



# A modified load apportionment model for identifying point and diffuse source nutrient inputs to rivers from stream monitoring data



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## SUMMARY

Determining point (PS) and diffuse source (DS) nutrient inputs to rivers is essential for assessing and developing mitigation strategies to reduce excessive nutrient loads that induce eutrophication. However, application of watershed mechanistic models to assess nutrient inputs is limited by large data requirements and intensive model calibration efforts. Simple export coefficient models and statistical models also require extensive primary watershed attribute information and further they cannot address seasonal patterns of nutrient delivery. In practice, monitoring efforts to identify all PSs within a watershed are very difficult due to time and economic limitations. To overcome these issues, based on the fundamental hydrological differences between PS and DS pollution, a modified load apportionment model (LAM) was developed relating the river nutrient load to nutrient inputs from PS, DS and upstream inflow sources while adjusting for in-stream nutrient retention processes. Estimates of PS and DS inputs can be easily achieved through Bayesian calibration of the five model parameters from commonly available stream monitoring data. It considers in-stream nutrient retention processes, temporal changes of PS and DS inputs, and nutrient contributions from upstream inflow waters, as well as the uncertainty associated with load estimations. The efficacy of this modified LAM was demonstrated for total nitrogen (TN) source apportionment using a 6-year record of monthly water quality data for the ChangLe River in eastern China. Aimed at attaining the targeted river TN concentration ( $2 \text{ mg L}^{-1}$ ), required input load reductions for PS, DS and upstream inflow were estimated. This modified LAM is applicable for both district-based and catchment-based water quality management strategies with limited data requirements, providing a simple, effective and economical tool for apportioning PS and DS nutrient inputs to rivers.

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## 1. Introduction

Excessive nutrients (e.g., nitrogen and phosphorus) in rivers is of increasing concern worldwide (Pieterse et al., 2003; Edwards and Withers, 2008; Howden et al., 2011), as it not only degrades riverine ecosystems and decreases the quality of water used for drinking, industry, agriculture, recreation, and other purposes (Bowes et al., 2010; Houser and Richardson, 2010), but also is a contributor to eutrophication and hypoxia in downstream lakes, estuaries and coastal waters (Diaz and Rosenberg, 2008; Gao and Zhang, 2010; Trevisan et al., 2012). To reduce excessive nutrient loads carried by rivers in an efficient and cost-effective manner, assessing nutrient input loads from point (PS) and diffuse sources (DSs) is required for developing watershed management and

control strategies, such as the Total Maximum Daily Load (TMDL) program (Freedman et al., 2008; Bowes et al., 2009; Chen et al., 2012).

Many numerical models, ranging from simple export coefficient models (Johnes, 1996), to statistical models such as SPARROW (Smith et al., 1997), to complex mechanistic models such as AGNPS, HSPF and SWAT (Borah and Bera, 2004), have been developed for assessing watershed-scale nutrient fate and transport and nutrient source apportionment. A major limitation of these watershed mechanistic models is that they require a large amount of data for calibration for a given watershed making their application difficult for the large number of watersheds requiring assessment (Shrestha et al., 2008; Shen and Zhao, 2010; Chen et al., 2012). For example, most states in the USA lack sufficient data to quantify DS loads, with no estimates of DS loads for 20% of watersheds undergoing TMDL development (Freedman et al., 2008). Similarly, export coefficient and statistical models require considerable information on primary watershed attributes (such as land-use,

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population, agricultural census data), and knowledge of nutrient discharge from PSs (e.g., sewage treatment works and industrial discharge) (Bowes et al., 2009). Although determining nutrient loads discharged from the PSs is relatively easy in concept, it is still difficult to capture all wastewater discharge within a watershed or a district in practice due to time and economic limitations, especially in developing countries with limited PS disposal regulations. Another important consideration is that export coefficient and statistical models usually operate on an annual time step, so they cannot easily be used to infer seasonal or storm-event patterns of nutrient delivery. Such temporal resolution is required to determine nutrient sources and loads during the most sensitive times of the year (e.g., typically the summer growing season) when eutrophication is most likely to occur in downstream water bodies (May et al., 2001; Bowes et al., 2008, 2009). Therefore a robust method is required for PS and DS nutrient source apportionment with high temporal resolution and limited data requirements.

Recently, the load apportionment model (LAM), which statistically quantifies the PS and DS nutrient inputs as a power-law function of the river discharge, has been proposed and successfully applied to a range of catchments of varying sizes, geologies and land uses (Bowes et al., 2008, 2009, 2010). LAM is based on the fundamental hydrological differences in the characteristics of nutrient inputs from PS and DS types. Point source nutrient input to the river is relatively constant and hydrologically independent. In contrast, DS nutrient inputs have a strong hydrologic dependence (Edwards and Withers, 2008). The LAM approach provides a simple and efficient tool for nutrient source apportionment with high temporal resolution based on routine stream monitoring data. With increasing concern for environmental water issues, many local authorities and states have carried out routine river monitoring programs (e.g., weekly, fortnightly, monthly, seasonally) to support water quality assessment and management plans (Bowes et al., 2008; Shrestha et al., 2008). These data sets, in conjunction with accompanying stream flow data, provide a fundamental data required to develop a LAM for a given watershed.

Existing LAMs still contain several important limitations. First, they assume that nutrients are relatively conservative during transport, rendering them unsuitable for rivers with high in-stream nutrient retention efficiency (Bowes et al., 2009). In-stream nutrient retention, which often accounts for an important fraction (1–80% for nitrogen and 20–70% for phosphorus) of the annual total nutrient load (Haag and Kaupenjohann, 2001; Grizzetti et al., 2005; Dierk and Michael, 2008; Chen et al., 2010), is significantly modified by river hydrological, morphological and ecological conditions (Alexander et al., 2000; Pieterse et al., 2003; Trevisan et al., 2012). Second, conventional LAMs usually assume that the PS inputs are constant throughout the year or a study period, which is generally true, but it may be not the case for some regions. For example, it is frequently observed that enterprises discharge sewage without permission or in excess of their discharge limits to maximize their profits in many regions of China (Sun and Yang, 2006; Qian et al., 2007), which could introduce a considerable error in LAM results. Third, nutrient inflows from upstream water bodies (representing the contribution from the upstream regions) are not addressed for a river-reach segment in LAM models; thus they do not satisfy requirements for district-based environmental water management. For example, in China a river is usually divided into several segments that are regulated by corresponding districts (Shang et al., 2012); thus the reach-end corresponding to the boundary between districts is commonly used as the compliance location for water quality management and regulation (Chen et al., 2009; Shang et al., 2012). Fourth, conventional LAMs often adopt a trial-and-error procedure for calibrating the model parameters, which is subjective and uncertain (Shen et al., 2006), considering the uncertainties involved in the observed data, model

parameters and model structure (Howden et al., 2011; Chen et al., 2012). Uncertainty is an important issue that requires addressing in water quality model development and application (NRC, 2001). To fully exploit available monitoring data for supporting water quality management, there is an excellent opportunity to modify conventional LAMs to address these identified limitations.

This study aimed to modify conventional LAMs to make them more robust and less sensitive to the previously mentioned model limitations (e.g., in-stream nutrient retention, temporal changes in PS discharge, boundary conditions, and uncertainty involved in calibrating model parameters). The modified LAM statistically relates the river nutrient load to nutrient inputs from PS, DS and upstream inflow sources while adjusting for in-stream nutrient retention processes. Beyond the commonly used trial-and-error procedure for calibration, the Bayesian statistical method coupled with the Markov Chain Monte Carlo (MCMC) algorithm, which optimally utilizes information from both prior knowledge and observed data (Shen and Zhao, 2010; Chen et al., 2012), was adopted for calibrating the model parameters and addressing the uncertainty associated with input load estimations. The efficacy of the modified model was demonstrated through application for total nitrogen source apportionment in the ChangLe River in eastern China using a 6-year record of monthly water quality data. The modified LAM was aimed at attaining the target total nitrogen concentration ( $2 \text{ mg L}^{-1}$ ) and at determining the required input load reductions for PS, DS and upstream inflow nutrient loads. This modified model adopts the merits but overcomes the limitations mentioned above for conventional LAMs and statistical models. It has limited data requirements and provides researchers and managers with a simple, effective and economical tool for apportioning PS and DS nutrient inputs to rivers.

## 2. Materials and methods

### 2.1. Study area

The ChangLe River watershed ( $120^{\circ}35'56''$ – $120^{\circ}49'03''$ E and  $29^{\circ}27'98''$ – $29^{\circ}35'12''$ N) is located in Zhejiang Province, eastern China (Fig. 1). The ChangLe River is one of the main tributaries of the Cao-E River, which ultimately flows into the Qiantang Estuary and East China Sea. The river system drains a total area of  $864 \text{ km}^2$  and flows about 70.5 km with a 0.36% gradient and a 40–70 m width. The portion of the watershed examined in this study contained two sub-catchments: Sub-catchment I corresponding to the reach from S1 (NanShan Reservoir) to S2 (midstream site) and sub-catchment II corresponding to the reach from S2 to S3 (downstream boundary or watershed outlet) (Fig. 1 and Table 1). The area represents a typical agricultural watershed in eastern China and is characterized by a subtropical monsoon climate. Long-term average annual rainfall is 1256 mm with more than 65% of rainfall usually occurring between May and September. The primary land-use categories are woodland and farmland (including paddy fields, uplands, and garden plots) (Table 1). Water input from NanShan Reservoir (S1) accounts for  $8 \pm 5\%$  of the annual cumulative discharge at S3 due to export for drinking water. Therefore catchment runoff from below S1 is the main water source ( $92 \pm 7\%$ ) at the watershed outlet.

### 2.2. Basic data collection

Total nitrogen (TN) concentrations at three sampling sites (S1–S3) along the ChangLe River were monitored monthly from January 2004 to December 2009 ( $n = 72$  samples per site) (Fig. 1). Water samples for chemical analysis were collected between 9 am and 2 pm in 2.5 L polyethylene bottles from 30 cm below the

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