



Source and Sink Strength of Carbon Dioxide, Methane and Distribution of Sulfate in Salt-marsh Soils at the Wadden Sea Coast of Northern Germany

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ABSTRACT: A field study was conducted at Schleswig-Holstein of Kiel in Germany to evaluate the factors controlling carbon and sulfate dynamics along a toposequence of coastal salt marsh soils. The soil at the top end of the salt marsh was salic silty to clayic Typic Sulfaquent (Salzrohmarsh) and the bottom end was sandy to silty Haplic Sulfaquent (Übergangsmischwatt). The mean (depth: 0-100 cm) values of pH and rH were 6.8-6.9; 6.8-7.0 and 17.3-8.1; 15-8.6 for the Typic Sulfaquent (TS) and Haplic Sulfaquent (HS) throughout the year, respectively. The net-emission of CO₂ was negative (-14.0 g m⁻² yr⁻¹) for the HS but highly positive (857 g m⁻² yr⁻¹) for the TS throughout the year. The annual emissions of CH₄ were almost 10 fold higher in HS (0.3 g m⁻² yr⁻¹) than that of the TS (0.03 g m⁻² yr⁻¹). The concentrations of CH₄ at different seasons showed almost inverse relationships with the concentrations of CO₂, varied significantly (p<0.05) with the seasons and depth function, and showed no dependence to temperature. The SO₄ contents were observed maximum in the TS than that of HS during all the seasons. There was no noticeable correlation (r=-0.09) between SO₄ and CH₄ concentrations. Moreover, even CH₄ was determined at depths where the SO₄ concentration was about 1200 mg SO₄ L⁻¹. @JASEM

Trace gases are becoming more and more interesting due to their possible contribution to an enhance greenhouse effects and global warming. The abundance of CH₄ has increased by 150% to 1.74 ppm and CO₂ by 31% to 367 ppm (IPCC, 2001) in the atmosphere since 1750. Methane contributes approximately 25% of global warming and closer to 10% of annual global emission and masks the mitigation potential induced by the reduction in CO₂-C emission (Mu et al., 2006). Soils are major global sources and sinks of methane and play an important role in regulating atmospheric CH₄ (Nakano et al., 2006). The fluxes of carbon from soil are an important part of the terrestrial carbon budget and the global carbon cycle. Coastal soils are emittants of CH₄ besides CO₂ due to having the oxidation-reduction processes as a result of frequent tidal influences but the extend of those fluxes to atmosphere is not yet clear.

There were large CH₄ concentrations in salting and in sulfate enriched hyper saline environments (Giani et al., 1996). Moreover, as soils differ in their characteristics, it is expected that soil CO₂ and CH₄ emissions also differ according to particular properties of the soils. Carbon dioxide develops under oxic as well as under anoxic soil conditions, while methane is produced in anaerobic environments through either CO₂ reduction. It is obvious that the factors affecting CH₄ and CO₂ emissions are complicated and interrelated.

The redox potentials of less than -200 mV are the favorable condition for CH₄ formation (Conrad, 1989). On the other hand, Holmer and Kristensen (1994) suggested that there is a coexistence of sulfate reducing and methane producing bacteria. Coastal salt marsh soils have high CH₄ concentration despite of the higher SO₄, play an important role in sequestration of carbon and emissions of trace gases (Schipper and Reddy, 1994). Investigations on the sources, sinks and factors regarding the trace gas emissions and their concentrations throughout the soil profiles especially in coastal or young marine soils are still insufficient. Accordingly, this investigation was done to evaluate the sources and sinks strength, and factors regulating the fluxes of carbon and sulfate dynamic in the coastal salt marsh soils of Northern Germany.

MATERIALS AND METHODS

Field study was conducted at Soenke-Nissen-Koog in Schleswig-Holstein of Kiel in Germany during 2000-2001. The possible factors (Table 1) controlling the parameters regarding C-dynamics along a toposequence of daily to seasonally flooded coastal salt marsh soils were studied. The toposequence of the site is divided into two sectors (Table 1). The soil at the top end of the salt marsh was classified as salic silty to clayic Typic Sulfaquent (Salzrohmarsh), which is flooded (with a height of 1.8 m above sea level: a.s.l. or 0.15 m above mean high tide) for about 180 times in a year. This sector is covered with small (3-20 cm) vegetation (*Puccinellia maritima* along with some

Halophytes), which is extensively grazed by sheep during summer. While the soil at the bottom end of the site is flooded (1.4 m a.s.l. or 0.25 m below mean high tide) for about 700 times in a year and the vegetations were mainly *Spartina anglica* and *Salicornia europea*. The soil in this section was classified as sandy to silty Haplic Sulfaquent (Übergangsmischwatt: Table 1).

Emissions of CO₂ and CH₄ from the surfaces of soils were measured in each month starting from May to October of the year. The data are presented based on the different seasons prevailing during May (Spring), June (Summer), August (Autumn) and September (Winter) of the year. The samples were collected from six spots as replications and then analyzed following chamber (made by acrylic glass with a base of 990 cm² and a content of 27 L) method (Hutchinson and Mosier, 1981). Concentrations of CO₂ and CH₄ in the soils and distribution of SO₄ were investigated at different depths of the soil profiles by using aluminum-tube with a diameter of 10 cm. The tubes were installed by pressing (somewhere drilling) towards the depths of about 100 cm as required leaving no space inside the tube. The soil monolith inside the Al-tube was sectioned at every 10 cm and the process of evacuation then collected the gases through a 100 mL syringe. The gas samples were analyzed with a Gas Chromatograph (Shimadzu GC-14 with LC10) equipped with FID and ECD. The SO₄ concentration was determined by the BaCl₂ method. Temperature sensors measured soil temperatures during 2000-2001 and mean annual temperature was determined from this measurement. The redox potentials were determined with stationary platinum electrodes against Ag/AgCl-electrodes at different depths of the studied soils.

RESULTS AND DISCUSSION

Redox potential and Soil temperature: The mean values of redox potentials (Eh) in the Typic Sulfaquent (TS) were ranged from -66 to +104 and were negative towards the deeper depth during all the seasons, except for the surface soils during summer, where the mean value was +104 mV (Table not shown). The standard deviations (SD) were also varied widely, especially summer and autumn. Mostly the Eh in the upper 3 cm of the surface soils of Haplic Sulfaquent (HS) were varied from 12 to 33 mV. The Eh values in the subsoils (50-100 cm) of both the TS and HS indicated that the methanogenesis can be occurred in the soils. The rH values (Table 1) in the epipedon of the HS and TS were ranged from 14.4 to 17.3 (corresponding with a grayish green to olive color of the soil matrix together with reddish brown holes), and in the sub-soils from 8.1 to 11.9 (corresponding with a black soil color due to sulfide). The rH values < 13 normally corresponds with a black color due to the formation of sulfide, and values < 10 make possible methane formation, whereas the values in between 13-20, Fe³⁺ can be reduced and FeII/FeIII oxides led to the formation of so called blue green rust, together with reddish brown ferrihydrite along holes of soil animals and plant roots. In the present study more than 500 mg SO₄ kg⁻¹ were determined in the surface soils of all the profiles in different seasons (Fig. 1), which might be due to the frequent addition of SO₄ from the highly SO₄ enriched sea waters. The rH values of the soils were found to have maintained the established relationships (Schlichting et al., 1995) of CH₄ productions and/or concentrations and emissions of the soils as reflected by figure 1 and table 2.

Table 1: Selected physico-chemical properties of salt marsh soils at the Wadden sea coast of North Germany. Typic Sulfaquent (Typische Salzrohmarsh): Vegetation – *Puccinellia maritima* along with some Halophytes, about 180 flooding per year, strongly stratified.

Depth (cm)	Horizon (US Tax.)	rH [#] value	pH water (1 : 2.5)	*ECe (mS cm ⁻¹)	Org.-C (g kg ⁻¹)	Total-N (g kg ⁻¹)	Bulk density (Mg m ⁻³)	Hydrolic con.(cm d ⁻¹)
0-14	Azh	17.3-10.4	6.9	37.1	35	3.8	0.73	70.8
14-29	Czrg		6.9	36.9	27	2.8	0.81	47.2
29-45	Czgr		6.9	40.7	23	2.4	0.79	48.5
45-70	Czr1	11.8-8.8	6.8	38.3	21	2.1	0.80	1.0
70-120	Czr2	10.7-8.1	6.8	39.7	18	1.8	0.76	0.4
Depth	Particle size (Int.) distribution (%)			CEC c mol kg ⁻¹	Water content (%) at different pF values			
	Sand	Silt	Clay		-∞	1.8	2.5	4.2
0-14	7.6	66.2	26.2	24.4	70.6	54.5	48.3	32.4
14-29	7.5	65.0	27.5	27.7	67.7	59.1	50.9	28.7
29-45	8.1	63.8	28.1	24.4	68.7	60.8	52.8	26.4
45-70	5.0	62.8	32.2	21.5	68.6	63.6	55.0	25.7
70-120	39.9	27.6	32.5	21.6	70.4	63.3	54.3	26.3

Haplic Sulfaquent (Marines Übergangsmischwatt): Vegetation – *Spartina anglica* and *Salicornia europaea*, about 600 flooding per year, strongly stratified.

Depth (cm)	Horizon (US Tax.)	rH [#] value	pH water (1 : 2.5)	*ECe (mS cm ⁻¹)	Org.-C (g kg ⁻¹)	Total-N (g kg ⁻¹)	Bulk density (Mg m ⁻³)	Hydric con.(cm d ⁻¹)
0-3	Az(h)	15-13.6	7.0	41.6	12.0	1.2	1.16	67.6
3-8	Czrg		7.0	39.8	6.0	0.7	1.16	67.6
8-22	Czr1	11.9-8.8	6.9	39.1	5.8	0.6	1.22	77.2
22-45	Czr2		7.0	42.4	5.9	0.6	1.17	95.7
45-85	Czr3	10.5-8.6	6.8	43.1	9.2	1.0	1.29	53.9

Depth	Particle size (Int.) distribution (%)			CEC c mol kg ⁻¹	Water content (%) at different pF values			
	Sand	Silt	Clay		-∞	1.8	2.5	4.2
0-3	45.0	48.0	7.0	17.3	55.3	46.4	33.3	13.2
3-8	58.7	39.8	1.5	13.6	55.3	46.4	33.3	13.2
8-22	51.7	44.9	3.4	15.3	53.3	45.5	27.6	8.0
22-45	46.8	46.1	7.1	16.5	55.2	46.4	30.5	8.0
45-85	33.6	52.3	14.1	27.2	50.4	46.8	29.4	7.4

The rH value has been calculated from the formula of $rH = (2 Eh/59) + 2$ pH: Schlichting et al. 1995); values of mean quarterary redox potential of Table 2. *ECe = Electrical conductivity of saturation extracts.

The mean values of soil temperatures were in the range of 9.0 to 9.8°C for TS at depths of 0 to 100 cm and 10.1 to 12.0°C for HS at depths of 0 to 50 cm throughout the year (Table 2). The higher temperatures at the later especially during summer seem to be influenced by daily flooding with relatively warmer seawater. The soil temperatures were almost 2°C higher in HS than that of the TS and it was also 2.5°C higher than the mean annual temperature of the soils (9.5°C), which might be due to higher absorption of solar radiation by the almost bare and more salt saturated surface of the HS soil and by the warmer surface sea water intrusions. The similar trend of increment of mean annual soil temperature was also observed for the soils of the National park in Schleswig-Holstein (Becker, 1998).

except for the summer. These negative values of CO₂ emissions were might be due to rapid growth and development of vegetations together with high translocation of emitted CO₂ by seawater. But in TS, the emissions of CO₂ were highly positive throughout the year (857 g m⁻² yr⁻¹) in comparison to the HS (-14.0 g m⁻² yr⁻¹), which might be due to high temperature, less flooding and the extremely low vegetation cover as well as the activities of both micro flora and fauna, soil physical parameters, etc. of the salt marsh soils. The present findings of CO₂ emissions can be compared with the results of Scala et al. (2000) who measured CO₂ emissions ranged from 137 to 3455 g m⁻² yr⁻¹ during November, 1998. Howard and Howard (1993) also proved that the emissions of CO₂ strongly depend on soil temperature and water content.

Emissions of CO₂: The net emissions of CO₂ (-14.0 g m⁻² yr⁻¹) were obtained negative (Table 3) for HS,

Table 2: Mean seasonal and calculated annual net emissions (mg m⁻² h⁻¹) of CO₂ and CH₄ from salt marsh soils in the Wadden sea coast of Northern Germany.

Soil unit	Spring		Summer		Autumn		Winter		Emission rates	
	SD		SD		SD		SD		mg m ⁻² d ⁻¹	g m ⁻² yr ⁻¹
Emissions of CO ₂ (mg m ⁻² h ⁻¹)										
Haplic Sulfaquent	-3.19	55	7.7	102	-2.0	56	-8.9	26	-38.4	-14.0
Typic Sulfaquent	33.6	93	116	193	196	234	45.5	86	2350	857
Emissions of CH ₄ (µg m ⁻² h ⁻¹)										
Haplic Sulfaquent	-3.36	67	24.9	41	67.7	147	47.1	131	0.82	0.3
Typic Sulfaquent	-5.7	59	8.8	27	18.3	29	-6.2	70	0.09	0.03

Level of Significance: U-Test	Haplic Sulfaquent				Typic Sulfaquent							
	Spring		Summer		Autumn		Spring		Summer		Autumn	
	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄	CO ₂	CH ₄
Summer	0.190	2.203**					1.650*	1.687*				
Autumn	0.468	1.137	0.158	0.574			3.193***	2.328**	1.507	1.240		
Winter	0.350	1.850*	0.008	1.957*	0.516	0.428	0.771	0.231	0.741	0.824	2.494**	1.703*

Level of Significance: 90 % = *, 95 % = **, and 99 % = ***

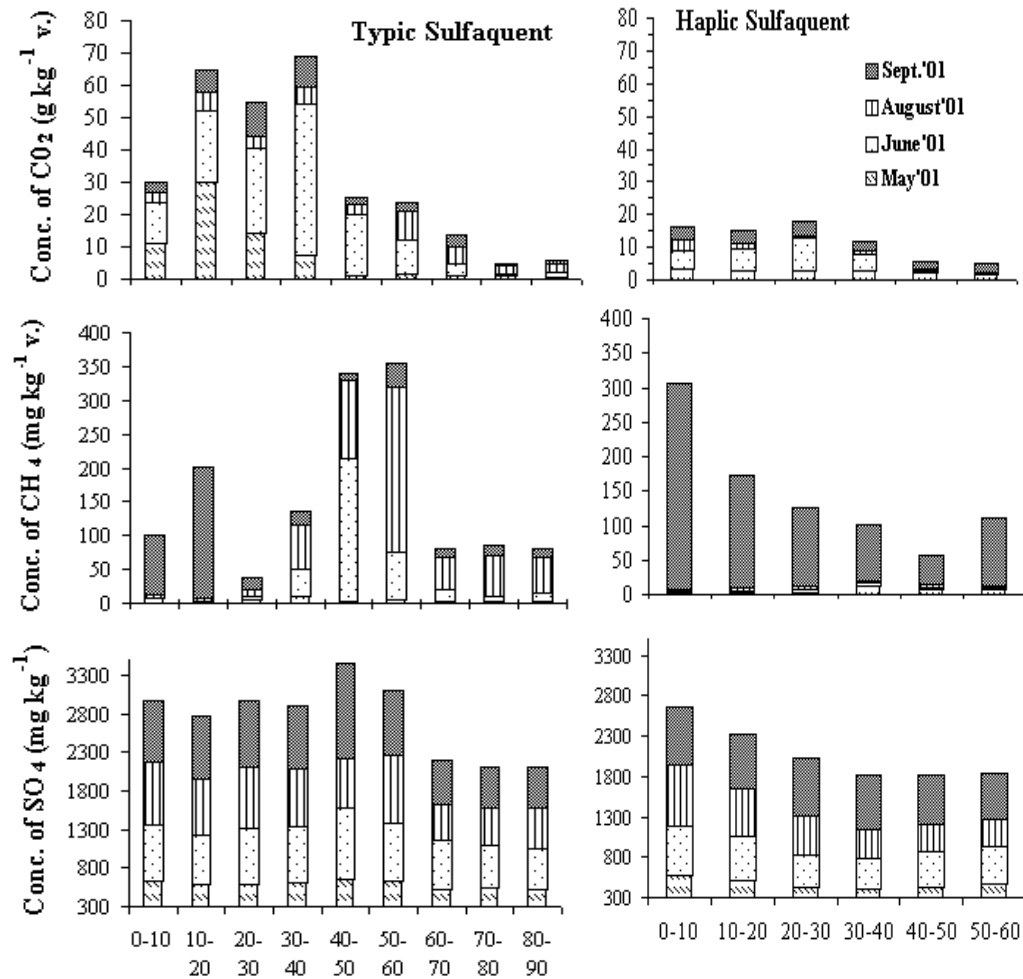


Fig. 1. Seasonal distribution of CO_2 , CH_4 and SO_4 in salt-marsh soils of North Germany

Emissions of CH_4 : The annual emissions of CH_4 were almost 10 folds higher ($0.3 \text{ g m}^{-2} \text{ yr}^{-1}$) in HS than that ($0.03 \text{ g m}^{-2} \text{ yr}^{-1}$) of the TS (Table 3). Methane emission rates varied seasonally, with the greatest in autumn and smallest in spring. The small mean efflux in both the soils during summer was probably due to dry weather, which favors oxidation of CH_4 during its movement from the subsoil to the topsoil. Biological activity was diminished due to low temperature during spring, which results the lowest emissions, while the emissions were increased during summer months of August and September. These increased emissions were not only caused by the higher temperature enhancing more bacterial productivity but also by the high transpiration rate with a direct efflux of CH_4 from the roots to the atmosphere

(Sebacher et al., 1985). Cracks in the soil as well as the loss of oxygen in the rhizosphere due to transport of air via aerenchyma enhances the oxidation. The increased rate of CH_4 may be compensated by stronger oxidation in summer and that might be the reason for the moderate variations in emissions of CH_4 between the seasons. The increased emissions of CH_4 during autumn reflects either the increased transpiration rates during these (August-September) months with plant enforced emission of CH_4 or the higher efflux has caused by cracks developed during summer, especially TS soil. The seasonal fluctuations of the CH_4 emissions were strongly significant for the HS, while the emissions of CO_2 were highly significant for the TS soil (Table 3). These suggest that vegetation cover; activities of soil flora, fauna, soil

temperature and organic matter status are the considerable controlling factors for these trace gases. The CH₄ emission rates during spring and winter were negative for TS, indicating that there was inconsiderable CH₄ efflux from these marsh soils. Despite of the potentials of CH₄ production according to reduction and content of soil organic matter (Table 1), the emissions of CH₄ were low (Table 3), which might be due to strong reducing effect of seawater SO₄²⁻.

Distributions of CO₂: The concentrations of CO₂ varied strikingly throughout the soil profiles and also with the seasons of the year. The maximum concentrations of CO₂ were measured during June'01 (16.1 g kg⁻¹ soil v.) followed by May'01 (7.4 g), Sept.'01 (4.5 g) and August'01 (4.4 g) in the TS Soil. But the sequences of concentrations of CO₂ were June'01 (4.8) > Sept.'01 (3.3) > May'01 (2.1) > August'01 (1.3 g kg⁻¹) for the HS soil and May'01 (2.2) > Sept.'01 (1.4 g) for HS soil. However, the maximum concentrations of CO₂ were detected within the surface soils of 20 to 40 cm in both the profiles regardless of the seasons, except for the August'01 where the maximum concentration was measured at a depth of 50 to 60 cm of the profile of TS soil. The high concentration of CO₂ in the TS soil might be the results of dissimilation processes of the plants as well as the higher activities of soil organisms. On the other hand, the stronger tidal influences at the HS soils lead to dissolution of produced CO₂ and thereby can be considered as a sink of CO₂. Partial water saturation of the soil leads to low air conductivity, which concentrated the gases in the soil air and soil solutions. The low CO₂ concentrations in the reduced subsoil of all the soil profiles reflect the low microbial activity.

Distributions of CH₄: The concentrations of CH₄ were distributed irregularly with the seasons and soil depths (Fig. 1). In the TS soil, the concentrations of CH₄ were measured maximum of 9 mg kg⁻¹ at depths of 30-40 cm followed by 7 mg at depth of 0-10 cm > 5 mg at depth of 50-60 cm during May'01. During June'01, the CH₄ concentration was 212 mg at depth of 40-50 cm followed by 71 mg at depths of 50-60 cm > 40 mg at depth of 30-40 cm. The CH₄ concentrations of 245 mg at depth of 50-60 cm > 114 mg at depth of 40-50 cm > 65 mg at 30-40 cm were measured during August'01. The concentrations of CH₄ of 193 mg at depth of 10-20 cm > 87 mg at 0-10 cm > 34 mg kg⁻¹ at 50-60 cm were detected during September'01 (Fig. 1). The sequences of concentrations of CH₄ as measured were August >

September > June > May for TS profiles; September > May > August > June for the profiles of HS. The concentrations of CH₄ at different seasons showed almost inverse relationships with the concentrations of CO₂. In all the profiles, the concentrations of CH₄ were very low and varied significantly ($p \leq 0.05$) varied with the seasons and soil depths. The concentrations of CH₄ showed no dependence to temperature.

High temporal and spatial variations in both the CO₂ and CH₄ emissions led to question whether are the emission pattern reflects the concentrations of these gases in the soil? The highest concentrations at each sampling times (up to 245 ppmv CH₄) were found in the TS soil and the lowest concentrations (till 1.4 ppm by volume) were in the HS soil, which were almost in the range of atmospheric concentration (CH₄: 1.65 ppmv). Water saturation and/or daily tides dilute the gases in HS soils resulting high gas concentrations to that of the TS, which mean not to build up. Stratification, aggregation and/or variable pore volume with different continuity may lead to an inhomogeneous soil with the formation of traps where produced CH₄ is accumulated leading to discontinuous release by bubbling (Takai and Wada, 1990). In the permanent saturated soils, tidal marine water mixes and dilutes the gas content in the soil solution twice a day. Therefore, the Haplic Sulfaquents have comparatively high gas concentrations to that of the Typic Sulfaquent, which mean not to build up. In the Haplic Sulfaquent soil, the shifting water table at rising tide may press CH₄ loaded water upward without exporting the soil solution from the system as the hydraulic conductivity of 68 to 96 cm d⁻¹ in the upper soil which is much lower than the rising tide, leading to an enclosure of the entrapped soil solution. The SO₄ concentrations in the studied soil solutions were a few folds higher (Fig. 1) than that of the seawater indicating a low exchange rate between the soil solutions and marine water.

Methane oxidation mediated mainly by methanotrophic bacteria is the responsible process, which is strongly inhibited by ammonium accessible for nitrification (Hütsch, 2001). Indeed, in many studies NH₄ was identified as a strong inhibitor for CH₄ oxidation (Bronson and Mosier, 1994). Root derived organic C can contribute to various C pools and become an origin of CH₄ emitted from flooded soils (Lu et al., 2000). In the total amount of atmospheric CH₄, the contribution of CH₄ from sulfate-rich soils is negligible, because sulfate-reducing and methane producing organisms compete for the same substrates

(H₂/CO₂, acetate-competitive substrates) but the sulfate reducers have the competitive advantage: they have stronger affinity to the competitive substrates and can use them to provide more energy than the methane producers (Schönheit et al., 1982). Increasing methane and decreasing sulfate concentrations (methane-sulfate transition) are found in some deeper depths, but not consistent with the seasons. Most of the soils contained more CH₄ in August-September than those of May-June, indicating no methane-sulfate transition, which means sulfate reduction is not limited and methanogenesis is suppressed down to that depth. As methanogenesis is strictly anaerobic, more CH₄ was expected in the more anaerobic soils. The soils under more anaerobic condition (near the sea) did not contain maximum methane nor did the more aerobic soil (far away from the sea) contain the least. This suggesting that there seemed to be no relation between methane concentration and state of soil development. There was also no evident relation between the redox properties and their corresponding methane concentrations.

Distributions of SO₄: There should be high SO₄ contents in the surface soils, which might be reduced to sulfide at subsoils. But in reality, the high SO₄ were determined in the subsoils of all the profiles in different seasons at depths of 40-50 cm followed by surface soils of TS (Fig. 1). The SO₄ contents were obtained maximum (up to 1236 mg kg⁻¹) in the TS followed by HS (up to 745 mg kg⁻¹) during all the seasons (Fig. 1), which was due to the result of the frequent intrusion of highly SO₄ (> 600 mg SO₄ L⁻¹) enriched seawaters. The concentrations of SO₄ throughout the profiles of all the soils were quite inversely distributed in relation to CH₄ concentrations throughout the year, except for the depths of 40-50 cm during June'01 and 50-60 cm during August'01 for the TS profile where both the CH₄ and SO₄ contents were high. These indicate that there may be a coexistence of sulfate reducing and methane producing bacteria as reported by Holmer and Kristensen (1994). There is no noticeable correlation ($r = -0.09$) was obtained between the SO₄ and CH₄ concentrations. Moreover, even CH₄ was determined at depths where the SO₄ concentrations were around 1200 mg SO₄ L⁻¹. This indicates that either CH₄ production is compatible even with the higher SO₄ concentration.

The present findings concluded that the fluxes of CO₂ and CH₄ from the coastal salt-marsh soils varied remarkably among the seasons and may act either as a sink or source of atmosphere depending

on the nature of the soils. Relatively high net emission of CO₂ was determined from the Typic Sulfaquent under salt meadow. The CH₄ emission was low resulting from low formation of CH₄, which was due to a high SO₄ input from SO₄ enriched seawater. There was also no significant concentration/efflux of CH₄, suggesting almost no contribution from the studied coastal salt marsh soils to the atmospheric methane. The heterogeneity in redox potential of the soils studied also indicate that CH₄ oxidation is likely to take place, and the CH₄ emission and concentration gradients suggest that the CH₄ produced is readily oxidized within the soils. Vegetation cover, temperature and organic matter status are the considerable controlling factors for these trace gases. Further research should be focused on more detailed look on the geophysical conditions of the soils, and CH₄ oxidation and SO₄ reduction rates.

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