

Oxidation of dissolved methane in a eutrophic, shallow lake: Lake Kasumigaura, Japan

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Abstract

Long-term seasonal variations in water-column methane oxidation were studied in the eutrophic, shallow Lake Kasumigaura, Japan. The entire water column was oxic throughout the year. Measurements were taken monthly from July 1991 to March 1996. The oxidation rates were derived from time-course dissolved methane concentration measurements during the bottle incubation at in situ temperature. The existence of methane oxidation activity in the oxic lake water was confirmed. Methane oxidation was distinctly seasonal, with low activity from January to April and high activity from August to November. Maximum methane oxidation was observed in late summer or early autumn, when the turnover time of dissolved methane was usually shorter than half a day. Methane oxidation activity does not depend on water temperature, dissolved methane concentration, dissolved oxygen, or dissolved inorganic nitrogen. The annual average methane consumption rate was $12.3 \pm 15.5 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$. Methane oxidation was the dominant methane sink, consuming an annual average of 74% of dissolved methane in the water column of Lake Kasumigaura.

Methane is the most stable carbon compound in anaerobic environments and is a very important intermediate in reactions that eventually lead to organic matter remineralization (Dagley 1978). Methane escapes from anaerobic environments to the atmosphere when it is not oxidized by methanotrophs. The oxidation of methane is now known to occur in both aerobic and anaerobic environments (Hanson and Hanson 1996). Previous study has suggested that freshwater environments are much more important as sources of methane to the atmosphere than are marine environments (cf. Ciccerone and Oremland 1988). The distribution of methane oxidation activity has been studied in several eutrophic lakes (e.g. Rudd et al. 1974; Harrits and Hanson 1980; Iversen et al. 1987), but most previous studies in lakes have been done at locations with permanently or seasonally anoxic hypolimnia, and the studies on methane oxidation in the water column have concentrated mainly on oxic–anoxic interfaces.

Aerobic methane oxidation is often at a maximum where methane and oxygen coexist in the oxic–anoxic transition zone, and it is thought that this serves to remove most of the methane diffusing from deep anoxic layers before it is transported to the atmosphere (Rudd and Hamilton 1975; Ward et al. 1987). If the lake has an anoxic hypolimnion,

methane produced in the anoxic sediment diffuses directly into the anoxic hypolimnion without aerobic methane oxidation. It is then consumed by methanotrophs at the oxic–anoxic interface in the water column (Jannasch 1975; Rudd and Hamilton 1975; Harrits and Hanson 1980; Scranton et al. 1993). On the other hand, if the lake does not have an anoxic hypolimnion, methane diffuses into oxic surface sediment, and is partially consumed by methanotrophs in the sediment (Lidstrom and Somers 1984; Kuivila et al. 1988; Frenzel et al. 1990; King et al. 1990). Only the residual portion of methane, then, can diffuse into the oxic hypolimnion. Because little attention has been directed to methanotrophs in the oxic water column, the knowledge about this type of passway in methane consumption is still very poor. Our measurement of methane oxidation in the oxic water column during the lake circulation period in a deep dimictic lake is one of the scarce reports (Utsumi et al. 1998).

In the case of large, shallow lakes, anaerobic bottom water is rarely observed because of wind-driven water mixing throughout the year. Although this type of lake is common, methane cycling in such environments has not been studied. This paper presents the results of surveys of methane oxidation activity in the water column of Lake Kasumigaura, a eutrophic, shallow lake in Japan. Along with long-term observation of methane concentrations in the lake (Nakamura 1997), we investigated various processes related to methane production, transport, and consumption. Measured methane oxidation rates were used to identify the factors controlling methane turnover in the water column. Finally, we evaluated the relative importance of a diffusive flux across the air–water interface vs. methane oxidation in the lake.

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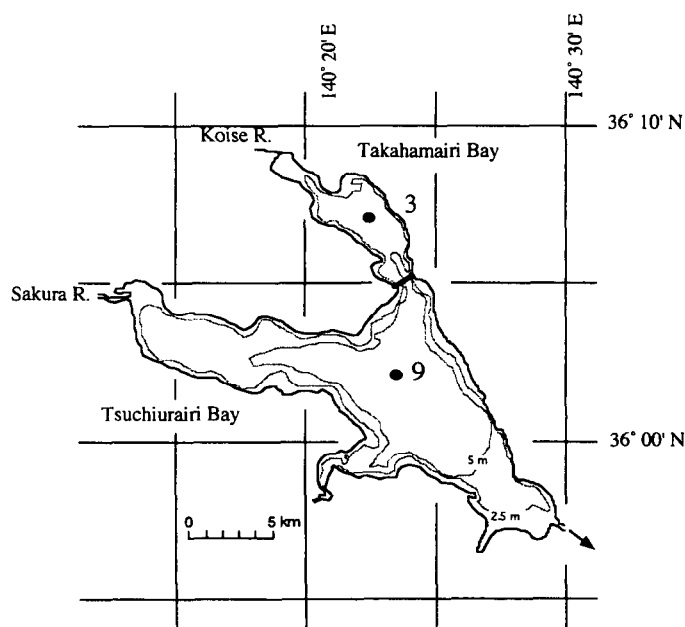


Fig. 1. Bathymetric chart of Lake Kasumigaura with station positions noted. Contour intervals are 2.5 and 5 m.

Materials and methods

Study site—Lake Kasumigaura (36°02'N, 140°24'E at the center of the lake) is the second largest lake (surface area, 168 km²) in Japan. It is a very shallow, with a maximum depth of only 7.3 m and a mean depth of 3.8 m (Fig. 1). The mean water renewal rate is 220 d (Otsuki et al. 1993). Differences in water temperature between the surface and bottom are minimal (<2°C, except for a few calm days in summer) because winds are usually sufficient to cause daily turnover. Consequently, the lake water is always well oxygenated.

The National Institute for Environmental Studies has conducted continuous monthly samplings of Lake Kasumigaura since 1977. Physical, chemical, and biological parameters including nutrients, primary production, and phytoplankton species composition have been measured (e.g. Takamura et al. 1992). The lake has been highly eutrophicated by the increasing population of the intake area. Recent levels of total nitrogen and total phosphorus in the lake water varied from 72 to 109 μM and 2.1 to 4.1 μM, respectively (Fig. 2).

Water samples for dissolved methane analysis were collected with a Go-Flo sampler (General Oceanics). Variations in dissolved methane concentrations within the lake have been reported elsewhere (Nakamura 1997). Additional sampling for the survey of methane was conducted, particularly during summer and autumn.

Methane oxidation rate measurements—Methane oxidation rates were measured at two stations, Sta. 3 in Takahamairi Bay and Sta. 9 in the center of the lake (Fig. 1). The depths of Sta. 3 and 9 are 3.7 and 5.8 m, respectively. Water samples for methane oxidation measurements were taken at

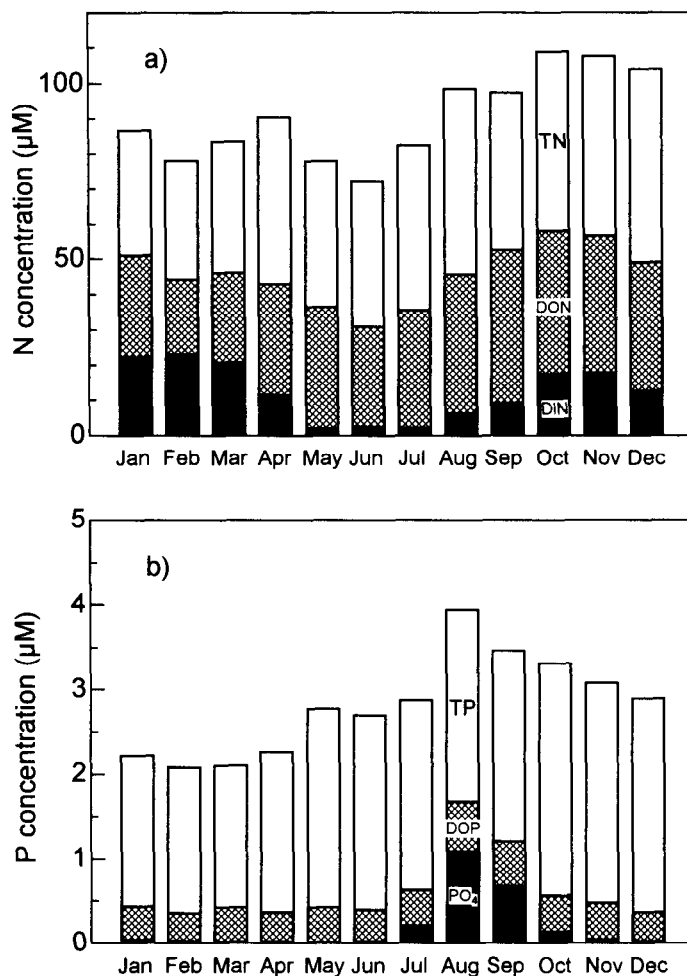


Fig. 2. Monthly average nitrogen (a) and phosphorus (b) concentrations (April 1990–March 1996 at Sta. 9, lake center) in Lake Kasumigaura.

~0.5-m depth with 10-liter Go-Flo samplers. Preliminary measurements of oxidation rate began in August 1990. From July 1991 to March 1996, measurements were taken at least monthly for both stations. For each station, 10–16 50-ml glass serum bottles were used for measuring methane oxidation rate. These glass bottles had been soaked in 3 N HNO₃ to remove contamination of trace metals (which are possible inhibitors for methanotroph), thoroughly washed with distilled water, and then sterilized by heat (170°C, 1 h). The water samples were immediately transferred to the incubation bottles onboard. Bottles were slowly overflowed with several volumes of water and sealed (without headspace). Each glass bottle was stoppered with an isoprene rubber septum and sealed with an aluminum crimp seal. After sealing, incubations were started on board at in situ water temperature in the dark. Because neither dilution nor addition of any nutrients was done, there was no reason to expect any time lag in the activity of the enclosed bacterial populations. The rates observed at the beginning of the incubation period could therefore be expected to mimic those occurring naturally.

The dissolved methane concentration was measured with

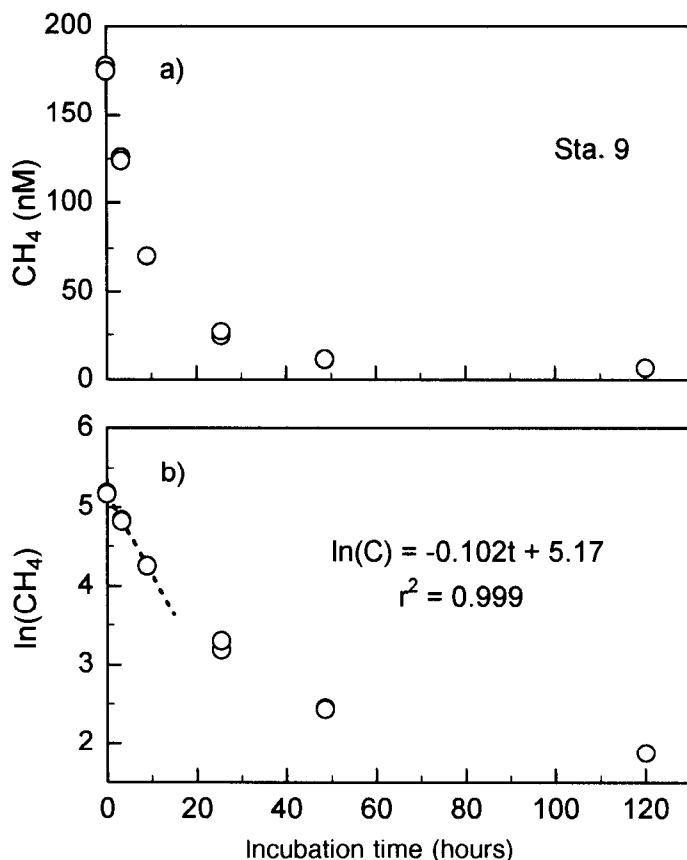


Fig. 3. Decrease in methane concentration during an incubation of a Lake Kasumigaura water sample (4 October 1995)—(a) linear ordinate; (b) logarithmic ordinate. The regression for the initial three time points can be expressed as $\ln(C) = -0.102t + 5.17$ ($r^2 = 0.9997$), where C is the concentration of dissolved methane (nM) and t is the time (h) elapsed after starting the incubation.

an automatic system consisting of a purge-and-trap apparatus and a gas chromatograph with a flame ionization detector (GC-FID) as previously described (Nakamura et al. 1994; Ishibashi et al. 1997). Analytical precision, expressed as the coefficient of one variation for repeated measurements of lakewater samples having 100 nM of methane concentration, was 0.6%.

Methane concentration was determined at three or four different time points, typically ranging from 2 to 24 h after the start of incubation. At each point, two bottles were sacrificed and methane oxidation was stopped by addition of 0.5 ml of 3.5% HgCl₂ solution, which completely inhibited methane consumption in the bottle. Controls consisted of samples to which HgCl₂ was added immediately after bottling onboard. The specific rate of methane oxidation was calculated by linear regression of the natural log of methane concentration against time; the specific rate of methane oxidation is equivalent to the slope in such a regression line (Fig. 3). Only samples in the period when the methane decrease was linear were used to determine methane oxidation rates. The specific oxidation rate is the first-order rate constant for methane oxidation (in units of h⁻¹). This rate is useful for comparing relative methane oxidation activities

Table 1. Standard error of slopes for specific methane oxidation rate measurements. Values are mean and standard deviation based on the sample number.

Turnover time (h)	Sta. 3		Sta. 9	
	<i>n</i>	error (%)	<i>n</i>	error (%)
<12	6	2.2±1.0	6	2.1±1.1
12–72	8	4.6±1.4	7	6.5±2.2
>72	4	10.2±6.1	5	10.4±1.7

The data period is from September 1994 to March 1996. Duplicate samples were sacrificed at each time point of the oxidation measurement. Before September 1994, only one bottle was sacrificed at each time point.

among samples with large differences in methane concentration because it is independent of methane concentration. The turnover time for methane is the inverse of the specific oxidation rate. When the turnover time is >1 week, an incubation period >24 h was necessary for the oxidation measurement. The precision of oxidation rate measurement, corresponding to the standard error of the slope, depends on the turnover time of dissolved methane (Table 1). Actual rates of methane oxidation (methane consumption rate; in units of nM CH₄ h⁻¹) can be calculated by multiplying the specific oxidation rate at any station by the measured ambient methane concentration.

Results

In this study, we used a method for measuring methane oxidation rate different from radiotracer methods (e.g. Rudd et al. 1974; Harrits and Hanson 1980; Griffiths et al. 1982; Ward et al. 1987; de Angelis and Scranton 1993; Scranton et al. 1993). Our method is an improvement on that of Janasch (1975) or Sansone and Martens (1978) in that we measured methane oxidation rate by monitoring decreases in methane concentration with time for natural lakewater samples incubated in glass bottles under near in situ conditions. Because of the high reproducibility of the GC-FID measurements with the purge-and-trap system, we were able to use short incubation periods with little consumption of dissolved methane, typically <20–30% of that initially present. These short incubation periods (usually <10 h) minimized both bottle effects and the change in methane and oxygen concentrations in the bottles during incubations. By using this method, it was thus possible to measure the rate close to the actual in situ methane oxidation activity in the water column without any extra operations or time lag.

The time course of dissolved methane concentration changes in the glass bottles (Fig. 3) can be described as exponential decay. The relationship between the logarithm of dissolved methane concentration and elapsed time was expressed with high linearity. No time lag in methane oxidation activity of the bacterial populations (methanotrophs) was observed in any bottle samples at the start of incubation, so the calculated specific rate would be expected to mimic those occurring naturally in the lake water. Furthermore, dissolved methane concentration was never observed to increase in the bottles during any of our incubations. Thus, in

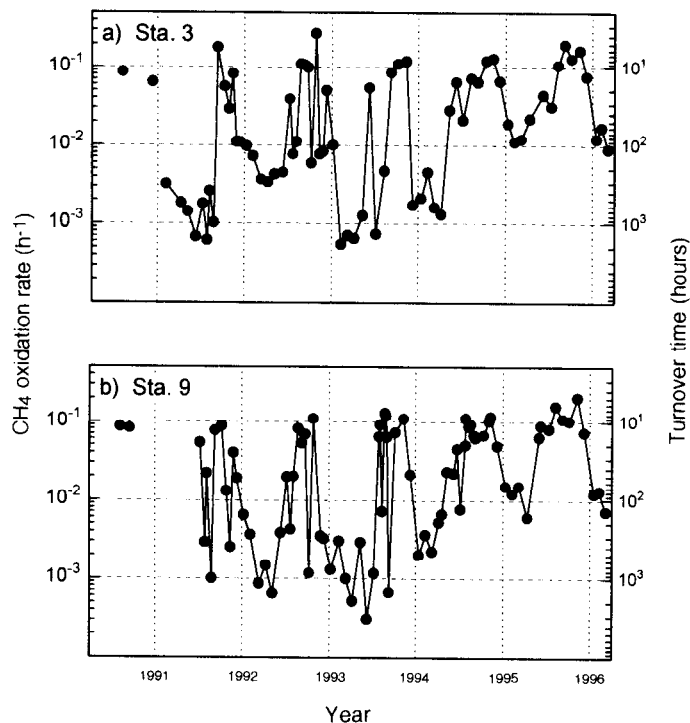


Fig. 4. Changes in specific methane oxidation rate and turnover time with respect to in situ oxidation for dissolved methane in Lake Kasumigaura—(a) Sta. 3; (b) Sta. 9.

the water column of Lake Kasumigaura, methane production might not occur because production of methane is strictly an anaerobic process (e.g. Rudd and Taylor 1980; Daniels et al. 1984).

The measured specific oxidation rates in the lake ranged from 0.0006 (10 February 1993; turnover time of 75 d) to 0.272 h^{-1} (27 October 1992; turnover time of 3.7 h) at Sta. 3 (Fig. 4a), and from 0.0003 (8 June 1993; turnover time of 134 d) to 0.204 h^{-1} (8 November 1995; turnover time of 4.9 h) at Sta. 9 (Fig. 4b). The dissolved methane depth profile was generally uniform (Nakamura 1997) because of the shallowness of the lake. Profiles of methane oxidation activity were correspondingly uniform, as confirmed by incubation of samples from several depths on several occasions.

Water-column methane oxidation activity had not previously been reported in shallow and nonstratified lakes like Lake Kasumigaura. Here, we evaluated the seasonal change in methane oxidation activity in oxic lake water for 5 years, confirming the existence of methane oxidation activity in oxic water columns. Note that this activity varied by as much as 3 orders of magnitude. Differences in activity between sampling times less than a month apart were sometimes as large as 2 orders of magnitude.

Methane oxidation activity was distinctly seasonal, with low activity from January to April and high activity from August to November at both stations (Fig. 5a, b). The period from May to July was characterized by a shift in water-column methane oxidation activity from low to high, while in December it shifted from high to low. Interannual variability (displayed as bars in Fig. 5) was very high in summer and autumn. Methane consumption rates in the water column

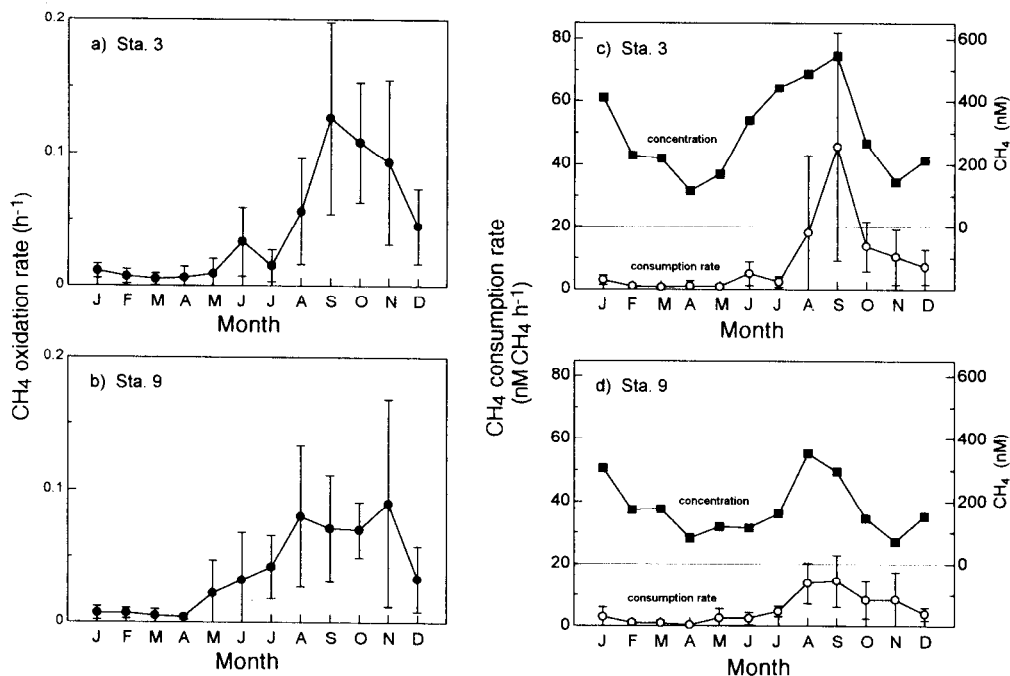


Fig. 5. Monthly average specific methane oxidation rates (a, Sta. 3; b, Sta. 9), methane consumption rates, and methane concentrations (c, Sta. 3; d, Sta. 9) in Lake Kasumigaura. Results represent the monthly averages for the observed years, 1990–1996, with bars indicating standard deviation (representing interannual variability).

Table 2. Methane oxidation rate in the water column, reported in various freshwater environments.

Location	CH ₄ oxidation rate		Period	Incubation	Reference
	nM h ⁻¹	h ⁻¹			
Lake 227 (Experimental Lakes Area, Canada)	0–390 40–720	— —	Stratification period Turnover period	Addition of [¹⁴ C]CH ₄	Rudd and Hamilton 1975
Lake Kivu (African rift lake)*	1.8–37	—	Unknown	Addition of CH ₄	Jannasch 1975
Lake Mendota (Wiscon- sin)	0–230 10–1,200	— —	Stratification period Turnover period	Addition of [¹⁴ C]CH ₄	Harrits and Hanson 1980
Lake Washington (Wash- ington)	~0	0	Monthly observation throughout year	Addition of [¹⁴ C]CH ₄	Lidstrom and Somers 1984
Lake Spirit (Oregon)	0.04–6	—	June–October	Addition of [¹⁴ C]CH ₄	Lilley et al. 1988
Hudson River (New York)	0.004–0.6	0–0.005 0.0004–0.03	March	Addition of [¹⁴ C]CH ₄	de Angelis and Scranton 1993
Lake Constance (Switzer- land)*	0.07–7 —	0.019	August February–November	Addition of CH ₄	Schmidt and Conrad 1993
Lake Nojiri (Japan)	0.3–710 1–300	0.004–0.012 0.006–0.042	Stratification period Turnover period	Without CH ₄ addition	Utsumi et al. 1998

* Data for Lake Kivi and Lake Constance are methane oxidation potentials (owing to appreciable amounts of methane addition for the incubation).

Table 3. Monthly average water temperature, specific methane oxidation rate, and methane consumption rate in Lake Kasumigaura (values are means ± 1 SD of interannual variation).

	Water temp. (°C)	Oxidation rate (h)	Consumption rate (nM CH ₄ h ⁻¹)	n
Sta. 3				
Jan	5.8±0.7	0.011±0.005	2.9±1.5	5
Feb	4.8±1.2	0.007±0.005	1.0±0.5	6
Mar	8.0±0.8	0.005±0.004	0.7±0.6	5
Apr	13.6±1.0	0.006±0.008	1.0±1.7	5
May	18.3±1.1	0.009±0.011	0.8±0.9	4
Jun	22.7±1.9	0.033±0.026	5.0±3.8	5
Jul	25.5±2.3	0.015±0.012	2.4±1.7	5
Aug	27.6±3.1	0.056±0.040	18.1±24.9	5
Sep	25.6±1.6	0.126±0.072	45.6±36.5	5
Oct	19.9±1.5	0.108±0.045	13.7±8.0	5
Nov	14.1±1.1	0.093±0.062	10.4±8.8	5
Dec	9.7±1.0	0.045±0.028	7.2±5.5	6
Sta. 9				
Jan	5.6±0.6	0.007±0.005	2.9±3.1	5
Feb	4.6±1.2	0.007±0.004	1.1±0.7	5
Mar	7.5±0.6	0.005±0.005	0.9±1.0	5
Apr	12.5±0.8	0.004±0.002	0.3±0.2	4
May	17.2±0.8	0.022±0.025	2.5±3.0	4
Jun	21.9±1.8	0.032±0.036	2.4±2.0	4
Jul	24.5±1.9	0.042±0.024	4.7±1.7	5
Aug	27.1±3.0	0.080±0.053	13.8±6.5	6
Sep	25.5±1.4	0.071±0.040	14.5±8.3	6
Oct	20.0±1.5	0.070±0.021	8.4±6.0	5
Nov	14.1±1.0	0.090±0.078	8.5±8.9	5
Dec	9.9±0.9	0.033±0.025	3.9±2.1	5

also fluctuated (Fig. 5c,d). Minimum and maximum average monthly methane consumption rates for the 5 years of observation were 0.7 ± 0.6 (March) and 45.6 ± 36.5 nM CH₄ h⁻¹ (September) at Sta. 3, and 0.3 ± 0.2 (April) and 14.5 ± 8.3 nM CH₄ h⁻¹ (September) at Sta. 9. The minimal consumption rates at Sta. 3 and 9 were almost the same; however, the maximal consumption rate at Sta. 3 was about threefold that at Sta. 9.

Discussion

Methane oxidation activity in the oxic water column of Lake Kasumigaura—The reported water-column methane oxidation rates are summarized in Table 2. Rudd and Hamilton (1978) reported annual changes in methane oxidation rates for dimictic Lake 227, in which an anaerobic hypolimnion was present during summer stratification. Methane oxidation activity strongly stratified and peaked at the oxic–anoxic interface during summer, and high rates of methane oxidation occurred throughout the water column during autumnal overturn. Lake Mendota and Lake Nojiri, having seasonally anoxic hypolimnion, are similar examples of high methane oxidation rates throughout the water column during autumnal overturn period (Harrits and Hanson 1980; Utsumi et al. 1998). However, seasonal changes in methane oxidation in monomictic Lake Washington, in which an aerobic hypolimnion is maintained throughout the year, were studied by Lidstrom and Somers (1984). They concluded that methane oxidation activity was negligible throughout the entire water column. From summer to autumn, we found high methane oxidation rates in Lake Kasumigaura water, even though the oxic–anoxic interface did not exist in the water column (Table 3). The measured oxidation rates in Lake Kasumigaura were similar to the oxidation rates of above-mentioned dimictic lakes during the fall overturn period; fur-

thermore, the methane oxidation activity in the lake was highly variable and seasonal.

The annual changes in dissolved methane concentrations at several stations in Lake Kasumigaura measured for 6 years by Nakamura (1997) followed a clear seasonal pattern with low concentrations from April to June, high concentrations from August to September, a minimum in November or December, and relatively high concentrations from January to March. The annual average dissolved methane concentration at the station in Takahamairi Bay (Sta. 3; 298 nM) was higher than that at the lake center station (Sta. 9; 179 nM), and the largest concentration differences between the two stations occurred in summer and autumn (Nakamura 1997). The large differences in methane consumption rates for summer and autumn are partially due to the large differences in dissolved methane concentrations and partially to the differences in specific methane oxidation rates (see Fig. 5c,d). The consumption rates from January to April were identical for both stations (Table 3).

Factors controlling methane oxidation activity in the water column—Various factors seem to control the activity of methanotrophs in different environments. As one would suspect in a biologically mediated process, temperature significantly affects biological reactions. Comparison, however, of pairs of the average specific oxidation rates at equivalent water temperature, e.g. April and November, or May and October, indicated significantly higher oxidation rates in autumn than those in spring at both stations (see Table 3). The effect of temperature on the specific methane oxidation rate could not be expressed as a significant regression in the Arrhenius plot (Fig. 6a). The seemingly minor role of temperature as a regulatory factor for methane oxidation activity differs substantially from the usually critical role it plays for other microbial processes.

Methane oxidation activity in the water column is dependent, in part, on the existence of two primary substrates, oxygen and dissolved methane. In Lake Kasumigaura, dissolved oxygen concentration was near saturation throughout the water column (6–12 mg O₂ liter⁻¹) owing to the shallowness of the lake and the prevailing wind-driven mixing. The dissolved oxygen concentration was significantly above the range of the estimated half-saturation constant for methane oxidation, K_m (0.5–0.8 mg O₂ liter⁻¹; Lidstrom and Somers 1984), or the reported optimum range of 0.1–1.0 mg O₂ liter⁻¹ (Rudd and Hamilton 1975) for microbial methane oxidation in the water column. In our observations, on the other hand, high methane oxidation activity was observed in the water column when dissolved oxygen concentrations were high from late summer to autumn. The dissolved oxygen concentration, then, should not be the major factor limiting methane oxidation activity. The seasonal change of specific methane oxidation rate (h⁻¹) had a different pattern from its dissolved methane concentration (Fig. 5). There was no statistically significant relationship between dissolved methane concentration and specific oxidation rate for data from all seasons (Fig. 6b).

Methane oxidation activity depends in part on dissolved inorganic nitrogen (DIN). Rudd and Hamilton (1978, 1979) concluded that the combination of requirements for fixed

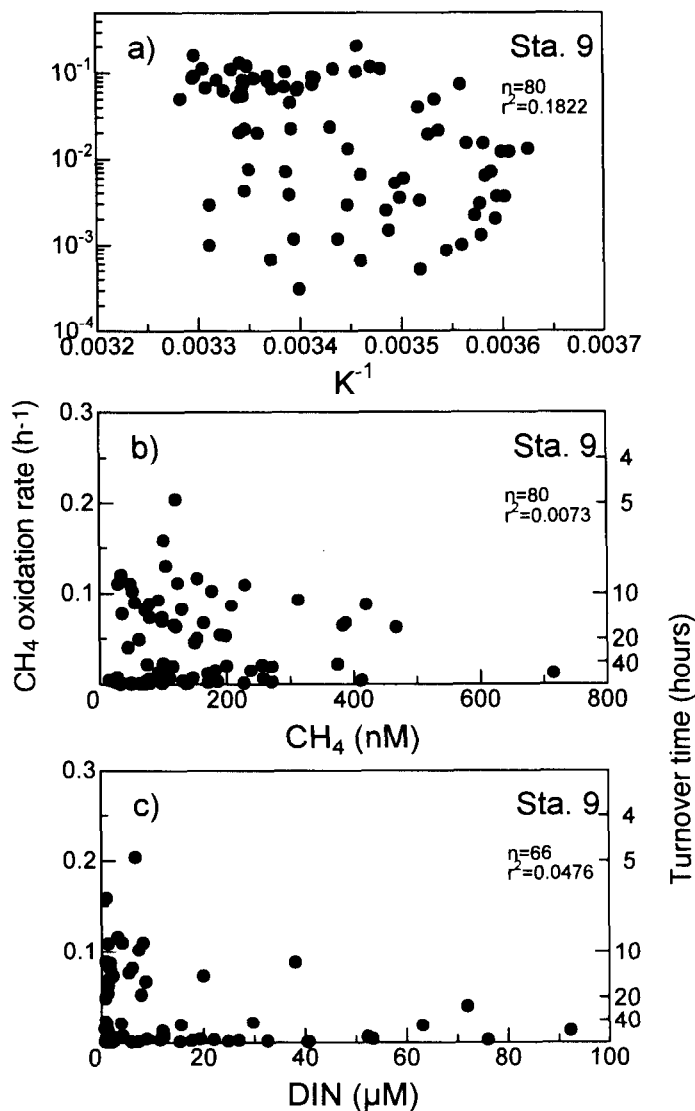


Fig. 6. Relationship between specific methane oxidation rate (h⁻¹) and lake water parameters—(a) water temperature (Arrhenius plot); (b) dissolved methane; (c) dissolved inorganic nitrogen.

nitrogen and oxygen constrained methane oxidation in Lake 227. According to their observations, methanotrophs fix nitrogen under low DIN conditions (<3 μM). Because nitrogen fixation is disrupted by high concentrations of dissolved oxygen, methane oxidation is confined to within a narrow zone at the oxycline. Methane oxidation activity in Lake 227 was thus limited during the period of lake stratification, and was widespread only during autumnal overturn under nitrogen-limited conditions.

In the nonstratified, shallow Lake Kasumigaura, DIN concentrations are sometimes <3 μM between mid-spring and autumn. Such low nitrogen concentrations there are mainly due to phytoplankton uptake, which is dominated by *Oscillatoria* (Takamura et al. 1992; Otsuki et al. 1993). DIN usually increases in late autumn after the end of the summer phytoplankton bloom (Fig. 2a). Even under low DIN and high dissolved oxygen conditions from summer to early au-

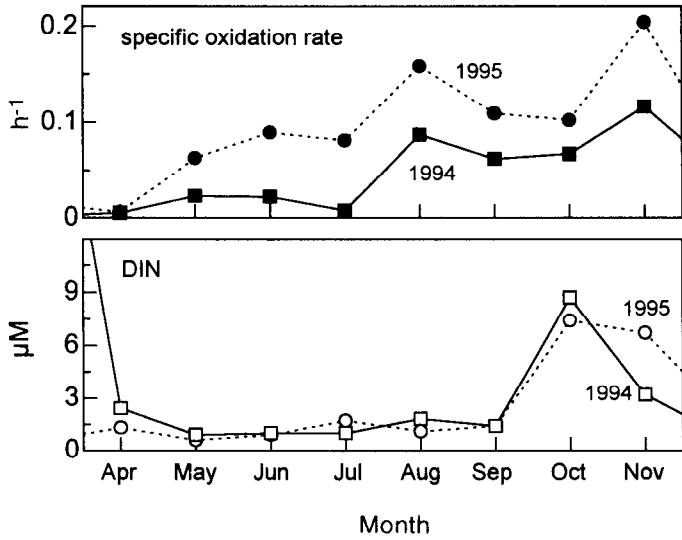


Fig. 7. Changes in specific methane oxidation rate and dissolved inorganic nitrogen (DIN) concentration at Sta. 9 in 1994 and 1995. ■, oxidation rate in 1994, □, DIN in 1994, ●, oxidation rate in 1995, ○, DIN in 1995.

tumn, methane oxidation activity increased in the water column (Fig. 7). There was no statistically significant relationship between DIN and specific oxidation rate for data from all seasons (Fig. 6c). Therefore, the DIN concentration did not affect the methane oxidation activity in the water column of Lake Kasumigaura, similar to the situation in Lake Mendota sediments (Harrits and Hanson 1980).

We failed to identify the environmental factors controlling the methane oxidation rate in Lake Kasumigaura. Monthly sampling might be not frequent enough for a lake with high methane oxidation rates and rapid variability (e.g. in varying daily wind and water temperature).

Water-column methane consumption and methane supply from sediments to water—Microbial oxidation is the only consumption process occurring within the water column that can prevent the eventual loss of methane to the atmosphere. Whole-lake rates of methane consumption were calculated by dividing the lake into boxes, multiplying the estimated methane consumption in each box by box volume, and integrating the consumption of all the boxes (Fig. 8; Nakamura 1997). The corresponding areal values were calculated by dividing the box-integrated totals by the surface area of the lake (168 km²). The minimum and maximum monthly average methane consumption rates were observed on April and August, respectively (Table 4).

The other important mechanism for methane loss from the water column is diffusive flux to the atmosphere across the air-water interface. The calculated diffusive fluxes of methane across the air-water interface correlated primarily with the seasonal change in dissolved methane concentrations (Nakamura 1997). The minimum and maximum monthly average fluxes were observed in November and September, respectively (Table 4). From February to May, when methane oxidation activity in the water column was low, methane consumption in the lake was lower than its diffusive flux to

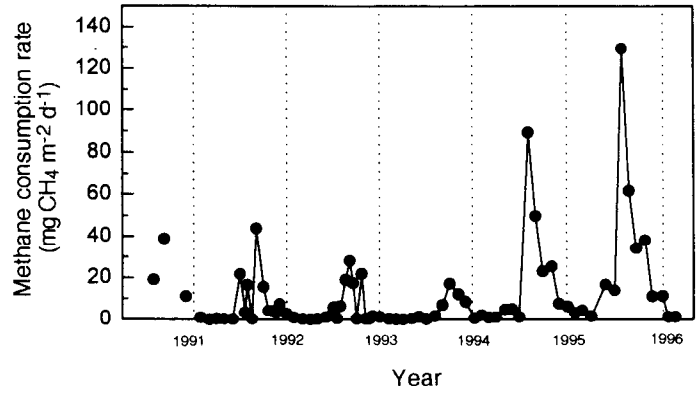


Fig. 8. Methane consumption rate in Lake Kasumigaura over 5 years.

the atmosphere, but in other seasons consumption rates were higher than the diffusive flux. Especially from late summer to autumn, when the highest methane oxidation activity was observed, microbial oxidation of methane was an important methane consumption process in the water column, destroying methane and thus reducing the transport of methane from the lake to the atmosphere. Hence, oxidation of methane by methanotrophs rather than diffusive fluxes across the air-water interface was the dominant methane removal process for Lake Kasumigaura. Microbial oxidation is the dominant sink in the lake, consuming an annual average of 74% of the dissolved methane in the water column.

The turnover time of dissolved methane in an average water column for the entire lake (Table 4) was calculated by dividing the methane concentration by the total methane sink (consumption rate plus diffusive flux) in the water column. The shortest and longest turnover times of dissolved methane were 0.3 (in November) and 3.7 d (in February), and the annual average turnover time was 1.6 d. The turnover

Table 4. Comparison of methane consumption rates and diffusive fluxes to the atmosphere as sinks for water-column methane and calculated turnover time of dissolved methane in Lake Kasumigaura (values are means ± 1 SD of interannual variation).

	Methane consumption rate		Diffusive flux		Turnover time (d)
	mg CH ₄ m ⁻² d ⁻¹	n	mg CH ₄ m ⁻² d ⁻¹	n	
Jan	4.4±4.0	5	2.6±1.5	6	3.0
Feb	1.4±0.9	6	2.1±1.8	6	3.7
Mar	1.2±1.5	6	2.7±1.1	6	2.9
Apr	0.6±0.6	5	1.9±0.8	6	2.2
May	1.4±1.9	4	3.1±0.8	6	1.9
Jun	4.9±6.2	5	3.7±1.6	6	1.0
Jul	6.2±5.9	5	4.5±6.4	6	1.2
Aug	48.2±51.5	5	12.0±7.1	6	0.4
Sep	37.0±19.6	5	10.8±4.8	6	0.4
Oct	19.0±8.9	5	4.5±1.6	6	0.5
Nov	15.9±14.1	5	1.1±0.6	6	0.3
Dec	7.2±3.1	5	1.9±1.4	6	1.3
Annual avg	12.3±15.5		4.2±3.5		1.6

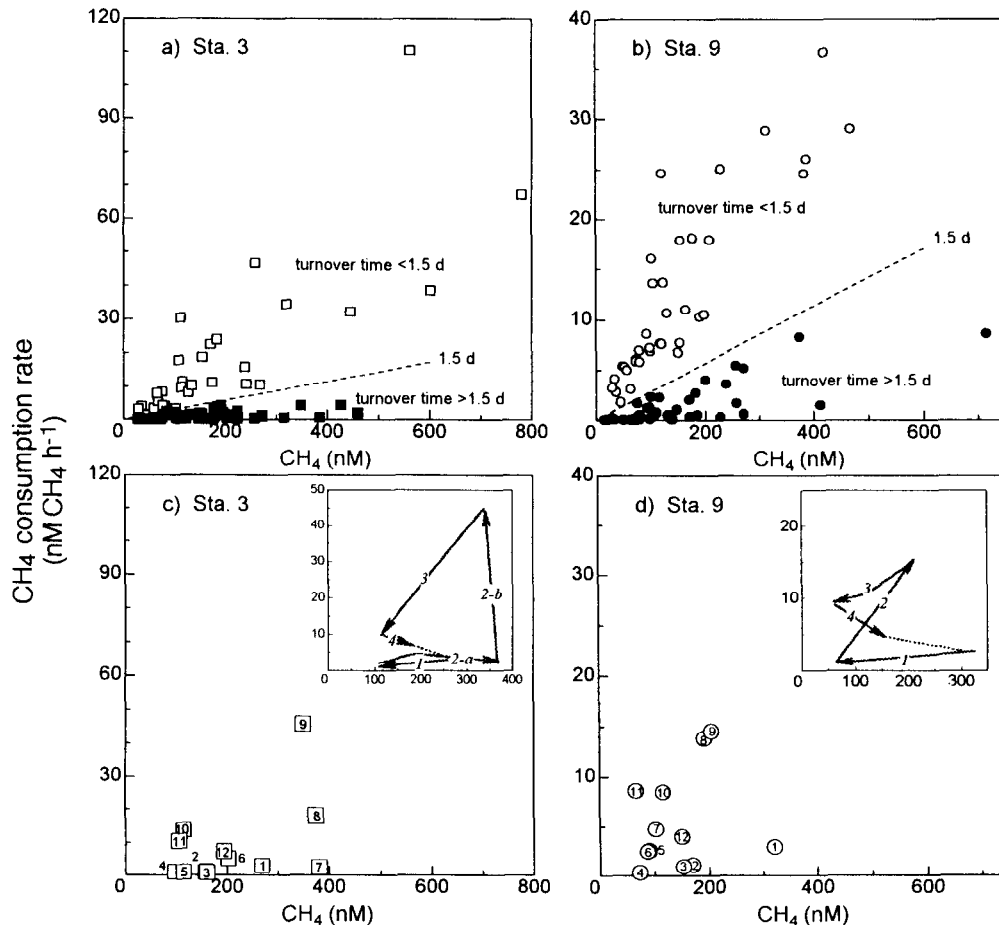


Fig. 9. Relationship between the concentration of dissolved methane and methane consumption rate at Sta. 3 (a) and Sta. 9 (b) (open symbols, turnover time <1.5 d; filled symbols, turnover time >1.5 d), and monthly averages at Sta. 3 (c) and Sta. 9 (d) in Lake Kasumigaura. Numbers in symbols indicate month. The typical trends with time are shown in the insertion (1, January–April; 2, April–September [2a, April–July and 2b, July–September in Sta. 3]; 3, September–November; 4, November–December).

time of dissolved methane is very short relative to the renewal time of the lake water (220 d; Otsuki et al. 1993). Hence, the concentration of dissolved methane in the water column was not controlled by the water budget of the lake.

When comparing all measured consumption rates with methane concentrations (Fig. 9a,b), the data for periods with shorter turnover times (<1.5 d) can be clearly distinguished from those with longer turnover times (>1.5 d) for both stations. The specific oxidation rate, which is the quotient of consumption rate by the concentration, has bizonal distribution. This is also noticeable in Fig. 6b, where scarce data points are found around the oxidation rate of 0.03 h^{-1} .

The monthly average plots for the same datasets reveal a systematic seasonality, which can explain the seasonal change in dissolved methane in Lake Kasumigaura (Fig. 9c,d). During the period from January to April, when methane consumption rates were usually lower than the diffusive flux, methane concentrations decreased substantially, with only a small decline in consumption rates, indicating decreasing methane supply from the bottom sediment. Diffusive flux was the dominant methane sink during this season.

Thus, the spring concentration minimum appears throughout the entire basin (Nakamura 1997).

From May to September, the trends in methane concentration and consumption rates at the two stations differed. At Sta. 3 from May to July, methane concentration increased markedly with very little concomitant rise in consumption rates, essentially reversing the January–April shift. This trend was due to an increase in methane supply from the bottom sediment while methane oxidation activity remained low, resulting in high summer methane concentrations occurring earlier in the season at Sta. 3 than at other stations (Fig. 5c,d). In contrast, the summer increase in methane concentrations at Sta. 9 occurred in August. The August point for Sta. 3 (see Fig. 9c) is based on the average of data for occasions with shorter and longer turnover times, both of which occurred in August during the 5 years of measurement. In September, high methane concentrations remained despite the higher methane oxidation activity that usually prevailed at that time, indicating the largest methane supply from the bottom sediment in the year. The transition from the spring minimum concentration (April) to the summer

maximum (August and September) at Sta. 9 (see Fig. 9d) proceeded more smoothly than that at Sta. 3 (see Fig. 9c), as the methane consumption rate increased concomitantly with methane concentration. The June or July concentration increase observed at Sta. 3 was rarely observed at Sta. 9 owing to the more rapid increase in methane oxidation activity at the lake center (Nakamura 1997).

From September to November, methane concentrations decrease until reaching the November minimum at both stations. High methane oxidation activity remains during this period, so we attribute the concentration minimum to gradually decreasing methane supply from the sediment. We attribute the methane concentration increases in the December–January period to rapidly decreasing methane oxidation activity and slowly decreasing methane supply from the sediment. These conditions lead to high methane concentrations in Lake Kasumigaura in January.

Conclusions

We have confirmed methane oxidation activity in the oxic water column of eutrophic, shallow Lake Kasumigaura. The level of activity fluctuated greatly from year to year. Because the average turnover time of dissolved methane (1.6 d) is much shorter than that of the lake water (220 d), the concentration of dissolved methane was not controlled by the water budget, but rather by the methane supply from the sediment, oxidation in the water column, and diffusion to the atmosphere. The activity was distinctly seasonal, with low specific rates from January to April and high specific rates from August to November, suggesting that different types of methanotrophs dominate during the different seasons.

A clear relationship between methane oxidation activity in the water column and routinely measured environmental factors was not observed. In freshwater lakes, methane oxidation is most active at interfaces between oxic and anoxic zones, where all necessary substrates for methane oxidation are available in sufficient quantities. An oxic–anoxic interface exists in the surface sediment in Lake Kasumigaura, so methane oxidation activity in this lake should be most active there. With the daily mixing of water, methanotrophs in the sediment are transported to the oxic water column and retain their activities in the planktonic state in the nonstratified shallow lake.

This phenomenon has similarity to that observed during the overturn period at Lake Nojiri, a stratified lake (Utsumi et al. 1998). Before the autumnal overturn period, active methane oxidation was observed within a limited depth zone in the hypolimnion at the oxic–anoxic interface. As lake overturn proceeded in December, methane stored in the anoxic hypolimnion during summer stratification was oxidized by methanotrophs throughout the water column as a result of oxygenated water mixing with anoxic water that contained high concentrations of methane. Methane oxidation activity in the surface layer was significantly increased by the water mixing, even in cold winter.

In Lake Kasumigaura, an increase in methane oxidation is observed, accompanied by maximum methane production

in late summer. The relationship between concentration and consumption rate suggests a certain time lag in the decline of oxidation activity with decreasing methane production in early autumn. The high oxidation activity continues through the end of autumn, when production of methane should have already decreased. The minimum water concentration is thus observed at late autumn. Methane oxidation is the dominant methane sink in Lake Kasumigaura, consuming an annual average of 74% of dissolved methane in the water column.

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