Austin Biometrics and Biostatistics



Austin Publishing Group

**Research Article**

# Extreme Methane Bubbling Emissions from a Subtropical Shallow Eutrophic Pond

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**Received:** September 29, 2014; **Accepted:** October 27, 2014; **Published:** November 01, 2014

**Abstract**

For the first time we report on real time diel bubble and diffusion gas fluxes lasting for 48 hours of a subtropical shallow pond. The averaged diffusion fluxes of methane and carbon dioxide were 0.074 and 62.70 mg•m$^{-2}$•h$^{-1}$, and the averaged ebullition fluxes of methane and carbon dioxide were 24.726 and 1.92 mg•m$^{-2}$•h$^{-1}$ respectively. Bubble emissions of $CH_4$ and $CO_2$ accounted for 99.7% of the total $CH_4$ emission and only 3.0% of the total $CO_2$ from the pond respectively. The $CH_4$ flux across the water-air interface of the pond was 595.20 mg•m$^{-2}$•d$^{-1}$ and equaled $CO_2$ flux of 14880.0 mg•m$^{-2}$•d$^{-1}$ by multiplying its global warming potential. Thus, the small pond added equivalent of 35.712 kg/d $CO_2$ emission by transferring $CO_2$ to $CH_4$ in the summer, in which process $CO_2$ was absorbed owing to alga propagation and $CH_4$ emission was derived from the anaerobic degradation of dead alga buried on its bottom.

**Keywords:** Methane; Carbon dioxide; Bubbling efflux; Diffusion; Eutrophic; Pond

## Introduction

Freshwaters can be substantial sources of $CO_2$ and $CH_4$ [1-3]. Lakes, ponds, and impoundments cover 3% of the earth's surface. Of them, natural lakes and ponds are estimated to cover about 4.2×10$^6$ km$^2$, whereas impoundments cover 2.6×10$^5$ km$^2$, and farm ponds cover about 7.7×10$^4$ km$^2$ [4]. Greenhouse gases emitted from lakes and reservoirs received respective attention [5-10]. However, less attention was paid to ponds, which are small in area and relatively shallow in depth but with various physical geographical characteristics and eutrophic situation. The importance of small lakes and ponds in the global lake area/number and in the global carbon cycle might have been underestimated [2,4,11]. Small wetland lakes and ponds that are often abundant in peat land areas can have high $CO_2$ and $CH_4$ emissions [12,13]. Moreover, it has been shown that the small areas of high $CH_4$ emissions, such as wetland ponds, can largely contribute to the landscape-scale $CH_4$ budgets in wetland regions and create a major uncertainty in the areal $CH_4$ emission estimates [14].

The rotation of the earth results in diel biogeochemical cycles, which are in response to the solar photocycle, particularly during stable hydrological conditions [15]. The amplitude of some of these diel changes can be as large as the changes occurring on annual timescales [15]. However, reports about gas fluxes on the diel timescale are scarce compared to those on the seasonal timescale. The former has received attention only more recently. To date, most reported gas ebullition rates were measured with bubble collectors [16-19], which could represent an average flux rate of a long monitoring period. No report could be found about the real-time gas ebullition emission. Study of diel variations is helpful to reveal which biogeochemical processes occur relatively rapidly in natural waters, and therefore which processes play an integral and important role in the normal functioning of natural water systems [15]. Data of diel greenhouse gas fluxes reported previously were at intervals of 3-4 h [20,21], and not continuous. Here, for the first time we present data of greenhouse gas flux from a field campaign lasting for 48 hours in the summer of 2013 regarding a subtropical hypertrophic pond. We hope the real time results can give some knowledge for recognizing the greenhouse gases emitting from small and shallow ponds both by ebullition and by diffusion.

## Materials and Methods

### Study area and monitoring sites

The pond (111°20'50.16"E, 30°44'30.978"N) is located at a suburban district of Yichang city, Hubei province, Central China. Yichang city is of a subtropical continental monsoon climate with a largely changing temperature in the spring, heavy rain and drought in the summer, wet weather in autumn and humid and snowy weather in the winter. The average annual temperature is 16.9 $^0$C, and the average rainfall is 1215.6 mm. The pond is about 2,500 m$^2$ with a maximum and mean water depth of 2.5 and 1.5 m respectively. It is surrounded by a small, locally known restaurant, and is a popular fishing area. The *total nitrogen and phosphorus concentrations* in waters are about 0.067 and 0.020 mg/L respectively. The pound bottom is covered with soft organic sediments (the total organic carbon content is 27.92 mg/g). The waters appeared green color owing to a great deal of the growing algae(mainly *Fragilaria, Scenedesmus quadricanda,* and *Oocystis*). We carried out diel flux measuring at two sites, which are 2 and 5 m away from its bank, and with depths of 1.2 and 1.5 m respectively (here marked as Site N and F). The field campaign lasted 48 hours from July 22 to 24, 2013. Here, we only report the results of Site N because of its representative water depth and location.




**Citation:** Xiao S, Liu W, Yang H, Liu D, Wang Y, Peng F, et al. Extreme Methane Bubbling Emissions from a Subtropical Shallow Eutrophic Pond. Austin Biom and Biostat. 2014;1(2): 6.

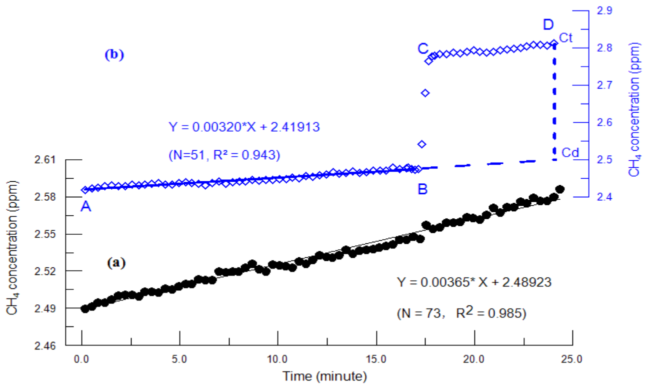


**Figure 1:** Curve Patterns of Changes in $CH_4$ concentration without bubble (a) and with bubble (b) during a single flux monitoring period.

### In situ sampling measurements and analysis

Surface and bottom water temperature (Ts and Tb), pH, air temperature (Ta), air pressure (Pa), intensity of illumination (Ii) and wind speed (Swi) were measured at the sites. Water temperature, the pH of water, and dissolved oxygen concentration (DO) in water were measured using the multi-parameter instrument Orion Star A329 (the United States). Water samples were taken from depths of 0.1 m below the water surface and 0.1 m above the bottom respectively for analysis of chlorophyll a concentration (Chl-a), and dissolved $CH_4$ and $CO_2$. Water samples (350 mL) were collected and transported to the laboratory for Chl-a analysis using the national standard method [22]. A headspace equilibration technique was used to quantify dissolved gas concentrations in water, and dissolved gas concentration in water is calculated according to the equation given by Johnson et al. [23].

### Water-to-air fluxes

A dynamic closed floating chamber system was used to measure diel $CH_4$ and $CO_2$ flux across the water-air interface. The chambers are non-transparent, thermally insulated tubes with a volume of 43.30 L and a surface area of 0.096 $m^2$ (diameter and height are 0.35 and 0.45 m respectively). Fans inside the chambers were applied to obtain better mixing of the air inside the chamber headspace. When we measured the emission flux of $CH_4$ and $CO_2$ across the water-air interface, one chamber was connected to a Los Gatos Research's Greenhouse Gas Analyzer (DLT-100) (Los Gatos Research, USA), which could monitor the $CH_4$ and $CO_2$ concentration inside the chamber continuously with 1Hz frequency. The DLT-100 is a cavity ring down spectrometer with high resolution (0.1 ppb) and precision (1% of reading the accuracy) and was already described in detail and used by previous researchers [8,9,24-28]. A single flux measurement was usually finished in 30 minutes. The chamber was then taken off the water surface to ensure adequate exchanging and mixing between gas inside the chamber and the environmental air. A separate chamber at the other site was connected to the DLT-100 Analyzer to continue the flux measuring. Thus, we alternated between monitoring sites N and F.

When there is no or little gas bubbles present in the chamber, concentrations of $CH_4$ and $CO_2$ gradually curve over time in a straight line due to the increase or decrease of gas concentrations in the chamber (Figure1a). Under this situation, a simple linear regression method is used to calculate the releasing rate and the flux of gases, which was described in detail by Lambert and Fréchette [29]. The gas concentration in the chamber will increase abruptly when bubbling occurs (Figure1b). We can separate the bubble and diffusion flux with data acquired by the DLT-100 Analyzer thanks to its high sampling frequency (See the captions of Figure 1b) in (b): AB & CD–diffusing, BC-bubbling. The equation ($Y = 0.00320*X + 2.41913$) is acquired by linear fitting based on data of AB; Ct is the measured $CH_4$ concentration at the monitored endpoint; Cd is the calculated $CH_4$ concentration at the monitored endpoint according to the fitted equation; and the surplus $CH_4$ concentration in the chamber resulted from bubble is equal to Ct minus Cd. The figure above displays the calculated averages derived from original data. Each data point is derived from 20 individual data entries.

## Results

### Variations in environmental factors

During the monitoring period, air temperature experienced a large net change of 12.3 °C (Table 1). However, surface and bottom water temperature changed only 5.7 and 4.4 °C respectively, and they showed a synchronal change (Figure 2). The differences between them were less than 2.0 °C due to the shallow water depth at the site. Surface and bottom water temperature had a similar average value, and was greater than the averaged air temperature.

Most environmental factors except the wind speed, the DO content in bottom water, and the pH of water appeared obviously diel changes, which corresponded to the earth's evolution and the solar radiation (Figure 2). Significant correlations between them were observed here.

An excess of nutrients, hot weather and motionless water resulted the algal bloom in the pond (Figure 2 and Table 1). Chl-a concentration in surface water was significantly positively correlated with Ii, which indicated the dependence of photosynthesis on solar

**Table 1:** Statistical description of main environmental factors.

| | Ta (°C) | Pa (KPa) | Swi (m/s) | L (Lux) | DO in water (mg/L) | | pH of water | | water temperature (°C) | | Chl-a (mg/L) | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | DOs | DOb | pH-s | pH-b | Ts | Tb | Chl-as | Chl-ab |
| Max | 37.1 | 98.97 | 0.99 | 78770 | 8.57 | 6.19 | 7.13 | 7.01 | 34.7 | 33.3 | 76.578 | 92.236 |
| Min | 24.8 | 98.52 | 0 | 0 | 2.44 | 2.23 | 6.07 | 5.52 | 29.0 | 29.9 | 48.724 | 45.232 |
| Average | 29.7 | 98.79 | 0.09 | 23412.88 | 5.02 | 3.99 | 6.60 | 6.63 | 31.1 | 31.1 | 60.021 | 64.640 |
| Stdev | 3.65 | 0.11 | 0.23 | 29538.56 | 1.61 | 0.98 | 0.23 | 0.27 | 1.17 | 0.82 | 7.50 | 11.28 |
| CV | 0.12 | 0.00 | 2.58 | 1.26 | 0.32 | 0.24 | 0.03 | 0.04 | 0.04 | 0.03 | 0.12 | 0.17 |

Stdev: standard deviation; CV: coefficient of variation. Dos and DOb: dissolved oxygen concentration in surface and bottom water; pHs and pHb: pH of surface and bottom water; Ts and Tb: surface and bottom water temperature; Chl-as and Chl-ab: Chl-a concentration in surface and bottom water respectively.

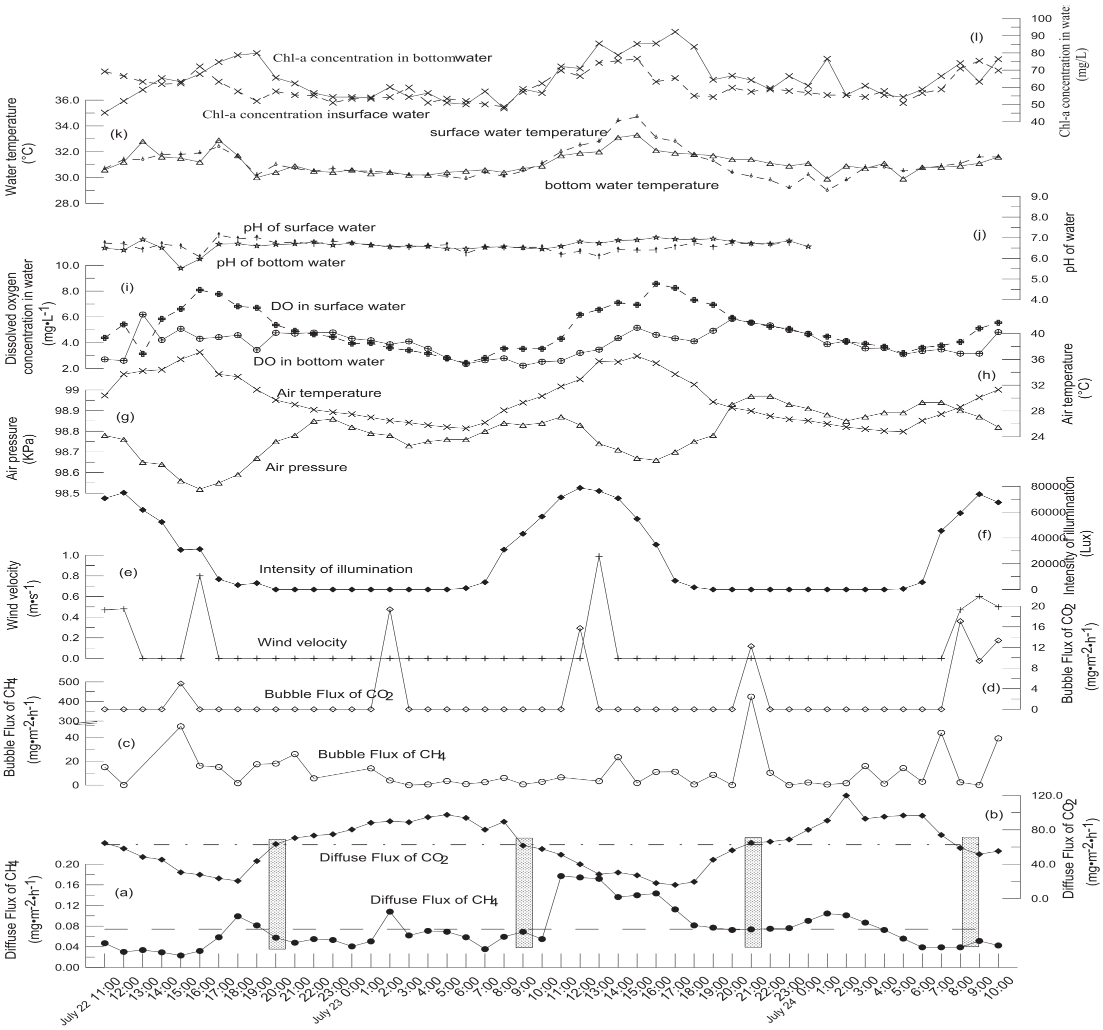


**Figure 2:** Diel $CH_4$ and $CO_2$ flux and environmental factors' changes during July 22-24, 2011in (a): dash-dotted and dashed lines represent the averaged diel diffusive $CH_4$ and $CO_2$ fluxes.

radiation. DO in surface water showed the similar changing tendency with the Chl-a concentration in surface water which resulted from the alternative of phytoplankton respiration of algae (Figure 2). Wind speed had the biggest coefficient of variation of all environmental factors, which might result from obstruction of low speed wind from the surrounding buildings.

## Variations in diel $CH_4$ and $CO_2$ flux

Both the diel diffusion $CH_4$ and $CO_2$ flux varied greatly during the monitoring period (Table 2). The diel diffusion $CO_2$ flux showed obviously synchronal changes with air temperature and surface temperature. The diel diffusion $CH_4$ flux fluctuated and changed gradually. The maximum values of diel diffusion $CH_4$ and $CO_2$ flux were 7.70 and 7.51 times of the respective minimum values. The natural variability of gas fluxes means that the number and time period of flux measurements is an important factor in obtaining an accurate average [30]. Here, the averaged values of both $CH_4$ and $CO_2$ flux occurred synchronously at around 9:00 and 20:00-21:00 of a day, which might give a referred time for single sampling when carrying out gas flux investigation in eutrophic water.

## $CH_4$ and $CO_2$ atmospheric fluxes

Both total $CH_4$ and $CO_2$ fluxes across the water-air interface at Site F were very high, and were 595.20 and 1550.79 mg•m$^{-2}$•d$^{-1}$ respectively. The $CH_4$ flux is 75.06 times of the overall average value across the water-air interface of the Three Gorges Reservoir [31] located in the same climate zone, and also much higher than the highest value (~150 mg•m$^{-2}$•d$^{-1}$) of multitude reservoirs documented from Lake Wohlen, a run-of-river hydropower reservoir located in Switzerland [32].

Bubble emissions of $CH_4$ accounted for 99.7% of the total $CH_4$ emission, which implied that $CH_4$ emitted into the air mainly through

**Table 2:** Variation of diel $CH_4$ and $CO_2$ fluxes.

| | Diffusive-$CH_4$ | Bubble-$CH_4$ | Diffusive-$CO_2$ | Bubble-$CO_2$ |
|---|---|---|---|---|
| Maximum (mg•m$^{-2}$•h$^{-1}$) | 0.177 | 424.284 | 119.84 | 19.37 |
| Minimum (mg•m$^{-2}$•h$^{-1}$) | 0.023 | 0.0 | 15.95 | 0.00 |
| Average (mg•m$^{-2}$•h$^{-1}$) | 0.074 | 24.726 | 62.70 | 1.92 |
| Stdev | 0.039 | 63.164 | 26.47 | 5.01 |
| CV | 0.525 | 2.555 | 0.42 | 2.61 |

**Table 3:** Correlation coefficients between *diel $CO_2$ and $CH_4$ diffusion flux* and the main environmental factors.

| | Ta | Pa | Swi | Ii | DOs | DOb | pH-s | pH-b | Ts | Tb | Diffusive-$CH_4$ | Diffusive-$CO_2$ | Chl-aSur | Chl-aBot |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Diffusive-$CH_4$ | 0.25 | 0.01 | -0.04 | 0.20 | .344 * | -0.02 | -.417 * | .454 ** | .435 ** | .389 ** | 1.00 | -.289 * | .304 * | .560 ** |
| Diffusive-$CO_2$ | -.904 ** | .588 ** | -0.26 | -.443 ** | -.856 ** | -.290 * | 0.09 | -0.13 | -.761 ** | -.738 ** | -.289 * | 1.00 | -.610 ** | -.749 ** |

**. Correlation is significant at the 0.01 level (2-tailed); *. Correlation is significant at the 0.05 level (2-tailed).

bubbles in the shallow eutrophic pond. However, bubble emissions of $CO_2$ only accounted for 3.0% of the total, which showed that the ebullition had no significance in the release of $CO_2$ from the pond. The result verifies that ebullition does not make a significant contribution to the release of $CO_2$ from shallow eutrophic freshwater systems to the atmosphere, in contrast to the situation for $CH_4$ where ebullition from them was significant [19]. The phenomenon was also observed in Petit Saut reservoir, where gas bubbles contained 50 to 80% $CH_4$ with few amounts (<1%) of $CO_2$ [16]. In the small hypertrophic freshwater Priest Pot, most $CH_4$ (96%) was also lost by ebullition, and most $CO_2$ (99%) by diffusive processes [33]. The ebullition $CH_4$ efflux was 593.42 mg•m$^{-2}$•d$^{-1}$, which is much bigger than reported lakes, reservoirs and ponds except that from the exposed Reservoir Lokka (seasonal average up to 656 mg•m$^{-2}$•d$^{-1}$) [34].

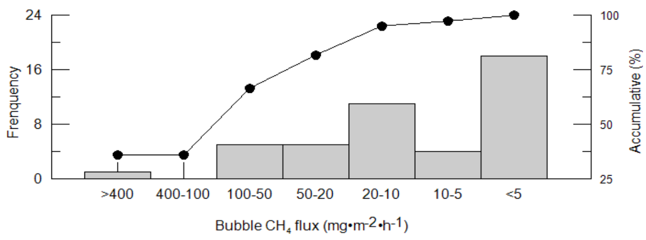


**Figure 3:** Frequency distribution of diel bubble $CH_4$ flux during July 22-24, 2011.

## Discussion

### Diel $CO_2$ and $CH_4$ diffusion flux

Diel $CO_2$ diffusion flux during the observing period was significantly positively correlated to the air pressure, and negatively correlated to the air temperature, intensity of illumination, DO in surface water, surface and bottom water temperature, and Chl-a in surface and bottom water (Table 3).

It's well known that the diel-scale inner coherence between air pressure, air temperature, water temperature and intensity of illumination, which results from the solar irradiation and the earth's evolution. Chl-a in surface water corresponded to the changing intensity of illumination with an alternative of photosynthesis and respiration. During the day when the rate of photosynthesis exceeds that of respiration, $CO_2$ is consumed and $O_2$ is produced. At night, in the absence of photosynthesis, respiration consumes $O_2$ and produces $CO_2$ [15]. Thus, significant correlations between the $CO_2$ diffusion flux and the Chl-a and DO in surface water were observed here.

### Diel $CH_4$ ebullition flux

No apparent time series regularity was found about changes of the diel ebullition $CH_4$ and $CO_2$ flux. $CH_4$ bubbled very frequently during the two days' field observing, and $CH_4$ ebullition occurred in 44 of all 48 monitored segmentations (91.67%). An especially big $CH_4$ bubbling event occurred at approximately 21:00 on July 23, 2013, which resulted in an abrupt increase of $CH_4$ concentration in the chamber from 6.64 to 539.93 ppm and the maximum bubble flux of 424.28 mg•m$^{-2}$•h$^{-1}$. All other ebullition $CH_4$ fluxes were lower than 85.0 mg•m$^{-2}$•h$^{-1}$ (Figure 3), and mainly located at 20-10 and <5 mg•m$^{-2}$•h$^{-1}$. Around 40.91% and 59.09% of total $CH_4$ flux was emitted in the first and second day respectively. $CH_4$ bubbling fluxes within 25% big rate accounted for 81.75% and the biggest $CH_4$ bubbling event accounted for 35.75% of the two days' whole bubbling $CH_4$ emission. This showed that ebullition was highly variable, and $CH_4$ ebullition fluxes measured by the chambers in a short time might underestimate real values severely. Otherwise, gas fluxes emitted during daytime were much lower than those during nighttime except diffusive $CH_4$ flux (Figure 4). So, it's necessary to extend monitoring time in frequent and episodic bubbling water system to acquire more exact gas fluxes.

The diel $CH_4$ flux was only significantly positively correlated to the DO in the bottom water (r=0.336, p<0.05, N=48), which might indicate that $CH_4$ was effectively oxidized by $O_2$. Although part $CH_4$ was consumed, $CH_4$ efflux was still high owing to the high $CH_4$ production resulted from high TOC content in sediments.

### C cycling mode of tropic shallow eutrophied impoundments

Overall, the growth of microalgae populations depends on three abiotic factors: available light, temperature, and level of nutrients such as nitrogen, phosphorus, and silicate (for diatoms) [35]. The present pond during monitoring periods satisfied all these three conditions. The good weather condition and the temperature of around 30 ºC were favorable to alga growth [36,37]. In case of many algae, maximal

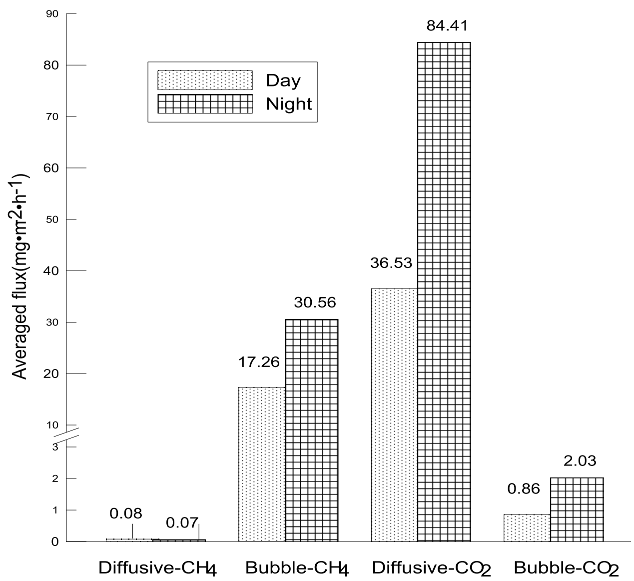


**Figure 4:** Comparison of gas fluxes between daytime (9:00-21:00) and nighttime (21:00-9:00).

growth rate are observed at optimum temperatures between 28 and 35° [38,39]. 30 °C had the best growth rates for both *Scenedesmus sp*. and *Chlorella* [40,41], which almost accorded with those observed here. The *total nitrogen and phosphorus concentrations* in waters are about 0.067 and 0.020 mg/L respectively. Nutrient loading increases autochthonous primary production in lakes, promoting oxygen consumption and anaerobic decomposition in the sediments [34].

Water mixing in the pond was well, and no vertical temperature stratification occurred. Sediments temperature during the observation was high and proximate to the surface water temperature, which could increase numbers of methanogenic bacteria and rates of methanogenesis [42,43].

Humans now strongly influence almost every major aquatic ecosystem, and their activities have dramatically altered the fluxes of growth-limiting nutrients from the landscape to receiving waters. On a global basis, strong correlations have been demonstrated between total phosphorus inputs and phytoplankton production in freshwaters, and between total nitrogen input and phytoplankton production in estuarine and marine waters [44]. Eutrophication causes predictable increases in the biomass of algae in lakes and reservoirs; streams and rivers; wetlands; and coastal marine ecosystems. Consistent and predictable Eutrophication-caused increases in algal blooms have been reported worldwide, which is a global problem [45-50]

Here, the $CH_4$ flux across the water-air interface of the pond was 595.20 $mg{\bullet}m^{-2}{\bullet}d^{-1}$, which equals $CO_2$ flux of 14880.0 $mg{\bullet}m^{-2}{\bullet}d^{-1}$ by multiplying its global warming potential of 25 over a time horizon of 100 yr [51]. Thus, the small pond added 35.712 kg/d $CO_2$ emission by transferring $CO_2$ to $CH_4$ in the summer. In eutrophic Reservoir Lokka, the relatively high $CH_4$ emissions were also primarily associated with the anaerobic decomposition of autochthonous, labile organic matter, rather than with decomposition of flooded old peat deposits [52]. Anoxia in eutrophic lakes favoring the $CH_4$ production is the major contributor to the atmospheric consequences of water Eutrophication [53].

Tropic eutrophic ponds satisfy all these three conditions (available light, temperature, and level of nutrients) to the growth of microalgae populations. Nutrient loading and anoxia in eutrophic lakes also promote oxygen consumption and anaerobic decomposition in the sediments, and together with high temperature favor the $CH_4$ production. Tropic shallow impoundments may serve as plants producing greenhouse gases by transferring $CO_2$ to $CH_4$ in the summer.

## Acknowledgment

This work was sponsored by National Science Foundation of China (No. 41273110, 51079163), and State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences.

## Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Austin Biom and Biostat - Volume 1 Issue 2 - 2014
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**Citation:** Xiao S, Liu W, Yang H, Liu D, Wang Y, Peng F, et al. Extreme Methane Bubbling Emissions from a Subtropical Shallow Eutrophic Pond. Austin Biom and Biostat. 2014;1(2): 6.