REPORT

Evaluating anthropogenic N inputs to diverse lake basins: A case study of three Chinese lakes

Wei Gao, Dennis P. Swaney, Bongghi Hong, Robert W. Howarth,
Yong Liu, Huaicheng Guo

Received: 28 August 2014 / Revised: 12 January 2015 / Accepted: 3 February 2015 / Published online: 20 February 2015

Abstract The environmental degradation of lakes in China has become increasingly serious over the last 30 years and eutrophication resulting from enhanced nutrient inputs is considered a top threat. In this study, a quasi-mass balance method, net anthropogenic N inputs (NANI), was introduced to assess the human influence on N input into three typical Chinese lake basins. The resultant NANI exceeded $10\,000\text{ kg N km}^{-2}\text{ year}^{-1}$ for all three basins, and mineral fertilizers were generally the largest sources. However, rapid urbanization and shrinking agricultural production capability may significantly increase N inputs from food and feed imports. Higher percentages of NANI were observed to be exported at urban river outlets, suggesting the acceleration of NANI transfer to rivers by urbanization. Over the last decade, the N inputs have declined in the basins dominated by the fertilizer use but have increased in the basins dominated by the food and feed import. In the foreseeable future, urban areas may arise as new hotspots for nitrogen in China while fertilizer use may decline in importance in areas of high population density.

Keywords Net anthropogenic N inputs (NANI) · Eutrophication · Lake · Nitrogen · Nonpoint source

INTRODUCTION

Cultural eutrophication resulting from enhanced anthropogenic nutrient inputs is the greatest stressor affecting freshwater ecosystems (Smith and Schindler 2009; Baulch

2013), especially for lakes with short residence times. Lakes are an important component of natural ecosystems in China, and play crucial roles in regulating floods, supplying drinking water, preserving biodiversity, and providing a freshwater fishery for food (Wang and Dou 1998). The latest lake survey found that 2693 natural lakes in China with an area of over 1.0 km^2 occupy $\sim 0.9\%$ of the country's territory (Ma et al. 2011). For larger lakes over 10 km^2 in area, freshwater lakes make up 35 % of the total lake area, and have an average depth of approximately 2 m. Before the 1980s, most freshwater lakes had relatively good water quality and were eligible for drinking (Le et al. 2010). However, since China's rapid economic development in the 1980s, human-induced nutrient inputs have increased substantially (Han et al. 2013, 2014), and a majority of freshwater lakes have experienced a substantial reduction in water quality and ecosystem functionality (Gao and Zhang 2010; Liu et al. 2012). One common problem for most lakes is elevated levels of total nitrogen (TN), total phosphorus (TP), and other organic pollutant concentrations, which are mainly caused by expanding populations and agricultural fertilizer inputs (Le et al. 2010).

Of the affected lakes, Lake Taihu, Lake Chaohu, and Lake Dianchi have suffered the most and were selected by the Chinese government in the 1990s as priority control areas due to their vital influence on local economic and social development. These three lakes differ significantly in their economic, social, and environmental conditions, providing a good study case for shallow freshwater lakes in China (Fig. 1). Despite their differences, all three lakes have suffered from serious N pollution (Liu et al. 2009; Frumin and Khuan 2012). The relationship between nutrient concentrations and eutrophication of lakes has been a debate for decades, focusing on whether N or P is the

Electronic supplementary material The online version of this article (doi:10.1007/s13280-015-0638-8) contains supplementary material, which is available to authorized users.

Fig. 1 Geographic location of the Lake Taihu, Lake Chaohu, and Lake Dianchi basins. These three lake basins are distributed across three economic and geographic regions of China. The Lake Taihu basin is located in Eastern China, the most urbanized area; the Lake Chaohu basin is located in an agricultural area; the Dianchi basin is in a middle economic situation

limiting factor for eutrophication (Paerl et al. 2004; Bergström and Jansson 2006; Schindler et al. 2008; Conley et al. 2009; Scott and McCarthy 2010). Generally, nutrient limitation for eutrophication of lakes is a case-specific question, depending on various factors such as temperature (Moss et al. 2013), N deposition (Elser et al. 2009), lake age (Moss et al. 2013), and nutrient loading (Paerl et al. 2011). For the three lake basins in this study, the high

concentration of N in water is considered to be one of the main stressors for eutrophication in Lake Taihu, Lake Chaohu, and Lake Dianchi since phytoplankton is dominated by non-N-fixing cyanobacteria such as *Microcystis* (Liu 1999; Paerl et al. 2011; Li et al. 2014; Ma et al. 2014). Recent studies on the relationship between nutrient concentrations and eutrophication show that N rather than P is the limiting nutrient in eutrophication of Lake Dianchi

(Yan et al. 2013; Liang et al. 2014). Furthermore, human-induced high NO_3^- concentrations in the groundwater have been observed in these lake basins (Gu et al. 2011; Li and Wang 2012). Despite significant efforts being made to mitigate pollution in these lakes, the TN concentrations are poorly controlled and have even increased (Pan and Gao 2010). Apparently, new N inputs from human action have largely compromised our efforts to eliminate or control pollution. To better understand and mitigate N pollution, N inputs from direct anthropogenic influence must be assessed.

Currently, N sources are quantified primarily at the watershed scale via emission inventories and watershed models. The inventory method is usually based on conventional pollution sources, such as sewage treatment plants, industrial output, urban runoff, and agricultural runoff (Singh and Bakshi 2013). Watershed models, such as SWAT, HSPF, and SPARROW, are also widely used to quantify nutrient emissions from different sources at various watershed scales (Akter and Babel 2012; Robertson and Saad 2013). These methods all focus on processes that deliver nutrients from the landscape to receiving waters and on identifying key sources by their contributions to the total nutrient load. One limiting factor of the above methods is data availability, especially for watershed models, which require significant monitoring to obtain calibration and validation data. In China, the history of environmental monitoring is relatively short and exists only for certain key areas. Furthermore, most of the monitoring data are not publicly available. Therefore, accurately assessing N pollution for many areas in China is difficult using current methods. Another shortcoming of these methods is their indirect relationship to human actions, which can hinder effective policy making and enactment. For example, the nutrients discharged from sewage and agricultural runoffs are both dependent on local food production and consumption. Ignoring the relationship between these factors may overemphasize a particular pollution source and mislead development of sound policies. In light of the above problems, a mass balance method with flexible data requirements may be a good alternative for assessing nutrients in China. In this study, the net anthropogenic N inputs (NANI) approach was used to assess quantitatively the anthropogenic influence on N inputs. The NANI model is a quasi-mass balance approach that estimates human-induced N inputs on the regional scale using conventional agricultural, social, and economic statistical data. This methodology was introduced by Howarth et al. (1996) and has been applied to various watersheds over several spatial scales and climatic regions (Howarth et al. 2012; Han et al. 2014).

The main objectives of this paper are to quantify the influence human activities have on N inputs into different

lake basins in China via the NANI method and to suggest environmental policies pertinent to managing the ultimate nitrogen sources. The three lake basins in this study vary considerably in their natural, economic, and social conditions and cover most types of shallow freshwater lake basins in China. By comparing the different NANI intensities, structures, spatial distributions, and historic changes between them, we illustrate the characteristics of N pollution in shallow freshwater lake basins. Our study of these three lake basins provides a better understanding of the current N pollution status, main pressure sources, and future trends for Chinese lake basins, potentially providing useful information that can benefit policy making and environmental management.

MATERIALS AND METHODS

Study area

Lake Taihu, Lake Chaohu, and Lake Dianchi are three of the largest freshwater lakes in China by area, ranking 3rd, 5th, and 6th, respectively. They are located in the Eastern, Middle, and Western regions of China, respectively (Fig. 1), which differ greatly in their socioeconomic development. These three lake basins represent different developmental stages in China based on general economic and social indices (Table 1). The Lake Taihu basin is located in the most developed area of China and has the highest GDP per capita and urbanization level. There are eight large cities in this basin: Shanghai, Suzhou, Changzhou, Wuxi, Hangzhou, Jiaxing, Zhenjiang, and Huzhou, comprising a total of 42 counties. In contrast, the Lake Chaohu basin is relatively undeveloped, with about a half of the Lake Taihu basin's GDP per capita and population density. The entire basin is located in central Anhui province and includes 14 counties. For this basin, agricultural income accounts for approximately one-fifth of the GDP, and most lands are croplands. These characteristics make the Lake Chaohu basin a good study case for the agricultural lake basin. The Lake Dianchi basin is the smallest, occupying only seven counties. The population density of this basin is similar to the Lake Taihu basin; however, its economic development is between the other two lake basins. The variety of social and economic conditions represented by these three lake basins make them good representatives of most of China's shallow freshwater lake basins.

The 1980s are considered to be a turning point in the water quality of most freshwater lakes in China (Le et al. 2010). Before then, industrial and agricultural production were in their primary stages with a limited effect on water quality. For example, China first used mineral fertilizers in

Table 1 A summary of the Lake Taihu, Lake Chaohu, and Lake Dianchi basins in 2010

Basin	Watershed area (km ²)	Lake area (km ²)	Percent cropland	Percent urban land	Population density (capita km ⁻²)	Percent of urban population	GDP per capita (\$)
Taihu	36 895	2445	51	22	1300	75	10 000
Dianchi	2920	300	20	16	1260	64	6400
Chaohu	13 486	820	63	10	790	54	4700

1978 with only 8.8 million tons, which was approximately 8 % of the total world consumption. By 2010, this quantity had increased to 55.6 million tons, and China had become the largest mineral fertilizer consumer in the world, accounting for 31 % of global consumption (Food and Agriculture Organization of the United Nations 2014). Lake Taihu, Lake Chaohu, and Lake Dianchi each had water quality acceptable for drinking water before the 1980s; however, they have degraded rapidly over the last 30 years, especially for TN and TP (Pan and Gao 2010; Frumin and Khuan 2012). Water pollution in these lakes has caused billions of dollars in economic loss and impeded social stability (Le et al. 2010). Based on multiple-year averages, riverine TN exports from watersheds to Lake Taihu (2007–2011), Lake Chaohu (1999–2008), and Lake Dianchi (2000–2010) are 1396, 824, and 3053 kg km⁻² year⁻¹, respectively (Wang et al. 2011; The 10th five year plan for water pollution control in Lake Chaohu basin; Integrated measures for water environmental repair in Lake Taihu basin 2013; Gao et al. 2014).

Net anthropogenic N inputs (NANI) calculations

The NANI calculation is based on the mass balance principle, and assumes that new N input into a region is mainly caused by atmospheric deposition, fertilization, crop N fixation, and net food and feed imports (Howarth et al. 1996). NANI is the net balance of anthropogenic N inputs (atmospheric deposition, fertilization, crop N fixation, and food and feed imports) and outputs (food and feed exports) in a given region. A positive NANI implies that excess N in the landscape can contribute to riverine nitrogen levels through runoff and leaching, while a negative NANI suggests a local deficit of N which is not sustainable over the long term. In contrast to terminal emission calculations, N discharged from sewage and animal wastes are not considered N sources in the NANI approach because they are included within the agricultural food and feed production and consumption. This method estimates the N flux directly from food production and consumption processes (Hong et al. 2013). To estimate N fluxes at the watershed scale from the county level, there are two methods available: the area-weighting method and land use-weighting method (Han and Allan 2008). The former method estimates N inputs to watershed using the fraction of county land that is

included within the watershed boundary; the latter method estimates the fraction of the relevant land use type, such as crop or urban land, lying within each catchment. In this study, the NANI calculation for the Lake Taihu and Lake Chaohu basins was estimated via the area-weighting method, while the NANI for the Lake Dianchi basin is based on the land use-weighting method which is cited from Gao et al. (2014).

Atmospheric N deposition

Nitrogen returning to the earth from atmospheric deposition is mainly in the form of NH_y, NO_y, and organic N. In the NANI calculation, only total NO_y deposition (both dry and wet) was considered as new N inputs because most of the NH_y and organic N would redeposit to the same watershed from where they originate (Howarth et al. 2006). In this study, we use the global NO_y deposition data from Lamarque et al. (2010) to estimate the total NO_y deposition (both dry and wet) of the Lake Taihu, Lake Dianchi, and Lake Chaohu basins. The database of atmospheric NO_y deposition was a model result for the 2000s at a horizontal resolution of 0.5° in latitude and longitude. The NO_y deposition in the Lake Taihu, Lake Dianchi, and Lake Chaohu basins was calculated using the area-weighting methodology.

Fertilizer N application

Globally, mineral fertilizer use is the single largest source of anthropogenic N inputs (Galloway et al. 2004; Howarth 2004). In China, mineral fertilizer data are compiled in yearbooks of single nutrient fertilizers (nitrogen, phosphorus, and potassium) and compound fertilizers. To calculate the total N fertilization usage, both the N fertilizer and compound fertilizer data were collected from local city or county yearbooks. The data cover selected years from 2000 to 2010 in each county (See Table S1, Electronic Supplementary Material). The data were then averaged to obtain a multi-year value. Manure fertilizers were not included based on the assumption that they are cycled within the same watershed (Howarth et al. 2012). Fertilizer for non-food crops is also included here since the fertilizer data yearbook reports only total fertilizers used.

Agricultural N fixation

Approximately 90 genera of specialized microorganisms have the ability to reduce atmospheric N₂ into ammonia via biological nitrogen fixation (Dilworth and Glenn 1991). Before industrial fixation was invented in the 1900s, crop N fixation was an important source of anthropogenic nitrogen (Galloway et al. 2004). There are two methods for estimating N fixation by crops. One is the area-based method which estimates N fixation from the harvested area and the N fixation rate of each crop type; the other is yield-based method which calculates it from yield data of each crop and its N content. Considering the latter method requires more parameters (such as percent of harvested N that can be attributed to fixation, and non-harvested N as the fraction of harvested N), each of which is subjected to uncertainty, and which are mostly not available to us; we estimated crop N fixation using the area-based method. In this study, four typical N fixation crops (peanuts, soybeans, snap beans, and alfalfa hay) were included in the calculation. The harvested areas for these four crops were directly determined using the local yearbook, and the N fixation rates were set as 8000 kg km⁻² year⁻¹ for peanuts, 9600 for soybeans, 9000 for snap beans, and 22 400 for alfalfa hay (Smil 1999; Boyer et al. 2002).

Net food and feed N imports

When locally produced food and feed cannot meet demands, they are imported from outside sources to balance the need, introducing new N to the region. The main components of the net food and feed imports are the N production and consumption of food and feed for human and livestock. The N consumption terms were estimated by multiplying the human and animal populations by their unit food N consumption. Human population data at the county scale were obtained from the yearbook, and their N consumption rates were estimated from Zhai et al. (2005). A total of eight animal populations were calculated (Table S2, Electronic Supplementary Material). Nitrogen production from food and feed was estimated for eleven main crops, four fruits, eleven animal products, and three types of fish, while their N content was derived from the Chinese food nutrient content (Table S3, Electronic Supplementary Material). In this study, the N for feeding fish is not included because the data are not available. The influence of feed N for fish on NANI is further discussed in the discussion section.

Riverine N export

Riverine N exports in Lake Dianchi basin are estimated by monthly flow data and monthly TN concentrations for 31 rivers covering the year 2001–2010 from Kunming monitoring center. Riverine N exports in Lake Chaohu

basin and Lake Taihu basin are derived from published references (Wang et al. 2011; the 10th five year plan for water pollution control in Lake Chaohu basin; Integrated measures for water pollution mitigation in Lake Taihu basin 2013).

RESULTS

Human influence on N input into diverse Chinese lake basins

Based on the average values from 2000 to 2010, the human-induced N input into the Lake Taihu, Lake Dianchi, and Lake Chaohu basins reached 21 800, 11 000, and 13 700 kg km⁻² year⁻¹, respectively (Fig. 2), which is much higher than the average for China and the world (Billen et al. 2013; Han et al. 2014). The N input into the Lake Taihu basin significantly outweighs the other two basins, with almost twice their values. As the most developed area in China, with an extremely high population density and economic output, the huge NANI value for the Lake Taihu basin is not surprising. This finding provides further evidence that the NANI in China is mainly driven by population and economic output (Han et al. 2014). However, the NANI calculations for the Lake Chaohu basin, which was the least developed, also yielded a relatively high N input. The NANI intensity in the Lake Chaohu basin was 25 % more than that for the Lake Dianchi basin despite the higher population density, urbanization, and economic output in the latter. This disparity between the NANI and economic output or population density indicates that the influence of economic development on N input is very complex and uncertain. Thus, both developed and less developed areas can suffer from high N input.

Main drivers behind N input from human activities

Food and energy production is the main driver of N alteration at many spatial scales (Galloway et al. 2004). In China, N fertilizer use is the single largest source of anthropogenic N input, dominating most areas in Eastern and Middle China (Han et al. 2014). In this study, mineral N fertilizers are still the most significant direct N inputs in all three basins (Fig. 2). For the Lake Taihu and Lake Dianchi basins, which have higher levels of urbanization and population density, N food and feed imports also comprise a significant share of N inputs. However, in the agriculturally dominated Lake Chaohu basin, the food and feed N imports account for less than 2 % of the total inputs, while N fertilizer consist of over 90 % of the NANI. The differing N sources between these basins indicate that

Fig. 2 Calculation and comparison of the NANI for the Lake Taihu, Lake Dianchi, Lake Chaohu basins. The NANI in these three lake basins are several times more than the averages for China and the world. Mineral N fertilization dominates for all of the basins, especially the Lake Chaohu basin. AGRIF, DEPO, FERT, and NFFI refer to agricultural N fixation, atmospheric N deposition, fertilizer N application, and net food & feed N imports, respectively

urbanization may substantially alter the structure of N inputs from human activities by introducing more N from food imports. Compared with other NANI components, atmospheric NO_y deposition is not significant in any of the three basins, varying from 3 to 7 % of NANI. Since most NO_y is transported from outside the basins due to its long atmospheric residence times, it is more likely related to anthropogenic activities outside the region than sources within the region.

Spatial characteristics of the NANI within the basins

The high NANI inputs are usually found in urban areas with high populations or farmlands with intense fertilizer use (Gao et al. 2014). Extremely large NANI values are found in urban centers or agriculturally dominated areas in these three lake basins (Fig. 3). The highest NANI inputs for the Lake Taihu basin were observed in the eastern part where the large urban centers, such as Shanghai, Suzhou, Changzhou, and Wuxi, are located. Apart from the southwest, where forests dominate, the NANI inputs in all other areas of the Lake Taihu basin are above $10\,000\text{ kg km}^{-2}\text{ year}^{-1}$. For the Lake Dianchi basin, the NANI inputs in the north are much larger than the south. The highest NANI were found in Wuhua County, which is in the center of Kunming City. The NANI for the Lake Chaohu basin were more heterogeneously distributed. The NANI in the north, which is the location of the Hefei city center, were above $40\,000\text{ kg km}^{-2}\text{ year}^{-1}$, while they were

below $8000\text{ kg km}^{-2}\text{ year}^{-1}$ in the southwest. Overall, the NANI in these basins are heterogeneously distributed and highly dependent on land use. Areas with extremely high NANI are usually located in urban areas, while predominantly forested areas often received less NANI.

DISCUSSION

Response of riverine N export to human actions

A positive NANI implies that N inputs are greater than outputs, suggesting a surplus of N in the watersheds. The surplus N generally ends in soil or groundwater storage, losses to the atmosphere by denitrification or volatilization, and river discharge. River discharge is usually the most significant pathway transferring N from land to lakes. On average 20–25 % of NANI is exported by river discharge from its basins (Howarth et al. 1996; Howarth et al. 2012; Swaney et al. 2012; Hong et al. 2013). The percentage of NANI exported in rivers may vary a lot in different watersheds, depending on catchment characteristics which control N retention in watersheds, including discharge and precipitation (Howarth et al. 2006; 2012). The NANI inputs in these three lake basins are very high, and the riverine N exported accounts for a much higher proportion (Fig. 4a). Approximately 38 % (95 % confidence interval of 29–47 %) of anthropogenic N inputs was exported by rivers, which imposes huge pressure on these lakes. The percentage

Fig. 3 Spatial distribution of NANI in the Lake Taihu, Lake Dianchi, and Lake Chaohu basins

observed in this study (38 %) is higher than the typical values of 20–25 % observed in temperate regions. When classifying all the rivers into rural and urban river categories, according to their distance to urban centers, response of riverine N export to NANI is quite different. The percentage of N exported is much closer to the range seen elsewhere for the rural rivers (Fig. 4b), but still very high for urban rivers (Fig. 4c). The difference between the two categories suggests that the transfer of NANI to rivers can be accelerated by urbanization. The higher percentage in urban rivers than rural rivers may be partly explained by the fact that significant impervious land cover in urban areas may accelerate N transport from watersheds to rivers, as tile drainage systems do in agricultural areas (McIsaac and Hu 2004). The high values of NANI in these areas may be also responsible for the larger proportions of NANI exported in rivers.

Research suggests that when NANI exceeds some threshold value (e.g., $1070 \text{ kg N km}^{-2} \text{ year}^{-1}$, Howarth et al. 2012), N loads may exceed the limited capacity for retention and denitrification in watersheds, resulting in higher percentage of riverine-exported NANI. The difference between urban rivers and rural rivers indicates that urbanization is an important factor in transporting N and urbanized areas are usually less effective in reducing N during its transportation in the environment.

Relationship between the socioeconomic indices and N inputs

Human actions, such as industrial N fixation, food trade, energy consumption, and diet choice have been shown to be significant drivers of N alteration at various scales

Fig. 4 Riverine N export relationship with net anthropogenic N inputs from subwatersheds in the Lake Taihu, Lake Dianchi, and Lake Chaohu basins. **a** All rivers; **b** rural rivers; **c** urban rivers. Approximately 38 % NANI was exported by rivers, and a higher percentage is observed in urban rivers than rural rivers

(Galloway et al. 2004; Howarth 2004; Lassaletta et al. 2014). However, when considering socioeconomic factors, which are mostly represented by population density, economic income, and industrial structure, the relationship to N inputs is still largely unknown. According to Han et al. (2014), total population density is the largest socioeconomic driver for changing NANI, while the gross domestic product also plays a significant role. Nevertheless, in this study, despite the Lake Chaohu basin having smaller population density, urbanization intensity, and economic income than the Lake Dianchi basin, its NANI inputs were 25 % higher than that for the Lake Dianchi basin, implying that the influence of socioeconomic factors on the NANI is very complex. Based on the relationship between NANI and the main socioeconomic factors in counties within these basins, the population density explains less than 50 % of the change in the NANI for the different counties, and the gross domestic product is less correlated (Fig. 5). Data from 354 cities in mainland China also indicate that economic production does not have a stable relationship with NANI, and that the economic structure has a complicated influence on the slope (Fig. S1, Electronic Supplementary Material). Therefore, socioeconomic indices may not be good predictors for NANI at the county or city scales. Since N input is better related to food production and consumption, NANI should be driven mainly by agriculture and relevant population factors.

Historical change in the N inputs within the lake basins

Over the last three decades, NANI in China increased by 112, and 77 % of this increase was from fertilizer (Han et al. 2014). However, this elevated NANI decreased by 40 % over the last decade compared to the 1980s, and

negative rates of change were found in places such as Beijing, Shanghai, and Shandong province (Fig. S2, Electronic Supplementary Material). Over the last decade, NANI in the Lake Taihu basin has declined at an average rate of $-423 \text{ kg km}^{-2} \text{ year}^{-1}$, and 75 % of the counties showed a declining trend (Fig. 6a). The main driver for this decline is the reduction in N fertilizer use. However, NANI increased in some southern counties, mostly driven by enhanced food and feed imports to support people migrating from rural areas. The Lake Dianchi basin generally featured an increasing NANI with 5 out of 7 counties exhibiting a growing NANI (Fig. 6b). Overall, NANI in the basin increased at $311 \text{ kg km}^{-2} \text{ year}^{-1}$, which was mainly due to mineral fertilizers (71 %). For the Lake Chaohu basin (Fig. 6c), the number of counties with increasing and decreasing trends was 5 and 7, respectively, and the NANI of the whole basin decreased at a rate of $-262 \text{ kg km}^{-2} \text{ year}^{-1}$, of which 93 % was due to fertilizer reduction. Similar to the Lake Taihu basin, fertilizer usage was the main driver behind the NANI reduction, and both fertilizer and food and feed imports were significant drivers in areas with increasing NANI. Further comparisons of the NANI components for 55 counties in these three basins are presented in Fig. S3, Electronic Supplementary Material. As more farmland is converted into urban land and fertilizer usage shrinks (Table S4, Electronic Supplementary Material), the increase in fertilizer use may cease in traditional agricultural areas of China, while N inputs for food and feed imports increase in the urban areas.

New NANI components and their impact

Since the first introduction of the NANI concept in 1996 (Howarth et al. 1996), alternative methods for calculating NANI have been proposed by numerous researchers (Hong

Fig. 5 Relationship between the NANI and the main socioeconomic factors, population density (a) and GDP (b), for counties within the Lake Taihu, Lake Dianchi, and Lake Chaohu basins

Fig. 6 Annual NANI rate of change in the Lake Taihu, Lake Dianchi, and Lake Chaohu basins from 2000 to 2010. The NO_y deposition rate is assumed to be stable over the period when calculating the annual NANI changes

Table 2 NANI with conventional components vs. NANI with additional components. Introduction of aquaculture and industry to calculation will change 3–9 and 1–6 % of NANI in the basins, respectively

Basin Name	Baseline	Adjustment by aquaculture		Adjustment by industry	
		NANI	Difference (%)	NANI	Difference (%)
Dianchi	10 974	10 657	−2.9	11 061	0.8
Taihu	21 819	23 748	8.8	23 023	5.5

et al. 2013). Two of the alternative methods proposed by Schaefer and Alber (2007) and Han et al. (2014) included N in non-food crops (such as cotton, tobacco, and crop seeds) in NANI calculations. However, both adjustments represented less than 1 % of the total N input. In this study, both Lake Dianchi and Lake Taihu basins are the largest horticulture (mainly flowers) and aquaculture regions of China, respectively. The role of these two new factors in NANI calculation should be assessed. We assumed that virtually all flowers produced in the Lake Dianchi basin are exported. The amount of N exported by flowers is estimated by multiplying harvested area of flowers by N content of harvested parts (Li et al. 2005). The amount of N used for feeding fish is estimated by multiplying aquaculture production by the virtual N factor for fish which is calculated by dividing the total N_r released to the environment from fish production process by the total N_r consumed by human beings from fishery production (Leach et al. 2012). Table 2 shows a comparison of NANI results between conventional components and new components. For the Lake Dianchi basin, inclusion of flowers exported in NANI has little influence on total N inputs, making up only 3 % of the total anthropogenic N inputs. For the Lake Taihu basin, introduction of feed for fish production increased the NANI by 9 %, implying that aquaculture is an important component for NANI calculation in this area. Although estimation of the role of horticulture and aquaculture in NANI is crude

because of poor data availability, it indicates that it may be important to include aquatic production in the calculation of NANI, which has been largely overlooked in past research.

Industrial point sources are also significant nutrient contributors in many regions of China. According to the first national pollution census bulletin, ammonia emission from industrial wastewater reached 3.04×10^8 kg in 2007 and 86 % is from non-food industries, such as chemical, nonferrous metal smelting, petroleum processing, and textile industry. However, Chinese industry varies a lot in spatial distribution and most industrial sources are located in Eastern China. Therefore, industrial contributions to N budgets are quite different from place to place. Based on multiple averages during the period 2000–2010, industrial N emissions in the Lake Taihu, Lake Dianchi, and Lake Chaohu basins are 1204, 87, and 258 $\text{kg km}^{-2} \text{ year}^{-1}$, respectively, only accounting for 1–6 % of total NANI. Overall, N from aquaculture and industry is not a large source of regional N inputs at relatively large spatial scales. However, they may contribute significantly at finer scales when they are not evenly distributed among different areas.

CONCLUSIONS

Excess N from human activities is responsible for degrading water quality in many freshwater lakes in China. Quantitative

estimates for the influence of human actions on N inputs into terrestrial ecosystem are needed to better understand human–nitrogen interactions. In this study, a quasi-mass balance method, net anthropogenic N inputs, was applied to three diverse lake basins in China to assess the anthropogenic influence on the N inputs. Extremely high N input intensities were found in all three basins, and mineral fertilizers were the main sources, suggesting that reducing N from nonpoint sources is an important task in these basins. A comparison of NANI across these three lake basins indicated that both developed and less developed areas could suffer from high NANI and that urbanization increases food and feed N imports, and thus, NANI. Because China is currently undergoing rapid urbanization, more rural lands will be converted into urban lands in the near future, stimulating food and feed imports in urban areas and accelerating N transport from watersheds to rivers. The spatial NANI distribution demonstrated the close relationship between N input, riverine N fluxes, and urban development.

Although connecting socioeconomic factors to N pollution is common for drawing policy-relevant conclusions, the results of this study indicate that socioeconomic indices only weakly explain the changes in NANI at relatively small scales. Regional N input abatement should focus on food production and consumption patterns. Over the last 10 years, the increasing NANI in most Chinese provinces has slowed or even reversed. In contrast to the Lake Dianchi basin, both the Lake Taihu and Lake Chaohu basins underwent significant NANI reductions, which imply that a turning point in the N input has occurred in these areas. Overall, these N input reductions are due to reduction in fertilizer use; however, food and feed imports are becoming significant drivers for increasing the NANI, implying that N pollution pressures are transported to food consumption regions. In the future, the N inputs from food and feed imports may continue to grow with rapid urbanization and increasing animal protein consumption. Thus, urban sewage and animal waste control will become increasingly important N sources, and recycling N from them to agricultural land will contribute to eliminating pressure from both synthetic fertilizer use and net food and feed imports. In agricultural area, some new factors such as aquaculture can significantly affect NANI calculations, implying that inclusion of regionally specific components of NANI may contribute to more accurate results.

Acknowledgments This study was funded by a grant from the China Scholarship Council, the Major Science and Technology Program for Water Pollution Control and Treatment of China (2013ZX07102), and National Natural Science Foundation of China (No. 41222002).

REFERENCES

- Akter, A., and M.S. Babel. 2012. Hydrological modeling of the Mun River basin in Thailand. *Journal of Hydrology* 452: 232–246.
- Baulch, H.M. 2013. Asking the right questions about nutrient control in aquatic ecosystems. *Environmental Science and Technology* 47: 1188–1189.
- Bergström, A.-K., and M. Jansson. 2006. Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. *Global Change Biology* 12: 635–643.
- Billen, G., J. Garnier, and L. Lassaletta. 2013. The nitrogen cascade from agricultural soils to the sea: modelling nitrogen transfers at regional watershed and global scales. *Philosophical Transactions of the Royal Society B-Biological Sciences* 368: 20130123.
- Boyer, E.W., C.L. Goodale, N.A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. *Biogeochemistry* 57: 137–169.
- Conley, D.J., H.W. Paerl, R.W. Howarth, D.F. Boesch, S.P. Seitzinger, K.E. Havens, C. Lancelot, and G.E. Likens. 2009. Ecology controlling eutrophication: Nitrogen and phosphorus. *Science* 323: 1014–1015.
- Dilworth, M.J., and A.R. Glenn. 1991. *Biology and biochemistry of nitrogen fixation*. Amsterdam: Elsevier Science Publishers.
- Elser, J.J., T. Andersen, J.S. Baron, A.K. Bergstrom, M. Jansson, M. Kyle, K.R. Nydick, L. Steger, et al. 2009. Shifts in lake N: P stoichiometry and nutrient limitation driven by atmospheric nitrogen deposition. *Science* 326: 835–837.
- Food and Agriculture Organization of the United Nations. 2014. FAO Statistical Yearbook 2011. Retrieved April 7, 2014, from <http://faostat.fao.org/site/291/default.aspx>.
- Frumin, G.T., and Z.Z. Khuan. 2012. Dynamics of nutrient concentrations in Lake Taihu. *Russian Journal of General Chemistry* 82: 2210–2215.
- Galloway, J.N., F.J. Dentener, D.G. Capone, E.W. Boyer, R.W. Howarth, S.P. Seitzinger, G.P. Asner, C.C. Cleveland, et al. 2004. Nitrogen cycles: Past, present, and future. *Biogeochemistry* 70: 153–226.
- Gao, C., and T.L. Zhang. 2010. Eutrophication in a Chinese context: Understanding various physical and socio-economic aspects. *AMBIO* 39: 385–393.
- Gao, W., R.W. Howarth, B. Hong, D.P. Swaney, and H.C. Guo. 2014. Estimating net anthropogenic nitrogen inputs (NANI) in the Lake Dianchi Basin of China. *Biogeosciences* 11: 4577–4586.
- Gu, Y., D.B. Sun, and Q.S. Wang. 2011. Studies on groundwater nitrate nitrogen distribution and its affecting factors in Chao Lake watershed. *Journal of Agricultural Science and Technology* 13: 68–74. (in Chinese).
- Han, H.J., and J.D. Allan. 2008. Estimation of nitrogen inputs to catchments: Comparison of methods and consequences for riverine export prediction. *Biogeochemistry* 91: 177–199.
- Han, Y., Y. Fan, P. Yang, X. Wang, Y. Wang, J. Tian, L. Xu, and C. Wang. 2014. Net anthropogenic nitrogen inputs (NANI) index application in Mainland China. *Geoderma* 213: 87–94.
- Han, Y.G., X.X. Yu, X.X. Wang, Y.Q. Wang, J.X. Tian, L. Xu, and C.Z. Wang. 2013. Net anthropogenic phosphorus inputs (NAPI) index application in Mainland China. *Chemosphere* 90: 329–337.
- Hong, B.G., D.P. Swaney, and R.W. Howarth. 2013. Estimating net anthropogenic nitrogen inputs to US watersheds: Comparison of methodologies. *Environmental Science and Technology* 47: 5199–5207.
- Howarth, R., D. Swaney, G. Billen, J. Garnier, B.G. Hong, C. Humborg, P. Johnes, C.M. Morth, et al. 2012. Nitrogen fluxes from the landscape are controlled by net anthropogenic nitrogen inputs and by climate. *Frontiers in Ecology and the Environment* 10: 37–43.
- Howarth, R.W. 2004. Human acceleration of the nitrogen cycle: Drivers, consequences, and steps toward solutions. *Water Science and Technology* 49: 7–13.

- Howarth, R.W., G. Billen, D. Swaney, A. Townsend, N. Jaworski, K. Lajtha, J.A. Downing, R. Elmgren, et al. 1996. Regional nitrogen budgets and riverine N&P fluxes for the drainages to the North Atlantic Ocean: Natural and human influences. *Biogeochemistry* 35: 75–139.
- Howarth, R.W., D.P. Swaney, E.W. Boyer, R. Marino, N. Jaworski, and C. Goodale. 2006. The influence of climate on average nitrogen export from large watersheds in the Northeastern United States. *Biogeochemistry* 79: 163–186.
- Lamarque, J.F., T.C. Bond, V. Eyring, C. Granier, A. Heil, Z. Klimont, D. Lee, C. Liousse, et al. 2010. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application. *Atmospheric Chemistry and Physics* 10: 7017–7039.
- Lassaletta, L., G. Billen, E. Romero, J. Garnier, and E. Aguilera. 2014. How changes in diet and trade patterns have shaped the N cycle at the national scale: Spain (1961–2009). *Regional Environmental Change* 14: 785–797.
- Le, C., Y. Zha, Y. Li, D. Sun, H. Lu, and B. Yin. 2010. Eutrophication of lake waters in China: Cost, causes, and control. *Environmental Management* 45: 662–668.
- Leach, A.M., J.N. Galloway, A. Bleeker, J.W. Erisman, R. Kohn, and J. Kitzes. 2012. A nitrogen footprint model to help consumers understand their role in nitrogen losses to the environment. *Environmental Development* 1: 40–66.
- Li, C.P., Y.B. Xu, Y.M. Li, Y. Zheng, W.L. Zhang, and H.B. Liu. 2005. The nutrient balance in the protected fields of vegetable and flower cultivation in Dian Lake front. *Journal of Yunnan Agricultural University* 20: 804–809. (in Chinese).
- Li, Q.G., and S.L. Wang. 2012. Nitrogen and oxygen isotopic tracing of nitrate pollution in the Dianchi Lake basin. *Earth and environment* 40: 321–327 (in Chinese).
- Li, Y.P., C.Y. Tang, Z.B. Yu, and K. Acharya. 2014. Correlations between algae and water quality: Factors driving eutrophication in Lake Taihu, China. *International Journal of Environmental Science and Technology* 11: 169–182.
- Liang, Z.Y., Y. Liu, H. Sheng, Y.H. Yang, H.C. Guo, L. Zhao, and B. He. 2014. Identification of water quality trends in Lake Dianchi using an integrated STL and RSI approach. *Acta Scientiae Circumstantiae* 34: 754–762 (in Chinese).
- Liu, L.P. 1999. Characteristics of blue algal bloom in Dianchi Lake and analysis on its cause. *Research of Environmental Sciences* 12: 36–37 (in Chinese).
- Liu, Z.H., X.H. Liu, B. He, J.F. Nie, J.Y. Peng, and L. Zhao. 2009. Spatio-temporal change of water chemical elements in Lake Dianchi, China. *Water and Environment Journal* 23: 235–244.
- Liu, W.Z., S.Y. Li, H.M. Bu, Q.F. Zhang, and G.H. Liu. 2012. Eutrophication in the Yunnan Plateau lakes: The influence of lake morphology, watershed land use, and socioeconomic factors. *Environmental Science and Pollution Research* 19: 858–870.
- Ma, R.H., G.S. Yang, H.T. Duan, J.H. Jiang, S.M. Wang, X.Z. Feng, A.N. Li, F.X. Kong, et al. 2011. China's lakes at present: Number, area and spatial distribution. *Science China-Earth Sciences* 54: 283–289 (in Chinese).
- Ma, J.R., J.D. Brookes, B.Q. Qin, H.W. Paerl, G. Gao, P. Wu, W. Zhang, J.M. Deng, et al. 2014. Environmental factors controlling colony formation in blooms of the cyanobacteria *Microcystis* spp. in Lake Taihu, China. *Harmful Algae* 31: 136–142.
- McIsaac, G.F., and X.T. Hu. 2004. Net N input and riverine N export from Illinois agricultural watersheds with and without extensive tile drainage. *Biogeochemistry* 70: 251–271.
- Moss, B., E. Jeppesen, M. Sondergaard, T.L. Lauridsen, and Z.W. Liu. 2013. Nitrogen, macrophytes, shallow lakes and nutrient limitation: Resolution of a current controversy? *Hydrobiologia* 710: 3–21.
- Paerl, H.W., L.M. Valdes, A.R. Joyner, and M.F. Piehler. 2004. Solving problems resulting from solutions: Evolution of a dual nutrient management strategy for the eutrophying Neuse river estuary, North Carolina. *Environmental Science and Technology* 38: 3068–3073.
- Paerl, H.W., H. Xu, M.J. McCarthy, G. Zhu, B. Qin, Y. Li, and W.S. Gardner. 2011. Controlling harmful cyanobacterial blooms in a hyper-eutrophic lake (Lake Taihu, China): The need for a dual nutrient (N & P) management strategy. *Water Research* 45: 1973–1983.
- Pan, M., and L. Gao. 2010. The influence of socio-economic development on water quality in the Dianchi Lake. *Engineering Sciences* 12: 117–122 (in Chinese).
- Robertson, D.M., and D.A. Saad. 2013. SPARROW models used to understand nutrient sources in the Mississippi/Atchafalaya River basin. *Journal of Environmental Quality* 42: 1422–1440.
- Schaefer, S.C., and M. Alber. 2007. Temperature controls a latitudinal gradient in the proportion of watershed nitrogen exported to coastal ecosystems. *Biogeochemistry* 85: 333–346.
- Schindler, D.W., R.E. Hecky, D.L. Findlay, M.P. Stainton, B.R. Parker, M.J. Paterson, K.G. Beaty, M. Lyng, et al. 2008. Eutrophication of lakes cannot be controlled by reducing nitrogen input: Results of a 37-year whole-ecosystem experiment. *Proceedings of the National Academy of Sciences of the United States of America* 105: 11254–11258.
- Scott, J.T., and M.J. McCarthy. 2010. Nitrogen fixation may not balance the nitrogen pool of lakes over timescales relevant to eutrophication management. *Limnology Oceanography* 55: 1265–1270.
- Singh, S., and B.R. Bakshi. 2013. Accounting for the biogeochemical cycle of nitrogen in input-output life cycle assessment. *Environmental Science and Technology* 47: 9388–9396.
- Smil, V. 1999. Nitrogen in crop production: An account of global flows. *Global Biogeochemical Cycles* 13: 647–662.
- Smith, V.H., and D.W. Schindler. 2009. Eutrophication science: Where do we go from here? *Trends in Ecology & Evolution* 24: 201–207.
- Swaney, D.P., B.G. Hong, C.P. Ti, R.W. Howarth, and C. Humborg. 2012. Net anthropogenic nitrogen inputs to watersheds and riverine N export to coastal waters: A brief overview. *Current Opinion in Environmental Sustainability* 4: 203–211.
- Wang, S.M., and H.S. Dou. 1998. *Lake records of China*. Beijing: Science Press (in Chinese).
- Wang, S.H., X. Jiang, and X.C. Jin. 2011. Classification and pollution characteristic analysis for inflow rivers of Chaohu lake. *Environmental Science* 32: 2834–2839 (in Chinese).
- Yan, X.P., Y.Z. Li, Y. Liu, Y.H. Yang, L. Zhao, and H.C. Guo. 2013. Identifying the influence of water chemistry on Chlorophyll a in Lake Dianchi: A structural equation modeling analysis. *Acta Scientiarum Naturalium Universitatis Pekinensis* 49: 1031–1039 (in Chinese).
- Zhai, F.Y., Y.N. He, Z.H. Wang, W.T. Yu, H.S. Hu, and X.G. Yang. 2005. The status and trends of dietary nutrients intake of Chinese population. *Acta Nutrimenta Sinica* 27: 181–184 (in Chinese).

AUTHOR BIOGRAPHIES

Wei Gao is a Doctoral Candidate at the College of Environmental Sciences and Engineering of Peking University. His research interests include the interaction between human activities and water environment, watershed integrated management and nutrient cycles. Address: College of Environmental Sciences and Engineering, The Key Laboratory of Water and Sediment Sciences, Ministry of Education, Peking University, Beijing 100871, China. e-mail: jacob@pku.edu.cn

Dennis P. Swaney is a Coordinator of Modeling at the Department of Ecology and Evolutionary Biology of Cornell University. His recent work focuses on nutrient accounting in large watersheds and the

relationship of NANI (Net Anthropogenic Nitrogen Inputs) to riverine nitrogen fluxes. Other research has included developing methods of estimating ecosystem metabolism in estuaries, modeling nutrient transport through watersheds to the coast, and modeling ecological responses of watersheds and coastal ecosystems to climate and land use changes.

Address: Department of Ecology and Evolutionary Biology, Cornell University, Ithaca, NY 14850, USA.
e-mail: dps1@cornell.edu

Bongghi Hong is a Research Associate at the Department of Ecology and Evolutionary Biology of Cornell University. His research interests include integrated assessment linking socioeconomic activities, land use change, and stream ecosystem health.

Address: Department of Ecology and Evolutionary Biology, Cornell University, Ithaca, NY 14850, USA.
e-mail: bh43@cornell.edu

Robert W. Howarth is a Professor at the Department of Ecology and Evolutionary Biology of Cornell University. He has worked extensively on environmental issues related to human-induced changes in the sulfur, nitrogen, phosphorus, and carbon cycles, the impacts of

global climate change, and interaction of energy systems and the environment.

Address: Department of Ecology and Evolutionary Biology, Cornell University, Ithaca, NY 14850, USA.
e-mail: howarth@cornell.edu

Yong Liu is a Professor at the College of Environmental Sciences and Engineering of Peking University. His research interests focus on ecological and water quality modeling, lake-watershed management, and environmental policy.

Address: College of Environmental Sciences and Engineering, The Key Laboratory of Water and Sediment Sciences, Ministry of Education, Peking University, Beijing 100871, China.
e-mail: yongliu@pku.edu.cn

Huaicheng Guo (✉) is a Professor at the College of Environmental Sciences and Engineering of Peking University. His research interests focus on basin integrated management and planning.

Address: College of Environmental Sciences and Engineering, The Key Laboratory of Water and Sediment Sciences, Ministry of Education, Peking University, Beijing 100871, China.
e-mail: guohc@pku.edu.cn