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Estimation of greenhouse gas fluxes in water based on convection–diffusion model

Xiufeng Quan[a], Ruijie Li[a,b,*], Dingguo Jiang[c,*], Huichao Dai[d], Feng Luo[a], Xiaoyan Fu[e]

[a]*College of Harbor Costal and Offshore Engineering, Hohai University, Nanjing 210000, China, email: rjli@hhu.edu.cn (R. Li)*
[b]*Key Laboratory of Coastal Disaster and Defense, Ministry of Education, Hohai University, Nanjing 210098, China*
[c]*China Three Gorges Corporation, Beijing 443002, China, email: jiangdingguo@aliyun.com (D. Jiang)*
[d]*College of Water Conservancy and Hydropower Engineering, Hohai University, Nanjing 210000, China*
[e]*College of Oceanography, Hohai University, Nanjing 210098, China*

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ABSTRACT

Due to the spatial and temporal differences in greenhouse gas emissions at the water–air interface of reservoirs, it is difficult to estimate the greenhouse gas emissions from the reservoir accurately. In order to explore a new method for estimating greenhouse gas fluxes from reservoirs, a vertical transport model of greenhouse gases in waters is constructed based on the convection–diffusion model, and a set of physical experiments are carried out to test the rationality of the new model. The results show that vertical distributions of gas concentrations measured in experiments are identified to simulation results of the convection–diffusion model, so it is necessary to consider the convection term in gas transport in waters. In addition, the gas fluxes calculated by the convection–diffusion model are compared with those measured by the static floating chamber method, and the maximum relative error is 21.43%.

Keywords: Greenhouse gas; Convection; Diffusion; Flux estimation

1. Introduction

Converting the potential energy of falling or fast-flowing water into mechanical energy, hydropower is a kind of renewable energy without consuming any fossil fuels, which could greatly reduce the emissions of greenhouse gases, such as CO_2 and CH_4 [1,2]. It is a green energy with obvious benefits of greenhouse gas emission reduction [3]. Under the environment of global warming, the hydropower development has become an important strategy for energy plans in various countries [4–7]. However, with disputes of greenhouse gases in reservoirs, the point that hydropower is a kind of green energy has been questioned [8,9], and development strategies of hydropower are also facing challenges [10].

Rudd first reported potential greenhouse gas emissions from reservoirs in 1993. And from then on, the issue of greenhouse gas emissions from reservoirs has been studied by many scholars [11–20]. Tropical reservoirs such as Balbina and Tucurui have been reported to have stronger greenhouse gas emissions than fossil-fuel power plants with equivalent levels [21–23]. Two schools have been conducting a 10-y academic debate on measurement schemes and uncertainties of greenhouse gas effects of reservoirs. Scholars have carried out plenty of long-term follow-up monitoring of greenhouse gas effects of reservoirs [24,25], which makes the technology of greenhouse gas monitoring fully developed [15]. Researches on greenhouse gas emissions from reservoirs with scientific methods are of great significance.

* Corresponding authors.

At present, greenhouse gas monitoring techniques are mainly divided into three categories [26]. The first type is to estimate the flux by measuring the concentration of dissolved gas in waters, such as water chemical equilibrium calculation, method of thin boundary layer and carbon isotope analysis [27–29]. These methods requiring sampling and analysis of the surface water in the monitoring water area and monitoring equipment with higher accuracy are the early methods used in practice. In addition, the corresponding post-calculation work is relatively heavy. The second-type is to calculate the flux by measuring the greenhouse gases passing through the water–air interface. The typical static floating chamber method is the most mature [30]. The third type is to calculate the flux by measuring the cumulative concentration of greenhouse gases above the water surface, such as TDLAS laser monitoring method and eddy covariance method [31–34]. These methods are seldom used in practice currently, because of their high demands of application environments and the sensitive of monitoring results to the disturbance on the water surface. Obviously, GHG flux estimation is generally conducted at the water–air interface. However, due to the temporal and spatial differences in greenhouse gas flux emissions at the water–air interface, there are uncertainties in the estimation results of these popular used methods [35]. The estimation results of various methods are difficult to be unified [36], especially the static floating chamber and the method of thin boundary layer which usually have significant differences [27]. Therefore, in order to reduce the disturbance of uncertain factors such as wind and waves at the water–air interface [35], new estimation methods of greenhouse gas of reservoirs need to be explored.

Taking the greenhouse gas carbon dioxide as a research object, a new method for estimating greenhouse gas emissions is put forward to study the vertical transport of greenhouse gases in waters by utilizing theoretical analyses and flume experiments together in the paper. First, based on the theoretical analysis, a convection–diffusion model for the vertical transport of greenhouse gases in waters is constructed. Then a comparative analysis of the new model with the typical diffusion model shows the new model is more rational in describing the process of the greenhouse gas transport [37]. In addition, flume experiments are designed and implemented to verify the existence of the vertical convection term in gas transport in waters. Finally, the greenhouse gas flux is calculated based on the new convection–diffusion model, and the accuracy of the new method is discussed.

2. Material and methods

2.1. Theoretical model

Assuming that the gas migrates and diffuses in a two-dimensional flow, for a water element of length Δx, height Δz, and unit thickness, the inflow and outflow mass of the gas in the time interval Δt can be expressed as shown in Fig. 1. The gas has a specific weight smaller than that of water, it will move upward with some velocity ω_0.

In Fig. 1, u and w are the instantaneous velocity of water in x- and z-direction, respectively. ε_x and ε_z are the gas diffusion coefficient in x- and z-direction, respectively. C is the concentration of gas in the water.

From the law of mass conservation, the concentration of gas in the control volume element varies with time if the inflow and outflow rates are not equal.

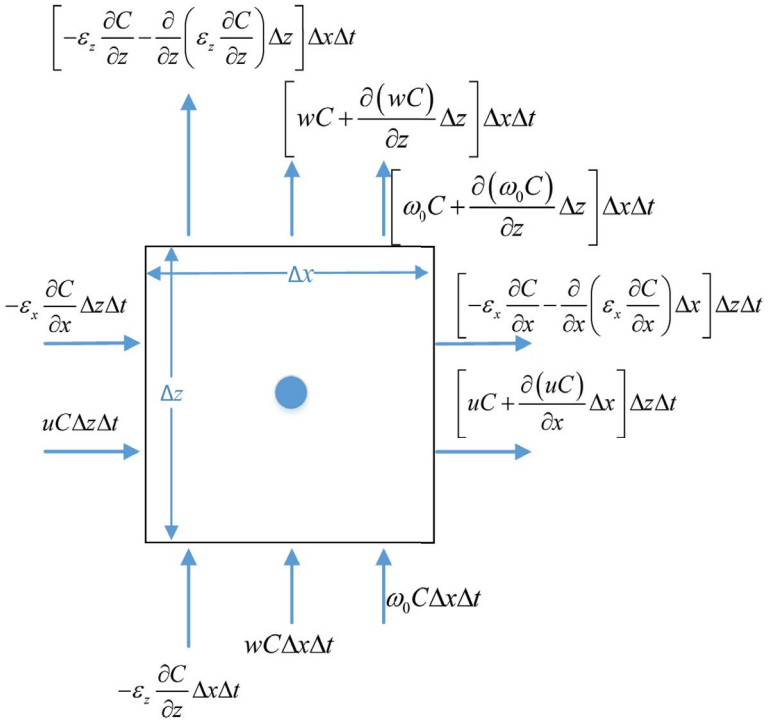

Fig. 1. Definition sketch for derivation of two-dimensional continuity equation of gas in a control volume element.

The net mass flux out of the control volume in x-direction:

$$\frac{\partial F_x}{\partial x} = u\frac{\partial C}{\partial x} - \varepsilon_x \frac{\partial^2 C}{\partial x^2} \tag{1}$$

The net mass flux out of the control volume in z-direction:

$$\frac{\partial F_z}{\partial z} = w\frac{\partial C}{\partial z} + \omega_0 \frac{\partial C}{\partial z} - \varepsilon_z \frac{\partial^2 C}{\partial z^2} \tag{2}$$

According to the law of conservation of mass:

$$\frac{\partial C}{\partial t} + \frac{\partial F_x}{\partial x} + \frac{\partial F_z}{\partial z} = 0 \tag{3}$$

Substituting Eqs. (1) and (2) into Eq. (3), yields:

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} + \left(w + \omega_0\right)\frac{\partial C}{\partial z} = \varepsilon_x \frac{\partial^2 C}{\partial x^2} + \varepsilon_z \frac{\partial^2 C}{\partial z^2} \tag{4}$$

Assuming $\varepsilon_x = \varepsilon_z = D$, Eq. (4) can be further simplified as:

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial x} + \omega\frac{\partial C}{\partial z} = D\frac{\partial^2 C}{\partial x^2} + D\frac{\partial^2 C}{\partial z^2} \tag{5}$$

When only the vertical direction is considered $\left(\frac{\partial C}{\partial x} = 0\right)$, Eq. (5) can be further simplified as:

$$\frac{\partial C}{\partial t} + \omega\frac{\partial C}{\partial z} = D\frac{\partial^2 C}{\partial z^2} \tag{6}$$

Eq. (6) is the equation of one-dimensional gas transport in vertical.

According to the one-dimensional vertical convection–diffusion equation, gas fluxes in waters can be expressed as the sum of convection and diffusion fluxes, as follows:

$$F = \omega C - D\frac{\partial C}{\partial z} \tag{7}$$

In this formula, F is the flux of dissolved gas in a certain depth of water. And when the vertical distribution of dissolved gas, the diffusion coefficient, and the vertical migration velocity are known, the flux of dissolved gas at any height in the water can be estimated according to the flux formula.

In Eq. (7), the vertical migration velocity ω includes the vertical velocity w of water and the vertical migration velocity of gas ω_0. In the still water state, the vertical velocity of water is zero, and the vertical migration velocity ω in Eq. (7) is the additional vertical migration velocity obtained by the gas due to buoyancy ($\omega = \omega_0$). At the same time, the diffusion coefficient D is constant in the steady state. It is worth noting that the object of flux estimation is not bubbles with significant size, but microscopic gas cores with very small diameter. The group effect caused by the migration of gas nuclei cannot be observed directly, but can only be verified indirectly by the change of gas concentration in water.

2.2. Experiment setup for vertical migration of gas in waters

The vertical migration experiment of greenhouse gases in waters was designed to verify the existence of the convection term. The object of the experiment was to collect the data of gas concentration at different water depth and gas flux at water–air interface in different periods after the initial concentration is uniformly distributed by mixing the gas in the water tank. Then the vertical distribution of gas in the water was analyzed and the gas fluxes at water surface was calculated and compared with those estimated by the vertical transport model, which verified the existence of the convection term.

The experiment equipment consisted of a gas generation module, a water gas mixing module and a monitoring and sampling module, as shown in Fig. 2. The basic operation steps were as follows: firstly, fill the water tank of a volume of 1 m^3 with water. Secondly, turn on the pressure-regulating valve on the gas storage tank to spread the gas CO_2 to the whole water tank through two nano gas disks with a diameter of 20 cm fixed at the bottom of the tank for 1 min (forming the supersaturation of the gas in the tank water) and then turn it off. Thirdly, turn on the small pump in the

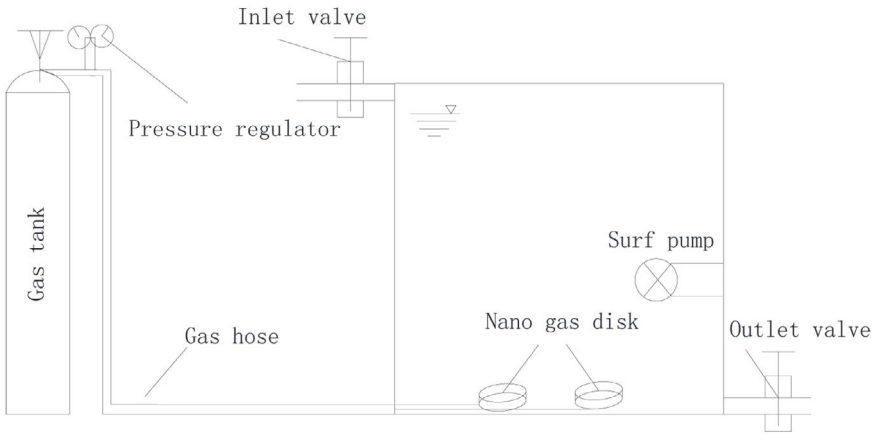

Fig. 2. Design of experiment devices.

water tank for a short period to ensure the water air was well mixed, and rest the water for 3 h. Finally start to collect the demand data after the water was stable.

The gas concentration in the water was measured by the headspace balance method. Ten sample points were set up along the water depth with an interval of 10 cm. The first sample point was at water surface. Two samples were taken from all the points at the same time to avoid operation errors which might affect the accuracy of experiments. Considering the stability of the experiment condition, the sampling interval was determined to be 3 h and 5 groups of samples were taken. The volume of water sample was 100 mL. After sampling, the sample bags were shaken with ultrasonic water bath for 15 min. After resting them for 24 h, the sample was analyzed by gas chromatograph and converted into the actual gas concentration in the water. The gas chromatograph (Agilent 7890a) was equipped with flame ionization detector (FID) and N2 was used as the carrier gas. The flow rate was 20 ml/min and the column temperature of TDX-O_2 column was 90°C. The detector temperature was 250°C, and the relative error was less than 1 nmol/L. During the sampling process, the gas flux at water surface was monitored simultaneously. The gas flux at water surface was measured by a static floating chamber. In the experiment, a flux chamber with height of 25 cm (the traditional flux chamber is generally 50 cm high) was used to speed up the measurement of the change rate of gas concentration. Before each sampling, the chamber opening was placed upward for about 5 min to ensure the fully exchange of the gas in the chamber with the ambient air. Then the chamber was placed at the water surface with its opening downward submerged in the water for sampling, which could ensure the gas in the chamber was isolated from the ambient air. The chamber was connected to an LGR-100 fast greenhouse gas analyzer of which measuring range of the greenhouse gas CO_2 is 200 × 10^{-6} to 4,000 × 10^{-6} with an accuracy of less than 1% of the reading number.

3. Results and analysis

3.1. Experimental results of water–air interface flux

The gas flux at water–air interface was measured by the static floating chamber. The results are shown in Fig. 3. The average flux of gas CO_2 at water–air interface is about 2.98 mmol·(m^2·h)$^{-1}$. The maximum fluctuation of the gas flux in the five groups is only 5.58%, indicating that the gas flux at the water air interface is relatively stable. The indoor experiment was carried out under no wind condition, because the measurement accuracy of the static floating chamber could be easily affected by winds. The measurement result of the static floating chamber is the actual flux.

The measurement accuracy of the static floating chamber is affected by the calibration of the change rate of gas concentration in the chamber. In the experiment, the change of gas concentration gradient in the chamber is given in Fig. 4. Compared with the linear regression fitting, the exponential regression fitting is more accurate for the change rate of gas concentration. The results are consistent with the conclusion of Xiao et al. [30], and the exponential regression fitting can improve the measurement accuracy of the static floating chamber. Therefore, in this experiment, exponential regression is used to fit the change rate of gas concentration in the process of flux measurement by static floating chamber method.

When the floating chamber method is used to estimate the gas flux at water–air interface, the gas concentration gradient shows an increasing trend with monitoring time (K_1 < K_2 < K_3), which does not conform to Fick diffusion law. If the convective term is negligible in gas transport in the water and only the diffusion term caused by concentration gradient exists, the difference between the gas concentration in the water and in flux box should gradually decrease with the flux exchange between water and air, and the rate of concentration accumulation will decrease. However, the flux monitoring data of carbon dioxide gas under actual working conditions is obviously inconsistent with it. Obviously, from the point of gradual increase of gas change rate in the flux box, there should be not only diffusion transport caused by concentration gradient, but also convective transport. The convective transport is not

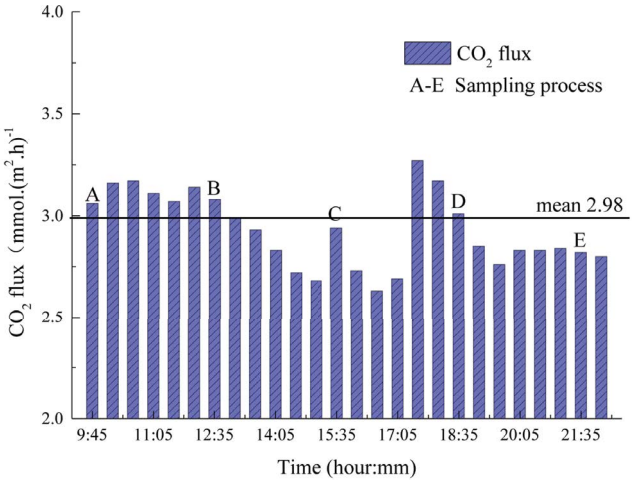

Fig. 3. CO_2 gas flux at water air interface.

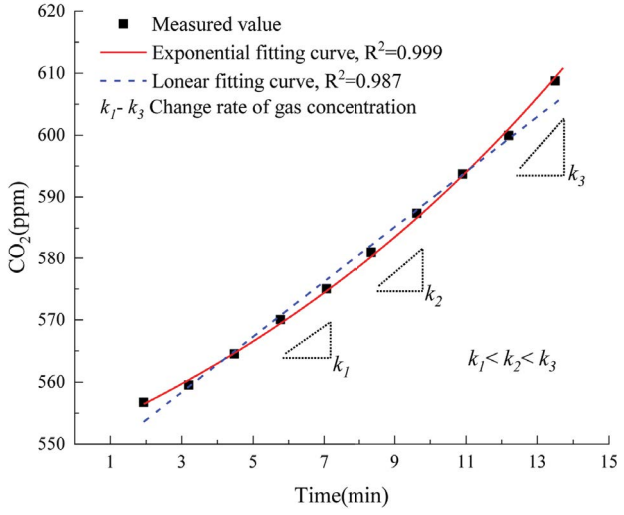

Fig. 4. Change rate of gas concentration in the chamber.

related to the concentration difference between the two sides of the water air interface, but may be related to the gas concentration in the water.

3.2. Experimental results of gas vertical migration in the water

The headspace equilibrium method was used to measure the gas concentration in water, and then the distribution of gas concentration was analyzed to verify the existence of the convection term. Results of the gas concentration in water are shown in Table 1.

The distribution of carbon dioxide concentration in the water has a significant regularity. At the initial moment, the distribution is uniform, and then the distribution of gas concentration changes gradually in the vertical. The gas concentration increases gradually with z to a certain height and then turns to decrease gradually toward the water–air interface. During the sampling process, even if the total amount of gas in the water is estimated with the maximum emission rate, the actual cumulative emission in 12 h is less than 0.038%, and the gas in the water is still in the state of supersaturation after a short period, which can be regarded as a stable state during the experiment.

The data of group E under the still water condition were selected for analysis. Assuming that the gas transport in the water conforms to the convection diffusion model, the convection coefficient and diffusion coefficient of the convection diffusion model are solved by the least square method, which are 0.0033 m/h and 0.0021 m^2/h, respectively, and the mean square error is 9.94. On the contrary, if the gas transport in the water does not include convection, the analytical solution of diffusion model is used to analyze the vertical distribution of gas concentration, and the mean square error is 930.56. Based on the first four groups of data to calibrate the coefficient, the concentration distribution fitting of group E is shown in Fig. 5. Obviously, compared with the diffusion model, the vertical distribution of gas concentration based on the diffusion model is more consistent with the measured results, and the convection term does exist in the vertical transport of gas in waters.

In order to analyze the influence of convection term on the vertical distribution of gas in the water, a numerical model of vertical transport of gas is built based on diffusion model and convection diffusion model, respectively, and the vertical distribution of gas concentration in the water is simulated under different levels. The second-order temporal and spatial precision crank Nicolson scheme is used to construct the numerical model. The initial distribution of gas concentration is set to 800 umol·L^{-1}. The emission flux is set to 10 mmol·$(m^2 \cdot h)^{-1}$ at water–air interface and zero at the bottom. The convection coefficient and diffusion coefficient are set with different values, respectively. The simulation results are as follows.

Obviously, there are differences in the distribution of gas concentration described by the model under the numerical simulation conditions that the vertical transport rate is in different proportion to the diffusion rate. When the convection coefficient is greater than or equal to the diffusion coefficient (as shown in Fig. 6a and b), the vertical distribution of gas in water changes significantly compared with that obtained under only diffusion. In addition, the concentration distribution of gas shows a trend of a small concentration at the upper and lower part of the water column and a larger concentration in the middle. In other words, when the gas concentration in the water is small in the upper and bottom layers and larger in the middle layer of the water column, the gas convection rate in the water should be considered at least at the same level as the diffusion rate,

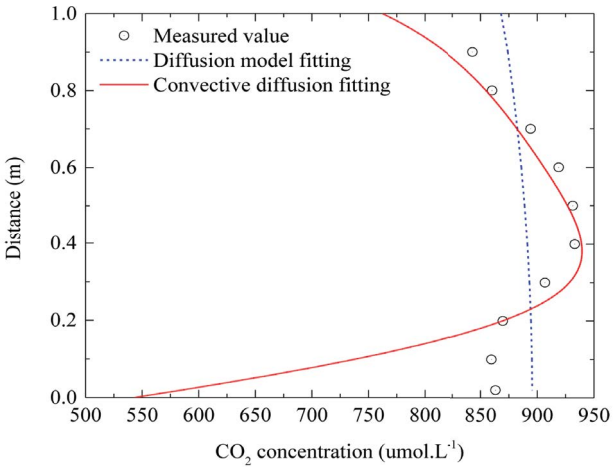

Fig. 5. Regression fitting of CO_2 gas concentration distribution.

Table 1
Concentration of CO_2 in the water measured during experiments (umol L^{-1})

Water depth (m)	*A*	*B*	*C*	*D*	*E*
0	860.3981	872.0716	816.9183	844.6716	842.8396
0.1	861.964	890.8328	823.1939	864.6971	860.1911
0.2	865.9965	909.5502	829.9926	890.5835	894.2038
0.3	864.6846	909.7614	839.1182	887.376	918.9184
0.4	851.981	914.5122	848.9410	866.2226	931.3086
0.5	845.5375	913.6658	867.5421	852.3673	933.0397
0.6	876.5691	887.2432	900.4848	855.5248	906.8379
0.7	904.9655	863.5438	906.5472	866.9511	869.6546
0.8	902.9132	882.8184	907.7233	867.0158	859.6427
0.9	901.0141	904.7122	918.399	860.5819	863.2178

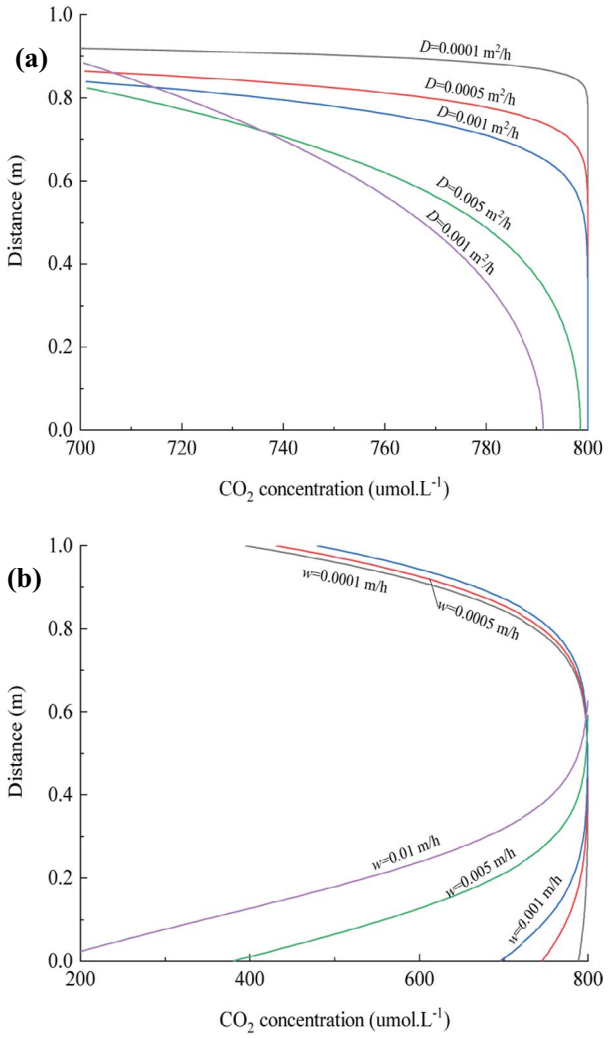

Fig. 6. Numerical simulation of vertical distribution of gas concentration. (a) Diffusion model and (b) convection–diffusion model.

which cannot be ignored. When analyzing the gas transport in the water, it is reasonable to consider the existence of convection term in theory.

3.3. Flux estimation results of different methods

The convection–diffusion model and the static floating chamber method are used to estimate the gas flux, as shown in Table 2. The average flux of gas at the water–air interface measured by the new method is 2.87 mmol·(m²·h)$^{-1}$, which has a maximum relative error of 3.92% compared with the flux determined by the static float chamber method.

4. Conclusions

The research in this paper shows that the transport of greenhouse gases in waters conforms to the convection–diffusion model. Through numerical simulation and flume experiments, it is verified that the convection term has a significant effect on the gas concentration distribution in the water. In particular, the convection term should not be ignored when estimating the gas fluxes in the water. Based on the convection–diffusion model, we propose a new method for estimating greenhouse gas fluxes in waters. The new method avoids the spatiotemporal uncertainty which may be caused in the traditional flux estimation methods, and the estimation error of the static floating chamber method is only 21.43%. This research can be provided as a support and basis for the accurate estimation of greenhouse gas emission from reservoirs.

Table 2
Flux estimation results of different methods

Sampling process	SFCM (mmol·(m²·h)$^{-1}$)	CDM (mmol·(m²·h)$^{-1}$)	Relative error
A	3.06	2.73	0.1083
B	3.08	3.13	0.0147
C	2.94	2.31	0.2143
D	3.01	3.46	0.1484
E	2.82	2.70	0.0411

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