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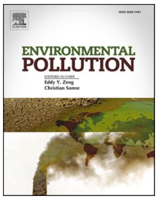

# Eutrophic levels and algae growth increase emissions of methane and volatile sulfur compounds from lakes☆

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ABSTRACT

Eutrophic lakes are hot spots of $CH_4$ and volatile sulfur compound (VSC) emissions, especially during algal blooms and decay. However, the response of $CH_4$ and VSC emissions to lake eutrophication and algae growth as well as the underlying mechanisms remain unclear. In this study, the emissions of $CH_4$ and VSCs from four regions of Lake Taihu with different eutrophic levels were investigated in four months (i.e., March, May, August and December). The $CH_4$ emissions ranged from 20.4 to 126.9 mg $m^{-2}$ $d^{-1}$ in the investigated sites and increased with eutrophic levels and temperature. $H_2S$ and $CS_2$ were the dominant volatile sulfur compounds (VSCs) emitted from the lake. The $CH_4$ oxidation potential of water ranged from 2.1 to 14.9 μg $h^{-1}$ $L^{-1}$, which had positive correlations with trophic level index and the environmental variables except for the $NH_4^+$-N concentration. Eutrophic levels could increase the abundances of bacteria and methanotrophs in lake water. α-*Proteobacteria* methanotroph *Methylocystis* was more abundant than γ-*Proteobacteria* methanotrophs in March and May, while the latter was more abundant in August and November. The relative abundance of *Cyanobacteria,* including *Microcystis*, *A. granulata* var. *angustissima* and *Cyanobium* had significantly positive correlations with temperature, turbidity, $SO_4^{2-}$-S, and total sulfur. Partial least squares path modelling revealed that the algal growth could promote VSC emissions, which had a positive correlation with $CH_4$ oxidation potential, likely due to the positive correlation between the $CH_4$ and VSC emissions from lakes. These findings indicate that water eutrophication and algae growth could increase the emissions of $CH_4$ and VSCs from lakes. Controlling algae growth might be an effective way to mitigate the emissions of $CH_4$ and VSCs from freshwater lakes.

## CRediT authorship statement

**Jing Wang**: Methodology, Data curation, Formal analysis, Writing - original draft, Investigation. **Zhi-Peng Wei**: Sampling, Methodology, Data curation; **Yi-Xuan Chu**: Data curation, Formal analysis, Investigation. **Guangmin Tain**: Conceptualization, Resources, Supervision, Writing - review & editing. **Ruo He**: Conceptualization, Methodology, Resources, Supervision, Project administration, Writing - review & editing.

## 1. Introduction

Methane ($CH_4$) is the second most important greenhouse gas after $CO_2$, accounting for approximately 20% of the greenhouse effect of global warming (Wuebbles and Hayhoe, 2002). Lake is an important natural source of $CH_4$ emissions, accounting for ~3% of the Earth's continental surface area and is responsible for approximately 6%–16% (8–48 Tg $yr^{-1}$) of global $CH_4$ emissions (Bastviken et al., 2004; Bastviken et al., 2011; Downing et al., 2006). Recently, due to anthropogenic activities and agricultural inputs of nutrients, algae growth and decay in eutrophic lakes have led to a large amount of organic matter degradation (Perga et al., 2016; Xiao et al., 2020) and $CH_4$ release (West et al.,

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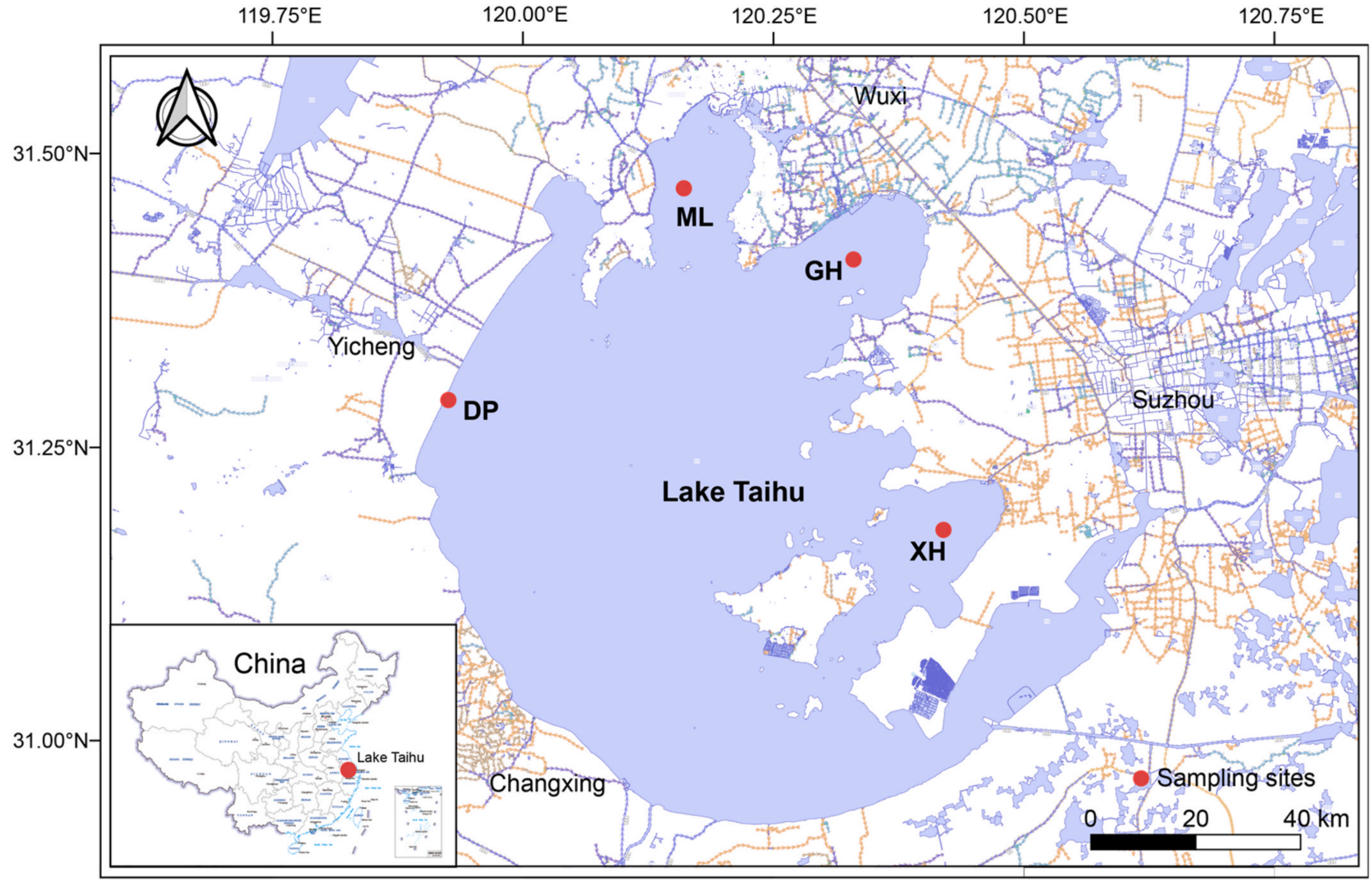

**Fig. 1.** Location of the four sampling positions, i.e., ML, DP, GH and XH, in Lake Taihu, China.

2016; Xiao et al., 2017). It is estimated that $CH_4$ emissions can be increased by 30%–90% with increase in eutrophication level from lakes and impoundments during the 21st century and can affect the C cycle and greenhouse effect in lacustrine ecosystems (West et al., 2016; Beaulieu et al., 2019).

Aerobic $CH_4$ oxidation is the major sink of $CH_4$ in freshwater lakes (Borrel et al., 2011). Methanotrophs are a unique group of bacteria that can use $CH_4$ as the sole carbon and energy source (Hanson and Hanson, 1996). It has been reported that methanotrophs can oxidize more than 90% of $CH_4$ produced from sediments and play an important ecological function in mitigating $CH_4$ emissions from freshwater lake systems (Deng et al., 2017; Michaud et al., 2017). Based on the phylogeny, morphological and physiological characteristics, aerobic methanotrophs can be divided into three clades: γ-*Proteobacteria*, α-*Proteobacteria* and *Verrucomicrobia* (Bowman, 2006; Semrau et al., 2010). The initial step of $CH_4$ oxidation to $CO_2$ is catalysed by methane monooxygenase (MMO), which includes particulate MMO (pMMO) and soluble MMO (sMMO) encoded by *pmoA* and *mmoX* genes, respectively (Smith and Murrell, 2009). The *pmoA* gene has been widely used as a biomarker to identify aerobic methanotrophs due to its existence in almost all known methanotrophs except *Methyloferula* and *Methylocella* (Dumont and Murrell, 2005).

The community structure and activities of methanotrophs vary with environmental conditions (i.e., water physiochemical variables) in lakes. Mayr et al. (2020) reported that *Methylosoma*, *Methylobacter* and *Crenothrix* were the main methanotrophs in the hypolimnion of Lake Rotsee in Switzerland, and their community varied with water temperature, nutrients, dissolved oxygen (DO) and $CH_4$ concentrations. The relative abundance of *Methylobacter* increase with decreasing water temperature and increasing DO concentration, while the relative abundance of *Crenothrix* increase with increasing $CH_4$ concentration (Mayr et al., 2020). Yang et al. (2019) found that lake eutrophication could increase the abundance of α-*Proteobacteria* methanotrophs, while the lower total nitrogen (TN) concentration in the water was suitable for the growth of *Methylobacter*. The $CH_4$ oxidation potential (MOP) is reported to be positively correlated with the oxidation–reduction potential (ORP) of overlying water and negatively correlated with the $NH_4^+$-N concentration of overlying water in Dianchi Lake, China (Yang et al., 2019). An increase in MOP is also observed to be coupled with the higher abundance of α-*Proteobacteria* methanotrophs (Lin et al., 2005). Thus, environmental variables are important factors influencing methanotrophic community and MOP, thereby resulting in variations in $CH_4$ release from lakes.

The growth and decay of algae in eutrophic lakes not only release a large amount of $CH_4$, but also produce many odour substances, mainly volatile sulfur compounds (VSCs), which greatly affect the water quality of lakes and the lives of the surrounding residents (Wu et al., 2021a). VSCs such as $H_2S$, $CH_3SH$, $(CH_3)_2S$ and $(CH_3)_2S_2$ can be produced during algae accumulation, bloom and decay (Liu et al., 2019; Liu et al., 2020). The VSCs in eutrophic lakes can cause odour pollution owing to their low olfactory threshold (Chen et al., 2010; Lee et al., 2017). In 2007, high concentrations of VSCs, including $CH_3SH$, $(CH_3)_2S$, $(CH_3)_2S_2$ and $(CH_3)_2S_3$, were responsible for the drinking water crisis occurred in Wuxi city, China (Zhang et al., 2010). Moreover, VSCs are toxic to microorganisms such as nitrifying bacteria and methanotrophs (Börjesson, 2001; Zhang et al., 2016). Previous studies have reported that $H_2S$, $CS_2$, $(CH_3)_2S$ and $CH_3SH$ can inhibit $CH_4$ oxidation in various environments (Börjesson, 2001; Lee et al., 2011). However, Kim et al. (2013) found that $(CH_3)_2S$ had a positive effect on methanotrophic performance in a biofilter. Eutrophic lakes are hot spots of emissions of $CH_4$ and VSCs, especially during algal blooms and decay. Recently, water eutrophication increased $CH_4$ emissions from lakes and the estimation of increased

$CH_4$ emissions have been reported (Zhou et al., 2020; Sun et al., 2021). However, the response of $CH_4$ and VSC emissions to lake eutrophication and algae growth as well as their correlations with the environmental variants, community structure and activities of methanotrophs remain unclear.

The objective of this study was to explore the impacts of eutrophic levels and algae growth on the emissions of $CH_4$ and VSCs from eutrophic lakes. Lake Taihu, a typical eutrophic lake in China, was selected for the research object. The physicochemical properties of water were detected in four regions of Lake Taihu and four months (i.e., March, May, August and November) to investigate temporal and spatial eutrophic levels. The emissions of $CH_4$ and VSCs were determined with an estimation of MOP in water. Additionally, the abundance and community structure of bacteria and methanotrophs in water were analysed as well as effects of environmental variables on microbial community and $CH_4$ emissions. The obtained results add to our understanding of the impacts of algal growth and decay on microbial metabolism, especially the activities and community structure of methanotrophs, and helps to reduce the emissions of $CH_4$ and VSCs from eutrophic lakes.

## 2. Materials and methods

### 2.1. Study sites and sampling

Lake Taihu is the third largest freshwater lake in China, with an average depth of ~1.9 m (Qin et al., 2007). Lake Taihu is a eutrophic lake with algae blooms from April to October each year (Ji et al., 2021). The trophic level in the northwestern area is much heavier than that in the southeastern area of Lake Taihu due to the higher concentrations of nutrients and prevailing southeast wind (Meng et al., 2021). We chose four regions with different trophic levels, including Meiliang Bay (ML), Gonghu Bay (GH), Xuhu Bay (XH) and the Dapu River mouth (DP), for sampling (30°55′40″-31°32′58″ N, 119°52′32″-120°36′10″ E, Fig. 1). ML is in the northern area of Lake Taihu with higher cyanobacterial biomass owing to the prevailing southeast wind (Ji et al., 2021). GH is in the northeastern area of Lake Taihu with more aquatic macrophytes, which serves as an important centralized source of drinking water for Suzhou and Wuxi cities in Jiangsu Province (Ding et al., 2020). XH is in the eastern area of Lake Taihu with less algae growth (Liu et al., 2016). DP is in the western area of Lake Taihu with a heavier eutrophic level owing to anthropogenic inputs of nutrients (Shao et al., 2018). Gas and water samples were collected from the four regions in March, May, August and November 2019. Three sampling sites with distances greater than 10 m from each other were conducted in each region. Gas and water samples in triplicate were taken from each sampling site.

Approximately 2 L of surface water at a depth of 0.5 m was collected from each site using a Plexiglass sampler. Approximately 200 mL water sample was taken for measuring AVS with the addition of 220 g $L^{-1}$ Zn $(Ac)_2$ to stabilize sulfide (Dong et al., 2012). Approximately 120 mL water sample was fully filled into a serum bottle to determine the dissolved concentrations of $CH_4$ and VSCs. The temperature, DO, pH, turbidity, Chl-a and phycocyanin concentrations in the surface water at a depth of 0.5 m were detected *in situ* using a multiparameter water quality sonde (YSI 6600, Yellow Springs, USA). The emissions of $CH_4$ and VSCs were measured using the floating chamber technique as described previously (Gonzalez-Valencia et al., 2014; Wang et al., 2022). The floating chamber consisted of a plexiglass cylinder with an open surface of 0.4 $m^2$ and a headspace volume of $6.5 \times 10^{-2}$ $m^3$. The floating chamber was equipped with a sampling port and connected with the anchored sampling ship through a rope. Gas samples were withdrawn using a syringe from the floating chamber every 20 min within a 1-h period, and then stored in pre-vacuumed 20 mL vials. Three floating chambers with 1-m interval were used to measure gas emissions in each sampling site.

### 2.2. MOP determination

A 20-mL water sample was put into a 100-mL sterile serum vial, which was purged with high purity air (99.9%) for ~5 min, and then sealed with a butyl rubber stopper. Purity $CH_4$ (99.9%) was injected to obtain a concentration of 0.1% ($v/v$) in the headspace of the vial. Autoclaved water was used as a control. Each water sample was performed in triplicate. The serum vials were placed on a shaker at 130 rpm and 25 °C. A 100-μL gas sample in the headspace of the serum vials was withdrawn to determine the $CH_4$ concentration in 12-h intervals. The MOP was calculated by a zero-order decrease in the $CH_4$ concentration in the headspace of the vials (Wang et al., 2011).

### 2.3. Analytical methods

A part of the water samples was used for analysing physicochemical variables, including concentrations of chemical oxygen demand (COD), total nitrogen (TN), $NO_3^-$-N, $NH_4^+$-N, total sulfur (TS), $SO_4^{2-}$-S, total iron (TFe) and total phosphorus (TP) to investigate temporal and spatial eutrophic levels. The TP concentration was detected as described by Xiang and Zhou (2011). The concentrations of COD, TN, $NH_4^+$-N and $NO_3^-$-N were determined as described previously (Eaton et al., 2005). The concentrations of TS, AVS and $SO_4^{2-}$-S were determined using the method described by He et al. (2018). The TFe concentration was detected as described by Yin et al. (2008). The detection limit for COD, TN, $NO_3^-$-N, $NH_4^+$-N, TS, $SO_4^{2-}$-S, AVS, TFe and TP are 0.2, 0.02, 0.06, 0.1, 0.01, 0.5, 0.003, 0.01 and 0.03 mg $L^{-1}$, respectively.

The concentrations of $CH_4$ and VSCs in the gas samples were measured using a gas chromatograph equipped with a thermal conductivity detector (GC-TCD) and a flame ionization detector (FID) (Wang et al., 2011), and HC-3 trace sulfur analyser equipped with a flame photometric detector, respectively (Chen et al., 2017). The GC-TCD was equipped with a GDX-104 column of 2 m with $N_2$ used as the carrier gas at a flow of 20 mL $min^{-1}$. The conditions for the GC-FID: DB-624 capillary column; the temperatures of the oven, injector and detector were 120, 250 and 220 °C, respectively. The HC-3 trace sulfur analyser was equipped with a flame photometric detector as described previously (Chen et al., 2017). The emissions of $CH_4$ and VSCs were calculated as described previously (Gonzalez-Valencia et al., 2014).

The dissolved concentrations of $CH_4$ and VSCs were determined in triplicate by the headspace equilibration technique (Jacinthe and Groffman, 2001; Xia et al., 2022). In brief, 50 mL purity $N_2$ was injected into the serum bottle (water sampling bottle) to displace an equal volume of water. The serum bottle was shaken vigorously for 5 min to equilibrate between the headspace and water phases. Then, the gas samples in the headspace were taken to determine the concentrations of $CH_4$ and VSCs. The dissolved concentrations of $CH_4$ and VSCs in water were calculated by Henry's law.

### 2.4. DNA extraction, qPCR analyses and MiSeq sequencing

Approximately 1 L of the water sample was filtered through a 0.22-μm microporous membrane to collect microorganisms in water. Then, the microporous membrane was used to extract total DNA using the E. Z. N.A.™ soil DNA extraction kit (Omega Bio-Tek, Inc., Norcross, USA). The DNA concentration was detected using a NanoDrop 2000. The abundances of the bacterial 16S rRNA gene and *pmoA* in the water samples were determined using qPCR as described by Wang et al. (2022) (Supplementary materials). The abundances of the bacterial 16S rRNA gene and *pmoA* were expressed by copies per litre (copies $L^{-1}$).

The microbial community structure were analysed by applying MiSeq sequencing. The fragment of the V3–V4 region of the bacterial 16S rRNA gene was amplified with the primer set 338F/806R as described previously (Wang et al., 2022; Supplementary materials). The PCR products were sequenced on an Illumina MiSeq platform at Shanghai Majorbio Bio-Pharm Technology Co., Ltd. The sequencing data

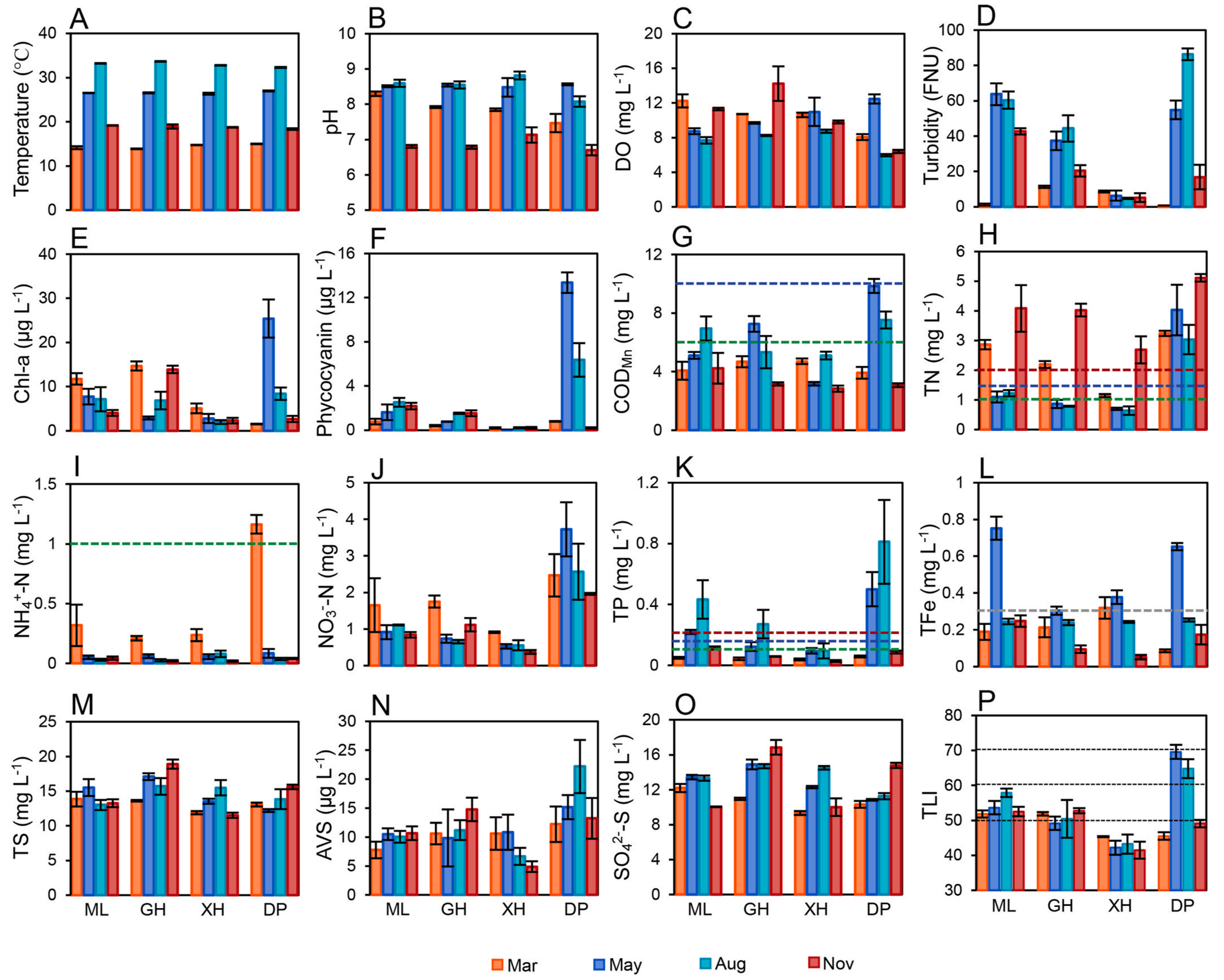

**Fig. 2.** Temporal and spatial variations of average T (A), pH (B), DO (C), turbidity (D), Chl-a (E), phycocyanin (F), $COD_{Mn}$ (G), TN (H), $NH_4^+$-N (I), $NO_3^-$-N (J), TP (K), TFe (L), TS (M), AVS (N), $SO_4^{2-}$-S (O) and TLI (P) in water at 0.5-m depth. Error bars indicate standard deviations (n = 9). Green, blue and red dashed lines indicate the limited values of Grade III, IV and V of the environmental quality standards for surface water in China (GB 3838–2002), respectively. Grey dashed line indicates the limited value ($\leq 0.3$ mg $L^{-1}$) of the supplementary standard of the environmental quality standards for surface water in China (GB 3838–2002). Black dashed lines indicate the trophic classification of lake freshwater based on the TLI values.

were analysed on the Majorbio Cloud platform (www.majorbio.com) as described by Wang et al. (2022).

The sequencing data of the bacterial 16S rRNA gene have been deposited in the National Omics Data Encyclopedia (NODE) under the accession number OER241134-241,181.

### 2.5. Data analysis

The differences among samples were analysed by analysis of variance (ANOVA) using SPSS 23.0. The correlations between the physiochemical variables of water and the microbial community were performed with the packages "corplot" and "psych" in R (4.1.0). Redundancy analysis (RDA) was carried out with the package "vegan" in R (4.1.0) to visualize the community structure dissimilarities and identify the key physiochemical variables.

Trophic lake index (TLI) was used to assess trophic levels of Lake Taihu in different months and regions. TLI was calculated with variables including Chl-a, TP, TN and $COD_{Mn}$ as described by Zhou et al. (2020) and shown in Eqs. (1)–(6).

$$TLI\left(\sum\right)=\sum_{j=1}^{m}W_j{*}TLI\,(j) \tag{1}$$

$$W_j=\frac{r_{ij}^2}{\sum_{j=1}^{m}r_{ij}^2} \tag{2}$$

$$TLI\,(Chla)=10{*}[2.5+1.086\,\ln(Chla)] \tag{3}$$

$$TLI\,(TP)=10{*}[9.436+1.624\,\ln(TP)] \tag{4}$$

$$TLI\,(TN)=10{*}[5.453+1.694\,\ln(TN)] \tag{5}$$

$$TLI\,(COD)=10{*}[0.109+2.66\,\ln(COD)] \tag{6}$$

Where $TLI\,(\sum)$ is the integrated trophic level index, $TLI(j)$ is the trophic level index of $j$, $W_j$ is the correlative constant for the TLI of $j$, $r_{ij}$ is the relative coefficient, which was described by Zhang et al. (2011). $j$ and $m$ represent the parameters (i.e., Chl-a, TP, TN and $COD_{Mn}$) and the number of parameters, respectively. Based on the TLI ($\sum$) value, the

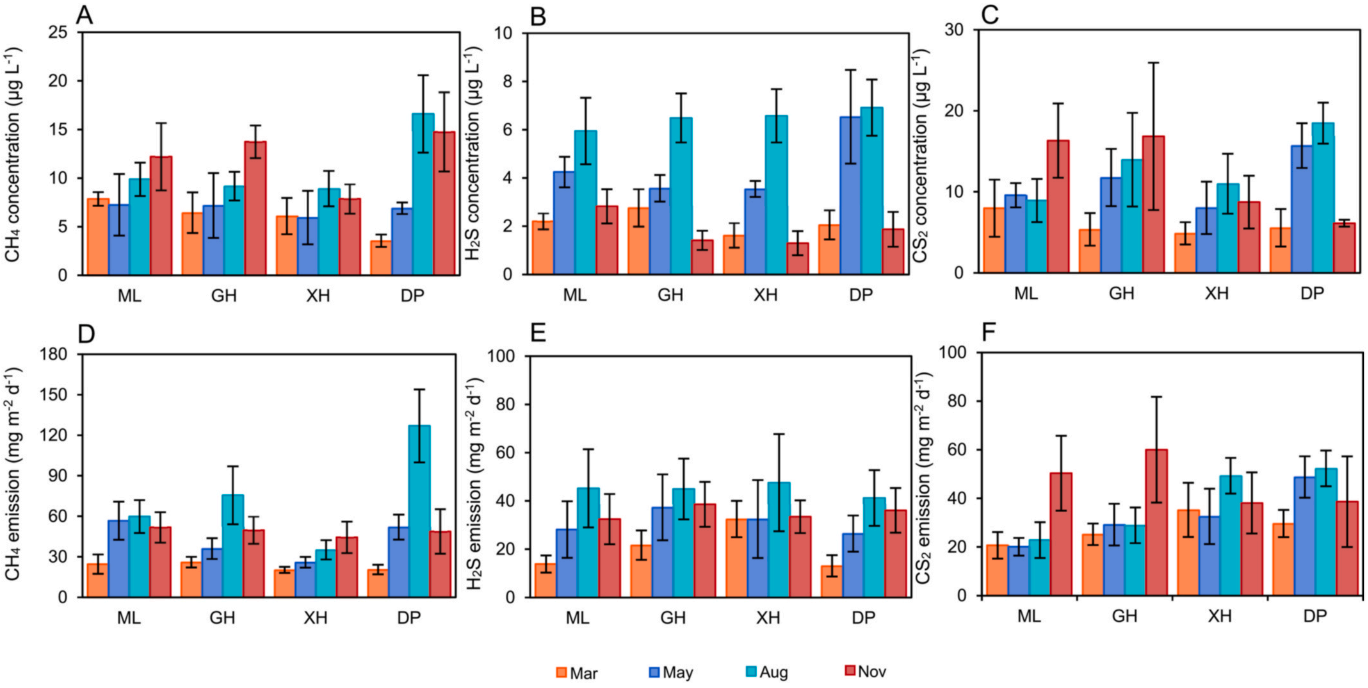

**Fig. 3.** Average concentrations and emissions of $CH_4$ (A, D), $H_2S$ (B, E) and $CS_2$ (C, F) from the four regions in Lake Taihu in four months (i.e., March, May, August and November), respectively. Error bars indicate standard deviations (n = 9).

lake trophic state was classified as oligotrophic (TLI (∑) < 30), mesotrophic (30 < TLI (∑) < 50), eutrophic (50 < TLI (∑) < 60), middle eutrophic (60< TLI (∑)< 70), and hypereutrophic (TLI (∑) > 70).

Partial least squares path modelling (PLS-PM) was performed using R (4.1.0) with the package “plspm” to explore the links among latent variables (Petersen et al., 2012). The latent variables in this study included algae growth, environmental variables, VSCs emission, methanotrophic abundance, methanotrophic community structure, MOP and $CH_4$ emissions. PLS-PM (1000 bootstraps) was used to estimate the path and determine coefficients in this model with the standardization of the variables. The $R^2$ of the latent variable of $CH_4$ emissions was estimated with the goodness of fit (GOF) statistic.

## 3. Results and discussion

### 3.1. Temporal and spatial variation of environmental variables and trophic levels

Lake Taihu is located in a subtropical region with the water temperatures of 14–34 °C at the 0.5-m depth from March to November during sampling period (Fig. 2). The water pH value ranged from 7.47 to 8.82 in GH, XH and ML, while it was slightly lower (6.70–7.13) in DP, mainly due to the degradation of organic matter (Wu et al., 2021b). The DO concentrations were lower at 5.97–8.73 mg $L^{-1}$ in the investigated sites in August than in the others, which could be mainly attributed to two reasons: 1) high temperature decreased oxygen solubility in water (Khangaonkar et al., 2012); and 2) algae decomposition consumed DO (Wang et al., 2020). Lower DO concentrations were detected in DP in August and November, mainly due to a large amount of oxygen consumed by the degradation of organic matter, such as decaying algae (Khangaonkar et al., 2012; Wang et al., 2015). The turbidity of water in ML, GH and DP was higher in May and August than in March and November, which could be attributed to sediment resuspension in the shallow lake by prevailing southeastern winds in summer (Wu et al., 2013). Among the four regions, the concentrations of Chl-a and phycocyanin were highest in DP, followed by ML and GH, and the lowest was detected in XH. This indicated that the water in the four investigated regions belonged to different eutrophic levels. Compared to Chl-a, the phycocyanin concentrations were lower in ML and GH in March and November, likely due to the different algae dominance in the four regions (Song et al., 2019). The $COD_{Mn}$ concentration ranged from 2.8 to 4.7 mg $L^{-1}$ in March and November, and increased 3.2–9.9 mg $L^{-1}$ in May and August, mainly owing to the high release from the degradation of organic matter in sediments at higher temperatures and the release of algae growth and metabolism (Wu et al., 2021b). The $COD_{Mn}$ concentrations in GH and DP in May and ML and DP in August were above 6 mg $L^{-1}$ (Grade III of the environmental quality standards for surface water in China (GB 3838–2002)). Among the four investigated sites, DP water had the highest nutrient concentrations including $NH_4^+$-N, $NO_3^-$-N, TN and TP, followed by ML, GH and XH. Of them, the concentrations of $NH_4^+$-N, TN and TP were above Grade III of the environmental quality standards for surface water in China (GB 3838–2002). The peak value of the $NH_4^+$-N concentration was observed at 1.2 mg $L^{-1}$ in DP in March with a low pH value and DO, likely due to high nitrification activities. Similarly, Xiao et al. (2019) also found the high $NH_4^+$-N level accompanied with lower DO and pH value in lake water. The TFe concentration was 0.3–0.8 mg $L^{-1}$ in the four regions in May, which was higher than the supplementary standard (≤0.3 mg $L^{-1}$) of the environmental quality standards for surface water in China (GB 3838–2002). Compared with the concentrations of nutrients and TFe, there was less difference in the concentrations of sulfur compounds including TS, AVS and $SO_4^{2-}$-S among the four regions. The $SO_4^{2-}$-S concentration ranged from 9.3 to 17.0 mg $L^{-1}$, accounting for 75.9%–94.8% of TS. This indicated that $SO_4^{2-}$-S was the predominant form of sulfur in water.

TLI calculated with the water variables including Chl-a, TP, TN and $COD_{Mn}$ showed that trophic levels of Lake Taihu varied with regions and seasons. The TLI values fluctuated within 49.2–57.9 in ML and GH, indicating the water in ML and GH was eutrophic. Compared with GH, the TLI values in ML were slightly higher in May and August. The TLI values in XH were less than 50, which belonged to mesotrophic level. The TLI values in DP were less than 50 in March and November, while they increased to 64.8 and 69.6 in May and August. These results showed that DP was suffering heavier eutrophication, followed by ML, GH and XH.

### 3.2. Higher eutrophic levels increase emissions of $CH_4$ and VSCs from lakes

The dissolved $CH_4$ concentration in DP was higher (11.1–20.5 μg $L^{-1}$) in August and November that might be attributed to high temperature and eutrophic levels (Fig. 3). However, the dissolved $CH_4$ concentration in ML and GH was higher in November than that in August, indicating eutrophic level was the important variable compared to temperature. The highest dissolved $H_2S$ concentration was detected in August ranging from 5.6 to 6.9 μg $L^{-1}$, followed by May, March and November. $H_2S$ production mainly comes from sulfate reduction and the degradation of organic matter under anaerobic conditions (Hargrave et al., 2008; Rathnayake et al., 2017). Chen et al. (2020) also found that microcystins generated by *Cyanobacteria* could induce desulfhydrase activity for $H_2S$ production. The dissolved $CS_2$ concentrations were 15.7 and 18.5 μg $L^{-1}$ in DP in May and August, respectively, which were significantly higher than those in ML and XH ($P < 0.05$), indicating high $CS_2$ production in DP with a higher eutrophic level.

The $CH_4$ emissions ranged from 20.4 to 126.9 mg $m^{-2}$ $d^{-1}$ in the investigated sites. A similar result was obtained by Yang et al. (2011) who reported that the $CH_4$ emissions ranged from 1.6 to 284.9 mg $m^{-2}$ $d^{-1}$ with an average $CH_4$ emission of 92.3 mg $m^{-2}$ $d^{-1}$ in Lake Taihu. Compared with the global average $CH_4$ emission of 9.9 ± 27.4 mg $m^{-2}$ $d^{-1}$ from lakes between 24° and 54° latitude (Bastviken et al., 2011), the $CH_4$ emissions were higher in Lake Taihu, likely due to the higher eutrophic levels (Yang et al., 2011). Among the four seasons, there was no significant difference in the $CH_4$ emissions in March and November. The $CH_4$ emission from ML was 56.8 mg $m^{-2}$ $d^{-1}$ in May, which was significantly higher than those from GX and XH ($P < 0.001$). Among the study sites, the highest $CH_4$ emission was detected in DP (126.9 mg $m^{-2}$ $d^{-1}$) in August, likely owing to the high eutrophic level and temperature. Similar results were also reported in previous studies that eutrophication and high temperature could increase $CH_4$ emissions from lakes, which were also positively correlated with Chl-a and TP in water (Xiao et al., 2017; Beaulieu et al., 2019). Since the $CH_4$ production from sediment is the main source of $CH_4$ emission from freshwater lakes, further studies such as $CH_4$ oxidation rate and $CH_4$ consumption models in vertical sediment and water profiles need to be conducted to evaluate $CH_4$ contribution of eutrophic water to the atmosphere.

VSC emissions including $H_2S$ and $CS_2$ were also detected in the lake. There was no significant difference in $H_2S$ emissions among the four regions. Among the four months, the $H_2S$ emissions were highest (41.2–47.6 mg $m^{-2}$ $d^{-1}$) in August and lowest (26.5–32.5 mg $m^{-2}$ $d^{-1}$) in March, indicating that temperature was an important factor influencing $H_2S$ emissions. This might be because temperature can affect $H_2S$ solubility in water owing to its weak acidity (Savary et al., 2012). $CS_2$ is mainly generated from action of $H_2S$ and organic matter, indirect photolysis of organic sulfur precursors, algae growth and biodegradation of organic sulfur compounds (Andreae and Jaeschke, 1992; Gharehveran and Shah, 2018). The highest $CS_2$ emissions were 50.3 and 60.0 mg $m^{-2}$ $d^{-1}$ from ML and GH in November, respectively, while they were 49.3 and 52.3 mg $m^{-2}$ $d^{-1}$ from XH and DP in August, respectively. This suggested that $CS_2$ emission could be affected by environmental variables such as sulfur-containing precursors, temperature, organic matter and $O_2$ concentration (Gharehveran and Shah, 2018). $CS_2$ emission can be increased with increasing temperature and organic matter content, while $O_2$ concentration can inhibit $CS_2$ formation (Gharehveran and Shah, 2018). Statistical analysis showed that $CH_4$ emissions were positively correlated with the emissions of $H_2S$ and $CS_2$ (Fig. S1), likely because they were mainly generated from anaerobic degradation of organic matter such as algae decomposition (Liu et al., 2019). Water TLI values were positively correlated with the $CH_4$ and $CS_2$ emissions, while they were not related with the $H_2S$ emissions. This might be mainly because $H_2S$ can dissolve easily in water, which can be influenced by environmental variables such as pH and temperature (Cholewinski et al., 2016).

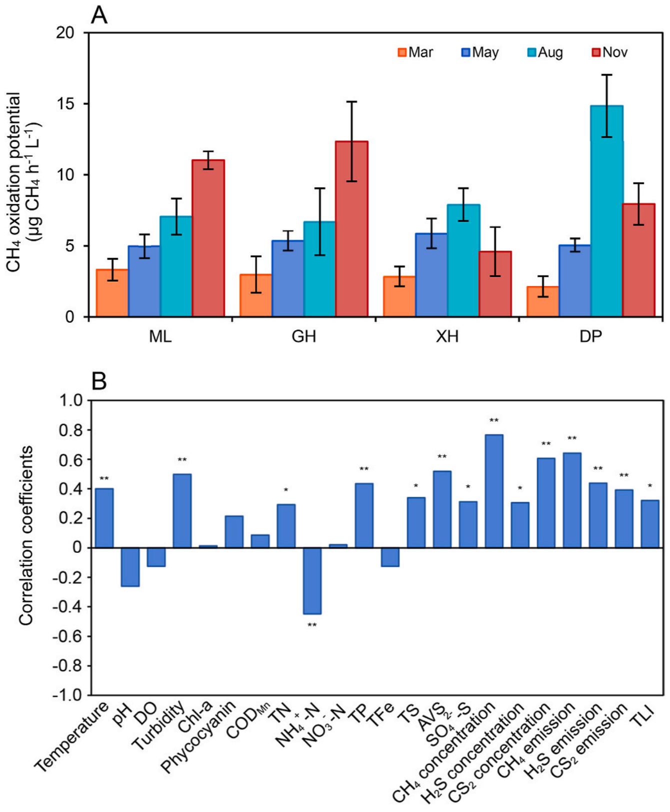

**Fig. 4.** Average $CH_4$ oxidation potential (MOP) of water at 0.5-m depth (A) and the correlation coefficients between MOP and environmental variables in Lake Taihu (B). * represents $P < 0.05$. ** represents $P < 0.01$.

### 3.3. Correlation between MOP and environmental variables

The MOP of water ranged from 2.1 to 3.3 μg $h^{-1}$ $L^{-1}$ in March, with no significant difference among the four regions (Fig. 4A). The MOP of water gradually increased to 11.0–12.3 μg $h^{-1}$ $L^{-1}$ in ML and GH in November. However, the highest MOP was detected at 7.9 and 14.9 μg $h^{-1}$ $L^{-1}$ in XH and DP in August, respectively. This was mainly because MOP was influenced by many factors, such as water quality, availability of trace metals, temperature, dissolved $CH_4$ and $O_2$ concentration (Schmale et al., 2015; Steinle et al., 2015; Crespo-Medina et al., 2014).

Statistical analysis showed that the MOP was significantly positively correlated with the environmental variables, including temperature, turbidity, TN, TS, AVS, $SO_4^{2-}$-S, TP, dissolved $CH_4$, $H_2S$ and $CS_2$ concentrations, while it was negatively correlated with $NH_4^+$-N concentration (Fig. 4B). Among the environmental variables, the MOP had the highest correlation coefficient with the dissolved $CH_4$ concentration. This was mainly because MOP followed Michaelis-Menten kinetics and increased with increasing $CH_4$ concentration before reaching substrate saturation (Wang et al., 2011). The MOP was positively correlated with the dissolved concentrations of $H_2S$ and $CS_2$. Previous studies have reported that VSCs such as $H_2S$, $(CH_3)_2S$, and $CH_3SH$ can inhibit $CH_4$ oxidation. The opposite result in this study might be attributed to 1) the relatively low VSC concentrations in water (Kim et al., 2013) and 2) the significantly positive correlation between the dissolved $CH_4$ and $CS_2$ concentrations, which indirectly induced the positive relation between $CS_2$ concentration and MOP. The concentrations of TN and TP can aggravate lake eutrophication and thus induce more $CH_4$ generation. Therefore, positive correlations were observed between MOP and the concentrations of TN and TP. Yang et al. (2020) indicated that the addition of $NH_4^+$-N could inhibit $CH_4$ oxidation in water and sediment. In this study, a high $NH_4^+$-N concentration was detected in the four water regions in March, mainly due to the low nitrification activity at low

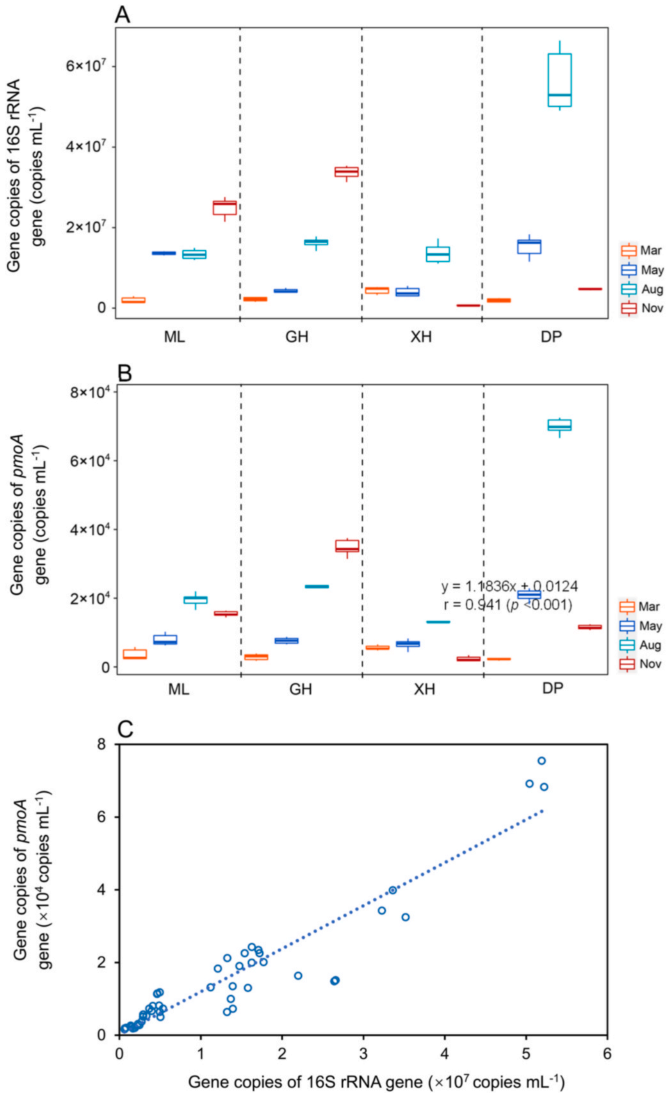

**Fig. 5.** Abundance of 16S rRNA gene (A) and *pmoA* (B) in water at 0.5-m depth and their relationship (C). Error bars indicate standard deviations (n = 27).

temperatures (Bruesewitz et al., 2009). Positive relationships were observed between the MOP and the emissions of $CH_4$ and VSCs, mainly because the dissolved $CH_4$ concentration was positively related to the emissions of $CH_4$ and VSCs in this study. Additionally, we also found the MOP was positively correlated with TLI. This indicated that lake eutrophication could promote $CH_4$ oxidation, likely owing to the high dissolved $CH_4$ concentration in eutrophic water.

### 3.4. Eutrophic levels increase abundance of bacteria and methanotrophs in water

The bacterial abundance varied with season in the investigated lake water (Fig. 5A). The bacterial abundance in ML and GH gradually increased from $1.95 \times 10^6$–$2.22 \times 10^6$ copies $mL^{-1}$ in March to $2.5 \times 10^7$–$3.4 \times 10^7$ copies $mL^{-1}$ in November. However, the highest bacterial abundance was observed in XH and DP in August. In November, the bacterial abundance decreased slightly in DP, likely due to the lowest DO concentration, which inhibited microbial growth (Yi et al., 2020). In XH, the bacterial abundance was relatively low with a slight increase in August, likely due to the high temperature simulating microbial growth. The difference in bacterial abundance was mainly attributed to temperature and/or eutrophic levels in the four study sites. Domaizon et al. (2013) reported that the abundance of *Cyanobacteria* increased with increasing temperature. Moreover, the abundance of algae increased with the eutrophic level of lakes, which might induce the higher bacterial abundance of ML and GH in November.

Compared with bacteria, the methanotrophic abundance was low in the lake water. Among the four investigated regions, the highest methanotrophic abundance was detected at $2.0 \times 10^4$ copies $mL^{-1}$ in DP in August, likely due to the high dissolved $CH_4$ concentrations in water (Fig. 5B). The methanotrophic abundance reached a peak value of $3.6 \times 10^4$ copies $mL^{-1}$ in GH in November. The methanotrophic abundance was lowest in XH due to the low $CH_4$ concentration and low eutrophic level. These results suggested that the methanotrophic abundance could be influenced by seasons (i.e., temperature) and dissolved $CH_4$ concentrations. Sundh et al. (2005) indicated that the methanotrophic abundance was higher in winter than in summer in Sweden, which was attributed to the lower temperature and the minimal inputs of phytoplankton-derived organic substrates that provided a conducive environment for methanotrophs while other bacteria were less competitive (Samad and Bertilsson, 2017). The highest methanotrophic abundance was observed in ML, XH and DP in August, which was consistent with the higher dissolved $CH_4$ concentration in water. A strong linear relationship was observed between the abundances of bacteria and methanotrophs (Fig. 5C), suggesting that the methanotrophic abundance varied with bacteria in the lake.

### 3.5. Microbial community succession in water

*Proteobacteria*, *Actinobacteriota*, *Cyanobacteria* and *Bacteroidota* were the predominant phyla in the lake water, accounting for 50.7%–96.7% of the total reads (Fig. 6). A similar result was found in East Lake in Wuhan, China, where the bacterial community was mainly composed of *Proteobacteria*, *Actinobacteria*, *Bacteroidetes* and *Cyanobacteria* with a relative abundance of 93.6% (Ji et al., 2019). The relative abundance of *Proteobacteria* varied considerably from 18.9% to 59.5% with a decrease to 12.9%–36.2% in GH and DP in August. In November, the relative abundance of *Proteobacteria* decreased to 9.3%–22.2% in ML and GH, while it increased slightly in DP. *Actinobacteriota* was also dominant in the freshwater system, probably due to their strong adaptability to the various environments (Neuenschwander et al., 2017). The relative abundance of *Actinobacteriota* in GH was 7.5%–20.0% in March and then increased to 24.8%–37.6% in August. The highest relative abundance of *Actinobacteriota* was 22.8%–40.5% in XH and DP in November. The relative abundance of *Cyanobacteria* in lakes reflects eutrophic levels (Ji et al., 2019). The relative abundance of *Cyanobacteria* in ML and GH was low in March and May and then increased to 38.9%–72.7% in November. The highest relative abundance of *Cyanobacteria* was 26.0%–51.3% in DP in August. Compared with the other three study sites, the relative abundance of *Cyanobacteria* was lower in XH. The seasonal variations in the relative abundance of *Cyanobacteria* were affected by temperature and available nutrients (Shan et al., 2019). The abundance of *Cyanobacteria* increased with increasing temperature and N and P concentrations (Xu et al., 2017; Tanvir et al., 2021), which was consistent with the results obtained in this study (Fig. 3H; Fig. 3K).

*Hgcl clade*, *Microcystis* PCC-7914 and *Aulacoseira granulate* var. *angustissima* were the most abundant genera in the lake water. Liu et al. (2015) also found that *hgcl clade* predominated in Pearl Estuary, China, likely due to its strong ability to utilize carbohydrates and compete for substrates. The relative abundance of *hgcl clade* was 1.1%–9.8% in March and lower than in the other months owing to the low temperature (Wang et al., 2021). The relative abundance of *hgcl clade* in ML, GH and XH increased to 12.1%–45.8% in May, while it decreased to 5.5%–22.0% in November. However, the relative abundance of *hgcl clade* increased to 19.5%–27.3% in DP in November owing to the high nutrients in the water (Xue et al., 2018) and their strong competition for organic matter (Liu et al., 2018). *Microcystis* is the predominant genus that induces water eutrophication (Cai et al., 2012). *Microcystis* PCC-7914 was not detected in March in the four investigated areas, but

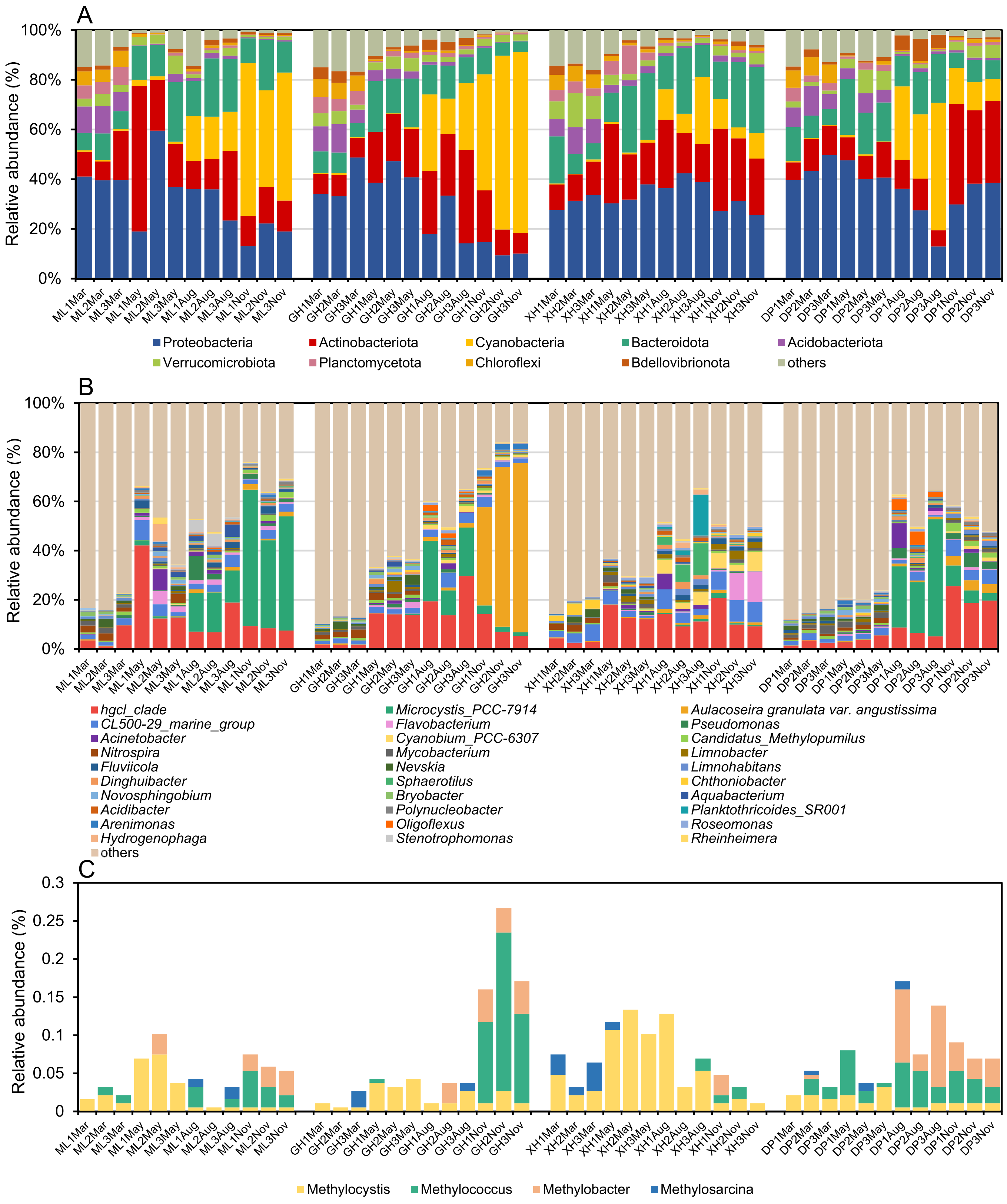

**Fig. 6.** The taxonomic classification of bacterial 16S rRNA gene reads at phylum (A) and genus (B) levels, and the relative abundance of methanotrophs in water at 0.5-m depth (C).

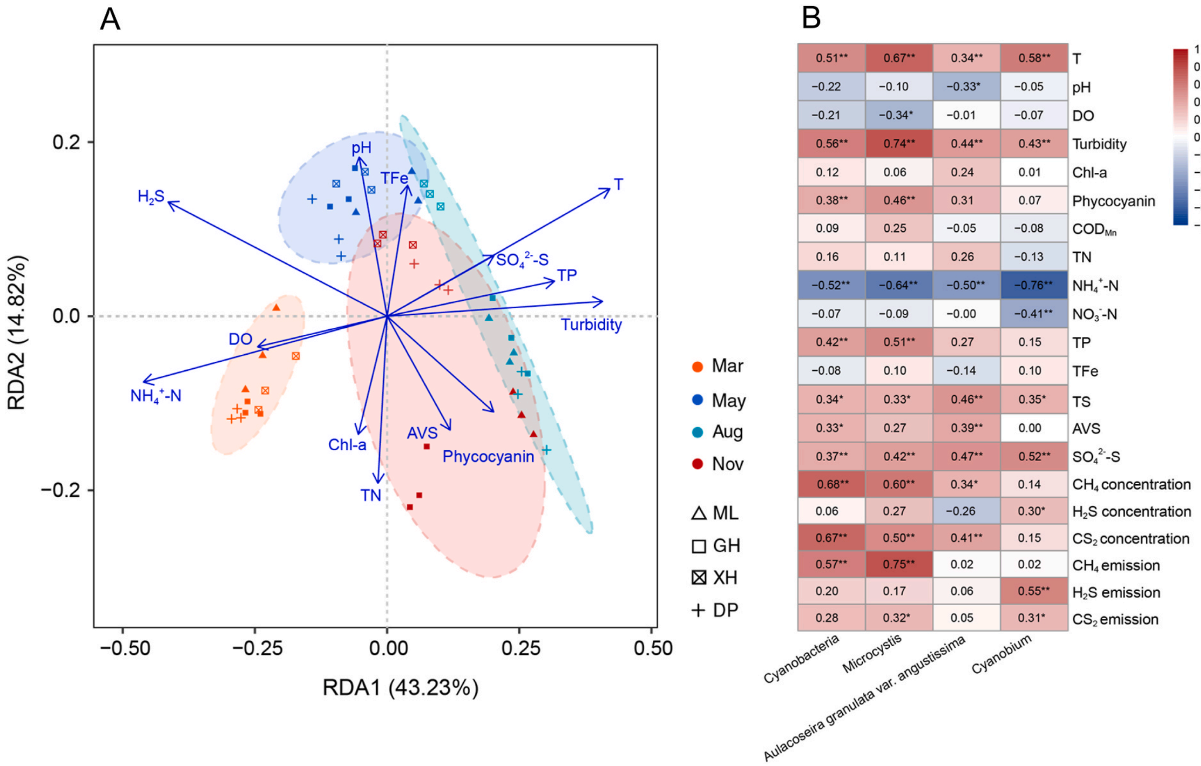

**Fig. 7.** Redundancy analysis (RDA) showing the relationships between microbial communities and environmental variables ($p < 0.05$) (A) and the correlations between the relative abundance of *Cyanobacteria*, *Microcystis*, *A. granulata* var. *angustissima* and *Cyanobium* and environmental variables (B). The numbers indicate the correlation coefficient. * represents $P < 0.05$. ** represents $P < 0.01$.

the relative abundance of *Microcystis* PCC-7914 increased to 0.1%–2.3% in May. Conroy et al. (2014) found that *Microcystis* could grow in March in Lake Erie and bloomed in May. Compared to Lake Erie, the growth and blooms of *Microcystis* were slightly late in Lake Taihu. The relative abundance of *Microcystis* PCC-7914 significantly increased to 10.7%–48.0% in August, indicating that the serious algae pollution appeared likely due to the high nutrients and suitable temperature (Song et al., 2017). *Aulacoseira granulata* var. *angustissima* is a diatom that can cause eutrophication in freshwater (Hynynen et al., 2004). The relative abundance of *A. granulata* var. *angustissima* was detected at 0.7%–70.1% in November, especially in GH, with the relative abundance of 41.6%–70.1%. Previous studies have also reported that the relative abundance of *A. granulata* var. *angustissima* increased in winter and was higher than that in the other seasons (Loudiki et al., 1994). Hynynen et al. (2004) found that the growth of *A. granulata* var. *angustissima* induced eutrophication in Lake Lievestuoreenjärvi with a relationship with the ratio of P/Si.

α-*Proteobacteria* methanotroph *Methylocystis* and γ-*Proteobacteria* methanotrophs *Methylococcus*, *Methylobacter*, *Methylosarcina* were detected in the water samples with low relative abundances of 0.01%–0.27%. Savvichev et al. (2021) also found that the relative abundance of methanotrophs was lower than 0.2% in lakes, and their community changed with seasons. In ML, GH and DP, α-*Proteobacteria* methanotroph *Methylocystis* was more abundant in March and May than γ-*Proteobacteria* methanotrophs, while it was less abundant in August and November. This might be because higher nutrients and organic matter are favourable for the growth of γ-*Proteobacteria* methanotrophs (Yang et al., 2011; Yang et al., 2016).

### 3.6. Influence of environmental variables on microbial community and $CH_4$ emissions

The influence of the environmental variables on the microbial community was estimated by RDA (Fig. 7A). The microbial community in the water samples grouped together in March, which separated from the other seasons, indicating an obvious difference in microbial communities in March. There was a positive relationship between the microbial communities and the concentrations of DO and $NH_4^+$-N in March. The microbial communities in May clustered together and were positively related to pH and the concentrations of TFe and $H_2S$. In August, the microbial communities in ML, GH, and DP grouped together, while those in XH were clustered nearly to May, probably due to the relatively low abundance of *Cyanobacteria* such as *Microcystis* PCC-7914. The microbial communities in November were relatively dispersed, which could be mainly attributed to the different eutrophic levels in the four sites.

The predominant *Cyanobacteria* were *Microcystis*, *A. granulata* var. *angustissima* and *Cyanobium* (Fig. 7B). Positive correlations were observed between the relative abundance of *Cyanobacteria* and temperature, turbidity, TS and $SO_4^{2-}$-S, indicating that these variables could promote algae growth. Yang et al. (2018) also found that an increase in temperature could prompt algae growth. In contrast, the $NH_4^+$-N concentration was negatively correlated with the algae abundance, which was in accordance with the result obtained by McCarthy et al. (2009). *Microcystis* is the main microorganism that induces algal blooms and red tides. There were negative correlations between DO and the relative abundance of *Microcystis*. In the initial stage of *Microcystis* growth, a large amount of $O_2$ could be produced due to the higher photosynthesis than respiration. However, excessive algae growth could prevent $O_2$ from entering water, and a large amount of $O_2$ was consumed during algae decay, which could decrease DO concentrations (Wang et al., 2021). There was a positive correlation between TP and the relative abundance of *Microcystis*, indicating that TP might be a key factor inducing algal blooms (Schindler et al., 2008; Song et al., 2017). The positive relationships between the dissolved $CH_4$ concentration and the relative abundances of *Cyanobacteria, Microcystis, A. granulata* var.

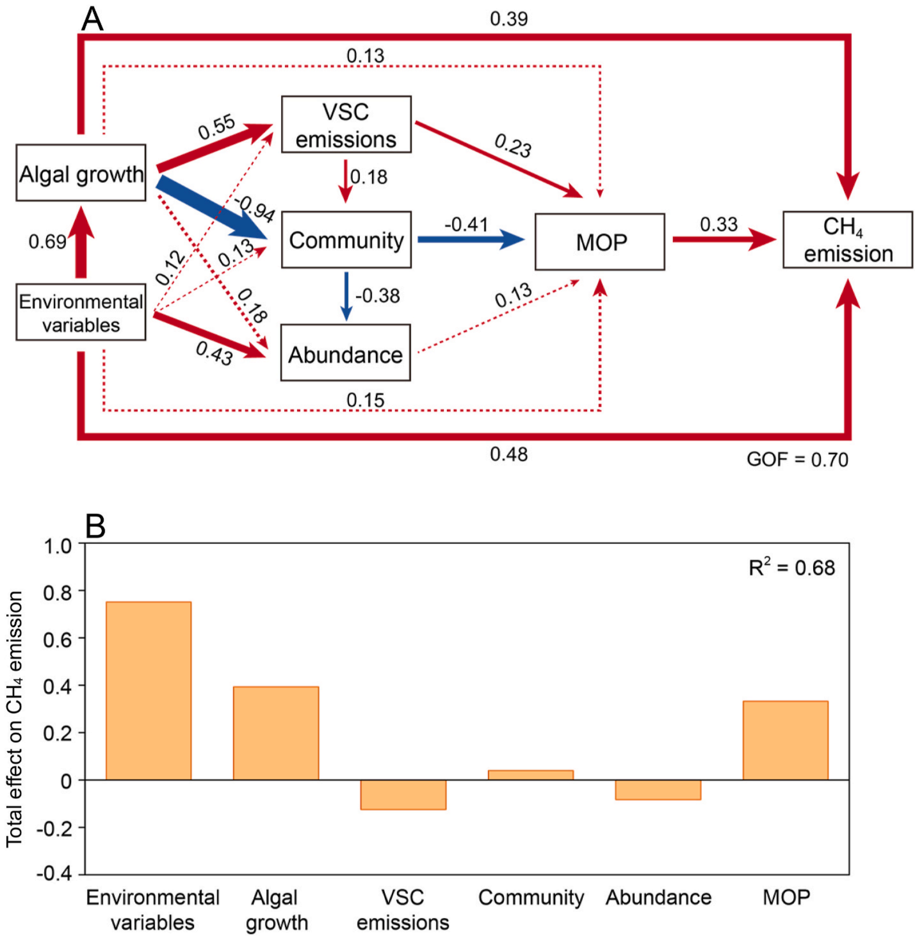

**Fig. 8.** Partial least squares path modelling (PLS-PM) presenting the direct and indirect effect of environmental variables and algae growth (i.e., the temporal and spatial relative abundance of algae obtained by MiSeq sequencing) (A) and the total effect of significant factors (B) on $CH_4$ emission from Lake Taihu. Red and blue arrows indicate positive and negative relationships, respectively. Continuous and dashed arrows indicate significant and nonsignificant relationships, respectively. The thickness of the lines indicates the strength of path coefficients. The values near the lines indicate the absolute values of path coefficients. $R^2$ indicates the values explained by the independent latent variables. Goodness of fit (GOF) indicates assessment of the model.

*angustissima* were observed. The dissolved $H_2S$ concentration was positively correlated with the relative abundance of *Cyanobium*, and the dissolved $CS_2$ concentration was positively correlated with the relative abundance of *Cyanobacteria*, *Microcystis* and *A. granulata* var. *angustissima*, indicating that different types of algae could affect VSC production. Similarly, we found there was significantly positive correlations between the relative abundance of *Cyanobacteria* and *Microcystis* and the $CH_4$ and VSC emissions. Of them, the relative abundance of *Cyanobium* were positively correlated with the $H_2S$ and $CS_2$ emissions, and the relative abundance of *Microcystis* was positively correlated with the $CS_2$ emission. These results indicated that algae growth could stimulate the $CH_4$ and VSC emissions. *Microcystis* and *Cyanobium* might be the predominant contributors to VSC emissions during algal blooms in freshwater lakes.

To deeply understand the influence of algae on $CH_4$ emissions, we conducted PLS-PM to explore the direct and indirect effects (Fig. 8). The model revealed that the environmental variables had positive effects on algal growth (i.e., the temporal and spatial relative abundance of algae obtained by MiSeq sequencing) with a path coefficient of 0.69, especially temperature, turbidity, $NH_4^+$-N and TP were significantly correlation with algal growth. The environmental variables and algal growth could both influence $CH_4$ emissions. Algal growth could promote VSC emissions, which had a correlation with MOP with a positive coefficient of 0.23. This might be because the $CH_4$ emission was positively related to the emissions of VSCs in this study (Fig. S1), and thus indirectly correlated with the MOP.

## 4. Conclusions

The $CH_4$ and VSC emissions from Lake Taihu increased with eutrophic levels and temperature. The MOP had significantly positive correlations with TLI and the environmental variables, including temperature, turbidity, TN, TS, AVS, $SO_4^{2-}$-S, TP, dissolved concentrations of $CH_4$, $H_2S$ and $CS_2$, while it was negatively correlated with the $NH_4^+$-N concentration. Eutrophic levels could increase the abundance of bacteria and methanotrophs in lake water. α-*Proteobacteria* methanotroph *Methylocystis* was more abundant in March and May than γ-*Proteobacteria* methanotroph, while it was less abundant in August and November. The relative abundance of *Cyanobacteria,* including *Microcystis*, *A. granulata* var. *angustissima* and *Cyanobium* had significantly positive correlations with temperature, turbidity, $SO_4^{2-}$-S and total sulfur. Algae growth could stimulate the $CH_4$ and VSC emissions. *Microcystis* and *Cyanobium* might be the predominant contributors to VSC emissions during algal blooms in freshwater lakes. These findings indicated improving water quality and controlling algal growth might be an effective way to reduce emissions of $CH_4$ and VSCs from lakes.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119435.

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