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## Diffuse export of nutrients under different land uses in the irrigation area of lower Beiyunhe River (China)

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### Abstract

Non-point source pollution is serious in the agriculture watershed of China. Understanding the characteristics of rainfall-runoff from agriculture watershed can provide theoretical support for controlling non-point source pollution. In this study, we investigated runoff characteristics of eight indices (dissolved total N,  $\text{NO}_3^-$ -N,  $\text{NH}_4^+$ -N, total phosphorus, dissolved total phosphorus, particulate phosphorus, total organic carbon, COD) from three types of land uses, including farmland, forest and village in the downstream irrigation area of the Beiyunhe River basin. The results showed that the event mean concentrations (EMCs) of total dissolved N in village, farmland and forestland were 17.81mg/L, 12.68mg/L and 3.14mg/L, respectively. EMC of total phosphorus in the order: farmland (0.44mg/L) > village (0.22mg/L) > forestland (0.17mg/L). EMC of COD in the order: farmland (45.07mg/L) > forestland (27.06mg/L) > village (18.03mg/L). The changes in the nutrients concentrations of the runoff water over a rainfall event indicated that the transports of the nutrients are similar among various land use types. The instantaneous concentrations of TN,  $\text{NH}_4^+$ -N, and  $\text{NO}_3^-$ -N were high in the initial period of runoff, tend to decreasing with rainfall continuing, and increase in later period. Phosphorus concentration with time variation was not obvious among three land use types. The phosphorus species with high proportion in the total phosphorus was particle P (accounting for 75%) in forestland, dissolve P (79%) in farmland, and particle P (48%) and dissolve P (52%) in village. The curves of COD and TOC had been shown as high in the initial period of runoff, tending to increasing with rainfall continuing, and decrease in the later period. First-flush of all the indices were obvious in all three land use types with the rank of village > forestland > farmland. In village, all of the pollutions have taken place the phenomenon of first flush, while in farmland, pollutions tended to uniformly distribute or dilution throughout the storm event.

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*Keywords:* land-use; runoff; pollution; loss; non-point source

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### 1. Introduction

Non-point source pollution (NPSP) has become an increasing concern among scientists, decision-makers, and the public at large, especially where point sources of pollution have been identified and remedied [1]. Agricultural sources of non-point pollutions often contribute the main share of NPSP.

Nutrients loss from arable land in dissolved and particle forms not only tend to impoverish soil nutrient stocks and lower soil productivity [2], but also cause eutrophication in water bodies [3]. So, understanding the output characteristics of non-point pollution was necessary to protecting the water bodies from pollution.

Source control may become the best choice of watershed best management projects (BMPs) until the mechanisms of rainfall-runoff were deeply discussed. Field monitoring under natural rainfall conditions and during long periods can help to improve our understanding of the underlying mechanisms and processes affecting rainfall-runoff-induced losses of nutrients.

A lot of these researches focus on the area characterized by high altitude or rainy climate [4-9]. But researches in North China Plain which characterized by semi-arid climate, plain and low altitude were scarcity. According to the "Report on the State of the Environment in China 2009" [10], the ranking of pollution level of the seven main water systems is Haihe, Liaohe, Yellow, and Huaihe, Songhua, Yangtze and Pearl rivers. Of the big rivers, Haihe River was measured as heavy pollution. In the low of Beiyunhe River as important components of Haihe River Basin, the pollution was extremely serious. With the recent control of point sources, non-point source pollution was increasing as prominent source [11].

This study focuses on the processes and the characteristics of nutrient species loss from different land use types in the agriculture watershed of North China Plain. The objectives of the present research were: (i) to figure out the characteristic of nutrients concentrations from the three land use types; (ii) to describe the process and characteristic of surface flow for a typical storm event; (iii) to analyze the strength and the effects of first flush on nutrients loss.

## 2. Materials and methods

### 2.1. Study area

The study area was located at irrigation area of lower of Beiyunhe River in the North China Plain (P.R. China) with an altitude that ranges from 1.5 to 13 m. The climate is semi-arid with an average annual precipitation of 590.7 mm, concentrated in July and August. The mean annual temperature is 11.6°C, and mean potential evapotranspiration reaches 1700 mm year<sup>-1</sup>, so the long-term water deficit is 1100mm. These conditions result in an aridic soil moisture and thermic soil temperature regime.

This study area is representative of the agricultural, socio-economic and environmental situation of many dry land farming areas in the region. For well-developed irrigation channels (there are three major rivers: Beiyunhe River, Longfenghe River and the Yongdinghe River), this irrigation area is become the main agricultural production region of North China Plain where the main crops of maize, and wheat are grown with intensive inputs. Use N fertilizer as an example, the consumption was 0.127 million tons in 2007, but fertilizers use efficiency is very low in this region [12]. The main limitation to agricultural and socio-economic development is not the shortage of water but pollution [13].

### 2.2. Monitoring area

In order to comprehensive survey non-point pollution and rainfall-runoff process of different land use types, improve the objectivity and accuracy of monitoring results, a 2-year experiment (rainy season: July to August of 2009 and 2010) had been carried out at two monitoring areas: *A* and *B*. Three monitoring sites for each monitoring area had been set up, covering three different land use types (i.e., farmland, woodland, villages).

Monitoring area "*A*" located at the town of Hebeitun, sparsely scattered, which may reflect the non-point source pollution of remote rural areas (Fig.1). Monitoring area "*B*" located at the Nanxinhuang village, residential concentration, nearby the main city, and closing to the intersection of Longfenghe and Beiyunhe River. "*B*" can reflect the outskirts of the rural non-point pollution.

The village had a fixed drainage, dried up before rain. Village monitoring sites had been set up in the outlet of drainage (Fig.1-A). For farmland, a field usually was divided into many small plots. Before rain, the ridges of a monitoring plot (near the channel) had been set with plastic baffles (baffle height is 25 cm, 10 cm in the underground), but a V-shaped groove, which easy to collect runoff samples, had been set up

in middle of the plot (Fig.1-A); Because of the corresponding catchment areas in forestland, the outlet of catchment areas had been set up with plastic baffles, similar to farmland monitoring site.

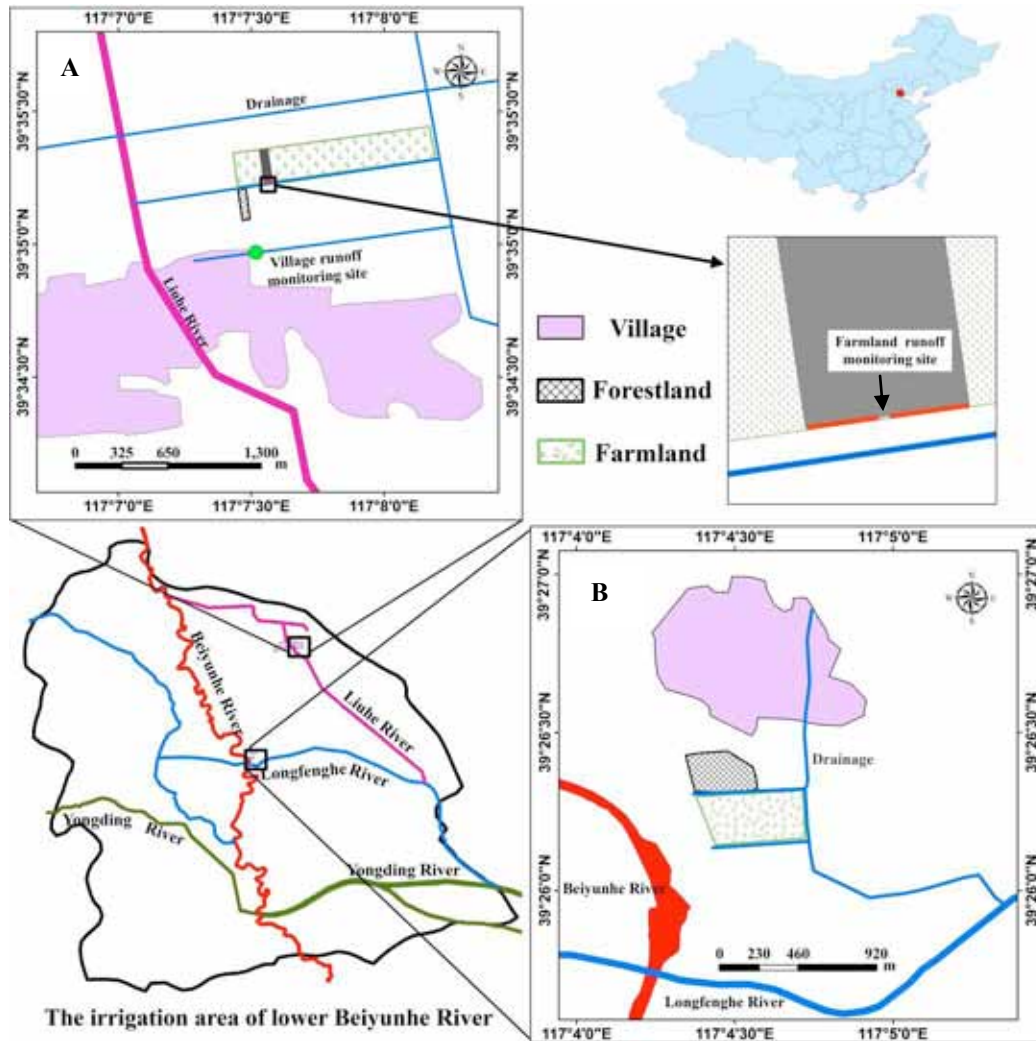


Fig.1 Rainfall-runoff monitoring sites

### 2.3. Sample collection and chemical analysis

When rainfall-runoff starts, samples were collected in accordance with 15~45min intervals. Village drainage runoff volume was measured by a Flow Meter nearby the discharge pipe. Runoff volume of farmland and forestland was both determined by estimating method, namely, calculating by requiring time to freighting a fixed volume container by the runoff. After each monitoring was completed, water samples were taken back to the laboratory immediately. Five precipitations have been monitored from July to August of 2009 and 2010 (Table 1) in two monitoring areas.

Table 1 Brief description of monitoring conditions during rainfall events in two monitoring areas

Sites	Date	The total sampling time /min	Rainfall/mm	Rainfall intensity/ mm • min <sup>-1</sup>	Number of samples
B	2009-07-22	65	94.8	0.136	6
	2009-08-06	45	7.9	0.043	4
	2009-08-19	240	24.7	0.064	7
A	2010-07-10	45	13.1	0.262	6
	2010-08-19	120	60.5	0.403	21

All runoff samples were immediately filtered (0.45 μ m) and stored at 4°C. Analysis was complete within 24 h of collection. Filtered samples of runoff were analyzed for NO<sub>3</sub><sup>-</sup>-N, NH<sub>4</sub><sup>+</sup>-N, dissolved total phosphorus (DP), total organic carbon (TOC) and dissolved total N (TN), unfiltered water samples were analyzed for COD and TP. NO<sub>3</sub><sup>-</sup>-N, COD, and ammonium nitrogen (NH<sub>4</sub><sup>+</sup>-N) concentrations were determined using the methods recommended by Ministry of Environment Protection of P.R. China [14]. An automatic total organic carbon (TOC) online analyzer (MultiN/C UV; Analytik Jena Inc., Germany) was used to determine TN and TOC concentrations. The analytical method of ammonium molybdate spectrophotometric was used to determine TP and DP concentrations. Particulate phosphorus (PP) concentrations were calculated as the difference between the total and dissolved concentrations.

#### 2.4. Data analysis

The use of an event mean concentration (*EMC*) is appropriate for evaluating the effects of rainfall-runoff on receiving waters. Receiving water bodies respond relatively slowly to storm inflows compared to the rate at which constituent concentrations change during a storm event. Thus, *EMC* is an important analytical parameter. The *EMC* represents a flow weighted average concentration computed as the total pollutant mass divided by the total runoff volume, for an event of duration  $t_r$  [15].

$$EMC = \frac{M}{V} = \frac{\int_0^{t_r} C_t Q_t dt}{\int_0^{t_r} Q_t dt} \cong \frac{\sum C_t Q_t \Delta t}{\sum Q_t \Delta t} \quad (1)$$

where *EMC*, event mean concentration (mg/l); *M*=total mass of pollutant over entire event duration(g); *V*, total volume of flow over entire event duration (m<sup>3</sup>); *t*, time (min); *C<sub>t</sub>*, time variable *t* concentration (mg/l); *Q<sub>t</sub>*, time variable flow (m<sup>3</sup>/min); and  $\Delta t$ , discrete time interval (min). The *EMC* is computed for the entire runoff duration, for a time less than the full runoff duration, a partial event mean concentration (*PEMC*) can be defined as

$$PEMC = \frac{m(t)}{v(t)} = \frac{\int_0^t C_t Q_t dt}{\int_0^t Q_t dt} \cong \frac{\sum C_t Q_t \Delta t}{\sum Q_t \Delta t} \quad (2)$$

where *m(t)*, pollutant mass transported up to time *t* (g); *v(t)*, flow volume up to time *t* (m<sup>3</sup>). From Eqs. (1) and (2) it follows that  $M=m(t_r)$ ,  $V=v(t_r)$ ; and  $PEMC(t_r)=EMC$ .

The cumulative load curves (CLCs) was used to examine the first-flush (the increasing rate of nutrient loads is above that of runoff) during the rainfall events [16-17]. This method plots cumulative runoff ratio on the horizontal axis and cumulative load ratio on the vertical axis.

The dimensionless normalized mass and flow volumes are needed from the storm runoff. Eqs. (3) and (4) represent the first flush phenomenon;

$$L = m(t) / M \quad (3)$$

$$F = v(t) / V \quad (4)$$

where *L*, dimensionless cumulative pollutant mass; *F*, dimensionless cumulative runoff volume. A first flush exists at time *t* if the dimensionless cumulative pollutant mass *L* exceeds the dimensionless cumulative runoff volume *F* at all instances during the storm events. A 45° line, when plotting *L* vs. *F*, indicates that pollutants are uniformly distributed throughout the storm events. If the data for a particular

storm falls above the  $45^\circ$  line, a first flush is suggested and  $PEMC(t_r) \geq EMC$ . Conversely, dilution is assumed to occur when the data falls below the  $45^\circ$  line and a first flush fails to occur. Every  $L$  curve can be fitted with  $F$  approximately by a power function [18]

$$L = F^b \quad (5)$$

where  $b$ , first flush coefficient.  $b$  indicates that the gap between the  $L$  curve and the bisector. A first flush occurred when this coefficient was smaller than 1. Dilution occurred when this coefficient was larger than 1. The strength of the first flush has an inverse relation to  $b$ .

### 3. Results and discussion

#### 3.1. The characteristic of runoff

Table 2 EMC of runoff from different land-use types

land use	Rainfall event	TP	DP	PP	COD	TOC	TN	NH <sub>4</sub> <sup>+</sup> -N	NO <sub>3</sub> <sup>-</sup> -N
Forestland	5	0.168	0.039	0.130	27.162	10.285	3.138	1.171	0.597
Farmland	3	0.442	0.338	0.134	45.067	19.674	12.679	1.686	5.091
Village	5	0.218	0.084	0.104	18.030	10.837	17.814	1.062	9.869

To compare characteristics of runoff water quality from different land uses (forestland, farmland, and village), the event mean concentrations of TP, DP, PP, COD, TOC, TN, NH<sub>4</sub><sup>+</sup>-N, and NO<sub>3</sub><sup>-</sup>-N were calculated. As can be seen from Table 2, there are great difference in EMCs of each indices, total phosphorus in the order: farmland > village > Forestland; COD in the order: Farmland > Forestland > Village; TN in the order: Village > Farmland > Forestland.

The EMCs of TP and TN at village and farmland were 2~5 times higher than those at forestland, suggesting that the two land-use types with high domestic pollution or intensive agricultural production greatly discharged these nutrients. The TP loss concentration of forestland and village were lower than that of farmland, indicating that the great loss of phosphate fertilizer in farmland. But the loss of particle phosphate was no significant difference among different land uses. Due to anaerobic conditions caused by surface vegetation coverage, the content of reducing substances were high, along with extensive use of organic fertilizer in farmland, EMC of COD at farmland was highest, followed by forestland and the village least.

In addition, statistical characteristics of instantaneous concentration of pollutants of several precipitations were carried out, also. From the box chart (Fig.2), we can figure out the pollutions in runoff are significant different from three land use types. For total nitrogen, Farmland and villages shared the relatively high instantaneous concentration of pollutions (instantaneous concentration were up to 20.68mg/L and 36.68mg/L, respectively), a wide range (4.73~20.68mg/L and 3.27~38.68 mg/L, respectively), mean (10.87mg/L, 13.50mg/L) and the median value (9.81mg/L, 11.40mg/L) are also relatively high. But forestland's instantaneous concentrations were relatively low (10.90mg/L), and the range was relatively narrow (1.62~10.90mg/L). The laws of NH<sub>4</sub><sup>+</sup>-N and NO<sub>3</sub><sup>-</sup>-N were similar to the total nitrogen. For total phosphate, particle phosphate, dissolve phosphate, COD, and TOC, and mean and median values of these indices from farmland were higher than in other land uses. In addition, the instantaneous concentrations from village had shown a narrow range.

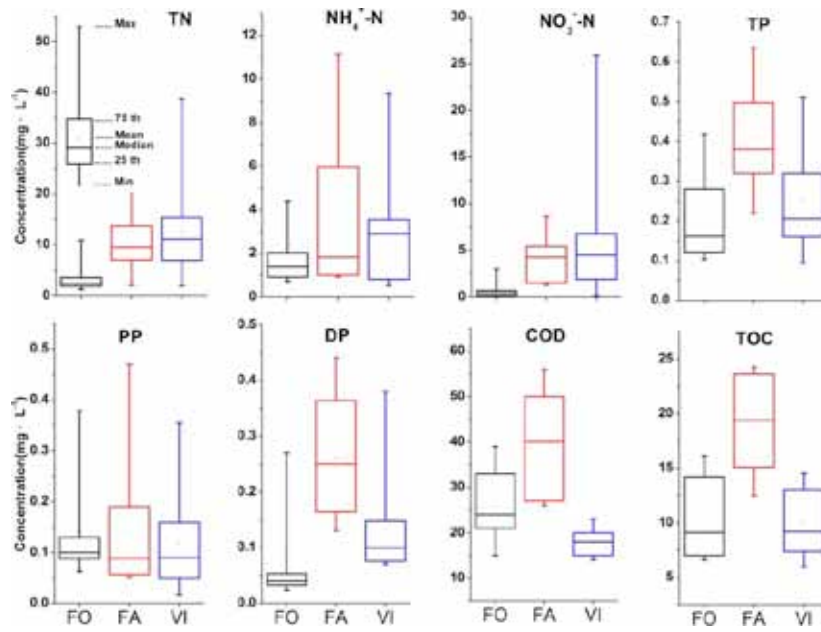


Fig.2 Comparison of runoff concentration in three different land-use types (Forestland, farmland and village were abbreviated as FO, FA, and VI, respectively).

3.2. Changes in pollution concentrations during the runoff process

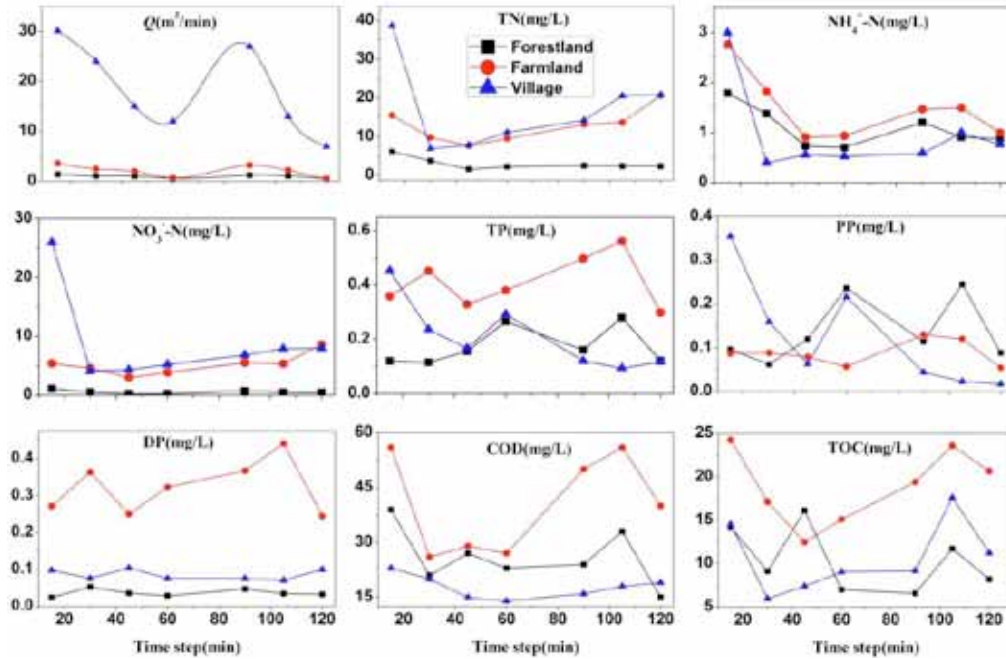


Fig.3 Nutrients concentrations during the rainfall-runoff event

Since the rainfall of August 19, 2010 was relatively large, and duration of runoff was long, the runoff characteristics of nutrient species were analyzed using nutrient concentrations and flow of this rainfall event. Time step of the horizontal axis was set at 0 when water sampling was started. Runoff flow has two peaks, at 90 minutes and 15 minutes, respectively.

When the runoff generated, the changes of nutrient concentrations loss from these land use monitoring sites can be seen from Fig.3. The flows at village, where instantaneous runoff flow can up to  $30.7\text{m}^3/\text{min}$ , were higher than that of farmland and forest. The runoff flow and the concentrations of TN,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , TP, PP, COD and TOC have two peaks and the location of the second peak of runoff flow preceded that of TN,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , TP, COD and TOC for this events. This preceding increase in nutrient concentrations can be interpreted by the theory of first-flush, which is examined in details in the next section.

The initial total nitrogen content of the village was  $38.68\text{mg/L}$ , and its concentration decreased to  $6.95\text{mg/L}$  after 30min, rapidly. Then the concentration rose slowly with time, and the concentration got to another peak in the end of runoff. In short, the density is high in the initial time, tends to decreasing with rainfall continuing, and increases in the later period. Farmland and forest land have also shown the same law, but relatively slow small fluctuations in forest. The same trend was reported by Li *et al.* [19] in Taihu basin(China). The curves of nitrogen compounds:  $\text{NO}_3^-\text{-N}$  and  $\text{NH}_4^+\text{-N}$  were similar to dissolve total nitrogen. And more information about the loss of nitrogen you can refer to our previous research [20].

The regularity of TP, PP and DP concentrations with runoff time was not obvious in different land use types. The same phenomenon was reported in previous literatures and it may be due to its adhesive properties with particulate material [21]. The two phosphate species: particle phosphate and dissolve phosphate loss proportion of total phosphate were significant different among the three land uses types (Fig.4). For forestland, as the absence of human inputs, the content of dissolve phosphate was relatively low, and also because of the absorption by vegetation, the particle phosphate became the dominant specie, accounting for more than 75% of total phosphate. For farmland, dissolve phosphate was the dominant specie, accounting for 79% of total phosphate. This may due to unreasonable application of phosphate fertilizer. For Village, the main loss form was particle phosphate in the early rainfall, and with the extensive of rainfall, the main loss form was dissolve phosphate. The form of dissolve and particle phosphate accounted for 52% and 48% respectively. The loss of particle phosphate may caused by the rain erosion in the early rainfall for low vegetation cover, with the rainfall-intensity and runoff flow decrease in the end of rainfall, dissolve phosphate became the dominate form of loss.

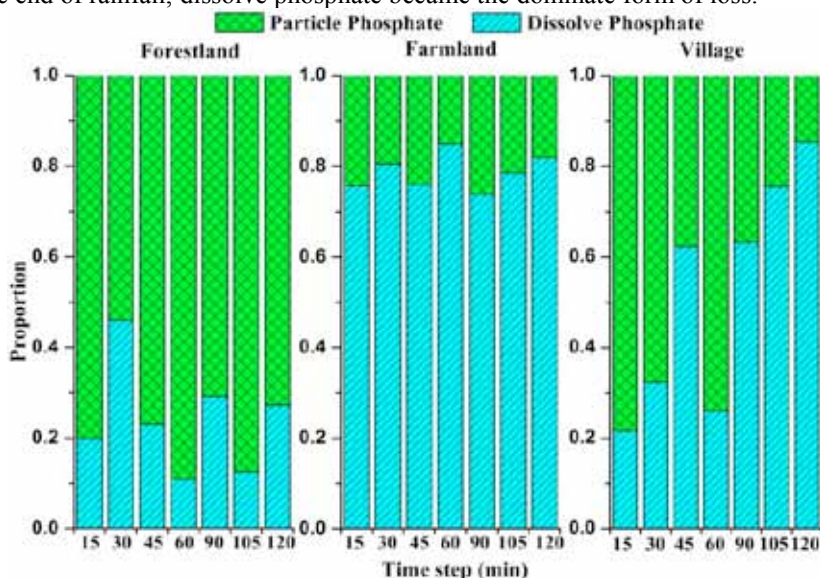


Fig.4 The loss ratio of dissolve and particle phosphate

The curves of COD and TOC in the three land use types had been shown as high in the initial time, tending to increasing with rainfall continuing, and decrease in the later period. Due to the vegetation residues enriched in the surface, along with organic fertilizers extensive used in farmland, the concentration in farmland and forestland were higher than village. As full interact for runoff and soil, the organic matter concentration gradually increased, and in the later period of producing runoff, the flow was small, these organic macromolecules tended to precipitate, and then the contents decrease eventually.

### 3.3. First-flush

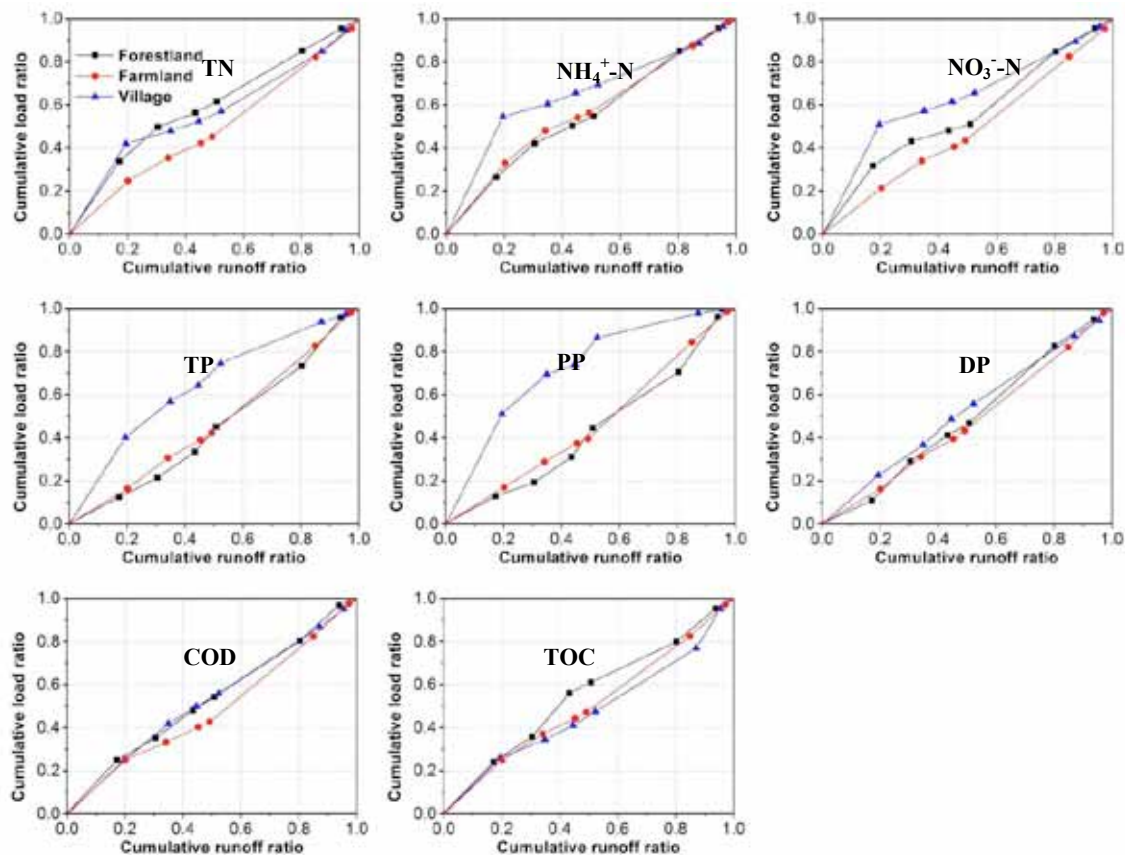


Fig.5 Cumulative load curves (CLCs) for a runoff event

The strength of first-flush was examined to clarify the differences in the transport of each nutrient. Fig. 5 shows the CLCs of nutrient species for a rainfall event. Most of the CLCs for TN,  $\text{NH}_4^+\text{-N}$ ,  $\text{NO}_3^-\text{-N}$ , TP, and PP showed a positive deviation, indicating that first-flush occurred, especially in village. The CLCs for DP, COD, and TOC were along the 1:1 straight line, indicating that first-flush was weak. Although the strength order of the first-flush was different among different land use types, on average it was  $\text{NH}_4^+\text{-N}$  (b values 0.39~0.76, mean 0.62) > TN (0.57~0.91, 0.70) >  $\text{NO}_3^-\text{-N}$  (0.45~0.99, 0.71) > TOC (0.81~0.89, 0.86) > COD (0.82~0.93, 0.87) > PP (0.39~1.24, 0.92) > TP (0.55~1.22, 0.97) > DP (0.91~1.22, 1.09). The strength order of the first-flush was consistent with the previous studies. Lee et al. [22] reported that the strength of the first-flush was in the order of TN > COD > DP for the residential area. Stutter et al. [21] analyzed the first-flush in two agricultural catchments in NE Scotland, and conclude that the strength of the first-flush was in the order of PP > DP. Huang et al. [23] studied the first flush effect from urban lawn rainfall runoff, conclude that strength of it was in the order of  $\text{NO}_3^-\text{-N}$  > COD > TP. Tasuku et al. [7] pointed out the first-flush in agriculture was in order of PP > TP > DP.



In addition, the first-flush of three different land use types were significant different. For the village, the relative strength of the first flush was  $PP > NH_4^+-N > NO_3^--N > TP > TN > COD > TOC > DP$ . For the forestland, the relative strength of the first flush was  $TN > NO_3^--N > NH_4^+-N > TOC > COD > DP > TP > PP$ . And for the farmland, the relative strength of the first flush was  $NH_4^+-N > TOC > TN > COD > NO_3^--N > DP > TP > PP$ . To consider all the nutrients, the strength order of the first-flush on average it was village (*b* values 0.39~0.91, mean 0.62) > forestland (0.61~1.24, 0.92) > farmland (0.70~1.15, 0.98).

In village, the first-flush of the particulate materials (PP) was stronger than that of other nutrients. Particulate materials held in drainage canal substrate sediments and in surface soils through overland flow are considered to be the major causes of first-flush [7]. The DP and  $NH_4^+-N$  loads were usually adsorbed by soil particles because Andosols have high capacity to hold phosphorus and the ammonium ion ( $NH_4^+-N$ ) or a cation is held by the negatively charged soil particles. Therefore, the DP and  $NH_4^+-N$  loads were thought to be desorbed from the soil particles by the rainwater over time and their runoff characteristics were different from other nutrient species. But for heavy domestic pollution and microbial activity in village,  $NH_4^+-N$  can easier enrich in the surface than phosphorus, means that the mass loads occurred in the early period of runoff. So, the strength first-flush was stronger. The *b* value of DP was close to 1. The transport pathway of DP, tending to uniformly distributed throughout the storm event.

But for forestland and farmland, which both covered by vegetation in the earth, the strength of first-flush were significant weaker than in village, especially for particulate material. This result may due to the buffer of runoff and possible impacts of the absorption and retention of vegetation. In farmland, all of the pollutions tended to uniformly distribute or dilution throughout the storm event expect for  $NH_4^+-N$ . Similar conclusions have also met with the research carried by Obermann *et al.* [24] in an agriculture watershed.  $NH_4^+-N$ , which was the main form of nitrogen fertilizer in this research area, accumulated in the surface of soil. So the strength of  $NH_4^+-N$  was stronger than that in forestland.

#### 4. Conclusions

The results suggested that the nutrient concentrations of runoff under different land uses in the irrigation area of lower Beiyunhe River were significant different, EMC of total phosphorus in the order: farmland > village > Forestland; EMC of COD in the order: Farmland > Forestland > Village; EMC of TN in the order: Village > Farmland > Forestland.

The changes in the nutrient concentrations of the runoff water over a rainfall event indicated that the transports of the nutrients are similar among different land use types. The instantaneous concentrations of TN,  $NH_4^+-N$ , and  $NO_3^--N$  were high in the initial period of runoff, tend to decreasing with rainfall continuing, and increase in later period. Due to adhesive properties with particulate material, phosphorus concentration with time variation was not obvious among three land use types. The phosphorus species with high proportion in the total phosphorus was particle P (accounting for 75%) in forestland, dissolve P (79%) in farmland, and particle P (48%) and dissolve P (52%) in village. The curves of COD and TOC in the three land use types had been shown as high in the initial time, tending to increasing with rainfall continuing, and decrease in the later period.

Among different land use types, the strength order of the first-flush on average it was  $NH_4^+-N > TN > NO_3^--N > TOC > COD > PP > TP > DP$ . A first-flush have taken place among all of pollutions except for DP. To consider all the nutrients, the strength order of the first-flush on average it was village > forestland > farmland. Due to the buffer of runoff and possible impacts of the absorption and retention of vegetation, the first-flush in forestland and farmland was close to each other and both weaker than that in village, especially for PP. In village, all of the pollutions have taken place the phenomenon of first flush, while in farmland, pollutions tended to uniformly distribute or dilution throughout the storm event.

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