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Methane emission and entrapment in flooded rice soils as affected by soil properties

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Abstract. Laboratory incubation experiments were conducted to study the effects of soil chemical and physical properties on CH_4 emission and entrapment in 16 selected soils with a pH range of 4.7–8.1, organic matter content of 0.72–2.38%, and soil texture from silt to clay. There was no significant correlation with CH_4 emission for most of the important soil properties, including soil aerobic pH (measured before anaerobic incubation), total Kjeldahl N, cation exchange capacity, especially soil organic matter, and soil water-soluble C, which were considered to be critical controlling factors of CH_4 emission. A lower CH_4 emission was observed in some soils with a higher organic matter content. Differences in soil Fe and Mn contents and their chemical forms contributed to the this observation. A significant correlation between the CH_4 emission and the soil organic C content was observed only after stratifying soils into subgroups according to the level of CH_4 emission in soils not amended with organic matter. The results also showed that the soil redox potential (Eh), anaerobic pH, anerobic pH, and biologically reducible Fe and Mn affected CH_4 emission significantly. Urea fertilization promoted CH_4 emission in some soils and inhibited it in others. This result appeared to be related to the original soil pH. CH_4 entrapment was positively correlated with soil clay content, indicating the importance of soil physical characteristics in reducing CH_4 emissions to the atmosphere.

Key words: Greenhouse gases – Methane emission – Methane entrapment – Redox potential – Rice fields – Soil organic carbon – Soil properties – Soil pH – Urea

CH_4 is one of the most important gases contributing to the so-called greenhouse effect. An accelerated increase in atmospheric CH_4 during the past 300 years has been observed (Khalil and Rasmussen 1985; Steele et al. 1987; Blake and Rowland 1988). This increase may be making an important contribution to an increase in global temperature, due to the relatively high absorption of infrared radiation by CH_4 (Bouwman 1990).

The main biotic sourced of atmospheric CH_4 are wetland rice cultivation, natural wetlands, ruminating animals, landfills, oceans and lakes, and biomass burning. Flooded rice fields are considered one of the major CH_4 emission sources and currently constitute approximately 20% of the total CH_4 budget (Bouwman and Sombroek 1990). CH_4 formation in flooded soils is a microbiological process affected by many environmental factors. However, the interactions between soil chemical and physical properties and CH_4 emission are not yet well understood. Information on the effects of different soil parameters on CH_4 emission and their quantitative relationship is necessary to provide a theoretical basis for controlling CH_4 emission in flooded rice soils. The objectives of the present study were to reveal the relationship between CH_4 emission and soil chemical and physical characteristics, including organic C, N, soil pH and Eh, soil cation exchange capacity, soil Fe, Mn, and their chemical forms, and the relationship between soil texture and CH_4 entrapment.

Materials and methods

A series of 16 flooded rice soils from USA, India, Thailand, and Liberia were selected. These soils were sampled from the surface layer (0–20 cm) and were air-dried, ground, and passed through a 1-mm sieve. The aerobic soil pH (soil and water ratio 1:1) varied from 4.7 to 8.1 and organic matter content from 0.72 to 2.38%. Soil texture ranged from silt to clay. Detailed information is given in Tables 1 and 2.

Incubation experiments were conducted to investigate CH_4 emission and production. Air-dried soil samples (10 g soil with and without 1% ground rice straw) were placed in 160-ml glass flasks, which were closed with a rubber septum to allow gas sampling from the headspace. The water : soil ratio was adjusted to 2:1. Before incubation, the headspace of each flask was purged with inert gas (He), to promote a quick anerobic environment. The flasks were incubated at 30 °C for 10 and 39 days. Six bottles from each treatment were removed at the end of each incubation period. Three of them were measured for CH_4 emission by sampling 1 ml of the accumulated gas in the headspace. The other three samples were shaken for 1 h to release CH_4 trapped in the soil. The

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Table 1. Main chemical characteristics of the soils studied

Location	Group	pH	OM (%)	H_2O-C (mg g^{-1})	FWC (%)	TKN (mg g^{-1})	CEC (mEq 100 g^{-1})	BR – (Mn + Fe) (mEq kg^{-1})
Barapani	1	5.3	1.28	0.69	9.29	0.75	3.53	25.8
Beaumont	1	5.4	2.29	1.86	14.00	1.18	27.32	28.8
Crowley	1	5.2	1.57	0.78	8.57	0.80	4.84	23.4
Desha	1	7.6	1.26	0.69	9.44	0.74	12.02	15.4
Dowling	1	6.6	2.38	1.52	11.01	1.52	36.55	37.2
Dundee	1	7.4	0.83	0.70	14.54	0.63	15.11	18.9
Sacramento	1	6.4	2.32	1.44	10.70	1.39	26.14	33.6
Sharkey	1	6.3	1.83	0.94	8.86	1.31	28.42	38.0
Tunica	1	7.3	1.54	1.04	11.64	1.11	25.90	28.5
Ballam	2	4.7	2.20	2.53	19.83	1.39	1.16	22.4
Coimbatone	2	8.1	1.44	0.93	11.13	0.78	27.26	11.2
Cuttack	2	5.7	1.02	1.07	18.27	0.63	9.03	21.3
Herbert	2	5.8	0.89	0.92	17.82	0.46	3.77	18.7
Ludhiana	2	6.5	0.72	0.86	20.59	0.52	3.50	8.1
Stockton	2	5.4	1.44	1.62	19.40	0.90	20.79	28.6
Yolo	2	7.0	2.00	1.49	12.84	1.29	21.10	16.4

OM, Organic matter; FWC, fraction H_2O-C of total organic C; TKN; total Kjeldahl N; CEC, cation exchange capacity; BR, biologically reducible. Significant differences in group means by *F*-test were found for the fraction of H_2O in total organic C ($P<0.01$) and biologically reducible Mn + Fe ($P<0.05$)

Table 2. Soil particle-size analysis

Location	Particle size (%)				
	1 – 0.05	0.05 – 0.01	0.01 – 0.005	0.005 – 0.001	<0.001
Ballam	63.92	5.15	2.58	2.58	25.77
Barapani	22.68	20.62	18.04	18.05	20.62
Beaumont	12.37	15.46	5.15	20.62	46.39
Coimbatone	48.45	6.45	6.45	2.58	38.45
Crowley	32.99	25.77	15.46	7.73	18.00
Cuttack	38.14	15.46	10.31	7.73	28.35
Desha	27.84	28.35	12.89	5.15	25.75
Dowling	16.53	0.50	0.50	28.35	54.12
Dundee	27.84	30.93	2.58	12.89	25.77
Herbert	71.65	2.58	2.58	7.22	15.98
Ludhiana	69.07	7.00	0.73	7.22	15.98
Sacrament	2.06	10.31	5.15	30.93	51.55
Sharkey	12.37	10.31	10.31	22.16	44.85
Stockton	9.79	10.31	2.58	21.65	55.67
Tunica	8.76	21.65	7.73	20.62	41.24
Yolo	20.10	7.73	28.35	18.04	25.77

same volume of gases accumulated in the headspace after shaking was sampled and measured. This result was considered as CH_4 production. The calculation used was: CH_4 entrapment (%) (I – CH_4 emission/CH_4 production) × 100.

The CH_4 concentration was analysed by a Perkin-Elmer 900 gas chromatograph equipped with a flame ionization detector, and expressed as CH_4 ng g^{-1} and μg g^{-1} of soil during 10 and 39 days of incubation, respectively.

The effects of urea fertilization on CH_4 emission were also studied in soils amended with 1% organic matter. The application rate of urea-N was 200 mg kg^{-1} of soil. The method described above was adopted, with soil samples incubated at 30 °C for 10 days.

We determined soil organic matter by the dry combustion method (Nelson and Sommers 1982) and total Kjeldahl N by distillation (Bremner and Mulvaney 1982). Soil cation exchange capacity was expressed as the sum of 1 *N* NH_4OAc (pH 7) extractable Na^+, K^+, Ca^{2+}, Mg^{2+}, and Al^{3+} analysed with inductively coupled argon plasma emission spectrometer (Atom Comp Series 800). Soil biologically reducible Fe and Mn were obtained by measuring 1 *N* NH_4OAc (pH 7) extractable Fe and Mn in soils which were anaerobically incubated (soil : water 1 : 2) for 10 days with the addition of 1% of rice straw at 30 °C. The filtrate was adjusted to pH 2 before the Fe and Mn analysis with ICP. The results are expressed as biologically reducible Fe and Mn (mEq kg^{-1}). Water-soluble C was measured by boiling 25 g soil for 2 h (water : soil ratio 4 : 1). The C in the filtrate was considered H_2O-C and was measured by an ultraviolet-promoted persulfate oxidation method (US Environmental Protection Agency method 415.2). Soil texture was measured by the hydrometer method. Soil pH and Eh were measured by a pH electrode and a platinum electrode with a calomel reference electrode, respectively.

Results and discussion

Methane emission from soils without rice straw amendment

Methanogenesis in soil is an energy transformation process, mediated by CH_4-generating bacteria. The amount

of soil organic matter and the decomposable C are generally considered critical controlling factors of CH_4 production. Increased CH_4 production following organic matter additions or temporary retardation of CH_4 production caused by fatty acids produced during organic matter decomposition have been observed in previous studies (Moraghan and Ayotade 1968; Yagi and Minami 1990). The decomposable C content in soil is considered to be more significantly related to CH_4 production than the total organic matter content because the latter provides instant energy to microorganisms. Van Cleemput et al. (1991) reported a linear relationship between H_2O-C and methane production.

Generally, the organic matter content of cultivated soils is considered an index of soil fertility. A cultivated soil with a relatively high amount of organic matter indicates a high level of microorganic activity. Therefore, a positive relationship between CH_4 emission and the soil organic C content if often expected.

Table 3 shows total CH_4 emitted from non-amended soils during 10 and 39 days of incubation. Variations in CH_4 emission in different soils were observed during both incubation periods (the range for 10 days of incubation was $2416-0=2416$ ng g^{-1}, and for 39 days $992-0=992$ µg g^{-1}). These observations reflected differences in the level of activity of methanogenic microorganisms in different soils, indicating differences in the CH_4 production potential of each soil. We attempted to correlate soil organic matter and H_2O-C contents with total CH_4 emitted during 10 and 39 days of incubation. Unexpectedly, neither a significant simple nor a significant multiple regression of CH_4 emission was established. This observation led us to stratify the 16 soils into different groups according to the amount of CH_4 emitted during the two incubation periods. Little CH_4 (less than 10 ng g^{-1}) was emitted during 10 days of incubation in 9 of the 16 soils (Table 3) and these soils are classified as group 1 (Fig. 1). The other seven soils (group 2) showed a emission range of $24-2416$ ng g^{-1} during the same period of incubation. At the end of 39 days, only three soils in group 1 (Beaumont, Dowling, and Tunica) showed a marked increase in CH_4 emission, as shown by the CH_4 emission ratio for the two periods (39 days: 10 days, Table 3). These results indicated that methanogenesis in the soils of group 1 was inhibited either for part or for the whole of the incubation period. The activity of CH_4-generating bacteria was therefore relatively low in these soils. More CH_4 emission was observed in the soils of group 2, indicating a higher methanogenic activity in these soils.

CH_4 formation in soil is a biological process mediated by obligate anaerobes at a soil redox potential lower than -140 to -160 mV (Jakobsen et al. 1981; Wang et al. 1993a). Before methanogenesis occurs, NO_3^-, Mn^{4+}, Fe^{3+}, and SO_4^{2-} in the soil must be reduced (Pannamperuma 1972; Patrick and Delaune 1977). All of these reductions are microbiological and energy-consuming. Therefore, they affect CH_4 formation to some extent. If a soil contains significant quantities of these oxidants, CH_4 formation is restricted, for the organic matter or the decomposable organic C would be first used by the other reducers than by the methanogenic bacteria. In this study, therefore, we measured biologically reducible Fe and Mn in the tested soils. The amounts varied greatly (Table 1). Most soils in group 2 contained more organic C or H_2O-C and less biologically reducible Fe + Mn than the soils in group 1. CH_4 formation in these soils, therefore, was not inhibited because the C source was available during the incubation period. Sufficient energy was provided both for the reduction of oxidants present in the soil and for CH_4 formation following this reduction.

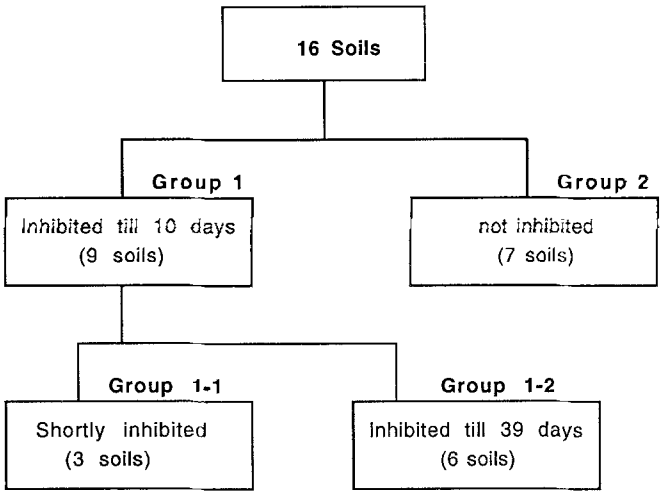

Fig. 1. Stratification of soils according to methane emission

We regressed CH_4 emission on organic matter, H_2O-C, biologically reducible Fe, biologically reducible Mn, biologically reducible Fe + Mn, total Kjeldahl N, and cation exchange capacity in the soils of group 2. The results showed a significant correlation between CH_4 and organic matter and the fraction of H_2O-C in the total

Table 3. CH_4 emission after 10 and 39 days of incubation

Group	Location	CH_4 (10 days) (ng g^{-1})	CH_4 (39 days) (µg g^{-1})	CH_4 ratio (39 days: 10 days)
1	Barapani	4.1	0.1	2.4×10^1
1	Beaumont	6.2	82.4	1.3×10^4
1	Crowley	0.9	0.1	1.1×10^2
1	Desha	0.9	1.1	9.9×10^2
1	Dowling	7.1	23.3	3.3×10^3
1	Dundee	2.3	0.2	8.7×10^1
1	Sacramento	2.0	0.3	1.5×10^2
1	Sharkey	2.1	0.1	4.7×10^1
1	Tunica	7.9	295.9	3.7×10^4
2	Ballam	142.0	921.5	6.5×10^3
2	Coimbatone	39.0	351.3	9.0×10^3
2	Cuttack	76.1	528.1	6.9×10^3
2	Herbert	98.2	620.0	6.3×10^3
2	Ludhiana	2416.0	518.0	2.1×10^2
2	Stockton	24.2	493.7	2.0×10^4
2	Yolo	152.2	448.3	2.9×10^3

F values by analysis of variance for group means of CH_4 emission were 2.06 ($P=0.173$) and 52.14 ($P<0.001$) for 10 and 39 days of incubation, respectively

organic C ($r = 0.93$*, $n = 7$, $P < 0.05$). This relationship is given in the following equation:

$$Y = -496.56 + 212.89X_1 + 79.21X_2 \quad (1)$$

where Y is CH_4 emission ($\mu g\ g^{-1}$), X_2 is organic matter content (%), and X_2 is the fraction of H_2O-C (%). The absolute values of the standard coefficients for X_1 and X_2 were 0.64 and 0.90, respectively. This indicates a bigger contribution from the percentage of H_2O-C in total organic C to CH_4 formation than from total organic matter. The other variables did not produce significant standard coefficients at $P < 0.05$, as tested by T values, and were excluded from the equation.

Although most of the soils in group 1-1 (Fig. 1) also contained relatively high levels of H_2O-C, the relatively high Fe and Mn levels in these soils inhibited CH_4 formation in the earlier period of incubation. The retardation of methanogenesis by soil Fe and Mn was lessened during the later period of incubation. This result indicates that after the reduction of soil Fe and Mn, sufficient C source remained available for CH_4 formation in these soils.

Most soils in group 1-2 were characterized by very low levels of H_2O-C ($0.69-0.78$ mg kg^{-1}). Methanogenesis did not seem to be induced significantly over the incubation period. The low level of soil decomposable C may have contributed to this observation.

Soil amended with rice straw

Soil Eh and pH

Table 4 shows CH_4 emission, soil pH, and soil Eh after 10 days of incubation in soils amended with 1% ground rice straw.

The addition of rice straw increased CH_4 emission greatly during the first 10 days of incubation compared with the CH_4 emission in non-amended soils, as shown in Table 3. However, the increases varied among soils. Following the addition of 1% rice straw, the supply of soil decomposable C should be no longer a limiting factor for CH_4 production, at least over a short period of incubation. This raised the question, therefore, of what soil variables could be contributing to the observation. We regressed the CH_4 emitted during 10 days of incubation in amended soils on the CH_4 emitted during 39 days of incubation in non-amended soils and found significant correlation ($r = 0.89$***, $n = 16$, $P < 0.001$). The relationship can be described by the following equation:

$$Y = -83.11 - 0.65X_1 + 0.05X_2 \quad (2)$$

Where Y is the CH_4 emitted during 10 days of incubation in soils amended with rice straw ($\mu g\ g^{-1}$), X_1 is the CH_4 emitted over 39 days in soils not amended with rice straw ($\mu g\ g^{-1}$), and X_2 is the soil Eh (mV). The absolute values of the standard coefficients of the variables X_1 and X_2 were 0.52 ($P < 0.001$) and 0.51 ($P < 0.002$), respectively.

Soil Eh decreases after the submergence of a soil following the consumption of O_2 and the reduction of NO_3^-, Mn^{4+}, Fe^{3+}, and SO_4^{-2}, and soil pH tends to approach neutral. We regressed soil Eh on biologically reducible Fe + Mn and pH, and found a significant relationship among these variables ($r = 0.73$***, $n = 16$, $P < 0.01$). The absolute values of the standard coefficient for biologically reducible Fe + Mn and pH were 0.34 and 0.59, respectively, indicating that soil pH had the stronger influence. The relationship is given by the following equation:

$$Y = 848.85 + 0.99X_1 - 148.81X_2 \quad (3)$$

where Y is soil Eh, X_1 is biologically reducible Fe + Mn, and X_2 is pH.

Figure 2a shows the relationship between CH_4 emission and soil pH measured in air-dried soils. As shown in Table 1, soil pH ranged from 4.7 to 8.1 in these 16 soils. There was scarcely any relationship between oxidized soil pH and CH_4 emission. However, soil pH converged to 7 after 10 days of anaerobic incubation, with a tendency for a near neutral pH ($6.9-7.2$) to be the optimum pH for CH_4 formation (Fig. 2b).

Conflicting observations on the effect of urea fertilization on CH_4 emission have been reported by many researchers. Schütz et al. (1989) reported a reduction in CH_4 emission after the incorporation of $(NH_4)_2SO_4$ or urea into the soil. Another report showed a stimulatory effect of urea fertilization on CH_4 emission (Lindau et al. 1991). We found that the effect of urea fertilization on CH_4 formation differed from soil to soil, and seemed to be related to its effect on soil pH. As shown in Table 5, in most acidic soils, the addition of urea stimulated CH_4 emission (compared with the CH_4 emission presented in Table 4), whereas CH_4 emission was retarded almost in all the non-acidic and alkaline soils.

It is well known that soil pH increases with the addition of urea fertilizer. In the acidic soils of the present study urea fertilization promoted a quick increase in soil pH. Previous work in the laboratory had shown that urea fertilization increased the pH of Crowley flooded rice soil

Table 4. Methane emission, soil pH and redox potential (Eh) after 10 days of incubation in soils amended with rice straw

Location	CH_4 ($\mu g\ g^{-1}$)	pH	Eh (mV)
Ballam	102.5	7.05	−188
Barapani	9.6	6.88	−142
Beaumont	40.2	7.08	−180
Coimbatone	55.1	7.22	−200
Crowley	10.4	6.80	−142
Cuttack	68.6	7.09	−189
Desha	22.8	6.98	−160
Dowling	43.1	7.08	−190
Dundee	14.2	7.07	−145
Herbert	63.5	7.08	−195
Ludhiana	58.1	7.08	−210
Sacramento	13.7	6.97	−143
Sharkey	11.4	7.05	−160
Stockton	42.4	7.01	−160
Tunica	50.2	7.18	−186
Yolo	111.3	7.04	−221

CH_4 values are means of triplicate assessments; pH and Eh values are means of duplicate assessments

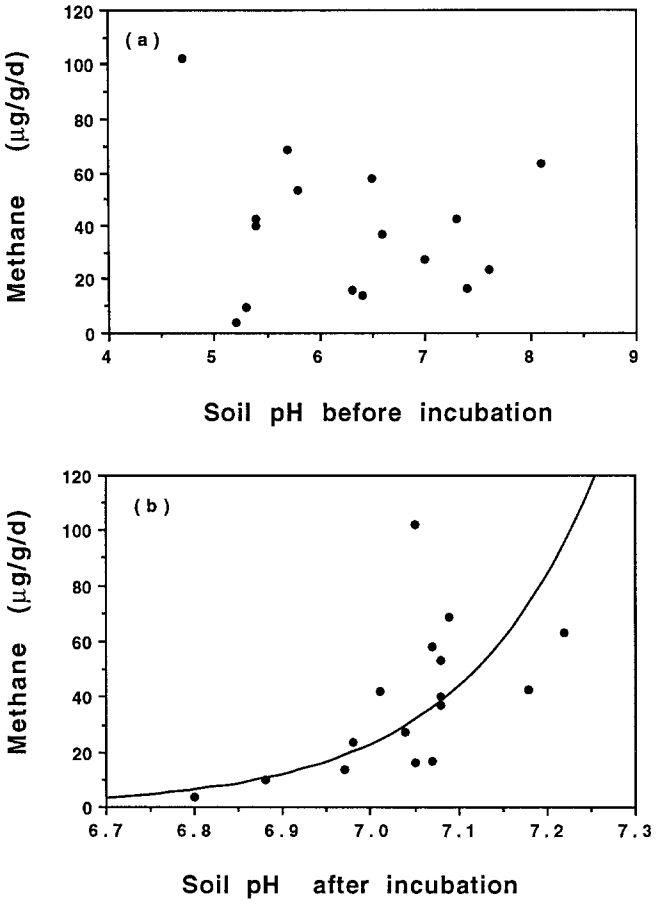

Fig. 2. Soil pH and methane emission in soils

Table 5. CH_4 emission in soils fertilized with urea and the relationship with oxidized soil pH

Location	CH_4 ($\mu g\, g^{-1}$)	pH	Effect of urea
Ballam	142.0	4.7	+
Barapani	15.8	5.3	+
Beaumont	54.1	5.4	+
Coimbatone	40.0	8.1	−
Crowley	13.9	5.2	+
Cuttack	111.2	5.7	+
Desha	5.2	7.6	−
Dowling	50.0	6.6	+
Dundee	36.2	7.4	+
Herbert	70.5	5.8	+
Ludhiana	83.6	6.5	−
Sacramento	18.9	6.4	+
Sharkey	34.7	6.3	+
Stockton	55.1	5.4	+
Tunic	39.8	7.3	−
Yolo	67.3	7.0	−

Effect of urea: CH_4 emission increased (+) and decreased (−) by urea application. For other explanations, see footnotes to Table 4

by more than 1 unit compared with the unfertilized soil after 1 week of urea fertilization (Wang et al. 1993b). This indicated that neutral soil pH, which is the optimum pH for soil methanogenesis, was promoted when urea was applied to the acidic soils before the pH was stabilized at neutral after submergence. However, in the non-acidic or alkaline soils, the increases in soil pH with urea fertilization could only inhibit methanogenesis. As CH_4 formation is critically controlled by soil pH, the variable effect of urea fertilization on CH_4 is influenced by its effects on soil pH.

Table 6. CH_4 entrapment after 10 days of incubation

Location	CH_4 entrapment (%)
Ballam	6.6
Barapani	0
Beaumont	80.6
Coimbatone	29.5
Crowley	94.4
Cuttack	40.2
Desha	0
Dowling	78.9
Dundee	7.2
Herbert	0
Ludhiana	12.1
Sacramento	67.8
Sharkey	98.5
Stockton	72.5
Tunica	52.6
Yolo	53.3

Table 7. Correlation coefficients for CH_4 entrapment and soil particle size

Soil particle sizes (mm)	r
1 − 0.05	−0.676**
0.05 − 0.01	−0.0359
0.01 − 0.005	−0.077
0.005 − 0.001	0.767**
< 0.001	0.863***

** $P<0.01$, *** $P<0.001$

The effect of soil physical properties on CH_4 entrapment

As shown in Table 6, CH_4 entrapment varied greatly from soil to soil, ranging from 0 to 99%, and was apparently affected by soil texture. Soils with a clayey texture entrapped more CH_4. We correlated CH_4 entrapment with soil particle size (Table 7) and found a significant negative correlation with soil particles of 1 − 0.05 mm, but a positive correlation with the clay (particle sizes 0.005 − 0.001 and < 0.001 mm) content of soil. Soil particles of 0.05 − 0.01 and 0.01 − 0.005 mm did not show a significant relationship to CH_4 entrapment. The more clay the soil had, the more CH_4 was trapped.

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