



On the contribution of atmospheric reactive nitrogen deposition to nitrogen burden in a eutrophic Lake in eastern China

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ABSTRACT

Although it has been demonstrated that atmospheric reactive nitrogen (i.e., Nr mainly including NH_3 , NH_4^+ , NO_x , NO_3^- and etc.) deposition has substantial impacts on nitrogen pools in remote and/or sensitive lakes, there is a scarcity of systematic evaluations regarding the impact on nitrogen burden in eutrophic lakes with riverine input as primary nitrogen source. Utilizing a regional atmospheric chemical transport model, combined with observation-based estimates of atmospheric nitrogen deposition fluxes and riverine nitrogen inputs, we investigate the contribution of atmospheric Nr deposition to the fifth largest freshwater lake located in eastern China, i.e., the Chaohu Lake which is facing frequent outbreaks of algal bloom. The results indicate that in the studied year of 2022, riverine total nitrogen (TN) input to the lake was $11553.3 \text{ t N yr}^{-1}$ and atmospheric TN deposition was $2326.0 \text{ t N yr}^{-1}$. For Nr species which are directly available for the biosphere supporting algae and plant growth, riverine NH_4^+ input was $1856.1 \text{ t N yr}^{-1}$ and atmospheric NH_x (NH_3 and NH_4^+) deposition was $824.5 \text{ t N yr}^{-1}$. The latter accounts for 30.8% of total NH_x input to the lake. For NO_y (HNO_3 and NO_3^-), riverine NO_3^- input was estimated as $2621.7 \text{ t N yr}^{-1}$, while atmospheric NO_y deposition was $629.3 \text{ t N yr}^{-1}$, accounting for 19.4%. In all, atmospheric Nr deposition accounts for 24.5 % of total Nr input to the lake. Our results suggest that even in regions with dense human activities with primary riverine N input, atmospheric deposition of Nr could also contribute significantly to the bio-available nitrogen in lake systems, and addressing eutrophication in Lake Chaohu and other eutrophic lakes will also need to consider the influence of atmospheric Nr deposition which is related to NH_3 and NO_x (i.e., $\text{NO} + \text{NO}_2$, the precursor of NO_y) emissions, in addition to the mitigation of riverine N input.

1. Introduction

Due to human population expansion and industrialization, anthropogenic processes have become the dominant force of reactive nitrogen (Nr, including NH_3 , NO_x , NO_3^- and etc.) production (Galloway et al., 2004). As a result, the total amount of Nr circulating within the biosphere has more than doubled compared to the pre-industrial period, and the global patterns of atmospheric Nr deposition have also been largely impacted (Galloway et al., 2008). There are numerous studies that demonstrate large increases in atmospheric Nr deposition (e.g., (Asman et al., 1998; Liu et al., 2022)), and many natural archives such as

ice cores, lake deposits and sea coral cores record the elevated global Nr deposition as a result of human activities (Mayewski et al., 1990; Holtgrieve et al., 2011; Ren et al., 2017). The Nr deposition is particularly large in regions with dense populations such as China which a hotspot of anthropogenic Nr emission. The deposited Nr is directly available for the biosphere (Galloway et al., 2003), and it affects nutrient levels and alters biogeochemical cycles, impacting primary productivity in marine and terrestrial ecosystems. In particular, atmospheric Nr deposition can stimulate the growth of phytoplankton in oligotrophic lakes and oceanic waters (Paerl, 1985; Bergström and Jansson, 2006), and this would also lead to changes in the

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nitrogen/phosphorus (N/P) stoichiometry and nutrient limitation in lakes away from direct human interference (Elser et al., 2009).

For eutrophication lakes in regions with dense population, riverine inputs in general are thought to be the dominant N source, while the contributions and the associated ecological effects of atmospheric Nr deposition have not been fully revealed and understood (Liu et al., 2011). For lakes, there are multiple sources of N, including surface and subsurface flow pathways, atmospheric Nr deposition, and N₂ fixed by cyanobacteria in lake (Sutton et al., 2011), of which riverine inputs and atmospheric deposition are in general considered as the major sources (Xu et al., 2021). For example, recent observations indicated that atmospheric inputs of N (including Nr and organic N) accounted for 16.0% in Lake Dianchi, Southwest China (Zhan et al., 2017), and 13.9%–27.3% in Lake Taihu, the third largest freshwater lake in China (Deng et al., 2023). Given the continuously improved wastewater treatment efficiency over the past decades (Tong et al., 2020), a distinct water quality improvement was demonstrated at the national scale (Huang et al., 2021). At the same time, although the implementations of nationwide air pollution mitigation strategies in the past few years have also reduced atmospheric aerosol concentrations, there have been no dramatic reductions in atmospheric nitrate and ammonium concentrations (i.e., the two main forms of atmospheric Nr) (Meng et al., 2022). Therefore, presumably the role of atmospheric Nr deposition in lake nitrogen burden may have become more important.

Accurately estimating the contribution of atmospherically deposited Nr to the total N input is crucial for comprehending the N budget and eutrophication mechanism of eutrophic lakes. Compared to the abundance of ground-based observations of air pollutant concentrations, deposition monitoring sites have been relatively sparse, especially for dry deposition, due to the difficulty and expense of direct measurement methods (e.g., eddy correlation, chambers) and indirect methods (e.g., inferential, gradient analysis) (Seinfeld and Pandis, 2006). Wet deposition from rain or snow is relatively easy to measure in existing networks such as CASTNET (Clean Air Status and Trends Network, United States), EMEP (European Monitoring and Evaluation Programme, Europe), EANET (Acid Deposition Monitoring Network in East Asia) and NNDMN (Nationwide Nitrogen Deposition Monitoring Network, China). Dry deposition fluxes, on the other hand, are usually derived from measured ambient concentrations of Nr species and calculated deposition velocities (Endo et al., 2011). Lake Chaohu is the fifth largest freshwater lake in China, located in a concentrated area of agriculture and industrial activities nearby the City of Hefei with over 10 million people. As a typical case of eutrophic lake with frequent outbreaks of algal bloom impacted by human activities, identifying the source of N in Lake Chaohu can provide insights into the pollution and eutrophication of aquatic ecosystems. However, by far the only estimate of atmospheric N input to Lake Chaohu came from single-point bulk deposition sampling method, reporting somewhat overwhelming contributions of atmospheric TN input, i.e., 39.6% and 48.4% of the total N input in Lake Chaohu in 2015 and 2019, respectively (Wei et al., 2018; Tian et al., 2022). However, the single-point bulk deposition sampling method is inadequately covering the entire lake and only captures wet deposition but unquantifiable dry deposition as well as deposition of gaseous nitrogen species (Liu et al., 2015), and thus the results should be treated with cautions. The NNDMN network containing 43 sites in China reveals that dry and wet/bulk Nr deposition fluxes were 20.6 ± 11.2 (mean \pm standard deviation) and 19.3 ± 9.2 kg N ha⁻¹ yr⁻¹ across China (Xu et al., 2015). But this network is currently not covering Lake Chaohu and any nearby regions. Atmospheric chemical transport models (CTMs) are often used to assess the deposition fluxes of atmospheric species including Nr at global and/or regional scales. The project of Model Inter-Comparison Study for Asia phase III (MICS-Asia III) reported that the mean Nr deposition flux over China under multiple CTMs in 2010 was 11.3 kg N ha⁻¹ yr⁻¹ (Ge et al., 2020). Similar result was also reported by using the MSC-W model (Meteorological Synthesizing Centre-West of EMEP) for 2008–2017, the annual mean Nr deposition was

simulated at 15.7 ± 0.9 kg N ha⁻¹ yr⁻¹ for China (Ma et al., 2023). Though the fluxes estimated by the models appear to be underestimated compared with the NNDMN observational results, the fraction of wet or dry deposition in total Nr deposition have shown consistent results with NNDMN.

In this study, in order to assess the contributions of atmospheric Nr deposition to N burden in Lake Chaohu, we employed a regional chemical transport model, the Weather Research and Forecasting Model coupled with Chemistry (WRF-Chem), to quantify atmospheric Nr deposition in Lake Chaohu in the winter of 2017/2018 and year of 2022 when river input of N can also be quantified using data from local water quality monitoring network. Observations of atmospheric aerosol NH₄⁺ and NO₃⁻ concentrations, as well as Nr deposition data in the city of Hefei are used to constrain the model results. This shall provide the first comprehensive dataset on the relative contribution of atmospheric deposition versus riverine inputs to N burden in Lake Chaohu, especially that of Nr. Based on this, nitrogen cycle and effects of nutrient supplies on the phytoplankton succession in Lake Chaohu as well as the associated environmental consequences can be evaluated.

2. Material and methods

2.1. Study area

Lake Chaohu (31°25′–31°42′ N, 117°16′–117°51′ E) is the fifth largest freshwater lake in China, located in eastern China nearby the megacity of Hefei with over 10 million population (Fig 1). The lake is characterized with a surface area of 780 km² and a mean depth of 2.89 m. It can be generally divided into two regions as Western Lake Chaohu (WLC) and Eastern Lake Chaohu (ELC). Severe eutrophication and harmful algal blooms have been a major environmental problem since 1995 (Shang and Shang, 2007), especially in the WLC. Although tremendous efforts have been made to restore its water quality, Lake Chaohu still faces mild eutrophication problem in recent years including 2023.

2.2. Potential sources of nitrogen inputs to Lake Chaohu

The Tenth Five-Year Plan of Water Pollution Prevention in Lake Chaohu indicated that as of 1999, riverine inputs accounted for 90% of the total nitrogen inputs, and the remaining 10% was from precipitation and groundwater, while atmospheric dry deposition was ignored (http://www.mee.gov.cn/gkml/zj/wj/200910/t20091022_172165.htm). As such, in the past decades, efforts from the local government have been focused on reducing nitrogen content in river water, and these efforts have improved the water quality of Lake Chaohu and made it stabilized at light eutrophication status. However, there has been no data to illustrate the contributions of atmospheric N deposition and if it is a significant source to the lake at least in this region. Meanwhile, the observations of atmospheric nitrogen deposition require much more complicated logistics. Thus, observations and assessments on atmospheric deposition are much ignored.

Therefore, in this study, we mainly focus on estimating the N inputs from the atmosphere to the lake in addition to riverine input.

2.2.1. Atmospheric nitrogen deposition modeling

The version (v3.5) of WRF-Chem, updated by the University of Science and Technology of China (USTC version of WRF-Chem), was used to simulate the emission, transport, chemical reactions, and dry and wet depositions of chemical species in this study. The model configurations used in this study are summarized in Table S1. In this study, simulations were performed at 36 km horizontal resolution with 150 (west–east) \times 150 (south–north) grid cells covering the southeast China. Two deposition types (dry and wet) and four Nr species (i.e., NH₃, HNO₃, NH₄⁺ and NO₃⁻) were included. These four species account for more than 90% of total atmospheric Nr deposition (Zhou et al., 2023) and are the main

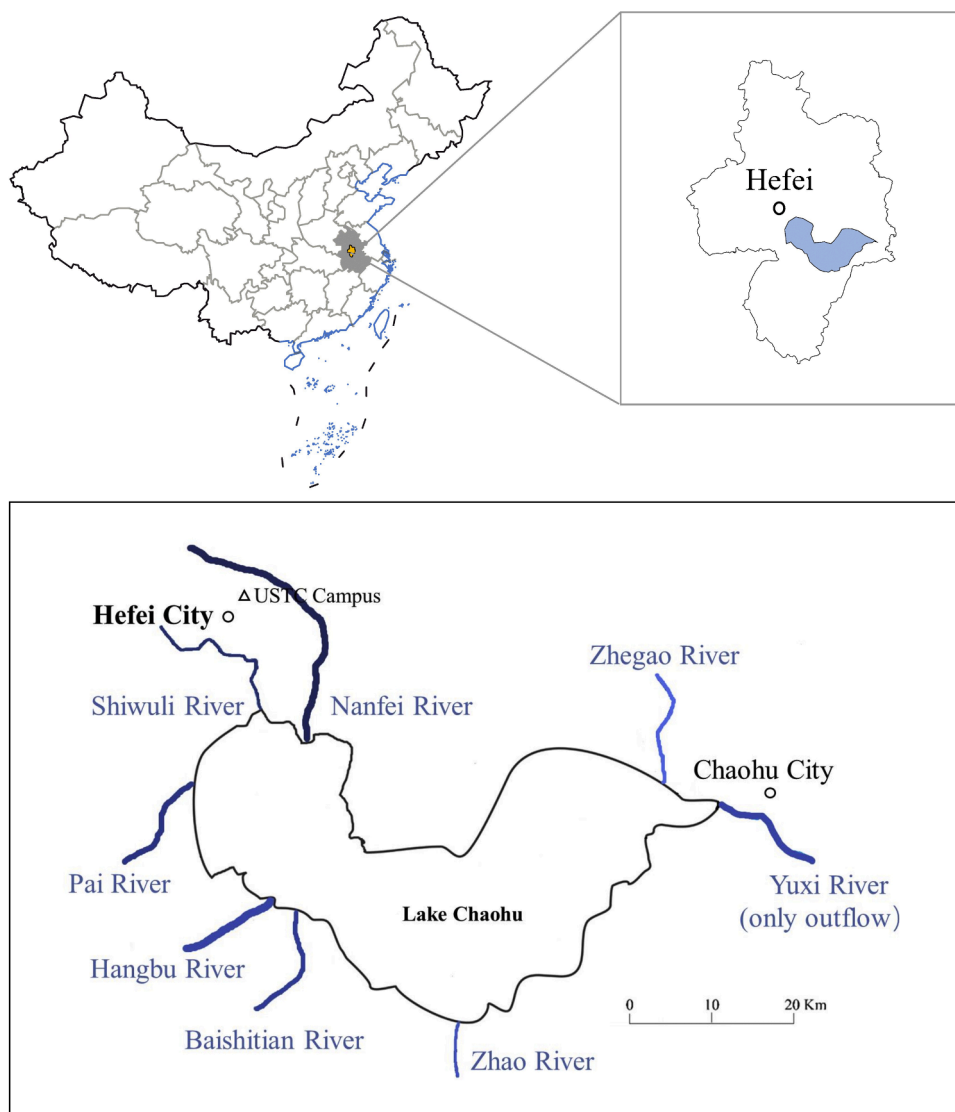


Fig 1. Geographic location of Lake Chaohu and its surrounding rivers. There are eight major rivers surrounding the lake, Yuxi River is the only outflow. The other seven inflow rivers were included in the estimation of riverine N inputs. The thickness of the line indicates the relative level of flow rates of these rivers, the shade of the color indicates the relative level of the total nitrogen concentrations of these rivers.

forms of nitrogen that can be directly used by plants and microbes. The simulations were performed in two periods, the winter of 2017/2018 representing by January 2018, and the year of 2022 representing by January, April, July and October to reflect the seasonal variations. Discontinuous simulations were conducted to save computation cost. From 2018 to 2022, air pollution mitigation strategies have reduced the emissions of NO_x which is the precursor of atmospheric nitrate (NH_3 emissions are not apparently affected) (Liu et al., 2023). Therefore, in order to reflect the potential effects of the reduced precursor emissions to atmospheric N_r deposition, we used the Multi-resolution Emission Inventory for China (MEIC, <http://meicmodel.org.cn>) 2015 inventory and 2020 inventory (Li et al., 2017; Zheng et al., 2018) for the 2018 and 2022 simulations, respectively.

In order to constrain the model results, we also collected daily ground observations of NH_4^+ and NO_3^- concentrations in $\text{PM}_{2.5}$ in January, April, July and October 2022. Those data directly constrain the modelled atmospheric NH_4^+ and NO_3^- concentrations which largely affect their deposition fluxes. Details can be found in the supplementary materials.

In addition, from April to August 2023, an automated wet and dry deposition sampler (ZR-3901, Qindao) was installed at the USTC

campus, which is 15.6 km away from the lake (Fig 1). The wet deposition sample was collected after each precipitation event and the dry sample was collected every two weeks to monitor atmospheric N deposition fluxes. The data in April and July 2023 are used to compare with the model results in the same months but in 2022. Although not exactly in the same year, but given the similar emission scenarios, and rainfall activities (50.1 mm and 134.2 mm in April and July 2022, respectively, 76.6 mm and 126.4 mm in April and July 2023, respectively), the deposition flux in 2022 and 2023 should not differ by a lot.

2.2.2. Estimation of riverine inputs

Data of river input flow rates and nitrogen concentrations were collected, and the riverine inputs of nitrogen was calculated as:

$$N_r = \sum f_i \cdot c_i \quad (1)$$

Where N_r is the total riverine N inputs to the lake, f_i and c_i are the flow rates and N concentrations of the rivers entering the lake, respectively.

There are seven main rivers flowing into Lake Chaohu (Fig 1). Monthly water quality data for the inflows of these seven rivers (i.e., river Baishitian, Hangbu, Nanfei, Pai, Shiwuli, Zhegao and Zhao) come

from the Chaohu Administration Bureau of Anhui Province, of which the four rivers in WLC (i.e., river Baishitian, Hangbu, Nanfei and Pai) account for 90% of the total flow entering the lake (Zhang et al., 1997), and according to current data the Nanfei river is the most polluted one. Ammonium nitrogen ($\text{NH}_4^+\text{-N}$) and total nitrogen (TN) concentrations in river water are monitored, while nitrate nitrogen ($\text{NO}_3^-\text{-N}$) is not within the scope of water quality monitoring. To complement the data, we used reported concentrations of NO_3^- and NH_4^+ in rivers flowing into Lake Chaohu (Wu et al., 2021) to estimate the ratio of NO_3^- and NH_4^+ in river water, and then we scaled the measured ratio of NO_3^- and NH_4^+ to periods with $\text{NH}_4^+\text{-N}$ data available, deriving the NO_3^- concentrations in river water. This way gives a first order of estimation on the riverine input of NO_3^- .

Among the seven rivers flowing into the lake, only the Hangbu and Zhao rivers have monitored flow rate data available from the Anhui Provincial Bureau of Hydrology. In order to get the flow rates of all other rivers including Nanfei which is the most polluted river and with the highest N contents, the method of hydrological analogy was used. This method is the simplest way to estimate flow rates for unmonitored river basins. It can give reasonable results where there are monitored rivers representative for regional hydrological, climatic, and land cover conditions (Shiklomanov et al., 2021) and has been used, for example, to estimate the freshwater discharge to the Arctic Ocean (Lammers et al., 2001). In brief, for the Chaohu Lake Basin, the meteorological conditions and subsurface factors are presumable the same or similar for all following in rivers, so that only the effect of drainage area (which for each river is available from the Anhui Provincial Bureau of Hydrology) is considered and the flow of unmonitored rivers can be calculated based on the ratio of flow and drainage area using the following equation:

$$f_{um} = \frac{D_{um}}{D_m} f_m \quad (2)$$

Where f_{um} and D_{um} are the flow and drainage area of unmonitored rivers, respectively, f_m and D_m are the flow and drainage area of monitored rivers, respectively. Based on the two rivers with available flow and drainage area data, we calculated the flows of other five rivers with their drainage area data. We note this method may induce uncertainties since differences in the meteorological conditions and subsurface factors of different river drainage area, it is currently the best way to estimate the flow rate which is necessary to calculate the N discharge.

3. Results and discussion

3.1. Temporal variations in riverine N input

Fig 2 displays the monthly amounts of riverine TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs of 2018 and 2022, along with the annual total inputs from 2018

to 2022. Fig 2a displays the results for the year of 2018, when monthly TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs ranged from 208.6 to 2417.1 t N month⁻¹, from 64.7 to 852.3 t N month⁻¹ and from 91.4 to 1203.8 t N month⁻¹, respectively. The monthly mean TN input was 1058.0 ± 610.6 t N month⁻¹, the mean $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs were 482.8 ± 263.8 and 682.0 ± 372.6 t N month⁻¹, respectively. In 2018, the changes in TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs were broadly consistent and all showed increases in March, May and July.

Fig 2b displays the results of the year of 2022, when monthly TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs ranged from 25.3 to 4271.8 t N month⁻¹, from 5.4 to 678.2 t N month⁻¹ and from 7.6 to 958.0 t N month⁻¹, respectively. The monthly mean TN input was 1011.8 ± 1149.4 t N month⁻¹, the mean $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs were 162.8 ± 179.5 and 230.0 ± 253.6 t N month⁻¹, respectively. Compared to the year of 2018, in 2022, TN input increased dramatically in March and July, however, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs were significantly less elevated than TN. In other words, the patterns of TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ in 2022 appeared to be diverged.

Fig 2c displays the annual total TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs from 2018 to 2022. As shown in the figure, TN inputs were relatively constant in these years except in 2020 when TN input increased by ~64% compared to other years. In contrast, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs display apparent decreasing trend especially after 2020, and by 2022 $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs had decreased by ~66% compared to 2018.

We also collected the data of annual riverine TN and $\text{NH}_4^+\text{-N}$ inputs to Lake Chaohu from literatures and compared with our study (Table S2). There are only a few relevant studies on Lake Chaohu, and most of which focus solely on TN with limited data available on $\text{NH}_4^+\text{-N}$. Nevertheless, our estimation is within the range of those reported in literatures.

The monthly riverine N input is either controlled by flow rate or N concentrations in river water, or both. It is noticed that in March 2018 and March 2022, TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs all increased. In March 2018, the TN input increased by ~242% and Nr input increased by ~146% compare to months before and after. In March 2022, the TN input increased by ~693% and Nr input increased by ~610% compare to months before and after. This is mainly due to the increased river flow rate caused by extensive rainfall at that time, which can also explain the increasing N input in July 2018. However, for July 2022 when TN input increased apparently by ~33%, flow rate decreased slightly by ~18%, compare to months before and after. The decreasing of flow rate may lead to high N concentrations, but additional effluents can also increase N concentrations and cause higher inputs. In July 2022, there was only one inflow river, Nanfei, which has evaluated TN concentrations. This suggests that in July 2022, there was additional effluent discharged into river Nanfei, probably large volumes of urban domestic sewage from Hefei city during summer. In addition, we examined the proportion of riverine N input accounted for by river Nanfei, which contributes ~66%

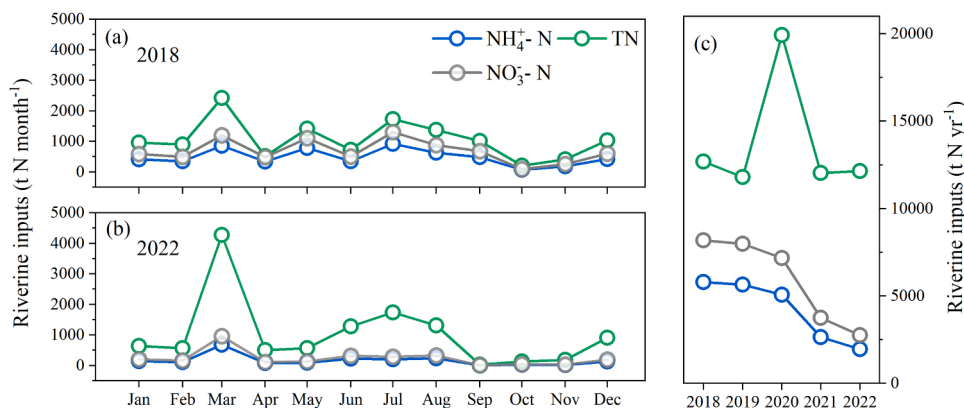


Fig 2. Riverine TN, $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ inputs to Lake Chaohu. (a) and (b) Monthly variation in 2018 and 2022 of riverine nitrogen inputs. (c) Annual average for the last five years.

and ~46% of total riverine TN and Nr inputs in 2018, and ~54% and ~43% in 2022, respectively. These results may be subject to uncertainties due to the assumptions of same or similar meteorological conditions and subsurface factors when estimating the flow rate, but they indicate that river Nanfei is the most important source of riverine N input to Lake Chaohu.

We also noted that in 2018, the monthly variations in TN and Nr were in general consistent, i.e., with similar degree of increases or decreases. But for the year of 2022, the monthly variations in TN and Nr were somewhat decoupled especially for summer when TN input appeared to be increased apparently more than that in Nr. In addition, this decoupling also occurred in river Nanfei. As mentioned above, the additional effluents in July 2022 increased the TN concentration significantly, while the $\text{NH}_4^+\text{-N}$ concentration of river Nanfei shown in the monitored data stayed stable at that time. Since there was no $\text{NO}_3^-\text{-N}$ data available, according to our estimation, the $\text{NO}_3^-\text{-N}$ concentration was close to $\text{NH}_4^+\text{-N}$. This implies that the effluent was likely to be dominated by organic N and therefore led the decoupling of TN and Nr concentrations. Thus, this decoupling in the year of 2022 may be a result of the increased coverage and efficiency of wastewater treatment in Hefei, removing inorganic N but may generate extra organic N in the effluents. In March 2020, revisions to the Chaohu Lake Water Pollution Control Regulations were published, which resulted in higher standards of effluent discharged into rivers and increased coverage and efficiency of sewage treatment in urban areas. This new regulation only set standards for TN and $\text{NH}_4^+\text{-N}$ in effluent discharges, while $\text{NO}_3^-\text{-N}$ and organic N were ignored. From our data, it is likely that after this new regulation $\text{NH}_4^+\text{-N}$ control was improved, while TN management was not. The latter could be due to the increase in organic N, and this may be associated with the mechanism of wastewater treatment (Supplementary Materials).

The annual riverine TN inputs are less variable compared to the monthly input, except in the year of 2020 when TN input increased by ~67% compared to the years before and after. This significant increase of TN input was likely due to elevated flow rate, which were attributed to a heavy rainfall event that lasted for two months in the summer of 2020, leading to ~135% increases in flow rate compared to the means of other years. In addition, the annual Nr inputs also appear to be decoupled with annual TN input since 2020, when Nr input decreased dramatically while TN input stayed constant. This decoupling can also be explained by the effects of the new regulations released in 2020. As

discussed earlier, the government's new control measures successfully reduced inorganic N concentrations in rivers but have little to no effects on TN probably due to the increases in DON concentration after treatment. Since DON can act as a substrate for microbial and algal growth in rivers and can be mineralized to bioavailable inorganic N, leading to eutrophication (Sattayatewa et al., 2009). Thus, maintaining high DON may hamper the efforts of Nr removal, and this should be addressed to further improve the water quality of Lake Chaohu.

In addition, we recommend that nitrate should be included in water quality monitor. Previous study measuring Nr in river water clearly indicates the presence of $\text{NO}_3^-\text{-N}$ with similar level to $\text{NH}_4^+\text{-N}$. Our study only made a first order estimation of river nitrate concentration and input without long-term observation. Currently, efforts to reduce $\text{NH}_4^+\text{-N}$ input have been successful but there is no observed $\text{NO}_3^-\text{-N}$ data to show how $\text{NO}_3^-\text{-N}$ has changed.

3.2. Atmospheric N deposition flux over Lake Chaohu

We first extracted the simulation results in the city Hefei and compared them with observations in the city to evaluate the model performance. Fig 3 displays the simulated wet and dry deposition fluxes in April and July 2022, along with the observed wet and dry deposition fluxes in April and July 2023. Although not in the same year, we note the model simulations would have used the same emission inventory (i.e., emission of 2020) for either 2022 or 2023 simulation, so that potential difference between the 2022 and 2023 simulation results could be due to differences in the frequency and magnitude of rainfalls in the two years that determines wet precipitations, and other meteorological parameters such as wind direction and speed. Nevertheless, the simulated Nr deposition fluxes in 2022 were 1.55 and 1.95 $\text{kg N ha}^{-1} \text{ month}^{-1}$, respectively. In comparison, the observed Nr deposition fluxes in 2023 were 1.08 and 1.86 $\text{kg N ha}^{-1} \text{ month}^{-1}$, respectively. Overall, the modeled and observed fluxes are at the same order of magnitude, though the modeled dry fluxes of NH_x appears to be much higher than the observations. The latter might be due to the well-known issue with the limitation of the flux observation method, i.e., in order to continuously capture dry deposition, the sampling bucket must be kept exposed to air and NH_x already collected in the sampling bucket may re-volatilize especially in high temperature seasons, resulting in lower dry NH_x deposition fluxes than real in practice (Pan et al., 2012). On the other hand, the model appears to underestimate wet deposition, and this

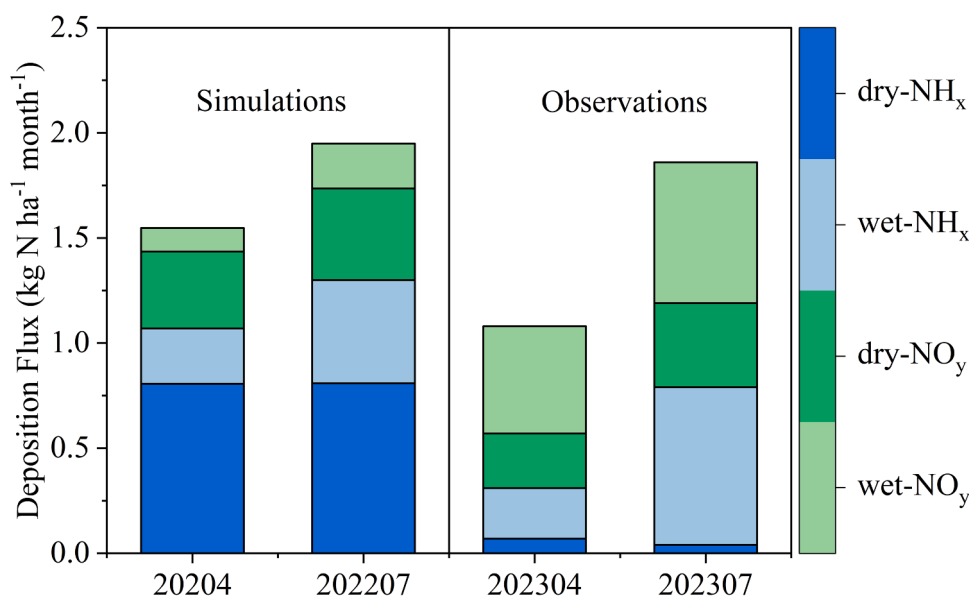


Fig 3. Comparison between the simulated and observed N deposition fluxes of Hefei. The deposition fluxes were divided into four parts according to the forms of N and types of depositions.

might be due to that CTMs have tended to underestimate wet deposition of nitrate-N in the Asian region because of the inaccurate modeled precipitations (Lu and Tian, 2014). Similar model-observation discrepancies are also noted by other studies. For example, Huang et al., (2015) simulated the Nr flux in Guangdong, China, indicating that the model overestimated dry depositions and underestimated wet depositions. Akter et al., (2023) found the Comprehensive Air Quality Model with Extensions (CAMx) model overestimates dry NH_x deposition flux but underestimates wet NO_y deposition flux in most regions of the United States. In all, we think the model at least captures the same order of magnitude of the Nr deposition fluxes, especially the underestimation of wet NO_y deposition offsets the overestimation of dry NH_x deposition, reducing the model-observation discrepancies.

The simulated Nr deposition fluxes in January 2018 and in the four months in 2022 over Lake Chaohu are displayed in Fig 4a. For January 2018, the total Nr deposition flux was $4.19 \text{ kg N ha}^{-1} \text{ month}^{-1}$, with $2.77 \text{ kg N ha}^{-1} \text{ month}^{-1}$ for NH_x and $1.42 \text{ kg N ha}^{-1} \text{ month}^{-1}$ for NO_y . For the four months in 2022, the total Nr deposition flux ranged from 1.15 to $1.81 \text{ kg N ha}^{-1} \text{ month}^{-1}$ and the mean Nr deposition flux was $1.55 \pm 0.32 \text{ kg N ha}^{-1} \text{ month}^{-1}$, of which the mean fluxes of NH_x and NO_y were 0.88 ± 0.29 and $0.67 \pm 0.12 \text{ kg N ha}^{-1} \text{ month}^{-1}$, respectively. The Nr deposition flux in January 2022 is $\sim 57\%$ lower compared to that in January 2018.

Averaging the results of these four months over the entire year, the mean Nr deposition flux in 2022 was estimated as $18.64 \pm 3.82 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, of which the mean fluxes of NH_x and NO_y were 10.57 ± 3.46 and $8.07 \pm 1.42 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, respectively. These annual results were comparable with other studies. For example, Itahashi et al., (2021)

applied the Community Multiscale Air Quality (CMAQ) model, returning values greater than $15 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ of Nr deposition in Eastern China in 2010. K. Zhou et al., (2023) developed a dataset with multiple statistical models that combine ground-level observations, chemistry transport simulations and etc., showing that the mean deposition fluxes of NH_x , NO_y and total Nr during 2005 to 2020 across China were 10.4, 14.4 and $24.9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, respectively.

3.2.1. Temporal variations in Nr deposition

Fig 4bc displays the simulated composition of particulate and gaseous Nr fluxes from wet and dry deposition. As shown in the figure, the lower Nr deposition flux in January 2022 compared to January 2018 mainly came from the decrease in dry NH_3 and wet particulate Nr deposition fluxes. Ambient Nr concentrations and deposition rates (dry deposition) or precipitation/ snowfall (wet deposition) can directly influence N deposition fluxes. For the decreased dry NH_3 flux, the simulated deposition rates are the same in January 2018 and 2022 in the model, so we checked the mean simulated concentrations of ambient NH_3 over Lake Chaohu in January 2018 and January 2022, which were 0.016 ppmv and 0.007 ppmv, respectively. This reduction of ambient NH_3 concentration is probably due to the lower Nr emissions, which can directly affect ambient Nr concentrations. We examined NH_3 emissions in the emission inventory, i.e. the MEIC 2015 and 2020 emission inventories used in simulation for 2018 and 2022, respectively. From 2018 to 2022, NH_3 emissions decreased by $\sim 29\%$, from 1.30 to $0.92 \text{ kg N ha}^{-1} \text{ month}^{-1}$. Consequently, lower NH_3 emissions resulted in lower ambient NH_3 concentration, leading to such decline of dry NH_3 deposition flux between January 2018 and January 2022. In January 2022, the wet

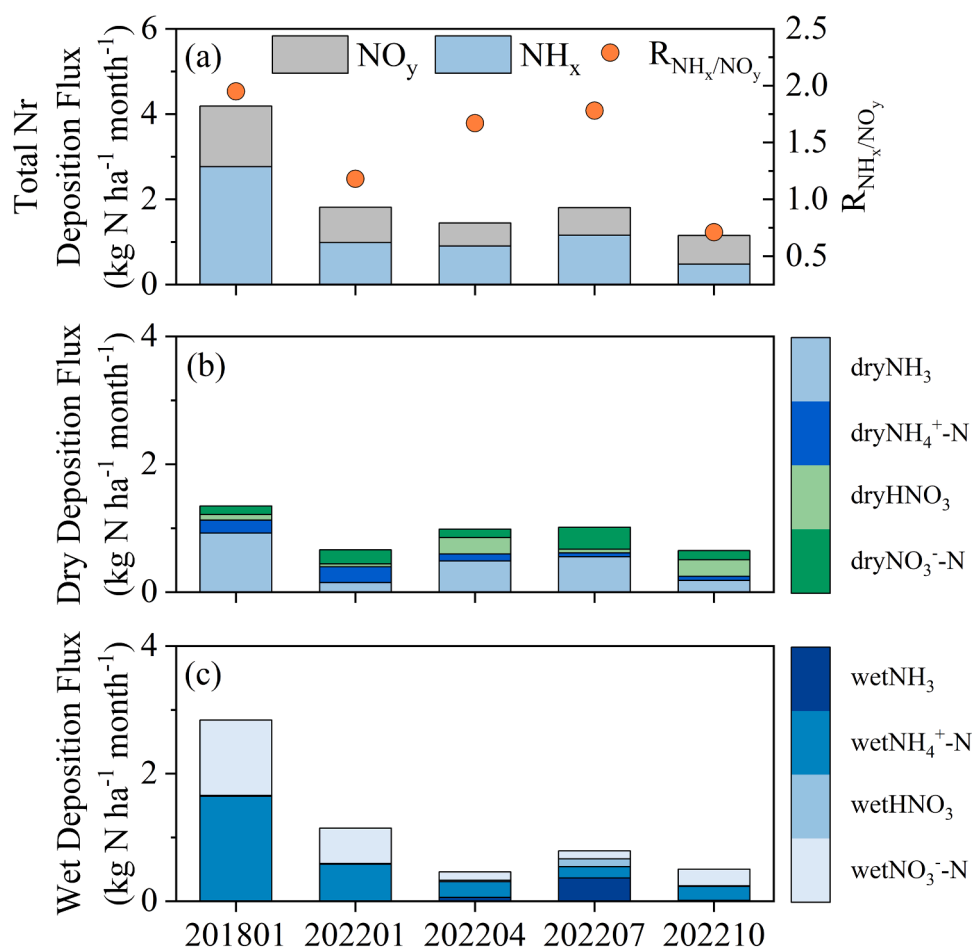


Fig 4. Simulated atmospheric deposition fluxes of Nr species over Lake Chaohu in the five months. (a) Monthly mean total Nr deposition fluxes over the lake, the orange dots represent the ratio between NH_x and NO_y deposition fluxes. (b) Monthly mean fluxes of dry deposition. (c) Monthly mean fluxes of wet deposition.

deposition flux of TN also decreased. We also first examined the monthly mean simulated concentrations. In January 2018, the NH_4^+ and NO_3^- concentrations were 7.62 and 20.95 $\mu\text{g m}^{-3}$, respectively. While in January 2022, the NH_4^+ and NO_3^- concentrations were 9.82 and 26.34 $\mu\text{g m}^{-3}$, respectively. Though the ambient particulate Nr concentrations didn't change significantly, there was a severe snowstorm in Hefei during January 2018. This heavy snowfall event can deposit a lot of Nr since snow can effectively scavenge atmospheric NH_x and NO_y (Murray et al., 2022). These may together explain why the deposition flux in January 2022 is low compared to January 2018.

For the year of 2022, we also noticed monthly variations which could be attributed to variations in Nr emissions and meteorological conditions (such as precipitation/snowfall and temperature). The major anthropogenic sources of NH_x are agricultural fertilizer use and human or animal waste, with relative low contributions from combustion-related processes (i.e., coal combustion, NH_3 slip, and vehicle exhaust) (Zhang et al., 2020), while NO_y mainly comes from industrial activities (e.g., coal power plant) and transportation (Pan et al., 2012). The high agricultural activities in spring and summer in nearby regions of Lake Chaohu may explain the high NH_x to NO_y ratio mainly in April and July, while industrial and transport emissions may become more important in January and October. In addition, some studies indicated that NH_3 volatilization from farmlands increases with air temperature in a nonlinear manner (Sommer et al., 2004). Elevated temperatures also promote the release of ammonia from plant leaves (Asman et al., 1998). So, the high NH_x to NO_y ratio, as well as the high dry NH_3 deposition fluxes in April and July as shown in Fig 4b are likely also associated with their relatively high temperature which facilitate NH_3 emissions from soil land. Other than changes in source emissions, frequency and intensity of precipitation or snowfall can affect wet deposition. As shown in Fig 4c, the high wet deposition flux in January 2022 could be attributed to winter snowfall, and the second highest wet deposition occurred in July, when rainfall was often more frequent and heavier.

3.2.2. Estimation of TN deposition flux

Apart from Nr (i.e., inorganic N), organic N also contributes to atmospheric TN deposition. On a global scale, Li et al. (2023) reported that the organic N contributed 21% of the atmospheric TN deposition, with the dominant depositing component was highly water-soluble and may potentially support algae growth in lakes. In our model, we didn't include the simulation of atmospheric organic N, but according to the two months deposition fluxes observations in Hefei (Details are shown in Table S3), we obtained the ratio of deposition fluxes of Nr/TN, which is 0.585. Assuming this ratio is representative of all yearlong scenario, we can estimate TN deposition flux according to the modeled Nr deposition flux. As a result, in 2022, TN deposition is 31.86 $\text{kg N ha}^{-1} \text{yr}^{-1}$ over Lake Chaohu, and among which organic N deposition flux was 13.22 $\text{kg N ha}^{-1} \text{yr}^{-1}$. We note this is only a very rough estimate, and need to be

further evaluated with more comprehensive model and observation efforts.

3.3. Contribution of atmospheric N deposition to N input to the lake

By multiplying the N deposition fluxes by the lake surface area, the N deposition inputs received by Lake Chaohu can be estimated. Fig 5 displays the monthly contributions of atmospheric N deposition to N burden in the lake for the five months with simulated and estimated N deposition fluxes. The mean monthly contributions were $42\% \pm 12\%$, $29\% \pm 19\%$ and $31 \pm 15\%$ for atmospheric NH_x (NH_3 and NH_4^+), NO_y (HNO_3 and NO_3^-) and TN deposition, respectively. There was no significant change in the contributions of N deposition from January 2018 to January 2022, indicating decline in both atmospheric deposition and riverine N inputs during these years. In 2022, the contributions peak in October and trough in July, corresponding to periods of high and low water abundance. The contribution in January is higher than that in October due to deposition input from winter snowfall. This indicates that atmospheric N deposition can overtake riverine input as the major source of N input to Lake Chaohu, especially Nr deposition during periods of low precipitation or snowfall.

As mentioned above, the TN deposition flux over Lake Chaohu was estimated to be 31.86 $\text{kg N ha}^{-1} \text{yr}^{-1}$. We can estimate the relative contributions of atmospheric vs. riverine input of TN and Nr to the lake at the annual scale. As shown in Fig 6a, the direct deposition of atmospheric TN to the lake would contribute 17.7% ($2485.1 \text{ t N yr}^{-1}$) of the TN loading ($11553.3 \text{ t N yr}^{-1}$) to the lake in 2022, aligning with previous studies of Lake Dianchi (16.0%, 2010–2011) and Lake Taihu (13.9%–27.3%, 2010–2021). However, this was lower than previous estimates for Lake Chaohu, which were 39.6% in 2015 ($3118.0 \text{ t N yr}^{-1}$) (Wei et al., 2018) and 48.4% in 2019 ($7661.2 \text{ t N yr}^{-1}$) (Tian et al., 2022). However, such previous estimates were obtained by single point measurements and may be subject to large uncertainties. For example, Tian conducted sampling in a typical rice-wheat rotation area which is 10 km away from the lake and with extensive soil N emissions and thus higher depositions also.

For Nr species, atmospheric NH_x deposition input contributes 30.8% ($824.5 \text{ t N yr}^{-1}$) of total NH_x input to the lake, while for NO_y species, the atmospheric contribution is 19.4% ($629.3 \text{ t N yr}^{-1}$) in 2022. In together, atmospheric Nr deposition contributes 24.5% of total Nr input to the lake (Fig 6b). These indicate the significant role of Nr deposition in supplying bioavailable N to the lake. NH_4^+ -N and NO_3^- -N can be directly assimilated by macrophytes, algae and bacteria (Camargo and Alonso, 2006), whereas only certain higher plant algal communities can utilize low molecular weight organic N (Sutton et al., 2011). In addition, NH_4^+ -N is the favorite N species utilized by cyanobacteria (Flores and Herrero, 2005; Muro-Pastor et al., 2005) and once the environmental ammonium is depleted, nitrate could be assimilated through the nitrate

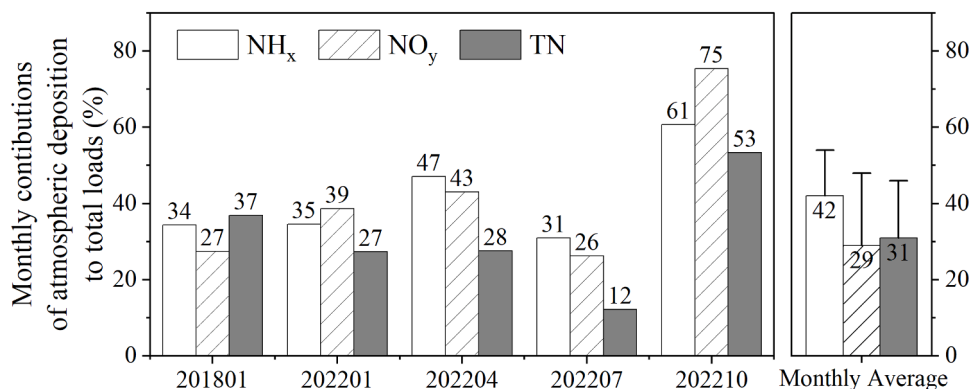


Fig 5. Monthly proportion of atmospheric nitrogen deposition to total nitrogen loads. The numbers on the bars represent the contribution of atmospheric deposition to the total loads for specific nitrogen species. The average contributions of the five months are shown on the right.

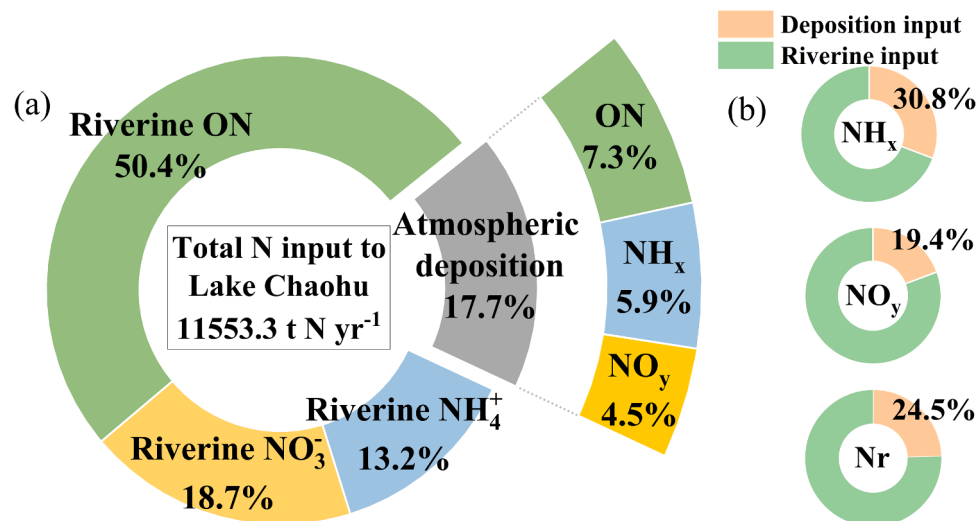


Fig 6. (a) Proportion of annual nitrogen inputs to Lake Chaohu in 2022. The numbers represent the contribution of organic nitrogen/NH_x/NO_y inputs to the total nitrogen loads. (b) Proportion of annual reactive nitrogen inputs to the lake.

assimilation pathway encoded by the *nirA* operon (Li et al., 2024). In summary, atmospheric Nr deposition represents a significant source of bioavailable N inputs, even in eutrophic lakes.

4. Conclusions and remarks

In this study, we investigated atmospheric N deposition flux to Lake Chaohu using the WRF-Chem model. The results indicate significant atmospheric contributions of N to the N burden in Lake Chaohu, especially Nr, which contributes 24.5% of total Nr input in 2022 and is directly available supplying algae growth. But we note that, at present, the contribution of atmospheric N deposition to the total N inputs remains uncertain. NO₂ and organic N were not yet included in our simulation. Furthermore, it is worth noting that the contributions discussed here were the direct portions of atmospheric deposition, considering the indirect portions of streamflow (sinks into rivers before entering the lake), the overall contribution may be even higher. In addition, the observations used to constrain the model results in this study were conducted in an urban area, which is affected by traffic emissions and differs significantly from those in the lake area, which faces agricultural emissions. Therefore, it is recommended to set monitor sites of atmospheric N deposition flux and N concentration around the lake area in the future.

For riverine inputs, although the results are subject to uncertainties since the flow rate needed to calculate the N discharge from the unmonitored rivers including the Nanfei river is estimated using the hydrological analogy method which assumes same meteorological conditions and subsurface factors for all rivers flowing into the lake, we find that ammonium input has decreased by 66% from 2018 to 2022 while TN input was stable despite new regulation controls, probably due to increasing DON concentration from urban effluents. Although the reduction of inorganic N input successfully mitigates the severe eutrophication problem, elevated DON may somewhat counteract the control effects of inorganic N since DON can be mineralized to bioavailable inorganic N. We estimated nitrate input to be comparable to ammonium, and it is recommended that nitrate should be included in water quality monitoring regulations. In addition, even after years of treatment, river Nanfei is still the main force of riverine N input to the lake, centralized management of river Nanfei may be the optimal solution. In summary, this study only provides a comprehensive estimation on the sources and amounts of N input to Lake Chaohu. To further figure out its effect on algae bloom outbreaks, in the future, the mechanisms of N utilization, transformation and DON mineralization within the water body of Lake

Chaohu need to be investigated. In addition, more hydrological parameters (e.g., flow rate) for rivers following into the lake need to be monitored to reduce the uncertainties in estimating the lake N burden.

CRediT authorship contribution statement

Weikun Li: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Xia Wang:** Software, Data curation. **Wei Song:** Resources, Investigation, Funding acquisition. **Zhongyi Zhang:** Funding acquisition. **Xueying Wang:** Resources. **Xiaodong Liu:** Resources. **Tianming Ma:** Resources, Investigation, Funding acquisition. **Qi Wang:** Resources, Investigation. **Yanli Zhang:** Resources, Investigation. **Xinming Wang:** Resources, Investigation. **Lei Geng:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in

the online version, at [doi:10.1016/j.watres.2024.122597](https://doi.org/10.1016/j.watres.2024.122597).

Data availability

Data will be made available on request.

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