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A seasonal study of methyl bromide concentrations in the North Atlantic (35°–60°N)

Ryszard Tokarczyk[1] and Robert M. Moore[1]

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[1] Methyl bromide concentrations in and over the North Atlantic were examined during spring, summer, and fall 2003. The results demonstrate that seasonality plays a great role in controlling methyl bromide fluxes from and into the ocean in this area. The North Atlantic acted as a sink of the atmospheric gas during the spring, a source during the summer, and a weak sink during the fall. The annual air-sea flux of methyl bromide from the North Atlantic area between 30°N and 60°N (approximately 15.4×10^6 km^2) was estimated to be in the range of -0.3 to -0.6 Gg y^{-1}, with the methyl bromide flux varying between $-4.0 \times 10^6 \pm 1 \times 10^6$ g d^{-1}, $1.6 \times 10^6 \pm 0.6 \times 10^6$ g d^{-1}, and $-0.6 \times 10^6 \pm 0.4 \times 10^6$ g d^{-1} in spring, summer, and fall, respectively. Methyl bromide production necessary to balance air-sea exchange with oceanic losses was greater in the southern part than in the northern part of the studied area; no oceanic production was necessary to balance methyl bromide loss from the Arctic waters around 60°N. While the regional contribution to the methyl bromide global oceanic flux is small, it is also complex and dynamic. Our data suggest that in this part of the ocean the flux is not so much dependent on sea surface temperature as it is on other, still unknown environmental variables.

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1. Introduction

[2] Methyl bromide (CH_3Br) is the main contributor of ozone-depleting bromine to the atmosphere. The ocean is both the largest known source and the second largest known sink of this compound [*Butler and Rodriguez*, 1996; *Yvon-Lewis and Butler*, 1997; *World Meteorological Organization*, 1999], and over the last decade significant efforts have been made to understand the role of the ocean in the atmospheric exchange of methyl bromide. Oversaturated areas were found in the coastal east Pacific and in temperate Atlantic waters [*Lobert et al.*, 1995, 1996; *Groszko and Moore*, 1998; *King et al.*, 2000]. Undersaturated areas were identified in the north Atlantic including the Gulf of Mexico, the Caribbean Sea [*Lobert et al.*, 1996; *Groszko and Moore*, 1998; *King et al.*, 2000], the Labrador Sea [*Groszko and Moore*, 1998], the Pacific Ocean [*King et al.*, 2000], and the Southern Ocean [*Lobert et al.*, 1997; *Yvon-Lewis et al.*, 2004]. The results of these studies suggest that polar and tropical oceans are likely to be undersaturated in methyl bromide for most of the time, while the saturation state of temperate ocean waters is less certain and harder to predict. The effect of large areas of undersaturation outweighs the oceanic source regions, implying that the world ocean acts as a sink for atmospheric methyl bromide; the net flux being estimated as -10 to -20 Gg yr^{-1} [*Lobert et al.*, 1995; *Groszko and Moore*, 1998; *King et al.*, 2000; *Yvon-Lewis et al.*, 2004]. However, despite continuing research on sources and sinks, the atmospheric budget of methyl bromide remains unbalanced, with known sinks outweighing sources by between 25% and 50% [*Kurylo et al.*, 1999; *Montzka et al.*, 2003]. The reason for this discrepancy could be either overestimated sinks or a missing, or underestimated source; or perhaps a combination of both of these factors. Oceanic expeditions often provide a limited knowledge of the region's contribution to the atmospheric budget, as rarely can the same area be visited several times. When an area is revisited, the results are often quite different; for example, *King et al.* [2000] found undersaturation of methyl bromide in the same regions of the North Atlantic that *Groszko and Moore* [1998] reported to be oversaturated a few years earlier, and oversaturation in the same area of the ocean that *Lobert et al.* [1996] found undersaturated. These discrepancies call for more systematic studies and are strong indications that seasonality plays an important role in methyl bromide distribution and its fluxes between the ocean and the atmosphere. *Wingenter et al.* [1998] reported a seasonal variation in tropospheric methyl bromide that cannot be explained on the basis of known oceanic or terrestrial inputs. *Baker et al.* [1999] investigated seasonal variations in the concentration of CH_3Br in coastal North Sea waters and found that they were supersaturated during the summer and undersaturated during the reminder of the year, suggesting the existence of a seasonal cycle. Our investigations undertaken as a part of a 2003 Canadian Surface Ocean Lower Atmosphere Studies (SOLAS) experiment present

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[1] Department of Oceanography, Dalhousie University, Halifax, Nova Scotia, Canada.

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Table 1. Station Locations and Biogeochemical Provinces After *Roy and Devred* [2005]

Station	Location	Spring 2003	Summer 2003	Fall 2003
L	43.5°N, 57.5°W	North West Continental Shelf	Slope waters	Slope waters
T1	39.0°N, 57.5°W	Gulf Stream	Gulf Stream	Gulf Stream
T2	36.8°N, 57.4°W	Subtropical Gyre	Subtropical Gyre	Subtropical Gyre
T3	42.3°N, 45.0°W	Slope waters	Slope waters	Subtropical Gyre
T4	46.5°N, 45.0°W	North West. Continental Shelf	North Atlantic Drift	Slope waters
T5	50.7°N, 45.0°W	North Atlantic Drift	North Atlantic Drift	North Atlantic Drift
T6	54.8°N, 45.0°W	Arctic waters	Arctic waters	Arctic waters
T7	59.5°N, 45.0°N		Arctic waters	Arctic waters

the results of 3 consecutive cruises and provide the first seasonal information about CH_3Br concentrations and production rates in several areas of the North Atlantic including subtropical, temperate and Arctic waters. Our results fall in line with the observations of *Baker et al.* [1999] from the North Sea. The results also suggest that the dependence between the flux and sea surface temperature (SST), first observed by *Groszko and Moore* [1998] and further developed by *King et al.* [2000, 2002], does not hold well in this particular region but seems to be masked by other environmental variables.

2. Methods

[3] This study was undertaken at 8 locations in the North Atlantic (Table 1). The approximate positions of the stations are shown in Figure 1. Data were collected during spring (24 April to 15 May), summer (9–23 July) and fall 2003 (14–27 October) from Canadian Coast Guard Ship (CCGS) *Hudson* and CCGS *Martha Black.* The area of study includes 6 different biogeochemical provinces [*Sathyendranath et al.*, 1995; *Longhurst*, 1998], the boundaries between which varied as the seasons progressed.

2.1. Measurements

[4] The concentration of dissolved methyl bromide was measured at each location in a depth profile from the surface to about 300 m. The measurements were done at depths corresponding to preselected levels of light intensity (90%, 50%, 30%, 15%, 5%, 1% and 0.2%) from surface to 100 m and at intervals of 50 to 100 m below that depth. Seawater was collected in a series of 15 L Niskin bottles mounted on a rosette, from which samples were drawn into several (typically 8 to 12) 100 mL borosilicate glass syringes without any contact with air. Methyl bromide was measured in each sample by a purge-and-trap method combined with gas chromatography/mass spectrometry (GC/MS). Trace gases were stripped from seawater with a stream of ultra-high-purity (UHP) helium, preconcentrated in a small stainless steel loop held at $-150°C$, separated in a pair of DB624 capillary columns (total length, 100 m; ID, 0.53 mm; film thickness, 3 μm) and detected by a quadrupole Finnigan Trace MS. CD_3Br was used as an internal standard to monitor instrument stability and ensure accuracy of the analysis. A blank was established each day by running the system without the water sample or by repeatedly purging a water sample so that it was free of dissolved methyl bromide. Typical blanks were 0.1 pmol L^{-1}. Peak areas were calibrated with a gravimetrically produced standard of methyl bromide in nitrogen contained in an Aculife treated aluminum cylinder. The standard was measured using Hamilton gas tight syringes, and volumes between 50 and 200 μL were injected via a septum port at the beginning and at the end of each experimental day. The resulting calibration curves were linear. The precision of this technique was in the range of 2% to 3%. Most samples were analyzed within a few hours of collection. However, during the fall season, because of an instrumental problem, samples collected at T3 and T4 locations were stored in the dark at 10°C in syringes submerged in a bucket of seawater for 48 hours, before being analyzed. The methyl bromide concentration of these samples was corrected for chemical loss, but it was not possible to correct for biological effects (production or loss). As the quality of these samples was somewhat compromised, the data points (two) in Figure 2 that refer to these measurements are larger to reflect larger analytical error and distinguish them from the others.

[5] Air samples were collected periodically during spring and summer cruises. Air was pumped into electropolished steel canisters with a steel diaphragm sampling pump at the

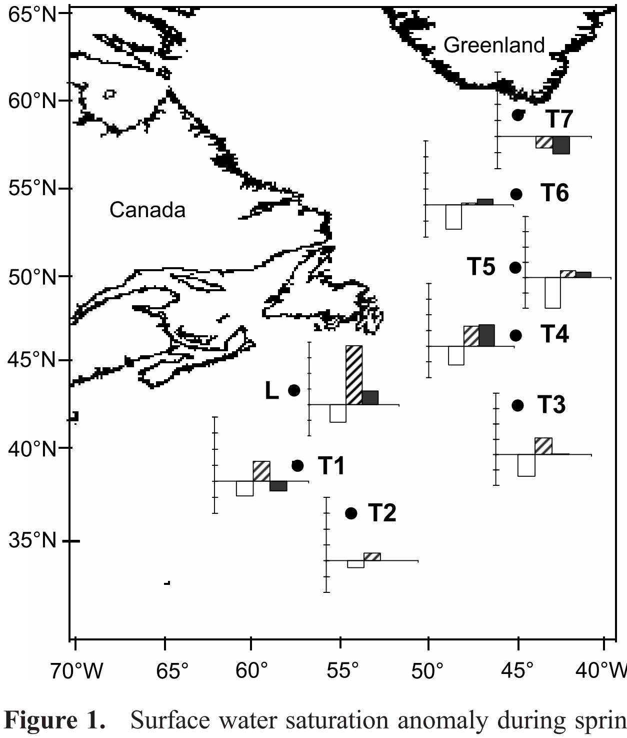

Figure 1. Surface water saturation anomaly during spring (open rectangles), summer (hatched rectangles), and fall (solid rectangles) of 2003. Vertical axis for all graphs extends from -50% to 100% (ticks marks are spaced every 25%). Solid circles represent positions of hydrographic stations (L to T7) during the experiment.

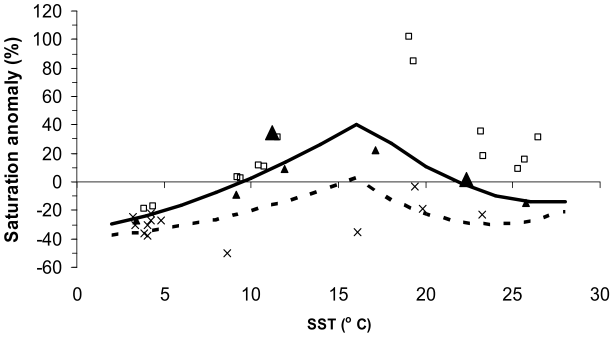

Figure 2. Methyl bromide saturation anomaly (SA) plotted against seawater temperature from spring (crosses), summer (squares), and fall (triangles) SOLAS 2003 cruises. The size of the symbol approximately equals the error of SA estimate (±5% for most data points). Solid line shows spring-summer empirical fit, and dashed line shows fall-winter empirical fit; both are from *King et al.* [2002]. The majority of our summer data points fall above the proposed spring-summer fit, while the majority of spring data points fall below this line; some even below the fall-winter empirical fit.

upwind rail of the ship. They were analyzed at the end of each cruise by cold trapping and GC/MS, using the same equipment as for the purge-and-trap samples but with a 200 mL stainless steel tubing loop in place of the purge vessel. Altogether we measured CH_3Br concentration in air 4 times during the spring and 11 times during the summer cruises, the precision of our measurements was within 2% to 4%. Further details of the sampling procedure and analytical equipment are described elsewhere [*Moore et al.*, 1996]. Air measurements are not available for fall 2003; therefore we have estimated atmospheric CH_3Br concentrations on the basis of the work of *Yokouchi et al.* [2002].

2.2. Calculations

[6] Fluxes (F) of CH_3Br between the ocean and the atmosphere were calculated from concentration differences across the air-sea interface (Δc) and a gas transfer velocity (K_w) according to the formula: $F = K_w \Delta c$, where: F = flux (nmol m^{-2} d^{-1}), K_w = gas transfer velocity (m d^{-1}) and $\Delta c = (c_w - p_{atm} \cdot H')$, where c_w (nmol m^{-3}) is the concentration of the gas in seawater, p_{atm} (atm) is the concentration of the gas in the atmosphere and H' (mol L^{-1} atm^{-1}) is the solubility of methyl bromide in seawater. A positive value of F indicates flux from the ocean to the atmosphere. We calculated methyl bromide solubility using the expression of *de Bruyn and Saltzman* [1997a], and the gas transfer velocity using the relationship $K_w = k\ u^2\ (S_c/660)^{-1/2}$ [*Wanninkhof*, 1992], where $k = 0.0936$ is a proportionality constant, u is the average wind speed and Sc is the Schmidt number. We used daily wind speed averages calculated for each particular location. We used the wind speed data from a physical oceanography database (PODAAC, Jet Propulsion Laboratory, http://poet.jpl.nasa.gov). The data are available at 0.25° resolution, wind speeds were averaged over 1° (with the station in the middle of 1° box) over the time prior and up to arrival on the site. In general, the wind speed data agreed well with the ship's log. We calculated Schmidt numbers for CH_3Br according to the expression $S_c = 2004 - 93.5t + 1.39t^2$, where t = sea surface temperature (°C) [*de Bruyn and Saltzman*, 1997b].

[7] By assuming a steady state, oceanic production can be estimated from the sum of the losses from seawater including flux to the atmosphere, downward mixing and chemical and biological loss. Estimates of the vertical diffusive flux from the mixed layer indicated that this loss process was comparatively small and was ignored for the purpose of our calculations. The chemical loss was calculated from seawater temperature and salinity using an empirical equation [*King and Saltzman*, 1997]. The biological loss was not measured during the SOLAS cruises, but on the basis of previous experiments [*Tokarczyk et al.*, 2001; *Tokarczyk and Saltzman*, 2001] we estimated it to average around 5% d^{-1} in the subtropical and the subarctic waters, and around 7% d^{-1} in the temperate waters. For the clarity of the conclusions we briefly discuss two possible production scenarios, one with no biological loss and one with biological loss at the levels indicated above.

3. Results and Discussion

3.1. Air Concentrations

[8] Methyl bromide mixes quickly within the troposphere and as a result its hemispheric concentration should remain relatively uniform. *Yokouchi et al.* [2002] reported a steady decline in atmospheric concentration at Alert from an average of ~11 ppt in 1996 to ~8.6 ppm in 2001. At Alert the highest concentrations were observed in late April, the lowest in early September with seasonal changes as high as ±1 ppt from the yearly average. Similar seasonal changes were observed at Barrow, Alaska, by *Wingenter et al.* [1998], who proposed that they may be explained by the seasonality of OH radicals that account for the main tropospheric sink of CH_3Br. Our results average at 9.3 ± 0.4 ppt in the spring and 8.3 ± 0.3 ppt in the summer, which are in very good agreement (>5%) with results reported by *Yokouchi et al.* [2002]. On the basis of this agreement we choose to use the literature value of 7.8 ppt [*Yokouchi et al.*, 2002] as representative for the fall CH_3Br atmospheric concentration in the studied area.

3.2. Seawater Concentrations

[9] Methyl bromide concentrations within the mixed layer varied from 1.0 to 2.4 pmol/L in spring, 1.3 to 3.6 pmol/L in summer, and 0.9 to 2.7 pmol/L in the fall. The highest concentrations were typically observed in the Continental Shelf and Slope waters, the lowest in the warm subtropical waters of the Gulf Stream and Sargasso Gyre. Concentration maxima were typically observed near the surface, between 0 and 20 m depths in spring/fall, but deeper (between 15 and 40 m) in the summer. The saturation level in the surface waters varied between seasons at most locations (Figure 2), which can only partially be explained by warming or cooling effects. In spring 2003 the entire area of the North Atlantic that we studied was undersaturated with methyl bromide. The saturation anomaly varied between biogeochemical provinces from −11% in the Sargasso Subtropical Gyre to −50% in the North Atlantic Drift waters (Table 2). As the seasons progressed most of the studied area became supersaturated, and, in the

Table 2. Methyl Bromide Saturation in the North Atlantic Biogeochemical Provinces (Spring-Fall 2003)[a]

		Saturation Anomaly		
Water Mass	SST	Spring	Summer	Fall
Continental Shelf (n = 7)	3°–5°C	−23% to −36%	–	–
Slope waters (n = 7)	11°–23°C	−35%	28–94%	22–34%
Gulf Stream waters (n = 4)	23°–27°C	−23%	31%	−15%
Subtropical Gyre/Sargasso (n = 5)	20°–25°C	−11%	12%	1%
North Atlantic Drift waters (n = 7)	9°–12°C	−50%	11–32%	9%
Arctic waters (n = 7)	3°–9°C	−38%	−19% to 3%	−9% to −27%

[a] The average value is reported when the data came from the same location, and a range is given when the data came from more than one station in a particular season. Uncertainty of reported saturation anomalies is ±4%. SST, sea surface temperature.

summer, positive saturation anomalies were observed everywhere except the northernmost location of 60°N (Figure 2). The largest supersaturations (up to 94%) were observed in the Slope waters in the vicinity of Nova Scotia (L station), the smallest (3%) at the southern edge of Arctic waters (55°N). The summer observations are in a qualitative agreement with earlier data of *Groszko and Moore* [1998], who reported supersaturation of temperate Nova Scotia shelf waters and warm subtropical waters of the Gulf Stream, and the Sargasso Sea in summer 1995. In the fall of 2003, saturation anomalies were lower than in summer; the coldest Arctic waters and the warmest Gulf Stream waters were undersaturated while Slope waters, North Atlantic Drift waters and Subtropical Gyre waters remained supersaturated but to a lesser degree than in the summer. On the basis of the observed trend we can speculate that the entire area would be undersaturated during the winter.

3.3. Sea Surface Temperature and Saturation Anomaly

[10] *Groszko and Moore* [1998] observed an empirical relationship between the saturation of methyl bromide and water temperature. Atlantic and Pacific Ocean waters were typically supersaturated at temperatures in the range 12° to 20°C, and undersaturated at temperatures outside this range, suggesting a simple model for predicting methyl bromide saturation on the basis of sea surface temperature alone [*Groszko and Moore*, 1998]. This relationship was further examined by *King et al.* [2000], who combined the data of Groszko and Moore with those from several NOAA cruises. They obtained a similar relationship with a slightly extended supersaturation range and concluded that on a global scale it may account for 40–70% of the observed CH_3Br variability. However, they also noticed that the relation cannot correctly reproduce changes at the regional scale, especially in the temperate ocean, where predicted values of saturation anomaly did not match the experimental data. In an attempt to overcome this difficulty, *King et al.* [2002] proposed treating the data collected during local spring-summer separately from those collected during fall-winter seasons and derived two sets of the "saturation – SST" equations. The new set of relations predict that during the fall-winter seasons ocean waters are undersaturated, while during the spring-summer seasons they are supersaturated within the temperature range from 10° to 22°C, and undersaturated outside this temperature range. Our seasonal observations in the North Atlantic suggest that this temperate region is likely even more complex and dynamic than previously thought. While our data fall within the scatter of the data used by *King et al.* [2002], they disagree with predictions given by their improved empirical equations. In the spring 2003 methyl bromide was undersaturated in the studied North Atlantic area over the entire SST range, which varied from 3° to 23°C (Figure 1). Only in the cold (3° to 5°C) continental shelf waters did the saturation data fit in the range proposed by spring-summer empirical equation (Figure 2), everywhere else they fell below that range, sometimes even below the fall-winter empirical line. In sharp contrast with spring conditions, only 2 months later, during the summer of 2003 most of the waters in the investigated area were supersaturated with methyl bromide, including warm subtropical waters (SST 27°C). Only the coolest (SST 4°C) Arctic waters at 60°N remained undersaturated with methyl bromide during this season. This time the measured saturation anomaly followed the predicted pattern within the temperature range of 3° to 15°C. In warmer waters saturation anomalies were higher, sometimes substantially than predicted (Figure 2). In the fall of 2003 the saturation anomaly in the studied region decreased from the summer values showing oversaturation between 10° and 22°C and undersaturation outside this temperature range. The fall pattern of saturation anomalies did not quite follow the fall-winter prediction suggested by *King et al.* [2002] but did fit the spring-summer one well (Figure 2). In our opinion these observations suggest that in temperate regions the relation between saturation anomaly and sea surface temperature is more dynamic than previously thought, heavily influenced by seasonality, and masked by other, still undefined environmental variables. From the changes in the saturation anomaly from spring to fall it seems likely that during winter the entire studied area becomes undersaturated in methyl bromide and that these conditions extend through the spring until April-May. By comparing our seasonal, limited sets of data with *King et al.* [2002] empirical fits, we can speculate that while in cold waters all data fall within the range of the King et al. proposed fit, they spread farther than current predictions allow when SST increases. The winter curve would likely be lower and flatter than proposed fall-winter fit, while the summer curve would likely be higher and steeper in its middle part than the proposed spring-summer fit (Figure 2). We suspect that the North Atlantic oscillates between two extremes, winter when waters over the entire area remain undersaturated and summer characterized by widespread supersaturation. Between these two extremes a relation between saturation anomaly and SST similar to these of *Groszko and Moore* [1998] and *King et al.* [2002] can be observed, but the temperature threshold separating over and undersaturated waters shifts seasonally. Consequently, in the temperate and perhaps even subtropical areas of the ocean where methyl bromide production and degradation processes are likely to

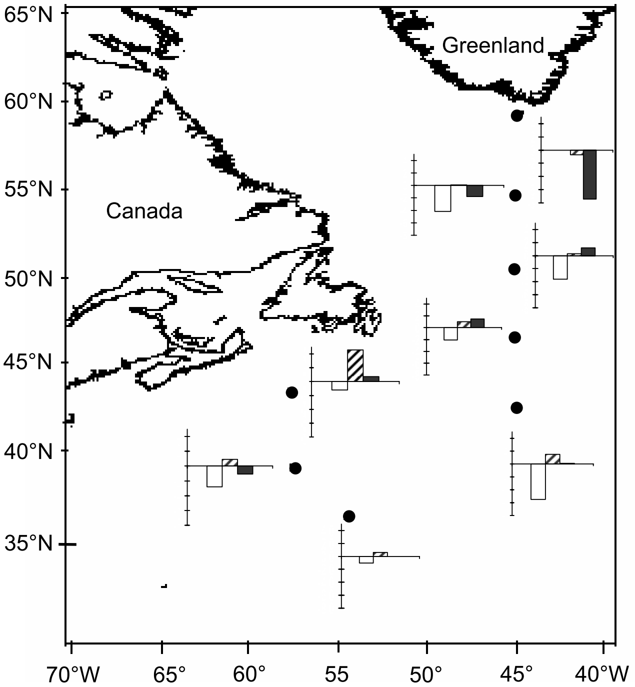

Figure 3. Fluxes (nmol m^{-2} d^{-1}) observed in the studied area between spring and fall 2003 (vertical scale extends from -8 to 5 nmol m^{-2}, tick marks are every 2 nmol m^{-2} d^{-1}). Positive values represent ocean-to-atmosphere flux, d^{-1}. Spring, open rectangles; summer, hatched rectangles; fall, solid rectangles.

be more dynamic than temperature changes, saturation of ocean waters cannot be predicted from the temperature data alone. Only the magnitude of these saturations can be estimated. Our observations support the earlier conclusion of *King et al.* [2000] that reliable prediction of air-sea fluxes in the Northern Atlantic will require information on additional variables.

3.4. Fluxes and Production

[11] During the spring of 2003 the North Atlantic was a sink for atmospheric methyl bromide (Figure 3). Negative fluxes were observed over the entire area. The lowest flux (-1 nmol $m^{-2}d^{-1}$) was observed in the Sargasso Subtropical Gyre. In the absence of biological degradation, no production would be needed to balance the loss of methyl bromide from the water anywhere except in the subtropical region. In the Sargasso Subtropical Gyre and the Gulf Stream, production in the range of 0.5 to 4 nmol $m^{-2}d^{-1}$ (respectively) was necessary to give mass balance. However, if biological degradation is assumed to be in the range of 5–7% d^{-1}, production in range of 1 nmol $m^{-2}d^{-1}$ (Sargasso Subtropical Gyre) to 7 nmol $m^{-2}d^{-1}$ (Gulf Stream and Arctic waters) would be necessary to balance oceanic losses.

[12] In the summer most of the North Atlantic area from 35°N to 55°N acted as a source of atmospheric methyl bromide. Only at the northernmost location (60°N) was the flux negative, indicating that this part of the ocean still acted as a sink. The highest fluxes (4–5 nmol $m^{-2}d^{-1}$) were observed in the area of station L. Net production in the range of 1–9 nmol $m^{-2}d^{-1}$ was required to balance the oceanic losses in the absence of biological degradation (1 to 11 nmol $m^{-2}d^{-1}$ with the estimated biological loss included). Required production was the highest in the subtropical waters of the Sargasso Gyre and the Gulf Stream, decreasing gradually toward the north. Virtually no production was required to balance the loss from the Arctic waters between 55°N and 60°N.

[13] During the fall, the oceanic source strength was weakening, with some areas (Arctic waters far north and subtropical Gulf Stream waters at the south) acting as sinks for atmospheric methyl bromide, while other areas remained weak sources. A strong negative (air to sea) flux up to -7.4 nmol $m^{-2}d^{-1}$ was observed near the Greenland coast (Figure 3), while a weaker negative flux was observed in the Gulf Stream region. In the rest of the area a small positive flux (0.1 to 1.5 nmol $m^{-2}d^{-1}$) was observed. In situ production needed to balance the oceanic losses was the highest of all the seasons, varying between 12 nmol $m^{-2}d^{-1}$ (15 nmol $m^{-2}d^{-1}$ with biological loss included) in the Gulf Stream area, and 2 nmol $m^{-2}d^{-1}$ in the temperate Slope and Atlantic Drift waters.

[14] Recent studies in the Southern Ocean [*Yvon-Lewis et al.*, 2004] and subsequent calculations that balance air-sea exchange with oceanic losses suggest that there is no methyl bromide production in cold Antarctic waters, raising the possibility that perhaps similar conditions are present in the northern hemisphere. Our observations in the Arctic waters suggest that this indeed may be the case. In the absence of biological degradation no production was required to balance the loss of CH_3Br from these waters in any of studied seasons. The addition of biological loss of about 6% d^{-1}, would require production in the range of 1 to 9 nmol $m^{-2}d^{-1}$ throughout the year to balance the loss at the southern edge of these waters (T6, 55°N), but still no production is required farther north at 60°.

[15] The area of the North Atlantic between 30°N and 60°N, is approximately 15.4×10^6 km^2, equivalent to about 20% of the Atlantic Ocean, or ~5% of the total World Ocean area. Assuming that our seasonal measurements are representative for that area we can try to estimate the magnitude and direction of seasonal fluxes in the region. The largest flux ($-4.0 \times 10^6 \pm 1 \times 10^6$ g d^{-1}) was observed during spring; at this time of the year the area was a sink for atmospheric methyl bromide. In the summer the direction of the flux changed and the region acted as source of methyl bromide to the atmosphere with a flux around 1.6×10^6 ($\pm 0.6 \times 10^6$) g d^{-1}. In the fall the area acted as a weak sink, the estimated flux was around -0.6×10^6 ($\pm 0.4 \times 10^6$) g d^{-1}. The winter flux has not been measured, however, making the assumption that it was in the range between fall and spring values would allow estimates to be made of the annual flux. If the winter flux is similar in magnitude to the fall or the spring flux, then the regional annual flux would be in the range of -0.3 Gg y^{-1} to -0.6 Gg y^{-1} respectively, which represents 3–6% of the global annual flux between the ocean and atmosphere estimated by *Groszko and Moore* [1998] or 1.5–3% of global annual flux estimated by *Lobert et al.* [1997] and *King et al.* [2002]. While the regional contribution to methyl bromide global oceanic flux is small, it is also one of the most complex and dynamic. This highlights the importance of seasonality in

the ocean-atmosphere exchange of biogenically produced gases and the need for more systematic studies in other parts of ocean.

[16] **Acknowledgments.** We thank the captains and the crews of CCGS *Hudson* and *Martha Black* for their cooperation during the 2003 cruises. We also thank chief scientists M. Levasseur, W. L. Miller, and M. Scarratt for their logistic support and Dalhousie Oceanography students Lu Wang and Stuart MacDonald for their assistance with sample collection. This research was supported by NSERC and the Canadian Foundation for Climate and Atmospheric Sciences.

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R. M. Moore and R. Tokarczyk, Department of Oceanography, Dalhousie University, Edsell Castle Circle, Halifax, NS, Canada B3H 4J1. (rmoore@dal.ca; rtokarcz@dal.ca)