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LETTER

Land use mediates riverine nitrogen export under the dominant influence of human activities

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Abstract

Riverine nitrogen (N) export is a crucial process that links upstream and downstream ecosystems and coastal zones. However, the driving forces of riverine N export that is closely related to water N pollution are still not well understood. In this study, we used a mass balance approach to quantify the sources of N discharge and analyzed the effect of land use composition on riverine N export, taking Zhejiang Province, China as a case study. We found that the total reactive N discharge to rivers in Zhejiang increased from 0.22 to 0.26 Tg yr−1 from 2000 to 2015. At the watershed scale, our estimate of N export agrees well with the monitored riverine N concentration in the eight major watersheds in Zhejiang. Direct discharge of domestic wastewater and effluents from wastewater treatment plants are dominant sources of riverine N export, followed by agricultural non-point sources. Although riverine N export increases with the increasing proportion of urban and agricultural land uses, we did not find any relationship between land use change and changes in riverine N export. This suggests that the dominant factor affecting riverine N export should be human activities (e.g. wastewater discharge and fertilization level), while land use only mediates riverine N export.

1. Introduction

Rivers play a pivotal role in the function of ecosystems and human lives. They are an important part of aquatic ecosystems, and of great importance to the socioeconomic sustainability of a region. Unfortunately, most rivers have been regarded as a means to dispose wastewater, and the concentration of nutrients such as nitrogen (N) in many rivers exceeds their self-purification capacity, resulting in adverse impacts on the water quality (Rashid and Romshoo 2013). To meet the societal demand for food and energy, human activities have more than tripled the reactive N (Nr) input to terrestrial ecosystems as compared to the pre-industrial era (Fowler et al 2013). A large proportion of this elevated Nr input is lost to the rivers, leading to riverine N pollution (Gu et al 2015). Meanwhile, other factors such as climate variability, land use and land cover change (LULC) are also being regarded as important drivers of riverine N export to ecosystems downstream (Kibena et al 2014, Ma et al 2015, Tian et al 2015). Therefore, many studies focused on the effect of Nr input on riverine N export such as the effort in developing the Net Anthropogenic N Input (NANI) model (Howarth et al 2012), the Dynamic Land Ecosystem Model (DLEM) (Yang et al 2015), and research on the effect of LULC on riverine N export (Wang et al 2014, Yin et al 2012). Nevertheless, how Nr input intensity and LULC interact to affect riverine N export is still unclear. Furthermore, NANI studies normally do not consider processes linking the N input into an ecosystem of a region and riverine N export, making it difficult to identify the sources
of Nr other than N input from N fertilizer, biological N fixation, atmospheric N deposition, and food/feed import.

Source apportionment provides information on major contributors of Nr and is crucial for the mitigation of riverine N export (Giri et al. 2016, Guo et al. 2004). Previous studies generally categorized sources of Nr into point and non-point sources (Carpenter et al. 1998, Zhang et al. 2015). Compared to point source pollution, nonpoint source pollution derived from agricultural production currently attracts more attention because it is affected by both anthropogenic and natural factors that cannot be easily measured or regulated (Chen et al. 2016, Zhang and Huang 2011). In fact, the point source is still serious, and about 450 km² of domestic wastewater flows into rivers worldwide annually, with nearly half of it untreated (Angyal et al. 2016). This suggests that both agricultural activities and urbanization are dominant driving forces of riverine N export (Firmansyah et al. 2017, Tong et al. 2017). In addition to agricultural and urban land uses, other land use types such as forestlands and wetlands also affect riverine N export (Su et al. 2013, Wang et al. 2014). However, the overall effect of these sources of Nr and land use types on riverine N export on the watershed or regional scale is poorly studied, as previous research addressed one or some of these sources (Guo et al. 2013, Liu et al. 2012), or land use types (Liu et al. 2013, Smith et al. 2013). Furthermore, source apportionment is usually conducted based on socioeconomic boundaries, while riverine N export is best studied based on the natural boundaries of watersheds. Integration of boundaries is always difficult for the analysis of riverine pollution that links socioeconomic and ecological processes.

In this study, we attempt to understand how land use composition and human activity intensity (e.g. N fertilization rate) contribute to riverine N export through integrating land use effects and source apportionment. A Coupled Human And Natural System (CHANS) N cycling model (Gu et al. 2015) was used to constrain N cycling on watershed and regional scales, and to improve the accuracy of source apportionment of riverine N export. In China, LULC has been changed drastically in recent decades with rapid urbanization, resulting in serious surface water pollution problems (Zhang et al. 2015). Based on land use and socioeconomic data, together with large scale monitoring of water quality, here we (1) quantified the sources of N discharge in eight watersheds and in the entire province of Zhejiang, China, where the LULC has also been changed drastically in recent decades; (2) explored the effect of the width of buffer zones on the relationship between land use composition and river water quality; and (3) identified the dominant contributor of riverine N export under land use change and human activities.

2. Materials and methods

2.1. Study area
In this study, we used Zhejiang province and eight watersheds (Tiaoxi, Grand Canal, Qiantang River, Yongjiang, Jiaojiang, Oujiang, Feiyun River and Aojiang) located in the province as case studies (figure 1). Zhejiang, one of the most developed provinces in China, lies in southeast China (27.2–31.5° N, 118–123° E) with an area of 105 500 km² and a typical subtropical monsoon climate. The annual average air temperature is 17.2 °C and the annual precipitation is 1490 mm. The total area of the watersheds ranges from 1545 (Aojiang) to 55 058 km² (Qiantang River) and encompasses a wide variety of land use types, population densities, and human activity intensities. The composition of land use among the watersheds in 2010 was 12%–44% cropland, 3%–25% urban land, 22%–80% forestland, 3%–9% wetland, and 1%–3% grassland (figure S2).

2.2. Watershed boundary
The Digital Elevation Model (DEM) of Zhejiang for 2010 with a resolution of 30 m was obtained from the geographic data cloud (www.gcloud.cn). Boundaries of the eight watersheds were identified using the hydrology tool in ArcGIS (version 10.3, Esri Global, Inc.) based on the DEM. The eight watersheds are located across more than one county, which is the unit for source apportionment in the CHANS model. We described how the source apportionment for each watershed was conducted in the following sections.

2.3. Land use and water quality monitoring
Land use maps in Zhejiang in the years of 2000, 2005 and 2010 with a resolution of 30 m were interpreted from the Landsat TM /ETM remote-sensing data (Wu et al. 2014). The land use changes in the eight watersheds from 2000 to 2010 were analyzed using the ArcGIS software. A high resolution land use map of 2010 was used to represent the period of 2010–2015 for the spatial analysis given that the Landsat data used to estimate the land use in 2010 normally covers the period 2010–2015.

To explore the relationships between land use composition and water quality, we collected water quality data from 106 monitoring sites (figure 1). These monitoring sites were operated by the Environmental Protection Bureau of Zhejiang province (http:// wms.zjemc.org.cn/wms/). Water quality parameters were all monitored using standard online techniques following the national environmental quality standards for surface water (GB3838-2002). Monthly water quality data for 2015, including pH, ammonium (NH₄⁺), total phosphorus (TP), dissolved oxygen (DO) and chemical oxygen demand (COD), of the 106 monitoring sites were obtained.
2.4. Analysis of the effect of buffer zone width

To understand how land use composition affects the water quality and how these relationships vary with the width of buffer zone that was set up for each monitoring site, Pearson’s correlation analysis was conducted separately for the following three buffer zone widths: 200, 500 and 1000 m riparian buffers surrounding the 106 monitoring sites. The land use composition within each of these buffer widths was calculated for analysis. We took a circular area surrounding the measurement point and determined the land use composition within different buffer zone widths to the specific monitoring point (Guo et al 2009, Xiao et al 2016). Annual mean water quality indicators were calculated for the analysis of the relationship between land use composition and water quality. All statistical analyses were performed using the SPSS statistical software (version 22.0, IBM Corp.).

2.5. Source apportionment from the CHANS model

We quantified the sources of riverine N export in the eight watersheds based on a mass balance method using the CHANS N cycling model (Gu et al 2015). The CHANS’ approach can estimate N fluxes from 14 different subsystems (cropland, forestland, grassland, industry, livestock, aquaculture, urban greenland, human, pets, wastewater treatment plants, solid waste treatment, surface water, groundwater, and atmosphere). We calculated the overall N balance of the 14 subsystems first, then the N fluxes to the surface water subsystem from the other 13 subsystems were extracted from the overall balance. These N inputs to surface water include sources such as N runoff from cropland due to the application of N fertilizer and manure. All these N fluxes were calculated on the county scale and allocated to different watersheds. The principle of the CHANS model is the mass balance of the whole system and each subsystem:

\[ \sum_{n=1}^{m} \text{IN}_n = \sum_{g=1}^{n} \text{OUT}_g + \sum_{k=1}^{p} \text{ACC}_k \]

where IN and OUT represent the N input and output, respectively, and ACC represents N accumulation. If there is N flow from one subsystem to another, the flux in the two related subsystems should be equal. This was used to constrain the estimation of N fluxes.

Data used in the source apportionment can be divided into two categories: (i) socioeconomic information for Zhejiang Province obtained from statistical information such as population, GDP, area under each land use type, fertilizer use, crop/livestock production, and energy consumption in different sectors; (ii) coefficients and parameters used for the calculation of N runoff to surface water, both obtained from the synthesis of peer-reviewed literature and field measurements. More details on budget calculation were described in the SI. The important coefficients and parameters can be found in Gu et al (2015).

County is the smallest administrative unit for data collection to conduct the source apportionment. The socioeconomic data of counties in Zhejiang province were collected from the statistical yearbooks at the provincial, city and county levels for the period of 2000–2015. If a county crosses multiple watersheds, its socioeconomic data were allocated to different...
watersheds using proxy variables. For example, Yuhang county is located across in Tiaoxi, Canal and Qiantang River watersheds. Fertilizer N input to cropland in Yuhang county was allocated to the three watersheds based on the proportion of cropland in these watersheds as a proxy variable.

3. Results

3.1. Effect of riparian buffer zone width on river water quality

The monitored water quality varied across the studied watersheds in Zhejiang (figure 2). High concentrations of NH$_4^+$, TP and COD were found in northeastern Zhejiang such as in the Canal watershed, where the percent of residential and agricultural land was large and the water quality was the worst among the eight watersheds. In contrast, the high DO concentrations were mainly found in the watersheds in southwestern Zhejiang, which has more forestland and wetland with a better water quality.

The Pearson’s correlation analysis indicated that buffer width affected the relationships between land use composition and water quality (table 1). The contribution of land use composition to water quality increases with the buffer width (increasing from 200 to 1000 m). Urban land was strongly and positively correlated with NH$_4^+$ concentration in the river regardless of the width of the buffer zone, with urban land having a stronger correlation with NH$_4^+$ concentration than cropland. The strength of the correlation sharply decreased with the decreasing width of the buffer zone. Low intensity land use systems including forestland, grassland and wetland was significantly and negatively correlated with all pollutants, including NH$_4^+$, TP and COD.

3.2. Source apportionment from the CHANS model

The sources of N discharge in Zhejiang or in its eight watersheds from 2000 to 2015 are presented in figure 3. More details of these emission sources are presented in table S1 available at stacks.iop.org/ERL/12/094018/mmedia. The total N discharge increased from 0.22 Tg N yr$^{-1}$ in 2000 to 0.26 Tg N yr$^{-1}$ in 2015 in Zhejiang with an annual growth rate of 1.2% (figure S3); a similar growth trend was found in the eight watersheds. Domestic wastewater including
Table 1. Pearson correlation coefficients for the relationship between land use and water quality with buffer zone width of 200–1000 m.

| Buffer width | Pollutant | Urban land | Cropland | Forestland | Wetland | Grassland |
|--------------|-----------|------------|----------|------------|---------|-----------|
|              | NH$_4^+$-N | 0.39$^b$   | 0.03     | −0.33$^b$  | −0.04   | −0.05     |
| 200 m        | TP        | 0.37$^b$   | 0.29$^b$ | −0.31$^b$  | −0.11   | 0.03      |
|              | COD       | 0.40$^b$   | 0.33$^b$ | −0.36$^b$  | −0.15   | 0.11      |
|              | DO        | −0.39$^b$  | −0.20$^b$| 0.52$^b$   | 0.02    | 0.02      |
|              | NH$_4^+$-N | 0.48$^b$   | 0.11     | −0.35$^b$  | −0.18   | −0.10     |
|              | TP        | 0.38$^b$   | 0.40$^b$ | −0.51$^b$  | −0.21$^a$| −0.09     |
| 500 m        | COD       | 0.44$^b$   | 0.42$^b$ | −0.55$^b$  | −0.24$^a$| −0.11     |
|              | DO        | −0.42$^b$  | −0.32$^b$| 0.51$^b$   | 0.14    | 0.16      |
|              | NH$_4^+$-N | 0.50$^b$   | 0.18     | −0.37$^b$  | −0.22$^a$| −0.03     |
|              | TP        | 0.37$^b$   | 0.52$^b$ | −0.49$^b$  | −0.30$^b$| −0.06     |
| 1000 m       | COD       | 0.40$^b$   | 0.53$^b$ | −0.56$^b$  | −0.23$^a$| −0.11     |
|              | DO        | −0.41$^b$  | −0.47$^b$| 0.55$^b$   | 0.16    | 0.14      |

$^a$ P < 0.05 (two-tailed).

$^b$ P < 0.01 (two-tailed). NH$_4^+$-N, ammonium nitrogen; TP, total phosphorus; COD, chemical oxygen demand; DO, dissolved oxygen.

effluent from wastewater treatment plants (WTP) and direct discharge, cropland, forestland and livestock production system were dominant sources of N discharge to surface water in 2015. The proportion of N discharge derived from domestic wastewater and forestland increased while that from industrial wastewater and cropland decreased in all eight watersheds. The contribution of other sources such as aquaculture, livestock production and urban lawn remained stable across the study period (figure 3).

The contribution of different sources to riverine N export varied in the eight watersheds. We classified the eight watersheds into three categories based on the sum of proportion of urban land and cropland (representing the intensity of human activities): low (Oujiang), moderate (Feiyun River, Qiantang River, Aojiang and Jiaojiang) and severe (Tiaoxi, Yongjiang and Canal). Forestland, domestic wastewater and livestock production were the major contributors of N discharge in the low, moderate and severe categories, respectively.

The calculated N concentrations (based on the total N discharge flux) were positively correlated ($R^2 = 0.51$, $P < 0.05$) with the monitored NH$_4^+$ concentrations at the watershed scale (figure 4), suggesting that the source apportionment based on mass balance very well captured the variations of riverine N export.

In order to analyze the degree of uncertainty of the calculation, we used a Monte Carlo ensemble simulation to quantify the variability of the riverine N export. Results showed that the N exports ranged from 214.0 to 217.4, 233.5 to 235.2, 256.0 to 258.0 and 262.4 to 263.9 Gg N in 2000, 2005, 2010 and 2015, respectively. More details can be found in the SI.
Figure 4. Relationship between the monitored and calculated N concentration at the watershed scale. The monitored N concentration is the average NH$_4^+$ concentration for all 106 monitoring sites, while the calculated N concentration is the riverine N export divided by the water volume in the corresponding watershed. The form of Nr in water bodies mainly includes NH$_4^+$ and NO$_3^-$, However, the provincial monitoring network does not take nitrate into consideration because nitrate was not regarded as a major pollutant in water, but NH$_4^+$ was. It is very difficult to quantify the conversion of NH$_4^+$ in the water after being lost from subsystems such as cropland. Thus, the calculated N concentration was larger than the monitoring N concentration.

Figure 5. The relationship between the monitored concentration of pollutants and the proportion of land area occupied by different land use types within the 1000 m wide buffer zone: (a) NH$_4^+$ vs urban land + cropland; (b) NH$_4^+$ vs forestland + wetland + grassland; (c) TP (total phosphorus) vs urban land + cropland; (d) TP vs forestland + wetland + grassland. NH$_4^+$-N, ammonium nitrogen; TP, total phosphorus.

3.3. Land use composition and water quality
To further quantify the contribution of different land use compositions on water quality, we selected NH$_4^+$ and TP as indicators to estimate their responses to changes in land use composition. Positive correlations between the sum proportion of urban land and cropland and NH$_4^+$ and TP were found, and water quality was better in watersheds with a greater proportion of forestland, grassland and wetlands (figure 5). NH$_4^+$ concentration was more strongly correlated with urban land than cropland, while the reverse was observed for TP (figure 6).

There was no significant relationship between land use change and the changes in riverine N export, such as urban land ($R^2 = 0.07, P = 0.359$) that has increased dramatically during the study period (figure S4). For example, the area of urban land in Tiaoxi was increased by 111%, but the riverine N export derived from wastewater discharged did not increase with urban land uses. Other land use type changes such as
cropland ($R^2 = 0.06, P = 0.475$) and forestland ($R^2 = 0.15, P = 0.087$) were not significantly correlated with the riverine N export either (figure S4).

4. Discussion

Our results from source apportionment based on the CHANS model agree well with those from land use based analysis on water quality, i.e. point source from urban areas (mainly direct discharge of domestic wastewater or effluent from WTP) was the dominated source of riverine N export in Zhejiang (figure 3, figure 6, figure S5). In fact, significant progress has been made on the control of point source pollution with the development of wastewater treatment technology and government investment in China (Qiu et al. 2010, Wang 2012). The proportion of urban wastewater being treated in Zhejiang increased from 33% in 2000 to 91% in 2015, significantly reducing the direct discharge of domestic wastewater to the rivers (EPBZ 2015). However, the effluent of WTP has become a dominant source of riverine N export derived from cropland mainly due to the reduction in crop area and N fertilizer input (ZPBS2016). The rapid urbanization in Zhejiang has occupied 12% of cropland area. The development of modern agriculture has improved the N use efficiency in cropland, reducing the amount of N fertilizer applied (from 600 000 to 460 000 tons in total during the research period in Zhejiang) and N loss to the rivers (ZPBS 2016). Thus, we found an increasing contribution to riverine N export from urban land use compared to that from cropland (figure 3).

Nitrogen export from forestland to rivers is mainly derived from N deposition to the forestland (Gu et al. 2015). Urbanization increases the use of fossil fuel and more NOx is emitted to the atmosphere that increases N deposition to natural ecosystems (Liu et al. 2013). These enhanced N depositions have resulted in N saturation in forestland and eventually increased the Nr runoff from forestland to rivers in Zhejiang (Chen et al. 2016), although the forestland area remained stable during the study period.
Systems with a low land use intensity (such as forestland) can also contribute to riverine N export (Ouyang et al 2010), but may also reduce N export through filtration, absorption and transformation of N before the N gets into waterways (Tong and Chen 2002). We found that pollutant concentrations substantially decreased with an increase in low intensity land use, but increased with land use dominated by human activities such as urban and cropland (figures 5 and 6). Water quality is closely associated with land use composition and the spatial configuration of urban, agricultural and forestland uses within a watershed (Lee et al 2009). Urban land use contributed more to riverine N export, while cropland contributed more to riverine TP export in Zhejiang (figure 6). Zhejiang province used less N fertilizer while the amount of P fertilizer applied remained stable from 2000 to 2015 (ZPBS 2016). Some environmental protection measures such as the use of non-phosphate detergent reduced P losses from urban areas, but no measures have been implemented on reducing N losses (EPBZ 2015). In addition, impervious surface in urban area rapidly transfers NH$_4^+$ to waterways, whereas the NH$_4^+$ in cropland is usually retained in the soil (Chen et al 2016).

The influence of land use on water quality varies with scale, e.g. the riparian buffer width (Guo et al 2009, Xiao et al 2016). White and Greer (2006) emphasized the role of grassland in improving water quality, while Cui et al (2016) suggested that the increasing proportion of grassland deteriorated water quality. In our study, the correlation between water quality and the proportion of grassland varied with buffer width. Grassland had the greatest impact on the reduction of NH$_4^+$ and TP on the buffer width of 500 m. Therefore, the effect of buffer zone width under different land uses should be considered to avoid the deterioration of water quality.

Water quality is affected by human activities and land use changes (Ahearn et al 2005, Wilson 2015); however, no relationship was found between land use change and riverine N export in the eight watersheds in this study. It appears that land use composition has little effect on riverine N export, because it is only the carrier of human activities, while human activities are the dominant factors affecting water quality. Some land use types such as forestland and wetland can reduce the quantity of Nr export to rivers to some extent (Silva and Williams 2001, Su et al 2013). In fact, other land use types such as urban land and cropland also have the ability to reduce Nr export to rivers. However, N fertilizer input or wastewater discharge exceeds the capacity of land to retain these Nr inputs, leading to Nr export to rivers. Therefore, human activities rather than land use change drive the riverine N export, whereas land use composition only mediates the amount of Nr export. Furthermore, other factors such as climate variability may also affect the riverine N export (Tian et al 2015, Yang et al 2015), which warrants further research.

5. Conclusion

This study analyzed the source apportionment of riverine N export in Zhejiang Province and its eight watersheds based on a CHANS model. Our results showed that point sources such as the effluents from WTP and direct discharge of domestic wastewater is the most important source of riverine N export in Zhejiang. The ‘Joint Control of Five Waters’ proposed by the local government focused on the treatment of wastewater is consistent with our results. Meanwhile, despite the reduction in the proportion of nonpoint sources such as agriculture and forest, their considerable contribution to the overall riverine pollution is worth attention. At the watershed scale, our estimate of N export agreed well with the monitored riverine N concentration in the eight major watersheds in Zhejiang. The dominant source of riverine N export varied among the watersheds, suggesting measures appropriate to each watershed are required to reduce N export in the watershed level. The strength of correlation between land use composition and water quality sharply decreased with decreasing width of the buffer zone. We concluded that human activities rather than land use change itself affect the N export and land use composition only mediated the amount of Nr export.

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