

Experimental study of the impacts of external disturbances on N₂O emission from water to air

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ABSTRACT

To obtain a comprehensive understanding of nitrous oxide (N₂O) emission from water to air, the impacts of external disturbances including wind, stirring and aeration on N₂O emissions were investigated by continuously monitoring N₂O concentration variations in water. The volumetric mass transfer coefficient of N₂O from water to air (K_{LaN_2O}) under different conditions was determined by using exponential regression to fit the monitoring data. The results showed that K_{LaN_2O} was 0.0017 min⁻¹ at the stable condition, and with the increase of wind velocity, stirring velocity and aeration rate, the diffusion of N₂O from water to air was enhanced. It was also observed that K_{LaN_2O} linearly increased with the increase of wind velocity and aeration rate, and exponentially increased with the increase of stirring velocity. The sequencing of the impacts of the three factors on N₂O emission was aeration > stirring > wind. As turbulence and the mixing intensity of the liquid phase under the aeration condition were more vigorous than those of the wind and the stirring, the impact of aeration on N₂O emission was greater than those of wind and stirring. When predicting N₂O emissions from water to air, external disturbances, either environmental factors or operational factors, should be taken into consideration.

Key words | emission, external disturbance, impact, nitrous oxide, volumetric mass transfer coefficient

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INTRODUCTION

As a potent greenhouse gas (GHG), nitrous oxide (N₂O) not only has a strong greenhouse effect, approximately 300-fold stronger effect than carbon dioxide (IPCC 2001), but it is also an important ozone-depleting substance (Ravishankara *et al.* 2009). In global non-CO₂ GHG emissions, methane (CH₄) was first, and N₂O second highest (USEPA 2012). The atmospheric abundance of N₂O in the mid-nineteenth century was around 275 ppb, while this value is more than 320 ppb at present (UNEP 2013). Human activities such as agriculture, biomass burning, industry and fossil fuel combustion, wastewater treatment, and so on, have lead to the consequent increase in N₂O emissions (UNEP 2013).

N₂O generation and emission vary considerably across different wastewater treatment plants (Kampschreur *et al.* 2009; Ahn *et al.* 2010; Foley *et al.* 2010). During wastewater treatment, N₂O can be produced by the various activities of microorganisms: (1) denitrification by ammonia-oxidizing bacteria during nitrification (Wrage *et al.* 2001); (2) oxidation of hydroxylamine (NH₂OH) (Yu *et al.* 2010); (3) reduction of

nitrite (NO₂) by heterotrophic bacteria under anoxic conditions (Wunderlin *et al.* 2013). N₂O emissions in aerobic zones were larger than those in anoxic zones (Ahn *et al.* 2010). Low dissolved oxygen (DO) concentration in the nitrification stage, increased nitrite concentration and low COD/N ratio in the denitrification stage caused an increase in N₂O emissions (Kampschreur *et al.* 2009).

As N₂O is produced in nitrification and denitrification, there are two pathways to reduce N₂O content in water. One is a bio-consumption process, and the other is a physical emission process. In the bio-consumption process, N₂O is reduced to dinitrogen (N₂) by microorganisms. In the physical emission process, N₂O transfers from a liquid phase to a gas phase via the liquid-gas interface. While differentiating the N₂O loss caused by diffusion and bio-consumption, it is important to evaluate N₂O emission from the biological wastewater treatment process and to explore effective methods for controlling N₂O emissions. The mechanism of the physical process can be

elucidated by the two-film theory (Lewis & Whitman 1924). The transferred mass of N₂O through the interface depends on two factors: the driving force causing N₂O transfer between the gas and liquid phases and the volumetric mass transfer coefficient of N₂O (K_{LaN_2O}). The driving force equals the differences between the concentrations of N₂O in the liquid phase (C_{N_2O}) and at the liquid–gas interface in equilibrium with the partial pressure of N₂O in the atmosphere (C_S). According to Henry's law, the solubility of a gas in water is proportional to the abundance of the gas in the atmosphere. As already noted, the atmospheric abundance of N₂O is very low (320 ppb), C_S can be taken as 0.0 mg/L, and the driving force equal to C_{N_2O} . The volumetric mass transfer coefficient of N₂O is a function of temperature, intensity of mixing and constituents in water (Tchobanoglous & Schroeder 1985).

In biological wastewater treatment, aeration and stirring can change intensities of mixing, resulting in differences in N₂O emissions between wastewater treatment processes to some degree. However, the impacts of aeration rates and stirring velocities on N₂O emissions are not quite clear. In order to distinguish the N₂O loss caused by the physical emission process from the total N₂O loss in biological wastewater treatment processes and to reveal the dynamics of N₂O emission, the impacts of external disturbances including aeration, stirring and wind on N₂O emission were investigated in this work.

MATERIAL AND METHODS

Batch experiments

Three batch experiments were conducted to investigate the impacts of wind (Test A), stirring (Test B) and aeration (Test C) on N₂O emission. All experiments were conducted in a glass beaker with a working volume of 1 L (Figure 1).

During all experiments, the ambient temperature was controlled at 20 °C by an air conditioner. The water level was kept at 5 cm below the top of the beaker and was maintained during the experiments.

Preparation of samples

Before starting the tests, an N₂O-saturated solution was prepared by aerating distilled water with high-purity N₂O gas (>99%, Huate Gas Co., Ltd, Foshan, China) for 20 minutes. Then, the N₂O-saturated solution was diluted to different initial concentrations with distilled water.

Test A: impact of wind

In order to study the impact of wind on N₂O emission, a fan was used to simulate the wind in Xi'an, Shaanxi Province, China. Wind velocities at the position of the beaker differed when the fan was placed at different distances from the beaker. The wind velocity was measured by a wind velocity meter (New Huayi Instrument Co., Ltd, Shenzhen, China). The wind velocity setting was based on the mean wind velocity in Xi'an, which was in the range of 1.3–2.6 m/s. In order to obtain a comprehensive understanding of the effects of wind velocity on N₂O emission, the range of wind velocity was enlarged, and 0.0, 0.5, 1.5, and 3.0 m/s were selected during test A. The results for wind velocity of 0.0 m/s were considered to represent the stable condition and were taken as the reference for all tests.

Test B: impact of stirring

Stirring velocity affected the floc size of activated sludge in the biological wastewater treatment process (Biggs & Lant 2000). In order to study the effects of stirring on N₂O emission in the anoxic unit of the biological wastewater treatment process, stirring velocities of 150, 200, 250, 300

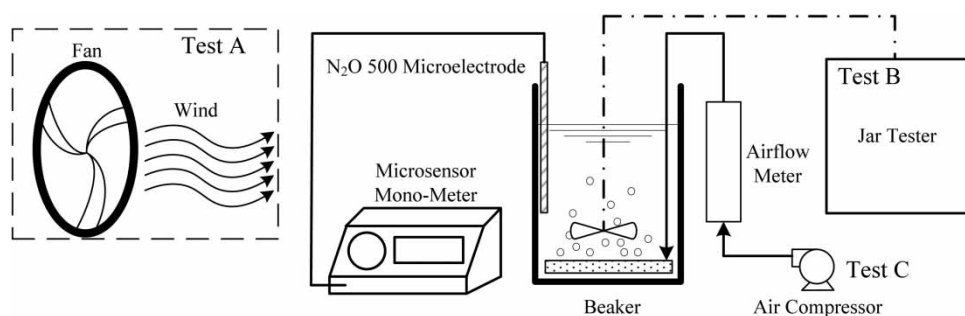


Figure 1 | Schematic diagrams of experiments. Test A: impact of wind; Test B: impact of stirring; Test C: impact of aeration.

and 350 r/min were selected during test B. These were controlled by a ZR4-6 jar tester (ZhongRun Water Industry Technology Development Co., Ltd, Shenzhen, China).

Test C: impact of aeration

As mentioned above, low DO concentration caused an increase in N₂O emission. Aeration rates in the oxic unit of the biological wastewater treatment process directly affected the DO concentration in the oxic unit. The effects of low and median aeration rates on N₂O emissions were examined in test C. Before starting test C, an aeration system was assembled. A diffuser was placed at the bottom of the beaker, and air was pumped to the beaker by an air compressor (Sobo Electrical Appliances Co., Ltd, Zhongshan, China). Aeration rates of 60, 70, 80, 90, 100, 110 and 120 L/h were selected during test C. The flow rate of air was monitored by an airflow meter (Yinhuan Flowmeter Co., Ltd, Yuyao, China).

N₂O measurement

N₂O concentration profiles in water were measured continuously using Clark-type micro electrodes. The N₂O 500 microelectrode and the microsensors monometer were purchased from Unisense (Arhus, Denmark). The microelectrode was calibrated with the two-point method according to the instructions provided by Unisense. In test A, as the N₂O concentration in water decreased very slowly, the time interval between measurements was not constant. In tests B and C, the time interval between measurements was 1 second.

Diffusion of N₂O from water to air

The two-film theory (Lewis & Whitman 1924), the penetration model (Higbie 1935) and the surface-renewal model (Danckwerts 1951) were used in the past to explain the mechanism of gas transfer across the liquid–gas interface. The most simple and commonly used was the two-film theory. In more than 95% of situations encountered, the results obtained by the two-film theory were essentially the same as those obtained with more complex theories (Tchobanoglous *et al.* 2003). In this work, the mechanism of N₂O transfer across the liquid–gas interface was elucidated by the two-film theory. When N₂O diffuses from water to air, the N₂O concentration in water can be calculated by Equation (1)

$$\frac{dC_{N_2O}}{dt} = -K_{LaN_2O} \cdot (C_{N_2O} - C_S) \quad (1)$$

where K_{LaN_2O} is the volumetric mass transfer coefficient of N₂O from water to air, min⁻¹; C_{N_2O} is the N₂O concentration in water, mg·L⁻¹; C_S is the concentration of N₂O at the liquid–gas interface in equilibrium with the partial pressure of N₂O in the atmosphere, mg·L⁻¹; t is time, min. As noted before, C_S can be taken as 0.0 mg/L. Equation (1) was rewritten as

$$\frac{dC_{N_2O}}{dt} = -K_{LaN_2O} \cdot C_{N_2O} \quad (2)$$

Solving Equation (2), C_{N_2O} equaled

$$C_{N_2O} = e^{-K_{LaN_2O} \cdot t + k_0} \quad (3)$$

where k_0 is an item that came from the indefinite integration of Equation (2). When t equaled 0, C_{N_2O} equaled the initial N₂O concentration in water, C_0 . As a result, k_0 can be solved and equaled to $\ln(C_0)$. Equation (3) was rewritten as

$$C_{N_2O} = C_0 \cdot e^{-K_{LaN_2O} \cdot t} \quad (4)$$

The volumetric mass transfer coefficient of N₂O, K_{LaN_2O} , was the pivotal parameter controlling N₂O emission from water to air. ‘-’ in the equations means that the N₂O concentration attenuated with the increase of time. Based on continuous monitoring, the N₂O concentration variations in water, C_0 and K_{LaN_2O} in Equation (4) were determined by using exponential regression to fit the monitoring data.

RESULTS AND DISCUSSION

Impact of wind velocities on N₂O emission

When wind velocity was 0.0 m/s, the N₂O concentration in water decreased very slowly. It took almost 7 hours for the N₂O concentration to decrease from 6.21 to 3.13 mg/L (Figure 2). By using exponential regression to fit the data obtained, C_0 and K_{LaN_2O} in Equation (4) at different wind velocities were determined. When the wind velocity increased from 0.0 to 3.0 m/s, K_{LaN_2O} increased from 0.0017 to 0.0074 min⁻¹. There was a linear relationship between the wind velocity and K_{LaN_2O} (Figure 3). When wind velocities were 0.0, 0.5, 1.5 and 3.0 m/s, the coefficients of determination (R^2) were 0.9695, 0.9941, 0.9926 and 0.9914, respectively, and the standard deviations (SD) were 0.1733, 0.0388, 0.0469 and 0.0490, respectively. Errors between the regression and the observation were small. K_{LaN_2O} obtained

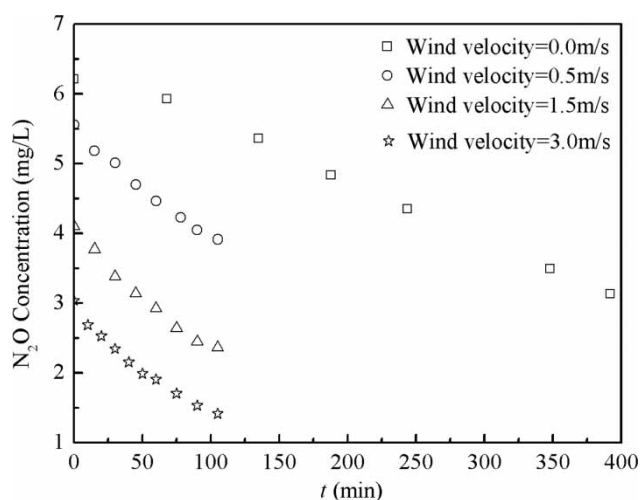


Figure 2 | N₂O concentration variations in water at different wind velocities.

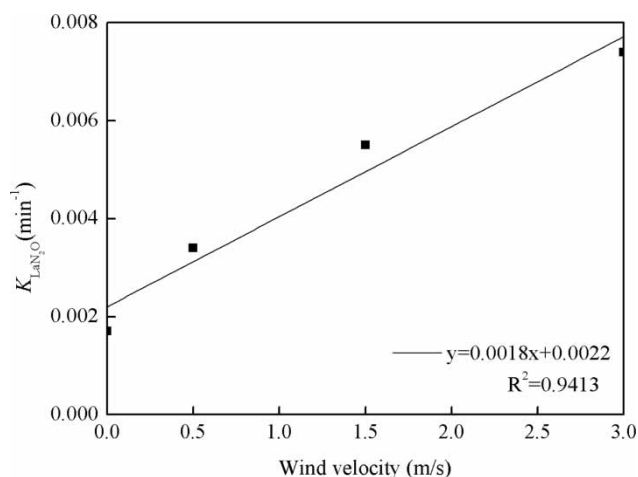


Figure 3 | Relationship between the wind velocity and the volumetric mass transfer coefficient of N₂O.

under the stable condition in this study (wind velocity 0.0 m/s) was smaller than that of the 0.0036 min^{-1} obtained by Zhang *et al.* (2012) in the non-aeration condition. Although there were some differences between the two values, K_{LaN_2O} was quite small under these two conditions, with both incidences being less than 0.01 min^{-1} .

Diffusion of N₂O from water to air was governed by the overall resistance to mass transfer, which equaled the sum of the gas and liquid phase resistance. The resistances were related to the thickness of the gas and liquid films, turbulence of the gas and liquid phases, the surface-renewal rate, and so on. Based on the two-film theory, as wind velocity increased, the gas phase was mainly influenced, turbulence of the gas phase was strengthened, and the thickness of the gas film decreased. However, the resistance in

the liquid phase still existed and did not decrease (Lewis & Whitman 1924). As a result, the gas phase resistance decreased and K_{LaN_2O} increased. When the wind velocity increased from 0.0 to 3.0 m/s, K_{LaN_2O} at different wind velocities remained in the same order of magnitude, and lower than 0.01 min^{-1} . Impact of the wind on N₂O emission was limited.

Impact of stirring velocities on N₂O emission

Similar to the impact of the wind on N₂O emission, stirring also promoted N₂O emission from water to air. The N₂O concentration variations in water coincided with Equation (4) (Figure 4). Taking K_{LaN_2O} at the stable condition as the reference, when the stirring velocity was increased to 350 r/min, K_{LaN_2O} increased from 0.0017 to 0.3263 min^{-1} . Differing from the linear relationship between the wind velocity and K_{LaN_2O} , there was an exponential relationship between the stirring velocity and K_{LaN_2O} (Figure 5). When the stirring velocity was below 200 r/min, K_{LaN_2O} increased slowly with the increase of the stirring velocity. However, when stirring velocity was higher than 200 r/min, K_{LaN_2O} increased rapidly with the increase of the stirring velocity (Figure 5). When stirring velocity was 150, 200, 250, 300 and 350 r/min, R^2 was 0.9992, 0.9986, 1.0000, 0.9999 and 0.9999 respectively, and SD was 0.0157, 0.0210, 0.0042, 0.0043 and 0.0059, respectively. Differences between the regression and the observation were also very small.

The wind mainly influenced N₂O emission in the gaseous phase. However, stirring affected the flow regime of the liquid phase. Consequently, N₂O emission in the liquid

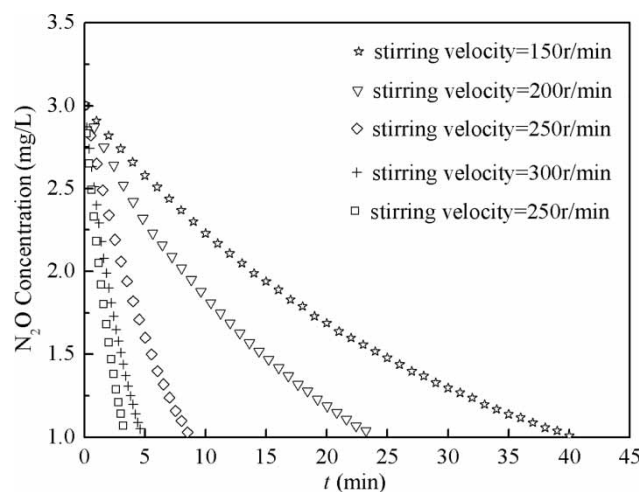


Figure 4 | N₂O concentration variations in water at different stirring velocities (as there were too many data points, all data points were plotted; however, some data points are not displayed in Figures 4 and 6).

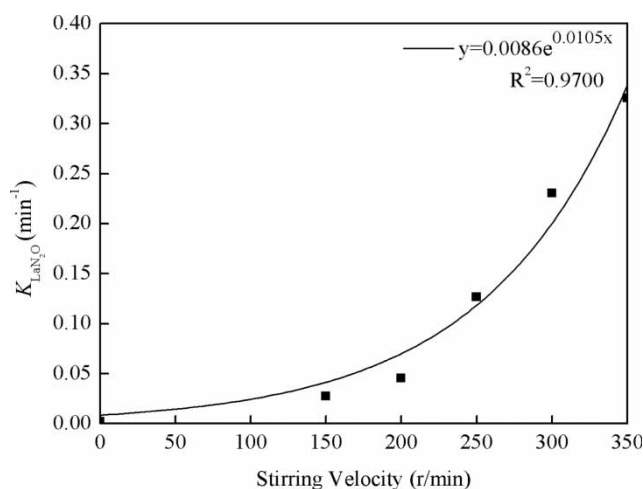


Figure 5 | Relationship between the stirring velocity and the volumetric mass transfer coefficient of N₂O.

phase was influenced. Different stirring velocities led to different mixing intensities and shears of the system. The mixing intensity and the shear of the system can be reflected by the value of the average velocity gradient (G , min^{-1}) (Parker *et al.* 1972; Oles 1992; Biggs & Lant 2000)

$$G = \left(\frac{\varepsilon}{\nu}\right)^{1/2} \quad (5)$$

where ν is the kinematic viscosity, $10^{-6} \text{ m}^2/\text{min}$; ε is the average turbulent energy dissipation rate, m^2/min^3 . ε equals

$$\varepsilon = \left(\frac{P_0 \cdot N^3 \cdot D^5}{V}\right) \quad (6)$$

where P_0 is the impeller power number; N is the impeller speed, r/min ; D is the impeller diameter, m ; V is the tank volume, m^3 . The larger the average velocity gradient was, the larger the mixing intensity and the shear of the system were. As ε was proportional to the third power of N , ε and G increased rapidly with the increase of stirring velocity. Consequently, the turbulence of water was enhanced, and the thickness of the liquid film decreased. Compared with the impact of wind on N₂O emission, the impact of stirring was greater. The values of $K_{\text{LaN}_2\text{O}}$ at different stirring velocities were 1–2 orders of magnitude larger than those at different wind velocities.

Impact of aeration rates on N₂O emission

When the aeration rate was increased from 60 to 120 L/h with a step size of 10 L/h, both R^2 were higher than 0.99,

and both SD were lower than 0.08. N₂O concentration in water decreased quickly when aeration started (Figure 6). Compared with $K_{\text{LaN}_2\text{O}}$ at the stable condition, when the aeration rate was increased to 120 L/h, $K_{\text{LaN}_2\text{O}}$ increased from 0.0017 to 0.5645 min^{-1} . Similar to the relationship between wind velocity and $K_{\text{LaN}_2\text{O}}$, there was a linear relationship between the aeration rate and $K_{\text{LaN}_2\text{O}}$ (Figure 7); $K_{\text{LaN}_2\text{O}}$ increased rapidly with the increase of the aeration rate. Quan studied the volumetric mass transfer coefficient of N₂O from water to air in clear water tests when the aeration rates were 12, 36 and 60 L/h (Quan *et al.* 2012). Compared with the results of 0.150 min^{-1} obtained by Quan, $K_{\text{LaN}_2\text{O}}$ was 0.3107 min^{-1} in this study when the aeration rate was 60 L/h. There is a large difference in $K_{\text{LaN}_2\text{O}}$ between the two studies, which is probably caused by different experimental conditions in the two studies. When determining $K_{\text{LaN}_2\text{O}}$ under aeration conditions, the velocity of air flow generated by the air compressor and the aperture of the diffuser of the aeration system could affect the results of $K_{\text{LaN}_2\text{O}}$. Different experimental conditions led to different $K_{\text{LaN}_2\text{O}}$ results with the same air flow rate of 60 L/h in different studies.

Differing from the impacts of the wind and stirring, aeration not only affected the flow regime of the liquid phase, but also enhanced the turbulence of the gaseous phase. When the aeration rate was increased, the thicknesses of the gas and liquid films were decreased simultaneously, turbulence of the gas and liquid phases were strengthened and the surface-renewal rate was also improved. The impact of aeration on N₂O emission was larger than the impacts of wind and stirring.

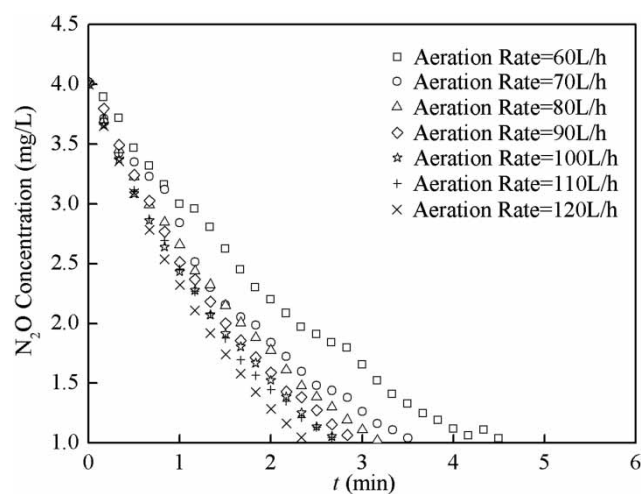


Figure 6 | N₂O concentration variations in water at different aeration rates.

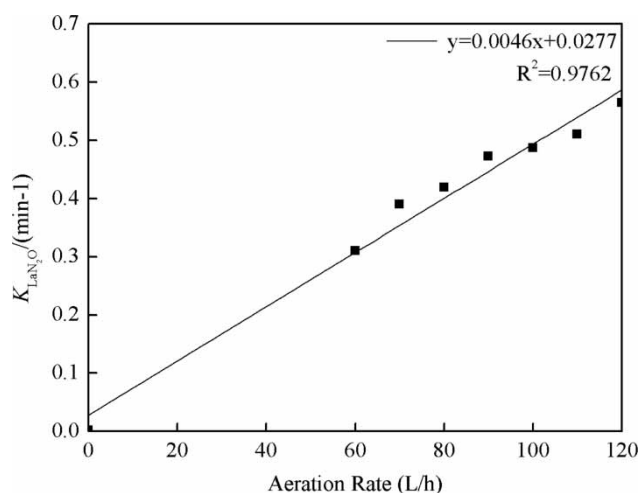


Figure 7 | Relationship between the aeration rate and the volumetric mass transfer coefficients of N₂O.

Comparison of the impacts of wind, stirring and aeration on N₂O emission

Compared with the stable condition, wind, stirring and aeration caused an increase of K_{LaN_2O} , and enhanced N₂O emission from water to air. Tables 1 and 2 exhibit the times for N₂O concentrations, dropping from 3 to 1 mg/L at different stirring velocities and aeration rates.

In the three factors, the impact of the wind on N₂O emission was the lowest (Figure 2) and the impact of the aeration on N₂O emission was the highest (Table 2). The impact of stirring on N₂O emission was more complicated. A threshold stirring velocity (200 r/min) seemed to exist. When stirring velocity was below the threshold stirring velocity, K_{LaN_2O} increased slowly with the increase of the stirring velocity; however, K_{LaN_2O} increased rapidly when the stirring velocity was higher than the threshold stirring velocity. The sequencing of the impacts of the three factors on K_{LaN_2O} was aeration > stir > wind. The impact of aeration on N₂O

Table 1 | Times for N₂O concentrations dropped from 3.0 to 1.0 mg/L at different stirring velocities

Stirring velocity (r/min)	150	200	250	300	350
Time (s)	2414	1436	521	287	203

Table 2 | Times for N₂O concentrations dropped from 3.0 to 1.0 mg/L at different aeration rates

Aeration rate (L/h)	60	70	80	90	100	110	120
Time (s)	217	158	154	137	132	130	111

emission was greater than the impacts of wind as turbulence and the mixing intensity of the liquid phase under the aeration condition was more vigorous than those for wind and stirring. Compared with the impact of stirring, values of K_{LaN_2O} at different aeration rates were higher than those at different stirring velocities, values of K_{LaN_2O} under two circumstances were in the same order of magnitude. The wind mainly influenced the N₂O emission in the gaseous phase, stirring affected the flow regime of the liquid phase, and aeration influenced both the gas and liquid phases. The turbulence and mixing intensity of the liquid phase were the restrictive factors for N₂O emission, and resistance against the mass transfer of N₂O was concentrated into the liquid phase. When turbulence and the mixing intensity of the liquid phase were enhanced, K_{LaN_2O} increased greatly.

CONCLUSIONS

- (1) Wind, stirring and aeration could promote N₂O emission from water to air. The volumetric mass transfer coefficient of N₂O from water to air, K_{LaN_2O} , increased with the increase of wind velocity, stirring velocity and aeration rate.
- (2) At stable conditions, K_{LaN_2O} was 0.0017 min⁻¹. K_{LaN_2O} linearly increased with the increase of wind velocity and aeration rate, and increased exponentially with the increase of stirring velocity.
- (3) The sequencing of the impacts of the three factors on K_{LaN_2O} was aeration > stirring > wind. The impact of aeration on N₂O emission was greater than the impacts of wind and stirring. Resistance against the mass transfer of N₂O was concentrated into the liquid phase. When predicting N₂O emissions from water to air, external disturbances, either environmental factors or operational factors, should be taken into consideration.

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