AN INVESTIGATION OF THE ATMOSPHERIC SOURCES AND SINKS OF METHYL BROMIDE

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Abstract. Methyl Bromide (CH_3Br) is a ubiquitous component of the atmosphere and has been implicated as an important player in the depletion of stratospheric ozone. Atmospheric CH_3Br abundances, interhemispheric gradients, oceanic concentrations, man-made emissions, and removal processes have been analyzed and interpreted with the help of a simple box model and a 2-D global photochemical model. Its calculated atmospheric lifetime (T) of 1.7-1.9 years, based on reaction with OH radicals, is consistent with a global source of 90-110 Gg (10^9 g)/year. Consequences of a much shorter lifetime of 1.2 years, due to possible deposition/hydrolysis losses, are also considered. Available data are used to estimate a CH_3Br source that is 35% (20-50%) man-made and 65% (80-50%) natural. Oceans are substantially supersaturated and provide the most important natural source of ≈ 60 (40-80) Gg/year. Within the oceans 200-300 Gg/year of CH_3Br may be produced. Indirect emissions from automobile exhaust and biomass burning can not be well quantified (1-10 Gg/year). A global trend of 0.1-0.2 ppt/year is predicted. Model results show significant vertical and seasonal variations in the atmospheric abundances and interhemispheric gradients of CH_3Br . Substantial uncertainties in calibrations, source estimates, and deposition processes are present.

Introduction

Methyl Bromide (CH_3Br) is a ubiquitous component of the atmosphere and has been measured in polluted and remote environments. It has been used as a soil, grain and space fumigant for a number of decades and has significant man-made sources [CIS, 1992; CMA, 1992]. Limited data show that ocean waters are supersaturated with methyl halides and a large marine source of CH_3Br is present [Lovelock, 1975; Singh et al., 1983]. Singh [1984] concluded that CH_3Br is largely natural in origin with a small anthropogenic component. More recently, it has been proposed that it may be predominately of man-made origin [Penkett et al., 1985; WMO, 1989]. This distinction is important because bromine is nearly 40 (30-120) times more effective than chlorine in stratospheric ozone destruction [Wofsy et al., 1975; Yung et al., 1980; McElroy et al., 1986; WMO, 1991; Mellouki et al., 1992]. It has been calculated that a 10% reduction in the emissions of CH_3Br is equivalent to a 2 to 3 year acceleration in the phase out of CFCs under the Montreal protocol [WMO, 1991]. Proposals have been put forth to regulate and eliminate the use of CH_3Br . In light of these considerations a better understanding of its sources and sinks is warranted.

Atmospheric and Oceanic Measurements of Methyl Bromide

Atmospheric measurements. A number of studies in recent years have provided atmospheric measurements of CH_3Br . Data from some 15 cities in the U. S. show elevated concentrations (130 ± 110 ppt). Exceptionally high values (several ppb) at some

California sites have also been observed [Singh et al., 1977; 1992]. In remote marine atmospheres CH_3Br mixing ratios of 5-30 ppt have typically been reported. Data collected from coastal Pacific sites do not show any discernible annual cycle or a long term trend [Singh et al., 1983; Cicerone et al., 1988]. In Table 1 we summarize the mean NH (northern hemisphere) and SH (southern hemisphere) surface air concentrations of CH_3Br measured by several different investigators. No similar free tropospheric data were available in the published literature. A global background concentration of 10-20 ppt, with a preference for the 10-13 ppt range, is present. In addition a north/south concentration gradient is consistently seen. This NH/SH ratio of 1.2-1.4 is largely independent of absolute calibrations and suggests a higher sources in the NH. Larger sinks (OH concentrations) in the SH compared to the NH have been suggested [Singh, 1977; Brenninkmeijer et al., 1992], and may also cause similar gradients. Recent models and methyl chloroform budget calculations do not support large hemispheric differences in the OH field [Prinn et al., 1992]. It is evident from Table 1 that measured CH_3Br concentrations differ to within a factor of two. Although part of this could be due to the presence of local sources and seasonal effects, much of it is due to uncertainties in calibration standards.

Oceanic measurements. The only available sea water data are from the eastern Pacific [Singh et al., 1983]. These show that (i) sea water is significantly supersaturated (mean 250%; median 180%); (ii) mean sea water concentrations in the NH (1.1 ± 0.8 ng/L) and SH (1.2 ± 0.7 ng/L) are nearly equal and (iii) sea water concentrations of CH_3Br and methyl chloride (CH_3Cl) are significantly correlated ($R=0.85$) pointing to a common oceanic source. In addition CH_3Cl , which is known to have a very small man-made sources, was nearly uniformly distributed in the two hemispheres. This implies that in the absence of man-made sources CH_3Br would also be similarly distributed. These data can suggest, but not prove, that the marine sources of CH_3Br in the two hemispheres may be more or less equal and the observed

Table 1: Mean methyl bromide mixing ratios (ppt) in the surface air of the northern and southern hemispheres

NH	SH	NH/SH	Data Year	Platform (Region)	Ref.
26	20	1.30 (1.15)*	1981-1982 (December)	Ship (Pacific)	(1)
15	11	1.40 (1.25)	1982-1983 (November)	Ship (Atlantic)	(2)
11	10	1.15 (1.10)	1985-1987 (Ann. Avg.)	Coastal (Pacific)	(3)
11	8	1.35 (1.25)	1983-1992 (Ann. Avg.)	Coastal (Pacific)	(4)
10	8	1.20 (1.15)	1988/1991 (December)	Coastal (Pacific)	(5)
14	-	-	1991/1992 (April)	Aircraft	(6)

* Annual mean hemispheric ratio, obtained by correcting surface ratio with the 2-D model. (1) Singh et al. [1983], (2) Penkett et al. [1985], (3) Cicerone et al. [1988], (4) R. A. Rasmussen-private comm., (5) F.S. Rowland- private comm., (6) L. Heidt- private comm.

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NH₃/SH gradients are largely due to man-made sources in the NH.

Mechanisms by which CH₃Br (or other methyl halides) may be produced in sea water are not well known. Zafiriou [1975] has proposed that Cl⁻ and Br⁻ ions in sea water may react with methyl iodide (CH₃I), produced by marine algae, to rapidly (several days) form CH₃Br and CH₃Cl (CH₃I+Br⁻ → CH₃Br+I⁻). Singh et al. [1983] were unable to find any correlation between sea water concentrations of CH₃I with those of CH₃Br or CH₃Cl, suggesting that this was probably not a dominant mechanism. Another possibility [White, 1982] may be either direct emission from organisms or by reaction between dimethyl sulfonium and halide ions in sea water [(CH₃)₂S⁺+R+Br⁻ → CH₃Br+CH₃SR].

Sources of Methyl Bromide

Man-made sources. Singh [1984] estimated that 15-20 Gg/year of CH₃Br is released from man-made sources alone. Recently, the Methyl Bromide Industrial Panel, an industry group, has prepared a new inventory of CH₃Br production, sales and use [CMA, 1992]. Table 2 shows the aggregated global sales of CH₃Br from 1965-1990. The 1965 and 1975 sales are obtained from CIS [1992]. An average annual growth rate of about 5% is indicated. An exponential function provides a best fit (R=0.99) to these data [Sales (Gg/year)=0.8646 exp(0.0477 (year-1900))]. Approximately 93% of the sales occur in the NH and 7% in the SH.

Nearly 95% of all CH₃Br sold is used in various soil, grain and space fumigation applications with a very small fraction (~5%) finding use as a chemical intermediate. Fumigation usages involves pre-planting (~75%), post harvesting (~15%) and structural (~5%) applications. CH₃Br is decomposed in soils at a rate of 5-10%/day via processes of hydrolysis and demethylation [Gentile et al., 1989]. The biggest use is pre-planting fumigation and here the actual fraction emitted is dependent on soil variables such as organic carbon, moisture, pH etc. and the manner of application. Numerical models of soil vapor migration, that take into account current use practices and typical soil types, have been used to calculate that 42±10% (30-60%) of the CH₃Br applied may be ultimately released to the atmosphere [Reible, D., private comm., 1992]. In space and grain fumigation applications a higher fraction (~80%) is probably released to the atmosphere. The current best estimate, based on studies by the Methyl Bromide Global Coalition, is that approximately 50% of CH₃Br sold is emitted to the atmosphere.

Indirect emissions of CH₃Br are also possible. There are preliminary indications that exhaust from automobiles, especially burning leaded gasoline, may be a source [Harsch and Rasmussen, 1977]. This is consistent with the high CH₃Br levels that have been reported in urban air. It is further possible that biomass burning, which is known to produce important quantities of CH₃Cl, may also be a source. No biomass burning data are available but, based on Br/Cl availability (Br/Cl<0.005), this source can be estimated to be quite small (<3 Gg/year). It is possible that such indirect sources could add up to a global emission of 1-10 Gg/year.

Table 2: Annual world-wide sales and emissions (Gg;10⁹ g) of man-made (MM) methyl bromide from 1965-1990

Year	65	75	84	85	86	87	88	89	90
Sales*	20	30	46	48	50	56	60	62	66
Emissions*	10	15	23	24	25	28	30	31	33
% MM*	14	20	28	29	29	32	33	34	36

*Sales(Gg/year)=0.8646exp[0.0477(year-1900)];R=0.99;
Emissions=50% of sales; Natural marine source=60 Gg/year.

Oceanic sources. Using the film model of Liss and Slater [1974] and based on the data collected in and over the eastern Pacific Ocean, Singh et al. [1983] calculated an average oceanic supersaturation of 180% (median) to 250% (mean) corresponding to a flux of 65-90 ng cm⁻² year⁻¹. Extrapolated to global oceans, this flux corresponds to a marine source of nearly 220-300 Gg/year. More recent information permits a reevaluation of these results. The first possible correction is due to disagreement in calibration standards. As can be seen from Table 1, all subsequent measurements are lower by a factor of about two. A second difficulty is in extrapolating the eastern Pacific data to global oceans. Productivity maps show that the sampled area is substantially (two to four times) more productive than the open Pacific Ocean [Koblentz-Mishke et al., 1970]. Another way to accomplish this extrapolation is to normalize CH₃Br flux to that of CH₃Cl which has been better studied and appear to have a common source with CH₃Br. CH₃Cl flux for this region is roughly double the global average flux. Scaling by normalizing to either productivity or CH₃Cl, suggests that the mean global CH₃Br flux is about half to one third that of the eastern Pacific Ocean. These considerations imply a global oceanic source of 40-80 Gg/year. The analogy with CH₃Cl also allows us to assume that this source is nearly equally distributed in the two hemispheres.

CH₃Br hydrolyzes in neutral water at a rate of 2%/day at 22°C and 3%/day at 30°C [Mabe and Mill, 1978]. It may also be decomposed in sea water by reaction with Cl⁻ ions at a comparable or faster rate [Elliot, 1985]. Assuming a removal rate of 4-6%/day within the mixed layer (~70 meters) of the world oceans the data of Singh et al. [1983], as interpreted above, can be used to calculate a CH₃Br production of 200-300 Gg/year within the oceans.

Data Analysis and Discussion

In this section we use available data to evaluate the lifetime and budget of CH₃Br. Data are first analyzed with the help of a 2-box model and further refined by using a global 2-D photochemical model [Kanakidou et al., 1991].

Methyl bromide lifetime. The reaction with hydroxyl radical is thought to be the principal atmospheric removal mechanism for CH₃Br. Recently Mellouki et al. [1992] have measured this OH rate constant (k_{OH}=2.35×10⁻¹² exp (-1300/T)) and calculate a CH₃Br lifetime (T) of 1.7 years based on this process alone. The 2-D model used in this study calculates a T of 1.9 years and a methyl chloroform lifetime of 6.2 years in reasonable agreement with its budget estimates [Talukdar et al., 1992; Prinn et al., 1992]. Because CH₃Br can hydrolyze as well as react with Cl⁻ ions and organic carbon, its loss via deposition/hydrolysis is possible. If oceans were substantially undersaturated, the dissolution of CH₃Br in the sea water followed by hydrolysis would provide an important sink with an atmospheric turn over rate of the order of 3 years. This corresponds to a uniform deposition velocity of 0.01 cm/s. No direct measurements of deposition have been made. Although Singh et al. [1983] found sea water supersaturation at all times, the possibility that parts of the oceans are under-saturated and are actual sinks can not be ruled out. Tropospheric photolysis and rainout/washout are calculated to be negligible sinks. Stratospheric loss can provide a turn over rate of about 30 years [WMO, 1991]. For subsequent calculations, we assume a CH₃Br lifetime of 1.7-1.9 years. Consequences of a much shorter lifetime of 1.2 years, possibly resulting from deposition/hydrolysis losses, are also discussed.

Two-box model. As a start a 2-box model [Singh, 1977] is used to describe the atmospheric distribution of CH₃Br.

$$dC_n/dt = F_n - (C_n - C_s)/t_e - C_n/T \quad (1)$$

$$dC_s/dt = F_s + (C_n - C_s)/t_e - C_s/T \quad (2)$$

$$F_n = 0.5 N_0 + (1-f) m e^{pt}$$

$$F_s = 0.5 N_0 + f m e^{pt}$$

Where C_N and C_S are mean (annual averages) CH_3Br atmospheric loadings in the NH and SH, t_e is the mean interhemispheric exchange rate, f is the fraction of the man-made source emitted in the SH, and p is a measure of temporal growth rate. By analogy with CH_3Cl , the total source in each hemisphere (F_N and F_S) is assumed to be made of an equal natural ($0.5 N_0$) and a man-made component fit to an exponential function. A steady state solution to the above equations leads to the following result.

$$(C_N - 0.5TN_0)/(C_S - 0.5TN_0) = t_e [p + 1 + (1-f)/T] \equiv \theta \quad (3)$$

$$TN_0/(C_N + C_S) = 2(\theta - C_N/C_S)/[(\theta - 1)(1 + C_N/C_S)] \quad (4)$$

Equation 4 is arranged to provide the fraction that is of natural origin. A t_e of 1.1-1.2 years, T of 1.7-1.9 years, p of 0.048 and f of 0.07 represent typical mean values. If one assumes that the natural source does not exist ($N_0=0$) then a C_N/C_S of 1.6-1.8 can be calculated from equation (3). (A ratio of 1.9 can be derived with a T of 1.2 years). The 2-D model also confirms the large north-south gradients associated with an exclusive man-made source set to 150% of sales (Figure 1a-I and I'). Sensitivity studies with a much smaller source (set to 50% of sales) showed that this gradient was smaller by only about 10%. This is significantly greater than the measured concentration ratios, corrected for season and altitude effects, of 1.1-1.25 (Table 1). This comparison supports the notion that significant sources of CH_3Br in the SH (presumably natural) must exist.

Using the mean C_N/C_S value of 1.1-1.25 (Table 1), equations 3 and 4 can be used to calculate a natural (or man-made) fraction that best fits the observations. For a T of 1.7-1.9 years, an average man-made fraction of about 35% (20-50%) can be calculated. An assumed T of 1.2 years results in a calculated

Fig. 1: Model calculated north-south ratios of CH_3Br abundances. Open and solid symbols are for deposition velocity (V_d in cm/s) of 0.0 ($T=1.9$ years) and 0.0125 ($T=1.2$ years), respectively. (a) (I) shows annual and column averaged (0-10 km) ratios with exclusive man-made source of 150% of sales, (I') is same as (I) but for surface level, (II) and (II') use emission source as described in Table 2; (b) surface ratios as a function of season, source same as in (II) and Symbols are Circles-March 30, Squares-June 30, Diamonds-September 30, Triangles-December 30.

Fig. 2: Model calculated mixing ratios of CH_3Br in the lower and upper troposphere for four seasons in 1985. Emission source from Table 2 and $V_d=0.0$. Symbols as in Fig. 1(b).

man-made fraction of 27% (20-35%). A global mean concentration of 12 ppt corresponds to a total CH_3Br atmospheric loading of 177 Gg and a source of 93 Gg/year ($T=1.9$ years), 61 natural and 32 man-made. However, if the lifetime of CH_3Br is as short as 1.2 year, then a much larger source of 147 Gg/year, 107 natural and 40 man-made, is also not entirely outside the bound of possibilities.

2-D model calculations. The above 2-box model provides a preliminary picture of the source distribution of CH_3Br . A 2-D model of the global atmosphere [Kanakidou et al., 1991] is used to more accurately assess its global distribution and budget. This is especially important since the data are nearly all from the surface and the lifetime of CH_3Br is short enough to result in seasonal and vertical gradients. This 2-D model produces an asymmetry in the OH field with NH values about 20% larger than SH. This has the net effect of under-predicting north-south ratios by about 5%. Loss of CH_3Br by mechanisms other than OH removal are poorly known but possible. Simulations were

Fig. 3: Predicted atmospheric growth of CH_3Br at 45°N and 45°S from 1970 to 1990. Annual and column averaged (0-10 km) mixing ratios. Emission source from Table 2.

performed assuming no deposition ($V_d=0.0$ cm/s) and a deposition velocity of 0.0125 cm/s over the oceans to simulate the hydrolysis loss rate. The global mean lifetime of CH_3Br as calculated by this model is 1.9 years ($V_d=0.0$ cm/s) and 1.2 years ($V_d=0.0125$ cm/s).

Model runs were performed from 1965 to 1990 to study both the distribution and trends of CH_3Br for the case of exclusive man-made sources and a combination of man-made and natural sources (Table 2). The man-made source of CH_3Br was distributed by 10° latitude belts based on the sales data [CMA, 1992]. This amounted to 93% emission in the NH and 7% in the SH. Nearly 80% of the global emissions occur within $30\text{--}60^\circ\text{N}$. An oceanic source of 60 Gg/year distributed equally in both hemispheres was used. Figure 2 shows the 1985 atmospheric distribution of CH_3Br as a function of latitude and height for four seasons and for the case of no deposition ($T=1.9$ years). As expected, both seasonal and vertical gradients are present but high precision measurements are required to identify these. Simulations with an oceanic deposition rate of 0.0125 cm/s ($T=1.2$ years) reduced the predicted atmospheric concentrations by about 40% and increased the hemispheric ratios by about 15% (Figure 1). Figures 1 and 2 clearly show that the north/south ratio is a strong function of altitude and season. In fact in a select season (e. g. NH winter) interhemispheric gradients could be created by the asymmetry in the removal processes alone. Unfortunately very little seasonal data are available and no seasonal cycles have been observed. The predicted north/south gradient of CH_3Br , based on the source described in Table 2, is in reasonable agreement with observations.

Figure 3 shows the annual mean mixing ratios (0 to 10 km average) at 45°N and 45°S as a function of time with and without deposition. A quadratic fit accurately describes this trend. From 1970 to 1990 the growth rate increases from 0.10 to 0.22 ppt/year at 45°N and 0.08 to 0.13 ppt/year at 45°S ($V_d=0.0$ cm/s). For the decade of the 80's, a mean global trend of about 0.1-0.2 ppt/year is predicted by the 2-D model.

Conclusions

CH_3Br is a globally distributed brominated organic chemical with substantial natural and man-made sources. A total global source of about 90 Gg/year is likely to be present. Present data can be interpreted to suggest that two thirds of this source is natural and one third is man-made. An important natural source is in the oceans which appear to be substantially supersaturated. Mechanisms by which CH_3Br (or other methyl halides) may be produced in sea water are not well known. The atmospheric lifetime of CH_3Br is 1-2 years with OH radical and deposition processes as the likely controlling factors. Uncertainties in its atmospheric abundance, man-made and natural emissions, and physical removal processes are considerable. Future studies are crucially needed in all of these areas to better quantify the sources and sinks of CH_3Br .

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