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Temporal variation of methane flux from Xiangxi Bay of the Three Gorges Reservoir

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Three diel field campaigns and one monthly sampling campaign during June 2010-May 2011 were carried out to investigate the CH_4 flux across the water-gas interface in Xiangxi Bay of the Three Gorges Reservoir, China. The average CH_4 flux was much less than that reported from reservoirs in tropic and temperate regions. The photosynthesis of phytoplankton dominated the diel gas fluxes during alga bloom in spring and summer. The maximum monthly flux occurred in June 2010 and corresponded to the lowest water level. Water temperature, sediment temperature, and TOC did not have significant correlation with the monthly CH_4 fluxes. Continuously decreasing hydrostatic pressure and the low water level resulted in more CH_4 emission at the sediment-water during the discharging period, and thus increases the CH_4 effluxes because the diffusion time through a thin water column is shorter and less CH_4 may be oxidized compared with that in a long water column.

CH_4 is an important atmospheric contaminant contributing to the greenhouse effect, almost 2/3 of the current CH_4 emissions are anthropogenic and the present CH_4 concentration of 1.77 ppmv is more than twice its preindustrial value[1,2]. Atmospheric CH_4 concentrations showed significant variations corresponding to the abrupt climate events[3,4]. Dam projects and freshwater reservoirs generate numerous impacts both on the region where they are located, as well as at an inter-regional, national and even global level (socioeconomic, health, institutional, environmental, ecological and cultural impacts)[5]. However, the conversion of land surface areas saturated by oxygen to anoxic sediments overlain by water results in CH_4 emissions from reservoirs under certain conditions[6–8].

Gas fluxes in natural ecosystems are known to be extremely variable, and sediment temperature, water temperature, DOC, NO_3^- availability and eutrophication level are strong regulators of greenhouse gas dynamics in the fluvial reservoir[7,9–11]. However, no relationship between GHG fluxes and DOC was observed in eutrophic water bodies[7,11], which might indicate that biogeochemical processes in the corresponding lakes/reservoirs are not C-limited[7]. Numerous investigations[12,13] did fail to find relationships between GHG fluxes and water temperature in aquatic ecosystems. CH_4 is exclusively formed in anaerobic environments[14], and therefore it is mostly produced in anoxic sediments[13]. This gas is then partially mineralized into CO_2 through aerobic oxidation by methanotrophic bacteria in the oxic layer of sediments or in the water column, and only the unoxidized fraction escapes to the atmosphere as CH_4[13,15]. In a summary, methane emissions from aquatic environments depend on methane formation and methane oxidation rates[16].

Biogenic CH_4 is produced by the activities of methanogens, a strictly anaerobic metabolic group belonging to the *Archaea*[17]. Many factors such as oxygen concentrations, competition for substrate acquisition, organic matter content and quality and temperature impacts CH_4 production rates either by affecting methanogens directly or indirectly by structuring the surrounding microbial community[17]. Sediments temperature is an important parameter influencing methanogenesis rates[9,10,16,18,19]. Numbers of methanogenic bacteria increase and rates of methanogenesis are correlated with increased sediment temperature during seasonal change[20].

Methane oxidation plays a vital role in controlling the flux of CH_4 from many ecosystems[8]. The efficiency of biological methane oxidation depends on physico-chemical conditions and on the means of methane transport[17]. Biological methane oxidation is carried out by methanotrophs which oxidize 30–99% of the CH_4 produced in freshwater lakes[21] and then plays a fundamental role in regulation of methane emissions. O_2 and CH_4

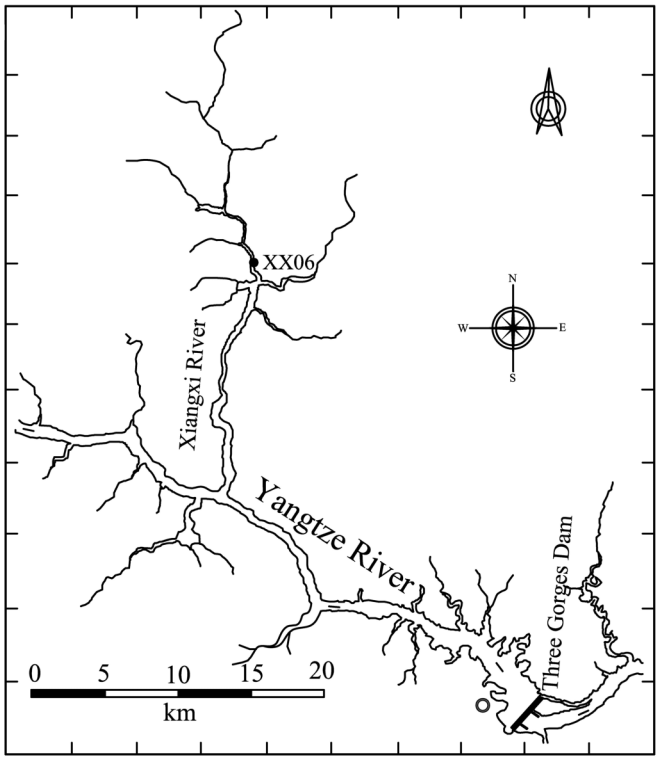

Figure 1 | Location of sampling site in the Xiangxi Bay of the Three Gorges Reservoir (Modified from Yang et al[30].).

concentrations, temperature, availability of nitrogen, and so on may all have influence on methane consuming[22]. How different environmental conditions affect the distribution, numbers, and activity of methanotrophs remains to be further studied[22].

As we known, the revolution and rotation of the earth results in diel and seasonal biogeochemical cycles, which are in response to the solar photocycle, particularly during stable hydrological conditions[23]. The amplitude of some of these diel changes can be as large as changes occurring on annual timescales[23]. However, less attention to the variation of CH_4 flux is paid on the diel timescale than that on the seasonal timescale. The former has received attention only more recently[23]. Study of diel variations is helpful to reveal which biogeochemical processes occur relatively rapidly in natural waters and therefore which processes play an integral and important role in the normal functioning of natural water systems[23].

Here, we present seasonal and diurnal variations of CH_4 flux across the water-gas interface in the Xiangxi Bay (XXB) of the TGR (Fig. 1). The bay suffers from serious alga blooms frequently. The goal of this paper is to disclose the temporal variation of CH_4 flux in the bay, and probe key factors which dominate the variation and possible reservoir operation to mitigate the CH_4 efflux.

Results

Diel CH_4 flux. Our CH_4 flux datum was much less than that from permanently flooded areas in the mainstream of the Yangtze River[24]. The diel CH_4 flux varied greatly during April 27–28, 2011 and October 4–5, 2010 (Fig. 2 & Fig. 3), and changed less during August 23–24, 2010. The average CH_4 flux during October 4–5, 2010 is ~0.081 mg m^{-2} h^{-1}, which is approximate to that of August 23–24 and much less than that during April 27–28, 2011 (Table 1). The diel average of flux observed during April 27–28,

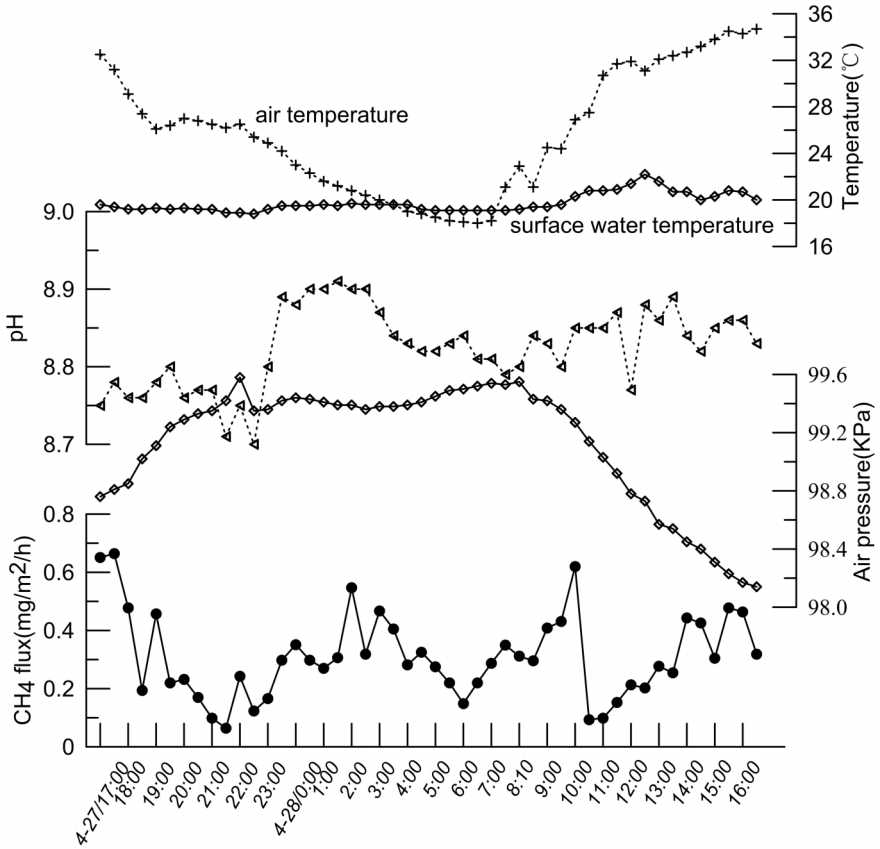

Figure 2 | Diel CH_4 flux and corresponding environmental parameters during April 27–28, 2011.

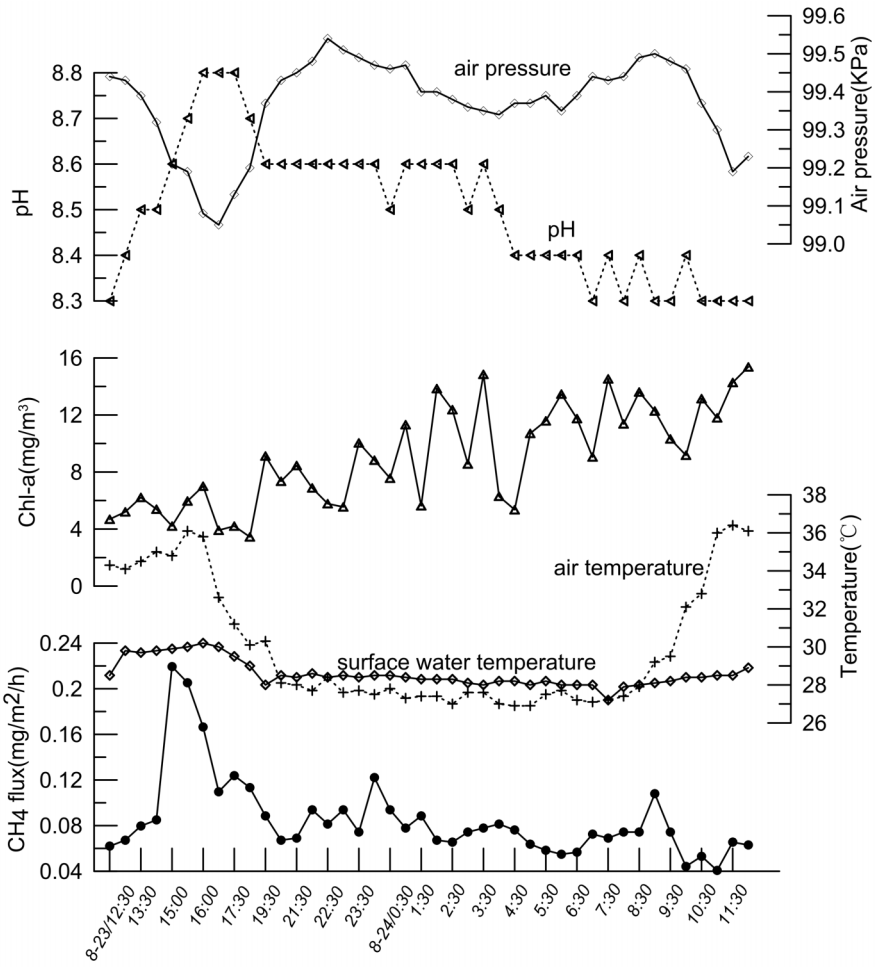

Figure 3 | **Diel CH_4 flux and corresponding environmental parameters during October 4–5, 2010.**

2011 was ~3.6 and ~3.8 times of that during August 23–24, 2010 (Fig. 4) and October 4–5, 2010 respectively.

Seasonal CH_4 flux. The surface water temperature changed with the air temperature synchronously as a whole (Fig. 5). Water depth varied from 19 to 48 m, and it was deepest during October 2010-January 2011. Total organic carbon (TOC) in surface water showed a fluctuating downward trend with time. Surface water pH ranged from 7.9 to 9.4, and the maximum occurred in March 2011. Eutrophication of the bay occurred frequently as a consequence of large influxes of nutrients in most time of the observation year. The four highest Chl-a occurred in the time of low water level and warm season.

Dissolved CH_4 content in surface water varied greatly from very low level to 6.32 μg L^{-1} with an average of 1.74 μg L^{-1}. The minimum occurred in August 2010, and the three highest occurred in June 2010, April 2011 and May 2011.

The CH_4 fluxes during the observation year ranged from −0.120 to 31.008 mg m^{-2} h^{-1} with an average of 3.288 mg m^{-2} d^{-1}, which was much less than that reported from reservoirs in tropic and temperate regions[13,25–27]. (Tucuruí (deep) and Samuel (shallow) reservoirs of Amazon hydroreservoirs released in average 13.82 ± 22.94 and 71.19 ± 107.4 mg CH_4 $m^{-2}d^{-1}$, respectively[25]. CH4 fluxes measured in three tropical reservoirs located in French Guiana (Petit Saut) and Brazil (Balbina and Samuel) were in the range of 48 ± 32 mg m^{-2} d^{-1}[26,27].) The maximum flux, which occurred in June 2010, was corresponding to the lowest water level. The CH_4 flux was less than 0 in July 2010 and March 2011, which indicated that water body absorbed CH_4 from the air in some degree.

Discussion

Environmental parameters have different influences on the diel CH_4 fluxes in different seasons. No significant correlation between the diel CH_4 flux during April 27–28, 2011 and corresponding

Table 1 | Comparison of diel CH_4 flux and some environmental factors in different seasons

	air temperature (°C)		surface water temperature (°C)			CH_4 flux	
time	range	variation amplitude	range	variation amplitude	surface water pH	Range (mg m^{-2} h^{-1})	Average (mg/m^2/d)
April 27–28, 2011	18.0 – 34.7	16.7	22.2 – 18.8	3.4	8.7–8.9	0.064–0.664	7.464
August 23–24, 2010	36.4 – 26.9	9.5	30.2 – 27.2	3.0	8.3–8.8	0.041–0.219	2.064
October 4–5, 2010.	27.3 – 15.7	11.6	24.4 – 23.2	1.2	7.6–8.2	0.000–0.202	1.944

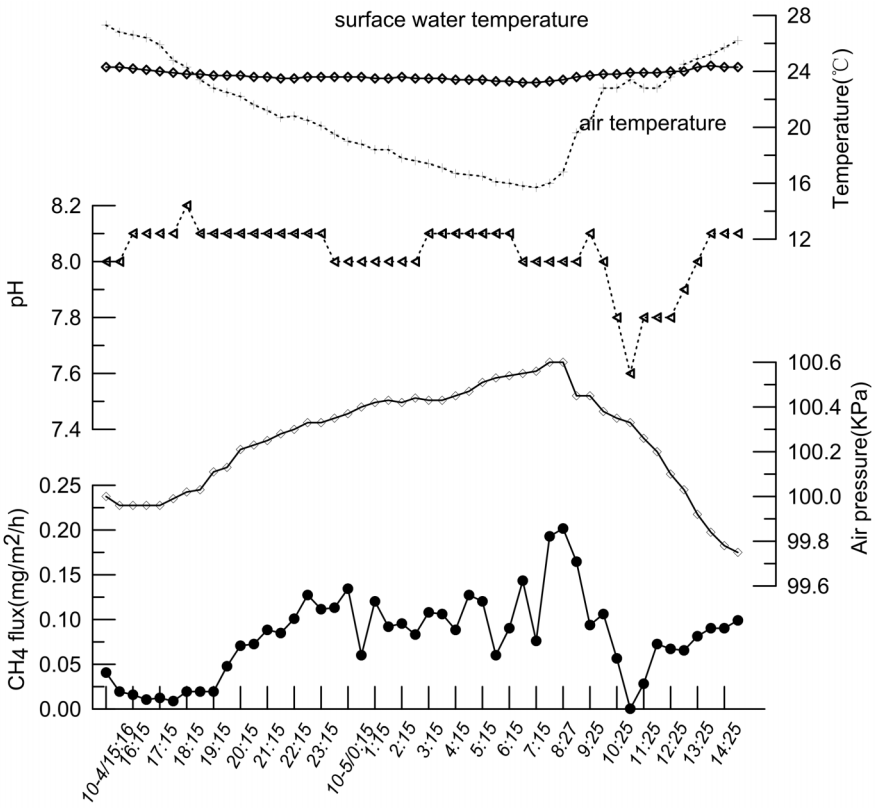

Figure 4 | Diel CH_4 flux and corresponding environmental parameters during August 23–24, 2010.

environmental parameters was observed. The diel CH_4 flux during August 23–24, 2010 was positively correlated with the air temperature, the surface water temperature and pH, and negatively correlated with the air pressure and Chl-a concentration of surface water. However, the diel CH_4 flux during October 4–5, 2010 was positively correlated with the air pressure and negatively correlated with the air temperature and the surface water temperature, which was in the opposite to the situation observed during August 23–24, 2010.

The photosynthesis of phytoplankton may dominate the diel gas fluxes during alga bloom, such as during August 23–24, 2010 and April 27–28, 2011. Because diel changes of Chl-a in surface water were not monitored on October 4–5, 2010 and April 27–28, 2011, the detailed process how the photosynthesis of phytoplankton influenced the diel CH_4 fluxes was difficult to be perceived. Average Chl-a content of surface water on October 4 and 5, 2010 was 0.82 and 1.49 μg L^{-1} respectively, which was much lower than that occurred on April 27–28, 2011 (21.10 and 9.78 μg L^{-1} on April 27 and 28 respectively) and August 23–24, 2010 (average 9.06 μg L^{-1}l). Relationship between the diel CO_2 and CH_4 flux was also dominated by the situation of eutrophication. Significant positive correlation between them was observed in low Chl-a level, however, significant negative relation in high Chl-a level.

A significantly positive correlation between the seasonal flux and the dissolved CH_4 content in the surface water was observed in present study. The seasonal CH_4 flux was also positively correlated with the air temperature and the surface water pH, and negatively correlated with the air pressure.

O_2 and CH_4 concentrations, temperature, availability of nitrogen, and so on may all have influence on methane consuming[22]. Correlation coefficient between seasonal CH_4 flux and CO_2 flux was -0.563(N = 12). Here the situation was thought to be resulted from the high levels of eutrophication in XXB. XXB is a productive system, and serious alga bloom is a frequent problem since the initial filling of the Three Gorges Reservoir in June 2003[28–31]. Alga bloom induced the increasing DO, and the latter was significantly positively correlated with pH (R = 0.882, P = 0.01), which was observed in many water ecosystems[32]. Here, the two biggest DO contents in water bodies occurred in June 2010 and April 2011 (Fig. 6), in which months the highest dissolved CH_4 content and CH_4 flux appeared during the observation year. It might indicate that DO was not the key factor that influenced the CH_4 flux in the bay. DOC and NO_3^- availability are strong regulators of GHG dynamics in the fluvial reservoir[7]. However, both the parameters did not have significant correlation with the monthly CH_4 flux. Nonetheless, the correlation coefficient between the CO_2 flux and TOC was 0.502(N = 10), which shows that there is some relationship between them and it's different from those eutrophic lakes/reservoirs.

The three highest bottom water temperatures were observed in August, September and October 2010 at Site XX06 (Fig. 5). However, the dissolved CH_4 content in the surface water and the CH_4 flux observed in the three months were not bigger than those in other months and at a low level. Meanwhile, the bottom water temperature in the months of June 2010, April 2011 and May 2011, when higher dissolved CH_4 content and/or bigger CH_4 flux occurred, was not higher than that in other months (Fig. 5). So, the main reason which dominated the seasonal CH_4 flux at the water-air interface might not be the sediment temperature. In fact, methane production rates in the shallow sediments should be more sensitive to seasonal variations of temperature than in the deep sediments[17,20]. On the contrary, increased water temperature could greatly increase the CH_4 oxidation rates[8], which dominated the CH_4 fluxes in deep reservoirs/lakes.

CH_4 in sediments enters into overlying water by diffusive and ebullitive transport. Bubble fluxes mainly occur in shallow parts of lakes and reservoirs where the hydrostatic pressure is not high enough to dissolve CH_4 in interstitial water[26,33]. However, bubbles can also be released from the sediment in deeper parts of lakes and reservoirs, but these bubbles tend to dissolve into the water during their transport through the water column and so do not reach the

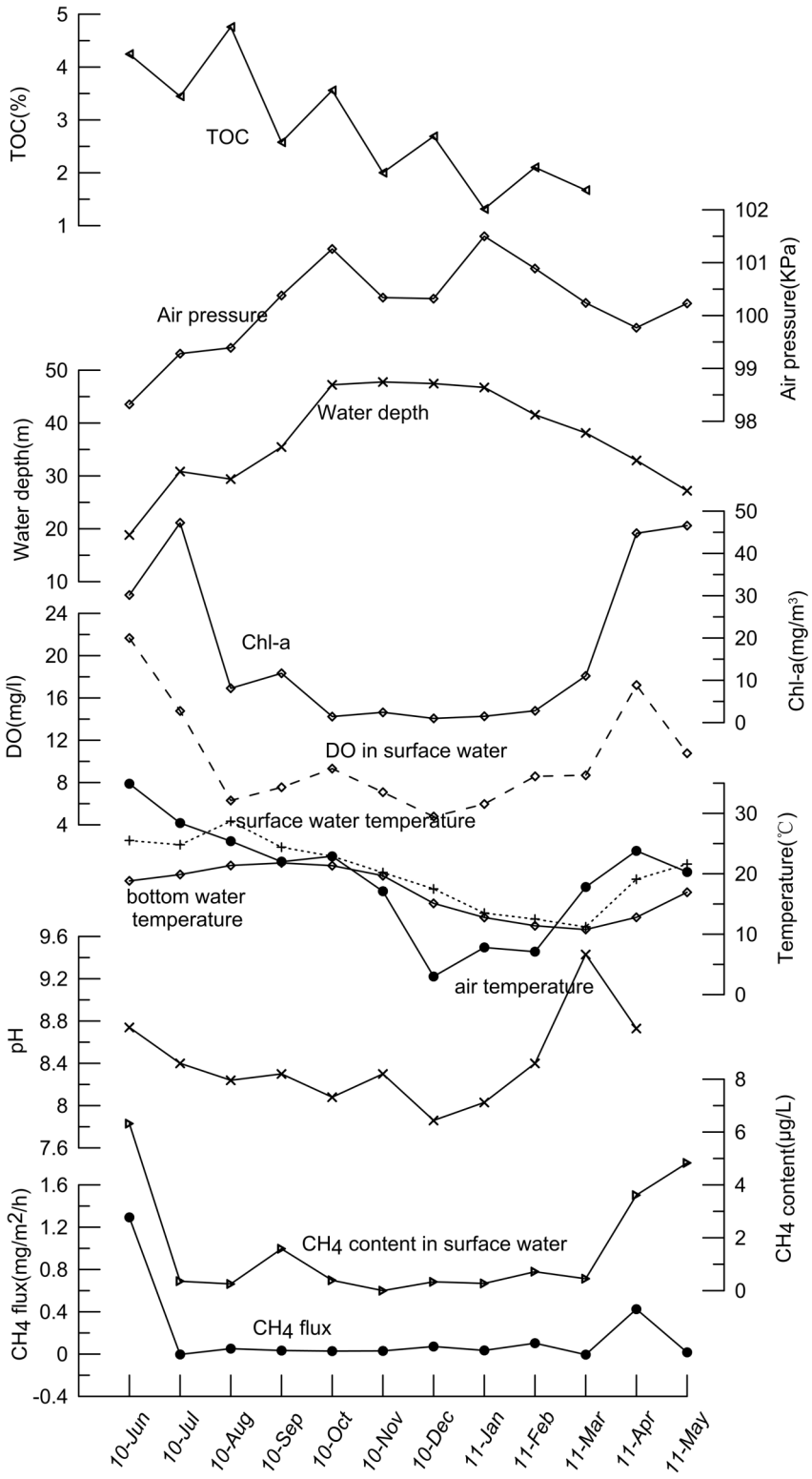

Figure 5 | Seasonal CH_4 flux and corresponding environmental parameters during the observation year.

atmosphere[34]. The release of bubbles can be triggered by variation of hydrostatic pressure associated with rapid changes of the water level above the sediment[35]. A decrease in the water level above the sediment and sediment pressure increases the CH_4 effluxes. The diffusion time through a thin water column is shorter and less CH_4 may be oxidized compared with that in a long water column[36,37].

The water level of TGR fluctuates from ~145 to ~175 m in order to control flooding, and it is usually continuously decreased from January to June (Fig. 7). The three biggest dissolved CH_4 contents of the surface water occurred in June 2010, April 2011 and May 2011, which were in the late stage of the discharging period of TGR. To the end of May 2011, the water level fell ~30 m, which resulted in the length of overlying water column reduced by ~60%. In this report, greatly and continuously decreasing water level caused that CH_4 efflux in June 2010 was ~33-fold higher than the averaged flux of the next 9 months. Continuously decreasing hydrostatic pressure

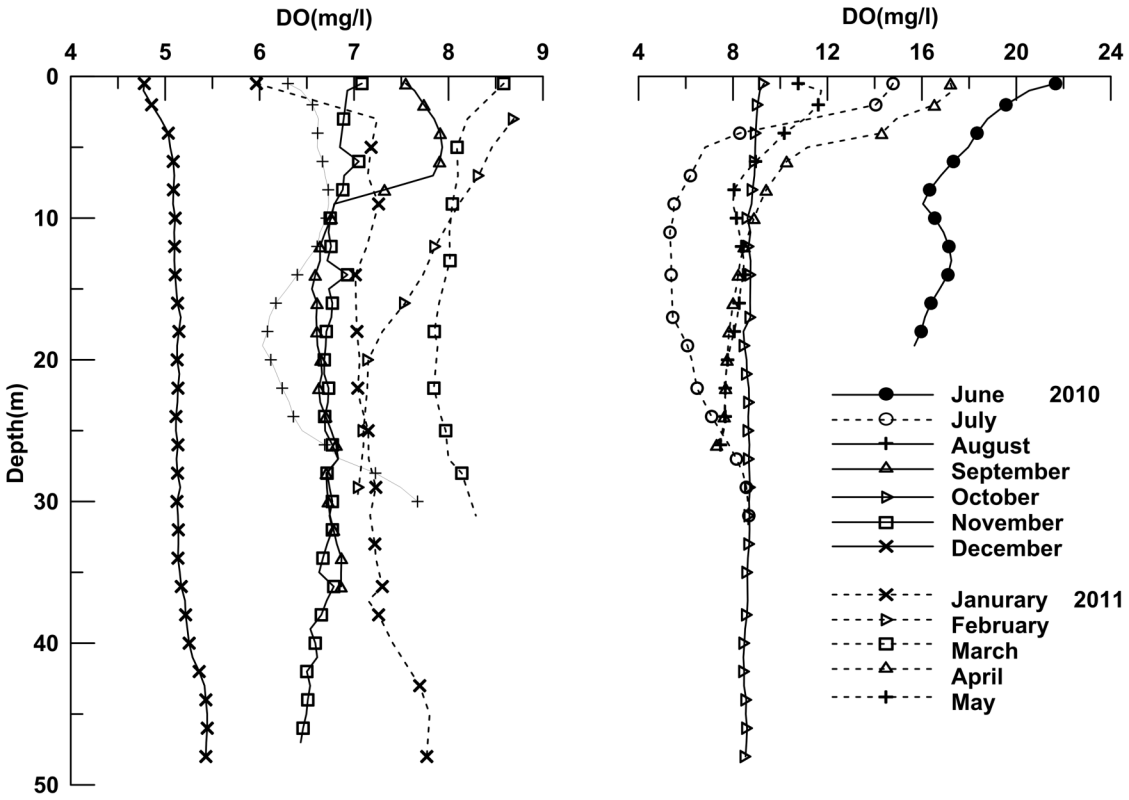

Figure 6 | DO in the water column at XX06 in the Xiangxi Bay during the observation year.

might result in more CH_4 emission. Meanwhile, the low water level in these three months was conducive to more CH_4 transported to surface water in stead of being oxidized in a long water column. Thus, a positive correlation between the water depth and CH_4 flux was observed here. Our study also shows that sediment-generated methane can easily evade the shallow reservoir, while the deep reservoir extends methanotrophic layer, oxidizing large quantities of methane coming from the sediments[25]. However, our explanation remains further proof owing to no data of the dissolved CH_4 content in the bottom water.

So, it's advised that several stages during the discharge period could be set to keep relatively stable water level (Fig. 7), in which more CH_4 would be emitted from sediments and oxidized in a long water column instead of reaching the atmosphere. If half of the

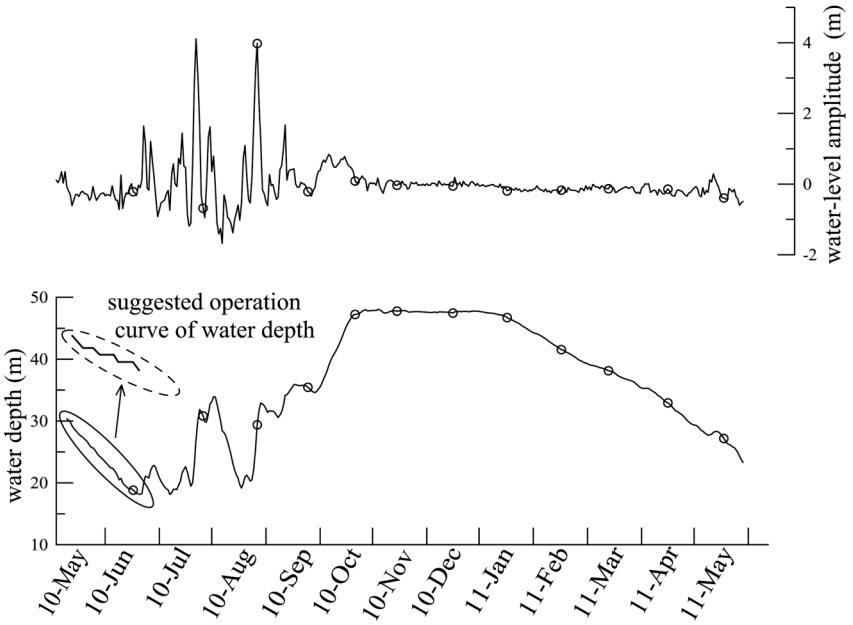

Figure 7 | The daily water depth curve at Site XX06. The data of water level were from the http://www.ctg.com.cn/inc/sqsk.php. The fold line surrounded by the dotted line is a sketch for suggested operation curve of water depth. *See the text.*

present CH_4 flux emitted in April, May and June were oxidized, a total of 683.93 t CH_4 would be consumed in water column supposed that the area of all bays accouns 1/3 of the Three Gorges Reservoir.

Methods

Study area and monitoring site. The TGR, which is located at the upper reach of the Yangtze River and resulted from the largest water-control projects in the world, is built for flood control, power generation, river navigation and drought prevention. The reservoir has a total capacity of 393 billion m^3 and a flood control capacity of 221.5 billion m^3 with the water elevation of 175 m. The operation of the reservoir depends on the temporal and spatial conditions and many other factors, and the impoundment elevation ranges from 145 to 175 m annually[38]. The TGR is a huge and typical fluvial reservoir, which is neither a lake nor a stream, but exhibits hydrologic behavior that is intermediary between these aquatic systems[21]. Meanwhile, its water lever is low during the summer rains and high during dry seasons for the sake of flood controlling and electricity generating. The variation of its water level is opposite to natural lakes. Primary reports of CH_4 emission flux varied greatly in different zones of the reservoir, for example, it was big at the marshes in the drawdown area[39] and very small in open water[24,40]. The Xiangxi River (XXR) is the largest tributary of the TGR in Hubei Province, and also a larger one close to the Three Gorges Dam (Fig. 1). It flows southwardly into the mainstream (the Yangtze River) of the reservoir at Xiangxi town, Zigui County. The main stream of the XXR is 94 km in length, and its drainage basin is located in 110°25′–111°06′E, 30°57′–31°34′N with an area of 3,099 km^2. The drainage area is of a sub-tropical continental monsoon climate with greatly changing temperature in springs and concentrated rainfalls in summers. It is often suffered from heavy rain and drought in summers, and the weather is rainy in autumns and snowy in winters. The vertical temperature changes significantly owing to the disparate terrain elevation difference and complicated landform. The average annual temperature is 16.6°C, and the average rainfall and runoff are 1,015.6 mm and 40.18 $m^3\ s^{-1}$. When the impoundment elevation of the TGR reaches 175 m, the backwater zone in XXR is about 40 km long[30]. The backwater zone is named as XXB, because it is similar to a lake but with characteristic hydraulic conditions. The Bay is thermally stratified most of the time[30]. XXB is a productive system, and serious alga bloom occurs frequently after the initial filling of the TGR in June 2003[28–31]. The monitoring site is located at the middle of XXB, ~16 km to the mainstream of the Yangtze River and marked as XX06 here (Fig. 1).

Sampling scheme. Three diel field campaigns, which were carried out on August 23–24 2010, October 4–5 2010 and April 27–28 2011 respectively, were undertaken at site XX06. A monthly sampling campaign was undertaken over a period of twelve months from June 2010 to May 2011, which was usually carried out at 9:00–10:00 of a day in the middle of each month.

In situ sampling measurements and analysis. Water temperature, pH, alkalinity, air temperature and wind speed were measured in situ. Water samples were taken from 0.5 m below the water surface for analysis of dissolved CH_4, CO_2, inorganic and organic carbon. Water temperature, pH, DO, and water depth were measured with a Hydrolab DS5 Multiparameter Sonde.

Water samples (100 mL) for dissolved gas analysis were collected into N_2-pre-flushed and pre-evacuated gas sampling bags with syringes and needles, and were then immediately treated with 0.1 mL saturated $HgCl_2$. In the laboratory, a headspace was created in the bags by injecting 200 mL of nitrogen gas. The bags were vigorously shaken, and left to equilibrate at ambient temperature for at least 2 h. 15 mL gas samples were sampled from the headspace with a gas-tight syringe and injected into an Agilent 7890A (Agilent Technologies, California, U.S.A) gas chromatograph equipped with a flame ionization detector. Dissolved gas concentration was computed as described in Johnson et al[41]. according to the Henry's law.

The water samples from the sites had been dealt and transported to lab to determine the concentrations of *Chl.a*, TP and TN, D-Si according to the "Water and wastewater monitoring and analysis methods (the fourth edition)"[42].

Water-to-air fluxes. Water-to-air CH_4 fluxes were determined by using floating chambers. The chambers are non-transparent thermally insulated tubs with a volume of 35.34 L and a surface area of 0.07 m^2 (radius and high are 0.15 and 0.5 m respectively). Fans were installed inside chambers to circulate air and homogenize GHG concentrations from the top to the bottom of the chambers.

A dynamic closed chamber system was used for diel CH_4 flux measurements. The chamber was connected to a Los Gatos Research's Greenhouse Gas Analyzer (DLT-100), which could monitor the CH_4 and CO_2 concentration inside the chamber continuously with 1 Hz frequency. The DLT-100 is a cavity ringdown spectrometer with high resolution (0.1 ppb) and precision (1% of reading the accuracy) and was already described in detail and used by previous researchers[43–46]. Single flux measurement is finished in 25 minutes. Then the chamber was taken off the water surface and put down again after the enough exchanging and mixing between gas inside the chamber and the environmental air.

Static closed chambers were used for monthly CH_4 and CO_2 flux measurements across the water-air interface. Gas samples from chamber headspace were taken with an interval of ~8 minutes for gas chromatograph (Agilent 7890A) using flame ionization detection (FID) and analyzed in two days. The details of the method are described by Wang and Wang[47]. The accuracy of the analyses was maintained by calibrating the gas chromatographs against a standard gas mixture after every 8 samples, which kept the coefficient of the replicated concentration determinations below 0.5%.

Calculation of the flux with laboratory analysis was described in detailed by Lambert and Fréchette[48].

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Author contributions

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Additional information

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