# Iodine 129/CFC 11 transit times for Denmark Strait Overflow Water in the Labrador and Irminger Seas

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[1] Iodine 129 discharged from nuclear fuel reprocessing plants in France and the United Kingdom is transported into the Nordic Seas on timescales of 3-5 years. Tracer <sup>129</sup>I is subsequently injected into intermediate waters that overflow the sills between Greenland, Iceland, and Scotland and ventilate the North Atlantic Deep Waters (NADW). During the early 1990s, discharges of <sup>129</sup>I increased by 600%, resulting in a large, well-resolved tracer "front" whose passage through the Nordic Seas is presently being observed by a time series of <sup>129</sup>I measurements on the WOCE (AR7W) section in the Labrador Sea. The highest <sup>129</sup>I levels were measured below depths of 3000 m in Denmark Strait Overflow Water (DSOW). These levels increased by about 300% between 1997 and 2001 to values  $>40 \times 10^7$  atoms/L, indicating that the leading edge of the tracer "front" from the early 1990s was being observed entering the NADW. Using a simple mixing/advection model, <sup>129</sup>I and CFC 11 results were used to calculate transit times of 0.4-2.6 years for the flow of DSOW from formation regions in the Nordic Seas to the Labrador Sea. Measurements on samples collected in the Irminger Sea in 2001 gave transit times of 0.3 years for core DSOW immediately south of Denmark Strait and about 2 years for locations off the southern tip of Greenland. An increase in DSOW transit times observed in the Labrador Sea between 1999 and 2001 probably reflects a more general weakening in the subpolar gyre of the North Atlantic during the 1990s.

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#### 1. Introduction

[2] The overflow of cold, dense water across the sills of the Denmark Strait and the Faroe-Shetland Channel is the principal mechanism for the ventilation of the deep North Atlantic Ocean and thereby has an important influence on the global thermohaline circulation. Most climate models predict that warming and freshening of water in the Nordic Seas will result from emissions of greenhouse gases and this could contribute to a slowing of circulation of North Atlantic Deep Water (NADW) [Delworth and Dickson, 2000]. Indeed, recent studies indicate that the overflow has been steadily freshening during the past 3-4 decades [Dickson et al., 2002] and that its rate of production may be decreasing as well [Hansen et al., 2001]. Despite the importance of the overflows in the prediction of climate change and many decades of study, the mechanisms governing their formation and subsequent transport through the deep North Atlantic are not yet fully understood. In

- [3] The 20th century invasion into the ocean of new anthropogenic substances, such as fallout from nuclear weapons tests in the atmosphere and chlorofluorocarbons (CFCs) from industrial sources, has provided valuable insights into the rates and pathways of intermediate and deep ocean ventilation [Smethie, 1993]. Recently, another tracer, <sup>129</sup>I, has been applied to studies of circulation in the Arctic [Smith et al., 1998, 1999] and North Atlantic [Raisbeck et al., 1995; Santschi et al., 1996; Edmonds et al., 2001]. Iodine 129 is a long-lived ( $t_{1/2} = 16$  million years) isotope that has been released to the ocean in large quantities (25 times the preexisting natural inventory) from European nuclear fuel reprocessing facilities since the late 1960s [Raisbeck et al., 1995]. Modern analytical innovations in accelerator mass spectrometry (AMS) mean that <sup>129</sup>I can now be measured on 1 L water samples throughout the Arctic and North Atlantic, thereby contributing to the development of 129I as a new and useful oceanographic tracer [Kilius et al., 1992; Edmonds et al., 1998].
- [4] Discharges of <sup>129</sup>I into the Irish Sea and English Channel from nuclear fuel reprocessing facilities at

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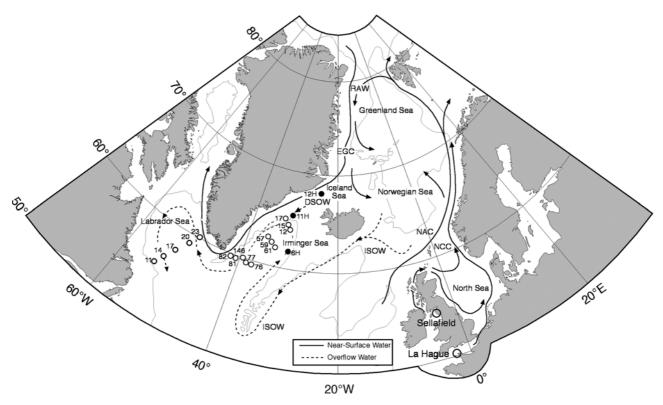
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order to evaluate the response of global thermohaline circulation to changes in climate indexes such as the North Atlantic Oscillation (NAO), it is necessary to constrain the magnitude and variability in the production rates of the overflow waters and establish accurate methods for following their downstream passage through subtropical and tropical regions [Curry et al., 1998; Dickson et al., 1999].

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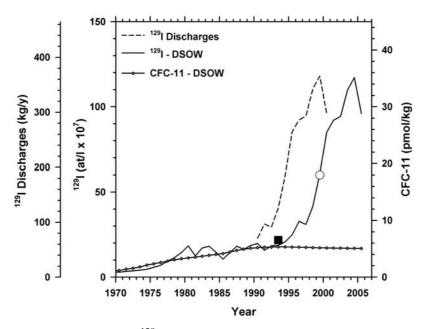


**Figure 1.** Iodine 129 tracer plume from Sellafield and La Hague passes through the North Sea, the Norwegian Coastal Current (NCC), and the Norwegian Atlantic Current (NAC) circulates cyclonically through the Nordic Seas and enters overflow waters that cross ridges north and east of Iceland. Iceland Scotland Overflow Water (ISOW) crosses the Mid-Atlantic Ridge, flows northward into Irminger Sea, and over-rides Denmark Strait Overflow Water (DSOW) descending across the Denmark Strait sill, and the two water masses flow together into the Labrador Sea. The five time series stations (1997–2001) occupied on the AR7W (WOCE) section in the Labrador Sea and the three sections occupied in the Irminger Sea in 2001 are indicated by open circles. Stations 6H, 11H, and 12H (closed circles) were occupied in 1993 by the CCGS *Hudson* [*Yeats and Measures*, 1998], while stations close to 11H and 12H were occupied by the R/V *Marion Dufresne* (GINS Project) in 1999 [*Raisbeck and Yiou*, 2002].

Sellafield (United Kingdom) and La Hague (France), respectively (Figure 1) are transported into the Nordic Seas on a timescale of 3-5 years [Edmonds et al., 1998; Raisbeck and Yiou, 2002]. The return flow of Atlantic Water southward in the East Greenland Current (and mixing associated with convection in the Greenland and Norwegian Seas [Rudels, 1995; Gascard et al., 2002]) also injects tracer 129 I into the intermediate waters that overflow the sills between Greenland and Iceland (Denmark Strait Overflow Water (DSOW)) and between Iceland and Scotland (Iceland Scotland Overflow Water (ISOW)) and thereby provides a potential ventilation tracer for the deep North Atlantic. Iodine 129 differs from traditional tracers such as CFCs and tritium, because it is discharged mainly from two point sources and therefore represents a tracer specific to European coastal water. The potential of 129 I as a circulation tracer is also favored by the timing of its discharges. Between 1990 and 2000, annual discharges of <sup>129</sup>I to the North Atlantic Ocean increased by 600% (Figure 2), mainly as a result of rapidly increasing releases from La Hague. Since the CFC 11 concentration in the atmosphere remained relatively constant over the same period [Walker et al., 2000], the <sup>129</sup>I/CFC 11 ratio in surface waters in the Nordic Seas also increased significantly during the past decade. As a result, measurements of these tracers in southward flowing NADW may provide a chronometer to determine transit times and ventilation ages for water masses formed after the mid-1990s with better resolution compared to that available from most other tracer methods. To test this hypothesis and to document the downstream transport of this recent large increase in the <sup>129</sup>I oceanic input function, a series of <sup>129</sup>I and CFC 11 sections were measured in the Labrador and Irminger Sea between 1997 and 2001. The results presented in this report provide the first time series for the entry of <sup>129</sup>I into the deep overflow waters of the North Atlantic and a potential new method for estimating transit times for DSOW in the Labrador and Irminger Seas.

#### 2. Sampling and Measurements

[5] One liter PVC bottles were filled with seawater from 10 L Niskin bottles deployed at stations along the AR7W transect (WOCE repeat line) during expeditions of the CCGS Hudson to the Labrador Sea in 1997, 1999 and 2001 and at stations in the Irminger Sea on Voyage 369 of the R/V *Oceanus* in 2001 (Figure 1). Iodine 129 analyses were performed on the 1 L samples by accelerator mass spectrometry [Kilius et al., 1992, 1994; Smith et al., 1998]



**Figure 2.** Dashed line is combined  $^{129}$ I discharge record for Sellafield and La Hague (1990–2000), with Sellafield discharges advanced by 2 years to be synchronized with La Hague. The input function for  $^{129}$ I in DSOW in southwestern Iceland Sea is calculated assuming transit times of 5 years and 7 years from La Hague and Sellafield and transfer factor (TF) of  $0.67 \times 10^{-12} \, \mu \text{mol L}^{-1}/\text{mol yr}^{-1}$  from each facility. The input function for CFC 11 in DSOW (extrapolated through 2005) is for 70% saturation [*Smethie and Fine*, 2001]. Iodine 129 data were measured in DSOW at stations 11H and 12H (Figure 1) in 1993 (solid square) by *Edmonds et al.* [2001] and in 1999 (open circle) by *Raisbeck and Yiou* [2002].

at the IsoTrace Laboratory at the University of Toronto. The sample data were normalized to IsoTrace Reference Material #2 ( $^{129}I/^{127}I = 1.174 \times 10^{-11}$  atom ratio). The blank (KI carrier added to distilled water) for this procedure is  $0.75 \pm$  $0.10 \times 10^7$  atoms/L and the standard deviation (one sigma) ranged from 5 to 10% [Edmonds et al., 1998]. Iodine 129 concentrations are reported here in units of atoms per liter. Other authors [e.g., Edmonds et al., 2001; Santschi et al., 1996] have reported their results in iodine units (IU) where 1 IU is equivalent to a molar ratio ( $^{129}\text{L}/^{127}\text{I}$ ) of  $10^{-10}$ . The use of iodine units is necessary for studies in freshwater or low-salinity marine regimes where iodine undergoes biogeochemical cycling, but is not essential in open ocean circulation studies where iodine uptake by biota is very limited [Truesdale, 1994]. At the nearly invariant oceanic iodine concentration of 0.47 µM [Wong, 1991], 1 IU is equal to  $2.9 \times 10^7$  atoms/L.

[6] Water samples for CFC measurements were collected from the same sampling bottles on the rosette used for the <sup>129</sup>I measurements. Samples were drawn into 100 ml glass syringes and sealed with stainless steel Luer caps. Samples were stored submerged in a bath of running seawater prior to analysis. An automated purge and trap system was used to strip halogenated carbon compounds (halogens) from seawater. A measured volume of seawater (~25 ml) was passed to a purge chamber, warmed to 20°C and purged with UHP Nitrogen. The analytes were then trapped on a chromatographic absorbent, packed in stainless steel tubing and then desorbed by heating the trap to 170°C. The halogens were separated using gas chromatography. An electron capture detector was employed for halogen analy-

ses, which was operated at a temperature of 350°C using UHP Nitrogen as a carrier gas.

#### 3. Results and Discussion

#### 3.1. Tracer Input Functions

[7] Radioactive tracers discharged from La Hague and Sellafield are transported through the North Sea and enter the Norwegian Coastal Current (NCC) on timescales of 2 years and 4 years, respectively [Livingston et al., 1982a, 1982b]. These tracers are then injected into the northward flowing Norwegian Atlantic Current (NAC) by lateral and vertical mixing between the NAC and the NCC, partly owing to the widespread formation of eddies [Kershaw and Baxter, 1995; Kershaw et al., 1997; Guegueniat et al., 1997; Yiou et al., 2002]. The NAC bifurcates north of Norway, with one branch flowing through the Barents Sea into the Arctic Ocean and the other flowing toward Fram Strait in the West Spitsbergen Current (Figure 1). Approaching Fram Strait, the AW flow splits again with the eastern branch flowing northward into the Arctic Ocean. The western branch undergoes recirculation both within and north of Fram Strait and joins the southward flowing East Greenland Current (EGC) as Return Atlantic Water (RAW [Fogelqvist et al., 2003]). The EGC includes Polar Water (PW) and modified Atlantic Water that has undergone transport through the Arctic Ocean [Rudels et al., 1999a]. During its southward transport along the Greenland margin, RAW has its salinity reduced by mixing both with modified Atlantic Water and with Arctic Intermediate Water (AIW) that has been formed in the Nordic Seas [Strass et al.,

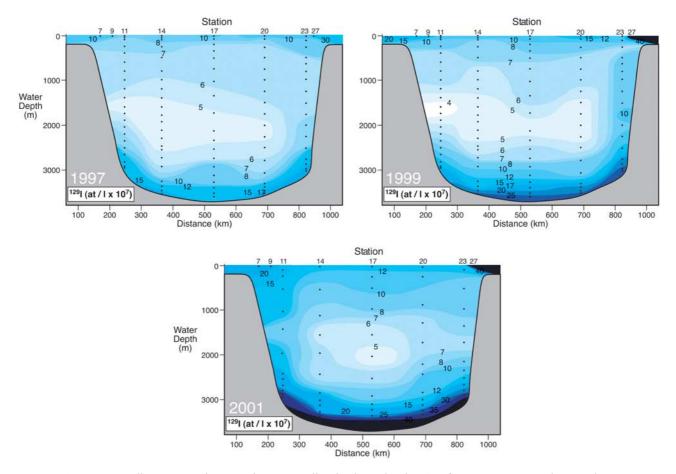
1993]. The East Greenland Current flows through the Denmark Strait with surface components passing south of Greenland and then northward into the Labrador Sea and Baffin Bay as the West Greenland Current (Figure 1). Deeper components of the EGC, including modified Atlantic Water and RAW contribute to DSOW that descends to the bottom of the Irminger Sea basin [Rudels et al., 1999b; Fogelqvist et al., 2003]. Tracer transit times estimated from the dispersion of <sup>134</sup>Cs and <sup>137</sup>Cs from the Sellafield Nuclear Fuel Reprocessing Plant in the 1970s and 1980s are approximately 1 year from the North Sea to the Barents Sea and West Spitsbergen Current, 3 years from the North Sea to the East Greenland Current (70°N) and the interior gyres of the Greenland and Icelandic Seas and 4 years to the West Greenland Current (70°N) in Baffin Bay [Aarkrog et al., 1983, 1987; Livingston et al., 1985; Smith et al., 1990; Kershaw and Baxter, 1995].

[8] ISOW and DSOW are formed north of the Greenland-Scotland ridges by processes that are still a subject of active investigation. ISOW is produced in the Norwegian Sea from a mixture of comparatively old overflow waters having low anthropogenic tracer concentrations and enters the eastern North Atlantic both through the Faroe Bank Channel and across the Iceland-Faroe Rise [Hansen and Osterhus, 2000]. ISOW entrains both Labrador Sea Water (LSW) and Northeast Atlantic Water during its southward passage toward the Charlie Gibbs Fracture Zone (CGFZ) and through the CGFZ into the western North Atlantic [Smethie et al., 2000; Fogelqvist et al., 2003]. The newly modified ISOW  $(\theta = 2.7^{\circ} - 2.9^{\circ} \text{C}, S \cong 34.92)$  then flows northward into the Irminger Sea where it overrides DSOW and flows cyclonically around the Irminger Basin into the Labrador Sea [Swift, 1984]. This water mass is also referred to as Northeast Atlantic Deep Water (NEADW) or Gibbs Fracture Zone Water (GFZW [Smethie et al., 2000]) in recognition of the substantial alteration of its hydrographic properties during transport to the Irminger Sea. However, in this paper it will be referred to as Iceland Scotland Overflow Water (ISOW), because the emphasis is on its source tracer properties.

[9] DSOW is formed north of the \approx 600 m deep Greenland-Iceland Ridge. Swift et al. [1980] argued that DSOW is formed mainly from upper Arctic Intermediate Water which is found in the upper few hundred meters of the Greenland and Iceland Seas where it outcrops at the surface during winter convection. Mauritzen [1996a] has argued that the volume of water produced in convection cells in the Nordic Seas is insufficient to supply the overflows for the deep North Atlantic. Instead, Mauritzen [1996b] has formulated a model in which DSOW is derived mainly from modified Atlantic Water which has been cooled and freshened during its passage through the Arctic Ocean and flows south through Fram Strait to replenish DSOW north of Denmark Strait. Rudels et al. [1999a] suggest that DSOW has a greater proportion of AIW derived from RAW that has been transformed in the Greenland Sea. In any case, DSOW  $(\theta = 0.7^{\circ} - 1.9^{\circ} \text{C}, S = 34.84 - 34.87)$  has been recently ventilated prior to its flow across the Denmark Strait sill and is labeled with elevated tritium and CFC levels that delineate the pathway of this water mass through the Irminger and Labrador Sea basins to subtropical and tropical regions of the deep Western North Atlantic [Smethie and Fine, 2001]. DSOW is also labeled by elevated levels of <sup>129</sup>I as shown by Edmonds et al. [2001] in water depth profiles measured in 1993 at several locations in the Irminger and Labrador Seas. The entry of the <sup>129</sup>I signal into the Deep Western Boundary Current (DWBC) and its transport as far south as 35°N by 1993, off Cape Hatteras has been confirmed by Santschi et al. [1996].

[10] Tracer input functions to the Nordic Seas can be estimated using transfer factors (TF [Aarkrog et al., 1987]). These are empirical quantities equal to the measured tracer concentration at a given location divided by the annual tracer discharge rate, T years earlier, where T is equal to the transit time [Aarkrog et al., 1987; Edmonds et al., 2001]. The <sup>129</sup>I input function at a reference location of 60°N in the NCC has been reconstructed by Smith et al. [1998, 1999] using a transfer factor equivalent to  $25 \times 10^{-12} \, \mu \text{mol L}^{-1/2}$ mol yr<sup>-1</sup> and transit times of 2 and 4 years from La Hague and Sellafield, respectively. Transfer factors estimated from  $^{137}\text{Cs}$  studies for Sellafield are  $10 \times 10^{-12} \, \mu\text{mol} \, L^{-1}$ mol yr<sup>-1</sup> to the Barents Sea and West Spitsbergen Current,  $1 \times 10^{-12} \ \mu mol \ L^{-1}/mol \ yr^{-1}$  to the East Greenland Current and  $0.25 \times 10^{-12} \, \mu \text{mol L}^{-1}/\text{mol yr}^{-1}$  to Baffin Bay [Aarkrog et al., 1983, 1987]. <sup>137</sup>Cs measurements in the Nordic Seas in 1981–1982 indicated that <sup>137</sup>Cs levels in DSOW source or precursor regions, northwest of Iceland, were about 2/3 of the levels in the boundary current, itself [Livingston, 1988; Edmonds et al., 2001]. Therefore the 129I input function for DSOW was calculated using a TF of  $0.67\times10^{-12}~\mu mol~L^{-1}/mol~yr^{-1}$ , equal to 2/3 of the TF for the East Greenland Current and transit times of 5 years from La Hague and 7 years from Sellafield. The tracer signal from the reprocessing sources is superimposed on a small 129I fallout background for Atlantic water  $(2.5 \times 10^7 \text{ atoms/L})$ [Smith et al., 1998; Edmonds et al., 2001]).
[11] The <sup>129</sup>I input function for DSOW is compared to the

combined Sellafield and La Hague discharge record [Smith et al., 1999] in Figure 2. To scale the time-dependent discharge record to the <sup>129</sup>I input function for DSOW it is necessary to assume (1) that the transit time and dilution (by mixing) of the <sup>129</sup>I plume over the lengthy pathway from Sellafield and La Hague to the Iceland Sea (Figure 1) are constant for the given time period and (2) that intra-annual mixing of different year classes of the tracer plume is negligible. Under these conditions, a direct relationship will be maintained between the reprocessing discharge signal (dashed line, Figure 2) and the DSOW input function (solid line, Figure 2). Also shown in Figure 2 are values for measurements of <sup>129</sup>I made at station 11H (Figure 1) in the core of DSOW located between 1400 m and 1500 m over the continental margin in 1993 by Edmonds et al. [2001] and in 1999 by Raisbeck and Yiou [2002]. These measured values of <sup>129</sup>I for DSOW in 1993 and 1999 are in good agreement with the calculated <sup>129</sup>I input function (Figure 2) and thereby validate the TF values. They also agree with <sup>129</sup>I values for precursor DSOW observed at water depths of about 300 m at station 12H immediately north of the Denmark Strait in 1993 [Edmonds et al., 2001; Yeats and Measures, 1998] and 1999 [Raisbeck and Yiou, 2002]. The input function for CFC 11 for DSOW (70% saturation) has been estimated by Smethie and Fine [2001] and is compared to that for <sup>129</sup>I in Figure 2. Until 1994 the input functions of both tracers varied approximately proportionally, but begin-



**Figure 3.** Iodine 129 sections on the AR7W line in the Labrador Sea for 1997, 1999, and 2001 show concentrations increasing each year in bottom water as the leading edge of the <sup>129</sup>I tracer "front" from the mid-1990s (Figure 2) is transported through the Labrador Sea in DSOW. Slightly elevated <sup>129</sup>I levels in surface water represent inputs from the West Greenland Current spreading horizontally into the Labrador Sea.

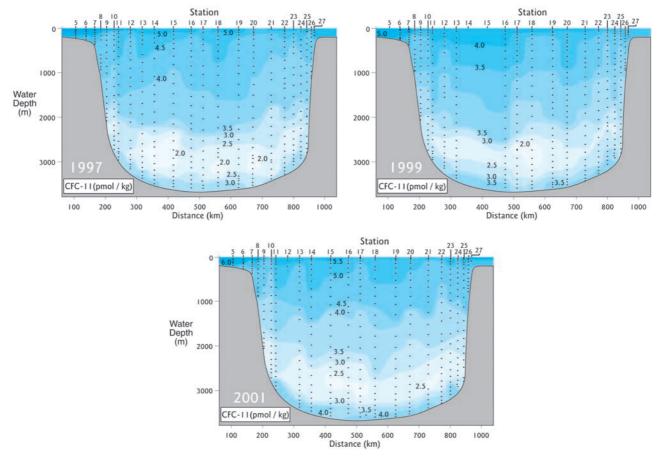
ning in 1994 the input function for <sup>129</sup>I began to increase relative to CFC 11 owing to enhanced reprocessing plant discharges five years earlier. This has resulted in a time varying change in the relative <sup>129</sup>I and CFC 11 concentrations in formation regions for DSOW, thereby providing a potential dual tracer experiment for measuring transit times and ventilation ages of water masses entering the deep North Atlantic.

#### 3.2. Labrador Sea

[12] Iodine 129 and CFC 11 results for the Labrador Sea section (stations 11–23 (Figure 1)) for 1997, 1999 and 2001 are illustrated in Figures 3 and 4. The highest <sup>129</sup>I levels were measured in DSOW, the densest water in the Labrador Sea, while the highest CFC levels were observed in the upper 1000 m. The depth profiles for <sup>129</sup>I and CFC 11 (Figure 5) and the <sup>129</sup>I/CFC 11 ratio (Figure 6) for station 17 from the central Labrador Sea are generally typical of those for other stations and can be used to delineate five main water masses (Figures 5 and 6) [Azetsu-Scott et al., 2003]. The highest concentrations of CFC 11 occur in the surface layer (<50 m) and are elevated due to direct gas exchange with the atmosphere. Labrador Sea Water (LSW<sub>new</sub>), newly ventilated during the previous winter is characterized by

levels of CFC 11 that decrease with increasing water depth to about 1500 m in each profile. Lower CFC 11 levels between 1500 m and 2300 m distinguish a second type of LSW (LSW<sub>old</sub>) that was ventilated during the 1992–1994 winters when convection depths reached 2300 m. Convection depths have become shallower in recent years and LSW<sub>old</sub> represents a remnant feature that is slowly eroding by mixing. ISOW lies immediately below LSW and is characterized by the lowest CFC 11 concentrations in the Labrador Sea and a maximum in salinity (Figure 6). DSOW constitutes the fresher, bottom water layer that flows into the Labrador Sea from the Irminger Sea at water depths below 3000 m and is characterized by higher levels of CFC 11.

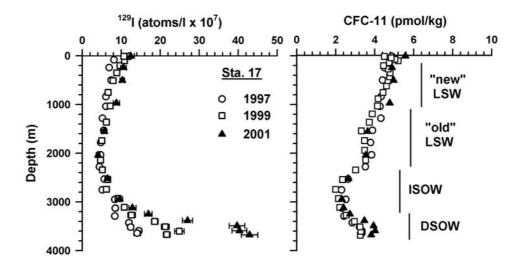
[13] Since the fallout background in the high-latitude North Atlantic is about  $2.5 \times 10^7$  atoms/L [Edmonds et al., 1998] and the lowest <sup>129</sup>I levels measured in any part of the Labrador Sea were greater than  $4 \times 10^7$  atoms/L, then the <sup>129</sup>I sections (Figure 3) indicate that by 1997 the entire Labrador Sea water column had been labeled by a European nuclear fuel reprocessing signal. The elevated levels of <sup>129</sup>I in both upper thermocline (0-1000 m) and bottom waters reflect the two main pathways for the entry of <sup>129</sup>I into the Labrador Sea. Iodine 129 is transported into upper interior waters (<1000 m) in association with the West Greenland



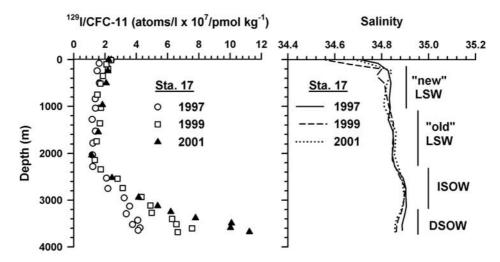
**Figure 4.** CFC 11 sections in the Labrador Sea for 1997, 1999, and 2001 exhibit their highest concentrations in the upper 1000 m and show only minor increases over this time period in bottom DSOW.

Current by mixing and advection [Clarke and Gascard, 1983]. A smaller component arrives from the north, via Baffin Bay by the recirculation of Atlantic water through Nares Strait [Ellis and Smith, 1999]. Pacific-origin water

passing southward through the Canadian Archipelago carries only a small, fallout <sup>129</sup>I signal [*Smith et al.*, 1999]. In addition, there may be a small input of <sup>129</sup>I to surface waters of the North Atlantic by airborne pathways resulting from



**Figure 5.** Iodine 129 water depth profiles for 1997, 1999, and 2001 at station 17 in the center of the Labrador Sea exhibit increases of 300% between 1997 and 2001 in bottom water concentrations, while CFC 11 levels show relatively little change over this time period.

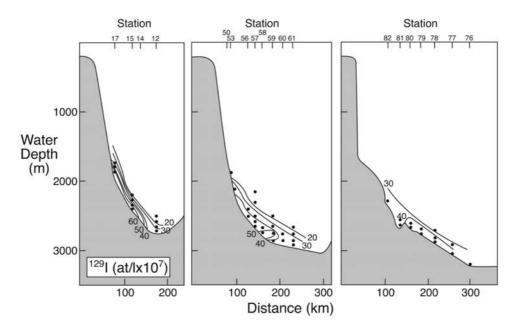


**Figure 6.** Iodine 129/CFC 11 water depth ratios at station 17 increased by about 300% in DSOW between 1997 and 2001, indicating that the increase in the <sup>129</sup>I concentration for this period reflects a source function change. Water depth profiles for salinity at station 17 exhibit a maximum in core ISOW and a slight freshening between 1997 and 2001 in bottom DSOW.

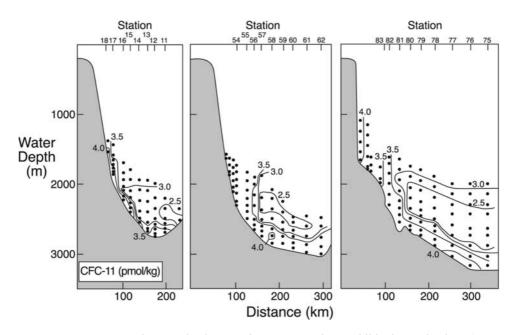
discharges to the atmosphere from Sellafield and La Hague, estimated to be as much as 10% of their total discharges of <sup>129</sup>I [*Moran et al.*, 1999]. The spreading of <sup>129</sup>I into the interior of the Labrador Sea combined with vertical mixing during winter convection produces a vertical gradient in <sup>129</sup>I, with the lowest <sup>129</sup>I concentrations located at water depths of about 2200 m corresponding to the recent (1992–1994) depth of convection. Although <sup>129</sup>I levels in LSW are presently low, they will steadily increase as a function of time as the recent <sup>129</sup>I "front" from European reprocessing plants passes through the Labrador Sea. In future years, <sup>129</sup>I may provide a chronological marker for tracing LSW water as it spreads through the North Atlantic and could prove

useful for validating the various transport times proposed by *Sy et al.* [1997] for North Atlantic intermediate waters. In contrast to CFC 11, the highest <sup>129</sup>I levels were observed in the bottom water layer of DSOW (Figures 3 and 5). This is a consequence of the geographical specificity of this tracer whose main route into the Labrador Sea is by the transport of overflow water from the Nordic Seas rather than by transport through the atmosphere.

[14] Although <sup>129</sup>I levels in the Labrador Sea increased only slightly in the surface mixed layer between 1997 and 2001 and remained virtually unchanged in LSW and ISOW, they increased by 300% in DSOW during this time period (Figure 5). Since CFC 11 concentrations exhibited only a



**Figure 7.** Iodine 129 concentrations on three sections occupied in 2001 in the Irminger Sea (Figure 1) are contoured to illustrate the southward flow of DSOW, delineated by elevated  $^{129}$ I levels (>40 at/L ×  $10^7$ ) along the Greenland continental slope.



**Figure 8.** CFC 11 concentrations on the three Irminger Sea sections exhibit elevated values (>4 pmol/kg) associated with DSOW as it flows southward and descends to depths below 3000 m on the Greenland continental slope.

small (<30%) increase over this time, these results cannot be ascribed to changes in mixing or circulation or by enhanced convection in the Nordic Seas. Rather, they must be the result of changes in the <sup>129</sup>I tracer input function. The relative inputs of <sup>129</sup>I and CFC 11 to the Labrador Sea between 1997 and 2001 are reflected by the depth distribution of the <sup>129</sup>I/CFC 11 ratio for station 17 (Figure 6). The <sup>129</sup>I/CFC 11 ratio increased only slightly in newer LSW between 1997 and 2001 as only a small inventory of <sup>129</sup>I entered the Labrador Sea in the upper 1000 m. However, the <sup>129</sup>I/CFC 11 ratio increased by 72% in DSOW between 1997 and 1999 and by an additional 72% between 1999 and 2001. This <sup>129</sup>I increase in the Labrador Sea, beginning in 1997, was caused by the arrival of the leading edge of the 129I tracer "front" that entered precursor DSOW, north of Denmark Strait in 1994-1995 (Figure 2). These results establish an upper limit of 3 years for the transit time for DSOW from the Icelandic to the Labrador Sea.

#### 3.3. Irminger Sea

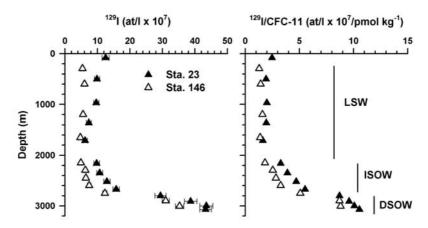
[15] Contours of <sup>129</sup>I and CFC 11 concentrations in DSOW on three sections normal to the flow of DSOW in the Irminger Sea Basin (Figure 1) are illustrated in Figures 7 and 8, respectively. The actual configuration of the DSOW core over the Greenland continental slope is variable in time and depends on a range of factors including climatological conditions upstream at distances at least as far as Fram Strait [Dickson et al., 1999]. However, from temperature and salinity data and from the positions of the highest tracer concentrations on each section, it appears that core DSOW passed through station 17 on the upstream section and near stations 57-59 and 76-77 on the two sections farther downstream. These results are generally consistent with current meter results from an array (TTO Array) deployed by the Fisheries Laboratory (Lowestoft, England) between 1990 and 1991 about 60 km downstream from the stations

between 1986 and 1990 on the stations 50-61 section (Dickson and Brown, 1994). The highest 129 I concentrations  $(>60 \times 10^7 \text{ atoms/L})$  were measured at station 17 in the fast flowing core of DSOW on the upstream section (stations 12-17) closest to Denmark Strait. The flow of DSOW through this region was tightly constrained to the Greenland continental slope with flow velocities that averaged 33 cm s<sup>-1</sup> on the TTO Array [Dickson and Brown, 1994]. Iodine 129 concentrations in core DSOW decreased by about 20% between the stations 12-17 and stations 50-61 sections and by an additional 20% between the stations 50-61 and 76-82 sections while flow velocities measured by the Lowestoft group [Dickson and Brown, 1994] decreased by a total of about 10-30% over these distances. These results reflect the entrainment of overlying ISOW having low <sup>129</sup>I concentrations that occurs as core DSOW descends the continental slope during its downstream flow. [16] Iodine 129 and <sup>129</sup>I/CFC 11 ratio profiles measured in 2001 at station 146 in the southwestern Irminger Sea are compared to those measured at station 23 in the eastern Labrador Sea in Figure 9. Remarkably, <sup>129</sup>I and <sup>129</sup>I/CFC 11 values are higher at all water depths at the Labrador Sea station compared to the Irminger Sea station, despite the fact that the <sup>129</sup>I initially passed through the Irminger Sea. Iodine 129 enters the upper 1000 m in both regions mainly by mixing offshore from the West Greenland and East Greenland Currents. The <sup>129</sup>I distributions suggest that these mixing processes are more effective in transporting <sup>129</sup>I into the basin interior in the Labrador Sea compared to the Irminger Sea. These observations are consistent with the intense mixing that takes place near Cape Farewell, at the southern tip of Greenland [Clarke, 1984] and with

the mixing generated by the Arctic surface outflow on the eastern and northwestern sides of the Labrador Sea. The

higher <sup>129</sup>I and <sup>129</sup>I/CFC 11 values in LSW at station 23

12-17 line and an array (Angmagssalik Array) deployed



**Figure 9.** Both <sup>129</sup>I profiles and <sup>129</sup>I/CFC 11 ratio profiles measured in 2001 at station 146 (open triangles) in the Irminger Sea exhibit lower values compared to 2001 data for station 23 (solid triangles) in the eastern Labrador Sea.

compared to station 146 reflect the fact that LSW is formed in the Labrador Sea and mixes with older LSW as it spreads into the Irminger Sea, thereby decreasing its <sup>129</sup>I concentration. The higher <sup>129</sup>I concentrations and <sup>129</sup>I/CFC 11 ratios in DSOW at station 23 compared to those at station 146 indicate that core DSOW has largely bypassed station 146 on the downslope side during its flow along the Greenland continental margin into the Labrador Sea. This is consistent with the position of the DSOW core at a water depth of about 3100 on the station 76–82 section compared to the slightly upstream and shallower position of station 146 at a water depth of 3005 m.

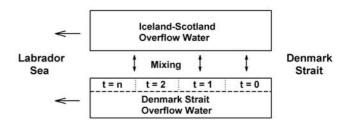
#### 3.4. Mixing Model

[17] A simple idealized mixing box model can be constructed to estimate transit times for DSOW using 129I and CFC 11 (Figure 10). Subsequent to its formation in the Nordic Seas where it is charged with atmospheric tracers and its <sup>129</sup>I transit time is initialized, DSOW passes over the Denmark Strait sill and descends to the bottom of the Irminger Sea Basin [Rudels et al., 1999a]. DSOW is separated into individual year classes and is assumed to undergo end-member mixing with overlying ISOW of the same year class as the two water masses pass cyclonically at the same flow velocity through the Irminger Sea into the Labrador Sea [Dickson and Brown, 1994; Smethie et al., 2000; Fogelqvist et al., 2003]. Horizontal mixing between year classes is assumed to be negligible in both DSOW and ISOW. The values of the <sup>129</sup>I and CFC 11 end-members for each year class of DSOW and ISOW are determined from the time-varying input functions for these tracers as outlined

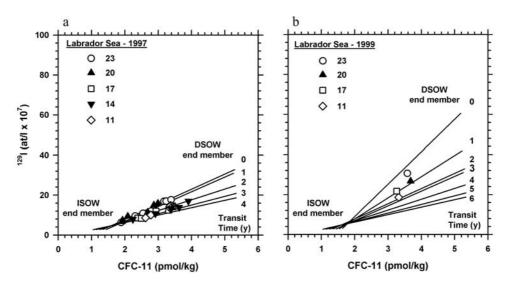
[18] The CFC 11 input function for ISOW encountering DSOW in the northern Irminger Sea, immediately downstream from Denmark Strait, at t=0 (Figure 10) can be estimated from CFC 11 time series measurements carried out on the western flank of the Mid-Atlantic Ridge in the eastern Irminger Sea from 1991 to 2001. The CFC 11 concentration in the core of ISOW increased from about 1 pmol/kg in 1991 [Smethie et al., 2000] to 2 pmol/kg in 2001 [Pickart et al., 2001]. The location of these measurements corresponds to a position approximately 1-2 years

upstream of Denmark Strait for the flow of ISOW. These results indicate that the CFC 11 concentration in ISOW in the Denmark Strait region increased by about 0.1 pmol/kg each year from 0.8 pmol/kg in 1991 to 1.8 pmol/kg in 2001. This is consistent with the CFC 11 input function of Smethie et al. [2000] for ISOW entering the eastern North Atlantic which eventually flows into the northern Irminger Sea basin on timescales of the order of 10 years [Smethie and Swift, 1989]. They estimated that this water is composed of 45% pure ISOW from intermediate depths in the Norwegian Sea, 35% LSW and 20% Northeast Atlantic Water. The 129 I input function for ISOW can also be calculated using the same water mass mixture. The 129I content of pure ISOW should be practically negligible, because this water has a relatively long residence time (≅45 years) at depths of 900 m behind the Iceland Scotland Ridge [Smethie and Swift, 1989; Smethie, 1993]. Prior to the 1990s, LSW will have been labeled mainly by fallout <sup>129</sup>I. Northeast Atlantic Water will have contained a small reprocessing input of <sup>129</sup>I in addition to fallout and probably have levels equivalent to those estimated by Edmonds et al. [2001] for Norwegian Sea surface water. Estimates of these various inputs provide a net <sup>129</sup>I content for modified

#### Mixing/Advection Model



**Figure 10.** A two-component, end-member mixing model is employed with mixing occurring between DSOW of different year classes (t = 0, 1, 2 years, etc.) and ISOW as the two water masses flow together from Denmark Strait into the Labrador Sea.

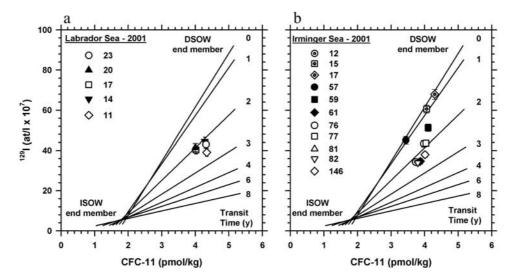


**Figure 11.** (a) Iodine 129/CFC 11 mixing curves for 1997 are calculated using end-member input functions (Figure 2) for DSOW and ISOW. Data from bottom 500 m plotted on the mixing curves give transit times of 0–2.5 years. (b) Data from core DSOW plotted on <sup>129</sup>I/CFC 11 mixing curves for 1999 correspond to transit times of 0.4–1.9 years. Dilution factors for DSOW, calculated from the position on the mixing curve, are in the range of 1.7–2.1 (Table 1).

ISOW of about  $4 \times 10^7$  atoms/L which is assumed to have been relatively constant during the late 1990s. This <sup>129</sup>I concentration is close to the value of  $3.5 \times 10^7$  atoms/L (1.2 IU) measured by *Edmonds et al.* [2001] in 1993 at station 6H (Figure 1) for ISOW flowing northward along the western flank of the Mid-Atlantic Ridge.

[19] The time varying, end-member values for  $^{129}$ I and CFC 11 in DSOW are simply given by their input functions in Figure 2. For ISOW, the  $^{129}$ I end-member value is assumed to be constant at  $4 \times 10^7$  atoms/L while the CFC 11 end-member concentration is assumed to increase linearly from 0.8 to 1.8 pmol/kg between 1991 and 2001, as noted above. Using these end-members, mixing curves can

be established for <sup>129</sup>I and CFC 11 in tracer concentration space for different year classes during the flow of DSOW from Denmark Strait to the sampling location. The mixing curves in Figures 11 and 12 are each calculated for the given year of sampling. Tracer data for DSOW plotted on the figure for the appropriate year of sampling provide an estimate of the transit time. The position of the data point on a given transit time curve also defines a dilution factor (DF), given as the total volume of DSOW [Smith et al., 1999]. The dilution factor is independent of the transit time and reflects the degree to which "core" DSOW has been sampled at each station. This modeling technique is a simplified



**Figure 12.** (a) 2001 tracer results for core DSOW in the Labrador Sea plotted on <sup>129</sup>I/CFC 11 mixing curves show transit times (2.0–2.6 years) that are greater than those for previous years. (b) Tracer results for core DSOW in the Irminger Sea show that the highest <sup>129</sup>I concentrations and smallest transit times (0.3–0.5 years) were measured at stations 15 and 17, immediately downstream from Denmark Strait.

Table 1. Transit Times and Dilution Factors for Core Denmark Strait Overflow Water at Labrador and Irminger Sea Stations

Station	1997		1999		2001	
	Transit Time (y)	Dilution Factor	Transit Time (y)	Dilution Factor	Transit Time (y)	Dilution Factor
			Labrador Sea			
11	1 - 2	1.9	1.9	2.0	2.6	1.3
14	1-2	1.6			2.1	1.3
17	1 - 2	1.9	1.0	2.1	2.1	1.5
20	0 - 1	2.4	0.9	1.7	2.0	1.5
23	0 - 1	2.0	0.4	1.8	2.3	1.3
			Irminger Sea			
12					2.2	1.7
15					0.5	1.5
17					0.3	1.3
57					0.5	2.0
59					1.4	1.4
61					2.4	1.7
76					1.8	1.5
77					1.9	1.5
81					2.2	1.6
82					2.4	1.7
146					2.2	1.5

version of that reported by Smith et al. [1998, 1999] for determining <sup>129</sup>I/<sup>137</sup>Cs transit times in the Arctic Ocean. It can also be used with one time varying tracer and a timeinvariant tracer such as temperature or salinity [Yeats and *Measures*, 1998]. In this regard, transit time curves for DSOW can be constructed using <sup>129</sup>I and either  $\theta$  or S with end-member concentrations for the latter tracers being defined by the initial properties of DSOW and ISOW. These modeling techniques avoid the commonly employed and usually unsupported assumption in double tracer studies that the tracer ratio (e.g., CFC 11/CFC 12 [Andrié et al., 2002]) remain constant in each volume element of water during transport to the sampling location. Note that since the <sup>129</sup>I input function for DSOW is referenced to the time and location at which the CFC 11 signal is initialized, then the <sup>129</sup>I/CFC 11 transit times are analogous to ventilation ages [Smethie et al., 2000].

#### 3.5. Transit Times and Dilution Factors: Labrador Sea

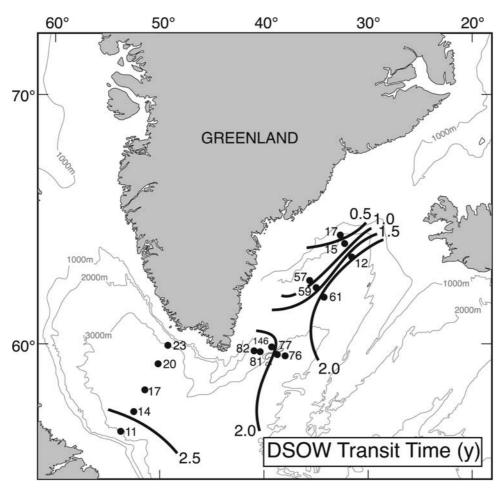
[20] Tracer data collected in 1997 for water depths extending from the core of ISOW (identified by a local salinity maximum) to the bottom are plotted in Figure 11a. This spans a water depth interval of about 500 m at each station. Tracer concentrations are highest in bottom water (core DSOW) while samples collected higher in the water column have a greater proportion of ISOW and consequently lower values of CFC 11 and <sup>129</sup>I. Figure 11a shows that tracer data from 1997 tend to converge with decreasing <sup>129</sup>I and CFC 11 concentrations on the ISOW end-member. This observation supports the assumption of the two-component, end-member mixing process that was adopted for the mixing model.

[21] The positions of maximum <sup>129</sup>I and CFC 11 values for each station give the transit time for core DSOW. Although the resolution between transit time curves is relatively poor for 1997, it can be seen in Figure 11a that transit times for DSOW have values of 0–1 years at the eastern Labrador Sea stations (stations 20 and 23) and 1–2 years at the central and western stations (stations 11, 14

and 17 (Table 1)). The dilution factors for DSOW are close to values of 2 which represent a 1:1 mixture of DSOW and ISOW. The 0 year and 1 year transit time curves for 1997 lie close together, because the 129I and CFC 11 inputs to DSOW varied little between 1996 and 1997 (Figure 2). The range of resolvable transit times for 1999 (Figure 11b) is expanded in tracer concentration space, because of the sharp increase in the <sup>129</sup>I input function (relative to CFC 11) for DSOW between 1997 and 1999 (Figure 2). For the purpose of clarity, only the tracer data for the core of DSOW are plotted in Figure 11b. Transit times for the flow of DSOW to stations in the eastern and central Labrador Sea are in the 0.4-1 years range, while flow to station 11 in the western Labrador Sea was characterized by a transit time of about 1.9 years (Table 1). Dilution factors were greater for flow to the westernmost station, station 11 (DF = 2.0) compared to the easternmost station, station 23 (DF = 1.8).

[22] The 2001 tracer results for the Labrador and Irminger Seas (Figures 12a and 12b) are also restricted for clarity to data from the core of DSOW at each station. The resolution of transit times increased in 2001 compared to 1999 because of the increase in the  $^{129}\mathrm{I}$  input function for DSOW source regions between 1999 and 2001 (Figure 2). Transit times estimated for 2001 (Figure 12a) are greater at each Labrador Sea station compared to those calculated in 1999 (Figure 11b). Transit times for the eastern (station 23) and central stations (stations 14, 17, 20) range from 2.0 to 2.3 years while that for the westernmost station (stations 11) is 2.6 years. Dilution factors for core DSOW average about 1.5 in 2001 compared to a mean value of about 2 in previous years. These results suggest that there was an increase of about 1 year in the transit time and a decrease in the dilution factor of about 25% for core DSOW in the Labrador Sea between 1999 and 2001.

[23] Transit times estimated from the mixing model are a function of the end-member concentrations used for <sup>129</sup>I and CFC 11. Tracer end-member concentrations are relatively small for ISOW and uncertainties in their values have only a minimal effect on estimates of transit times. Uncertainty in



**Figure 13.** Contours outlining <sup>129</sup>I/CFC 11 transit times for DSOW at stations in the Labrador and Irminger Seas show that the lowest values (<0.5 years) were measured close to the Denmark Strait sill, and the highest (>2.5 years) were measured in the western Labrador Sea.

the CFC 11 input function for DSOW is associated mainly with uncertainty in the percent saturation of CFC 11 in DSOW and has been estimated by *Smethie and Fine* [2001] to be about 20%. Uncertainties in the <sup>129</sup>I input function for DSOW are governed by uncertainties in the reprocessing plant annual discharge rates, the transfer factor (TF) and the transit time of the <sup>129</sup>I plume from the North Sea to Denmark Strait. Errors in reported reprocessing plant discharge rates are probably negligible, but it is difficult to set theoretical limits on errors in transfer factors and transit times for the 129I plume. However, the two values of 129I measured in precursor DSOW in 1993 and 1999 (Figure 2) agree to within 20% of the calculated <sup>129</sup>I input function which is based solely on historical data sets [Aarkrog et al., 1983, 1987; Edmonds et al., 2001]. Therefore if it is assumed that uncertainties for both the CFC 11 and 129I input functions for DSOW are about 20%, then uncertainties in the transit time and dilution factor are estimated to be about 0.5 years and 0.4, respectively, for the 2001 data sets.

[24] If CFC 11 is replaced by S as the second tracer and transit time curves are constructed using end-members of S = 34.84 and S = 34.92, for DSOW and ISOW, respectively [Smethie et al., 2000; Fogelqvist et al., 2003], then similar values of the transit time and dilution factor are determined for each station in the Labrador Sea. Similar results are also obtained using potential temperature ( $\theta$ ) as

the second tracer and end-member values of  $\theta = 0.8^{\circ} C$  and  $\theta = 2.9^{\circ} C$ , for DSOW and ISOW, respectively. This exercise shows that DSOW transit times can be determined from <sup>129</sup>I measurements and source water hydrographic properties alone, providing the latter remain constant during the time of the study.

[25] The transit times of 0.5-2.6 years reported here for DSOW in the Labrador Sea are smaller than ventilation ages of about 6 years reported by Azetsu-Scott et al. [2003] based on CFC 12 data. The ventilation ages reported by Azetsu-Scott et al. [2003] are determined from comparisons of the CFC 12 atmospheric input function with CFC 12 concentrations in the mixture of DSOW and older, less well ventilated water masses at the bottom of the Labrador Sea. Therefore the Azetsu-Scott et al. [2003] ventilation ages represent a type of mean age for the mixture of DSOW and mainly, ISOW that flows through the Labrador Sea. This is in contrast to the present model in which transit times are estimated only for the DSOW component of that mixture. The lower transit times determined here reflect the removal of the ISOW component (having longer transit times) from the calculation.

#### 3.6. Transit Times and Dilution Factors: Irminger Sea

[26] Transit times and dilution factors for core DSOW at the Irminger Sea stations, determined from tracer plots shown in Figure 12b, are tabulated in Table 1. The distribution of transit times for DSOW in the Labrador and Irminger Seas is contoured in Figure 13. Low transit times (<0.5 years) characterize the fast flowing, core of DSOW at station 17 on the upstream section, close to regions of DSOW formation in the Iceland Sea. Transit times increase to values >2 years at downstream stations in the Irminger Sea. Dilution factors for core DSOW (Table 1) increase from 1.3 at station 17 south of Denmark Strait to 1.5 at the downstream stations, 76 and 77 owing to mixing between DSOW and modified ISOW during downstream transport. These results are consistent with the initial rapid flow of DSOW southward through Denmark Strait and along the continental slope followed by a decrease in current velocities as the flow steers cyclonically around the southern tip of Greenland [Dickson and Brown, 1994].

[27] Surprisingly, transit times (and dilution factors) for DSOW in the central and eastern Labrador Sea are similar to those in the southern Irminger Sea. For one dimensional, horizontal flow this would imply extremely rapid transport of DSOW from the Irminger Sea to the Labrador Sea. However, it is also possible that there is some recirculation of DSOW in the Labrador and Irminger Seas. Although this would violate the model assumption of negligible horizontal mixing between separate year classes of DSOW, it would have the effect of reducing both the <sup>129</sup>I and CFC 11 concentration gradients in bottom water between the two basins and differences in <sup>129</sup>I/CFC 11 transit times. The recirculation of middepth ( $\cong$ 700 m) cyclonic boundary currents in the Labrador Sea and Irminger Seas has been observed using subsurface floats by *Lavender et al.* [2000], but has not yet been reported for the deep overflows.

[28] DSOW in the Labrador Sea in 2001 was significantly fresher by about 0.02 salinity units than DSOW in the Irminger Sea, as has generally been the case for the past four decades [*Dickson et al.*, 2002]. This freshening is probably caused by mixing between DSOW and low salinity, Lower Deep Water (LDW) south and west of Cape Farewell. LDW forms a cyclonic loop at the head of the Western Basin, flowing north along the bottom over the western flank of the Mid-Atlantic Ridge [*McCartney*, 1992]. LDW is derived from the northward spread of Antarctic Bottom Water (AABW) across the equator and has low concentrations of <sup>129</sup>I and CFC 11. As a result, mixing of LDW and DSOW will not significantly alter transit times.

## 3.7. Timescales for Denmark Strait Overflow Water Formation

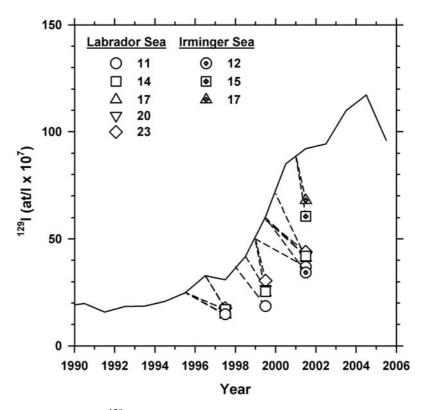
[29] Transit times in the present study of 0.3–2 years for the flow of DSOW through the Irminger Sea and 0.5–2.5 years for the Labrador Sea are in agreement with previous studies that traced DSOW from its origin in the Greenland/Iceland Seas. *Smethie and Swift* [1989] reported an upper limit of 1.7 years for the ventilation age of DSOW in the vicinity of stations 15 and 17 in the Irminger Sea based on tritium and krypton 85 tracer data. They also noted that the ventilation age included the residence time of precursor DSOW behind the Greenland-Iceland Ridge and estimated this latter parameter to be in the range of 0–1.8 years. *Livingston et al.* [1985] estimated an upper limit of 2 years for the transport of <sup>137</sup>Cs in DSOW from surface regions of the Nordic Seas

to Irminger Sea stations slightly downstream from stations 15 and 17. Yeats and Measures [1998] estimated a transit time of 2 years for the transport of DSOW from just north of Denmark Strait to the central Labrador Sea using CFC 11 and hydrographic data. Dickson et al. [1999] drew similar conclusions based on current meter measurements from moorings over the continental slope off Greenland. They showed a strong time series correlation between the temperature of DSOW near station 57 in the Irminger Sea and the temperature of Atlantic Water on a section across the West Spitsbergen Current in the eastern Fram Strait, 3 years earlier. Their study indicated that a 3 year advectivediffusive time lag existed for the transport of heat across the Nordic Seas from eastern Fram Strait to the depths of the Irminger Sea. As noted above (section 3.1) the <sup>129</sup>I transit time from Fram Strait to formation regions for DSOW, north of Denmark Strait is about 2 years. Therefore the 1 year transit time for DSOW at station 57 is equivalent to a net tracer transit time of about 3 years between the eastern part of Fram Strait and the Irminger Sea, in agreement with the results of *Dickson et al.* [1999].

[30] Recently, Häkkinen and Rhines [2004] have interpreted measurements of sea surface height calculated from TOPEX/Poseidon altimeter data to suggest that the subpolar gyre circulation has significantly weakened during the 1990s. This weakening is associated with a shift in the NAO from high positive values in the early 1990s to an extreme negative value in 1996. This interpretation is supported by a 19 month current meter record (1996-1998) from a mooring positioned close to station 11 in the Western Labrador Sea that indicates a decrease in the flow of DSOW of the order of 10-20% for the duration of the record [Häkkinen and Rhines, 2004]. The values for <sup>129</sup>I/CFC 11 transit times for DSOW in the Labrador Sea are poorly resolved for 1997, but transit times clearly increased from 0.4-1.9 years in 1999 to 2.0-2.6 years in 2001 (Table 1). These results indicate that the weakening of the flow of DSOW directly observed in the Western Labrador Sea during 1996–1998 continued through to at least 2001 for other regions of the Labrador Sea as well. This apparent weakening of the DSOW may have been accompanied by weakening of the flow along the entire <sup>129</sup>I pathway between the European reprocessing plants and the Nordic Seas. If this proves to be the case, then it will become evident in future studies by differences between measured 129I levels in precursor DSOW and predicted values based on transfer factors.

### 3.8. Comparison With <sup>129</sup>I Input Function

[31] Iodine 129 results for core DSOW in the Labrador and Irminger Seas are compared to the <sup>129</sup>I input function for DSOW in the Iceland Sea in Figure 14. Each data point is connected to a point on the <sup>129</sup>I input function representing the initial concentration of <sup>129</sup>I in precursor DSOW that was diluted by mixing to give the measured <sup>129</sup>I concentration in the Labrador or Irminger Seas. The time lag between the <sup>129</sup>I input function and the data points represents the transit time for the flow of DSOW to each station. These results show that the 1997 data from the Labrador Sea recorded the arrival of the leading edge of the <sup>129</sup>I tracer "front" as it passed through the Iceland Sea in about 1995. The 1999 and 2001 results recorded the arrival of the steep



**Figure 14.** Open symbols for  $^{129}\text{I}$  concentrations in DSOW in the Labrador and Irminger Seas are connected by a dashed line to points on the  $^{129}\text{I}$  input function (continuous solid line) representing the initial concentration of  $^{129}\text{I}$  in precursor DSOW. The time lag between the data points and the input function represents the DSOW transit time, and the vertical concentration difference (y axis) reflects the dilution factor. The 1997-2001  $^{129}\text{I}$  results reflect the passage of the steep, leading edge of the  $^{129}\text{I}$  tracer "front" into the Labrador and Irminger Seas.

middle portion of the <sup>129</sup>I input "front" with an approximately one year lag between stations 23 and 11 for each sampling year. By 2001, more than 70% of the main <sup>129</sup>I tracer signal increase had entered the Irminger Sea and the steepest part of the leading edge was spread out laterally through the Irminger and Labrador Seas. Further, about 30–40% of the <sup>129</sup>I signal had already been transported southward past station 11 in the western Labrador Sea by 2001.

[32] One implication of these observations is that the steepest part of the leading edge of the <sup>129</sup>I tracer "front" has already passed through the Nordic Seas or is undergoing recirculation and mixing in the interior gyres. As a result, 129I levels in the Nordic Seas should slowly attain a maximum value in the next few years and then level off or even decline, depending on the future nature of the 129I input function. The steep leading edge of the 129I tracer "front" is presently distributed laterally across the Labrador Sea and southward toward the Newfoundland Basin. As the 129I tracer "front" passes through the Newfoundland Basin, part of it will enter the southward flow of DWBC around the Grand Banks and into the subtropical North Atlantic and part will be recirculated in the subpolar basins [Stramma et al., 2004]. Smethie et al. [2000] indicate that the mixing of DSOW and ISOW is sufficiently great that by the time flow in the DWBC has entered the subtropical North Atlantic the two water masses cannot be further distinguished from one another. CFC 11 ventilation ages of 15-30 years reported

by Smethie et al. [2000] for the overflow waters in the tropical and subtropical North Atlantic therefore represent a mean value for the combined water masses and are probably greater than that for DSOW alone. Recently, Fine et al. [2002] and Steinfeldt and Rhein [2004] have noted some of the problems associated with determining accurate ages in the DWBC and have estimated effective ages of 20-23 years for North Atlantic Deep Water at the equator. Measurements of the leading edge of the 129I "front" should provide better resolution between the two overflow water components and a more accurate estimate of the dilution factor and ventilation age for the DSOW component of the DWBC. Measurements of the position of the 129I "front" along the path of the DWBC during the next 5-10 years represent a unique opportunity to establish an upper limit for the speed at which high-latitude climate signals are communicated to the deep North Atlantic.

#### 4. Conclusion

[33] A 300% increase in the <sup>129</sup>I concentration in DSOW in the Labrador Sea observed between 1997 and 2001 indicates that the leading edge of a large <sup>129</sup>I tracer "front" associated with discharges from European nuclear fuel reprocessing plants during the 1990s has passed through the Nordic Seas and is presently entering the NADW via the Labrador Sea. A two-component model that invokes mixing

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between DSOW and ISOW during bottom water flow through the Irminger and Labrador Seas was used to estimate transit times and dilution factors for DSOW. Iodine 129 and CFC 11 tracer data collected in 2001 in the Labrador Sea provide estimates of about 2 years for DSOW transit times and values of about 1.4 for DSOW dilution factors. The former correspond to transit times of about 7 years and 9 years for the entire pathway from the La Hague (France) and Sellafield (United Kingdom) nuclear fuel reprocessing plants, respectively to the deep Labrador Sea. DSOW transit times to the Labrador Sea increased from values in the range of 0.4-1.9 years in 1999 to 2.0-2.6 years in 2001. These results are consistent both with a general weakening observed in the subpolar gyre of the North Atlantic in the late 1990s and direct measurements of decreased DSOW flow rates in 1996-1998 in the western Labrador Sea [Häkkinen and Rhines, 2004]. The relatively large magnitude of the <sup>129</sup>I tracer "front" and its geographic specificity means that <sup>129</sup>I could be a useful tracer for observing temporal variations in the supply of DSOW to the deep North Atlantic on annual and decadal timescales.

[34] Acknowledgments. We would like to thank A. Clarke for organizing the CCGS Hudson missions to the Labrador Sea, F. Zemlyak and B. Gershey for the CFC measurements, and C. Soto and X.-L. Zhao for preparing and analyzing the <sup>129</sup>I samples. We also thank R. Pickart for planning the R/V *Oceanus* 369 cruise and E. Gorman, D. LeBel, and J. Cheery for the CFC measurements on this cruise. E. P. Jones was partially supported by the Canadian Panel on Energy Research and Development. S. B. Moran was supported by NSF grant OCE-9730257 and W. M. Smethie Jr. by NSF grant OCE07-18127. This is LDEO contribution number 6697. The operation of the IsoTrace Laboratory is partially supported by NSERC MFA funds.

#### References

- Aarkrog, A., H. Dahlgaard, L. Hallstadius, H. Hansen, and E. Holm (1983), Radiocesium from Sellafield effluents in Greenland waters, *Nature*, 304, 49–51.
- Aarkrog, A., S. Boelskifte, H. Dahlgaard, S. Duniec, L. Hallstadius, E. Holm, and J. N. Smith (1987), Technetium-99 and cesium-134 as long distance tracers in Arctic waters, *Est. Coast. Shelf Sci.*, 24, 637–647.
- Andrié, C., M. Rhein, S. Freudenthal, and O. Plähn (2002), CFC time series in the deep water masses on the western tropical Atlantic, 1990–1999, *Deep Sea Res., Part I*, 49, 281–304.
- Azetsu-Scott, K., E. P. Jones, I. Yashayaev, and R. M. Gershey (2003), Time series study of CFC concentrations in the Labrador Sea during deep and shallow convection regimes (1991–2000), *J. Geophys. Res.*, 108(C11), 3354, doi:10.1029/2002JC001317.
- Clarke, R. A. (1984), Transport through the Cape Farewell–Flemish Cap section, Rapp. P. V. Réun. Cons. Int. Explor. Mer, 185, 120–130.
- Clarke, R. A., and J.-C. Gascard (1983), The formation of Labrador Sea water. part I: Large-scale processes, *J. Phys. Oceanogr.*, 13, 1764–1797
- Curry, R. G., M. McCartney, and T. M. Joyce (1998), Oceanic transport of subpolar climate signals to mid-depth subtropical waters, *Nature*, *391*, 575–577.
- Delworth, T. L., and K. W. Dickson (2000), Implications of the recent trend in the Arctic/North Atlantic Oscillation for the North Atlantic thermohaline circulation, *J. Clim.*, 13, 3721–3727.
- Dickson, R. R., and J. Brown (1994), The production of North Atlantic Deep Water: Sources, rates and pathways, *J. Geophys. Res.*, 99, 12,319–12,341
- Dickson, B., J. Meincke, I. Vassie, J. Jungclaus, and S. Osterhus (1999), Possible predictability in overflow from the Denmark Strait, *Nature*, 397, 243–246
- Dickson, B., I. Yashayaev, J. Meincke, B. Turrell, S. Dye, and J. Holfort (2002), Rapid freshening of the deep North Atlantic Ocean over the past four decades, *Nature*, 416, 832–837.
- Edmonds, H. N., J. N. Smith, L. R. Kilius, H. D. Livingston, and J. M. Edmond (1998), <sup>129</sup>I in archived seawater samples: Source functions and tracer comparisons, *Deep Sea Res.*, *Part I*, *45*, 1111–1125.

- Edmonds, H. N., Z. Q. Zhou, G. M. Raisbeck, F. Yiou, L. Kilius, and J. M. Edmond (2001), Distribution and behaviour of anthropogenic <sup>129</sup>I in water masses ventilating the North Atlantic Ocean, *J. Geophys. Res.*, 106, 6881–6894.
- Ellis, K. M., and J. N. Smith (1999), The flow of radionuclides through the Canadian Archipelago, in *Proceedings of IAEA Marine Pollution Sympo*sium, IAEA Tech. Doc. 1094, pp. 462–464, Monaco.
- Fine, R. A., M. Rhein, and A. Chantal (2002), Using a CFC effective age to estimate propagation and storage of climate anomalies in the deep western North Atlantic Ocean, *Geophys. Res. Lett.*, 29(24), 2227, doi:10.1029/2002GL015618.
- Fogelqvist, E., J. Blindheim, T. T. Tanhua, S. Osterhus, E. Buch, and F. Rey (2003), Greenland-Scotland overflow studied by hydro-chemical multivariate analysis, *Deep Sea Res.*, *Part I*, *50*, 73–102.
- Gascard, J., A. J. Watson, M. Messias, K. A. Olsson, T. Johannessen, and K. Simonsen (2002), Long-lived vortices as a mode of ventilation in the Greenland Sea, *Nature*, 416, 525–527.
- Guegueniat, P., P. Kershaw, J. Hermann, and P. Bailly du Bois (1997), New estimation of La Hague contribution to the artificial radioactivity of Norwegian waters (1992–1995) and Barents Sea, *Sci. Total Environ.*, 202, 249–266.
- Häkkinen, S., and P. B. Rhines (2004), Decline of subpolar North Atlantic circulation during the 1990s, Science, 304, 555–559.
- Hansen, B., and S. Osterhus (2000), North Atlantic Nordic Sea exchanges, Prog. Oceanogr., 45, 109–208.
- Hansen, B., W. R. Turrell, and S. Osterhus (2001), Decreasing overflow from the Nordic Seas into the Atlantic Ocean through the Faroe Bank channel since 1950, *Nature*, 411, 927–930.
- Kershaw, P. J., and A. J. Baxter (1995), The transfer of reprocessing wastes from NW Europe to the Arctic, *Deep Sea Res., Part II*, 42, 1413–1448.
- Kershaw, P., P. Gurbutt, D. Woodhead, K. Leonard, and J. Rees (1997), Estimates of fluxes of <sup>137</sup>Cs in northern waters from recent measurements, Sci. Total Environ., 202, 211–223.
- Kilius, L., A. E. Litherland, J. C. Rucklidge, and N. Baba (1992), Accelerator mass spectrometric measurements of heavy long-lived isotopes, J. Appl. Rad. Isotop., 43, 279–287.
- Kilius, L. R., J. C. Rucklidge, and C. Soto (1994), The dispersal of <sup>129</sup>I from the Columbia River estuary, *Nucl. Instrum. Methods*, *B92*, 393–397.
- Lavender, K. A., R. E. Davis, and W. B. Owens (2000), Mid-depth recirculation observed in the interior Labrador and Irminger Seas by direct velocity measurements, *Nature*, 407, 66–69.
- Livingston, H. D. (1988), The use of Cs and Sr isotopes as tracers in the Arctic Mediterranean Seas, *Philos. Trans. R. Soc. London, Ser. A*, 325, 161, 176
- Livingston, H. D., V. T. Bowen, and S. L. Kupferman (1982a), Radionuclides from Windscale discharges I: Non-equilibrium tracer experiments in high latitude oceanography. *J. Mar. Ros.* 40, 253–272
- in high latitude oceanography, *J. Mar. Res.*, 40, 253–272. Livingston, H. D., V. T. Bowen, and S. L. Kupferman (1982b), Radionuclides from Windscale discharges II: Their dispersion in Scottish and Norwegian coastal circulation, *J. Mar. Res.*, 40, 1227–1258.
- Livingston, H. D., J. H. Swift, and H. G. Ostlund (1985), Artificial radionuclide tracer supply to the Denmark Strait Overflow between 1972 and 1981, *J. Geophys. Res.*, *90*, 6971–6982.
- Mauritzen, C. (1996a), Production of dense overflow waters feeding the North Atlantic across the Greenland-Scotland Ridge, part I: Evidence for a revised circulation scheme, *Deep Sea Res.*, *Part I*, *43*, 769–806.
- Mauritzen, C. (1996b), Production of dense overflow waters feeding the North Atlantic across the Greenland-Scotland Ridge, part 2: An inverse model, *Deep Sea Res., Part I, 43,* 807–836.
- McCartney, M. S. (1992), Recirculating components to the deep boundary current of the northern North Atlantic, *Prog. Oceanogr.*, 29, 283–383.
- Moran, J. E., S. Oktay, P. H. Santschi, and D. R. Schink (1999), Atmospheric dispersal of <sup>129</sup>Iodine from nuclear fuel reprocessing plants, *Environ. Sci. Technol.*, *33*, 2536–2542.
- Pickart, R. S., F. Straneo, W. M. Smethie Jr., and D. LeBel (2001), R/V *Oceanus* 369 cruise summary: Irminger Sea circulation and convection, unpublished cruise report, Woods Hole Oceanogr. Inst., Woods Hole, Mass.
- Raisbeck, G. M., and F. Yiou (2002), Use of <sup>129</sup>I as an oceanographic tracer in the Nordic Seas, paper presented at the 5th International Conference on Environmental Radioactivity in the Arctic and Antarctic, St. Petersburg, Russia, 16–20 June.
- Raisbeck, G. M., F. Yiou, A. Q. Zhou, and L. R. Kilius (1995), <sup>129</sup>I from nuclear fuel reprocessing facilities at Sellafield (U.K.) and La Hague (France): Potential as an oceanographic tracer, *J. Mar. Syst.*, 6, 561–570.
- Rudels, B. (1995), The thermohalocline circulation of the Arctic Ocean and the Greenland Sea, *Philos. Trans. R. Soc. London, Ser. A*, 352, 287–299.
- Rudels, B., H. J. Friedrich, and D. Quadfasel (1999a), The Arctic circumpolar boundary current, *Deep Sea Res., Part II*, 46, 1023–1062.

- Rudels, B., P. Eriksson, H. Grönvall, R. Hietala, and J. Launiainen (1999b), Hydrographic observations in Denmark Strait in fall 1997 and their implications for the entrainment into the overflow plume, *Geophys. Res. Lett.*, 26, 1325–1328.
- Santschi, P. H., D. R. Schink, O. Corapcioglu, S. Oktay-Marshall, U. Fehn, and P. Sharma (1996), Evidence for elevated levels of iodine-129 in the Deep Western Boundary Current in the Middle Atlantic Bight, *Deep Sea Res.*, *Part I*, 43, 259–265.
- Smethie, W. M., Jr. (1993), Tracing the thermalhaline circulation in the western North Atlantic using chlorofluorcarbons, *Prog. Oceanogr.*, 31, 51–99
- Smethie, W. M., Jr., and R. A. Fine (2001), Rates of North Atlantic Deep Water formation calculated from chlorofluorocarbon inventories, *Deep Sea Res.*, Part I, 48, 189–215.
- Smethie, W. M., Jr., and J. H. Swift (1989), The tritium: Krypton-85 age of Denmark Strait Overflow Water and Gibbs Fracture Zone Water just south of Denmark Strait, J. Geophys. Res., 94, 8265–8275.
- Smethie, W. M., Jr., R. A. Fine, A. Putzka, and E. P. Jones (2000), Tracing the flow of North Atlantic Deep Water using chlorofluorocarbons, *J. Geophys. Res.*, 105, 14,297–14,323.
- Smith, J. N., K. M. Ellis, and E. P. Jones (1990), Cesium-137 transport into the Arctic Ocean through Fram Strait, *J. Geophys. Res.*, 95, 1693–1701.
  Smith, J. N., K. M. Ellis, and L. R. Kilius (1998), <sup>129</sup>I and <sup>137</sup>Cs tracer
- Smith, J. N., K. M. Ellis, and L. R. Kilius (1998), <sup>129</sup>I and <sup>13</sup>/Cs tracer measurements in the Arctic Ocean, *Deep Sea Res.*, *Part I*, 45, 959–984.
- Smith, J. N., K. M. Ellis, and T. M. Boyd (1999), Circulation features in the central Arctic Ocean revealed by nuclear fuel reprocessing tracers from Scientific Ice Expeditions 1995 and 1996, J. Geophys. Res., 104, 29,663–29,677.
- Steinfeldt, R., and M. Rhein (2004), Spreading velocities and dilution of North Atlantic Deep Water in the tropical Atlantic based on CFC time series, J. Geophys. Res., 109, C03046, doi:10.1029/2003JC002050.
- Stramma, L., D. Kieke, M. Rhein, F. Schott, I. Yashayaev, and K. P. Kolterman (2004), Deep water changes at the western boundary of the subpolar North Atlantic during 1996 to 2001, *Deep Sea Res., Part I*, *51*, 1033–1056.
- Strass, V. H., E. Fahrbach, U. Schauer, and L. Sellmann (1993), Formation of Denmark Strait Overflow Water by mixing in the East Greenland Current, J. Geophys. Res., 98, 6907–6919.

- Swift, J. H. (1984), The circulation of the Denmark Strait and Iceland Scotland overflow waters in the North Atlantic, *Deep Sea Res.*, *Part I*, 31, 1339–1355.
- Swift, J. H., K. Aagaard, and S. V. Malmberg (1980), The contribution of the Denmark Strait Overflow to the deep North Atlantic, *Deep Sea Res.*, Part A, 27, 29–42.
- Sy, A., M. Rhein, J. Lazier, K. Koltermann, J. Meincke, A. Putzkal, and
   M. Bersch (1997), Surprisingly rapid spreading of newly formed intermediate waters across the North Atlantic Ocean, *Nature*, 386, 675–679.
   Truesdale, V. W. (1994), Distribution of dissolved iodine in the Irish Sea, a

temperate shelf sea, Est. Coast. Shelf Sci., 38, 435–446.

- Walker, S. J., R. F. Weiss, and P. K. Salameh (2000), Reconstructed histories of the annual mean atmospheric mole fractions for the halocarbons CFC-11, CFC-12, CFC-113, and carbon tetrachloride, *J. Geophys. Res.*, 105, 14,285–14,296.
- Wong, G. T. F. (1991), The marine geochemistry of iodine, *Rev. Aquat. Sci.*, 4, 45–73.
- Yeats, P. A., and C. I. Measures (1998), The hydrographic setting of the second IOC contaminants baseline cruise, *Mar. Chem.*, *61*, 3–14.
- Yiou, F., G. M. Raisbeck, G. C. Christensen, and J.-C. Gascard (2002), Temporal and spatial records of <sup>129</sup>I input to the Arctic from the nuclear fuel reprocessing facilities at Sellafield (UK) and La Hague (France), paper presented at the 5th International Conference on Environmental Radioactivity in the Arctic and Antarctic, St. Petersburg, Russia, 16–20 June.
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