FLUX ESTIMATES FROM SOIL METHANOGENESIS AND METHANOTROPHY: LANDFILLS, RICE PADDIES, NATURAL WETLANDS AND AEROBIC SOILS

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Abstract. Present and future annual methane flux estimates out of landfills, rice paddies and natural wetlands, as well as the sorption capacity of aerobic soils for atmospheric methane, are assessed. The controlling factors and uncertainties with regard to soil methanogenesis and methanotrophy are also briefly discussed.

The actual methane emission rate out of landfills is estimated at about 40 Tg yr⁻¹. Changes in waste generation, waste disposal and landfill management could have important consequences on future methane emissions from waste dumps. If all mitigating options can be achieved towards the year 2015, the CH₄ emission rate could be reduced to 13 Tg yr⁻¹. Otherwise, the emission rate from landfills could increase to 63 Tg yr⁻¹ by the year 2025. Methane emission from rice paddies is estimated at 60 Tg yr⁻¹. The predicted increase of rice production between the years 1990 and 2025 could cause an increase of the CH₄ emission rate to 78 Tg yr⁻¹ by the year 2025. When mitigating options are taken, the emission rate could be limited to 56 Tg yr⁻¹. The methane emission rate from natural wetlands is about 110 Tg yr⁻¹. Because changes in the expanse of natural wetland area are difficult to assess, it is assumed that methane emission from natural wetlands would remain constant during the next 100 years. Because of uncertainties with regard to large potential soil sink areas (e.g. savanna, tundra and desert), the global sorption capacity of aerobic soils for atmospheric methane is not completely clear. The actual estimate is 30 Tg yr⁻¹.

In general, the net contribution of soils and landfills to atmospheric methane is estimated at 180 Tg yr⁻¹ (210 Tg yr⁻¹ emission, 30 Tg yr⁻¹ sorption). This is 36% of the global annual methane flux (500 Tg yr⁻¹).

1. Introduction

Because of its effective absorption of infra-red radiation, atmospheric methane (mainly found in the troposphere) plays a direct role as a greenhouse gas in our global climate. Methane accounts for about 15% to the current commitment to global warming (OTA, 1991). Its global warming potential (GWP) is 11 times the GWP of CO₂ when calculated over a 100-year period (Watson *et al.*, 1992). Atmospheric methane also exerts a strong influence on atmospheric chemistry. Mainly in the troposphere, it is oxidized by hydroxyl radicals (OH) via formaldehyde (CH₂O) to carbon monoxide (CO) and subsequently to carbon dioxide (CO₂). Methane oxidation yields the following net results for atmospheric chemistry. In nitric oxide (NO)-poor environments a net loss of OH and O₃ is found (Crutzen, 1993). Besides being an important regulator of OH radicals, methane is also a primary sink for C1 atoms in the atmosphere. Methane is also a major source of H₂ and stratospheric

water vapour (Tyler, 1991). Its atmospheric lifetime is 8-12 years (Watson et al., 1992).

The current atmospheric methane concentration of about 1720 ppbv is the result of both natural (e.g. wetlands) and anthropogenic emissions (e.g. rice fields and landfills) as well as of CH₄ oxidation in aerobic soils and in the atmosphere. According to Khalil and Rasmussen (1990), atmospheric methane has been increasing during the 1980s at a rate of about $1.02 \pm 0.02\%$ per year. However, at the beginning of the 1990s, it was shown that this increase was slowing down and even dropped below zero in the Northern Hemisphere by the end of 1992 (Bekki *et al.*, 1994; Dlugokencky *et al.*, 1994).

The global annual CH₄ emission rate is estimated to be about 500 Tg yr⁻¹ (IAEA, 1992), with an uncertainty between 10 and 20% (Khalil, 1992). Natural wetlands contribute for about 22%, while rice paddies and landfills contribute respectively for 12 and 6%. Carbon availability, temperature/moisture interactions, soil type, pH, Eh and nutrient dynamics are the key variables controlling methane emission from anaerobic soil systems. Aerobic soils (e.g. forest, upland agricultural soils), on the other hand, can be a sink for atmospheric methane. Methane oxidation by soils is estimated at about 6% of the global annual methane emission into the atmosphere (IAEA, 1992). Methane sorption by aerobic soils is mainly influenced by nitrogen input, moisture content/soil type interactions and pH.

The importance of methane as a greenhouse gas and as an atmospheric reagent, as well as its changing atmospheric concentration warrant the importance of source and sink determinations. This paper deals with the general aspects of present and future methane emission rate estimates out of landfills, rice paddies and wetlands as well as with the oxidation rate in aerobic soils.

2. Estimates of CH₄ Emission out of Landfills

Landfills have been used for many years as a common and economical manner of refuse disposal. On a global scale, approximately 653 Tg of waste is landfilled (Thorneloe *et al.*, 1993). Projections for the year 2000 suggest that, due to an increase in population and waste generation, landfills could become a major source of atmospheric methane (Kreileman and Bouwman, 1994). The most reliable estimates of methane emission from landfills are situated around 40 (10–70) Tg yr⁻¹ (Table I).

Buried refuse is gradually broken down by a complex interaction of microbial processes. Three groups of bacteria obtain their metabolic energy from organic waste conversion. The first group are *hydrolytic* and *fermentative bacteria*. These organisms convert organic polymers into monomers, which are subsequently fermented to secondary metabolites. This part of organic waste conversion is restricted to the aerobic lifetime of the landfill and tends to decrease the pH. The secondary metabolites are converted to CH₄ and CO₂ by a second group of micro-organisms,

Tg yr ⁻¹	Year	Reference
36-93ª	1994	Kreileman and Bouwman (1994)
2025	1993	Matthews et al. (1993)
21–57	1993	Peer et al. (1993)
1565	1993	Lelieveld and Grutzen (1993)
2070	1 992	Watson et al. (1992)
1050	1992	Bogner (1992)
3070	1988	Cicerone and Oremland (1988)
3070	1987	Bingemer and Crutzen (1987)

 Table I

 Estimates of methane emission rates out of landfills

^a Predicted emission for the year 2050.

the obligate proton-reducing acetogens and sulphate-reducing bacteria in direct association with a third group, the H_2 removing methanogens. The removal of H_2 is important to permit active growth of the acetogenic bacteria, which are inhibited by H_2 accumulation. When a landfill has reached steady state conditions, landfill gas contains approximately 55% CH₄ and 45% CO₂ (Farquhar and Rovers, 1973), as well as some trace gases which are responsible for the bad odour.

Methane formation in landfills is mainly influenced by the moisture content and pH of the refuse. Methanogenesis in landfills occurs between a very narrow pH range (6.8–7.4) (Thorneloe, 1993) and is stimulated when the moisture content of the refuse increases (Gurijala and Suflita, 1993). The physical parameters of the refuse, as well as the landfill design and age can have an important influence on methane production. Not all methane produced is emitted into the atmosphere. Methane oxidation and gas flaring have a strong mitigating effect on the methane emission out of landfills. Soils covering a landfill can rapidly oxidize CH_4 (Whalen *et al.*, 1990) and a gas flaring system can recover most of the methane produced (Aitchison, 1993).

As can been seen in Table I, emission rate estimates from landfills vary between 10 and 70 Tg yr⁻¹, with an average emission rate of 40 ± 10 Tg yr⁻¹. The high emission rates mentioned in this table (70 Tg yr⁻¹) are probably overestimated, because of the underestimation of the high oxidation capacity of landfill cover soils (Boeckx and Van Cleemput, 1994; Jones and Nedwell, 1993; Peer *et al...*, 1993; Smith *et al.*, 1994; Whalen *et al.*, 1990) and the increasing installation of gas flaring systems, especially in developed countries (Thorneloe *et al.*, 1993). Future trends in waste management are very important in estimating CH₄ emission from landfills. In the discussion on future waste generation, landfilling and CH₄ emission out of landfills, a difference between developed and less developed countries appears (Torneloe *et al.*, 1993). In the OECD countries less waste is landfilled. There is also a trend to reuse, recycle and separate the waste as well as to waste incineration. Landfill sites shift from smaller towards larger and better controlled sites (gas

recovery, protection of groundwater infiltration). From this it can be expected that, in these countries, CH_4 emission from waste generation will diminish or at least not increase during the next decades. However, in East European countries and in the former USSR, landfilling is continuing and no gas recovery systems are used. In the Asian countries, landfilling is constant, but the increase of the population could cause an increase in methane production. In the South American region, landfilling tends to increase, together with an increasing interest in waste recycling and gas recovery. In the African countries, waste recovery is necessary for daily life. In all these countries methane emission from landfills is expected to increase.

An emission rate scenario for methane out of landfills can be made assuming first order kinetics for the degradation of organic waste (equation (1) (Hoeks, 1983):

$$\frac{\mathrm{d}P}{\mathrm{d}t} = -k \cdot P \tag{1}$$

where P is the amount of biodegradable waste (kg).

In general, 18% of the landfilled waste is biodegradable (van Amstel *et al.*, 1993; Thorneloe *et al.*, 1993). The degradation coefficient (k) is estimated between 0.5 yr⁻¹ for mixed waste and 0.05 yr⁻¹ for paper, wood and textile (Hoeks, 1983). Van Amstel *et al.* (1993) calculated a degradation coefficient of 0.1 yr⁻¹ for the period 1945–1995 and 0.365 yr⁻¹ for 1995–2010. Integration of equation (1) gives equation (2), with *Po*, the initial amount of biodegradable waste present in the landfill (t = 0) and *Pt* the amount of biodegradable waste present at any time t(yr):

$$Pt = Po \cdot e^{-k \cdot t} \tag{2}$$

Stoichiometrically, 0.373 m³ CH₄ can be produced from 1 kg glucose ($C_6H_{12}O_6$) (equation (3)). However, in general, one does not deal with defined chemicals but with an unknown mixture. The amount of oxygen required to oxidize organic matter by chemical means ($K_2Cr_2O_7$) is expressed as chemical oxygen demand (COD). Generally, about 1 kg of COD is required to oxidize 1 kg of degradable organic matter (equation (4)) and 1 kg COD corresponds to 0.350 m³ CH₄ (equations (3), (4) and (5)).

$$C_6H_{12}O_6 \to 3 CH_4 + 3 CO_2$$
 (3)

$$C_6 H_{12} O_6 + 6 O_2 \to 6 CO_2 + 6 H_2 O \tag{4}$$

$$\frac{3\text{mol CH}_4 \times 22.4 \frac{1}{\text{mol}}}{6\text{mol O}_2 \times 32 \frac{g}{\text{mol}}} = \frac{0.3501\text{ CH}_4}{g\text{ COD}} = \frac{0.350\text{m}^3\text{ CH}_4}{\text{kg COD}}$$
(5)

Thus, from 1 kg of biodegradable waste approximately 0.350 m³ CH₄ can be produced. By Equation (6), the CH₄ production (α , m³ yr⁻¹) can be calculated. By integrating equation (6), the accumulative gas production (α_{acp}) can be found

Year of maximum	90% recovery	50% recovery	10% recovery
emission	$(Tg yr^{-1})$	$(Tg yr^{-1})$	$(Tg yr^{-1})$
	1% Increase	e of landfilled wa	ste per year
1995 + 25	8.2	40.8	73.4
2000 + 25	8.6	42.9	77.2
2005 + 25	9.1	45.1	81.1
	507 :	f. I	
	5% increase	e of fanolitied wa	ste per year
1995 + 25	8.2	40.8	73.4
2000 + 25	10.4	51.1	93.7
2005 + 25	13.3	66.5	119.6
	1% decreas	e of landfilled wa	ste per vear
1995 + 25	8.2	40.8	73.4
2000 + 25	7.8	38.8	69.8
2005 + 25	7.4	36.9	66.4
	5% decrease	e of landfilled wa	ste ner vear
1005 + 25	2) accreas		72 /
1993 + 23	0.2	40.0	13.4
2000 + 25	6.3	31.6	56.8
2005 + 25	4.9	24.4	44.0

Calculated global estimates of methane emission rates (α_{ace}) out of land-fills

By integrating equation (6), the accumulative gas production (α_{acp}) can be found (equation (7). The total global landfilled waste is 653×10^9 kg (Thorneloe *et al.*, 1993) and about 18% is biodegradable. So $Po = 653 \times 10^9 \times 0.18$ kg).

$$\alpha = -0.35 \frac{\partial P}{\partial t} = 0.35 \times k \times Poe^{-k \cdot t}$$
(6)

$$\alpha_{\rm acp} = 0.350 \times Po \times (1 - e^{-k \cdot t}) \tag{7}$$

$$\alpha_{\rm ace} = NOX \times 0.350 \times Po \times (1 - e^{-k \cdot t}). \tag{8}$$

The accumulative methane emission rate (α_{ace}) can be calculated by equation (8), wherein *NOX* is the percentage of methane which is not oxidized or recovered. Considering an active life time of a landfill of 25 years (Aitchison, 1993), a typical plot for methane emission rate scenarios can be drawn. In Figure 1 the accumulative emission rate is shown for k values between 0.1 and 0.5 yr⁻¹. An oxidation and recovery potential of 50% was taken into account. In Table II some global emission rate predictions are shown. To calculate these data it was assumed that the total landfilled waste either increases or decreases at a constant rate of 1 or 5% yr⁻¹,



Figure 1. Methane emission rate scenarios for landfills, considering a range of degradation coefficients, a lifetime of 25 years and a methane/recovery efficiency of 50%.

 $k = 0.3 \text{ yr}^{-1}$ and that respectively 90%, 50% and 10% of the produced methane is recovered or oxidized. A total time span of 35 years was considered. From Table II it is clear that reducing the amount of landfilled waste as well as efficient gas recovery or oxidation are important mitigating options. Installing gas recovery systems or enhancing methane oxidation seems to be more effective than reducing the amount of landfilled waste, at least for the next 35 years.

For larger landfills, gas recovery systems, together with a daily soil covering during the landfilling period, are recommended (Aitchison, 1993; Oonk, 1993). Large amounts of landfill gas are produced and emitted during the exploitation. A daily cover can substantially reduce the methane emission before the gas flaring system is operational. The recovered gas can either be used directly (heating, gas boiler), used for power generation (electricity) or the gas can be upgraded to natural gas quality and injected in the gas distribution net (Oonk, 1993). For smaller and older landfills a gass recovery system would not give an important economic benefit. Because of the large number of such small sites they contribute significantly to methane emission. A cheaper and very effective option to reduce methane emission from small and old landfills is methane oxidation by soils covering the landfill (Aitchison, 1993). For small sites, Nozhevnikova *et al.* (1993) found a total oxidation of methane in the aerobic cover without any release into the atmosphere, while for larger dumps an oxidation of 50% was observed. Boeckx and Van Cleemput (1994) found an oxidation capacity of 95% in a Belgian landfill

scenari	os out o	r landnii	s for the
2000	2025	2050	2100
42	63	93	109

	Table III			
Regional yearly CH4	emission scenarios	out of	landfills	for the
coming 100 years				

CH₄ emission (Tg)

North America

Pacific-OECD

Eastern Europe

Latin America

World

Asia

Africa

Europe

cover soil. However, in the meanwhile, waste reduction has to be achieved, because it is a more economical and beneficial option. Source reduction of organic waste, by separately collecting and handling organic waste garden, fruit and kitchen waste (GFK) could also substantially reduce future landfill emissions.

In Table III, an overview is given for the emission rate scenarios in different parts of the world, according to the Intergovernmental Panel on Climate Change (IPCC) for the year 1990 until 2100 (van Amstel *et al.*, 1993). The prediction of the global methane emission rate from landfills by the year 2025 agrees well with the estimations presented in Table II for this year, where it was assumed to have a yearly increase of the total landfilled waste with 5% and a gas recovery of 50%. Although, at this moment, important improvements are made towards landfill management, it is believed that towards the year 2050 the methane emission rate will increase to 81 Tg yr⁻¹ (Kreileman and Bouwman, 1994) or even up to 93 Tg yr⁻¹ (van Amstel *et al.*, 1993). Without mitigating options, methane emission will become the third source of atmospheric methane.

Potential emission from landfills can be reduced by about 50–90% (van Amstel *et al.*, 1993). If all potential reduction options (such as gas recovery, CH₄ oxidation and waste reduction) could be achieved by the year 2025 the global methane emission from landfills is estimated at 13 Tg yr⁻¹ by the year 2100 (van Amstel *et al.*, 1993).

A number of uncertainties still remain in estimating methane emission from landfills. Methane emission from landfills is highly variable and heterogeneous. Both physical and chemical characteristics of the waste, landfill design and age (influence on CH_4 production), as well as the efficiency of gas recovery systems and methane oxidation (influence on the emission) are responsible for these fluctions. More field emission data should be collected to establish a reliable emission estimate.

3. Estimates of CH₄ Emissions out of Rice Paddies

Rice paddies are the third anthropogenic source of atmospheric metahne (IPCC, 1992; IAEA, 1992). The actual harvested rice area is approximately 140 million hectares (Neue *et al.*, 1994b). More than 90% of the rice paddies are situated in South-East Asia (Zuidema *et al.*, 1994). Actually the most reliable estimate for CH₄ emission out of rice paddies is \pm 60 (20–150) Tg yr⁻¹ (IAEA, 1992; IPCC, 1992; Kreileman and Bouwman, 1994; Minami, 1993; Watson *et al.*, 1992).

Methane is formed during microbial decomposition of organic matter in the anaerobic zone of the rice paddies. The available organic matter in rice paddies is decomposed by several microbial species to fulfil the nutrient requirements of the methanogenic bacteria. Methane production in rice fields is influenced by a large set of parameters. The amount and 'quality' of the available carbon source either from amended organic matter or root exudates importantly influences the CH4 production (Renneberg et al., 1992). The fluctuations of the soil temperature and the rice plant-growing activities importantly contribute to the diurnal fluctuations in methane emission (Wang, 1993). The seasonal variations are explained by the change in available substrate and by the water regime of the paddies (Mianami and Neue, 1994). Methane production in rice fields is further influenced by soil type, pH and Eh. There is a wide variability between different soil types. The soil type mainly influences the time between flooding and the onset of methanogenesis (Minami, 1993). Indeed, soil properties govern electrochemical and chemical changes that occur upon flooding. The key parameter, however, controlling CH₄ production is the redox potential (Eh) of the soil (Gaunt, 1994; Wang et al., 1993). Inorganic fertilizer additions do not have a clear influence on methane emission (Neue et al., 1994a).

Methane, produced at depth, is not entirely released into the atmosphere. Methane is emitted by three different pathways: diffusion, ebullition (10%) and transport through the aerenchyma of the plant (90%) (Schütz *et al.*, 1989). So, the biomass of the rice plants is a very important factor determining the amount of methane emission out of rice fields (Savent *et al.*, 1994). A considerable part of the formed methane can also be oxidized at the aerobic part of the root zone or when passing through the oxidized layer of the flooded soil.

The evolution of the global estimates of methane emission out of rice paddies for the last 10 years is shown in Table IV and Figure 2. From Figure 2 it is clear that, in general, the emission rate estimates are declining towards the currently accepted estimate of about 60 Tg yr⁻¹ (IAEA, 1992; Kreileman and Bouwman, 1994). The evolution of these estimates can be explained by the fact that more flux measurements became available, as well as by an improvement of the accuracy of the measurement techniques. Also more flux measurements are being conducted in Asia where most of the rice fields are situated. However, extrapolation from individual flux measurements to a global budget still remains very difficult. A better understanding of the controlling factors on CH₄ production and emission

Table	I۷
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Year	Average (Tg yr ⁻¹)	Range (Tg yr ⁻¹)
1984ª	166	142–190
1985°	160	120-200
1986 ^a	120	70-170
1988 ^a	115	60-70
1989 ^a	100	50-150
1989 ^a	53	30–75
1989 ^a	100	6040
1989ª	120	70–170
1990 ^a	100	50-150
1990 ^a	90	60-120
1990 ^a	43	25-60
1990 ^a	84	53-114
1990 ^a	48	22-73
1990 ^a	98	25-170
1992 ^a	60	20-120
1992 ⁶	100	50-150
1993 ^a	63	12-113
1993 ^c	55	40-70
1993 ^d	70	20-120
1994 ^e	60	

Different estimates of methane emission rates out of rice paddies over the last 10 years

^a After Minami (1993); ^b After Watson et al. (1992);

^c After Matthews *et al.* (1993); ^d After Lelieveld and Crutzen (1993); ^e After Kreileman and Bouwman, (1994).

could provide better emission estimates. Obviously, the CH_4 emission from rice paddies was overestimated in the early 1980s, due to few measurements, and are now becoming more accurate.

The wide range in the emission rate estimate (Table IV) can be explained by the high spatial variably of CH₄ emission. Schütz and Seiler (1989) reported CH₄ emissions between 0.02 and 200 mg m⁻² day⁻¹. Another factor explaining the changing estimates and wide emission range are seasonal and diurnal emission fluctuations. Minami and Neue (1994) reported, for different rice fields, a seasonal emission range from 6 up to 1700 kg CH₄ ha⁻¹.

Differences between emission estimates can also be explained by the estimation technique used. Bachelet and Neue (1993) calculated results of emission estimates from Asian rice fields using three different methods. The FAO method uses country statistics and crop calendars to determine the land area under rice cultivation each month. Assuming a constant emission rate, Asian rice fields were considered to emit 82 Tg yr⁻¹. The second method assumes that methane production represents



Figure 2. Methane emission rate estimates out of rice fields, for the period 1984-1994.

a constant fraction of the primary production and uses empirical relationships between net primary production, temperature and precipitation records. Using this method an emission rate of 57 Tg yr⁻¹ was found. The third method is related to methane production and rice grain production. It involves the total organic matter added to paddy soils and assumes that a constant fraction is emitted as methane. This assumption leads to an estimate of 63 Tg yr⁻¹. Taking the data of Tables IV and V into account, it is clear that the first method overestimates the methane flux.

In order to feed the growing human population, rice production will have to increase during the next 20–30 years. Bouwman (1991) reported that global rice production may increase by 65% between 1990 and 2025. This increase will cause an increased methane emission of 42% over the next 35 years ($1.2\% \text{ yr}^{-1}$). Anastasi *et al.* (1992) predicted an average increase of methane emission of $1.1\% \text{ yr}^{-1}$. According to Lindau (1993), increased rice production may lead to an increased CH₄ emission of 20% over the next 10 years ($2\% \text{ yr}^{-1}$).

Because the area of arable land is highly limited in the rice-producing countries, increased production has to be achieved mainly by intensified cropping (2–3 crops per season), rather than by an expansion of the cultivation area (Sass *et al.*, 1992). So the increase of methane emission from rice fields will be attributed to a more intensive use of the areas. It has also been pointed out by climate models that, if positive temperature feedbacks do materialize, methane emissions from paddy soils may increase drastically (den Elzen and Rotmans, 1993). In Table V an overview is given of the emission scenarios for the next 100 years, according to the IPCC (van Arnstel *et al.*, 1993). The emission increase is almost completely attributed to the Asian countries.

CH ₄ emission (Tg)	1990	2000	2015	2050	2100
World	60	66		87	84
North American	1	1	1	1	1
Pacific-OECD	1	1	1	2	2
Eastern Europe	0	0	0	0	0
Asia	55	61	72	80	77
Latin America	1	1	1	1	1

2

0

2

0

2

0

2

0

Table V Regional yearly CH₄ emissions scenarios out of rice fields for the coming 100 years

2

0

Africa

Europe

Reduction of methane emission from rice fields does not seem to be easy to achieve, because reducing measures should not affect rice yield. Both water managing during certain periods of the growing season and reducing the organic matter amendments aim at reducing methane emission. An intermitted irrigation could reduce the CH₄ emission by 15-59%, and at the same time increase production by 10-20%. However, intermitted irrigation may favour the emission of another, much more powerful greenhouse gas: nitrous oxide (N_2O). Small emissions of N_2O may offset the gains from reducing CH₄. Rational organic and inorganic fertilizer applications is also an important reducing option (Sicui Liang and Geng Yang, 1994). It is shown that an excess of organic matter can substantially reduce the grain yield (Sass et al., 1991). Ridge cultures decrease methane emission by 39– 43% and increase production by 20-30%. Dry seeding showed a great potential for reducing methane emission, but the yield is also reduced. According to the IPCC (van Amstel et al., 1993) a maximum reduction of 30% could be achieved by the year 2025. This would result in a global methane emission rate from rice fields of 56 Tg yr^{-1} .

There is significant information available concerning the factors controlling CH₄ emission, the location area and cultivation period of rice fields. However, sources of uncertainties to improve the estimations from rice paddies are: effects of local soil temperatures, soil type, application of non-commercial fertilizers, seasonal and annual variations in water management and the effect of soil chemistry on the methane emission potential (Matthews et al., 1993).

4. Estimates of CH₄ Emissions out of Natural Wetlands

Wetlands are the main natural source of atmospheric methane (IAEA, 1992). The major wetlands of the world can be subdivided into northern, temperate and tropical/subtropical wetlands. About 50% of the total wetland area is situated between

Tropical (Tg yr ⁻¹)	Temperature $(Tg yr^{-1})$	Boreal–Arctic Tg yr ⁻¹)	Global (Tg yr ⁻¹)	References
90	66		156	Khalil and Rasmussen (1983)
			48	Bolle et al. (1986)
34	12	65	111	Matthews and Fung (1987)
			115	Cicerone and Oremland (1988)
45	11	24	80	Aselmann and Crutzen (1989)
55	17	29	111	Barlett et al. (1990)
			100	Schütz <i>et al.</i> (1990)
71	12	32	115	Fung et al. (1991)
			150	Watson et al. (19982)
			115	IAEA (1992)
66	5	38	109	Bartlett and Harriss (1993)
			108	Matthews et al. (1993)
			125	Lelieveld and Crutzen (1993)
			111	Kreileman and Bouwman (1994)

Table VI Different estimates of methane emission rates out of natural wetlands over the last 10 years

50 and 70° N. Another major part of the global wetland area (35%) is found between 20° N and 30° S (Bartlett and Harris, 1993). Although the global wetland area is assumed to be relatively constant between 530 (Matthews and Fung, 1987) and 570 million hectares (Aselmann and Crutzen, 1989), the areal coverage may alter during one season (Matthews, 1993) and the extend of inundation can significantly change during the year. Also, wetlands can easily shift from being a source to a small sink for atmospheric CH₄ when they get dried up (Bartlett and Harris, 1993). This can cause high spatial and temporal variability of methane emission out of wetlands. But despite that, it is generally agreed that the annual methane emission is fairly constant. The most reliable estimate is 110 (100–200) Tg yr⁻¹ (IAEA, 1992; Kreileman and Bouwman, 1994; Roulet and Matthews, 1993). Although wetlands are responsible for 22% of the global annual CH₄ emission, the increase of atmospheric methane of the last decades cannot be attributed to them (Roulet and Matthews, 1993).

High carbon availability under anaerobic conditions favours methanogenesis. Methanogenic bacteria decompose organic matter residues of fermentative bacteria using CO_2 as an electron acceptor in the absence of oxygen or other electron acceptors. Factors controlling methane emission are similar to those in rice fields. Thus, a complex set of parameters influences the CH_4 emission from natural wetlands: soil water status, temperature, soil type, pH, Eh, land use and vegetation, nutrient input and organic matter accumulation and characteristics (Matthews, 1993). The methane emission from tropical/subtropical wetlands is generally governed by large scale precipitations and flood cycles. High latitude seasonal methane emission



Figure 3. Methane emission rate estimates out of natural wetlands, for the period 1983–1994.

sions are controlled via interactions between temperature and changes in water tables (Matthews, 1993). Emission rate estimates are shown in Table VI and Figure 3. From this table it can be concluded that $53 \pm 11\%$ of the total methane emission originates in the tropical regions. Individual wetlands show great spatial and temporal variability in methane emission due to different environmental factors (mainly temperature and water status fluctiations, as well as some episodic events). Although the spatial and temporal variability it is clearly demonstrated in Table VI and Figure 3 that the calculated global methane emission rates from wetlands have varied relatively little during the last 10 years. The differences are attributed to the different distributions of the various wetland ecosytems which were used in the calculation procedures by the authors. This results in different total areas of specific types. Of course, extrapolation of methane emissions out of these habitats causes different results.

Based on a positive temperature feedback mechanism, it is to be expected that methane emission from wetlands would increase in the future due to a direct temperature response of the bacteria, an increased length of the emission season and an expansion of the potential wetland area (den Elzen and Rotmans, 1993). However, other factors (nutrient limitations and declining water supply) may moderate this response (Matthews, 1993). Predictions of hydrologic perturbations over the next 50–100 years are highly uncertain. Changes in wetland areas are difficult to describe (Zuidema *et al.*, 1994) and to quantify (Matthews, 1993). Therefore, global methane emission rates from wetlands are assumed to be constant in time. It

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CH ₄ oxidation (Tg yr ⁻¹)	Year	Reference
20	1984	Seiler (1984)
32 ± 16	1987	Seiler and Conrad (1987)
23-56	1990	Schütz et al. (1990)
15-45	1992	IAEA (1992)
30 ± 25	1993	Lelieveld and Crutzen (1993)
50	1993	Ojima et al. (1993)
30 ± 15	1994	Smith et al. (1994)

Table VII Different estimates of methanothrophy in aerobic soils over the last 10 years

is still an open question whether wetlands will become a smaller or larger methane source or whether parts of it could shift to methane sink areas.

Although there is a general agreement that the global annual methane flux from wetlands is approximately 111 ± 26 Tg yr⁻¹ (derived from Table VI), some uncertainties remain. The area, distribution and environmental characteristics of wetlands as well as general seasonal dynamics of water and temperature regimes are well characterized. However, larger uncertainties are related to organic inputs, seasonal dynamics of methane production periods and inter-annual variations in wetland area, as well as the amount of methane ebullition (Matthews *et al.*, 1993). Therefore, extrapolation from one wetland region to another may cause considerable estimation errors. By filling in the currently leaking emission data (tropical Asia, Siberia, Africa), more accurate measurements and uniform wetland classification may lead to a better methane emission assessment (Bartlett and Harriss, 1993).

5. Estimates of CH₄ Oxidation in Aerobic Soils

Oxidation of atmospheric methane has been reported in a variety of aerobic soils. Methane oxidation is found in forest soils (Steudler *et al.*, 1989; Schnell and King, 1994), agricultural soils (arable land and grassland) (Hütsch *et al.*, 1994), desert soils (Striegl *et al.*, 1992), tundra soils (Whalen *et al.*, 1992) and taiga soils (Whalden *et al.*, 1991). Both *methanotrophic* and *nitrifying bacteria* are responsible for the oxidation of atmospheric methane (Bédard and Knowles, 1989). Methane oxidation in soils in influenced both by physical and chemical paramaters. Soil moisture and soil compaction (porosity) control the diffusion rate of CH₄ into the soil. Also pH, nitrogen dynamics (NH₄⁺ and NO₂⁻ turnover, Schnell and King, 1994) and soil temperature can have an important influence on the oxidation capacity of upland soils (Ojima *et al.*, 1993; Hütsch *et al.*, 1994).

In Table VII estimates from different sources concerning methane oxidation in soils are presented. The most reliable estimate of the oxidation rate of atmospheric methane in soils is approximately 34 ± 9 Tg yr⁻¹ (Table VII). Because the role of large potential methane oxidizing areas (e.g., savannas, deserts, tundra) is still unknown, the estimates of soil methanotrophy remain very uncertain (Batjes and Bridges, 1992). Recent studies have shown that land use changes may possibly decrease the CH₄ uptake and also increase the release of another greenhouse gas (N₂O) in the near future (Ojima *et al.*, 1993). To tighten the estimates of soil methanotrophy more measurements are required.

6. Conclusions

With almost 36% of the total annual methane emission, landfills, natural wetlands and rice paddies contribute importantly to the increase of atmospheric methane. In comparison to atmospheric methane oxidation, aerobic soils are a rather small sink for atmospheric methane (6% of the annual emission). However, the actual knowledge about the sink function of large upland soil areas is still poor.

The methane emission rate from landfills is estimated to be about **40 Tg yr**⁻¹. Although waste separation and recycling are becoming more common, it is predicted that landfilling will increase at least for the next 20 years. If no reducing measures are taken (gas recovery, CH₄ oxidation, waste reduction) the estimated methane emission rate out of landfills will be **63 Tg yr**⁻¹ towards the year 2025. If all possible mitigating options could be achieved towards the year 2025, landfills could emit only **13 Tg yr**⁻¹ in 2100.

Rice paddies contribute about 60 Tg yr⁻¹ to the global methane emission. To feed the growing population of the earth, rice production should increase with 65% between 1990 and 2025. Consequently this will lead to an intensified use of the rice production areas and could cause a methane emission rate of about 78 Tg yr⁻¹ by the year 2025. Mitigating options are difficult to achieve because they should not affect the grain yield. If all possible reducing measures can be executed the methane emission rate would drop to 56 Tg yr⁻¹ by the year 2025.

Natural wetlands are the largest natural source of atmospheric methane. Yearly, they emit about **110 Tg** into the atmosphere. With regard to future climate feedbacks, it is not yet clear whether natural wetland areas will increase or decrease. It is assumed that the global methane emission from wetlands will remain fairly constant for the next decades.

Present and future emission rate estimates, taking a positive and a negative scenario into account, are shown in Figure 4.

In general, it is agreed that aerobic soils oxidize about 30 ± 15 Tg yr⁻¹. However, the quantity of methane oxidation in large potential areas (e.g. savannas, tundra, deserts) is still unclear. At this moment, the net contribution of soils and landfills to the atmospheric methane concentration is estimated at 180 Tg yr⁻¹ (210



Figure 4. Present and future methane flux estimates for landfills, rice paddies and natural wetlands.

 yr^{-1} emission, 30 Tg yr^{-1} sorption). This is 36% of the global annual CH₄ flux (500 Tg yr^{-1}).

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