

# Effects of Hydrological Processes on Surface and Subsurface Nitrogen Losses from Purple Soil Slopes

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## Research Article

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# 1   **Effects of hydrological processes on surface and subsurface** 2   **nitrogen losses from purple soil slopes**

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17   **Abstract:** Relationships of hydrological processes via surface flow (SF) and  
18   subsurface flow (SSF) to nitrogen (N) losses from sloping farmlands have been rarely  
19   researched. In this study, laboratory experiments were conducted to investigate  
20   ammonia nitrogen (NH<sub>4</sub>-N), nitrate nitrogen (NO<sub>3</sub>-N) and total nitrogen (TN) losses  
21   from purple sloped soils due to SF, SSF and sediment (S). Effects of rainfalls and  
22   slope gradients on N losses were also studied. Three rainfall intensities ( $0.4 \pm 0.02$ ,  
23    $1.0 \pm 0.04$  and  $1.8 \pm 0.11$  mm min<sup>-1</sup>) and four slope gradients (5° , 10° , 15° and

20° ) were designed in experiments. Larger SF discharges occurred with increasing rainfall intensities while SSF was prone to happen under low rainfall intensities. Although  $r^2$  of regression results were low, both N loss concentrations and loads coincided positively with discharges except for a negative relation between N concentrations and SF discharges. In comparison, smaller SSF discharges produced substantial N loads with higher N concentrations especially for NO<sub>3</sub>-N. NH<sub>4</sub>-N, NO<sub>3</sub>-N, and TN losses were dominated by S, SSF and SF, respectively. Furthermore, linear increases in loss loads with increasing discharges revealed that distributions of N loss loads were compatible with flow distributions in stormwater. 10° may be a critical slope gradient for SSF discharge and nutrient export.

**Key words:** Nitrogen loss; Flow discharge; Purple soil; Slope gradient; Rainfall intensity

## 1. Introduction

There is increasing recognition of the need to consider nutrient losses as an important problem causing pollution in water environment of watershed. (Bechmann, 2014; Jia et al., 2007; Kumwimba et al., 2017; Panagopoulos et al., 2011). Nitrogen (N) losses from sloped soils also have become a serious problem restricting sustainable and ecological friendly developments of sloping farmlands (Diaz et al., 2010; Ding et al., 2017; Kahl et al., 2007). Understanding N, including ammonia nitrogen (NH<sub>4</sub>-N), nitrate nitrogen (NO<sub>3</sub>-N), and total nitrogen (TN) loss mechanisms, is essential for ecological sustainability and efficient agricultural production.

Typically, processes of N losses are inextricably correlated with precipitation, soil properties and structures, and terrain factors (Kleinman et al., 2006; Ouyang et al.,

2017; Tremblay et al., 2011). In sloping farmlands, N losses occur mainly by means of surface flow (SF), subsurface flow (SSF), and sediment (S) (Pionke et al., 1999; Zhao et al., 2015). The SF and S are prone to happen due to gradients of sloping farmlands and rainfalls from local climate (Liu et al., 2015). Also, SSF has been found to be an important part of hydrological processes of sloping farmlands (Hewlett and Hibbert, 1967; Peyrard et al., 2016), especially for purple sloped soils (Jia et al., 2007; Xie et al., 2019). Jiang et al. (2009) reported that early N transport and losses were dominated by runoff and rainfall intensities, and during the later stages N losses occurred mainly in forms of nitrate from hilly areas of purple soils. It has been widely proved that purple soils are formed by purple bedrock weathering, thus the loose and thin soil layers and distinctive dualistic structures enhance SSF generations (He et al., 2009; Tang et al., 2012; Wei et al., 2006). Additionally, purple soil slopes account for 70% of the total farmlands in the Three Gorges Reservoir Area along the Yangtze River (Ma et al., 2016). There is evidence that  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN from sloping farmlands of purple soils can be exported in large numbers subjected to various slope gradients and rainfall intensities (Ding and Zhang, 2009; Jia et al., 2007; Xie et al., 2020; Xu et al., 2011; Zhao et al., 2015). Xu et al. (2011) reported that N losses due to sediment transport accounted for 53%~62% of total N losses from field observations. Several studies emphasized impacts of the flow processes on N losses (Qian et al., 2017; Xu et al., 2007), especially for the  $\text{NO}_3\text{-N}$  losses via SSF. The slope gradient has been analyzed in many studies of N losses from purple soil slopes. Several studies showed that N losses correlated positively with slope gradients (Ding et al., 2017; Huo et al., 2013), while others found that N losses tended to increase initially and then decrease with increasing slope gradients (Ding and Zhang, 2009; Li et al., 2010).

Recent studies have attempted to clarify how hydrology impacts N losses, especially the relationships between N concentrations and flow (Zhao et al., 2015). Billy et al. (2013) noted that surface runoff discharges related positively to nitrate concentrations, and underlined the major influences of hydrological processes on nutrient losses in an agricultural watershed. In contrast, Armstrong et al. (2011) reported that nitrate concentrations in runoff correlated negatively with discharges from laboratory rainfall experiments using soil flumes. These correlations are region and scale specific with diversities. However, relationships between the subsurface hydrological process and nutrient losses were less described in literatures. For purple sloped soils, previous studies about N losses highlighted effects of rainfalls on N losses, but no relation between water flow and N losses was revealed (Ding and Zhang, 2009; Ding et al., 2017; Zhu et al., 2009). Ding et al. (2017) presented negative relations between TN loads in runoff and rainfalls. They further noted that more N losses happened due to increased runoff from increasing rainfalls. Zhu et al. (2009) reported that both interflow discharges and nitrate leaching losses were dependent on rainfall amounts. Meanwhile, Jia et al. (2007) indicated that increasing volumes of subsurface outflow enhanced nitrate losses from purple soils using simulated rainfalls. It can be seen that studies on effects of hydrological processes such as flow discharges on N losses from sloped purple soils are less frequent despite the importance that has been suggested.

The literatures highlighted above identify knowledge gaps about effects of hydrological processes on N losses due to both SF and SSF, and N losses that can be expected from SF, SSF and S in a rainfall event. Additionally, it appears that site-specific experimental data may be required to model and analyze these losses efficiently.

To better understand linkages between rainfalls, hydrological processes, terrains, nutrient exports, water qualities and environment services, we must synthesize both new and existing knowledge and make it available in a useful and functional format for better management practices that reduce environmental risks. Specially, better and refined models are needed for evaluation of pollutant transport following the hydrological processes considering terrain factor. As a result of regional diversities in climate, terrain, and soil properties, etc., such result bases need to be advanced and applied on regional basis. The objectives of our study were to evaluate effects of hydrological processes on N losses by modeling relationships between N loss (concentrations and loads) and flow discharges via SF and SSF, and investigate N loss loads from SF, SSF and S, as well as effects of rainfall intensities and slope gradients on N losses from sloped soils.

## **2. Materials and methods**

### **2.1. Soil**

Experimental soils used in this study was collected from small Wangjiaqiao watershed (110°42' E, 31° 5' N) in Zigui County, Yichang City, Hubei Province, China (Figure 1). The study site is located besides the middle-upper Yangtze River and consists of sloping lands under economic fruit cultivation. It is dominated by a sub-tropical monsoon climate having an average annual rainfall of 1100 mm, 81.9% of which falls intensely between June and October (Xie et al., 2017). Surface slopes of this study site are approximately from 5° to 20°.

Purple soils predominate sloping farmlands of the Wangjiaqiao watershed. Formed from rock weathering, the structure of purple soils is loose and the soil layer

is thin. Experimental soils were collected from the top 40-cm soil layer of the sloping farmlands at the Wangjiaqiao watershed (Figure 1B). The basic physical and chemical properties of the soil are listed in Table 1. This purple soil is classified as loam and Entisol in USDA taxonomy, representing the most common agrotype in the Wangjiaqiao watershed. Collected soils were air-dried and then passed through a 10-mm sieve.

## 2.2. Experimental setup

The screened soil samples were backfilled into a soil tank (Figure 2) in 5-cm increments and compacted to a bulk density of  $1.35 \text{ g cm}^{-3}$ . The bottom of the tank was paved with a 10-cm relatively waterproof layer of cement to simulate purple bedrocks. The contact surfaces between the soils and the tank were encased in an amino-plastic web, to prevent adverse boundary influences. The soil tank was equipped with a hydraulic pressure slope adjustment facility to simulate different slopes, and four slopes were designed ( $5^\circ$ ,  $10^\circ$ ,  $15^\circ$ ,  $20^\circ$ ) in this study. At slope foot of the tank, two V-angled grooves were set to collect surface and subsurface flow, at the soil surface and the interface between the underside of soils and the waterproof layer, respectively. The V-angled grooves were sheltered from rainfalls avoiding inaccuracy in flow collections. Soil tank experiments were conducted in a laboratory hall at the Institute of Soil and Water Conservation, Changjiang Water Resources Commission from June to October, 2017. Twelve scenarios of soil tank experiments, involving four slope gradients and three rainfall intensities, were performed in duplicate.

Rainfall events were simulated by a roughly 9-m high stationary artificial rainfall device with 85% degree of rainfall uniformity (Figure 2). Three rainfall intensities

(0.4±0.02, 1.0±0.04 and 1.8±0.11 mm min<sup>-1</sup>) were set and each rainfall intensity was calibrated by tests before experiments. Durations of all rainfall events were 60 minutes. To keep the same initial nutrient content of tested soils in the tank, tank was backfilled with fresh soils from the same screened soil samples after running each scenario. Moreover, initial soil moisture contents were controlled at about 0.25 cm<sup>3</sup> cm<sup>-3</sup> with pre-precipitations in accordance with soil moisture contents of sloping farmlands in Wangjiaqiao watershed.

### 2.3. Water sample collection and analysis

Manual samplers were applied to collect surface runoff and subsurface water flow every 2 ~ 3 minutes and 5 ~ 6 minutes, respectively, during each rainfall event, and the time and duration of sample collections were documented. 16 surface water samples and 10 ~ 16 subsurface flow water samples were collected in each event. Volumes of all water samples were measured by gradations on flow collectors. Afterwards, the water samples were left to stand for 3 hours and then 0.05 L of each supernatant water sample was transferred into clean polyethylene bottles. Bottles of water samples were stored at 4° C in a refrigerator and chemical determinations were completed within 48 hours. NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN concentrations were determined via SmartChem Discrete Auto Analyzer (SmartChem 200, Alliance, France). N loss loads of water flow in our study were calculated according to Equation (1). Similar methods have been utilized in previous studies (Alva et al., 2006; Wang et al., 2011).

$$Q = \sum \frac{C_i \times q_i}{S \times 10^5} = \sum \frac{C_i \times q_{vi} \times t_i}{S \times 10^5} \quad (1)$$

Where  $Q$  (kg N ha<sup>-1</sup>) is the N loss load,  $S$  (m<sup>2</sup>) is the surface area of the soil



tank,  $i$  is the sequence of surface runoff or subsurface flow samples,  $C_i$  ( $\text{mg L}^{-1}$ ) is the N concentration,  $q_i$  ( $\text{mL}$ ) is the discharge volume of surface runoff or subsurface flow,  $q_{vi}$  ( $\text{mL s}^{-1}$ ) is the discharge of surface runoff or subsurface flow computed by dividing  $q_i$  ( $\text{mL}$ ) by duration of corresponding water sample collection (s) and  $t_i$  (s) is the time interval of two adjacent sample collections.

Collectors of surface flow were dried overnight in an oven at  $105^\circ \text{C}$ , followed by weighing using an electronic balance to obtain mass. The empty collectors were weighed beforehand and their mass were recorded in order to obtain net weights of sediments. During one rainfall event, due to needs for pretreatment of soils before determining N concentrations, the soil sample mass of each single collector was inadequate. Hence, the samples of sediments were gathered together into a sealed bag for determination of N concentrations. In total, 24 rainfall events produced 24 bags of soil erosion samples, and  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN concentrations of each soil erosion sample were measured as mean concentrations of lost sediment under the corresponding rainfall events. Sediment mass of all collections for each event were divided by durations to obtain sediment discharge rates. Then, sediment loads were calculated by multiplying sediment discharge rates and intervals of sample collections. The sediment loads for each event were multiplied by the mean N concentrations to obtain N loss loads from soil erosion.

## 2.4. Statistical analysis

Linear regression relationships between N losses and flow were evaluated subjected to different rainfall intensities and slope gradients. The relationships between N export characteristic indexes [concentration ( $\text{mg L}^{-1}$ ) and loss load ( $\text{Kg}$

ha<sup>-1</sup>)] and flow discharges were examined through linear regressions of the natural logarithm of N indexes as function of flow discharges (cm<sup>3</sup> s<sup>-1</sup>). N concentrations were estimated by linear interpolation between adjacent measured data when measured data were not available. IBM SPSS Statistics 20 (IBM Corp., USA) was applied to compare regression lines, also was used to test for statistical differences between N concentrations via SF and SSF, N loss loads via SF, SSF and S using ANOVA at a significance level of 0.05. EXCEL (Microsoft Corp., WA, USA) and Origin 8.5 (Origin Lab Corp., USA) were used for curve fitting and graphing as appropriate.

### 3. Results

#### 3.1. Flow discharge

As Figure 3 shows, surface flow discharges only coincided positively with rainfall intensities with small variances. However, SSF producing mechanisms were complicated with larger variances than SF. Moreover, the SF initiated about 0.5 to 1 hour earlier than SSF. Low rainfall intensity induced relatively larger SSF discharges, especially at 15° (0.88 cm<sup>3</sup> s<sup>-1</sup> under 0.4 mm min<sup>-1</sup> rainfall intensity). It is also worth noting that SSF flow discharges presented significant differences at 10°. At 10° , SSF flow discharge appeared minimum values subjected to low (0.4 mm min<sup>-1</sup>) and moderate (1.0 mm min<sup>-1</sup>) rainfall intensities versus maximum value under high rainfall intensity (1.8 mm min<sup>-1</sup>). The SF discharges were significantly larger than SSF (4.00 versus 0.36 cm<sup>3</sup> s<sup>-1</sup>, 12.49 versus 0.23 cm<sup>3</sup> s<sup>-1</sup> and 21.98 versus 0.23 cm<sup>3</sup> s<sup>-1</sup> under 0.4, 1.0 and 1.8 mm min<sup>-1</sup> rainfall intensities, respectively) and the SF discharges tended to increase at initial stage of runoff generation and then remained

constant until the end of rainfall events (Figure 4). SSF discharges indicated peak values at middle stages of generation processes with large fluctuation (Figure 4). These findings are in agreement with several published studies (Jia et al., 2017; Montenegro et al., 2013; Gao et al., 2010).

### 3.2. Nitrogen loss concentration

NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN concentrations from all samples were summarized by rainfall intensities, flow producing patterns, and slope gradients in Figure 5, and a statistical summary is exhibited in Table 2. All mean N concentrations of SSF were remarkably greater than corresponding values of SF, especially for NO<sub>3</sub>-N and TN, and differences were 5.0 to 71.7-fold. For 5° of slope gradient and 0.4±0.02 mm min<sup>-1</sup> of rainfall intensity, means from SF (n=32) and SSF (n=30) were 0.12 and 0.81 mg L<sup>-1</sup>, for NH<sub>4</sub>-N, 1.6 and 114.7 mg L<sup>-1</sup>, for NO<sub>3</sub>-N, and 5.8 and 128.2 mg L<sup>-1</sup> for TN, respectively. It also indicated a significant negative correlation between N concentrations and rainfall intensities, especially for SSF (Table 2). There was almost no regular patterns in N concentrations as a response to slope gradients.

In this study, linear regressions were conducted to examine the spread of the natural logarithm transformed NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN concentrations as function of water flow discharge (Figure 6 and parameters in Table 3). The intercepts (negative for NH<sub>4</sub>-N but positive for NO<sub>3</sub>-N and TN) of all cases were different from zero ( $p < 0.0001$ ). Additionally, in each case, intercepts of SSF were greater than that of SF. The flow-related model for SF was highly significant ( $p < 0.0001$ ) with low  $r^2$  (0.051 and 0.225 for NH<sub>4</sub>-N and NO<sub>3</sub>-N, respectively) under all slopes for NH<sub>4</sub>-N and NO<sub>3</sub>-N concentrations. For SSF, parameters in response to all land slope gradients were

insignificant ( $p = 0.033$ ,  $0.033$  and  $0.071$  for  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and  $\text{TN}$  respectively).  $r^2$  was only a small fraction of what it was for the flow-related relationship (Endale et al., 2017). Slopes in Table 3 were mostly negative for SF but positive for SSF. Overall,  $r^2$  were negatively related to slope gradients.

### 3.3. Nitrogen loss load

#### 3.3.1. Loss features in response to rainfall intensities and slope gradients

Statistical summary for all data of mean loss loads in  $\text{Kg ha}^{-1}$  of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and  $\text{TN}$  by means of SF, SSF and S are displayed in Table 2. The data existed differences among SF, SSF and S according to significance analysis at 95% level of probability ( $p < 0.05$ ). All N loss loads via SF and S tended to increase with increasing rainfall intensities (Figure 7). Total  $\text{NH}_4\text{-N}$  loss load had a significantly positive correlation with rainfall intensity as shown in Figure 7A, and average loads via S accounted for 44.5~70.9% of total loads under 1.0- and 1.8-mm  $\text{min}^{-1}$  rainfall intensities. In comparison, mean  $\text{NH}_4\text{-N}$  loss loads due to SSF were only  $0.010 \text{ Kg ha}^{-1}$  (23.9% of total loss loads),  $0.004 \text{ Kg ha}^{-1}$  (4.0% of total loss loads), and  $0.002 \text{ Kg ha}^{-1}$  (1.40% of total loss loads), in response to 0.4, 1.0, and 1.8  $\text{mm min}^{-1}$  rainfall intensities, respectively (Table 2). Moreover, slope gradient at  $15^\circ$  mostly induced maximum  $\text{NH}_4\text{-N}$  loss loads (Figures 7A and 8A). For  $\text{NO}_3\text{-N}$ , low precipitation intensity was much more likely to cause loss loads by means of SSF (Figure 7B) and loads via S were negligible with a mean proportion of 0.73% of total loss loads. Mean  $\text{NO}_3\text{-N}$  loads from SSF listed in Table 2 were  $1.790 \text{ Kg ha}^{-1}$  (81.7% of total loss loads),  $0.885 \text{ Kg ha}^{-1}$  (54.7% of total loss loads), and  $0.520 \text{ Kg ha}^{-1}$  (28.1% of total loss loads), in response to 0.4, 1.0, and 1.8  $\text{mm min}^{-1}$  rainfall intensities, respectively.

Overall,  $\text{NO}_3\text{-N}$  loads occurred in largest numbers by means of SSF compared with  $\text{NH}_4\text{-N}$  and TN especially under light rainfalls (Figure 7). The results indicated occurrence of N loss differences at the slope gradient of  $10^\circ$  (Figure 8B). For TN, as a response to low rainfall intensity, losses happened mainly by means of SSF. However, TN loads through SF tended to increase and accounted for 47.5 ~ 70.1% of total loss loads under moderate and high rainfall intensities. Mean total loads from these two rainfall intensities ( $1.0$  and  $1.8 \text{ mm min}^{-1}$ ) were respectively 2.0 and 2.7 times greater than that from the low rainfall intensity ( $0.4 \text{ mm min}^{-1}$ ). Mean loss loads of TN showed dominant differences among the four slope gradients, especially between the  $5^\circ$  and the other three slope gradients (Figure 8C). According to differences of SSF discharges at  $10^\circ$  mentioned in section 3.1, N loss loads similarly occurred distinct differences at the slope gradient of  $10^\circ$ . In conclusion,  $\text{NH}_4\text{-N}$  and TN losses mostly happened through the soil surface while  $\text{NO}_3\text{-N}$  loads depended on SSF. It appears that combined effects of rainfall and terrain are needed to evaluate the general regular patterns of N loss from sloped soils.

### 3.3.2. Loss processes and relationships to flow

Cumulative loss loads of nitrogen due to SF and SSF both presented linearly increasing trends with cumulative flow amounts (Figure 9), but increasing trends of  $\text{NH}_4\text{-N}$  were not revealed in Figure 9 due to its negligible loads compared with  $\text{NO}_3\text{-N}$  and TN. Loss rates of TN were highest and differences between surface loss rates of TN and  $\text{NO}_3\text{-N}$  were enlarged as rainfall intensity enhanced. SSF induced minor differences between loss rates of TN and  $\text{NO}_3\text{-N}$  for all scenarios. These differences for loss processes of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN were related to the loss distributions as presented in section 3.3.1. Linear regressions (data not shown) of

280 cumulative N loss load versus flow subjected to twelve scenarios showed a strong  
281 positive correlation ( $p < 0.0001$ ) in high  $r^2$  values ( $> 0.9$ ), and slopes were all positive  
282 and significantly different from zero ( $p < 0.0001$ ). Similar results have been found by  
283 other researches on surface runoff and subsurface drainage (Kanwar et al., 2005). It  
284 implied that N loss distributions were compatible with flow distributions in  
285 stormwaters according to the linear increases shown in Figure 9.

286 In this study, natural logarithms of N loss loads are depicted as linearly varying  
287 with flow discharges (Figure 10). Parameters from linear regression of ln-transformed  
288 N loss load versus flow discharge are shown by slope gradient in Table 4. Figure 10  
289 shows plots based on data pooled over all slope gradients for SF and SSF, and N loss  
290 loads via SF and SSF both positively correlated with flow discharges. There were  
291 differences in parameters between SF and SSF.

292 Model parameters of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN were significant for all scenarios.  
293 For  $\text{NH}_4\text{-N}$ ,  $r^2$  at  $5^\circ$ ,  $10^\circ$ ,  $15^\circ$ ,  $20^\circ$  and all slope gradients were greater from SF (0.571,  
294 0.513, 0.492, 0.670 and 0.521) than SSF (0.348, 0.260, 0.102, 0.502 and 0.305), and  
295 presented a decreasing trend with increment in slope gradients except for  $20^\circ$ .  
296 Slopes were positive for all scenarios, and values varied between 0.757 and 1.837  
297 (1.558 pooled) of SSF versus 0.026 to 0.040 (0.035 pooled) of SF. For  $\text{NO}_3\text{-N}$ ,  $r^2$  at  
298  $5^\circ$ ,  $10^\circ$ ,  $15^\circ$ ,  $20^\circ$  and all slope gradients were greater for SF (0.541, 0.565,  
299 0.513, 0.506 and 0.491) than for SSF (0.403, 0.449, 0.127, 0.461 and 0.338), and  
300 presented a decreasing trend with increment in slope gradient for SF. The slopes were  
301 positive under all scenarios, and values varied between 0.720 and 1.944 (1.599 pooled)  
302 for SSF versus 0.022 to 0.035 (0.027 pooled) for SF. For TN,  $r^2$  at  $5^\circ$ ,  $10^\circ$ ,  $15^\circ$  and all  
303 slope gradients were greater for SF (0.666, 0.523, 0.715, and 0.565) than for SSF

(0.525, 0.417, 0.276, and 0.411) except for 20° (0.368 and 0.424, respectively). The slopes were positive under each scenario, and values varied between 0.962 and 1.532 (1.421 pooled) for SSF versus 0.025 to 0.051 (0.040 pooled) for SF.

## **4. Discussion**

### **4.1. Flow discharge**

Positive correlations between SF discharge and rainfall intensity can be explained by runoff yield with excess infiltration. In contrast, impacts of slope gradients on SF discharges were negligible in this study. Furthermore, subsurface flow processes from sloped soils were more complicated in response to rainfalls (Dunne and Black, 1970; Yin et al., 2006). Low discharges of SSF were prone to be observed in purple soils due to the impermeable purple bedrock underlying the shallow soil matrix and slow water movement of the fine-textured soils (Persson and Saifadeen, 2016). Larger SSF discharges under light rainfalls might result from less loss of rainfall amounts via SF (Huang et al., 2013). Huang et al. (2013) also indicated that a slope gradient of 17.6% promoted best soil infiltration using a soil box with variable slope gradients from 8.8% to 36.4%. However, maximum discharges occurred under different gradients in this study hence no determinately sole slope gradient was indicated for the largest outflow of SSF. Also, mean SSF discharges presented significant differences at 10°. 10° is a critical slope gradient for SSF generation from purple sloping lands as indicated by Ding and Zhang (2009). In spite of the small discharge, prolonged SSF could still cause large water losses and thereby lead to large amounts of N loss loads in high N concentrations.

## 4.2. Nitrogen losses

### 4.2.1 Comparisons between SF and SSF

Much higher concentrations of loss nitrogen via SSF than those via SF have been reported by studies of N losses in various sites from sloped soils (Bechmann, 2014; Jia et al., 2007; Zheng et al., 2017). The regression results of this study showed that loss concentrations of N via SF decreased with increasing flow discharges while loss concentrations of N via SSF increased. Nevertheless, the predictive power of regression results was low given the highly variable N dynamics. These results can be explained by that SF discharges ranging from 1.33 to 29.09 cm<sup>3</sup> s<sup>-1</sup> in response to three rainfall intensities, and SSF discharges mostly concentrated on 0.2~0.3 cm<sup>3</sup> s<sup>-1</sup>, therefore N concentrations in SF were deliquated by large amounts of runoff (Bechmann, 2014; Kleinman et al., 2006; Veizaga et al., 2015). Additionally, nutrients in soils were less effectively transferred towards surface runoff by raindrop splash compared to leaching by subsurface water from rainfall infiltration through soils. Hence, large SF discharges enhanced dilutions of N concentrations and low SSF discharges enhanced N leaching losses with high N concentrations. Zheng et al. (2014) reported that NO<sub>3</sub>-N concentrations were over 5.73 times greater than NH<sub>4</sub>-N concentrations in SSF because oxidations transform NH<sub>4</sub>-N into NO<sub>3</sub>-N in soils and NO<sub>3</sub>-N is negatively charged which is compatible with negative charges of soil colloid. Hence, large numbers of NO<sub>3</sub>-N are prone to leach via subsurface flow. Moreover, in their study, maximum NO<sub>3</sub>-N concentrations in SSF appeared under 0.5-mm min<sup>-1</sup> rainfall intensity, which is in accordance with our experimental results.

Although loss patterns among NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN varied (Figure 7), differences were only revealed by parameters of intercepts in linear regressions of



ln-transformed N loss loads versus flow discharges. The slope and  $r^2$  were both similar among  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN. In contrast, differences in parameters of  $r^2$ , intercept and slope between SF and SSF were attributed to the larger errors from complex subsurface processes, different N loss patterns and smaller SSF discharges. Compared with average N loss loads reported in Zhao et al. (Zhao et al., 2001) as follows:  $0.152 \text{ Kg ha}^{-1}$  (73.1%) and  $0.056 \text{ Kg ha}^{-1}$  (26.8%) of  $\text{NH}_4\text{-N}$  via SF and SSF, respectively, and  $0.076 \text{ Kg ha}^{-1}$  (15.7%) and  $0.409 \text{ Kg ha}^{-1}$  (84.4%) of  $\text{NO}_3\text{-N}$  via SF and SSF, respectively. The loss distributions for  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  in our study were in agreement with their study considering similar load values (Table 2). Given lower  $\text{NH}_4\text{-N}$  concentration values compared with  $\text{NO}_3\text{-N}$  in SSF as displayed in the section 3.2,  $\text{NH}_4\text{-N}$  loads were smaller than  $\text{NO}_3\text{-N}$  loads and mainly occurred by means of S.

#### 4.2.2 Relations to hydrological processes

Water discharge as a soil hydrological property plays an important role in interpreting N losses (Silva et al., 2005). Several studies (Iital et al., 2014; Mueller et al., 2016) have indicated that nitrogen loads correlated positively with discharges in river catchments, which is in agreement with results in this study, but detailed relationships were not revealed for  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN losses via surface and subsurface processes separately in their studies. As increasing discharge intensified loss concentrations of N, loss loads of N were much more likely to be enhanced by SSF discharges. Also, linear increases of N loads with increasing outflow amounts of SSF highlighted that the small outflow amounts contributed to subsurface N loss. Furthermore, porosity and soil hydraulic characteristics as heterogeneity properties of soils were reported to impact solute transport (Persson and Saifadeen, 2016). Given the backfilled loose purple soils with low hydraulic conductivity, hysteretic

subsurface N loss occurred along with hysteretic outflow producing as shown in Figure 4. Previous studies also reported significant positive relationships between nutrient concentration and subsurface flow discharge (Dunne et al., 2005; McNamara et al., 2008; Wang et al., 2014), but these relationships existed diversities based on study sites as well as rainfall events. We also note that experimental set up and design may have impacts on the initial conditions such as soil moistures and nutrient contents. Nevertheless, the current experimental set up has allowed us to examine processes under similar initial conditions and provide comparisons of N loss patterns between SF and SSF.

Slopes of regressions between ln-transformed N concentrations and flow discharges were negative versus positive slopes of regressions between ln-transformed N loads and flow discharges for SF. Loss load consists of concentration and flow discharge during the same period as defined in Equation (1), hence we can hypothesize that flow discharge had a greater positive impact on N loss loads from soil surface despite dilutions of N loss concentrations via SF. Mueller et al. (2016) indicated, agricultural land use produced a strong correlation between flow discharge and nitrate load, therefore further studies considering effects of vegetation tillage need to be undertaken in the researches of hydrological impacts on N losses.

#### 4.2.3 Responses to rainfall intensities and slope gradients

From laboratory simulated rainfall events, Ding and Zhang (2009) showed that TN loss loads via sediment from the sloped purple soils coincided positively with rainfall intensities which is in accordance with our results, but the proportion (49.59%~88.68%) was much larger than the data (6.5%~27.8%) in our study. These differences may have been due to the larger magnitudes of soil erosion in their study

and different initial TN contents in soil samples. In essence, positive correlations between N losses and rainfall intensities via SF and S can be related to discharge amounts and N transfer towards surface runoff. Conversely, significantly negative relationships between N losses via SSF and rainfall intensities were found in our study and previous studies (Jia et al., 2007; Kanwar et al., 2005).

A study by Ding et al. (2017), investigating effects of rainfall and terrain on TN losses, provides an interesting comparison to our results. These authors conducted 20 experimental scenarios with five slope gradients (5°, 10°, 15°, 20°, and 25°) combined with four rainfall intensities (0.5, 1, 1.5, and 2 mm min<sup>-1</sup>). Ding et al. (2017) indicated positive correlations between rainfalls and TN loads but negative correlations between rainfalls and TN concentrations for SF, which is consistent with our experimental results. However, a significantly positive correlation between slope gradients and TN loads via surface runoff was revealed due to stronger scouring effect of steeper slopes according to their studies. The scouring effect from slope gradients may have relations to the intersection angle between the raindrop and downslope soil surface directions. This effect was mainly embodied by flow discharge to some degree. Given the impacts of gradients on flow as shown in section 3.1 and the responses of N loads to slope gradients in this study, N loads was compatible with flow process which occurred variations at 10° with impacts of rainfalls. We reckon that the 10° may be a critical slope gradient for SSF discharges and nitrogen exports since similar findings have been reported by others (Ding and Zhang, 2009; Li et al., 2010).

## 5. Conclusions

We studied surface and subsurface N losses from purple soil slopes subjected to

four slope gradients ( $5^{\circ}$ ,  $10^{\circ}$ ,  $15^{\circ}$ , and  $20^{\circ}$ ) and three rainfall intensities ( $0.4 \pm 0.01$ ,  $1.0 \pm 0.02$ , and  $1.8 \pm 0.11$  mm min<sup>-1</sup>) by laboratory experiments. In conclusion, our experiments showed that surface and subsurface outflows as simultaneous hydrological processes were highly different. Overall, N load distributions of both SF and SSF were concordant with outflow distributions in stormwaters. In terms of loss concentrations and loads, positive correlations with outflow discharges were found by linear regressions except for N loss concentrations via SF. We believe that this negative impact of surface flow on N loss concentrations resulted from dilutions from large discharges of SF. Flow discharges play a more important role in N losses via SF. In contrast, smaller SSF discharges with higher N loss concentrations produced substantial N loads especially for NO<sub>3</sub>-N. Loss loads of NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN occurred mainly by means of S, SSF, and SF, respectively. Responses of N loads to rainfall intensities were positive via surface flow while negative via subsurface flow. Slope gradient of  $10^{\circ}$  was reported as a critical gradient for SSF processes and N losses. We attribute N loss patterns to raindrop splashes, scouring effects and discharges.

Given our findings we suggest that the role of hydrological process, as a key driver of N losses, is reassessed when constructing models to reduce nitrogen losses and establish fertilizer applications in sloping farmlands. We also believe that the impact of soil slope gradient on N loss, dependent from rainfall intensity, needs to be investigated further.

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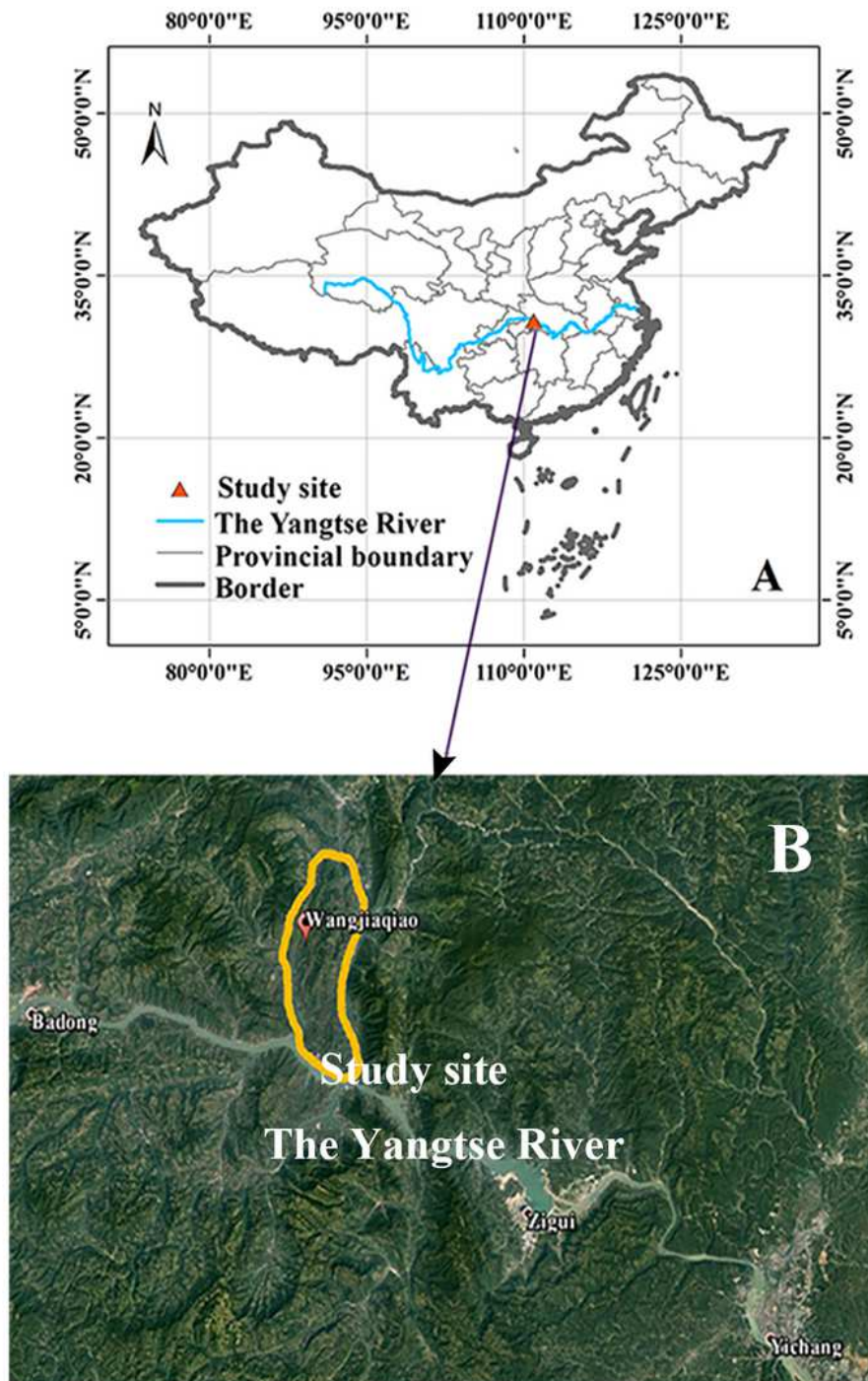
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# Figures

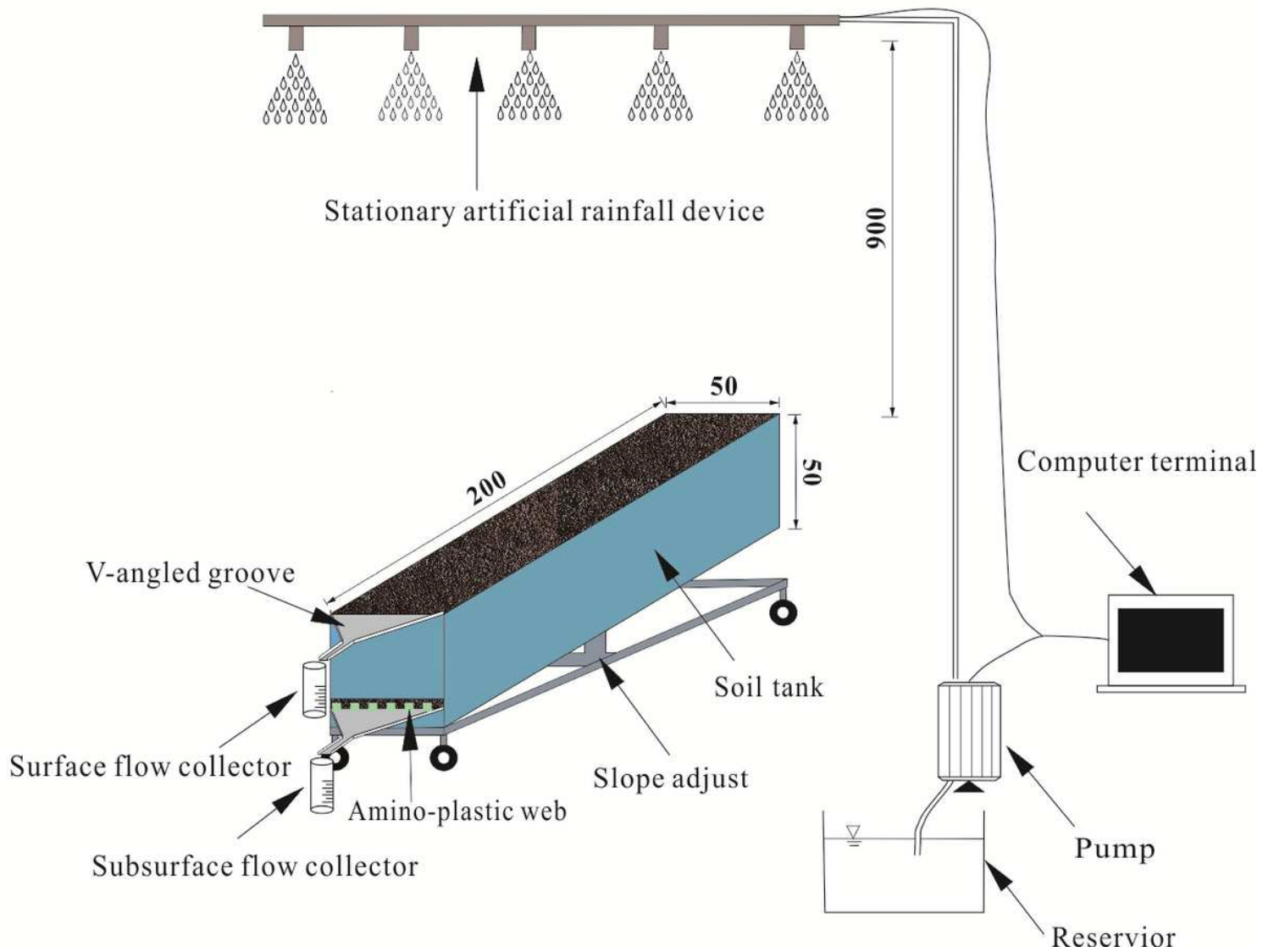


**Figure 1**

Location of the study site in Hubei Province, China (A), and of the soil collection site at Wangjiaqiao watershed (B). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal

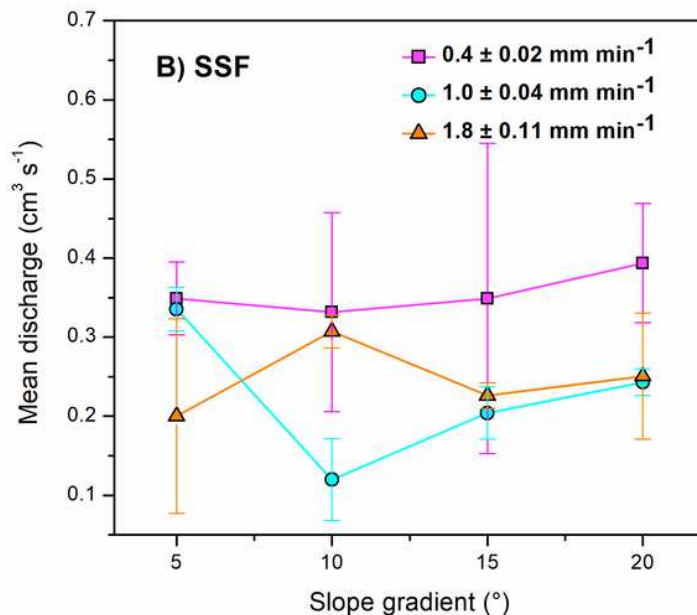
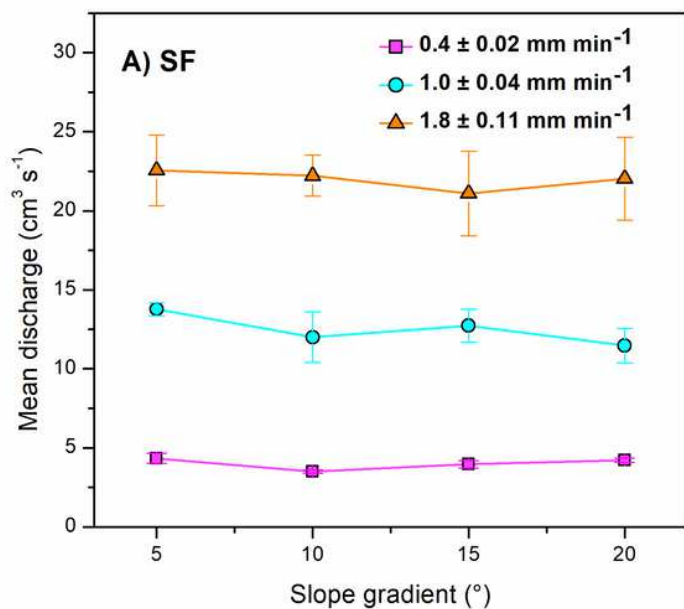
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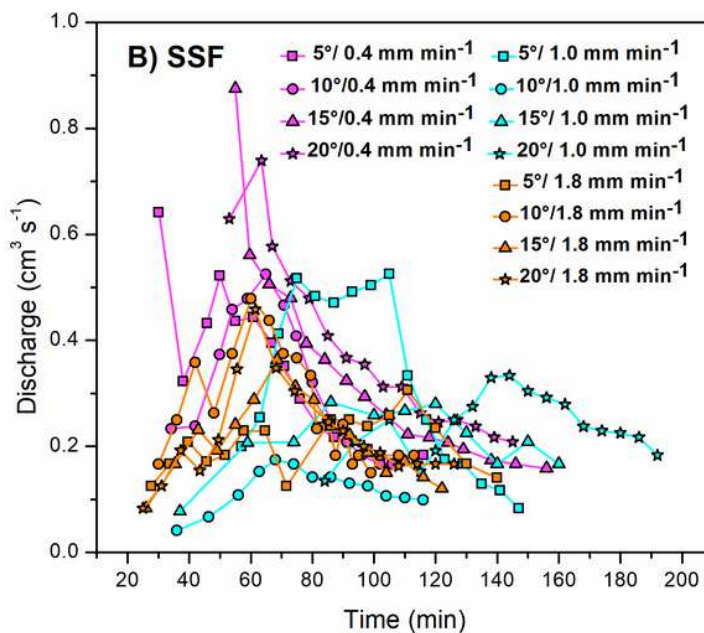
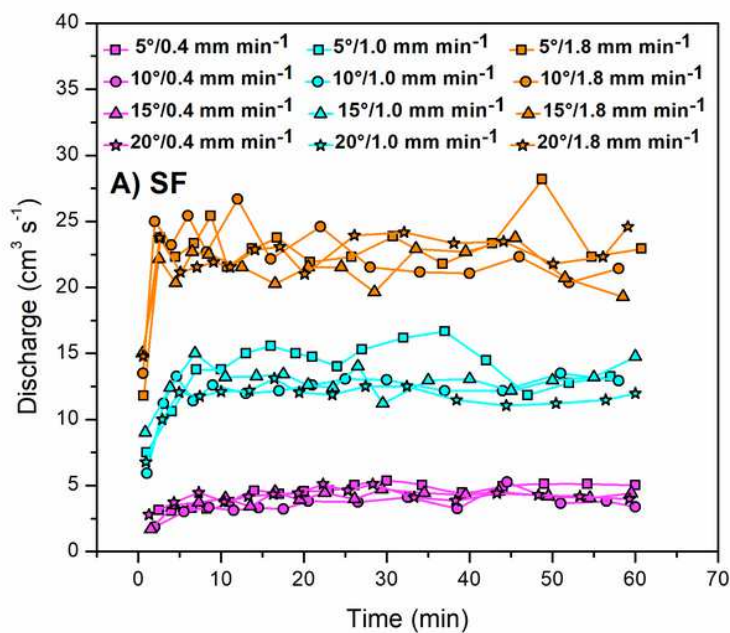
**Figure 2**

Schematic diagram of the experimental setup.



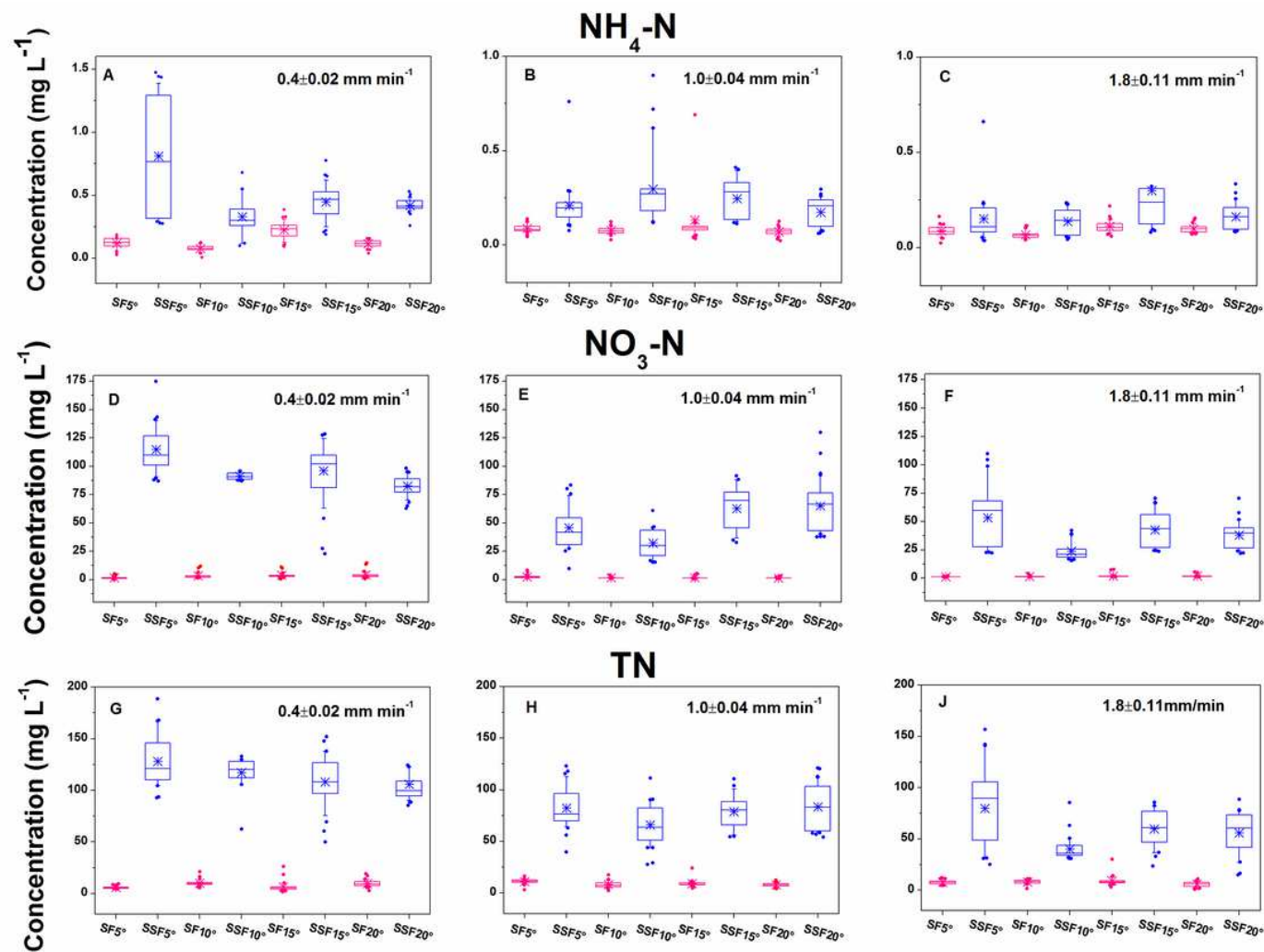
**Figure 3**

Flow discharges of (A) surface flow (SF) and subsurface flow (SSF) as response to rainfall intensities and slope gradients.



**Figure 4**

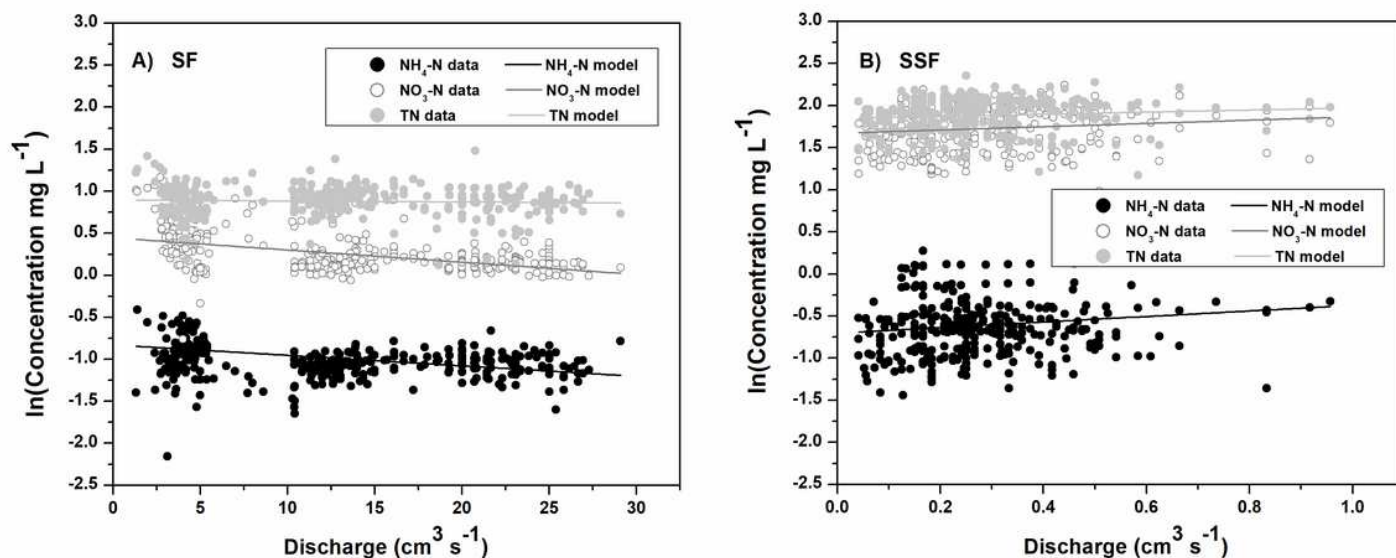
Dynamics of flow discharge via (A) SF and (B) SSF for twelve treatments.



**Figure 5**

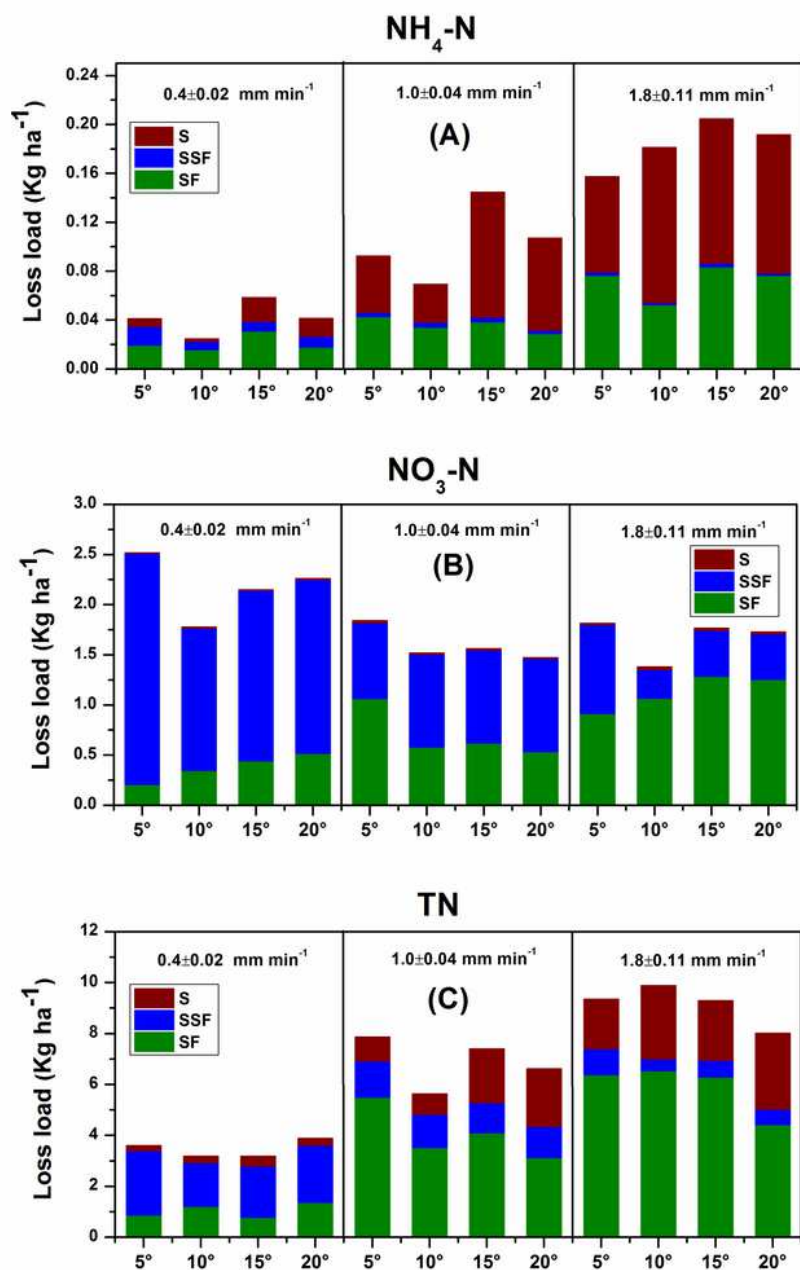
Box plots showing distributions of (A, B and C) NH<sub>4</sub>-N, (D, E and F) NO<sub>3</sub>-N and (G, H and I) TN for SF and SSF. The boxes bound the 25th and 75th percentiles. Solid lines inside boxes represent medians and asterisk symbols represent means. Whiskers represent the 10th and 90th percentiles and dots represent outliers.





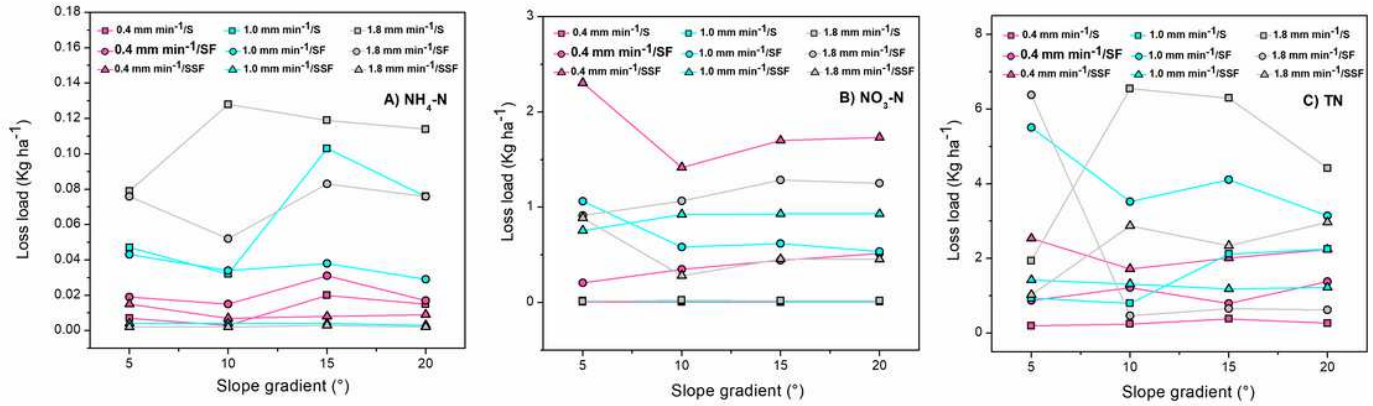
**Figure 6**

Relationships between the natural logarithm ( $\ln$ ) of concentration ( $\text{mg L}^{-1}$ ) of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN and flow discharge ( $\text{cm}^3 \text{s}^{-1}$ ) shown as the observed data and the corresponding linear regression model for (A) SF and (B) SSF.



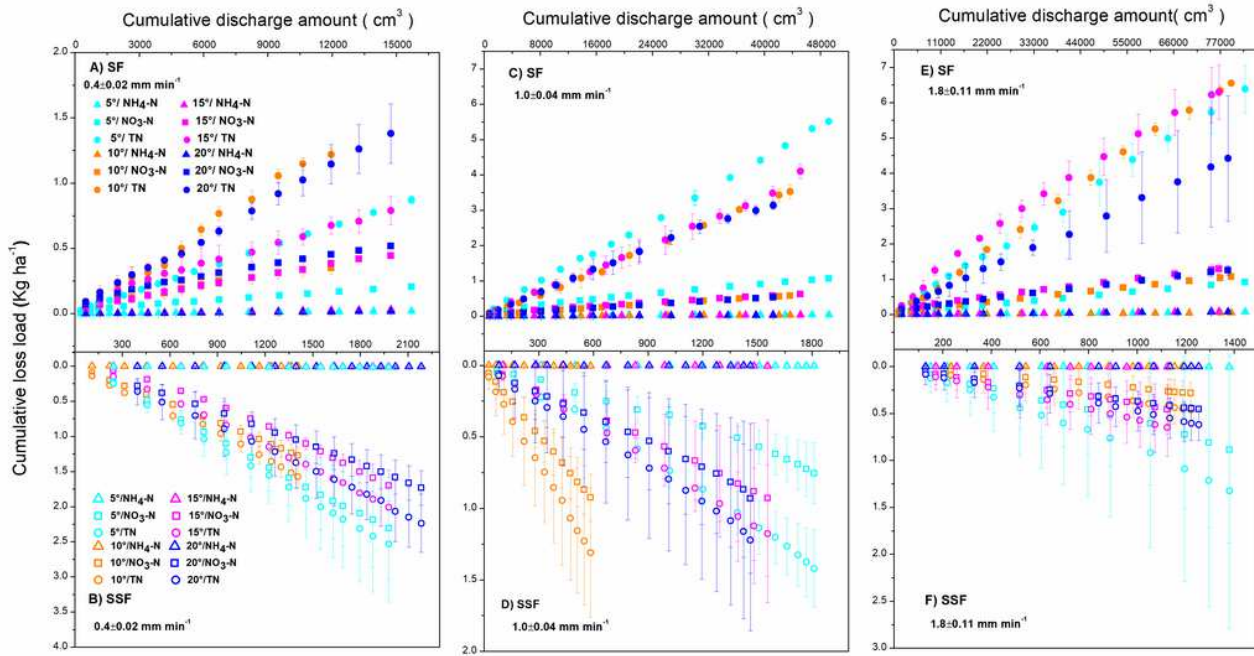
**Figure 7**

Bar plots showing (A) NH<sub>4</sub>-N, (B) NO<sub>3</sub>-N and (C) TN loss loads by means of sediment (S), surface flow (SF), and subsurface flow (SSF) of each event subjected to different slope gradients and rainfall intensities.



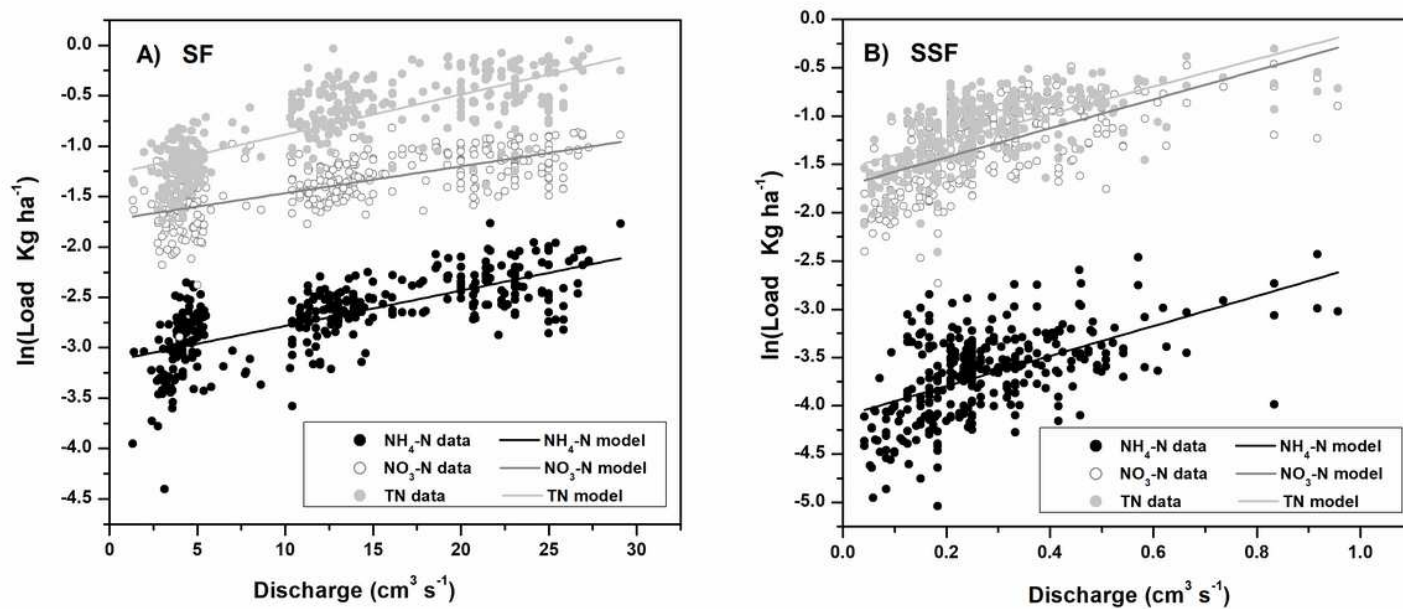
**Figure 8**

Mean loss loads of (A) NH<sub>4</sub>-N, (B) NO<sub>3</sub>-N and (C) TN as a function of slope gradient under three rainfall intensities for sediment (S), surface flow (SF) and subsurface flow (SSF).



**Figure 9**

Cumulative loss loads of NH<sub>4</sub>-N, NO<sub>3</sub>-N and TN at different slope gradients as a function of cumulative flow volume for surface flow (SF) and subsurface flow (SSF) under (A and B) low, (C and D) moderate, and (E and F) high rainfall intensities.



**Figure 10**

Relationship between the natural logarithm ( $\ln$ ) of load ( $\text{Kg ha}^{-1}$ ) of  $\text{NH}_4\text{-N}$ ,  $\text{NO}_3\text{-N}$  and TN and flow discharge ( $\text{cm}^3 \text{s}^{-1}$ ) shown as the observed data and the corresponding linear regression model for (A) SF and (B) SSF.

## Supplementary Files

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