Nitrous oxide and methane in the Atlantic Ocean between 50°N and 52°S: Latitudinal distribution and sea-to-air flux

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\begin{abstract}
We discuss nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) distributions in 49 vertical profiles covering the upper ~300 m of the water column along two ~13,500 km transects between ~50°N and ~52°S during the Atlantic Meridional Transect (AMT) programme (AMT cruises 12 and 13). Vertical N\textsubscript{2}O profiles were amenable to analysis on the basis of common features coincident with Longhurst provinces. In contrast, CH\textsubscript{4} showed no such pattern. The most striking feature of the latitudinal depth distributions was a well-defined “plume” of exceptionally high N\textsubscript{2}O concentrations coincident with very low levels of CH\textsubscript{4} located between ~23.5°N and ~23.5°S; this feature reflects the upwelling of deep waters containing N\textsubscript{2}O derived from nitrification, as identified by an analysis of N\textsubscript{2}O, apparent oxygen utilization (AOU) and NO\textsubscript{3} concentrations. In the Atlantic, N\textsubscript{2}O and CH\textsubscript{4} would have made a somewhat larger contribution to marine-derived atmospheric CH\textsubscript{4} than previously thought.

\end{abstract}

\section{1. Introduction}

Nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) both strongly influence Earth’s climate and atmospheric chemistry. They have relatively long atmospheric lifetimes and are infrared-active; together they account for ~20% of enhanced greenhouse forcing (IPCC, 2001). N\textsubscript{2}O participates in stratospheric O\textsubscript{3} regulation via NO\textsubscript{x} generation (Nevison and Holland, 1997) and CH\textsubscript{4} is involved in the formation of stratospheric water and in photochemical reactions that regulate tropospheric OH and O\textsubscript{3} (Crutzen, 1991). The atmospheric inventories of N\textsubscript{2}O and CH\textsubscript{4} are currently increasing, but at variable rates that are not well understood (DLugokencky et al., 1998, 2001; Khalil and Rasmussen, 1992; Prinn et al., 1990) hence their global source–sink functions are the subject of intense scrutiny (IPCC, 2001).

The marine sources of N\textsubscript{2}O and CH\textsubscript{4} are not well constrained. For CH\textsubscript{4}, one estimate sets this at 0.4 Tg CH\textsubscript{4} yr\textsuperscript{-1} (Bates et al., 1996) although most recent syntheses converge at around 11–18 Tg CH\textsubscript{4} yr\textsuperscript{-1}, about 2–3% of the global total (e.g. Bange et al., 1994; Lelieveld et al., 1998). Importantly, Bange et al. (1994) attribute ~75% of the latter estimate to estuarine and shelf seas. For N\textsubscript{2}O the uncertainty is no better; recent estimates suggest 6.28 Tg N\textsubscript{2}O yr\textsuperscript{-1} (range 1.99–10.68 Tg N\textsubscript{2}O yr\textsuperscript{-1}) (Nevison et al., 1995) and 4.71 Tg N\textsubscript{2}O yr\textsuperscript{-1} (range 1.57–7.85 Tg N\textsubscript{2}O yr\textsuperscript{-1}) (Mosier et al., 1998; Krooe et al., 1999) against a global source total ~25.8 Tg N\textsubscript{2}O yr\textsuperscript{-1} (IPCC, 2001). However, Bange (2006) suggests that these estimates of the oceanic N\textsubscript{2}O source strength are too low and that ~11 ± 6.28 Tg N\textsubscript{2}O yr\textsuperscript{-1} is more realistic. As for CH\textsubscript{4}, this higher estimate is believed to be dominated by coastal N\textsubscript{2}O sources (Bange et al., 1996; Bange, 2006). The uncertainty surrounding the marine sources of N\textsubscript{2}O and CH\textsubscript{4} reflects a paucity of targeted sampling in key marine provinces. Perhaps somewhat surprisingly, much of the South Atlantic Ocean remains poorly sampled with respect to subsurface N\textsubscript{2}O and CH\textsubscript{4}. Weiss et al. (1992) measured N\textsubscript{2}O in 6 N. Atlantic, 4 Tropical Atlantic, and 9 S. Atlantic surface transects between 1978 and 1990. N\textsubscript{2}O was highest in the tropical and Benguela upwellings (~120–130% saturation), whereas the N and S Atlantic ranged from mildly undersaturated to moderately supersaturated in N\textsubscript{2}O (Weiss et al., 1992). Butler et al. (1995) collected 40 Atlantic N\textsubscript{2}O depth profiles,
35 of which were in the South Atlantic between the equator and ~50°S, along a south-westerly transect between ~25° and ~30°W. Concentrations throughout the top 100 m were in the range ~5–10 nmol N₂O L⁻¹ (Butler et al., 1995; Nevison et al., 2003). For CH₄, Conrad and Seiler (1988) profiled from 35°S to 50°N; CH₄ saturations were in the range 101–158%. Rhee (2000) measured surface underway N₂O and CH₄ between the UK and Uruguay; both were mildly saturated with the exception of higher N₂O and CH₄ in the W. African Upwelling between the equator and 20°N. To our knowledge Rhee (2000) is the only study to date to include CH₄ measurements in the South Atlantic Gyre (SAG); however, these data are restricted to the upper 10 m (Rhee, 2000). Most other recent work has tended to focus on the tropical and sub-tropical North Atlantic (e.g. Oudot et al., 1990, 2002; Seifert et al., 1999; Morell et al., 2001; Walter et al., 2004, 2006). With the exception of surface data (e.g. Weiss et al., 1992; Rhee, 2000) N₂O and CH₄ distributions in the Atlantic Ocean remain incompletely described. This is important given that the Atlantic incorporates a range of oceanographic regimes, including both coastal and equatorial upwelling, oligotrophic gyres, high-nutrient low-chlorophyll (HNLC) waters, and high productivity waters with strong seasonal effects such as in the North Atlantic Spring bloom.

The Atlantic Meridional Transect Programme (AMT), which exploits the annual transit of RRS James Clark Ross between the UK and Antarctica in September–October and the return leg (Antarctica–UK) in April–May, offered a unique opportunity to investigate the distributions of N₂O and CH₄ in a range of Atlantic waters down to ~300 m, including temperate shelf seas, upwelling regions, and oligotrophic mid-ocean gyres (Hooker et al., 2000). The rationale and methodology of the AMT programme (2002–2006) and an overview of hydrographic conditions along the cruise tracks are presented elsewhere (Robinson et al., 2006). Here we report the distributions of N₂O and CH₄ in 49 vertical profiles covering the upper ~300 m of the water column along two ~13,500 km transects between ~50°N and ~52°S (AMT cruises 12 and 13). Importantly our measurements include novel data from the SAG. Our complete data set provides a basis for deriving gyre scale sea-to-air fluxes of N₂O and CH₄ and hence for re-evaluating the contribution from the Atlantic Ocean to the atmospheric budgets of these climatically important gases.

2. Methods

2.1. Cruise tracks

The AMT12 (12 April–17 May 2003) and AMT13 (10 September–14 October 2003) cruise tracks (Fig. 1) were designed to meet the overarching AMT objectives of evaluating intra- and inter-annual variability in biogeochemical processes in the mid-North and South Atlantic Gyres, to compare ecosystem functioning between the gyres, and to sample climatically active trace gases (Robinson et al., 2006). The two cruises together crossed a total of 7 biogeographical provinces defined by Longhurst (1998) on the basis of satellite (CZCS) imagery supported by near-surface hydrography, light and nutrient distributions, plankton ecology, and other measurements (Fig. 1, Table 1): South Subtropical Convergence (SSTC, 45°–42°S); South Atlantic Gyral (SATL, 42°–6°S); Western Tropical Atlantic (WTSA, 6°S–11°N); North Atlantic Tropical Gyral (NATR, 11°–26°N); North Atlantic Subtropical Gyral—East (NAST(E), 26°–44°N); North Atlantic Drift (NADR, 44°–58°N); Eastern (Canary) Coastal (CNRY, 13°–26°S). Common to both cruises was sampling between the equator and 30°S primarily along 25°W in the SAG, which affords some degree of seasonal data comparison for this region. In contrast, the northern hemisphere cruise tracks were very different. While AMT12 sampled along a SW–SE zigzag into the North Atlantic Gyre (NAG) with the most westerly station at 35.83°W, AMT13 targeted coastal upwelling off the Moroccan and Mauritanian shelf (Fig. 1). General hydrographic aspects and some relevant biogeochemical features along the cruise tracks have already been reported (Robinson et al., 2006).

2.2. Sampling

Water samples for dissolved N₂O, CH₄, O₂, and NO₃⁻ were collected predawn (0200–0400 h, local time) with a standard CTD (Sea-Bird 911 plus) rosette (Ocean Test Equipment: 24 × 20 L Niskins). Sampling dates, locations and depths are summarized in Table 1; sampling always routinely included the chlorophyll maximum and the following percentage irradiances: 97, 55, 33, 14, 1.0 and 0.1 (Robinson et al., 2006).

Sub-samples for dissolved gas analyses were always the first to be drawn from the CTD (typically 9–10 per cast); collection was via silicon tubing. In each case care was taken to avoid air entrainment and the sample was allowed to overflow by three volumes. Single samples for N₂O and CH₄ analyses were collected in 1 L volumetric flasks and immediately poisoned with 200 µL of 0.25M aqueous HgCl₂. Selected dissolved O₂ samples were
collected in gravimetrically calibrated 120 mL borosilicate glass bottles and immediately fixed with MnSO4 and NaOH+Nal; these samples were used to calibrate an O2 sensor (Sea-Bird Electronics, SBE 43) mounted on the CTD frame and used for routine dissolved O2 measurements. Sub-samples for NO3 analysis were collected in acid-cleaned high-density polyethylene screw cap bottles following flushing with sample. Sample storage for N2O and CH4, and NO3 was in the dark in a 5 °C cold room. Pre-analysis storage never exceeded 8 h for N2O and CH4, and 4 h for NO3. Samples for O2 analysis were stored under water and analysed within 4-8 h of collection.

2.3. Analysis

Dissolved N2O and CH4 were analysed by single-phase equilibration gas chromatography, with electron capture detection (ECD) for N2O and flame ionization detection (FID) for CH4. Routine calibration was with a mixed secondary standard (361 ppbv N2O, 2000 ppbv CH4) prepared by pressure dilution (Upstill-Goddard et al., 1990, 1996) and independently calibrated against two mixed primary standards with certified accuracies of ±1 % (10 ppmv N2O, 5 ppmv CH4 and 20 ppmv N2O, 8 ppmv CH4; BOC Special Gases, UK). Due to difficulties in obtaining mixed primary standards with suitably low N2O mixing ratios we also used a 1.05 ppmv primary N2O standard (certified accuracy 7 %) and independently calibrated our system against two mixed primary standards with certified accuracies of ±2 % (http://www.nplindia.org/). Method analytical precision (1σ), determined from repeat analyses (n = 10) of the mixed secondary standard, was ±5 %.

Equilibrated mixing ratios corrected for phase partitioning during analysis (Upstill-Goddard et al., 1996) were converted to percent saturations using atmospheric N2O and CH4 mixing ratios obtained from the National Oceanic and Atmospheric Administration/Earth System Research Laboratory.
Table 2
Mixing ratios of N\textsubscript{2}O and CH\textsubscript{4} at atmospheric monitoring stations during AMT12 and AMT13.

<table>
<thead>
<tr>
<th>Station</th>
<th>Latitude</th>
<th>Longitude</th>
<th>Parameter</th>
<th>Atmospheric mixing ratio (ppbv)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>CH\textsubscript{4} (June)</td>
<td>CH\textsubscript{4} (September)</td>
</tr>
<tr>
<td>Northern stations</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Alert</td>
<td>82.27</td>
<td>-62.31</td>
<td>CH\textsubscript{4} N\textsubscript{2}O</td>
<td>1821</td>
</tr>
<tr>
<td>Summit</td>
<td>72.35</td>
<td>-38.29</td>
<td>CH\textsubscript{4}</td>
<td>1815</td>
</tr>
<tr>
<td>Heimaey</td>
<td>63.20</td>
<td>-20.70</td>
<td>CH\textsubscript{4} N\textsubscript{2}O</td>
<td>1826</td>
</tr>
<tr>
<td>Mace Head</td>
<td>53.20</td>
<td>-09.54</td>
<td>CH\textsubscript{4} N\textsubscript{2}O</td>
<td>1820</td>
</tr>
<tr>
<td>Harvard Forest</td>
<td>42.54</td>
<td>-72.18</td>
<td>N\textsubscript{2}O</td>
<td>-</td>
</tr>
<tr>
<td>Terceia Island</td>
<td>38.46</td>
<td>-27.23</td>
<td>CH\textsubscript{4}</td>
<td>1800</td>
</tr>
<tr>
<td>Tudor Hill</td>
<td>32.16</td>
<td>-64.53</td>
<td>CH\textsubscript{4}</td>
<td>1770</td>
</tr>
<tr>
<td>Tenerife</td>
<td>28.18</td>
<td>-16.29</td>
<td>CH\textsubscript{4}</td>
<td>1777</td>
</tr>
<tr>
<td>Ragged Point</td>
<td>13.10</td>
<td>-59.26</td>
<td>CH\textsubscript{4}, N\textsubscript{2}O</td>
<td>1792</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td></td>
<td>1803 ± 21</td>
</tr>
<tr>
<td>Southern stations</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ascension Island</td>
<td>-07.55</td>
<td>-14.25</td>
<td>CH\textsubscript{4}</td>
<td>1723</td>
</tr>
<tr>
<td>Gobabeb</td>
<td>-23.35</td>
<td>15.02</td>
<td>CH\textsubscript{4}</td>
<td>1679</td>
</tr>
<tr>
<td>Cape Point</td>
<td>-34.21</td>
<td>18.29</td>
<td>CH\textsubscript{4}, N\textsubscript{2}O</td>
<td>1708</td>
</tr>
<tr>
<td>Tierra Del Fuego</td>
<td>-54.52</td>
<td>-68.29</td>
<td>CH\textsubscript{4}</td>
<td>1708</td>
</tr>
<tr>
<td>Palmer Station</td>
<td>-64.55</td>
<td>-64.00</td>
<td>CH\textsubscript{4}</td>
<td>1703</td>
</tr>
<tr>
<td>Halley Bay</td>
<td>-75.35</td>
<td>-26.30</td>
<td>CH\textsubscript{4}</td>
<td>1700</td>
</tr>
<tr>
<td>South Pole</td>
<td>-89.59</td>
<td>-24.48</td>
<td>CH\textsubscript{4}, N\textsubscript{2}O</td>
<td>1702</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td></td>
<td>1703 ± 13</td>
</tr>
</tbody>
</table>

All data are from the NOAA/ESRL with the exception of Cape Point where mixing ratios were obtained from SAWS.

Table 3
Mean mixed layer concentration of N\textsubscript{2}O and CH\textsubscript{4} by Longhurst province for AMT12 and AMT13.

<table>
<thead>
<tr>
<th>Province</th>
<th>AMT12</th>
<th></th>
<th>AMT13</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean N\textsubscript{2}O % Saturation nmol\textsubscript{L}\textsuperscript{-1}</td>
<td>Mean CH\textsubscript{4} % Saturation nmol\textsubscript{L}\textsuperscript{-1}</td>
<td>Mean N\textsubscript{2}O % Saturation nmol\textsubscript{L}\textsuperscript{-1}</td>
<td>Mean CH\textsubscript{4} % Saturation nmol\textsubscript{L}\textsuperscript{-1}</td>
</tr>
<tr>
<td>SSTC</td>
<td>107 ± 6</td>
<td>9.7 ± 0.5</td>
<td>157 ± 40</td>
<td>3.6 ± 0.9</td>
</tr>
<tr>
<td>SATI</td>
<td>104 ± 11</td>
<td>6.8 ± 1</td>
<td>162 ± 31</td>
<td>3 ± 0.5</td>
</tr>
<tr>
<td>WTRA</td>
<td>109 ± 21</td>
<td>5.9 ± 1.3</td>
<td>170 ± 43</td>
<td>3.5 ± 0.8</td>
</tr>
<tr>
<td>NATR</td>
<td>104 ± 3</td>
<td>6.1 ± 0.5</td>
<td>194 ± 43</td>
<td>3.7 ± 1</td>
</tr>
<tr>
<td>NAST (E)</td>
<td>105 ± 8</td>
<td>7.1 ± 0.7</td>
<td>185 ± 50</td>
<td>3.7 ± 0.9</td>
</tr>
<tr>
<td>NADR</td>
<td>98 ± 10</td>
<td>7.6 ± 0.9</td>
<td>147 ± 17</td>
<td>3.3 ± 0.4</td>
</tr>
<tr>
<td>CNRY</td>
<td>n.s.</td>
<td>n.s.</td>
<td>n.s.</td>
<td>n.s.</td>
</tr>
</tbody>
</table>

n.s. refers to not sampled.

Mixed layer depths required for estimating sea-to-air fluxes of N\textsubscript{2}O and CH\textsubscript{4} were estimated from profiles of \sigma_\theta and temperature (Hooker et al., 2000). We thus defined the base of the mixed layer as coinciding with the start of the thermocline as indicated by three out of four successive gradients in \sigma_\theta and/or temperature exceeding 0.035 m\textsuperscript{-1} and/or 0.1 °C m\textsuperscript{-1}, respectively. For situations where the thermocline was too weak to be identified with this approach we assumed its top to be represented by a change of 0.1 in \sigma_\theta or 0.5 °C in temperature relative to the corresponding surface value (after Hooker et al., 2000) (Table 1).

3. Results and discussion

3.1. N\textsubscript{2}O and CH\textsubscript{4} in the upper 300 m

Table 3 lists mean mixed layer concentrations and percent saturations of N\textsubscript{2}O and CH\textsubscript{4} based on the mixed layer depth estimates listed in Table 1.
Groups of individual N$_2$O profiles show common features that allow them to be conveniently grouped into several “types” coincident with Longhurst (1998) provinces (Fig. 1, Table 1). In contrast, the CH$_4$ data are not amenable to such analysis; the profiles show far greater similarity in shape and span smaller concentration ranges.

3.1.1. N$_2$O and Longhurst provinces

Vertical N$_2$O profiles are shown in Fig. 2. SSTC and NADR were only encountered during AMT12 and CNRY was only encountered during AMT13. All other provinces were sampled during both cruises (Table 1). The N$_2$O profiles for SSTC and NADR (surface high-latitude south and north Atlantic, respectively) were similar in general shape, being characterized by small increases in concentration with depth between the surface and the base of the mixed layer; overall increases were $\approx$0.5–2 nmol L$^{-1}$ at SSTC, and $\approx$0.3–0.8 nmol L$^{-1}$ at NADR (mixed layer means given in Table 3). Below the mixed layer in both cases N$_2$O was essentially constant down to $\approx$300 m (SSTC: $\approx$13 ± 1.3 nmol L$^{-1}$, 124 ± 7% saturation; NADR: $\approx$8.9 ± 1.3 nmol L$^{-1}$, 106 ± 13% saturation). For comparison the ranges of N$_2$O concentrations reported by Walter et al. (2006) for the cold temperate N. Atlantic between 40 and 50 N along 10–50 W (mixed layer, $8.6 \pm 1.4$ nmol L$^{-1}$; sub-thermocline: $11.3 \pm 1.5$ nmol L$^{-1}$) are not significantly different from our data for NADR during AMT12.

The highest mean mixed layer N$_2$O encountered during AMT was in the CNRY province (Table 3); here the maximum surface N$_2$O concentration was 9.7 nmol L$^{-1}$ ($\approx$150% saturation; 2 m depth, station AMT13_28). Below the CNRY mixed layer N$_2$O concentrations increased rapidly during AMT13, reaching an approximately constant $30 \pm 5$ nmol L$^{-1}$ ($\approx$370 ± 40% saturation) between 100 and 300 m. Corresponding NO$_3^-$ concentrations generally exceeded $35 \mu$mol L$^{-1}$ (mean $37 \pm 2.9 \mu$mol L$^{-1}$) and O$_2$ decreased significantly below the mixed layer to a mean $\approx$60 $\mu$mol L$^{-1}$ (range 39–78 $\mu$mol L$^{-1}$) below $\approx$70 m depth (data not shown). To our knowledge these vertical profile data are the first to be reported for the Mauritanian upwelling region within the CNRY province. N$_2$O in the CNRY mixed layer (Table 3) was significantly higher than previously reported values ($\approx$113–118% around the southern boundary of CNRY adjacent to the Guinea Shelf between 7 and 12 N (Oudot et al., 2002; Walter et al., 2006). In contrast, Weiss et al. (1992) found 130–140% saturation and Rhee (2000) found 102–115% saturation, in surface waters between 10 and 20 N close to 20 W.

![Fig. 2. Vertical profiles of N$_2$O concentration during AMT12 (clear triangles) and AMT13 (filled triangles) grouped in relation to Longhurst (1998) province.](image-url)
The SATL is the largest of the provinces encountered during AMT, spanning approximately 36° of latitude. Mean mixed layer \( N_2O \) (Table 3) was very close to the ranges found by Weiss et al. (1992), Butler et al. (1995), and Rhee (2000) (~98–108% saturation) during surface surveys in this region. Vertical \( N_2O \) profiles south of ~26°S were similar during both cruises, exhibiting a rather mild linear increase in concentration between the surface and 300 m of 0.7–6.8 and 0.9–6.4 nmol L\(^{-1}\) for the austral autumn and austral spring, respectively (Fig. 2). These stations showed a weak trend of increasing \( N_2O \) at 300 m depth toward lower latitudes. North of 26°S within the SATL \( N_2O \) in the upper 150 m was similar to surface levels further south. However, below this depth there was a progressive increase in \( N_2O \) concentration toward the northern SATL boundary during both seasons. During the austral autumn \( N_2O \) in the deepest sample from AMT12_23 (301 m, Table 1) was 12.3 nmol L\(^{-1}\) (149% saturation). Further north (AMT12_29) this reached 22.4 nmol L\(^{-1}\) (272% saturation) at 302 m. Similarly, during the austral spring \( N_2O \) in the deepest sample from AMT13_53 (304 m, Table 1) was 12.6 nmol L\(^{-1}\) (148% saturation) and this increased to 28.8 nmol L\(^{-1}\) (295% saturation) at AMT13_44 (302 m) further north. The northward increase in \( N_2O \) below ~150 m continued into WTRA; \( N_2O \) reached 31.6 nmol L\(^{-1}\) (340% saturation) at ~302 m at AMT12_40 and 33 nmol L\(^{-1}\) (360% saturation) at ~300 m at AMT13_34. Across the WTRA the concentrations of \( N_2O \) were consistently high; ~22–33 nmol L\(^{-1}\) (265–360% saturation) during AMT12 and ~21–33 nmol L\(^{-1}\) (240–360% saturation) during AMT13. Walter et al. (2006) reported their highest \( N_2O \) concentration (37.3 nmol L\(^{-1}\) at 400 m depth) adjacent to the Guinea Dome, located approximately equidistant between AMT12_40 and AMT13_34 (Fig. 1) and Oudot et al. (2002) found ~60 nmol L\(^{-1}\) at ~400 m in the eastern WTRA. In addition Walter et al. (2006) found shallower \( N_2O \) maxima (240–280 m depth) further south towards the equator corresponding to \( \sigma_0 = 26.6–27.0 \); this range in \( \sigma_0 \) is identical to that for the \( N_2O \) maxima observed in the tropical Atlantic during AMT12 and AMT13. The trend in northerly increasing \( N_2O \) at depth also persisted into NATR; 36 nmol N\(_2\)O L\(^{-1}\) (397% saturation) was recorded at ~302 m at station AMT12_42. However, further north \( N_2O \) at depth again began to decrease; the corresponding 300 m concentration at AMT12_51 being only 8.7 nmol L\(^{-1}\) (119% saturation). Profiles from NAST(E) were similar during both cruises, being characterized by a mild, approximately linear increase in \( N_2O \) with depth from the base of the mixed layer to the deepest sample of ~1.9–3.7 and 3.8–6.7 nmol L\(^{-1}\) for
AMT12 and AMT13, respectively. The differences between the magnitude of increase in N$_2$O with depth between AMT12 and AMT13 is most likely a result of spatial variability as the two cruise tracks were significantly different within the NAST(E). In addition some undersaturations of a few percent were observed in some near-surface samples; however, most samples showed supersaturation (Table 3). For comparison Weiss et al. (1992) and Rhee (2000) found saturations close to atmospheric equilibrium with NAST(E).

Generally our mean percent N$_2$O saturations calculated by Longhurst province for the tropical Atlantic are more variable than those previously reported. The largest variations occur in the WTRA; $109 \pm 21\%$. However, it must be noted that despite the larger variations, the mean for this region is close to mean surface saturations previously reported; $\sim 104–108\%$ saturation (Oudot et al., 2002; Walter et al., 2006).

3.1.2. Vertical distribution of CH$_4$

In contrast to the situation for N$_2$O, the main features of the observed CH$_4$ distributions do not clearly correspond to Longhurst provinces. However, in order to facilitate comparisons mean mixed layer concentrations and percent saturations of CH$_4$ are nevertheless summarized on this basis (Table 3).

The vertical CH$_4$ profiles are shown in Fig. 3. Within the SAG ($\sim 26$ to $6^\circ$S) mixed layer CH$_4$ was rather variable, both within and between the individual cruises; $3.7 \pm 1.7$ nmol CH$_4$L$^{-1}$ ($201 \pm 98\%$ saturation) for AMT12 and $2.8 \pm 0.3$ nmol CH$_4$L$^{-1}$ ($155 \pm 17\%$ saturation) for AMT13 displaying considerable seasonal variation and highlights that the SAG is a significant source of CH$_4$ to the atmosphere. This is also a feature when the mean mixed layer CH$_4$ concentration is calculated for the SATL province as a whole (Table 3). Previous underway analyses of near-surface SAG waters (sample inlet at 6 m depth) during AMT7 (September–October 1998) ranged from a few % undersaturation to approximately 108% supersaturation (Rhee, 2000). Notwithstanding the fact that the AMT12, AMT13 and AMT7 cruise tracks were somewhat different, AMT7 was more westerly than either AMT12 or AMT13, it is perhaps not surprising that the CH$_4$ saturations found on AMT7 (Rhee, 2000) were much lower than those from either AMT12 or AMT13. Underway equilibrated CH$_4$ concentrations are frequently significantly lower than those obtained from CTD samples collected simultaneously from the same depth (Bange, pers. comm.). The discrepancy presumably reflects solubility and response time considerations for CH$_4$, and appears not to be a problem for N$_2$O (Bange, pers. comm.).

Below the mixed layer within the SATL province north of $26^\circ$S CH$_4$ saturation decreased toward lower latitudes on both cruises, the opposite of the situation for N$_2$O. Despite the comparative mixed layer variabilities mean CH$_4$ at 300 m depth between $\sim 26$ and $6^\circ$S during AMT12 ($2.7 \pm 0.4$ nmol L$^{-1}$, range $2.4–3.3$ nmol L$^{-1}$; $119 \pm 20\%$ saturation) and AMT13 ($2.5 \pm 0.5$ nmol L$^{-1}$, range $1.9–3$ nmol L$^{-1}$; $110 \pm 25\%$ saturation) was not significantly different; in each case the lower ends of the above ranges represent lower latitude stations. The mean CH$_4$ concentrations at $\sim 300$ m between $6^\circ$S and $23.5^\circ$N were in contrast, significantly different: $2.8 \pm 0.4$ nmol L$^{-1}$ (range $2.3–3.4$ nmol L$^{-1}$; $121 \pm 19\%$ saturation) during AMT12 (AMT12_33 to AMT12_51) and $1.7 \pm 1.3$ nmol L$^{-1}$ during AMT13 (AMT13_23 to AMT13_41) and $1.6 \pm 0.7$ nmol L$^{-1}$ during AMT12_52.
(range 1.3–1.9 nmol L\(^{-1}\), 71 ± 14% saturation) during AMT13 (AMT13_24 to AMT13_40). Oudot et al. (2002) similarly, reported CH\(_4\) saturations ∼50–90% for a latitudinal transect at 43.0°S. North of 25°N during AMT12 mean mixed layer CH\(_4\) (3.5 ± 0.7 nmol L\(^{-1}\), range 2.6–4.8 nmol L\(^{-1}\); 164 ± 42% saturation) was considerably higher than during AMT13 (2.3 ± 0.2 nmol L\(^{-1}\), range 1.9–2.6 nmol L\(^{-1}\); 120 ± 12% saturation). Similarly, below the mixed layer the boreal spring mean was overall higher than during autumn: AMT12; 3 ± 0.4 nmol L\(^{-1}\) (range 2.4–4 nmol L\(^{-1}\); 139 ± 18% saturation), AMT13; 2.3 ± 0.2 nmol L\(^{-1}\) (range 1.8–2.9 nmol L\(^{-1}\); 107 ± 13% saturation). The higher mixed layer mean during AMT12 reflects several near surface samples with CH\(_4\) saturations in excess of 200% saturation at stations north of the tropics. Generally previous CH\(_4\) data for the surface waters of the subtropical North Atlantic agree more closely with the data from AMT13 than those from AMT12 with saturation values in the range 96–150% (e.g. Scranton and Brewer, 1977; Conrad and Seiler, 1988; Seifert et al., 1999; Rhee, 2000) with values generally increasing on the approach towards continental shelf regions.

Tropical Atlantic mixed layer CH\(_4\) was considerably higher in the central region (AMT12: 3.7 ± 0.8 nmol L\(^{-1}\), 183 ± 43% saturation) than further east (AMT13: 2.3 ± 0.5 nmol L\(^{-1}\), 128 ± 26% saturation). The mean CH\(_4\) saturations in eastern tropical Atlantic mixed layer during AMT13 agree closely with previously reported values for tropical Atlantic open ocean studies; ∼100–140% (Conrad and Seiler, 1988; Rhee, 2000; Oudot et al., 2002). Previously Conrad and Seiler (1988) reported CH\(_4\) concentrations ∼50–93 nL L\(^{-1}\) for the upper 20 m of the tropical Atlantic between 3°N and 2°S along the 22°W. These data translate to approximate CH\(_4\) saturations ∼130–250% in the uppermost 4 m and ∼140–220% at 20 m depth; Moreover, Oudot et al. (2002) report ∼220% CH\(_4\) saturations measured in surface waters around 4°W, 4.3°S. The mean mixed layer CH\(_4\) saturations reported here for the Central Tropical Atlantic (183 ± 43%) are among the highest values thus far reported. Our data confirm the Central Atlantic Ocean as a potentially significant source of atmospheric CH\(_4\).

3.1.3. Latitudinal-depth contrast in N\(_2\)O and CH\(_4\)

The latitudinal-depth distributions of N\(_2\)O and CH\(_4\) saturation were strikingly different from each other and broadly persistent across the two seasons (Figs. 4 and 5). The most striking feature of both the AMT12 and AMT13 N\(_2\)O distributions (Fig. 4) may best be described as a very well-defined “plume” of exceptionally high supersaturations ∼140–340% N\(_2\)O located between about 23.5°S and 23.5°N, extending from 20 to 50 m below the surface to below the deepest waters sampled, and broadly coincident with the lowest observed values of CH\(_4\) saturation (Fig. 5). The same feature is readily discernable in corresponding salinity and temperature data (Robinson et al., 2006), and can also be clearly seen in the BLASTII N\(_2\)O data set presented in Nevison et al. (2003). Outside the plume N\(_2\)O was everywhere at, or very close to, atmospheric equilibrium. Outside the regions of comparatively low CH\(_4\) saturation associated with the N\(_2\)O plumes, CH\(_4\) was essentially everywhere quite strongly supersaturated. To what extent the plume suppresses the CH\(_4\) signal in the near-surface
waters above it remains unclear, however, higher near-surface CH₄ encountered outside the plume implies some degree of mixing with waters above 20–50 m. Indeed a major CH₄ feature during AMT12 was a surface region ~25 m deep between ~25 S and 35 N (i.e. outside the “plume”) in which CH₄ saturation exceeded 200% and with two localized maxima around 15–20 S and 20–30 N, each with >250% CH₄ saturation. This feature was much weaker during AMT13; only a sub-surface region of exceptionally high CH₄ saturation was discernable, centred at ~25 m depth around 16 N. Oudot et al. (2002) observed CH₄ undersaturation and N₂O supersaturation coincident at about 400 m depth in the eastern and western basins of the tropical Atlantic around 5 S, accompanying a well-documented O₂ minimum (Reid, 1989). It seems likely that this is part of the feature identified in Figs. 4 and 5.

The contrasting distributions of N₂O and CH₄ in the upper 300 m of the Atlantic water column reflect the relative sources and sinks of these two gases in oceanic waters, coupled with upward transport.

### 3.2. Source of the upwelled N₂O

Further insight into the production mechanisms of N₂O in the subsurface ocean may be derived by examining the relationships between ∆N₂O and AOU (Yoshinari, 1976; Suntharalingam and Sarmiento, 2000; Nevison et al., 2003) and between ∆N₂O and NO₃ (Cohen and Gordon, 1979; Walter et al., 2006), where ∆N₂O is the difference between N₂O measured in situ and its theoretical concentration equivalent to 100% saturation. It is generally agreed that a strong positive correlation between these variables is evidence for nitrification as the main source of N₂O (e.g. Yoshinari, 1976; Elkins et al., 1978; Cohen and Gordon, 1979; De Wilde and Helder, 1997; Patra et al., 1999; Oudot et al., 2002; Nevison et al., 2003; Walter et al., 2006). The relationships between ∆N₂O and AOU vary considerably between different ocean regions and as a function of depth (e.g. Yoshinari, 1976; Elkins et al., 1978; Cohen and Gordon, 1979; Butler et al., 1989; Law and Owens, 1990; Oudot et al., 2002; Nevison et al., 2003; Walter et al., 2006) and may reflect the sensitivity of nitrifiers to varying ambient O₂ (e.g. Goreau et al., 1980; Poth and Focht, 1985).

For example De Wilde and Helder (1997) found a significant increase in N₂O at O₂ concentrations below 15–20 µmol L⁻¹ in the Somali Basin.

We investigated the mechanism of N₂O production within the tropical Atlantic by examining the relationships between ∆N₂O and AOU and between ∆N₂O and NO₃ for sub-mixed layer waters during AMT12 and AMT13 (Fig. 6). For the tropical Atlantic, i.e. the region between 23.5 S and 23.5 N, strong correlations between ∆N₂O and AOU (AMT12: \( r^2 = 0.82, n = 36, p < 0.001 \); AMT13: \( r^2 = 0.85, n = 29, p < 0.001 \)) and between ∆N₂O and NO₃ (AMT12: \( r^2 = 0.86, n = 36, p < 0.001 \); AMT13: \( r^2 = 0.68, n = 29, p < 0.001 \)) suggest a nitrification source for the upwelled N₂O in agreement with previous studies in the tropical Atlantic (e.g. Oudot et al., 2002; Walter et al., 2006). The relationship between ∆N₂O and AOU within the tropical Atlantic is remarkably similar for both its central (AMT12) (1) and eastern sectors (AMT13) (2):

\[
\Delta N_2O = 1.135 + 0.121AOU
\]  

\[
\Delta N_2O = 1.047 + 0.106AOU
\]  

This similarity implies a common N₂O source across much of the tropical Atlantic Ocean. In contrast, Walter et al. (2006) report a somewhat different relationship for tropical Atlantic water above 500 m (\( > \sigma_T = 27.1 \)):

\[
\Delta N_2O = 2.4381 + 0.0785AOU
\]  

The discrepancy between the AMT ∆N₂O:AOU relationships and those of Walter et al. (2006) is likely related to the different depths over which these relationships were derived.
During AMT highest N$_2$O in the tropical Atlantic was associated with $\sigma_o$ values consistent with South Atlantic Central Water (SACW) (e.g. Emery and Meincke, 1986; Poole and Tomczak, 1999) (Fig. 7) and the majority of highest N$_2$O concentrations are associated with $\sigma_o = 26.6–27.4$, suggesting that the N$_2$O may be produced predominantly in this water mass. Similarly, Walter et al. (2006) found highest N$_2$O concentrations in the eastern basin within SACW: 37.3 nmol L$^{-1}$, which prompted them to suggest nitrification as the primary N$_2$O production mechanism in the tropical Atlantic. Due to the similarity of our tropical Atlantic AN$_2$O:AOU relationships for AMT12 (central basin) and AMT13 (eastern basin) we propose that the elevated N$_2$O in these regions (Fig. 7) is primarily a consequence of nitrification within the SACW.

3.3. Source of CH$_4$ in the mixed layer

Within the well-oxygenated ocean surface mixed layer, CH$_4$ supersaturations are a common feature (e.g. Lamontagne et al., 1973; Scranton and Brewer, 1977; Scranton and Farrington, 1977; Owens et al., 1991; Patra et al., 1998) and have been ascribed to methanogenesis by O$_2$-tolerant methanogens inside anoxic “microniches” maintained by bacterial activity (Oremland, 1979). This view is supported by the isolation of an “oxic” methanogen from coastal waters (Cynar and Yayanos, 1991) and the identification of methanogens in marine zooplankton guts and particles (Martý et al., 1997). Although some studies have reported close associations between CH$_4$ and indicators of primary productivity such as chlorophyll-a (e.g. Conrad and Seiler, 1988; Oudot et al., 2002), other studies have found either only weak correlations or no correlation at all (e.g. Upstill-Goddard et al., 1999; Holmes et al., 2000), and we found no such correlation during either AMT12 or AMT13, which qualitatively tends to favour the “anoxic microniche” hypothesis. Although total suspended particle concentrations with which to directly test this were unavailable during AMT, particulate organic carbon and nitrogen (POC and PON) were measured. However, we found no significant relationships between CH$_4$ and either POC or PON (Forster, 2006). Similarly, although Traganza et al. (1979) report a correlation between CH$_4$ and zooplankton ATP, during AMT neither zooplankton numbers or biomass (San Martin et al., 2006) showed any clear correlation with CH$_4$ (Forster, 2006).

The regions of low CH$_4$ within the plume reflect upwelling of deep water in which CH$_4$ is depleted by bacterial oxidation (e.g. Ward and Kilpatrick, 1993; Ward et al., 1987).

3.4. Sea-to-air emissions fluxes

Sea-to-air emission flux densities ($F$, mol m$^{-2}$ d$^{-1}$) of N$_2$O and CH$_4$ at individual stations were estimated from their measured partial pressures in seawater and air, using

$$F = k_w \Delta P,$$

where $k_w$ is the gas transfer velocity for N$_2$O or CH$_4$ (cm h$^{-1}$), $L$ is the appropriate gas solubility (mol cm$^{-3}$ atm$^{-1}$) at ambient temperature and salinity, and $\Delta P$ is the gas partial pressure difference (natum.) across the sea–air interface. The empirical relationships of Liss and Merlivat (1986) and Wanninkhof (1992) were used to quantify the wind speed dependence of $k_w$; we derived $k_w$ for N$_2$O and CH$_4$ from the corresponding values for CO$_2$ using appropriate Schmidt numbers, $Sc$ (Wanninkhof, 1992). For Liss and Merlivat (1986), $k_w$ for CO$_2$ was multiplied by ($Sc$/600)$^{0.5}$ ($n = -0.67$ for $U_{10}$ = 3.6 m s$^{-1}$, $n = -0.5$ for $U_{10}$ > 3.6 m s$^{-1}$, $U_{10}$ is the 10 m wind speed). For Wanninkhof (1992), $k_w$ was multiplied by ($Sc$/660)$^{0.5}$. Alternative wind speed related parameterizations of $k_w$ (e.g. Erickson, 1993; Nightingale et al., 2000) yield air–sea fluxes that are intermediate between these values; hence using the relationships of Liss and Merlivat (1986) and Wanninkhof (1992) provides upper and lower boundaries to our flux estimates. In situ wind speeds were recorded at 60 s intervals using an anemometer located on the ship’s forecastle ~22 m above sea level. These were subsequently corrected for ship speed and course, and lateral flow distortion (Yelland et al., 1998), and converted to $U_{10}^0$, the equivalent wind speed at 10 m above the sea surface for neutral atmospheric stability, as described in Nightingale et al. (2000). Wind speeds used in estimating individual station fluxes are the means of values recorded within 0.05° of the station to allow for off-station drift during sampling. Values of $\Delta P$ for N$_2$O and CH$_4$ are mean values determined over the mixed layer depths (Table 1).

Fig. 7 shows the individual station N$_2$O and CH$_4$ emission flux densities ($\mu$mol m$^{-2}$ d$^{-1}$) and the corresponding mean mixed layer partial pressures (natum) as functions of latitude. Both gases showed a high degree of inter-station flux variability. The overall ranges of variability were two orders of magnitude for N$_2$O and two to three orders of magnitude for CH$_4$; these ranges primarily reflect large differences in ambient wind speeds between stations rather than variability in mixed layer gas inventories (Fig. 7). Mean flux densities derived from the individual station fluxes for each Longhurst province, and the corresponding province-based emissions fluxes (Tg N$_2$O or CH$_4$ yr$^{-1}$) are summarized in Table 4.

Table 4

<table>
<thead>
<tr>
<th>Province</th>
<th>Surface area (x 10$^6$ km$^2$)</th>
<th>AMT12</th>
<th>AMT13</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>N$_2$O</td>
<td>CH$_4$</td>
<td>N$_2$O</td>
</tr>
<tr>
<td>STTC</td>
<td>4.1</td>
<td>0.79–1.57</td>
<td>3.88–6.81</td>
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<tr>
<td>SATL</td>
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<td>0.97–1.62</td>
<td>3.63–5.89</td>
</tr>
<tr>
<td>WTRA</td>
<td>5.4</td>
<td>1.17–2.13</td>
<td>3.92–6.43</td>
</tr>
<tr>
<td>NATR</td>
<td>8.3</td>
<td>0.60–0.87</td>
<td>6.14–9.69</td>
</tr>
<tr>
<td>NADR</td>
<td>4.4</td>
<td>0.72–1.05</td>
<td>1.91–3.65</td>
</tr>
<tr>
<td>CNRY</td>
<td>3.5</td>
<td>–0.04 to –0.08</td>
<td>0.46–0.90</td>
</tr>
<tr>
<td>Total</td>
<td>44.3</td>
<td>2.73–4.65</td>
<td>2.31–4.04</td>
</tr>
</tbody>
</table>

Upper and lower limits are representative of fluxes and emissions calculated using the models of Liss and Merlivat (1986) and Wanninkhof (1992), respectively. n.s. refers to not sampled.

* Annual flux calculated from samples collected during austral fall.

* Annual flux calculated from samples collected during boreal fall.
Table 4. In each case the lower and higher figures in each range refer to estimates deriving from the $k_w$–wind speed relationships of Liss and Merlivat (1986) and Wanninkhof (1992), respectively. Although, we have previously summarized a subset of these data (Robinson et al., 2006), we below present the first complete summary analysis of the full AMT data set for N$_2$O and CH$_4$ emissions.

Due to differences in the northern hemisphere cruise tracks between the two cruises only the emissions fluxes from SATL are amenable to a seasonal comparison. This province had the highest emissions fluxes for both gases, principally as a consequence of its comparatively large surface area (\~40% of the total, Table 4) rather than as a consequence of high individual flux densities. Interestingly, although SATL N$_2$O emissions were around 4 times higher during AMT12 than during AMT13, there was no comparable seasonality in CH$_4$ emissions; the difference for N$_2$O cannot therefore be explained in terms of the ambient wind speed distributions. Rather, mixed layer N$_2$O showed comparatively high inter-station variability during AMT12 (Fig. 7), causing a bias towards higher emissions values. To the best of our knowledge our N$_2$O and CH$_4$ emissions flux estimates for SATL (Table 4) are the only such estimates reported for this large Atlantic province (Fig. 8).

CNRY had exceptionally high individual station flux densities for N$_2$O but due to its comparatively small surface area its contribution to the total Atlantic N$_2$O flux is rather small (Table 4). Previously Nevison et al. (2004) estimated a total atmospheric N$_2$O source due to coastal upwelling globally \~0.31 \pm 0.2 Tg yr$^{-1}$. 

![Fig. 8. Estimated sea–air flux of N$_2$O for stations sampled during AMT12 (A) and AMT13 (B) and for CH$_4$ during AMT12 (C) and AMT13 (D). Fluxes are estimated using the models of Liss and Merlivat (1986) (empty circles and grey dashed line) and Wanninkhof (1992) (empty triangles and solid grey line). The average partial pressure in the mixed layer at each station is shown by the black filled squares and black solid line).](image)
Adjusting for the larger surface area of CNRY relative to the area of East African upwelling defined by Nevison et al. (2004), our estimate for CNRY is, on a unit area basis: in good agreement with the Nevison et al. (2004) estimate. We can conclude that CNRY represents an approximately average N2O source strength as compared to other regions experiencing significant upwelling. However, for CH4 CNRY appears to be a comparatively weak source. NADR appears to be a similarly weak CH4 source but it is approximately neutral or a very weak sink with respect to N2O (Table 4). For WTRA our emissions estimates show good agreement with those derived both by Oudot et al. (2002) and Walter et al. (2004).

Considering the large sizes of the individual Longhurst provinces (Table 4) and our restriction to an essentially two-dimensional transect on each cruise, our emissions flux estimates deriving from these are, as with other studies, subject to uncertainties arising from the fact that our ability to account for within-province spatial variability, and to a lesser extent temporal variability, was necessarily limited. Clearly such uncertainties are rather difficult to evaluate without additional data. Importantly, however, our mixed layer concentration means for both N2O and CH4 are in fact rather close to values found in previous work at other locations in these areas. This gives us confidence in our resulting sea-to-air emissions flux estimates.

The Longhurst provinces detailed in Table 4 together make up ~42% of the total surface area of the Atlantic Ocean, rendering our data the most extensive contiguous surveys of both N2O and CH4 for this ocean basin. An earlier estimate for the total oceanic CH4 emission of 0.4 Tg yr$^{-1}$ (Bates et al., 1996) was derived from latitudinal transects of the open-ocean Pacific, and so is similar in this respect to the AMT cruise tracks in that it involved minimal sampling of coastal and/or shelf waters. Nevertheless our total estimate for 42% of the Atlantic Ocean exceeds this (Table 4), bringing into question the validity of this previous global estimate for CH4. More recent estimates of total marine source strengths are, for N2O, 4.71–6.28 Tg yr$^{-1}$ (Mosier et al., 1998; Kroeze et al., 1999; Nevison et al., 1995), and for CH4, 11–18 Tg yr$^{-1}$ (Bange et al., 1994; Leieveld et al., 1998). Based on these estimates our data imply that the Atlantic Ocean might account for ~6–15% and 4–13%, respectively, of the total marine sources of atmospheric N2O and CH4. Bange et al. (1994) derived an estimate of the total Atlantic CH4 emission ~0.9–1.4 Tg yr$^{-1}$: this is close to our estimate for 42% of the Atlantic Ocean. Based on our data and making the assumption that our flux estimates are indeed representative of the whole Atlantic, this ocean basin could be a 2-fold larger atmospheric CH4 source than previously thought. However, testing this assumption and further refining our flux estimates will require initiating a more detailed sampling strategy both spatially and seasonally, than was possible during AMT. Notwithstanding the uncertainties in our data set, given that the Atlantic Ocean accounts for around 20% of the global ocean surface, on a unit area basis it appears that the Atlantic may be a slightly weaker source of atmospheric N2O than other open ocean regions but it could make a somewhat larger contribution to marine-derived atmospheric CH4 than previously thought.

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