

This discussion paper is/has been under review for the journal *Atmospheric Chemistry and Physics (ACP)*. Please refer to the corresponding final paper in *ACP* if available.

**Recent trends in  
atmospheric methyl  
bromide**

S. A. Yvon-Lewis and  
E. S. Saltzman

# Recent trends in atmospheric methyl bromide: analysis of post-Montreal Protocol variability

S. A. Yvon-Lewis<sup>1</sup> and E. S. Saltzman<sup>2</sup>

<sup>1</sup>Department of Oceanography, Texas A&M University, College Station, TX 77843, USA

<sup>2</sup>Department of Earth System Science, University of California Irvine, Irvine, CA, USA

Received: 9 February 2009 – Accepted: 23 February 2009 – Published: 10 March 2009

Correspondence to: S. A. Yvon-Lewis (syvon-lewis@ocean.tamu.edu)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Abstract

The atmospheric methyl bromide ( $\text{CH}_3\text{Br}$ ) burden has declined in recent years, in response to the phaseout of agricultural and structural fumigation consumption under the amendments to the Montreal Protocol. The timing and magnitude of this decrease represents an opportunity to examine our current understanding of the  $\text{CH}_3\text{Br}$  budget, the phaseout schedule, and recent estimates of interannual variability in biomass burning and global OH. In this study, simulations obtained from a time-dependent global model of atmospheric  $\text{CH}_3\text{Br}$  emissions and uptake are compared to observations from the NOAA flask network. The model includes an updated global methyl bromide source inventory that includes biofuel combustion emissions estimated at  $6.1 \pm 3 \text{ Gg yr}^{-1}$  globally. The phaseout of  $\text{CH}_3\text{Br}$  production for agricultural uses began in 1998, concurrent with the pulse in biomass burning associated with the 1998 El Niño. The combined effects of three factors (biomass burning, global OH, and anthropogenic phaseout) appear to explain most of the observed atmospheric methyl bromide trend over the 1997–2005 period. The global budget remains imbalanced, with a large missing source. These results suggest that more than 80% of the missing source does not exhibit significant interannual variability during the phaseout period and, therefore, does not result from underestimating agricultural  $\text{CH}_3\text{Br}$  emissions.

## 1 Introduction

$\text{CH}_3\text{Br}$  is a stratospheric ozone depleting substance that, unlike many other ozone-depleting compounds, has both natural and anthropogenic sources. The use of  $\text{CH}_3\text{Br}$  as an agricultural fumigant was banned according to the amendments to the Montreal Protocol (UNEP, 1995), and phaseout began in 1998. Although many sources and sinks of  $\text{CH}_3\text{Br}$  have been identified and quantified, the global budget of atmospheric  $\text{CH}_3\text{Br}$  for the pre-phaseout atmosphere is not balanced (WMO, 2003). The best estimate annual global budget for 1996 had an imbalance of nearly 37 Gg between sources

### Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

---

**Recent trends in  
atmospheric methyl  
bromide**S. A. Yvon-Lewis and  
E. S. Saltzman

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

and sinks (Table 1). This imbalance is clearly a result of our incomplete knowledge, rather than a real difference between sources and sinks, because the atmosphere was not rapidly changing at that time (Yokouchi et al., 2002; Montzka et al., 2003). The imbalance may reflect uncertainty in both natural and anthropogenic components of the budget. Knowledge of the emissions from terrestrial plants and ecosystems is quite limited (WMO, 2003; Gan et al., 1998; Dimmer et al., 2001; Redecker et al., 2000; Rhew et al., 2001; Varner et al., 1999). Even the anthropogenic emissions of CH<sub>3</sub>Br from agricultural fumigation are not easily quantified, since soil uptake during fumigation can be significant, and may vary with soil conditions and application technique (MBTOC, 2006).

The decline in atmospheric CH<sub>3</sub>Br burden resulting from the phaseout of agricultural emissions presents an opportunity to test our understanding of global CH<sub>3</sub>Br cycling. Yokouchi et al. (2002) and Montzka et al. (2003) detected the atmospheric decline using measurements from flask air networks. Those authors, as well as Reeves (2003), noted that the initial decline during 1998–2002 was roughly twice as large as expected. They suggested that the observations could be explained if the anthropogenic component of the total emissions was larger than previously thought. This assertion also required an atmospheric lifetime  $\geq 14\%$  longer than the best estimate of 0.7 yr based on the known CH<sub>3</sub>Br sinks (reaction with OH, photolysis, uptake by the ocean, and uptake by soils) (Yvon-Lewis and Butler, 1997). In this study, we revisit the atmospheric decline during phaseout using a slightly longer data set, covering 1995–2005 (Montzka et al., 2003 – with updates from NOAA/GMD <ftp://ftp.cmdl.noaa.gov/hats/methylhalides/ch3br/flasks/>), and an updated, time-dependent global model of CH<sub>3</sub>Br emissions and uptake.

## 2 Global model, budget and data sources

This study was carried out using a numerical model that simulates the major processes involved in the global biogeochemical cycle of CH<sub>3</sub>Br, including oceanic production

and consumption, soil uptake, terrestrial plant emissions, fumigation, gasoline combustion, biomass burning, biofuel emissions, atmospheric loss via reaction with OH and photolysis in the stratosphere. The model is described in detail by Saltzman et al. (2004). It consists of two well-mixed atmospheric boxes representing northern and Southern Hemispheres. Transport between the hemispheres is governed by an interhemispheric exchange time constant of 1 year. Surface fluxes of methyl bromide are seasonally and spatially variable. For the oceans, monthly,  $1^\circ \times 1^\circ$ , average ocean parameters from the NOAA/GFDL Global Oceanographic Data Set Atlas (available at <http://dss.ucar.edu/datasets/ds279.0>) were used to characterize oceanic temperatures, salinities, mixed layer depths, and 10 m wind speeds. Oceanic  $\text{CH}_3\text{Br}$  production rates are specified based on the observed saturation state of the pre-phaseout ocean (King et al., 2002). Each surface ocean cell maintains a mass balance for  $\text{CH}_3\text{Br}$  and interacts independently with the atmosphere. Land surface sources and sinks are distributed using the International Satellite Land Surface Climatology Project (ISLSCP) (Meeson et al., 1995; Sellers et al., 1995) and Matthews (1983) databases as described in Saltzman et al. (2004).

The pre-phaseout source and sink fluxes are shown in Table 1 and reflect a revision in the budget described by Montzka and Fraser (2003). The sinks and corresponding oceanic emission terms were adjusted for the revised tropospheric burden of 137 Gg (Montzka et al., 2003). Separate peatland and rice paddy emissions are not shown as the databases do not distinguish them from wetlands (Saltzman et al., 2004). The newly quantified biofuel emissions were added, and seasonal variations in biomass burning were included. This budget is not balanced, and the missing source is discussed below. The details of the sources and sinks used in this model are described in Saltzman et al. (2004). Changes made since then are discussed below.

Monthly fire distributions for 1997–2005 have been determined from satellite observations by van der Werf et al. (2006).  $\text{CH}_3\text{Br}$  emissions from these fires are scaled to the CO fluxes calculated for the fires using emissions factors from Andreae and Merlet (2001). The resulting  $\text{CH}_3\text{Br}$  emissions from biomass burning for 1997–2005 range

---

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

from 11 to 16 Gg yr<sup>-1</sup>. This range is lower than the previous best estimate of 20 Gg for biomass burning emissions in the Montzka and Fraser (2003) budget. There is an initial increase in biomass burning emission in 1998 with a subsequent decline in emissions through 2004 (Fig. 1). For the model simulations prior to 1997, the biomass burning emissions used are the monthly averages of the 1997–2005 data excluding the highest and lowest values for a given month.

The pseudo first order loss rate constant for CH<sub>3</sub>Br by reaction with OH was computed using the temperature-dependent rate constant from Sander et al. (2006) and the OH and temperature fields from Spivakovsky et al. (2000). The pseudo first order rate constants are averaged for each hemisphere for each month to generate a seasonal hemispheric OH sink for CH<sub>3</sub>Br. Interannual variability in global OH is simulated by scaling OH in the model to the variations inferred by Prinn et al. (2005) from the ALE-GAGE network measurements of methyl chloroform. According to those estimates, global OH decreased by approximately 13% during 1997/1998, most likely as a result of the increase in biomass burning during that time. Subsequently, the OH levels rebounded to values approaching those during 1995/1996.

Previous budget estimates do not include emissions from combustion of biofuels. While some of these emissions can be accounted for through the satellite fire database described for the biomass burning emission distributions, the emissions associated with burning of biofuels inside houses and structures cannot be observed from satellites. Emissions from biofuels, minus the outdoor burning that would be a part of the biomass burning calculations, equal approximately 6.1 Gg yr<sup>-1</sup> and are primarily in the Northern Hemisphere (Andreae and Merlet, 2001; Yevich and Logan, 2003). Yevich and Logan (2003) caution that their biofuel emission estimates could have an uncertainty of ±50%. This uncertainty directly applies to the CH<sub>3</sub>Br emissions from biofuels, as we used emission factors from Andreae and Merlet (2001) to convert the Yevich and Logan (2003) biofuel emissions to CH<sub>3</sub>Br emissions.

The total emissions from fumigation of soils, durables, perishables, and structures was 43.3 Gg yr<sup>-1</sup> in 1996 (Table 1). The hemispheric distribution, seasonality, and an-

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

nual trend of the agricultural fumigation source are based on government and industry statistics on consumption (MBTOC, 2006) (Fig. 2). The quarantine and preshipment (QPS) emissions remained constant at  $12.3 \text{ Gg yr}^{-1}$  from 1995 through 1998 (Buffin, 2004). The trend in QPS emissions from 1999 through 2005 is based on the QPS emission trends reported in MBTOC (2006). There was an increase in QPS emissions in 2005 (Table 1) that reflected the increased use to meet new guidelines for regulating wood products in international trade (IPPC, 2002; Federal Register, 2004.).

### 3 Results and discussion

#### 3.1 Model initialization and scenarios

The model is initialized by running it for several years with the 1996  $\text{CH}_3\text{Br}$  budget and Spivakovsky et al. (2000) OH levels, until a repeatable annual cycle in atmospheric  $\text{CH}_3\text{Br}$  is obtained. As noted earlier, there is a significant imbalance in the “best estimate”  $\text{CH}_3\text{Br}$  budget (Table 1), with atmospheric losses exceeding the known sources by  $\sim 37 \text{ Gg yr}^{-1}$  after adjusting to the  $137 \text{ Gg}$  tropospheric burden. The magnitude and seasonality of the unknown source(s) were adjusted in the model by trial and error to achieve agreement with the mean 1995-1998 tropospheric hemispheric mixing ratios ( $\text{NH}=10.2 \text{ ppt}$ ,  $\text{SH}=8.0 \text{ ppt}$ ) and seasonalities (Montzka et al., 2003) (Fig. 3a). This resulted in annually averaged emissions for the unknown source of  $\sim 19 \text{ Gg yr}^{-1}$  in the Northern Hemisphere and  $\sim 17 \text{ Gg yr}^{-1}$  in the Southern Hemisphere. This approach is the same as that used in Saltzman et al. (2004), with slightly different emissions reflecting the updated source inventory.

This model reproduces pre-phaseout atmospheric variations quite well, considering the crude seasonal parameterizations for the terrestrial sources and soil sink, and the fact that interhemispheric exchange is based on mean hemispheric levels. While the lack of atmospheric dynamics in this model precludes investigation of the spatial distribution of the missing source, the model can be used to examine the nature of the

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

missing source through interannual variations in sources and sinks and the impact of a change in lifetime.

Eight model scenarios are discussed here (Table 2). The first four cases are used to examine the impact of interannual variations in selected sources and sinks. The calculated hemispheric missing source strengths and seasonalities were fixed for the duration of Scenarios 1-4. Scenarios 5-8 are used to examine the nature of the missing source and the possibility of a longer atmospheric lifetime for CH<sub>3</sub>Br. The model is also used to investigate the impact of the atmospheric variability on the saturation state of the ocean.

### 3.2 Interannual Variability in biomass burning, atmospheric OH, and anthropogenic CH<sub>3</sub>Br emissions

The first scenario examines the impact of interannual variability in biomass burning CH<sub>3</sub>Br emissions on the atmospheric CH<sub>3</sub>Br burden (Table 2, Figure 4a). During the 1997-1998 period, the emissions from biomass burning increased by 3.0 Gg y<sup>-1</sup> (NH) and 5.6 Gg y<sup>-1</sup> (SH) over the mean values used for the pre-phaseout conditions. This case yields an increase in the annually averaged atmospheric CH<sub>3</sub>Br of 0.27 ppt (NH) and 0.26 ppt (SH) over the mid-1997 to mid 1998 period that is consistent with the observations. The effect of increased biomass burning was much larger (on a relative basis) in the Southern Hemisphere, both because the increase in emissions was larger than in the Northern Hemisphere, and because biomass burning is larger fraction of the total emissions in the Southern Hemisphere. Given the mean lifetime of about 0.7 years, roughly one third of the emissions in each hemisphere is transported to the other hemisphere. As noted by Saltzman et al. (2004) the approach of using hemispherically averaged atmospheric boxes may underestimate the interhemispheric mixing of low latitude sources, like biomass burning. Hemispheric averaging also undoubtedly slightly overestimates the atmospheric lifetime of low latitude emissions which are released in the region of highest OH and atmospheric reactivity.

The second scenario examines the effect of interannual variability in global OH (Ta-

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ble 2, Fig. 4b). Model OH levels are fixed at the Spivakovsky et al. (2000) values from 1995–1996. For model years 1997–2004, OH is scaled by the annual variations given by Prinn et al. (2005). For the remainder of the simulation, the 2004 OH scaling factor was used. The decline in OH concentrations during 1998 results in a increase in the atmospheric burden. The subsequent increase in OH back to pre-1998 values results in a decrease and subsequent leveling off of the atmospheric CH<sub>3</sub>Br.

The third scenario examines the effect of anthropogenic phaseout (Table 2, Fig. 4c). The phaseout imposed by the Montreal Protocol and its amendments resulted in a nearly 86% reduction in emissions from agricultural fumigation from 1996 budget to 2007. Critical Use Exemptions (CUEs) have allowed the emissions to decrease at a slower rate than originally specified by the amendments to Montreal Protocol (Fig. 2). For this model case, biomass burning was fixed at the 1996 values for the entire run. The model results show that the decline in anthropogenic emissions results in a significant decline in atmospheric CH<sub>3</sub>Br levels from 1996 to 2007 CH<sub>3</sub>Br. The phaseout alone appears to account for most of the overall change in atmospheric burden from 1996 to 2007. However, this scenario does not accurately capture the initial increase in CH<sub>3</sub>Br in 1997/1998, and it does not result in low enough atmospheric values in 2007.

The fourth scenario examines the combined effect of interannual variability in biomass burning, OH, and anthropogenic phaseout (Table 2, Fig. 4d). This case does a good job of simulating Northern Hemisphere variability, capturing both the 1998 increase and subsequent decline of CH<sub>3</sub>Br. In the Southern Hemisphere, the simulation overestimates the initial increase in CH<sub>3</sub>Br during late 1997 and early 1998. After the peak, southern hemispheric levels in the model run decreased rapidly, bringing the simulation back into agreement with the atmospheric measurements for several years. From 2004 through 2007, the model shows a leveling off of Southern Hemisphere CH<sub>3</sub>Br levels, while the atmospheric measurements show a continued decline until 2007. Both of the overestimates occur in the Southern Hemisphere and could be a result of the distributions of biomass burning near the equator and the location of the inter-tropical convergence zone.

---

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



---

**Recent trends in  
atmospheric methyl  
bromide**S. A. Yvon-Lewis and  
E. S. Saltzman

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

One of the limitations of this study is the inability to evaluate other biogeochemical impacts of the 1998 El Niño or other natural variations on other CH<sub>3</sub>Br sources and sinks. In particular, the droughts associated with the El Niño could have significantly affected both terrestrial emissions and the soil sink. Widespread changes in carbon cycling occurred as a result of the 1998 El Niño, as evidenced by the subsequent increase in atmospheric CO<sub>2</sub>. Increased CO<sub>2</sub> at that time was caused, at least in part, by a decrease in vegetative growth and an increase in soil respiration (Cramer et al., 2001; Nemani et al., 2003; Erbrecth and Lucht, 2006). However, there is insufficient information at present with which to assess how such changes affected CH<sub>3</sub>Br cycling.

The results discussed above suggest that the phaseout was superimposed on an unusually elevated background CH<sub>3</sub>Br level caused by the combined increase biomass burning associated with the 1998 El Niño (van der Werf et al., 2004) and the decrease in OH associated with the increased biomass burning emissions. This caused the initial decrease in CH<sub>3</sub>Br associated with the phaseout to appear larger than expected from the change in agricultural emissions.

### 3.3 The “Missing CH<sub>3</sub>Br Source” and atmospheric CH<sub>3</sub>Br lifetime

The model results discussed above demonstrate that the major features of the recent variability in atmospheric CH<sub>3</sub>Br can be simulated by simultaneously imposing inter-annual variability in biomass burning, global OH, and anthropogenic emissions on the 1996 best-estimate global budget including an unknown seasonally varying natural source that exhibits no interannual variability. Yokouchi et al. (2002) and Montzka *et al* (2003) detected the rapid decline in atmospheric CH<sub>3</sub>Br from 1998 to 2002. They suggested that the rapid decline could indicate that the anthropogenic component of the budget was larger than previously thought. This required either an upward revision of the anthropogenic emissions (as part or all of the missing source) or an upward revision of the atmospheric lifetime to  $\geq 0.8$  yr (Montzka et al., 2003) from the current best estimate of 0.7 yr (Yvon-Lewis and Butler, 1997).

In model scenarios 5–8, the magnitude and nature of the missing source is exam-

ined. The cause of the imbalance between sources and sinks is one of the most puzzling aspects of the global  $\text{CH}_3\text{Br}$  budget. This imbalance has persisted for more than a decade in all global budgets, despite continued research on both sources and sinks. It is generally suspected that the missing source is related to emissions from terrestrial vegetation. The modeling results of Warwick et al. (2006) suggest that the missing source is from a combination of tropical and subtropical plants and biomass burning. To date, only limited emissions surveys from terrestrial plants and ecosystems have been carried out, and it can be argued that the seasonality of these sources is not well documented (WMO, 2003; Gan et al., 1998; Dimmer et al., 2001; Redecker et al., 2000; Rhew et al., 2001; Varner et al., 1999).

Model scenario 5 examines the possibility that the missing source is really just an underestimate of the non-QPS anthropogenic emissions (Table 2, Fig. 4e). The missing source along with its seasonality is added to the agricultural emissions in each hemisphere increasing these emissions, and no additional missing source is included. The biomass burning trend, OH trend, and phaseout rate are maintained as in scenario 4. Including the missing source in the non-QPS anthropogenic emissions results in a model-predicted decline during phaseout more rapid than observed, and ending in lower concentrations than observed (Fig. 4e). For the missing source to be entirely anthropogenic, and for the model to reproduce the atmospheric trend, this source must consist of QPS emissions which are not subject to the phaseout. It is unlikely that QPS emissions were underestimated by nearly 100%.

Model scenario 6 examines the effect of increasing the lifetime of  $\text{CH}_3\text{Br}$  to reduce the “missing source” to half of the amount required in the cases above (Table 2, Fig. 4f). This is done by reducing both the missing source and the soil uptake by in each hemisphere relative to the best estimate budget. This results in an increase in the atmospheric lifetime to 0.81 yr (~16% increase over the best estimate). With the longer lifetime, the model also underestimates the seasonal amplitude of atmospheric  $\text{CH}_3\text{Br}$ , particularly in the Northern Hemisphere. Increasing the atmospheric lifetime still further to account for the entire missing source would lead to an even larger discrepancy

---

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

in terms of the amplitude of the seasonal cycle. The phaseout of anthropogenic emissions results in an underestimate of atmospheric CH<sub>3</sub>Br levels after 2002 due to the loss of the remaining missing source, as it was included as an anthropogenic non-QPS emission in this scenario.

Scenario 7 assumes that the reduced global missing source is assumed to be natural and does not decline with the anthropogenic phaseout (Table 2, Fig. 4g). Other conditions are as in scenario 6. The model results exhibit fairly good agreement with the observations. However, as in scenario 4, the model overpredicts the atmospheric values after 2004 slightly. As is scenario 6, seasonal amplitudes pre-phaseout and post 2004 are underestimated.

While the interannual variations in OH introduced in case 2 have an effect on the atmospheric CH<sub>3</sub>Br lifetime, this effect is very small and is largest for 1997/1998 with an approximately 0.04 yr (<6%) increase in the lifetime for those two years (Fig. 4b). After 1998, the CH<sub>3</sub>Br lifetime in the model gradually returns to the initial 0.7 yr value. An increase in the lifetime large enough to account for only half of the missing source results in a seasonal amplitude that is smaller than observed pre-phaseout (Fig. 4f and g). The missing source is not the result of overestimated sinks.

Scenario 8 involves increasing the escape rate of agricultural CH<sub>3</sub>Br to 60% of the amount applied rather than the 50% assumed in scenarios 1–4 (Table 2, Fig. 4h). The lifetime is the same as in scenarios 1–5 (~0.7 yr). The missing source in each hemisphere is reduced by the amount that the agricultural emission increased and seasonality was adjusted to match the 1995–1998 tropospheric mixing ratios (NH=10.2 ppt, SH=8.0 ppt) and seasonalities (Montzka et al., 2003; Fig. 3b). This revised pre-phaseout budget is given in Table 1. The prescribed interannual variations in biomass burning emissions, reaction with OH, and anthropogenic emissions are the same as in scenario 4. By maintaining the original lifetime, the seasonal amplitude of the atmospheric CH<sub>3</sub>Br is closer to the observed seasonality for the entire simulation. The 1997 Southern hemispheric concentrations are elevated compared to observations, as in scenario 4. The predicted concentrations from 2002–2007 are in better agree-

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

ment with the observations than in any other scenario. The budget for 2007 from this scenario is shown in Table 1.

### 3.4 Ocean saturation state

In this study, oceanic  $\text{CH}_3\text{Br}$  production (magnitude, geographic distribution, and seasonality) was calculated for 1996 conditions. This oceanic production was held constant for the duration of all model runs, as were the kinetic parameters affecting oceanic  $\text{CH}_3\text{Br}$  cycling (wind speeds, mixed layer depths, surface ocean temperature, and biological degradation rate constants). Although oceanic production was held constant, the saturation state of the oceans with respect to the overlying atmosphere varies in the model runs as a function of the atmospheric  $\text{CH}_3\text{Br}$  levels, and the internal cycling of  $\text{CH}_3\text{Br}$  within the oceans. Figure 5a shows the time series of oceanic  $\text{CH}_3\text{Br}$  saturation state obtained from scenario 8. The decrease in saturation anomaly for 1997 and 1998 results from the increase in atmospheric burden caused by increased biomass burning emissions. The saturation state of the oceans increased substantially during the phaseout period, shifting from a saturation anomaly (deviation from equilibrium) of  $-11.7\%$  in 1996 to  $-6.0\%$  in 2005 (Fig. 5a). The saturation anomaly decreased slightly during 2005 due to the increase in atmospheric  $\text{CH}_3\text{Br}$ , most notably in the Southern Hemisphere. Before phaseout, the Northern Hemisphere was more undersaturated than the Southern Hemisphere. This reversed during the first year of phaseout, likely as a result of the much greater decline in emissions from the Northern Hemisphere.

The model results indicate that the oceans in both hemispheres, should now be closer to equilibrium than prior to the phaseout. In fact, the model results indicate that the net flux is now positive (from sea to air) in the summer in each hemisphere. The global average net flux in 2007 is predicted to be  $-6.6 \text{ Gg yr}^{-1}$ , less than half the pre-phaseout global average of  $-14.0 \text{ Gg yr}^{-1}$ . Most of the published saturation anomaly measurements for  $\text{CH}_3\text{Br}$  in the open ocean were conducted during pre-phaseout conditions (King et al., 2002), and there are insufficient data from the phaseout period for comparison. Saturation anomaly measurements across the major ocean basins dur-

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

ing different seasons should be carried out to test this prediction and validate current models of oceanic cycling.

## 4 Conclusions

The observed decrease in atmospheric  $\text{CH}_3\text{Br}$  resulting from the Montreal Protocol places new constraints on the global  $\text{CH}_3\text{Br}$  budget. This study suggests that a portion ( $\sim 7 \text{ Gg yr}^{-1}$  or about 20%) of the  $\text{CH}_3\text{Br}$  missing source is likely the result of underestimated anthropogenic fumigation emissions. The remainder ( $\sim 31 \text{ Gg yr}^{-1}$  or about 80%) cannot be due to underestimation of the agricultural emissions and is not likely to be the result of overestimated sinks (underestimated lifetime). Increasing agricultural sources beyond a 60% escape rate does not yield agreement with the observed atmospheric rate of decline even when the lifetime is increased (Fig. 4e and f). The best estimate lifetime (0.7 years) and larger missing source are necessary to maintain the large observed seasonal amplitude both before the phaseout and after 2002. Assuming constant production of  $\text{CH}_3\text{Br}$  in the oceans, the decreasing atmospheric levels imply a substantial increase in the saturation state of the oceans. The resulting change in air/sea flux represents an additional  $7.4 \text{ Gg}$  in net flux to the atmosphere in 2007, relative to prephaseout conditions.

We conclude from this study that the behavior of the atmospheric  $\text{CH}_3\text{Br}$  burden over the phaseout period is reasonably consistent with our current understanding of the global budget which includes an unidentified source. Interannual variations in the atmospheric burden are sensitive not only to the anthropogenic phaseout but also to variations in biomass burning emissions and OH radical concentrations. There is no evidence from this study that the anthropogenic contribution to the global budget has been underestimated or that the atmospheric lifetime of  $\text{CH}_3\text{Br}$  is appreciably longer than previously estimated. There remains a significant imbalance in the global budget, which is likely due to unknown or underestimated natural sources. Additional field and modeling studies are needed in order to better constrain the  $\text{CH}_3\text{Br}$  source from

### Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



terrestrial ecosystems, particularly in the tropics. This study also highlights the need for new measurements of oceanic CH<sub>3</sub>Br saturation anomalies, to provide a comparison of pre- and post-phaseout conditions and to validate current models of oceanic cycling.

*Acknowledgements.* We would like to thank Jim Butler, Dan King, Ryszard Tokarczyk, and Michael Prather for innumerable insightful discussions, and Guido van der Werf and Jim Ranson for use of their biomass burning estimates. We particularly thank Steve Montzka for his helpful criticism and healthy skepticism. This research was supported by the National Science Foundation and the National Oceanic and Atmospheric Administration.

## References

- 10 Andreae, M. O. and Merlet, P., Emission of trace gases and aerosols from biomass burning: Global Biogeochem. Cy., 15(4), 955–966, 2001.
- Buffin, D.: Methyl bromide exemptions flout rules of Montreal Protocol, Pesticides News, 64, 18–19, 2004.
- 15 Cramer, W., Bondeau, A., Woodward, F. I., et al.: Global response of terrestrial ecosystem structure and function to CO<sub>2</sub> and climate change: results from six dynamic global vegetation models, Global Change Biol., 7, 357–373, 2001.
- Dimmer, C. H., Simmonds, P. G., Nickless, G., and Bassford, M. R.: Biogenic fluxes of halomethanes from Irish peatland ecosystems, Atmos. Environ., 35, 321–330, 2001.
- 20 Erbrecht, T. and Lucht, W.: Impacts of large-scale climatic disturbances on the terrestrial carbon cycle, Carbon Balance Management, 1:7, doi:10.1186/1750-0680-1-7, 2006.
- Federal Register: Department of Agriculture; Animal and Plant Health Inspection Service; Importation of Wood Packaging Material, 69 Federal Register 179, 16 September, 55719–55733, 2004.
- 25 Gan, J., Yates, S. R., Ohr, H. D., and Sims, J. J.: Production of methyl bromide by terrestrial higher plants, Geophys. Res. Lett., 25, 3595–3598, 1998.
- IPPC, International Standards for Phytosanitary Measures. ISPM No.15. Guidelines for Regulating Wood Packaging Material in International Trade. Secretariat of the International Plant Protection Convention, FAO, Rome, Italy, 2002.
- King, D. B., Butler, J. H., Yvon-Lewis, S. A., and Cotton, S. A.: Predicting oceanic methyl bro-

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

wide saturation from SST, *Geophys. Res. Lett.*, 29(24), 2199, doi:10.1029/2002GL016091, 2002.

Lee-Taylor, J. M. and Holland, E. A.: Litter decomposition as a potential natural source of methyl bromide, *J. Geophys. Res.*, 105, 8857–8864, 2000.

5 MBTOC, United Nations Environment Programme (UNEP) Methyl Bromide Technical Options Committee (MBTOC) 2006 Assessment of the Alternatives to Methyl Bromide. United Nations Environment Programme, Nairobi: 453 pp., (online available at: <http://ozone.unep.org/teap/Reports/MBTOC/index.shtml>), 2006.

10 Matthews, E.: Global vegetation and land use: New high-resolution databases for climate studies, *J. Clim. Appl. Meteorol.*, 22, 474–487, 1983.

Meeson, P. J., Corprew, F. E., McManus, J. M. P., Mayers, D. M., Closs, J. W., Sun, K.-J., Sunday, D. J., and Sellers, P. J.: ISLSCP Initiative I-Global data sets for land-atmosphere models, 1987–1988 (CD-ROM), vol. 1–5, USA\_NASA\_GDAAC\_ISLSCP\_001-USA\_NASA\_GDAAC\_ISLSCP\_005, <http://daac.gsfc.nasa.gov/>, Distrib. Active Arch. Cent., Greenbelt, MD, USA, 1995

15 Montzka, S. A., Butler, J. H., Hall, B. D., Mondeel, D. J., and Elkins, J. W.: A decline in tropospheric organic bromine, *Geophys. Res. Lett.*, 30, 1826, doi:10.1029/2003GL017745, 2003.

20 Montzka, S. A. and Fraser, P. J. (Lead Authors), Butler, J. H., Connell, P. S., Cunnold, D. M., Daniel, J. S., Derwent, R. G., Lal, S., McCulloch, A., Oram, D. E., Reeves, C. E., Sanhueza, E., Steele, L. P., Velders, G. J. M., Weiss, R. F., and Zander, R. J.: Controlled substances and other source gases, Chapter 1 in *Scientific Assessment of Ozone Depletion: 2002*, Global Ozone Research and Monitoring Project-Report No. 47, World Meteorological Organization, Geneva, Switzerland, 2003.

25 Nemani, R. R., Keeling, C. D., Hashimoto, H., Jolly, W. M., Piper, S. C., Tucker, C. J., Myneni, R. B., and Running, S. W.: Climate-Driven Increases in global terrestrial net primary production from 1982 to 1999, *Science*, 300, 1560, doi:10.1126/science.1082750, 2003.

30 Prinn, R. G., Huang, J., Weiss, R. F., et al.: Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, *Geophys. Res. Lett.*, 32, L07809, doi:10.1029/2004GL022228, 2005.

Redeker, K. R., Wang, N. Y., Low, J. C., McMillan, A., Tyler, S. C., and Cicerone, R. J.: Emissions of methyl halides and methane from rice paddies, *Science*, 290, 966–969, 2000.

Reeves, C. E.: Atmospheric budget implications of the temporal and spatial trends in methyl

---

**Recent trends in atmospheric methyl bromide**

S. A. Yvon-Lewis and  
E. S. Saltzman

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- bromide concentration, *J. Geophys. Res.*, 108, 4343, doi:10.1029/2002JD002943, 2003.
- Rhew, R. C., Miller, B. R., and Weiss, R. F.: Natural methyl bromide and methyl chloride emissions from coastal salt marshes, *Nature*, 403, 292–295, 2000.
- Rhew, R. C., Miller, B. R., Vollmer, M. K., and Weiss, R. F.: Shrubland fluxes of methyl bromide and methyl chloride, *J. Geophys. Res.*, 106, 20875–20882, 2001.
- Saltzman, E. S., Aydin, M., De Bruyn, W. J., King, D. B., and Yvon-Lewis, S. A.: Methyl bromide in preindustrial air: Measurements from an Antarctic ice core, *J. Geophys. Res.*, 109, D05301, doi:10.1029/2003JD004157, 2004.
- Sander, S. P., Finlayson-Pitts, B. J., Friedl, R. R., Golden, D. M., Huie, R. E., Keller-Rudek, H., Kolb, C. E., Kurylo, M. J., Molina, M. J., Moortgat, G. K., Orkin, V. L., Ravishankara, A. R. and Wine, P. H.: “Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies, Evaluation Number 15”, JPL Publication 06-2, Jet Propulsion Laboratory, Pasadena, USA, 2006.
- Sellers, P. J., Meeson, B. W., Closs, J., et al.: An overview of the ISLSCP Initiative I global data sets for land-atmosphere models, 1987–1988, vol. 1–5: USA\_NASA\_GDAAC\_ISLSCP\_001, OVERVIEW.DOC, <http://daac.gsfc.nasa.gov/>, Distrib. Active Arch. Cent., Greenbelt, MD, USA, 1995.
- Shorter, J. H., Kolb, C. E., Crill, P. M., Kerwin, R. A., Talbot, R. W., Hines, M. E., and Harriss, R. C.: Rapid degradation of atmospheric methyl bromide in soils, *Nature*, 377, 717–719, 1995.
- Spivakovsky, C. M., Logan, J. A., Montzka, S. A., Balkanski, Y. J., Foreman-Fowler, M., Jones, D. B. A., Horowitz, L. W., Fusco, A. C., Brenninkmeijer, C. A. M., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Three-dimensional climatological distribution of tropospheric OH: update and evaluation, *J. Geophys. Res.*, 105, 893–8980, 2000.
- Thomas, V. M., Bedford, J. A., and Cicerone, R. J.: Bromine emissions from leaded gasoline, *Geophys. Res. Lett.*, 24, 1371–1374, 1997.
- UNEP, Report of the seventh meeting of the parties to the Montreal Protocol on Substances that Deplete the Ozone Layer, United Nations Environmental Program, World Meteorol. Org. (WMO), Geneva, Switzerland, December 1995.
- van der Werf, G. R., Randerson, J. T., Collatz, G. J., Giglio, L., Kasibhatla, P. S., Arellano Jr., A. F., Olsen, S. C., and Kasischke, E. S.: Continental-Scale Partitioning of Fire Emissions During the 1997 to 2001 El Niño/La Niña Period, *Science*, 303, 73–76, 2004.
- van derWerf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano, Jr., A. F.: Interannual variability in global biomass burning emissions from 1997 to 2004,

---

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



Atmos. Chem. Phys., 6, 3423–3441, 2006,  
<http://www.atmos-chem-phys.net/6/3423/2006/>.

Varner, R. K., Crill, P. M., and Talbot, R. W.: Wetlands: a potentially significant source of atmospheric methyl bromide and methyl chloride, *Geophys. Res. Lett.*, 26(16), 2433–2436, 1999.

Warwick, N. J., Pyle, J. A., and Shallcross, D. E.: Global modeling of the atmospheric methyl bromide budget, *J. Atmos. Chem.*, 54, 133–159, doi:10.1007/s10874-006-9020-3, 2006.

World Meteorological Organization, Scientific Assessment of Ozone Depletion: 2002, Rep. Global Ozone Res. And Monitor. Proj., vol. 47, 498 pp., World Meteorol. Organ., Geneva, Switzerland, 2003.

Yevich, R. and Logan, J. A.: An assessment of biofuel use and burning of agricultural waste in the developing world, *Global Biogeochem. Cy.*, 17(4), 1095, doi:10.1029/2002GB001952, 2003.

Yokouchi, Y., Toon-Sauntry, D., Yazawa, K., Inagaki, T., and Tamaru, T.: Recent decline of methyl bromide in the troposphere, *Atmos. Environ.*, 36, 4985–4989, 2002.

Yvon-Lewis, S. A. and Butler, J. H.: The potential effect of oceanic biological degradation on the lifetime of atmospheric CH<sub>3</sub>Br, *Geophys. Res. Lett.*, 24, 1227–1230, 1997.

Yvon-Lewis, S. A., and Butler, J. H.: Effect of oceanic uptake on atmospheric lifetimes of selected trace gases, *J. Geophys. Res.*, 107, 4414, doi:10.1029/2001JD001267, 2002.

ACPD

9, 6515–6539, 2009

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Table 1.** Global CH<sub>3</sub>Br budgets for 1996 and 2005. The 1996 budget is modified from WMO (2003) and includes the average 1997–2005 biomass burning. The italicized numbers highlight budget terms that changed between 1996 and 2007. Superscripts indicate references.

	Best Estimate (Gg/y) Best		
	1996	1996 (60% Ag)	2007
Sources			
Ocean	42.0 <sup>*1</sup>	42.0 <sup>*1</sup>	42.0 <sup>**</sup>
Fumigation-Quarantine and Preshipment	12.3 <sup>2</sup>	12.3 <sup>2</sup>	<i>13.82</i> <sup>3</sup>
Fumigation-Soils and Other	31.0 <sup>3</sup>	<i>36.6</i> <sup>3</sup>	<i>5.2</i> <sup>3</sup>
Gasoline	5.7 <sup>4</sup>	5.7 <sup>4</sup>	5.7 <sup>4</sup>
Biomass Burning	11.4 <sup>5,6</sup>	11.4 <sup>5,6</sup>	11.4 <sup>5,6</sup>
Biofuel	6.1 <sup>6,7</sup>	6.1 <sup>6,7</sup>	6.1 <sup>6,7</sup>
Wetlands	4.6 <sup>8</sup>	4.6 <sup>8</sup>	4.6 <sup>8</sup>
Salt marshes	14.6 <sup>9</sup>	14.6 <sup>9</sup>	14.6 <sup>9</sup>
Shrublands	1 <sup>10</sup>	1 <sup>10</sup>	1 <sup>10</sup>
Rapeseed	6.6 <sup>11</sup>	6.6 <sup>11</sup>	6.6 <sup>5</sup>
Fungus	1.7 <sup>12</sup>	1.7 <sup>12</sup>	1.7 <sup>12</sup>
Subtotal Sources	137	<i>143</i>	<i>113</i>
Sinks			
Ocean	-56 <sup>13,14</sup>	-56 <sup>13,14</sup>	<i>-48.6</i> <sup>13,14</sup>
OH and hv	-77 <sup>14,15</sup>	-77 <sup>14,15</sup>	<i>-63.6</i> <sup>14,15,16</sup>
Soils	-41 <sup>14,17</sup>	-41 <sup>14,17</sup>	<i>-34.0</i> <sup>14,17</sup>
Plants			
Subtotal Sinks	-174	-174	<i>-146</i>
Total (Sources+Sinks)	-37 <sup>***</sup>	<i>-31</i> <sup>***</sup>	<i>-32</i> <sup>***</sup>

<sup>1</sup>King et al. (2002), <sup>2</sup>Buffin (2004), <sup>3</sup>MBTOC (2006), <sup>4</sup>Thomas et al. (1997), <sup>5</sup>van der Werf et al. (2006), <sup>6</sup>Andreae and Merlet (2001), <sup>7</sup>Yevich and Logan (2003), <sup>8</sup>Varner et al. (1999), <sup>9</sup>Rhew et al. (2000), <sup>10</sup>Rhew et al. (2001), <sup>11</sup>Gan et al. (1998), <sup>12</sup>Lee-Taylor and Holland (2000), <sup>13</sup>Yvon-Lewis and Butler (2002), <sup>14</sup>Saltzman et al. (2004), <sup>15</sup>Spivakovsky et al. (2000), <sup>16</sup>Prinn et al. (2005) <sup>17</sup>Shorter et al. (1995).

\* Net ocean flux = -14.0 Gg/y

\*\* Net ocean flux = -6.6 Gg/y

\*\*\* Assumed to be a natural missing source

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Table 2.** Model scenarios used in this study. If not explicitly stated, sources and sinks are maintained as in the 1996 model year.

Scenarios to examine interannual variations in sources/sinks:	
1	Interannual variability in biomass burning only
2	Interannual variability in OH only.
3	Interannual variability in non-QPS anthropogenic emissions only due to phaseout.
4	Interannual variability in biomass burning, OH and non-QPS anthropogenic emissions.
Scenarios to examine missing source and atmospheric lifetime:	
5	missing source term treated as agricultural emissions and allowed to decrease with phaseout. Interannual variability in biomass burning, OH and non-QPS anthropogenic emissions included.
6	missing source reduced by 50%, and atmospheric lifetime of CH <sub>3</sub> Br increased to 0.81 yr. Remaining missing source adjusted to match the observed pre-phaseout seasonality and treated as agricultural. Interannual variability in biomass burning, OH and non-QPS anthropogenic emissions included.
7	missing source reduced by 50%, and atmospheric lifetime of CH <sub>3</sub> Br increased to 0.81 yr. Remaining missing source adjusted to match the observed pre-phaseout seasonality and treated as natural. Interannual variability in biomass burning, OH and non-QPS anthropogenic emissions included.
8	Agricultural emissions increased to 60%, and atmospheric lifetime kept as it was in scenarios 1–5. missing source reduced by the amount of the agricultural increase. Interannual variability in biomass burning, OH and non-QPS anthropogenic emissions included.

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

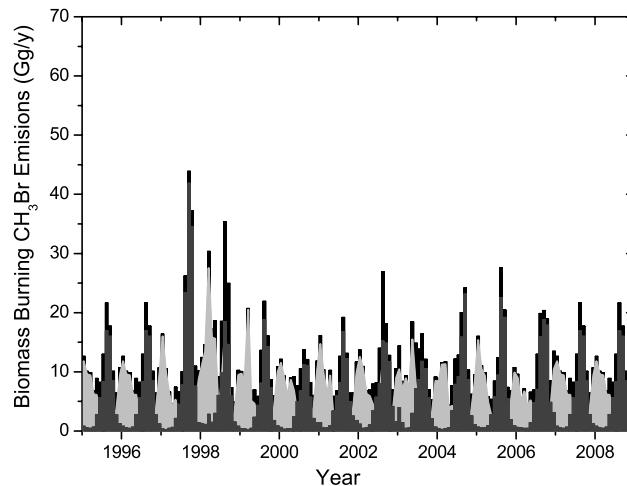
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman

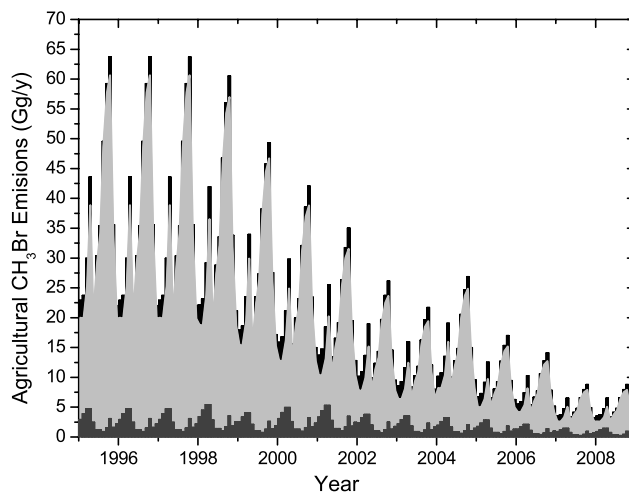


**Fig. 1.** The trend in biomass burning emissions of CH<sub>3</sub>Br for the Southern Hemisphere (dark gray), the Northern Hemisphere (light gray), and the globe (black). Prior to 1997, the biomass burning emissions are shown as the monthly averages of the 1997–2005 data excluding the highest and lowest values for a given month.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman



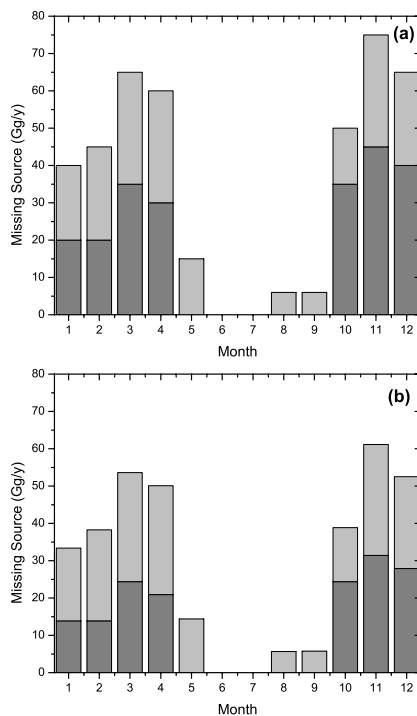
**Fig. 2.** The trend in agricultural emissions of CH<sub>3</sub>Br from 1995 through 2008 for the Southern Hemisphere (dark gray), the Northern Hemisphere (light gray), and the globe (black).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

---

**Recent trends in  
atmospheric methyl  
bromide**S. A. Yvon-Lewis and  
E. S. Saltzman

---

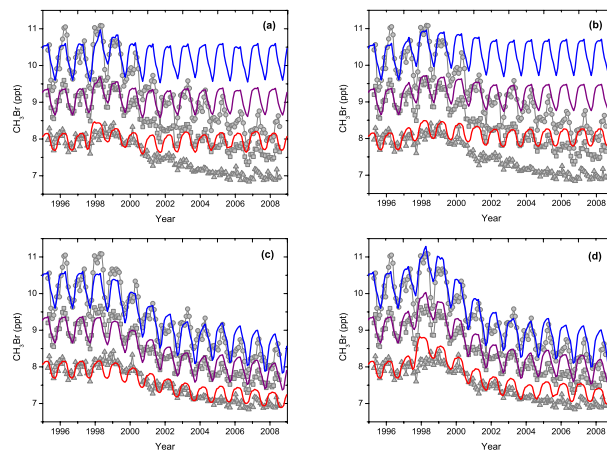


**Fig. 3.** Seasonal cycle in the missing source for the Southern Hemisphere (light gray) and the Northern Hemisphere (dark gray) for **(a)** scenarios 1–5 and **(b)** scenario 8.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman



**Fig. 4.** Modeled and measured trends in atmospheric  $\text{CH}_3\text{Br}$  concentrations for 1995–2006. In all plots, the data points for Southern Hemisphere ( $\blacktriangle$ ), the Northern Hemisphere ( $\bullet$ ), and the globe ( $\blacksquare$ ) are observations from NOAA/GMD (<ftp://ftp.cmdl.noaa.gov/hats/methylhalides/ch3br/flasks/>). Model results are shown for the Southern Hemisphere ( $-$ ), the Northern Hemisphere ( $-$ ), and the globe ( $-$ ) for several scenarios: **(a)** biomass burning trend with no anthropogenic phaseout, **(b)** OH trend with no trend in biomass burning and no anthropogenic phaseout, **(c)** the anthropogenic phaseout only, **(d)** anthropogenic phaseout with biomass burning and OH trends, **(e)** anthropogenic phaseout with biomass burning and OH trends with the missing source included in the agricultural emissions, **(f)** the anthropogenic phaseout with trends in biomass burning and OH with an increase in the lifetime and the remainder of the missing source, with the seasonality optimized for pre-phaseout conditions, included in the agricultural emissions, **(g)** the anthropogenic phaseout with trends in biomass burning and OH with an increase in the lifetime and the remainder of the missing source included as a natural source with the seasonality optimized for pre-phaseout conditions, and **(h)** agricultural emissions increased to 60% with the remainder of the missing source included as a natural source with the seasonality optimized for pre-phaseout conditions and the lifetime the same as in scenarios 1–5.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

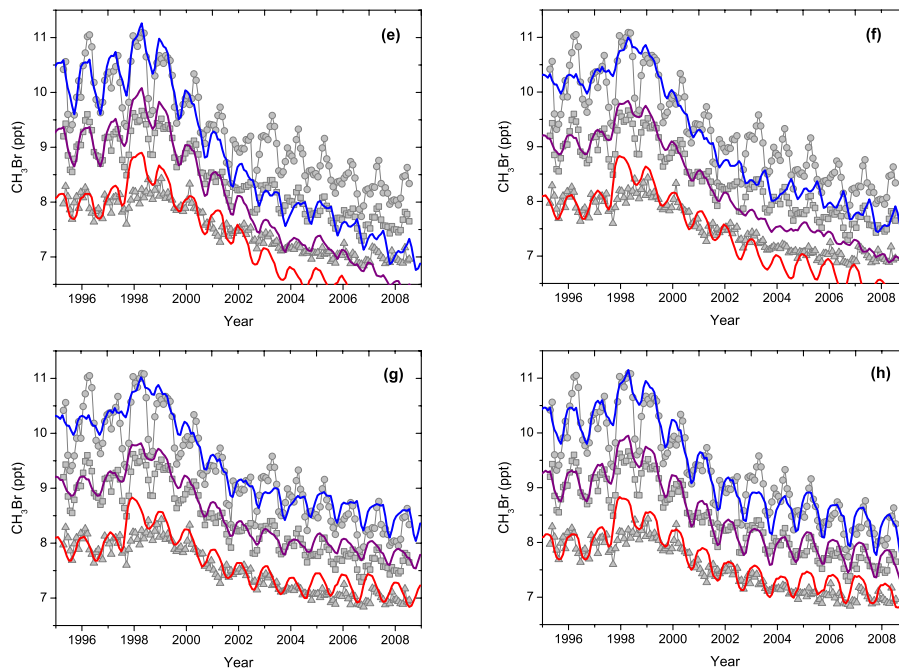
Back

Close

Full Screen / Esc

Printer-friendly Version

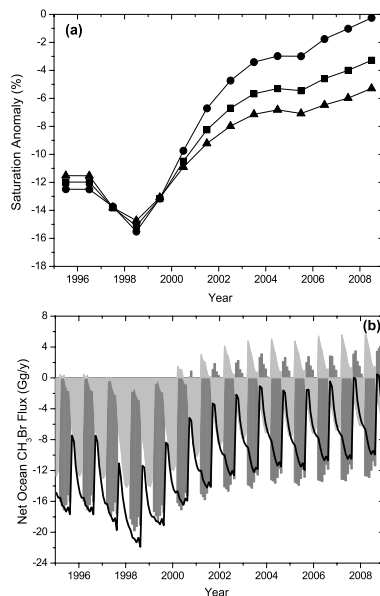
Interactive Discussion

**Recent trends in  
atmospheric methyl  
bromide**S. A. Yvon-Lewis and  
E. S. Saltzman**Fig. 4** Continued.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



## Recent trends in atmospheric methyl bromide

S. A. Yvon-Lewis and  
E. S. Saltzman



**Fig. 5.** From scenario 8, model predicted **(a)** annual average saturation anomaly (difference from equilibrium) of  $\text{CH}_3\text{Br}$  in the ocean for the Southern Hemisphere ( $\blacktriangle$ ), the Northern Hemisphere ( $\bullet$ ), and the globe ( $\blacksquare$ ) and **(b)** net ocean fluxes for the Southern Hemisphere (light gray), Northern Hemisphere (dark gray), and the globe ( $-$ ).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)