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Exploring Dynamics of Riverine Phosphorus Exports under Future Climate Change using a Process-based Catchment Model

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Abstract

A quantitative understanding of riverine phosphorus (P) export in response to shifts in anthropogenic inputs, terrestrial retention, and climate is important for developing mitigation measures at a watershed scale. In this study, we simulated a decadal change in the riverine P export in a human-dominated watershed from a cold climatic region located in Northeast Asia. A process-based catchment model nested within a delicate land P module was applied to simulate the dynamics of P retentions and its exports in the watershed. We found that the terrestrial P retention capacities declined for 2008–2017, and the decline rates would accelerate under three representative concentration pathways (i.e. RCP2.6, RCP4.5 and RCP8.5). The P released from the diffused source and historical legacy could partly offset the effort through point-source P reduction by the improved wastewater treatments. Climate changes (e.g., duration and frequency of extreme rainfall event) could accelerate P deliveries from the P legacy retained in soils. We suggest that a long-term watershed P management strategy should be targeted to reduce historical P legacy input into river rather than solely focusing on the short-term changes in the riverine P concentration.

Keywords

Anthropogenic P input; terrestrial retention; process-based catchment model; river P loading; climate change; cold climate regions

1. Introduction

Anthropogenic activities have substantially altered phosphorus (P) cycling on the lands (Elser and Bennett, 2011; Powers et al., 2016; Tong et al., 2017a). Agricultural

P-fertilizer application, domestic wastewater discharge, and the other human activities have released substantial P amounts into the watershed, which subsequently aggravate P loadings into river and lake (Bussi et al., 2016; Liu et al., 2018a; Tong et al., 2017b). Of late, freshwater systems are constantly under stressors of changing nutrient cycles (Best, 2019). The increasing riverine P loading due to anthropogenic inputs has gained wide attention globally (Han et al., 2011; Hu et al., 2020; Tong et al., 2017b; Zhang et al., 2017). In the stream, a total P (TP) of 76 $\mu g \cdot L^{-1}$ or above could represent a highly productive status (Dodds et al., 1998). Stackpoole et al., (2019) has investigated 173 watersheds in the United States of America (USA), and found that over half of them had TP concentration exceeding this threshold. During 2000–2014, a continental-scale increase in the stream P was observed in relatively undisturbed catchment in the USA, and the underlying mechanism has not been clearly understood (Stoddard et al., 2016). High P levels could result in the deterioration of water quality by eutrophication, algal blooms, coastal hypoxia and dead zones (Breitburg et al., 2018; Damania et al., 2019; Griffith and Gobler, 2020).

Understanding the long-term dynamics of P input and output balance at a watershed scale is an essential step in regulating the human activity and improving water quality (Stoddard et al. 2016; Chen et al., 2018; Goyette et al., 2018). Increasing evidence has suggested that watershed has capacity to modulate downstream P exports by retaining the excess P on lands (Gomez-Velez et al., 2015; Powers et al., 2016). As a lithophile element, P is relatively immobile after being adsorbed by Al, Fe, or Ca (Grenon et al., 2021). Land P retention may regulate its delivery rate into water and creates time lags between the human P input and its concentration in water (Kusmer et al., 2019). In the short term, P retention may reduce its input into rivers and lakes (Goyette et al., 2018). In the long term, P accumulated in the soil over time could create a large P legacy that would act as a chronic source for the downstream regions even after the P inputs are reduced (Haygarth et al., 2014; Macintosh et al., 2018; Withers et al., 2017). However, once the accumulated P exceeds the soil storage capacities, the soil to water transport would accelerate substantially as revealed in a study in North America (Goyette et al., 2018). Simulating dynamics of soil P retention is essential to improve the watershed scale P management that is mainly achieved by regression analysis between long-term P inputs and river export (e.g. the net anthropogenic P input (NAPI) method, Hu et al., 2020; Metson et al., 2017) or simplified soil-P process modelling (e.g. dynamic P pool simulator) (Mogollón et al., 2018; Sattari et al., 2012).

Climate change is reshaping nutrient deliveries from soil to water due to changes in hydrological process (Kalcic et al., 2019; Madakumbura et al., 2021; Ockenden et al., 2017). Since the 1900s, surface air temperature on Earth has increased by 0.9 °C, with five of the warmest years occurring after 2010 (UNESCO, 2020). Even small changes in air temperatures could have large impacts on the rainfall events and river runoffs (Pachauri et al., 2014). Intense rainfalls could intensify the mobilisation of nutrients retained in soil and increase input into the receiving waters body through runoff (Fang et al., 2020; Mukundan et al., 2020). Subsequent shift in hydrology (e.g. water volume, river flow rate) might impact the in-stream retentions and delivery along the upstream to downstream continuum (Doyle et al., 2003; Zhang et al., 2017). For instance, shifts in the magnitude and frequency of extreme high flows and floods could reduce river P retention capacity and in-channel loss under eutrophic conditions (Withers and Jarvie, 2008). Crossman et al. (2013) revealed that shifts in rainfall and air temperature could result in an increase of up to 30% in the P loadings into Lake Simcoe which offset the P reduced through improved agricultural practices. Bosch et al. (2014) revealed that with climate change, there would be a 17% increase in the runoff and a 23% increase in nutrient yield in the Lake Erie basin in the USA.

To the best of our knowledge, a detailed description of the processes related to the anthropogenic P inputs into the watershed, terrestrial P retentions and climate changes from a holistic perspective is lacking. We attempted to explore their dynamics using a process-based catchment P model with a delicate soil module in this study. A human dominated watershed in a cold climatic region in Northeast Asia is illustrated. From a methodological viewpoint, this application attempted a potential solution to simulate the dynamics of soil P retentions and riverine export using a process-based catchment model. From a water management perspective, we suggest that a long-term watershed P management plan should be targeted for reducing the historical P legacy inputs into the rivers, rather than solely focusing on short-term changes in river P concentrations and loading.

2. Materials and methods

2.1 Study area and riverine P monitoring

The Songhua River (SR) is one of the largest rivers in Northeast Asia, with a length of 1,927 km and a streamflow of 7.6×10^{10} m^3 per year (Wang et al., 2015). The catchment of the Songhua River Basin (SRB, E: 119°52’–132°31’, N: 41°42’–51°38’, Figure 1) has an area of 5.6×10^5 km^2 and a population of approximately 54 million (Williams et al., 2015). It is a foothills-fed river, and the catchment receives water mainly from summer monsoon with a certain amount of snowfall in the high elevation streams (Li et al., 2014). The watershed has a large difference in the topography and is surrounded by the highly elevated mountain regions (above 1000 m, Figure S1). The main stream is divided into three sections: an upper section from the head stream to Songyuan, a middle section from the Songyuan to Tonghe, and a lower section from Tonghe to Tongjiang (Figure 1). The major tributaries include the Nenjiang, Mudan, Hulan, Lalin, and Mayi rivers (Figure S1). According to the locations of the tributaries and the elevations of the basin, a total of 10 sub-basins were delineated, ranging from forests to crop lands, and including cities such as Harbin and Jilin (Figures S1–S2 and Table S3). SRB has varied climatology, mainly due to differences in topography. The monthly mean air temperature in the basin ranged from -17.6 °C (January) to 20.6 °C (July), with an annual average of 3.1 °C (Figure S3). The annual average rainfall was 438–642 mm for 2008–2017, with the maximum in July (129 mm) and the minimum in January (only 4.9 mm). An average of 110 rain days occurs in one year (Figure S3). Since the 1960s, the air temperature in the watershed has been increasing at the rate of 0.3–0.4 °C·decade^{-1} (Jiang and Wang, 2019). More frequent extreme rainfall events have occurred in summer and autumn (Gou et al., 2020; Song et al., 2015; Yu, 2012), resulting in a stronger intra-annual variation in the stream flow (Wang et al., 2015).

The watershed is primarily agricultural, with some main cities located in the middle stream (Figure S2). The river receives nutrients via the point (e.g. wastewater effluent discharge) and the diffuse sources (e.g. land runoff) (Tong et al., 2017b). A monitoring program was initiated in 2006 to identify the dynamics of riverine P loadings across the SRB. Due to the high costs associated with the sampling and nutrient analysis, long-term and consistent water quality monitoring is rare in large rivers (Rowe et al., 2013; Sprague et al., 2017). A total of 10 sites were monitored in the upper, middle, and lower streams (1 site each), and major tributaries (7 sites) monthly in the 2008–2017 (Figure 1), and their results are summarised in Table 1. These monitoring sites were generally set at the outlet for each sub-basin to incorporate the overall impacts of anthropogenic P inputs into the basins. The procedure for water monitoring remained consistent during the study period, which was based on the 'technical specification requirement for the monitoring of surface waters in China'. The sampling was usually conducted during the early days of the month. An unfiltered surface water sample (1 L) was collected from the middle stream of the channels. Because of the difficulties in P species analysis for such a long period, only TP (the sum of dissolved and particulate P) was determined using the persulfate digestion-automated colorimetric analysis with a method detection limit (MDL) of 10 μg·L^{-1}. In the subsequent data analysis, all TP values lower than the MDL were set to 1/2 of the MDL.

2.2 Modelling soil P delivery and retention in the watershed

In this study, a process-based catchment P model (INCA-P) nested within a delicate soil module was explored to model the soil P retentions and estimate P loadings from human inputs to river (Jackson-Blake et al., 2016). INCA is a semi-distributed process based model that uses a mass-balance approach to track flow and nutrient pathways in both terrestrial and aquatic compartment. The model is based on a series of first-order chemical equations that are solved by using the Runge-Kutta method. Original version was developed by Wade et al. (2002), and it was modified by incorporating sediment erosions and the delivery process and modelling dissolved and particulate P separately instead of simulating bulk P (Jackson-Blake et al., 2016; Lazar et al., 2010). Different land use types were identified for each sub-basin and all the land-based processes (i.e. P input, storage and output) were calculated based on the 1 × 1 km grids for each type at sub-basin levels, and then scaled up to the actual areas (Jackson-Blake et al., 2016). INCA-P is capable of revealing the dynamic P delivery processes in soils (e.g. erosion, sorption/desorption and weathering) based on currently well-known chemical reaction (Jackson-Blake et al., 2016). INCA-P achieved successes in modelling land P delivery and retention in the catchments in Europe (e.g. Northeast Scotland, Jackson-Blake and Starrfelt, 2015) and Asia (e.g. North India, Pathak et al., 2018). INCA-P uses external time-series for surface air temperatures, rainfalls, hydrologically effective rainfall and soil moisture deficit generated by the PERSiST model (Futter et al., 2014). PERSiST is a watershed-scale and semi-distributed hydrological model at a daily time-step that is designed primarily for usages with the INCA model. A summary of the major input parameters of the model is provided in Table S1. A detailed sensitivity analysis of the model can be found in Pathak et al. (2018) and Wade et al. (2002).

In the modelling, soil P pool is divided into anthropogenic P inputs (e.g., fertilisers, crop residue) and historical storages (Figure 2). Based on their bioavailability to crops, three P forms were assumed to exist in soil: inactive, labile and bio-available (Goyette et al., 2018; Grenon et al., 2021; Sattari et al., 2012). Labile P can exchange with the bio-available P through desorption and sorption. The exchanges between inactive and labile P can occur through immobilisation and weathering (Jackson-Blake et al., 2016). P flow pathways into rivers include direct runoff, soil waters and groundwater (Figure 2). A summary of the dynamic P exchanges among the three forms could be described as follows: $\frac{dP_T}{dt} = P_{LIN} - \frac{dP_S}{dt} - P_{UP} - \frac{P_T}{V_S} q_S$

Equation (1)

$$\frac{dP_L}{dt} = P_{SIN} + \frac{dP_S}{dt} + C_W C_T P_I - m_l \frac{P_L}{M_S} - C_I C_T P_L$$

Equation (2)

$$\frac{dP_I}{dt} = C_I C_T P_I - m_l \frac{P_I}{M_S} - C_W C_T P_L$$

Equation (3)

Where P_T, P_L, P_I, and P_S refer to the mass of total dissolved, labile, inactive P, and the part of P_T involved in the exchanges with P_L; P_{LIN} and P_{SIN} refer to the P inputs through liquid or solid fertiliser, P_{UP} refers to the P uptake by crops; and M_S refers to the soil mass. V_S refers to the soil water volume; C_W and C_T refer to the weathering and temperature factors; m_L refers to the reach sediment inputs per unit area; and C_I refers to the chemical immobilisation factor. Detailed descriptions of these processes and associated parameters were provided by Jackson-Blake et al. (2016). In this study, difference between diffused-source P input, crop P uptake, and soil output was used to estimate the P retention in land as follows:

$$\mathrm{F}_R = \frac{P_{IN} - P_{UP} - P_{LO}}{P_{IN} - P_{UP}} \times 100\%$$

Equation (4)

where P_{IN} refers to diffused source P input; P_{UP} refers to the P output with crop production; P_{LO} refers to the sum of P losses through the direct runoff, soil water, and underground water; F_R refers to the P fraction of the diffused source input which was retained in the soil.

As an essential model input, we analysed the main anthropogenic P inputs into the basin by cropping, livestock operation, municipal/rural wastewater, and aquaculture, which are believed to be the dominant P sources (Liu et al., 2016; Tong et al., 2020). This dataset was a part of our national data set calculated by the hierarchical national P cycling model which was applied to evaluate the impact of human P inputs on water

TP concentrations (Tong et al., 2017b; Tong et al., 2020). To address spatial variations of human activities in the basin, a raster dataset based on gridded population densities (Li et al., 2019), livestock (Potter et al., 2010) and land use types (Liu et al., 2014) was developed. The output was a high-resolution map (at a resolution of 1 × 1 km) of anthropogenic P inputs with the five major sources for each year from 2008 to 2017 (Figure S4 and Dataset S1). P output through crop harvests in the watershed was also estimated based on regional statistics and re-distributed into 1 × 1 km grids according to the distribution of crop land.

2.3 Climate change scenario and modelling evaluation

Hydrological processes on land are largely controlled by climates which impact the soil P deliveries into the streams (Dudula and Randhir, 2016; Mukundan et al., 2020). To evaluate potential impacts, climate projections by MIROC5 based on the CMIP5 global climate model (Forster et al., 2013) were introduced into the model. MIROC5 was selected because of its good performance against historical air temperatures and rainfall records in the East Asia (Chen et al., 2021; Fu et al., 2020). We followed the previous studies to select the typical climate change projections (Jia et al., 2021; Fan and Bai, 2021). Three representative concentration pathways (i.e. RCP2.6, RCP4.5, and RCP8.5) had been selected to represent future GHG emission scenarios. RCP2.6 scenario is a peak scenario, indicating that the radiative forcing level would reach 3.1 W/m^2 by the mid-century but would return to 2.6 W/m^2 by 2100. RCP4.5 is the GHGs control strategy that is required to meet the 2.0 °C aspiration that is set under the Paris

Accord. RCP8.5 is a business-as-usual scenario in which the GHG emissions continue to increase throughout the 21st century. Daily air temperature and rainfall data under the three RCP scenarios were extracted from specific grids in the watershed. The extracted data was used to compare with the historical records and data from 2020 to 2050 was applied for the future simulations. In the scenario analysis of the future P reductions and climate changes, we first assumed that human P inputs (i.e., point- and diffuse- source inputs, Figure 2) in the SRB remained at the same level as that in 2017 and simulated the P input into the river from the soil in 2020–2050 (with the results in Figure 6). Three climatic scenarios (RCP2.6, RCP4.5 and RCP8.5) were introduced to simulate the impacts of the climate changes. Second, we have set up two P reduction scenarios (e.g. 10%, 20% and 30% reduction in point or diffuse-source inputs relative to the 2017) and simulate their responses in future riverine P loadings. Point-source reduction refers to reduction on municipal wastewater input, while the diffuse-source refers to fertilizer use, livestock farming and rural wastewater. We assume a reduction (10%, 20% and 30%, respectively) for all the diffuse-sources regardless of the types.

In this study, 2006 and 2007 were considered as a ‘warming up’ period to eliminate the initial biases (such as initial quick flows, initial soil water flow, etc.) in modelling. The monthly monitoring data obtained from water sampling sites and hydrological stations in the SRB were applied for calibration and validation. The models were calibrated by adjusting the INCA-P model parameters, such as flow velocity and soil P process parameters to fit the flow and water quality records for the time period from

2008 to 2012. The major parameters potentially impacting the modelling performance has been summarized by the previous studies (Whitehead et al., 2019). The model was subsequently validated for the time period of 2013–2017. The performance on the hydrodynamics was examined by stream flow rates in the mainstreams and tributaries (Figure S5). The performance in predicting the riverine P was examined by the TP monitoring results and riverine P loadings (see the section of Results, Figures S6–S7). Due to the lack of data on water P species, only TP concentration was used for applied. The Nash-Sutcliffe efficiency (NSE) and Pearson correlation coefficients (R) between the simulated and monitoring results were calculated (Ervinia et al., 2020; Wu et al., 2017). The river flow data was obtained from hydrological stations at the outlet of the sub-watershed. The monthly TP concentrations were obtained from the decadal river P monitoring project. The riverine P export was calculated based on the measured river flow rate and P concentration. Initial soil P storage (including the labile and inactive P) in each sub-basin was obtained from the China Soil Database (Shangguan et al., 2013) (Figure S8). In general, the initial soil inactive P in the basin ranged from 150 to 2085 mg/kg, but fluctuated in different land use types (e.g. 736 mg/kg in crop land and 985 mg/kg in forest). Initial soil labile P contents ranged from 5.7 to 88.7 mg/kg. Daily temperatures and rainfall data for 2008–2017 were used to simulate the hydrological process that was obtained from the weather stations in the basin (http://data.cma.cn). Spatial distribution data on the land use types, soil property, elevation, and population density in the basin were provided by Resources and Environment Science and Data Center, China.

3. Results

3.1 Temporal trend in the river P concentration and loading

Based on the monitoring data set of 2008–2017, we found that the TP concentration in the main streams of the SR and its tributaries remained relatively high (summarized in Table 1). In general, a water TP concentration of 76 $\mu g \cdot L^{-1}$ or higher could suggest a highly productive or eutrophic river system (Dodds et al., 1998; Stackpoole et al., 2019). In 2017, the annual average TP concentrations in the upper, middle, and lower streams of river were 123.0 ± 22.1, 101.4 ± 36.6 and 86.9 ± 16.5 $\mu g \cdot L^{-1}$, respectively, which were all higher than the threshold. Large variations in TP concentrations were observed among the different tributaries (Table 1). The lowest TP was observed in the Nenjiang River with the annual average of 65.1 ± 27.3 $\mu g \cdot L^{-1}$, whereas the highest TP value was observed in the Woken River with an annual average of 166.2 ± 68.1 $\mu g \cdot L^{-1}$. A clear distinction was observed regarding the seasonality of TP concentrations, with higher TP occurring in the wet season and lower TP in the dry season (Figure S9). An overall decline in the river TP concentrations was observed in the main streams (at a rate of 0.3–0.9 μg/L per month, $p < 0.01$, Figure 3) and some tributaries (Figure S6). However, rare changes were observed in riverine P export from 2008–2017 ($p > 0.05$, Figure 3). In 2017, the annual TP export from the SR was approximately 6,600 Mg (at a rate of 13.2 $kg \cdot km^{-2}$). Among all the tributaries, the Nenjiang River contributed the largest P input into the main stream, with an annual input of 1,250 Mg (accounting for 19–40% of the lower stream P export, Figure 4A and Table S3). TP loadings into the main stream from other tributaries were relatively small (e.g., 113 Mg by the Woken River and 170 Mg by the Mayi River). A strong seasonal variation in riverine P export was observed with the P loadings in summer (~3,500 Mg) and autumn (~1,400 Mg) occupying about 74% of the annual export (Figure 4B).

3.2 INCA-P modelling results and the performance evaluation

Figure 3 also shows comparisons between the observed and simulated stream flows, TP concentration, and river P loading for the entire study period. Model performance was evaluated using R and NSE. An NSE of 1 corresponds to a perfect match between the modelled and the observed data. An NSE of 0 indicates that the model prediction is as accurate as the average of the observed data. In general, the model showed good agreements between the observed and modelled flow, TP concentration, and P loading at all the sites in the main stream (Figure 3), with the R values ranging between 0.90–0.92, 0.71–0.82, and 0.82–0.88 and NSE values between 0.59–0.69, 0.19–0.22, and 0.46–0.50 for the river flow, TP concentration, and river TP loadings, respectively. For the tributaries, the NSE values ranged from 0.59 to 0.80 for river flows, from 0.03 to 0.20 for TP concentrations, and from 0.22 to 0.68 for river P loadings (Figures S5–S7). In particular, the model successfully depicted the decadal trends and seasonal changes (e.g., the flooding and dry seasons) of river flows and river P exports during the study period (Figures 3B and S7).

3.3 Anthropogenic P input and the land retention

In contrast to the insignificant changes in river P export in the watershed (Figure 3), we found that the reductions in anthropogenic P input in the basin (Dataset S1) did not result in a decrease in the riverine P export. An overall decline was observed in human P inputs into SRB in 2008–2017, from 1.6×10^5 Mg in 2008 to 1.3×10^5 Mg in 2017. Substantial P reduction was observed in the point-source input from 4,450 Mg in 2008 to 1,754 Mg in 2017. This reduction was mainly achieved by the rapid construction of WWTPs in the watershed where the number of operational WWTPs increased from 71 in 2008 to 214 in 2017. In 2017, over 90% of the local wastewater discharge was treated before the discharge.

The relative impacts of diffuse and point-source on receiving water were different as diffused source P input is retained in land before reaching the stream (Chen et al., 2015). Land retention, defined as the fraction of diffused source P input retained in land, could regulate the rates of P delivery into surface water, thereby influencing the relative P contribution from diffused sources into rivers (Goyette et al., 2018; Powers et al., 2016; Stackpoole et al., 2019). The modelling results indicated that the P input from the diffused sources and historical storage became increasingly dominant over time (Figures 5 and S10). From 2008 to 2017, the annual P inputs into the rivers from diffused sources increased from 7,600 to 8,900 Mg in the entire SRB at a rate of 133.8 $\mathrm{Mg \cdot year^{-1}}$. With decrease in point-source P input, the fraction of point-source input in total river P loadings declined from approximately 38.8 ± 10.7% in 2008 to only 18.9

± 7.7% in 2017 (Figure 5). We found that the watershed had a strong capacity to retain P in the lands. In 2017, the land retained approximately 93.6 ± 1.6% of the diffused source P inputs (see Figure S10). The entire watershed retained approximately 189 kg P·km^{-2} from the diffused source input in 2017, and 2600 kg P·km^{-2} was retained in the SRB during the study period. In particular, our modelling has also indicated that the P retention capacity across the SRB showed slow but substantial decrease during 2008–2017 (Figure S10).

3.4 Response in the land P retention under climate change scenarios

Under the climate change scenarios (RCP2.6, RCP4.5 and RCP8.5), air temperature and rainfall in the watershed is expected to change significantly. Significant increases in daily surface air temperature and rainfall can occur during 2020–2050 (Figure S11), with an annual increase of 0.01–0.04 °C in air temperatures and an annual increase of 5.96–7.86 mm in the rainfall for three RCP scenarios. Occurrences of heavy rainfall events (> 25 mm in one day) would be more frequent under all three climate change scenarios, from 1–5 days per year for 2020–2030 to 4–9 days per year for 2040–2050. Consequently, annual river flows of the SR would increase by 8–15% relative to 2020, from 563 × 10^{8} m^{3} in 2017 to (1232–1359) × 10^{8} m^{3} during 2040–2050 (Table 2). We simulated the impacts of climate change on land P export to river and found that under the climate change scenarios, P inputs from diffused sources and historical storage to river can increase substantially for 2020–2050 (Figure S12). Even if anthropogenic P input in the SRB remained at the same levels as those in 2017, the P contributions into rivers from the land could increase from 13.3 kg·km^{-2} in 2017 to 22.6–25.8 kg·km^{-2} in 2040–2050.

4. Discussion

4.1 A summary about soil P retention in the watershed

P retained in soils (forming a ‘legacy’) is an underestimated pool that determines P exports to the downstream waterbodies (Hu et al., 2020; Powers et al., 2016; Sharpley et al., 2013; Stackpoole et al. 2019). However, it is difficult to be estimated accurately because of complicated soil P mechanism. Soil P retention could reduce the discharge into rivers and lakes in the short term; while in the long-term, the accumulated P over time can act as a chronic P source even after the external input is reduced (Macintosh et al., 2018; Withers et al., 2017). In general, the P retention fraction of the diffused source input in the basin was to the value estimated by the global NEWS model in this basin (approximately 90–95%) (Tysmans et al., 2013). This fraction was higher than those in the Yangtze River Basin (40–50%) (Liu et al. 2018b) and Ganges River Basin (only 3%) (Tysmans et al., 2013) and was approaching to those in the Lake Erie Basin (90%), the Lake Michigan Basin (95%) and the Mississippi River Basin (91%)(Han et al., 2011). This indicates that a large fraction of the diffused source P inputs is the part of the historical P legacy.

The capacity to retain P in the land is supposed to be finite, with a threshold beyond which riverine P export could increase substantially (Goyette et al., 2018). In a survey targeting 23 watersheds in North America, researchers have suggested a low threshold of 2.1 t P km^{-2}. Beyond this, further P inputs into watersheds could cause a significant acceleration of P losses with runoff (Goyette et al., 2018). In several regions (e.g. East Asia and North America), historical agricultural P usage has been substantially greater than the actual needs of the crops (Macdonald et al., 2011). Thus, a large amount of P originating from the historical P inputs has accumulated in the soils over time (Grenon et al., 2021; Macintosh et al., 2018). Soil P retentions would explain the differences in the responses of riverine P exports to anthropogenic P inputs into the watershed (Chen et al., 2015; Jarvie et al., 2013; Kusmer et al., 2019). In general, the human-dominated basins may undergo a prolonged but finite accumulation period when P input exceeds the agricultural need. The accumulated P could continue to mobilise long after the P inputs are reduced (Powers et al., 2016). However, different basins could have varying responses to anthropogenic P inputs depending on factors, such as climatic conditions, historical storage, and soil properties (Hu et al., 2020; Kusmer et al., 2019).

4.2 Watershed P management under climate change scenarios

Climatic factors have important implications for terrestrial P retentions (Chen et al., 2015; Fischer et al., 2021; Goyette et al., 2016). A previous study observed a positive relationship between rainfall, drainage outflow, and increased P loading in the runoffs (Williams et al., 2015). In a recent study in the Western Lake Erie Basin in the USA, it was proposed that climate change would result in less nutrient runoff due to increased evaporation and decreased snowfall, in spite of the increases in the overall amounts of precipitation (Kalcic et al., 2019). However, some other researchers hold the view that with the climate changes, riverine P export would be enhanced largely due to the more frequent extreme rainfall events, as indicated in agricultural regions in the Midwestern region of the USA (Carpenter et al., 2018) and three diverse agricultural catchments in the UK (Ockenden et al., 2016). They further concluded that the impacts of climate changes would mainly occur in winter, with an increase of up to 30% in the P loading by 2050. To mitigate these impacts, a 20–80% reduction in the agricultural P inputs is required (Ockenden et al., 2017). In the SRB, hydrological characteristics may change substantially in the future with climate change (summarised in Table 2). We simulated two P input reduction scenarios (point and diffused-source reductions) under RCP2.6, RCP4.5, and RCP8.5 for 2020–2050. We found that, in the future, compared with the no-reduction scenarios (Figure 6), reductions of diffused source P inputs could have a greater effect on reducing the riverine P exports than the point-source reduction. With a 50% reduction in diffused source input, riverine P exports may decrease from 10564 ± 641 Mg (in the scenarios without P reductions) to 8872 ± 641 Mg under the three climate scenarios for 2040–2050. With a 50% reduction in point-source P input, there would be a limited reduction in the riverine P loading for 2040–2050 (Figure 7).

4.4 Mitigation in agricultural practices in the watershed

Potential challenges revealed in the SRB could be present in many other watersheds globally (Ockenden et al., 2017). Considering increasing contributions from diffused sources and historical P legacy, the mitigation in the agricultural practices is required to reduce soil P loss under climate change scenarios (Arneth et al., 2019). We suggest that three strategies should be considered in the future watershed P management. First, the historical P store should be taken into account, and the P-fertilizer usage should be reduced. In fact, after a long history of P-fertilizer applications, an increase in surface soil P content has occurred in many regions (Ma et al., 2019). For agricultural systems located in Europe and Canada, a reduction in the P fertilizer usage has been shown to have very few negative impacts on crop yield (Bast et al., 2009; Mogollón et al., 2021; Sattari et al., 2012). However, it would take a long period for recovery to the critical legacy P stock levels in the catchment (Chen et al., 2019). For the regions (e.g., USA and Europe) with substantial P legacies due to the extensive P fertiliser usages during 1970–2000, soil P legacies would allow crop productions without new P additions for approximately six decades (Mogollón et al., 2021; Sharpley et al., 2013). Second, soil erosion should be alleviated by changing land use (e.g. afforestation and reforestation of cultivated land). The majority of soil P loss through runoff has been in the form of particulates (Chen et al., 2015). Since the 1950s, due to the intensified farming in the SRB, the depth of the organic soil layers has decreased from 60–70 cm in the original cultivation period to only 20–30 cm (Fang and Sun, 2017). Increased soil erosions can intensify the riverine P exports (Alewell et al., 2020). Thus, soil conservations and the controls of sediment delivery into waters are beneficial (e.g., reducing tillage intensity and construction of gully control) (Shao et al., 2021; Yan et al., 2018). Third, mineral fertilisers have been used to intensify terrestrial P retentions. Compared with the other watersheds, many regions in the SRB have relatively high soil organic contents which provide the releases of a large pool of P over time. Soluble phosphate available for crop uptake could form the inactive ionic complexes with the Al and Fe, as well as the precipitation with Ca (Grenon et al., 2021). Thus, the application of mineral fertilisers could introduce Ca or Fe to these fields (Grenon et al., 2021). Organic soils with the greater Fe and Al stocks could show a higher retention of P in the lands (Larsen et al., 1958). However, this practice would not mitigate the soil 'P-legacy' issues, but only reduce the delivery rates into the rivers.

4.4 Uncertainty in modelling and limitations

There are always issues related to model uncertainties and model limitations in any flows and water quality modelling studies (Choa et al., 2020; Jia et al., 2021). The fact that the system is dynamic and that the chemical processes are kinetic in nature and interact with the hydrology makes the modelling a complex process. To the best of our knowledge, this study is one of the few attempts to apply a process-based model to simulate land P retention. A similar but simpler case could be found in Sattari et al. 's (2012) study, which applied a two-pool soil P model to depict the soil P residues. This study has some limitations. First, unlike the traditional methods to model the soil P retention, a process-based model was developed. INCA-P has progressed significantly in the soil P modules, but soil P dynamic processes are still simple. For example, the roles of carbon and the other nutrients in the biological P uptakes, mineralisation, and immobilisation rates were not included. Second, long-term water monitoring data with the high frequency (e.g. daily or even hourly), particularly under high flow conditions, are essential for validating the model. However in this study, only monthly monitoring data were used to validate the modelling result. Third, future water monitoring should also include the other P species, such as total dissolved and particulate P, in addition to TP. Most of the P in the streams exists in the form of particulate P, while dissolved P is more bioavailable. Future monitoring should reflect potential impacts of different P species in rivers.

5. Conclusions

In summary, this study applied a process-based catchment P model with a delicate soil module to characterise the nexuses among P inputs from human activities, land P retention, and climate change. We proposed that the increasing P contributions from diffused sources or historical legacy counteracted P reductions by improved sanitation in the SRB. Under climate change scenarios, P input into rivers from diffused sources and historical legacy would increase substantially which could constitute a challenge for water quality improvement. We suggest that a long-term perspective of watershed control strategy should target the controls and reduction of P inputs from the historical legacy, rather than solely focusing on short-term changes in riverine P concentrations and loadings.

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Figure captions

- Figure 1. Location of the Songhua River Basin, and the distributions of water sampling sites and hydrological stations in the watershed;
- Figure 2. Diagram showing the major processes influencing land P deliveries in the model;
- Figure 3. Comparisons between the measured and simulated river flow (A), TP (B), and P loading (C) in the main stream during 2008–2017;
- Figure 4. Riverine P loadings in the Songhua River Basin (A) and the seasonal variations in 2017;
- Figure 5. Changes of P inputs into the rivers from the point and diffused sources during the study period;
- Figure 6. Potential changes of river P loading without the P input reduction under climate change scenarios (RCP2.6, RCP4.5, and RCP8.5);
- Figure 7. Response in riverine P loading in lower stream under diffused sources and point-source reduction scenarios with future climate change;

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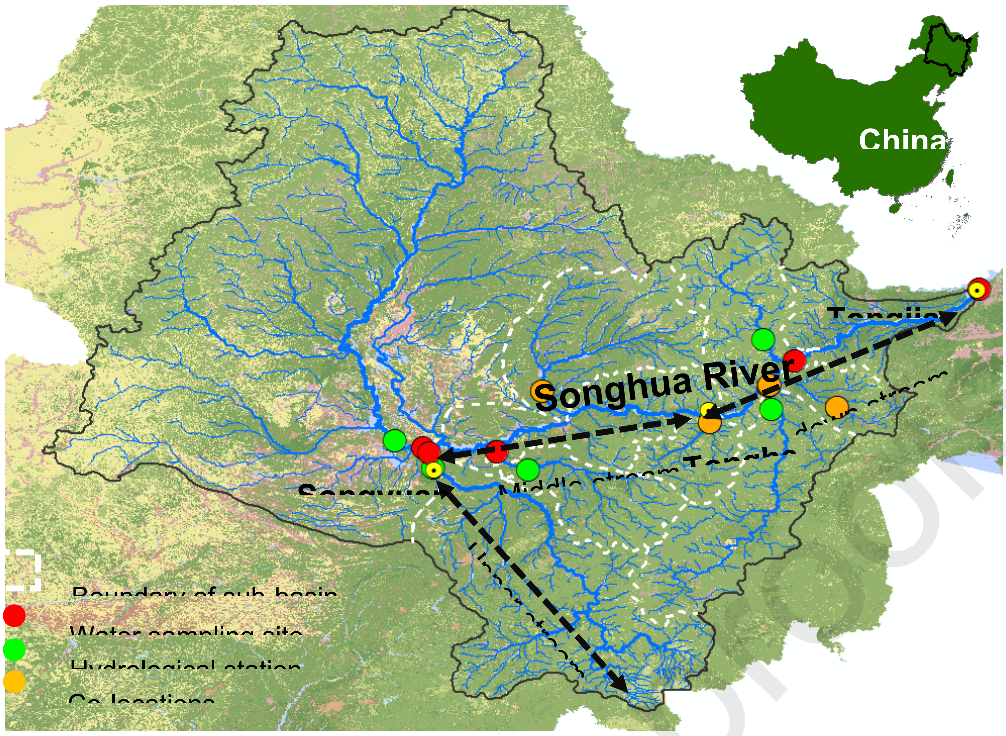


Figure 1. Location of the Songhua River Basin, distributions of water sampling sites and hydrological stations in the watershed [a]

[a] Locations of the tributaries and the associated watersheds was provided in Figure S1.

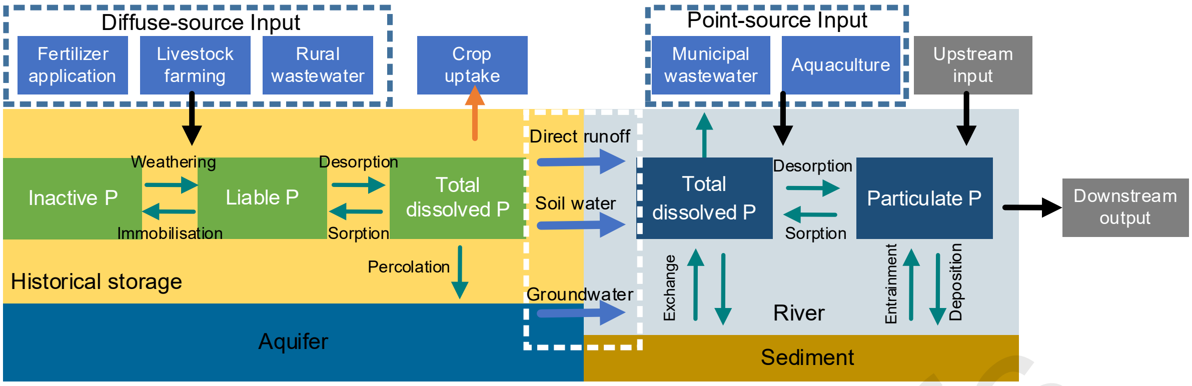
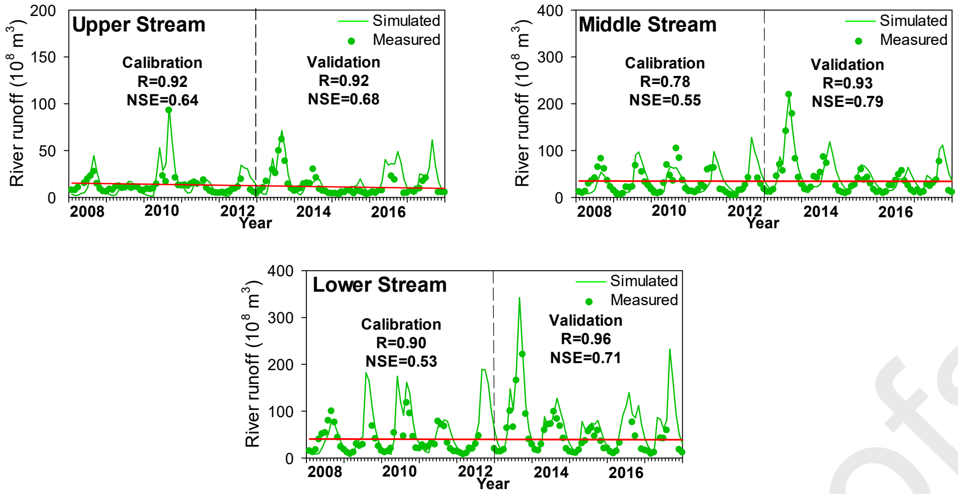
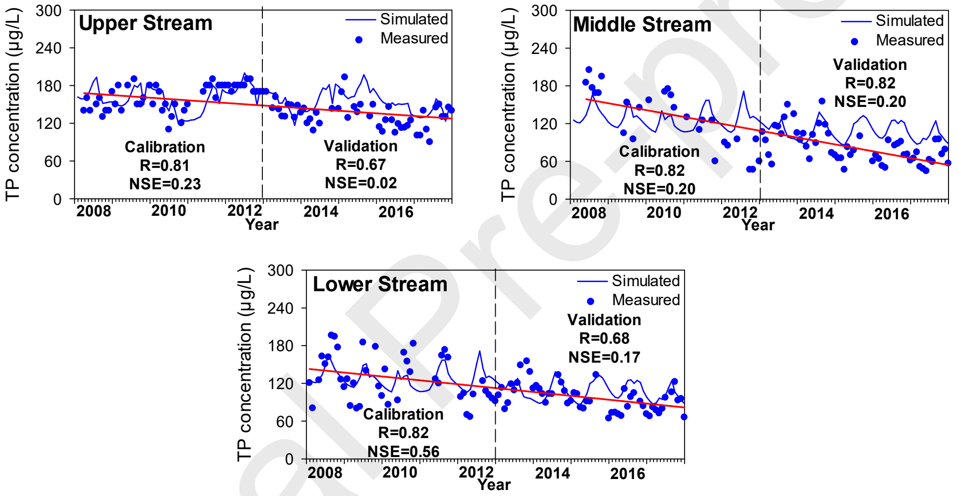


Figure 2. A diagram about the major processes influencing land P deliveries [a]

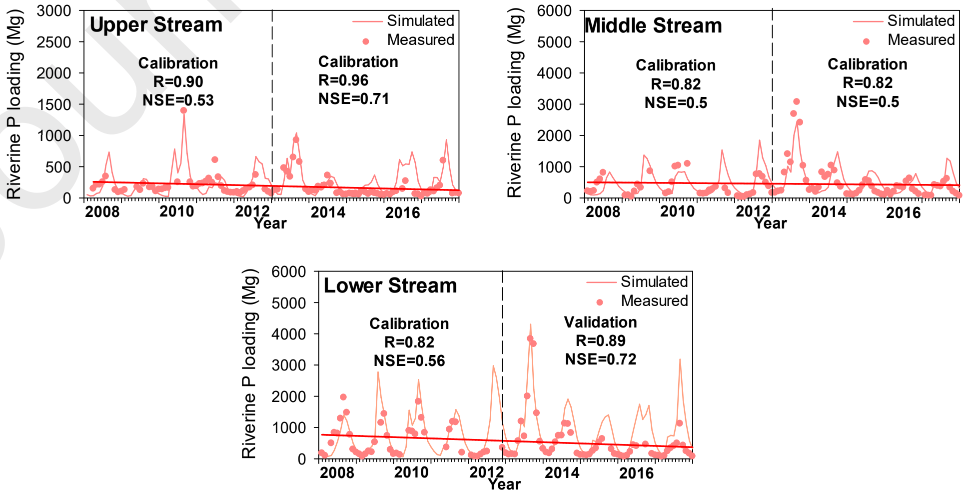
[a] A detailed description about these processes and their calculations was provided in Jackson-Blake et al., 2016, Environmental Modeling & Software, 83, 356-386.


Upper Stream
Simulated
Measured
Calibration
R=0.92
NSE=0.64
Validation
R=0.92
NSE=0.68
River runoff (10^8 m^3)
200
150
100
50
0
Middle Stream
Calibration
R=0.78
NSE=0.55
Validation
R=0.93
NSE=0.79
400
300
200
100
0
Lower Stream
Calibration
R=0.90
NSE=0.53
Validation
R=0.96
NSE=0.71
2008
2010
2012
2014
2016
Year

(A)


Upper Stream
Simulated
Measured
Calibration
R=0.81
NSE=0.23
Validation
R=0.67
NSE=0.02
TP concentration (μg/L)
300
240
180
120
60
0
Middle Stream
Validation
R=0.82
NSE=0.20
Calibration
R=0.82
NSE=0.20
Lower Stream
Validation
R=0.68
NSE=0.17
Calibration
R=0.82
NSE=0.56
2008
2010
2012
2014
2016
Year

(B)

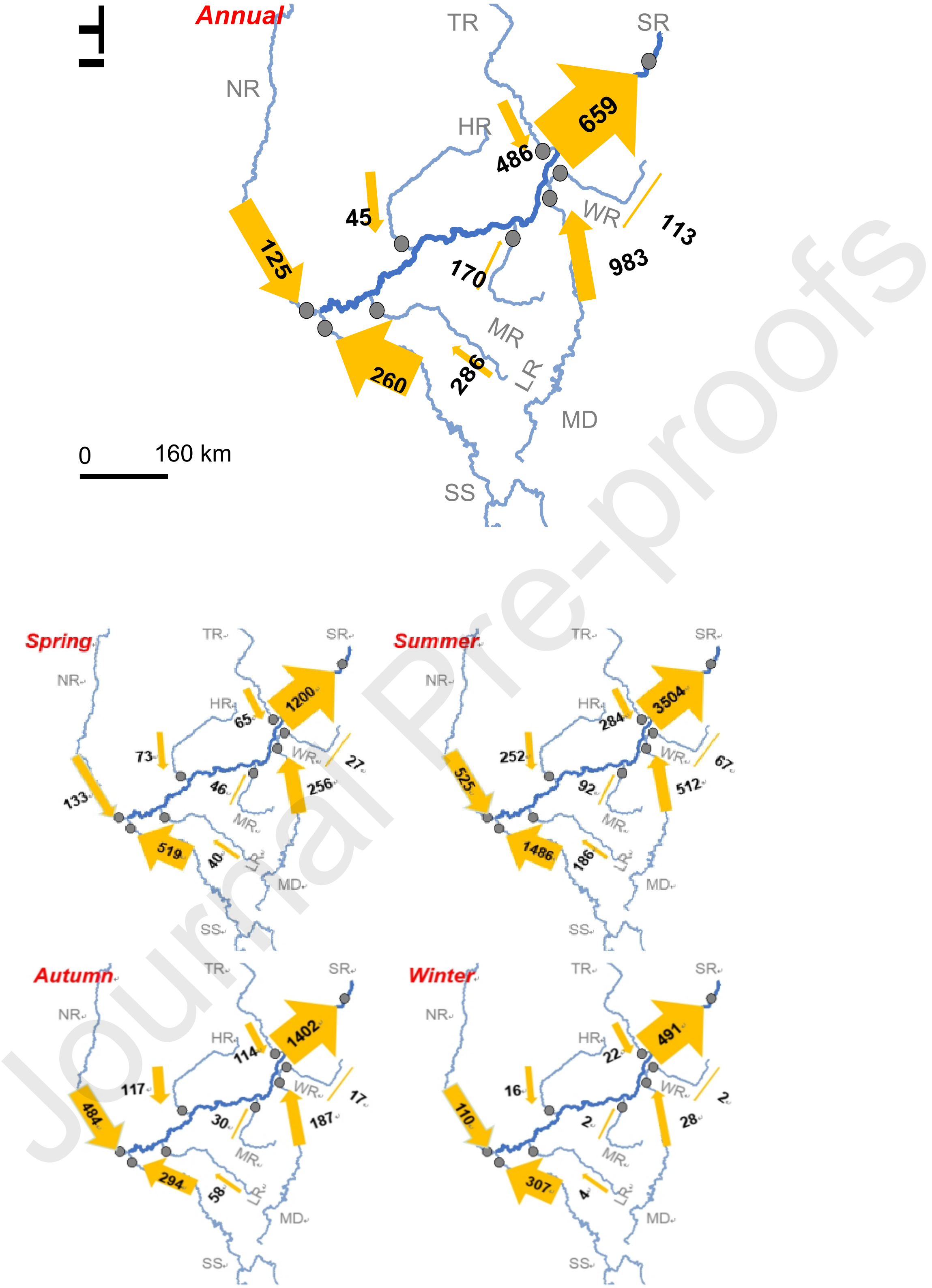

Upper Stream
Simulated
Measured
Calibration
R=0.90
NSE=0.53
Calibration
R=0.96
NSE=0.71
Riverine P loading (Mg)
3000
2500
2000
1500
1000
500
0
Middle Stream
Calibration
R=0.82
NSE=0.5
Calibration
R=0.82
NSE=0.5
6000
5000
4000
3000
2000
1000
0
Lower Stream
Calibration
R=0.82
NSE=0.56
Validation
R=0.89
NSE=0.72
2008
2010
2012
2014
2016
Year

(C)

Figure 3. A comparison between measured and simulated river flow (A), TP (B) and P loading (C) in main stream in 2008–2017[a, b]

[a] k in figure represents the slope of the linear regressions between river flow (or TP concentration or P loading) and sampling time. p represents the significance in the regression analysis.

[b] A comparison between the measured and simulated results in tributaries was provided in Figures S5–S7.


Annual
TR
SR
NR
HR
659
486
45
125
170
WR
113
983
MR
260
286
LR
MD
SS
0
160 km
Spring
TR
SR
NR
HR
1200
65
73
27
WR
46
256
133
MR
519
40
LR
MD
SS
Summer
TR
SR
NR
HR
3504
284
252
67
WR
92
512
525
MR
1486
186
LR
MD
SS
Autumn
TR
SR
NR
HR
1402
114
117
17
WR
30
187
484
MR
294
58
LR
MD
SS
Winter
TR
SR
NR
HR
491
22
16
2
WR
2
28
110
MR
307
4
LR
MD
SS

(A)

(B)

Figure 4. Riverine P loadings in the Songhua River Basin (A) and seasonal variations in 2017 (Mg) (B)[a, b]

[a] Gray dots represented the locations of outlets in the sub-watershed.

[b] Riverine P loading in the other years was provided in Supplementary Dataset S2.

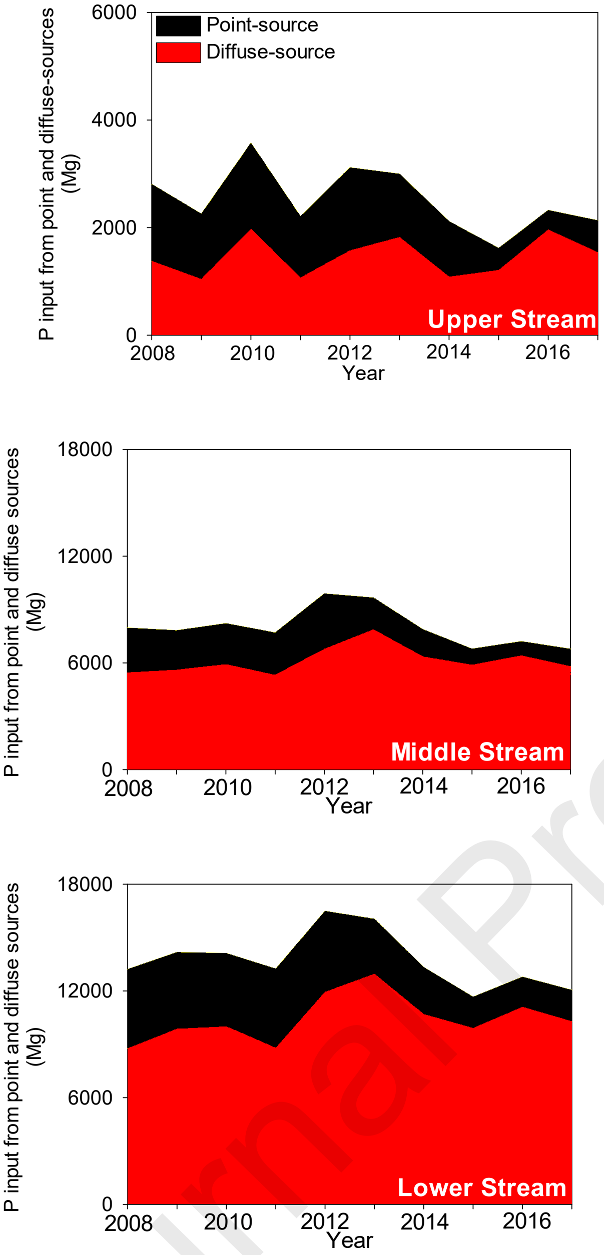


Figure 5. Changes of land P inputs into the mainstream from the point and diffuse-sources during the study period

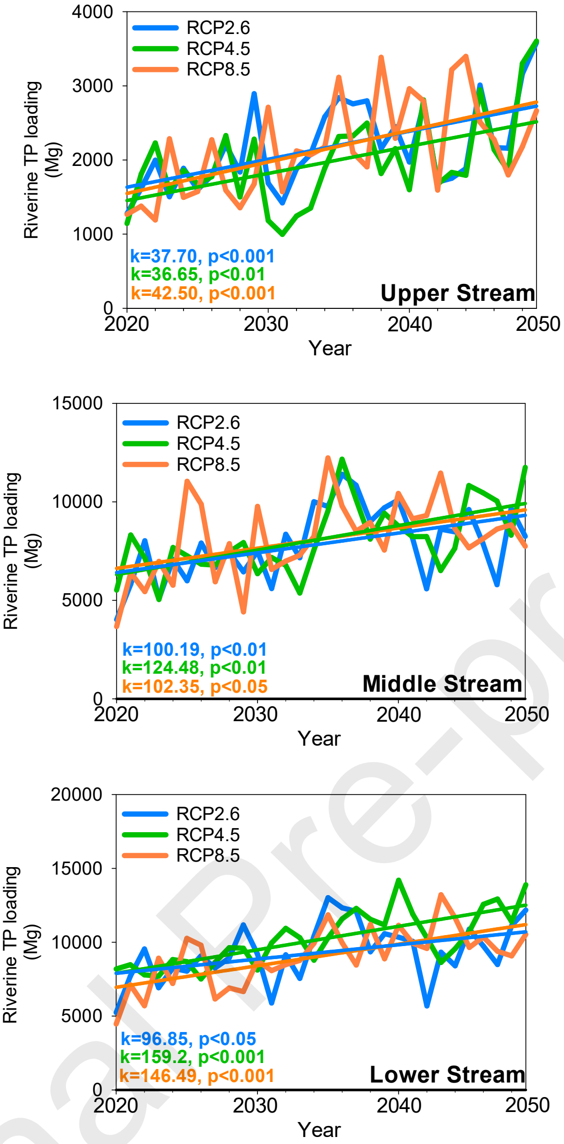
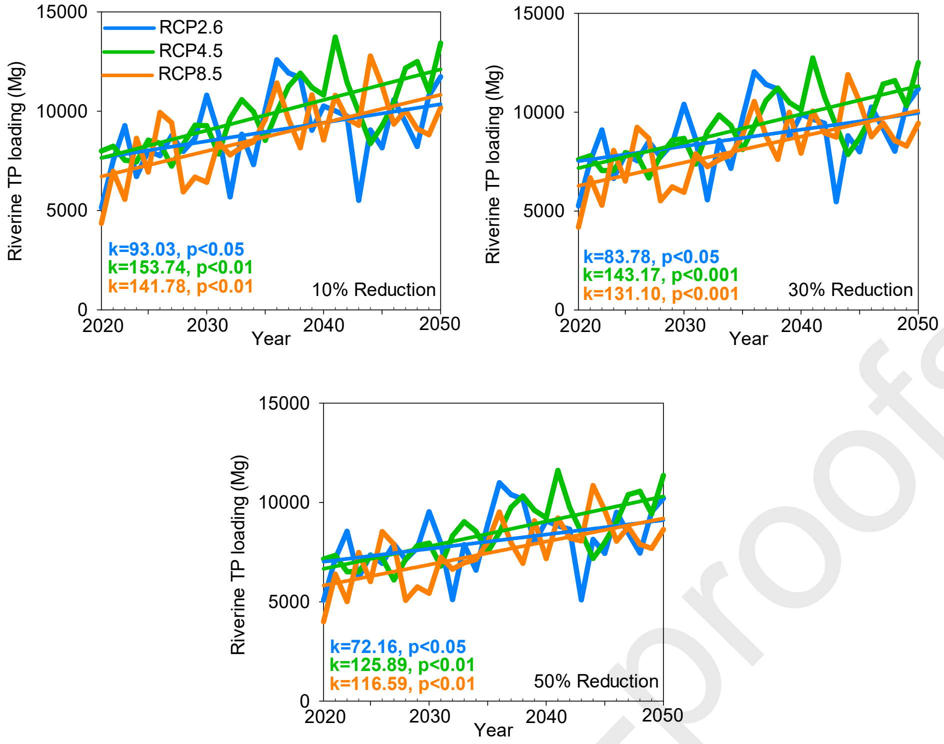
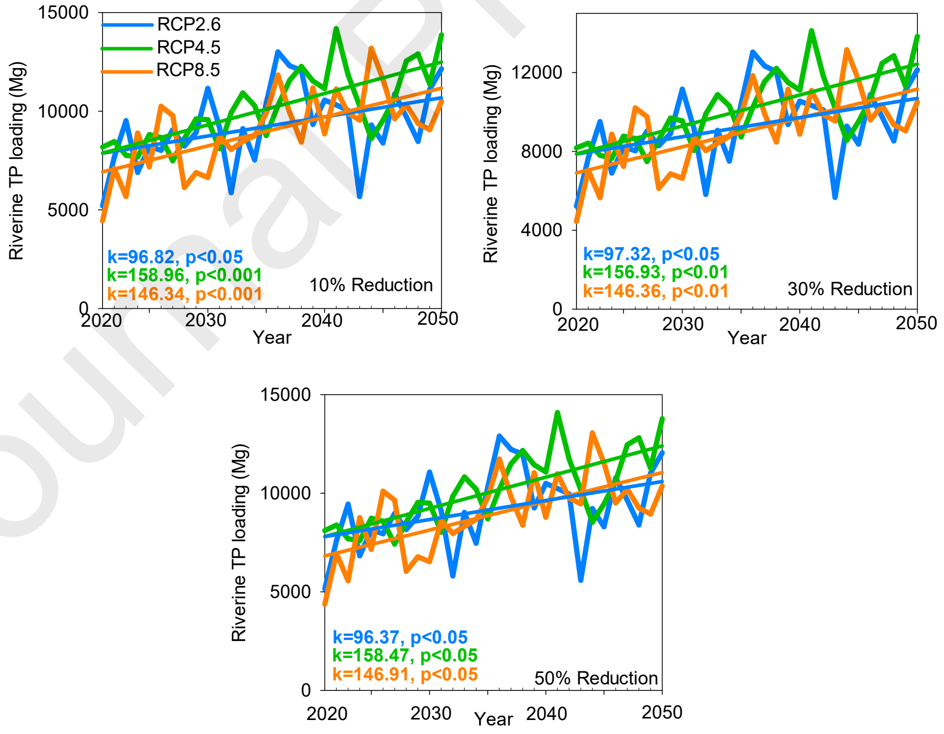


Figure 6. Potential changes of river P loading without the P input reduction under climate change scenarios (RCP2.6, RCP4.5, and RCP8.5)

[a] The human P input into the watershed is assumed to remain the same as that in 2017.



(A) Diffused-source input reduction



(B) Point-source input reduction

Figure 7. Response in riverine P loading in lower stream under diffuse source and point source reduction scenarios with climate changes [a]

[a] We have set up two P reduction scenarios (e.g. 10%, 20% and 30% reduction in diffuse or point-source input relative to the 2017) and model their responses in riverine P loadings.

[b] Diffuse-sources refer to the sum of fertilizer application, livestock farming and rural wastewater. Difference among the specific sources is not distinguished.

Table 1. TP concentrations and loadings in the main stream and tributaries of the Songhua River[a, b]

River	**Longitude**	**Latitude** [b]	**Period**	**n**	**Area** (km^2)	**Rainfall** (mm·year-1)	**Annual runoff** (10^8 m^3)	**TP** **(**$\mu g \cdot L^{-1}$**)**	**TP input** ($kg \cdot km^{-2} \cdot year^{-1}$)	**TP output** ($kg \cdot km^{-2} \cdot year^{-1}$)
Upper stream	125.93	44.77	2008—2017	116	74355	712.5	164	123.0 ±	313.85	15.09

								22.1		
Middle stream	128.16	45.92	2008—2017	97	450077	495.4	308	101.4 ± 36.6	131.88	10.45
Lower stream	132.52	47.7	2008—2017	109	523580	593.1	407	86.9 ± 16.5	202.80	13.32
Nenjiang River	123.7	46.8	2008—2017	100	291128	493.7	76	65.1 ± 27.3	124.72	12.30
Lalin River	125.71	45.39	2012—2017	49	18339	569.2	24	82.7 ± 19.5	437.86	18.22
Hulan River	126.34	46.25	2014—2017	31	27736	632.8	13	115.2 ± 19.0	673.32	15.30
Mayi River	128.72	45.81	2016—	17	10425	583.9	15	71.1 ±	329.45	20.46

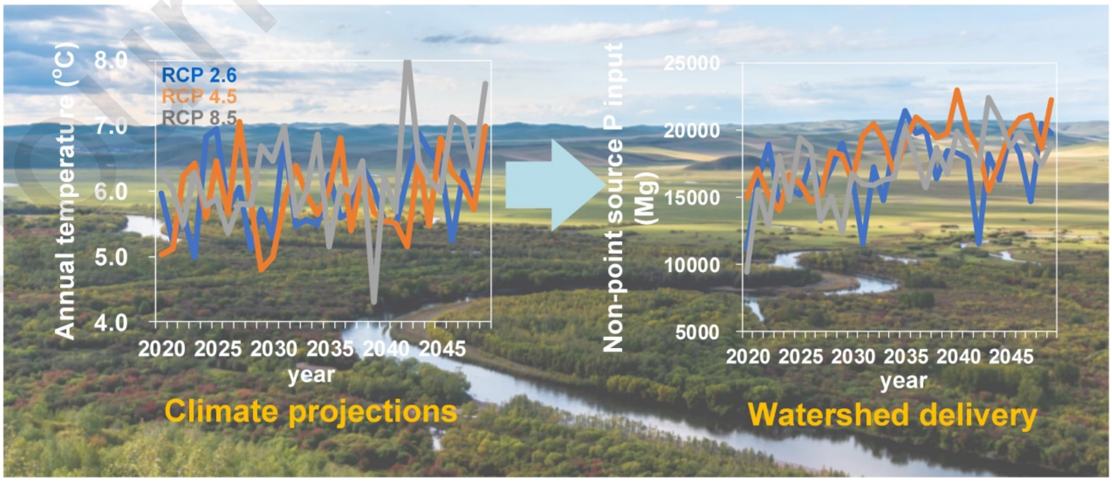
			2017					30.6		
Mudan River	129.55	46.32	2008—2017	103	35683	573.9	72	108.8 ± 24.6	160.44	10.77
Woken River	130.52	46.02	2013—2017	42	10820	615.1	3	166.2 ± 68.1	326.05	20.87
Tangwang River	129.91	46.67	2012—2017	47	20557	663.3	45	66.6 ± 19.1	86.75	18.44

[a] Result in the year of 2017 was provided in the table.

[b] Longitude and latitude of water sampling sites at the outlet for each sub-watershed were provided.

Table 2. Changes of hydrologic conditions in the baisn under climate change scenarios

Variables	RCP2.6 scenario			RCP4.5 scenario			RCP8.5 scenario		
	2020	2030	2040	2020	2030	2040	2020	2030	2040
Precipitation (mm^{-1})	882.5	881.9	973.7	937.4	858.5	1198.5	738.6	792.4	972.8
Snowfall (mm^{-1})	86.6	86.6	183.9	128.2	100.6	159.7	146.4	104.2	125.4
Snowmelt (mm-1)	44.7	138	87.1	56.7	109.2	168	73	79.3	103.8
Evapotranspiration (mm-1)	568.56	575.76	578.64	501.72	560.64	574.8	569.76	579.24	575.64
Surface runoff (mm-1)	298.96	369.36	329.04	366.48	334	575.68	194.8	234.32	383.36



Highlights

- A process-based catchment model with a delicate soil P module was attempted to estimate P export and land retention.
- P released from diffuse source and historical legacy may offset efforts brought by the point source controls.
- Rapid climate change in the watershed could further deplete the terrestrial P retention capacities.
- A long-term perspective of P control strategy should be targeted at the reductions of historical P legacy inputs.

Conflict of interests

The authors declare no conflict of interests.

Credit Author Statement

Tong Y., Wang, Y., and Wang X. designed the study and wrote the paper. Chen Z. and Wen, Y performed the calculations. Qi, M., Zhu, M., Wang, R.and Sha, J collected the raw data for the calculation. Zhang, H. and Lin, Y revised the manuscript.