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Using river sediments to analyze the driving force difference for non-point source pollution dynamics between two scales of watersheds

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ABSTRACT

The formation and transportation processes of non-point source (NPS) pollution varied among the studied watersheds in the Northeastern China, so we hypothesized that the driving force behind NPS pollution followed the spatial scale effect. With a watershed outlet sedimentary flux analysis and a distributed NPS pollution loading model, we investigated the temporal dynamics of NPS and the differences in driving forces. Sediment core samples were collected from two adjacent watersheds, the smaller Abujiao watershed and the larger Naoli watershed. The natural climatic conditions, long-term variations in the distribution of land use, soil properties and tillage practices were the same in the two watersheds. The vertical distributions of total nitrogen, total phosphorus, Zn and As at 1-cm intervals in the section showed clear differences between the watersheds. There were higher concentrations of total nitrogen and total phosphorus in the larger watershed, but the heavy metals were more concentrated in the smaller watershed. Lead-210 (^{210}Pb) analyses and the constant rate of supply model provided a dated sedimentary flux, which was correlated with the corresponding yearly loading of NPS total nitrogen and total phosphorus in the two watersheds. The total phosphorus showed a stable relationship in both watersheds with an R^2 value that ranged from 0.503 to 0.682. A rose figure comparison also demonstrated that the pollutant flux in the sediment was very different in the two watersheds, which had similar territorial conditions and different hydrological patterns. Redundancy analysis further indicated that expanding paddy areas had a large impact on the sedimentary flux of nitrogen and phosphorus in the smaller watershed, but precipitation had a direct impact on NPS loading in the larger watershed. We concluded that the spatial scale effect affected the NPS pollution via the transport processes in the waterway, which was mainly influenced by branch length and drainage density.

1. Introduction

Watershed terrestrial non-point source (NPS) pollution and transportation in the aquatic system are the main threats to water quality (Cho et al., 2016; Perakis and Hedin, 2002). NPS pollution is affected by diverse factors, which may include land use type and distribution, rainfall patterns, agricultural tillage practices and hydrological response features (Azzellino et al., 2006; Ouyang et al., 2010). The individual or combined impacts of these factors on NPS pollution dynamics have been studied in many watersheds (Bois et al., 2013). However, the driving forces of these factors are not same or stable across different sizes and types of watersheds; furthermore, the interactions of these factors can vary temporally within the same watershed (Kelsey et al., 2008; Liu et al., 2015). Therefore, it is interesting to distinguish the interactions of these diverse factors and their influence on NPS pollution in watersheds with different spatial scales (Vitro et al., 2017). The integrated application of the watershed NPS pollution model and chronological sedimentary patterns is an innovative method that can provide the direct evidence for this hypothesis.

Many watersheds in the world suffer from NPS pollution, especially in watersheds where pollution from industrial or municipal point sources have been well-managed (Culbertson et al., 2016). In rural watersheds agricultural NPS runoff

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predominates including significant runoff of nutrients, microbes and pesticides where as in urban watersheds petroleum hydrocarbons, trace metals, pesticides, microbes and nutrients generally predominate, which is the most challenging issue for watershed water quality around the world (Wilson and Xenopoulos, 2009). The contribution of agricultural NPS pollution to watershed water pollution varies from 40% to 60% in China, and there has been a similar situation in the U. S. in recent decades (Dubrovsky et al., 2010; Ouyang et al., 2017a). The booming pressure caused by increases in NPS pollution increases the standard of related regulations and prompts improvements in the research level in this field. As a result, there has been widespread implementation of Best Management Practices (BMPs) to prevent loading (Giri et al., 2012). Effective watershed pollution control requires the identification of NPS pollution processes and procedures or methods to control such pollution to a practical extent. Furthermore, diverse principles and methods have been developed and applied for NPS pollution source identification, process control and risk assessment (Wei et al., 2016).

Many distributed hydrological models have been developed to describe NPS pollution, these models can assess the influence of interactions between potential factors that control NPS pollution loading and performances of selected BMPs in watersheds of varying scales (Gebremariam et al., 2014; Ouyang et al., 2013). The Soil and Water Assessment Tool (SWAT) is an effective hydrological model that has been used for evaluating the interaction of different types of NPS pollution factors with nitrogen or phosphorus dynamics under an extensive range of environmental conditions for watersheds (Sommerlot et al., 2016; Woznicki et al., 2016). This model system contributes to the understanding the complicated natural impacts and/or anthropogenic activities on the formation and transportation of NPS pollution. Hydrological processes of the SWAT model can simulate evapotranspiration (ET), lateral flow, surface run-off, percolation and watercourse transmission losses (Kim et al., 2017). As a result, soil erosion and nutrient pollution transport for watersheds can be simulated when territorial and hydrological patterns are considered (Furl et al., 2015; Nasta et al., 2017).

NPS loading of sediment at a watershed outlet can record pollution transportation dynamics, which is also the fingerprint for the watershed pollution process (Wilkinson et al., 2014). Quantification of sediment flux can characterize the water pollution transport processes, temporal patterns and the main driving forces (Jiao et al., 2014; Teshager et al., 2017). Estimates of sediment age, deposition rates and deposition flux are of great importance in revealing environmental change and quantifying the relationship between watershed erosion and deposition in the river course (Gevao et al., 2016). Modern sediment dating and calculations of deposition rate comprehensively reflect the deposition process and help determine the quantitative indicators of depositional history (Mabit et al., 2014). ^{210}Pb, ^{137}Cs, and other short half-life radioisotopes, radionuclide tracing and dating have become a significant means to study pollution depositional trends (Kirchner, 2011), which also help to identify the vertical distributions of pollution flux and influential factor identification over different time scales using multiple radioactive isotopes (Caillet et al., 2001; Hancock et al., 2014).

Therefore, understanding long-term NPS pollution dynamics and responses to human disturbances and/or natural characteristics of the watershed are very valuable for optimizing watershed water pollution control, planning, management and policy, particularly in watersheds with expanding agricultural and urban development (Atkinson et al., 2009; Ercan and Goodall, 2016). Given this foundation, the goal of this study is to distinguish the interactions between different impact factors of NPS pollution using modeling and sedimentary methods. We investigated watersheds of two scales. The specific objectives of this study were as follows: (1) to explicitly identify the vertical distribution of the fluxes of four pollutants between two watersheds with different spatial scales; (2) to quantify the correlation differences of watershed NPS pollution loads with the dated flux of TN and TP in sediment with the same temporal patterns; and (3) to compare the different driving forces and spatial scale effects on NPS pollution loading and sediment flux between watersheds with rose figures and RDA analyses.

2. Materials and methods

2.1. Study areas

To test our hypothesis, two adjacent watersheds in an agricultural development area in the Heilongjiang Province, Northeast China were selected as a study area (Fig. 1). The regional climate is a continental monsoon climate in a cold temperate zone. The annual average rainfall is 583.18 mm, and the rainfall is concentrated in May through September (Ouyang et al., 2012). The smaller watershed, the Abujiao (ABJ) watershed, has a drainage area of 141.5 km^2. The elevation of the watershed decreases from west to east. The larger Naoli (NL) watershed is located in southern part of China. These two adjacent agricultural watersheds have the same natural climatic conditions, tillage practices, population density and agricultural development.

2.2. Sediment core collection and analysis

Sediment cores were collected from the estuary of the Abujiao (C1) and Naoli watersheds (C2) in July 2015 (Fig. 1). Sediment cores were collected with a columnar sampler (PVC tube, 7.5 cm in diameter) and were then sliced into disks at a vertical interval of 1 cm. Samples were transferred to polyethylene bags and were freeze-dried, slightly crushed, passed through a 0.147-mm sieve and stored in sealed bags. Due to sedimentary features, the C1 core produced 24 slices and the C2 core produced 30 slices (Ouyang et al., 2017b).

To highlight the pollution sedimentary dynamics, the concentrations of total nitrogen (TN), total phosphorus (TP), Zn and As were analyzed for each slice. TN was measured using the semimicro Kjeldahl method and TP was analyzed using the NaOH melting–Mo and Sb colorimetry method. The two heavy metals were measured using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) with the detection limit of 0.06 mg/kg (IRIS Intrepid II XSP, Thermo Electron, USA) (Lintern et al., 2016). The dry sediment samples were digested with HNO3–HF–HClO4 mixture. The standard reference material GBW-07402 was used simultaneously for analytical quality assessment and the obtained average recoveries ranging from 96.34% to 101.47% (Jiao et al., 2014).

2.3. Sediment core dating method

To determine temporal trends of sedimentary processes, Lead-210 (^{210}Pb) was used for sediment flux dating (Townsend and Seen, 2012). ^{210}Pb$_{tot}$ and ^{226}Ra activity were determined to calculate the activity of ^{210}Pb$_{ex}$ (^{210}Pb$_{ex}$ = ^{210}Pb$_{tot}$-^{226}Ra). Samples were stored in a sealed container for a month for stabilization before measurement. A low-background HPGe (High Purity Germanium) γ spectrometer was used for measurements. ^{210}Pb$_{tot}$ activity was determined using a 46.5-keV γ-ray and ^{226}Ra activity, in equilibrium with the ^{210}Pb$_{su}$ activity, which was determined using 95.2 keV and 351.9 keV γ-rays. The constant rate of supply (CRS) model assumes that the fallout of ^{210}Pb from the atmosphere to the