



Effects of straw and biochar amendment on hydrological fluxes of dissolved organic carbon in a subtropical montane agricultural landscape[☆]

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ABSTRACT

Straw and biochar amendments have been shown to increase soil organic carbon (SOC) stocks in arable land; however, their effects on hydrological fluxes of dissolved organic carbon (DOC), which may offset the benefits of C sequestration amounts remain uncertain. Therefore, we conducted a three-year field study that included four treatments (CK, control with no fertilizer; NPK, synthetic N fertilizer; RSDNPK, synthetic N fertilizer plus crop residues; BCNPK, synthetic N fertilizer plus biochar of crop straw) to investigate the effects of straw and biochar amendment on DOC losses through hydrological pathways of overland flow and interflow from a wheat-maize rotation system in the subtropical montane agricultural landscape. We detected substantial intra- and inter-annual variations in runoff discharge, DOC concentration, and DOC fluxes for both overland flow and interflow pathways, which were primarily attributed to variations in rainfall amount and intensity. On average, the DOC concentrations for interflow (2.98 mg C L⁻¹) were comparable with those for overland flow (2.71 mg C L⁻¹) throughout the three-year experiment. However, average annual DOC fluxes for interflow were approximately 2.60 times greater than those for overland flow, which probably related to higher runoff discharges of interflow than overland flow. Compared to the control, on average, the N fertilization treatments significantly decreased the annual DOC fluxes of overland flow and significantly increased annual DOC fluxes of interflow. Relative to the application of synthetic N fertilizer only, on average, crop straw amendment practice significantly increased annual DOC fluxes of interflow by 28.7%, while decreasing annual DOC fluxes of overland flow by 12.0%; in contrast, biochar amendment practice decreased annual DOC fluxes of interflow by 25.3% while increasing annual DOC fluxes of overland flow by 44.6%. Overall, considering both overland flow and interflow, crop straw amendment significantly increased hydrological DOC fluxes, whereas biochar had no significant effects on hydrological DOC fluxes throughout the three-year experiment. We conclude that crop straw incorporation strategies that aim to increase SOC stocks may enhance hydrological losses of DOC, thereby in turn offsetting its benefits in the subtropical montane agricultural landscapes.

1. Introduction

The soil organic carbon (SOC) pool is the largest C pool in terrestrial ecosystems, and plays an essential role in sustaining soil fertility and crop productivity in agricultural landscapes (Marx et al., 2017). Hydrological loss of dissolved organic carbon (DOC) is an important process in soil C cycling and a critical component of terrestrial C budgets. However, hydrological DOC loss is often an overlooked pathway of SOC losses in various terrestrial ecosystems (Froberg et al., 2005; Lohse et al.,

2009). Recent global estimates suggest that the global terrestrial DOC leaching fluxes of 0.28 Gt C yr⁻¹ account for 15% of terrestrial net ecosystem productivity (Nakhavali et al., 2021). Moreover, due to its high mobility and biological activity (Mandal et al., 2019), DOC also plays an important role in regulating the transformation and transport of nutrients and pollutants in the environment (Sane et al., 2016).

Some studies have investigated hydrological DOC loss from forest, grassland, and peatland ecosystems, while the relatively little attention has been paid to intensively managed agroecosystems (Michalzik et al.,

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2003; Qassim et al., 2014; Qiu et al., 2012). With respect to agroecosystems, positive (Camino-Serrano et al., 2016; Whittinghill et al., 2012), negative (Hagedorn et al., 2012; Lu et al., 2013), and neutral effects (Lovett et al., 2013; McDowell et al., 2004) of N additions have been reported for DOC losses. For example, ammonium-based N fertilizer additions could enhance DOC retention in the soil matrix and consequently decrease hydrological DOC loss, while sodium nitrate addition is associated with increases in DOC leaching loss in soils (Evans et al., 2008; Lu et al., 2013). Nevertheless, the manner in which the magnitude of hydrological DOC loss responds to N fertilization practices in agroecosystems remains uncertain.

Crop straw incorporation into the soil as organic fertilizer is a well-established strategy to increase SOC stocks in croplands (Malhi et al., 2012; Monaco et al., 2008; Niu et al., 2011). It also affects hydrological DOC losses via overland flow and interflow due to changes in soil C biogeochemical processes and hydrological properties (Liu et al., 2014). For instance, Prosdocimi et al. (2016) demonstrated that crop straw mulching practices decrease hydrological DOC losses, most likely related to decreases in the discharge of sediment and runoff in cropland. Similarly, some studies indicated that crop straw amendment could control soil erosion from hillslope croplands, thereby decreasing hydrological DOC losses (Rahma et al., 2017; Shi and Schulin, 2018). In contrast, previous studies reported enhancement of hydrological DOC loss following crop straw amendment because the application of crop straw could increase soil DOC availability and stimulate soil pore and hydrological path formulation (Bhattacharyya et al., 2009; Blanco-Canqui and Lal, 2007; Chirinda et al., 2010; Dong et al., 2018). Thus, the effects of crop straw amendment on hydrological DOC loss from agricultural soils still have a relatively high uncertainty.

Biochar is a carbon-rich residue generated by the pyrolysis of waste biomass under O₂-limiting conditions (Sohi, 2012). Because of its long residence time in soil, biochar has been recommended to increase C sequestration in terrestrial ecosystems (Huang et al., 2018; Joseph et al., 2010). However, biochar amendment can affect hydrological DOC loss from agricultural soils (Haeefele et al., 2011; Jones et al., 2012; Yang et al., 2017), which in turn may be beneficial for C sequestration in agricultural soils. For example, biochar application could influence soil hydraulic conductivity and porosity, thereby altering soil hydrological processes and the associated partitioning of precipitation between different hydrological pathways (Schaetzl, 2002). In addition, biochar application can affect the soil sorption capacity of DOC and consequently hydrological DOC loss (Eykelbosh et al., 2015; Haeefele et al., 2011; Smebye et al., 2016). For instance, some studies found that biochar application could significantly increase DOC concentrations in agricultural soils due to the enhanced sorption capacity of DOC in soils (Haeefele et al., 2011; Smebye et al., 2016). In contrast, Eykelbosh et al. (2015) found that biochar application decreased DOC availability in the soil matrix and consequently hydrological DOC loss. These inconsistent patterns suggest that further studies are needed to determine whether biochar amendment would induce greater hydrological DOC loss in the subtropical montane agricultural landscapes.

To date, few experimental studies have produced a detailed understanding of the effects of organic amendments on hydrological DOC losses through continuous multi-year field continuous measurements, particularly in subtropical agricultural landscapes. To address this gap in knowledge, we conducted a three-consecutive-year field study to examine the effects of straw and biochar amendment on hydrological DOC losses in both overland flow and interflow in a subtropical montane agricultural landscape in southern China. The objectives of our study were to i) quantify hydrological DOC losses with simultaneous multi-year simultaneous field measurements of overland flow and interflow, consequently exploring the main hydrological regulators, and ii) evaluate the effects of crop straw and biochar amendments on hydrological DOC losses in subtropical montane agricultural landscapes. We further hypothesized that amendments of crop straw and biochar might enhance hydrological DOC losses which may somewhat offset the

benefits of SOC stocks.

2. Materials and methods

2.1. Study site and experimental design

The study was conducted at the Yanting Agro-Ecological Station of Purplish Soil (31°16'N, 105°27'E), which belongs to the Chinese Ecosystem Research Network (CERN), in Sichuan Province, southern China. The experimental site has a subtropical climate, with a mean annual temperature of 17.5 °C, and annual precipitation of 846 mm (values given are for the period 1981 to 2018 using meteorological observations obtained at this site). Approximately 70% of the annual precipitation occurs from May to September each year.

The experimental soil is known locally as 'purple soil' due to its color and is classified by the Eutric Regosol in accordance with the FAO Soil Classification. The soil profile of the cropland in the purple soil area is shallow (20–80 cm), and beneath the shallow soil profile is the bedrock with low water conductivity (Li et al., 1991). The relatively thin soil profile can be easily saturated by precipitation water following rainfall events. The vertical infiltrating water in the soil profile can be easily informed as interflow at the soil-bedrock interface and move out of the soil along the slope. Rain-fed wheat-maize rotation is a common cropping system in this region. The physicochemical properties of topsoil (0–20 cm) can be summarized as follows: pH of 8.3 (H₂O: soil of 2.5:1.0), bulk density of 1.32 kg m⁻³, soil organic C content of 8.75 g kg⁻¹, total N content of 0.62 g kg⁻¹, and saturated hydraulic conductivity of 18.6 mm h⁻¹.

The field study included 12 free-drainage lysimeter experimental plots. Each lysimeter experimental plot had an area of 32 m² (4m × 8m) with a slope of 6.5°, which allows the simultaneous monitoring of overland flow and interflow (Fig. 1). To avoid horizontal water and matter leaching, each plot was isolated by cement with dividing walls that extended down to the bedrock to at least 60 cm. Detailed information on the design and construction of the lysimeter plots can be also found in Zhu et al. (2009). In the present study, there were four experimental treatments, including one control and three fertilizer treatments: control (no fertilization, CK), synthetic N fertilizer only (conventional N fertilization practice, NPK), synthetic N fertilizer plus crop straw (RSDNPK), and synthetic N fertilizer plus biochar (BCNPK). The form of the applied synthetic N fertilizer was ammonium bicarbonate. Wheat (maize) straw, with average N content and C:N ratio of 6.4 ± 0.6 (8.8 ± 0.7) g N kg⁻¹ and 66 ± 2 (48 ± 4), respectively, was cut into small pieces (length: approximately 5 cm) and incorporated in the plots of RSDNPK treatment prior to planting maize (wheat) (Zhou et al., 2016). The selected biochar was a crop straw made from the slow pyrolysis of wheat straw at 500 °C in a fluidized bed furnace (Sanli New Energy Company, Henan, China), and the details of its properties are shown in Liu et al. (2019). The biochar was incorporated at a depth of 20 cm with an application rate of 16 t ha⁻¹ yr⁻¹. Moreover, in accordance with the recommended N application rates for Chinese cereal systems, all fertilization treatments in the present study received equal N amounts of 280 kg N ha⁻¹ yr⁻¹ (in terms of mass, i.e. 130 kg N ha⁻¹ in the wheat season and 150 kg N ha⁻¹ in the maize season), and equal N amounts of calcium superphosphate (90 kg P₂O₅ ha⁻¹ equivalent) and potassium chloride (36 kg K₂O ha⁻¹ equivalent). All fertilizers were homogeneously incorporated into a soil depth of 20 cm as basal fertilization on the same day of crop planting.

2.2. Hydrological discharge and DOC loss measurements

We measured the discharges of overland flow interflow and took the water samples following each rainfall event over the three-year duration of the experiment. After the discharge measurement, 500-ml water samples were taken and stored in polyethylene-bottles at 4 °C prior to the DOC concentration analysis (within one week). The water samples

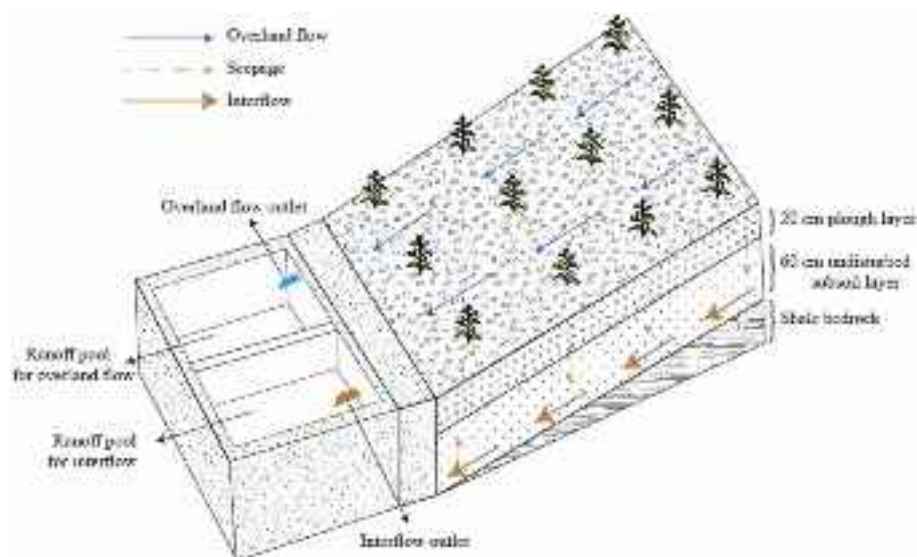


Fig. 1. Schematic illustration of runoff plot structure on the sloping upland of purple soil. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

were filtered through a 0.45 μm polyethersulfone membrane (Whatman[®]) and analyzed for DOC concentrations using a continuous flow analyzer with a chemical oxidation module (Bran + Luebbe, Nordstedt, Germany) by using the methods based on the conversion of all dissolved organic carbon into CO_2 by an ultraviolet digester.

2.3. Auxiliary measurements

Topsoil samples (0–15 cm) were collected from each plot using a soil auger twice per week, and the visible stones, roots, and other litter were removed manually before mixing completely. The mixed samples were then extracted with 0.50 M K_2SO_4 solution (soil: solution = 1:5 w/v), shaken for 1 h, then centrifuged for 10 min at 4000 rpm and filtered through a 0.45 μm polyethersulfone membrane (Whatman[®]) filter. The extracts were analyzed for NH_4^+ , NO_3^- , and DOC content using a continuous flow analyzer with a chemical oxidation module (Auto Analyzer 3, SEAL Analytical, Germany). The temperature and moisture content of the topsoil (0–5 cm) were monitored using a manual thermometer (JM624, Jinming Instrument Co. Ltd, Tianjin, China) and a portable frequency domain reflector probe (MP-406B, Zhongtian Precision Instruments Co. Ltd, Nantong, Jiangsu, China), respectively. Daily precipitation, air pressure, and temperature were monitored by an automatic meteorological station at the research station approximately 100 m from the experimental plots. At harvest, crop grain yields were measured by three replicate plots for each treatment with sampling areas of 0.25 m^2 each for wheat, and 1 m^2 each for maize. Crop grains were harvested and oven dried at 70 $^\circ\text{C}$ for 48 h to measure the crop grain yields.

2.4. Data analysis and statistical analysis

The DOC loss flux for each rainfall event was calculated as follows:

$$Q_i = C_i \times q_i / 100$$

where Q_i is the DOC loss flux (kg ha^{-1}), C_i is the runoff water DOC concentration (mg L^{-1}) and q_i is the runoff discharge (mm).

The annual cumulative DOC loss flux was calculated as:

$$Q = \sum_{i=1}^n Q_i$$

where Q indicates the annual cumulative DOC loss flux (kg ha^{-1}), $i = 1-n$

(n is the number of runoff events in a year).

Soil DOC content was calculated as following:

$$w = \frac{\rho V(1+x)}{m}$$

where w is the soil DOC content (mg C kg^{-1}), ρ is the soil DOC concentration (mg C L^{-1}), V is the volume of the extract liquid, x is the soil absolute water content (%), and m is the soil mass (g).

All statistical analyses were performed using SPSS (version 22.0; IBM, Inc., USA). One-way analysis of variance was used to test the effects of the four treatments on soil DOC, soil NO_3^- -N, soil NH_4^+ -N and hydrological DOC fluxes, followed by the least significant difference test (LSD, $p < 0.05$). Regression analysis was performed to explore the relationships between rainfall amount and intensity with DOC concentrations and DOC fluxes of different hydrological pathways. Moreover, we employed Origin 2021 (Student version, Origin Lab Corporation, USA) for figure preparation.

3. Results

3.1. Climate condition, environmental variables, and crop productivity

During the three-year experiment, annual precipitation varied from 629 mm to 887 mm, with approximately 77% of the annual precipitation occurring from May to October (Fig. 2). Daily air temperature ranged from -1.5 $^\circ\text{C}$ to 31.7 $^\circ\text{C}$, with annual mean air temperature of 16.8 $^\circ\text{C}$.

The soil water content ranged from 5.15% to 25.54% (mean: 16.91%) for CK treatment, 3.31%–32.13% (mean: 18.23%) for NPK treatment, 5.53%–38.26% (mean: 20.01%) for RSDNPK treatment, and 5.37%–29.80% (mean: 19.60%) for BCNPK treatment (Fig. 3).

Soil DOC contents were in range of 24.89 mg C kg^{-1} to 107.26 mg C kg^{-1} (mean: 61.23 mg C kg^{-1}) for CK treatment, 45.73 mg C kg^{-1} to 123.91 mg C kg^{-1} (mean: 82.96 mg C kg^{-1}) for NPK treatment, 55.58 mg C kg^{-1} to 152.33 mg C kg^{-1} (mean: 103.21 mg C kg^{-1}) for RSDNPK treatment, and 46.39 mg C kg^{-1} to 125.88 mg C kg^{-1} (mean: 81.24 mg C kg^{-1}) for BCNPK treatment (Fig. 4). Soil DOC content, on average, was significantly greater for RSDNPK treatment than those for the control and other two treatments. Soil NO_3^- -N contents ranged from 0.01 mg N kg^{-1} to 14.98 mg N kg^{-1} (mean: 7.28 mg N kg^{-1}) for CK treatment, 0.01 mg N kg^{-1} to 22.57 mg N kg^{-1} (mean: 10.87 mg N kg^{-1}) for NPK treatment, 0.01 mg N kg^{-1} to 32.17 mg N kg^{-1} (mean: 14.23 mg N kg^{-1}) for RSDNPK treatment and 0.01 mg N kg^{-1} to 26.08 mg N kg^{-1} (mean:

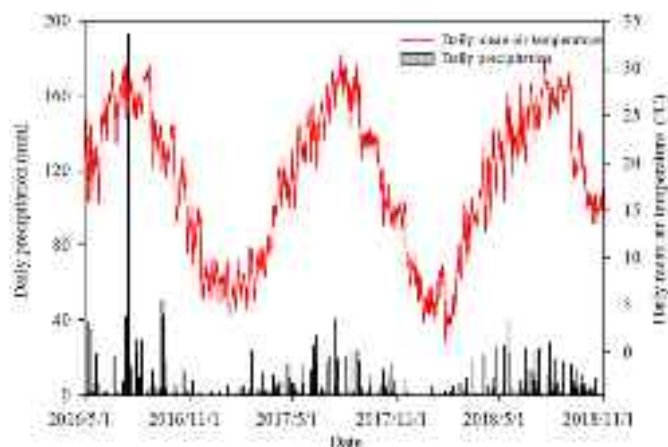


Fig. 2. Daily average precipitation and air temperature during the experimental period of May 2016 to October 2018.

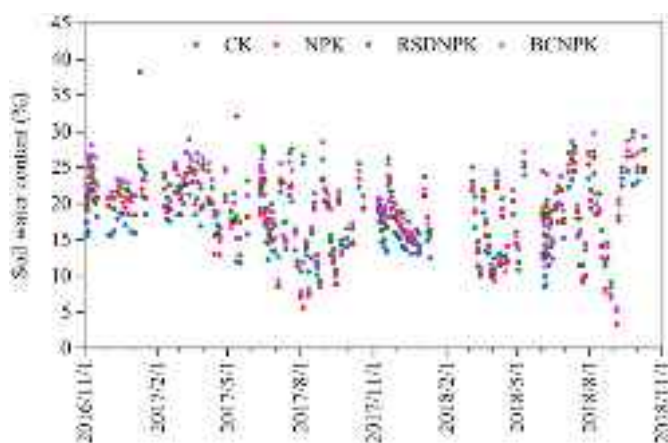


Fig. 3. Dynamic of soil gravimetric water content for all treatments during the experimental period.

10.59 mg N kg⁻¹) for BCNPK treatment (Fig. 4). Soil NH₄⁺-N contents ranged from 0.01 mg N kg⁻¹ to 2.85 mg N kg⁻¹ (mean: 1.11 mg N kg⁻¹) for CK treatment, 0.01 mg N kg⁻¹ to 3.67 mg N kg⁻¹ (mean: 1.63 mg N kg⁻¹) for NPK treatment, 0.01 mg N kg⁻¹ to 3.89 mg N kg⁻¹ (mean: 1.70 mg N kg⁻¹) for RSDNPK treatment and 0.01 mg N kg⁻¹ to 3.54 mg N kg⁻¹ (mean: 1.61 mg N kg⁻¹) for BCNPK treatment (Fig. 4).

Grain yields ranged from 0.27 to 1.22 Mg ha⁻¹ (mean: 0.66 Mg ha⁻¹)

for CK treatment, 3.03–5.54 Mg ha⁻¹ (mean: 4.16 Mg ha⁻¹) for NPK treatment, 3.50–6.32 Mg ha⁻¹ (mean: 5.23 Mg ha⁻¹) for RSDNPK treatment and 2.96–5.30 Mg ha⁻¹ (mean: 4.09 Mg ha⁻¹) for BCNPK treatment (Fig. 5). On average, the application of N fertilizer significantly increased crop yields as compared with the control, while there were no significant differences in crop yield among the three fertilization treatments.

3.2. Discharges of overland flow and interflow

During the three-year experiment, annual cumulative discharges of overland flow ranged from 29.78 to 170.78 mm (mean: 87.72 mm) for CK treatment, 14.20–52.88 mm (mean: 28.23 mm) for NPK treatment, 15.90–39.30 mm (mean: 23.70 mm) for RSDNPK treatment and 14.29–91.09 mm (mean: 31.06 mm) for BCNPK treatment (Table 1). The fertilization treatments significantly decreased overland flow discharges compared to the control.

Annual cumulative discharges of interflow ranged from 50.55 to 136.74 mm (mean: 83.08 mm) for CK treatment, 31.26–160.59 mm (mean: 87.02 mm) for NPK treatment, 26.26–174.14 mm (mean: 83.84 mm) for RSDNPK treatment and 23.29–124.37 mm (mean: 78.71 mm) for BCNPK treatment (Table 1). There were no significant differences in the annual cumulative discharges of interflow across the experimental treatments.

3.3. Hydrological DOC loss pathways and fluxes

During the three-year experiment, annual mean DOC concentrations of overland flow ranged from 1.74 to 3.51 mg C L⁻¹ (mean: 2.57 mg C L⁻¹) for CK treatment, 2.04–3.81 mg C L⁻¹ (mean: 2.90 mg C L⁻¹) for NPK treatment, 1.93–3.41 mg C L⁻¹ (mean: 2.67 mg C L⁻¹) for RSDNPK treatment and 1.92–3.52 mg C L⁻¹ (mean: 2.68 mg C L⁻¹) for BCNPK treatment (Fig. 6). There were no significant differences in the annual mean DOC concentrations of overland flow among the different treatments. Annual cumulative DOC fluxes of overland flow ranged from 0.77 to 4.26 kg C ha⁻¹ (mean: 2.12 kg C ha⁻¹) for CK treatment, 0.46–1.56 kg C ha⁻¹ (mean: 0.83 kg C ha⁻¹) for NPK treatment, 0.40–1.31 kg C ha⁻¹ (mean: 0.73 kg C ha⁻¹) for RSDNPK treatment and 0.44–2.73 kg C ha⁻¹ (mean: 1.20 kg C ha⁻¹) for BCNPK treatment (Fig. 6). Although there were substantial intra- and inter-annual variations in DOC loss via overland flow, the fertilization treatments significantly decreased the DOC fluxes of overland flow compared to the control.

Annual mean DOC concentrations of interflow ranged from 1.72 to 2.49 mg C L⁻¹ (mean: 2.19 mg C L⁻¹) for CK treatment, 2.52–3.77 mg C L⁻¹ (mean: 3.24 mg C L⁻¹) for NPK treatment, 2.95–4.24 mg C L⁻¹ (mean: 3.55 mg C L⁻¹) for RSDNPK treatment and 2.15–3.50 mg C L⁻¹

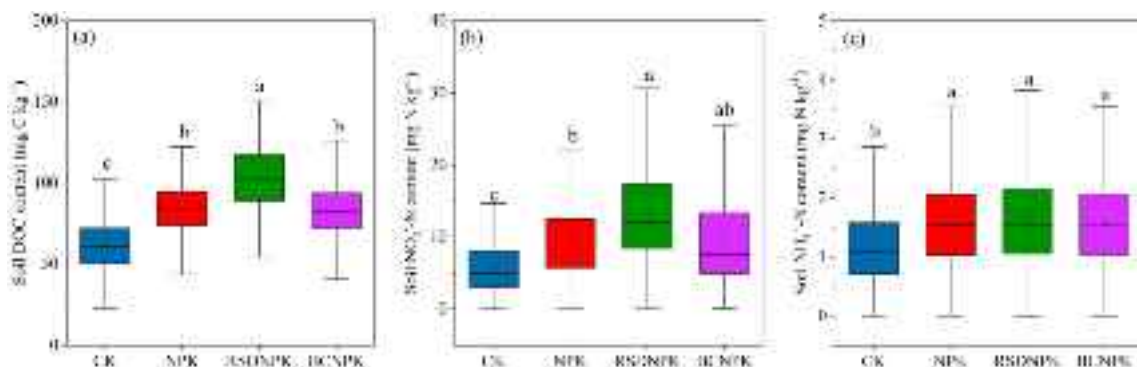


Fig. 4. The boxplot of soil DOC content (a), soil NO₃⁻-N content (b) and soil NH₄⁺-N content (c) among different treatments during the experimental period. Different lowercase letters indicate significant differences among treatments (*P* < 0.05). The hollow square in the box plot is the average value and the solid line is the median value. The upper and lower boundary of the box indicates the 75th and 25th percentile. The error bars above and below the box indicate the maximum and minimum values, respectively.

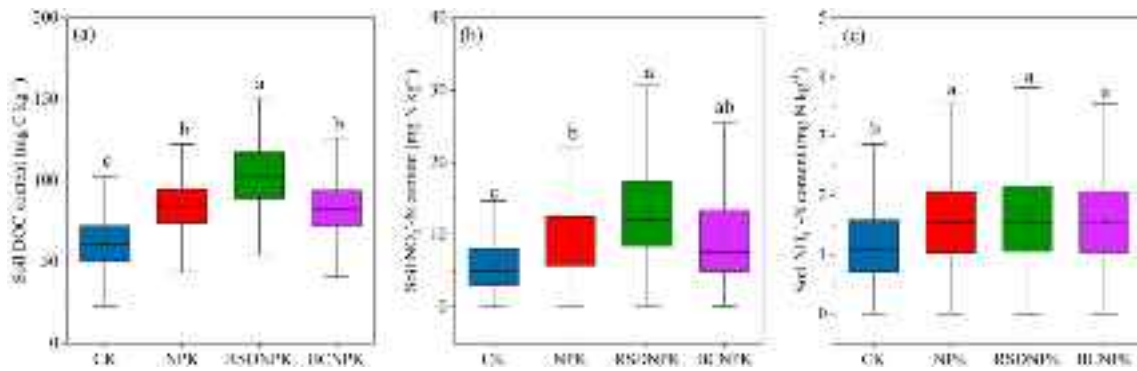


Fig. 5. The grain yields in 2016–2018 among different treatments. Different lowercase letters indicate significant differences among treatments ($P < 0.05$).

Table 1

Annual accumulative discharge, DOC concentration and DOC flux through overland flow and interflow for four treatments during the experimental year. Different lowercase letters and capital letters indicate significant differences among treatments and study years ($p < 0.05$), respectively. Values are means \pm standard error ($n = 3$).

Year	Treatment	Discharge(mm)		DOC concentration (mg C L ⁻¹)		DOC flux (kg C ha ⁻¹)	
		Overland flow	Interflow	Overland flow	Interflow	Overland flow	Interflow
2016	CK	170.78 \pm 16.66 aA	136.74 \pm 12.00 aA	1.74 \pm 0.12 aB	2.37 \pm 0.21 cA	4.26 \pm 0.21 aA	3.29 \pm 1.03bA
	NPK	52.88 \pm 5.91 cA	160.59 \pm 7.29 aA	2.04 \pm 0.23 aB	3.77 \pm 0.20abA	1.56 \pm 0.01 cA	6.25 \pm 0.34abA
	RSDNPK	39.30 \pm 5.71 cA	174.14 \pm 32.44 aA	1.93 \pm 0.16 aC	4.24 \pm 0.12 aA	1.31 \pm 0.32 cA	9.54 \pm 2.09 aA
	BCNPK	91.09 \pm 10.61bA	124.37 \pm 2.90 aA	1.92 \pm 0.17 aB	3.20 \pm 0.35bcA	2.73 \pm 0.60bA	4.64 \pm 0.55bA
2017	CK	33.47 \pm 8.62 aB	50.55 \pm 2.11 aB	3.51 \pm 0.44 aA	2.49 \pm 0.19 aA	0.77 \pm 0.09 aB	1.47 \pm 0.15 aB
	NPK	14.20 \pm 1.74bB	31.26 \pm 2.71abB	3.81 \pm 0.56 aA	3.43 \pm 0.50aAB	0.47 \pm 0.07bB	1.22 \pm 0.16 aB
	RSDNPK	15.90 \pm 3.53bB	26.26 \pm 14.34abB	3.41 \pm 0.15 aA	3.45 \pm 0.00 aB	0.49 \pm 0.08bB	1.15 \pm 0.61 aB
	BCNPK	14.29 \pm 0.47bB	23.29 \pm 2.64bB	3.52 \pm 0.29 aA	3.50 \pm 0.60 aA	0.45 \pm 0.03bB	0.85 \pm 0.07 aB
2018	CK	29.78 \pm 2.52 aB	61.95 \pm 4.51 aB	2.46 \pm 0.22 aB	1.72 \pm 0.64 aA	1.33 \pm 0.60 aB	1.71 \pm 0.58 aB
	NPK	17.62 \pm 1.33 aB	69.21 \pm 3.56 aB	2.83 \pm 0.18aAB	2.52 \pm 0.21 aB	0.46 \pm 0.01 aB	2.96 \pm 0.41 aB
	RSDNPK	15.90 \pm 1.97 aB	51.12 \pm 14.62 aB	2.68 \pm 0.16 aB	2.95 \pm 0.22 aB	0.40 \pm 0.04 aB	2.75 \pm 0.48 aB
	BCNPK	17.80 \pm 0.53 aB	58.47 \pm 4.37 aB	2.58 \pm 0.14 aB	2.15 \pm 0.43 aA	0.44 \pm 0.04 aB	2.31 \pm 0.53 aB
Mean (2016–2018)	CK	87.72 \pm 17.49a	83.08 \pm 5.24a	2.57 \pm 0.24a	2.19 \pm 0.31b	2.12 \pm 0.21a	2.16 \pm 0.55b
	NPK	28.23 \pm 2.63b	87.02 \pm 2.14a	2.90 \pm 0.17a	3.24 \pm 0.29 ab	0.83 \pm 0.03b	3.48 \pm 0.11 ab
	RSDNPK	23.70 \pm 3.68b	83.84 \pm 19.98a	2.67 \pm 0.07a	3.55 \pm 0.11a	0.73 \pm 0.14b	4.48 \pm 1.06a
	BCNPK	31.06 \pm 3.81b	78.71 \pm 2.73a	2.68 \pm 0.07a	2.95 \pm 0.45 ab	1.20 \pm 0.20b	2.60 \pm 0.38 ab

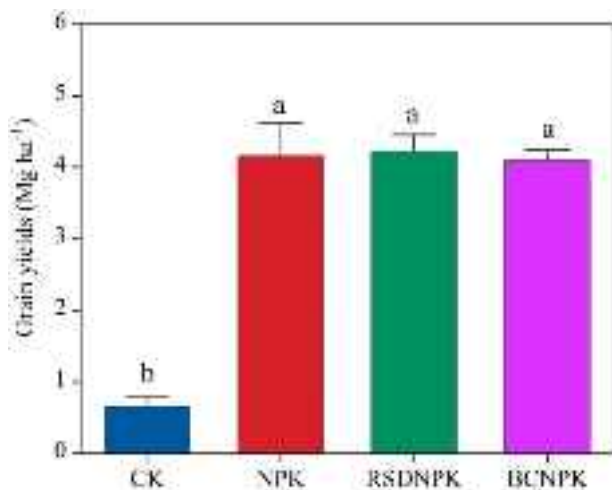


Fig. 6. Seasonal variations in runoff discharge (a), DOC concentration (b) and DOC flux (c) via overland flow during the experimental period. Values are means \pm standard error ($n = 3$).

(mean: 2.95 mg C L⁻¹) for BCNPK treatment (Fig. 7). The corresponding annual cumulative DOC fluxes of interflow ranged from 1.47 to 3.29 kg C ha⁻¹ (mean: 2.16 kg C ha⁻¹) for CK treatment, 1.22–6.25 kg C ha⁻¹ (mean: 3.48 kg C ha⁻¹) for NPK treatment, 1.15–9.54 kg C ha⁻¹ (mean: 4.48 kg C ha⁻¹) for RSDNPK treatment and 0.85–4.64 kg C ha⁻¹ (mean:

2.60 kg C ha⁻¹) for BCNPK treatment (Fig. 7). The annual cumulative DOC fluxes of interflow for the RSDNPK treatment on average were significantly higher than those for the control and NPK treatments.

The DOC concentrations of the overland flow and interflow were positively correlated with the rainfall amount and rainfall intensity (Fig. 8). Similarly, DOC loss fluxes through either overland flow or interflow exhibited positive relationships with rainfall amount and rainfall intensity (Fig. 9).

4. Discussion

4.1. Hydrological fluxes of DOC loss

Hydrological DOC loss in terrestrial ecosystems is an interaction between biogeochemical carbon processes and hydrological processes (van Verseveld et al., 2009). In this study, the annual DOC loss fluxes ranged from 0.40 to 4.26 kg C ha⁻¹ yr⁻¹ (mean: 1.22 kg C ha⁻¹ yr⁻¹) through overland flow and 0.85–9.54 kg C ha⁻¹ yr⁻¹ (mean: 3.18 kg C ha⁻¹ yr⁻¹) through interflow during the three-year experiment (Table 1). The annual hydrological DOC fluxes of either interflow or overland flow in our current study almost all fell in the range of 0.45–27.50 kg C ha⁻¹ yr⁻¹ obtained in different agricultural landscapes, e.g., managed montane grassland (Fu et al., 2019), upland cropland (Hua et al., 2014) as well as rice paddy field (He et al., 2017).

Nevertheless, we found that there were significant intra-annual and inter-annual variations in DOC losses through hydrological paths of both overland flow and interflow (Table 1, Figs. 6–7). It should be noted that the hydrological DOC loss events were concentrated during the maize

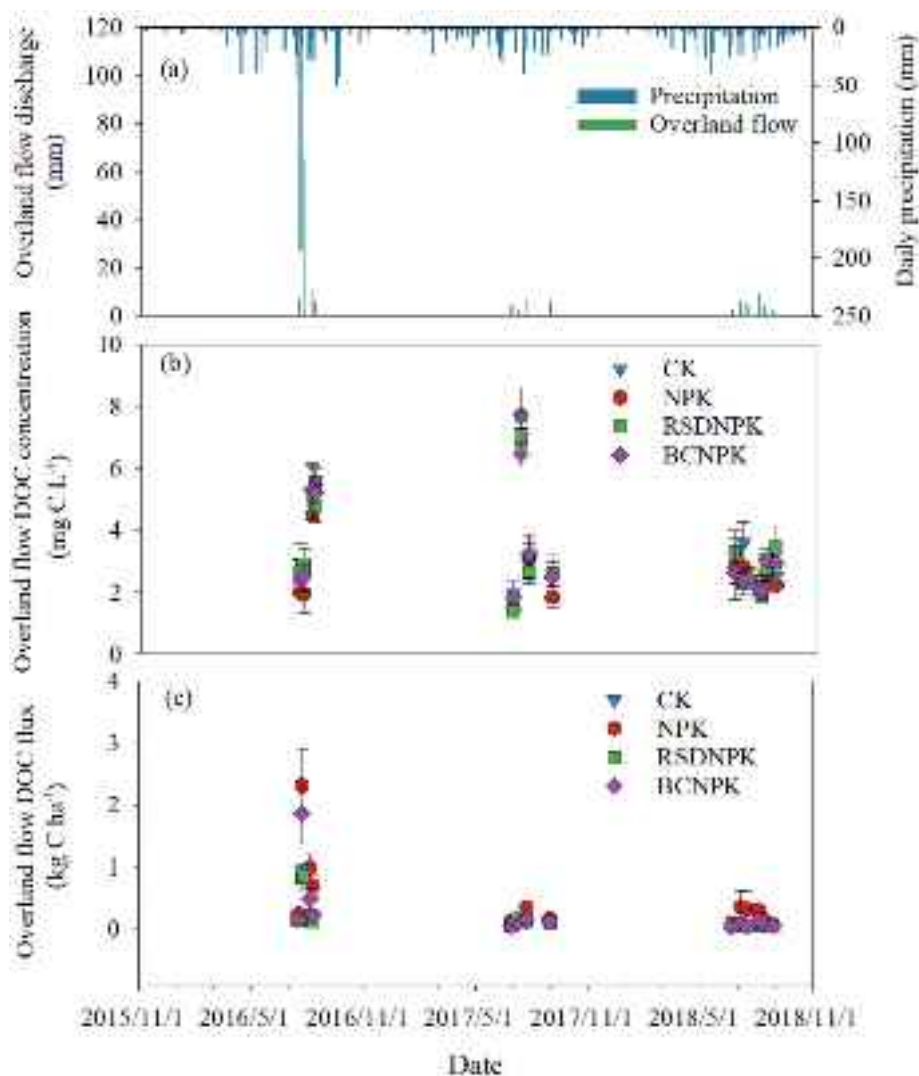


Fig. 7. Seasonal variations in runoff discharge (a), DOC concentration (b) and DOC flux (c) via interflow during the experimental period. Values are means \pm standard error ($n = 3$).

growing season (i.e., rainy season, May to September), while no DOC loss events occurred in the dry wheat growing season (Figs. 6–7). Thus, one possible reason for these variations is the great temporal dynamics of rainfall patterns (e.g., rainfall amount and rainfall intensity). Rainfall is the main driver of runoff and may directly impact hydrological DOC fluxes from soils from ecosystem to watershed scales (Wang et al., 2019). This explanation is further supported by the significant correlations between the rainfall amount and intensity and the DOC concentrations, as well as the significantly positive correlations between rainfall amount and intensity and DOC fluxes for both overland flow and interflow (Figs. 8–9). These results suggest that rainfall patterns are likely the main controller of DOC loss from a hydrological perspective, which is in line with previous studies that found rainfall patterns control not only the magnitude but also the temporal pattern of hydrological DOC losses in various ecosystems (Bah et al., 2020; Ma et al., 2018; Smemo et al., 2007; Tian et al., 2012; Williams et al., 2017). For instance, Fei et al. (2019) demonstrated that rainfall could easily detach DOC adsorbed on topsoil, thereby inducing greater DOC leaching losses via subsurface runoff. Froberg et al. (2006) also illustrated that rainfall could enhance soil microbial activity, soil organic carbon decomposition, and soil DOC availability, thereby increasing the potential for hydrological DOC loss.

On average, the average annual DOC loss in overland flow ($1.22 \text{ kg ha}^{-1} \text{ yr}^{-1}$) accounted for approximately 27% of the total hydrological

DOC loss, while DOC loss in interflow ($3.18 \text{ kg ha}^{-1} \text{ yr}^{-1}$) accounted for 73% of the total hydrological DOC loss (Table 1). Similar to the present study, a three-year field study also showed that annual DOC fluxes through interflow were over four times greater than for those through the path of overland flow in the same study region (Hua et al., 2014). Previous studies have also shown that overland flow accounts for only a small part of the total runoff in this area (Wang and Zhu, 2011), while interflow is the major hydrological path in the study region (Zhu et al., 2009). The soil depth is relatively shallow (i.e., less than 80 cm), and thus soil can be easily saturated even when daily precipitation is over 20 mm (Zhou et al., 2012; Zhou et al., 2016). Thus, the percolated water can quickly reach the soil-bedrock interface and formulate lateral flow, thereby resulting in a high discharge of interflow relative to overland flow (Hua and Zhu, 2018). This is a possible explanation for the greater DOC fluxes through interflow because a larger discharge of runoff usually drives greater hydrological DOC loss (Ma et al., 2014). Nevertheless, hydrological DOC loss is controlled not only by hydrological processes but also by biogeochemical processes. Further comprehensive studies with careful consideration of interactions between hydrological and biogeochemical processes are urgently needed.

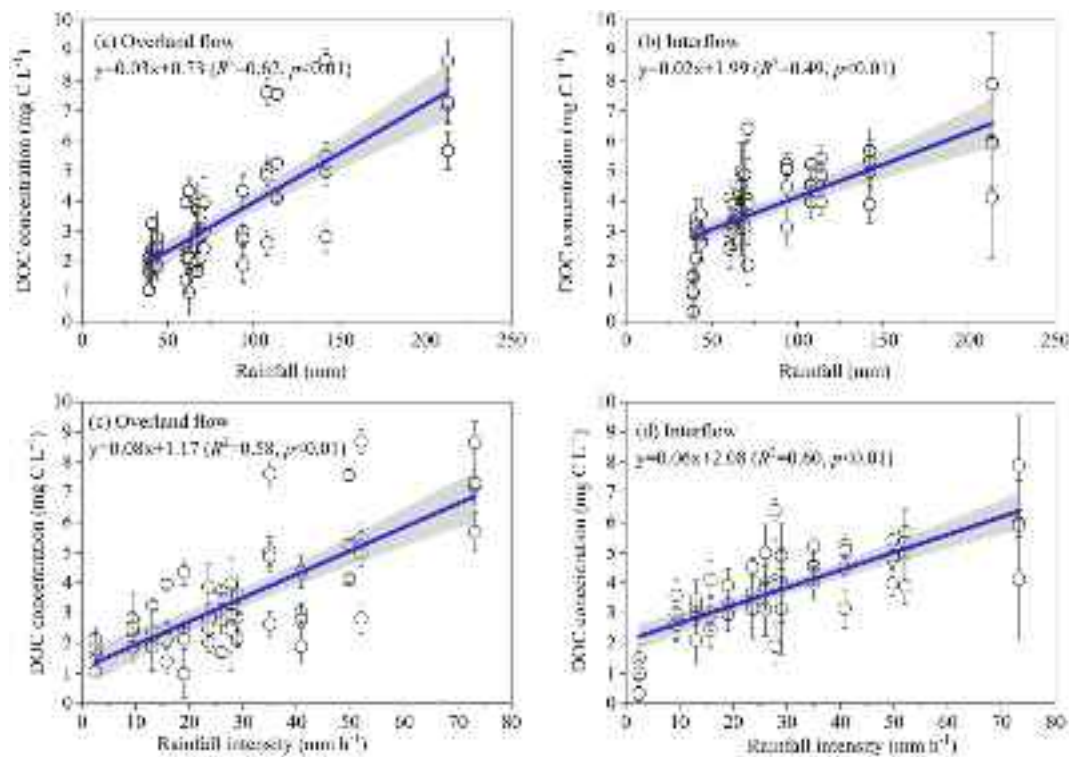


Fig. 8. Correlations between rainfall amount with DOC concentration (a–b), and rainfall intensity with DOC concentration (c–d), via overland flow and interflow during the experimental period. The shaded area represents 95% confidence interval. Values are means ± standard error (n = 3).

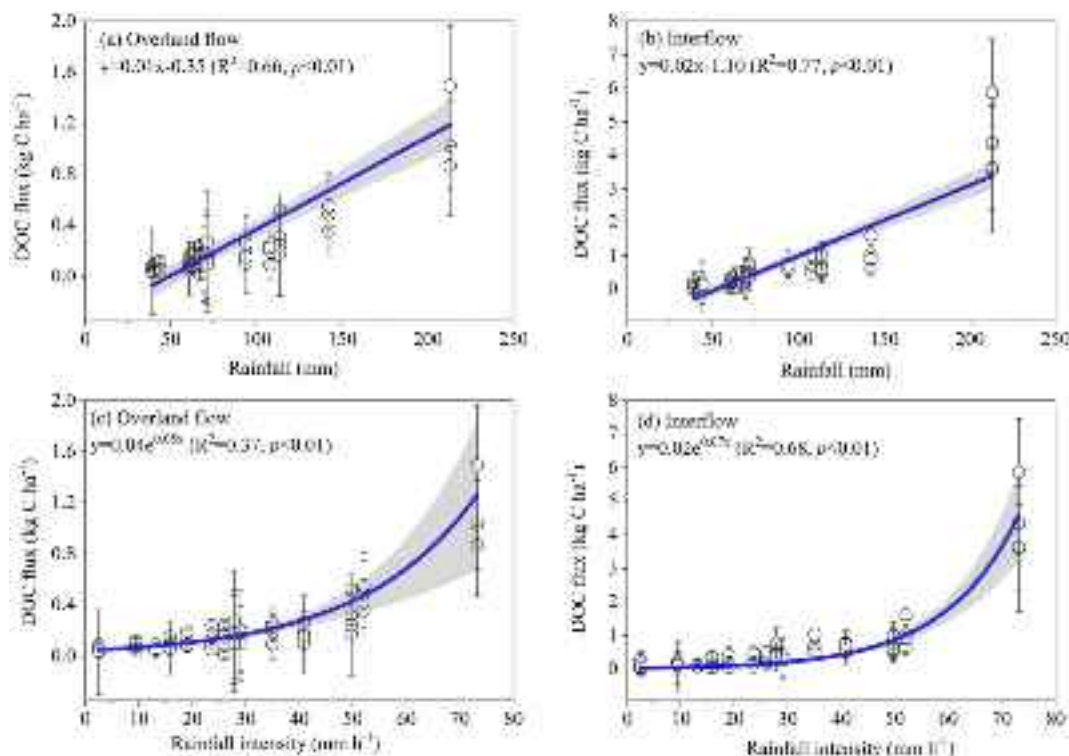


Fig. 9. Correlations between rainfall amount with DOC flux (a–b), and rainfall intensity with DOC flux (c–d), via overland flow and interflow during the experimental year. The shaded area represents 95% confidence interval. Values are means ± standard error (n = 3).

4.2. Effects of N fertilizer application on hydrological DOC loss

Given that addition of N has been identified as a key regulator of

hydrological DOC loss in terrestrial ecosystems, the direction and magnitude of N fertilization effects on hydrological DOC losses in agricultural soils are uncertain. In the present study, N fertilizer application

significantly decreased DOC fluxes of overland flow while increasing DOC fluxes of interflow, that is, fluxes of DOC leaching (Table 1). The decrease in DOC fluxes of overland flow could be well explained by the lower discharge of overland flow for the control relative to N fertilization treatments ($P < 0.05$, Table 1), which is in line with the findings of previous studies. With respect to the increases in DOC loss of interflow for N fertilization treatments observed in our current study, previous studies also found that the application of N fertilizer could enhance soil DOC leaching fluxes in arable land related to the increased plant productivity. Similarly, in the present study, the application of N fertilizer significantly increased crop grain yields (Fig. 5), which in turn could stimulate fresh plant-derived DOC production, thereby increasing DOC fluxes of interflow relative to the control. In addition, the application of N fertilizer could increase bioavailable N for soil microbes (Fig. 4), which could consequently enhance the decomposition of soil organic matter and DOC production in soil (Liu et al., 2014), thereby increasing the potential of hydrological DOC loss in interflow.

4.3. Effects of biochar amendment on hydrological DOC loss

Amendment of biochar practices can modify soil physical, chemical, biological, and mechanical properties (Mukherjee et al., 2014; Sohi et al., 2010). This is one of the few studies that have assessed the long-term impact of biochar on DOC under field conditions (Lu et al., 2014; Zhang et al., 2017). Biochar contains some labile fractions of organic C (Cross and Sohi, 2011; Singh and Cowie, 2014; Smith et al., 2010), which can increase DOC concentrations after amendment. Zhang et al. (2017) found that applications of 4 t ha^{-1} to 8 t ha^{-1} of biochar could increase DOC concentrations from 84 mg kg^{-1} to 144 mg kg^{-1} in 2 years. Smebye et al. (2016) reported that the DOC concentration was 15 times greater in a biochar-amended soil than in control (non-amended) soil 48 h after biochar application. In contrast, biochar may reduce DOC concentration by sorption (Lu et al., 2014). DOC is an active fraction of SOC; hence, a one-time measurement cannot reflect the real impact of biochar on this soil property. We analyzed soil DOC concentrations and fluxes over three years (2016–2018) in this study (Table 1) and found that biochar did not significantly influence DOC loss via overland flow or interflow. Similar results have been reported previously for silt loam soil (Dong et al., 2019) and sandy loam soil (Nelissen et al., 2015). There are four possible explanations for this phenomenon. First, except for the inherent soil carbon, any additional carbon was determined by the amount of labile biochar carbon, as previously reported for by Wardle et al. (2008). Cross and Sohi (2011) showed a decrease in labile C content with increasing biochar pyrolysis temperature. In the carbonization process of biomass, small molecules and soluble organic matter enter the smoke and steam with water, leaving behind the complex stable structure of the carbonized matter (Xie et al., 2015). Therefore, biochar decomposition is very slow due to its very high stability, and the decomposition rate decreases with time (Kuzyakov et al., 2014); even microbially-utilized biochar compounds have a much slower turnover within microorganisms compared to other C sources (Siedt et al., 2021), indicating that biochar addition inputs almost no fresh or labile carbon to the soil. Second, the DOC turnover rate can be high, as DOC is in the active soil C pool (Kalbitz et al., 2000), while biochar sorption capacity may be limited. Moreover, sorption and desorption may reach a balanced state within a few weeks to months (Chen et al., 2011; Jiang et al., 2012); therefore, biochar would not decrease DOC by sorption. Third, because of the lack of impact of biochar addition or its application rates on aboveground biomass yields, belowground C input (Fang et al., 2016; Weng et al., 2017) and consequently soil DOC content may remain unaltered by biochar (Fig. 4). Finally, the priming effect of biochar on SOC mineralization may decrease over time due to the depletion of labile SOC or stabilization of labile organic matter by biochar-induced organo-mineral interactions (Singh and Cowie, 2014; Weng et al., 2017), thus presenting the non-significant effects of biochar amendment on soil DOC availability and hydrological DOC fluxes relative to the

practices of synthetic N fertilizer application only.

4.4. Effects of crop straw incorporation on hydrological DOC loss

The hydrological DOC loss of overland flow was significantly decreased by the incorporation of crop straw (RSDNPK treatment) relative to other experimental treatments in the present study (Table 1). This finding agreed with a previous study that practices of crop straw amendment significantly decreased the annual hydrological DOC loss of overland flow by approximately three times as compared with the control (Hua and Zhu, 2018). Moreover, amendments of crop straw can facilitate the formation of fungal hyphae and root exudates and increase the cementation of microaggregates into macroaggregates (Zhang et al., 2017), which may improve soil structure and increase water infiltration, thereby decreasing DOC loss through overland flow. Indeed, in the present study, the mass percentages of $0.25\text{--}2 \text{ mm}$ aggregates were decreased by incorporation of crop straw ($P < 0.05$, Fig. S1), which could somewhat explain the decrease in the hydrological DOC fluxes of overland flow. Similarly, some studies found that straw mulch cover resulted in a decrease in the proportion of surface runoff and an increase in the proportion of infiltration, in contrast to biochar addition (e.g., Li et al., 2019). This may be because crop mulch directly protects the soil surface from raindrop impact, and consequently improves water infiltration by preventing crust formation and enhancing interflow runoff (Rahma et al., 2017).

In contrast with hydrological DOC fluxes of overland flow, the incorporation of crop straw increased hydrological DOC fluxes of interflow throughout the three-year experiment (Table 1). Similarly, Johnston et al. (2009) also found that soil DOC content significantly increased, thereby accelerating DOC leaching losses, following a 17-year continuous straw incorporation in the cropland of England. These findings are likely related to the stimulation of organic matter decomposition for DOC production in soil and an increase in soil water infiltration capacity by the incorporated crop straw, which consequently exacerbated DOC leaching loss via interflow. Moreover, our findings also agreed with previous reports that the incorporation of crop straw increased DOC leaching loss relative to the amendment of biochar in rice paddy fields (Liu et al., 2021). Compared to biochar, crop straw can be more easily decomposed by soil microorganisms, thereby increasing the availability of DOC following the incorporation of crop straw in soils (Kubar et al., 2020; Yang et al., 2017). Moreover, the relatively higher soil water content would also explain the greater DOC fluxes of interflow for crop straw incorporation (Fig. 3). Previous studies have demonstrated that higher soil moisture conditions can enhance soil microbial activity (Hueso et al., 2012) and increase labile and active organic C pools (e.g., DOC) in soils (Liu et al., 2014; Ponizovsky et al., 2006), thereby enhancing DOC loss via hydrological pathways of interflow.

5. Conclusions

The present study found that interflow was the main pathway of hydrological DOC loss relative to overland flow in subtropical montane agricultural landscapes. The rainfall amount and intensity were the regulators of hydrological DOC loss, and their intra- and inter-annual variations were mainly attributed to the temporal dynamics of precipitation. Since hydrological DOC loss is not only controlled by hydrological processes but also by biogeochemical processes in various terrestrial ecosystems worldwide, further research is needed to illustrate the biogeochemical DOC process and the underlying mechanisms. Nevertheless, compared to application of synthetic N fertilizer only, incorporation of crop straw significantly increased total hydrological DOC fluxes of interflow and overland flow while no significant effects were detected for amendment of biochar in subtropical calcareous agricultural soils. Overall, stimulation of hydrological DOC losses by crop straw incorporation strategy may offset the benefits of increasing SOC stocks

in the subtropical montane agricultural landscapes, suggesting that hydrological DOC loss should be carefully considered to evaluate the potential of C sequestration for a given management strategy in agricultural landscapes.

Credit author statement

Nan Jiang: Formal analysis, Methodology, Writing – original draft. Hamidou Bah: Investigation, Data curation. Minghua Zhou: Conceptualization, Funding acquisition, Writing – review & editing. Peng Xu: Validation, Writing – review & editing. Bowen Zhang: Validation. Bo Zhu: Supervision

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2021.118751>.

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