way. In determining DOB's it is assumed that ^{90}Sr and ^{137}Cs concentrations in the tracer-free volume are below the minimum detectable limits and probably of the order of 0.05 dpm/100 l (Kupferman and Livingston, 1979). The tracer-free volume up to 1973 was below 1000 m and N of 40S in the South Atlantic and below 2000 m and S of 30N in the North Atlantic. The total inventory of ^{137}Cs below 2000 m and S of 30N in the North Atlantic, determined from Table 3 is 53 ± 43 kCi (the uncertainty is the propagated uncertainty for the 10° bands represented). This is equivalent to a mean ^{137}Cs concentration in this region of 0.26 ± 0.22 dpm/100 l. Thus it would appear that the DOB procedure results in corrections that are of the order of 0.26 dpm/100 l too small in this region. It is unlikely that the mean concentration in this region actually is this large since WHOI ^{90}Sr and ^{137}Cs analyses from the region (Kupferman and Livingston, 1979) result in a mean ^{137}Cs concentration of -0.05 dpm/100 l where the standard deviation of these WHOI values is about 0.5 dpm/100 l and the standard error of the mean about ± 0.1 dpm/100 l. The problem arises because the analyses values which were used to determine the contractors' DOB were, in the South Atlantic, systematically smaller than in the North Atlantic. This has resulted in a DOB value that is slightly too small in the North Atlantic and slightly too large in the South Atlantic. (There also appears to be an east-west trend, which was discussed earlier.) If it is accepted that the DOB procedure has resulted in a mean value of the ^{137}Cs concentration for the entire North Atlantic that is too high by 0.26 ± 0.22 dpm/100 l, then the total inventory in the North Atlantic has been overestimated by 160 ± 135 kCi or about $5.0 \pm 4.2\%$. This is certainly an upper limit since UD values to which this DOB based estimate does not apply were used in the 0 to 1000 m layer but not in the deeper layers so that values in the entire North Atlantic cannot be said to be high by 0.26 dpm/100 l.

It would appear that the systematic errors discussed here and in (1) could result in an overestimate in the total amount of ^{137}Cs in the North Atlantic Ocean of no more than 8% or 260 kCi.

(3) *Uncertainty resulting from sampling error.* When a population is sampled an estimate of any population parameter calculated from this sample will differ from the mean value obtained by averaging the estimates obtained from a large number of samples. This difference is known as the sampling error. If a sample is not sufficiently representative of the population and if this possibility is not taken into account in the procedure used for calculating the estimate the sampling error can be appreciable.

We have estimated the total amount of ^{137}Cs in the North Atlantic from a sample consisting primarily of 55 ^{137}Cs profiles. We have attempted to minimize the sampling error by dividing the ocean into Marsden Squares and estimating the total from the amount in each square. This amounts to using a stratified sampling plan although our sample was given, not selected with such a plan in mind. Any residual sampling error was roughly taken into account by using the uncertainty in profile inventory due to lateral variation to estimate the total statistical uncertainty in the ^{137}Cs inventory in Marsden Squares (for example, see 2e(2)). It is possible to make an explicit estimate of sampling error. To do this we have made the following two assumptions: 1) The principal variation of weighted mean ^{137}Cs concentration is with latitude. This is true in the two deeper layers and in the surface layer also if we exclude the slope water and upwelling areas. 2) For any layer the weighted mean ^{137}Cs concentration can for each 10° latitude band be reasonably well approximated by a linear function of latitude. With these assumptions, the sampling error in a layer in a 10° latitude band depends only on the slope (that is the change in weighted mean ^{137}Cs concentration with latitude) of the linear function and on the mean latitude of all the profiles in the latitude band (where the mean is a weighted one, weighted by the relative amount of volume assigned to each profile in the band). If the slope is zero or the mean latitude of the profiles is the mid latitude of the band, the sampling error will be equal to zero. The amount of sampling error depends on the degree of departure of the slope from zero and of the mean latitude from the mid latitude. We have determined mean latitudes as indicated above and the slopes by comparing the mean ^{137}Cs concentration in a 10° latitude band with those in adjacent bands (This mean was calculated from weighted mean ^{137}Cs concentrations of the profiles weighted by the relative amount of volume assigned to each). We have used these quantities to estimate the sampling error in each 10° band with the exception of the 30-40N band in the 1000 to 2000 m layer and the 40-50° band in the 2000 m to bottom layer. These bands were excluded since their inventories were calculated initially from a linear regression of weighted mean ^{137}Cs on latitude.

We have summed the calculated sampling errors for the 10° bands to find the sampling error for each layer and for the entire North Atlantic. The values are: 0 to 1000 m layer, 31 kCi; 1000 to 2000 m layer, -13 kCi; 2000 m to bottom, -16 kCi; entire North Atlantic water column, 2 kCi. These estimates in all cases are smaller than the comparable statistical uncertainties shown in Table 3. This indicates that using uncertainties due to lateral variation to estimate total statistical uncertainty was an appropriate procedure.

j. Total amount of ^{137}Cs in the North Atlantic and the uncertainty in this value. The component and total ^{137}Cs inventories and their statistical uncertainties are shown in Table 3. If the total statistical uncertainty is combined with the systematic uncertainties estimated in subsections i(1) and i(2) we arrive at the asymmetric estimate of total uncertainty shown in the table.

3. Procedures for estimating the inputs of ^{137}Cs to the North Atlantic and the uncertainties in these inputs

We wished to compare the estimates of the amounts of ^{137}Cs in the ocean in 1972, shown in Table 3, with additions and losses to the North Atlantic to see how well these account for the total amount found. It was therefore necessary to estimate the inputs from direct fallout on the ocean surface, advection (including river input) and the disposal of radioactive wastes containing ^{137}Cs and ^{90}Sr in forms that make them readily available to the sea water or open ocean sediments.

a. Direct fallout. Direct fallout has been the largest single input to the ocean. ^{90}Sr and ^{137}Cs enter the ocean in precipitation either in a dissolved state or on particles that make them available, in the dissolved state very soon after entry. This has been inferred, for ^{90}Sr , from direct observations in rain (Martel, 1956) and for ^{137}Cs on the basis of indirect evidence relating to the similarity of the $^{137}\text{Cs}/^{90}\text{Sr}$ ratio in fallout debris and in ocean waters, and the generally similar nonrefractory chemical characteristics of these isotopes in fallout (Volchok *et al.*, 1971, p. 71, 2).

The traditional method of estimating the amount of fallout into the oceans has been to assume that average input rates in 10° latitude bands are the same over the ocean as measured on land by the U.S. global precipitation sampling network (for example, Feely, 1977). In 1976 there were 72 stations in this network. The first stations began operating in 1954. The number and location of stations have varied. During the period when the input rate was greatest (1961-65) there were typically 140 to 150 stations. ^{90}Sr is one of the principal fission products that this network monitors.

Except for a few special experiments it has not been possible to measure directly the amount of fallout into the oceans. The majority of such direct measurements were made on ocean weather ships in the North Atlantic Ocean. They appear to indicate that there is no systematic bias toward increased fallout of ^{90}Sr into the oceans, compared to land (Volchok, 1973).

We have calculated the input of ^{137}Cs ($^{90}\text{Sr} \times 1.45$) as direct fallout into the North Atlantic Ocean, to the end of 1972, by 10° latitude bands using data from the over-land precipitation network (Volchok and Toonkel, Tables 2 and 3). This is shown in Table 6. We have not included the area of the Caribbean or Gulf of Mexico in the area of the North Atlantic. However, part of the return flow of the Gulf Stream Gyre passes through these seas so it is possible that some of the fallout deposited there has been advected into the North Atlantic proper. This would increase the input to the North Atlantic by no more than 169 kCi, or about 8% of that shown in Table 6. Recent measurements in the Caribbean Sea show that the total ^{137}Cs inventory per km^2 is about what would be found if the Caribbean were completely closed (Bowen *et al.*, to be published). It would appear that at any time, the balance between N. Equatorial and S. Equatorial Current fallout concentrations supplied to

Table 6. Input of ^{137}Cs to North Atlantic as fallout^a.

Latitude Band	^{137}Cs (kCi)
0-10	144
10-20	183
20-30	364
30-40	480
40-50	490
50-60	334
60-66	70
Total to North Atlantic Ocean	2065

^a To the end of 1972 assuming that rate in a 10° latitude band is the same as that determined from precipitation network on land. Inputs in mCi/km^2 ^{90}Sr for 10° latitude bands calculated from Volchok and Toonkel 1974, converted to ^{137}Cs by multiplying by 1.45.

the Caribbean results in an inventory that closely approximates that predicted (using Volchok and Toonkel, 1974) from direct fallout to its surface. The Gulf of Mexico might be expected to behave similarly. For this reason, we have not included inputs to these two adjacent seas in the total input to the North Atlantic.

b. Transport by circulation processes and river water. The principal transports of water into and out of the North Atlantic and their relation to its structure have recently been discussed (Worthington, 1976). We will use the information contained in this monograph plus river-flow information, along with ^{137}Cs concentration data, to estimate the net movement of ^{137}Cs into the North Atlantic in 1972 by these transports.

In Table 7 are listed the seawater transports into and out of the North Atlantic, based on Worthington (1976). We have given the temperature range of the layer of the ocean to which the water is delivered (for inflows), or which is the origin of the water for the transport (for outflows). The source or destination is mentioned also. The East Greenland Current and the flow through the Canadian Archipelago are believed to provide the source waters for the Labrador Current (Worthington, 1970). We have used these currents in preference to the Labrador Current transport used in Worthington (1976), (the transport values are the same), because the distribution of our ^{137}Cs profiles is more favorable for estimating ^{137}Cs concentrations in the East Greenland Current. We assume that the mean ^{137}Cs concentration in the Current through the Canadian Archipelago is equal to that in the East Greenland Current since their sources (the Arctic Ocean) are believed to be the same (Dorsey and Peterson, 1976).

The transports in Worthington's (1976) monograph are known with varying degrees of precision. Most of the values he gives are in good agreement with other estimates. His transports are constrained by the requirement that they leave con-

Table 7. ^{137}Cs Transports into and out of the North Atlantic Ocean in 1972.

Sense of Flow	Rate ($10^9\text{m}^3/\text{Sec}$)	Temperature	Source (In) or Destination (Out)	Characteristics	$^{137}\text{Cs} \pm \text{SD}^{\text{f}}$ (dpm/100 l)	Stations	^{137}Cs Transport (kCi/yr) $^{\text{h}}$
		Range of Layer in N.A. ($^{\circ}\text{C}$)		Used in Selecting ^{137}Cs Samples		Selected From $^{\circ}$	
In	5	>17	South Atlantic	$\sigma_T < 26.6\%_{\text{oo}}^{\text{a}}$	$11.8 \pm 1.5(9)$	113, c, d	8.4
	1	7-12	South Atlantic	$T = 4.0 \text{ to } 7.2^{\circ}\text{C}^{\text{b}}$	$1.1 \pm 0.4(8)$	e, f	0.2
	1	<2	South Atlantic	$T < 2.0^{\circ}\text{C}^{\text{g}}$	$0.8 \pm 1.0(4)$	114, 113, b	0.1
	3	12-17	E. Greenland and	$S < 34.7\%_{\text{oo}}^{\text{b}}$	$55.8 \pm 7(4)^{\text{r}}$	123	31.9
	1	7-12	Canadian Archipelago Currents				
	4	<4	Denmark Strait	$T = 0 \text{ to } 1.5^{\circ}\text{C}^{\text{e}}$	$17.6 \pm 5.3(3)$	123	10.1
	2	<4	Iceland-Scotland Ridge	$T = 0 \text{ to } 1.5^{\circ}\text{C}^{\text{e}}$	$18.3 \pm 6.0(4)$	97, 120	5.2
	1	12-17	Mediterranean Sea	$D = 450\text{-}750 \text{ m}^{\text{t}}$	$12.4 \pm 7.6(2)$	86	1.8
0.25 $^{\text{v}}$	—	Rivers		$96.4 \pm 43.0^{\text{v}}$	—	3.4	
					Total In	61.5	
Out	8	<4	South Atlantic	$T < 4^{\circ}\text{C}^{\text{g}}$	$0.4 \pm 0.7(6)$	113, a	0.4
	1	4-7	Norwegian Sea	$T = 4 \text{ to } 7^{\circ}\text{C}^{\text{t}}$	$19.7 \pm 1.6(12)$	124, 125	2.8
	8	7-12	Norwegian Sea	$T = 7 \text{ to } 12^{\circ}\text{C}^{\text{k}}$	$23.7 \pm 0.9(22)$	98, 99, 119	27.1
	1	12-17	Mediterranean Sea	$T = 12 \text{ to } 17^{\circ}\text{C}^{\text{l}}$	$33.1 \pm 4.6(6)$	102	4.7
					Total Out	35.0	

^a Sverdrup *et al.*, 1942, p. 639.

^b Temperatures chosen so σ_θ of water from stations used is equal to σ_θ of water from the western North Atlantic with T in the range 7 to 12°C.

^c Worthington, 1976, p. 30, 31.

^d Sverdrup *et al.*, 1942, p. 65; Worthington, personal communication.

^e Worthington, 1970; Worthington, personal communication.

^f Roether and Weiss, 1975.

^g Estimated from Malkus, 1962, p. 137, Table 10, assuming one half of continental runoff to the Atlantic enters the North Atlantic.

^h Worthington, 1976, p. 38.

ⁱ *Ibid.*, p. 61.

^k *Ibid.*, p. 67.

^l *Ibid.*, p. 74.

^m Uncertainty in these transports and in total transport assumed to be $\pm 50\%$, see text.

ⁿ Estimated from ^{90}Sr and ^{137}Cs in New York City Tap Water (HASL 321, 1977, Appendix, pp. 65, 66). Since this is reservoir water, we assume this value to be indicative of the concentration in river water. This is probably an upper limit. Uncertainty = $1/2|^{137}\text{Cs} + ^{90}\text{Sr} \times 1.45|$.

^o () = number of samples averaged.

^p For station locations, see Figure 1. Cruise and station numbers in Appendix 1.

^q Weighted average from depth profile based on estimate (V. Worthington, personal communication, 1977) that 75% of the transport of the East Greenland Current is composed of water with $S < 34\text{‰}$. The upper and lower limits in the ^{137}Cs uncertainty are calculated assuming that 100% and 50% of the transport is composed of water with $S < 34\text{‰}$ respectively.

stant the volume of water in each of five layers and in the entire North Atlantic. We have assumed that the long term mean value of each of these transports is within 50% of the value given by Worthington, shown in Table 7. In view of the agreement with other estimates and the constrained internal consistency, we feel that this is reasonable and very possibly an overestimate of the uncertainty.

We have also included river input in Table 7. This transport, although small, is of interest as a possible source of fission products originally deposited on the land (Bowen *et al.*, 1974; Bowen *et al.*, 1970). Worthington, who balanced his transports only to the nearest $10^6 \text{ m}^3/\text{sec}$, did not include this input.

(1) *Estimate of transport in 1972.* We have adopted a simple and brief procedure which is reasonable given the precision with which each of the individual water transports is known. We chose ^{137}Cs profiles which contain water samples characteristic of each transport and have taken the values in these samples to derive the mean concentration of ^{137}Cs in the transport. The standard deviation we assume is an estimate of the uncertainty in this mean value. Since this uncertainty was for the principal transports much smaller than 50% of the mean value, we ignored it in assessing the overall uncertainty in the amount of ^{137}Cs carried by each transport. We assumed *this* to be determined predominantly by the uncertainty in the water transport values.

Figure 1 shows the locations of the profiles. The letters represent profiles not included in the WHOI-UD combined data set. Cruise and station numbers for these profiles are also given in Appendix I. The profiles used are listed in Table 7 as are the ^{137}Cs values, the amounts of ^{137}Cs transported in 1972, the principal characteristic used in selecting the samples, the references used in selecting these characteristics and the number of samples used in determining each mean. Some of the samples we used were collected in 1968. We do not think that their use significantly affects the uncertainties of the transport estimates.

From Table 7 we can see that the major transport of ^{137}Cs into the North Atlantic was from the East Greenland-Canadian Archipelago currents. These currents carry water containing high concentrations of fallout nuclides from the Arctic Ocean (Dorsey and Peterson, 1976). The Denmark Strait overflow runs a distant second although, along with the Iceland-Scotland ridge overflows, it is the principal direct source of ^{137}Cs to the deeper layers from outside of the Atlantic.

The principal transport of ^{137}Cs out of the North Atlantic is by the flows into the Norwegian Sea.

Taking all transports into consideration, we calculate a net transport of 35 kCi of ^{137}Cs into the North Atlantic Ocean in 1972 (Table 7).

(2) *Estimate of transport since the start of large scale nuclear weapons testing.* As was mentioned in the previous subsection, the East Greenland-Canadian Archipelago currents were, in 1972, responsible for the major input of ^{137}Cs to the North

Atlantic while the flows into the Norwegian Sea were responsible for the principal outputs. All of these currents are essentially near-surface currents. At the boundaries of the North Atlantic, the highest ^{137}Cs concentrations are found in the near-surface layers. Since fallout enters the oceans at the surface, this is not an unexpected result; it indicates moreover that these currents which were in 1972 the principal transporters of ^{137}Cs , were probably generally the principal transporters of ^{137}Cs .

Bowen *et al.* (1968) have shown evidence of a transient increase in ^{90}Sr transport by the South Equatorial Current (and probably by the East Greenland Current). In 1963, the ^{90}Sr concentration in the 0-10°N latitude band increased by about 62 dpm/100 l and remained at the higher level for about six months. If we assume that the entire transport from the South Atlantic above 17°C (Table 7) contained this increased concentration of ^{90}Sr , the additional input of ^{137}Cs to the North Atlantic would have been 32 kCi. This is more than an order of magnitude smaller than the transport into the North Atlantic that is estimated later in this section. Since only a part of the transport from the South Atlantic is South Equatorial Current water, we concluded that this, and similar transient events, have resulted in a negligible net transport of ^{137}Cs into the North Atlantic.

We will attempt to estimate the ^{137}Cs carried by the principal transporting currents in the years previous to 1972 and use these to calculate the net transport of ^{137}Cs into the North Atlantic since the start of large scale nuclear weapons testing.

The Danish government has collected surface water samples in mid-summer yearly since 1962 in the waters along the east coast of Greenland (Aarkrog *et al.*, 1963; Aarkrog and Lippert, 1964, 1965, 1966, 1967, 1968, 1969, 1970, 1971, 1972, 1973, 1974). The three locations where samples are collected are Danmarks-havn (77N, 19W), Angmakssalik (66N, 39W), and Prins Christians Sund (60N, 41W). Except for some extremely high values in the summers of 1963 and 1964, (over 100 dpm/100 l ^{90}Sr) which could be attributed to the high rate of fallout in these years, concentrations of ^{90}Sr in water of near oceanic salinities (27‰ and above) have remained nearly constant at 52.2 ± 6.5 dpm/100 l (Fig. 5) for the years 1962 through 1972. Aarkrog and Lippert (1966) reported an uncertainty of $\pm 15\%$ for their ^{90}Sr values for 1965. We have used this percentage uncertainty for the uncertainties in the Danish values after 1965, shown in Figure 5. The mean salinity in the 10 Danish samples averaged was $29.9 \pm 2.3\%$. Satisfactory experience in intercalibrations between the Danish AEC laboratory, RISØ and WHOI lends confidence to our use of these values. The Danish mean value furthermore is close to the ^{90}Sr value found in the 7 m samples at GEOSECS station 14 (65°55.7'N, 27°27.0'W) of 51.3 ± 2.3 dpm/100 l at salinity 30.054‰ and in the 83 m sample, of 47.9 ± 0.2 dpm/100 l at salinity 33.102, in August of 1972, also shown in Figure 5. This station was used in estimating the transport of ^{137}Cs by the East Greenland-Canadian Archipelago Currents shown in Table 7. We will therefore assume that the ^{137}Cs transport for these currents in 1972 shown in Table 7 was

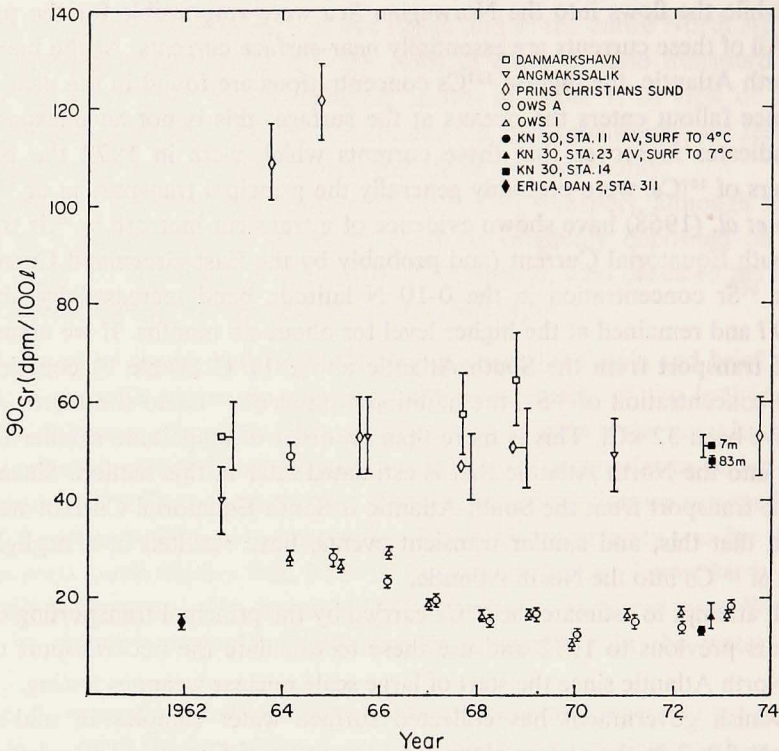


Figure 5. Surface and near surface ^{90}Sr concentrations as a function of time at stations along the east coast of Greenland and in the northern North Atlantic. For locations of stations and for salinities see text.

close to the annual mean transport since 1962. This would give a total input for the period 1962 through 1972 of 310 kCi (decay corrected to 1972). A near surface (DOB corrected) ^{90}Sr concentration measured at Erika Dan station 311 in March of 1962 at $59^{\circ}31'N$, $43^{\circ}32'W$ in the vicinity of Prins Christians Sund was 15.3 ± 2.5 dpm/100 l, at salinity 33.806 (Fig. 5). While this low ^{90}Sr value could be the result of winter mixing, the salinity is indicative of surface water. Since the major fallout input transient began in 1961, we will assume that this low value was characteristic of concentrations in the East Greenland Current around that time, and that the transport of fission products before 1962 was small compared to the transports afterward.

We also assume that the amount of transport before 1962 is related to the total amount of transport in the same proportion that the cumulative input of global fallout in 1961 is related to cumulative input in 1972 (see Volchok and Toonkel, 1974, Table 4). The pre-1962 quantity we decay corrected to 1972 as if the entire

transport in the earlier years occurred in 1956. Thus we estimate a pre-1962 ^{137}Cs transport of 90 kCi, and total of 400 kCi.

We will also assume that the total input by currents and rivers was always in the same proportion as it was to the East Greenland-Canadian Archipelago input in 1972. This is probably an overestimate of the other inputs since surface currents must respond more quickly to the input of fallout than do the overflows. With this assumption, the estimated total input to the North Atlantic by currents and rivers since the start of large scale nuclear testing, decay corrected to 1972 is 770 kCi ^{137}Cs .

Figure 5 also shows the minimum winter surface water ^{90}Sr concentration at ocean weather stations (OWS) A (62N, 33W) and I (59N, 19W), those closest to the flows from the North Atlantic into the Norwegian Sea. ^{90}Sr samples were collected at OWS on a monthly basis for WHOI since the early 1960's (Bowen *et al.*, 1968). Also shown in Figure 5 is the mean ^{90}Sr concentration (effective $^{137}\text{Cs}/1.45$) at the GEOSECS stations closest to these OWS. For station 119, (km 30 Sta. 23) closest to OWS I, the eight samples from the surface to the 7°C isotherm were averaged. For station 124 (km 30 Sta. 11), closest to OWS A the seven samples from the surface to the 4°C isotherm were averaged. The temperatures correspond to those thought to be the lowest in the water entering the Norwegian Sea, from the respective areas (Worthington, 1976, p. 61, 67). The uncertainties shown for these mean values are the standard deviations for the samples averaged.

The mean values are very close to the OWS surface values for 1972. This agreement, and the slope of the post 1966 curve of OWS ^{90}Sr vs. time indicate that vertical mixing in this region of the ocean is relatively rapid, and that the surface OWS values are valid indicators of the mean ^{137}Cs concentration entering the Norwegian Sea from the North Atlantic, except perhaps during the high fallout years in the early 1960's. For the years 1962-65, we believe the better approximation is to estimate transport from the mean ^{90}Sr concentration of the winter minimum in 1965, 1966 and 1967. With these assumptions, and treating the outflows before 1972 in the same fashion as we treated the inflows, the total outflow of ^{137}Cs from the North Atlantic due to currents, since the start of large scale nuclear testing, decay corrected to 1972, is 530 kCi.

The net transport by currents and rivers is 240 kCi *into the North Atlantic*. We assume the uncertainty in this value is the same as the uncertainty assumed for the individual currents, $\pm 50\%$ or ± 120 kCi. Alternatively, if we assume that the mean ^{137}Cs concentration in the inflow is uncertain by 25%, with the same uncertainty for the mean concentration in the outflow, then the uncertainty in the net input, based on the propagated uncertainty in the input and output transports of ^{137}Cs is ± 230 kCi. The 1972 uncertainties in the input and output ^{137}Cs concentrations are in general about 10% (Table 7). We can only approximately estimate the uncertainties inherent in extending our 1972 estimate back to the start of large scale nuclear test-

ing. We feel that ± 200 kCi is not an unreasonable value for the uncertainty in the net transport of ^{137}Cs , and will use this in further discussions.

c. Discharge of low level wastes from nuclear facilities. Input of soluble fission products comes from the discharge of low level liquid wastes by nuclear facilities. The largest known contribution has come from the nuclear reprocessing plant at Windscale on the Cumbrian Coast of Great Britain. This plant has had the largest known reprocessing capacity during the period 1964-1975 (Franklin, 1975). Through 1972, the total input of ^{137}Cs was 4% of the total ^{137}Cs in the North Atlantic (Kupferman *et al.*, 1979). It is of interest to note that the $^{137}\text{Cs}/^{90}\text{Sr}$ ratio during 1974-76 was close to 11 in contrast to the ratio of 1.45 found in samples collected in the open North Atlantic and about 2 in the earlier discharges. This anomalously high ratio should make the Windscale effluent easy to identify in regions where dilution with water having the normal ratio is not too great. The presence of ^{134}Cs ($T_{1/2} = 2.07$ y) in the effluent can also be used to identify this source (Livingston and Bowen, 1977).

Most of the material released by Windscale has not entered the open North Atlantic directly but has been carried around the north coast of Scotland, through the North Sea and into the Norwegian Sea by the coastal currents (Kupferman *et al.*, 1979).

As mentioned in a previous section, the anomalously high ^{137}Cs inventory in profile 99 (an anomaly of about 15 mCi/km²) 360 km west of Scotland in the 0 to 1000 m layer may be due to that portion of the discharge from Windscale which has entered the North Atlantic directly. A plot of isolines of surface ^{137}Cs concentration west of Scotland in 1974 (Hetherington, 1976) indicates that increased ^{137}Cs concentrations associated with the Windscale discharge have on occasion reached the vicinity of station 99 at 57°30'N 12°03'W.

Since the total release of ^{137}Cs by Windscale through 1972 was only 4% of the total in the North Atlantic and since most of this has not entered the North Atlantic directly, we will not include the input of this or of other nuclear facilities. The inclusion of profile 99 in assessing the total amount of ^{137}Cs in the 0 to 1000 m layer results in a negligible increase (8 kCi, or 0.3%) in the total.

4. Results

Table 8 presents a summary of the results of the computations. The difference between inputs and amount present is far too large to be accounted for by the uncertainties in our calculations. It is inconceivable to us that our estimate of ^{137}Cs in the ocean can be high by 970 kCi or that our estimate of net transport into the North Atlantic can be low by a factor of 4. Note that no uncertainty has been quoted for the amount of ^{137}Cs introduced by direct fallout. This is because Volchok and Toonkel (1974) did not estimate their uncertainty. For some periods removal from the stratosphere has been found to balance the delivery to the earth's surface to

Table 8. Comparison of calculated amount of ^{137}Cs present in the North Atlantic in 1972 and estimates of known inputs.

Amount of ^{137}Cs present in North Atlantic in 1972	3272	$\begin{matrix} +84 \\ -344 \end{matrix}$	kCi
Inputs through 1972			
Direct fallout			2065 kCi
Net transports by currents			240 ± 200 kCi

within 10%, although the probable error of the estimate of total worldwide fallout is quoted as about $\pm 25\%$ (Volchok and Krey, 1967). Another method of assessing the uncertainty in the total fallout to the North Atlantic will be presented in the discussion.

5. Discussion

a. Uncertainties in the estimate of direct fallout. The values in Table 8 show the excess of the measured oceanic inventory over the input inferred from measurements both of fallout in precipitation overland and of ocean current transports. To achieve balance with the inventory, the input from fallout would have to be higher than that inferred from overland measurements by an amount in the range 970 to 620 kCi (47% to 30% higher than the fallout input shown). The first value is the difference between the measured oceanic inventory and the inputs; the latter value is the former value further reduced by the sum of the combined statistical and systematic uncertainties assigned to the inventory estimate.

We have used two approaches to estimate the probable uncertainty in the land fallout data. First we have selected a subset of 16 precipitation collection stations near the boundaries of or on islands in the North Atlantic Ocean (Monrovia, Liberia; Lagos, Nigeria; San Juan, Puerto Rico; Coral Gables, Florida; Miami, Florida; Kindley Air Force Base, Bermuda; Sidi and Rabat, Morocco; Columbia, South Carolina; Sterling, Virginia; New York, New York; Westwood, New Jersey; Harmon Air Force Base, Newfoundland; Goose Bay, Labrador; Prestwick, Scotland; Oslo, Norway; Keflavik, Iceland). There is at least one station in each 10° latitude band. We have excluded stations sited at altitude higher than 100 m above sea level to minimize orogenic effects. There are eight stations on the western side, four on the eastern and four on islands. The fallout into the North Atlantic determined using these stations, for the high fallout years from 1962 through 1966 is 25% higher than that found using the entire network of precipitation stations (estimated from Volchok and Kleinman, 1971). The precipitation is only 11% higher. Second, the input of ^{137}Cs to the North Atlantic has been calculated from global soil inventory data using two methods. Using soil ^{90}Sr inventories at 1 January 1971 (Hardy *et al.*, 1972), -1.6% inventory change by the end of 1972 and a value of 1.45 for the ratio $^{137}\text{Cs}/^{90}\text{Sr}$, the North Atlantic ^{137}Cs input from direct fallout was calculated to

be 2341 kCi. Calculation of the ^{137}Cs input from $^{239,240}\text{Pu}$ data for the same samples and a conversion ratio for $^{239,240}\text{Pu}/^{137}\text{Cs}$ of 0.022 {from the stratospheric ratio estimate of 0.018 (Harley, 1975, p. I-106) corrected to 0.022 by the end of 1972 to allow for ^{137}Cs decay and input of fresh debris of similar ratio} resulted in an estimated value of 2702 kCi. The average of these two estimates, 2521 kCi, is 22% higher than that determined using the precipitation network. These two results indicate the strong possibility that a systematic error will result if it is assumed that mean fallout rates in 10° latitude bands are the same over the ocean as are those determined overland from the precipitation network.

It has been inferred from measurements of fallout in precipitation at four ocean weather ships during the period 1963 to 1971 that no systematically higher fallout occurred at these stations compared to adjacent land masses (Volchok, 1973). The results indicate that the amount of fallout measured at these stations was 61% and the rainfall 63% of that on land in the same latitude bands. If the amount of fallout is increased by a factor of 0.17 to correct for a systematic bias between the column collectors used on the ships, and the standard pot collectors (Volchok, 1968) the result still indicates less fallout at the ocean stations than on land. Since precipitation is notoriously difficult to measure at sea (WMO, 1976; Spinnanger, 1958; Skaar, 1955) perhaps more to the point is that the concentration of ^{90}Sr in precipitation at the weather ships is 1.25 times that measured on land (corrected for the column-pot systematic bias, Volchok, 1968). On the ships fallout and precipitation are measured by two different collectors enclosed within the same windscreen. This arrangement may be less susceptible to systematic errors in measurement of the concentration than in the amounts of precipitation, or of fallout. The weather ship results, taken together with the results in Table 8, may be stronger evidence for the equality of rainfall on the North Atlantic and on land than of equal fallout. Historical climatic estimates of the worldwide mean overocean: overland rainfall ratio (for example, from Jacobs, 1951, p. 67, 69 and Malkus, 1962, p. 129) have been in the range of 1.1 to 1.3.

The available evidence indicates to us that the imbalance seen in Table 8 results primarily from a systematic underestimate of the input of direct fallout.

b. Renewal of deep water. In the water below 1000 m the amount of ^{137}Cs calculated from Table 3 is $830 \begin{matrix} +70 \\ -264 \end{matrix}$ kCi. The limits of uncertainty include the statistical and systematic uncertainties, the latter adjusted for the relative volume below 1000 m. The amount of ^{137}Cs below 1000 m is 25% of the total in the ocean. A major portion of this was, we feel, delivered by the overflows from the Norwegian Sea and the northern North Atlantic water entrained by these overflows (Worthington, 1976, p. 35). Other sources of ^{137}Cs to subsurface water from outside the North Atlantic are small compared to this one (Table 7). Sinking along σ_T surfaces which

outcrop in the northern North Atlantic in winter (Iselin, 1939; Rooth and Östlund, 1972) has been suggested as a mechanism for delivery of soluble fallout nuclides to the deep ocean. However our data as presented here do not speak to this question. We propose to organize the data set to support critical examination of this hypothesis and to present the results in an early report.

Peterson and Rooth (1976) argue by implication that the overflow waters are formed above the permanent pycnocline of the Norwegian Sea. Winter minimum surface ^{90}Sr concentrations at OWS M (66N, 2E) in the Norwegian Sea are, for those years for which data are available (1965 through 1970), about 15% higher on average than those at OWS A and I shown in Figure 5. They decrease with time at about the same rate as at stations A and I. We will assume that ^{90}Sr concentrations at OWS A, I and M were essentially equal for the period 1962 through 1972. Tritium (Östlund *et al.*, 1976) and ^{137}Cs concentrations (Bowen *et al.*, unpublished) from GEOSECS stations 18 (70°00.4'N, 00°00.7'W) and 19 (64°10'N, 5°36.5'W) in the Norwegian Sea in August 1972 show that these tracers were well mixed vertically to the $\sigma_T = 28.06\text{‰}$ isopycnal surface at 700 and 800 m respectively. From these data we infer, as in the case of the flows into the Norwegian Sea (see 3b(2)) that vertical mixing in this region was relatively rapid and also that surface ^{90}Sr concentrations at OWS M are at least roughly indicative of concentrations in the overflow water at about the same time. If the water entrained by the overflows contained similar ^{90}Sr concentrations then it is possible to estimate the sinking rate of near surface water (i.e., water above 1000 m) if it is assumed that the water, once it sinks below 1000 m is not returned above this level within decades. We will also assume (as in 3b(2)) that the amount of ^{137}Cs transported for the 10 years previous to 1962 is related to the total amount transported through 1972 in the same proportion that the cumulative input of global fallout as of 1961 is related to the cumulative input as of 1972.

In order to transfer 25% of the ^{137}Cs in the ocean to depths below 1000 m, with these assumptions, the mean sinking rate of near surface water for the 20 years prior to 1972 must have been close to $14 \times 10^6 \text{ m}^3/\text{s}$. The uncertainty estimated from the uncertainty in the total amount of ^{137}Cs below 1000 m is $+1.2, -4.4 \times 10^6 \text{ m}^3/\text{s}$.

If newly descended water returns to the near surface on a less than decade time scale the sinking rate must be higher. The sinking rate may have been underestimated for another reason. The North Atlantic water entrained by the overflow water is 67% of the transport of the water that overflows the sills (calculated from Worthington, 1976, p. 35). Entrained water must come from water which is near or below the depth of the sills. This water, from 600 m and below, may have contained lower ^{137}Cs concentrations than water higher in the water column. A higher transport rate would then be necessary to carry the estimated amount of ^{137}Cs below 1000 m.

The rate of $14 \times 10^6 \text{ m}^3/\text{s}$ is in reasonable agreement with other estimates of the renewal rate of the deep waters. Worthington (1976) estimates $10 \times 10^6 \text{ m}^3/\text{s}$ over-

flow and entrained North Atlantic water pass below the 4°C isothermal surface. The rate we calculated would be expected to be higher than Worthington's because the temperature at the 1000 m level ranges up to 11°C and is warmer than 4°C over 80% of its surface area (U.S. Naval Oceanographic Office, 1967). Thus the volume below 1000 m contains a substantial volume of water warmer than 4°C.

6. Conclusions and implications for future work

a) Compared to earlier estimates (for example, see Volchok *et al.*, 1971), the input of ⁹⁰Sr and ¹³⁷Cs and the North Atlantic inventory of these isotopes in 1972 is found to be much more closely balanced, the former being about 20 to 30% less than the latter. The difference does not appear to be irreconcilable.

b) For the first time in a calculation of this type, the influence of vertical and horizontal sampling intensity on the overall uncertainty has been quantitatively assessed. It has been found that uncertainties arising from this source are of the same order as systematic uncertainties, as may be seen in Table 3. Thus, increasing the number of samples substantially would not have materially increased the precision or accuracy of the results. For the 1972 sampling intensity such increase would have been most effectively brought about by an evaluation of the deep ocean blank in the North Atlantic alone, which can still be accomplished, and in further interlaboratory comparisons.

c) The mean sinking rate of water to depths below 1000 m for the 20 years prior to 1972 must have been close to 14×10^6 m³/s. This is in agreement with Worthington's (1976) result that 10×10^6 m³/s of overflow and entrained near surface North Atlantic water sink through the 4°C isothermal surface.

d) In defining the limits of the tracer-free volume (Kupferman and Livingston, 1979) we found no convincing evidence for a Western Boundary Undercurrent containing anomalously high tracer concentrations south of 30N. Nor did we find any evidence of deep penetration from the north, by eddies or currents.

e) An evaluation of advective inputs of ⁹⁰Sr and ¹³⁷Cs to the North Atlantic indicates that the East Greenland Current and the current through the Canadian Archipelago are responsible for the greatest input followed by the overflows at the Denmark Strait and the Iceland Scotland ridge. The greatest output is due to flows into the Norwegian Sea. In comparison the transport of ⁹⁰Sr and ¹³⁷Cs by all other flows including river flow is small.

f) The computational approach applied above when combined with the larger amount of high quality data available to us has resulted in a degree of agreement between the inputs of ⁹⁰Sr and ¹³⁷Cs to the North Atlantic and the North Atlantic inventory considerably improved over those resulting from earlier attempts to derive mass balances. Therefore, we believe that we understand the pathways by which the ⁹⁰Sr and ¹³⁷Cs found in 1972 entered the North Atlantic Ocean. With this

understanding of the overall balance we will begin to apply this approach to examine the internal consistency of the various models that have been proposed to explain the exchange of near surface and deeper ocean waters and the time scales associated with this exchange.

The data discussed in this paper will be released in future publications whose format will be better suited for this purpose.

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APPENDIX 1

Cruise, station number and date corresponding to profile serial number or letter

Serial Number	Cruise	Station	Date	Serial Number	Cruise	Station	Date
80	AII-49	1496	VII-7-69	125	KN-30	5	VIII-8-72
88	AII-56	1653	III-3-70	126	KN-30	3	VII-27-72
91	AII-59	1759	XII-1-70	133	Gillis 7205-2	25	X-7-72
92	AII-59	1758	XI-27-70	134	Gillis 7205-2	43	X-15-72
93	AII-59	1757	XI-15-70	135	Eastwd. 19B	4	X-31-72
94	AII-59	1756	XI-10-70	136	Eastwd. 19B	5	XI-1-72
95	AII-59	1755	XI-7-70	137	Gillis 7205-4	73	XI-10-72
97	CH-105	1294	VIII-8-72	138	Gillis 7205-4	77	XI-12-72
98	CH-105	1293	VIII-6-72	139	Gillis 7205-4	87	XI-17-72
99	CH-105	1292	VIII-5-72	140	Gillis 7205-4	94	XI-20-72
100	CH-105	1291	VIII-1/2-72	141	Gillis 7205-4	102	XI-24-72
101	CH-105	1290	VII-29-72	142	Trident 132	3	II-27-73
102	CH-105	1288	VII-15-72	143	Gallatin	OWS E	III-19-73
103	CH-105	1287	VII-10-72	144	Trident 135	3	IV-23-73
104	CH-105	1286	VII-8-72	145	Trident 135	9	IV-29-73
105	CH-105	1285	VII-4-72	146	Trident 135	15	V-3-73
106	CH-105	1284	VI-29/30-72	151	Trident 151	14	IV-27-74
108	KN-25	223	V-1/2-72	153	Bibb	OWS E	IX-15-72
109	KN-30	117	III-21-73	154	Gillis 7205-4	107	XI-27-72
110	KN-30	115	III-15-73	155	Trident 151	7	IV-19-74
111	KN-30	113	III-5-73	156	Trident 135	11	V-1-74
112	KN-30	111	III-1-73	157	Eastwd. 19B	3	X-31-72
113	KN-30	40	X-19-72	158	Eastwd. 19B	6	XI-2-72
114	KN-30	37	X-13-72	159	Eastwd. 19B	7	XI-3-72
115	KN-30	33	IX-26-72	160	Trident 132	7	III-3-73
116	KN-30	31	IX-22-72	a	Cr 165	2499	III-29-68
117	KN-30	29	IX-17-72	b	Argo Circe	246	XII-1-68
118	KN-30	27	IX-12-72	c	KN-30	48	X-26-72
119	KN-30	23	VIII-28/29-72	d	KN-30	49	X-29-72
120	KN-30	19	VIII-25-72	e	KN-30	54	XI-9-72
123	KN-30	14	VIII-13-72	f	KN-30	58	XI-18-72
124	KN-30	11	VIII-15-72				

APPENDIX 2
Dimensional information for North Atlantic Ocean, by Marsden Squares

Latitude (°N)	Marsden Square	Outside 100 m Isobath						Inside 100 m Isobath		
		Surface	Volume (10 ⁶ km ³)				Surface	Volume 10 ⁶ (km ³)	Depth ^α (m)	
		Area (10 ⁶ km ²)	Surface to 1000 m	1000 m to 2000 m	2000 m to Bottom	Surface to Bottom	Depth ^α (m)			Area (10 ⁶ km ²)
0-10	036	0.578	0.578	0.578	0.642	1.798	3114	0.049	0.004	82
	001	0.590	0.590	0.590	1.242	2.422	4102	0.012	0.001	100
	002	1.054	1.054	1.054	2.515	4.623	4386	0.037	0.002	57
	003	1.225	1.225	1.225	2.685	5.135	4192			
	004	1.225	1.225	1.225	2.347	4.797	3916			
	005	1.065	1.065	1.065	2.039	4.169	3915	0.135	0.005	40
	006	0.366	0.366	0.366	0.279	1.011	2759	0.147	0.005	31
	007							0.012	0.001	50
	Total 10°	6.103	6.103	6.103	11.749	23.955		0.392	0.018	
10-20	038	0.368	0.368	0.368	0.302	1.038	2818	0.072	0.001	18
	039	1.189	1.189	1.189	2.866	5.244	4410			
	040	1.189	1.189	1.189	4.015	6.393	5377			
	041	1.189	1.189	1.189	2.541	4.919	4137			
	042	1.189	1.189	1.189	3.273	5.651	4753			
	043	0.307	0.307	0.307	0.626	1.240	4041			
		Total 10°	5.431	5.431	5.431	13.623	24.485		0.072	0.001
20-30	074	0.577	0.577	0.577	0.304	1.458	2528	0.011	0.000	20
	075	1.117	1.117	1.117	3.340	5.574	4990			
	076	1.117	1.117	1.117	3.565	5.799	5192			
	077	1.117	1.117	1.117	2.290	4.524	4050			
	078	1.117	1.117	1.117	3.926	6.160	5515			
	079	1.117	1.117	1.117	3.910	6.144	5500			
	080	0.981	0.981	0.981	1.271	3.233	3296	0.011	0.000	10
	081	0.057	0.044			0.044	770	0.022	0.001	30
		Total 10°	7.200	7.187	7.143	18.606	32.936		0.044	0.001
30-40	109	0.181	0.181	0.178		0.359	1987	0.010	0.000	15
	110	1.012	1.012	1.012	2.054	4.078	4030			
	111	1.012	1.012	1.012	2.031	4.055	4007			
	112	1.012	1.012	1.012	1.231	3.255	3216			

	113	1.012	1.012	1.012	2.501	4.525	4471			
	114	1.012	1.012	1.012	3.307	5.331	5268			
	115	1.012	1.012	1.012	2.860	4.884	4826			
	116	0.654	0.654	0.654	1.004	2.312	3537	0.100	0.003	30
	117							0.031	0.001	28
	Total 10°	6.907	6.907	6.904	14.988	28.799		0.141	0.004	
40-50	145	0.390	0.385	0.377	0.171	0.933	2393	0.110	0.006	54
	146	0.876	0.875	0.875	2.093	3.843	4390			
	147	0.876	0.876	0.875	1.070	2.821	3223			
	148	0.876	0.876	0.875	1.658	3.409	3894			
	149	0.841	0.817	0.765	1.353	2.935	3489	0.034	0.003	80
	150	0.647	0.550	0.355	0.757	1.662	2569	0.111	0.008	72
	151	0.311	0.282	0.224	0.123	0.629	2024	0.118	0.010	86
	152							0.067	0.003	48
	Total 10°	4.817	4.661	4.346	7.225	16.232		0.440	0.030	
50-60	181	0.102	0.072			0.072	705	0.248	0.014	55
	182	0.697	0.604	0.419	0.300	1.323	1899			
	183	0.712	0.712	0.684	0.690	2.086	2932			
	184	0.712	0.712	0.678	0.606	1.996	2805			
	185	0.712	0.710	0.708	0.987	2.405	3380			
	186	0.561	0.469	0.285	0.315	1.069	1904	0.022	0.001	38
	187	0.040	0.008			0.008	195	0.020	0.002	83
	Total 10°	3.536	3.287	2.774	2.898	8.959		0.290	0.017	
60°N to	217	0.034	0.018			0.018	508			
Northern	218	0.236	0.194	0.110	0.022	0.326	1384	0.006	0.001	100
Boundary of	219	0.290	0.234	0.123	0.017	0.374	1294	0.022	0.001	53
Atlantic Ocean	220	0.313	0.284	0.228	0.137	0.649	2072			
	221	0.069	0.051	0.017	0.006	0.074	1082	0.017	0.001	47
	222	0.270	0.238	0.176	0.116	0.530	1963			
	223	0.138	0.063			0.063	457			
	Total 10°	1.350	1.082	0.654	0.298	2.034		0.045	0.003	
Totals										
North Atlantic Ocean		35.344	34.658	33.355	69.387	137.400		1.424	0.074	

α Depth = $\frac{\text{volume, surface to bottom}}{\text{area}}$ (volume and area to more significant figures than shown in table).