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Research papers

Long-term agricultural non-point source pollution loading dynamics and correlation with outlet sediment geochemistry

Wei Ouyang [a,*], Wei Jiao [a], Xiaoming Li [a], Elisa Giubilato [b], Andrea Critto [b]

[a] State Key Laboratory of Water Environment Simulation, School of Environment, Beijing Normal University, Beijing 100875, China
[b] Department of Environmental Sciences, Informatics and Statistics, Ca' Foscari University of Venice, Venice 30123, Italy

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ABSTRACT

Some agricultural non-point source (NPS) pollutants accumulate in sediments in the outlet sections of watersheds. It is crucial to evaluate the historical interactions between sediment properties and watershed NPS loading. Therefore, a sediment core from the outlet of an agricultural watershed was collected. The core age was dated using the ^{210}Pb method, and sedimentation rates were determined using the constant rate of supply (CRS) model. The total nitrogen (TN), total phosphorus (TP), Cd, Pb, Cu, Ni and Cr accumulations in the sediment generally showed fluctuating increases, with the highest sedimentation fluxes all occurring in approximately 1998. The measurement of specific mass sedimentation rates reflected a record of watershed soil erosion dynamics. Using SWAT (Soil and Water Assessment Tool) to simulate long-term watershed agricultural NPS pollution loadings, the historical interactions between sediment properties and NPS loadings were further evaluated. The N leaching process weakened these interactions, but the historical accumulations of TP and heavy metals in sediments generally correlated well with watershed NPS TP loading. The regression analysis suggested that Pb and Cr were the most suitable indexes for assessing long-term NPS TN and TP pollution, respectively. Assessing the NPS loading dynamics using the vertical characteristics of sediment geochemistry is a new method.

1. Introduction

Sedimentation processes usually occur during the transport of various pollutants into surface waters (Ansari et al., 2000). Consequently, sediment geochemistry is considered a useful indicator of environmental changes and anthropogenic impacts (Chatterjee et al., 2007; Nath et al., 2000). It has been widely recognized that intensive agricultural development can increase the watershed soil erosion load and release more associated non-point source (NPS) pollutants, such as nitrogen, phosphorus and heavy metals (Jain, 2002; Jiao et al., 2014). Some NPS nitrogen (N) and phosphorus (P) discharged by upland processes accumulate in river sediments. Although some fine particles are lost during the sedimentation process, it is hypothesized that some characteristics of the vertical sediment geochemistry are correlated with the NPS loading history (Migani et al., 2015). Research regarding the sediment property responses to long-term agricultural NPS pollution dynamics remains scarce.

Over the past few decades, the study of sediment cores has proven to be an excellent approach for establishing the effects of anthropogenic and natural processes on the sedimentary environment (Shotyk, 2002). Vertical profiles of pollutant species in sediment cores are commonly used as "historical pollution records" of whole watersheds (Harikumar and Nasir, 2010). To inversely identify the pollution history, it is essential to estimate the sedimentation rates and sediment ages. The results also provide valuable information regarding the soil erosion dynamics of a watershed (Mabit et al., 2014), which are directly related to agricultural NPS pollution. In this context, the application of radiometric methods to sedimentary chronology has developed rapidly and with considerable success (Kumara et al., 1999). In particular, the half-life of ^{210}Pb (22.3 years) makes it an ideal radioisotope for dating sediments from the past 100–150 years. To date, this method has been used extensively in different sedimentary environments, including wetlands, lakes, reservoirs, flood plains, estuaries and coastal areas (Mabit et al., 2014).

Effective control of NPS pollution in agricultural watersheds is required to meet the high standards of water management. Consequently, studies seeking a better understanding of watershed management have expressed increasing concern regarding the quantification of NPS pollutant loading (Dechmi and Skhiri, 2013; Heathwaite et al., 2005). Thus, a number of water quality models at the watershed scale have been developed and applied. Among

* Corresponding author.
E-mail address: wei@itc.nl (W. Ouyang).

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these models, the Soil and Water Assessment Tool (SWAT) is frequently used to assess NPS pollution over long timescales and at diverse spatial scales in agricultural watersheds (Laurent and Ruelland, 2011; Ouyang et al., 2010). However, the modeling methods are usually quite time consuming due to the input database import and parameter validation processes (Choi and Blood, 1999). Using sediment geochemistry to indicate agricultural NPS pollution is a potential way to provide the knowledge needed to support routine management, especially in data-sparse or ungauged watersheds.

Watershed sediment analyses are widely accepted indicators of environmental water quality (Somura et al., 2012). Based on the hypothesis of the interaction between sediment and NPS pollution loading, we present a new approach to validate sediment geochemistry indicators that can function on long-time scales. The interaction principle was achieved by integrating SWAT modeling and a ^{210}Pb-dated sediment analysis. The primary objectives of this study were as follows: (1) to analyze and date the total nitrogen (TN), total phosphorus (TP), Cd, Pb, Cu, Ni and Cr accumulations in sediment at a watershed outlet; (2); to identify the long-term NPS nitrogen and phosphorus loading in an agricultural watershed; and (3) to evaluate the historical interactions between sediment properties and watershed NPS TN and TP loading, thus, identifying the proper indicators.

2. Materials and methods

2.1. Study area description

The study area is located on the Sanjiang Plain, Northeast China, encompassing a total watershed area of 24,863 km^2 (Fig. 1). In addition to intensive regional agricultural development, approximately half of the natural wetlands, forests and grasslands have been reclaimed and converted into paddy land and upland since the 1950s. Rice and maize are the two main types of crops being cultivated. This watershed has a frigid, temperate, continental monsoon climate, with an average annual temperature of 1.91 °C. The mean annual precipitation is approximately 600 mm, most of which falls between May and September (Ouyang et al., 2014). The local river, with pH values ranging from approximately 0.5 to 7.1, is characterized by a seasonal hydrological regime, generally flowing from southwest to northeast.

2.2. Watershed NPS total nitrogen and phosphorus loading simulations

The SWAT model was applied to estimate watershed NPS TN and TP loads from 1977 to 2013. NPS nitrogen (N) was simulated in ammonia, organic and nitrate forms, and their sum represented the total N (TN). NPS phosphorus (P) was modeled in organic, soluble and sediment forms, and their sum represented the total P (TP). The SWAT databases were prepared and imported; including topography (1:250,000); land cover in 1979, 1992, 1999 and 2009 (1:1,000,000); climate information; and soil properties (1:1,000,000) (Fig. 1). The climatic features include daily historical monitoring data (minimum and maximum temperature, wind speed, precipitation and solar radiation) obtained from three local weather stations between 1973 and 2015. After field investigations, the agricultural practices of paddy rice and maize over three decades were taken into account to improve the modeling efficiency (Ouyang et al., 2013).

After a sensitivity analysis using the SWAT-CUP tool, the SWAT model was calibrated with monitoring data obtained in the first twenty-four months and later validated with data from an additional two years. The model was validated based on the order of streamflow and soil concentrations (Fig. 2). Using the streamflow and sediment yield routing and monitoring data, the sixteen dominant parameters (CN_2.mgt, SMTMP.bsn, ALPHA_BF.gw, TIMP.bsn, GW_DELAY.gw, CMN.bsn, CH_N_2.rte, NPERCO.bsn, CH_K_2.rte, Spexp.bsn, SOL_AWC(1-2).sol, SPCON.bsn, SOL_BD(1).sol, RSDCO.bsn, CANMX.hru, AI1.wwq, ESCO.hru, BC1.swq, GWQMN.gw, BC2.swq and BC3.swq) that affect these two indicators were validated (Fig. 3). The modeling performances of the streamflow and sediment were evaluated using the coefficient of determination (R^2) and the Nash-Sutcliffe efficiency (E_{NS}), which was larger than 0.698. The watershed NPS N and P loadings were validated with the defined parameter values due to the lack of stable nutrient concentration monitoring data. The validated parameters associated with TN and TP (Cmn, Psp, Pperco, Phoskd, BC1, BC2, BC3, BC4, AI1, AI2 and Rsdco) were predefined according to data in a similar watershed. Details of the SWAT validation process in this watershed can be found in our earlier paper (Ouyang et al., 2014). Linear

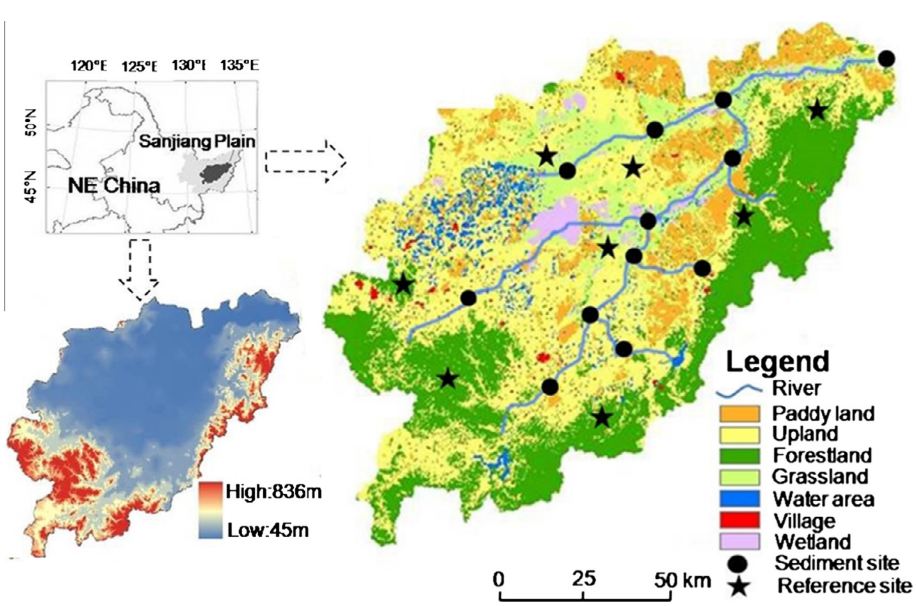

Fig. 1. Location of study area showing topography, land uses and sampling sites.