Coastal reservoirs as a source of nitrous oxide: Spatio-temporal patterns and assessment strategy

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ABSTRACT

Coastal reservoirs are widely regarded as a viable solution to the water scarcity problem faced by coastal cities with growing populations. As a result of the accumulation of anthropogenic wastes and the alteration of hydroecological processes, these reservoirs may also become the emission hotspots of nitrous oxide (N2O). Hitherto, accurate global assessment of N2O emission suffers from the scarcity and low spatio-temporal resolution of field data, especially from small coastal reservoirs with high spatial heterogeneity and multiple water sources. In this study, we measured the surface water N2O concentrations and emissions at a high spatial resolution across three seasons in a subtropical coastal reservoir in southeastern China, which was hydrochemically highly heterogeneous because of the combined influence of river runoff, aquacultural discharge, industrial discharge and municipal sewage. Both N2O concentration and emission exhibited strong spatio-temporal variations, which were correlated with nitrogen loading from the river and wastewater discharge. The mean N2O concentration and emission were found to be significantly higher in the summer than in spring and autumn. The results of redundancy analysis showed that NH4+-N explained the greatest variance in N2O emission, which implied that nitrification was the main microbial pathway for N2O production in spite of the potentially increasing importance of denitrification of NO3−-N in the summer. The mean N2O emission across the whole reservoir was 107 μg m−2 h−1, which was more than an order of magnitude higher than that from global lakes and reservoirs. Based on our results of Monte Carlo simulations, a minimum of 15 sampling points per km2 would be needed

to produce representative and reliable N2O estimates in such a spatially heterogeneous aquatic system. Overall, coastal reservoirs could play an increasingly important role in future climate change via their N2O emission to the atmosphere as water demand and anthropogenic pressure continue to rise.

Keywords: Nitrous oxide (N_2O); Spatial heterogeneity; Spatially resolved measurement; Wastewater discharge; Subtropical reservoir; IPCC

1. Introduction

Nitrous oxide (N_2O) is a potent greenhouse gas with a global warming potential nearly 300 times that of carbon dioxide (CO_2) on a mass basis over a 100-year time horizon (IPCC, 2013). N_2O is also considered to be one of the major substances that can destroy ozone in the stratosphere (Ravishankara et al. 2009; Shaaban et al., 2018). The atmospheric averaged N_2O concentration reached 331.1±0.1 ppbv in 2018, which is approximately 23% higher than the pre-industrial (before 1750) levels (World Meteorological Organization, 2019). There has been a steady increase in atmospheric N_2O at a rate of 0.7–0.8 ppb yr^{-1} over the past three decades (Davidson 2009; Saikawa et al. 2014; Xiao, et al., 2019a). Quantifying the potential source strength of various ecosystems is fundamental for predicting future N_2O emission and climate change (Yang et al., 2020).

Aquatic ecosystems are considered as important sources of N_2O emission, contributing for approximately 25%–30% of global N_2O emissions (Zhou et al., 2019). As an important component of the Earth's surface water systems, man-made reservoirs, which include those for hydropower, flood management, water supply, and navigation purposes, are now considered as significant contributors of the atmospheric N_2O (Beaulieu et al., 2019; Cheng et al., 2019; Guérin et al., 2008; Wang et al., 2017; Yang & Flower 2012). Recent estimates indicated that the amount of N_2O emitted from global reservoirs was approximately 0.03−0.07 Tg N yr^{-1} (Deemer et al., 2016; Maavara et al., 2019). However, the estimate of N_2O emission from reservoirs, especially from the ones with anthropogenic disturbances, remains highly uncertain

due to the data limitation. Additionally, N_2O emissions from reservoirs in different climatic zones exhibited high spatial-temporal variations across and within the systems (Beaulieu et al., 2015; Cheng et al., 2019; Liang et al., 2019; Liu et al., 2011a, 2015, 2017; Musenze et al., 2014; Shi et al., 2020). This is another source of uncertainty in reservoir N_2O budget. Thus, more *in situ* measurements with high spatiotemporal resolution are needed to reduce the uncertainty and to develop more accurate approaches for upscaling to whole reservoir N_2O emissions and further large-scale assessments of reservoir N_2O fluxes.

The N_2O production in reservoir is known to mainly derive from the nitrification and denitrification of terrestrial nitrogen (N). Human activities in the drainage basin, particularly land use change, sewage discharge and agriculture fertilization, have severely altered the N transport of from the terrestrial ecosystem to the aquatic ecosystem (Davidson, 2009; Hosen et al., 2014; Williams et al., 2016), and influenced the N_2O production in reservoirs (Wang et al., 2017). Syvitski et al. (2005) estimated over 100 billion metric tons of sediment and up to 3 billion metric tons of organic matter were trapped in the reservoirs in the last five decades. Hence the allochthonous material load by human activities would play a much more important role for accelerating N_2O emission from reservoirs. To date, some efforts have been made on the response of N_2O emission from reservoirs to the human activities (e.g., agriculture, urbanization sewage discharges) in the basin, especially in China (e.g., Liu et al., 2011a; Wang et al., 2017; Xiao et al., 2019b). However, the data of N_2O emissions from reservoirs under the impact of human activities are disproportionately scarce in

97 comparison to the amount and area of reservoirs. More importantly, the majority of
98 existing studies only focused on inland freshwater reservoirs, but few studies have
99 mentioned N_2O emission from coastal reservoirs. As coastal reservoirs often
100 experience a higher salinity and inputs of anthropogenic carbon and nutrients than
101 inland ones, N_2O production and emission rates might differ between the reservoirs in
102 these two zones. However, there is currently a lack of empirical data to test this
103 hypothesis and advance our understanding of reservoirs in the global N_2O cycle.

104 In order to fill these knowledge gaps, this study researched high spatial
105 resolution measurements of dissolved N_2O concentrations in a subtropical coastal
106 reservoir in southeast China over three seasons of a year to estimate the N_2O fluxes
107 across the water-atmosphere interface. The specific objectives of this study are to (1)
108 quantify the magnitude of N_2O emission fluxes from the coastal reservoirs, (2) assess
109 the spatial variations in diffusive N_2O fluxes both within system and among systems,
110 (3) explore the drivers of the spatial variations of coastal reservoir N_2O emissions;
111 and (4) examine how many measurements sities in space were needed to
112 representatively cover the observed spatial variability in coastal reservoir N_2O
113 emissions. The results of the present study will provide the scientific basis for the
114 development of global biogeochemical models and national GHG inventories through
115 characterizing N_2O emission from the reservoirs in coastal areas.

116 ## 2. Materials and Methods

117 *2.1. Study Area*

118 The study was conducted in Wenwusha Reservoir (25°49′36″–25°54′00″ N,

119°35′12″–119°38′11″ E), a subtropical reservoir in the coastal area of Fujian Province, Southeast China (Figure 1). The reservoir was built for irrigation and flood-prevention. It has a surface area of 5.2 km^2, a total volume of 3.20×10^8 m^3, drainage area of 275 km^2, and mean depth of 9.0 m. The reservoir is influenced by a subtropical monsoon humid climate, with a mean annual temperature of 19.3 °C. The average annual precipitation is 1390 mm, most of which (approximately 75%) occurs in the wet season from May to September.

The Wenwusha Reservoir was created by two dams on the Nangyangdong River estuary. We compartmentalized the reservoir into two reservoir zones according to the two dam constructed time, topographic feature, and trophic status (Figure 1). The northern reservoir zone (NRZ) was constructed in 1957, with a surface area of 1.9 km^2 and a total volume of 1.40×10^8 m^3. The NRZ is located in highly urbanized districts and is heavily impacted by human activities, e.g. the sewage discharge from aquacultural, industrial, and municipal activities. The NRZ also received large amounts of nutrient input from Nanyangdong river. The southern reservoir zone (SRZ) was constructed in 2004, with a surface area of 3.3 km^2 and a total volume of 1.69×10^8 m^3. Around 70% of its catchment is used for agricultural activities (e.g., aquaculture and farming), but its immediate surroundings are partly forested and wetland (Figure 1). The water salinity in NRZ is much lower than that in SRZ (0.4–1.3‰ versus 0.4–3.7‰), as a result of freshwater dilution caused by the surface runoff.

2.2. Sampling strategies

Considering the reservoir size, topographic feature, and pollution source, a total of 21 transects (10 in the NRZ and 11 in the SRZ) were selected to collect samples to characterize the variation across the whole reservoir. According to the lengths of the transects, there were 3-10 sampling sites in the transects (Figure 1). The total number of sample sites was 103 (56 in the NRZ and 47 in the SRZ). These sites were distributed in 5 different sewage loading sectors, namely the industrial effluent loading sector (Sector-I, n = 4), town sewage loading sector (Sector-T, n = 6), river input sector (Section-R, n = 7), aquaculture sewage loading sector (Section-A, n = 22), and non-wastewater loading sector (Sector-N, n = 64). At each site, surface-water samples were collected at a depth of 20 cm. Three *in situ* whole reservoir surveys were conducted in mid–November 2018, mid–March and mid–June 2019. Each whole reservoir survey was completed in two consecutive days to reduce the biases caused by day-to-day variation. More details about the sampling transects are presented in supporting information Table S1.

2.3. Measurement of dissolved N_2O concentration and diffusive flux

2.3.1. Dissolved N_2O concentration

Surface water samples for dissolved N_2O concentration analysis were collected using a 100-mL syringe equipped with three-way stopcocks. After sample was collected from reservoir, water was quickly transferred into a 55-mL gas-tight glass serum bottle. Prior to sealing the bottle, 0.2 mL saturated $HgCl_2$ solution was injected into the sample bottle for inhibiting microbial activity. The bottle was immediately sealed with an open-topped screw cap equipped with a halobutyl rubber septum to

exclude any air bubble (Borges et al., 2018; Xiao et al., 2019a; Yang et al., 2020). All water samples were stored in an ice box, transported to the laboratory within 6 hr, and analyzed within two days of collection.

Dissolved N_2O concentrations in the collected water samples were measured following the headspace equilibration method described by Yu et al. (2013, 2017). In the laboratory, ultrahigh purity N_2 gas (99.999%) was injected into the sample bottle via a syringe to create a 25-mL headspace. The bottles were then shaken vigorously for 10 min in an oscillator to allow the dissolved N_2O diffuse out and gases reach equilibrium between the liquid phase and headspace. After waiting for 0.5 hr, approximately 5 mL air sample was drawn from the headspace using a syringe equipped with three-way stopcocks. N_2O concentrations in the headspace air were measured using gas chromatography (GC-2014, Shimadzu, Kyoto, Japan) equipped with an electron capture detector (ECD). The detection limit for the N_2O analysis was 0.02 ppm, and the relative standard deviations of N_2O analyses were ≦5.0% (Yang et al., 2020). Based on the equilibrium temperature, salinity–dependent Henry's law constant, and the measured headspace gas concentration, dissolved N_2O concentration was calculated using the formula provided by Weiss and Price (1980).

2.3.2. N_2O flux from the transfer coefficient method

The N_2O flux ($F_{W\text{-}A}$, μmol m^{-2} h^{-1}) across the water–atmosphere interface was calculated using the classic boundary-layer model (equation (1)), which has been widely used for N_2O emissions in the lentic ecosystem (e.g., lakes, reservoir and ponds) (e.g., Cole and Caraco, 1998; Liang et al., 2019; Musenze et al., 2014; Xiao et

al., 2019b; Yang et al., 2015a).

$$F_{\text{W-A}} = k \times (C_{obs} - C_{eq}) \quad \text{(Eq1)}$$

where C_{obs} is the measured dissolved N_2O concentration (nmol L^{-1}) in the surface water (20 cm depth); C_{eq} in water that is in equilibrium with the atmosphere at the *in situ* temperature (nmol L^{-1}); and k is the gas transfer velocity (m h^{-1}).

In the lentic ecosystem, the N_2O transfer velocity is mainly driven by wind speed because no surface water flow (Xiao et al., 2019b). k in the present study was calculated using a wind-dependent formula (equation (2)) derived from a small shallow lake as follows (Cole and Caraco, 1998):

$$k = (Sc/660)^{-n}(2.07 + 0.215 \times U_{10}^{1.7}) \quad \text{(Eq2)}$$

where n = 0.66 and 0.50 for wind speeds ≤3 and >3 m s^{-1}, respectively; Sc is the Schmidt number for N_2O and is dependent on the temperature (T, ºC); and U_{10} is the frictionless wind speed at 10 m high expressed in m s^{-1}.

The Sc and U_{10} were estimated using the following equations (Crusius and Wanninkhof, 2003; Wanninkhof, 1992):

$$Sc = 2055.6 - 137.11T + 4.3173T^2 - 0.054350T^3 \quad \text{(Eq3)}$$

$$U_{10} = U_z[1 + \frac{(C_{d10})^{1/2}}{K}\ln(\frac{10}{z})] \quad \text{(Eq4)}$$

where U_z is the wind speed (m s^{-1}) at height z (2.0 m in this study) above the water surface at which wind speed was measured; C_{d10} is the drag coefficient at 10 m above the water surface (0.0013 m s^{-1}); and K is the von Karman constant (0.41).

2.4. Measurement of environmental variables

During each campaign, from 103 sites in the reservoir was also sampled for

measuring chemical properties. Surface water samples were collected at a depth of 20 cm using a 5 L organic glass hydrophore, and transferred into 150 mL polyethylene bottles. Approximately 0.5 mL saturated $HgCl_2$ solution was added to each bottle to inhibit microbial activities (Taipale and Sonninen 2009; Zhang et al., 2013). All samples were preserved in an ice box, and transported to the laboratory for measurement. After filtering the samples through a 0.45 μm cellulose acetate filter (Biotrans™ nylon membranes), the concentration of total dissolved nitrogen (TDN), nitrate-nitrogen ($N–NO_3^-$), and ammonium-nitrogen ($N–NH_4^+$) was measured using a flow injection analyzer (Skalar Analytical SAN^{++}, The Netherlands). The detection limits for TDN, $NN–NO_3^-$, and $N–NH_4^+$ were 3.0, 0.6 and 0.6 μg L^{-1}, respectively. The relative standard deviations of TDN, $NN–NO_3^-$, and $N–NH_4^+$ analyses were ≦2.0%, ≦3.0% and ≦3.0%, respectively.

Water temperature (W_T), dissolved oxygen (DO), pH, electrical conductivity (EC), and salinity at a depth of 20 cm were also measured *in situ* at each sampling site. W_T and pH were measured using a portable pH/mV/Temperature meter (IQ150, IQ Scientific Instruments, USA). DO, EC and salinity were determined using a multiparameter water quality probe (550A YSI, USA), an electrical conductivity meter (2265FS EC, Spectrum Technologies, USA) and a salinity meter (Eutech Instruments-Salt6, USA), respectively. The relative standard deviations of DO, pH, EC, and salinity analyses were ≦2.0%, ≦1.0%, ≦1.0% and ≦1.0%, respectively. Meanwhile, the meteorological factors, including the air temperature (A_T), atmospheric pressure (A_P) and 2 m height wind speed (W_S), were recorded using a

portable weather meter (Kestrel-3500, USA).

2.5. Statistical analysis

The analysis of variance (ANOVA) to test for significant (p<0.05) effects of reservoir zone and sewage sector water depth on surface water physicochemical properties, dissolved N_2O concentration and N_2O diffusive fluxes using the SPSS 17.0 statistical software package (SPSS Inc., USA). Pearson correlation coefficients were used to examine the relationships between water physicochemical parameters and N_2O concentration/ fluxes. Principal component analysis (PCA) was also performed to analyze the relationships between the N_2O fluxes and surface water physicochemical properties to show their pattern at different sampling campaigns. For quantifying spatial heterogeneity effects by randomizing the high-resolution N_2O measurements, a Monte Carlo analysis was used to evaluate the effect of sample size on the N_2O estimation of the whole reservoir. Without replacements, the N_2O measurements are resampled from the 103 sites (n = 10, 20, 30…, 100). The resampling process then was repeated 10000 times and the overall mean and standard deviation of N_2O emissions were calculated for each sample size. The results were presented as means ± SE. Statistical plots were generated using OriginPro 7.5 (OriginLab Corp. USA) and ArcGIS 10.2 (ESRI Inc., Redlands, CA, USA). Conceptual diagrams were drawn using EDraw Max version 7.3 (EdrawSoft, Hong Kong, China).

3. Results

3.1. Water Quality Parameters in Reservoir

There were substantial spatial variations in water chemical parameters (e.g., salinity, DO, and nutrient concentrations) between both reservoir zones on all three sampling occasions (p<0.01; Table S2), with lower levels of DO (Figure 2a-c), EC (Figure 3g-i and Figure S1d) and salinity (Figure 2j-l and Figure S1c) and higher concentrations of N–NO_3^- (Figure 2d-f and Figure S2a-c), N–NH_4^+ (Figure 2g-i and and Figure S2d-f), and TDN (Figure 2j-l and Figure S2g-i) in NRZ than in SRZ. However, the W_T (Figure 3a-c) and pH (Figure 3d-f) were relatively constant across the reservoir (p>0.05; Figure S1a and b), with the coefficients of variation only ranged from 4% to 7%, and 1% to 7% over the three sampling campaigns, respectively.

The mean DO, salinity, N–NO_3^-, N–NH_4^+ and TDN also differed significantly among the five sewage loading sectors over the three sampling campaigns (Table S3). Mean N–NO_3^-, N–NH_4^+ and TDN concentrations were significantly lower in Sector-N than the other sectors (p<0.05;Table S3), and were generally highest in Sector-R. In contrast, mean DO concentration and salinity was generally greater in Sector-N than the other four sectors (Table S3).

The main water quality parameters also showed strong temporal variation across the whole reservoir (p<0.001, Two-way ANOVAs; Table S2). The water temperature was highest in summer (29.3 ºC), followed by spring (23.1 ºC) and autumn (18.3 ºC) (Figure 3a-c). The seasonal patterns of N−NO_3^- (Figure 2d-f), TDN (Figure 2j-l), pH (Figure 3d-f) and salinity (Figure 3j-l), concentrations were contrary to that of water temperature (Figure 3a-c). The DO varied seasonally, with the peak in spring (Figure 2a-c). In contrast, N−NH_4^+ concentrations were significantly lower in spring (Figure

273 2g-i).

274 *3.2. Spatial Variation in Surface Water N_2O Concentration*

275 Across all sampling sites in the reservoir, dissolved N_2O concentration in
276 November 2018, March 2019 and June 2019 ranged from 8.99 to 86.58, 6.11 to
277 110.37, and 7.33 to 261.27 nmol L^{-1} (Figure 4), respectively, with mean N_2O
278 concentration of 46.42±2.03, 58.96±5.90, and 43.96±2.69 nmol L^{-1} in the three
279 sampling campaigns. Across the three sampling campaigns, the mean N_2O
280 concentration were significantly greater in NRZ than in SRZ (p<0.05, Two-way
281 ANOVAs; Table 1 and Figure S3a-c). Meanwhile, dissolved N_2O concentration also
282 showed remarkably differences betwee the wastewater loading area and the
283 non-wastewater loading area (p<0.05; Table S4 and Figure S4a-c). Across the three
284 sampling campaigns, specifically, the mean N_2O concentration were significantly
285 lower in Sector-N than the other sectors (p<0.05; Figure 5a), and were generally
286 highest in Sector-R (Figure 5a).

287 *3.3. Spatial Variation in Diffusive N_2O Fluxes Across the Water-Air Interface*

288 Across all sampling sties, the N_2O fluxes ranged from 2.61 to 415.61, from
289 -10.83 to 254.01, and from 2.04 to 711.50 μg m^{-2} h^{-1} in November 2018, March 2019
290 and June 2019, respectively, with mean N_2O fluxes of 110.41±9.31, 78.48±6.27, and
291 153.12±8.61 μg m^{-2} h^{-1} in the three sampling campaigns. Overall, the Wenwusha
292 Reservoir acted as a constant source of N_2O to the atmosphere.

293 N_2O fluxes (Figure 6) showed a similar spatial variability to N_2O concentration
294 (Figure 4). Relatively higher N_2O fluxes were found in the NRZ of the Wenwusha

295 Reservoir. Across all sampling campaigns, the N_2O fluxes in the NRZ ranged from
296 6.87 to 711.50 μg m^{-2} h^{-1}, with an average of 153.12±8.61 μg m^{-2} h^{-1}, which was
297 almost 3 times higher than those from the SRZ (p<0.001, Two-way ANOVAs; Table 1
298 and Figure S3d-f).

299 Significant differences in mean N_2O emission fluxes were also observed among
300 the wastewater loading area and the non-wastewater loading area (p<0.05; Table S4
301 and Figures S4d-f). Across the three sampling campaigns, specifically, Sector-N had
302 significantly lower mean N_2O fluxes than the other four wastewater loading sectors
303 over the whole study period (Figure 5b), while Sector-R had relatively higher mean
304 N_2O fluxes than other sectors (Figure 5b).

305 *3.5. Water Quality Drivers of Variation in N_2O Concentrations and Fluxes*

306 The simple regressions analysis demonstrated thatthe spatial variation of N_2O
307 concentration and fluxes was positively related to the $N{-}NO_3^-$ (r^2 = 0.20−0.87,
308 p<0.01), $N{-}NH_4^+$ (r^2 = 0.17−0.42, p<0.01 or <0.05) and TDN concentrations (r^2 =
309 0.13−0.88, p<0.01) within each sampling occasion (Table S5, and Figures S5 and S6),
310 respectively, and negatively correlated with that of salinity and DO (p<0.05 or <0.01;
311 Table S5). Principal component analysis (PCA) was also employed to explore the
312 relationships among all these environmental variables over whole reservoir survey
313 during each sampling campaign (Figure 7a-c). The first component (PCA I),
314 explaining 93.1% (Figure 7a), 95.8% (Figure 7b) and 95.7 (Figure 7c) of the total
315 variances during the autumn, spring and summer surveys, respectively, was
316 significantly correlated with $N{-}NO_3^-$, $N{-}NH_4^+$ and TDN. The first component can be

317 considered as the influence of water pollution, reflecting the spatial variation. The
318 second component (PCA II), explaining 3.6%, 3.8% and 4.2% of the variances during
319 the autumn, spring and summer surveys, respectively, was correlated with the EC,
320 salinity and DO. In addition, PCA was employed to explore the temporal correlation
321 between the N_2O concentration (or fluxes) and environmental variables. The first and
322 second components together explained 98.2% of the total variance (Figure 7d). In
323 particular, the PCA I contributed 93.8% of the total variance and was correlated with
324 the $N{-}NO_3^-$, $N{-}NH_4^+$, TDN and temperature (Figure 7d).

325 *3.6. Effects of Different Sampling Size on the Estimation of Whole-Reservoir N_2O*
326 *Fluxes*

327 The results of Monte Carlo analysis showed that the accuracies of average
328 diffusive N_2O fluxes across the whole reservoir increased considerably with the
329 expansion of numbers of sampling sites included in flux estimations (Figure 8). For
330 each sample size, the resampling process was repeated 10,000 times to create the
331 boxplots of N_2O resampling means (Figure 8a) and the standard deviation of
332 resampling means (Figure 8b).

333 In Figure 8a, the light grey, grey and dark grey shaded areas show the respective
334 70%, 95%, and 99% of the Highest Probability Density (HPD) ranges which indicate
335 the variance of standard errors for approximate coverages of the resampling means
336 from 10,000 simulations for each sample size. With the increase of sample sizes, the
337 converging HPD ranges of possible resampling mean N_2O values indicated the
338 improving estimation accuracy of N_2O flux. In addition, with the expansion of sample

sizes, the biases of the resampling N_2O estimations declined, and the standard deviations of resampling means decreased exponentially (Figure 8b). Notably, the results of Monte Carlo analysis showed that increase of the sample size from 10 to 100 markedly reduced the relative standard deviation of mean diffusive N_2O flux from 21% to 1.2%.

The number of samples in Figure 8 could be used to determine an optimal sample size to produce reasonable flux estimates. However, to achieve less than 5% of error, more than 80 samples over the whole reservoir were still needed (ca. 15 samples per km^2), due to reservoir heterogeneity. If the sample sizes were less than 80, the uncertainty range of the resampling simulations increases because of missing some measurements from the N_2O hotspots. Therefore, it is important to conduct high spatial resolution sampling to include all N_2O hotspots to improve the accuracy of flux estimates.

According to the 10,000 simulations of only 10 samples, a relative standard deviation of the estimated mean diffusive N_2O flux was 22.6% (Figure 8b). Despite a large range, these means had a median value of 107 μmol m^{-2} h^{-1} (Figure 8a). The bias of the resampling median is small compared to the mean of all samples. Therefore, the high spatial resolution measurements from different transects can effectively reduce the estimation bias of N_2O flux in the current study.

Moreover, the standard deviations of resampled N_2O mean diffusive flux decreased dramatically from less than 10 μmol m^{-2} h^{-1} for the sample size of 40 to 1.29 μmol m^{-2} h^{-1} for the sample size of 100 (Figure 8b). The relative standard

deviation of mean diffusive flux reduced to only 1.2% for the sample size of 100, indicating that the estimated whole-reservoir N_2O emissions approached the actual mean, with much less variation. Increasing the sample size could effectively reduce the estimation bias of the whole-reservoir N_2O emissions.

4. Discussion

4.1. Subtropical Coastal Reservoir are Atmospheric N_2O Sources

Results from our spatially intensive sampling showed that the average N_2O emission flux from Wenwusha Reservoir were 107.0 μg m^{-2} h^{-1}. Although, the average N_2O fluxes in the Wenwusha Reservoir were smaller than those in some tropical reservoirs (Table S6), for example, Tucuruiand reservoir, Samuel reservoir (Lima et al., 2002), Petit Saut reservoir and Fortuna reservoir (Guérin et al., 2008), the N_2O fluxes in our reservoir were one to two orders of magnitude higher than those in many reservoirs in subtropical (Liu et al., 2011a; Musenze et al., 2014; Shi et al., 2020; Yu et al., 2018) and temperate regions (Cheng et al., 2019; Descloux et al., 2017; Huttunen et al., 2002) (Table S6). The mean N_2O emission rates in our reservoir were also substantially higher than the median emission rate (7.2 μg m^{-2} h^{-1}) in global lakes/reservoirs (Hu et al., 2016), and were approximately 2.5 times higher than the average of China's reservoirs (42.3 μg m^{-2} h^{-1}) (Li et al., 2018). Compared with lakes worldwide (Table S6), our reservoir had a larger N_2O emission fluxes. For example, the mean N_2O emission rates in our reservoir was approximately 8.7 times higher than the average of China's lakes (12.2 μg m^{-2} h^{-1}) (Li et al., 2018). In addition, the mean N_2O emission from our reservoir was approximately 3.9 and 1.9 times higher than

383 emissions from *Cyperus malaccensis* marsh (Wang et al., 2018) and aquaculture
384 ponds (Yang et al., 2020) in the same subtropical coastal region. Futhermore, high
385 N_2O emission fluxes has been reported in other subtropical coastal reservoirs studies
386 (e.g., Cheng et al., 2014; Zhang, 2012). These results together indicate that subtropical
387 costal reservoirs could be important sources of atmospheric N_2O and therefore more
388 attention is needed.

389 *4.2. External Sewage Input Affects the Spatial Pattern of N_2O Emission in the*
390 *Reservoir*

391 There are some studies on the small-scale spatial variations in N_2O flux across
392 the water-air interface in inland aquatic ecosystems (e.g., reservoirs, lakes) (Cheng et
393 al., 2019; Musenze et al., 2014; Zhao et al., 2013; Xiao et al., 2019a). However, such
394 knowledge is still lacking in coastal reservoirs. In the current study, our intensive
395 sampling in multiple sites shed light on the marked spatial variation in N_2O emissions
396 in Wenwusha Reservoirin (Figure 6), and the coefficients of variation changed from
397 81% to 110% in the three sampling campaigns. Notably, the wastewater loading area
398 only took up 30% of the whole reservoir area, but it contributed approximately 60%
399 of N_2O emissions in the whole reservoir.

400 Some scholars have found that the microbial processes of N_2O production can be
401 promoted by large provision of of N substrate, subsequently increasing water N_2O
402 concentration and emission to atmosphere (Herrman et al., 2008; Huttunen et al., 2003;
403 Liu et al., 2011a; Xiao et al., 2019a, 2019b; Yu et al., 2013; Zhao et al., 2014). In this
404 study, some environmental parameters are relative stable in the whole reservoir, for

example, air temperature, atmospheric pressure, solar radiation, and wind speed (wind-dependent gas transfer coefficient) , but some parameters had marked spatial variations, such as the concentrations of $N\text{-}NH_4^+$, $N\text{-}NO_3^-$, and TDN (Figure 2). Located in a catchment with intensive human activity, the Wenwusha Reservoir catchment is heavily affected by anthropogenic activities, particularly wastewater discharge from domestic and industrial sources, and drainage from mariculture ponds, (Figure 1). Results in our reservoir indicated the the highest average concentrations of $N\text{-}NH_4^+$, $N\text{-}NO_3^-$, and TDN from the river input sector, suggesting the important effect of the upstream area in providing nitrogen to the waterbody. Different from other wastewater loading sectors, non-wastewater loading sector had much lower $N\text{-}NH_4^+$, $N\text{-}NO_3^-$, and TDN concentrations and obviously higher DO level (Table S3), suggesting a improved water quality without the impact of sewage input. Therefore, the changing levels of sewage discharge was one of the primary factors controlling the large spatial variation in N_2O emissions (Figure 6).

The changes of N_2O fluxes between the two reservoir zones (Figure S3) further confirmed the role of sewage discharge in controlling N_2O emissions from the Wenwusha Reservoir. During our study period, N_2O concentrations (Figure 4 and Figure S3a) and emission fluxes (Figure 6 and Figure S3a) in the NRZ were much higher than those in the SRZ. This is likely due to that the markedly higher concentrations of $N\text{-}NH_4^+$, $N\text{-}NO_3^-$, and TDN (Figure 2) enhanced N_2O production in NRZ, as a consequence of more input from wastewater (Figure 1). The relationship between sewage input, N substrates, and N_2O emission has been found in lakes (e.g.,

427 Xiao et al., 2019a), coastal river network (Yu et al., 2013), and inland reservoirs (e.g.,
428 He et al., 2017; Liu et al., 2011a, 2017). In the current study, N_2O production data is
429 unavailable. However, the significantly positive relationships between the N_2O
430 concentration (NO_2 emission fluxes) and N concentrations in Wenwusha reservoir
431 ($p<0.05$ or <0.01; Table S4 and Figures 5-7) confirmed the above explanation.

432 The dissolved N_2O concentrations in the wastewater of municipal sewage
433 drainage channels, aquaculture ponds, and rivers adjacent to the reservoir were about
434 3–5 times higher than those in the reservoir surface water, and this caused a direct
435 input of dissolved N_2O into the Wenwusha Reservoir, with a consequence of steeper
436 gradients of N_2O concentrations between the surface water and atmosphere and
437 large N_2O diffusive emissions. Despite some environmental protection measures, a
438 large amount of sewage was still discharged into the reservoir. This external N_2O
439 input can support N_2O emissions continuously from water surface into air, even lack
440 of internal N_2O production inside the water ecosystem. Certainly, the contributions of
441 sewage discharge to the direct input of N_2O into the waterbody should not be
442 overlooked. Effective measures are urgently needed to reduce and finally stop the
443 discharge of sewage into the reservoir.

444 *4.3. Role of Salinity in the Spatial Variation in Reservoir N_2O Emission*

445 Except for N availability, the spatial variation in N_2O concentrations and fluxes
446 in the Wenwusha Reservoir may be influenced by the difference in salinity. Salinity
447 controls the mineralization rate of N through its effect on microbial activity, enzyme,
448 and metabolism, and consequently regulates the rates of N_2O production,

consumption, and emission into the atmosphere. Some studies suggested that high salinity can reduce microbial diversity (Francis et al., 2003; Li et al., 2020; Moin et al., 2009; Mosier and Francis, 2008) and activity (Wang et al., 2010), especially the ammonia-oxidizing bacteria (AOBs), with the consequence of reducing sediment / water N_2O production (Rysgaard et al., 1999; Wang et al., 2018). The negative relationship between salinity and N_2O concentrations (flux) has been reported in coastal wetlands and aquatic ecosystems (Liu et al., 2015; Sun et al., 2015; Wang et al., 2018; Welti et al., 2017). In the present study, salinity in the surface water showed remarkable spatial variations (Figure 3j-l), with the NRZ exhibiting significantly lower salinity than the SRZ (p<0.01; Figure S3c), as a result of freshwater dilution caused by the river runoff (Figure 1). The salinity (Figure 3j-l) and N_2O concentration (or flux) (Figure 4 and 6) exhibited opposite spatial patterns in the reservoir (p<0.01; Table S5), indicating that salinity might be an important environmental factor influencing N_2O dynamics between the two reservoir zones. Yet, salinity was likely not a dominant factor in governing the spatial variations of N_2O emission fluxes in each zone, since the spatial heterogeneity of water salinity within each zone was very low (Figure 3j-l). Meanwhile, the substantial differences in salinity beween two reservoir zones also provide additional evidence for that the large contribution of external N loading to the spatial variation in N_2O fluxes within Wenwusha Reservoir. Further studies merit to explore the exact impacts of salinity on N_2O production and emission.

4.4. Temporal Variation in N_2O Emission Fluxes Across the Coastal Reservoir

In this study, the N_2O flux exhibited clear seasonal variation (p<0.01; Table 1), with significant higher values in summer (Figure 5). Similar seasonal pattern was also observed in inland freshwaters (Musenze et al., 2014; Xiao et al., 2019a, 2019b; Zhu et al., 2013). Temperature is generally considered as the most important factor driving N_2O emission temporal patterns due to its stimulation of microbial activity related to N_2O production (Beaulieu et al. 2010; Harrison and Matson, 2003; Hinshaw and Dahlgren 2013). Larger N_2O fluxes in summer than autumn and spring (Figure 5) coincided with higher warmer temperatures in summer (Figure S1a) across the whole reservoir. However, an interesting outcome was that, although the water temperature was higher in spring than autumn (Figure S1a), the N_2O concentration and flux (Figures 3 and 5) showed an opposite patterns with water temperature. This was probably related to the disturbance of precipitation. In our study period, several continuous rain events occurred during the spring surveys. The large precipitation not only dramatically increased water depth in the reservoir, but also increased inflow of water and soil drained into the reservoir by rain falling on surrounding land, ultimately causing a dilution effect on many of the reservoir's biological and chemical parameters. Thus, instead of higher temperature, the dilution effect from large rainfall and river flows may be responsible for the lower N_2O concentration in spring than in autumn. Similar results were reported by He et al. (2017) and Outram and Hiscock (2012). In addition, annual drainage is a typical management activity that is practiced in autumn period after mariculture harvest, and it is a way to export aquaculture effluent. Thus, the N_2O concentrations and emission fluxes in autumn were larger than

493 those in spring also might be related to the aquaculture effluent discharge, which
494 provides a large amount of N substrate for reservoir's N_2O production. This
495 hypothesis was supported by the evidence that N_2O concentrations and emission
496 fluxes showed significantly positive relationships with $N{-}NO_3^-$, $N{-}NH_4^+$ and TDN
497 (p<0.01; Table S5), respectively. These results together indicated that the seasonal
498 variations in the N_2O concentrations and emission fluxes in the Wenwusha Reservoir
499 were regulated synthetically by temperature, precipitation and external N input.

500 *4.5. Implications of Spatially Resolved Measurements of Reservoir N_2O Emission*

501 In the past twenty years, scholars have gradually realized the large N_2O emission
502 from reservoirs to atmosphere (e.g., Deemer et al., 2016; Descloux et al., 2017;
503 Guérin et al., 2008; Li et al., 2018; Maavara et al., 2019; Wang et al., 2017). However,
504 the accurate contribution of reservoirs to the global N_2O budget remains highly
505 uncertain. Our results show that subtropical costal reservoirs are important
506 atmospheric N_2O sources with substantive spatial variability. The large spatial
507 variability in N_2O fluxes within the reservoir implies a large uncertainty of the whole
508 reservoir estimate of N_2O fluxes provided by earlier studies that were conventionally
509 based on single site or limited number of site measurements. The results of Monte
510 Carlo analysis further showed that increasing the sample size from 10 to 100
511 significantly reduced the relative standard deviation of mean diffusive N_2O flux from
512 22.6% to 1.6% (Figure 8). In the process of extrapolating small-scale N_2O fluxes to
513 the whole reservoir, therefore, our results highlight the importance of conducting high
514 spatial resolution sampling (>15 samples per km^2) to minimize the bias.

Our high spatial resolution sampling data show significant larger N_2O emissions from the wastewater loading area, indicating the important role of wastewater input for promoting N_2O emissions from reservoirs. With the world entering anthropocene, human activities, particularly wastewater discharge, have severely increased the input of anthropogenic nutrients to aquatic ecosystems, particularly in developing countries with poor wastewater treatment system (Hosen et al., 2014; Williams et al., 2016; Yang et al., 2013; Yu et al., 2013), but insufficient attention has been given to anthropogenic activities as an accelerator influencing the N_2O and other greenhouse gas emissions from water (He et al., 2017; Wang et al., 2017; Xiao et al., 2019a; Xing et al., 2006; Yang et al., 2008). Clearly, strengthening environmental protection, particularly wastewater treatment and discharge, is the key to mitigate water pollution and greenhouse emission in aquatic environment (Yang, 2014; Yang et al., 2015b).

This study, to the best of our knowledge, represents the first attempt to quantify N_2O emission fluxes from costal reservoir at such a high spatial resolution. Overall, the results provide a valuable data source for model development and validation to predict N_2O emission fluxes from costal reservoirs suffering from intensive human disturbance.

4.6. Limitations and Future Research

Similar to most studies, there are limitations in the current study. First, diurnal variations in N_2O flux have been reported in inland freshwaters (Baulch et al., 2012; Laursen and Seitzinger, 2004; Wu et al., 2018; Xiao et al., 2019a; Yang et al., 2011), and some researches found larger fluxes during nighttime compared with daytime

(Baulch et al., 2012; Wu et al., 2018; Yang et al., 2011). However, the day–night pattern of N_2O fluxes in the Wenwusha Reservoir is unknown, which may lead to estimate uncertainty of seasonal or annual N_2O budget. Therefore, future long-term observations along with diurnal temporal scales can further reduce the estimate bias of N_2O emission budget. Moreover, thermal stratification is a phenomenon frequently happening in reservoirs, particularly in deep reservoir (Straskraba, 1999; Thornton, 1990). The stratification could create barriers to the exchange of material (e.g., nutrient and DO) between the epilimnion and the hypolimnion (Hayes et al., 2107; Yu et al., 2015), ultimately resulting in a change of the main pathways of N_2O production in the water profile (Beaulieu et al., 2015; Liang et al., 2019; Salk et al., 2016) and the subsequent emission to the atmosphere. However, the vertical pattern of N_2O dynamic within reservoir in the current study is unknown, which limits the understanding of N_2O production and spatial patterns. In the future work, therefore, there is an urgent need for utilizing advancing technologies (e.g., isotopic tracing) to investigate the vertical profile of N_2O dynamic in coastal reservoirs. Furthermore, this study examined N_2O concentrations and fluxes, and basic environmental factors *in situ*, while the contributions from other N_2O production pathways, and microbial abundance and activity (nitrifier, denitrifier, and ammonifier) are unclear. Thus, the future detailed investigation of the more production pathways and microbial mechanisms of N_2O in coastal reservoirs will be helpful for understanding the mysterious N cycle in aquatic ecosystems.

5. Conclusions

We estimated N_2O emissions from Wenwusha Reservoir based on high spatial resolution data. Total surface water diffusive N_2O fluxes was estimated to be 4.8 Mg yr^{-1}, with an annual average CH_4 flux of 107.0 μg m^{-2} h^{-1}. Fluxes in N_2O were much greater in sectors in which sewage is discharged to the reservoir, due to greater inputs of anthropogenic N. As a result of high levels of nutrient and organic matter loading, areas of wastewater loading (comprising 30% of the reservoir area) acted as potential N_2O emission hotspots, and made a disproportionate contribution (60%) to whole-reservoir N_2O emissions. Our results illustrate that coastal reservoirs are an important source of atmospheric N_2O, where spatial variation in emissions is driven by anthropogenic activity. To produce representative, accurate estimates of N_2O emissions from coastal reservoirs suffering anthropogenic disturbance, it is essential they are based on highly spatially resolved data.

Declaration of interest

None

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