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RESEARCH ARTICLE

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Influence of land use on the persistence effect of riverine phosphorus

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1 | INTRODUCTION

Although technological and infrastructure deployment provide an effective method to address point source pollution, non-point source nutrient pollution remains a problem in many rivers (Huang & Jun, 2014; Roy & Bickerton, 2014). Excess P concentrations and fluxes have several harmful effects on riverine ecosystems leading to eutrophication/hypoxia of reservoirs, estuaries, and coastal waters (Roy & Bickerton, 2014; Chen, Hu, Wang, et al., 2015). Watershed

Abstract

The persistence effect contribution of legacy nutrients is often cited as a reason for little or no improvement in water quality following extensive implementation of watershed nutrient mitigation actions, yet there is limited knowledge concerning factors influencing this response, often called the "persistence effect." Here, we adopted detrended fluctuation analysis and Spearman analysis methods to assess the influence of land use on the watershed phosphorus (P) persistence effect, using monthly water quality records during 2010-2016 in 13 catchments within a drinking water reservoir watershed in eastern China. Detrended fluctuation analysis was used to calculate the Hurst exponent α to assess watershed legacy P characteristics ($\alpha \approx 0.5$, $\alpha > 0.5$, and $\alpha < 0.5$ indicate white noise, persistence, and anti-persistence, respectively). Results showed weak to strong P persistence (0.60-0.81) in the time series of riverine P in the 13 catchments. The Hurst exponent α had negative relationships with agricultural land (R = -.47, p = .11) and developed land (R = -.67, p = .01) and a positive relationship with forest land cover (R = .48, p = .10). The persistence effect of riverine P was mainly determined by retention ability (biogeochemical legacy) and migration efficiency (hydrological legacy). A catchment with strong retention capacity (e.g., biomass uptake/storage and soil PO₄ sorption) and low migration efficiency results in a stronger persistence effect for riverine P. In practice, source control is more effective in catchments with weak persistence, whereas sink control (e.g., riparian buffers and wetlands) is preferred in catchments with strong persistence effects.

KEYWORDS

legacy phosphorus, memory effect, Hurst exponent, detrended fluctuation analysis, Spearman analysis

management practices to target non-point source pollutants might fail to achieve expected improvements in water quality, even after extensive implementation of mitigation measures (Jarvie et al., 2013; Meals, Dressing, & Davenport, 2010; Onderka, Mrafková, Krein, & Hoffmann, 2012; Stålnacke, Grimvall, Libiseller, Laznik, & Kokorite, 2003; Van Meter & Basu, 2015). A major reason cited to explain the lack of water quality improvement is the contribution of legacy P, transiently stored P (e.g., vegetation, soils, sediments, and groundwater) that has accumulated from years of human P inputs within the watershed (Jarvie et al., 2014; Sharpley et al., 2013; Chen, Hu, Wang, et al., 2015). It should be mentioned that the retention of P within watershed

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and the release of legacy P are strongly impacted by catchment buffering capacity (Doody et al., 2016). For example, soils and sediments will buffer changes in soil P concentrations and release to run-off depending on mineral composition, organic matter content, and redox condition (McDowell, Monaghan, & Morton, 2003). Once the catchment buffering capacity reach a threshold (saturation) level, the rerelease of legacy P along with hydrological process will be more rapid. Hence, the release of legacy P can adversely impact river water quality for long time periods (e.g., years to decades) resulting in a "persistence effect" or "memory effect" for riverine P. Identifying the significance of the persistence effect is essential for effective environmental decision-making and scientific research, yet it is difficult to assess given our incomplete understanding of controls on nutrient dynamics following changes in land-use or management practices (Van Meter & Basu, 2015).

River water quality monitoring data integrate the effects of natural factors and human activities at the watershed scale and therefore contain information concerning the persistence effect, which is the representation of the legacy effect (Dong, Mei, Shang, Huang, & Huang, 2016; Mei et al., 2016). As early as 1951, Hurst (1951) demonstrated that the annual run-off record from the Nile River displayed a long-term persistence (self-similar), and he developed the "Hurst exponent" to characterize the long-term persistence. Subsequently, longterm persistence has been recognized in many time series, such as economic (Muchnik, Bunde, & Havlin, 2009), climatic (Orun & KoçAk, 2009), and water quality (Shi, Liu, Huang, Zhang, & Su, 2010) data sets. Calculation methods for the Hurst exponent include detrended fluctuation analysis (DFA) (Onderka et al., 2012), wavelet transforms (Simonsen, Hansen, & Nes, 1998), and rescaled range analysis (Nnaji, 2011). In the case of a nonstationary time series, represented by a trend and additive fractal noise, more accurate evaluation is generally obtained using the DFA method (Kirichenko, Radivilova, & Deineko, 2011). Data mining based on the Hurst exponent has great potential to characterize persistence in water quality and the legacy effect (Onderka et al., 2012). Shi et al. (2010) used DFA to analyse pH time series in stream water above and below Lake Poyang, China, and found that the pH time series had a long-term persistence effect. Onderka et al. (2012) analysed long-term records (~20 years) of riverine nitrate concentrations at three stations on the upper Váh River (Slovakia) by wavelet transforms and DFA and found varying degrees of persistence among the sites. Mei et al. (2016) adopted the rescaled range analysis to address the persistence effect for riverine nitrogen in 13 Chinese rivers and found a significant persistence for total nitrogren concentrations in 11 of the 13 rivers. Most previous studies focused on nitrogen; however, there are few reports about the persistence effect for P, which displays several contrasting biogeochemical characteristics compared to nitrogen.

The persistence effect is a major factor affecting long-term water quality at the watershed scale, and therefore, defining the strength and factors regulating the P persistence effect are important in pollution prevention and control. For watershed non-point source pollution, hydrological processes is playing a central role that is affecting various processes including pollution loading amount, retention capacity of landscape, and the migration efficiency of pollutants (Amin, Veith, Collick, Karsten, & Buda, 2017; Ding, Zhou, Lei, Liao, & Wang, 2013; Shan, Yin, & Li, 2002). Meanwhile, land use/land cover is important

factor influencing hydrological processes (Shi et al., 2013; Wijesekara et al., 2012). Furthermore, changes in watershed land use can result in transformation of the watershed from a "sink" to a "source" by directly and indirectly impacting the migration and transformation of nutrients (Onderka et al., 2012). Therefore, changes in land use/land cover would be expected to be an important factor influencing the persistence effect (Mei et al., 2016). However, there has been little research concerning the factors influencing the persistence effect for riverine P as a function of land-use and watershed characteristics. Shanxi Reservoir is a drinking water source watershed in the mountainous region of Zhejiang Province, eastern China. The watershed has been strongly impacted by human activities in recent decades, including extensive land-use change and atmospheric deposition. On the basis of monthly riverine P concentration records during 2010-2016 in 13 catchments within this watershed, this study adopted the DFA and Spearman analysis methods to determine if the riverine P time series experience a P persistence effect and to assess the influence of land use on the persistence effect.

2 | MATERIALS AND METHOD

2.1 | Study area and data source

The Shanxi watershed is located in the uplands of the Feiyun River watershed in Zhejiang Province, China (Figure 1). This watershed has a total watershed area of 2,303 km² supplying a multi-annual regulating reservoir (total storage capacity of 1.8×10^9 m³) with a water diversion project located 30 km downstream. The reservoir is the major drinking water source for more than 7 million people in the local region. The study area has a subtropical monsoon climate with mean annual precipitation of 1,870 mm and temperature of 17 °C. Mean watershed elevation is 573 m (range 9–1,663 m). The watershed is dominated by highly weathered, iron oxide-rich, red soils with land use dominated by forest (75%) and cultivated (15%) lands. Population density within the watershed is 236 persons/km², and the remaining major nutrient inputs are from non-point sources (Huang et al., 2017).

Total phosphorus (TP) is a key water quality concern for potential eutrophication and harmful algal blooms in Shanxi Reservoir (Dong et al., 2016). Water quality in 13 catchments within the watershed was monitored by the local Water Resources Bureau on an approximately monthly basis during 2010-2016 (Figure 1). TP was measured following persulfate digestion using the ammonium molybdate tetrahydrate spectrophotometry method. The catchment areas ranged from 10.1-284.2 km², and the average TP concentrations of the 13 catchments ranged from 0.02 \pm 0.01 to 0.10 \pm 0.10 mg/L (Table 1). According to local drinking water source protection regulations, all of the sampling sites were located in the second protection zone, in where water quality should not be worse than Class II. Average TP concentrations in most catchments were Class II whereas in SXK was Class III (Table 1). Land-use/land-cover data were provided by the local land ministry and was subsequently classified into four categories of agricultural, forest, developed, and unused lands, whose proportions were 3.3-53.0%, 44.7-95.4%, 0.4-3.7%, and 0.0-2.3%, respectively

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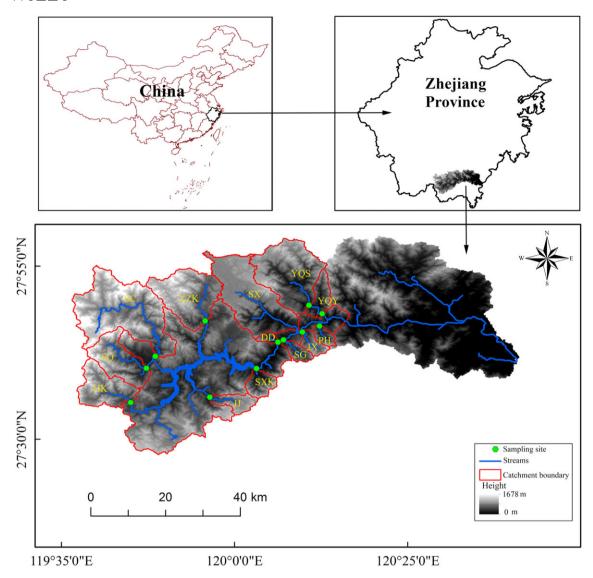


FIGURE 1 Geographic location of study area and sampling sites within Shanxi watershed

(Table 1). Land-use distribution did not conform to a normal distribution on the basis of kurtosis and skewness (Table 1).

2.2 | Detrended fluctuation analysis

DFA was developed to assess the occurrence of long-range correlations (persistence) in noisy and nonstationary data with embedded noisy and unknown trends (Peng, Havlin, Stanley, & Goldberger, 1995). For a time series of riverine P concentrations x(i), i = 1, 2, ..., n, DFA comprises three calculation steps.

The first step is the establishment of a new "profile" series y(i), i = 1, 2, ..., n by calculating the accumulated deviation of x(i).

$$y(i) = \sum_{i=1}^{n} [x(i) - \overline{x}], \qquad (1)$$

where x(i) is the *i*th element in the time series and \overline{x} is the mean of all x(i).

The second step is the removal of the local trend. The new "profile" series y(i) is divided into m = int[n/s] non-overlapping segments v(i), i = 1, 2, ..., m of equal length *s* (scale length). A local trend for each segment v(i) is determined as the best linear fit, and the local trend series is p_v (i). Then, a detrended new series y_v (i) is calculated by

$$y_{v}(i) = y(i) - p_{v}(i).$$
 (2)

The third step is the calculation of the root mean square fluctuation of the detrended series $F^2(n, v)$ by subtracting the departures y_v (*i*) from the profile:

$$F^{2}(n, \mathbf{v}) = \frac{1}{n} \left[\sum_{i=1}^{n} \left(\mathbf{y}(i) - \mathbf{y}_{\mathbf{v}}(i) \right)^{2} \right]^{2}.$$
 (3)

This procedure is repeated for all time scales (n lengths of segments) to provide a relationship between F(n) and the length of segments:

$$F(n) = \frac{1}{m} \left[\sum_{v=1}^{m} F^2(v, n) \right]^{\frac{1}{2}}.$$
 (4)

A linear relationship between F(n) and n in log–log space ($F(n) \propto n^{\alpha}$) indicates the presence of scaling:

TABLE 1 Shanxi watershed riverine TP concentrations and land-use types across the 13 catchments

	TP	Land-use classification (%)				Catchment		
Catchment	Average concentration (mg/L)	Water quality grade	Agricultural land	Forest land	Developed land	Unused land	area (km ²)	
SQ	0.04 ± 0.03	ll ^a	11.1	87.2	0.9	0.8	119.1	
SC	0.02 ± 0.01	II	3.3	95.4	0.4	1.0	115.6	
НК	0.03 ± 0.02	II	18.5	79.7	0.8	0.9	176.9	
JJ	0.03 ± 0.02	II	26.1	70.6	3.3	0.0	48.7	
XZK	0.04 ± 0.04	II	18.6	78.5	0.6	2.3	284.2	
SX	0.08 ± 0.05	II	41.1	53.6	3.7	1.6	243.8	
DD	0.04 ± 0.03	II	38.3	59.2	2.4	0.2	10.4	
JX	0.03 ± 0.03	II	38.3	60.0	1.7	0.0	25.7	
PH	0.05 ± 0.03	II	42.3	55.9	1.8	0.0	19.8	
SXK	0.10 ± 0.10	III	19.6	77.2	2.8	0.3	38.8	
SGX	0.08 ± 0.04	II	53.0	44.7	2.3	0.0	24.0	
YQT	0.03 ± 0.02	II	32.2	64.9	1.8	1.2	262.4	
QYS	0.05 ± 0.03	II	32.7	64.7	1.5	1.1	182.3	
М	0.05	-	28.8	68.6	1.9	0.7	119.4	
SD	0.02	-	14.1	14.5	1.0	0.7	100.6	
CV (%)	49.18	-	48.9	21.1	56.2	100.2	84.3	
Kurtosis	0.50	-	-0.56	-0.53	-0.78	0.04	-1.39	
Skewness	1.19	-	-0.18	0.27	0.30	0.77	0.48	

Note. TP = total phosphorous.

^aIn the environmental guideline of national quality standards for surface waters, China (GB3838-2002), TP concentrations (mg/L) <0.02, <0.1, <0.2, <0.3, and <0.4 indicate Classes I, II, III, IV, and V, respectively.

 $\log F(n) \approx \alpha \log n, \tag{5}$

where α is the slope.

For 0 < α < 1, the slope α is identical to the well-known Hurst exponent (Onderka et al., 2012). An $\alpha \approx 0.5$ indicates that the time series is white noise, whereas α > 0.5 or 0.5 < α indicates that the time series contains long-term persistence or anti-persistence, respectively.

3 | RESULTS AND DISCUSSION

3.1 | Relationships between land use and riverine P concentration

In many watersheds, riverine phosphorus originating from non-point sources is strongly impacted by land use (Chen, Hu, Wang, et al., 2015). Due to the lack of a normal distribution (Table 1), Spearman rank correlation analysis was adopted to examine relationships between riverine phosphorus concentration and land use in the 13 catchments (Table 2). Riverine TP concentration showed a weak to moderate positive correlation with agricultural land (R = .51, p = .07) and developed land (R = .51, p = .08). Fertilizer and manure P is applied

to cultivated lands as a nutrient source and is susceptible to run-off/ leaching to the river system in particulate and dissolved forms. As sewage collection and treatment are still incomplete in the rural areas and small communities, developed land is also a potential source of P to the river system. In contrast, forest land is generally recognized as a sink for non-point source pollution because the vegetation and extensive rooting system of forests have a strong ability to absorb and assimilate nutrients (Miserendino et al., 2011; Tu, 2011). In Shanxi watershed, riverine TP concentration had a negative correlation with forest land (R = -.54, p = .06), consistent with forests serving as a sink for P inputs. The unused lands, consisting of grass land, pit-ponds, and barren land, showed no significant relationship with riverine TP concentration (R = -.03, p = .92). Similarly, there was no significant correction between riverine TP concentration and catchment area (R = -.15, p = .61).

In general, the agricultural and developed lands appear to be the primary sources of TP in the Shanxi watershed, whereas forest lands appear to be a sink. These relationships between land use and riverine phosphorus concentration are consistent with the previous studies (Miserendino et al., 2011; Tu, 2011; Chen, Hu, Wang, et al., 2015; Huang, Chen, Zhang, Zeng, & Dahlgren, 2014).

 TABLE 2
 Relationships between TP concentration, persistence effect exponent α , and land use

Land use		Agricultural land	Forest land	Developed land	Unused land	Catchment area
TP concentration	R	.51	54	.51	03	15
	p	.07	.06	.08	.92	.61
Exponent α	R	47	.48	67	.17	.04
	p	.11	.10	.01	.58	.89

3.2 | Relationship between land use and riverine P persistence effect

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The release and migration of legacy nutrients are influenced by many factors (i.e., soil, land use, climate, landform, and river/groundwater networks; Onderka et al., 2012; Worrall & Burt, 1999), and therefore, the persistence effect for riverine P in different catchments may be appreciably different. The persistence effect exponent α for the 13 catchments calculated by DFA ranged from 0.60 to 0.81 indicating persistence effects for riverine P across all catchments (Figure 2). We defined 0.5 < α < 0.65, 0.65 ≤ α < 0.75, and 0.75 ≤ α < 1 as weak, moderate, and strong persistence effects, respectively (Dong et al., 2016).

In Shanxi watershed, catchments with strong, moderate, and weak persistence effects accounted for 46.2%, 38.5%, and 15.3% of the catchments, respectively (Figure 2). There was no significant relationship of catchment area with the riverine P persistence effect (R = .04, p = .89). The area of a catchment may affect hydrologic networks (surface runoff and groundwater), and therefore, it might be expected that larger catchments have longer flowpaths resulting in greater transient storage capacity and travel times. The lack of a strong relationship with catchment size suggests that land use/land cover may be an important factor regulating the persistence effect.

The relationship between land use and the persistence effect exponent α for riverine P was assessed using Spearman rank

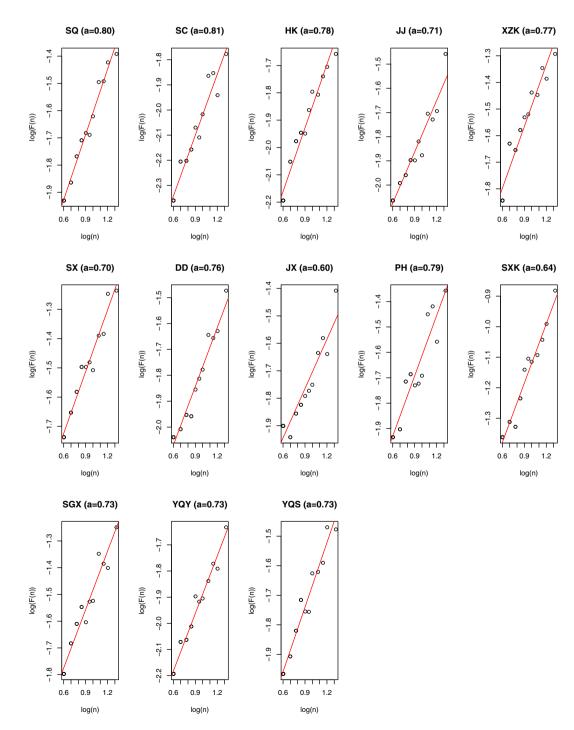


FIGURE 2 Persistence effect exponent of total phosphorous time series

correlation analysis (Table 2). The exponent α had a weak negative relationship with agricultural land (R = -.47, p = .11). Phosphorus application to these agricultural lands through fertilizer and manure additions may be strongly retained by these iron oxide-rich soils (PO₄ sorption) and therefore not efficiently transferred to the river system. Before buffering capacity of these agricultural lands reaches a threshold (saturation) level (Doody et al., 2016; McDowell et al., 2003), this strong retention capacity may change very slowly over time resulting in any long-term changes in riverine P concentrations not being reflected in the 7-year riverine P record (Mei et al., 2016; Van Meter & Basu, 2015). Some field observations indicated that a 48-90% reduction in Olsen-P levels in agricultural soils required 8-14 years (Sharpley et al., 2013) and subsequently required a decade or more to "draw down" soil P reserves to levels where dissolved P in run-off was substantially reduced (Jarvie et al., 2013). In contrast, the persistence effect exponent α had a significant negative relationship with developed land (R = -.67, p = .01). The impervious area associated with developed land is high resulting in the transport of phosphorus mainly via overland run-off. The strong mobilization capability results in the retention of phosphorus in the developed land area were weak. The persistence effect exponent α had a positive relationship with forest land (R = .48, p = .10). Due to the high vegetative cover and extensive rooting system of forests, these ecosystems have a strong capacity to enhance nutrient retention and reduce nutrient mobilization and contribute to legacy P that is able to influence riverine P persistence (Chen, Hu, Guo, et al., 2015). The source strength and retention ability of P in unused lands were insignificant, resulting in no appreciable influence on the persistence effect for riverine P (R = .17, p = .58).

Conceptually, the catchment-scale persistence effect for riverine P is determined by both retention capacity (e.g., biomass uptake and soil PO₄ sorption) and migration efficiency (e.g., surface run-off and groundwater networks). For a given catchment, a larger land area having strong retention ability and lower migration efficiency would result in a stronger persistence effect for riverine P. In this study, the effects of catchment size did not play an obvious role in regulating the riverine P persistence effect. However, the contrasting land uses of forest (positive—R = .48) versus developed land (negative—R = -.67) highlight the potential importance of land use in regulating the P persistence effect for riverine P may be influenced by many factors in addition to land use, such as soils, climate, landform, and river/groundwater network characteristics (Mei et al., 2016).

3.3 | Implication of persistence on riverine phosphorus dynamics

Due to the persistence effect, riverine P concentrations and fluxes are the product of both current year P input and legacy P inputs, leading to a potential time lag in the response of riverine P concentrations to improvements in watershed nutrient management and pollution control. Neglecting the role of the persistence effect may lead to an underestimation of the effectiveness of past mitigation efforts and mislead future watershed nutrient management and pollution control activities (Worrall & Burt, 1999; Onderka et al., 2012). For a catchment with a weak persistence effect, the water quality response to implementation of conservation measures for source control should be highly effective. In contrast, catchments having a strong persistence effect will have a delayed response in water quality to implementation of conservation measures. In this case, source control would be less effective, and sink control by means of plant uptake/harvest and interception processes (e.g., riparian buffer strips and wetlands) would result in a stronger impact on stream water quality. Furthermore, knowledge of the persistence effect allows environmental managers to inform the public of how conservation measures are likely to impact water quality and set reasonable expectations. A comprehensive understanding of the persistence effect for riverine P is essential to watershed nutrient management and pollution control efforts to improve phosphorus-use efficiency and riverine P exports (Condron, Spears, Haygarth, Turner, & Richardson, 2013; Sharpley et al., 2013).

Understanding the persistence effect for riverine P is a scientific research area of great importance that will have valuable practical applications for watershed nutrient management. The generation, transformation, and migration components of non-point source pollutants are very complex, and it is currently not possible to directly monitor all these processes at the watershed scale. Hence, the quantitative responses of riverine water quality to nutrient inputs, climate, and land use, which are an important basis for implementation of conservation measures, are mainly predicted by watershed models (i.e., AGNPS and SWAT) (Nasr et al., 2007; Van Meter & Basu, 2015). However, these models generally assume that current riverine nutrient concentration and flux result directly from current human activities (i.e., nutrient input and land-use change) and natural factors (i.e., rainfall and temperature). Thus, the potential impacts from legacy activities in previous years are generally not considered. Most lumped watershed models (i.e., SPARROW and GlobalNEWS), as well as the net anthropogenic nitrogen and phosphorus inputs mass balance approach, assume a steady-state nutrient cycle, either on a yearly basis or over a multiyear period, such that stream export is a fixed percentage of net annual inputs (Hong et al., 2012; Van Meter & Basu, 2015). Obviously, the legacy effect (or persistence effect) must be taken into account in watershed nutrient modelling efforts to fully inform watershed management activities. The few studies that have made efforts to quantify legacy P effects found that it is a very important component of riverine nutrient fluxes and that additional future research is necessary to better understand the mechanisms involved across a wide range of watershed conditions (Chen, Hu, Guo, et al., 2015; Chen, Hu, Wang, et al., 2015; Chen et al., 2017).

4 | CONCLUSIONS

This study assessed the influence of land use on the persistence effect for riverine P in a drinking water source reservoir in eastern China. The persistence effect exponent α for the time series of riverine P in 13 catchments ranged from 0.60 to 0.81, indicating strong, moderate, and weak persistence effects (P legacy effects) for 46.2%, 38.5%, and 15.3%, respectively, of the catchments. Thus, the 13 catchments all demonstrated appreciable P legacy effects on current and future riverine P concentrations and loads. The persistence effect for riverine P is determined by retention ability (biogeochemical legacy) and migration ¹²⁴ ∣ Wiley

efficiency (hydrological legacy). A catchment with strong retention capacity (e.g., biomass uptake and soil PO₄ sorption) and low migration efficiency (e.g., groundwater vs. surface run-off flowpaths) results in stronger persistence effects for riverine P. Given the moderate to strong P persistence effects in Shanxi watershed, sink controls (e.g., riparian buffers and wetlands) would be expected to have a stronger immediate impact on riverine P concentrations than source controls (e.g., nutrient management). A comprehensive understanding of the persistence effect for riverine P is essential to guide watershed nutrient management activities and set a reasonable timeline for meeting pollution control targets.

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