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**WATER QUALITY AND PROTECTION:
ENVIRONMENTAL ASPECTS**

Soil Erosion and Pollutant Transport during Rainfall-Runoff Processes¹

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Abstract—The pollutant from land surface applied to agricultural chemicals is one of the major sources of contamination in water bodies. The pollutant transport within a watershed is profoundly influenced by the rainfall-runoff processes, especially the associated upland erosion and sediment transport processes because most of pollutant can be dissolved into water or attached to the soil particles. A set of soil experiments in laboratory was conducted in this paper to investigate the impacts of upland erosion and sediment transport on pollutant loads. The soil utilized for the experiments was the silty sand collected from Loess Plateau, China; and ammonium bicarbonate was applied on the soil surface as the pollutant source. Runoff discharge, soil loss, and ammonia- and nitrate-nitrogen concentrations were measured to establish the relationships which can help the numerical model to predict the pollutant losses coupled with upland soil erosion during the rainfall-runoff processes. The experimental results indicate the ammonia-nitrogen concentration in runoff reaches the peak at the initial stage of the overland flow generation, and quickly decreases and approaches to the steady state. The ammonia-nitrogen transported by the soil loss also makes contributions to the nitrogen loss; and its amount mainly depends on the soil transport rate. The ammonia-nitrogen dissolved in overland flow is dominant due to the strong aqueous solution of ammonium bicarbonate during the first storm right after its application.

Keywords: pollutant transport, rainfall-runoff, soil erosion, sediment transport, sorption

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INTRODUCTION

Agricultural chemicals (pesticides and nitrogen fertilizers) are widely used in agricultural fertilization activities. The pollutant from agricultural chemicals is often transported into creeks, rivers, and lakes through the rainfall-runoff process since most pollutants can be dissolved in water or attached to the soil particle, which may cause non-point source (NPS) pollution within a watershed [28]. The studies related to the agricultural NPS pollution have been fervent in many fields since 1990s [4, 7, 8, 14, 17, 20, 25, 26, 41]. The transport and transformation mechanism of agricultural pollutant is a complex process. Pollutant, sediment, rainfall, runoff, infiltration, groundwater are interlaced in the above process. Rainfall, for instance, mainly affects the soil through raindrop splash erosion and runoff sheet erosion, including detaching the topsoil into soil particles and transporting soil particles by runoff. Soil erosion appears in quick succession since more particles have been carried away during the activity of rainfall-runoff [39]. The pollutant in topsoil is influenced by eluviation, runoff, and soil erosion since most of them could be attached on soil particles

or dissolved in water as the rains continue. As the pollutant is transported into surrounding waters by runoff in suspended state and dissolved state, it can contaminate the waters through advection and diffusion [9, 23, 30, 37]. Sediments from the erosion process are also the source of pollution since they not only absorb and transport contaminants but also affect the hydro system by erosion and deposition. The aforementioned process has been recognized to induce destruction of farmland, siltation of rivers, and depression of water ecosystems [6, 36].

The individual characteristics of soil erosion and pollutant transport during the rainfall-runoff process have been studied with different experiments and numerical models. The dynamic process of between soil erosion and rainfall-runoff has been well studied by many scientists [3, 15, 24, 34]. Wang et al. (1990) found the characteristic of nitrogen loss of cultivated loessial soil under erosion condition. Many researchers have realized the characteristics of soil erosion and pollutant transport are susceptible to many factors such as rainfall intensity, soil type, plants, land topography, etc. Zhang et al. (2004) and You et al. (2012) found vegetation could affect soil erosion and pollutant transport a lot since it effectively reduced soil ero-

¹ The article is published in the original.

Fig. 1. (a) Schematic diagram of A- and B-zones, (b) experimental set-up, intensity = 90 mm/h.

sion and pollutant loading compared to bare soil. Shi (2009) also realized the characteristics of nitrogen loss on different slopes under rainfall were quite different. In addition, some numerical models were developed to analyze the characteristics of soil erosion and/or pollutant transport during the rainfall-runoff process at different scales [1, 10, 12, 27, 33]. The dynamics between soil erosion and pollutant losses, however, is less considered in the previous studies. The quantified relationships necessary for the practice and the numerical modeling should be established to understand the mechanism of pollutant transport and transformation for the watershed management and predict the pollutant losses in numerical simulations. Therefore, in this paper a set of laboratory experiments is provided to obtain the relationships between upland erosion, sediment transport, and pollutant transport during rainfall-runoff processes. The relationships are then analyzed in the form of linear formulations which can be simply applied in water managements and numerical models.

MATERIALS AND METHODS

The laboratory experiments were conducted with the soil flume which was made with the metal material in 3 m (L) $\times$ 1 m (W) $\times$ 0.5 m (H). The slope of the flume was adjustable by the hydraulic devices. A steel partition was then applied to evenly divide the soil flume into two zones, labeled as A-zone and B-zone (Fig. 1a). The settings for both zones were the same for all tests. The main purposes to separate the flume are: (a) to demonstrate the similarity of the experiment; and (b) to avoid the effect of asymmetric soil surface. The soil type used for experiments was the silty sand and classified as relatively loess from China's Loess Plateau. The soil diameter analysis showed that the soil texture consists of 12.9% clay, 58% silt, and 29.8% sand, which accorded to the loess properties. Because of the characteristics of the sand, the preparation of sand bed in the flume was carefully and gradually layered into the flume by adding 0.01 m thickness each time. The finalized thickness was 0.05 m for all

tests. In addition to the sand layer on the bottom of soil flume, there were two apertures formed at the bottom of soil flume to allow soil moisture and seepage to freely infiltrate, as shown in Fig. 1b. Considering the maneuverability and simplicity of this experiment, the tested slopes in this study were set to 15° and 20°, which were general slopes for farmlands on the Loess Plateau in China.

The rainfall for tests was simulated by the simulator which is composed of a pressure irrigation sprinkler and inflow pipes (Fig. 1b). Three nozzles were installed 2 m above the soil surface with the distance between each other of 1 m to ensure the uniformity of the rainfall distribution. The uniformity was optimized and calibrated with the previous setting. The intensity of the rainfall simulator was designed for 90 mm/hour and the duration was set to 35 minutes. In the tests, the rainfall with short duration and high strength was to simulate the scenarios for the local characteristics. The simulation of ammonium bicarbonate was targeted to replicate the real agricultural activities which the farmers spread it uniformly on the soil surface. The average amount applied in this experiment was 50 g/m². The runoff data were collected with metal containers at the downstream end of the soil flume where a plastic shed was fixed to prevent the containers from collecting the rainwater outside of the flume in the process of experiment. The collecting time of the runoff samples and the frequency are shown in table.

Runoff samples collected from A-zone and B-zone were properly preserved until the sediments were fully deposited. The clear water above the sediment in each sample was then carefully extracted by the needle tubing for the analysis. All of the weight of sediment was measured when sediments were fully dried. The runoff

The collecting time of runoff data

Time	0~2 min	2~35 min
Collecting period	10 s	20 s
No-collecting interval	10 s	40 s

Fig. 2. (a) Runoff rate along the rainfall simulation, intensity = 90 mm/h; (b) erosion rate along the rainfall simulation, intensity = 90 mm/h; (c) sediment concentration along the rainfall simulation, intensity = 90 mm/h; (d) Relationship between Soil erosion and Runoff rate under 15° slope ($y = -0.14x + 4.21$) and 20° slope ($y = -0.11x + 3.75$), intensity = 90 mm/h.

volume (clear water) was then calculated with the subtracting the weight of sediments from the total weight. The clear water was mainly used for measuring the pollutant contents in runoff and the sediment was used to analyze the absorptive effect of soil particles on agricultural pollutant. Ammonia-nitrogen and nitrate-nitrogen concentrations were analyzed in different methods. The concentration of ammonia-nitrogen was measured with Indophenol Blue Colorimetric Method (IBCM). The test sample was mixed with potassium chloride (KCl) solution in an electronic oscillator and then reacted with hypochlorite and phenol in the strong alkaline medium. As the soluble dye indophenol blue was generated in the reaction process, the concentration of ammonia-nitrogen could be measured via colorimetric analysis in the spectrophotometer. Ultraviolet spectrophotometry was used to measure the nitrate-nitrogen concentration. Similarly, the test sample was mixed with KCl and oscillated for 1 h, then the absorbance at 220 and 275 nm of leaching solution could be determined with the spectrophotometer. The concentration of nitrate-nitrogen was then measured according to dual-wavelength ultraviolet spectrophotometry [18].

RESULTS AND DISCUSSION

The experiments were designed to understand the transfer mechanism of soil erosion, pollutant transport, and runoff process. Variable values of pollutant concentration, sediment yield and runoff volume were measured to characterize the pollutant transport on loess under different slopes. For each test, two flume slopes (15° and 20°) were conducted and all simulated scenarios were simultaneously and constantly applied to A- and B-zones.

Runoff and Soil Erosion Experiment

In this test the amount of the runoff and the soil erosion rate were collected through the time to construct the relationship. For 15° flume slope A- and B-zones show that the runoff appeared at 3.5 min after the rainfall started and quickly increased up to inflection point 26 mL/s at 6.3 min (Fig. 2a). The runoff increased rapidly in the beginning is the effect of bare soil surface crusting. The crust was observed and formed into a thin but dense layer on the top after the raindrops fell on the loess. This crusting layer reduced rainwater infiltration and increased overland flow. Cai et al. (1990) showed that runoff on loess surface with crusting was an order of magnitude higher than the

one without crusting. Once the surface crusting occurs, the runoff may greatly increase. In this test, the runoff gradually reached a steady state with a constant value after the rapid increment in the beginning. The similar result was obtained in the 20° test. The runoff also appeared at 3.5 min and quickly reached inflection point 27 mL/s at 8.3 min because of surface crusting and then kept steady until the end of the simulation. The delayed time between two different slopes was 2 minutes. This is because the force component of raindrop in the perpendicular direction on the soil surface is smaller when the slope is steeper. The steeper slope weakens the effect of splash erosion on loess and decelerates the formation of the crust. The runoff rate in 20° increased more slowly than in 15°. However, the runoff rate in the 20° was larger in the later stage. It is because the steeper slope created large kinetic energy of runoff in the trial.

The measurements collected from two zones in the above test were consistent which indicates the experiment was controlled. Therefore, the average results of soil erosion, ammonia-nitrogen, and nitrate-nitrogen concentration obtained from both zones are presented in the following sections. The soil erosion measurement through time is presented in Fig. 2b. It shows that the erosion occurred at 3.5 min as the same of the runoff appeared and reached to the peak in 20 s, and subsequently decreased to a steady rate of 0.3 g/s. The results can derive that splash erosion was dominant before the surface crusting was formed in the initial stage of the storm, and then the soil particles were quickly rushed away by the runoff. Comparing to the time at inflection point of runoff test, the sediment yield reached the peak value before the runoff yield did. This indicates the crusting constrain the sediment yield in the beginning stage of the rainfall-runoff process. Liu et al. (2004) showed that surface soil would be dense and solid after the crust was formed. This would cause the decreasing erosion rate. The peak values of the erosion rate between 15° and 20° show the erosion is proportional to soil slopes within a certain range of slopes. The steeper loess slope induces the bigger flow velocity and erosion rate [29, 32].

The sediment concentration changing with time acted similar to the erosion rate (see Fig. 2c), while the sediment concentration during the later stage was stable with a low value and was different from the fluctuating situation of erosion rate. As the erodible particles were washed away, the sediment concentration decreased over time until it reached a stable but low state.

In Fig. 2d, a negative correlation between the erosion rate and the runoff rate in both slopes is obtained and represented as $\text{Erosion rate} = A \times \text{runoff rate} + B$, where A and B are the parameters. The erosion rate progressively decreased with the increase of runoff rate. This linear relationship indicates the effect of splash erosion on loess was dominant compared to

sheet erosion. The erosion pattern measured by Kothari et al. (2004) is different from Fig. 2d, which is due to the fact that the erosion pattern is susceptible to soil types, rainfall intensity and soil slopes [3, 15]. The absolute values of the slopes of the linear equations were almost the same for different slopes (see Fig. 2d), which means the changing of the flume slope in this experiment made little difference to the relationship between erosion rate and runoff rate.

Runoff and Pollutant Transport Experiment

One of the pollutants applied on the soil in the study was ammonium bicarbonate. Ammonia-nitrogen represented the basic state of ammonium bicarbonate undergoing erosion due to the action of rainfall and runoff. The peak of ammonia-nitrogen concentration from runoff, occurring at 5 min after the experiment began to rain. Its value was 500 mg/L and then was immediately down to a constant value of 20 mg/L (Fig. 3a). The main reason is that most of ammonium bicarbonate was likely washed away in the first phase of the storm due to its high soluble property. Nitrate-nitrogen was conducted in this test as the other indicator. The time series result of the nitrate-nitrogen concentration significantly differed from ammonia-nitrogen. The peak of the concentration was occurred at the moment that the runoff appeared (see Fig. 3b). The nitrate-nitrogen concentration was at low ebb while the ammonia-nitrogen reached the peak at 5 min. This is related to the property of ammonium bicarbonate and the mechanism of nitrate-nitrogen transport since the concentration of nitrate-nitrogen and ammonia-nitrogen were in a poor correlation.

The pollutant transport rate was estimated by nitrogen loss. The dissolved ammonia-nitrogen loss in runoff and the dissolved nitrate-nitrogen loss in runoff were estimated in the study. The performance of the dissolved ammonia-nitrogen loss in runoff was similar with the concentration case which increased to the peak at initial period then rapidly decreased to the stable phase (Fig. 3a). The trend of dissolved nitrate-nitrogen loss in runoff, on the other hand, did not decrease over time (Fig. 3b) which was quite different to its concentration case. It is because that the ammonia-nitrogen loss in runoff is mainly determined by its concentration in runoff, while the nitrate-nitrogen loss is determined by runoff volumes. The average pollutant transport rate in runoff was then calculated by the total ammonia-nitrogen and nitrate-nitrogen loss in unit time in the overland flow. The average pollutant transport rate was 2.21 mg/s for 15° and 1.75 mg/s for 20°. The above result confirmed that the lower velocity of runoff on flatter slope enhances time contact for the dissolution reaction of pollutants [21]. The correlation between ammonia-nitrogen concentration and runoff rate was shown in a linear relationship (see Fig. 3c), which is similar with the correlation between nitrate-

Fig. 3. (a) NH_4^+-N concentration and loss rate in runoff along the rainfall simulation, intensity = 90 mm/h; (b) NO_3^--N concentration and loss rate in runoff along the rainfall simulation, intensity = 90 mm/h; (c) relationship between NO_3^--N concentration and runoff rate under the 15° slope ($y = -0.47x + 20.75$) and 20° slope ($y = -0.23x + 13.25$), intensity = 90 mm/h; (d) relationship between NO_3^--N concentration and Runoff rate under the 15° slope ($y = -0.47x + 20.75$) and 20° slope ($y = -0.23x + 13.25$), intensity = 90 mm/h.

nitrogen concentration and runoff rate (see Fig. 3d). The 15° shows the steeper slope than the 20° which means the ammonia-nitrogen and nitrate-nitrogen on lower slope might be more easily influenced by runoff compared to the one on upper slope.

Soil Erosion and Pollutant Transport Experiment

The transfer mechanism of the pollutant under the rainfall-runoff process should consider the pollutant dissolved in water and soil. The aforementioned experiments reflect effect of the runoff rate. The soil and pollutant amount absorbed by the soil are discussed in this section. For the ammonia-nitrogen absorbed by the soil, the peak of the concentration appeared at the initial runoff event and rapidly fell to approximating to zero value (Fig. 4a). The concentration of ammonia-nitrogen absorbed by soil was of great correlation with soil erosion rate as shown in Fig. 4b. The adsorption effect of sediment on ammonia-nitrogen was more obvious under the steeper soil slope since the steeper slope produced larger sediment

yield. Figure 5 shows the concentration ratios of ammonia-nitrogen dissolved in runoff to that absorbed by sediment under 15° and 20° slopes. The result demonstrates the ammonia-nitrogen dissolved in runoff was dominant during the storm after its application since ammonium bicarbonate can be easily dissolved in water. The ratio under 20° slope demonstrates the concentration of ammonia-nitrogen dissolved in runoff was larger under steeper slope.

The first differentiation of all measured variables to time was estimated (Fig. 6). The measured data were normalized before the differentiation. The results indicated variables were mostly affected by the rainfall in the initial stage (<10 min), while they were less affected in the later stage (>10 min) for both slopes. The nitrate-nitrogen concentration compared to others was less fluctuated through the time for both slopes. It follows that nitrate-nitrogen concentration were less affected by the process of rainfall over time.

Fig. 4. (a) Concentration of $\text{NH}_4^+\text{-N}$ absorbed by sediment along the rainfall simulation, intensity = 90 mm/h; (b) relationship between $\text{NH}_4^+\text{-N}$ absorbed by sediment and Erosion rate under the 15° slope ($y = 21.19x - 3.53$) and 20° slope ($y = 43.23x - 18.30$), intensity = 90 mm/h.

CONCLUSIONS

The contamination of agricultural chemical washed from the land surface applied to the watershed is the important problem for the environmental protection. The primary transfer mechanism of rainfall-runoff pollutant from land to water is a complex network process. The watershed characteristics and the precipitation process are interacted and complicated which obstacle the understanding of rainfall-runoff pollutant directly from the field measurements. This paper conducted a set of laboratory experiments with simplifying the control factors to understand this complex pollutant transfer process during the rainfall-runoff situation. The results show that the dissolution in runoff and adsorption by soil particles are the main transport means for pollutants during the rainfall-runoff on loess slopes. In this study, a set of rainfall experiments have been conducted to examine the impacts of rainfall-runoff and upland erosion on pollutant loads and transport by using a soil flume with two different slopes. They show the runoff promptly increases and reaches the steady state because the effect of bare soil surface crusting. The erosion rate, on the other hand, is in an opposite correlation with runoff rate since the erosion rate progressively decreases with the increase of runoff rate.

The quantity of pollutants occurs in the beginning stage of the storm when runoff and sediment erosion rates reach to the peak values, and then decreases to a relatively steady value. Comparing to the nitrate-nitrogen, the dissolved ammonia-nitrogen is dominant in overland flow during the first storm after being applied to the soil surface. The concentration of ammonia-nitrogen absorbed by soil particles has a correlation with soil erosion rate and its value increase with the soil rate. The average runoff and sediment yield under steeper slopes are generally higher than that under flatter slopes. However, the relations between average concentration of pollutants and the

loess slopes are not particularly distinct in this experiment. This study shows that the dissolved ammonia-nitrogen is the important factor to the surrounding water environment when there is a strong rainfall, the nitrogen absorbed by soils could not be ignored since it has an important contribution to pollutant losses during the rainfall-runoff and erosion processes.

Finally, this study delivers the quantitative relationship between pollutant losses and upland soil erosion under two slopes (15° and 20°). These linear equations can provide a reasonable method to consider the role of sediment when modeling of pollutant losses in rainfall-runoff processes for the local scale of problems in China. The special treatments to measure the change of pollutant concentration for numerical modeling where the soil erosion is considered can also be avoided by applying the linear equations.

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Fig. 5. Ratios of $\text{NH}_4^+\text{-N}$ dissolved in runoff to $\text{NH}_4^+\text{-N}$ absorbed by sediment under the 15° and 20° slopes, intensity = 90 mm/h.

Fig. 6. The first difference of the normalization time series data of each variable under the 15° (a) and 20° (b) slope, intensity = 90 mm/h.

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