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RESEARCH ARTICLE

A lagged variable model for characterizing temporally dynamic export of legacy anthropogenic nitrogen from watersheds to rivers

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Abstract Legacy nitrogen (N) sources originating from anthropogenic N inputs (NANI) may be a major cause of increasing riverine N exports in many regions, despite a significant decline in NANI. However, little quantitative knowledge exists concerning the lag effect of NANI on riverine N export. As a result, the N leaching lag effect is not well represented in most current watershed models. This study developed a lagged variable model (LVM) to address temporally dynamic export of watershed NANI to rivers. Employing a Koyck transformation approach used in economic analyses, the LVM expresses the indefinite number of lag terms from previous years' NANI with a lag term that incorporates the previous year's riverine N flux, enabling us to inversely calibrate model parameters from measurable variables using Bayesian statistics. Applying the LVM to the upper Jiaojiang watershed in eastern China for 1980–2010 indicated that ~97 % of riverine export of annual NANI occurred in the current year and succeeding 10 years (~11 years lag time) and ~72 % of annual riverine N flux was derived from previous years' NANI. Existing NANI over the 1993–2010 period would have required a 22 % reduction to attain the target TN level (1.0 mg N L^{-1}), guiding watershed N source controls considering the lag effect. The LVM was developed with parsimony of model structure and parameters (only four parameters in this study); thus, it is easy to develop and apply in other watersheds. The LVM provides a simple and effective tool for quantifying the lag effect of anthropogenic N input on riverine export in support of efficient development and evaluation of watershed N control strategies.

Keywords Watershed · Lag time · Legacy nutrients · Water quality model · Nitrogen saturation · Eutrophication

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Introduction

Anthropogenic activities have substantially increased nitrogen (N) availability in many terrestrial ecosystems to an extent that exceeds the ability to assimilate N, often resulting in large increases in riverine N fluxes to coastal waters (Bouraoui and Grizzetti 2014; Du et al. 2014). Various management efforts have been implemented to control excessive N in riverine ecosystems, as it degrades aquatic ecosystem health, decreases water quality for several beneficial uses, and causes eutrophication and hypoxia in downstream freshwater and coastal waters (Sun et al. 2013; Li et al. 2014). Despite a significant decline in anthropogenic N inputs with the implementation of the Clean Water Act (1972) in the USA and the Nitrate Directive (1991) in Europe, riverine N concentrations continue to increase in many areas, such as the Mississippi River, Chesapeake Bay, North Sea, and Baltic Sea (Worrall et al. 2009; Dubrovsky and Hamilton 2010; Bouraoui and Grizzetti

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2014). Nitrogen leaching from legacy sources has been recognized as a primary reason for these limited results, due to the long lag time (ranging from years to decades) elapsed between watershed N inputs and riverine export (Sebilo et al. 2013; Sanford and Pope 2013; Tesoriero et al. 2013). Exceedance of water quality targets has prompted regulatory agencies to reevaluate nutrient management standards, measures, and guidelines (Sharpley et al. 2013). This requires quantitative information concerning the lag effect for anthropogenic N inputs on changes in riverine N export to optimize pollution control expenditures, strategies, and schedules, as well as to set appropriate expectations for the public (Sanford and Pope 2013; Bouraoui and Grizzetti 2014).

Various lumped watershed models (e.g., export coefficient models, SPARROW, and PolFlow) and mechanistic models (e.g., AGNPS, HSPF, and SWAT) (De Wit et al. 2003; Moriasi et al. 2007; Li et al. 2014; Du et al. 2014), as well as N budgeting approaches such as net anthropogenic nitrogen inputs (NANI) (Han et al. 2011; Hong et al. 2012; Howarth et al. 2012), are available for quantifying the impact of anthropogenic N inputs on riverine N fluxes. However, the calibration procedures for these models contain considerable uncertainty since measured riverine N fluxes are a mixture of N and water sources with different ages and the lag time is often larger than the temporal extent of available calibration data (Meals et al. 2010; Bouraoui and Grizzetti 2014). Due to the complexities of understanding transit time and biogeochemical mechanisms for N passing through the soil profile, vadose zone, and groundwater to the river network (Meals et al. 2010; Hamilton 2012; Sebilo et al. 2013), the N leaching lag effect is not well addressed and formulated in most current watershed mechanistic models (Meals et al. 2010; Sanford and Pope 2013; Bouraoui and Grizzetti 2014). For example, values for groundwater residence time in the SWAT model range from 0 to 500 days, which is much lower than estimated residence times (years to decades) derived from stable isotope tracers (mainly ^{18}O and ^{3}H) (Iqbal 2002; Phillips and Lindsey 2003; Tesoriero et al. 2013). Similarly, the HSPF model does not consider such long residence times for groundwater (Sanford and Pope 2013).

While lumped watershed models and the NANI budgeting approach are not able to assess the fine temporal resolutions (daily, monthly, and seasonal) available from mechanistic models, they are widely applied to quantify the relationship between anthropogenic activities and riverine N exports due to their simple structure and limited data requirements (Shen and Zhao 2010; Howarth et al. 2012; Bouraoui and Grizzetti 2014). Lumped models and the NANI budgeting approach generally assume that the N status of soils, aquifers, and biomass is at steady state (at least over a multi-year period). Thus, they are commonly applied to predict riverine N flux using a multi-year average temporal resolution to avoid the uncertainty derived from the N leaching lag effect (De Wit et al. 2003; Howarth et al. 2006; Swaney et al. 2012). However, a major challenge remains in determining the appropriate length of the multi-year period that should be used to estimate N source inputs to satisfy the steady-state assumption. Furthermore, lumped models and the NANI budgeting approach cannot explicitly address the lag effect of N leaching to rivers.

Overall, current watershed models do not effectively represent the lag effect of anthropogenic N inputs on riverine export, resulting in a paucity of quantitative knowledge concerning such a lag effect at the watershed scale. This study developed a lagged variable model (LVM) for determining temporally dynamic exports of watershed NANI by rivers and to quantify the contribution of NANI from any previous year (i.e., legacy N sources) to annual riverine TN export. We introduced a Koyck transformation approach from the economic literature to deal with models having an indefinite number of lag terms (Ravines et al. 2006). This approach is able to transform the indefinite number of lag terms from previous years' NANI in the LVM into a lag term incorporating the previous 1 year's riverine TN flux, enabling us to inversely calibrate the unknown parameters in the LVM from measurable variables using Bayesian statistics. The efficacy of the model was demonstrated for a 31-year water quality record (1980–2010) for TN fluxes from the upper Jiaojiang watershed in eastern China. Furthermore, the maximum allowable NANI and the required NANI reduction for attaining a target riverine TN concentration (such as 1.0 mg N L^{-1}) were inversely estimated by the calibrated LVM. The model results will inform water resource research and management efforts to improve watershed N modeling and N control strategies.

Materials and methods

Watershed description

The upper Jiaojiang watershed is located in the highly developed Taizhou region of Zhejiang Province, China (Fig. 1). The sampling location for this study was 55 km upstream of Taizhou Estuary. The river drains a total area of 2474 km^2 and has an average annual water depth of 5.42 m and discharge of 72.9 m^3 s^{-1} at the sampling location. There is no river regulation, such as dams and transboundary water withdrawal facilities. The climate is subtropical monsoon having an average annual temperature of 17.4 °C and average annual precipitation of 1400 mm. Rainfall mainly occurs in May–September with a typhoon season in July–September. Agricultural land (including paddy field, garden plot, and dry land) averaged ~12 % of total watershed area in 1980–2010 (Table 1), with developed land (including rural and urban residential lands, roads, and mining and industry lands), forest, and barren land (including surface waters, wetlands, rock, and natural reservation land) contributing ~3, ~67, and ~18 %, respectively. The

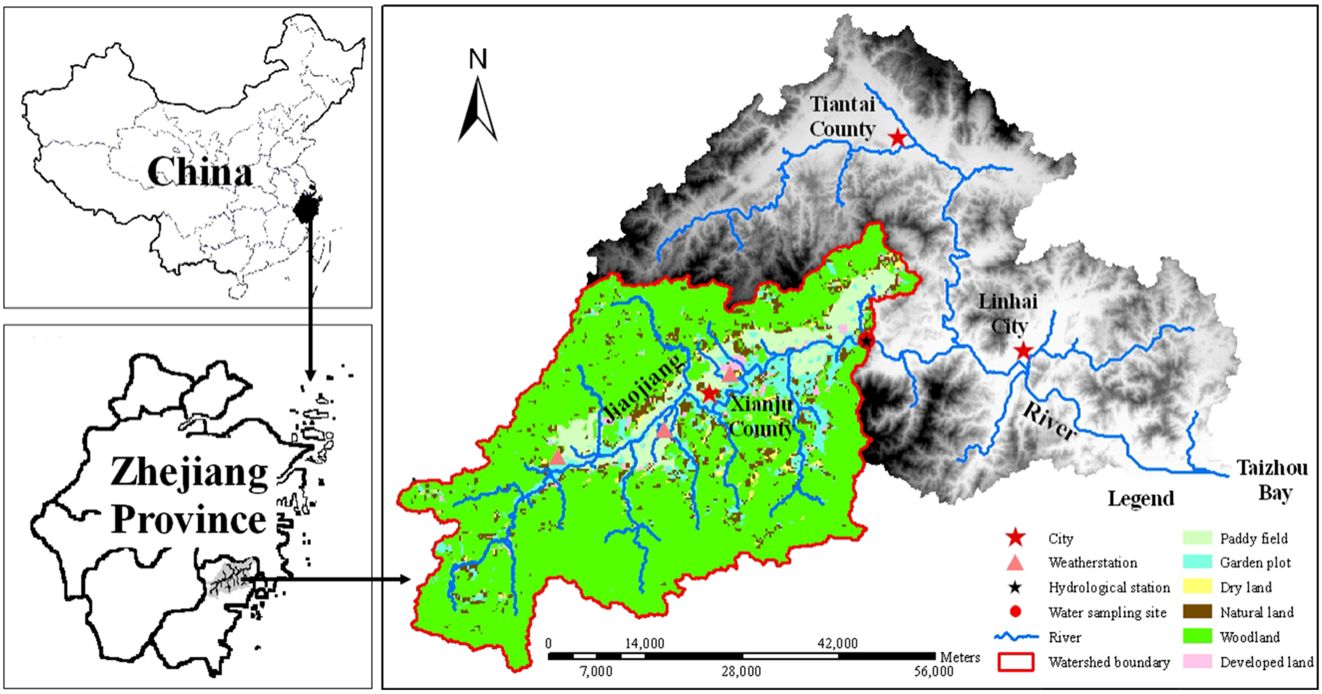

Fig. 1 Location of upper Jiaojiang watershed in China and Zhejiang Province and the river hydrology and water quality sampling site

economic role of agriculture has been increasingly replaced by industry since the 1990s, resulting in a remarkable reduction (~40 %) in chemical N fertilizer application since 2000. Agricultural land area irrigated and drained with cement channels and pipes increased by ~2-fold since 2000 (Electronic supplementary material A).

Evaluation of change in agricultural soil available nitrogen

Extensive soil samples (1 composite sample/15 ha for plain regions and one composite sample per 25 ha for hilly regions) were collected from the top 20-cm layer of agricultural lands (paddy field, dry land, and garden plot) in the watershed by the local Agriculture Bureau in 1984 and 2009. Available N was measured in the upper 20-cm layer at the same location (n=195) in both years and used to evaluate changes between 1984 and 2009 (Electronic supplementary material B).

Riverine TN flux estimate

River water samples were collected once every 4 to 8 weeks during the 1980–2010 study period (n=250 sampling times) (Fig. 1). Data for river TN and nitrate concentrations, daily discharge, and daily precipitation were obtained from the local Environment Protection Bureau, Hydrology Bureau, and Weather Bureau, respectively. Daily river discharge records in 1980–2010 were divided into high (0–30 %), medium

Table 1 Characteristics of land-use distribution, population, domestic animal, and hydroclimate for the upper Jiaojiang watershed over the 1980–2010 period

Periods	1980s	1990s	2000s
Agricultural land	11 %	11 %	13 %
Developed land	2 %	3 %	3 %
Forest	68 %	68 %	67 %
Barren land	18 %	18 %	17 %
Annual area planted to crops (km^2)	636.6	600.9	469.5
Precipitation (m year^{-1})	1.37	1.42	1.42
River water discharge (m^3 s^{-1})	72.2	77.5	70.7
Population density (capita km^{-2})	248	266	288
Animal density (capita km^{-2})	107	80	99
Drained agricultural land area percentage	19 %	18 %	29 %

The number of each type of animal is converted into the number of pigs according to their nitrogen excretion rates as shown in Table D4 in the Electronic supplementary material. All values are the average for each time period

(30–70 %), and low flow (70–100 %) regimes using the duration curve method (Chen et al. 2012). To estimate annual TN flux based on the discrete TN concentration monitoring data, the LOADEST model (Sun et al. 2013) was applied for predicting daily TN concentration and flux, resulting in high R^2 (R^2=0.73 for concentration and R^2=0.90 for flux, n=250, p<0.001) and low average relative error (±5 % for concentration and ±3 % for flux) between measured and modeled values (Electronic supplementary material C).

NANI estimation and uncertainty analysis

NANI was estimated as the sum of four major components: atmospheric N deposition, commercial fertilizer N application, agricultural N fixation, and net food, feed, and seed N input (Han et al. 2011, 2014; Hong et al. 2012; Howarth et al. 2012). The net food and feed N input were calculated as the sum of human and livestock N consumption minus the sum of livestock and crop N production (Electronic supplementary material D).

To gain insight into the uncertainty of NANI estimation, an uncertainty analysis was performed using Monte Carlo simulation (Electronic supplementary material D). In performing the Monte Carlo simulation, we assumed that all the model parameters in the NANI estimation followed a normal distribution with a coefficient of variation of 30 % for each of the parameters (Yan et al. 2011; Ti and Yan 2013; Chen et al. 2014). A total of 10,000 Monte Carlo simulations were performed to obtain the mean and 95 % confidence interval for annual NANI values.

Development of the lagged variable model for riverine TN export

The model developed in this study was inspired by previous watershed statistical models (De wit et al. 2003; Li et al. 2014; Bouraoui and Grizzetti 2014), where the riverine N flux was modeled using a non-linear regression equation based on N sources and watershed attributes at a multi-year average resolution. NANI were used instead of various N sources (i.e., fertilizer, animal waste, domestic sewage, and atmospheric deposition) as model input to avoid the introduction of new uncertainty with estimates of N flows among human, animal, and vegetation biomass within the watershed. Furthermore, the model was developed at a yearly resolution rather than at a daily/monthly/seasonal resolution, since watershed data concerning N sources and sinks are usually only available for annual time steps.

NANI has been widely recognized as an effective predictor of riverine N fluxes and their relationship is commonly described using an exponential function due to the effects of progressive N saturation in landscapes (McIsaac et al. 2001; Han et al. 2009; Hong et al. 2012; Chen et al. 2014). In addition, fractional export of NANI by rivers is strongly related as a power function to various watershed attributes, including hydroclimate (e.g., water yield, precipitation, and temperature) (McIsaac et al. 2001; De wit et al. 2003; Han et al. 2009; Howarth et al. 2012; Li et al. 2014), land use (Groffman et al. 2004; Han et al. 2009), and agricultural management (e.g., drainage systems, fertilizer application, and tillage) (Sobota et al. 2009; Kopáček et al. 2013; Chen et al. 2014). Current models are generally established using a multi-year average temporal resolution to reduce the uncertainty derived from the N leaching lag effect (Howarth et al. 2006; Hong et al. 2012; Swaney et al. 2012; Li et al. 2014). However, annual riverine TN export originates from both the current year's NANI and legacy N sources derived from previous years' NANI that are transiently stored in the watershed (e.g., soils and aquifers) (Meals et al. 2010; Hong et al. 2012; Tesoriero et al. 2013; Bouraoui and Grizzetti 2014; Chen et al. 2014). Therefore, we developed the following lagged variable model (LVM) for annual riverine TN flux at the watershed outlet (F_t, kg N ha^{-1} year^{-1}) that incorporates both NANI from the current year and from any previous year (lag terms) based on the observed relationships between NANI and riverine N flux in previous studies (McIsaac et al. 2001; Han et al. 2009; Swaney et al. 2012; Chen et al. 2014):

$$F_t = a\prod_{j=1}^{m}\theta_{t,j}^{b_j}\exp\left[\beta_0\mathrm{NANI}_t + \beta_0\sum_{i=1}^{n}\left(\rho_{t-i}\mathrm{NANI}_{t-i}\right)\right] \qquad (1)$$

where subscript t denotes the tth year, NANI_{t-i} denotes NANI in the previous ith year, and $\theta_{t,j}$ denotes the normalized value of the jth explanatory variable that influences fractional export of NANI to rivers in the tth year. Unknown parameters a and b_j denote the response magnitude of N export efficiency to changes in explanatory variables (e.g., meteorology, hydrology, and land use). Unknown parameter β_0 represents the export fraction coefficient for the current year's NANI_t and historical (legacy) NANI_{t-i} to the river, which is independent of watershed temporal attributes and reflects the influence of inherent watershed geological and geomorphologic characteristics. Unknown parameter ρ_{t-i} denotes the residual coefficient of historical NANI_{t-i}. Expressing Eq. (1) in a linear form after a logarithmic transformation yields:

$$\ln(F_t) = \ln(a) + \sum_{j=1}^{m}\left[b_j\ln\left(\theta_{t,j}\right)\right] + \beta_0\mathrm{NANI}_t + \beta_0\sum_{i=1}^{n}\left(\rho_{t-i}\mathrm{NANI}_{t-i}\right) \qquad (2)$$

Due to year-to-year N removals via riverine export, denitrification, biomass uptake/storage, and wood product export (Van Breemen et al. 2002), the remaining NANI from the previous years in the watershed (e.g., soils, aquifers, and sediments) decreases over time. Therefore, the historical NANI_{t-i} residual coefficient ρ_{t-i} was assumed as a power decay function of relevant years and explanatory variables:

$$\rho_{t-i} = \lambda^{\sum_{i=1}^{n}\left(\prod_{k=1}^{n} R_{t-i,k}\right)} \tag{3}$$

where λ is the decay coefficient for historical NANI_{t-i} and denotes the response magnitude of decay to changes in explanatory variables, $R_{t-i,\,k}$ denotes the normalized value of the kth explanatory variable that influences N removal through riverine export, denitrification, and forest biomass uptake/storage/export. Based on Eq. (3), Eq. (2) can be further expressed as:

$$\ln(F_t) = \ln(a) + \sum_{j=1}^{m}\left[b_j \ln(\theta_{t,j})\right] + \beta_0\left(\mathrm{NANI}_t + \lambda^{\prod_{k=1}^{n} R_{t-1,k}}\mathrm{NANI}_{t-1} + \ldots + \lambda^{\prod_{k=1}^{n} R_{t-1,k}} + \ldots + \prod_{k=1}^{n} R_{t-i,k}\mathrm{NANI}_{t-i}\right) \tag{4}$$

Equation (4) has an indefinite number of lag terms from previous years' NANI; thus, it cannot be solved directly. A Koyck transformation approach that is commonly applied in economic analyses (Ravines et al. 2006) was adopted to express the indefinite number of lag terms from any previous year's NANI in Eq. (4) with a lag term. Based on Eq. (4), riverine TN flux in the $t-1$th year (F_{t-1}) can be estimated as:

$$\ln(F_{t-1}) = \ln(a) + \sum_{j=1}^{m}\left[b_j \ln(\theta_{t-1,j})\right] + \beta_0\left(\mathrm{NANI}_{t-1} + \lambda^{\prod_{k=1}^{n} R_{t-2,k}}\mathrm{NANI}_{t-2} + \ldots + \lambda^{\prod_{k=1}^{n} R_{t-2,k}} + \ldots + \prod_{k=1}^{n} R_{t-i,k}\mathrm{NANI}_{t-i}\right) \tag{5}$$

Multiplying $\lambda^{\prod_{k=1}^{n} R_{t-1,k}}$ to both sides and subtracting Eq. (5) from Eq. (4) yields:

$$\ln(F_t) = \ln(a) + \sum_{j=1}^{m}\left[b_j \ln(\theta_{t,j})\right] + \beta_0\mathrm{NANI}_t + \lambda^{\prod_{k=1}^{n} R_{t-1,k}}\left[\ln(F_{t-1}) - \ln(a) - \sum_{j=1}^{m}\left[b_j \ln(\theta_{t-1,j})\right]\right] \tag{6}$$

The resulting Eq. (6) based on the Koyck transformation involves a finite number of variables (NANI_t, $\theta_{t,\,j}$, $\theta_{t-1,j}$, $R_{t-1,\,k}$, and F_{t-1}) and parameters (a, b_j, β_0, and λ) that can be calibrated. Most importantly, this model addresses the potential contribution from any previous years' NANI through incorporating one lag term and overcomes the difficulty raised from determining how many previous years of NANI should be considered in modeling riverine N export.

To calibrate the unknown parameters a, b_j, β_0, and λ in the LVM based on Eq. (6), a Bayesian approach coupled with the Markov Chain Monte Carlo algorithm and a Gibbs sampler was adopted using WinBUGS 1.4 (Chen et al. 2012). This methodology has been widely and successfully applied for calibrating the lagged variable model parameters in economic analyses (Ravines et al. 2006). A detailed description of the Bayesian calibration approach for relevant water quality model parameters is available in Shen and Zhao (2010) and Chen et al. (2012). According to parameter values from previous relevant studies (McIsaac et al. 2001; Howarth et al. 2006; Han et al. 2009; Chen et al. 2014), the prior distribution of the four unknown parameters was assumed to follow a normal distribution, i.e., a, 5.0±5.0; b_j, 1.0±1.0; β_0, 0.018±0.018; and λ, 0.5±0.5. The WinBUGS code for Bayesian calibration is available in Electronic supplementary material E. To obtain the best-fit posterior model for parameters a, b_j, β_0, and λ, two Markov chains were initiated at different arbitrary initial values as well as being independent of each other using WinBUGS 1.4. The generated posterior distributions of parameters were all based on 10,000 MCMC interactions (Chen et al. 2012) until the model successfully converged according to a visual inspection of the marginal trace plots and Monte Carlo errors <10 % of standard deviation (Chen et al. 2012). After model convergence, a total of 1000 samples for each unknown quantity were randomly taken from the following iterations to reduce autocorrelation (Shen and Zhao 2010). The agreement for annual TN fluxes between those estimated by LOADEST and those modeled by Eq. (6) was evaluated using correlation (R^2) and Nash–Sutcliffe coefficient metrics (Moriasi et al. 2007; Du et al. 2014).

To determine the major watershed temporal attributes as the most efficient explanatory variables for $\theta_{t,j}$ and $R_{t-1,k}$ in Eq. (6), firstly, only water yield (W, m year^{-1}) was adopted in both $\theta_{t,j}$ and $R_{t-1,k}$ for calibrating unknown parameters a, b_j, β_0, and λ using Bayesian statistics. Then a correlation analysis was adopted to determine the autocorrelations among the potential explanatory variables. For the upper Jiaojiang watershed, there were significant correlations among temperature (T, °C), drained agricultural land area percentage (DA), and developed land area percentage (D; i.e., r=0.55 for T vs DA, r=0.84 for T vs D, and r=0.92 for DA vs D, p<0.01), while no significant correlations were found between W and each of these factors. However, there was a close relationship between W and precipitation (r=0.99, p<0.01). To avoid the influence of overlap among introduced explanatory variables, the following combinations of independent explanatory variables were further adopted in $\theta_{t,j}$ and $R_{t-1,k}$, i.e., $\theta_{t,j}$, W and DA, W and D, and W and $1/T$; $R_{t-1,k}$, $W\times$DA, $W\times D$, and $W\times T$, resulting in eight alternative formats for the model (Electronic supplementary material F). Finally, the best set of explanatory variables for Eq. (6) was determined according to model agreement among the alternative model formats. All individual explanatory variables and their products used for calibration were reduced to the same scale using the maximum scaling method (Chen et al. 2012), i.e., $X_{t,i}/X_{m,i}$, where $X_{t,i}$ is the value of the ith explanatory variable or the product of several variables in tth year and $X_{m,i}$ is the maximum of the ith explanatory variable or product of several variables among the 31 years.

Based on the calibrated posterior parameters, export of NANI by rivers in the current year (FNANI$_t$, kg N ha^{-1} year^{-1}) and in the succeeding years (FNANI$_{t+1}$, kg N ha^{-1} year^{-1}) can be estimated according to Eq. (1) as follows:

$$F\mathrm{NANI}_t = a\prod_{j=1}^{m}\theta_{t,j}^{b_j}\exp(\beta_0\mathrm{NANI}_t) - a\prod_{j=1}^{m}\theta_{t,j}^{b_j}\ \text{(Current year)} \tag{7}$$

$$F\mathrm{NANI}_{t+1} = a\prod_{j=1}^{m}\theta_{t+1,j}^{b_j}\exp\left(\beta_0\lambda^{\prod_{k=1}^{n}R_{t,k}}\mathrm{NANI}_t\right) - a\prod_{j=1}^{m}\theta_{t+1,j}^{b_j} \tag{8}$$

(Succeeding years)

Estimation of required NANI reduction

To demonstrate the application of the lagged variable model for water quality management, the maximum allowable NANI and required NANI reduction were determined using the coupled posterior a, b_j, β_0, and λ values. The maximum allowable NANI (NANI$_{t,m}$, kg N–ha^{-1}–year^{-1}), which is the allowable anthropogenic N amount that can be input to the watershed and still meet the desired water quality target (i.e., TN=1.0 mg N L^{-1} in this study, which is designated by the local environment protection agency) at the river outlet in the tth year, can be inversely estimated from Eq. (6):

$$\mathrm{NANI}_{t,m} = \frac{\ln(F_{t,m}) - \ln(a) - \sum_{j=1}^{m}\left[b_j\ln(\theta_{t,j})\right] - \lambda^{\prod_{k=1}^{n}R_{t-1,k}}\left\{\ln(F_{t-1,m}) - \ln(a) - \sum_{j=1}^{m}\left[b_j\ln(\theta_{t-1,j})\right]\right\}}{\beta_0} \tag{9}$$

where $F_{t,m}$ and $F_{t-1,m}$ are the maximum allowable riverine TN flux in the tth and $t-1$th years, respectively, which were calculated by dividing the product of target TN concentration and river discharge by watershed area. Required NANI reduction was then estimated as the difference between NANI$_{t,m}$ and existing NANI$_t$.

Results

Riverine TN export in relation to NANI, hydroclimate, and land use

Over the 1980–2010 study period, although there were no significant temporal trends in either precipitation or water yield (p>0.05; Fig. 2a), estimated riverine TN flux increased by 91 % from an average 8.5 kg N ha^{-1} year^{-1} in the 1980s, to 10.1 kg N ha^{-1} year^{-1} in the 1990s, and to 13.0 kg N ha^{-1} year^{-1} in the 2000s (Fig. 2b). Annual mean TN concentration steadily increased by 120 % from 1980 (0.94 mg N L^{-1}) to 2010 (2.10 mg N L^{-1}). Since 1993, annual mean TN concentration has exceeded the regulatory TN concentration target of 1.0 mg N L^{-1} that is set by the local environmental protection agency. Estimated NANI increased from 38.0 kg N ha^{-1} year^{-1} in 1980 to 76.9 kg N ha^{-1} year^{-1} in 1999 followed by a decline from 77.6 kg N ha^{-1} year^{-1} in 2000 to 67.3 kg N ha^{-1} year^{-1} in 2010 (Fig. 2c). Since 2000, NANI showed a decreasing trend due to decreased chemical N fertilizer application and agricultural biological N fixation from decreased crop cultivation area (~44 %).

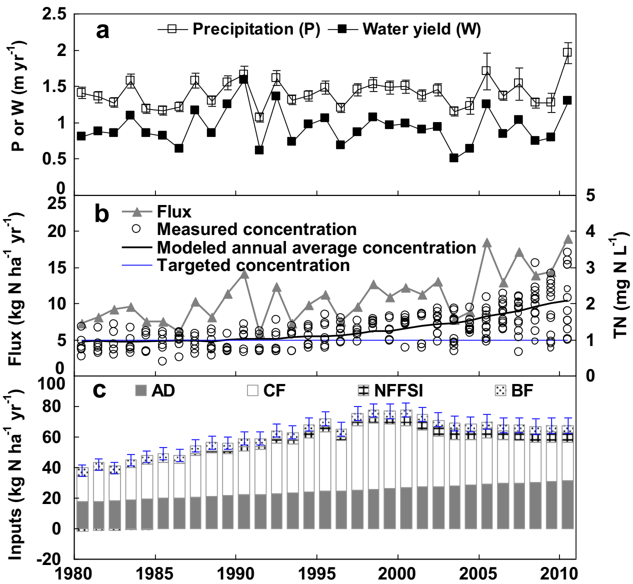

Fig. 2 Historical trends of precipitation, water yield (**a**), riverine TN flux, measured concentration, modeled annual average concentrations, and targeted TN concentration in the upper Jiaojiang watershed (1980–2010) (**b**), and N inputs from atmospheric deposition (*AD*), chemical fertilizer (*CF*), biological fixation (*BF*), and net food/feed/seed input (*NFFSI*) to the watershed (**c**). *Error bars* in (**a**) and (**c**) denote standard deviations of measurements from three weather monitoring sites within the watershed and 95 % confidence interval of NANI, respectively

Annual riverine TN flux was positively correlated with NANI, and their relationship was best fit with an exponential function (Table 2). Exponential relationships were also found between TN exports and net food, feed and seed input, and between TN export and atmospheric deposition, while no significant relationships were found between TN export and other individual inputs. Compared with NANI, water yield (45 vs 26 %), precipitation (57 vs 26 %), drained agricultural land area (32 vs 26 %), and developed land (47 vs 26 %), explained a larger fraction of variability for annual riverine TN export, which were best described by power functions. These analyses support the exponential and power functions adopted in developing the lagged variable model Eq. (1).

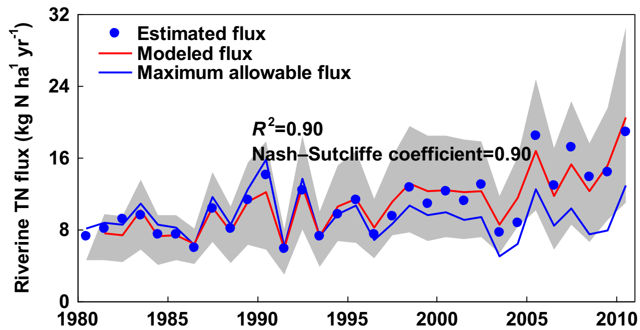

Fig. 3 LOADEST estimated vs modeled riverine TN flux by the lagged variable model and maximum allowable TN flux (i.e., by multiplying the target TN level by river water discharge) in 1980–2010 in the upper Jiaojiang watershed. *Shadow area* denotes 95 % confidence interval of modeled riverine TN flux

The lagged variable model calibration

When only annual water yield was considered as the explanatory variable for $\theta_{t,\ j}$ and $R_{t-1,\ k}$ in Eq. (6), the Bayesian calibrated parameters a, b_j, β_0, and λ had small Monte Carlo errors (<5 % of standard deviation; an estimate of the difference between the mean of the sampled values and the true posterior mean) (Table F1 of Electronic supplementary material F). During the 10,000 iterations, the trace plots for the four parameters successfully converged after 4000 iterations, and the posterior samples of the model parameters were kept for further inference (Fig. F1 of Electronic supplementary material F). The calibrated posterior parameters yielded high agreement between modeled riverine TN fluxes using Eq. (6) and LOADEST estimated values (Fig. 3) with a high R^2 value (0.90), high Nash–Sutcliffe coefficient (0.90), and low relative errors (<5 %). Further consideration of drained agricultural land area, developed land area, and temperature as explanatory variables for $\theta_{t,\ j}$ and $R_{t-1,\ k}$ decreased the model's predictive capability, as indicated by lower R^2 values (0.53–0.88) and Nash–Sutcliffe coefficients (0.52–0.88) than the model only considering water yield (Table F1 of Electronic

Table 2 Results of regression analysis between year-to-year riverine TN flux (y, kg N ha^{-1} year^{-1}) and various independent parameters (x) in 1980–2010 (n=31)

Independents	Regression equations	R^2
NANI (kg N ha^{-1} year^{-1})	$y=4.424e^{0.0136x}$	0.26**
Atmospheric deposition (kg N ha^{-1} year^{-1})	$y=3.076e^{0.0459x}$	0.44**
Net food, feed, and seed input (kg N ha^{-1} year^{-1})	$y=7.964e^{0.06x}$	0.40**
Precipitation (m year^{-1})	$y=5.763x^{1.755}$	0.57**
Water yield (m year^{-1})	$y=11.149x^{0.815}$	0.45**
Drained agricultural land (%)	$y=28.577x^{0.660}$	0.32**
Developed land (%)	$y=1499.9x^{1.382}$	0.47**

**p<0.01, significant level

supplementary material F). Therefore, this study used water yield (W) as the optimal explanatory variable resulting in Eq. (1) being explicitly expressed as:

$$F_t = 2.653 W_t^{0.684} \exp\left[0.0075\mathrm{NANI}_t + 0.0075\sum_{i=1}^{n}\left(0.600^{\sum_{i=1}^{n} W_{t-i}}\mathrm{NANI}_{t-i}\right)\right] \tag{10}$$

Dynamic export of annual NANI by river over years

From Eq. (7) and the calibrated parameters shown in Eq. (10), estimated mean riverine export of annual NANI from the current year was 1.11 kg N ha^{-1} year^{-1} on average (range, 0.50–1.73 kg N ha^{-1} year^{-1}; Fig. 4a), which represented only 1.7 % (range, 1.2–2.6 %) of the corresponding year's NANI. From Eq. (8), estimated mean export in the succeeding 1–30 years ranged from 0.002 to 3.5 %/year (i.e., 2.2–17.5 % for cumulative export), with 95 % of export occurring in the succeeding 10 years (Fig. 4a). Total cumulative export over the study period was 269.0 kg N ha^{-1}, which represented 14 % of total NANI. Over the 1980–2000 period, cumulative riverine export of annual NANI in the current and succeeding 10–30 years ranged from 4.42 to 13.9 kg N ha^{-1} year^{-1} and demonstrated a significant upward trend (R^2=0.82, p<0.01; Fig. 4b). Correspondingly, the cumulative riverine export fraction of NANI varied from 10.3 to 17.6 %, which also exhibited a significant increasing trend with time (R^2=0.85, p<0.01; Fig. 4b). The decreasing trends observed for cumulative export flux and fraction over the 2001–2010 period (Fig. 4b) result from their estimates using progressively fewer years (1–10 years) of future years' export data; thus, NANI from 2001–2010 will continue to export a considerable flux to the river in the 2011–2020 period. If we assume that annual water yield (0.57 m year^{-1}) and NANI (69.9 kg N ha^{-1} year^{-1}) in 2011–2020 remain at their average values for 2000–2010 (Fig. 2), the model predicted that annual riverine TN flux will increase ~1.2 % each year between 2011 and 2020.

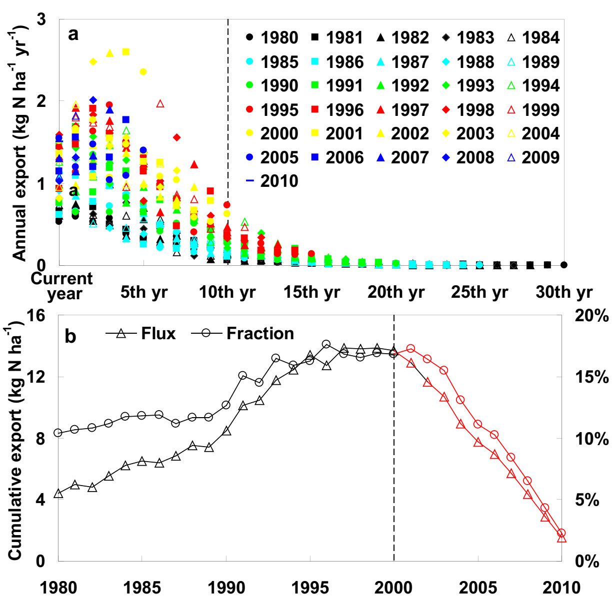

Fig. 4 Dynamic (**a**) and cumulative (**b**) export of annual NANI by river for current year and succeeding 1–30 years in the upper Jiaojiang watershed. Note: Decreasing cumulative export flux and fraction over the 2001–2010 period (indicated as *red circles and triangles*) result from their estimates using progressively fewer years (1–10 years) of future year's export

Riverine TN source apportionment

The natural background N export was determined by setting NANI$_t$ and NANI$_{t-i}$ simultaneously to zero in Eq. (10) (i.e., $2.653W^{0.684}$; W is maximum scaled annual water yield) (Han et al. 2009). Over the 1980–2010 period, estimated mean background riverine TN export was 1.84 kg N ha^{-1} year^{-1} on average (range, 1.20–2.65 kg N ha^{-1} year^{-1}) and contributed 18.0 % (range, 10.9–23.9 %) of the observed annual riverine TN export. As estimated by Eq. (7), current year's NANI only contributed 1.11 kg N ha^{-1} year^{-1} (range, 0.50–1.73 kg N ha^{-1} year^{-1}) and 10.3 % (range, 7.2–13.3 %) of the annual riverine TN flux (Fig. 5a). Previous years' NANI (legacy N sources) contributed 7.88 kg N ha^{-1} year^{-1} (range, 3.68–16.1 kg N ha^{-1} year^{-1}) and 71.7 % (range, 64.2–81.1 %) of the annual riverine TN flux (Fig. 5a). Of annual riverine TN flux, 67.6 % was from the previous 10 years' NANI on average (Fig. 5b). As expected, the contribution of previous years' NANI decreased with increasing time due to removal via denitrification, biomass uptake/storage/export and riverine export, decreasing from 11.5 % for the previous 1st year to 2.1 % for the previous 10th year (Fig. 5b).

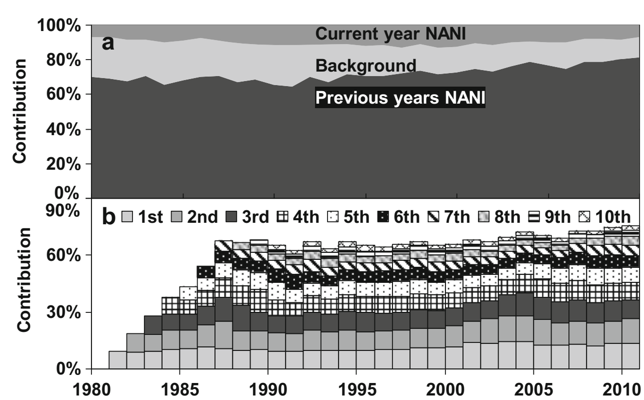

Fig. 5 Contributions of current-year NANI, previous-year NANI, and natural background sources (**a**), and each of previous 1–10 year's NANI (**b**) to observed annual riverine TN export in the upper Jiaojiang watershed

Required NANI reduction to meet water quality targets

From Eq. (9), estimated mean maximum allowable NANI to meet the water quality standard of TN=1.0 mg N L^{-1} ranged from 29.8 to 85.7 kg N ha^{-1} $year^{-1}$ with an average of 56.0 kg N ha^{-1} $year^{-1}$ for the upper Jiaojiang watershed over the 1980–2010 period (Fig. 6). The maximum allowable NANI increased with increasing water yield (R^2=0.72, p<0.01) resulting in larger and smaller values during high and low runoff years, respectively. When comparing the maximum allowable NANI with the existing NANI, the resulting NANI reductions ranged from −26.8 to 39.3 kg N ha^{-1} $year^{-1}$ (Fig. 6) and exhibited an increasing trend over the 1980–2010 study period (R^2=0.47, p<0.01). Negative values denote an allowable increase of NANI to the watershed that would still allow attainment of the TN target, which mainly occurred prior to 1992, consistent with the observed lower river TN concentration for the 1980–1992 period (<1 mg N L^{-1}; Fig. 2b). In contrast, for the 1993–2010 period, existing NANI would need to be reduced by 1 % (wetter years) to 57 % (drier years) to attain the target riverine TN level.

Discussion

Efficiency of the lagged variable model

The lagged variable model (LVM) that only considered water yield as the explanatory variable for $\theta_{t,\, j}$ and $R_{t-1,\, k}$ provided the best performance for the upper Jiaojiang River watershed (Table F1 of Electronic supplementary material F). Small Monte Carlo errors (Table F1 of Electronic supplementary material F, <5 % of standard deviations) and well-mixed posterior samples after the 4000th iteration for parameters a, b_j, β_0, and λ (Fig. F1 of Supporting Information F) indicate that the Bayesian model converged well and the posterior parameters were robust and unique (Shen and Zhao 2010; Chen et al. 2012). The strong agreement observed between the LVM modeled and LOADEST estimated riverine TN fluxes (R^2 and Nash–Sutcliffe coefficient=0.90; Fig. 3) suggest that the calibrated results are highly consistent, especially considering

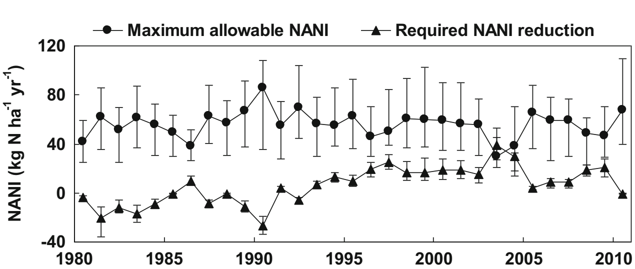

Fig. 6 Maximum allowable NANI and required NANI reduction to attain the targeted riverine TN concentration of 1.0 mg N L^{-1} in the upper Jiaojiang watershed. *Error bar* denotes 95 % confidence interval of estimated maximum allowable NANI and required NANI reduction

the complexities of N delivery across watershed landscapes to rivers. These calibrated results equal or exceed those obtained with other watershed N simulations using mechanistic models such as SWAT, AGNPS, and HSPF and lumped watershed models such as the export coefficient model, SPARROW and PolFlow model, as well as statistical models developed between NANI and riverine TN flux (Nash–Sutcliffe coefficient varied between 0.65 and 0.90 (>0.65 is considered very good as reviewed by Moriasi et al (2007)) and R^2 varied between 0.70 and 0.96 (McIsaac et al. 2001; De wit et al. 2003; Li et al. 2014; Han et al. 2009; Chen et al. 2014). Although this LVM lacks the ability to predict seasonal/monthly/daily riverine N export (it is difficult to evaluate watershed N budgets at finer temporal resolutions) compared with mechanistic models, it has the advantage of simplicity and importantly can quantify the contribution of legacy N sources (the lag effect of NANI) to riverine N exports. Other potential watershed attribute variables (e.g., temperature, developed land area, and drained agricultural land area), which can also influence riverine N export through increasing denitrification and wood product export and non-harvested biomass uptake/storage and decreasing N retention capacity (Groffman et al. 2004; Han et al. 2009; Sobota et al. 2009; Howarth et al. 2012; Chen et al. 2012; Kopáček et al. 2013), had no significant impact on the LVM efficiency in the upper Jiaojiang watershed (Table F1 of Electronic supplementary material F). This suggests that the influence of these N removal/storage pathways on riverine N export might be stable and has been averaged and incorporated by fitting coefficients a, b_j, β_0, and λ for the studied watershed. Furthermore, the calibrated posterior export fraction coefficient β_0 that reflects the influence of inherent watershed geological and geomorphologic characteristics was generally steady among the alternative formats of the LVM containing various explanatory variables (Table F1 of Electronic supplementary material F). These findings further support the robustness of the developed LVM.

From the calibrated LVM, estimated average natural background export of 1.84 kg N ha^{-1} $year^{-1}$ (range, 1.20–2.65 kg N ha^{-1} $year^{-1}$) is comparable with estimated results from previous studies (i.e., 0.7–2.8 kg N ha^{-1} $year^{-1}$) (Han et al. 2009; Howarth et al. 2012). Estimated cumulative export fraction of total NANI by the river over the study period was ~14 % (Fig. 4), which falls within the range of previous estimates (10–40 %) for export fractions of multi-year averaged NANI (Han et al. 2009; Howarth et al. 2012; Swaney et al. 2012). The 86 % imbalance between riverine export and NANI is believed to primarily result from denitrification, wood product export and forest biomass uptake/storage, as well as storage in soil, vadose zone, and groundwater (Van Breemen et al. 2002). Field studies conducted in surrounding regions estimated that soil storage and leaching to groundwater could account for ~20 % of the applied chemical and manure N (Yan et al. 2011; Ti and Yan 2013). This is consistent with the 105 kg N ha^{-1} of net available N accumulation (accounted for ~6 % of cumulative NANI over the 1984–2009 period) observed in upper 20-cm layer of agricultural soils between 1984 and 2009 (Fig. 7b) and the 2.7-fold increase of flow-adjusted nitrate concentration (nitrate represented 55 % of measured TN in 1980–2010 on average) from 1980 to 2010 during the baseflow period (70–100 % flow duration interval, when discharge is mainly supplied by groundwater inputs, Fig. 7a) in the studied watershed. Considering the large percentage of woodland (~67 %) and the high atmospheric N deposition rate (Fig. 2c) in the upper Jiaojiang watershed, wood product export, and forest biomass storage may account for a considerable proportion of the N imbalance. Assuming that ~15 % of NANI (or 33 % of atmospheric N deposition) was exported by wood products and forest biomass storage as observed in eastern China (Sheng et al. 2014), denitrification would by difference account for the fate of ~51 % of NANI in the upper Jiaojiang watershed. This denitrification percentage is similar to the sum of agricultural land denitrification (i.e., 36–4 % of total N applied) (Yan et al. 2011; Ti and Yan 2013) and in-stream denitrification (i.e., 10–35 % of total N input to rivers) (Yan et al. 2011; Chen et al. 2012) in the surrounding region. These comparisons further verify the efficacy of the calibrated LVM.

It should be pointed out that the function types used in the LVM, which were supported by the observed relationships between riverine TN flux and each of the relevant influencing factors in this study (Table 2), might be directly applicable to other watersheds with similar characteristics but may require optimization for application in other watersheds. The developed LVM is subject to the uncertainty derived from the assumption that new NANI and legacy N sources experience the same hydrological pathway to rivers. With increasing time, more legacy N might be distributed into subsurface soils, vadose zone, and groundwater, while newly added NANI might be retained in the surface soils and sediments, resulting in a potential difference in hydrological drivers influencing new NANI vs legacy N export to rivers. To improve the efficiency and universality of the model in the future, it may be possible to separate runoff components (e.g., surface runoff, subsurface flow, and groundwater recharge indices or baseflow) as explanatory variables $\theta_{t,\,j}$, considering the differences in transit times among groundwater (decades), surface runoff (days to months), and soil water (months to years) (De wit et al. 2003; Sanford and Pope 2013; Tesoriero et al. 2013). The LVM assumes annual NANI removal via riverine export, denitrification, biomass uptake/storage, and wood product export as a power decay function with time, which might be inefficient in matching the complexities of N transport and transformation processes. It may be possible to refine these functions to better reflect their response to changes in watershed attribute variables. The legacy N originates primarily from non-point sources (Meals et al. 2010); thus the model could be further refined through distinguishing point source vs non-point source model inputs. Due to the limited information available in previous studies, the prior distribution for the three unknown coefficients a, b_j, β_0, and λ was assumed to follow normal distributions for Bayesian calibration, which might not fully reflect the reality and therefore require more quantitative knowledge for future improvement. It is not possible to verify modeled results by direct observations due to the unavailability of reliable and efficient approaches for measuring N delivery processes across various landscapes to rivers, as well as the lag time at the watershed scale. Measurements of ^{18}O, ^{15}N, and ^{3}H isotopes (Sanford and Pope 2013; Tesoriero et al. 2013), which can be used to address the riverine N sources and the ages of groundwater and surface water, are required to verify the modeled results in the future. Extensive information from the literature and direct measurements for the examined watershed, including changes of soil and groundwater N levels, denitrification and forest N uptake efficiencies, can also contribute to indirectly verify various components of the model results.

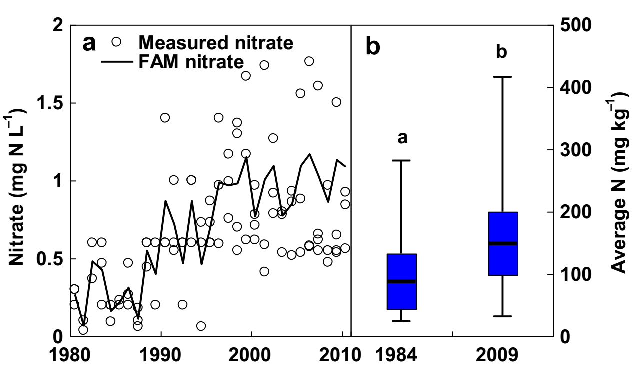

Fig. 7 River nitrate concentration (n=99, *black line* denotes flow-adjusted concentration (*FAM*)) during the low flow regime (70–100th interval) in 1980–2010 (**a**) and soil available nitrogen (n=195) content in the upper 20 cm of agricultural soils between years 1984 and 2009 in the upper Jiaojiang watershed. *Capital letters above bars* denote significant differences (p<0.01)

Cause of the lag effect on riverine N export

The LVM determined a considerable lag effect of NANI on changes in riverine TN flux (Fig. 4a), which is responsible for the contrasting trend between decreasing NANI and increasing riverine TN export in the 2000s in the upper Jiaojiang River watershed (Fig. 2b, c). Long transit times for N passing through the progressively N-saturated landscapes to the river are expected to be a major cause of the temporal lag effect of NANI on riverine export. Annual riverine TN flux was exponentially correlated with NANI, atmospheric deposition, and net food, feed and seed input (Table 1), suggesting that small changes in N input may lead to relatively large changes in riverine N flux, as the biological N assimilation capacity of the watershed becomes progressively more N saturated

(McIsaac et al. 2001; Howarth et al. 2006; Worrall et al. 2009; Swaney et al. 2012). In temperate forest ecosystems, a threshold value of ~10.7 kg N ha^{-1} year^{-1} has been suggested for N saturation, above which significantly higher N export fluxes to rivers were observed (Howarth et al. 2012; Swaney et al. 2012). Similarly, riverine N exports in forest ecosystems increased dramatically and non-linearly as atmospheric deposition exceeded 10 kg N ha^{-1} year^{-1} in forests of southern China (Fang et al. 2009). Observed NANI (38.0–77.6 kg N ha^{-1} year^{-1}) and atmospheric N deposition (17.5–31.3 kg N ha^{-1} year^{-1}) over the 1980–2010 period in the upper Jiaojiang watershed (Fig. 2c) far exceed these thresholds, suggesting progressive N saturation of the terrestrial and aquatic systems. Due to progressive N saturation, a considerable proportion of annual NANI and historical N cannot be retained in the watershed and passes through the soil and groundwater to rivers over long transit times, resulting in the increasing riverine TN export even following a 13 % decrease of NANI in the 2000s (Fig. 2b).

In the upper Jiaojiang watershed, the LVM estimated 2.2–17.5 % of annual NANI was exported by the river in the succeeding 1–30 years (Fig. 4a). This result is consistent with a long-term field study using a ^{15}N tracer that showed 8–12 % of the applied ^{15}N fertilizer was exported to the hydrosphere in the succeeding 30-year period (Sebilo et al. 2013). The majority (~95 %) of riverine export of annual NANI occurred in the succeeding 10 years (Fig. 4a), implying at least an 11-year lag time of NANI (current year plus succeeding 10 years) to riverine export. This estimated N leaching lag time is supported by trends in annual mean TN concentration/flux (Fig. 2b) and flow-adjusted nitrate concentrations during the low flow regime (Fig. 7a). Between 1980 and 1989, despite a 50 % increase of NANI (Fig. 2c), riverine TN concentration/flux and baseflow nitrate concentrations did not increase. Beginning in the early 1990s (~11-year lag), both of these riverine N species experienced increasing concentrations. These increases of TN concentration/flux and nitrate concentrations continued through 2010, in spite of the 13 % decline in NANI that was observed from 2000 to 2010. These temporal riverine N dynamics support the decadal length lag effect between NANI and riverine N export. Comparable results were observed in the Mississippi River watershed, where current-year NANI was estimated to influence riverine nitrate fluxes for 2–9 years (McIsaac et al. 2001). The lag time of N leaching to rivers is dependent on hydrological and biogeochemical processes in the watershed (Hamilton 2012). Stable isotopic tracers (mainly ^{3}H) have shown that delivery times for surface runoff, soil water/shallow groundwater, and groundwater to river systems are on the order of months, years, and decades, respectively (Iqbal 2002; Phillips and Lindsey 2003; Sanford and Pope 2013; Tesoriero et al. 2013). Incorporation of N into soil organic matter and subsequent release of this N for potential leaching to the hydrosphere is also estimated to require several years to decades (Mulvaney et al. 2001; Sebilo et al. 2013). Estimated magnitudes of lag time by the LVM (~11 years) and by the statistical model developed for the Mississippi River watershed (~9 years; McIsaac et al. 2001) are an average of all processes delaying N delivery from the watershed to the river outlet.

Influence of lag effect for watershed nitrogen modeling

Due to the lag effect of N leaching to the river, observed annual riverine TN flux in the upper Jiaojiang River was mainly derived from legacy N sources, i.e., the previous years' NANI (~71.7 %, Fig. 5a). This result is consistent with results observed in field and watershed scale studies where 25–80 % of annual N loss originated from mineralization of soil organic matter (Kopáček et al. 2013; Chen et al. 2014) and nitrate exported from groundwater or baseflow accounted for 30–40 % of riverine nitrate flux (Iqbal 2002; Lindsey et al. 2003; Sanford and Pope 2013). Similarly, the significant increase of nitrate concentration observed from 1980 to 2009 in the studied watershed during the low flow regime (Fig. 7a) and the net available N accumulation observed in upper 20-cm layer of agricultural soils between 1984 and 2009 (Fig. 7b) imply an increasing contribution of legacy N from soil and groundwater to the river N flux. These results stress the need to consider the lag effect in watershed models to better understand and simulate N delivery lag times often observed between N applications to landscapes and riverine export (McIsaac et al. 2001; Meals et al. 2010; Bouraoui and Grizzetti 2014). The greatest contribution (~95 %) to annual riverine TN flux originated from the current year plus previous 10 years' NANI (Fig. 5b), suggesting that the 11-year weighted moving average (Fig. 4a) would be appropriate for N source inputs to lumped models in this watershed. For watershed mechanistic models, an appropriate model calibration should contain at least an 11-year record of continuous monitoring data to demonstrate riverine N flux changes in response to changes in watershed N inputs. These findings are consistent with Howden et al. (2011), who suggested that a monitoring period of ≥12 years is required to fully determine the response of river N fluxes to watershed management measures.

Implications for N pollution control

The developed lagged variable model provides a quantitative method that is sensitive to lag times for controlling aquatic N pollution with a focus on N source reductions without changing current land use at the watershed scale. Estimated annual maximum allowable NANI increased with increasing water yield (R^2=0.72, p<0.01), which results primarily from increasing dilution capacity with increasing river discharge (Chen et al. 2012). To attain the target river TN level

(1.0 mg N L^{-1}), existing NANI over the 1993–2010 period would have required a 22 % reduction to reduce the TN flux by 26 % on average (Fig. 3). This finding is consistent with modeling results for the Mississippi River watershed where a 14.2 % reduction in NANI was predicted to lead to a 33 % reduction in annual riverine nitrate flux (McIsaac et al. 2001). This results from the exponential response in riverine export (Table 1) to lowering the degree of N saturation in the watershed (Howarth et al. 2012; Swaney et al. 2012).

Due to the lag effect, such a predicted disproportionate reduction in riverine TN flux from cutting NANI results from successive NANI reductions over several years (i.e., cumulative effect of previous years' NANI reduction). Export of annual NANI by the river is limited in the current year but will contribute to riverine N fluxes for the succeeding 10 years (Fig. 4a), implying that the effect of NANI reduction can be expected to take at least one decade to fully reach its cumulative effect in terms of riverine TN export. This result is consistent with observed results in other studies, where the reduction in riverine N load following cuts to N inputs required at least one decade to several decades to be fully realized (Sebilo et al. 2013; Sanford and Pope 2013; Bouraoui and Grizzetti 2014). These findings emphasize that increasing trends for riverine N fluxes in N saturated landscapes of the world result from both current and legacy activities over the past decades (Tesoriero et al. 2013). It is thus important to consider such time delay when developing and evaluating aquatic N pollution mitigation or restoration measures. Required NANI reduction was negatively correlated with water yield or river water discharge in 1993–2010 (R^2=0.55, p<0.01), suggesting that NANI reductions would have a more pronounced effect in years with higher water yields due to a higher export fraction.

Considering high chemical N fertilizer application rates to agricultural lands in the upper Jiaojiang watershed (~263 kg N ha^{-1} $year^{-1}$, Fig. 2c), the required reduction in NANI can be most efficiently achieved by reducing nitrogen fertilizer application rates, since N fertilizer-use efficiency is generally very low (~36 %) in this study watershed as well as in surrounding areas (30–40 %, Yan et al. 2011; Ti and Yan 2013). Additional nitrogen fertilizer reductions could be achieved by more efficient recycling of animal and domestic wastes for land application. Considering the increasing net food/feed/seed N input (Fig. 2c), domestic sewage should be increasingly treated by sewage treatment plants to avoid direct inputs of N into rivers. While atmospheric N deposition was also high in this watershed over the 2000–2010 period (~30 kg N ha^{-1} $year^{-1}$, Fig. 2c), its reductions are not easily achieved by local management efforts. Furthermore, given the high contribution of legacy N sources to the riverine TN flux (Fig. 5), alternative measures for intercepting surplus N leaching from landscapes to rivers, including wetlands, riparian buffers, and ecological ditches, would be beneficial for obtaining a more rapid response in riverine TN export reduction (Chen et al. 2012).

Conclusion

This study developed a lagged variable model to quantify the temporally dynamic export of watershed NANI by rivers over time. The model can determine the contribution of NANI from the current year and any previous year (i.e., legacy N sources), as well as natural background sources, to annual riverine TN export. The identified lag time elapsed between watershed N inputs and riverine export offers important knowledge for determining how many years of monitoring data are required for calibrating watershed mechanist models and lumped models. Using this model, it is further possible to estimate the maximum allowable NANI and required NANI reduction necessary for attaining a river TN target level, which provides a quantitative method for guiding watershed N source controls without changing current land use. The model was developed with parsimony of model structure and parameters (only four parameters in this study); thus, it is easy to develop and apply in other watersheds. Application to the upper Jiaojiang watershed demonstrates the efficacy of the model for use by water resource researchers and managers as a simple and effective tool for quantifying the lag effect of watershed N leaching to rivers. In the future, the modeling approach might be improved by adopting various runoff components (surface runoff vs groundwater) as separate explanatory variables, specifying net N inputs for different sources (point vs non-point sources) as model inputs, and individually parameterizing N fates (e.g., denitrification, biomass uptake, and wood export). This study demonstrates the need to consider the lag effect as an improvement to current watershed models and for developing and assessing N pollution control measures.

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