Temporal evolution of the tracer signal in the Deep Western Boundary Current, tropical Atlantic

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Abstract. Four World Ocean Circulation Experiment (WOCE) repeat cruises (October 1990 to March 1994) in the tropical Atlantic off Brazil are used to study the spatial and temporal evolution of the chlorofluorocarbon (CFC) (components CFC-11 and CFC-12) and tritium signal in the upper North Atlantic Deep Water (NADW). Its shallowest part, located in the tropical Atlantic around 1600-m depth, is the shallow upper North Atlantic Deep Water (SUNADW). It is characterized by a distinct tracer maximum, which is presumably received through winter time convection in the subpolar North Atlantic. Here we discuss the tracer fields and the temporal evolution of the tracer signal of the SUNADW in the tropical Atlantic along two meridional sections at 44° and 35°W and two zonal sections at 5° and 10°S off Brazil. The spatial and temporal development of the tracer field in the tropical Atlantic as well as the correlation with hydrographic parameters show that the temporal tracer change being due to the arrival of "younger" water is disturbed by other processes. In particular, the impact of variable mixing and spreading pathways on the observed tracer variability in the SUNADW is evident in the observations.

1. Introduction

Interhemispheric exchange of deep water masses in the Atlantic occurs predominantly through the Deep Western Boundary Current (DWBC). Flowing along the continental margin of the American continent, the DWBC exports deep water of northern origin (North Atlantic Deep Water (NADW)) to the South Atlantic in a depth range between 1200 and 4000 m. In shallower layers a reverse flow of warmer southern origin water occurs. The near–bottom flow is also northward and carries cold, fresh Antarctic Bottom Water (AABW) into the northern hemisphere.

Four water masses can be distinguished in the DWBC (Table 1), the shallow upper NADW (SUNADW), the Labrador Sea Water (LSW), the lower NADW – old water mass (LNADW–old), and the overflow lower NADW (OLNADW). Two of these water masses are characterized by high concentrations of transient tracers like tritium (^3H) and chlorofluorocarbons (CFCs), and the two tracer maxima are nowadays present along the DWBC from the source region of the deep water masses in the northern North Atlantic to about 5° and to 20°S, respectively [*Wallace et al.*, 1994; *Rhein et al.*, 1995] (hereinafter, *Rhein et al.* [1995] is referred to as

RSS95). The deeper maximum characterizes the deepest part of the NADW, the OLNADW [RSS95; *Plähn and Rhein*, 1998], which is found around 3800-m depth in the tropical Atlantic. The high tracer signal reflects the convective renewal of one of the northern source water masses, and its oxygen and silica signal can also be followed along the western boundary [*Speer and Mc-Cartney*, 1991].

The upper tracer maximum belongs to the shallowest part of the NADW around 1600-m depth in the tropical Atlantic. This water mass, named shallow upper North Atlantic Deep Water (SUNADW) by RSS95, was not identified before the collection of transient tracer measurements and is named in other papers as upper LSW (ULSW). The tracer maximum indicates that it has been ventilated recently in the suggested formation region in the southern part of the Labrador Sea [*Pickart*, 1992; *Pickart et al.*, 1996]. Frequent observations since the 1970s suggest that it is a persistent feature of the DWBC. South of about 30°N, the salinity maximum of this water mass is correlated to elevated concentrations of tritium and CFCs all along the western boundary [e.g., *Weiss et al.*, 1985; *Fine and Molinari*, 1988; *Pickart*, 1992; *Molinari et al.*, 1992; RSS95], and the salinity and tracer maxima decrease farther downstream. The enhanced tracer levels of SUNADW indicate that it participates in the "fast response" of the meridional overturning cell.

The comparatively low CFC and tritium signal of the Labrador Sea Water (LSW), found around 2100-m depth below the SUNADW, has been attributed to incomplete convection during the 1960s and the 1970s [e.g., *Wallace and Lazier*, 1988; *Read and Gould*, 1992;

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Table 1. Limits and Characteristics of the Water
Masses in the Deep Western Atlantic.

Density Boundary	Property	Range	Approximate Depth, m
	SUNADW		
$\sigma_1 = 32.15$, $\sigma_{1.5} = 34.42$			1200
	Θ maximum	3.4°–4.5°C	
	CFC-11 maximum		
	S maximum	34.80–35.00	
	LSW		
$\sigma_2 = 36.94$, $\sigma_{1.5} = 34.70$			1900
	Θ	2.8°–3.4°C	
	O_2 maximum		
	S	34.94–34.98	
	LNADW-old		
$\sigma_2 = 37.00$, $\sigma_{1.5} = 34.755$			2400
	Θ	2.2°–2.8°C	
	O_2 maximum		
	CFC-11 minimum		
	S	34.91–34.95	
	OLNADW		
$\sigma_4 = 45.83$			3450
	Θ	1.75°–2.2°C	
	CFC-11 maximum		
	O_2 maximum		
	S	34.85–34.91	
	AABW		
$\sigma_4 = 45.90$			3900
	Θ	<1.75°C	
	decrease in O_2		
	S	<34.85	
	CFC-11 increase		

From *Rhein et al.* [1995]. Approximate depth is a mean at 35°W, 5°S. SUNADW is shallow upper North Atlantic Deep Water, LSW is Labrador Sea Water, LNADW-old is lower NADW-old water mass, OLNADW is overflow lower NADW, and AABW is Antarctic Bottom Water. Θ is potential temperature, S is salinity, O_2 is oxygen, CFC-11 is chlorofluorocarbon.

Sy et al., 1997]. Between the LSW and the OLNADW, there is a low tracer layer between temperatures of 2.2°–2.8°C. This water, the LNADW–old, leaves the formation region with low concentrations [*Smethie and Swift*, 1989; *Rhein*, 1994] and is, in general, more associated to lower velocities than to the zones with a tracer maximum [e.g., *Fine and Molinari*, 1988; *Watts*, 1991; *Richardson and Schmitz*, 1993].

Large variability was observed in the DWBC transports. *Fischer and Schott* [1997] found a seasonal cycle at the equator at 44°W, while *Vaughan and Molinari* [1997] described variability with a 2–3–year period off the Caribbean. Thus the DWBC cannot be regarded as a continuous constant flow.

In this study we discuss the spatial distributions and the temporal evolution of the CFC and tritium signal in the SUNADW, using four World Ocean Circulation Experiment (WOCE) repeat cruises with R/V *Meteor* in the years 1990–1994 along the meridional sections at 44° and at 35°W and along the zonal sections at 5° and at 10°S (Figure 1). The 10°S section was only occupied in November 1992 and in March 1994.

2. Methods

2.1. CTD Data

During all four cruises a conductivity–temperature–depth (CTD) model Neil Brown Mark III was used.
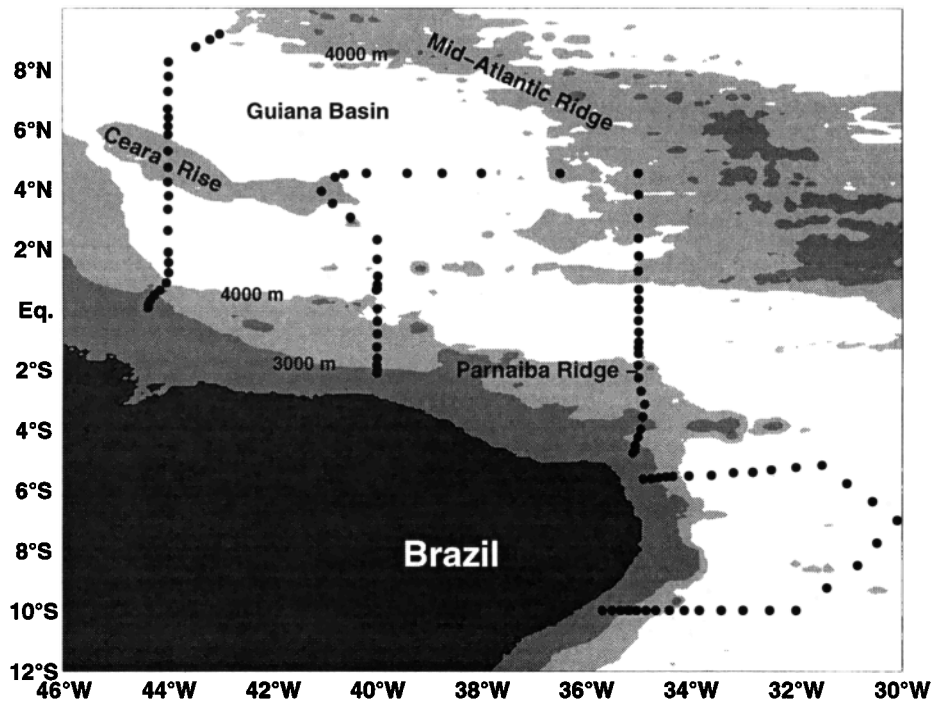
Figure 1. World Ocean Circulation Experiment (WOCE) repeat sections in the tropical Atlantic. The hydrographic and tracer data have been obtained during the R/V *Meteor* cruises in October 1990 (M14), May–June 1991 (M16), November 1992 (M22), and February–March 1994 (M27).

Water samples were collected to calibrate the conductivity sensor. The temperature accuracy is estimated to be 0.002°C for M14 (October 1990), M22 (November 1992), and M27 (February–March 1994) and estimated to be 0.003°–0.004°C for M16 (May–June 1991). The salinity accuracy is of the order of 0.003 for all cruises. The pressure calibration had a standard deviation of ±3 dbar compared to the thermometric pressure readings.

The temperature–salinity (Θ–S) plots in the range from 2.0° to 2.8°C show a bias in the deep Θ–S properties. In contrast to the SUNADW and OLNADW, this water layer is believed to be mostly unaffected by temporal changes in the tropical Atlantic because it is located in a zone with sluggish velocities [*Richardson and Schmitz*, 1993]. At 44°W the three *Meteor* cruises M14 (October 1990), M16 (June 1991), and M27 (March 1994) overlay. However, the salinities of these cruises are fresher by about 0.003 than M22 (November 1992). This offset is also visible in the salinity bottle data, where the M14, M16, and M27 data coincide well but are fresher than the M22 values by about 0.003. The M22 values from 1992, however, overlay in this temperature range with the bottle salinities taken by five cruises which were carried out with the R/V *Baldrige* between February 1990 and September 1991 (R.L. Molinari, personal communication, 1995). We believe the offset to be due to standard seawater differences of the salinometer calibration. In order to obtain an internally consistent data set of the four *Meteor* cruises the offset of 0.003 was subtracted from the salinities of the M22 cruise.

2.2. Chlorofluorocarbons

The CFCs were collected using precleaned 10-L Niskin bottles mounted on a CTD unit. They were analyzed on the ship using a gas chromatographic technique similar to that of *Bullister and Weiss* [1988]. During all cruises, only water below 700 m was sampled in order to get a better vertical resolution on the DWBC water masses. In the density range of the SUNADW, about 140 CFC-11 and 65 CFC-12 values were obtained in October 1990 (M14). In May–June 1991 the coverage was about 60 CFC-11 and 60 CFC-12 samples. In November 1992, about 190 CFC-12 samples and 250 CFC-11 values were achieved, and in February–March 1994 the numbers increased to 565 CFC-11 and 560 CFC-12 samples. The increase in 1994 was due in part to the additional sections along 40°W and 4°30'N.

The accuracy (±0.003–0.005 pmol kg^{-1}) of the CFC data was checked by closing two or more bottles at one depth. The blanks were cruise dependent between 0.003 and 0.009 pmol kg^{-1} and all data have been corrected by these blanks. The absolute calibration was done using gas standards kindly provided by R. Weiss, Scripps Institution of Oceanography (SIO); the data are reported relative to the SIO86 scale. To convert the data to the new SIO93 scale, divide the CFC-12 values by 0.9874 and the CFC-11 values by 1.0251 [*Cunnold et*

al., 1994]. For the low values found in the deep water in the tropical Atlantic, these differences between the scales are negligible.

2.3. Tritium, Helium

The sampling of the tritium and helium data was mainly done in 1990 on the sections along 44°W, 35°W, and 5°S. The temporal evolution of the tritium signal was only resolved along 5°S, where data from three cruises exist (October 1990 (M14), November 1992 (M22), and February–March 1994 (M27)). On each cruise, about 30 tritium samples were obtained in the SUNADW depth range at 5°S. In 1990, about 40 samples were achieved at 44°W and 30 samples at 35°W.

The samples were taken from the standard Niskin samplers and stored in pinched–off copper tubes for measurement of helium isotopes. Samples for low–level tritium analysis are collected in 1–L glass bottles. For ^{3}He analysis, samples are degassed in a special vacuum extraction system, and the extracted gasses are transferred into an ampoule made from low–helium–permeability glass. For low–level tritium measurement, about 500 cm^3 of each sample was degassed and stored for several months in low–permeability glass bulbs for ^{3}He ingrowth due to tritium decay. The measurements were performed using a dedicated helium isotope mass spectrometer following the procedures described by *Bayer et al.* [1989]. Usually, ^{3}He data are reported using a delta notation, where δ^3He is the percent deviation of the ^{3}He/^{4}He ratio of a water sample from that of atmospheric air (R_{air}=1.384x10^{-6}); ^{3}He is measured with a precision of the ^{3}He/^{4}He ratio measurement on the order of about ±0.2%. Precision of the tritium analysis is about ±1-2% with a detection limit of 3–5 mTU (1 TU means a tritium/hydrogen ratio of 10^{-18}; 1 mTU means 10^{-3} TU). To correct for a nontritiogenic ^{3}He component in the glass bulbs produced during ingrowth time by secondary cosmic rays (mainly neutrons [*Kurz and Brook*, 1994]), a correction of 10 mTU was applied to the tritium data. Including this correction, the overall accuracy of the tritium data reported in this contribution is estimated to be 10 mTU or better.

3. Tracer Background

During the past several decades the atmosphere and the upper ocean have been tagged with CFCs that had previously not existed in the environment. These tracers are introduced into the deep ocean in regions of deep water formation. The CFC concentrations increased exponentially till the mid–1970s, thus the CFC-11/CFC-12 ratios increased too. After 1975 the CFC increase was linear, and the ratios remained constant However, the annual percentage change in the CFC concentrations decreased with time [e.g., *Smethie*, 1993]. If the temporal evolution of the CFC signal in the SUNADW is dominated by the temporal evolution of the surface CFC concentrations, one would expect to find a CFC increase with time in the tropical Atlantic,

albeit smaller than observed at the surface and delayed by the transit time the SUNADW needs to arrive in the tropical Atlantic. However, the annual percent changes in the CFC concentrations should decrease with time.

Tritium, the radioactive isotope of hydrogen (half–life 12.43 years) was delivered to the atmosphere mainly during the surface nuclear weapon tests in the early 1960s and enters the ocean surface waters by water vapor exchange, precipitation, and river runoff [e.g., *Weiss and Roether*, 1980]. The anthropogenic "bomb" tritium signal masked the natural tritium level in ocean surface waters (about 0.2 TU) by several orders of magnitude. The maximum tritium concentrations in the convective regions of the northern North Atlantic observed in 1963 were about 18 TU [*Dreisigacker and Roether*, 1978]. After this time the atmospheric input to the ocean surface decreased continuously, and present–day tritium concentrations of subpolar North Atlantic surface waters are below 2 TU (Heidelberg Tritium Laboratory, unpublished data, 1997). If the temporal evolution of the tritium signal in the SUNADW is mainly determined by the temporal evolution of the atmospheric input into the surface water and the water started in the formation region after the stop of the atmospheric nuclear weapon testing, one would expect to find a tritium decrease with time in the tropical Atlantic. The tritium change is expected to be smaller than observed at the surface and delayed by the transit time that the SUNADW needs to arrive in the tropical Atlantic.

Tritium decays to ^{3}He with a half–life of 12.43 years [*Unterweger et al.*, 1980]. Tritiogenic ^{3}He elevates the ^{3}He/^{4}He ratio of the waters in the ocean (typical δ^3He values are between ~0% near the ocean surface and, depending on the tritium content, about 10-20% in the lower thermocline). For much of the ocean the tritiogenic ^{3}He component can be separated from the other helium components found in ocean waters (mainly atmospheric and mantle helium). In these cases an apparent ^{3}H/^{3}He age can be calculated. This apparent age reflects the true age of a water parcel only in cases where eddy diffusion is negligible compared to advection. Whenever mixing with waters bearing tritium and ^{3}He cannot be excluded, appropriate mixing models are needed to develop a ventilation age or mean residence time of a water mass from the ^{3}H/^{3}He data (for details of the ^{3}H/^{3}He dating method, see, e.g., *Schlosser and Smethie* [1995]). In the tropical Atlantic the SUNADW mixes with the overlying Upper Circumpolar Water (UCPW) [*Stramma*, 1991], which is ^{3}H and CFC poor but contains a prominent portion of nontritiogenic ^{3}He, limiting the application of ^{3}H/^{3}He ages to date the formation of SUNADW.

4. Results

4.1. Tracer Distributions

The sections along 44°W for the 1990, 1992, and 1994 cruises present the main features of the tracer fields in the tropical Atlantic (Figure 2). The SUNADW around
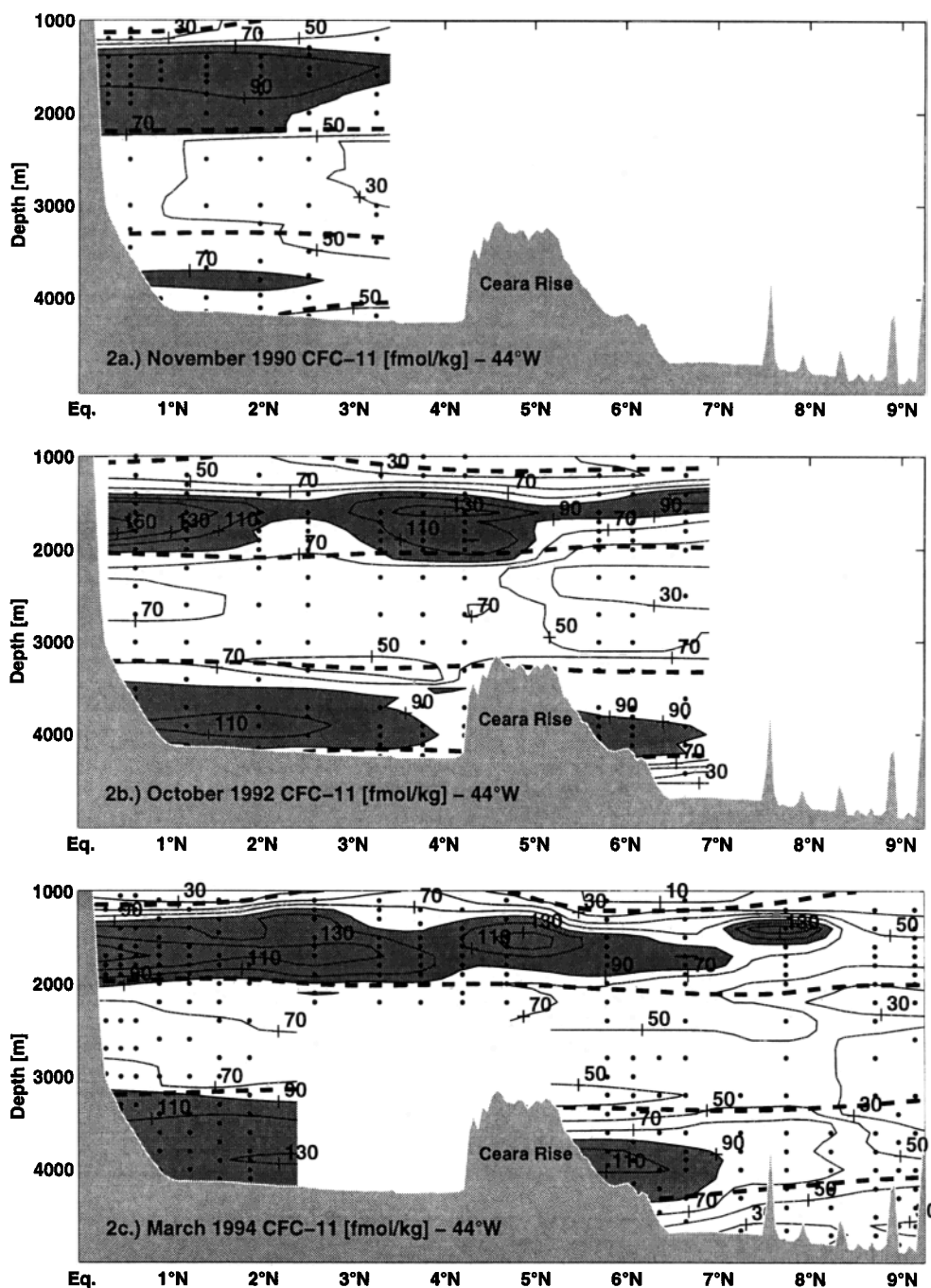
Figure 2. CFC-11 distribution along 44°W for (a) October 1990, (b) November 1992, and (c) March 1994. The 1991 data are not shown because only three stations were sampled for CFCs. Note the different horizontal extension of the sections. The CFC maxima of the shallow upper North Atlantic Deep Water (SUNADW) and the overflow lower North Atlantic Deep Water (OLNADW) are shaded. Tritium distribution along (d) 44°W, October 1990; (e) 35°W, October 1990; (f) 5°S, October 1990; (g) 5°S, November 1992; and (h) 5°S, March 1994. The tritium maxima >100 mTU of SUNADW and OLNADW are shaded. Note the coarser spatial resolution of the tritium data compared to the CFC data set. The boundaries of the SUNADW are indicated by the isopycnals $\sigma_{1.5} = 34.45$ and $\sigma_{1.5} = 34.70$ (dashed lines), and the boundaries of the OLNADW are indicated by the isopycnals $\sigma_4 = 45.83$ and $\sigma_4 = 45.90$ (dashed lines).
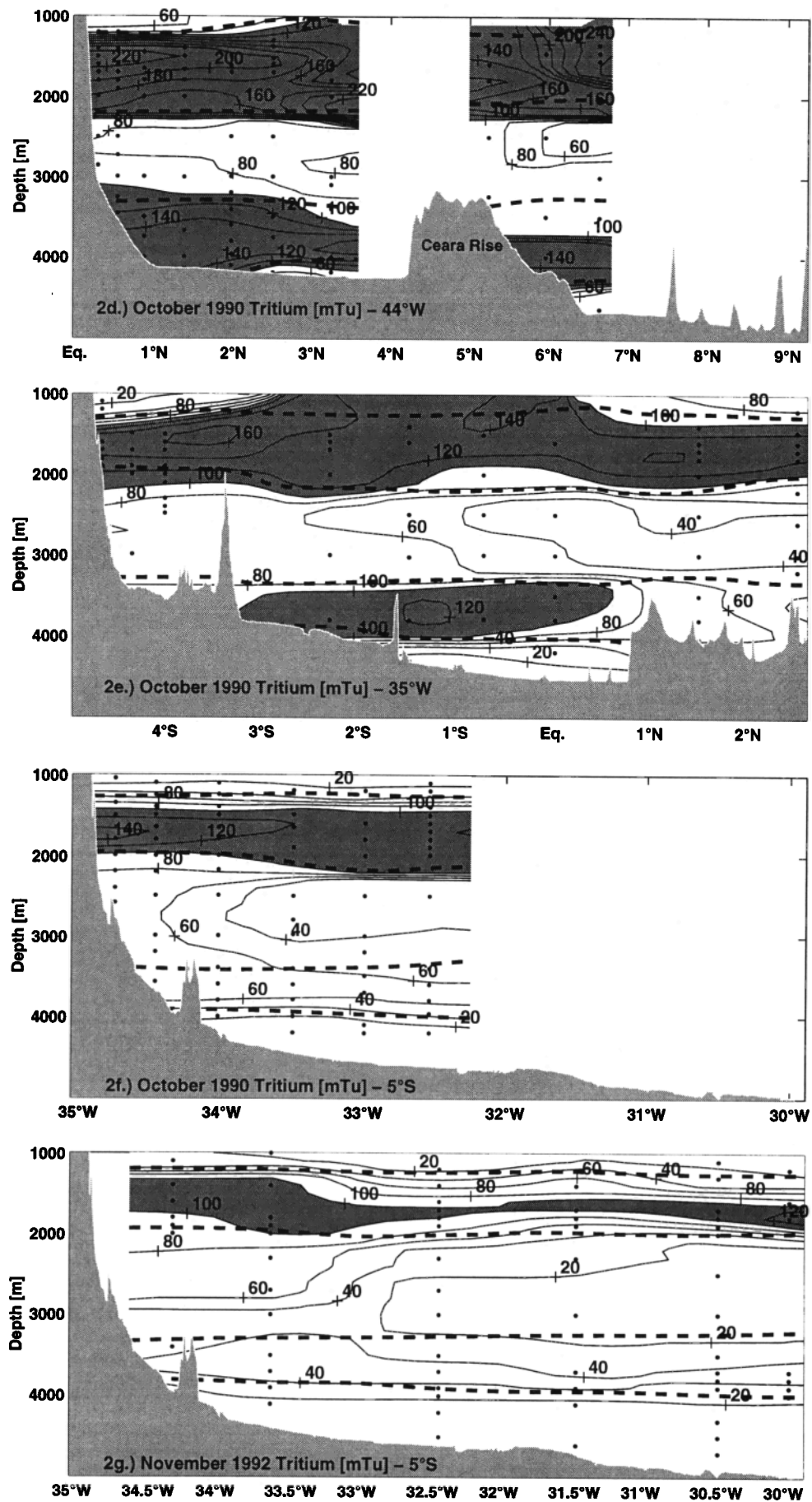
2d.) October 1990 Tritium [mTu] – 44°W

2e.) October 1990 Tritium [mTu] – 35°W

2f.) October 1990 Tritium [mTu] – 5°S

2g.) November 1992 Tritium [mTu] – 5°S
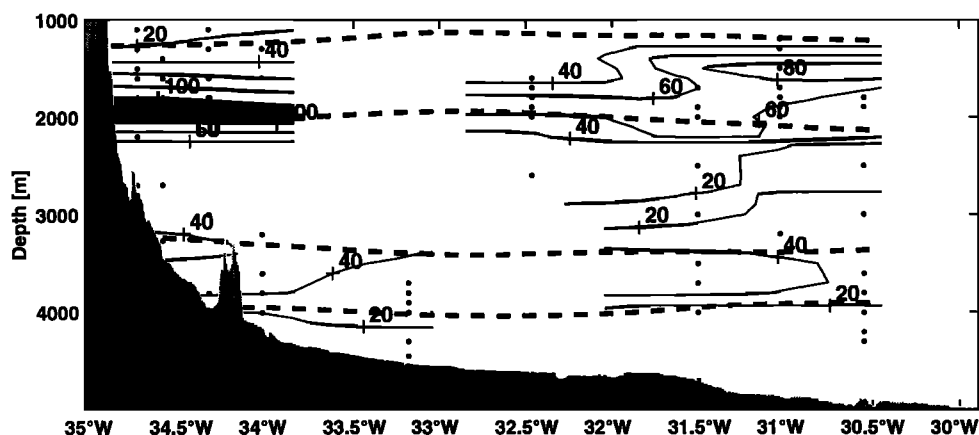
Figure 2. (continued)

Figure 2. (continued)

1600-1800-m depth and the OLNADW around 3800-m depth are characterized by CFC-11 maxima. The upper maxima are not bounded to the topography but extend farther north (Figures 2b and 2c), presumably caused by the recirculation of this water mass in the tropical Atlantic [*Richardson and Schmitz*, 1993]. The OLNADW core at 3800 m is split by the Ceara Rise at 5°N (Figures 2b and 2c), and the CFC-11 concentrations on the northern side of the Ceara Rise are only slightly lower than in the DWBC. This pattern is consistent with the flow field [*Schott et al.*, 1993], characterized by eastward flowing cores south of Ceara Rise and at the northern flank of the rise.

The 1990 tritium section along 44°W (Figure 2d) delineates the same basic features as obtained from CFC-11. Compared to the surrounding water masses, SUNADW and OLNADW are characterized by elevated tritium concentrations, which reflect their convective renewal in the northern North Atlantic. In detail, the signature of tritium, CFC-11, and salinity correlates well both in the upper and in the lower maximum. Highest tritium values are found in the DWBC at the continental slope (SUNADW, 240 mTU at about 1600 m; OLNADW, 161 mTU at about 3750 m). In the OLNADW located north of the Ceara Rise the maximum tritium concentrations are only moderately lower (158 mTU). Below the SUNADW the tritium values decrease continuously, reaching a minimum (<80 mTU) in the LNADW-old located directly above the OLNADW.

The CFC-11 distributions as well as the flow fields in the DWBC along 35°W, 5°S, and 10°S have been discussed by RSS95, and the tracer field and circulation of the AABW in 1994 is studied by *Rhein et al.* [1998]. The temporal evolution of the CFC concentrations in the OLNADW and the recirculation in the Guiana Basin are discussed by *Plähn and Rhein* [1998]. Thus only the tritium distribution at 35°W and at 5°S is described in the following.

In Figure 2e the tritium distribution on the 35°W section as obtained in 1990 is depicted. Again, high tritium values >150 mTU in the depth range of 1500-

1800 m, extending all along the transect, reflect the comparatively good ventilation of the SUNADW. Two cores with maximum tritium concentrations bounded at the continental slope and at about 1.5°N give evidence for recirculation processes and spreading of SUNADW in different branches. The core of OLNADW with a tritium content of ~120 mTU is centered at 3600-3800-m depth. The decline of the concentrations in the intermediate layers comprising LSW and LNADW-old corresponds to the tritium distribution obtained at 44°W.

The zonal section along 5°S was sampled for tritium in the years 1990, 1992, and 1994 (Figures 2f-2h). Again, the tritium distribution resembles the vertical stratification of water masses obtained farther north and west and reflects the main features of the circulation. SUNADW shows two distinct cores at about 1550-1800-m depth: one situated at the continental slope and the other located near 31°W. The lower tritium maxima indicating the OLNADW in the depth range of 3400-3800 m cover the full section with the highest tritium values detected in the DWBC close to the continental slope. The temporal decrease of the tritium concentrations in SUNADW and OLNADW is clearly visible (see section 4.2).

4.2. Intercomparison of the CFC Data Sets

To check the consistency of the CFC data set of the four cruises is a rather difficult task. Because of the atmospheric CFC increase with time, one expects an increase in the CFC concentrations of the various deep water masses, most significant in the SUNADW and the OLNADW. The increase is presumably higher nearest to the formation region of the water masses but is also present farther downstream. The increase is unknown and could be obscured, for instance, by the variability in the horizontal mixing with "older" and thus CFC poorer water masses. Furthermore, vertical mixing could also enhance the tracer signal of the CFC poor water masses located between the two tracer maxima.

In our data set the smallest impact of the transient nature of the CFC signal is most likely found in the
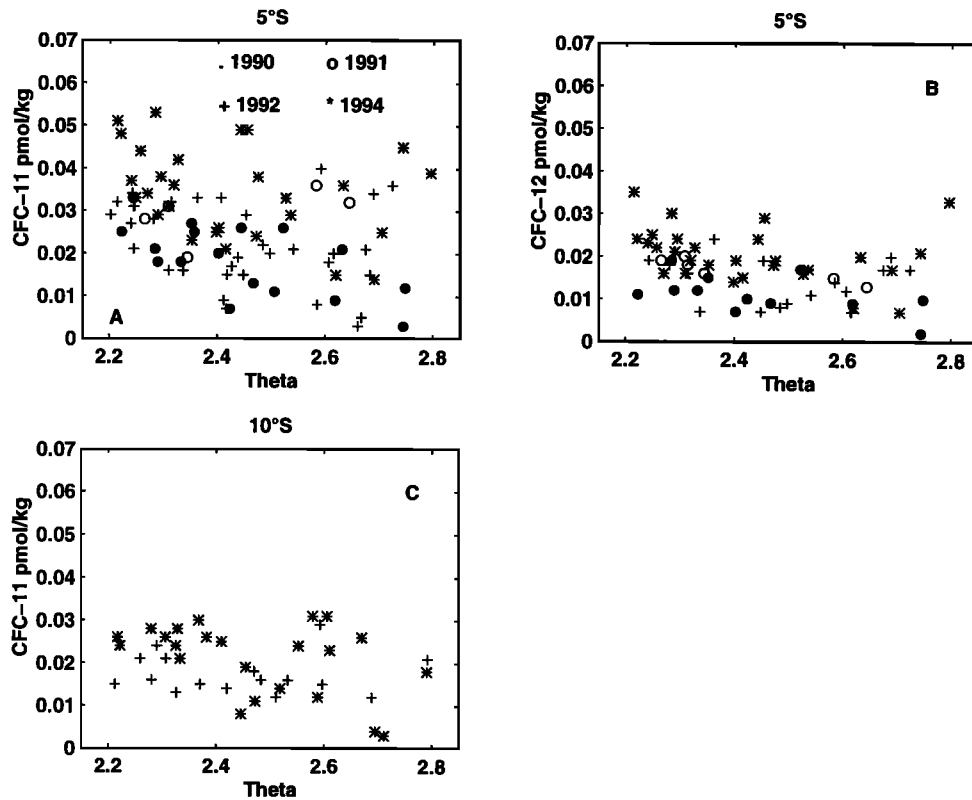
Figure 3. Potential temperatures for (a) CFC-11 at 5°S, (b) CFC-12 at 5°S, and (c) CFC-11 at 10°S for LNADW–old.

tracer poor layer between 2.2° and 2.8°C (about 2400–3400-m depth) at 5° and 10°S. This LNADW–old is located above the OLNADW, which is characterized by a CFC maximum, but the signal is relatively low at these two sections. At 5°S the CFC-11 and CFC-12 data sets of all four cruises can be compared (Figures 3a and 3b).

Only the 1994 data differ from the previous cruises and are higher by about 0.008 pmol kg^{-1} CFC-11 and 0.007 pmol kg^{-1} CFC-12 for temperatures lower than 2.45°C. At 10°S, only the CFC-11 concentrations measured in 1992 and 1994 can be compared, because no CFC-12 data exist for the 1992 cruise. Here (Figure 3c) no offset between the 1992 and 1994 data set is observed for temperatures higher than 2.45°C, whereas for lower temperatures the 1994 values are slightly higher. The CFC-11 signal of OLNADW at 5° and 10°S increased from 1990 to 1994, thus the lower temperature range of the LNADW–old could be influenced by this increase through vertical mixing.

To conclude, the four CFC data sets seem to be consistent, and the weak increase from 1992 to 1994 in the LNADW–old could be caused by the CFC increase with time. On the other hand, we cannot exclude the possibility that the 1994 data set might show an offset compared to the other three previous cruises by about 0.008 pmol kg^{-1} CFC-11 and 0.007 pmol kg^{-1} CFC-12. However, the temporal CFC increase in the SUNADW and OLNADW is significantly higher, so that we ne-

glect this possibility in the following. At the low CFC levels present in the DWBC in the tropical Atlantic ($<$0.1 pmol kg^{-1}) the blank of the measurements is the dominant error, and the intercomparison at the CFC poor level (Figure 3) shows that the blank of the various cruises has been determined reasonably well to obtain an internally consistent data set. The two SIO86 calibration standards used during the four cruises are accurate on a 1% level (for our data $<$0.001 pmol kg^{-1}). Errors in the volume determination of the sample loops used for the calibration are presumably smaller than 5%. On all four cruises the same sample loops have been used, so that this possible error is not relevant for the presented data set but might have to be taken into account when compared to data sets from other labs.

4.3. Mean Tracer Distributions

In order to compare the tracer and salinity distributions of the SUNADW along a section from different times, the data along a section were interpolated on a regular grid in density space by objective analysis (horizontal grid: 0.6°; vertical grid: 0.05 $\sigma_{1.5}$ units). The mean tracer concentration on each horizontal grid point was then calculated by averaging the tracer concentrations between the density limits of the water mass. The densities $\sigma_{1.5} = 34.55$ and $\sigma_{1.5} = 34.70$ are chosen as upper and lower boundaries. We choose the $\sigma_{1.5} = 34.55$ as the upper limit instead of $\sigma_{1.5} = 34.45$
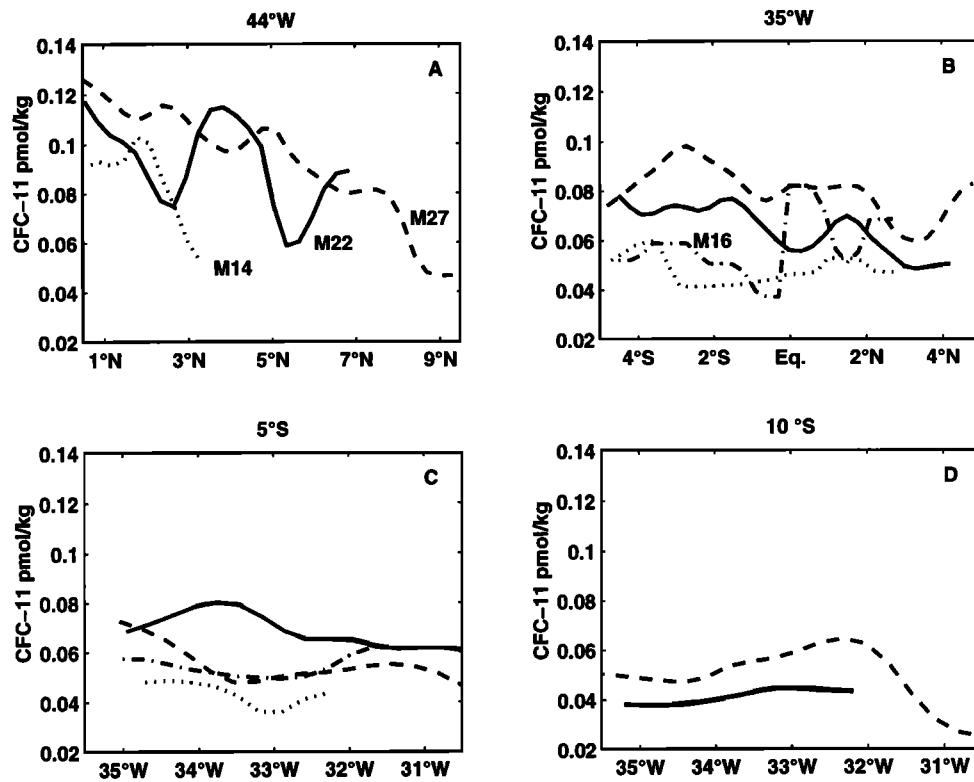
Figure 4. Horizontal distribution of the vertically averaged CFC-11 data (pmol kg^{-1}) of SUNADW for the cruises M14, October 1990 (dotted line); M16, May–June 1991 (dashed–dotted line); M22, November 1992 (solid line); and M27, February–March 1994 (dashed line) (a) along 44°W, (b) along 35°W, (c) along 5°S, and (d) along 10°S. Because of the very coarse resolution at 44°W in June 1991 (M16) with only three CFC profiles, only the data for the 35°W and the 5°S section are presented. Upper and lower boundaries for SUNADW are $\sigma_{1\,5} = 34.55$ and $\sigma_{1\,5} = 34.70$, respectively.

(RSS95) to avoid the strong vertical tracer gradients separating the low tracer zone of the Upper Circumpolar Water from the SUNADW-tracer maximum.

At 44°W the highest mean CFC-11 values (Figure 4a) are found near the Brazilian continental slope in the DWBC [*Schott et al.*, 1993]. The decrease to the north is not continuous due to the presumed recirculation in the Guiana Basin [*McCartney*, 1993]. In general, the CFC-11 concentrations increase from 1990 to 1994. High CFC-11 in the SUNADW is correlated with high salinity (Figure 5). The highest CFC-11 and salinity values were observed in November 1992 located close to the Brazilian continental slope. This feature is obscured in Figures 4a and 6a by the interpolation of the objective analysis and the vertical averaging. As mentioned above, the salinity maximum of SUNADW decreases downstream in the subtropical and tropical Atlantic by mixing, as reflected in the salinity decline from 44°W to 10°S (Figure 6).

At 35°W the mean CFC-11 concentrations in 1991 just north of the equator are as high as observed in 1994

(Figure 4b). The high CFC-11 values are connected to the highest mean salinity at 35°W (Figures 5b and 6b). Except for the region just north of the equator at 35°W, CFC-11 mainly increased with time (Figure 4b).

At 5°S the CFC-11 signal in 1992 (M22) is higher everywhere compared to 1994 (Figure 4c), similar to the salinity distribution; that is, the higher salinities observed in November 1992 compared to March 1994 are correlated with higher CFCs (Figure 6c). As for the 44°W and the 35°W sections (Figures 5a and 5b), a linear correlation between salinity and CFC-11 seems to exist for the SUNADW (Figure 5c). The lowest mean CFCs and salinities are found in 1990; the observations from 1991 and 1994 are slightly higher. Apparently, the temporal CFC increase in the surface waters, which should be mirrowed by an appropriate temporal CFC increase in the SUNADW from 1990 to 1994, is obscured at 5°S by other processes, most likely mixing with the overlying UCPW as discussed below. The salinity and CFC decrease of SUNADW at 5°S from November 1992 to March 1994, however, was limited to this water mass.
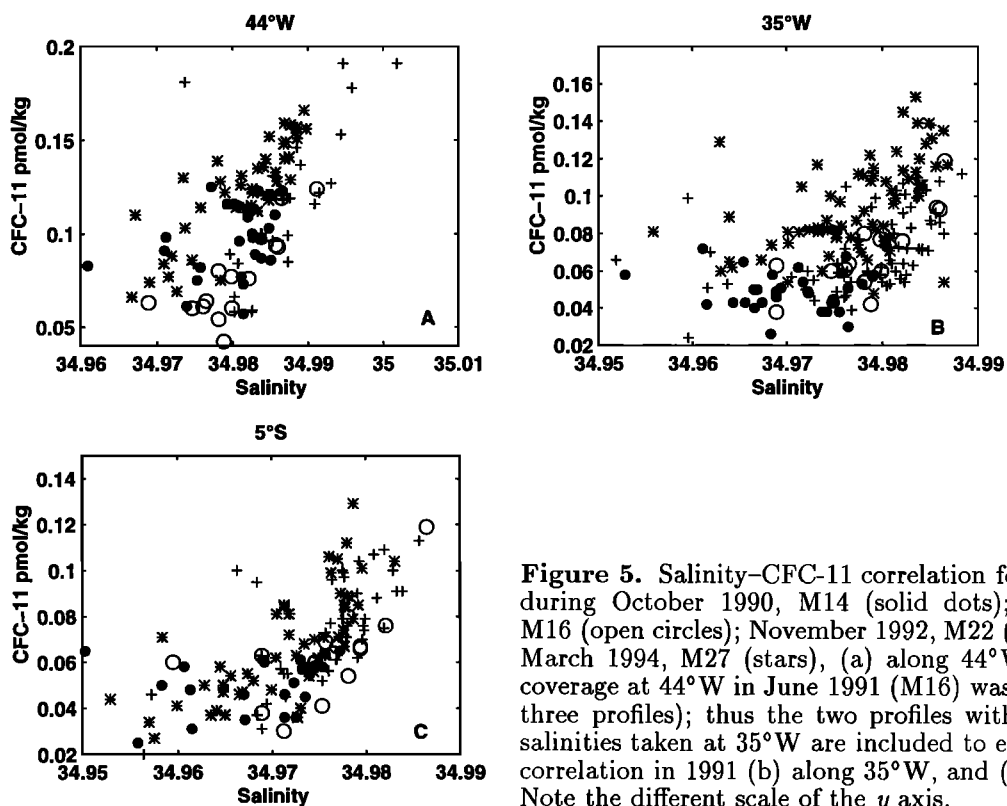
Figure 5. Salinity–CFC-11 correlation for SUNADW during October 1990, M14 (solid dots); June 1991, M16 (open circles); November 1992, M22 (crosses); and March 1994, M27 (stars), (a) along 44°W. The CFC coverage at 44°W in June 1991 (M16) was sparse (only three profiles); thus the two profiles with the highest salinities taken at 35°W are included to emphasize the correlation in 1991 (b) along 35°W, and (c) along 5°S. Note the different scale of the y axis.
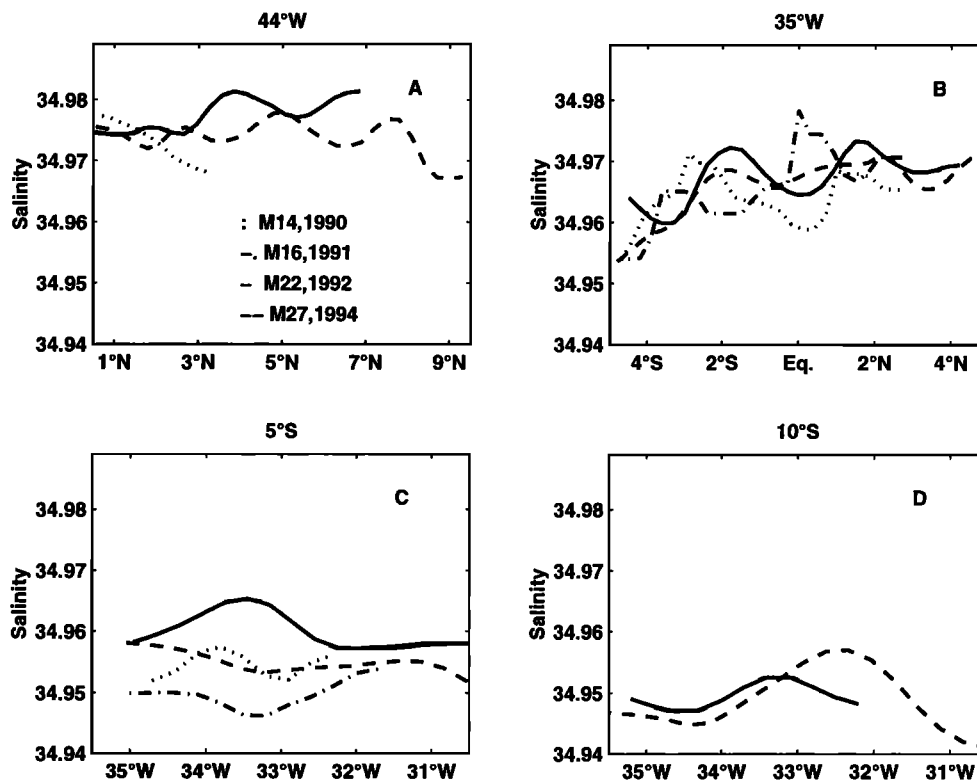
Figure 6. Horizontal distribution of the vertically averaged salinity of SUNADW for the cruises M14, October 1990 (dotted line); M16, May–June 1991 (dashed–dotted line); M22, November 1992 (solid line); M27, and February–March 1994 (dashed line) (a) along 44°W, (b) along 35°W, (c) along 5°S, and (d) along 10°S. Upper and lower limits for SUNADW are $\sigma_{1.5} = 34.55$ and $\sigma_{1.5} = 34.70$, respectively.

The CFC maximum at about 3800-m depth observed in March 1994 was everywhere higher in March 1994 than in November 1992 [*Plähn and Rhein*, 1998].

In March 1993, *Andrie* [1996] found between 1600- and 1800-m CFC-11 levels, between 0.03 and 0.11 pmol kg^{-1} at 35°W and between 0.03 and >0.075 pmol kg^{-1} at 5°S. In November 1992, i.e., four months earlier, we observed at 35°W and at 5°S, CFC-11 concentrations in the same depth level between 0.06 and 0.13 pmol kg^{-1}; that is, the two data sets are compatible for the SUNADW. They are also compatible for the deep tracer core, the OLNADW [*Andrie*, 1996; *Plähn and Rhein*, 1998]. No temperature or salinity data of the SUNADW are reported in the two aforementioned papers, preventing a more detailed comparison of the two CFC data sets.

The tritium concentrations decrease from 1990 to 1994 (Figure 7a), as expected from the temporal decline of the atmospheric fluxes into the ocean. If we take the radioactive decay of tritium into account and relate the measured tritium values to a common date (we chose the year 1981 to refer to the data obtained during the (TTO-NAS) program; decay corrected tritium data are reported as TU81), the temporal gradient steepens considerably between November 1992 and March 1994 (Figure 7b). This indicates that a more intensive mixing with tritium poor water masses affects the tritium values in March 1994, coincident with the findings from the CFC distributions. The UCPW is characterized by low ^{3}H (about 23 mTU at 5°S in 1990) values. That means that the tritium decline expected

from the atmospheric fluxes into the ocean is amplified by mixing with UCPW, whereas for the CFCs, more intensive mixing with UCPW counteracts the expected temporal increase.

The apparent ^{3}H/^{3}He ages of the SUNADW at 5°S varied from 38±4.5 years (1990) and 44.1±7.1 years (1992) to 47.8±9.6 years (1994). If one interprets theses values as true ages, the SUNADW observed between 1990 and 1994 in the tropical Atlantic would have been formed in the 1950s. However, most of the ^{3}He excess of the UCPW is due to the contribution of mantle helium and not produced by tritium decay. A straightforward conversion of the mean δ^3He signal (about 2% at 5°S in 1990) into tritium units results in a tritium equivalent of the ^{3}He excess in the order of 860 mTU, a value much higher than the average tritium concentration of UCPW in 1990 (23 mTU, see above), showing clearly that within a reasonable precision the tritiogenic ^{3}He component cannot be separated. Mixing of SUNADW with UCPW therefore limits the application of the ^{3}H/^{3}He age to date the formation of the SUNADW. However, as *Schlosser and Smethie* [1995] pointed out, in some cases the CFC-11/^{3}H ratio might be a useful parameter for the dating of young water masses formed between the mid-1970s and present. In Figure 7c the temporal evolution of the CFC-11/^{3}H ratio (calculated using the tracer input functions for the North Atlantic, tritium is decay-corrected to 1981) is depicted. Apparently, during the last decades the ratio increases as a function of time at a rate of several percent per year, yielding a time resolution of about ±1 to
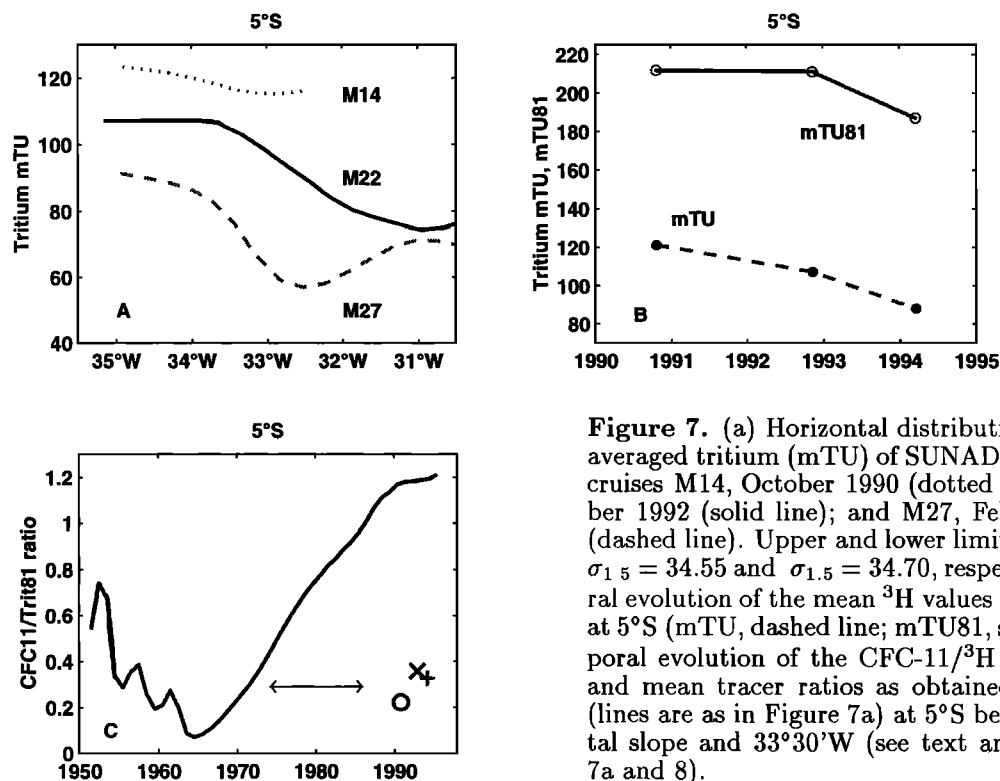
Figure 7. (a) Horizontal distribution of the vertically averaged tritium (mTU) of SUNADW along 5°S for the cruises M14, October 1990 (dotted line); M22, November 1992 (solid line); and M27, February–March 1994 (dashed line). Upper and lower limits for SUNADW are $\sigma_{1.5} = 34.55$ and $\sigma_{1.5} = 34.70$, respectively. (b) Temporal evolution of the mean ^{3}H values of SUNADW values at 5°S (mTU, dashed line; mTU81, solid line). (c) Temporal evolution of the CFC-11/^{3}H ratio of SUNADW and mean tracer ratios as obtained in different years (lines are as in Figure 7a) at 5°S between the continental slope and 33°30'W (see text and compare Figures 7a and 8).

±2 years. Also shown in Figure 7c are the mean decay-corrected CFC-11/^{3}H ratios obtained in different years at 5°S between the continental slope and 33°30'W. Evidently, the age information derived by this procedure is ambiguous as the temporal trend of the CFC-11/^{3}H ratio is characterized by a minimum in the mid-1960s due to maximum input of bomb tritium. However, we may combine the information obtained from the tracer ratio and from the temporal trend of the tritium concentration and conclude that the SUNADW observed at 5°S must have been formed in the early 1970s after the maximum tritium input occurred. The time span between the respective years of formation and observation (represented by an arrow in Figure 7c) of the order of about 20 years may be interpreted as propagation time from the area of SUNADW formation to the tropical Atlantic. The bias in the range of ±1 years as observed in Figure 7c probably is due to different mixing histories of the SUNADW and the procedure used to estimate the CFC-11/^{3}H ratios.

At 10°S the mean CFC-11 values increased by about 0.01 pmol kg^{-1} from 1992 to 1994 (Figure 4d), and the mean salinities changed slightly (Figure 6d). The CFC-12 distribution repeats the discussed features of CFC-11, although the database is somewhat coarser.

5. Discussion

The SUNADW is formed in the subpolar North Atlantic, where it obtains the tracer signal by contact with the atmosphere. After formation the water mass joins the DWBC and spreads continually to the south and ultimately into the South Atlantic. Because of the rising atmospheric CFC concentrations since the 1930s, one expects the CFC signal of SUNADW to increase with time; "younger" water, which carries a higher CFC load, subsequently enters the tropical Atlantic. The CFC-11/CFC-12 ratios [e.g., Weiss et al., 1985] and the annual CFC increase with time [e.g., Smethie, 1993] can be used to calculate the year a water parcel left the surface and started its journey in the deep ocean.

These CFC dating methods calculate velocities of the order of 1 cm s^{-1} [e.g., Smethie, 1993], which would point to about 30 years that the water needs to propagate to the tropical Atlantic. There is evidence that the propagation is significantly faster. At 26.5°N in the subtropical Atlantic an abrupt CFC increase in the LSW was observed in July 1996 [Molinari et al., 1998]. CFC-11 increased from 0.5 pmol kg^{-1} in 1994 to >1.0 pmol kg^{-1} in 1996. The CFC increase was accompanied by a cooling and thickening of the LSW layer (M. McCartney, personal communication, 1996), and these features are very likely caused by the arrival of LSW, which was formed after the onset of intensified deep convection in the Labrador Sea in 1988. Similar surprisingly rapid spreading rates for LSW have been found across the subpolar North Atlantic [Sy et al., 1997].

The discrepancy between the calculated propagation and the observed ones is presumably caused by shortcomings in the assumptions used in the CFC dating methods [Pickart et al., 1989], leading to an underestimation of the spreading velocity. Using more realistic boundary conditions for the formation of OLNADW and SUNADW and taking into account self-mixing in the DWBC and mixing with its surrounding water masses, the box models tuned by CFC and tritium observations in the DWBC point to mean velocities of the tracer-bearing water masses in the DWBC of 5–10 cm s^{-1} [Pickart et al., 1989; Rhein, 1994; Rhein et al., 1996].

The tracer propagation speed is nevertheless significantly lower than direct measurements [e.g., Schott et al., 1993]. From current meter moorings in the tracer bearing cores of the DWBC, annual mean velocities as high as 25 cm s^{-1} were reported [e.g., Fischer and Schott, 1997]. As the tracer distributions are not only influenced by advection but also by turbulent mixing on different spatial and temporal scales (e.g., recirculation gyres, eddy processes, self-mixing, and mixing with surrounding water masses), even the mean flow field of the DWBC derived from current meter moorings might overestimate the net Lagrangian parcel motion.

The longer the time that the water propagates, the greater is the influence of mixing with surrounding water masses and, through the recirculation gyres, with water masses formed in different years and probably with different hydrographic characteristics. The recirculation gyres and thus the pathways of the water masses might vary with time. Even the spreading along the DWBC might be subject to temporal variability, similar to the variability observed in the DWBC transports. These have timescales from 40 to 60 days [Pickart and Watts, 1990; Johns et al., 1993; Schott et al., 1993]. The current meter moorings in the DWBC at 44°W show a seasonal cycle, ranging from 7 Sv during September/October to about 25 Sv during January/February [Fischer and Schott, 1997]. In regions with high along-current tracer gradients the pulsing might influence the tracer signal at a certain location.

Thus the straightforward interpretation of a CFC increase with time at a certain location in the boundary current being only caused by the arrival of younger water might be too simple. The temporal variability in the mixing histories and/or recirculation could influence or even obscure the increase of the CFCs on an individual location in the boundary current as observed at 5°S between November 1992 and March 1994. The CFC distribution of the SUNADW in the tropical Atlantic gives evidence to the significant impact of the above mentioned processes.

The presumed recirculation of SUNADW in the Guiana Basin [e.g., Richardson and Schmitz, 1993; McCartney, 1993] stretches the high tracer values to the northern limit of our 44°W section (Figure 2). The correlation between the CFC-11 and salinity maxima (Fig-

ure 5) and the tritium maxima lead us to the conclusion that the maxima represent less diluted SUNADW. The maxima probably reflect in an indirect manner the recirculation, which has been observed directly with sound fixing and ranging (SOFAR) floats [*Richardson and Schmitz*, 1993].

The salinity and tracer tongues spreading east in the equatorial region [*Weiss et al.*, 1985; *Kawase and Sarmiento*, 1986; *Andrie et al.*, 1998] suggest that one part of the NADW follows the coastline of South America and one part flows parallel to the equator. The tracks of SOFAR floats show reversing equatorial currents with timescales of the order of months [*Richardson and Schmitz*, 1993]. High variability is present in the hydrographic and tracer fields in the equatorial band at 35°W. Similar to the 44°W section, the CFC–salinity correlation reveals the importance of variable mixing and changing pathways on the observed CFC concentrations. The complex flow field in the equatorial band might also be responsible for the advent of high-salinity and high-CFC water in 1992 followed by a decline of both parameters in 1994.

Another problem might be the choice of the horizontal extent of the boundary current. Figures 3–7 and the velocity fields presented by RSS95 illustrate the difficulties of defining the horizontal limit of the DWBC and thus the region to calculate the mean tracer concentrations. The values presented in Figure 8 are averages from the continental slope to 2°N at 44°W, to 2°S at 35°W, and to 33°30'W at 5°S and at 10°S. Extending the areas offshore to 3°N at 44°W, to 2°N at 35°W, and to 32°W at 5°S and 10°S, the general features of Figure 8 remain the same except for minor details. The CFC-11 increase for the mean values presented in Figure 8 varies for the different sections and time intervals between 23% per year and -14% per year. The annual percent change in the CFC concentrations reveals no spatial or temporal pattern. If the annual percent change in the SUNADW in the tropical Atlantic would be dominated by the atmospheric trend, one would, in general, expect to find decreasing percent changes with time [*Schlosser and Smethie*, 1995].

Another striking feature of Figures 8a and 8b is that the 5°S CFC-11 and CFC-12 values are almost equal to the mean concentrations found at 35°W for the years 1990 (M14), 1991 (M16), and 1992 (M22); only in spring 1994 (M27) a CFC decrease from 35°W to 5°S was observed. This behavior is in contrast to the salinity dis-
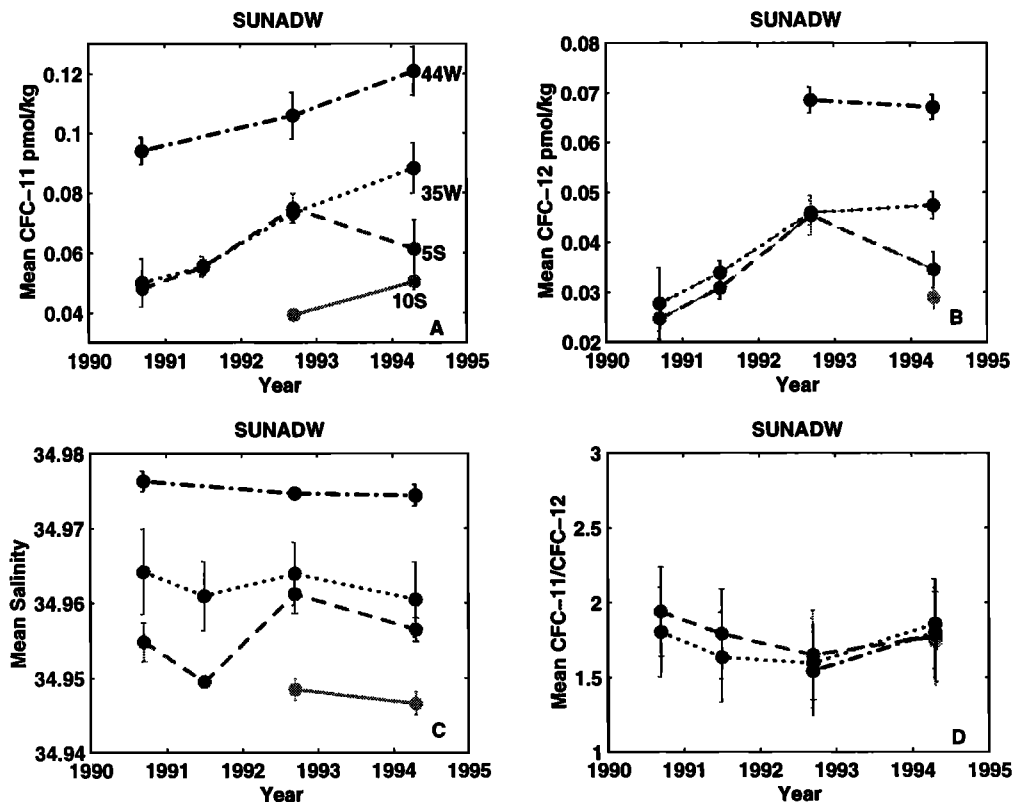

Figure 8. Temporal evolution of the mean (a) CFC-11, (b) CFC-12, (c) salinity, and (d) CFC-11/CFC-12 ratios; averaged from the continental slope off Brazil to 2°N at 44°W, to 2°S at 35°W, and to 33°30'W at 5°S and at 10°S. The values are calculated from the spatial distributions presented in Figures 4 and 6; the standard deviations calculated from averaging these data are included.

tributions. The mean salinity is lower at 5°S than at 35°W during all four cruises (Figure 8c). One would expect that the downstream salinity decrease from the 35°W to the 5°S section is accompanied by a downstream tracer decrease, as evident in the salinity and tracer decrease from 44° to 35°W (Figures 8a–8c).

Apparently, this is not the case for the mean CFC concentrations at 35°W and at 5°S in the years 1990, 1991, and 1992, and this is not the case for the mean oxygen and tritium concentrations at 35°W and 5°S. There is no difference in the oxygen values (about 5.5 mL L^{-1}) at 35°W and at 5°S in 1991 (M16) and in 1992 (M22), but in 1994 (M27), like the CFCs, the mean oxygen concentration was lower at 5°S (5.3 mL L^{-1}) than at 35°W (5.5 mL L^{-1}). Oxygen data were not taken in October 1990 (M14), but at this cruise, tritium was analyzed on all sections. There was a mean tritium decrease from 44° (355 mTU81, south of 2°N) to 35°W. The mean tritium concentration at 35°W (260 mTU81, south of 2°S), however, was only partly higher than at 5°S (212 mTU81, west of 33°30'W). These independent oxygen and tritium measurements support the findings from the CFC data. To explain these features, more understanding of the spatial and temporal variability of the flow field, the mixing processes, and the temporal variability of the SUNADW characteristic are needed.

The mean CFC-11 and CFC-12 concentrations of the DWBC are low in the tropical Atlantic. If the CFC-11 and CFC-12 concentrations are 0.06 and 0.03 pmol kg^{-1}, respectively, an accuracy of ±0.003 pmol kg^{-1} for each of the components leads to an uncertainty in the CFC-11/CFC-12 ratio of 2.0±0.3. Within these uncertainties the ratios did not change significantly between 1990 and 1994 in the research area (Figure 8d).

6. Conclusions

The spatial and temporal evolutions of the CFC and tritium distributions in the SUNADW of the tropical Atlantic have been studied using the observations on four WOCE repeat cruises carried out between October 1990 and March 1994. The atmospheric CFC increase with time should be reflected by a CFC increase in the SUNADW in the tropical Atlantic, although diminished by mixing with older SUNADW and with surrounding water masses on the way from the formation region to the tropical Atlantic. At 44°W, at 35°W, and at 10°S the mean CFC values in the SUNADW off Brazil increase with time along the individual sections. However, at 5°S between November 1992 and March 1994 the CFC concentrations decreased, indicating more intensive mixing in March 1994 with CFC poor UCPW. The decay–corrected temporal ^{3}H decrease was almost negligible between October 1990 and November 1992. A significant decrease (about 12 mTU81 per year) was observed in the time period November 1992 to March 1994, also indicating that the 1994 data are influenced

by more intensive mixing with ^{3}H poor UCPW. Mixing with CFC poor and ^{3}H poor UCPW augments the temporal decline of the ^{3}H values of SUNADW but counteracts the expected temporal increase of the CFC values. On the contrary, mixing of SUNADW and UCPW should not dramatically affect the CFC-11/^{3}H ratio of the SUNADW due to the low CFC and ^{3}H concentrations of the UCPW. Our observations at 5°S point to a ventilation age of SUNADW of the order of 20 years, which corresponds to a mean transfer velocity from the area of SUNADW formation to the tropical Atlantic of about 1.5 cm s^{-1} and gives further evidence that the propagation of SUNADW is faster than before believed.

The locally different linear correlations of the CFC maxima with salinity maxima in the SUNADW suggest that variability in the mixing history and in the spreading paths of the SUNADW have a significant impact on the tracer field in the tropical Atlantic. The arrival of more saline and thus less diluted SUNADW increases the observed CFC concentrations. Variability in spreading and mixing might obscure the expected CFC increase caused by the arrival of water, which was formed more recently.

The ^{3}H distributions support the conclusions drawn from the CFC observations. The diminishing atmospheric input of tritium since the mid-1960s, in general, is depicted in the spatial and temporal distribution of this tracer in the sections occupied. The tritium values show a good correlation to the CFC data and salinity, giving further evidence for the assumption that at least part of the observed variability is caused by mixing processes and changeable spreading of SUNADW.

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