

# Experimental study of the impacts of external disturbances on N<sub>2</sub>O emission from water to air

Bo Hu, Shan He, Jianqiang Zhao and Ying Chen

## ABSTRACT

To obtain a comprehensive understanding of nitrous oxide (N<sub>2</sub>O) emission from water to air, the impacts of external disturbances including wind, stirring and aeration on N<sub>2</sub>O emissions were investigated by continuously monitoring N<sub>2</sub>O concentration variations in water. The volumetric mass transfer coefficient of N<sub>2</sub>O 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<sup>-1</sup> at the stable condition, and with the increase of wind velocity, stirring velocity and aeration rate, the diffusion of N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O emission was greater than those of wind and stirring. When predicting N<sub>2</sub>O 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

**Bo Hu**

**Shan He**

**Jianqiang Zhao** (corresponding author)

**Ying Chen**

School of Environmental Science and Engineering,  
Chang'an University,  
Xi'an,  
China  
E-mail: jqzhao@chd.edu.cn

**Bo Hu**

Key Laboratory of Environmental Protection &  
Pollution and Remediation of Water and Soil of  
Shaanxi Province,  
Xi'an,  
China  
and  
Chang'an University,  
Middle Section of the South 2nd Ring Road,  
710064,  
Xi'an,  
Shaanxi Province,  
China

## INTRODUCTION

As a potent greenhouse gas (GHG), nitrous oxide (N<sub>2</sub>O) 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-CO<sub>2</sub> GHG emissions, methane (CH<sub>4</sub>) was first, and N<sub>2</sub>O second highest (USEPA 2012). The atmospheric abundance of N<sub>2</sub>O 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 N<sub>2</sub>O emissions (UNEP 2013).

N<sub>2</sub>O 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, N<sub>2</sub>O 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 (NH<sub>2</sub>OH) (Yu *et al.* 2010); (3) reduction of

nitrite (NO<sub>2</sub><sup>-</sup>) by heterotrophic bacteria under anoxic conditions (Wunderlin *et al.* 2013). N<sub>2</sub>O 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 N<sub>2</sub>O emissions (Kampschreur *et al.* 2009).

As N<sub>2</sub>O is produced in nitrification and denitrification, there are two pathways to reduce N<sub>2</sub>O content in water. One is a bio-consumption process, and the other is a physical emission process. In the bio-consumption process, N<sub>2</sub>O is reduced to dinitrogen (N<sub>2</sub>) by microorganisms. In the physical emission process, N<sub>2</sub>O transfers from a liquid phase to a gas phase via the liquid-gas interface. While differentiating the N<sub>2</sub>O loss caused by diffusion and bio-consumption, it is important to evaluate N<sub>2</sub>O emission from the biological wastewater treatment process and to explore effective methods for controlling N<sub>2</sub>O emissions. The mechanism of the physical process can be

elucidated by the two-film theory (Lewis & Whitman 1924). The transferred mass of N<sub>2</sub>O through the interface depends on two factors: the driving force causing N<sub>2</sub>O transfer between the gas and liquid phases and the volumetric mass transfer coefficient of N<sub>2</sub>O ( $K_{LaN_2O}$ ). The driving force equals the differences between the concentrations of N<sub>2</sub>O in the liquid phase ( $C_{N_2O}$ ) and at the liquid–gas interface in equilibrium with the partial pressure of N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O emissions between wastewater treatment processes to some degree. However, the impacts of aeration rates and stirring velocities on N<sub>2</sub>O emissions are not quite clear. In order to distinguish the N<sub>2</sub>O loss caused by the physical emission process from the total N<sub>2</sub>O loss in biological wastewater treatment processes and to reveal the dynamics of N<sub>2</sub>O emission, the impacts of external disturbances including aeration, stirring and wind on N<sub>2</sub>O 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 N<sub>2</sub>O emission. All experiments were conducted in a glass beaker with a working volume of 1 L (Figure 1).

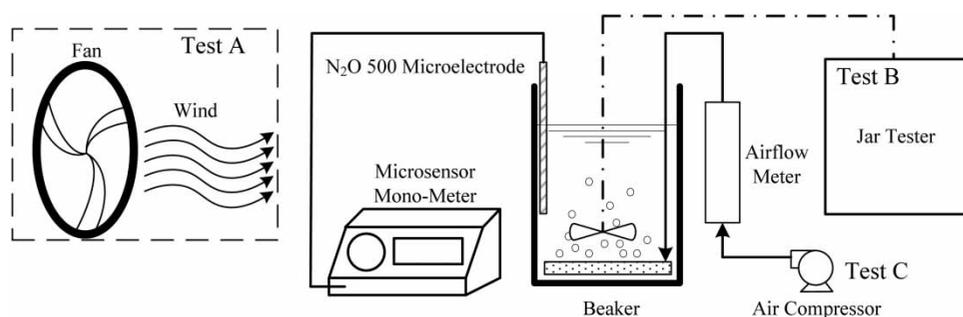


Figure 1 | Schematic diagrams of experiments. Test A: impact of wind; Test B: impact of stirring; Test C: impact of aeration.

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 N<sub>2</sub>O-saturated solution was prepared by aerating distilled water with high-purity N<sub>2</sub>O gas (>99%, Huate Gas Co., Ltd, Foshan, China) for 20 minutes. Then, the N<sub>2</sub>O-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 N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O emission in the anoxic unit of the biological wastewater treatment process, stirring velocities of 150, 200, 250, 300

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 N<sub>2</sub>O 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 N<sub>2</sub>O 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).

### N<sub>2</sub>O measurement

N<sub>2</sub>O concentration profiles in water were measured continuously using Clark-type micro electrodes. The N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O transfer across the liquid–gas interface was elucidated by the two-film theory. When N<sub>2</sub>O diffuses from water to air, the N<sub>2</sub>O 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 N<sub>2</sub>O from water to air, min<sup>-1</sup>;  $C_{N_2O}$  is the N<sub>2</sub>O concentration in water, mg·L<sup>-1</sup>;  $C_S$  is the concentration of N<sub>2</sub>O at the liquid–gas interface in equilibrium with the partial pressure of N<sub>2</sub>O in the atmosphere, mg·L<sup>-1</sup>;  $t$  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 N<sub>2</sub>O 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 N<sub>2</sub>O,  $K_{LaN_2O}$ , was the pivotal parameter controlling N<sub>2</sub>O emission from water to air. ‘-’ in the equations means that the N<sub>2</sub>O concentration attenuated with the increase of time. Based on continuous monitoring, the N<sub>2</sub>O 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 N<sub>2</sub>O emission

When wind velocity was 0.0 m/s, the N<sub>2</sub>O concentration in water decreased very slowly. It took almost 7 hours for the N<sub>2</sub>O 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<sup>-1</sup>. 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

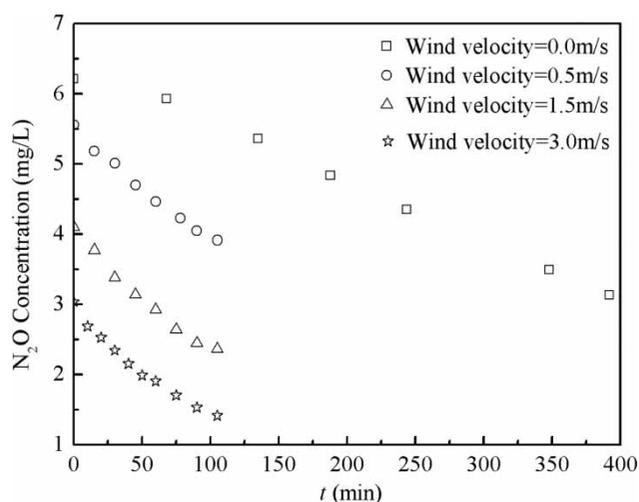


Figure 2 | N<sub>2</sub>O concentration variations in water at different wind velocities.

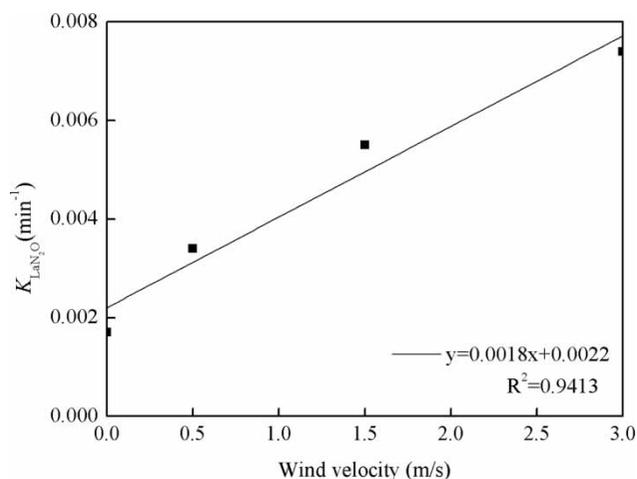


Figure 3 | Relationship between the wind velocity and the volumetric mass transfer coefficient of N<sub>2</sub>O.

under the stable condition in this study (wind velocity 0.0 m/s) was smaller than that of the  $0.0036 \text{ min}^{-1}$  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  $0.01 \text{ min}^{-1}$ .

Diffusion of N<sub>2</sub>O 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  $0.01 \text{ min}^{-1}$ . Impact of the wind on N<sub>2</sub>O emission was limited.

### Impact of stirring velocities on N<sub>2</sub>O emission

Similar to the impact of the wind on N<sub>2</sub>O emission, stirring also promoted N<sub>2</sub>O emission from water to air. The N<sub>2</sub>O 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  $0.3263 \text{ min}^{-1}$ . 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 N<sub>2</sub>O emission in the gaseous phase. However, stirring affected the flow regime of the liquid phase. Consequently, N<sub>2</sub>O emission in the liquid

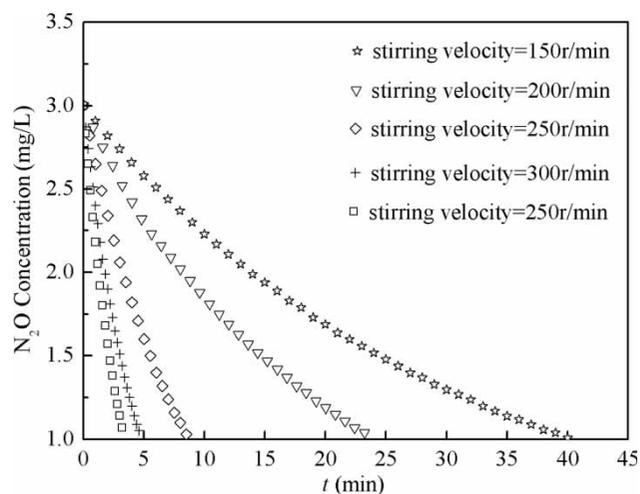
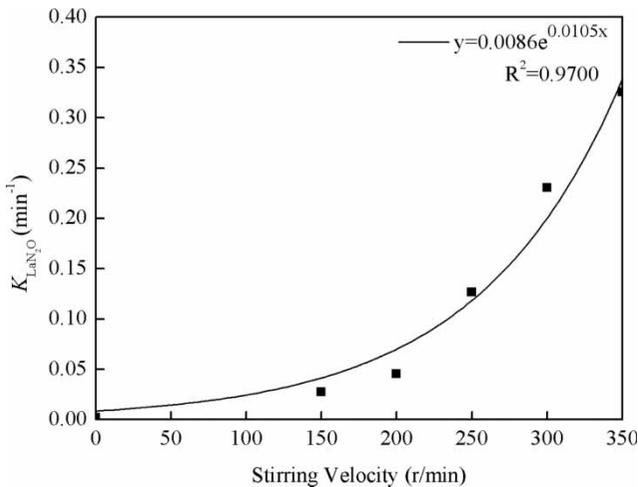


Figure 4 | N<sub>2</sub>O concentration variations in water at different stirring velocities (as there were too many data points, all data points were plotted; however, some data points are not displayed in Figures 4 and 6).



**Figure 5** | Relationship between the stirring velocity and the volumetric mass transfer coefficient of N<sub>2</sub>O.

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 ( $G$ ,  $\text{min}^{-1}$ ) (Parker *et al.* 1972; Oles 1992; Biggs & Lant 2000)

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

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

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

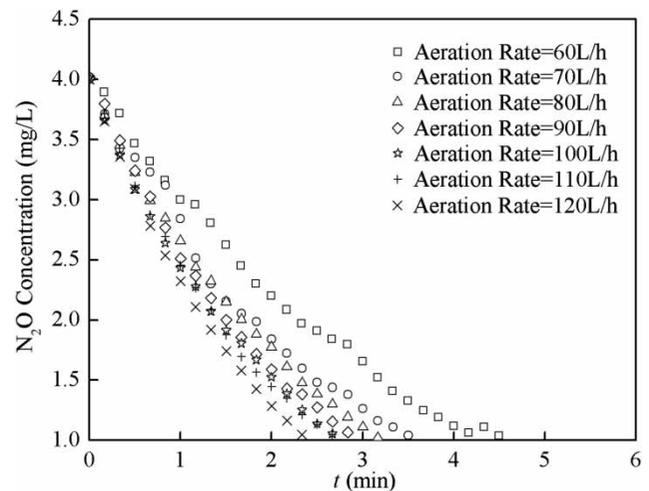
where  $P_0$  is the impeller power number;  $N$  is the impeller speed,  $\text{r}/\text{min}$ ;  $D$  is the impeller diameter,  $\text{m}$ ;  $V$  is the tank volume,  $\text{m}^3$ . The larger the average velocity gradient was, the larger the mixing intensity and the shear of the system were. As  $\varepsilon$  was proportional to the third power of  $N$ ,  $\varepsilon$  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 N<sub>2</sub>O 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 N<sub>2</sub>O emission

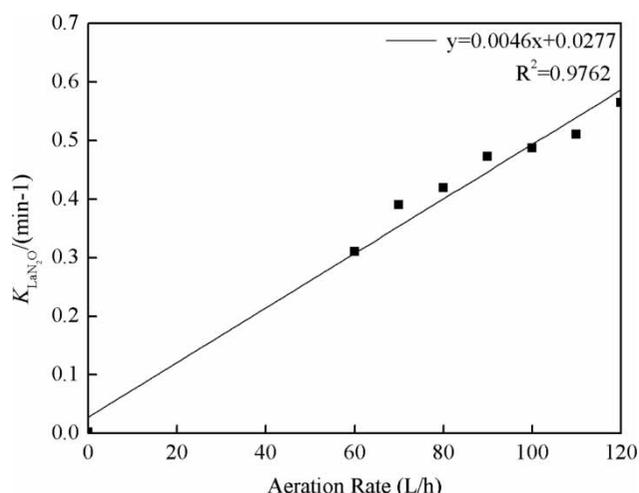
When the aeration rate was increased from 60 to 120 L/h with a step size of 10 L/h, both  $R^2$  were higher than 0.99,

and both  $SD$  were lower than 0.08. N<sub>2</sub>O 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  $\text{min}^{-1}$ . 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 N<sub>2</sub>O 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  $\text{min}^{-1}$  obtained by Quan,  $K_{\text{LaN}_2\text{O}}$  was 0.3107  $\text{min}^{-1}$  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 N<sub>2</sub>O emission was larger than the impacts of wind and stirring.



**Figure 6** | N<sub>2</sub>O concentration variations in water at different aeration rates.



**Figure 7** | Relationship between the aeration rate and the volumetric mass transfer coefficients of N<sub>2</sub>O.

### Comparison of the impacts of wind, stirring and aeration on N<sub>2</sub>O emission

Compared with the stable condition, wind, stirring and aeration caused an increase of  $K_{LaN_2O}$ , and enhanced N<sub>2</sub>O emission from water to air. Tables 1 and 2 exhibit the times for N<sub>2</sub>O 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 N<sub>2</sub>O emission was the lowest (Figure 2) and the impact of the aeration on N<sub>2</sub>O emission was the highest (Table 2). The impact of stirring on N<sub>2</sub>O 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 N<sub>2</sub>O

**Table 1** | Times for N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O 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 N<sub>2</sub>O emission, and resistance against the mass transfer of N<sub>2</sub>O 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 N<sub>2</sub>O emission from water to air. The volumetric mass transfer coefficient of N<sub>2</sub>O 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<sup>-1</sup>.  $K_{LaN_2O}$  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 N<sub>2</sub>O emission was greater than the impacts of wind and stirring. Resistance against the mass transfer of N<sub>2</sub>O was concentrated into the liquid phase. When predicting N<sub>2</sub>O emissions from water to air, external disturbances, either environmental factors or operational factors, should be taken into consideration.

### ACKNOWLEDGEMENT

This work was supported by the Fundamental Research Funds for the Central Universities (Grant No. 2013G3292017; Grant No. 2013G1291066).

### REFERENCES

- Ahn, J. H., Kim, S., Park, H., Rahm, B., Pagilla, K. & Chandran, K. 2010 N<sub>2</sub>O emissions from activated sludge processes,

- 2008–2009: results of a national monitoring survey in the United States. *Environ. Sci. Technol.* **44** (12), 4505–4511.
- Biggs, C. A. & Lant, P. A. 2000 Activated sludge flocculation: on-line determination of floc size and the effect of shear. *Water Res.* **34** (9), 2452–2550.
- Danckwerts, P. V. 1951 Significance of liquid film coefficient in gas absorption. *J. Ind. Eng. Chem.* **43** (6), 1460–1467.
- Foley, F., de Haas, D., Yuan, Z. G. & Lant, P. 2010 Nitrous oxide generation in full-scale biological nutrient removal wastewater treatment plants. *Water Res.* **44** (3), 831–844.
- Higbie, R. 1935 The rate of absorption of pure gas into a still liquid during short periods of exposure. *Trans. Am. Inst. Ind. Chem.* **31**, 365.
- IPCC 2001 *Climate Change 2001: The Scientific Basis*. Cambridge University, Cambridge.
- Kampschreur, M. J., Temmink, H., Kleerebezem, R., Jetten, M. S. M. & van Loosdrecht, M. C. M. 2009 Nitrous oxide emission during wastewater treatment. *Water Res.* **43** (17), 4093–4103.
- Lewis, W. K. & Whitman, W. G. 1924 Principles of gas absorption. *Ind. Eng. Chem.* **16** (12), 1215–1220.
- Oles, V. 1992 Shear induced aggregation and breakup of polystyrene latex particles. *J. Colloid Interface Sci.* **154** (2), 351–358.
- Parker, D. S., Kaufman, W. & Jenkins, D. 1972 Floc breakup in turbulent flocculation processes. *Jo. Sanit. Eng. Div. Proc. Am. Soc. Civil Eng.* **98** (SAI), 79–99.
- Quan, X. C., Zhang, M. C., Lawlor, P. G., Yang, Z. F. & Zhan, X. M. 2012 Nitrous oxide emission and nutrient removal in aerobic granular sludge sequencing batch reactor. *Water Res.* **46** (16), 4981–4990.
- Ravishankara, A. R., Daniel, J. S. & Portmann, R. W. 2009 Nitrous oxide (N<sub>2</sub>O): the dominant ozone-depleting substance emitted in the 21st century. *Science* **326** (5949), 123–125.
- Tchobanoglous, G. & Schroeder, E. D. 1985 *Water Quality: Characteristics, Modeling, Modification*. Addison-Wesley Publishing House, MA, USA.
- Tchobanoglous, G., Burton, F. L. & Stensel, H. D. 2003 *Wastewater Engineering: Treatment and Reuse*. 4th edn, Metcalf & Eddy, McGraw-Hill, Inc., USA.
- UNEP 2013 *Drawing down N<sub>2</sub>O to protect climate and the ozone layer*. A UNEP Synthesis Report. United Nations Environment Programme (UNEP), Nairobi, Kenya.
- USEPA 2012 *Summary Report: Global anthropogenic non-CO<sub>2</sub> greenhouse gas emissions: 1990–2030*. US Environmental Protection Agency, Washington, DC, USA.
- Wrage, N., Velthof, G. L., van Beusichem, M. L. & Oenema, O. 2001 Role of nitrifier denitrification in the production of nitrous oxide. *Soil Biol. Biochem.* **33** (12–13), 1723–1732.
- Wunderlin, P., Lehmann, M. F., Siegrist, H., Tuzson, B., Joss, A., Emmenegger, L. & Mohn, J. 2013 Isotope signatures of N<sub>2</sub>O in a mixed microbial population system: constraints on N<sub>2</sub>O producing pathways in wastewater treatment. *Environ. Sci. Technol.* **47** (3), 1339–1348.
- Yu, R., Kampschreur, M. J., van Loosdrecht, M. C. M. & Chandran, K. 2010 Mechanisms and specific directionality of autotrophic nitrous oxide and nitric oxide generation during transient anoxia. *Environ. Sci. Technol.* **44** (4), 1313–1319.
- Zhang, M. C., Lawlor, P. G., Li, J. P. & Zhan, X. M. 2012 Characteristics of nitrous oxide (N<sub>2</sub>O) emissions from intermittently-aerated sequencing batch reactors treating the separated liquid fraction of anaerobically digested pig manure. *Water Air Soil Pollut.* **223** (5), 1973–1981.

First received 7 May 2014; accepted in revised form 4 August 2014. Available online 28 August 2014