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Properties of CO_2 and CH_4 fluxes across water–air interface at the Three Gorges Reservoir, the mainstream of Yangtze River from Zhutuo to Wanzhou, China

Yu Qin[a,*], Zhengmian Liu[a], Zhe Li[b], Boxiao Yang[a]

[a]*Key Laboratory of Hydraulic and Waterway Engineering of the Ministry of Education, Chongqing Jiaotong University, Chongqing, 400074, China, email: qinyu54001@163.com (Y. Qin)*
[b]*Key Laboratory of Reservoir Environment, Chongqing Institute of Green and Intelligence Technology, Chinese Academy of Sciences, Chongqing 400714, China*

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ABSTRACT

The Three Gorges Reservoir is one of the most important hydraulic projects of the Yangtze River in China, so it is vital to monitor p (CO_2), p(CH_4) and CO_2, CH_4 fluxes in water–air interface in a long term. Five monitoring sites (Zhutuo, Mudong, Fulin, Zhongxian and Wanzhou) were chosen from the mainstreams of the Yangtze River in this study. CO_2 and CH_4 were monitored monthly for period of 1 year from May of 2016. Results indicated that in the mainstream of the Yangtze River p (CO_2) and CO_2 fluxes were 1,246.99–4,495.20 μatm and (41.29 ± 4.46) mmol m^{-2} d^{-1}, respectively. The p(CH_4) and CH_4 fluxes were 21.63–588.28 μatm and (0.168 ± 0.028) mmol m^{-2} d^{-1}. The p(CH_4) had a positive correlation with water temperature, while negative correlation with dissolved oxygen (DO). The p (CO_2) also had a significant positive correlation with water temperature, but negative correlation with conductivity, DO, pH and wind speed. Water–air CH_4 and CO_2 fluxes mainly impacted by p(CH_4), p (CO_2), water temperature, DO, conductivity and pH.

Keywords: Three Gorges Reservoir; CO_2; CH_4; Fluxes of air–water interface; Environmental factors

1. Introduction

There is an argument on the role of reservoirs and the influence of greenhouse gas emitted from reservoirs. According to previous research revealed, some biogenic elements such as carbon, nitrogen, phosphorus, silicon are unstable in the river-reservoir system [1–4]. Reservoirs have significantly altered flux and physical composition of biogenic substances in river system, and even have impacts to the balance of global substance flow and recycling and ocean ecology system. With flooding of terrestrial ecosystems by the reservoirs, the chemistry of flood soils will be altered that causes the new increased input of carbon and the nutrients to water column [5]. The carbon recycling in the interior of the reservoir is a sophisticated process, where biogenic elements go through chemical and biological changes, causing significant changes of substances at different depth of river, while precipitation of rot terrestrial plants and submergence of flooded soil during impoundment will increase the nutrient loading in reservoirs [6,7]. Additionally, it is common that impoundment of reservoir results in a degradation of flooded soil organic matter and plant biomass as well as a decrease of transport fluxes [8]. Researchers have studied this field by different sites. Tremblay [24] measured the fluxes of over 280 Canadian reservoirs, rivers and natural lakes and found that younger reservoirs emitted higher greenhouse gas. A previous scholar assessed the fluxes of reservoirs and compared with characters of fluxes of reservoirs in different climate zones [9]. Other scholars believe that the location of climate zone and the reservoir ages are the key to affect the level of gross carbon flux worldwide [10].

The Yangtze River (6,300 km) is the largest river in Asia and the third largest river in the world. The Three Gorges

* Corresponding author.

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Dam, at the mainstream of the Yangtze River, causes a decline of water flow speed and creates a large eroded area by impoundment. The carbon input from riversides increases, leading to a variation of the ecological environment at the two sides of the river as well as at the backwater of the reservoir [11,12]. However, the study about the emission of greenhouse gases from hydro-reservoirs just commenced in China. So it lacks data which can support further analysis and research. This paper presents the data investigated from sites at the upstream of the Three Gorges Reservoir and aims to provide an essential record of primary environmental factors data in different seasons and a systematical methodology for the further study [13–17].

2. Materials and methods

2.1. Study site

Based on hydrological characteristics and geography of The Three Gorges Reservoir, in this study, five monitoring points are chosen to collect samples, which are typical sampling spots at The Three Gorges Reservoir. Additionally, ZT and MD are located in a non-annual backwater, while FL, ZX and WZ are in the area of an annual backwater. The location of sampling spots is shown in Table 1 and Fig. 1.

Fig. 1. Water system of sampling sites.

2.2. Sampling methodology

Water samples were taken monthly at the five monitoring points of the Yangtze River from May 2016 to February 2017. The data of sampling ranged from 15th to 20th in every month during a period from 10 am to 3 pm. Water was taken under 0.5 m of water by 5 L water collecting equipment, in which water samples were sealed in headspace glass vials at a submerged state and then stored with cryopreservation immediately. Within 48 h, all experiments must be finished.

Data, such as water temperature, dissolved oxygen (DO), pH, conduction, temperature, atmospheric pressure and wind speed, were detected on site. Chlorophyll-a (Chl-a) was obtained by spectrophotometry. Based on a study, the concentration of CO_2 and CH_4 in water was detected by static headspace gas chromatography with a flame ionization detector [18]. The two-layer film model derived from Fick's first law and promoted by a group of scholars was used in this paper to measure the flux of CO_2 and CH_4 across the water–air interface [19]. It is shown as below:

$$\text{Flux} = K_x\left(C_1 - C_2\right) \tag{1}$$

K_x is an exchange constant for the gas and liquid phases. C_1 is the concentration of gas (μatm min^{-1}) obtained before by static headspace gas chromatography, while C_2 is the equilibrium concentration of gas (μatm min^{-1}) in water.

Table 1
Location of sampling spots

Sampling spot	Latitude and longitude
Zhutong (ZT)	N29°1′00″ E105°51′00″
Mudong (MD)	N30°30′15″ E106°02′48″
Fulin (FL)	N29°48′00″ E107°27′00″
Zongxian (ZX)	N30°24′57.63″ E108°12′40.86″
Wanzhou (WZ)	N30°46′26.66″ E108°24′46.74″

2.3. Data process and analysis

All data were statistically analyzed through SPSS® and Origin® for correlation analysis between gas fluxes and concentrations. Other environmental indicators, such as pH, DO, water temperature, Ch-a and so on, were analyzed to reveal the potential influencing factors.

3. Result

3.1. Partial pressures of CO_2 and CH_4 analysis

The results of the partial pressure of CO_2 are shown as Fig. 2.

Through a whole monitoring year, at three of all sampling points, MD, FL and WZ, the p(CO_2) of those had a similar fluctuation and reached a peak on July 2016 (MD: 3,959.48 μatm, FL: 4,216.47 μatm and WZ: 3,724.60 μatm). Although the point of ZT showed a unique fluctuation and had two peaks (4,484.08 μatm in July and 4,495.21 μatm in November 2016) in the sampling year compared with the rests, the trend of ZT point was quite consistent with trends of MD, FL and WZ if the second peak of October and December is not considered. After July of 2016, the overall trend of five places started to decline accompanied by some fluctuations until the December of 2016, and reached to the lowest p(CO_2) during this year (ZT: 1,246.99 μatm, MD: 1,249.86 μatm, FL: 1,256.00 μatm, ZX: 1,292.09 μatm and WZ:

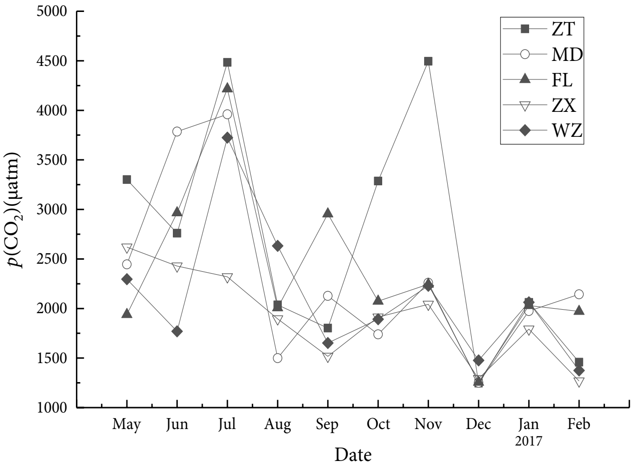

Fig. 2. Results of the partial pressure of CO_2.

1,476.56 µatm). Additionally, the range of p(CO_2) on two sites in annual backwater area, ZX and WZ, were narrower than that of MD and ZT in the non-annual backwater, which means that the carbon input in the area of annual backwater had no significant increase and was more stable. Thus, it is reasonable to prove the point of FL as a boundary between annual and non-annual backwater. The second peak of ZT was probably attributed to its location located at the end of backwater where it had a less impact on the impoundment of the Three Gorges Reservoir.

The evolution of p(CH_4) on five sites is shown in Fig. 3.

From the first month to start monitoring, every point of partial pressure of CH_4 went down obviously except WZ due to the insufficient oxidation, attributed to methane produced in sediment diffused more quickly than high water level. Overall, from September to February the partial pressure of CH_4 remained at a low level except for ZT which appeared the second peak similar to regular pattern on the partial pressure of CO_2. The maximum of ZX in August needs further research to consider the primary cause because the data was probably out of a tolerance scope due to the mistake during field sampling.

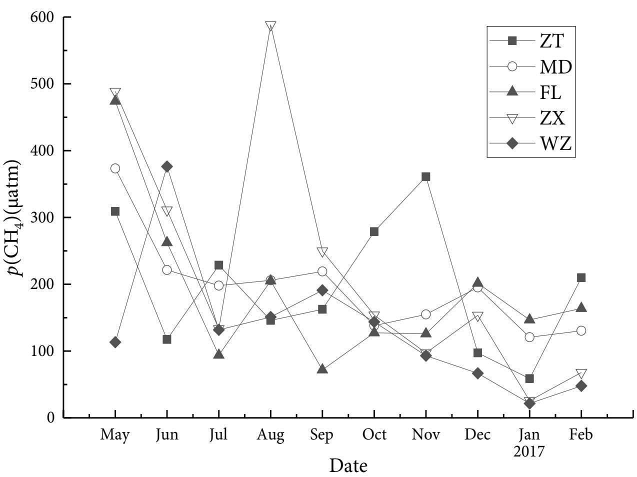

Fig. 3. Results of the partial pressure of CH_4.

3.2. Annual average partial pressures of CO_2 and CH_4

Fig. 4 shows the results of the annual average partial pressures of CO_2 The overall trend of the partial pressure of CO_2 was declining from the upstream, ZT, to downstream, WZ. The maximum annual average in five sites was ZT which was 2,693.48 µatm, while the minimum in those was FL, 1,908.78 µatm. The observation points of MD, FL, WZ were 2,318.40, 2,365.89, 2,110.79 µatm, respectively. It is apparent that the range of partial pressures of CO_2 in WZ and ZX was more stable compared with that of FL, MD, ZT all of which were located in the non-annual flooded area or transition zone between annual flooded area and non-annual flooded area. The increased amount of phytoplankton within the area of the annual flooded area caused their photosynthesis enhance apparently so CO_2 in water decreased in the process.

Fig. 5 shows the results of the annual average partial pressures of CH_4. The annual average partial pressures of CH_4 had a same regular pattern with that of CO_2 except the observation points ZX, where range of variation and annual average partial pressures of CH_4 (226.87 µatm) were higher than the others. WZ was still at the lowest level of annual average partial pressures of CH_4, 133.61 µatm. The figure of ZT, MD and FL were, respectively, 196.87, 195.68 and 187.24 µatm.

3.3. Flux of CO_2 and CH_4

The flux of CO_2 is shown in Fig. 6.

The trend of fluxes CO_2 has a similarity with partial pressure generally because the most of observation points reached the peak in July 2016 and in November, a second peak appeared as well (Fig. 6). All of the data had positive value which means the source of CO_2 came from water and emitted to the air. Additionally, the months with warm temperature had a higher CO_2 emission than the months in winter. The point of ZT was still specially high because of the second peak in November. This occurrence may be caused by that in the area of non-annual backwater the carbon input was not regular similar to the area of the annual backwater.

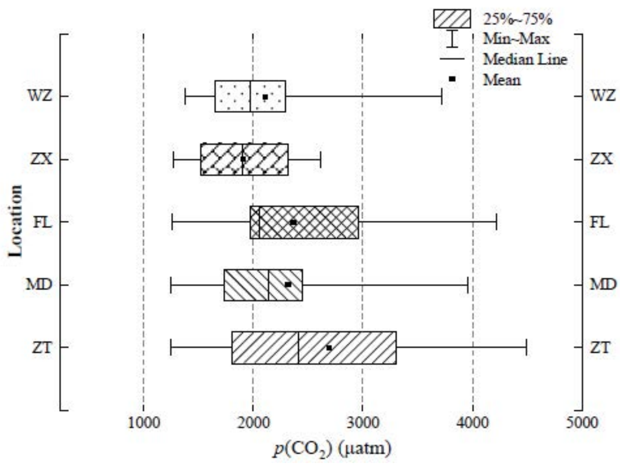

Fig. 4. Results of the annual average partial pressures of CO_2.

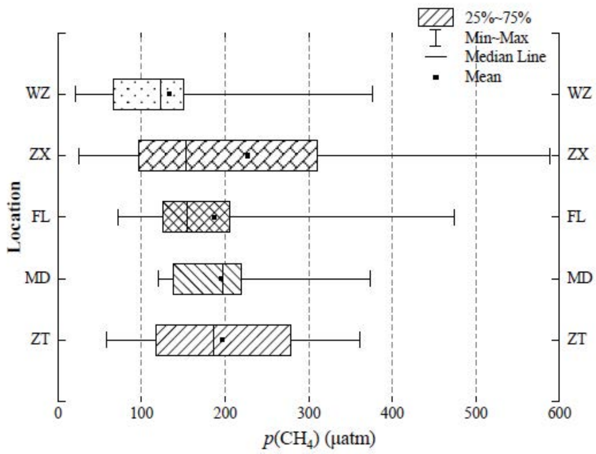

Fig. 5. Results of the annual average partial pressures of CH_4.

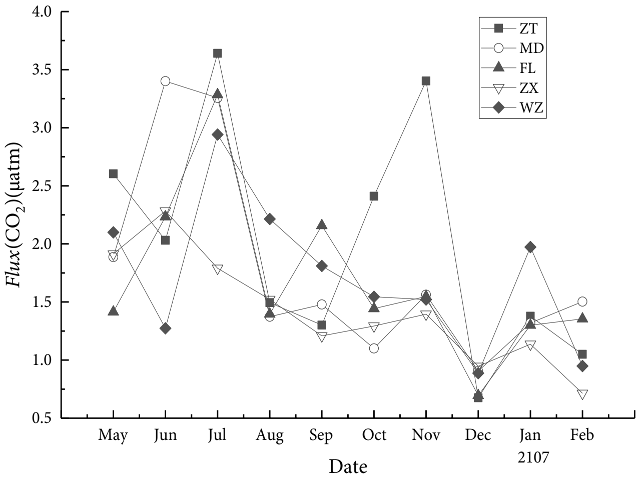

Fig. 6. Results of the air–water CO_2 fluxes.

The general trend of CH_4 fluxes in between the air–water interface was decreased from May 2016 to January 2017 in Fig. 7, which was relative to the overture of water level in the Three Gorges Reservoir except the data of ZT in August 2016. Apparently, in May 2016, the water level of the reservoir was low due to a low storage of reservoir operations, raising the speed of diffusion of CH_4 from sediment to the interface between air–gas caused a low-efficient oxygenation for CH_4 [20–24]. After January 2017, the water level of the reservoir started to decrease, probably led to the fluxes of CH_4 to increase.

3.4. Annual average fluxes of CO_2 and CH_4

As in Fig. 8 indicate, from the detective results of five observation sites, the reservoir was the source of CO_2 in the atmosphere due to the positive value of fluxes CO_2, the annual average of 41.29 ± 4.46 mmol m^{-2} d^{-1}. The maximum of CO_2 annual average fluxes in five points was ZT, 1.99 mmol m^{-2} h^{-1}, while the minimum of that was ZX with the figure of 1.42 mmol m^{-2} h^{-1}. The points MD, FL and WZ were, respectively, 1.78, 1.68 and 1.72 mmol m^{-2} h^{-1}. The rise

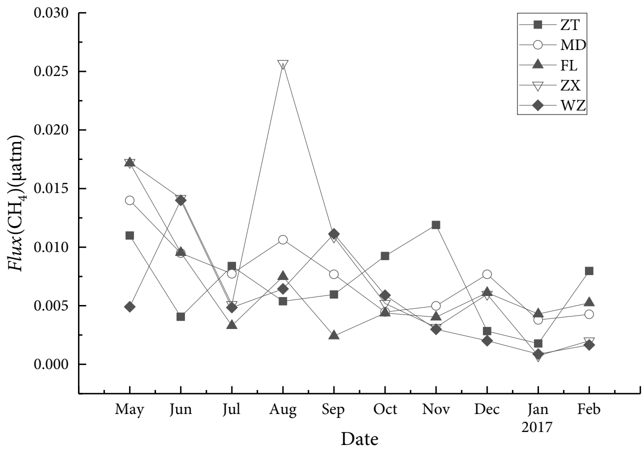

Fig. 7. Results of the air–water CH_4 fluxes.

in WZ may be caused by the denser populated city which has the most population in five sites [25,26].

It can be proved the CH_4 emission from the reservoir because all values of the data were positive [27]. There was no obvious regular pattern among five sites in Fig. 9. The maximum of CH_4 annual average flux was ZX, 0.009,0 mmol m^{-2} h^{-1}, while the minimum of that was WZ, 0.005,5 mmol m^{2} h^{-1}. The values of CH_4 annual average fluxes in ZT, MD, FL were, respectively, 0.006,8 mmol m^{-2} h^{-1}, 0.007,4 mmol m^{-2} h^{-1}, 0.006,4 mmol m^{-2} h^{-1}.

3.5. Environmental factors influencing fluxes of CH_4 and CO_2

3.5.1. Temperature of surface water

During the 10 months of observation, the temperature of five sites had a similar regular pattern which was rising from May to August and then declining until January 2017 (Fig. 10). In August 2016, the temperature was highest (ZT: 25.1°C, MD: 25.9°C, FL: 29.4°C, ZX: 31.8°C and WZ: 33.7°C), while the lowest temperature appeared in December 2016 and January 2017 (ZT: 13.7°C, MD: 14.1°C, FL: 13.0°C, ZX: 13.5°C and WZ: 13.6°C). The temperature of the surface water would affect the gas emission speed between water–air interface in some degree [28].

3.5.2. Dissolved oxygen

In Fig. 11, the DO of surface water in the 10 months observation in five points kept rising, which was opposite to the trend of water temperature of the surface. This may be due to the warmer water temperature which negatively affects the DO. Additionally, the nearer the Three Gorges Dam to the site, the lower the values of DO had, the slower water flow was in the reservoir [29]. ZT as the furthest upstream of site had the obvious higher values, while values of DO in ZX and WZ, much closer to the Three Gorges Dam, were at low level. The warmer temperature of the water would create a suitable environment for algae to grow, the DO as a source of growth would be absorbed. The range of DO in five points was 7.86–10.64 mg L^{-1} (ZT), 7.51–9.60 mg L^{-1} (MD), 7.41–9.87 mg L^{-1} (FL), 6.71–9.83 mg L^{-1} (ZX), 6.97–9.24 mg L^{-1} (WZ).

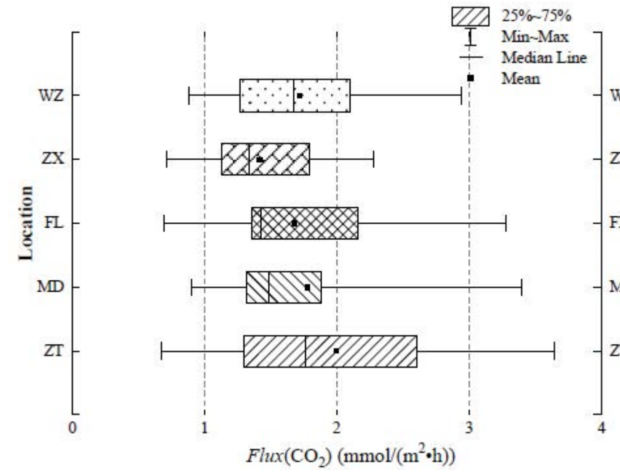

Fig. 8. Results of air–water CO_2 annual average fluxes.

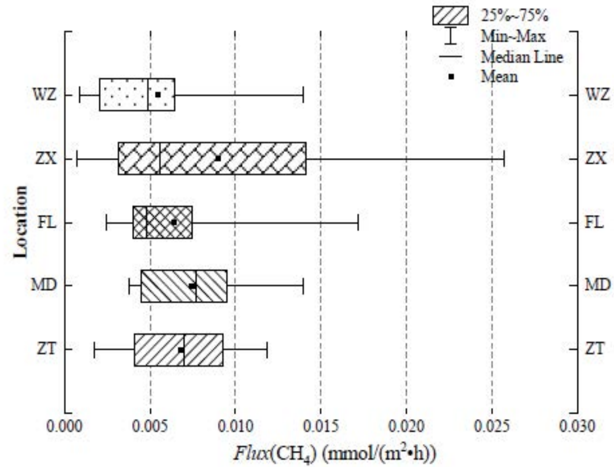

Fig. 9. Results of air–water CH_4 annual average fluxes.

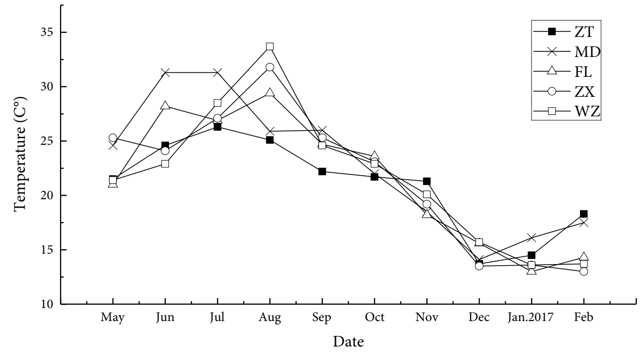

Fig. 10. Distribution of temperature of surface water.

3.5.3. pH

The overall pH of the surface water was alkaline, and the fluctuation of pH was not significant except for the point of ZT in July 2016 (Fig. 12). In winter from December to May, pH had a higher value ranging from 8.0 to 8.65. The pH range

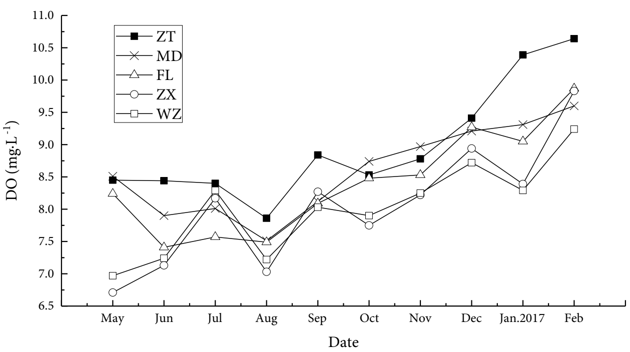

Fig. 11. Distribution of dissolved oxygen.

in ZT, MD, FL, ZX and WZ were, respectively, 7.71–8.61, 8.03–8.50, 8.01–8.61, 7.94–8.62 and 7.97–8.63.

3.5.4. Chl-a

The distribution of Chl-a in sites of ZT, MD, FL, which had peak values between October and November, had a similarity, and the rest of the months remained stable and at low level (Fig. 13). The concentration of Chl-a on the site of ZX, which ranged from 0.3 to 4.0 mg L^{-1}, had a peak in winter, while that of WZ, ranged from 1.5 to 4.3 mg L^{-1}, reach the highest summit in May 2016. It is apparent that the points ZX and WZ, nearer to dam were in areas of low-speed water flow, which caused a fewer distribution of fluctuation but in the area farther away from the dam, ZT, MD and FL, the fluctuation of Chl-a distribution was more important due to the faster water flow. It is not suitable for the growth of phytoplankton, so there were such different regular patterns.

3.6. Correlation between indicators and fluxes of CH_4, CO_2, pressures of CO_2, CH_4

The data on water temperature, DO, pH, Chl-a, conductivity, wind speed and fluxes of CH_4, CO_2, pressures of CO_2, CH_4 were conducted the analysis of correlation. The results are shown in Table 2.

4. Discussion

The results indicated that except data of Chl-a, the rest of the environmental indicators was significantly related to fluxes and pressure of CO_2, CH_4 in every site. Due to the change of water temperature which influenced the value of CH_4 pressure in water, CH_4 raised more easily from the bottom of the river to surface water when water temperature was high. Simultaneously, a suitable water temperature makes a contribution to methanogens to produce methane as a metabolic byproduct in river sediments with anoxic conditions. Therefore, the pressure of CH_4 has a significant positive correlation with water temperature and is negatively related to DO.

The pressure of CO_2 was positively related to water temperature while it has a negative correlation with conductivity, DO, pH, wind speed. The impact of water temperature

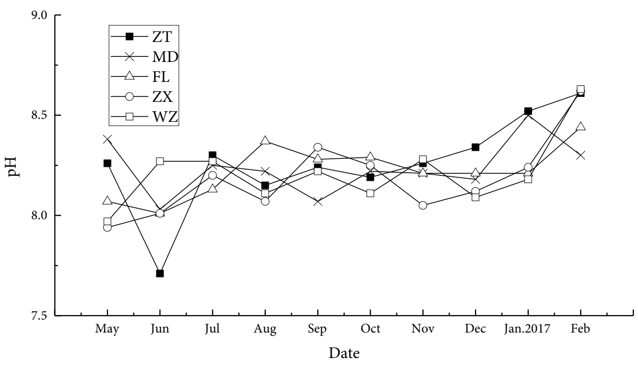

Fig. 12. Distribution of pH.

Table 2
Correlation analysis between indicators and fluxes of CH_4, CO_2, pressures of CO_2, CH_4

	CH_4 pressures	CO_2 pressures	CH_4 fluxes	CO_2 fluxes
CH_4 pressures	1	–	0.971**	–
CO_2 pressures	–	1	–	0.879**
Water temperature	0.448**	0.543**	0.515**	0.612**
Conductivity	–	–0.353*	–0.324*	–0.360
DO	–0.310**	–0.373*	–0.420**	–0.487**
pH	–	–0.280*	–	–0.317**
Wind speed	–	–0.344*	–	–
Chl-a	–	–	–	–

Note: $^{**}p \leq 0.01$, indicating extremely significant correlation; $^{*}p \leq 0.05$, indicating significant correlation.

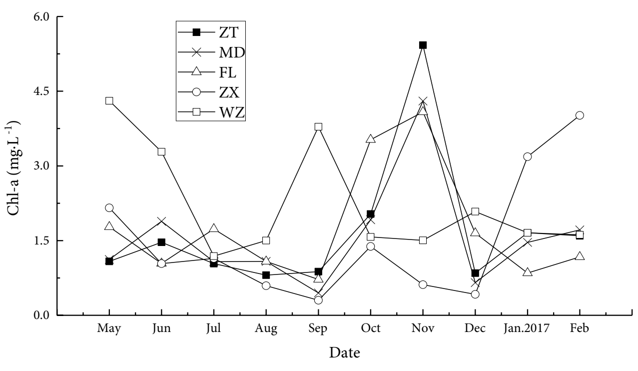

Fig. 13. Distribution of Chl-a.

on the pressure of CO_2 has a similar characteristic with that of CH_4. A higher value of water conductivity means high quality of total dissolved solids, which therefore means dissolved solids including carbon in water may be at a low level. The respiration and decomposition of microorganism will consume oxygen and carbon in water, so the more DO is consumed, the more CO_2 will be produced. There is an equilibrium reaction with hydrogen ion, bicarbonate ion and dioxygen [30]. When pH value ascends, the dioxygen attaches hydrogen ion with a positively charged ion from a bicarbonate ion, causing a decrease of pressure of CO_2 in water. Wind speed makes a disturbance at the surface water will results in a more efficient diffusion of gas from water to air.

Fluxes of CH_4 have a positive correlation with CH_4 pressure and water temperature and are negatively related to DO and conductivity. Due to the calculation of our research which is based on the Fick's law of diffusion – the flux diffuses from regions of high concentration to low, the higher the pressure of CH_4 present in water, the more the fluxes will diffuse between water–air interface. It is apparent that a higher water temperature makes a positive contribution to the flux from water to the atmosphere. During CH_4 diffusing from sediment to surface water, it will oxidize if there is adequate value of DO. The more dissolved solids in the water, the less consumption of oxidized carbon, so CH_4 will less diffuse to the atmosphere.

Fluxes of CO_2 have a negative correlation with the pressure of CO_2 and water temperature while negatively relate to DO and conductivity. The same principle applies with fluxes of CH_4 which corresponds to own pressure in water and water temperature. The value of DO results in the less decomposition of dissolved carbon in water so that the production of CO_2 by decomposition will be less. Lastly, the hydrogen ion will reduce diffusion of CO_2 as the previous analysis.

5. Conclusions

- The range of pressure of CO_2 and CH_4 was 1,246.99–4,495.20 μatm and 21.63–588.28 μatm, respectively, in the surface water within the research sites. The main trend pressure of CO_2 in the direction of water flow is decreasing, while that of CH_4 has no significant character. The pressure of CO_2 and CH_4 in summer was higher than those in winter. The decrease of fluxes of CO_2 between water–air interface with the area from non-annual backwater to permanent backwater indicated that the Three Gorges Reservoir has a reductive impact on the diffusion of CO_2 in the Yangtze River, while those of CH_4 has no difference in the area.
- Within a major part of operation overturn of the Three Gorges Reservoir from May 2016 to February 2017, the value of fluxes of CO_2 was positive average with 1.75 mmol m^{-2} h^{-1} which meant that reservoir is a source of CO_2. Fluxes of CH_4 were the same conclusion with fluxes of CO_2 with the average of 0.0070 mmol m^{-2} h^{-1}, and the trend of the pressure of both is consistent with fluxes of those.
- However, fluxes of CH_4 had a positive correlation with the pressure of CH_4 and water temperature while is negatively related to conductivity and DO in our research sites. Additionally, the correlation of fluxes dioxygen with the pressure of CO_2 and water temperature is positive while with DO, conductivity and pH is significantly negative. Other environmental indicators have no obvious impact and correlation, which need further research to unveil potentials.

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