METHANE FLUX FROM AN EUTROPHIC LAKE KASUMIGAURA, JAPAN

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To estimate methane flux from inland water environment, measurements of methane concentration, production, air–water exchange and oxidation in Japanese fresh water lakes have been started.

An automated methane analyzer for natural water samples, utilizing a purge and trap/FID–GC technique, was developed and applied. Sub nano–mol/l levels of dissolved methane can be measured so as to accurately determine the equilibrated concentration with atmospheric methane.

Lake Kasumigaura is a shallow eutrophicated freshwater lake, having surface area of 171 km^2, 2nd largest in Japan, and the average depth of 4 m. Monthly observation has been continued since 1976 for the monitoring of lake water eutrophication and for limnological studies by our institute. Dissolved concentration of methane and its distribution in Lake Kasumigaura have been measured monthly since Apr. 1990. Because of the shallow depth, the lake water is not stratified all over the year. The lake water methane concentration was far higher than the equilibrated concentration (2–4 nmol/l) with the atmospheric methane during the whole measurement period. Higher concentrations were observed in the arm of the lake, where the organic carbon contents in the water and in the sediment are higher. Vertical distribution of dissolved methane is usually uniform, that is consistent with the results for other chemical components. The variation of averaged concentration for 5 or 6 of monthly observed stations in the lake is shown in Fig.1. The annual averaged surface water concentration was 150 nmol/l (Apr.1990–Mar.1991). High concentrations were observed at early autumn and winter. The maximum in early autumn was coincided with the season of maximum decomposition of organic carbon produced in the lake. The winter maximum may related to the low methane oxidation rate in the lake water and smaller air–water exchange flux.

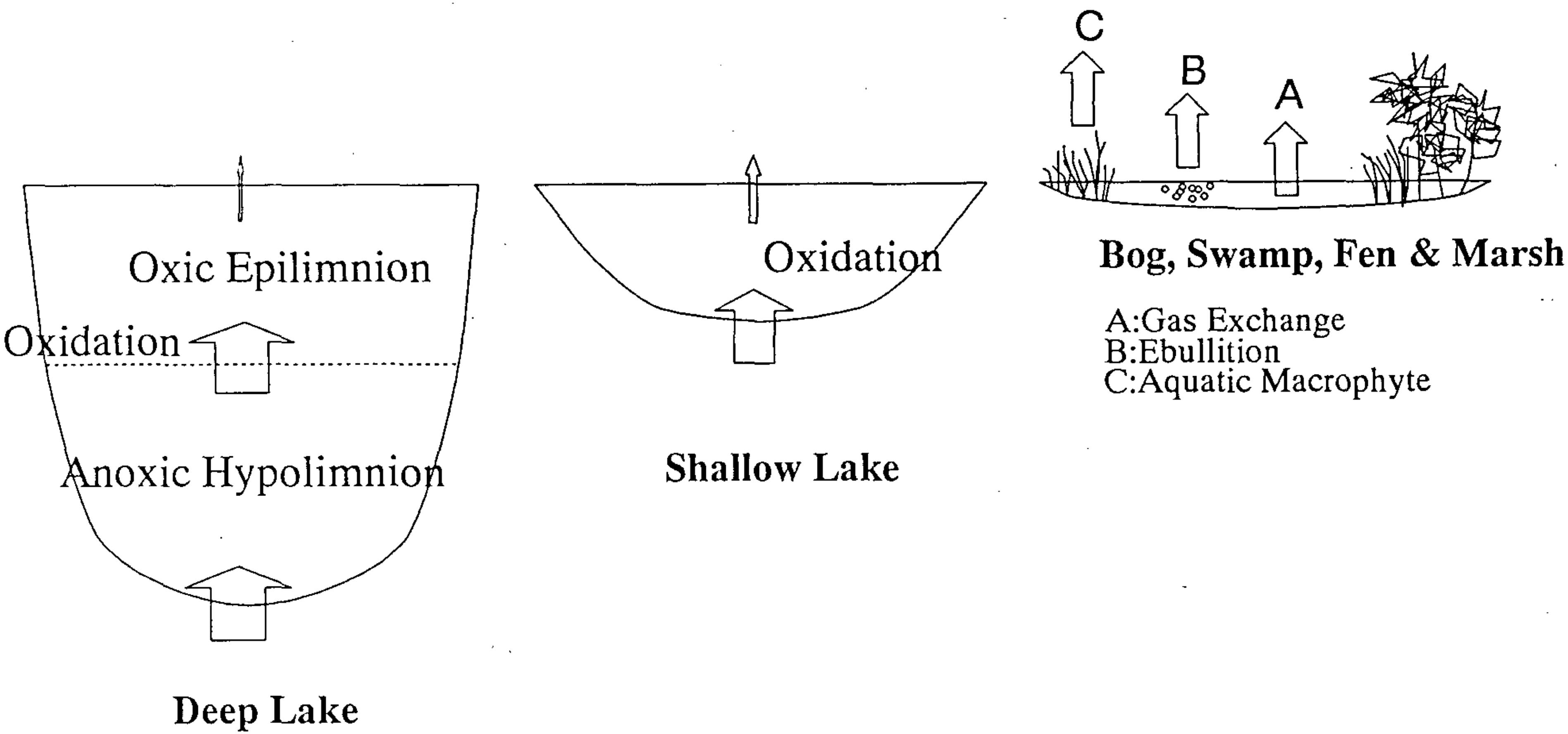
As oxygen is not depleted from Lake Kasumigaura water all over the year, production of methane in water would be quite small, then the major source of the dissolved methane should be the biological process in the bottom sediment and diffused to lake water. The lake water concentration is controlled by the diffusive flux from the sediment, oxidation in the lake water and air–water exchange. The bacterial oxidation rates of methane in the water were measured for various seasons. Usually, the oxidation of methane followed the equation of 1st order reaction. The observed rate in Aug.1990 were about 0.086 1/h, which corresponds the half life of 8 hour for dissolved methane. In this condition, air–water exchange flux is supposed to be smaller than the decomposition in the water column. While, the oxidation rate in the following winter and spring decreased drastically.

The flux of methane across the air–water interface was calculated with wind velocity, water temperature and empirical gas exchange model (Sebacher et al., 1983). Estimated annual flux was 1.1 g $CH_4/m^2/y$ (Apr.1990–Mar.1991), and the seasonal variation is shown in Fig.1. The annual flux from Lake Kasumigaura is about 1/10 of that reported from freshwater lakes in Florida (Barber et al., 1988). Observed methane concentrations from Japanese oligotrophic lakes are far lower than that in Lake Kasumigaura. A close relationship between methane concentration and lake productivity was observed with our results. We are continuing to estimate total methane flux from Japanese wetlands. To elucidate the relationship between lake environment and methane flux is the purpose of this study. The global flux estimation for methane from freshwater environment must be examined with the knowledge of the methane production, oxidation, gas–exchange process, and with precise field data.

Methane flux from freshwater lakes

Flux mg CH_4/m^2/day	Range	Month	Location	Authors	Year
49	24–74	summer mean	Manitoba	Rudd & Hamilton	1978
37	5–250	yearly mean	Louisiana	DeLaune *et al.*	1983
18.4	1.9–180	Aug.–Nov.	S.California	Cicerone & Shetter	1981
27	22–32	July/Aug.	Central Amazon	Bartlett *et al.*	1988
120	60–180	July/Aug.	Amazon	Devol *et al.*	1988
4.2	0.8–26	yearly mean	Kasumigaura	This work	1991

compiled by Aselmann & Crutzen (1989)

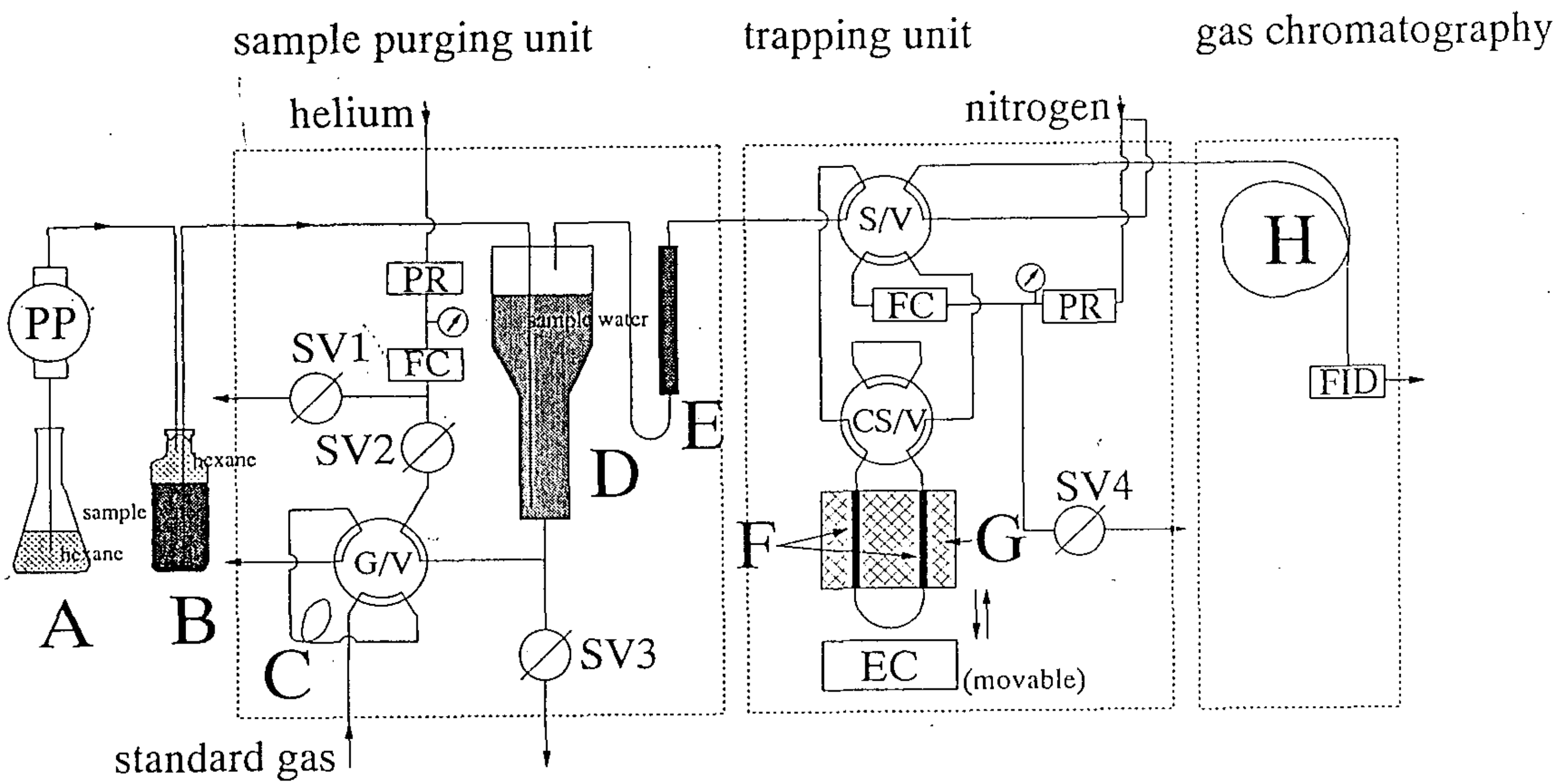


Methane Flux from Natural Wetlands

Production = △ (Concentration) + Oxidation + Diffusive flux to air

sediment — water column — water column — water surface

Schematic Diagram of Automated Analyzer for Dissolved Methane



A : hexane bottle,
B : glass sample vial,
C : loop for standard gas,
D : sample purging vessel,
E : silica gel drying column,
F : charcoal trapping column,
G : Al block with electric heater,
H : column for GC,
SV : solenoid valves,
G/V : standard gas sampling valve,
S/V: sample injection valve,
CS/V: concentrated gas sampling valve,
PR : pressure regulator,
EC : electric cooling unit,
PP : plunger pump,
FID : flame ionization detector.

Limnological parameters of Lake Kasumigaura

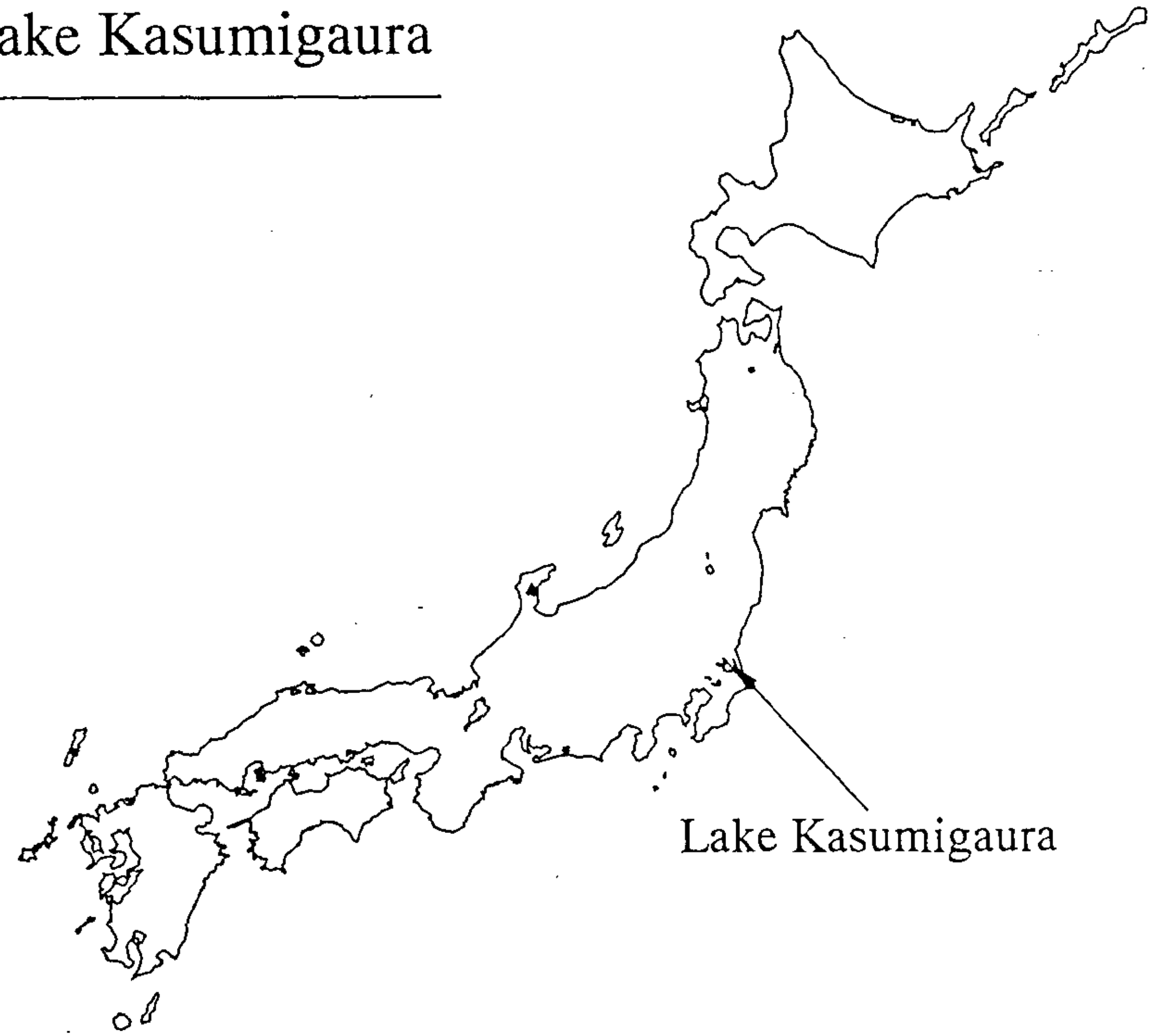
Location, 36° 0'N, 140° 30'E

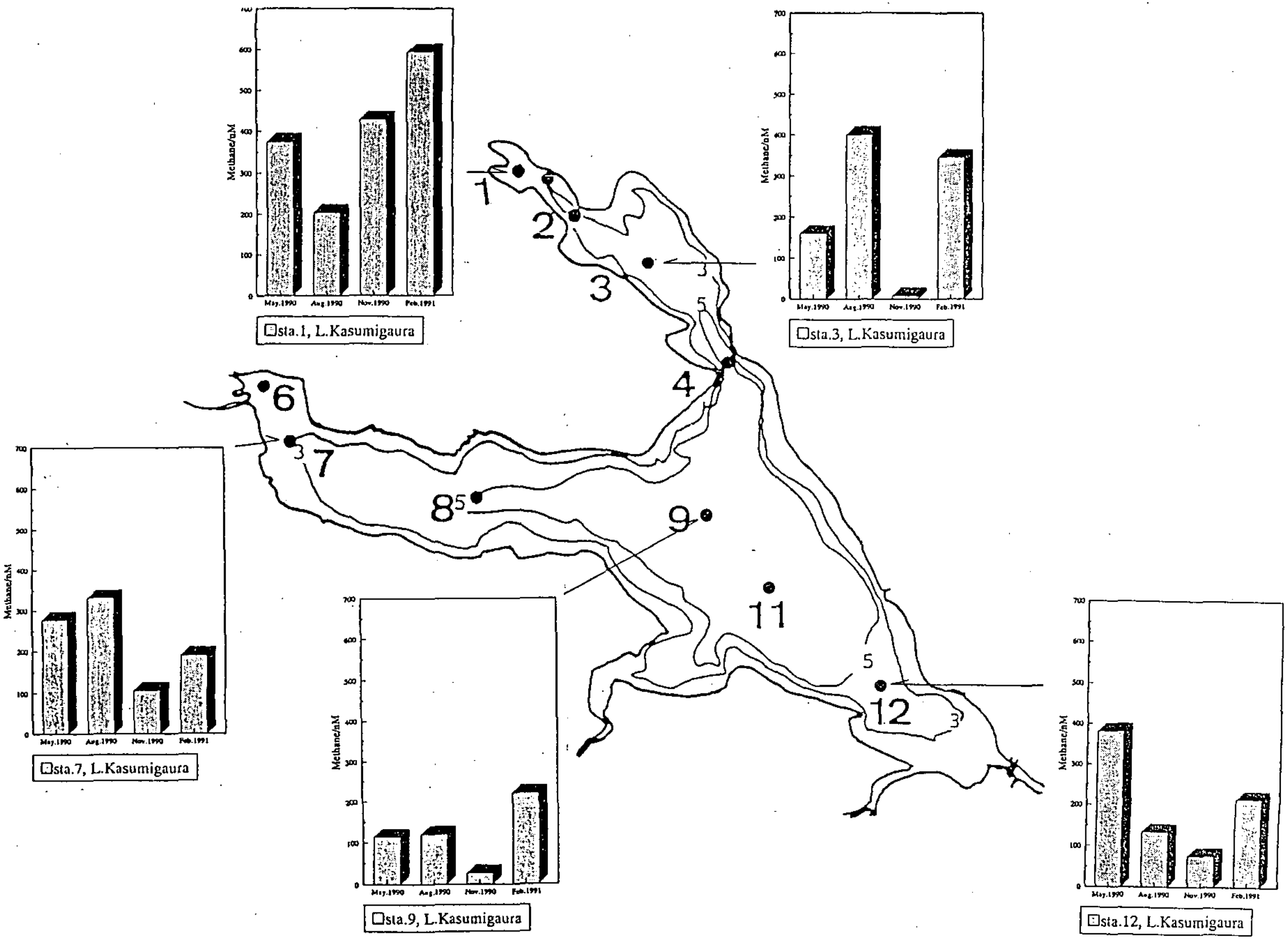
Area, 171 km^2 (second largest in Japan)

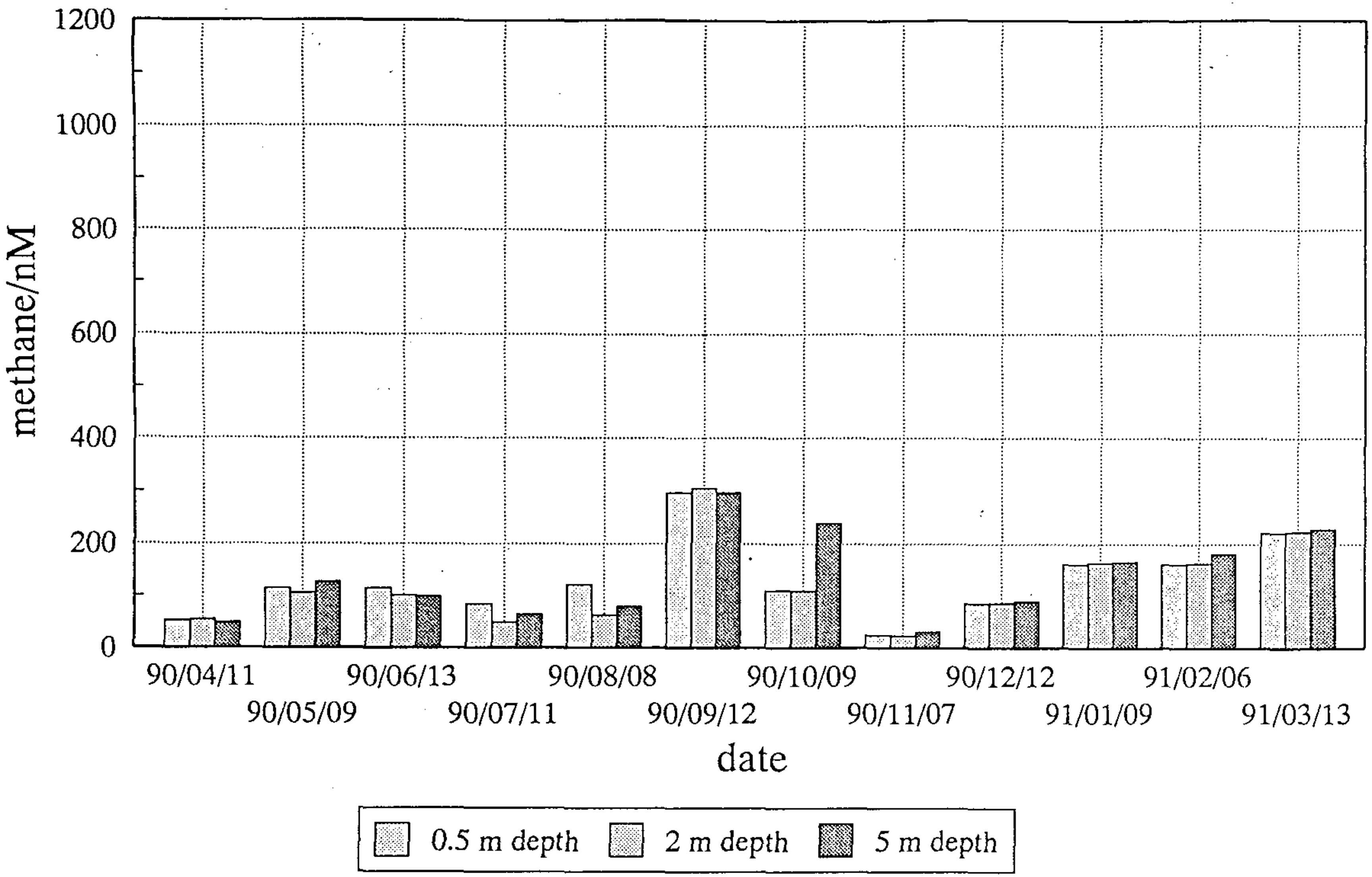
Mean depth, 3.9 m

Water volume, 0.66 km^3

Trophic level, Eutrophicated

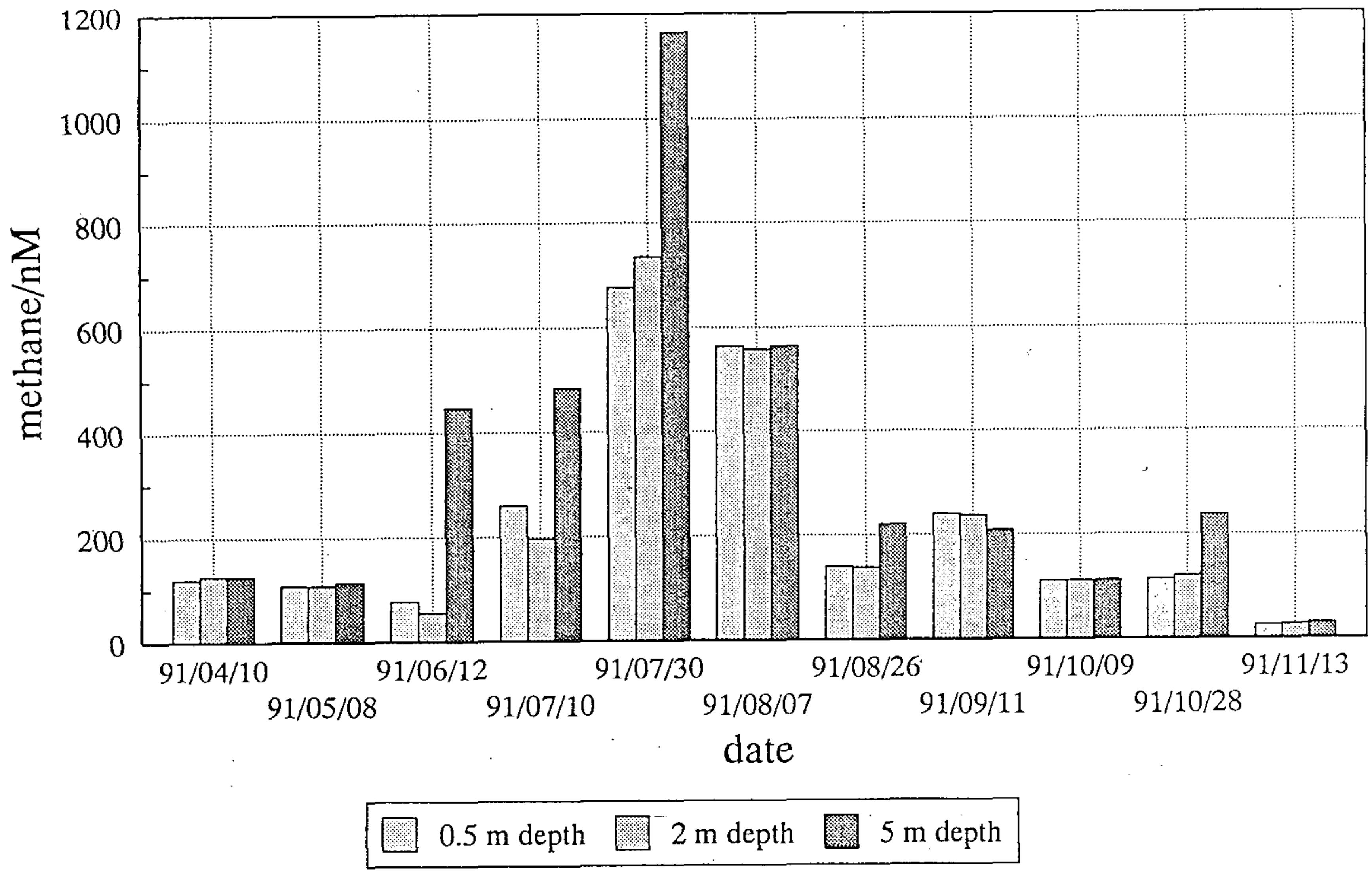



□sta.1, L.Kasumigaura
□sta.3, L.Kasumigaura
□sta.7, L.Kasumigaura
□sta.9, L.Kasumigaura
□sta.12, L.Kasumigaura
Methane/nM
May.1990
Aug.1990
Nov.1990
Feb.1991
1
2
3
4
6
7
8
9
11
12



Variation of methane concentration in water

Center of Lake Kasumigaura (sta.9), from Apr.1990–Nov.1991


methane/nM
1200
1000
800
600
400
200
0
91/04/10
91/05/08
91/06/12
91/07/10
91/07/30
91/08/07
91/08/26
91/09/11
91/10/09
91/10/28
91/11/13
date
0.5 m depth
2 m depth
5 m depth

Theory for air–water gas exchange

$$\text{Flux} = k_l (C_l - C_a / H)$$

k_l :piston verocity, C_l:conc. in water, C_a :conc. in air, H :Henry's law const.

From chamber experiment by Sebacher et al. (1983);

$$k_l = 1.1 + 1.2\, v^{1.96} \qquad \text{(cm/h)}$$

v :wind velocity (m/s) at 2 cm height from water surface

Temperature effect on k must be corrected by viscosity of water.

Wind velocity must be corrected for observation height.

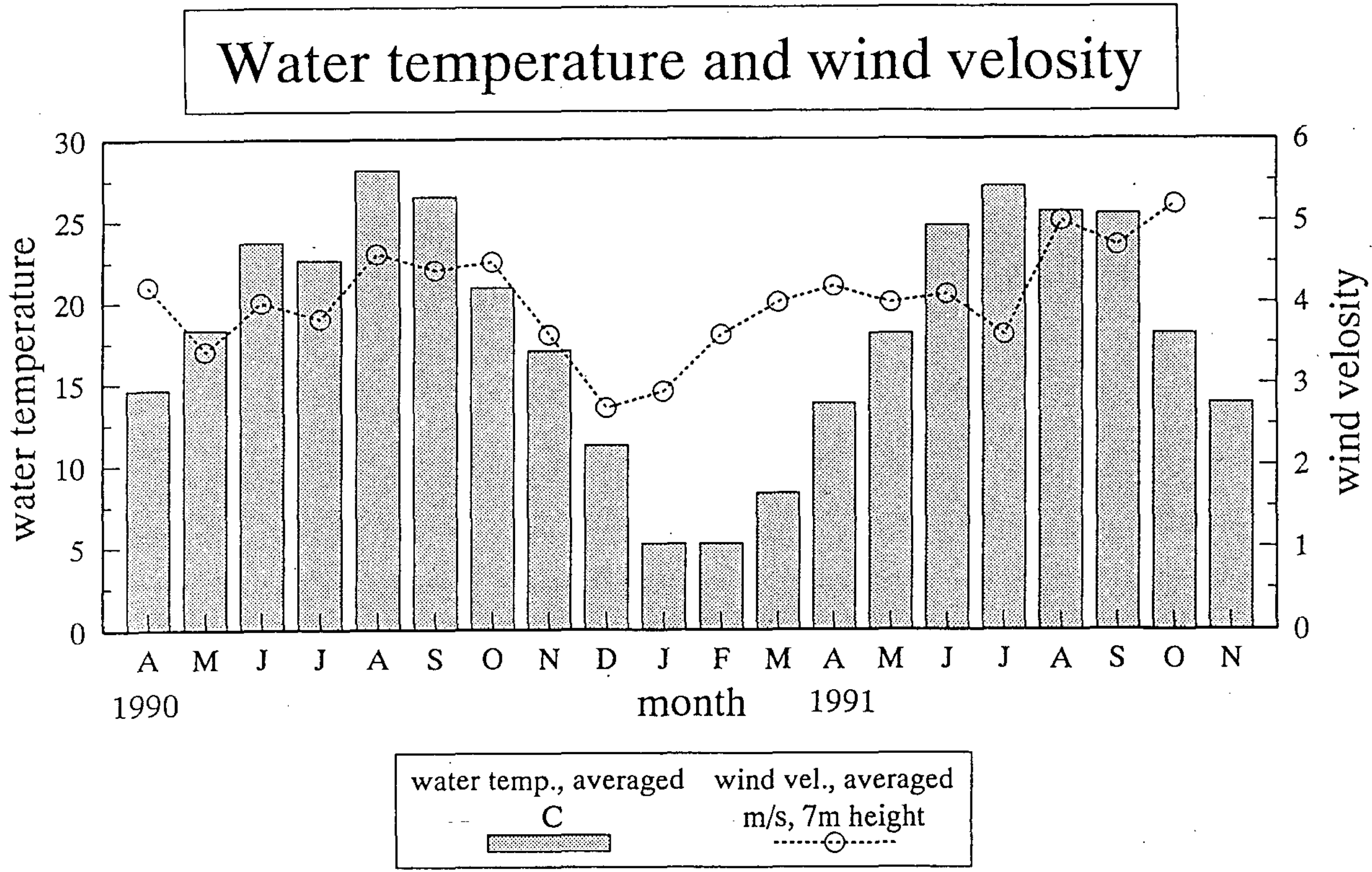
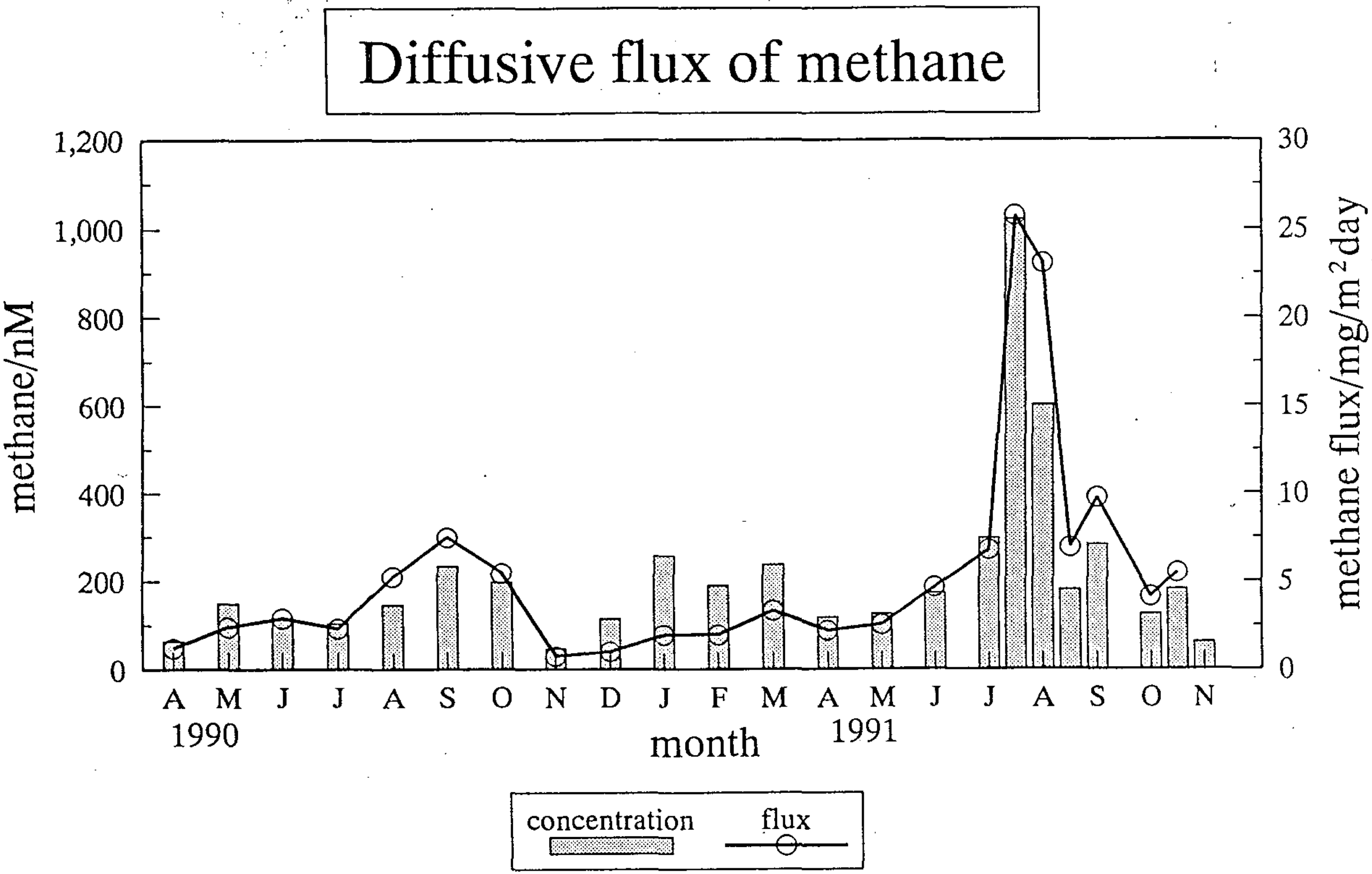
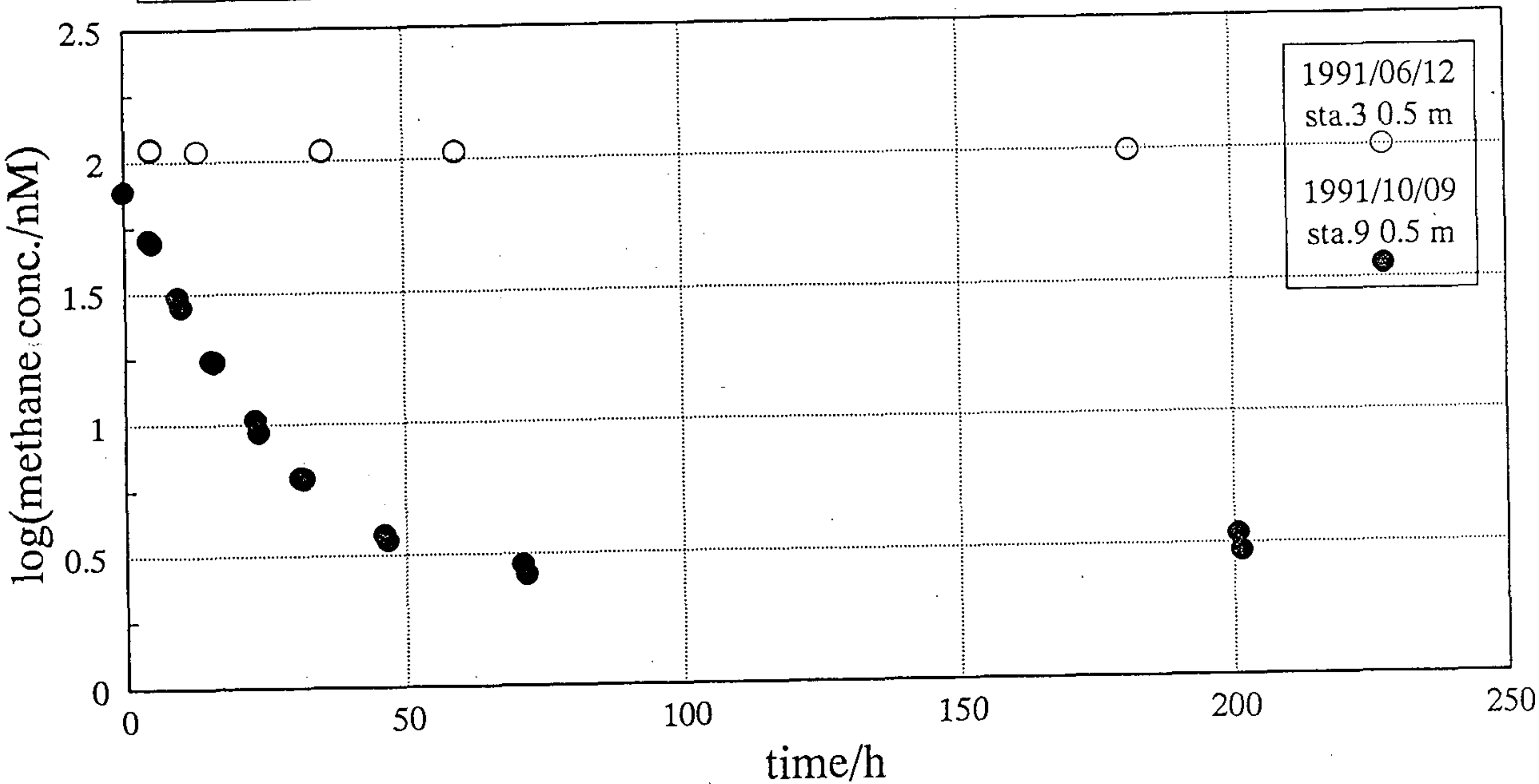

Water temperature and wind velosity
water temperature
wind velosity
30
25
20
15
10
5
0
6
5
4
3
2
1
0
A M J J A S O N D J F M A M J J A S O N
1990
month
1991
water temp., averaged
C
wind vel., averaged
m/s, 7m height

Fig. 1. in abstract



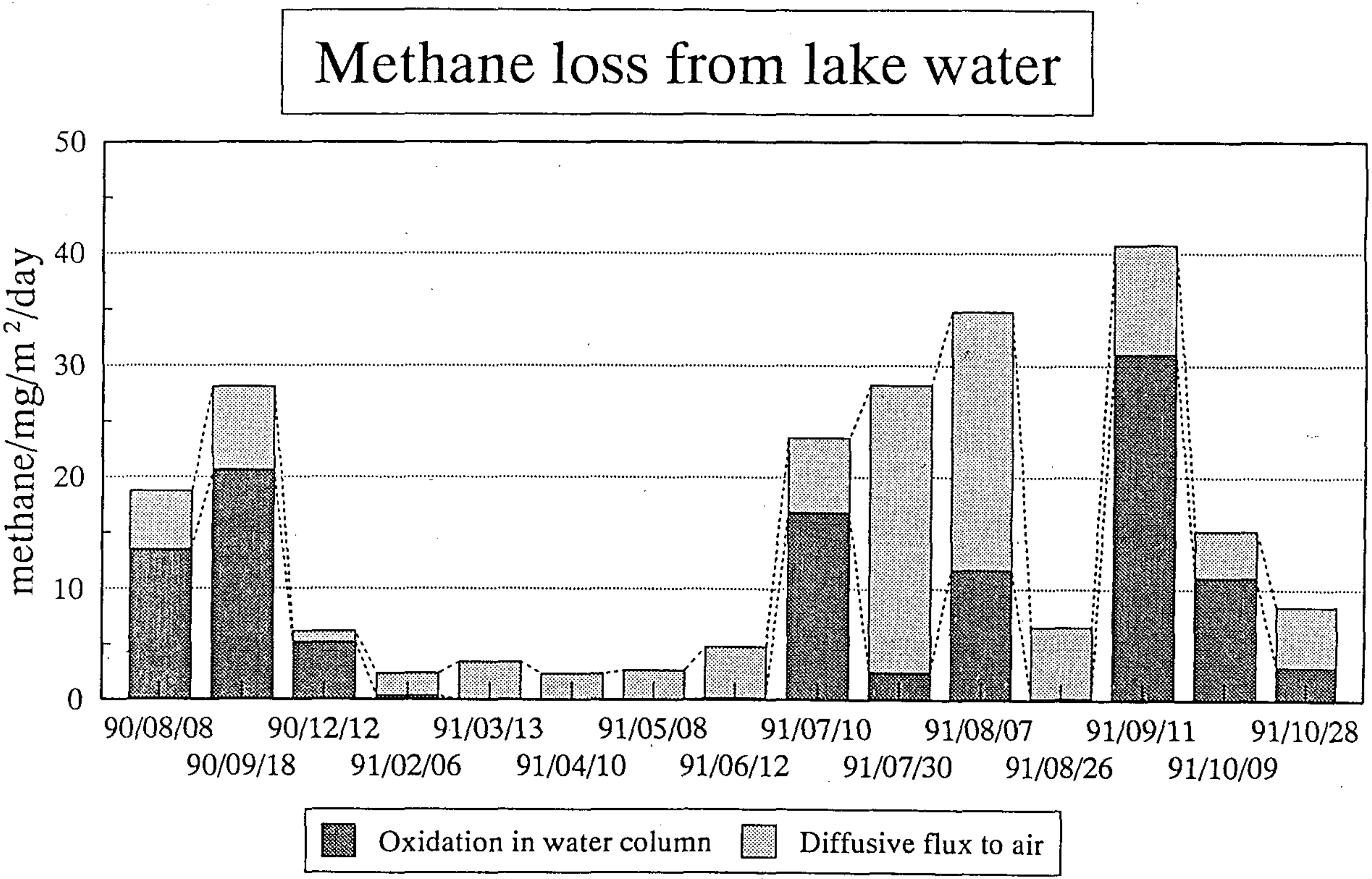
Example of methane oxidation experiment

Incubation at lake water temperature



Methane oxidation rate in Lake Kasumigaura water

date	station	T (C)	Ini.conc. (nM)	0 order (nM/h)	1st order (1/h)	half life (h)
90/08/09	3	27	781	54	0.086	8
	9	27	208	8.4	0.086	8
90/09/18	9	26	130	6.3	0.082	8
90/12/12	3	11	174	5.5	0.042	17
90/02/06	3	5	272	0.81	0.0018	390
90/03/13	3	8	296	0.034	0.00012	5800
90/04/10	3	16	42	0.023	0.00061	1100
90/05/08	3	18	122	0.064	0.00041	1700
90/06/12	3	28	110	0.10	0.00063	1100
90/07/10	3	26	316	0.36	0.00086	810
	9	26	191	7.5	0.059	12
90/07/30	3	29	2417	1.4	0.00062	1100
	9	29	186	0.53	0.0029	240
90/08/07	3	26	961	2.3	0.0026	270
	9	26	375	5.2	0.022	32
90/08/26	3	29	223	0.19	0.00087	800
	9	29	137	0.14	0.0010	660
90/09/11	3	26	259	22	0.18	4
	9	26	36	2.1	0.077	9
90/10/09	3	20	29	2.1	0.056	12
	9	20	79	5.6	0.088	8
90/10/28	3	17	42	1.2	0.029	24
	9	17	97	1.5	0.013	53



Methane loss from lake water
methane/mg/m 2/day
50
40
30
20
10
0
90/08/08
90/09/18
90/12/12
91/02/06
91/03/13
91/04/10
91/05/08
91/06/12
91/07/10
91/07/30
91/08/07
91/08/26
91/09/11
91/10/09
91/10/28
Oxidation in water column
Diffusive flux to air

Emission of methane from wetland

	Emission rate $mg/m^2/day$	Area $10^{12} m^2$	Mean prod. days	Global emission Tg/y
Bog	15	1.87	178	5
Fen	80	1.48	169	20
Swamp	84	1.13	274	26
Marsh	253	0.27	249	17
Floodplain	100	0.82	122	10
Lake	43	0.12	365	2
Global total		5.69		80

Aselmann & Crutzen (1989)

Fifteen % of global metahne emission by this estimation.
By Ciceron & Oremland (1988),
21 % of global emission was attributed to natural wetland.