Development and application of watershed-scale indicator to quantify non-point source P losses in semi-humid and semi-arid watershed, China

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A B S T R A C T

Quantifying non-point source (NPS) phosphorus (P) pollution loads is essential for watershed nutrients management. This study intended to develop a NPS P indicator which (1) was suitable in semi-humid and semi-arid watersheds of Northern China; (2) integrated the key NPS P loss factors and constructed them in a simple and physically understandable way and (3) kept P loss forms distinctively separate. An inverse distance-based delivery ratio was proposed to count for the P delivery efficiency from source to watercourses. We applied this P indicator in Luan River Watershed (LRW) of northern China under typical hydrological years and seasons. Results demonstrated that this NPS P indicator predicted reasonable NPS TP loads using simple methods and readily obtainable inputs. The sub-watersheds located in the south of LRW were recognized as the high risk areas of NPS P loss to Panjiakou reservoir. The upland and paddy fields near the river channels were particularly posing high risk and thus should be treated with prioritized management practices such as soil conservation and recommended fertilization. Rainfall-runoff related variables rather than P source variables explained more of the spatial variation in NPS P load and percentages. Using this tool could give policy makers insight into the component and location of NPS P pollution that needs to be the focus of policy at watershed scales before sophisticated studies were conducted in smaller scales.

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1. Introduction

Phosphorus (P) is widely regarded as a limiting nutrient for primary production in aquatic systems, and in excess it may impair water quality by accelerating the production of algae and aquatic plants (Correll, 1998). Reducing nutrient inputs into water bodies is considered as the most effective strategy for controlling water pollution and sustaining high ecological status (Conley et al., 2009). Owing to the effective control of point sources in the past decades, non-point sources (NPS), particularly agricultural activities have become the leading contributor to water quality degradation worldwide. In 2010, a national pollution census first indicated that in China the agriculture’s contribution to total P loads in receiving waters was up to 67.4% (Zhang, 2010a), which has raised extensive interests on NPS pollution from both government and scientists.

Quantifying loads of NPS pollution and its components is essential for watershed or regional nutrient management planning. Approaches including export coefficient models (ECM) (Ding et al., 2010; Johnes, 1996) and mechanistic models (e.g., SWAT, HSPF) (Mishra et al., 2007; Shen et al., 2014) have been applied world widely in NPS pollutant load estimation. The ECM has the advantage of requiring less data and has fewer parameters, but doesn’t consider the influence of spatial heterogeneity of rainfall and underlying surfaces (Ding et al., 2010). The mechanistic models can provide accurate results, but often encounter difficulties in application because of their complicated structures and strict requirements for the input data (Zhang, 2010b). This phenomenon was particularly prominent in countries without long term and spatially dense monitoring data and basic or empirical field studies, such as China (Ongley et al., 2010; Shen et al., 2012).

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The Chinese researchers have developed several local NPS P quantification methodologies (Chen et al., 2008, 2010; Han et al., 2011; Yang et al., 2012). Chen et al. (2008, 2010) proposed a model framework to analyze the biogeochemical P flows in agroecosystem on the basis of emission inventory and full P balance calculation. The P inputs included P brought by mineral fertilizer, livestock manure, organic fertilizers, precipitation, seedling and irrigation, and the outputs included plant uptake, runoff and leaching. Han et al. (2011) introduced a concept of net anthropogenic phosphorus accumulation and calculated it in Beijing by deriving data from fertilizer use, human food and animal feed, non-food P and riverine P. Yang et al. (2012) estimated the potential P loads from cropping, animal production and rural living in a typical county of Northern China Plain. These methodologies were mostly established on a P-balance basis and the outputs were generally indicative of the potential losable P in source. Whereas the NPS P loss potential was not determined by source potential alone, but also by transport potential (Gburek et al., 2000). Few of these studies took consideration of the role of P transport factors (e.g., runoff, field to stream distances) in determining P delivery routes and efficiency, except for Yang et al. (2012) who used scenarios of delivery coefficients to count the likely P delivery ratios from source areas to watercourses. In addition, the terrain P enters watercourses as dissolved P (DP) carried by surface and subsurface runoff and particulate P (PP) associated with eroded soils (Sharpley et al., 2003). They vary greatly in their relevance to aquatic eutrophication because DP is readily bio-available and PP potentially bio-available (Bostrom et al., 1988). Separating TP loads into these forms and pathways would allow a detailed examination of the P load composition and of the factors influencing both the relative and absolute magnitude of these P components.

The main objective of this work was to develop an indicator to quantify the NPS P losses by integrating a range of contributory factors determining NPS P loss from terrain to water within a single calculation system. Our target is an estimator which follows the principle that the coincidence of high source potential and high transport potential determined the actual high NPS pollution risk; keeps P transport forms (e.g., DP and PP) and processes (e.g., surface runoff, subsurface runoff and soil erosion) distinctly separate; and is structured in a way that the individual parameters of calculation have physical meaning. A second objective of this study was to apply NPS P indicator in a semi-humid and semi-arid watershed of China and characterize the variations in NPS P predictions in response to hydrological condition changes in order to obtain important insights into P management in watershed scale.

2. Methods and methodology

2.1. Site description

Luan River watershed (LRW) is a sub-basin of Hai River Basin (HRB) in Northern China. The Luan River originates near the border between Hebei Province and Inner Mongolia, flowing through plateau, mountain and plain from west to east. This study mainly investigated the mountainous part of LRW located at the upstream of an important Panjiakou Reservoir (Fig. 1).

Due to the data availability, nine sub-watersheds with hydrometric stations installed in the outlets were applied with LRW NPS P indicator (LRW-Pi). These sub-watersheds were drained by Budeng river, Yixun river and Yimatu river in the north, by Wulie river, Lao-niu river and Xingzhou river in the middle and by Pu river, Liu river and Sa river in the south (Fig. 1). Sub-watershed boundaries were delineated by ArcSWAT tool in ArcGIS 9.3 software (ESRI, Inc., Redlands, CA) based on a digital elevation model (DEM) expressed as a 90 m raster and the geographic coordinates of hydrometric stations.

The LRW is characterized by a semi-humid and semi-arid monsoon climate and an annual average rainfall of 520 mm. Influenced by the topography, rainfall decreases from the south to the north. More than half of the annual rainfall occurred between June and September. Percentages of different soil types or land uses in each

![Fig. 1. Locations of Luan River Watershed and the studied sub-watersheds.](image)
sub-watershed were derived from the land use and soil maps. The dominant soil types are brown or cinnamon soil depending on sub-watersheds, and the topsoil texture is mainly loamy sandy according to particle size compositions (Table 1). Forest covers the largest area (34.8–65.6%) in all sub-watersheds, followed by grass land and dry land, and paddy land covers the least (Table 1). Dry lands and paddy lands are generally fragmented and located in hill sides, terraces and flood plains along the river channel and the regular upland crops are corn and vegetables.

The excessive application of mineral fertilizers has long been recognized as an important contributor to NPS nutrient pollution in Chinese agricultural lands. Recently livestock production and rural living have been identified as another one (Du et al., 2014; Yang et al., 2012). In LRW, livestock (mainly pigs, poultry, cattle and sheep) either graze in grassland or are raised in individual farms or centralized feedlots. The latter accounts for the majority of the total stock, being from 85 to 100% depending on counties and animal types. The waste produced by rural living and livestock production are either treated in limited numbers of disposal facilities, used for biogas producing, or applied to cropland as fertilizers, leaving the rest being washed into watercourses by storm runoff or stable-cleaning water (Zhu, 2011). This P indicator only took the waste applied to land into consideration as a source of NPS pollution.

### 2.2. Development of LRW-PI

Non-point source P loss is a function of source factors (e.g., soil P status, fertilizer and manure application) and transport factors (e.g., erosion, surface and subsurface runoff, connectivity between the land and the waterbody) (Drewry et al., 2011). Processes determining NPS P loss forms and magnitudes included in this methodology are P solubilization and PP transfer. PP transfers were closely associated with erosion of soil particles or manure dry matter; while DP transfers represented the solubilization of DP from soils, fertilizers and manure and their mobilization driven by surface and subsurface runoff. These processes were used to estimate NPS P loads mobilized in and ready to leave each calculation cell, which represented the potential losable P from terrain sources. A parameter reflecting the degree of hydrological connectivity between source cells and surface waters were further multiplied to obtain the final delivered PP and DP loads. The total NPS P loads entering watercourses were quantified as the sum of delivered PP and DP. Tables 2 and 3 respectively listed the components and equations for LRW-PI computation and Table 4 listed the data sources. All inputs and outputs were eventually expressed as 90 m raster files and the raster calculations were performed by raster calculator in ArcGIS 9.3 software.

#### 2.2.1. Particulate P loads leaving the cell

Particulate P leaving the cells included P associated with eroded sediments and manure solids mobilized from the source to the cell edge. We quantified the PP loss from soil (PPsolid, kg ha−1) by multiplying eroded sediment rates (A, ton ha−1) calculated by revised universal soil loss equation (RUSLE) (Renard et al., 1997; Wischmeier and Smith, 1978), topsoil TP concentrations (TPtop, mg kg−1) (Vadas et al., 2009) and P enrichment ratio (ER, unitless) (Sharpley et al., 2002; Vadas et al., 2009) (Eq. (1) in Table 3). The RUSLE inputs include rainfall erosivity (R), soil erodibility (K), slope (S), slope length (L), crop cover-management factor (C) and support practices factor (P) which were respectively derived from long term daily rainfall data, soil maps with the attributes of particle sizes and organic matter, digital elevation model, land cover and management practices (Eq. (4) in Table 3). Considering the local applicability, this study adopted a daily rainfall erosivity model developed for the mountainous area in Beijing (Bi et al., 2006) to estimate the rainfall erosivity factor. The soil erodibility factor K followed the method proposed by Williams et al. (1983) in EPIC model in view of data availability. The length and slope factor (LS) was calculated by the equations from Liu et al. (1994) and McCool et al. (1987). The cover-management factor C and support practice factor P adopted values from previous studies in the similar region of China (Bi et al., 2006; Men et al., 2007). Since eroded sediments were enriched with P compared to source surface soil due to the preferential transport of finer, more sorptive soil and organic
Table 2
Components of LRW-PI computation.

<table>
<thead>
<tr>
<th>Components</th>
<th>Determination method</th>
<th>Data required</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particulate P leaving cells</td>
<td>Revised Universal Soil Loss Equation</td>
<td>DEM, rainfall, land use, soil map, management practices</td>
</tr>
<tr>
<td>Sediment loss rates</td>
<td></td>
<td>Soil OlsenP and organic matter, enrichment ratio calculated based on sediment loss rates</td>
</tr>
<tr>
<td>Sediment P concentration</td>
<td>Empirical equations to determine sediment P concentration from soil total P and using routine soil test information to estimate soil total P</td>
<td>Sediment P concentrations; sediment loss rates</td>
</tr>
<tr>
<td>Particulate P loads contributed from soils</td>
<td>Empirical coefficients to estimate PP loads from manure</td>
<td></td>
</tr>
<tr>
<td>Particulate P loads contributed from manure</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dissolved P leaving cells</td>
<td>Base flow separation</td>
<td>Measured daily flow volumes, sub-watershed area</td>
</tr>
<tr>
<td>Depths for surface runoff, subsurface runoff and total runoff</td>
<td>Classification according to frequency distribution</td>
<td>Total runoff depths Depths for surface runoff, subsurface runoff and total runoff</td>
</tr>
<tr>
<td>Total runoff depth rank</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Distribution ratios of surface runoff or subsurface runoff in total runoff</td>
<td></td>
<td>Soil Olsen P, empirical coefficients</td>
</tr>
<tr>
<td>Dissolved P concentration contributed from soil</td>
<td>Empirical coefficients to estimate dissolved P concentration in both surface and subsurface runoff from topsoil test P</td>
<td></td>
</tr>
<tr>
<td>Dissolved P loads contributed from manure/fertilizer</td>
<td>Estimation using census data, empirical coefficients and runoff data</td>
<td>P fertilization rates, livestock numbers and rural population, P excretion coefficients, manure land-applied rates, P utilization rates, total flow ranks, distribution ratios of runoff/P concentration</td>
</tr>
<tr>
<td>Cell to water delivery</td>
<td>Inverse distance weights</td>
<td>River networks</td>
</tr>
<tr>
<td>Distance to streams</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

particles, ER was used for estimating TP in eroded sediments (Eq. (3) in Table 3). Topsoil TP concentrations were not readily available and were calculated based on soil organic matter and OlsenP concentrations using Eq. (2) in Table 3.

The PP loss contributed by manure application was also considered presuming that part of the applied fecal P was in particulate form. A regional survey in LRW suggested that around 23% of the urine and 40% of feces excreted by rural dwellers and livestock were applied to crop lands (Bai, 2010). The manure PP leaving the cell (PPman, kg ha\(^{-1}\)) was estimated by summing the land-applied PP in each county according to census data and empirical coefficients cited from worldwide studies, and then dividing by the cropland areas in each county (Eqs. (5) and (6) in Table 3; Table 5). Other land uses were considered to be without manure and fertilizer P inputs.

2.2.2. Dissolved P loads leaving the cell

Dissolved P leaving the cell comprised the soluble parts of soil P, fertilizer P and manure P which were available for delivery from source area to cell edges by surface and subsurface runoff. DP contributed by soils was expressed as the DP mass extracted from soil by runoff, and that from manure and fertilizer were obtained by apportioning the total losable DP from manure or fertilizer to surface and subsurface pathways according to distribution ratios of runoff and P concentration between two pathways. Since runoff parameters were involved in the computation, they were introduced prior to load estimation.

2.2.2.1. Runoff analysis. Runoff analyses were performed on the daily flow records during the 1995–2008 period in studied sub-watersheds (Tables 2 and 4). A digital filter technique was first applied to separate the surface and subsurface runoff from the total flow (Nathan and McMahon, 1990). Volumes of the total, surface and subsurface flows were then aggregated and divided by the sub-watershed area to obtain runoff depths (referred as RDf, RDsurf and RDsub). Ratios of surface or subsurface runoff depths in total runoff depths were also calculated to reflect the runoff distribution between surface and subsurface pathways (DRsurf, DRsub). RDsub and RDsurf would be used for the estimation of DP loss contributed by soils, and RDf, DRsurf and DRsub for the estimation of DP loss by manure and mineral fertilizer. The RDf values were not directly used for the computation but were converted into a unitless parameter named total flow ranks (TF\(_{\text{rank}}\)). The TF\(_{\text{rank}}\) were assigned with a series of values ranging from 0 to 1 and were determined according to the distribution of the annual or seasonal RDf in all sub-watersheds during 1995 and 2008. The RDf values at quantiles of 5%, 15%, 30%, 45%, 60% and 75% were set as the boundaries to divide RDf into 7 classes. Mean values of the maximum and minimum quantiles in each distance class were then assigned as the TF\(_{\text{rank}}\) value of each runoff class, which were respectively 0.025, 0.1, 0.225, 0.375, 0.525, 0.675 and 0.875. This parameter reflected the relative magnitudes of a certain RDf among studied sub-watersheds in a certain time period, allowing the comparisons of DP losses across sites and time.

2.2.2.1.1. Soil DP mobilized within each cell. Soil DP mobilized within each cell (DPsol, surf, DP sol, sub, kg ha\(^{-1}\)) was estimated by multiplying runoff depths in each pathway and runoff DP concentrations released by soils (Eqs. (7) and (8) in Table 3). We used a constant coefficient of 0.004, as many other studies did (Sharpley et al., 2002; Vadas et al., 2009), to convert soil extractable P concentrations (Olsen\(\text{P}_{\text{surf}}, \text{mg kg}^{-1}\)) to runoff DP concentrations (mg L\(^{-1}\)). Another coefficient, \(\text{DR}_{\text{surf}}\), was used to describe P concentration ratio between subsurface and surface pathways. This study assigned a constant value of 1, assuming no difference between P concentrations in two pathways.

2.2.2.1.2. Fertilizer DP mobilized within each cell. China consumed high amount of fertilizers but has a relatively low utilization efficiency, being 10–20% for P (Chen et al., 2008). We estimated the losable fertilizer DP loads in the cropland of each county and then allocated them to different runoff pathways (Eqs. (9) and (10) in Table 3). \(P_{\text{fert}}\) represented the amount of mineral fertilizer P applied to the unit cropland area. It was calculated in county level using census records and cropland area (upland and paddy land). The calculation of mobilized fertilizer DP presumed: (1) all fertilizer P was in soluble form; (2) methods and timing of P application were uniform in all sub-watersheds; (3) only 25% of the applied fertilizer P could be taken up by crops or retained by soils annually and the rest 75% would be ready for release and (4) 60% of the fertilizer P was applied during the flood season which covered part of the crop season and 40% during the non-flood season. We further used the
Table 3
Equations for LRW-PI computation.

P loads delivered to cell edge

| PP losses from soil | PP_{soi} = TP_{soi} \times \text{ER} \times A \times 0.001 | (1) |
| DP_{soi} = [13 + (2.7 \times \text{OM}_{soi}) + (0.06 \times \text{OlsenP}_{soi})]^2 | (2) |
| \ln(\text{ER}) = 2.2 - 0.25 \times \ln(1000 \times A) | (3) |

A = \text{RESCP}

• PP_{soi}: PP loads leaving the cell contributed by soils (kg ha\(^{-1}\))
• TP_{soi}: topsoil TP concentrations (mg kg\(^{-1}\)) estimated from topsoil OlsenP concentrations (OlsenP_{soi}, mg kg\(^{-1}\)) and organic matter contents (OM_{soi}, %)
• ER: P enrichment ratios in eroded soils relative to source soils
• A: eroded sediment rates (ton ha\(^{-1}\))

from manure

PP_{man} = 40 \times PD_{focal}/\text{AREACrop} | (5) |

PP_{focal} = N_{human} \times (\text{ECU}_{human}) \times (1 - (\text{PER}_{human}) \times \sum_{i=1}^{n} (N_{livestock})_{i} \times (\text{ECU}_{livestock})_{i} \times (1 - (\text{PER}_{i})) | (6) |

• i: livestock types
• PP_{man}: manure PP loads ready to leave the cells (kg ha\(^{-1}\))
• PP_{focal}: particulate fecal P produced in each county (kg)
• 40%: percentage of excreted feces applied to land
• PER_{human}: percentage of water extractable P in fecal P (%) (Table 5)
• AREACrop: cropland area including dry land and paddy land in each county (ha)
• N_{human}, N_{livestock}: numbers of rural dwellers and livestock
• ECU_{human}, ECU_{livestock}: P excretion coefficients in feces (Table 5)

DP loads from soil

DP_{soi,ur} = OlsenP_{soi} \times 0.004 \times DR_{soi} \times 0.01 | (7) |

DP_{soi,sub} = OlsenP_{soi} \times 0.004 \times DR_{soi} \times DR_{sub} \times 0.01 | (8) |

DP_{soi,ur,sub}: soil DP loads leaving the cell via surface or subsurface runoff (kg ha\(^{-1}\))

• DR_{soi}: depth of surface or subsurface runoff (mm)
• DR_{focal}: ratio of P concentration in subsurface runoff to that in surface runoff. DR_{focal} equals 1 in this study
• 0.01: ensures kg ha\(^{-1}\) unit for runoff DP

from fertilizer

DP_{focal} = P_{focal} \times 75% \times TF_{focal} \times DR_{focal} | (9) |

DP_{focal} = P_{focal} \times 75% \times TF_{focal} \times DR_{focal} | (10) |

• DP_{focal}: fertilizer DP loads leaving the cell via surface or subsurface runoff (kg ha\(^{-1}\))

• P_{focal}: fertilizer P application rates in unit cropland area (kg ha\(^{-1}\))

• TF_{focal}: total flow ranks, values ranging from 0 to 1.

• DR_{focal}: DR_{soi}: distribution ratio of surface or subsurface runoff in total runoff.

• 75%: percentage of losable fertilizer P recommended by Yang et al. (2012)

from manure

P_{man} = N_{human} \times (\text{ECU}_{human}) \times \sum_{i=1}^{n} (N_{livestock})_{i} \times (\text{ECU}_{livestock})_{i} | (11) |

DP_{focal} = N_{human} \times (\text{ECU}_{human}) \times (\text{PER}_{human}) \times \sum_{i=1}^{n} (N_{livestock})_{i} \times (\text{ECU}_{livestock})_{i} \times (\text{PER}_{i}) | (12) |

DP_{man,ur} = P_{man} \times 75\% \times TF_{man} \times DR_{man} | (13) |

DP_{man,ur,sub} = DP_{man} \times 75\% \times TF_{man} \times DR_{sub} | (14) |

• P_{man}: total urinary P produced in each county (kg)

• P_{focal}: dissolved fecal P produced in each county (kg)

• ECU_{human}, ECU_{livestock}: P excretion coefficients in urine (Table 5)

• DP_{man}: manure DP applied to cropland (kg ha\(^{-1}\))

• 23%: percentage of excreted urine applied to land

• 40%: percentage of excreted feces applied to land

• DP_{man,ur}, DP_{man,ur,sub}: Manure DP loads leaving the cell via surface or subsurface runoff (kg ha\(^{-1}\))

P loads delivered from cell edge to watercourses

\[ D = \frac{100}{(d_{max})} \]  

• (d_{max}): the highest distance in distance class i.

• 100: a constant to constrain the order of magnitudes.

\[ \text{TPP} = (PP_{soi} + PP_{man}) \times D \]  

\[ \text{TPD}_{ur} = (DP_{soi,ur} + DP_{man,ur} \times \text{TF}_{ur} \times \text{DR}_{ur}) \times D \]  

\[ \text{TPD}_{sub} = (DP_{soi,sub} + DP_{man,sub} \times \text{TF}_{sub} \times \text{DR}_{sub}) \times D \]  

\[ \text{TPD} = \text{TPP} + \text{TPD}_{ur} \]  

\[ \text{TPD}_{ur} + \text{TPD}_{sub} \]  

\[ \text{TPD}_{ur} + \text{TPD}_{sub} \]  

• \text{TPP}, \text{TPD}: total delivered PP or DP loads from each cell (kg ha\(^{-1}\)).

• P, D: total delivered P loads from each cell (kg ha\(^{-1}\)).

products of DR_{soi}, TF_{soi} and runoff distribution ratio per pathway to apportion the losable fertilizer DP into surface and subsurface pathways.

2.2.2.1.3. Manure DP mobilized within each cell. Manure DP was assumed to consist of urinary P and the water-soluble part of fecal P. The excreted urine P (P_{urine}, kg) and dissolved fecal P (DP_{focal}, kg) were estimated based on census data regarding rural population and livestock production and empirical excretion coefficients (Eqs. (11) and (12) in Table 3). As stated previously, only 23% of the urine and 40% of the excreted feces were applied to crop lands. This calculation also presumed that 75% of the applied DP_{man} was considered to be losable to watercourses after crop uptake and soil retention. The losable manure DP was distributed into each pathway as did for fertilizer DP (Eqs. (14) and (15) in Table 3).
### Table 4
Types, sources and description of the data used in the case study.

<table>
<thead>
<tr>
<th>Data type</th>
<th>Data source</th>
<th>Data description</th>
</tr>
</thead>
<tbody>
<tr>
<td>DEM</td>
<td>International scientific data service platform, Chinese Academy of Science</td>
<td>A grid size of 90 m × 90 m</td>
</tr>
<tr>
<td>Soil P</td>
<td>Soil and fertilizer institute of Chengde city, Hebei province</td>
<td>Soil Olsen P in 90 m × 90 m grid</td>
</tr>
<tr>
<td>Soil properties</td>
<td>National second soil survey</td>
<td>Soil texture, organic matter (1:250,000)</td>
</tr>
<tr>
<td>Land Use</td>
<td>Data sharing infrastructure of earth system science, China</td>
<td>Land uses classification (1:100,000)</td>
</tr>
<tr>
<td>Runoff</td>
<td>Chengde branch of Hebei provincial bureau of hydrology and water resources survey</td>
<td>Daily runoff from 9 hydrometric stations (1995–2008)</td>
</tr>
<tr>
<td>P inputs</td>
<td>Annual census published by each county’s statistical bureau</td>
<td>Fertilizer application, rural population, livestock production (2005)</td>
</tr>
</tbody>
</table>

### Table 5
P excretion coefficients cited from multiple literature sources.

<table>
<thead>
<tr>
<th>Categories</th>
<th>ECU&lt;sup&gt;a&lt;/sup&gt; (kg P individual&lt;sup&gt;−1&lt;/sup&gt; yr&lt;sup&gt;−1&lt;/sup&gt;)</th>
<th>ECF&lt;sup&gt;b&lt;/sup&gt; (kg P individual&lt;sup&gt;−1&lt;/sup&gt; yr&lt;sup&gt;−1&lt;/sup&gt;)</th>
<th>PER&lt;sub&gt;app&lt;/sub&gt;&lt;sup&gt;b&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cattle/horses</td>
<td>1.1</td>
<td>6.46</td>
<td>30%</td>
</tr>
<tr>
<td>Hogs</td>
<td>0.79</td>
<td>4.36</td>
<td>50%</td>
</tr>
<tr>
<td>Sheep</td>
<td>0.38</td>
<td>1.88</td>
<td>60%</td>
</tr>
<tr>
<td>Poultry</td>
<td>0.25</td>
<td>0.31</td>
<td>25%</td>
</tr>
<tr>
<td>Human</td>
<td>0.21</td>
<td>0.31</td>
<td>30%</td>
</tr>
</tbody>
</table>

<sup>a</sup> P excretion coefficients in urine and feces (Cao, 2002; NEPD, 2002).

<sup>b</sup> Percentage of water soluble P in excreted fecal P (Ajiboye et al., 2004; Hainze et al., 2004; Kleinman et al., 2005; Turner and Leytem, 2004).

### 2.2.3. Cell to water delivery

Particulate and dissolved P loads mobilized in each cell as calculated above were further weighed by the proximity of cells to stream channel to obtain the delivered P loads (Tables 2 and 3). Many phosphorus indexes considered the distance from source to watercourses as an effective proxy for runoff risk (Buczko and Kuchenbuch, 2007; Drewry et al., 2011). They assumed that the closer to streams or rivers, the higher potential to be delivered to watercourses. Distances to watercourses were divided into groups and rating values were assigned to each distance group to reflect the potential of P delivery. For example, rating values were 1 and 0.2, respectively for distance groups ≤50 m and >50 m in field study (Bechmann et al., 2009) and 1, 0.9, 0.8, 0.6 and 0.3, respectively for distances ≤500 m, 500–1500 m, 1500–3000 m, 3000–5000 m and >5000 m in watershed-scale study (Zhou and Gao, 2011). These values were generally determined based on experiences or expert judgements. The LRW-PI adopted the inverse-distance weight approach, which could mathematically compute the delivery ratios and was proposed by King et al. (2004), to determine the cell-to-water delivery ratios of P at various distances. The linear distances (d, m) between the center of each cell and rivers were calculated by Euclidean distance tool in ArcGIS 9.3 software based on the river networks. Then the distances were divided into unequal interval distance classes: 0–250 m, 251–500 m, 501–1000 m, 1001–2000 m, 2001–5000 m, 5001–10000 m and 10001–20000 m and the maximum distance in each distance class (<i>d</i><sub>max</sub>) were used to calculate the inverse distance-based delivery ratio (<i>D</i>) following Eq. (16). The constant 100 was used to constrain the order of results magnitudes. The total delivered PP, DP and TP loads were finally calculated following Eqs. (17)–(21).

### 2.3. Application of LRW-PI in typical hydrological conditions

Five typical hydrological conditions including annual average (AA), average flood season (AFS), average non-flood season (ANFS), wet year (WY) and dry year (DY) were selected to characterize the variations in NPS P predictions in response to hydrological condition changes. Based on the analysis over the 14-year rainfall records in the sub-watersheds (see Appendix 1), the year 1995 and year 2000 respectively presented relatively higher rainfall amount and relatively lower rainfall amount than any other years and therefore were chose to respectively represent the WY and DY. Considering that more than 75% of the annual rainfall occurred from June to September, this study defined the period from June to September as the flood season and the rest of the year including January to May and October to December as the non-flood season. LRW-PI was applied in studied sub-watersheds on ArcGIS platform. The output NPS TP, TDP<sub>sub</sub>, TDP<sub>TP</sub>, TDP and TPP in years or seasons were finally expressed as 90 m × 90 m raster layers and the mean values in each sub-watershed were derived. Cross-site relationships between NPS P loads or percentages and selected input variables were examined to explore which factors were influencing the spatial variation in NPS P predictions. All statistical analysis including Pearson correlations, Mann–Whitney U test were carried out by R software (R Core Team, 2012).

### 3. Results and discussions

#### 3.1. Observations of P inputs, soil P status, rainfall and runoff

**Fig. 2a** showed clear spatial variation in P inputs and soil P status. Budeng sub-watershed received the highest average P inputs per hectare of sub-watershed area due to owning the highest percentage (35%) of area covered by dry land (**Fig. 2a; Table 1**). Liu and Sa sub-watersheds ranked the second and Laoniu and Wuile sub-watersheds received the lowest P inputs. Mineral fertilizer, rather than manure, was the major source of P inputs (38–63%). Topsoils in all sub-watersheds except Xingzhou were enriched with OlsenP (≥20 mg kg<sup>−1</sup>), indicating a higher potential to lose more P, either in dissolved form or sediment-associated form, into runoff.

**Fig. 2b** displayed that the annual average rainfall and runoff depths tended to decrease from the south to the north. The significantly higher rainfall and runoff depths in the southern sub-watersheds (e.g., Sa, Liu and Pu) than in the northern sub-watersheds (e.g., Budeng, Yimatu and Yixun) suggested a much higher P transport potential in the southern sub-watersheds. DR<sub>sub</sub> ranged from 46% to 95%, highlighting the dominance of subsurface pathway in runoff transfer. This might also suggest a high possibility for the occurrence of nutrients transfer via subsurface pathway, reaching groundwater or returning to streams as latetal flow or springs. We found that DR<sub>sub</sub> had a close relationship with cinnamon soil percentages (<i>R</i><sup>2</sup> = 0.45, <i>p</i> = 0.046) which were strongly correlated with soil sand proportions (<i>R</i><sup>2</sup> = 0.88, <i>p</i> < 0.001). The sandy texture of soils might have favored water infiltration.

### 3.2. Annual average total loads

As LRW-P indicator estimated, the 14-year annual average total NPS P loads in studied sub-watersheds ranged from 0.49 to 1.22 kg ha<sup>−1</sup> and averaged at 0.79 kg ha<sup>−1</sup> (**Fig. 2c**). These values
felt within or closely to previously reported TP loads from predominantly agricultural basins in Europe (<0.1–6 kg ha⁻¹) (Kronvang et al., 2007; Withers and Jarvie, 2008), United States (median value of 2.25 kg ha⁻¹) (Beaulac and Reckhow, 1982) and northern China plain (0.8–1.6 kg ha⁻¹ at delivery ratios of 5% and 10%) (Yang et al., 2012). The average annual NPS TP loads showed clear spatial variation (Fig. 2c), with the southern sub-watersheds (e.g., Sa, Liu and Pu) having apparently higher values than those in other sub-watersheds. This was expected since Sa, Liu, Pu sub-watersheds were characterized by high rainfall and runoff, relatively high P inputs and soil OlsenP contents and steeper slopes (Fig. 2a and b; Table 1). As one of the northern sub-watersheds with less rainfall and runoff, Budeng sub-watershed presented unexpected higher TP loads than its adjacent Yixun and Yimatu sub-watersheds (Fig. 2c). This might be mainly attributed to the highest P inputs per unit of watershed area and the highest percentage of dry land (Fig. 2a; Table 1).

We also compared the annual NPS P loads in six sub-watersheds during the period 2000–2008 simulated by LRW-P indicator and generalized watershed loading function (GWLF) model. The accuracy of the GWLF model simulation was overall acceptable and the model calibration and verification details were described in Du et al. (2014) and Du (2014). Fig. 3 showed that despite of the observable differences in Xingzhou, Wulie and Pu sub-watersheds, both approaches captured consistent pattern in the relative magnitudes of NPS TP loads among sub-watersheds (Fig. 3). Mann–Whitney U test indicated no significant difference between the two groups of means (p = 0.24).

3.3. Composition of annual average NPS P loads

Analysis of the NPS P load composition could highlight P sources and pathways, as well as the potential influence of P on water quality. As LRW P indicator estimated, NPS TP consisted of 35–65% of PP, 25–45% of DP in subsurface runoff and 10–25% of DP in surface runoff (Fig. 4a). TDP exceeded TPP in either loads or percentages in most of the sub-watersheds except Wulie and Laoniu (Fig. 4a). This suggested that majority of the lost NPS P would be readily available for the aquatic organisms in the downstream rivers or reservoir. While there has been a widely-hold assumption that most P loss from agricultural land occurs via surface runoff in association with soil particles (Heathwaite and Dils, 2000). The significant contribution from DP loss in subsurface runoff explained this contrast. No measurements of either P distribution in soil profile or P concentrations in subsurface flow were made to provide direct evidence of subsurface P loss in studied sub-watersheds. However, a 7-year monitoring of well water in 13 sites of LRW indicated that TP concentrations in well water (0.05–0.93 mg L⁻¹) already reached critical level which may promote surface water eutrophication (0.02 mg L⁻¹) (Correll, 1998). This indirectly evidenced the occurrence of substantial subsurface P loss. We hypothesized three reasons for this significant subsurface DP loss: (1) subsurface flow was the major flow pathway, as previously discussed; (2) the sandy soils generally had lower P adsorption capacity (Buczko and Kuchenbuch, 2007); and (3) the long term application of manure and fertilizer had elevated P saturation degree in subsoils and consequently facilitated the P downward or lateral movement.

Fig. 4b revealed that mineral fertilizer and soils were the major sources of NPS TP pollution. Mineral fertilizer was mainly contributing to DP loss while soils to PP loss (Fig. 4b). Unlike the findings of Chen et al. (2010) which identified livestock feeding and rural life as the leading source of agricultural P loads (57%) in a national scale, our results seemed to underestimate the contribution of livestock production and rural living (<25%) to NPS P pollution. The results differed because we defined that NPS
P pollution only count the land-applied part of waste produced by rural living and livestock feeding and treated the rest as point sources. In recent years, the pollution in rural areas of China due to livestock and rural dwellers has raised increasing attention from both governments and scientific communities. However there hasn't been a clear definition of non-point source pollution in these areas up to date. As to the spatial correlation between inputs and NPS P predictions, only P inputs were positively correlated with TDP and associated TDP/TP percentage (Table 6). No significant correlations were found between soil OlsenP and any type of predicted activities.

3.4. Annual average NPS P loads influenced by land uses

Variations in NPS TP loads with land uses reflected the influence of human activities. Dry land had the highest TP loads, ranging from 1.9 kg ha\(^{-1}\) to 8.9 kg ha\(^{-1}\). These values fell within the ranges reported for arable lands in United States and Yongding River basin, China (Fig. 5, Table 7). Paddy land was second to dry land and had TP loads ranging from 1.2 kg ha\(^{-1}\) to 2.5 kg ha\(^{-1}\). These two land use types had relatively high TP loads due to the application of manure and mineral fertilizer. The grassland and forest had relatively lower TP loads, which were comparable to previously documented values in American and European countries (Fig. 5, Table 6). Considering that croplands in this study were generally located in sloped uplands and riverside areas as well as the influence of distance on P transfer, the dry and paddy lands near river channels were

Table 6

<table>
<thead>
<tr>
<th>Datasets</th>
<th>Input variables</th>
<th>TP</th>
<th>TDP</th>
<th>TDP_sub</th>
<th>TDP_net</th>
<th>TDP</th>
<th>TDP/TP</th>
<th>TDP_net/TP</th>
</tr>
</thead>
<tbody>
<tr>
<td>AA dataset</td>
<td>P(_{total})</td>
<td>0.51 ns</td>
<td>0.12 ns</td>
<td>0.64 ns</td>
<td>0.66 ns</td>
<td>0.68 ns</td>
<td>0.043*</td>
<td>0.69 ns</td>
</tr>
<tr>
<td>OlsenP</td>
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<td>0.63 ns</td>
<td>0.37 ns</td>
<td>0.42 ns</td>
<td>0.08 ns</td>
<td>-0.19 ns</td>
<td>0.64 ns</td>
<td></td>
</tr>
<tr>
<td>Grass land %</td>
<td>0.21 ns</td>
<td>0.27 ns</td>
<td>-0.02 ns</td>
<td>0.27 ns</td>
<td>0.08 ns</td>
<td>-0.11 ns</td>
<td>-0.4 ns</td>
<td></td>
</tr>
<tr>
<td>Forest %</td>
<td>0.46 ns</td>
<td>-0.65 ns</td>
<td>-0.25 ns</td>
<td>-0.25 ns</td>
<td>-0.26 ns</td>
<td>0.25 ns</td>
<td>-0.05 ns</td>
<td></td>
</tr>
<tr>
<td>Dry land %</td>
<td>0.86 0.002***</td>
<td>-0.86 0.006**</td>
<td>-0.64 ns</td>
<td>-0.72 0.04*</td>
<td>-0.7</td>
<td>0.39 ns</td>
<td>0.05 ns</td>
<td></td>
</tr>
<tr>
<td>RD(_{soil})</td>
<td>0.9 0.011***</td>
<td>0.88 0.002**</td>
<td>0.71 0.013*</td>
<td>0.78 0.016*</td>
<td>0.18 ns</td>
<td>-0.18 ns</td>
<td></td>
<td></td>
</tr>
<tr>
<td>RD(_{fertilizer})</td>
<td>0.91 &lt;0.001***</td>
<td>0.87 0.002**</td>
<td>0.8 0.01*</td>
<td>0.68 0.04*</td>
<td>0.8 0.01</td>
<td>-0.26 ns</td>
<td>0.06 ns</td>
<td></td>
</tr>
<tr>
<td>DR(_{soil})</td>
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<td>0.03 ns</td>
<td>0.07 ns</td>
<td>-0.5 ns</td>
<td>-0.12 ns</td>
<td>0.03 ns</td>
<td>1 &lt;0.001***</td>
<td></td>
</tr>
<tr>
<td>Rainfall</td>
<td>0.86 0.003***</td>
<td>0.9 0.001***</td>
<td>0.7 0.037*</td>
<td>0.62 ns</td>
<td>0.7 0.034*</td>
<td>0.04 ns</td>
<td>0.08 ns</td>
<td></td>
</tr>
</tbody>
</table>

| Full dataset | | | | | | | | |
| | RD\(_{soil}\) | 0.83 <0.001*** | 0.79 <0.001*** | 0.72 <0.001*** | 0.75 <0.001*** | 0.79 <0.001*** | 0.23 ns | 0.16 ns |
| | RD\(_{fertilizer}\) | 0.80 <0.001*** | 0.80 <0.001*** | 0.61 <0.001*** | 0.79 <0.001*** | 0.70 <0.001*** | 0.16 ns | -0.32 ns |
| | RD\(_{fertilizer}\) | 0.83 <0.001*** | 0.80 <0.001*** | 0.69 <0.001*** | 0.78 <0.001*** | 0.75 <0.001*** | 0.21 ns | -0.23 ns |
| | RD\(_{fertilizer}\) | 0.31 ns | -0.34 ns | -0.09 ns | -0.51 ns | -0.25 ns | 0.047 ns | 1 <0.001*** |
| | Rainfall | 0.91 <0.001*** | 0.94 <0.001*** | 0.71 <0.001*** | 0.82 <0.001*** | 0.78 <0.001*** | 0.06 ns | -0.45 ns |

* Pearson r, not squared.
** Spatial correlation between dry land % and NPS P loads including Budeng sub-watershed.
*** Spatial correlation between dry land % and NPS P loads excluding Budeng sub-watershed.
* Full dataset includes data in five hydrological conditions (AA, WY, DY, AF, ANFS).
extremely susceptible to runoff P loss and thus required special attention and prioritized management practices.

The percentage of crop land in a watershed has been considered as an important explanatory variable for nutrient loss concentrations or loads. Taranu and Gregory-Eaves (2008) found a significant and positive cross-study correlation between Agricultural land % and lake TP loads at a broad spatial scale ($r = 0.53$, one tailed $p = 0.021$). However no such significant correlations were found in our study (Table 6). We found that Budeng sub-watershed was an outlier and excluding it significantly improved the correlation significance particularly between dry land % and TP, TPP or TDP sub. Yet the negative correlation coefficients suggested that the correlation-ship was contrary to what was previously reported. This contrast, as well as the limited correlations between P inputs and P loads, might indicate that the impacts of P source on the spatial variation in NPS P load prediction had been overwhelmed by those of other factors such as rainfall and runoff. Morse and Wollheim (2014) reported that the inter-annual climate and associated runoff variability had masked the influences of land use changes on nutrient fluxes in a suburbanizing watershed. While the exception of Budeng sub-watershed did evidence the determining effect of P source factors (high dry land % and P inputs) on TP loss loads.

![Diagram](image_url)

Fig. 6. Dynamics of rainfall, runoff characteristics and predicted NPS P loads and percentages in studied sub-watersheds under typical hydrological conditions. Red dash line represented the AA baseline. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
3.5. NPS P predictions varied with hydrological conditions

Fig. 6 displayed the dynamics of rainfall, runoff and predicted NPS P loads and percentages in WY, DY, AFS and ANFS. As many studies demonstrated (Ahmadi et al., 2014; Du et al., 2014; Jeppesen, 2009), the wet year with high rainfall and runoff increased NPS TP losses by 18–45% relative to the AA baseline depending on sub-watersheds, and the dry year with low rainfall and runoff reduced NPS TP loads by 10–60%. These TP changes were mainly attributed to runoff DP changes in both transport pathways and eroded PP losses. It is worthy pointing out that we observed unchanged TPP loads in northern sub-watersheds in both WY and DY. How PP losses from soils were calculated was responsible for this. The soil PP loss was the product of TP_{soi}, erosion rates and P enrichment ratios (Eq. (1) in Table 3). The erosion rates were positively correlated with rainfall while ER was inversely correlated with erosion rates due to the selective transport of sediment particles (Eqs. (3) and (4) in Table 3). The unchanged rainfall amount in Yixun, Budeng and Yimatu resulted in unchanged TPP predictions. While the counteraction between erosion rates and P enrichment ratio was responsible for the unchanged TPP prediction in DY. Since we didn’t consider the runoff effects on PP loss, TPP estimation heavily relied on rainfall ($r = 0.94, P < 0.001$, Table 6). Therefore the application of LRW-P indicator in sites or years with relatively low runoff/rainfall ratios might overestimate PP loss. Another important implication from what we observed in the DY was that reduced runoff may reduce sediment erosion rates but not necessarily reduce PP loss, and vice versa. Between the hydrological seasons, around 60–70% of the annual NPS TP, TPP and TDP_{sub} and TDP losses occurred in the flood season (Fig. 6f; Fig. 6g; Fig. 6h; Fig. 6i), corresponding to that more than 70% of the annual runoff occurred in the flood season (Fig. 6b). Yet TDP_{sub} loads maintained at comparable levels in flood season and non-flood season (Fig. 6i). This could be explained by the relatively constant subsurface runoff depths in the two seasons (Fig. 6c).

We examined the cross-site correlations between NPS P predictions and rainfall-runoff variables based on either the AA dataset or the full dataset including five hydrological conditions. Rainfall-runoff variables demonstrated the most and the strongest correlations with NPS P loads or percentages, even though some of them could have been influenced by multi-collinearity among variables (Table 6). By comparing the correlation coefficients and significance, we observed stronger spatial correlations between NPS P loads or percentages and rainfall and runoff variables than between NPS P loads or percentages and P source variables ($P_{total}$, OlsenP, land use %). The significant and positive correlations between NPS P loads and rainfall-runoff variables implied that: (1) sub-watersheds characterized by high rainfall, runoff and P transport potential such as Liu, Sa and Pu should be listed as high risk areas for NPS P load; (2) high rainfall or runoff periods (e.g., wet year; flood season) should be the critical time targeting NPS P losses and (3) management practices that reduce runoff depths could lower P losses. Another important finding was that DR_{sub} played a minor role in determining NPS P loads, but well determined the percentages of TDP_{sub} in TDP (Table 6).

3.6. Highlights and uncertainties

The delivery of P from field edge to watercourses was an important process for the modeling approach and in reality was controlled by complex and dynamic interactions between rainfall characteristics and soils and surface properties. It was particularly difficult to quantify since there were few sources of data on which to base the coefficients when developing a simple indicator (Heathwaite et al., 2003). This study treated this processes as a “black box” and used a delivery ratio based on distances. We compared the delivery ratios assessed as the inverse distance weights in LRW-PI and as a function of the watershed area (km2) in GWLF model (Eq. (22)) (Haith and Shoenaker, 1987).

$$SDR = 0.451 \times (\text{Area}_{basin})^{-0.298} \quad (22)$$

Delivery ratios by the two methods generally fell within a narrow range from 0.04 to 0.08 (Fig. 7). In Budeng and Sa sub-watersheds they were perfectly matched. Greater D values than SDR values in sub-watersheds of Laoniu, Pu, Wuie and Yimatu were mainly attributed to that these sub-watersheds had larger areas and SDR values were adversely correlated with areas. Liu sub-watershed had similar area with Sa sub-watershed but lower D values, probably due to sparser river networks in Liu sub-watershed. Using the area of entire watershed may underestimate the delivery ratios since only part of the watershed area could contribute to the delivery of nutrients and sediments (Lane et al., 2009). This phenomenon was particularly prominent in regions with low or discontinuous hydrological connectivity due to low rainfall and/or high infiltration. In our study which was located in semi-humid and semi-arid regions and had relatively high infiltration, the inverse distance-based delivery ratio should be more suitable for NPS P loads estimation.

We are also conscious that as a watershed-scale indicator, this methodology unavoidably would encounter uncertainties. The
coefficients cited from worldwide studies could be one source. For example, this study used a constant coefficient to convert soil Olsen P to runoff P concentrations assuming that soil P extractability was similar among soils. Yet the regression between soil test P and runoff P could vary with soil types and management (Sharpley et al., 2002). Another coefficient worth mentioning is DRpc. This study assigned a value of 1, assuming no difference in P concentrations existing between surface and subsurface runoff. Yet this is the ideal situation where there is no P recession during transport through soil matrix or surface landscape. Many studies reported lower TP or DP in subsurface runoff and the differences varied with studies (Heathwaite et al., 2003; Mittelstett et al., 2011). Therefore the current settings for DRpc in this methodology might overestimate the contribution from subsurface runoff in soils which were less permeable or had higher P sorption capacity. A slight variation in DRpc might exert significant influences on the outputs, for example, P composition. Therefore detailed local studies regarding soil P-runoff P relationship and P recession during transport should be conducted in the future to fill this knowledge gap and refine DP loss formulation.

4. Conclusions

The LRW-PI was capable to estimate annual and seasonal NPS P loads at sub-watershed scales using simple methods and readily obtainable inputs. The application of LRW-PI in 9 sub-watersheds in five hydrological conditions identified the southern sub-watersheds were posing high risk of NPS P loss to water quality in Panjakou reservoir and the critical timing for NPS P controlling were wet year and flood season. The dry land and paddy fields near the river channels should be treated with prioritized management practices. Considering the significant contribution of subsurface P loss and the difficulty in reducing or controlling subsurface runoff volume, the overall P reduction in studied sub-watersheds should focus on PP loss control and P source control. Conservation practices such as terraced fields, filter strips would greatly intercept the sediment and PP delivery to streams. The recommended fertilization based on soil P test and crop P requirement would effectively reduce DP in both surface and subsurface pathways, as well as PP eroded from soils. The application of an inverse distance-based delivery ratio well reflected the hydrological characteristics in semi-humid and semi-arid regions but greatly reduced the complexity and difficulty to obtain the values. We are conscious that some parts of this P indicator would be improved once local studies fill the knowledge gap. But we strongly recommend using such indicator to help policy makers to screen out high priority areas or time for watershed-scale P management actions before sophisticated research being conducted in smaller scales.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecolind.2015.12.002.

References


