Response to bg-2017-488-RC1

We appreciate for anonymous referee comments concerning our manuscript entitled "Characteristics of wet dissolved carbon deposition in a semi-arid catchment at the Loess Plateau, China"(ID: bg-2017-488). We have studied comments carefully and have made corrections. The main corrections in the paper according to the referee's comments are as follows:

Comment 1. The authors do not have good definition of what is studied, "wet carbon deposition" make readers confused, which also includes particle carbon. Please change to "wet dissolved carbon deposition" for the whole article.

Response: Thanks for your suggestions. We have changed "wet carbon deposition" to "wet dissolved carbon deposition" in this whole manuscript.

Comment 2. The research direction is interesting. However, the authors do not provide strong evidences the reason of doing study at LPC. Furthermore, due to the studied area is dry and have heavy dust storms, so that dry deposition should also accounts for large part, which need to be at least pointed out in detail in the article.

Response: Thanks for your suggestions. Because the topic of this manuscript is about wet dissolved carbon deposition, thus the dry deposition is not mentioned although it may account for a large part of carbon deposition. Thanks for your advices and we will focus the dry and wet carbon deposition in the future experimental research.

Comment 3. The introduction part is long and need to be cut short. Meanwhile, the logic of introduction is not clear and some similar ideas appear at different part. It is good to cite studies have been done in China, but the author need to point out their potential connection to this study.

Response: Thanks for your suggestions. The introduction part has been rewrite and the details show in Line 25-85 of this manuscript:

Line 25-85:

Wet carbon deposition is recognized as the rainfall scavenging of aerosols and gas phase organic compounds, which originated from biogenic and anthropogenic sources (Duarte et al., 2006; Houghton, 2003; May et al., 2013). Dissolved carbon is a ubiquitous component of rainwater in many regions around the world (Dachs et al., 2005). Global scale model simulations show that DOC concentrations ranging from 1 to 10 mg C L^{-1} with a total of 188 Tg C yr^{-1} of wet dissolved organic deposition flux occurred during the terrestrial rainfall, which equals approximately to 43% of the total particulate organic carbon transported by rivers to oceans (Galy et al., 2015). A modeling study conducted by Safieddine and Heald (2017) estimated that the total global wet deposition of DOC was 273 Tg C yr^{-1}. Similarly, Willey et al. (2000) found that the concentration of DOC in terrestrial precipitation was 1.93 mg C L^{-1}, which was greater than nitric and sulfuric acid combined. Thus, the global flux of DOC and DIC via precipitation can be estimated as 430 and 80 Tg C yr^{-1} (Willey et al., 2000). Furthermore, as a labile and bioavailable fraction of dissolved carbon, wet deposition of dissolved carbon may provide a substantial input of nutrient source for terrestrial and aquatic ecosystems. For example, Mladenov et al. (2012) used a long-term dataset of weekly DOC deposition and demonstrated that atmospheric wet deposition of dissolved carbon represented a significant source to an alpine catchment in the Rocky Mountains of Colorado, USA. 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 a semi-arid catchment in the LPR. Consequently, the large magnitude of dissolved carbon flux via rainfall played an important role in ecological processes and acted as one of the key driving forces of the global carbon biogeochemical cycle. Thus, it is urgent to improve the associated knowledge and understanding of dissolved carbon deposition in rainfall.

With regard to relevant studies on nitrogen and phosphorus in atmospheric wet

deposition, only a few quantitative studies are available on the atmospheric wet deposition of dissolved carbon. Iavorivska et al. (2016a) summarized that there were only 83 contemporary studies over the last three decades focused on the concentration and flux of dissolved organic carbon via rainfall at a worldwide scale. These available datasets were sparsely measured and only a handful of studies maintained a long-term monitoring. Dissolved carbon deposition concentration and flux in rainfall are not general parameters in monitoring networks, such as the National Atmospheric Deposition Program (USA), European Monitoring Evaluation Program (European Union) and Chinese Ecosystem Research Network (China) (Iavorivska et al., 2016a). In general, only few studies have assessed the dissolved carbon deposition via rainfall, such as the 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 (0.1-4.8 mg C L^{-1}) (Kieber et al., 2002), Korea (0.2-9.4 mg C L^{-1}) (Yan and Kim, 2012), and Poland (4.7-5.1 mg C L^{-1}) (Siudek et al., 2015). Until recently, the only measurement data available were those related to wet dissolved carbon deposition in the northern and Tibetan region of China (Li et al., 2016; Pan et al., 2010). Pan et al. (2010) reported that significant seasonal differences in DOC concentrations and deposition fluxes are found in northern China. The corresponding annual average concentration and deposition flux of DOC from the atmosphere ranged from 2.4 to 3.9 mg C L^{-1} and from 1.4 to 2.7 g C m^{-2} yr^{-1}, respectively. Li et al. (2016) also reported that the DOC concentration of seasonal precipitation varied between monsoonal and non-monsoonal periods and the average deposition of DOC was 1.1 mg C L^{-1}. The annual deposition flux of DOC was about 0.6 g C m^{-2} yr^{-1} in Tibetan Plateau of China. These results indicated that wet dissolved carbon concentration and deposition fluxes were commonly found in coastal, forested and alpine regions and showed a spatial and temporal variation between different regions. The differences were attributed to rainfall, meteorological conditions and were related to the regional source of carbon. In addition, due to the lack of quantitative measurement in dissolved carbon concentration and associated fluxes, including corresponding sources, chemical composition and variation patterns, the carbon

exchange between the atmosphere and terrestrial ecosystem have not been incorporated into the current regional or global carbon cycle models (Jurado et al., 2008; Kieber et al., 2002). Thus, wet dissolved carbon deposition in rainfall remains unsolved and highlights the need to acquire more information.

Previous studies have provided insights on the magnitude and importance of wet dissolved carbon deposition in rainwater worldwide. However, studies that have explored atmospheric wet deposition of dissolved carbon are rarely found in the target research area, which is a semi arid catchment in the LPR. The LPR (N 35-41°, E 102-114°), which has an area of 6.4×10^5 km^2, is situated in the middle stream of the Yellow River. The plateau is covered by an average thickness of 100 m of loess. Loess is formed by the accumulation of wind-blown silt (Ding et al., 2002). Meanwhile, the fine-grained particles may serve as nuclei to form a rain droplet or cloud condensation. Consequently, it is worth noting that atmospheric dust scavenged by rainfall may be another source of dissolved carbon, which might differ from other regions. Thus, attention should be paid to the wet deposition processes of DOC and DIC, which were a potential external input of carbon to the semi-arid catchment in the LPR. Until this point, knowledge of wet dissolved carbon deposition and the associated flux has not been fully explored in the LPR. Thus, it is necessary to investigate the DOC and DIC deposition via rainfall, where little information is available.

In this study, we measured DOC and DIC concentrations in samples collected during 16 rainfall events from July to September in the Yangjuangou catchment, which is a semi arid-catchment of LPR. Therefore, the primary goal of this study is to investigate the variations of DOC and DIC fluxes from the atmosphere to understand the magnitude of dissolved carbon deposition during the concentrated rainfall season in LPR. Specifically, the three objectives of this study were to examine the concentration of DOC and DIC and the associated variations in a rainfall event or monthly periods, to quantify the deposition fluxes of DOC and DIC, and to explore the relationships between dissolved carbon, rainfall properties and rainwater chemical characteristics. These results will provide evidence of wet dissolved carbon deposition,

which may be important for understanding the carbon cycle and ecosystem response in a semi-arid catchment in the LPR

Comment 4. I am sorry to find that the English of this article is poor and some mistakes are made mainly because of carelessness.

Response: Thanks for your suggestions. The manuscript has been thorough improved by an English edition company (American Journal Experts: https://www.aje.cn).

Comment 5. The method of the study is not good expressed and only some samples of three months were collected, which I think is not enough to study the precipitation characteristics of study area. What kind of bottle and what material made of? which is important for doing study of DOC, if the containers were put outside for long time before the rainfall, the final data should also include some dry deposition.

Response: Thanks for your suggestions. These questions about rainwater sampling method have been added in detail in Line 103-123. And Line 126-130. And we also added a photo of the field station in this catchment in Figure 1.

Line 103-123:

In the Yangjuangou catchment, the experimenters resided in the field observatory station and treated the samples immediately after a rainfall event to ensure the dissolved carbon in the rainwater did no microbiologically biodegrade. Because the common sampling frequency is monthly or weekly at this field observatory station, this sampling method may result in dissolved carbon in rainwater that is degraded by microbial activities (Kieber et al., 2002; Willey et al., 2000). This is also the reason for the measurement scarcity of wet dissolved carbon deposition concentrations and fluxes in the existing ecosystem monitoring networks worldwide. In this study, rainfall samples were collected from July to September 2015. The use of an

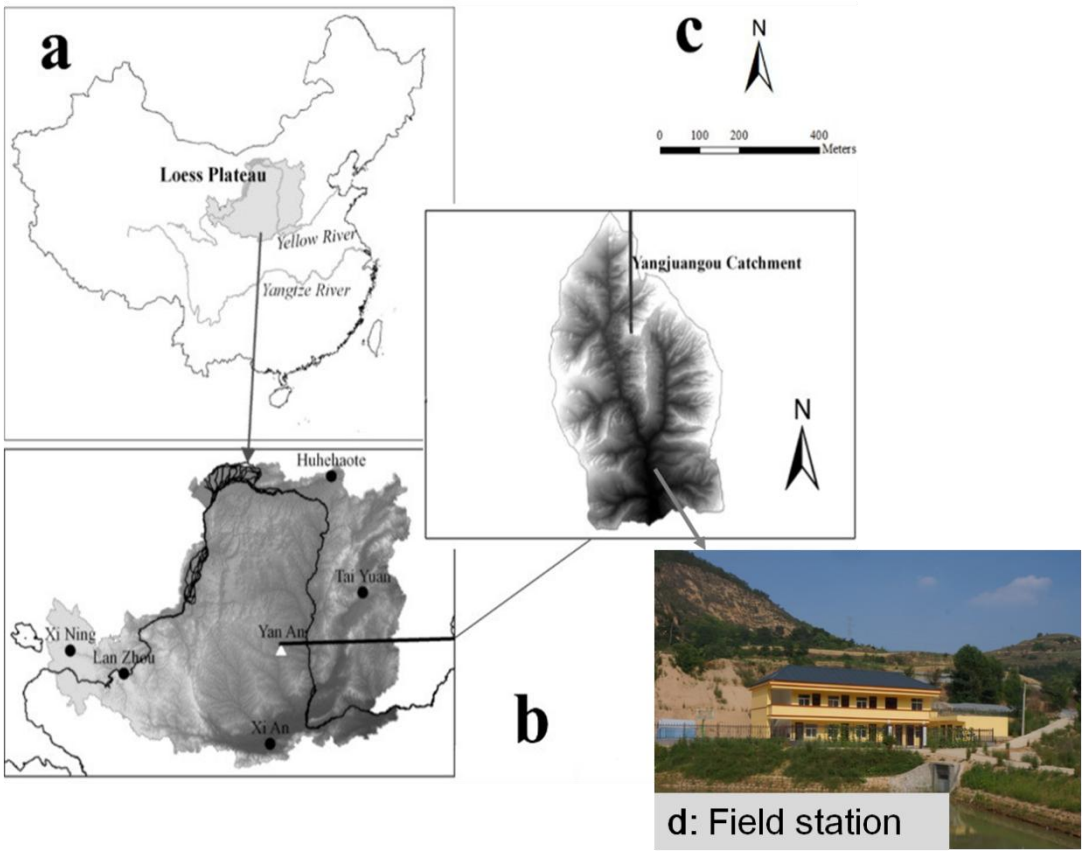
open-to-the-atmosphere collector is a common approach for collecting atmospheric wet deposition during an individual rainfall event. A rain gauge was installed on the roof of the building at the sampling site to determine the rainfall amount and can be used for collecting rainfall samples during a rainfall event. Two additional duplicate rainfall samples were collected using a steel bucket (d=29 cm). During the field collection, the rainwater samples were poured into high-density polyethylene bottles at the end of a rainfall event. All rainfall containers were cleaned with deionized water after a collection and returned to the sampling sites for the next rainfall sampling. Information on the rainfall events that occurred and were sampled during the study period is summarized in Table 1 and Figure 2. There were 37 rainfall events in total and a total rainfall amount of 102.4 mm over the sampling period of July to September, of which the individual rainfall amount varied from 0.2 mm to 13.6 mm. The rainfall event collections in July, August, September were 4, 7, and 5, respectively, whereas the corresponding total rainfall events were 6, 12, and 7 in each month. Therefore, we sampled 15.8, 22.4 and 32.9 mm of rainfall that occurred in each month, respectively. The sampled rainfall accounted for 69.4% of the total amount of rainfall over the sampling period. Some events were not sampled due to the experimenter transporting the samples to the State Key Laboratory of Urban and Regional Ecology in Beijing for indoor analysis, and thus, there is no experimenter was at the field station during those events.

Line 126-130:

In this study, there are three steps for conducting in situ and laboratory measurements. First, the TDS (total dissolved solids) and 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) and stored in high-density polyethylene bottles. Before use, the bottles were strictly cleaned with a 10% hydrochloric acid solution, and then, these bottles were soaked in deionized water and air-dried. The bottles filled with 200 ml filtrates were stored in a freezer (4℃). After

completing the rainwater filtration, all filtered membranes were soaked in 80℃ deionized water and maintained at this temperature for 12 h to clean and air-dry for the next use.

Figure 1:



Comment 5. Due to only three months were studied, it is far-fetching to discuss decreasing or increasing trend of concentrations.

Response: Thanks for your suggestions. We have changed into compare decreasing or increasing trend only during three sampling months in this manuscript.

Comment 6. Not enough to study the precipitation characteristics of study area during three months.

Response: Thanks for your suggestions. We have added discussion information

between the rainfall characteristics and the concentrations and fluxes of dissolved carbon in Line 165-179 and Line 190-197. Correspondingly, we also added the interval of rainfall in Table 1 and the Figure 3 shown the relationship between the rainfall amount and dissolved carbon concentrations and fluxes. The following will show in detail:

Line 165-179:

The variation in the dissolved carbon concentration of the rainwater could be attributed to the differences of rainfall amount and frequency, carbon sources, and meteorological conditions (Iavorivska et al., 2017a; Iavorivska et al., 2017b; Iavorivska et al., 2016b). As shown in Figure 3-a&b, the concentration of DOC and DIC in the rainwater generally decreased with a single rainfall amount. Similar relationships between the dissolved carbon concentration and rainfall amount have also been found at other sites (Heartsill-Scalley et al., 2007; Pantelaki et al., 2018; Santos et al., 2013). However, DOC concentrations can be high or less associated with a smaller rainfall amount during a single event (Figure 3-a&b). These findings suggested that the concentration of DOC was not followed a dilution behavior. Elevated DOC concentrations were observed in July, which could be attributed to aerosols emitted from biogenic and anthropogenic activities and dust particles (Kieber et al., 2002; Mladenov et al., 2012). In addition, the frequency of rainfall events is lower and shows a flush effect. For example, when the rainfall event occurred on July 17, it had been 10 days since last rainfall, suggesting that DOC tends to be effectively flushed from the atmosphere with a large rainfall amount. However, the decreased DOC concentrations in August and September may be attributed to a reduced aerosol source (Witkowska and Lewandowska, 2016). Moreover, the frequency of rainfall events was increased, and the rainfall intervals ranged between 0 and 5 days, and thus, this may lead to a reduction in the DOC concentration (Figure 2-a).

Line190-197:

A higher DOC deposition flux was found in July with a significantly higher DOC concentration, especially the rainfall that occurred on July 19. However, there is a

reduced DOC deposition flux, even with higher DOC concentration associated with a lesser rainfall amount, such as the single rainfall events on July 29 and 31. These results indicated the lesser rainfall amounts may overwhelm the effects of a high DOC concentration. Meanwhile, the DIC deposition flux appeared in accordance with variations in the associated rainfall amount for each month. Thus, the DOC concentrations in rainfall may have a major impact on the DOC deposition flux, which is supported by the results shown in Figure 3-c & d. These results show that the different variations in DOC and DIC occur during sequential rainfall events in the concentrated rainfall season in the LPR.

Table 1 Characterization of rainwater chemistry from the rainfall events that occurred during the sampling period of July to September.

Sample date	NH_4^+-N (mg L^{-1})	NO_3^--N (mg L^{-1})	TDS (mg L^{-1})	pH	Rainfall (mm)	Rainfall interval days (d)
Jul.-17	0.04	3.24	103.37	6.34	3.80	10
Jul.-19	0.04	3.05	104.00	6.99	9.00	0
Jul.-29	0.05	1.18	253.70	6.54	0.60	7
Jul.-31	0.04	2.57	63.79	5.70	2.40	1
Aug.-2	0.02	1.09	48.87	6.53	10.00	0
Aug.-3	1.07	1.27	36.53	6.54	0.80	0
Aug.-10	0.86	1.88	103.32	7.24	2.20	5
Aug.-11	1.16	1.09	47.90	6.76	4.40	0
Aug.-13	0.77	1.53	68.17	6.72	0.60	0
Aug.-25	1.25	1.74	115.37	6.80	4.20	0
Aug.-30	0.14	0.96	97.94	6.86	0.60	1
Sep.-4	0.32	0.75	15.45	6.29	13.00	0
Sep.-8	0.39	0.65	19.13	6.04	5.40	3
Sep.-10	0.58	0.79	10.70	6.17	13.60	0
Sep.-22	0.54	0.91	44.65	6.74	0.50	2
Sep.-27	0.63	0.86	36.70	6.64	0.50	4

Figure 3 Relationships between measured parameters and rainfall amounts in a single rainfall event during July to September: (a) DOC concentration, (b) DIC concentration, (c) DOC flux and (d) DIC flux.