Science of the Total Environment 533 (2015) 196–204

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Reconstructing historical changes in phosphorus inputs to rivers from point and nonpoint sources in a rapidly developing watershed in eastern China, 1980–2010

Dingjiang Chen [a,b,*], Minping Hu [a,c], Yi Guo [a,d], Randy A. Dahlgren [b]

[a] *College of Environmental & Resource Sciences, Zhejiang University, Hangzhou 310058, China*
[b] *Department of Land, Air, and Water Resources, University of California, Davis, CA 95616 USA*
[c] *Zhejiang Provincial Key Laboratory of Subtropical Soil and Plant Nutrition, Zhejiang University, Hangzhou 310058, China*
[d] *Ministry of Education Key Laboratory of Environment Remediation and Ecological Health, Zhejiang University, Hangzhou 310058, China*

HIGHLIGHTS

- Long-term TP input loads from nonpoint and point sources were determined by the LAM.
- The LAM produced river total TP loads were verified by the outputs from the LOADEST.
- In-stream retention/remobilization component of point source P load was estimated.
- Both nonpoint and point source TP input loads increased rapidly over the past 31-yr.
- Nonpoint source input dominated TP load, especially for the summer high-flow period.

ARTICLE INFO

Article history:
Received 14 April 2015
Received in revised form 26 May 2015
Accepted 20 June 2015
Available online xxxx

Editor: Eddy Y. Zeng

Keywords:
Eutrophication
Agriculture
Sewage
Nutrient
Point source
Diffuse (nonpoint) source
Load apportionment model

ABSTRACT

Quantifying point (PS) and nonpoint source (NPS) phosphorus inputs to rivers is critical for developing effective watershed remediation strategies. This study reconstructed PS and NPS total phosphorus (TP) inputs to the Yongan River in eastern China in 1980–2010 using a load apportionment model (LAM) from paired riverine TP concentrations and river discharge records. Based on the fundamental hydrological differences between PS and NPS pollution, the LAM statistically quantified their individual inputs as a power-law function of river discharge. The LAM-estimated monthly/annual riverine TP loads were in good agreement with results derived from a regression model, Load Estimator (LOADEST). The annual TP load increased from 18.4 to 357.0 Mg yr^{-1} between 1980 and 2010. The PS input contributed 7–45% of annual total TP load and increased 23-fold, consistent with a 20-fold increase in flow-adjusted average chloride concentration during the low flow regime (a proxy for wastewater inputs), as well as measured increases in population, poultry, and industrial production. Inferring from observed TP and chloride ratios, as well as total suspended solids (TSS) and river discharge dynamics, temporally retained P load within the river during the low flow regime was estimated to contribute 18–65% of the annual PS input load. NPS inputs consistently dominated the annual riverine TP load (55–93%) and increased 19-fold, consistent with the strong correlation between riverine TP and TSS concentrations, increasing developed land area, improved agricultural drainage systems, and phosphorus accumulation in agricultural soils. Based on our analysis, TP pollution control strategies should be preferentially directed at reductions in NPS loads, especially during summer high-flow periods when the greatest eutrophication risk occurs.

1. Introduction

Enrichment of phosphorus (P) is a major cause of waterbody eutrophication and associated impairments to water quality and aquatic ecosystems (Bowes et al., 2014; Sharpley et al., 2015). To reduce the negative impacts of excess P, watershed managers develop strategies to attain a target P level that can satisfy the designated use of a water body through control of anthropogenic sources (Freedman et al., 2008; Chen et al., 2012; Bowes et al., 2015). Setting P source reduction targets and identifying remedial options rely on appropriate and accurate apportionment of P loads to point (PS) and nonpoint sources (NPS) (Bowes et al., 2009a; Jarvie et al., 2012; Chen et al., 2013).

---

* Corresponding author at: College of Environmental & Resource Sciences, Zhejiang University, 866# YuHangTang Road, Hangzhou, 310058 Zhejiang Province, China.
E-mail address: chendj@zju.edu.cn (D. Chen).

http://dx.doi.org/10.1016/j.scitotenv.2015.06.079

Many numerical models, ranging from simple export coefficient models, to statistical models such as SPARROW, to complex mechanistic models such as AGNPS, HSPF and SWAT (Moriasi et al., 2007; Shrestha et al., 2008; Chen et al., 2013), to Material Flow Analysis (Schaffner et al., 2009), are available for assessing watershed-scale nutrient source apportionment. A major limitation of these watershed mechanistic models as well as Material Flow Analysis method is that they require a large amount of detailed data for calibration of a given watershed making their application challenging for the large number of watersheds requiring assessment (Freedman et al., 2008; Shrestha et al., 2008; Chen et al., 2013). Lumped models, which usually provide a simplified understanding of nutrient sources and transfer dynamics from the watershed to rivers, also require information on primary watershed attributes (e.g., land use, population, and agricultural census data), as well as nutrient discharge from sewage treatment facilities and industries. However, accurate datasets spanning a number of decades are relatively rare for most watersheds, especially in developing countries experiencing the most severe nutrient pollution (Freedman et al., 2008; Bowes et al., 2009a). The source apportionment results determined from current models are also subject to considerable uncertainty since measured riverine P loads are a mixture of P-rich waters having different ages (Sharpley et al., 2013; Jarvie et al., 2013). Very often the lag time (ranging from several years to decades) between legacy P inputs and riverine export is much larger than the temporal extent of available watershed attributes and river monitoring data (Meals et al., 2010; Kleinman et al., 2011). Therefore, these models still encounter several challenges for determining long-term changes in PS and NPS contributions.

Based on the fundamental hydrological differences between nutrient inputs from PS and NPS to rivers, a load apportionment model (LAM) may be developed for statistically quantifying point and nonpoint source nutrient inputs as a power-law function of river discharge (Bowes et al., 2008, 2009a, 2009b, 2010, 2014; Greene et al., 2011; Chen et al., 2013). The LAM does not require detailed watershed attribute information and can potentially be applied to any dataset comprised of paired P concentrations and river discharge measurements. This approach has been successfully applied to a wide range of watersheds in England, Ireland, U.S.A. and China (Bowes et al., 2008, 2009a, 2009b, 2010, 2014; Greene et al., 2011; Jarvie et al., 2012; Chen et al., 2013). In terms of long-term riverine P source apportionment, however, existing LAM applications encounter some important limitations. First, point source P inputs can be temporally retained within the river network via a range of abiotic and biotic processes (e.g., sorption to stream sediments, periphyton uptake) during the low flow regime, with the retained P being subsequently remobilized via erosion of fine bed sediments, advective release of dissolved P from pore waters, or scouring of biologically incorporated P from benthic sources during storm events (Schaffner et al., 2009; Bowes et al., 2010, 2015; Jarvie et al., 2012, 2013; Sharpley et al., 2013; Hayakawa et al., 2015). In conventional applications of LAMs, the remobilization of P from the river channel is incorrectly assigned to the nonpoint source input load, which leads to overestimation of P from NPS and underestimation of PS. A second limitation for conventional LAM application is that they are usually validated from the results of other models, commonly export coefficient models (Bowes et al., 2008, 2009a, 2009b, 2010). However, it remains a challenge to verify LAM results using export coefficient based models for many watersheds since long-term specific P export coefficients are not available or rapidly changing over time. Due to time and economic constraints, it is difficult to capture all wastewater discharges within a watershed, especially in developing countries where limited wastewater collection and treatment is practiced (Chen et al., 2013). This results in difficulty for verifying LAM point source load estimates by direct comparison to wastewater discharge records.

Since the 1980s, many Chinese rivers have experienced a significant increase in nutrient concentrations and loads associated with intensive crop cultivation, livestock production, human population expansion, and urbanization (Liu et al., 2003; Li et al., 2007; Yu et al., 2010). Although eastern China has experienced the most rapid economic development since the 1980s, little information concerning long-term changes in the distribution of PS and NPS nutrient pollution is available for rivers in eastern China. Coastal waters along the East China Sea, as well as many inland lakes and reservoirs, have excessive nutrient inflows from upstream rivers resulting in increased hypoxia/anoxia and risks to drinking water safety from harmful algal blooms (Yang et al., 2013; Huang et al., 2014). Accordingly, it is urgent to establish quantitative knowledge for PS and NPS P inputs to rivers in eastern China over the past several decades to guide effective control strategies for rapidly increasing P pollution.

Using the LAM approach, this study reconstructs the historical (1980–2010) changes in PS and NPS total phosphorus (TP) loads in the Yongan River watershed in eastern China. To deal with the challenges and limitations for conventional LAM applications, this study: (1) addresses the instream retention/remobilization P load from PS during the low flow regime based on the TP:chloride ratio to calibrate the LAM apportioned PS and NPS P inputs; and (2) employs a regression model, Load Estimator (LOADEST, Runkel et al., 2004), that can estimate long-term change in riverine nutrient loads as functions of water discharge and time to verify the LAM results. To conform to the LAM assumption that changes in PS and NPS pollution are primarily regulated by hydrological conditions, the 31-yr record was statistically divided into 5 time periods according to changing trends in flow-adjusted TP and chloride concentrations for separate calibration the LAM. Novel aspects of this study include improvements of the LAM to provide more accurate apportionment of riverine P loads and reconstructing the first historical quantification of PS and NPS pollution for a typical river in eastern China. The results of this study advance our quantitative knowledge of long-term riverine P pollution dynamics and inform P management strategies to effectively assess and control pollution at the watershed scale.

2. Materials and methods

2.1. Watershed description

The Yongan River watershed (120° 13′ 46.065″–121° 0′ 52.464″E and 28° 28′ 10.118″–29° 2′ 22.156″N; elevation ~15–1000 m) is located in the highly developed Taizhou City area of Zhejiang Province, China (Fig. 1). The Yongan River is the third largest river of Zhejiang Province and flows through Xianju County and Linhai City to the Taizhou Estuary and the East China Sea. The sampling location (Fig. 1) for this study was 55 km upstream of Taizhou Estuary at an elevation of ~15 m. 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 $\text{m}^3\ \text{s}^{-1}$ (mean summer vs winter discharge = 122 vs 30 $\text{m}^3\ \text{s}^{-1}$) at the sampling location. The climate is subtropical monsoon having an average annual temperature of 17.4 °C and average annual precipitation of 1400 mm (Supplementary material: Fig. S1a). The rainfall mainly occurs in May–October with a typhoon season occurring in July–September. Total population within the watershed increased from ~590,000 in 1980 to ~740,000 in 2010 (Supplementary material: Fig. S1b), with ~67% in the Xianju County and 33% in the Linhai City. Over the 31-year study period, domestic livestock production (pig, cow, sheep and rabbit) decreased by ~25% while poultry production (chicken and duck) increased by 4.8-fold. Agricultural land (including paddy field, garden plot and dryland) averaged ~12% of total watershed area in 1980–2010, with developed land (including rural and urban residential lands, roads, and mining and industry lands) and natural land (woodland, water surface, swamp, rock, and natural reservation land) contributing ~3% and ~85%, respectively (Supplementary material: Fig. S1c). The economic role of agriculture has been increasingly replaced by industry (with a 200-fold increase in industrial gross domestic production, Fig. S1b), resulting in a remarkable reduction (~37%) in chemical P fertilizer application since 2000. The agricultural land area with