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requests for materials
should be addressed to
S.B.X. (shangbinx@
163.com)

Methane formation and consumption processes in Xiangxi Bay of the Three Gorges Reservoir

Chenghao Wang¹, Shangbin Xiao^{1,2}, Yingchen Li¹, Huayao Zhong¹, Xuechen Li¹ & Feng Peng¹¹College of Hydraulic and Environment, China Three Gorges University, Yichang, 443002, China, ²Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, 710075, China.

Indoor simulation experiment was carried out to evaluate the formation and consumption rates of methane (CH₄) in Xiangxi Bay of the Three Gorges Reservoir (TGR), China. The results show that both the CH₄ formation and consumption rates were significantly positively correlated with temperature. CH₄ efflux decreased with rising temperature due to its potential increasing oxidation rate. CH₄ oxidation in surface sediments accounted for 51.8% of the total production and it even reached to 77.4% at 35 °C. The methane oxidation rate in water column ranged from 1.26 to 4.65 mg/(m²h), of which the average and greatest rate accounted for 46.7% and 73.9% of CH₄ production respectively under the condition of 30 m water column and 35 °C. The methane oxidation may increase by 41.04 mg/(m²h) under average water level of TGR (160 m), and most methane resulted from sediments can be oxidized in the water column.

CH₄ is an important atmospheric contaminant contributing to the greenhouse effect¹. Owing mainly to anthropogenic production, atmospheric methane concentrations have doubled from 850 ppbv to approximately 1750 ppbv over the last 150 years.

Hydropower resulted from reservoirs has been thought as a kind of clean and renewable energy. However, the conversion of land surface areas saturated by oxygen to anoxic sediments overlain by water results in CH₄ emissions from reservoirs under certain conditions². Hydroelectric reservoirs do not have a negligible carbon footprint, in other words, they are not carbon-free³. CH₄ emissions from hydroelectric reservoirs might have an influence on the anthropogenic CH₄^{4–11}. Moreover, some reservoirs emit more carbon than fossil-fuel based electricity¹². Other greenhouse gas (GHG) emissions like CO₂ and N₂O are also found in many lakes and reservoirs¹³, although seasonal mean N₂O fluxes were generally low⁶.

At the very beginning, CH₄ is exclusively formed in anaerobic environments^{10,14,15}. This gas is then partially mineralized into CO₂ through aerobic oxidation by methanotrophic bacteria in surface sediments or water column. Only the unoxidized parts escape to the atmosphere as CH₄ emissions^{10,16}. CH₄ emissions from aquatic environments depend on both CH₄ formation and CH₄ oxidation rates, and the latter is also known as consumption process¹⁷. According to previous reports, CH₄ emission from reservoirs not only linked to reservoir age and latitude³, but also related to both temperature and sediment characteristics (eg. C: N ratio). CH₄ formation, which results from a temperature-controlled mineralization process of organic matter¹⁸, is more temperature sensitive than CH₄ oxidation¹⁷. The rate of methanogenesis varies with sediment site and depth. It decreases with increasing sediment depth due to the number of methanogens in deeper sediment¹⁹. Moreover, substrate concentrations on temperature sensitivity of methanogenesis will also influence CH₄ formation, and it reduces with decreasing substrate concentrations^{17,20}. Further studies are also needed to explore the distribution of methanogens and methanotrophs under different environment conditions²¹.

The TGR, which is located in the subtropical zone and resulted from the biggest water-control projects in the world, is a huge and typical fluvial reservoir. However, the greenhouse gas fluxes from TGR were reported scarcely. Data reported varied greatly and resulted in a tremendous controversy^{22–24}. This paper aims to quantitatively evaluate the processes of CH₄ formation and consumption in Xiangxi Bay of TGR based on simulation experiment. The relationship between temperature and contributions of oxidation process in surface sediments and water column are reported here.

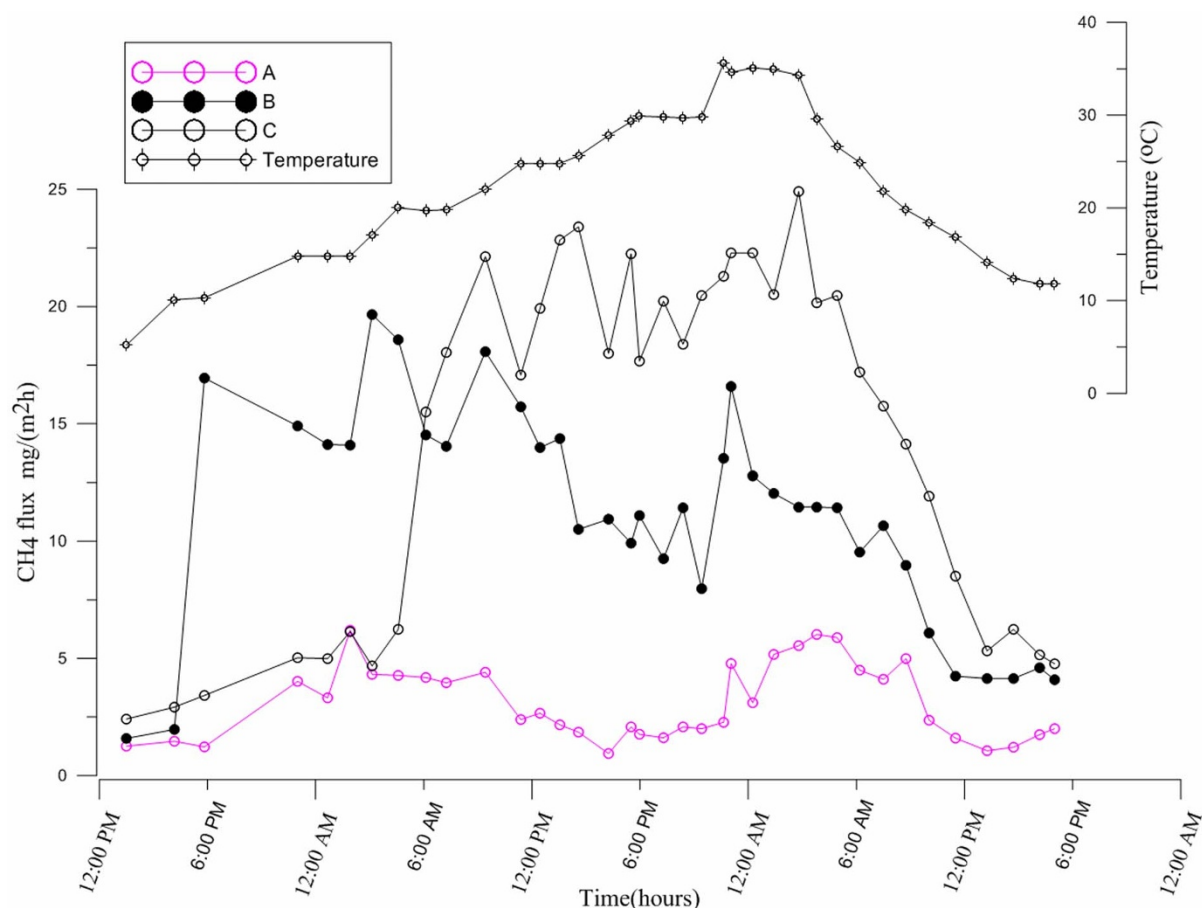


Figure 1 | CH₄ flux under different temperatures in Tube A, B and C. Tube A, B and C are experimental controls. CH₄ fluxes in Tube A, B and C simulate CH₄ formation with oxidation consumption of surface sediments and water column, CH₄ formation only with oxidation consumption of surface sediments, and CH₄ formation process without aerobic oxidation, respectively. CH₄ flux didn't increase continuously during the rapid warming process except Tube C from 20 to 30°C, while it decreased continuously during the rapid natural cooling processes in all tubes. When the temperature remained constant, CH₄ flux in each tube is fluctuating around a certain value.

Results

Based on the experimental design, CH₄ fluxes of Tube A, B and C respectively simulate CH₄ efflux with oxidation in surface sediments and water column, CH₄ efflux only with oxidation in surface sediments, and CH₄ efflux process without aerobic oxidation (Fig. 1 & Table 1).

A good correlation between CH₄ flux and temperature for each tube (Fig. 2, Supplementary Table S1 & Supplementary Table S2) is observed here. During the rapid natural cooling process, CH₄ flux decreased continuously in all tubes. However, CH₄ flux didn't increase continuously with the rapid warming except Tube C, and the flux in Tube A and B decreased with temperature increasing from 20 to 30°C. When temperature remained changeless, CH₄ flux in each tube fluctuated around a certain value.

In general, the CH₄ efflux is the biggest in Tube C, and is the smallest in Tube A. The latter may be due to CH₄ oxidation in surface sediments and its water column.

When temperature remained changeless, the CH₄ flux changed little (Fig. 2). There is a positive correlation between the CH₄ flux and

temperature according to Fig. 1 and Fig. 2 if we rule out the decreasing CH₄ with the rapid warming from 20 to 30°C.

Discussion

The Bunsen solubility coefficient for CH₄ is 0.03–0.05, which is expressed as ml of CH₄ (STP, Standard Temperature and Pressure) dissolved in 1 ml of H₂O²⁵. So the quantity of dissolved CH₄ can be ignored here. There is a positive correlation in some degree between the CH₄ flux and temperature (Supplementary Table S1). However, the correlation coefficients of Tube A and B (0.30 ~ 0.40) are lower than that of Tube C (>0.90), which may indicate that the CH₄ production in Tube C is significantly positively correlated with temperature. Better correlation between the CH₄ flux and temperature is observed when the average values of both are used (Supplementary Table S2).

CH₄ fluxes of Tube C are higher than B on the whole except at the beginning of rapid warming period. The difference between CH₄ fluxes of Tube C and B resulted from the aerobic CH₄ oxidation in surface sediments. Average CH₄ fluxes were used during rapid

Table 1 | Variations of CH₄ fluxes of three tubes

Tube	Maximum mg/(m ² h)	Minimum mg/(m ² h)	Average mg/(m ² h)	Standard deviation	C.V
A	6.20	0.94	3.02	1.59	0.53
B	19.66	1.59	10.70	4.86	0.46
C	26.83	2.41	13.61	7.77	0.57

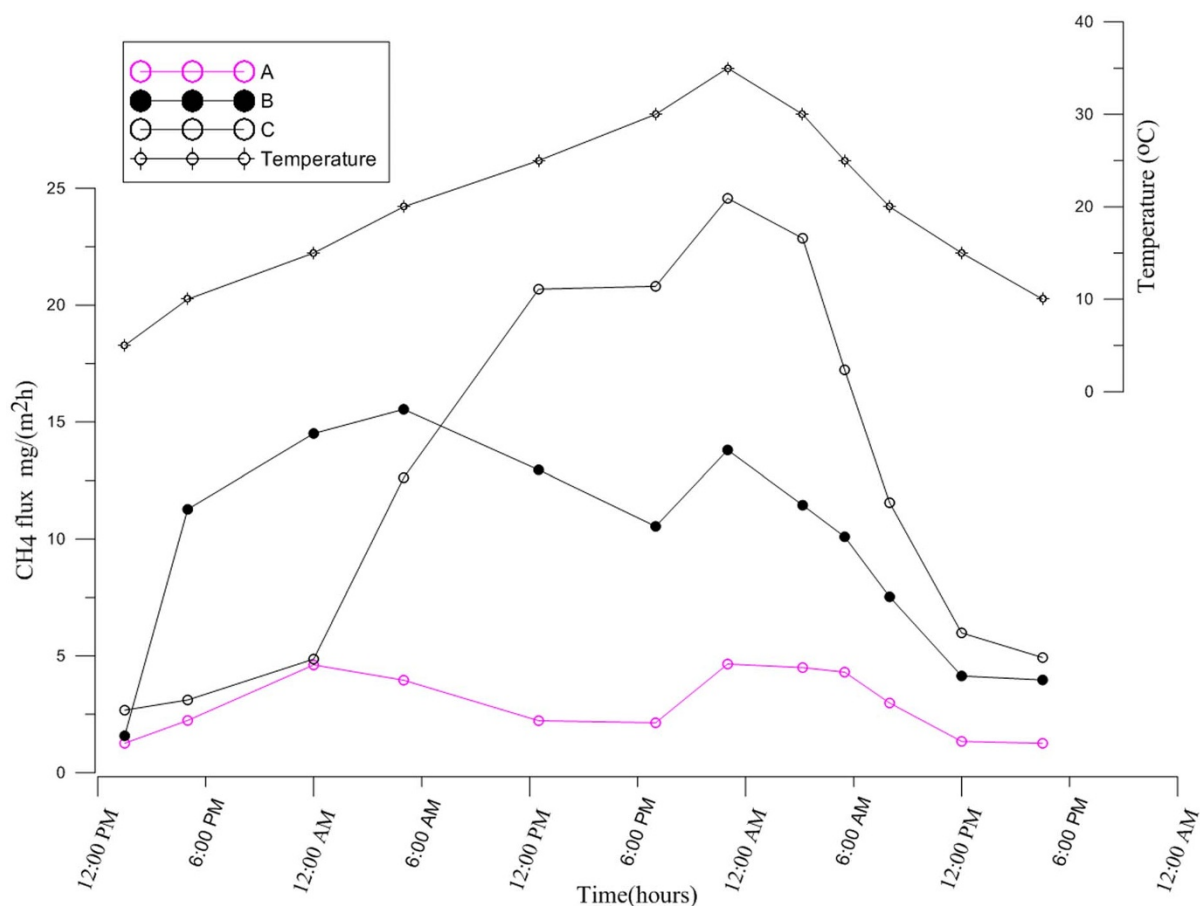


Figure 2 | Average CH₄ flux vs. each constant temperature. The wave property and some experimental error effects can be weakened when we use average value. Meanwhile, CH₄ consumption processes can be clear. CH₄ flux of Tube C is higher than B except a high fluxes process at the beginning. On the whole, CH₄ flux has a decline trend during this gaseous diffusion process when CH₄ emits from sediments to atmosphere via surface sediments and water.

warming process from 5 to 20°C to evaluate this process. The decreasing CH₄ flux, which resulted from its oxidation, is shown as Fig. 3. The CH₄ oxidation rate is less sensitive to temperature according to previous reports^{17,26}. However, our results do show a significantly positive correlation between the CH₄ oxidation rate and temperature, and the correlation coefficient is 0.94 (it can even be improved to 0.96 when average values are used, Fig. 3 & Supplementary Table S3).

The linear fitting between the CH₄ oxidation rate (initial value and average value) with temperature have different slopes (Supplementary Table S3). During each period with changeless temperature, CH₄ efflux varies little, so the average value is used here in order to better understand the potential process in CH₄ formation and oxidation.

CH₄ fluxes of Tube B are higher than that of Tube A on the whole except in the beginning. Therefore, its average value was also used during the rapid warming process from 5 to 20°C. The CH₄ flux difference between Tube B and A resulted from extra CH₄ oxidation in water column of Tube A. A positive correlation between CH₄ oxidation rate in water column and temperature ($R = 0.68$) was observed here, and the correlation coefficient can be improved to 0.90 when the average values were used (Fig. 3 & Supplementary Table S4).

CH₄ oxidation rate in both surface sediments and water column increased with temperature. A high correlation coefficient between CH₄ oxidation rate and its fluxes of Tube A and B in the warming process (20–30°C) is observed (Supplementary Table S5). Increased CH₄ oxidation rate under high temperature is bigger than increased CH₄ formation rate^{17,18}, which results in the similar decreasing CH₄ flux in Tube A and B during the warming process.

The average CH₄ oxidation in surface sediments accounts for 51.8% of the total CH₄ production with a range of 26.1–77.4%. Therefore, CH₄ oxidation played a vital role in controlling the CH₄ flux². Considering the different CH₄ consumption contribution of water at different water temperatures (Table 2), average CH₄ oxidation in water column accounts for about 46.7% of the total oxidation, and the value reaches to 73.9% when temperature is 35°C. Our data accords well with previous reports, in which water column oxidized 51 ~ 80% of the CH₄ produced in freshwater lakes²⁷, and also played a fundamental role in regulation of CH₄ emissions. In Lake Kasumigaura, CH₄ oxidation consumed an annual average of 74% of dissolved CH₄ in the water column. This lake is very shallow with a maximum depth of only 7.3 m and a mean depth of 3.8 m¹⁴.

The bigger the reservoir water depth is, the more CH₄ is oxidized in water column. The change amplitude of water depth in the TGR is about 30 m when its water level changes from 145 m to 175 m²⁸, which can influence CH₄ consumption largely (Table 3). The oxidation can be increased by 41.0 mg/(m²·h) in 30 m water column when the average CH₄ oxidation contribution rate, 1.37 mg/(m³·h), was used to calculate the CH₄ oxidation in water column.

Here, higher temperature is conducive to decreasing the CH₄ flux in Xiangxi Bay of TGR, which is quite different from the results from eight lakes of boreal and northern regions in central Sweden¹⁷. According to the latter, CH₄ emissions would increase if the climate gets both warmer and more variable. Further work is expected to explore the potential mechanism between CH₄ formation and oxidation in fresh aquatic systems. Many factors can determine various CH₄ formation and consumption process²⁷. For instance, the trophic

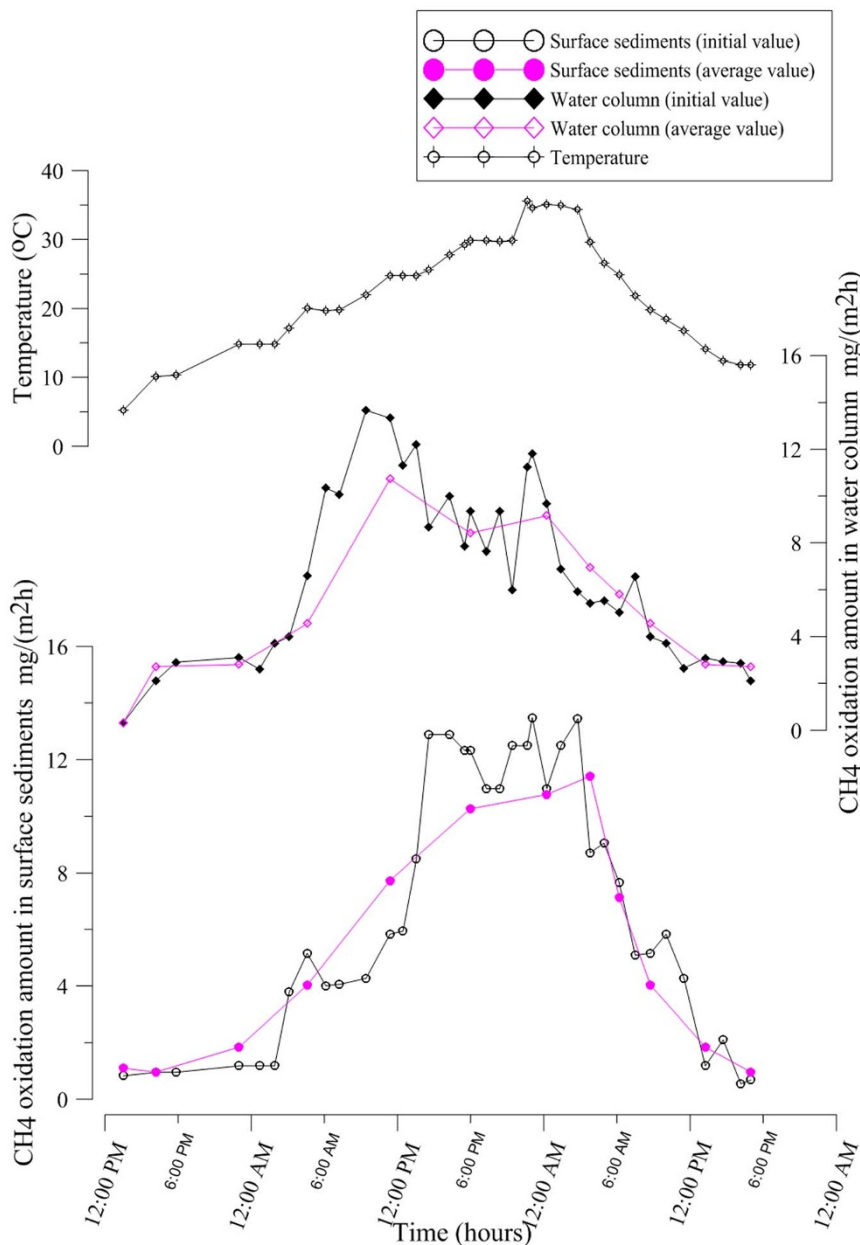


Figure 3 | Changes of CH₄ oxidation amount with temperature in surface sediments and water column (initial and average values). Average value was also used during the rapid warming process from 5 to 20°C. The difference between fluxes of Tube C and B is the decrease of CH₄ flux due to oxidation consumption in surface sediments, while the difference between B and A results from oxidation consumption in water column. Average values of CH₄ fluxes were used during rapid warming process from 5 to 20°C. The change of decrease of CH₄ flux can be interpreted as oxidation amount. Oxidation rate, which is the oxidation amount in unit time, is positively correlative to temperature due to the temperature-controlled formation process. Faster increase rate of oxidation process results in a decrease process of the total CH₄ emission.

state of water, state method, sediments origin, climatic region and so on. Whether or not consumption rate dominates the whole process will lead to the opposite results.

Deeper water and higher temperature, whether in summer or winter, can decrease the CH₄ emission in Xiangxi Bay of TGR according to the results of our research. CH₄ effluxes range from 1.26 to 4.65 mg/(m²h) under different water depth and temperature

conditions. The highest CH₄ oxidation rates are located at oxic/anoxic interfaces where opposite fluxes of CH₄ and O₂ occur²⁹. A large fraction of CH₄ diffusing from the anoxic zone is oxidized at the sediment surface (66 ~ 95%²⁹⁻³¹) and in the water column (45 ~ 100%³²). CH₄ consumption in Xiangxi Bay of the TGR is mostly determined by the surface sediments oxidation effect of about 51.8%, and water bodies of about 46.7%.

Xiangxi Bay of TGR has an approximate CH₄ flux to the Middle Yangtze River²⁴, both of which are in the subtropical climate zone. CH₄ fluxes in the TGR are much smaller than those data from water bodies of tropical zone such as Petit Saut and Miranda^{33,34}, but larger than those from the reservoirs in temperate zone and frigid zone such as Québec and Lokka^{6,11}. This verifies that CH₄ emissions were correlated to latitude³.

Table 2 | CH₄ consumption contribution rate of water at different water temperatures

Water temperature(°C)	5	10	15	20	25	30	35
C (mg/(m ³ ·h))	0.09	0.73	0.76	1.22	2.23	2.07	2.47

Table 3 | CH₄ consumption rate in water column at different water depth

Water depth(m)		0	5	10	15	20
C (mg/(m ² ·h))	Values range	0	0.43 ~ 12.36	0.86 ~ 24.71	1.29 ~ 37.07	1.72 ~ 49.42
	Average value	0	6.84	13.68	20.52	27.36
Water depth(m)		25	30	35	40	45
C (mg/(m ² ·h))	Values range	2.15 ~ 61.78	2.58 ~ 74.13	3.01 ~ 86.49	3.44 ~ 98.84	3.87 ~ 111.20
	Average value	34.20	41.04	47.88	54.72	61.56

Xiangxi Bay of TGR is a human-made freshwater system, and its CH₄ flux may decrease as time goes on according to the negative correlation between gas flux and reservoir age³. At the same time, the earth is experiencing a global warming process obviously, particularly after 1980s³⁵. The air temperature of TGR area also has a rising tendency³⁶, which increased by 0.2–1.0 °C in 2003–2009 when compared to the average value in 1996–2002³⁷. Thus, CH₄ flux in Xiangxi Bay of TGR may decline to some degree over the next decade.

Methods

TGR is an artificial fresh river-channel reservoir which formed after the construction of the Three Gorge Project (TGP) with an area of 1084 km². It has a total reservoir capacity of 39.3 billion m³ with a normal water level elevation of 175 m³⁸. The reservoir is located in a sub-tropical continental monsoon climatic region. The spring has a plenty of rain with temperature changing frequently; midsummer drought occurs frequently in summer with burning hot and concentrated rainfall; the steady raining is continual in autumn and winter; moreover, the winter also has early frost. The annual change range of water level is about 30 m which is influenced by flood control operation of TGR³⁹. TGR has a steep-sided gorge characteristic rather than a relatively shallow basin characteristic when compared with the boreal and tropical reservoirs⁴⁰.

Xiangxi River, as the largest tributary flowing into the Yangtze River in Hubei of TGR area, is located in the western Hubei province of China⁴¹. The river is 94 km and its estuary is about 32 km from the dam site. The geographical coordinates of the River are 30°57' ~ 31°34'N, 110°25' ~ 111°06'E^{42,43}.

Surface sediments and water samples used here were both taken from the Xiangxi Bay. The sediments were mixed gently in order to decrease spatial variability of sediments inside and the destruction of the microbial communities. These communities are the main part of CH₄ formation processes^{44,45}. Sediments used for experiment were well mixed^{46,47} and equally divided into thirds (each weighed about 2.9 kg), and then were transferred into three 1.0 m long acrylic round tubes (signed A, B and C, and the length of sediments in each tube was 35 cm). The free space of tubes was filled with water, and the sediments were immersed more than 72 hours prior to the experiment.

Tube A, B and C were designed to simulate three different situations from CH₄ formation to oxidation. Tube C is designed to simulate CH₄ formation without any aerobic oxidation. Tube C was filled with pure nitrogen to maintain anaerobic condition in Tube C 3 days ago before the experiment. Then pure nitrogen flow was injected into the tube continually for 12 hours so as to avoid CH₄ accumulating and equilibrate the pressure inside with outside atmospheric pressure before the experiment. Tube A is designed to simulate CH₄ formation with oxidation in surface sediments and water column, and B is designed to simulate CH₄ formation only with oxidation in surface sediments. Water was injected into Tube A to column of 27 cm before experiment in order to simulate CH₄ oxidation in water column.

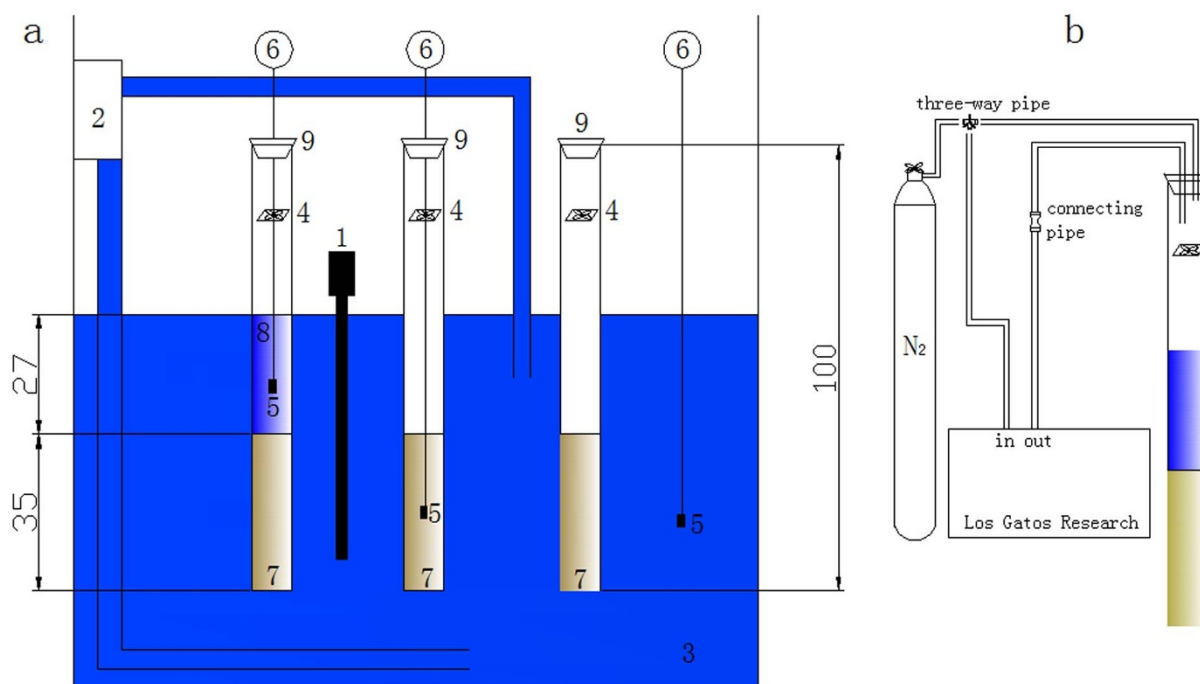


Figure 4 | Experimental equipment layout. (a) The whole experiment system consists of a big water tank with electric heater (1), micro-pump (2), small fans (4), temperature transducers (5) and electronic thermometers TP3001 (6), some three-way and connecting pipes. The environment water (3) in the tank was darkened by ink to shield sediments from light. Three 1.0 m long acrylic round tubes were signed A, B and C as experimental controls, and the length of sediments from Xiangxi Bay (7) in each tube was 35 cm. Tube A also has a water column of 27 cm from Xiangxi Bay (8). All of these tubes were sealed by gum stoppers (9) with two small two measurement holes. Free space of Tube C was filled with pure nitrogen, while Tube A and B were filled with air. (b) Experimental equipment of Tube A. Three-way pipes, connecting pipes and other pipes connect the tube with nitrogen cylinder and LGR. The Greenhouse Gas Analyzer (CH₄, CO₂, H₂O) LGR from Los Gatos Research was used to measure the concentration of greenhouse gases.

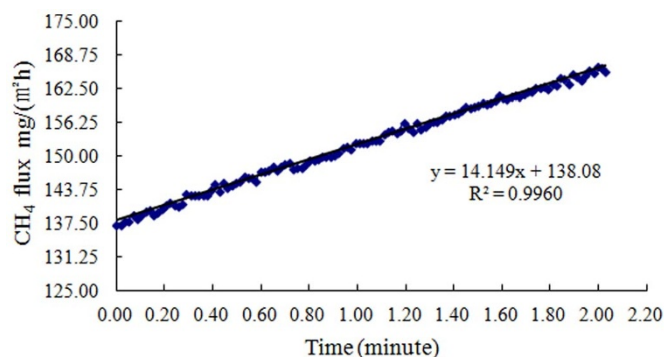


Figure 5 | The diagram illustrates the calculation of CH₄ flux. The blue line is measured values. The black line is a trend line. The R² (squared correlation coefficient) and linear formula of it is shown.

The whole experiment system consists of a big water tank with three tubes inside, electric heaters, a micro-pump, a few of pipes, some three-way and connecting pipes, some small fans, an air pump, barometers, stabilizer, gum stoppers and a greenhouse gas analyzer (Fig. 4). The Greenhouse Gas Analyzer (CH₄, CO₂, H₂O) LGR (Los Gatos Research, USA, main technical parameters: CH₄: 1 ppbv, CO₂: 0.2 ppmv, H₂O: 100 ppmv; the accuracy: <1%; measuring range: CH₄: 0.1–20 ppmv, CO₂: 200–4000 ppmv, H₂O: 7000–70000 ppmv; precision is better than 1%) was used to measure the concentration of greenhouse gases in the tubes.

The continuous experiment process included a warming period and a cooling one, during which the temperature ranged from 5 to 35 °C. In warming process, the temperature of 5, 10, 15, 20, 25, 30 and 35 °C were maintained for about 3 hours. Micro-pump was used here to mix the water of tank well. The whole experiment lasted for more than 60 hours. Once the gas contents in each tube were measured, pure nitrogen was injected to lower the concentrations of CH₄ and CO₂ in the tube right away. The dissolve oxygen (DO) concentration of water in Tube A was measured every two hours. Average DO of the experiment was kept around 9.27 mg/L.

When calculating CH₄ fluxes, a series of scattered points were fitted into a linear function model only when the R² (squared correlation coefficient) is bigger than 0.995 in most cases. As an example, the diffusion rate (k)⁴⁸ of CH₄ in Tube C at 5 °C is shown as Fig. 5.

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Author contributions

C.H.W. and S.B.X. prepared the primary manuscript and figures. Y.C.L. and X.C.L. prepared the simulation experiment. F.P. and H.Y.Z. collected the samples from Xiangxi Bay. All authors reviewed and discussed the manuscript.

Additional information

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