

An Empirical Model of Soil Chemical Properties that Regulate Methane Production in Japanese Rice Paddy Soils

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To understand which soil chemical properties are the best predictors of CH₄ production in rice paddy soils, a model was developed with empirical data from nine types of rice soils collected around Japan and anaerobically incubated at 30°C for 16 wk in laboratory conditions. After 1, 2, 4, 8, and 16 wk of incubation, CO₂, CH₄, and Fe(II) were measured to understand soil organic matter decomposition and iron (Fe) reduction. Available N (N_{ava}) was also measured at the end of incubation. The results showed that decomposable C and reducible Fe are two key parameters that regulate soil CH₄ production (P_{CH₄}). There was a significant relationship between decomposable C and available N (N_{ava}) ($r^2 = 0.975^{**}$). Except for a sandy soil sample, a significant relationship between total Fe (Fe_{total}) and reducible Fe was found. From this experiment, a simple model of soil CH₄ production was developed: $P_{CH_4} = 1.593N_{ava} - 2.460Fe_{total}/1000$ (each unit was mg kg⁻¹ soil). After simulated CH₄ production by two soil chemical properties as above, there was a significant consistency between model simulation and actual measurement ($r^2 = 0.831^{**}$).

ATMOSPHERIC methane (CH₄) is one of the most important greenhouse gases because it is responsible for approximately 20% of the anthropogenic global warming effect. Most atmospheric CH₄ is produced by microbial activity in extremely anaerobic ecosystems, such as natural and cultivated wetlands, sediments, sewage, landfills, and the rumen of herbivorous animals. Rice paddies account for about 17% of the anthropogenic CH₄ sources (IPCC, 1995). The amount of CH₄ produced and emitted from flooded rice soil is primarily determined by the availability of methanogenic substrates and the influence of environmental factors. The environmental factors affecting soil CH₄ production include soil properties, climate, and agricultural practices (Mer and Roger, 2001).

Theoretically, CH₄ production in flooded rice soils begins when more energetically favorable terminal electron acceptors, such as NO₃⁻, Mn(IV), Fe(III), and SO₄²⁻, have been completely reduced as a result of organic matter decomposition, at which point the soil Eh has typically decreased to -150 mV (Inubushi et al., 1984; Wang et al., 1993). In the processes of microbial decomposition of soil organic matter, organic matter or its fermentation product H₂ are the electron donors, and NO₃⁻, Mn(IV), Fe(III), and SO₄²⁻ are inorganic electron acceptors (Takai, 1970; Inubushi et al., 1984). Because reducible Fe(III) is one of the most abundant electron acceptors in paddy soils, it plays a key role in the regulation of CH₄. An important role is also played by abundance of labile soil organic matter as the electron donor (Gaunt et al., 1997; Wasmann et al., 1998; Yao et al., 1999; Mer and Roger, 2001; Huang et al., 2002; Mitra et al., 2002; Van Bodegom et al., 2003).

Rice is harvested on 150 million ha globally, with about 90% grown in Asia (IRRI, 1993). Methane released from paddies is released through the rice plants, and net CH₄ emission is determined by the balance between CH₄ production and CH₄ oxidation in rice plant and paddy soil ecosystems (Mer and Roger, 2001). The ability to model CH₄ emissions from rice paddy soils is limited by an understanding of the interaction between soil properties and CH₄ production. To estimate CH₄ emission from different regions with different soil types and to evaluate mitigation options for CH₄ emis-

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sion from paddy soils, it is necessary to identify soil parameters that are predictive of CH₄ production. The objective of this study was to relate various soil chemical properties to CH₄ production in a wide variety of Japanese rice paddy soils.

Material and Methods

Soil Collection

Nine kinds of soils were collected from seven prefectures of Japan (Table 1, latitude from 35°01' N to 39°41' N and longitude from 135°37' E to 141°00' E). One soil was an Andisol, five were Inceptisols, and three were Entisols. The Andisols, Inceptisols, and Entisols represent 10, 37, and 31% of all Japanese paddy fields, respectively (Kyuma et al., 1984). Soils were collected from the plow layer (top 20 cm) of the rice fields before rice transplanting season. All soils were air-dried and sieved (2 mm) before the experiment.

Soil Analysis and Pre-incubation

Air-dried soils were used to measure total C and N, soil bulk density, pH, available Fe, and total Fe and Mn by standard methods according to soil normal analysis methods (JSSSPN, 1986). Total C and N were analyzed by a CN-900 Analyzer (Sumika Chemical Analysis Service). Available Fe was analyzed by the methods of Asami and Kumada (1959). Total Fe and Mn were extracted by treatment with HF and HClO₄, and analyzed by inductively coupled plasma–optical emission spectroscopy (ICP–OES) (Varian VISTA-PRO).

All soil samples were pre-incubated at 40% WFPS (water-filled pore space) for 2 wk before the anaerobic incubation experiment started. Water filled pore space was calculated as: WFPS = [(gravimetric water content × soil bulk density)/total soil porosity], where soil porosity is [1 – (soil bulk density/2.65)] and 2.65 is the assumed particle density of the soil (Aulakh et al., 1991; Cheng et al., 2004).

After 2 wk of pre-incubation, the mineral N (NH₄⁺-N and NO₃⁻-N) and ferrous iron (Fe(II)) in all soil samples were determined. Ammonia and NO₃⁻ in soil were extracted with 10% KCl solution for 30 min. The extracted solutions were filtered and stored in a deep freezer (–18°C) until analysis. Ammonia and NO₃⁻ concentrations were measured by a colorimetric method, using TRAACS 2000. Iron(II) was extracted with 1 mol L⁻¹ pH 2.8 acetic acid (5 g soil by 50 mL solution) and measured by the method of Asami and Kumada (1959).

Anaerobic Incubation Experiment

Each soil sample (5 g on an oven-dried basis) that was pre-incubated at 40% WFPS for 2 wk was weighed into a 68-mL serum bottles and amended with 10 mL distilled water. All bottles were capped with a butyl rubber stopper and the air replaced with pure N₂ gas by vacuum and gas replaceable equipment (Freezemobile and Unitop HL, Virtis Company, New York). All bottles were incubated at 30°C. After 1, 2, 4, 8, and 16 wk of incubation, three replicate bottles for each soil sample were taken from the incubator, first for measuring headspace concentrations of CO₂ and CH₄. Iron(II) production in submerged soil samples with 10 mL submerged water was determined after extraction with 40 mL

Table 1. Collection sites and characteristics of nine kinds of Japanese paddy soil samples.

No.†	Soil classification	Management‡	Collection site	Latitude, longitude	Bulk density Mg m ⁻³	Total C g kg ⁻¹	Total N g kg ⁻¹	C/N	pH (H ₂ O)	Total Fe g kg ⁻¹	Total Mn g kg ⁻¹	Available Fe g kg ⁻¹	NH ₄ ⁺ -N mg N kg ⁻¹	NO ₃ ⁻ -N mg N kg ⁻¹
1	Andisols	Convention	Iwate	39°40' N, 141°00' E	0.649	80.3	5.33	15.08	6.08	39.5	0.6	8.83	47.0	103.5
2	Inceptisols	Convention	Yamagata	38°16' N, 140°19' E	0.933	21.6	1.70	12.69	5.91	53.3	1.0	10.11	11.0	16.1
3	Inceptisols	Manure appl.	Yamagata	38°16' N, 140°19' E	0.937	25.8	1.96	13.21	6.58	53.6	0.9	10.58	15.4	16.0
4	Entisols	drainage equipped	Niigata	37°26' N, 138°53' E	0.893	18.9	1.69	11.18	5.75	36.6	0.6	5.63	30.7	35.5
5	Entisols	Convention	Niigata	37°26' N, 138°53' E	0.773	27.1	2.43	11.14	5.20	38.0	0.5	5.84	70.8	3.8
6	Inceptisols	Convention	Fukushima	37°24' N, 140°21' E	0.989	17.7	1.56	11.37	6.61	40.6	0.6	8.64	20.7	10.1
7	Entisols	Convention	Chiba	35°33' N, 140°28' E	1.251	8.1	0.86	9.42	6.24	35.3	0.7	2.68	4.1	56.6
8	Inceptisols	Convention	Kyoto	35°01' N, 135°37' E	1.011	17.4	1.70	10.20	7.12	30.9	1.1	7.94	0.7	57.6
9	Inceptisols	Manure appl.	Kyoto	35°01' N, 135°37' E	1.013	22.5	2.14	10.49	7.12	29.6	1.0	7.85	1.3	80.7
Means‡					0.939	26.6	2.15	11.64	6.29	39.7	0.8	7.57	22.4	42.2
CV (%)§					17.8	78.6	58.8	15.0	10.1	21.7	28.1	32.7	105.2	82.1

† Number was according collection sites listed from north to south of Japan; soils were named as U.S. Soil Taxonomy.

‡ The different managements for No. 3, 9, and 4 indicate a soil sample from paddy fields with cattle manure application and drainage equipment.

§ Mean and CV (coefficient of variation) were for all nine kinds of soils.

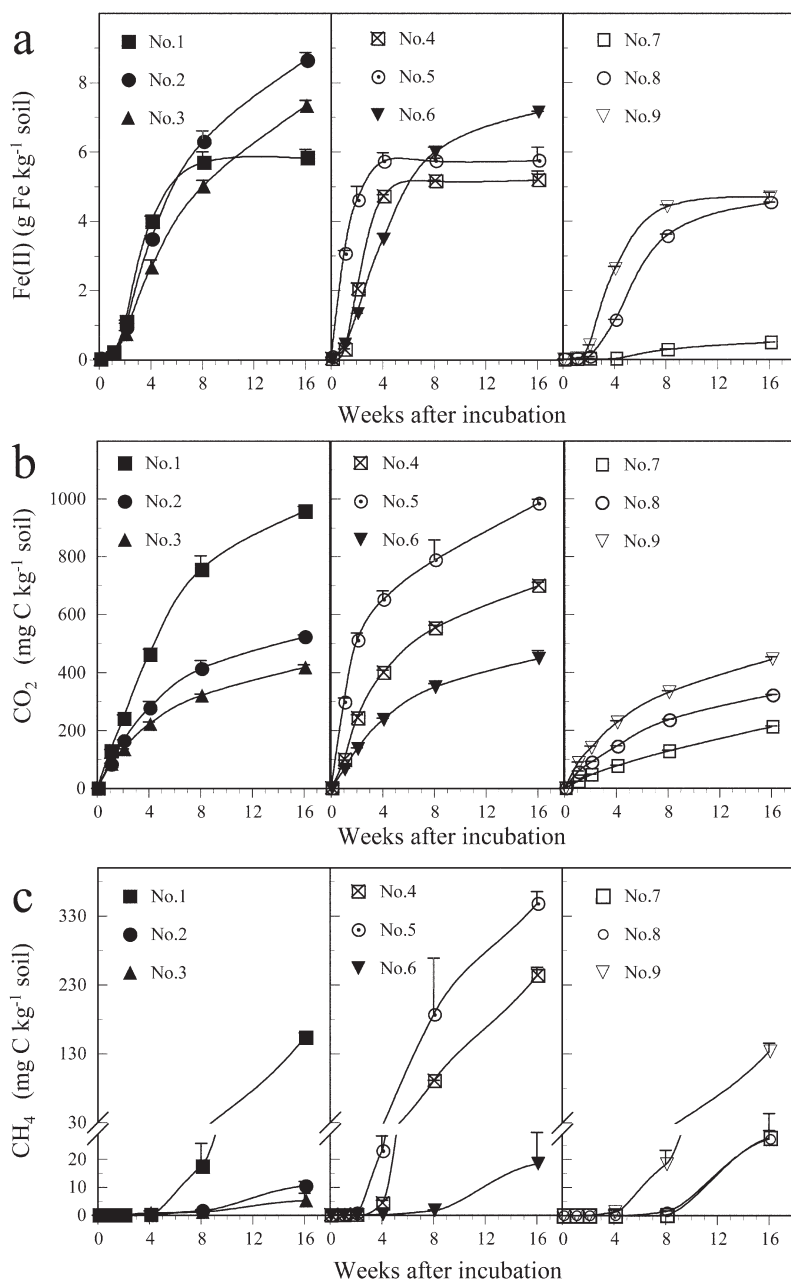


Fig. 1. The changes in amounts of Fe(II) (a), CO₂ (b), and CH₄ (c) from nine kinds of soils during 16 wk anaerobic incubation. Soil numbers were the same as in Table 1. Bars indicated standard deviations from three replicates.

acetic acid solution (1.25 mol L⁻¹ pH 2.8). The last three replicates for each of the soil samples were extracted with 10% KCl solution for 30 min to measure NH₄⁺ production after 16 wk of incubation.

Carbon dioxide and CH₄ concentrations were determined on a gas chromatograph (GC-7A; Shimadzu) equipped with thermal conductivity and flame ionization detectors, respectively (Cheng et al., 2005).

Calculations

Carbon dioxide and CH₄ concentrations in the headspace were used to calculate CO₂ and CH₄ production after correcting for gaseous dissolution in the incubation water by Bunzen's

law (Cheng et al., 2005). Methane consumption was assumed to be negligible in the highly anaerobic incubations; therefore, CH₄ fluxes were described as CH₄ production throughout this study. Decomposed carbon was calculated as CO₂-C and CH₄-C during the 16-wk incubation. We defined Fe(II) produced after the 16-wk incubation as reducible Fe and NH₄⁺ produced after the 16-wk incubation (corrected for the pre-incubation [NH₄⁺]) as available N.

Results and Discussion

Properties of Nine Japanese Agricultural Soils

The properties of the nine kinds of soils from seven prefectures of Japan varied widely (Table 1). The Andisols had the highest total C (80.3 g kg⁻¹) and the lowest bulk density, while one Entisol (a sand paddy with a suborder name of Psammets) had the lowest total C (8.1 g kg⁻¹) and the highest bulk density. As expected, total C and N were closely related ($r^2 = 0.984^{**}$). Mean soil pH (in water) was 6.29 with a coefficient of variation (CV) of 10.1%, showing that all soils were neutral or acidic. The mean contents of total Fe and total Mn were 39.7 and 0.8 g kg⁻¹ soil with CV of 21.7 and 28.1%, respectively. Mean available Fe was 7.57 g kg⁻¹ soil with a CV of 32.7%. Mean NO₃⁻-N was 42.2 mg kg⁻¹ soil with a CV of 105.2%. Among these compounds, NO₃⁻, Mn(IV), and Fe(III) are used as terminal electron acceptors for microbial respiration (Takai, 1970; Inubushi et al., 1984). Concentration data would suggest that Fe is the most important of these electron acceptors, because the amount of total Fe is larger than total Mn by a factor of 50 and NO₃⁻ by a factor of 1000 (Table 1).

Iron(II), Carbon Dioxide, and Methane Production during Anaerobic Incubation

Cumulative increases in Fe(II) and CO₂ (Fig. 1a and b) indicated that production rates declined over the course of the 16-wk incubations. Headspace CO₂ accumulated rapidly in soils 1, 4, and 5, and more slowly in soils 7 and 8. This pattern in CO₂ production among the nine soils was similar to the pattern of Fe(II) produced by the respiration of Fe(III)-reducing bacteria. Initial rates of Fe(II) production were highest in soil 5. In soils 1, 4, 5, and 9, Fe(II) production ceased after 8 wk of incubation; in soil 5 production ceased after 4 wk of incubation (Fig. 1a). High rates of CH₄ production coincided with the cessation of Fe(III) reduction in soils 1, 4, 5, and 9 (Fig. 1a and c). Also, the CO₂ and CH₄ production ratio (in molar units) began to trend toward 1:1 after Fe(III) was completely reduced (data not shown).

Carbon and Nitrogen Mineralization after 16 wk of Anaerobic Incubation

Carbon dioxide and CH₄ originate from the decomposition and fermentation of soil organic carbon. After the 16-wk incubation, the total decomposed C (CO₂-C + CH₄-C) was significantly different among the soils ($P < 0.01$). There was not a significant relationship between total decomposed C and total C ($r^2 = 0.328$). Mean mineralized N averaged 123.62 units with a CV of 59.9%. The mean ratio of total decomposed C to mineralized N was 5.62 with a low CV of 19.1%, which is comparable to Redfield ratio (C/N = 5.67) and soil microbial biomass C and N ratio in rice paddies reported by Shibahara and Inubushi (1995). Compared to the mean total C/N (11.64, Table 1), the ratio of decomposed C to mineralized N in rice soil was lower than that of the bulk soil organic matter pool, which indicated that the easily decomposed organic matter was from soil microbial biomass and humified organic matter in the soils. Also, the relationship between total decomposed C and mineralized N was significant (Fig. 2, $r^2 = 0.975^{**}$).

Relationships among Total Iron, Available Iron, and Reducible Iron

Relationships among total Fe, available Fe, and reducible Fe were shown in Fig. 3. Available Fe measured by the method of Asami and Kumada (1959) was almost equal to reducible Fe in soils 4 and 5, two Entisols samples (Table 1). There was a strong linear relationship between total Fe and reducible Fe that held for all the soils except soil 7 ($r^2 = 0.837^{**}$ without soil 7). We cannot fully explain this result because the total Fe content was similar to the other soils, but soil 7 was a sand paddy with the lowest organic matter content in the group (8.1 mg kg⁻¹, Table 1). Because soil organic matter can stabilize Fe minerals in a poorly crystalline (i.e., reducible) form (D'Angelo, 2005), the low soil organic matter content of soil 7 could have allowed iron minerals to crystallize more completely than in the other soils. Additionally, sand paddy occupies a very small area in Japan for cultivated rice compared with the other soil types.

Parameterized Soil Chemical Properties for Evaluating Soil Methane Production

The goal of this study was to develop predictive relationships between soil CH₄ production and the soil characteristics we measured. Based on our results, we reasoned that CH₄ production would occur only when the amount of labile organic carbon (i.e., electron donor) exceeded the amount of reducible Fe(III) (i.e., the primary terminal electron acceptor). We used a ratio of 1 mole carbon mineralized to 4 moles of Fe reduced to convert Fe(III) to carbon units. A significant relationship was found between CH₄ production and difference of decomposed C and Fe(II) production (Fig. 4a, $r^2 = 0.888^{**}$) and is expressed as a simple equation below:

$$P_{\text{CH}_4} = 0.302(C_{\text{edc}} - 1/4\text{Fe}_{\text{red}}) \text{ (in mol units)} \quad [1]$$

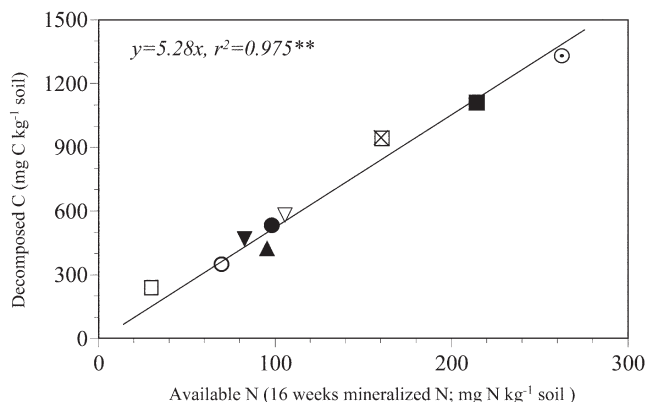


Fig. 2. Relationship between decomposed C (CO₂-C + CH₄-C) and available N (mineralized N) after 16 wk incubation. The ** indicates significant correlations at $P < 0.01$. The symbols represent different soils, as in Fig. 1.

where P_{CH_4} is CH₄ production, C_{edc} is easily decomposable C, and Fe_{red} is reducible Fe.

Available N can be referred to from the database in Japan, while the easily decomposable C cannot. Easily decomposable C had the following simple zero-order relationship with available N (Fig. 2):

$$C_{\text{edc}} = 5.28 N_{\text{ava}} \text{ (in mass units)} \quad [2]$$

where N_{ava} is mineralized or available soil N. So we use N_{ava} to substitute C_{edc} in Eq. [1].

Usually, the total Fe is reported in soil databases, while available Fe is not. Therefore, total Fe should be a more useful parameter than available Fe for estimating CH₄ production at a landscape scale. Reducible Fe also can be predicted with a simple zero-order equation (Fig. 3) as:

$$\text{Fe}_{\text{red}} = 0.152 \text{Fe}_{\text{total}} \text{ (in mass units)} \quad [3]$$

where Fe_{total} is total Fe of the soil.

Combining Eq. [2] and [3], CH₄ production can be expressed as:

$$P_{\text{CH}_4} = 1.593N_{\text{ava}} - 2.460\text{Fe}_{\text{total}}/1000 \text{ (units of mg kg}^{-1}\text{)} \quad [4]$$

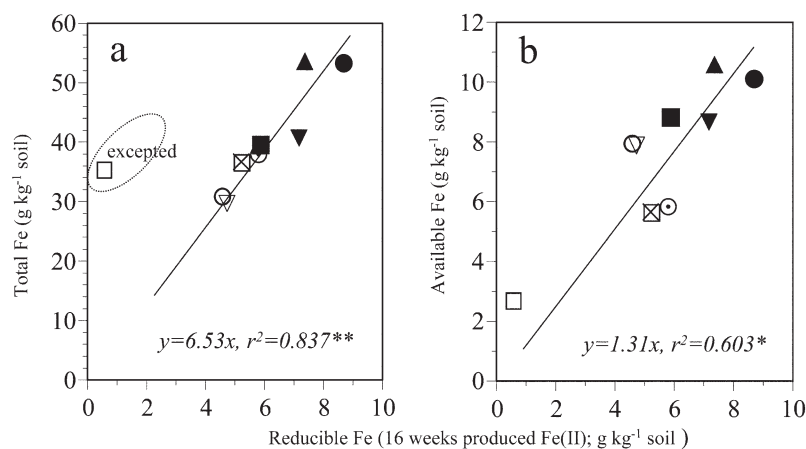


Fig. 3. Relationships between reducible Fe and total Fe (a), and between reducible Fe and available Fe (b). The * and ** indicate significant correlations at $P < 0.05$ and 0.01 . The symbols represent different soils, as in Fig. 1.

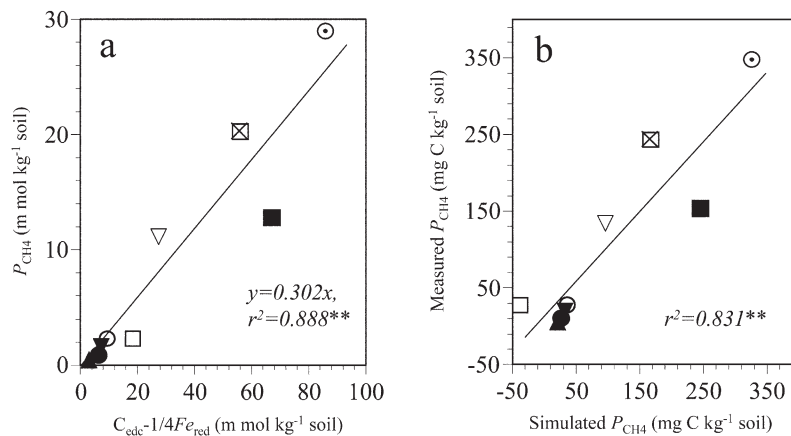


Fig. 4. Relationship between CH_4 production and difference from decomposed C to reducible Fe (a); and between CH_4 production by measurement and by simulation through a simple model as: $P_{\text{CH}_4} = 1.593N_{\text{ava}} - 2.460Fe_{\text{total}}/1000$ (b). The ** indicate significant correlations at $P < 0.01$. The symbols represent different soils, as in Fig. 1.

The two soil properties, available N and total Fe, were strong predictors of CH_4 production (Fig. 4b, $r^2 = 0.831^{**}$).

Conclusions

Several previous studies have related soil properties to CH_4 production for the purpose of simulating CH_4 production and emission from rice fields (Wassmann et al., 1998; Huang et al., 1998; Yao et al., 1999; Mitra et al., 2002; Yan et al., 2003; Li et al., 2004). However, many of the soil properties that have been measured, such as redox potential, reducible Fe and Mn, and decomposable C, are not easily extrapolated to the landscape scale because they are not reported in soil databases. The purpose of our laboratory incubation experiment was to relate CH_4 production to soil parameters that are more readily available in such databases. The results showed that CH_4 production is positively related to available N and reducible Fe in a group of nine Japanese soils that collectively represent most of the paddy soils in Japan. Reducible Fe was significantly related to total Fe. From this experiment, a simple CH_4 production model was proposed (Eq. [4]).

It should be noted that our results were from a simple laboratory experiment without rice plants. The only source of decomposable C was from the soil organic matter, while in rice paddies, most decomposable C comes from rice plant residues and root exudates. Therefore, a field-based model should be developed based on the parameterization of soil chemical properties.

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