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Impact of a global warming on biospheric sources of methane and its climatic consequences

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ABSTRACT

Most of atmospheric methane originates by bacterial processes in anaerobic environments within the soil which are found to become more productive with increases in ambient temperature. A warming of climate, due to increasing levels of industrial gases resulting from fossil fuel burning, is thus likely to increase methane abundance within the atmosphere. This may lead to further heating of the atmosphere, since both methane and ozone (which is generated in the troposphere from reactions of methane) have greenhouse effects. We have explored this feedback mechanism using a coupled climate-chemical model of the troposphere, by calculating the impact of the predicted global warming due to increased emissions of carbon dioxide and other industrial gases on the biospheric sources of methane. Although we find this climate feedback to be, by itself, relatively minor, it can produce measurable increases in atmospheric CH_4 concentration, a quantity which should additionally increase as a consequence of increasing anthropogenic emissions of CO and CH_4 itself. It would thus seem useful to carefully monitor future atmospheric CH_4 concentrations.

1. Introduction

The consequences of industrial activity upon global climate have been considered mainly in terms of the carbon dioxide greenhouse effect. Recently it has been suggested that the observed higher concentrations of ozone in the northern hemisphere, which presumably arise because of anthropogenic activity, are responsible for a climatic warming of ~0.2 K (Fishman *et al.*, 1979). Also, calculations of tropospheric chemical changes due to likely future increases in the anthropogenic emissions of CO, CH_4 and NO_x in a coupled climate-chemical model have shown that significant climatic warming can occur from the resulting enhancement of tropospheric methane and ozone (Hameed *et al.*, 1980). Here we consider the natural sources of methane which, according to present estimates, constitute roughly 80% of the total methane input into the atmosphere. Field experiments by Baker-Blocker *et al.* (1977) and King and Wiebe (1978) indicate that methane generation in natural environments increases with temperature. A climatic warming due to the greenhouse effects of CO_2, CH_4 and O_3 may be enhanced because of the resulting amplification of these biospheric methane sources. Moreover, such an additional enhancement of atmospheric CH_4 should make methane a useful trace gas to monitor in the future, as one means of gauging the impact of industrial activity upon the atmosphere and climate.

It is important to point out that because the cycles of CH_4, CO and NO_x are only crudely understood at the present time, the calculations presented here should be regarded as indicative rather than definitive. Our aim is to roughly gauge the relative magnitudes of some effects in the climate-chemical system with model calculations which are consistent with our present understanding of the system. It may be noted that carbon monoxide and nitrogen oxides are also produced by

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natural sources which may show sensitivity to temperature, but we have ignored such effects in the present study. Any changes produced in the coupling between the troposphere and the stratosphere are also not considered.

In the following section we review the sources of atmospheric methane and present a parameterization for the dependence of natural sources of methane upon surface air temperature. We then incorporate this parameterization into a modified version of the climate-chemical model employed by Hameed *et al.* (1980), in order to study the response and feedback of natural sources of methane to a global warming induced by a doubling of atmospheric CO_2. In addition to increased CO_2, we also present two scenarios for the interactive effects of increased industrial emissions of CO, CH_4 and NO_x.

2. Sources of atmospheric methane

Methane is emitted into the atmosphere by natural as well as man-made sources. Most of the natural sources are believed to be located in flooded soils. Based on CH_4 fluxes from incubated soil by Koyama (1963), Ehhalt and Schmidt (1978) have estimated the global natural source of methane as 570–850 million tons per year (MT/yr) of which 100–220 MT/yr is assigned to enteric fermentation in animals. De Haven (1980) has utilized the methane flux measurement of Baker-Blocker *et al.* (1977) in Michigan wetlands, together with geographical locations of flooded soils, to construct a global distribution of such sources. He estimates the total flux to be about 650 MT/yr of which 100 MT/yr is from enteric fermentation. The estimates for methane from animal digestive systems are based on a study by Hutchinson (1948). Another estimate of the methane sources has been presented by Sheppard *et al.* (1982).

Measurements of methane fluxes from wet soils have also been made more recently by King and Wiebe (1978), Cicerone and Shetter (1981) and Harriss and Sebacher (1981). These studies indicate much variability in methane fluxes between different locations.

Methane is also introduced into the atmosphere by man-made sources. A study by Hitchcock and Wechsler (1972), quoted by Ehhalt and Schmidt (1978), gives atmospheric methane sources from coal mining, industrial losses and automobile emissions to be between 140 and 210 MT/yr. Venting and flaring of natural gas is also a source of atmospheric methane (Tiratsoo, 1967) but no estimates of its magnitude are available. Since fossil-fuels are free of ^{14}C, the contribution of such sources to the atmospheric methane can, in principle, be determined by measuring the isotope fraction $^{14}C/^{12}C$ in methane and comparing it with the normal biospheric value. Because of the low abundance of atmospheric methane, and because $^{14}C/^{12}C \sim 10^{-12}$, direct measurements of this ratio have not been made. However, studies on samples of liquified air have been reported (see Ehhalt and Schmidt, 1978). These were performed between 1949 and 1960 and showed $^{14}C/^{12}C$ values in methane to be, on the average, 80% of that in living wood with considerable scatter about this value. A more recent measurement by Volz *et al.* (1981) (page 5165) also gives this value as 80%. We will therefore consider the contribution of biospheric methane sources to be 80% of the total for the present atmosphere. Estimates of the total methane source can also be made by using the mass balance argument in atmospheric chemical models which compute the sinks of atmospheric methane. Such estimates depend on the OH abundance in these models and are generally lower than the direct estimates of Ehhalt and Schmidt (1978) and De Haven (1980). Examples of total methane sources assessed by photochemical models are 147–230 MT/yr (Crutzen and Fishman, 1977), 278 MT/yr (Hameed *et al.*, 1979) and 474 MT/yr (Hameed *et al.*, 1980).

Table 1 lists the latitudinal distribution of the methane sources used in the present study. Column 2 gives the natural source distribution which is based on the compilation by De Haven (1980), while Column 3 gives the anthropogenic sources with the assumption that they constitute 20% of the total source and are distributed latitudinally in the same manner as fossil-fuel consumption. The total magnitude of the source is calculated in the chemical model to obtain tropospheric methane concentrations close to the observed methane abundance in the present atmosphere. These are listed in column 4 and their latitudinal variation is in satisfactory agreement with measurements of Ehhalt and Schmidt (1978) and Heidt *et al.* (1980). The chemical reactivity of methane in the different

Table 1

Latitude zone	Biospheric source MT/yr	Fossil-fuel source MT/yr	Calculated CH_4 concentration	Chemical lifetime of CH_4 in years
90°–80° S	0	0	1.57	69
80°–70° S	0	0	1.57	57
70°–60° S	0	0	1.57	35
60°–50° S	0.2	0	1.57	23
50°–40° S	1.0	0	1.57	12
40°–30° S	8.7	1.6	1.57	8.8
30°–20° S	25.5	0.8	1.58	8.7
20°–10° S	10.2	0	1.58	8.7
10°–0° S	55.9	0.8	1.61	8.1
0°–10° N	70.9	0.8	1.64	6.3
10°–20° N	49.3	1.6	1.65	7.1
20°–30° N	68.7	3.0	1.66	8.5
30°–40° N	18.8	17.4	1.67	11
40°–50° N	11.0	30.9	1.67	19
50°–60° N	9.5	20.7	1.67	33
60°–70° N	0.06	4.3	1.67	71
70°–80° N	0	0.8	1.67	94
80°–90° N	0	0	1.67	109

latitude zones is compared in column 5 where the calculated chemical lifetimes are given.

The microbial generation of methane in flooded soils can be expected to depend on a number of environmental variables. Studies in incubated soils indicate sensitivity of methane producing bacteria to temperature, pH, the redox potential and the abundance of several chemical species including sulfates and nitrates (Mah *et al.*, 1977). Some or all of these factors at a given location may be subject to change with time due to natural or anthropogenic causes. Thus the change in pH of soils due to acid rain over large areas of the industrialized countries may cause a long-term change in the rate of methanogenesis. Studies aimed at detecting such a change have not been reported in the literature. Cicerone and Shetter (1981) have found that addition of nitrogen fertilizer to rice paddies significantly increases the rate of methane emission. The large scale use of chemical fertilizers could therefore constitute a major intervention in the methane cycle. Here we are concerned with the variation of methane emissions from soils with ambient air temperature which has been measured by Baker-Blocker *et al.* (1977) and King and Wiebe (1978). Baker-Blocker *et al.* have reported methane fluxes from two farm ponds and a swamp in Michigan during the period May–September 1975. We have fitted their data with the expression

$$F = 138 \times 10^{-5} \exp(0.117\ T)\ \mathrm{g\,m^{-2}h^{-1}} \qquad (1)$$

where T is in °C. The correlation coefficient is 0.69. King and Wiebe measured methane release from a salt marsh in Georgia during the period April 1975 to February 1977. They reported methane fluxes from three parts of the marsh distinguished by the height of vegetation. In all three cases the rate of methane release was lower than that observed by Baker-Blocker *et al.* The most copious fluxes were observed at the site with stalks of less than 0.5 m. Their data for the last 12 months of observations can be represented as

$$F = 12.8 \times 10^{-5} \exp(0.143\ T)\ \mathrm{g\,m^{-2}h^{-1}} \qquad (2)$$

The correlation coefficient is 0.92. A similar analysis of their mid-marsh data yields

$$F = 0.78 \times 10^{-5} \exp(0.088\ T)\ \mathrm{g\,m^{-2}h^{-1}} \qquad (3)$$

with the correlation coefficient = 0.61. The rate of methane release from the third site (with stalks > 1 m) was also small and does not appear to have a systematic variation with T (we calculate the correlation coefficient as 0.33). Noting that eq. (1) represents a much higher flux than (2) and (3), and also that the average of the exponents in (2) and (3) is 0.116, we have adopted the following model for the temperature variation of natural methane fluxes

$$F = F_0 \exp(0.12\ T) \qquad (4)$$

where, for subsequent application, F_0 is the present biospheric flux of methane (column 1 of Table 1).

3. The climate-chemical model

The coupled climate-chemical model constitutes a modification of that previously employed by Hameed *et al.* (1980). This comprises the coupling of a latitudinally dependent energy-balance climate model with a latitudinally dependent tropospheric chemical model, both being annual models. Several changes have been made with respect to the earlier version of the climate model, such as the subdivision of poleward advective transfer into sensible and latent heat components, and these changes are discussed elsewhere (Coakley *et al.*, 1982). For a

doubling of atmospheric CO_2, the climate model produces an increase in equilibrium global mean surface temperature of 3.1 °C, in good agreement with general circulation models of the annual climate (Manabe and Wetherald, 1975; Manabe and Wetherald, 1980). We note, however, that recent simulations employing seasonal general circulation models (Manabe and Stouffer, 1980; Wetherald and Manabe, 1981) suggest that an annually-averaged seasonal model might be less sensitive to climatic forcing than an annual model.

The chemical model calculates the latitudinal variation of tropospheric trace species by numerically solving the coupled species continuity equations under mean annual conditions. The formulation of the model is described by Hameed *et al.* (1980) and Hameed and Stewart (1979). Species with chemical lifetimes shorter than a few days (OH, HO_2, H, NO, NO_2, NO_3, HNO_2, N_2O_5, O, $O(^1D)$, CH_3, CH_3O_2, CH_3O, HCO, H_2CO) are assumed to be in local chemical equilibrium and their north-south transport is not calculated. The present atmosphere is simulated by prescribing the latitudinal concentration distributions of CH_4, CO, H_2, O_3 and HNO_3 at their observed values and calculating their sources from the continuity equations. The procedure is straightforward except for odd nitrogen for which the prescribed concentrations of HNO_3 (which are the mean values for each latitude zone of the data of Huebert and Lazrus (1980)) are related to NO_x sources. The sources calculated for CH_4 are shown in Table 1. The global sources obtained for the other gases and their assumed anthropogenic components are shown in Table 2. The model is then used to calculate the impact of changes in the sources on the tropospheric chemical balance. Specifically, we consider variations in the natural CH_4 source according to eq. (4) in response to climate change induced by a doubling of atmospheric CO_2 together with possible increases in the anthropogenic sources of CO, CH_4 and NO_x. The concentrations of OH obtained in the model are shown in Fig. 5 of Hameed *et al.* (1980).

Table 2. *Model estimates of net global sources of tropospheric constituents*

Tropospheric constituent	Global source in MT/yr	Anthropogenic component MT/yr	Comments
CH_4	412	82	
CO	2823	700	CO production from CH_4 calculated in model
NO_x	36	20	As mass of nitrogen
O_3	787	—	This source represents injection from stratosphere
H_2	−11	—	Anthropogenic sources exist but not included in this model

4. Methane and climate change

If a change in global climate occurs it can alter the chemical composition of the atmosphere in a variety of manners of which we discuss two in this study:

(i) *Change in the natural flux of methane.* As discussed earlier the rate of methane release from soils increases with temperature. Thus with a global warming an increase in atmospheric methane abundance may be expected. But decomposition of methane in the atmosphere is a major source of several species such as O_3, CO, H_2, H_2CO etc., and the abundance of these gases may also be expected to rise. Since CH_4 and O_3 have significant greenhouse effects, a positive feedback to climate change is possible due to the initial impact on natural CH_4 sources.

(ii) *The climate-chemical feedback.* Simulations of climate change in general circulation models indicate that atmospheric water vapor density changes with climate such that local relative humidity is approximately conserved (Wetherald and Manabe, 1975). This is of significance to the tropospheric chemical balance because the hydroxyl (OH) radical is produced mostly through the reaction:

$$O(^1D) + H_2O \rightarrow OH + OH$$

and OH is the major tropospheric sink of CH_4, CO and H_2. The density of O_3 is affected by the change in CH_4 concentration. Also the chemical destruc-

tion of O_3 in the troposphere is dominated by water vapor and its derivates through the reactions:

$$O_3 + HO_2 \rightarrow OH + O_2 + O_2$$

$$O_3 + OH \rightarrow HO_2 + O_2$$

$$O(^1D) + H_2O \rightarrow OH + OH$$

Considerable amounts of O_3 are also destroyed at the earth's surface, any changes in which are not considered in the present study. However, it is clear that an increase in tropospheric temperature is likely to reduce the concentrations of CH_4 and O_3, and this in turn provides a negative feedback to the change in climate. A further discussion of this feedback is given by Hameed *et al.* (1980).

5. Global warming due to the release of CO_2, CH_4, CO and NO_x

The magnitude of the likely increase in global temperature because of enhanced CO_2 in the atmosphere due to fossil-fuel burning has been the subject of much discussion in the literature. Recently we have suggested that the climatic consequences of the release of other gases from fossil-fuels may also be significant (Hameed *et al.*, 1980). This analysis assumed an increase by a factor of 1.7 in atmospheric CO_2, together with an increase in fossil-fuel use by between a factor of 4 (low estimate) and a factor of 8 (high estimate). Thus we have explored the consequences of fourfold and eightfold increases in the anthropogenic emissions of CO, CH_4 and NO. Calculations show that these industrial gases are likely to contribute significantly to the greenhouse heating of the atmosphere in spite of the moderating influence provided by the climate-chemical negative feedback.

Our present results are illustrated in Table 3 for a doubling of atmospheric CO_2, and for which we have incorporated the effect of temperature-dependent methane emission from natural sources. In line 1 we double CO_2 but do not increase either anthropogenic emissions of CH_4, CO and NO_x or natural emissions of CH_4. This corresponds to the standard CO_2 doubling scenario with the model producing a global warming of 3.13 °C. In line 2 negative climate-chemical feedback has been included, producing a modest decrease in global warming (2.93 °C), consistent with our prior study. Line 2 includes positive feedback due to natural sources of CH_4 and, interestingly enough, this feedback virtually compensates the negative climate-chemical feedback. One reason for the small magnitude of this positive feedback is that most of the natural sources of CH_4 are located in tropical regions where our climate model produces minimum warming. For example, with a doubling of CO_2 the model predicts tropical warming of 2.2 °C as compared with global warming of 3.1 °C. Thus the increase in natural methane flux is smaller than would be predicted using a globally averaged model.

Although the two feedbacks which have been incorporated in going from line 1 to line 3 in Table 3 produce compensation from a climate standpoint, it is interesting to note that they produce an altered chemical composition (compare line 3 to line 1), with the warmer Earth having less tropo-

Table 3. *Calculated changes in tropospheric O_3 and CH_4 concentrations and increases in global mean surface temperature for a doubling of atmospheric CO_2, together with increases in anthropogenic sources of CH_4, CO and NO_x*

Factor increase in anthropogenic sources of CH_4, CO, NO_x	Increase in natural CH_4 flux	Climate/chemical feedback	$\times O_3$	$\times CH_4$	ΔT_s(°C)
1	No	No	1.00	1.00	3.13
1	No	Yes	0.89	0.83	2.93
1	Yes	Yes	0.93	1.13	3.11
4	No	Yes	1.45	1.33	3.69
4	Yes	Yes	1.51	1.74	3.88
8	No	Yes	1.93	2.25	4.38
8	Yes	Yes	2.00	2.89	4.59

spheric O_3 but more tropospheric CH_4. The opposite directions of change in the concentrations of O_3 and CH_4 are of interest in the historical relationship between these atmospheric gases. Because ozone is produced photochemically from methane, it has been suggested that they have increased or decreased together in the past (Walker, 1977). In Table 3 we see that this relationship can be reversed when the climate-chemical interaction is included.

In Table 3 we additionally include fourfold and eightfold increases in the man-made sources of CO, CH_4 and NO_x. The increases in ΔT_s, in the absence of an increase in the natural flux of CH_4, are consistent with our prior study (Hameed *et al.*, 1980), while inclusion of positive feedback due to the natural flux of CH_4 adds roughly 0.2 °C to the warming. What is perhaps most significant is the fact that Table 3 suggests significant increases in CH_4 concentration, with this concentration being enhanced by two separate mechanisms. The first is an increase due to interactive tropospheric chemistry resulting from future possible increases in anthropogenic emissions of CO, CH_4 and NO_x, while the second is a result of an increase in the natural flux of methane due to global warming resulting from anthropogenic increases of CO_2, CH_4 and O_3. It is thus interesting to note that Rasmussen and Khalil (1981) have recently suggested that the atmospheric concentration of methane is increasing at the rate of approximately 2% per year.

6. Discussion

The calculations presented here cannot be called accurate because of considerable gaps in our knowledge of the cycles of tropospheric gases. However, the climatic and atmospheric compositional effects which we calculate due to increased fossil-fuel emissions of CO, CH_4 and NO_x, together with increases in the natural methane sources, point to the necessity of closer investigation of the sources, sinks and atmospheric concentrations of these trace gases. Currently carbon monoxide and nitrogen oxides are monitored mostly as local air pollutants. A better understanding of the magnitudes, distributions and potential variability of their sources is required. Because CH_4 is not regarded as an air pollutant, its cycle has attracted even less attention; no current direct estimate of its anthropogenic sources exists in the literature. Our understanding of atmospheric methane is far from complete, but the calculations presented here suggest that the effects due to increases in fossil fuel combustion and the climatic impact on methanogenesis in soils are likely to be significant in determining future trends of atmospheric methane concentration.

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