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Dynamics of dissolved O_2, CO_2, CH_4, and N_2O in a tropical coastal swamp in southern Thailand

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Abstract. We studied the distribution of dissolved O_2, CO_2, CH_4, and N_2O in a coastal swamp system in Thailand with the goal to characterize the dynamics of these gases within the system. The gas concentrations varied spatially and seasonally in both surface and ground waters. The entire system was a strong source for CO_2 and CH_4, and a possible sink for atmospheric N_2O. Seasonal variation in precipitation primarily regulated the redox conditions in the system. However, distributions of CO_2, CH_4, and N_2O in the river that received swamp waters were not always in agreement with redox conditions indicated by dissolved O_2 concentrations. Sulfate production through pyrite oxidation occurred in the swamp with thin peat layer under aerobic conditions and was reflected by elevated SO_4^{2-}/Cl^- in the river water. When SO_4^{2-}/Cl^- was high, CO_2 and CH_4 concentrations decreased, whereas the N_2O concentration increased. The excess SO_4^{2-} in the river water was thus identified as a potential indicator for gas dynamics in this coastal swamp system.

Introduction

Wetlands provide active sites for exchange of carbon, nitrogen, and sulfur between terrestrial ecosystems and atmosphere through various biochemical pathways. Tropical and sub-tropical wetlands are important for the exchange of radiatively active gases (CO_2, CH_4 and N_2O) between the land surface and the atmosphere. Tropical wetlands are thought to be an important source for atmospheric CH_4 (Khalil & Rasmussen 1983; Mathews & Fung 1987; Cicerone & Oremland 1988; Fung et al. 1991), the concentration of which has nearly tripled in the last four-hundred years (Rasmussen & Khalil 1984). Under highly reduced conditions, submerged soils act as a sink for tropospheric N_2O (Richey et al. 1988), although the sink is smaller than that of stratospheric decomposition (Crutzen 1970). Most of the data on exchange of CH_4 and N_2O were collected in central and southern America and in equatorial Africa. Information on dynamics of the above gases in tropical wetlands in other locations, in particular tropical Asia, is restricted (Bartlett & Harriss 1993).

According to Aselmann and Crutzen (1989), 27% of the world's wetlands (5.7 million km^2) occurs in tropical areas, two thirds of which are in lowlands of Southeast Asia. Peat swamps, one landform of the tropical wetlands, are found along coasts of insular Southeast Asia including Malay Peninsula, Sumatra, Kalimantan, and other islands, and comprise approximately 0.3 million km^2 (Driessen 1978). Tropical peats are characteristically forested, in contrast to temperate and boreal peats which are mostly covered by peat moss (sphagnum) and herbaceous plants.

The tropical peat swamp ecosystems play an important role in the hydrologic cycle and act as a local climatic regulator. However, a considerable portion of the swamp areas in tropical Asia has recently been developed and populated. The remainder may be reclaimed to meet with an increasing food demand. Swamp reclamation is commonly accompanied by extensive drainage and forest clear-cutting followed by burning, which drastically alters the cycles of water and bioelements in the swamp system. For instance, vegetation clearance may result in the input of fresh organic matter to soil and subsequent soil subsidence due to burning and rapid decomposition of peat materials under aerobic condition (Armentano & Menges 1986). Such a change of material balance must affect the rates of production and consumption of O_2, CO_2, CH_4, and N_2O in the swamp system.

The Thailand–Japan cooperative study on a tropical coastal peat swamp system was launched in 1990 to investigate the recent changes of tropical peat swamp forests in Southeast Asia from different perspectives, such as ecology, biogeochemisty, and agriculture, and to come up with a realistic solution for appropriate utilization or rehabilitation of the disturbed swamps (Vijarnsorn et al. 1995). During a period of the project, basic information on gas exchange between the swamp system and the atmosphere was collected. Here, we report the results from our surveys on distributions of dissolved O_2, CO_2, CH_4, and N_2O together with the chemical compositions of swamp, river, and ground waters. The objectives of this paper are to characterize the dynamics of these gases in the coastal swamp system, and to elucidate a potential indicator for redox condition controlling the gas dynamics. As this paper will be the first report for the gas dynamics in a swamp system of tropical Asia, the data sets presented may help to fill the gap of geographically biased information on gas exchange between wetlands and atmosphere.

Materials and methods

Study area

Our study sites were located in Narathiwat Province, adjacent to the border of Malaysia (Figure 1). The mean temperature and annual precipitation for a period of 1961 and 1990 were 27.3 °C and 2,465 mm, respectively. The east-west monsoon from the Gulf of Thailand brings more than 60% of the annual rainfall and divides a year in two seasons: the dry season: January through August, and the wet season: September through December.

The total area of peat swamp in Thailand amounts to 453 km^2, 60% of which is found in the study area (Vijarnsorn 1996). The peat swamps including two major complexes belonging to the To Daeng and Bacho Swamps (Figure 1) have been developed on the areas that were formed due to the sea level decline after the last hypsithermal period. They include former lagoons and embayments cut off from the sea by sandy plains or dunes where brackish vegetation including mangrove forests were replaced by freshwater swamp forests. Because of this, the woody peat layers are underlain by marine deposits rich in pyrite (mud clay), and acid sulfate soils commonly appear in transition areas between the sandy plains and the swamps. Such geographic setting and peat characteristics are typical of the swamp systems along the east coast of the Malay Peninsula.

A fairly large portion of the To Daeng Swamp is preserved and water-logged and still carries a climax evergreen peat swamp forest (Niyomdham 1986; Smitinand et al. 1991). The thickness of peat layer is 1–3 m (Vijarnsorn 1992). This swamp is fed by rainfall and river run-off from mountainous areas in the west. The swamp water is discharged into the Gulf of Thailand through the Bang Nara River (Figure 1). The river water flows both north and south from the middle reach near R8. Since 1990, the river water level has been kept above sea level by operating tidal gates at the two river mouths (NTG, STG) and at the end of a man-made canal connecting the middle reach to the sea (CTG). These tidal gates are fully opened at heavy rain events to avoid flooding.

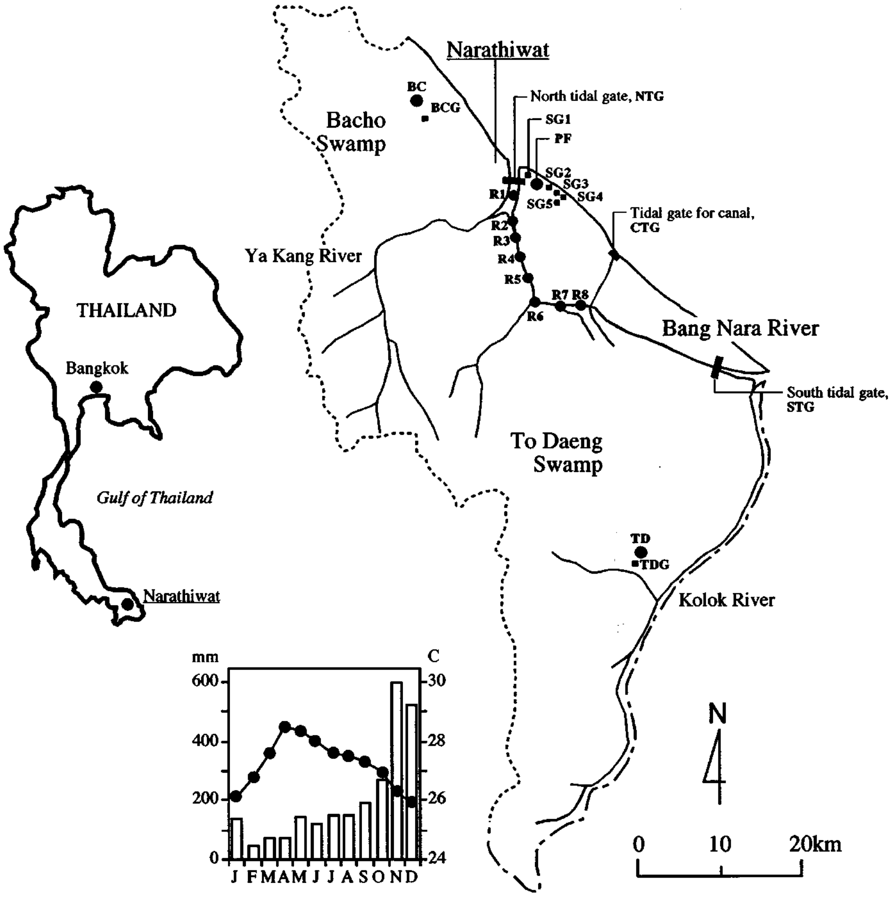

Figure 1. Study area. Dotted areas denote swamps. Dashed and broken lines present the border to Malaysia and the district boundary, respectively. Information on sampling sites is provided in Table 1. Temperature [●] and rainfall [vertical bars] in Narathiwat Province in the panel are means for thirty year records beginning in 1961 by Meteorological Department of Thailand.

The peat layer in the Bacho Swamp (Figure 1) is thinner than 1 m (Vijarnsorn 1992). This swamp has been drained extensively, and cleared since 1975 for agricultural land use. However, most of the reclaimed area has been abandoned because it is unsuitable for crop production, due to extremely low hydraulic conductivity, oligotrophy, and acidity. An acid tolerant tree (*Melaleuca cajuputi*), reed (*Lepironia articulata*), and fern (*Blechnum indicum*) occupy the swamp. The water management results in this swamp being water-logged only in the wet season. The water table is located at 0.5–0.8 m below the surface in the hot and dry period of March through April (Vijarnsorn 1992).

Table 1. List of sampling sites.

Symbol	Location	Sampling depth (m)	Remarks
TD	To Daeng Swamp (SRNSC[1])	<0.1	Preserved swamp forest
BC	Bacho Swamp	<0.1	Reclaimed swamp
PF	PRDSC[2]	<0.1	Experimental paddy fields
R1–R8	Bang Nara River	0.5	Natural and reclaimed swamp
NTG	North mouth of Ban Nara River	<0.2	Tidal gate
CTG	Month of man-made canal	<0.2	Tidal gate
TDG	To Daeng Swamp (SRNSC), EP[3]	30[5]	Edge of To Daeng Swamp
BCG	Boo Kon Sue Door Primary School, HP[4]	5[5]	Sandy plain fringing Bacho Swamp
SG1	Private house at Ban Bang Manoa, EP	5[5]	Sandy plain, arable land, residence
SG2	Private house at Ban Khao Tan Yong, EP	5[5]	Sandy plain, arable land, residence
SG3	Khao Tan Yong Primary School, EP	5[5]	Sandy plain, arable land, residence
SG4	Isram Burapa School at Ban Mai, EP	5[5]	Sandy plain, arable land, residence
SG5	Private house at Ban Mai, EP	4[5]	Sandy plain, arable land, residence

[1] Sirindhorn Research and Nature Study Center
[2] Pikunthong Royal Development Study Center
[3] EP; electric-pump well
[4] HP; hand-pump well
[5] Depth of strainer

Sampling sites

Field expeditions were carried out in the wet season (December, 1990; December, 1991; November–December, 1992; December, 1993) and in the dry season (July, 1992; August, 1993). Sampling locations are listed in Table 1 and denoted in Figure 1. The paddy field site (PF) was located in an experimental paddy field of the Pikunthong Royal Development Study Center (PRDSC). The Bang Nara River samples were collected at midstream of R1 through R8. The river depth ranged from 3 to 5 m except at the shallowest site R3 (1.5m). Additional samplings were carried out at NTG and CTG at 1–2 months intervals in 1993 to monitor the seasonal variation in chemical composition of the river water. Well sites were selected in swamp area (TDG, BCG) and in sandy plain area (SG1–SG5).

Sample collection

Waters for gas and ion analyses were sampled principally at each site during the wet and dry season observations, except for the observation in December, 1990 in which samples for ion analyses were not collected. All samples in the paddy field were taken in December, 1990. Swamp and paddy waters were collected by submerging containers very gently by hand with minimum soil agitation. River waters were taken from a motorboat by a 250-ml glass syringe in December, 1990 and by Van Dorn sampler at other times. A roped bucket was used to collect river water during the monitoring of ion concentrations at the tidal gates in 1993. At well sites, waters were first pumped out until water temperatures became stable in order to minimize influence of temperature fluctuation on dissolved gas concentrations. The well waters were once collected without bubbling in a vessel, and sub-samples for each measuring components were prepared.

Waters for O_2 concentration measurements were collected in 100-ml BOD bottles and were fixed immediately in the field by the Winkler method. Waters for pH measurements were carefully collected in the same BOD bottles without inclusion of air bubbles. Samples for measuring CO_2, CH_4, and N_2O concentrations were collected in 30-ml glass vials. The vials were sealed by butyl rubber stoppers without head space, after adding 0.1 ml of saturated mercuric chloride solution. Waters for ion analyses were filtered by pre-combusted glass fiber filters (Whatman GF/C) in the laboratory at the PRDSC, then kept in a freezer.

A plot of 80×100 m, divided in 10×10 m grids was laid out near the BC site on 30 November 1993 to determine the spatial variation of dissolved gases in the swamp. Waters for measuring gas concentrations were collected at ninety-nine intersections of the grid lines on 3 December 1993. Depth and temperature of water were recorded at each point. On the same day, CH_4 emission rates were measured at seven locations randomly selected around the grid plot, using floating Plexiglas chambers (20×20×13h cm). Two or three chambers were gently placed on the water surface and left for 1 hour. Septum-fitted glass ampoules which had been filled with CH_4-free water were used to collect ambient air samples. The air was collected by a gas-tight syringe and injected into the ampoule that was mounted with a separate needle to let water out. Chamber air samples were sucked through septa of the chambers and filled in the ampoules in the same manner. A time-dependent linear increase of CH_4 concentration in the chamber was confirmed for the first 3 hours from a preliminary experiment. The CH_4 flux was calculated from the difference in mixing ratio between the ambient and the chamber samples.

Chemical analysis

Dissolved O_2 concentration and pH were determined at the PRDSC by titration and a pH meter with a glass electrode during the wet and dry season observations. Only during the grid observation, these variables were measured on site by handheld meters. The following analyses were performed in Japan. The concentration of dissolved CH_4 was analyzed using a head space technique. Head space was made in a vial by replacing sample water by 10 ml of N_2 ($>$99.999%). For calibration, 10 ml of cylinder gas (10.4 ppmv CH_4) each was injected to vials previously filled with CH_4-free water, prepared by boiling and subsequent purging by N_2. Standards and samples were vigorously shaken by hand for 2 minutes and placed in a temperature-stat water vessel for 12 hours to ensure gas-liquid equilibrium in the vials. The head space gas was withdrawn by a gas-tight syringe, then injected to an FID gas chromatograph (Shimadzu GC-9A) equipped with a 3-m Molecular Sieve 5A column. Concentrations of CH_4 in samples for the flux determination were measured using the same gas chromatograph and standard gas as for the head space samples. Concentrations of dissolved N_2O were determined by an ECD gas chromatograph (Shimadzu GC-7A or GC-8A) equipped with a 3-m Molecular Sieve 5A column and a purge-trap gas extracting device. For measuring dissolved CO_2, total inorganic carbon was extracted as CO_2 by stripping an aliquot of water by N_2 after adding 3% phosphoric acid to the sample. The CO_2 evolved was determined by a nondispersive infrared gas analyzer (Shimadzu UR-101). The concentration of free CO_2 was calculated using temperature and pH data (Stumm & Morgan 1981). Samples collected at Bang Nara River in August, 1993 could not be analyzed due to instrument failure. Concentrations of anions (NO_2^-, NO_3^-, SO_4^{2-}, Cl^-) were determined by ion chromatography (Yokogawa IC-200, IC-7000S). Cations (Na^+, K^+, Mg^{2+}, Ca^{2+}) were analyzed by ion chromatography or by atomic absorption spectrometry (Shimadzu AA-670). Concentrations of NH_4^+ in most samples were measured by colorimetry (Solorzano 1969), others by ion chromatography.

Statistical analysis

Differences of mean values were examined by *t*-test. Significance of regression equations was evaluated by *F*-test.

Results and discussion

Inter-annual variation in precipitation

Monthly mean precipitation from 1990 to 1993 and for the 30-year average are shown in Figure 2. In 1990, monthly precipitation was higher than the 30-year average in April, July, and September and lower in November to December, but annual rainfall was nearly equal to the 30-year average. Monthly and annual precipitation in 1991 were similar to the 30-year average. Seasonal pattern of precipitation in 1992 resembled that in the 30-year

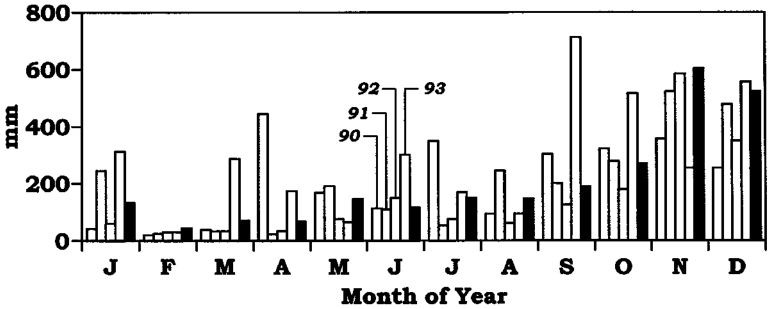

Figure 2. Monthly precipitation over the study period (white bars) together with 30-year average since 1961 (filled bars) in Narathiwat Province. Data were provided by Meteorological Department of Thailand.

average, although the annual precipitation was 30% lower. Low precipitation in 1992 brought severe drought over the country, most probably due to the El Niño-Southern Oscillation (ENSO). Annual precipitation in 1993 was 1.4 times the 30-year average, which was due to increased precipitation in January, March, April, June, September and October, although precipitation in November was half of the 30-year average. Variable amounts of precipitation in different seasons and on year to year basis may affect gas dynamics in the study area.

Swamp and river waters

Swamp water was characterized as a whole by its low pH value (Table 2). At the BC site, surface water was available only during wet seasons. It was brownish in color and always more acidic than that of the TD, with a significantly lower mean pH ($P < 0.01$). Concentrations of dissolved organic carbon were extremely high (>50 mgC liter^{-1}) at BC and relatively high (~6 mgC liter^{-1}) at TD (Yoshioka et al. in preparation). As very low SO_4^{2-} concentrations were found in the waters at these two sites, organic acids from peat probably caused the acidity of swamp water.

The total concentrations of inorganic anions and cations steadily increased from the swamp sites to R1 to R8. Mean SO_4^{2-} concentration at R8 was two orders of magnitude higher than those of the swamp sites in both wet and dry seasons. The SO_4^{2-} concentration decreased from R8 towards R1 during each observation, even though temporal variations resulted in a wide range of SO_4^{2-} concentrations. These SO_4^{2-} concentration gradients probably reflect mixing of waters draining acid sulfate soil areas surrounding the peat swamps and neutral waters draining alluvial plains under cultivation. This view was supported by the observations that mean pH value in R8 was significantly

Table 2. Summary of observations in surface waters.

	TD		BC	R1		R8	
	Dry period	Wet period	Wet period	Dry period	Wet period	Dry period	Wet period
Temp °C	24.8	25 (24–25)	27 (25–27)	28.9 (28.8–29.0)	27.1 (26.5–27.8)	29.6 (29.0–30.1)	27.9 (27.0–29.0)
EC μS cm^{-1}	40 (25–55)	22 (19–25)	56 (24–79)	87 (82–92)	75 (29–117)	110 (85–136)	235 (70–356)
pH	4.6	5.1 (4.6–5.7)	4.1 (4.0–4.3)	6.0 (6.0–6.0)	5.6 (4.4–6.4)	4.3 (4.2–4.4)	4.0 (3.7–4.4)
Anions and cations μeq $liter^{-1}$							
Cl^-	NA	66 (51–75)	125 (63–172)	487 (434–539)	358 (117–497)	476 (293–641)	950 (309–2050)
NO_2^-	NA	nd (nd–nd)	nd (nd–nd)	0.4 (nd–0.7)	nd (nd–nd)	nd (nd–nd)	nd (nd–nd)
NO_3^-	NA	0.7 (nd–2)	nd (nd–nd)	1 (nd–2.5)	5 (2.2–7.3)	0.6 (nd–1.2)	nd (nd–nd)
SO_4^{2-}	NA	3 (2–5)	5 (nd–10.4)	132 (111–152)	219 (41–483)	374 (287–460)	992 (257–1850)
NH_4^+	NA	0.3 (nd–0.6)	5 (nd–10)	0.8	3 (2.6–3.2)	7	20 (11–28)
Na^+	NA	68 (62–78)	155 (74–215)	487 (434–539)	326 (144–442)	467 (293–641)	746 (278–1590)
K^+	NA	18 (13–22)	21 (6–45)	44 (40–48)	50 (31–78)	34 (25–42)	52 (28–67)
Mg^{2+}	NA	10 (7–12)	36 (23–43)	120 (91–149)	83 (34–130)	181 (82–280)	241 (74–476)
Ca^{2+}	NA	14 (5–23)	17 (14–23)	61 (34–87)	70 (35–129)	95 (37–152)	115 (32–221)
Dissolved gases							
O_2 μM	125	35 (9–72)	55 (6–140)	158 (149–166)	127 (87–164)	84 (70–98)	41 (6–113)
CO_2 μM	29 (2–55)	345 (28–600)	306 (46–1090)	61	92 (26–204)	112	177 (26–376)
CH_4 μM	0.01	51 (0.1–108)	7 (0.01–50)	0.2 (0.2–0.3)	0.7 (0.01–1.9)	1.7 (0.01–3.4)	12 (0.2–31)
N_2O nM	9.6	5 (2–12)	7 (2–12)	12 (9–14)	8 (3–14)	10 (9–12)	10 (5–14)

All variables are expressed as mean and range. ‘nd’: not detected; ‘NA’: not available.
Data for dissolved gas concentrations in TD and BC include measurements in areas nearby the sites.

lower than that in R1 ($P < 0.01$) in both wet and dry seasons, and that NO_3^- concentration was always higher in R1 than R8 with the exception in July, 1992.

Dissolved gas concentrations in swamp and river waters varied widely at different dates and sites. As the swamp data in dry season were limited, we attempted to characterize the gas distributions in surface waters by using the data set in wet season. While the mean CH_4 concentration of TD was significantly higher than that of BC ($P < 0.01$), the difference between TD and R8 was insignificant. Differences in mean concentrations of O_2, CO_2, and N_2O between TD and R8 were statistically insignificant. These observations do not contradict the data on SO_4^{2-} which suggests that site R8 receives waters from the swamp area. In the Bang Nara River, the concentration of O_2 was always higher, whereas the concentrations of CO_2 and CH_4 were always lower in R1 than in R8. Differences in means between R1 and R8 were significant for O_2 ($P < 0.01$) and CO_2 ($P < 0.05$). The changes in gas concentrations from the upper to the lower sites of the river were partially due to the mixing of waters drained from the swamps and alluvial plains as was indicated by the SO_4^{2-} distributions.

Ground water

Low O_2 concentrations in the swamp well waters, TDG and BCG, suggest that the corresponding aquifers are anaerobic during both wet and dry seasons (Table 3). Differences in mean concentrations of all ionic and gaseous species between wet and dry seasons were statistically insignificant in these wells. When the measurements in the extraordinarily dry year of 1992 were excluded, however, the mean CH_4 concentration was significantly higher during wet seasons than during dry seasons in both TDG and BCG ($P < 0.05$). The N_2O concentrations at these wells in wet seasons were below atmospheric equilibria, with an exception of an over-saturation at BCG (11 nmoles liter^{-1}) in 1992. In the dry seasons, the N_2O concentrations of these wells slightly exceeded atmospheric equilibria. These observations suggest that N_2O is reduced to N_2 in peat during wet seasons. The low levels of CH_4 at TDG and BCG and the N_2O over-saturation at BCG in the wet season of 1992 might have been caused by suppression on methanogenesis and denitrification in peat under relatively oxic conditions as a result of the drought in this year.

Very low NO_3^- concentrations at TDG and BCG both in wet and dry seasons (Table 3), and high concentrations of NH_4^+, particularly in TDG during wet seasons (mean of 159 μeq liter^{-1}), suggest dissimilatory nitrate reduction or little nitrification in the aquifer. The SO_4^{2-} concentration in TDG water was about two orders of magnitude lower than that in the BCG in wet and dry seasons. The low SO_4^{2-} concentration in the TD water was possibly

Table 3. Summary of observations in ground waters.

	TDG		BCG		SG1 through SG5	
	Dry	Wet	Dry	Wet	Dry	Wet
Temp °C	26.0 (26.0–26.0)	26.8 (26.0–28.1)	28.4 (27.5–29.2)	28.9 (28.8–29.0)	28.6 (26.5–31.3)	28.1 (26.5–29.9)
EC μS cm^{-1}	108	111 (100–124)	96 (84–108)	104 (66–135)	196 (32–557)	161 (34–386)
pH	6.2 (6.1–6.3)	6.1 (6.1–6.1)	6.2 (6.0–6.4)	6.1 (5.8–6.4)	5.3 (4.1–6.4)	5.5 (4.7–6.9)
Anions and cations μeq liter^{-1}						
Cl^-	213 (140–285)	248 (199–292)	146 (118–174)	208 (175–228)	1020 (127–2260)	567 (75–1900)
NO_2^-	nd (nd–nd)	nd (nd–nd)	nd (nd–nd)	nd (nd–nd)	nd (nd–nd)	nd (nd–nd)
NO_3^-	nd (nd–nd)	nd (nd–nd)	4 (nd–8.7)	nd (nd–nd)	804 (60–2690)	669 (41–2160)
SO_4^{2-}	0.6 (nd–1.1)	0.6 (nd–1.0)	91 (51–130)	53 (49–58)	818 (28–4780)	172 (19–579)
NH_4^+	89	159 (109–209)	6	12 (10–15)	11 (2–39)	7 (0.8–26)
Na^+	521 (375–666)	575 (429–746)	233 (188–277)	224 (179–281)	1030 (138–2320)	502 (141–1240)
K^+	56 (41–70)	54 (40–72)	71 (68–75)	67 (63–73)	260 (26–710)	147 (25–403)
Mg^{2+}	79 (78–81)	69 (56–92)	79 (71–87)	63 (49–72)	181 (22–372)	89 (15–252)
Ca^{2+}	73 (34–112)	67 (38–121)	45 (37–45)	60 (35–95)	363 (33–751)	213 (25–463)
Dissolved gases						
O_2 μM	0.7 (nd–1.3)	6.5 (nd–14)	nd (nd–nd)	nd (nd–nd)	129 (49–196)	146 (89–248)
CO_2 μM	412 (329–494)	1150 (301–2320)	521 (509–533)	1300 (240–3290)	36 (1–124)	297 (15–941)
CH_4 μM	0.9 (0.4–1.5)	226 (0.01–417)	0.04 (0.01–0.08)	48 (0.01–87)	0.01 (0.01–0.04)	0.48 (0.01–2.5)
N_2O nM	10 (9–10)	6 (2–12)	10 (8–13)	8 (nd–12)	89 (9.7–225)	765 (48–3310)

All variables are expressed as mean and range. ‘nd’: not detected.

due to sulfate reduction in the TDG aquifer or the absence of SO_4^{2-} in surface water (Table 2). The SO_4^{2-} in the BCG might be produced by an oxidation of pyrite beneath the peat layer in the Bacho Swamp during periods without water-logging. These observations suggest that the subsurface environment of the To Daeng Swamp was more reduced than that of the Bacho Swamp, in agreement with a larger CH_4 accumulation in TDG than BCG during both dry and wet seasons except for the 1992 wet season.

The ground waters under the sandy plains (SG1 through SG5) were oxygenated (Table 3). Mean concentrations of CO_2, N_2O, and CH_4 were significantly ($P < 0.05$) higher in wet seasons than in dry seasons. However, electrolyte concentrations tended to be higher in dry seasons than in wet seasons. The differences were statistically significant ($P < 0.05$) in cases of Na^+, Mg^{2+}, and Ca^{2+}. These observations suggest a rapid turnover of the aquifer waters in less than one year. Even in wet seasons, the mean CH_4 concentration was much lower than that at TDG and BCG. Levels of N_2O were over-saturated with respect to the atmosphere both during wet and dry seasons. The high N_2O concentrations were accompanied by high levels of NO_3^- (means of 818 and 172 μmoles liter^{-1} in dry and wet periods, respectively). As cropping fields and residential districts occupied a large part of the sandy plains, elevated NO_3^- and N_2O in the sandy plain ground waters is probably due to the anthropogenic input through fertilization and sewage disposal (Ueda et al. 1993). The temporal variations in concentrations of dissolved gases and ionic species may be explained by lateral recharge of the sandy plain aquifers by the swamp ground waters as a result of flooding during high precipitation period.

Excess gases over equilibria in surface waters

The measured and equilibrium gas concentration can be expressed as follows:

$$\Delta \text{gas} = [\text{gas}]_{\text{meas}} - [\text{gas}]_{\text{eq}},$$

where $[\text{gas}]_{\text{meas}}$ and $[\text{gas}]_{\text{eq}}$ are respectively a measured and an equilibrium concentration, and Δgas is the deviation from the equilibrated concentration for the gas of interest. The $-\Delta O_2$ value, also known as an apparent oxygen utilization (AOU), is plotted against ΔCO_2 in Figure 3(a). The $-\Delta O_2$ was positive in most cases but negative at the site PF where the photosynthesis by micro algae on the soil surface was active. Chlorophyll-a concentrations in TD, BC, and Bang Nara River waters were below the detection limit (0.1 μg liter^{-1}) in December, 1991 (T. Yoshioka, unpublished data). These environments lacked submerged macrophytes. Therefore, positive $-\Delta O_2$ values in swamp and river waters may be a result of excess respiration over photosynthesis.

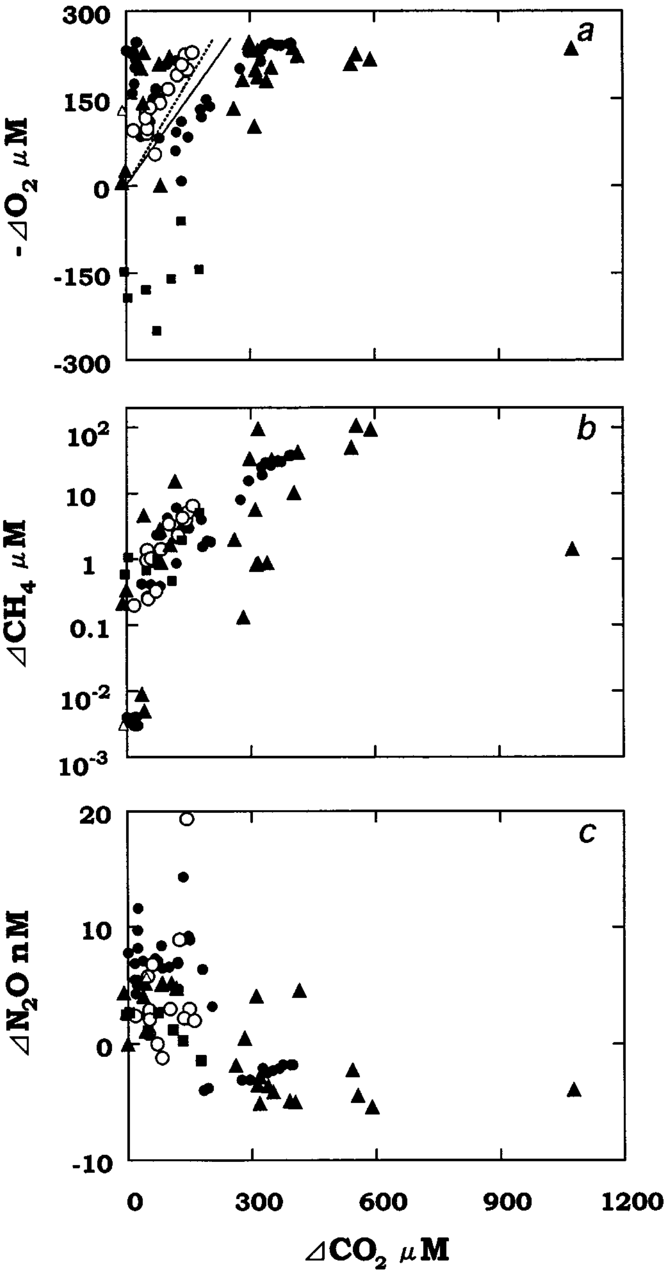

Figure 3. Plots for (a) $-\Delta O_2$, (b) ΔCH_4, and (c) ΔN_2O versus ΔCO_2 in swamps (triangle), paddy fields (square), and Bang Nara River (circle). Wet and dry seasons data are denoted by closed and open plots, respectively. See text for definition of Δ-value. Solid and broken lines in $-\Delta O_2$ versus ΔCO_2 diagram respectively present the theoretical ratios given by Devol et al. (1995) and Richey et al. (1988).

Since one mole of CO_2 evolves per mole of O_2 respired aerobically, ΔCO_2 is a function of $-\Delta O_2$ in strictly aerobic mineralization. The $-\Delta O_2/\Delta CO_2$ is expressed as D_{O_2}/D_{CO_2} where D presents molecular diffusion coefficient. Richey et al. (1988) defined D_{O_2}/D_{CO_2} as RQ and estimated RQ = 1.2 based on D_{O_2} (2.6×10^{-5} cm^{-2} s^{-1}) and D_{CO_2} (2.1×10^{-5} cm^{-2} s^{-1}) given by Broecker and Peng (1974). Devol et al. (1995) applied RQ = 1.0 for their analysis based on a different data set of molecular diffusion coefficient. The smaller $-\Delta O_2/\Delta CO_2$ than the above idealized ratios would therefore indicate CO_2 production by anaerobic respiration, which corresponds the points to the right of the lines in the diagram. An excess CO_2 production over O_2 consumption could also be attributed to lateral exchange of water between river and floodplain where anaerobic processes and root respiration produced CO_2 without consumption of O_2 (Devol et al. 1995). A considerable number of plots to the left of the lines in Figure 3(a) ($-\Delta O_2/\Delta CO_2 >$ RQ) points to O_2 consumption without CO_2 evolution, such as the oxidation of sulfide. We will discuss some of the evidence from this process in later sections.

The value of ΔCH_4 increased logarithmically as ΔCO_2 increased (Figure 3(b)), as was typically observed in reduced aquatic environments (e.g. Richey et al. 1988; Devol et al. 1988; Happell & Chanton 1993). The value of ΔN_2O tended to decrease as ΔCO_2 increased (Figure 3(c)). The negative values of ΔN_2O corresponded with the samples with large ΔCH_4 and ΔCO_2. This fact suggests that the swamp system acts as a sink for atmospheric N_2O when the surface water is highly reduced.

Spatial variation of dissolved gases in Bacho swamp

The measurements obtained in the grid plot at the Bacho Swamp are summarized in Table 4. Since water was stagnant in and around the grid plot as a result of calm conditions during sampling, variations in measurements can hardly reflect an advective water movement. Concentrations of O_2 were below those of CO_2, CH_4, and N_2O which were above those dictated by atmospheric equilibria. Concentrations of these gases varied irrespective of water depth. The concentrations of O_2, CO_2, and CH_4 were not significantly related to pH of water, most probably due to small variation in the pH values. However, the N_2O concentrations correlated positively with the pH values ($r = 0.23$, $n = 91$, $P < 0.05$). A significant positive correlation between CO_2 and CH_4 ($r = 0.23$, $n = 97$, $P < 0.05$) was observed in aggregated data for surface waters (Figure 3(b)). Concentrations of CH_4 and N_2O were negatively related to O_2 concentration ($r = -0.30$, $n = 99$, $P < 0.01$ for CH_4; $r = -0.29$, $n = 91$, $P < 0.01$ for N_2O). The N_2O concentration was correlated with CH_4 concentration ($r = 0.55$, $n = 92$, $P < 0.01$). These results suggest that the CH_4 and N_2O concentrations in the water are regulated more by their production

Table 4. Results of intensive observation in the grid plot of Bacho swamp.

	Mean	sd	Median	*n*	Min	Max
Water depth cm	15	6	15	99	4	38
pH	4.2	0.1	4.2	99	3.9	4.4
O_2 μM	166	20	165	99	120	207
CO_2 μM	126	25	124	97	77	210
CH_4 μM	1.33	0.67	1.19	99	0.44	3.84
CH_4 μmoles m^{-2} h^{-1}	70	72	29	20	12	788
N_2O nM	10.9	0.9	10.7	92	9.3	13.8

than their consumption. The measurements in Table 4, however, may not be representative for the wet season because of an atypical rainfall pattern. Total precipitation in September and October of 1993 was 2.7 times greater than that of 30-year average, while that in November prior to the sampling was less than half of the 30-year average.

The SE/MEAN per number of measurements for O_2, CO_2, and N_2O never exceeded 10% (Figure 4), so spatial variations in concentrations of these gases in the 0.8 hectare area of the grid are much smaller than those observed at different times and sites. Larger site to site variation for the CH_4 concentration probably reflects spatial heterogeneity in methanogenesis in the peat soil (Figure 5). However, the CH_4 concentrations in the grid plot are less variable than the measurements for different sites during wet and dry seasons. The SE/MEAN in CH_4 concentrations decreases slowly with the addition of more samples, reaching roughly 8% of the mean beyond about 20 measurements (Figure 4). The SE/MEAN for CH_4 flux measurements was higher (54% of the mean at 20 samples) than that for the concentration of dissolved CH_4. High variability in the CH_4 flux was observed in the Everglades, Florida (Bartlett et al. 1989) and in the Hudson Bay lowland (Moore et al. 1994), with the SE/MEAN ratios in 30–60% range for forty measurements. Episodic ebullition accounts for such large variations in CH_4 emissions from wetlands (Crill et al. 1988; Bartlett et al. 1988, 1990; Devol et al. 1988, 1990; Wassmann et al. 1992; Shurpali et al. 1993).

Diffusive gas exchange between water and atmosphere (F) can be expressed by an empirical boundary layer model, $F = D\Delta gas/Z$, in which D is the coefficient of molecular diffusion, Δgas the excess concentration over atmospheric equilibrium, and Z the thickness of a stagnant boundary layer (Liss & Slater 1974). Here, $D_{CH_4} = 2.3 \times 10^{-5}$ cm^{-2} s^{-1} (Broecker & Peng 1974) was applied to calculate diffusive emission. The Z was assumed at 310 μm which was appropriate for low wind-speed condition (Devol et al. 1988).

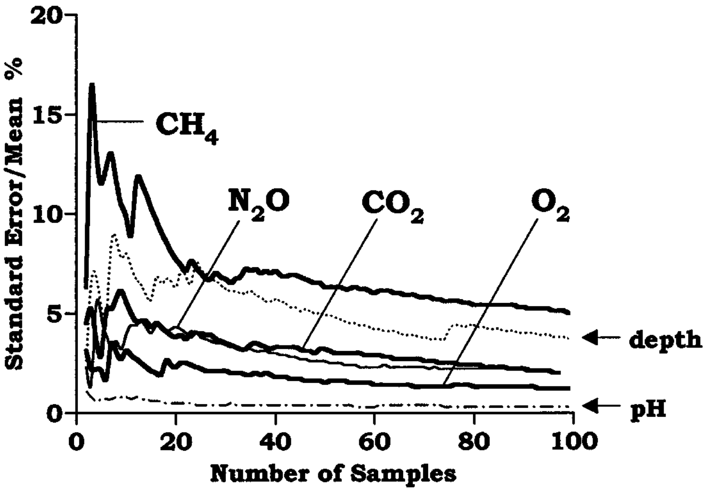

Figure 4. Relationship between standard error of the mean, as a percent of the mean, and number of measurements for concentrations of dissolved gases, water depth, and pH in the grid plot of Bacho Swamp.

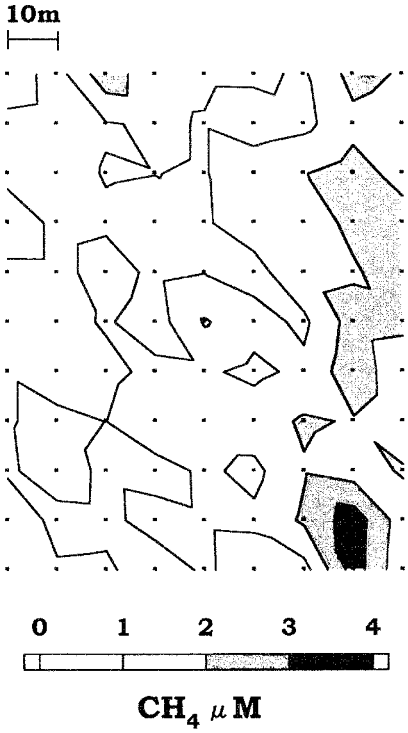

Figure 5. Distribution of dissolved CH_4 in the Bacho Swamp intensive grid. Black points indicate locations of sample collection.

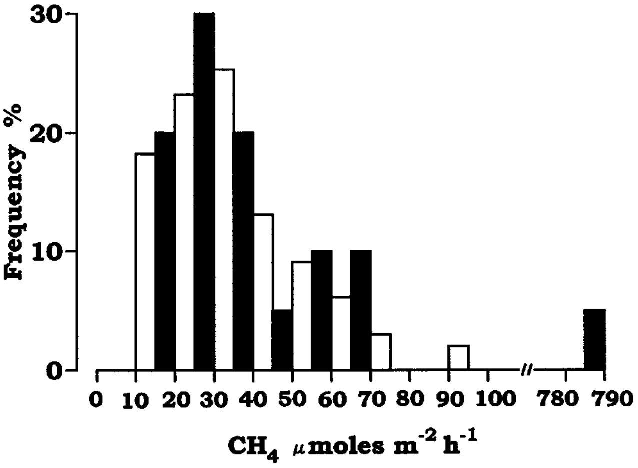

Figure 6. Percent frequency distribution of the CH_4 flux measured by static chamber [black bars, $n = 20$] and calculated by boundary layer model [white bars, $n = 99$].

Frequency distribution of the diffusive CH_4 flux estimated from the surface dissolved CH_4 concentrations by the boundary-layer model resembles that of measured CH_4 flux by the chamber method with mean of 67±169 (1 σ) μmoles m^{-2} h^{-1} (Figure 6). The largest CH_4 flux measured by the chamber was more than an order of magnitude greater than the rest of the measurements, and was apparently due to ebullition. When this measurement was excluded, the mean CH_4 flux by the chamber method became 33±16 μmoles m^{-2} h^{-1}, very close to that by the boundary-layer model (36±14 μmoles m^{-2} h^{-1}). We can estimate the diffusive component of the measured fluxes and thus by subtraction estimate the contribution made by ebullition. However, the one-time detection of ebullition in twenty measurements is insufficient to evaluate the contribution of ebullition in CH_4 transport to the atmosphere.

Temporal difference in gas concentration in Bang Nara River

Since the Bang Nara River receives water from the surrounding swamps, the riverine gas concentrations yield the information on the gas dynamics in the entire swamp system. The O_2 concentration in the Bang Nara River was highly variable during the period of sampling along the river (Figure 7). Generally, O_2 concentrations were low at 18 km upstream of R1, and tended to increase as water flowed down to R1. Dissolution of the atmospheric O_2 and the discharge of oxygenated water from tributaries flowing the upland area are likely to have caused such an increase in the O_2 concentration. The lateral input of the oxygenated water is conspicuous at R6, a junction of a tributary.

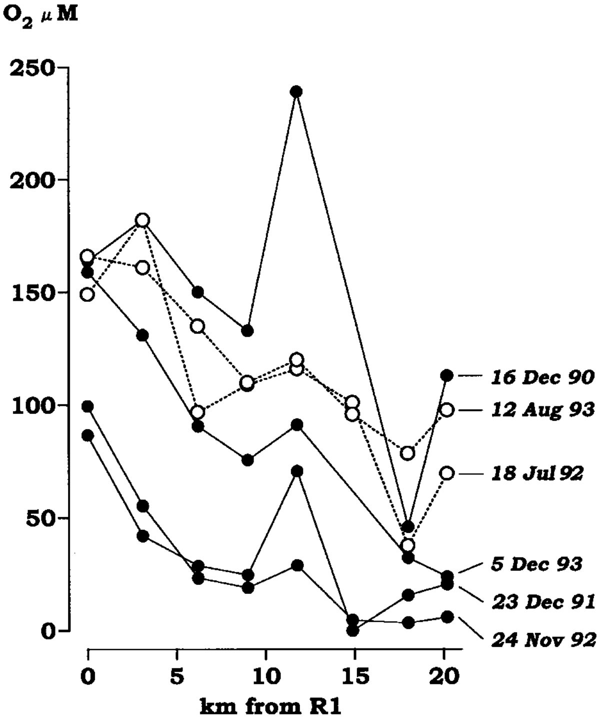

Figure 7. Concentration of dissolved O_2 in the Bang Nara River. Data obtained during wet and dry seasons are denoted by filled and open symbols, respectively.

The results of the river survey fell into three groups. The first group comprises the results obtained during the wet seasons of 23 December, 1991 and 24 November, 1992. Both have relatively low dissolved O_2. The O_2 concentrations were around 100 μmoles liter^{-1} at R1, and were less than 30 μmoles liter^{-1} in the 15–20 km upstream water. The second group, with observations made during the dry seasons of July, 1992 and August, 1993, showed higher levels of dissolved O_2. The third group comprises measurements on 16 December, 1990 and 5 December, 1993. Although made during the wet season, these had higher O_2 levels than those of the first group and more closely resembled those measured during the dry season. These results indicate that the balance between oxidation-reduction conditions shown by dissolved O_2 concentrations in the Bang Nara River is not simply determined by the cycle of wet and dry seasons.

The variation of concentrations of dissolved gases and NO_3^- along the river course differed between 23 December, 1991 and 24 November, 1992 (Figure 8), while the O_2 distributions were similar among those times (Figure 7). The CO_2 as well as CH_4 concentrations were higher on 23 December,

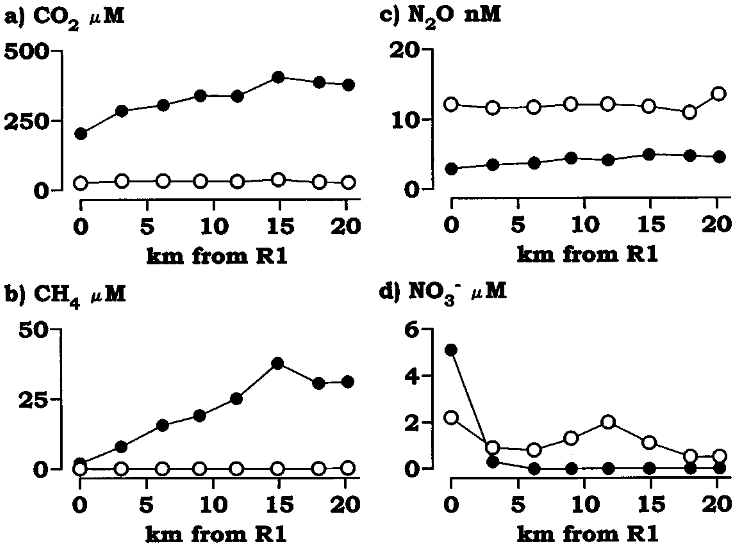

Figure 8. Distributions of (a) CO_2, (b) CH_4, (c) N_2O, and (d) NO_3^- in the Bang Nara River on 23 December 1991 [●] and 24 November 1992 [○].

1991 than those on 24 November, 1992 (Figures 8(a, b)). The opposite was the case for dissolved N_2O (Figure 8(c)). The N_2O concentrations were below atmospheric equilibria (ca. 6 nmoles liter^{-1}) on 23 December 1991. The under-saturation of N_2O accompanied by undetectable levels of NO_3^- in R3 through R8 on 23 December, 1991 suggests the *in situ* reduction of these compounds by denitrification. Low levels of CO_2 and CH_4 and over-saturation of N_2O were characteristics for 24 November, 1992, possibly due to enhanced aerobic mineralization and subsequent oxidation in the river and watershed under the drought condition. Similar O_2 distribution between the two observations suggests that the O_2 levels depended more on the atmospheric exchange and discharge through the tributaries than on riverine metabolisms. As O_2 and the other dissolved gases were apparently controlled by different mechanisms, the use of the dissolved O_2 levels is a poor predictor for the concentrations of dissolved CO_2, CH_4, and N_2O in the Bang Nara River. Similar independency of O_2 and CO_2 or CH_4 was observed in other swamp systems where rainfall inputs oxygenated the flood water and diluted CO_2 and CH_4 (Happell & Chanton 1993).

Excess SO_4^{2-} as oxidation-reduction indicator

The redox condition in the swamp system should be reflected not only in gas distribution but also in water chemistry. In the Bang Nara River, the

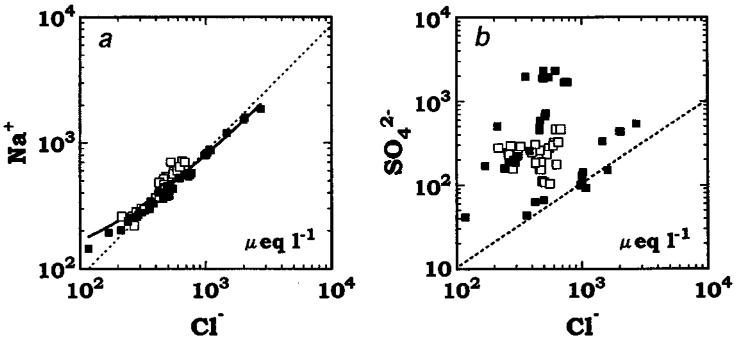

Figure 9. Diagrams for (a) Na^+ and (b) SO_4^{2-} versus Cl^- in the Bang Nara River. ■-wet period data (23 December 1991; 24 November 1992; 5 December 1993). □-dry period data (18 July 1992; 12 August 1993). Broken lines present Na^+/Cl^- and SO_4^{2-}/Cl^- equivalent ratios of sea water (Willson 1975). Solid line in Na^+ versus Cl^- diagram denotes linear regression for pooled data set of wet and dry periods; $[Na^+] = 0.71[Cl^-]+97$ ($r = 0.98$, $n = 67$).

concentrations of Cl^- and Na^+ are well correlated ($r = 0.98$, $n = 67$) and scatter around the theoretical dilution line (TDL) of sea water (Figure 9(a)). Clearly sea salt is the dominant source for Cl^- and Na^+, and these solutes can be applied as conservative tracers in the watershed. By contrast, significant sources for the SO_4^{2-} other than the sea salt are suggested by concentrations of SO_4^{2-} located above the TDL (Figure 9(b)). This fact suggests that there are significant sources for the SO_4^{2-} other than sea salt dissolution in the watershed.

The excess SO_4^{2-} over Cl^- in the Bang Nara River contrasts with the observation in a salt marsh along a tidal creek where SO_4^{2-} falls below the TDL due to an active sulfate reduction (Bartlett et al. 1987). In temperate salt marsh systems, sulfur cycling is active through periodical intrusion of seawater SO_4^{2-}, FeS_2 production by SO_4^{2-} reduction during the high temperature period, and FeS_2 oxidation to release SO_4^{2-} during the low temperature period (Luther et al. 1982, 1986). In our coastal peat swamp, however, FeS_2 now buried beneath the thick peat layer was produced in the past when the swamp was under brackish condition, while the intrusion of seawater SO_4^{2-} is no longer present.

The FeS_2 in the mud clay can be oxidized under aerobic conditions to produce SO_4^{2-} by the following reactions (Van Breemen 1978).

$$FeS_2 + 15/4O_2 + 7/2H_2O \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4H^+,$$

$$FeS_2 + 15/4O_2 + 5/2H_2O + 1/3K^+ \rightarrow 1/3KFe_3(SO_4)_2(OH)_6$$
$$+ 4/3SO_4^{2-} + H^+.$$

In addition to pyrite oxidation, SO_4^{2-} is produced through hydrolysis of the jarosite ($KFe_3(SO_4)_2(OH)_6$) via the following reaction.

$$KFe_3\,(SO_4)_2(OH)_6 \rightarrow 3FeOOH + 2SO_4^{2-} + K^+ + 3H^+.$$

Oxidation of the reduced sulfur compounds takes place where peat layers are thin, and water table are seasonally low. In fact, straw yellow mottles of jarosite was observed in the transitional areas from swamps to sandy plains. So FeS_2 oxidation in the swamp fringe is probably a source for the excess SO_4^{2-} in the Bang Nara River water. Oxygen consumption by oxidation of FeS_2 in the Bang Nara River watershed may also partly account for the differences between the $-\Delta O_2$ vs ΔCO_2 diagram presented here (Figure 3(a)) and those reported for the Amazon floodplain (Richey et al. 1988; Devol et al. 1988; Devol et al. 1995). To further test this hypothesis, an accurate oxygen budget at the watershed level is required.

The excess SO_4^{2-} amount in the river water would fluctuate according to flooding depth, if oxidation of FeS_2 releases the SO_4^{2-}. Temporal changes in SO_4^{2-}/Cl^- at the north and central tidal gates of the Bang Nara River, NTG and CTG (Figure 10) tend to follow that of precipitation, but the correlation is not significant ($r = 0.44$, $n = 9$). This is probably because the SO_4^{2-}/Cl^- is a single time point while the precipitation is a monthly aggregate. However, SO_4^{2-}/Cl^- peaked in September, after heavy rain (713 mm month^{-1}) following two dry months (264 mm in total). The SO_4^{2-}/Cl^- was also high during a wet June (302 mm month^{-1}) after a dry May (65 mm month^{-1}). The following sequences could explain such fluctuation in SO_4^{2-}/Cl^-: (1) SO_4^{2-} is produced through FeS_2 oxidation in the fringe of the swamp during a dry period, (2) SO_4^{2-} is flushed into the river by high precipitation, and (3) depressed SO_4^{2-} production during flooding of the watershed.

If the excess SO_4^{2-} reflects the amount of drainage of the catchment, the variations of dissolved gases also produced in soils should be related to the change in SO_4^{2-}/Cl^-. Significant correlations indeed exist between the SO_4^{2-}/Cl^- and CO_2 (Figure 11(a), $P < 0.01$), CH_4 (Figure 11(b), $P < 0.01$), and N_2O (Figure 11(c), $P < 0.01$) in the Bang Nara River. When the river SO_4^{2-}/Cl^- was close to the equivalent ratio in sea water (0.104), in other words the swamp fringe would have been kept in reduced condition, the CO_2 and CH_4 concentrations were high (Figures 11(a, b)). The CO_2 and CH_4 concentrations decreased as the SO_4^{2-}/Cl^- increased, which was probably due to suppression of anaerobic production of these gases under oxic condition. The N_2O concentration was below atmospheric equilibrium (ca. 6 nmoles liter^{-1}) when the SO_4^{2-}/Cl^- approached to the sea water ratio, and decreased as the SO_4^{2-}/Cl^- decreased (Figure 11(c)). This points to N_2O

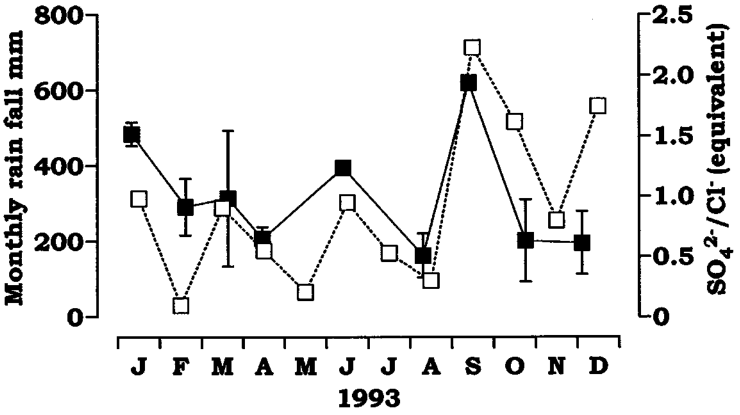

Figure 10. Mean with range of SO_4^{2-}/Cl^- at NTG and CTG [■], and monthly precipitation recorded by Meteorological Department of Thailand near R1 [□].

reduction to N_2 under highly reduced condition. The O_2 concentrations were poorly correlated with SO_4^{2-}/Cl^- (Figure 11(d)), in agreement with the view that the O_2 level did not always indicate the oxidation-reduction condition in the watershed. This is because that precipitation supplies O_2 to the water but hardly alters SO_4^{2-}/Cl^-. The SO_4^{2-}/Cl^- in the Bang Nara River was a useful indicator for the analysis of the dynamics of CO_2, CH_4, and N_2O in the study area.

Conclusion

Concentrations of different gases in a coastal swamp system in Thailand varied widely in swamp water, river water, and ground waters: 0–248 μmoles O_2 liter^{-1}; 1–3290 μmoles CO_2 liter^{-1}; 0.01–417 μmoles CH_4 liter^{-1}; 0–3.3 μmoles N_2O liter^{-1}. The CO_2 and CH_4 concentrations were high in reduced environments, particularly in swamp ground water. N_2O was probably consumed in both surface and ground waters under highly reduced conditions, and accumulated only in the nitrate-contaminated aerobic ground waters of the sandy plains surrounding the swamps. Temporal variations in gas concentrations could be attributed to the fluctuation of oxidation-reduction conditions within the system as determined by seasonal variations in precipitation. The release of SO_4^{2-} through pyrite oxidation between swamp and sandy plain caused high value of SO_4^{2-}/Cl^-, which correlated well with concentrations of CO_2, CH_4, and N_2O. Therefore, the SO_4^{2-}/Cl^- in the river water could be used to characterize redox conditions in the watershed.

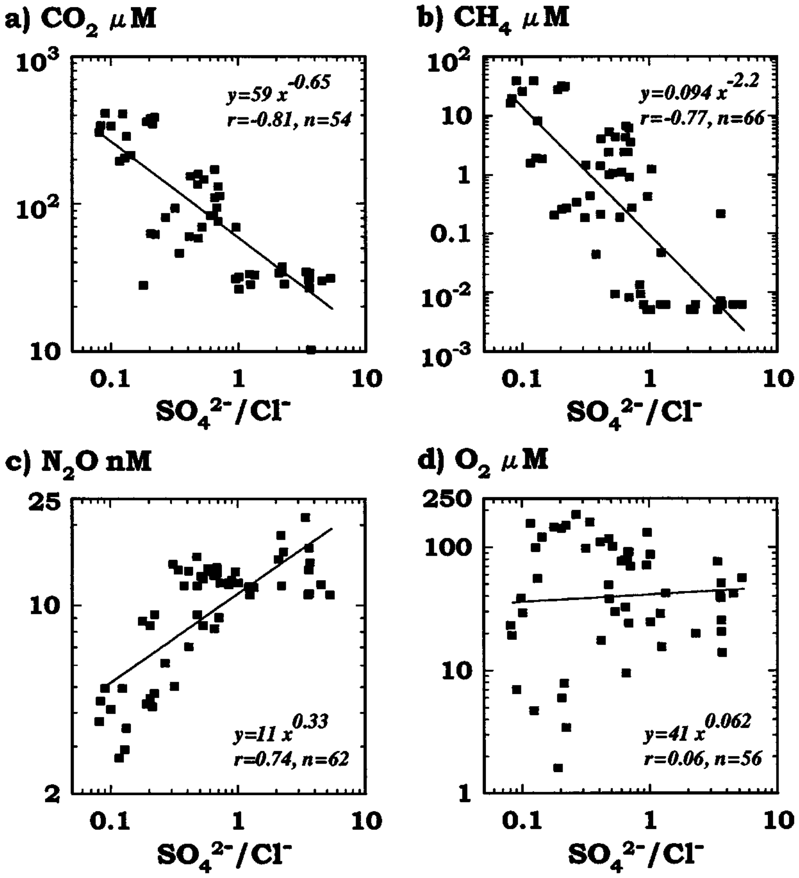

Figure 11. Regressions of (a) CO_2, (b) CH_4, (c) N_2O, and (d) O_2 concentrations to the SO_4^{2-}/Cl^- in the Bang Nara River.

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