

## Effects of environmental factors on the methane and carbon dioxide fluxes at the middle of Three Gorges Reservoir

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### ABSTRACT

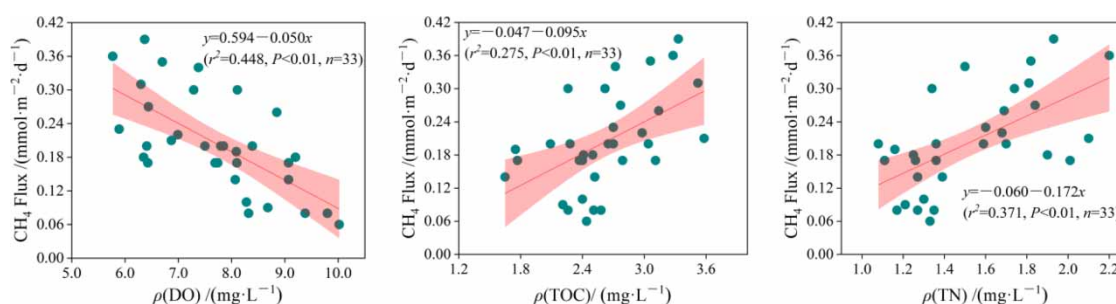
In recent years, scientists have paid special attention to the greenhouse gas emissions of the Three Gorges Reservoir. This study took Fuling to Wanzhou, located at the middle section of the Three Gorges Reservoir, as the research locations. From August 2017 to August 2018, the partial pressure of carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) and the flux of CO<sub>2</sub> and CH<sub>4</sub> at the water–gas interface were studied. Spearman's correlation between the partial pressure and discharge flux of CO<sub>2</sub> and CH<sub>4</sub> in water and environmental variables was analyzed. The research results showed that the variation of partial pressure and flux was consistent. At different sample locations, there was no statistical difference in CO<sub>2</sub> and CH<sub>4</sub> fluxes, but under different operating periods, the CO<sub>2</sub> and CH<sub>4</sub> fluxes were significantly different. The highest values happened during the drainage period as well as the low-water period, respectively. Total organic carbon and total nitrogen were significantly positively correlated, and dissolved oxygen was extremely negatively correlated. The high value of CH<sub>4</sub> flux in the middle of the reservoir area was related to the spatial distribution of the sediment and the amount of sediment deposition.

**Key words:** GHG flux, influencing factors, middle section of reservoir area, sediment deposition, surface waters, Three Gorges Reservoir

### HIGHLIGHTS

- This paper focuses on the greenhouse gas change in the middle section of the Three Gorges Reservoir area, which has received relatively little attention at present.
- The research time span of this paper is long, and the analysis of greenhouse gas (GHG) is comprehensive.

### GRAPHICAL ABSTRACT



## 1. INTRODUCTION

The study of greenhouse gas (GHG) source and sink changes in reservoirs has been an interdisciplinary frontier and hotspot in the field of global river ecological environment research and water conservancy and hydropower engineering in the past 20 years (Abril *et al.* 2014; Sawakuchi *et al.* 2014). According to some studies, reservoirs emit about 70 million tons of methane (CH<sub>4</sub>) and 100 million tons of carbon dioxide (CO<sub>2</sub>) every year, and reservoirs have become an important source of GHG in the atmosphere; what is more, the amount of CH<sub>4</sub> released from reservoirs accounts for 20% of the total CH<sub>4</sub> emissions from human activities (McCully 2002). The infrared absorption function of CH<sub>4</sub> molecule is stronger than that of CO<sub>2</sub>, which

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makes the global warming efficiency of CH<sub>4</sub> 20–32 times that of CO<sub>2</sub> (Rodhe 1990; Lemer & Roger 2001), and plays an important role in global warming. At present, the preliminary consensus is that the reservoir will have a high GHG flux in the initial 10–20 years and gradually stabilize after 20 years (Tremblay *et al.* 2004). Therefore, the main factors affecting the change of GHG source and sink of reservoirs are the age of reservoirs and the climate zone of reservoirs (Barros *et al.* 2011; Kumar *et al.* 2019). At the same time, the differences in rainfall, air temperature, wind speed and relative humidity among the crops on the two sides verified by the SAWT model also have indirect effects on the reservoir (Dash *et al.* 2021).

Dissolved oxygen (DO), chlorophyll concentration, temperature, surface water temperature, salinity and turbidity are known as water quality variables (Shamshirband *et al.* 2019). According to the existing studies, the release form of carbon is mainly CO<sub>2</sub> in the case of abundant oxygen in water, while the form is mainly CH<sub>4</sub> and CO<sub>2</sub> in the case of anoxic or anaerobic conditions. Therefore, an important indirect factor in the production, diffusion and release of GHG is the concentration of DO in water. In the tropical reservoirs, part of CH<sub>4</sub> generated in sediments rose to the water surface through diffusion (Rosa *et al.* 2003). However, due to the gradual increase in the concentration of DO in the rising process, most of the CH<sub>4</sub> was oxidized to CO<sub>2</sub> by CH<sub>4</sub>-oxidizing bacteria on the anaerobic–aerobic critical surface. As a result, the CH<sub>4</sub> release from the water–gas interface was significantly reduced, while the CO<sub>2</sub> release was increased. Changes in concentrations of C and N in water also affect the primary productivity of aquatic plants and the metabolic process of plankton, and the concentration of pollutants changes along the river, thus indirectly affecting the concentrations of CO<sub>2</sub> and CH<sub>4</sub> in water and the release fluxes of CO<sub>2</sub> and CH<sub>4</sub> at the water–air interface (Yang *et al.* 2013; Kargar *et al.* 2020).

After the completion of the Three Gorges Reservoir, several kilometers to tens of kilometers of river water were inundated to form slow or still water reaches in the lower reaches of the mainstream and tributaries of the Yangtze River, and river water was significantly reduced in the tributaries. The original diversified and fast-flowing river system disappeared, and a complex lake–river system with slow or even stationary water flow appeared (Nilsson & Berggren 2000). It is worth noting that the Three Gorges Reservoir is a typical channel canyon reservoir. After the establishment of the reservoir, the water level of the natural channel increases, and the flow velocity gradually slows down. The changes of the natural river hydrodynamic environment brought about by the construction of the dam and the water impoundment process also lead to the accumulation of sediment in the reservoir with the increase of the operating years. While the self-purification capacity of water body is weakened, for the river–lake transitional water body, the carbon which is from biogeographic transformation and GHG source and sink in the reservoir also presents transition and high heterogeneity in time and space (Morales-Pineda *et al.* 2014). For example, in the river area at the tail of the reservoir, the reservoir mainly degrades carbon from different sources because the upstream river has more organic matter from different sources. However, in the lake area at the head of the reservoir, due to the decrease of flow rate, the enhancement of light transmittance and the growth of algae, the reservoir mainly presents the process of endogenous carbon synthesis. The differences described above will, to some extent, lead to gradient changes in the source and sink of GHG in reservoirs in time and space (Li 2018; Li *et al.* 2018b). In the past 10 years, many teams in China have been monitoring and analyzing GHG and reservoir carbon cycle in the Three Gorges Reservoir (Li *et al.* 2016a; Liu *et al.* 2017; Li 2018). The main views are as follows: (1) The GHG source and sink volume of the water body in the Three Gorges Reservoir does not have the characteristics of high release flux as previously reported. On the contrary, it is at a low level in the global case base (Li *et al.* 2018b). (2) The mainstream of the Three Gorges Reservoir mainly degrades substances of different sources, while the algae growth in the backwater area of tributaries may present a local carbon sink phenomenon (Qin *et al.* 2017a).

At present, there are few reports on the characteristics and influencing factors of GHG sources and sinks in the middle section of the Three Gorges Reservoir. The middle section of the Three Gorges Reservoir (Fuling–Wanzhou section), as a transition section from ‘fluvial facies’ to ‘lacustrine facies’, is different from rivers and lakes in terms of channel characteristics (‘S’ type) (Li *et al.* 2016b), habitat characteristics, ecological structure and function, and key source factors (Li *et al.* 2018b). In addition, the current literature reports mainly focus on the macro characteristics of reservoir GHG sources and sinks, and the relevant literature reports focus on the spatial heterogeneity of reservoir GHG and its influencing factors are still limited. Therefore, the study of the typical sections of the Three Gorges Reservoir can effectively improve the understanding of GHG emissions in the reservoir ecosystem and clarify its impact on the global carbon cycle, which is of self-evident importance. In the preliminary investigation, the gas concentration in the water body of the Three Gorges Reservoir was initially tracked and monitored. The study found that the highest concentration of GHG in the water body appeared in the middle section of the reservoir, and there was no significant change in the concentration along with the depth of the river. It is preliminarily judged that this characteristic is related to the spatial distribution of sediment deposition in the mainstream of the

Three Gorges Reservoir to some extent and may be affected by the operation of the reservoir ‘storing clear water and discharging muddy water’ (Maeck *et al.* 2013).

Therefore, this study selected three typical sections of Fuling (FL), Zhongxian (ZX) and Wanzhou (WZ), which are in the middle section of the Three Gorges Reservoir, as the objects to carry out a 1-year *in situ* monitoring experiment. Combined with environmental factors such as meteorological and climatic conditions, the study revealed the main factors affecting the change of GHG source and sink in the middle section of the Three Gorges Reservoir. This study provides theoretical and data support for further study of GHG effects of the reservoir and provides a basis for further revealing the biogeochemical process of GHG source and sink changes in the middle section of the Three Gorges Reservoir under the operation of the reservoir.

## 2. MATERIALS AND METHODS

### 2.1. Study site

The length of the Three Gorges Reservoir is about 663 km. The average water surface width of the mainstream reservoir section is 1,100 m, and the water area is about 1,084 km<sup>2</sup> (Li *et al.* 2013; Luo *et al.* 2018). According to the operational plan of the Three Gorges Project about ‘storing clear and releasing muddy’, the water level of the reservoir will be lowered to 145 m before the flood season begins at the end of June and gradually be increased to 175 m after the flood season ends at the end of September. The range is between 145 and 175 m. The drainage period is from February to May, while the low-water-level operation period is from June to September, and from October of each year to January of next year is the period of impoundment and high-water-level operation (short for ‘high-water-level period’). When the water level is at a low running stage, the backwater of the mainstream reaches the downstream of Chongqing Longevity area, 524 km away from the leading edge of the dam, which is called the ‘perennial backwater area’ (Li *et al.* 2015a).

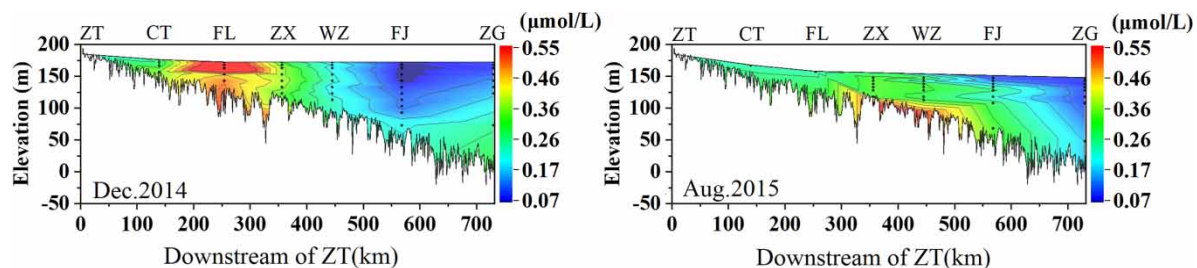
In the preliminary investigation, our team tracked and monitored the gas concentration in the water body of the Three Gorges Reservoir. The study found that the highest concentration of GHG in the water body appeared in the middle section of the reservoir (Wanzhou–Fuling section), and there was no significant change in the concentration along the depth of the river (Figure 1). Based on the above analysis, in this study, we chose three sections, Fuling (FL), Zhongxian (ZX) and Wanzhou (WZ), as key research areas in the ‘perennial backwater area’ mentioned to carry out *in situ* observation for 1 year.

FL, which is located in the middle of Chongqing and located at the conjunctions of the Yangtze River and the Wujiang River, is 120 km from Chongqing above and 482 km from the Three Gorges Dam below. This reach, with a flow of 86 km and an average annual flow of about 11,200 m<sup>3</sup>/s, is a typical mountainous river course in the upper reaches of the Yangtze River. The annual runoff distribution of this reach is extremely uneven, with an average flow of 1,050 m<sup>3</sup>/s from June to October in the flood season, and its runoff accounts for 74.5% of the annual runoff.

ZX, 190 km above Chongqing and 386 km below the Three Gorges Dam on the Yangtze River, is located in the eastern and central part of Chongqing. The reach is flat and wide, with a uniform flow of 88 km and an average annual flow of about 12,400 m<sup>3</sup>/s.

WZ, located in the northeast of Chongqing, is a central city in the upper reaches of the Yangtze River. It is 328 km from Chongqing and 283 km from the Three Gorges Dam. This reach is a stable section of large mountain rivers, with a relatively straight course of 84.3 km and an average annual flow of about 13,700 m<sup>3</sup>/s.

The data obtained by the study will be divided according to the above three water-level running periods to analyze the changes of GHG flux source and sink in the Three Gorges Reservoir section. Total amount of water will influence the



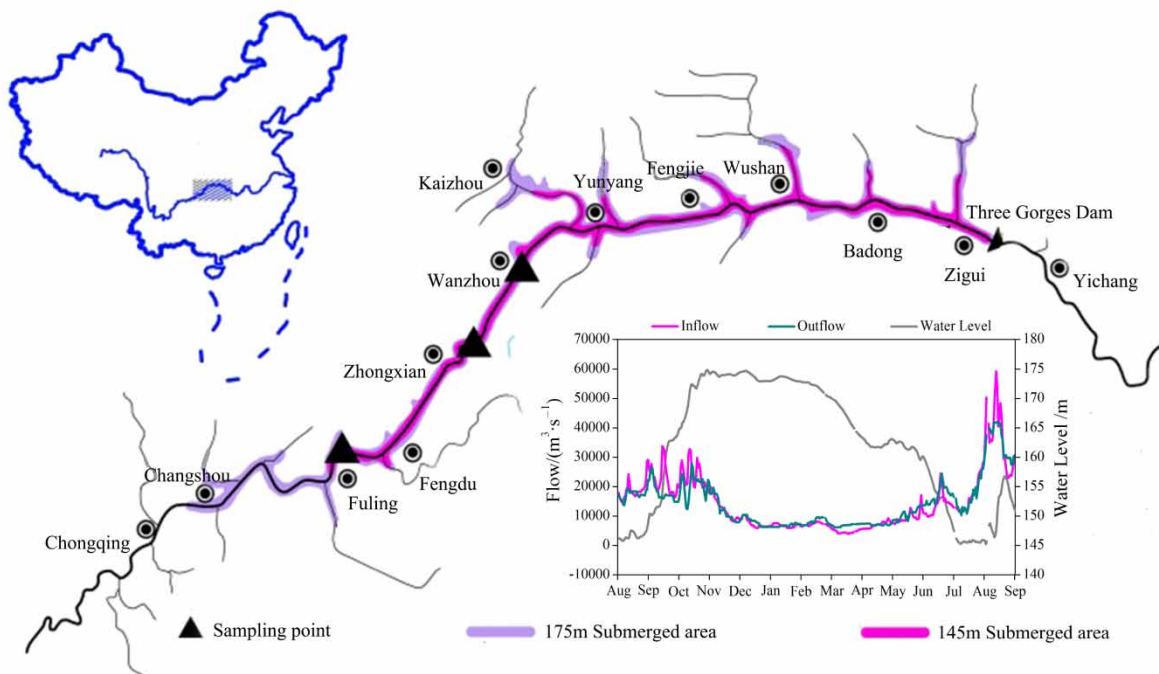
**Figure 1** | Concentration distribution of CH<sub>4</sub> in water of the Three Gorges Reservoir (Li *et al.* 2020).

turbidity of the reservoir (Alizadeh *et al.* 2018). From August 2017 to August 2018, the water level changes during the study period of the Three Gorges Reservoir (Figure 2) (source: <https://www.ctg.com.cn/sxjt/sqqk/index.html>).

## 2.2. Sampling and analysis

From August 2017 to August 2018, this study conducted monthly monitoring and analysis on three sampling sites for a period of 12 months. The monitoring time of all sampling sites was controlled from 20th to 22nd of each month. Massive amounts of GHG gathering research will be at 9:00–11:00 (Wu 2016) as the representative of the flux of the day time, Wu (2012) to choose 9:00–11:00 in the morning in the Three Gorges Reservoir with monitoring, Zhao *et al.* (2011) monitored GHG emissions in the Xiangxi River Basin of the Three Gorges Reservoir from 9:00 to 14:00. Due to spatiotemporal differences in GHG fluxes, GHG fluxes at the same point and at different times will also change. To obtain more accurate data, the sampling time of this study is as much as possible controlled at 9:00–11.00 am. The sampling tool is a 5-L water sampler to collect the 0.5 m surface water of the above three sections, respectively (Gong *et al.* 2019; Liu *et al.* 2019), the water samples will be sent back to the laboratory immediately on the same day, all the test and analysis work will be completed within 48 h, and the relevant water environmental indicators and the characteristics of GHG flux at the water–air interface will be monitored simultaneously.

At present, there are two commonly used methods for monitoring the gas emission flux at the water–gas interface: one is static chamber method – gas chromatography; the other is the combination of headspace balance method – gas chromatography and thin boundary layer (TBL) model estimation method. Static box-gas chromatography is usually only able to obtain flux data at points, with high labor intensity and high cost of gas analysis, which makes it unsuitable for large area and long-term observation. Therefore, static box-gas chromatography is only suitable for the observation of static water bodies (Tremblay *et al.* 2005). However, the model estimation method is most commonly used in field monitoring due to its simplicity, flexibility and easy operation (Qin *et al.* 2017b). Considering the strong fluidity of the water body, the long distance between sampling points, the long duration of monitoring and the poor geographical and climatic conditions of the points, the feasibility of the static box method is weak. In addition, due to the disturbance of water outside the box, the flux data obtained by the static box method is relatively discrete. The comparative experiment shows that the flux results obtained by the TBL estimation method are generally more stable than the former method (Yao *et al.* 2015). Therefore, in this study, the GHG flux at the water–gas interface in the middle section of the Three Gorges Reservoir was obtained by using the headspace balance TBL model estimation method.



**Figure 2** | Sampling point and flow water-level map of the Three Gorges Reservoir; sample list is showed in Table 1.

**Table 1** | Basic information of the sampling sites

| Sample sites | Location                      |
|--------------|-------------------------------|
| FL           | 29°48'00"N, 107°27'00"E       |
| ZX           | 30°24'57.63"N, 108°12'40.86"E |
| WZ           | 30°46'26.66"N, 108°24'46.74"E |

At the sampling place, the headspace bottle is placed in a water collector filled with water and sealed in a completely submerged state. It is confirmed that there is no bubble in the headspace, put it upside down and send it back to the laboratory for analysis. The bottle is sealed in a completely submerged state. At the same time, it is confirmed that there is no bubble in the head empty bottle, it is placed upside down, and sent back to the laboratory for analysis. After the headspace bottle filled with water sample is taken out, two injection needles are inserted into the headspace bottle at the same time at room temperature, to the headspace bottle injection of 5 mL of high-purity nitrogen mechanical shock after 20 min, after waiting for two phase of water-gas balance to extract the upper air bottle, into an agilent7820A gas chromatograph, after TDX-01 chromatographic column separation in an Ni catalyst with a FID detector test, after obtaining the CO<sub>2</sub> and CH<sub>4</sub> concentration, Equation (1) is plugged in, to get water samples under test of CO<sub>2</sub> and CH<sub>4</sub> partial pressure (Kolb & Ettre 2006), the specific method to calculate the GHG see literature (Qin *et al.* 2017b):

$$P(\text{Gas}) = \frac{(P_{\text{final}} \times K_{\text{equilibrium}}) + \left( \left( \text{HS}/S \right) \times \frac{(P_{\text{final}} - P_{\text{initial}})}{V_m} \right)}{K_{\text{sample}}} \quad (1)$$

where  $P(\text{Gas})$  is the gas partial pressure in the water sample to be measured,  $\mu\text{atm}$ ;  $P_{\text{initial}}$  and  $P_{\text{final}}$  are the partial pressure of the gas to be measured in the air above the bottle before and after equilibrium,  $\mu\text{atm}$ ; HS/S is the volume ratio of the gas in the bottle to the water;  $K_{\text{sample}}$  and  $K_{\text{equilibrium}}$  are the corresponding solubility of the gas to be measured at the time of sampling and water temperature in the bottle before sample analysis, respectively,  $\text{mol}\cdot\text{L}^{-1}\cdot\text{atm}^{-1}$ ;  $V_m$  is the mole of the gas volume,  $\text{mol}\cdot\text{L}^{-1}$ ; the unit of partial pressure is uniformly converted into Pa,  $1 \mu\text{atm} = 0.101325 \text{ Pa}$ .

According to Fick's law (Rantakari *et al.* 2015), for freshwater, the gas exchange flux at the water-air interface can be calculated by the following equation:

$$\text{Flux} = k_x(C_{\text{water}} - C_{\text{air}}) \quad (2)$$

where Flux is the GHG diffusion flux,  $\text{mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ;  $k_x$  is the gas exchange coefficient,  $\text{cm}\cdot\text{h}^{-1}$ ;  $C_{\text{water}}$  is the GHG concentration in water,  $\mu\text{mol}\cdot\text{L}^{-1}$ ;  $C_{\text{air}}$  is the GHG concentration in the atmosphere,  $\mu\text{mol}\cdot\text{L}^{-1}$ .

For the gas exchange coefficient  $k_x$ , the empirical model formula (Rik 1992) is adopted, the instantaneous wind speed ( $U_1$ ) above the water surface measured in the field is used and the wind speed at 10 m above the water surface ( $U_{10}$ ) is converted according to the following equation:

$$U_{10} = 1.22 \times U_1 \quad (3)$$

At the sampling place, the water temperature and DO were measured by YSI® Pro ODO (the accuracy of 0.1 °C, 0.01  $\text{mg}\cdot\text{L}^{-1}$ , respectively); the pH value was measured by the multi-parameter water quality analyzer HACH® MS5 (the accuracy of 0.01); the instantaneous wind speed above the water surface was measured by AR-826 anemometer (the accuracy was 0.01  $\text{m}\cdot\text{s}^{-1}$ ); the temperature was measured by the thermometer on site (the accuracy was 0.1 °C); the atmospheric pressure is measured by a barometer on site (with an accuracy of 0.01 bpa); the field measuring instruments have passed the standard calibration procedures before sampling; and at the end of the sampling field test, the water samples from each section are taken back to the laboratory to complete the analysis and testing of other indicators, including total organic carbon (TOC) and total nitrogen (TN), within 48 h (Yang & Lv 2016). Inside the TOC was determined by Whatman GF/F glass fiber

filter membrane, which was dried at 450 °C for 4 h, and the TN was determined by alkaline potassium persulfate oxidation ultraviolet spectrophotometry.

### 2.3. Data analysis method

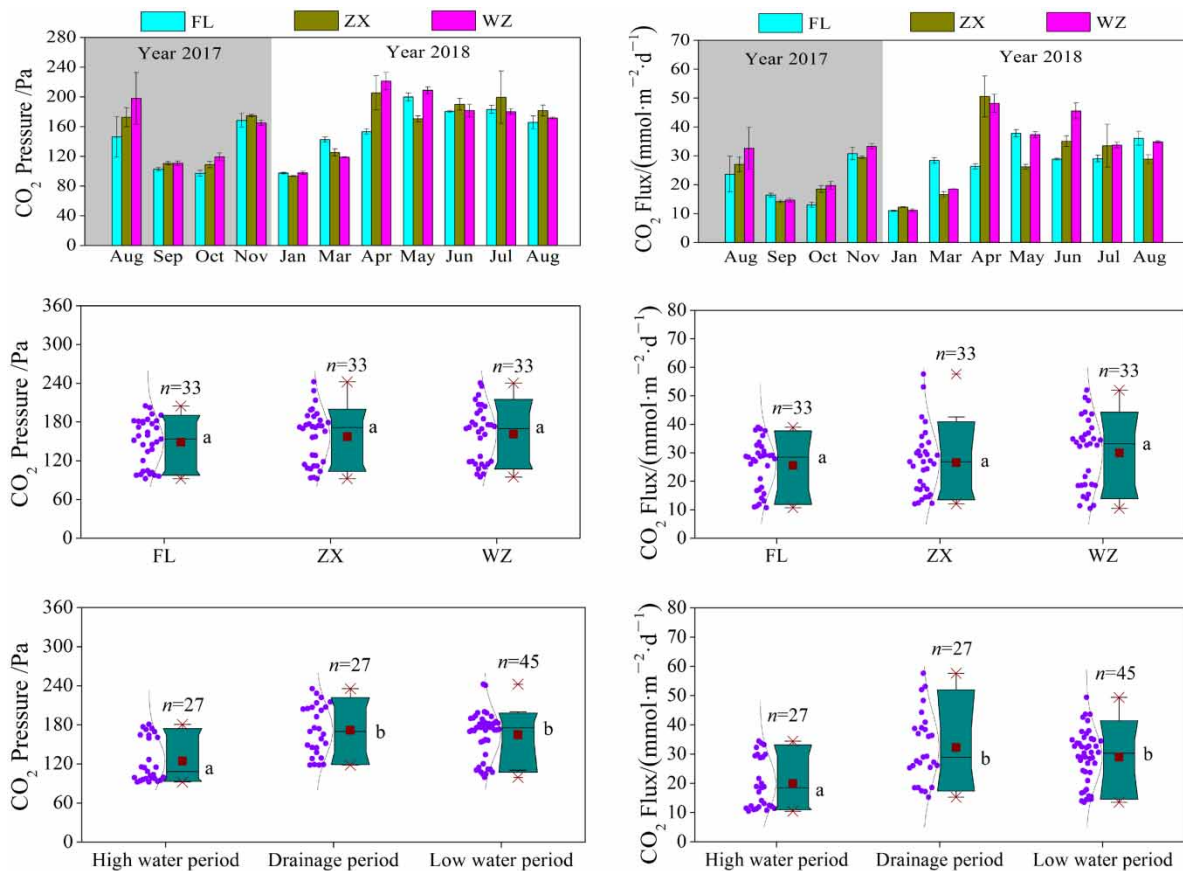
In this study, all the experimental and calculated data were entered into SPSS® or Origin® for statistical analysis, and one-way ANOVA was used to explain the differences of the data in different periods and spaces. Spearman's correlation analysis was used to analyze the correlation between CO<sub>2</sub> and CH<sub>4</sub> concentration and various environmental factors (DO, TOC and TN), or Origin was used for linear regression analysis, indicating the linear correlation between data changes. There are also Pearson's correlation analysis methods for correlation analysis, but Spearman's analysis is more suitable and more accurate because GHGs belong to non-normal distribution. Pearson's correlation analysis is more suitable for data with normal distribution (Dash *et al.* 2019).

## 3. RESULTS AND DISCUSSION

### 3.1. Results

#### 3.1.1. Changes of CO<sub>2</sub> partial pressure in water and flux at the water-air interface

As shown in Figure 3, the average annual partial pressure of CO<sub>2</sub> in the middle of the Three Gorges Reservoir is  $155.8 \pm 38.2$  Pa. The average annual partial pressure of CO<sub>2</sub> at FL point was  $148.8 \pm 34.3$  Pa, reaching its maximum value in May of the next year; the average annual partial pressure of CO<sub>2</sub> at ZX point was  $157.5 \pm 38.3$  Pa, reaching its maximum value in April of the next year; and the average annual partial pressure of CO<sub>2</sub> at WZ point was  $161.2 \pm 40.7$  Pa, reaching its maximum value in April of the next year. The monthly changes of FL, ZX and WZ were similar. There was no significant difference in CO<sub>2</sub> partial pressure at each point (ANOVA,  $P > 0.05$ ). The mean partial pressures of CO<sub>2</sub> in the high-water



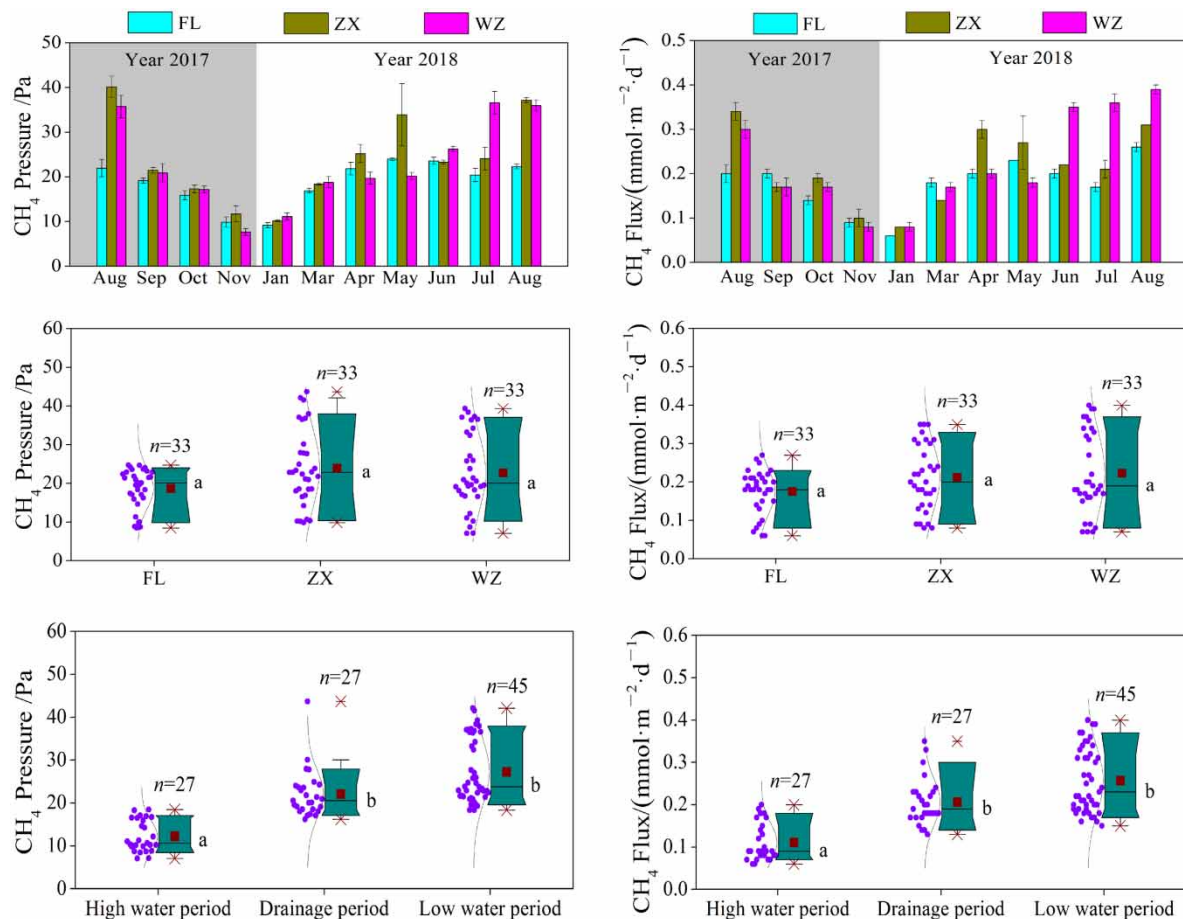
**Figure 3** | CO<sub>2</sub> partial pressure of water and CO<sub>2</sub> flux at the water-air interface. (Note: a and b indicate differences between groups.)

stage, drainage stage and low-water stage were  $124.6 \pm 32.5$ ,  $171.8 \pm 36.3$  and  $165.1 \pm 31.1$  Pa, respectively. Under different operating conditions, the partial pressure of  $\text{CO}_2$  changed significantly (ANOVA,  $P = 0.03 < 0.05$ ), and the drainage stage and the low-water stage were significantly higher than the high-water stage.

The average annual  $\text{CO}_2$  flux in the middle of Three Gorges Reservoir is  $27.4 \pm 10.8 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The average annual  $\text{CO}_2$  flux at FL point was  $25.6 \pm 8.6 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in May of the following year; the average annual  $\text{CO}_2$  flux at ZX point was  $26.6 \pm 11.0 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in April of the next year; and the average annual  $\text{CO}_2$  flux at WZ point was  $30.0 \pm 11.9 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in April of the next year. The annual  $\text{CO}_2$  flux is positive, which is the ‘source’ of atmospheric  $\text{CO}_2$ . The monthly changes of FL, ZX and WZ were similar. There was no significant difference in  $\text{CO}_2$  flux at each point (ANOVA,  $P > 0.05$ ). The average  $\text{CO}_2$  fluxes were  $19.9 \pm 8.5$ ,  $32.2 \pm 11.6$  and  $28.9 \pm 9.0 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  in high-water stage, discharge period and low-water stage, respectively. Under the conditions of different operating periods,  $\text{CO}_2$  flux changes significantly (ANOVA,  $P = 0.03 < 0.05$ ), and the drainage stage and low-water stage are significantly higher than the high-water stage, which are similar laws to the partial pressure of  $\text{CO}_2$ .

### 3.1.2. Changes of $\text{CH}_4$ partial pressure in water and flux at the water–air interface

As shown in Figure 4, the average annual  $\text{CH}_4$  partial pressure in the middle period of the Three Gorges Reservoir is  $21.7 \pm 8.4$  Pa. The average annual  $\text{CH}_4$  partial pressure at FL point was  $18.6 \pm 4.9$  Pa, reaching its maximum value in May of the next year; the average annual  $\text{CH}_4$  partial pressure at ZX point was  $23.8 \pm 9.3$  Pa, reaching its maximum value in August; and the average annual  $\text{CH}_4$  partial pressure at WZ point was  $22.7 \pm 9.4$  Pa, reaching its maximum value in August of the next year. There was no statistical difference in  $\text{CH}_4$  partial pressure at each point (ANOVA,  $P > 0.05$ ). The mean  $\text{CH}_4$  partial



**Figure 4** |  $\text{CH}_4$  partial pressure of water and  $\text{CH}_4$  flux at the water–air interface. (Note: a and b indicate differences between groups.)

pressure in high-water stage, drainage stage and low-water stage was  $12.2 \pm 3.4$ ,  $22.0 \pm 4.8$  and  $27.2 \pm 7.1$  Pa, respectively. Under different operating conditions, there were significant differences in  $\text{CH}_4$  partial pressure (ANOVA,  $P = 0.000 < 0.05$ ), and the low-water stage and drainage stage were significantly higher than the high-water stage.

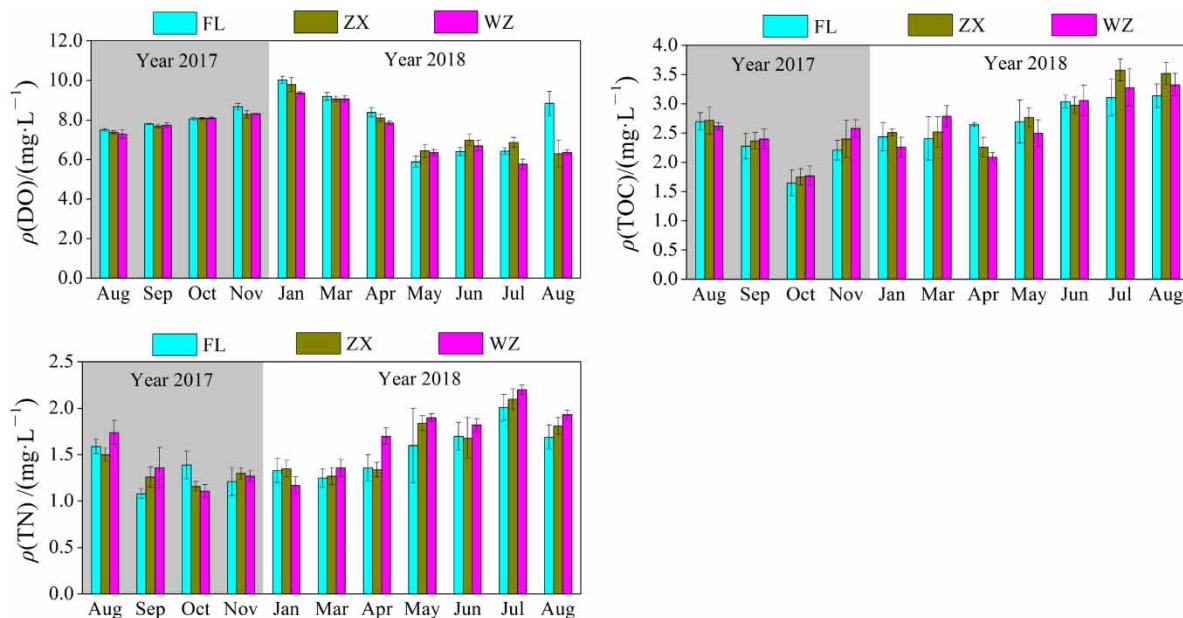
The average annual  $\text{CH}_4$  flux in the middle of Three Gorges Reservoir is  $0.20 \pm 0.09 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The average annual  $\text{CH}_4$  flux at FL point was  $0.17 \pm 0.05 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in August of the next year; the annual average  $\text{CH}_4$  flux at ZX point was  $0.21 \pm 0.08 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in August; and the annual average  $\text{CH}_4$  flux at WZ point was  $0.22 \pm 0.10 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , reaching its maximum value in August of the next year. The annual  $\text{CH}_4$  fluxes of FL, ZX and WZ are all positive and are the 'source' of atmospheric  $\text{CH}_4$ . There was no statistical difference in  $\text{CH}_4$  flux at each point (ANOVA,  $P > 0.05$ ). The average  $\text{CH}_4$  flux in high-water stage, drainage stage and low-water stage is  $0.11 \pm 0.04$ ,  $0.20 \pm 0.04$  and  $0.25 \pm 0.07 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , respectively. In different operating periods,  $\text{CH}_4$  fluxes were significantly different (ANOVA,  $P = 0.000 < 0.05$ ), the low-water level and drainage stage were significantly higher than the high-water-level stage and the partial pressure of  $\text{CH}_4$  fluxes and  $\text{CH}_4$  presented similar laws.

### 3.1.3. Changes in physical and chemical indices of water quality

The change rules of DO, TOC and TN in the middle of the Three Gorges Reservoir at different points and different operating periods are shown in Figure 5. At different points, DO, TOC and TN showed similar variation patterns, in which DO reached a higher value the next January, and TOC and TN both reached a higher value the next July. There was no significant difference in annual DO, TOC and TN values at FL, ZX and WZ. In different operation periods, DO reaches the highest value in the high-water stage and the lowest value in the low-water stage. Both TOC and TN reached the lowest value in the high-water stage and the highest value in the low-water stage.

### 3.1.4. Main influencing factors of GHG flux at the water-gas interface

The correlation between the partial pressure of GHG and the GHG flux at the water-gas interface and environmental indicators in the middle section of the Three Gorges Reservoir is shown in Table 2. Correlation analysis shows that the partial pressures of  $\text{CO}_2$ ,  $\text{CH}_4$  and fluxes of  $\text{CO}_2$  and  $\text{CH}_4$  at the water-gas interface in the middle part of the Three Gorges Reservoir are significantly positively correlated with TOC and TN, and extremely significantly negatively correlated with DO (Table 2). Correlation analysis of DO, TOC, TN and  $\text{CH}_4$  fluxes is shown in Figure 6.



**Figure 5** | Distribution of DO, TOC and TN in the water body of the middle part of the Three Gorges Reservoir.

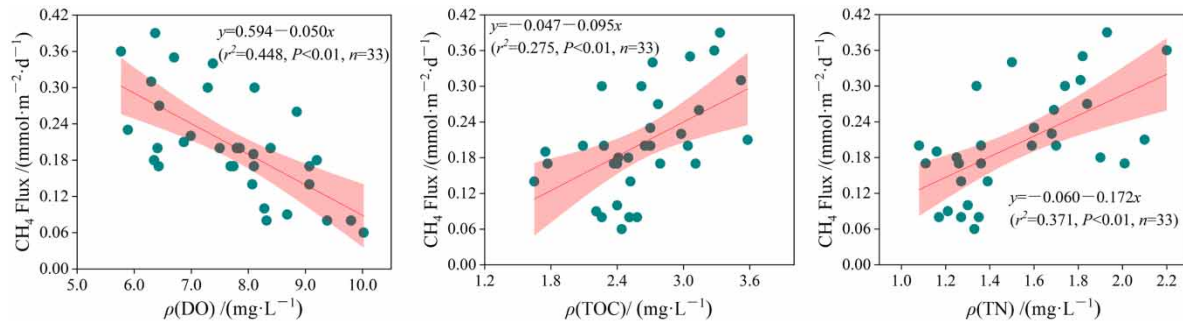


**Table 2** | Correlation between CO<sub>2</sub> and CH<sub>4</sub> partial pressure and flux and various physical and chemical indicators

|     | CO <sub>2</sub> partial pressure | CH <sub>4</sub> partial pressure | CO <sub>2</sub> flux | CH <sub>4</sub> flux |
|-----|----------------------------------|----------------------------------|----------------------|----------------------|
| DO  | -0.632**                         | -0.752**                         | -0.481**             | -0.682**             |
| TOC | 0.425*                           | 0.658**                          | 0.380*               | 0.572**              |
| TN  | 0.662**                          | 0.699**                          | 0.549**              | 0.621**              |

\*indicates  $P \leq 0.05$ , indicating significant correlation.

\*\*indicates  $P \leq 0.01$ , indicating extremely significant correlation.

**Figure 6** | Correlation between TOC, TN and CH<sub>4</sub> fluxes.

## 3.2. Discussion

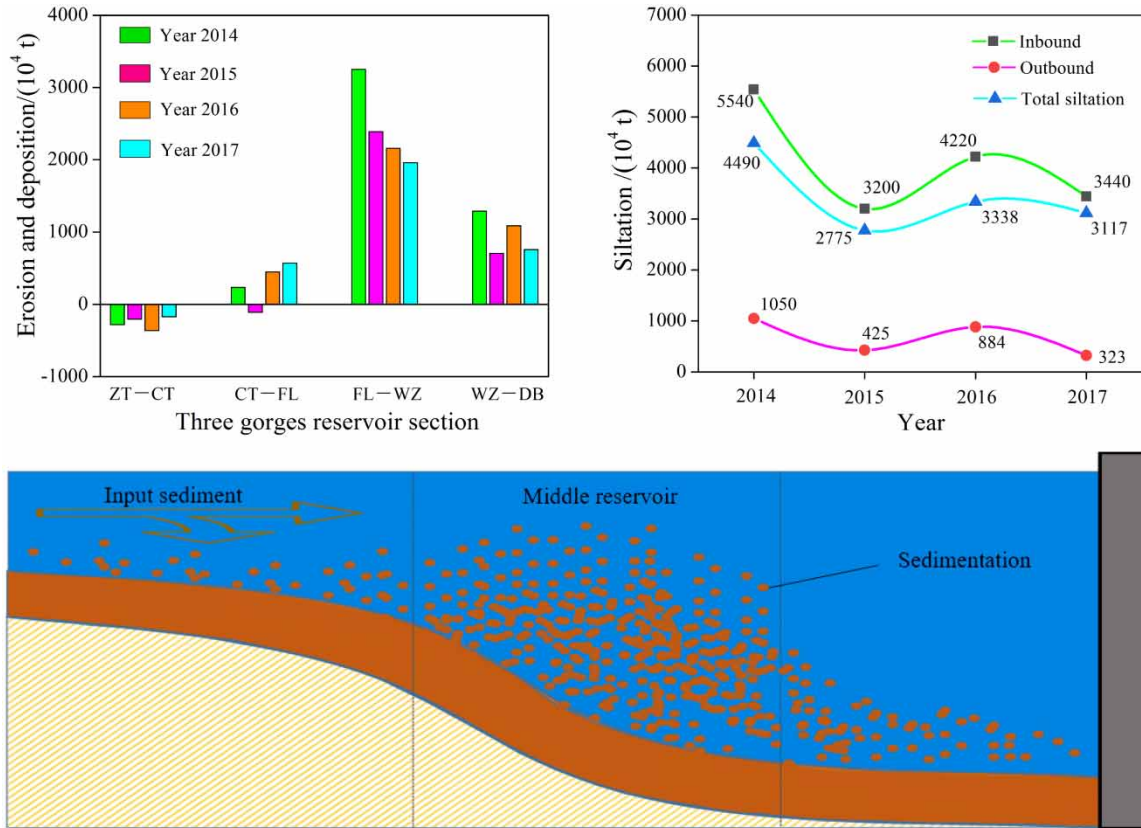
### 3.2.1. Sediment deposition and its effects

More than 90% of suspended sediment in the Three Gorges Reservoir has been deposited in the perennial backwater area and in the ‘point’ form in the wide valley and curved reach in the past 10 years (Hu *et al.* 2013). Sediment deposition is related to river plane morphology, average flow rate, hydraulic residence time, sand-carrying capacity and relative sediment concentration (Li *et al.* 2016b). Sand-carrying capacity is greater than sediment concentration, resulting in scour, and conversely, sedimentation (Li *et al.* 2015b).

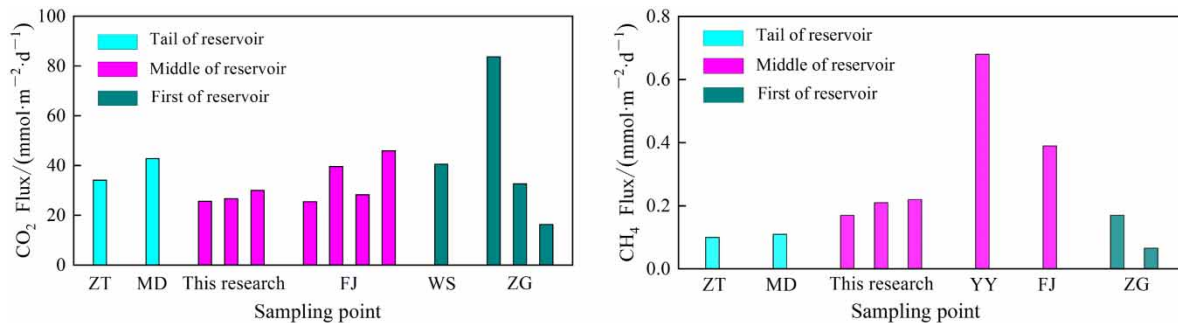
As shown in Figure 7, in the Zhutuo–Cuntan section of the tail river of the reservoir, the hydraulic retention time is short, the flow rate is large and the sand-carrying capacity is greater than the sediment content. Therefore, scour is dominant in this section. The Cuntan–Fuling section belongs to the fluctuating backwater area, and the sand-carrying capacity is weakened. In the Fuling–Wanzhou section, the perennial backwater area of the reservoir, the average flow velocity decreased, the hydraulic retention time increased and the sand-carrying capacity greatly weakened. In addition, the overall shape of the channel was ‘S’, resulting in a large number of sediment deposition, which was the most serious section in the reservoir area (Hu *et al.* 2013; Liu *et al.* 2014; Li *et al.* 2015b, 2016b). In the perennial backwater area of the first reservoir, affected by the impoundment of the Three Gorges Reservoir, the change of water level is basically synchronous with the water level in front of the Three Gorges Dam, the sand-carrying capacity is greatly reduced, the sand-carrying capacity is less than the sediment concentration and the sediment deposition is large.

According to the research (Yang *et al.* 2016), the average cumulative sediment deposition in the trunk stream of the Three Gorges Reservoir area was  $3,430 \pm 644.0$  tons. In the Zhutuo–Cuntan section of the reservoir end river area, the sediment caused scour and the sediment flushing amount was  $255.2 \pm 734,000$  tons. In the Cuntan–Fuling section of fluctuating backwater area, there was a small amount of silt deposition, which was  $285 \pm 2.588$  million tons. In the perennial backwater area of the first part of the reservoir, the sediment state is deposition, the deposition amount is  $960 \pm 2.485$  million tons, but the deposition is lower than that in the middle part of the reservoir. As shown in Figure 7, from 2014 to 2017, due to the comprehensive influence of water conservancy projects’ sand blocking, temporal and spatial distribution changes of rainfall, sand mining in river channels and other factors, the sediment transport quantity decreased significantly and the deposition rate slowed down somewhat.

The high CH<sub>4</sub> flux in the middle section of the Three Gorges Reservoir (Fuling–Wanzhou section) is correlated to the spatial distribution and deposition of sediment (Maeck *et al.* 2013). Compared to published literature, as shown in Figure 8,



**Figure 7** | Schematic diagram of sedimentation, sedimentation and sedimentation in the Three Gorges Reservoir.



**Figure 8** | Comparison of CO<sub>2</sub> and CH<sub>4</sub> fluxes in the first, middle and tail sections of the Three Gorges Reservoir area (Yang *et al.* 2012; Yang 2017; Yao *et al.* 2012; Wei *et al.* 2013; Li *et al.* 2014; Fu *et al.* 2016; Zhao 2016).

CO<sub>2</sub> and CH<sub>4</sub> fluxes at the water–gas interface in the Three Gorges Reservoir are  $27.4 \pm 10.8$  and  $0.20 \pm 0.09$  mmol·m<sup>-2</sup>·d<sup>-1</sup>, respectively. Compared to the published literature, as shown in Figure 7, CO<sub>2</sub> and CH<sub>4</sub> fluxes at the water–gas interface in the reservoir area compare with the reservoir head of the Three Gorges Reservoir (Yang *et al.* 2012; Yao *et al.* 2012; Zhao 2016) and the tail of the reservoir (Yang 2017). There was no significant difference in CO<sub>2</sub> flux, but the CH<sub>4</sub> flux in the middle of the reservoir was higher than the flux at the head and tail of the reservoir. The above relationship characteristics are also supported in the correlation analysis of physical and chemical indexes of water quality. As more sediment deposition in the middle section of the reservoir leads to a high CH<sub>4</sub> flux, it is suggested that sediment deposition can be removed by dredging to slow down the CH<sub>4</sub> emission.

### 3.2.2. Influence of DO

DO has an important effect on the discharge of CH<sub>4</sub> in the reservoir (Schrier-Uijl *et al.* 2011). The content of DO in water determines the process of organic matter degradation in water. Usually, in the process of the carbon cycle, the sediments CH<sub>4</sub> produced in anaerobic conditions, through pore water into the sediment and water interface, and then enter the water upward diffusion, part of CH<sub>4</sub> in the process of diffusion was oxidized to CO<sub>2</sub>, and during the period of water diffusion, CH<sub>4</sub> is further oxidation, only a small amount of CH<sub>4</sub> discharge into the atmosphere (Chen *et al.* 2009). Studies have shown that CO<sub>2</sub> flux is significantly negatively correlated with DO in water (Wang *et al.* 2012). Similarly, the concentration of CH<sub>4</sub> is significantly negatively correlated with the concentration of DO in water (Wang *et al.* 2009), indicating that the higher the DO in water, the more CH<sub>4</sub> is oxidized. Biological activities in the water can affect DO concentration in the water, that is, phytoplankton can increase DO in the water through photosynthesis; on the contrary, respiration of aquatic plants and decomposition of organic matter can consume DO in the water. DO produced by photosynthesis also affects GHG production and emissions (Arend *et al.* 2011). In this study, there was a significant negative correlation between DO and CH<sub>4</sub> flux (Furlanetto *et al.* 2012), and CH<sub>4</sub> flux increased with the decrease of DO. In the middle part of the Three Gorges Reservoir, the DO in the low-water-level period is significantly lower than that in the high-water-level period, and the content of DO in the water body decreases, which results in the CH<sub>4</sub> produced by the sediment at the bottom, cannot be fully oxidized during the rising process. Therefore, the content of CH<sub>4</sub> in the low-water-level period is higher than that in the high-water-level period.

### 3.2.3. Influence of TOC

The decomposition or mineralization of organic matter in lakes or reservoirs is essential for the production of CO<sub>2</sub> and CH<sub>4</sub>. The main source of organic carbon in water is related to the system biological residues and exogenous input. Organic carbon is the direct carbon source of methanogenic bacteria decomposition and CO<sub>2</sub> production, and its content has an important influence on the production and emission of CO<sub>2</sub> and CH<sub>4</sub> in the reservoir (Sobek *et al.* 2012). Under anaerobic conditions, the decomposition of most organic matter at the bottom of the reservoir promotes the formation of CH<sub>4</sub> (Roland *et al.* 2017). DOC affects the CO<sub>2</sub> concentration of water to some extent, and this effect varies depending on the concentration, source and geographic location of the DOC (Zhao *et al.* 2008). During the silting process in the middle section of the Three Gorges Reservoir, a large amount of organic carbon is carried, providing a carbon source for methanogens at the bottom. Under the action of the methanogens and the DO in the water column, CO<sub>2</sub> and CH<sub>4</sub> are generated and released into the atmosphere (Jacinthe *et al.* 2012). When the content of organic matter in sediments and water is high, the number of methanogenic bacteria increases, and the increase of CH<sub>4</sub> production leads to the increase of emission flux. In this study, TOC in water was significantly positively correlated with CH<sub>4</sub> flux (Martinez-Cruz *et al.* 2017). As shown in Figure 6, CH<sub>4</sub> flux increased with the increase of TOC (Li *et al.* 2018a). In northern reservoirs with high DOC concentrations, CO<sub>2</sub> concentrations are usually strongly correlated with DOC concentrations, and aquatic metabolism may be the main driver of CO<sub>2</sub> oversaturation (Saidi & Koschorreck 2017).

### 3.2.4. Influence of TN

TN is often used to indicate the degree of water pollution by nutrients. The increase of water nutrient level is conducive to CH<sub>4</sub> production, mainly due to the increase of nutrient elements and organic carbon content transported from water to sediment, methanogens get more substrates and nutrients, dissolved nitrogen in rivers is a potential contributor to GHG emissions (Qu *et al.* 2017). Previous studies have shown that CH<sub>4</sub> production in lake and reservoir sediments increases with the increase of lake nutrient level (Davidson *et al.* 2015). On the one hand, phytoplankton will absorb nitrogen nutrients in the water under photosynthesis, resulting in a decrease in CO<sub>2</sub> concentration in the water, and atmospheric CO<sub>2</sub> will enter the water (Jiang *et al.* 2012). On the other hand, the increase of nitrogen concentration in water will improve primary productivity, increase water respiration and lead to the increase of CO<sub>2</sub> concentration in water. Thus, the transition leads to changes in the CO<sub>2</sub> 'source' and 'sink' processes between photosynthesis and respiration. The increase of nitrogen nutrients in the reservoir ecosystem (Morgane *et al.* 2019) can stimulate primary production and carbon sequestration (Knoll *et al.* 2013). The high concentration of nutrients can provide substrates for aquatic organisms to accelerate respiration, reduce DO in water and form an anaerobic environment, which is conducive to the generation and discharge of CH<sub>4</sub> in water. In this study, TN and CH<sub>4</sub> flux showed a very significant positive correlation, as shown in Figure 6, and CH<sub>4</sub> flux increased with the increase of TN content. The results show that the TN content in the water body is the environmental factor affecting the seasonal variation of CH<sub>4</sub> emission.

#### 4. CONCLUSIONS

1. The average annual CO<sub>2</sub> flux in the middle of the Three Gorges Reservoir is  $27.4 \pm 10.8 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . There was no statistical difference in the CO<sub>2</sub> flux between the measured points. The CO<sub>2</sub> flux during the drainage period and the low-water-level period was significantly higher than that during the high-water-level period, and reached the lowest value during the high-water-level period. The average annual CH<sub>4</sub> flux in the middle of the reservoir area is  $0.20 \pm 0.09 \text{ mmol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . There is no statistical difference in CH<sub>4</sub> flux between the measured points, and CH<sub>4</sub> flux in the drainage stage and the low-water stage is significantly higher than that in the high-water stage.
2. In the middle part of the Three Gorges Reservoir, there is a certain correlation between the spatial distribution and deposition amount of sediment and the emission of carbon flux of GHG. With the increase of sediment deposition, the emission of carbon flux of GHG also tends to increase. The partial pressure and flux of CO<sub>2</sub> and CH<sub>4</sub> in water were significantly positively correlated with TOC and TN and significantly negatively correlated with DO.
3. The high value of CH<sub>4</sub> flux at the water–gas interface in the middle section of the reservoir is related to the spatial distribution and deposition of sediment. In future studies, the GHG emission in the middle section of the reservoir should be further studied in combination with the physical and chemical characteristics of sediment.
4. This research focuses on the macroscopic exploration of the middle section of the Three Gorges Reservoir area (Wanzhou–Fuling section). However, there is still a lack of microscopic observations in this field, such as microbes, phytoplankton and zooplankton related to GHGs, which need to be further studied.
5. The research is based on long-term observation and is heavily influenced by field conditions. Further research can be carried out in the area of laboratory simulation experiment.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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