Characteristics of wet carbon deposition in a semi-arid catchment at Loess Plateau, China

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10   **Abstract.** Wet carbon deposition is a critical node of the global carbon cycle, but little is known about dissolved organic and
inorganic carbon (DOC and DIC) variation and flux in semi arid area of the Loess Plateau Region (LPR). In this study,
concentration of DOC and DIC in rainfall was monitored in the period of July to September 2015 at Yangjuangou catchment
in the LPR. Results showed that the Volume-Weighted Mean (VWM) concentration of DOC and DIC were 24.62 and 4.30
(July), 3.58 and 10.52 (August), 1.01 and 5.89 (September) mg C L^{-1}. VWM concentrations of DOC and DIC for the
15   concentrated rainy season (July - September) in the studied region were 7.06 and 7.00 mg C L^{-1}, respectively. In addition, the
monthly deposition flux of DOC and DIC were 541.64/94.60, 131.03/385.03, and 44.44/259.16 mg C m^{-2} for July, August
and September. The estimated annual wet carbon depositions were 1.91, 1.89 g C m^{-2} yr^{-1} for DOC and DIC, which were
higher than those of other sites and lower than those in the tropical and sub-tropical sites. Furthermore, the loess dust
deposition process provides soil parental material in soil formation process and might be another source of carbon at the LPR.
20   Therefore, the given results reflect characteristics of wet carbon deposition process during concentrated rainfall season in a
semi-arid catchment of the LPR. Our preliminary results suggest that further investigation is needed on carbon source and
deposition flux from atmosphere at long term temporal and large scale for revealing the global carbon cycle.

1 Introduction

It has been known that carbon stored in atmosphere is an important component of the global carbon pool (Raymond,
25   2005; Willey et al., 2000). Carbon wet deposition from atmosphere is one of the key driving forces of carbon and
biogeochemical cycle, which represents massive exchanges of physical processes between terrestrial and atmospheric
environment. Dissolved organic and inorganic carbon (DOC and DIC) is a major component of rainwater in many regions

around the world (Dachs et al., 2005). Consequently, precipitation is known to be a significant source of DOC and DIC in global carbon cycle. Concentration of DOC in precipitation in marine (0.28 mg C L^{-1}) and continental (1.93 mg C L^{-1}) is

30   greater than nitric and sulfuric acid combined. There, the global flux of DOC and DIC via precipitation can be estimated at 4.3×10^{14} and 0.8×10^{14} g C yr^{-1} (Willey et al., 2000). It is equivalent to 9% of the excess carbon released by fossil combustion and cement production (Schimel et al., 1996). Consequently, the huge magnitude of carbon flux via precipitation drives relevant studies on wet carbon deposition as one of the key driving forces of global cycle. There is thus urgency to improve associated knowledge and understanding of atmospheric carbon wet deposition.

35   For relevant work in comparisons of various criteria on nitrogen and phosphorus of atmospheric deposition, only few quantitative studies are available on the atmospheric wet carbon deposition. Similarly, studies conducted in exploring atmospheric deposition can be rarely found in the target research area, the Loess Plateau Region (LPR). In addition, carbon exchange between atmosphere and land has not been incorporated into current regional or global carbon cycle models (Jurado et al., 2008; Kieber et al., 2002). Thus, the lack of measurement and quantitative knowledge, such as corresponding sources,

40   chemical composition and variation pattern, has been shown the need for substantial information. Around the world, only few quantitative studies can be found on carbon wet deposition, such as in Lithuania (Armalis, 1999), Poland (Siudek et al., 2015), USA (Iavorivska et al., 2017), New Zealand (Kieber et al., 2002) and Brazil (Coelho et al., 2008), which DOC flux in rainfall ranged 1.2 to 5.4 g C m^{-2} yr^{-1}. Not until recently, the only measurement data available were those related to wet deposition in northern and Tibetan region of China (Li et al., 2016; Pan et al., 2010). Pan et al. (2010) reported that significant seasonal

45   differences in DOC concentrations and deposition fluxes can be found in northern China. The corresponding annual average concentration and deposition flux of DOC from atmosphere ranged from 2.4-3.9 mg C L^{-1} and 1.4-2.7 g C m^{-2} yr^{-1}. Li et al. (2016) also reported that the DOC concentration in seasonal precipitation varied between monsoon and non-monsoon period and the average deposition of DOC was 1.1 mg C L^{-1} that almost 1/3 was contributed by fossil fuel combustion. The annual deposition flux of DOC was about 0.63 g C m^{-2} yr^{-1} in Tibetan Plateau of China. These results indicated that a considerable

50   spatial and temporal variation in wet carbon concentration and deposition flux between different regions. The differences were attributed to precipitation, air mass origin and also related to the source of carbon.

Atmospheric wet carbon deposition may occur by precipitation scavenging of aerosol-bound organic compounds, which was originated from biogenic (vegetation, biomass burning, etc) and anthropogenic sources (fuel combustion, industrial emission, etc) and carbon dioxide (Duarte et al., 2006; Houghton, 2003; May et al., 2013). Furthermore, it has been

55   demonstrated that emissions of incomplete fossil fuel combustion were a key source of DOC in precipitation, such as in northern and Tibetan of China (Li et al., 2016; Pan et al., 2010) , USA (Mladenov et al., 2012; Raymond, 2005), Europe (Siudek et al., 2015) and New Zealand (Willey et al., 2000). For instance, contribution of fossil fuel combustion in DOC was around 22-28% in North America, European, and Tibetan of China. Therefore, spatial-temporal variation patterns and sources of wet carbon deposition contribute to carbon exchange between atmosphere and terrestrial environment. Despite a large

60 extent of wet carbon deposition is characterized at other sites, there are few studies explained the DOC and DIC in rainfall, which is a critical step in understanding carbon cycle in LPR.

Previous studies have provided insights on the magnitude and importance of wet carbon deposition via precipitation worldwide. However, characteristics of DOC and DIC on the LPR have been rarely studied. The LPR (N35-41°, E102-114°), with area of 6.4×10^5 km^2, situated in the middle stream of the Yellow River. The plateau is covered by an average thickness

65 of 100m loess. Loess is formed by the accumulation of wind-blown silt (Ding et al., 2002). Meanwhile, Zhu and Zhu (1990) shown that rain-out dust process refers fine-grained loess being washed down onto the land by precipitation and identified as an source of loess parent material in soil forming process. The fine-grained particle may serve as nuclei to form a rain droplet or cloud condensation. Consequently, it is worth noting that atmospheric dust scavenged by precipitation may be another source of organic carbon, which might differ from other regions. Moreover, Mladenov et al. (2012) used a long term dataset

70 of weekly DOC deposition collected at Colorado Rocky Mountains, USA, to demonstrate that atmospheric carbon deposition represented a significant source to an alpine catchment. Wang et al. (2017) reported that atmospheric wet deposition might be a large source of DOC in stream water based on isotopic characteristics of carbon in Yangjuangou catchment. Thus, it leads to pay attentions on wet deposition process of DOC and DIC in LPR. In addition, the Chinese government initiated "Grain-for-Green" project in 1999, which remarkably increased vegetation coverage and restored ecological environment. Until this

75 point, knowledge of wet carbon deposition and associated process has not been fully explored in the LPR. Thus, it is necessary to investigate the DOC and DIC deposition via precipitation, where little knowledge is available.

In this study, data were collected from monitoring stations of rainfall quality in atmospheric wet deposition at a catchment of LPR. The primary goal of this study is to evaluate the magnitude of DOC and DIC flux from atmosphere so as to understand atmospheric wet deposition process during concentrated rainfall season in LPR. Specifically, three objectives

80 were investigated: (1) to examine the relationship of DOC and DIC concentration in rainfall, (2) to clarify the variations in concentrated precipitation season (July to September); and (3) to estimate the wet deposition flux of DOC and DIC in LPR. These results will provide evidence of wet carbon deposition and fill a gap of the regional rainfall carbon cycle at a semi arid catchment in LPR.

2 Materials and Methods

2.1 Description of the sampling site

As shown in Figure 1, the field sampling was performed at Loess Plateau Ecological Restoration and Soil and Water Conservation Monitoring Station located in Yangjuangou catchment (N36° 42', E109° 31') in Yan'an City, Shaanxi Province. This catchment covers 2.02 km^2 in area and characterized by a typical loess hilly and gully topography with a gully density of

2.74 km/km^2 (Wang et al., 2011). As shown in Table 1, the main land use types in this catchment are forest, shrub, grassland,

90   orchard and farmland. The major forest species are *Robinia pseudoacacia*, *Salix spp.* and *Populus spp.* The *Artemisa argyi*,

Stipa Bungeana trin., *Bothriochloa ischaemum*, *Lespedezadavurica schindl.*, and *Artemisia sacrorum* are classified as

grassland. The major orchards are *Prunus armeniaca L., Malus pumila Mill.*,and *Juglans rejia L.* The major crops are *Setaria*

italica, Zea may L. Glycinemax (L) Merr. Panicum miliaceum L. and *Solanum tuberosum* (Fu et al., 2014). The catchment

have a semi arid continental monsoon climate and annual mean minimum and maximum temperatures of 6.5℃ (January) and

95   22.9℃ (July). Mean annual precipitation is 535 mm, whereas the concentrated precipitations occurred in the rainy season

from June to September with large inter-annual fluctuations. A meteorological station is used to monitoring rainfall amount,

air temperature, moisture and wind velocity, etc. The soil in this area is classified as typical loess with a fine silt texture and

weakly resistant to detached by raindrops or runoff. The average erosion rate is 7715.5 t km^{-2} yr^{-1} between 2006 and 2009 (Fu

et al., 2014).

2.2 Collection of rainfall sample

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~~We collected~~ rainfall samples from July to September, 2015. The use of open to the atmosphere collector is a

common approach to collect atmospheric wet deposition ~~during each precipitation event~~. A rain gauge was installed on the

roof of ~~the~~ building in the sampling site for determining precipitation volume and also can be used for collecting rainfall

samples ~~during a precipitation event~~. Another two duplicate rainfall samples were collected at each site, using a steel bucket

105   (d=29 cm). After a precipitation event, the ~~data of~~ samples were transferred into a p⬚ bottle. All of rainfall containers

were clean with deionized water after a collection and return to sampling site for next rainfall sam⬚. All of the samples

were collected according to the precipitation amount. The collected rainfall events in July, August, September is 4, 7, 5,

respectively, whereas the corresponding total rainfall events were 6, 12, 7 in each month.

2.3 In situ and laboratory measurements

110   In this study, there are three steps in conducting in situ and laboratory measurements. First, the TDS (Total

Dissolved Solids), pH of unfiltered rainfall samples were immediately tested using a portable Ultrameter 6PFC (MYRON L.

Company, USA) after each rainfall collection. Then, each sample was filtered by a 0.45 μm membrane filter (Whatman, GE,

USA) to removal particulates and 200 ml filtrates were stored at freezer to prevent biological activities. Third, the frozen

samples were transported to laboratory in the State Key Laboratory of Urban and Regional Ecology (RCEES-CAS) for

115   further analyze. Total dissolved carbon (TDC) and DIC concentration was determined by Vario (Elementar, Germany),

which included a high-temperature combustion furnace, self-contained acidification module and a highly sensitive CO$_2$

detector. Prior to measurement, the instrument should dose 125 ml of 1% H$_3$PO$_4$ solution (phosphoric acid) in acidification

module, and then validation conducted by analyzing various concentrations of a TDC standard solution for achieving

accurate results. TDC can be automatically measured by the combustion of a sample while DIC can be measured after acidification of a sample. The distilled water was also tested at every 50 samples for ensuring results quality. Considering TDC is recognized as the sum of DIC and DOC component, thus the DOC was the difference between TDC and DIC for each sample (DOC=TDC-DIC). Meanwhile, NO_3^- and NH_4^+ were measured by the FUTURA Segmented Continuous Flow Analysis (Alliance, France).

2.4 Data processing

In the present study, the DIC and DOC concentration in rainfall for a month or a rainy season were Volume-Weighted Mean (VWM) concentration, which is commonly used in wet deposition studies for minimizing the average effects of small rainfall amount (Li et al., 2017; Pan et al., 2010; Santos et al., 2011; Santos et al., 2014). The calculation of VWM concentration and wet deposition flux were defined in the following equations:

$$VWM = \left. \sum_{i=1}^{n} Ci \times Pi \middle/ \sum_{i=1}^{n} Pi \right.$$

$$F = VWM \times \sum_{i=1}^{m} Pi$$

where, Pi (mm) is the rainfall amount corresponding to each sample, Ci (mg C L^{-1}) is the DOC and DIC concentration in an individual rainfall sample. n, m are the number of sampled and total rainfall events in a time period; F (mg C m^{-2}) is the wet deposition flux of DOC and DIC in a month or rainy season in the study region. In order to analyze their potential relationships among DIC, DOC, NH_4^+-N, NO_3^--N, TDS, and pH, the Pearson's test (P<0.05) was performed using SPSS(Statistics Package for Social Science) (IBM, 2010). The corresponding figures were developed by using Sigma Plot 10.0 (Systat, 2008).

3 Results and Discussions

3.1 Concentrations and flux of DOC and DIC

~~Concentrations of DOC and DIC were determined in rainfall samples collected from July to September, which is the rainy season (38% of total average annual precipitation).~~ As illustrated in Figure 2-a, concentrations of DOC and DIC were demonstrated in individual rainfall event. Overall, noticeable differences of concentrations can be found between individual rainfall events. Concentrations of DOC exhibited large variations ~~and ranged between~~ 0.56 to 28.71 mg C L^{-1}, whereas the maximum and minimum concentrations were found in July and September. Ranges of DOC concentrations in July, August

and September were 15.14-28.71, 0.73-6.75, and 0.56-1.86 mg C L^{-1}, respectively (with a decreasing trend). In addition, DIC concentrations generally varied from 0.5 to 13.6 mg C L^{-1} from July to September. In each month, DIC concentrations were 3.47-6.86 (July), 6.77-17.49 (August), 4.99-10.35 (September) mg C L^{-1} (with increasing trend). It is worth noting that DOC

145 concentrations exhibited substantial variations with much higher concentrations than DIC in July and then decreased with a lower concentration than DIC in August and September.

For more detailed investigations to examine the differences of concentrations in DOC and DIC and to minimize average effect in a slight rainfall event, VWM concentrations were calculated in each month. VWM concentrations of DOC in July, August and September were 24.62, 3.58, 1.01 mg C L^{-1}, and the corresponding DIC concentrations were 4.30, 10.52,

150 5.89 mg C L^{-1}. Overall, the DOC concentration in July was highest with a significant difference than August and September, whereas the DIC was lower than the other two months. These results indicated that the different variation patterns of DOC and DIC during the rainfall events at rainy season in the LPR. Therefore, rainfall events may be considered as an important driver in translating atmospheric carbon to land.

As shown in Figure 2-b, carbon deposition flux in individual rainfall event and the proportion of DOC and DIC were

155 demonstrated during sampled period. In general, DOC deposition flux ranged from 0.21-258.36 mg C m^{-2}, whereas higher and lower deposition flux can be found in July (15.27-258.36 g C m^{-2}), September (0.46-16.81 mg C m^{-2}) . In contrast to DOC, lower DIC deposition flux was found in July (4.12-42.32 mg C m^{-2}) and then increased at August (2.6-67.66 mg C m^{-2}) and September (3.10-89.81 mg C m^{-2}). In addition, DOC contributed primarily to wet carbon flux, with a proportion ranged from 78.8 to 85.9 % and an average was 82.9% in July. Then, the proportion of DOC contribution decreased in August and

160 September, which was ranged from 7.6-41.8% with an average of 17%. Otherwise, DIC contribution in July was lower and then increased at August and September. It was calculated that DOC deposition flux in July, August, September were 541.64, 131.03, and 44.44 mg C m^{-2}. Meanwhile, DIC in each month were 94.60, 385.03, and 259.16 mg C m^{-2}. The highest DOC deposition flux was found at July with a relatively small rainfall amount. With higher amount of precipitation, DOC deposition flux decreased at August and September. Thus, it can be assumed that DOC concentrations in rainfall may have

165 major impact on DOC deposition flux, which may be related to the source of DOC in this region. Meanwhile, DIC deposition flux appears in accordance with variations of the associated rainfall amount in each month. Based on the DOC and DIC concentrations and rainfall amount during the rainy season (July-September), VWM concentrations of DOC and DIC were 7.06 and 7.00 mg C L^{-1}. Therefore, it was estimated the annual wet deposition of DOC and DIC were 1.91 and 1.89 g C m^{-2} yr^{-1}, respectively.

170 **3.2 Correlation analysis**

Correlations between DOC/DIC concentrations and NH$_4^+$-N, NO$_3^-$-N, TDS, pH in rainfall events samples were shown in Table 3. The concentrations of NH$_4^+$-N, NO$_3^-$-N, TDS and pH in terms of regression functions of DOC and DIC in

rainfall are presented in Figure 3. For DOC, significant positive correlation between DOC concentration and NO_3^--N, TDS in rainfall was obtained with correlation coefficient of 0.756, 0.565 (P<0.01), whereas a negative correlation between DOC concentration and NH_4^+-N with correlation coefficient of -0.669 (P<0.01). Positive correlation was found between DOC concentration and NO_3^- might be due to the source of vehicle emissions, which was a direct source of organic acids and NO_3^- (Figure 3-a) , as also stated by Santos et al. (2014) and Willey et al. (2000). Meanwhile, DOC was also positively correlated with TDS and indicated that dissolved solids may contribute to organic carbon in rainfall, which probably related to the loess particle (Figure 3-b). As shown in Table 2, the observed TDS varied with an extensive ranged from 10.70-253.70 mg L^{-1} and the average value was 73.10 mg L^{-1} for all the rainfall events occurred in study period. The average TDS in July, August and September were 131.22, 74.01, 25.33 mg L^{-1}, which coincided with variations of DOC concentrations in each month. TDS in rainfall in studied region was much higher than other regions of China. As reported by Hao et al. (2017) in the Xiangxi river catchment at eastern China, TDS was ranged from 40.63 to 70.71 mg L^{-1} on average of 55.26 mg L^{-1} in rainfall. Therefore, the presence of higher TDS may contribute to organic carbon, which may explain why higher DOC concentrations were found in rainfall at Yangjuangou catchment. Furthermore, negative correlation between DOC and NH_4^+-N concentrations indicated that lower concentration of NH_4^+-N and pH with a higher DOC were found in July (Figure 3-c). It disagreed with the conclusions made by Santos et al. (2014) and Santos et al. (2011), who mentioned that a positive correlations obtained between DOC and NH_4^+-N. Dissolved NH_4^+ was likely related with dust particles originated from agriculture and may have a contribution to acid neutralization (Lohse et al., 2008). Santos et al. (2014) concluded that the acidity of deposition depends on the concentration of acid-forming ions and alkaline species. Therefore, higher DOC concentrations in rainfall may diminish dissolved NH_4^+, thus a lower NH_4^+-N concentrations and pH in rainfall in July. Nevertheless, the lower DOC concentration may lead to more NH_4^+-N, which may neutralized acidity, and consequently with a higher NH_4^+-N concentration and pH in August and September. In addition, a significant positive correlation between DIC concentration and NH_4^+-N, pH was found with correlation coefficient of 0.691, 0.615 (P<0.01), respectively (Figure 3-d & 3-e). Dissolved inorganic carbon was originated from carbon dioxide dissolved and as a form of carbonic acid in rainfall. NH_4^+-N in rainfall and formed ammonia or ammonium nitrate, which may be neutralize the carbonic acid. Thus, it may be resulted in a higher DIC concentration with a higher NH_4^+-N concentration and pH.

3.3 Comparisons wet carbon deposition with other sites

Owing to various sources, meteorological condition, seasons and sampling time in different regions, DOC and DIC in precipitation exhibited spatial and temporal variations. It should be noted that the investigation on DOC and DIC deposition via precipitation has not been conducted in the LPR. Moreover, to our knowledge, there were two similar observations were performed for other sites, northern and Tibetan of China (Li et al., 2016; Pan et al., 2010). In this study, VWM DOC concentrations were ranged from 1.01-24.62 mg C L^{-1} during July to September. Meanwhile, VWM DOC

concentrations in the rainy season (from July to September) were 7.06 mg C L^{-1}, which suggested that higher wet deposition

205   of atmospheric organic carbon in the LPR. DOC concentrations measured in rainfall at LPR were much higher than those

from Beijing (3.90 mg C L^{-1}) (Pan et al., 2010), and Lhasa (1.10 mg C L^{-1}) (Li et al., 2017) , capital of Tibet Autonomous

Region in China. The carbonaceous aerosol particles and soluble organic gases in the atmosphere may have a major impact

on DOC concentrations in precipitation. Pan et al. (2010) found that the relatively serious air pollution by industrial and

urban traffic activities corresponded to a higher DOC concentration in Beijing than those in other northern of China. Also, Li

210   et al. (2016) reported that the anthropological activities play an important role in increasing DOC concentration by providing

the organic gaseous and particles into atmosphere, such as intensively burning biomass for religious, vehicle transportation.

The Yangjuangou catchment located at the rural area with less potential pollution sources from transportation system, but it

exhibited higher level of DOC compared to Beijing and Lhasa. As it may be seen in Figure 2, the DOC deposition fluxes

were the predominant proportion and higher concentrations in July, while DIC deposition fluxes became higher than DOC

215   and associated with larger DIC concentrations in August and September. This might explain by a higher contribution of

anthropological emission in July. These results highlighted the simultaneous gaseous organic carbon (OC) source and rainfall

events influence on the DOC and DIC deposition fluxes during wet deposition process. A higher relative proportion of DOC

deposition fluxes with positive correlation observed between DOC and NO_3^--N concentrations may prove a higher

contribution of anthropological emissions in July, as also reported by Santos et al. (2011). Furthermore, NO_3^--N dissolved in

220   rainwater and formed acid condition may reduce CO_2 dissolution, which observed lesser DIC concentration in July rainfalls.

As rainfall amount and frequency increased, the occurrence of dilution effect and reduced gaseous OC may lead to a

decreased DOC concentration and deposition fluxes in August and September. Indeed, Santos et al. (2014) suggested that

probably the acid neutralization due to the presence of NH_4^+-N, which was beneficial for CO_2 dissolution in the rainwater.

This might be another aspect that further supported by the positive relationship between DIC concentrations and pH, NH_4^+-N

225   concentrations, as showed in Figure 3-d & 3-e. Regarding the carbon source, rainfall characteristics and the interactions

between ions in rainwater, a seasonal effects on wet carbon deposition process was observed in LPR.

       In addition, VWM concentrations of DOC in rainfall from the LPR was higher than observations reported for other

sites in the world, such as USA (1.1-2.9 mg C L^{-1}) (McDowell and Likens, 1998; Quideau and Bockheim, 1997; Willey et al.,

2000), Brazil (3.3-4.1 mg C L^{-1}) (Coelho et al., 2008), New Zealand (0.12-4.81 mg C L^{-1}) (Kieber et al., 2002), Korea (0.18-

230   9.36 mg C L^{-1}) (Yan and Kim, 2012), and Poland (4.72-5.10 mg C L^{-1}) (Siudek et al., 2015). Such differences between the

present studied region and other regions might be partly explained by the various carbon sources, meteorological type,

sampling time scale and methods in mainland or coastal regions. Furthermore, the monthly VWM concentration of DOC

ranged from 24.62 mg C L^{-1} (July) to 1.01 mg C L^{-1} (September). In this study, the highest DOC concentration occurred in

July might relate to abundance sources, such as aerosol, agricultural activities, and the meteorological conditions.

235   Furthermore, large variations of DOC concentrations may be attributed to loess dust scavenged by raindrops and might be a

source of organic carbon. These results agreed with Mladenov et al. (2012), who found that a high carbon deposition was associated with dust in Colorado Rocky Mountains, USA. Consequently, the role of loess dust deposition related to the wet carbon deposition cannot be negligible and be worthy special attention on characterizing the carbon deposition in this region. Due to the limited sampling period, we did not determine the annual variation pattern of DOC, however, it reflected the DOC
240 concentration in rainfall during the rainy season and highlighted the importance of investigating wet carbon deposition in the LPR.

Figure 2 showed the wet carbon concentration, deposition flux in July, August, and September. Therefore, it was estimated that annual wet deposition of DOC and DIC were 1.91 and 1.89 g C m^{-2} yr^{-1}, which were much higher than that of other reported regions in China. For instance, the annual wet deposition flux of DOC and DIC was 1.9, 0.7 g C m^{-2} yr^{-1} in
245 northern China (Pan et al., 2010) and 0.63 g C m^{-2} yr^{-1} of DOC flux observed in Tibetan Plateau (Li et al., 2016). With regard to the worldwide sites, the estimated values in present study were much lower than reported in Brazil (Ribeirao Preto-4.8 g C m^{-2} yr^{-1}, Araraquara-5.4 g C m^{-2} yr^{-1}) (Coelho et al., 2008), USA (North Carolina-2.87 g C m^{-2} yr^{-1}) (Willey et al., 2000). Moreover, it was comparable to the annual wet deposition flux of DOC in Seoul, South Korea (1.90 g C m^{-2} yr^{-1}), which was mostly originated from emissions by fossil-fuels combustion (Yan and Kim, 2012). Therefore, atmospheric wet carbon
250 deposition at semi arid catchment of LPR may have considerable differences among domestic or worldwide regions. It is further implied that the atmospheric wet carbon deposition exhibit a less precipitation with high concentration and represent a substantial carbon input of a watershed in semi arid area of LPR. Even though the annual wet carbon deposition flux was estimated and may have uncertainty due to limited samples, evidence of its significant role was revealed. Hence, further investigations should be long-term undertaken to verify the spatial-temporal variation pattern of atmospheric carbon
255 deposition process in semi arid region in relation to the global carbon cycle.

4 Conclusion

In this study, concentration of DOC and DIC in rainfall was measured in period of July to September 2015 at Yangjuangou catchment in the LPR. It was found that VWM concentrations of DOC in rainfall of July, August, September were 24.62, 3.58, 1.01 mg C L^{-1} respectively, presenting a decreasing trend. However, the VWM concentrations of DIC in
260 rainfall of July, August, September was 4.30, 10.52, 5.89 mg C L^{-1}, respectively. Moreover, the monthly deposition flux of DOC and DIC were 541.64/94.60, 131.03/385.03, and 44.44/259.16 mg C m^{-2} for July, August and September. In addition, VWM concentrations of DOC and DIC for the concentrated rainy season (July-September) in the studied region were 7.06 and 7.00 mg C L^{-1}. The estimated annual wet carbon deposition was 1.91, 1.89 g C m^{-2} yr^{-1} for DOC and DIC. Wet carbon deposition of concentration and flux exhibited strong variation, which may derive from carbon source. Besides the biogenic
265 and anthropologic emission source, the loess dust deposition, which a source for soil parental material in soil formation

process at the LPR, might be another source of carbon in precipitation and merit further relevant work. Although the present work provide an estimate of wet carbon deposition, further investigation should be conducted on carbon source and deposition flux from atmosphere at long term temporal and large scale for revealing the global carbon biogeochemical cycle. Nevertheless, our primary results indicated that atmospheric deposition represents a substantial contribution to a watershed in 270   semi arid area of LPR.

Acknowledgments

This work was financially supported by the National Nature Science Foundation of China (41571130083 and 41390464), the State Key Laboratory of Urban and Regional Ecology Open Fund, the Research Center (SKLURE2015-2-2) for Eco-Environmental Sciences, the Chinese Academy of Sciences (CAS), and the Youth Innovation Promotion Association, CAS.

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Table 1. The land use types and it's area in Yangjuangou catchment

Soil type	Forest	Grass	Shrub	Agricultural	Terrace	Orchard	Residential	Water
Area (km²)	0.649	0.528	0.512	0.101	0.186	0.011	0.03	0.003
Ratio (%)	32.14	26.17	25.32	0.501	9.22	0.53	1.46	0.15

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Table 2. Characterization of each rainfall sample and temperature, wind during sampling period in Yangjuangou catchment.

Sample date	NH_4^+-N (mg L^{-1})	NO_3^--N (mg L^{-1})	TDS (mg L^{-1})	pH	Rainfall (mm)	Temperature (℃)		Wind speed (m s^{-1})	
						Range	Mean	Range	Mean
Jul.-17	0.04	3.24	103.37	6.34	3.80				
Jul.-19	0.04	3.05	104.00	6.99	9.00	13.6-33.5	22.2	0.2-3.3	0.43
Jul.-29	0.05	1.18	253.70	6.54	0.60				
Jul.-31	0.04	2.57	63.79	5.70	2.40				
Aug.-2	0.02	1.09	48.87	6.53	10.00				
Aug.-3	1.07	1.27	36.53	6.54	0.80				
Aug.-10	0.86	1.88	103.32	7.24	2.20				
Aug.-11	1.16	1.09	47.90	6.76	4.40	10.7-34.0	22.6	0.2-2.1	0.37
Aug.-13	0.77	1.53	68.17	6.72	0.60				
Aug.-25	1.25	1.74	115.37	6.80	4.20				
Aug.-30	0.14	0.96	97.94	6.86	0.60				
Sep.-4	0.32	0.75	15.45	6.29	13.00				
Sep.-8	0.39	0.65	19.13	6.04	5.40				
Sep.-10	0.58	0.79	10.70	6.17	13.60	9.1-31.3	17.5	0.2-1.9	0.37
Sep.-22	0.54	0.91	44.65	6.74	0.50				
Sep.-27	0.63	0.86	36.70	6.64	0.50				

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Table 3. Correlation coefficients between DOC, DIC, NH_4^+-N, NO_3^--N, TDS, and pH (n=48).

DOC	DOC	NH_4^+-N	NO_3^--N	TDS	pH
DOC	-	-0.669**	0.756**	0.656**	-0.086
NH_4^+-N	-	-	-0.439	-0.176	0.325
NO_3^--N	-	-	-	0.388*	-0.106
TDS	-	-	-	-	0.371*

DIC	DIC	NH_4^+-N	NO_3^--N	TDS	pH
DIC	-	0.691**	-0.325	0.161	0.615**
NH_4^+-N	-	-	-0.439*	-0.176	0.325
NO_3^--N	-	-	-	0.388*	-0.106
TDS	-	-	-	-	0.371*

Note: ** ($P<0.01$), * ($P<0.05$)

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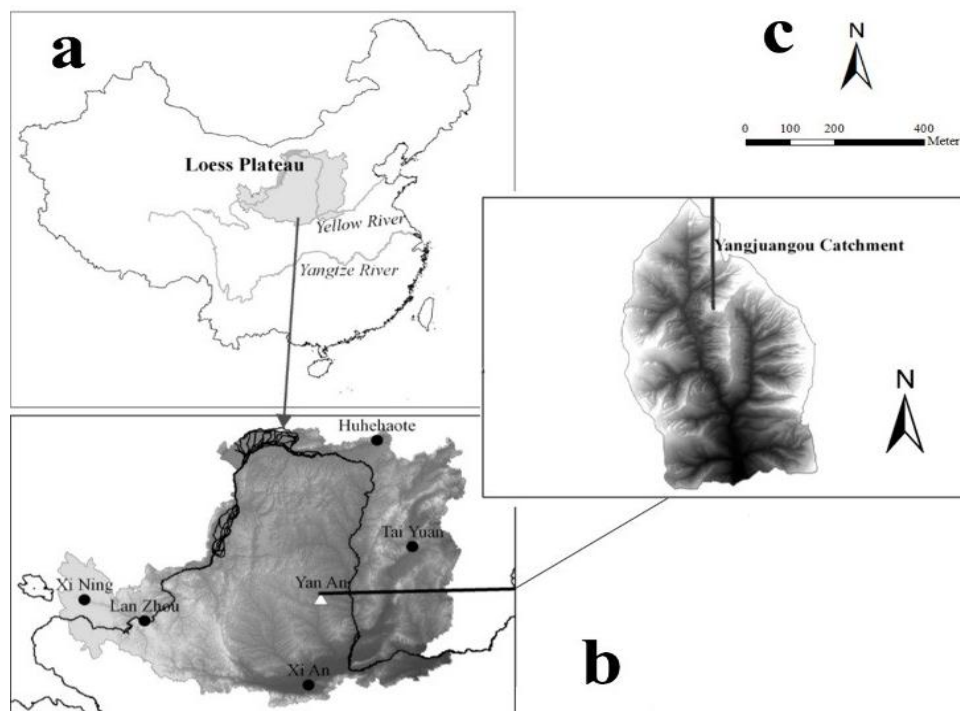
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Figure 1 Geographic location of the Yangjuangou catchment (c) in Loess Plateau region (b) of China (a).

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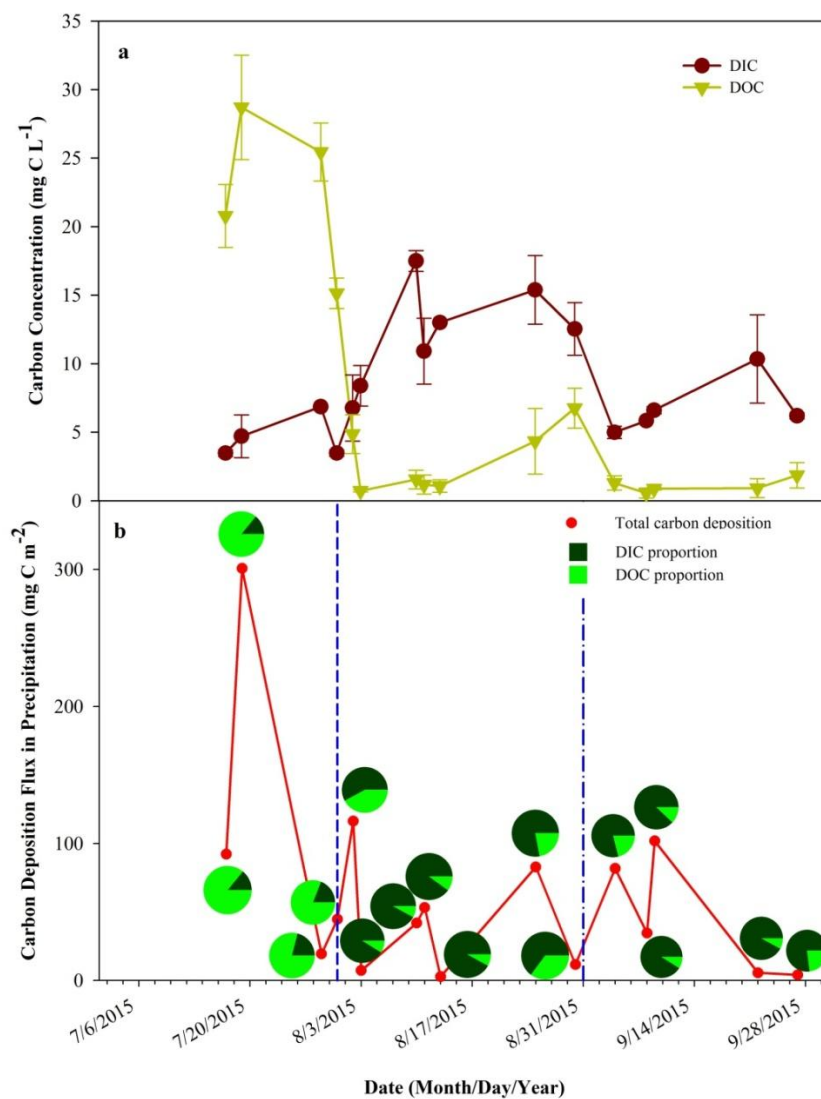
Figure 2 The carbon concentration: (a), deposition flux and the proportion of DOC and DIC: (b) in individual rainfall event during July to September.

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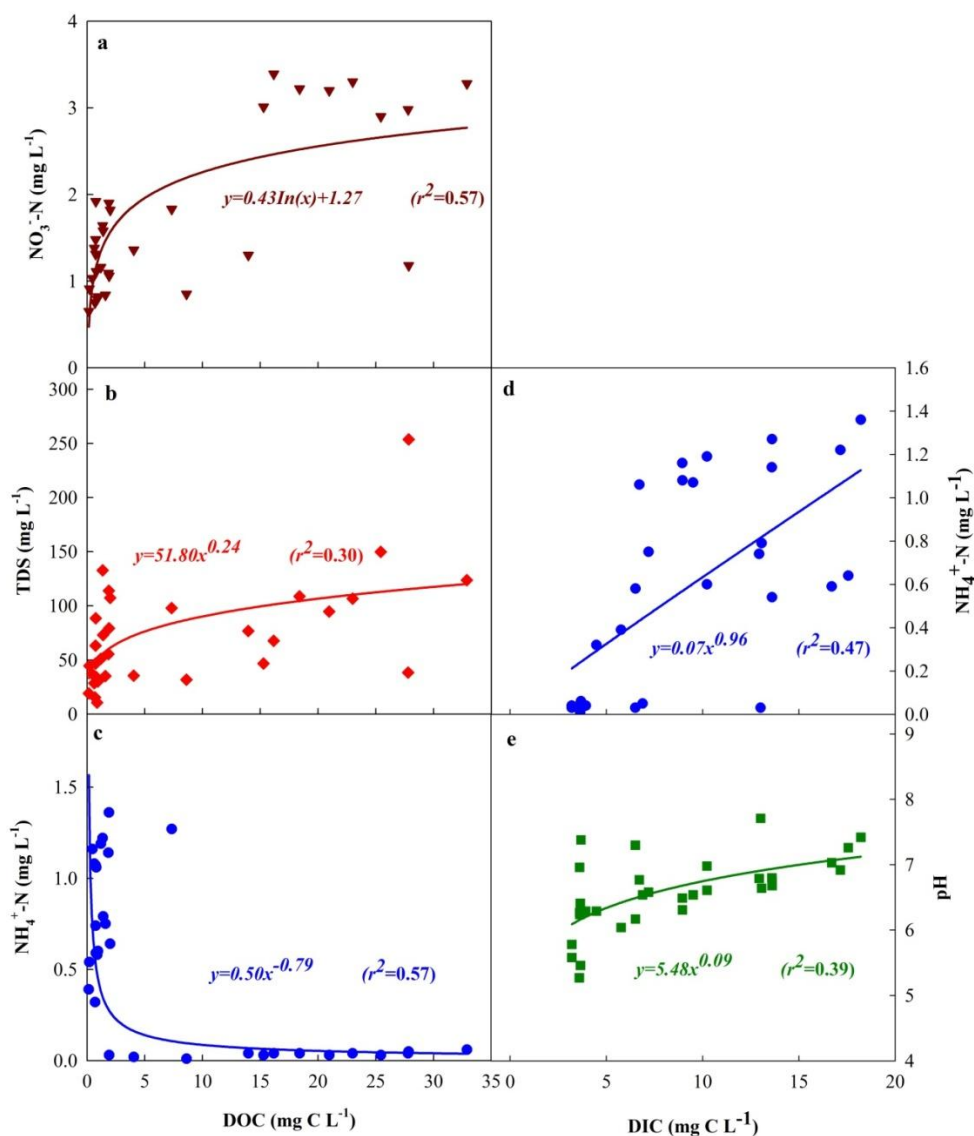
Figure 3 Significant correlations between DOC concentrations and various rainwater variables: (a) NO_3^--N, (b) TDS, (c) NH_4^+-N, in rainfall event samples collected during July-September. In addition, correlations between DIC concentrations with variables of: (d) NH_4^+-N, (e) pH, are also demonstrated accordingly.