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Constraints on the global sources of methane and an analysis of recent budgets

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ABSTRACT

Recent observations show that the concentration of methane before being affected by human activities was about 650 ppbv; now it is about 1690 ppbv and has increased at an average rate of 16.5 ppbv/yr over the last decade. The present lifetime of methane is 8–12 years. These facts produce constraints on the global budgets of methane and particularly on the anthropogenic fraction (ratio of anthropogenic to total emission rates). Taking into account the possibilities that the lifetime of methane has gotten longer over the last century because of a possible decline in the natural sinks and that emissions from natural sources have also changed, we show that the anthropogenic fraction should be between 40%–70%, and the total present emissions should be between 420–620 Tg/yr. Budgets that do not meet these conditions would be inconsistent with one or more of the observations mentioned above. When we analyzed 11 budgets of methane published over the last decade, we found that only 2 meet these conditions.

1. Introduction

In the last decade, some 11 complete budgets of methane have been published, most of them after it was discovered that methane was increasing in the atmosphere (Rasmussen and Khalil, 1981; Blake et al., 1982; Fraser et al., 1981; Khalil and Rasmussen, 1983). The increasing concentrations are thought to be caused by anthropogenic processes related to the production of more and more food and energy for a growing population and possibly by a reduction in the oxidizing capacity of earth's atmosphere and soils to remove methane (Khalil and Rasmussen, 1985; Levine et al., 1985; Thompson and Cicerone, 1986; Steudler et al., 1989). The many proposed budgets of methane are very different from each other, and not all can be right even when the extremes of the stated uncertainties are taken into account. This paper has 2 goals: (1) to develop a criterion for a budget to be compatible with the observed features of the global cycle of methane and (2) to analyze the 11 published budgets to see which satisfy the criterion. It turns out that only two can explain the combination of present observations.

2. A criterion for global methane budgets

2.1. The observed features of the global cycle of methane

The criterion we will develop is a range of values for the ratio of present annual anthropogenic emissions to the total emissions. The following observations are the foundation for the criterion.

(i) Based on global atmospheric measurements, the present (late 1988 to early 1989) average concentration of tropospheric methane is $C_p = 1690$ ppbv or about 4600 Tg in the atmosphere (Blake and Rowland, 1988; Khalil and Rasmussen, 1989). About 2.72 Tg in the earth's atmosphere are equivalent to 1 ppbv in the troposphere. This factor includes a correction for the reduced concentration of methane in the stratosphere (Khalil and Rasmussen, 1985).

(ii) The concentration before human activities affected the global cycle of methane was about $C_o = 650$ ppbv or 1770 Tg (Khalil and Rasmussen, 1982; Craig and Chou, 1982; Rasmussen and Khalil, 1984; Stauffer et al., 1985; Raynaud et al., 1988). This value is obtained from measurements of CH_4 in air trapped in polar ice.

(iii) The average rate of increase, also determined from global measurements, has been $dC/dt = 16.5$ ppbv/yr or 45 Tg/yr over the last decade (Khalil and Rasmussen, 1986; Blake and Rowland, 1988; Rasmussen and Khalil, 1986; Khalil and Rasmussen, 1989).

(iv) The present lifetime of methane is between 8 and 12 years. This value is based mostly on the reaction of CH_4 with OH radicals. The estimates of the effective average OH concentrations are based on the global budget of the man-made trace gas methylchloroform (CH_3CCl_3) (see, for example, Khalil and Rasmussen, 1984). A lifetime in this range is generally supported by most recent estimates including those in the papers of Khalil and Rasmussen (1983, 1984, 1985) and Cicerone and Oremland (1988).

We believe that these four conditions are well established by direct observations of the global cycle of methane and have been verified by several independent observers. While there are uncertainties in these observations, they are small compared to the uncertainties in the budgets we are about to describe and have no practical effect on the conclusions.

In addition to these observations, there are 3 other factors that must be included in developing a global budget of methane.

(v) It is likely that during the last decade the total anthropogenic emissions have increased: $dS_a/dt \geqslant 0$.

(vi) The natural emissions may have changed over the last century or two: $dS_n/dt \neq 0$.

(vii) The sinks of CH_4 may have weakened over the last century and particularly in recent decades.

Anthropogenic emissions are taken to be: cattle, rice agriculture, biomass burning, landfills, coal mining, production of natural gas, and urban areas. Natural sources included in the published budgets are wetlands, oceans, freshwaters, tundra, termites, and tropical forests.

There are good reasons to believe that anthropogenic emissions, particularly domestic cattle, rice paddies, and natural gas use, have increased tremendously over the last century, but whether there have been significant increases over the last decade is not as well known. FAO figures suggest that cattle populations and area of rice fields have not increased much over the last decade compared to increasing trends in earlier years (FAO, 1977–1986).

Reaction with OH radicals is believed to be the principal mechanism for removing CH_4 from the atmosphere, although some is removed by soils and stratospheric processes.

There has been much speculation as to whether OH concentrations may have declined in recent times compared to 100–200 years ago because of increasing global levels of CO and CH_4 from anthropogenic sources (Sze, 1977; Chameides et al., 1977; Khalil and Rasmussen, 1985; Levine et al., 1985; Thompson and Cicerone, 1986; Isaksen and Hov, 1987). Some of the increase of CH_4 could therefore be caused by a net decline of the OH sink. While there are reasons to believe that OH should be declining, there is no compelling evidence to support this hypothesis. Recently Steudler et al. (1989) have suggested that the capacity of soils to remove CH_4 may also be declining because of increased deposition of nitrogen compounds. In our model we cannot distinguish the effect of changes in individual sinks; we can only assess the effect of a net change in the removal rate. The present understanding of the methane cycle tends to support a generally declining removal rate, although it is not clear whether this change is insignificantly small or a substantial ($\geqslant 10\%$) contributor to the trend of methane.

Finally, it is possible that even the natural sources have changed. With increasing population, some natural wetlands have been drained and the numbers of wild ruminants may have declined. These possibilities would tend to decrease natural emissions. On the other hand, since global temperatures have increased slightly, the emission rates from the remaining wetlands may have increased. Whether these and other processes have led to a net change in the natural emission rates is essentially another unknown factor.

2.2. The mass balance model

The global mass balance of methane is expressed by eqs. (1), (2):

$$dC(t)/dt = S(t) - C(t)/\tau(t), \quad (1)$$

$$S(t) = S_n(t) + S_a(t), \quad (2)$$

where C is the globally averaged concentration of methane (ppbv), S are the emissions (ppbv/yr), and τ is the lifetime (years). The subscript "n" is for natural and "a" is for anthropogenic. We define δ, ε, and λ as the following ratios:

$$\delta = \tau_o/\tau_p, \quad (3)$$

$$\varepsilon = S_{np}/S_{no}, \quad (4)$$

$$\lambda = \delta/\varepsilon. \quad (5)$$

In eqs. (3), (4) the subscript "p" is to indicate present conditions, and "o" is for the conditions in the atmosphere of 100 or more years ago.

Eqs. (1)–(5) result in the following expressions for the present anthropogenic and natural emission rates:

$$S_p = \beta + C_p/\tau_p, \quad (6)$$

$$S_{pn} = \varepsilon C_o/(\delta\tau_p), \quad (7)$$

$$S_{pa} = S_p - S_{pn}, \quad (8)$$

where β is $(dC/dt)_p$, the present rate of increase of concentrations. We define the present anthropogenic fraction of emissions as:

$$F = S_{pa}/S_p = 1 - S_{pn}/S_p. \quad (9)$$

Eqs. (3)–(8) are used to write F as:

$$F(\tau_p, \lambda) = 1 - C_o/[\lambda(\alpha + \beta\tau_p)]. \quad (10)$$

2.3. The criterion

To establish the limits of the anthropogenic fraction consistent with conditions (i)–(vii) discussed in Subsection 2.1, we consider F as a function of present lifetime of methane (τ_p) and the ratio (λ) of effect on lifetime from a slowdown in the removal rate (δ) and changes of natural sources (ε). We then define a region of possible values of F, in the space of F (vertical axis) and τ_p (horizontal axis), that are consistent with the observations stated in Subsection (2.1). The horizontal extent of this region is given by the expected range of present lifetime (8–12 years) as stated in condition (iv). The vertical extent of the region is determined from eq. (10) for the observed values of the pre-industrial levels, present concentrations, and trends (conditions (i)–(iii) in Subsection (2.1)) and for various possible values of λ, which describes conditions (v)–(vii).

The 2 possibilities, a general reduction in the rate of removal and a decline of natural emissions, have opposite effects on the anthropogenic fraction F. For instance, if the present lifetime is 20% more than the past lifetime ($\delta = 0.8$) and the natural emissions are also only 80% of the past emissions, then $\lambda = 1$ and the resulting F is the same as it would be if there were no changes in OH, soil sink, and natural sources. Therefore, we set the limits on λ between 1.1 and 0.6. These limits include the possibility of a combined change of OH and other sinks of up to 40% ($\delta = 0.6$) during the last century without any change of natural emissions ($\varepsilon = 1$). This limit is consistent with the maximum possible OH depletion estimated by Khalil and Rasmussen (1985), Levine et al. (1985), and Thompson and Cicerone (1986). The limit of 1.1 is to include the possibility that there has been little change in OH or the soil sink but natural emissions have decreased. Because the slowing down of removal processes and reduced natural emissions have opposite effects on F, in our opinion the range for λ between 0.6 and 1.1 includes all combinations of changes in OH, soil sink, and S_n consistent with the present understanding of the CH_4 cycle.

The region R, for the anthropogenic fraction F consistent with the conditions (i)–(vii), is defined by:

$$R = \{(F(\tau_p, \lambda), \tau_p) | 8 \leqslant \tau_p \text{ (years)} \leqslant 12; \; 0.6 \leqslant \lambda \leqslant 1.1\}. \quad (11)$$

The criterion a budget must satisfy is that the anthropogenic fraction must lie in the region R defined by eq. (11). If a budget does not satisfy this criterion, then it must be incompatible with one or more of the conditions (i)–(vii) stated in Subsection 2.1. Because the conditions (i)–(vii) are broad, satisfying the criterion in eq. (11) is a necessary but not a sufficient condition for a budget to be correct. Meeting the criterion says nothing about whether the budget is accurate in allocating the total natural and the total anthropogenic emissions among the various individual sources.

Table 1. *Estimates of methane emissions from various anthropogenic and natural sources: 1978–1988*

	Ehhalt and Schmitt (1978)		Donahue (1979)		Shep- pard et al. (1982)	Crutzen (1983)		Khalil and Ras- mussen (1983)	Blake (1984)		Seiler (1984)		Crutzen (1985)		Seiler (1986)		Bingemer and Crutzen (1987)		Cicerone and Oremland (1988)	
ruminants	100–220	160	100–220	160	90	60	60	120	70–160	115	70–100	85	60	60	70–100	85	70–80	75	65–100	80
rice paddy fields	280	280	140–280	210	39	30–60	45	95	149–189	165	30–75	53	120–200	160	70–170	120	18–91	54	60–170	110
biomass burning					60	30–110	70	25	25–110	68	50–100	75	20–70	45	55–100	78	30–100	65	50–100	55
landfills																	30–70	50	30–70	40
coal mining	8–28	18	16–50	33							30	30	34	34	35	35	35	35	25–45	35
natural gas flaring					50	20	20	40			20–30	25	33	33	30–40	35	0–35	18	25–50	45
automobiles	1	1													1–2	2				
other anthropogenic	7–21	14	110–210	160	100			40	62–100	81										
d*S*	0–50	25	0–50	25	18	0–25	13	20	0–25	13	0–30	15			0–10	5				
swamps and marshes	190–300	245	200–300	250	39	30–220	25	150	120–190	155	15–60	38	70–90	80	25–70	48	26–137	82	100–200	115
lakes	1–25	13			35			10	13	13	1–7	4			15–35	25			1–25	5
oceans	1–17	9			30			13	5–21	13									5–20	10
tropical forests					767				60–400	230										
tundra	0–3	2	3–50	27				12												
other natural						150	150	48			5–15	10					0–30	15	10–100	40
total anthropogenic	396–550	473	366–760	563	339	140–250	195	320	297–560	429	200–335	268	267–397	332	261–447	354	183–411	297	255–535	365
total natural	192–345	269	203–350	277	871	180–370	275	233	198–624	411	21–82	52	70–90	80	40–105	73	26–167	97	116–345	170
total	588–895	741	569–1110	840	1210	320–620	470	553	495–1184	840	221–417	319	337–487	412	301–552	427	209–578	394	371–880	535
F	67–61	64	64–68	67	28	44–40	41	58	60–47	51	90–80	84	79–82	81	87–81	83	87–71	75	69–61	68

Notes: The first column under each budget is the range of estimated emissions, the second column is the central value. *F* is the ratio of anthropogenic emission to total emissions.

3. Analysis of budgets

The 11 budgets we analyzed are given in Table 1 (with some rounding of emission rates from individual sources). An earlier budget of Ehhalt (1974) was not analyzed separately since it is similar to the budget of Ehhalt and Schmidt (1978). The budgets titled Crutzen—1985 and Seiler—1986 are taken from Bolle et al. (1986), and the budget of Crutzen (1983) is taken from Warneck (1988). In their budget, Bingemer and Crutzen (1987) did not separate the emissions from rice fields and wetlands. We estimated the apportionment between these sources as in Table 1.

The anthropogenic emissions in these budgets may be increased by between 0–5 Tg/yr^2 for the period when the budget was put together and the present. This would represent the range of possible increases in anthropogenic emissions up to 1988 so that all budgets can be referenced to that year. This term is included in Table 1 as dS and is at most 50 Tg/yr for the older budgets. It does not change the conclusions and is not included in the totals given in Table 1 or in further calculations.

From the budgets in Table 1, we can estimate the central value and the uncertainty of the anthropogenic fraction (F) and see whether it is compatible with our criterion (eq. (11)).

Most budgets include a range of emission estimates from each source. Often the estimated minimum and maximum of the total emissions are taken to be the sum of the minima and the sum of the maxima of emissions from individual sources. We believe that the ranges of natural, anthropogenic, and total emissions calculated by this process are unrealistically large (see Khalil, 1989, for a statistical solution to this problem). Nonetheless, to provide the broadest possible range of values for the anthropogenic fraction compatible with each published budget, we have used this method of adding minima and adding the maxima to estimate the minimum and maximum respectively of the range of total natural or total anthropogenic emissions. We calculated the percent of anthropogenic emissions and uncertainties as the following limits:

$$F_1 = 100\%\ S_{aL}/(S_{aL} + S_{nU}), \tag{12a}$$

$$F_2 = 100\%\ S_{aU}/(S_{aL} + S_{nU}), \tag{12b}$$

$$F_3 = 100\%\ S_{aL}/(S_{aL} + S_{nL}), \tag{12c}$$

$$F_4 = 100\%\ S_{aU}/(S_{aU} + S_{aU}), \tag{12d}$$

$$F_m = 100\%\ (S_{aL} + S_{aU})/(S_{aL} + S_{aU} + S_{nL} + S_{nU}), \tag{12e}$$

where (S_{aL}, S_{aU}) are the lower and upper limits of the total anthropogenic emissions and (S_{nL}, S_{nU}) are the limits of the total natural sources.

When plotted on a graph of present lifetime of methane and F, these budgets and their uncertainties appear as polygons in which the central or "best" estimate (F_m) for each budget is at the intersection of the two diagonals.

The region defined by eq. (11) is plotted in Fig. 1 as the "target" for consistency with the observed features of the global methane cycle. The effects of OH depletion, a changing soil sink, and changing natural emissions are represented by λ and are plotted in increments of 0.1 between $\lambda = 0.6$ to 1.1.

We put the 11 budgets in three classes. In Fig. 1a, we plotted the 5 budgets that have no overlap with the criterion region R (Sheppard et al., 1982; Seiler, 1984; Bolle, 1986; and Crutzen, 1985). Fig. 1b shows the 5 budgets, which, in their extreme limits, have some overlap with the criterion region (Ehhalt and Schmidt, 1978; Donahue, 1979; Blake, 1984; Bingemer and Crutzen, 1987; Crutzen, 1983 in Warneck, 1988). In Fig. 1c we show the 2 budgets that are substantially within the criterion region with central estimates also within the criterion region (Khalil and Rasmussen, 1983; Cicerone and Oremland, 1988).

4. Discussion and conclusions

The earlier budgets tended to have large total global emissions of 600–1200 Tg/yr (Ehhalt and Schmidt, 1978; Donahue, 1979; Sheppard et al., 1982; Blake, 1984). Our budget (Khalil and Rasmussen, 1983) had the lowest total emissions (550 Tg/yr) compared to the other budgets of the time. The budgets after this time tended to have much smaller total global emissions (350–400 Tg/yr) and a very high fraction of anthropogenic emissions (Seiler, 1986; Seiler, 1984; Crutzen, 1985; Bingemer and Crutzen, 1987). The recent budget of Cicerone and Oremland (1988) brings the total global emissions back up to around 550 Tg/yr.

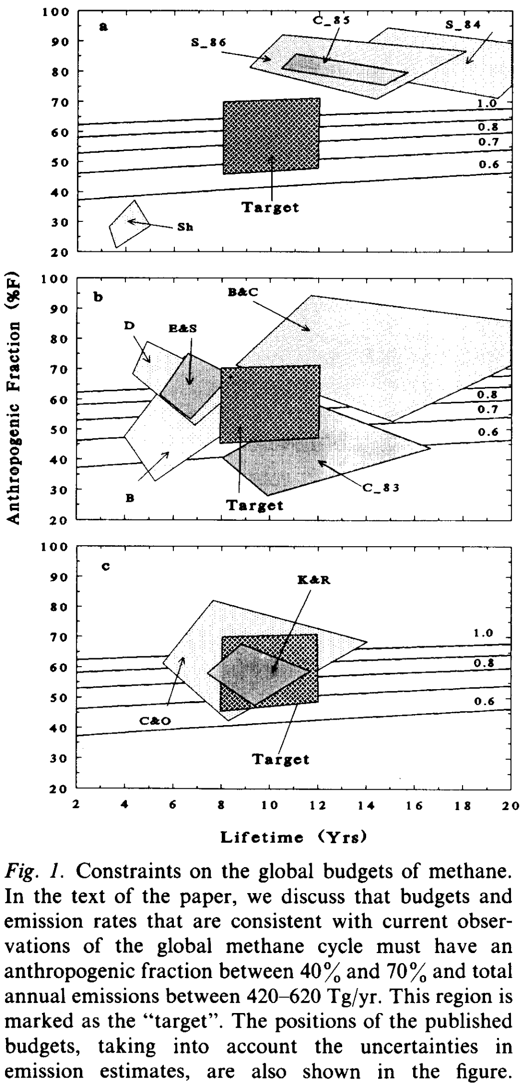

Fig. 1. Constraints on the global budgets of methane. In the text of the paper, we discuss that budgets and emission rates that are consistent with current observations of the global methane cycle must have an anthropogenic fraction between 40% and 70% and total annual emissions between 420–620 Tg/yr. This region is marked as the "target". The positions of the published budgets, taking into account the uncertainties in emission estimates, are also shown in the figure. Budgets in (a) are not consistent with the criterion, budgets in (b) may be made consistent if some extreme values for emission rates are adopted, and in (c) we show the 2 budgets that meet the criterion. The solid lines are for $\delta = 0.6$ to 1.1, which represents the possible decreases of CH_4 sinks or decreases of natural emissions over the last several hundred years.

The budgets in Fig. 1a are generally the extreme examples of the progression from emission rates that are too large to emission rates that are too small. Even if we relax all constraints on the present lifetime of methane, none of these budgets can explain the ice core data.

The budgets in Fig. 1b can be made consistent with the criterion, but only by taking some of the more extreme values of emissions from individual sources. Of all the budgets we studied, the Bingemer and Crutzen (1987) budget has the largest uncertainties in the anthropogenic fraction, which reflects the very large ranges of possible emissions included in this budget. The large uncertainties make this budget very imprecise, so it includes all possibilities, both realistic and unrealistic. Some of these possibilities are bound to be consistent with any criteria we set on the broad features of the budget. The Crutzen (1983) budget meets the criterion in some of its limits primarily because it includes 150 Tg/yr emissions from termites. Since the time when this budget was put together, the consensus has shifted to much lower emission rates from termites of only 5–30 Tg/yr (Seiler et al., 1984; Fraser et al., 1986).

The budget in Blake (1984) includes large ranges of emissions from all sources to account for the budgets that had been published before. Later in his thesis Blake favors the minimum values of emission rates for each range as reported in our Table 1. These minimum values produce a budget with an anthropogenic fraction of about 60%, a lifetime of 10 years, and total emissions around 500 Tg/yr. In that limit the budget meets the criterion of eq. (11).

The only budgets that fit well with the criterion both in their best estimates and in the uncertainties are very similar in their broad features (Khalil and Rasmussen, 1983; Cicerone and Oremland, 1988). In both budgets the total emissions are 550 Tg/yr and the average anthropogenic fraction is 60–70%.

If the uncertainties are interpreted statistically, the ranges of total natural emissions (S_{nL}, S_{nU}) and total anthropogenic (S_{aL}, S_{aU}) emissions are estimated so that there is a 90% chance that emissions lie within these limits, called 90% confidence rectangles (Khalil, 1989). When the 90% confidence rectangles of natural and anthropogenic emissions are taken to represent the uncertainties, all the budgets in Fig. 1b become less consistent with the criterion region. This result shows that the budgets in Fig. 1b can be consistent only if highly improbable limits of the budgets are adopted. Budgets in Fig. 1a shrink further away from the criterion region, and budgets in Fig. 1c retain the substantial overlap with the criterion region. It should be noted that the treatment of uncertainties affects only the size of the polygon representing a budget; it does not affect the central value of F implicit in the budget.

The main conclusion of this analysis is that global budgets of methane should have total emission rates between 420–620 Tg/yr (consistent with a lifetime between 8 and 12 years) and an anthropogenic fraction of between 40–70%. If these conditions are not met, then the budgets will not be able to explain the pre-industrial levels, the present concentrations and trends, the estimates of the present lifetime, or some combination of these observations. If one believes that there has been no substantial change in the rate of removal of methane, or that its effect on the anthropogenic fraction has been compensated by decreasing natural emissions, then the anthropogenic fraction should be near 70%.

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