Response to bg-2017-488-RC2

Thanks for your suggestions. 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. A major issue is the use of English. The manuscript needs a major revision in terms of grammar and phrasing.

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

Comment 2. Four and five significant figures are given for the numbers! Probably the uncertainty associated with these measures would not allow to use more than 2 or 3.

Response: Thanks for your suggestions. The figures in this manuscript has been changed no more than 2 or 3, such as in table 3.

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	-	-0.67**	0.76**	0.66**	-0.09
NH_4^+-N	-	-	-0.44	-0.18	0.33
NO_3^--N	-	-	-	0.39*	-0.11
TDS	-	-	-	-	0.37*

DIC	DIC	NH_4^+-N	NO_3^--N	TDS	pH
DIC	-	0.69**	-0.33	0.16	0.62**
NH_4^+-N	-	-	-0.44*	-0.18	0.33
NO_3^--N	-	-	-	0.39*	-0.11
TDS	-	-	-	-	0.37*

Comment 3.The three last sentences of the abstract need to be re-written.

Response: Thanks for your suggestions. These sentences of the abstract have been re-written in Line 20-24.

Line 20-24:

The estimated annual wet carbon depositions were 1.91 and 1.89 g C m^{-2} yr^{-1} for DOC and DIC, respectively. The results of this study suggest the variation in concentrations and fluxes of DOC and DIC and explored that these variation may be related to the dissolved carbon source and the rainfall characteristics during the concentrated rainfall season in the semi-arid catchment of the LPR. Furthermore, these results also suggest that dissolved carbon may be an important external input of carbon into terrestrial ecosystems.

Comment 4. Line 26 "massive exchange of physical processes" means nothing. Review the use of English (grammar and meaning).

Response: Thanks for your suggestions. "massive exchange of physical processes" has been deleted and the whole manuscript has been reviewed and improved an English edition company.

Comment 5. Line 52. In addition to scavenging aerosols, rain scavenges the gas phase organic compounds.

Response: Thanks for your suggestions. We have changed in the Line 26-28.

Line 26-28:

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).

Comment 6.The writing of the introduction needs to be improved so it flows better. There are some sentences that do not make sense in this context (for example lines 73-74).

Response: Thanks for your suggestions. The introduction part has been rewritten 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 7. Which were the blanks for DOC and DIC?

Response: Thanks for your suggestions.

Line 135-138:

TDC is automatically measured by the combustion of a sample, whereas DIC is measured after acidification of a sample. The distilled water blanks were also tested every 50 samples for ensuring the quality of results. TDC is recognized as the sum of the DIC and DOC components, and thus, the DOC was the difference between TDC and DIC for each sample (DOC=TDC-DIC).

Comment 8. Lines 151-152. It may be important, but its relevance should be demonstrated in terms of a mass balance or comparison with other sources and/or fluxes.

Response: Thanks for your suggestions. This details show in Line 166-179 of this manuscript:

Line 166-179：

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).

Comment 9. Try to be concise and not repeat data and ideas in the discussion. For example, at lines 232-233, these concentrations were given before. The discussion could be shortened significantly, and the message would be more clear.

Response: Thanks for your suggestions. The discussion part has been concisely shorten and did not repeat data and ideas in line 226-264.

Line 226-264:

Owing to various sources, meteorological conditions, seasons and sampling times in different regions, the DOC and DIC of the rainfall exhibited spatial and temporal variations. Investigations on the DOC and DIC deposition via rainfall have not been previously conducted in the LPR. Moreover, to our knowledge, two similar observations were performed for other sites in northern and Tibetan of China (Li et al., 2016; Pan et al., 2010). In this study, the DOC concentrations ranged from 1.01to 24.62 mg C L^{-1} over the sampling period of July to September. Meanwhile, DOC concentrations in the rainy season (from July to September) were 7.06 mg C L^{-1}, which suggested a higher wet deposition of dissolved organic carbon in the LPR. DOC concentrations measured in the rainfall in the 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). The carbonaceous aerosol particles and soluble organic gases in the

atmosphere may have a major impact on the DOC concentrations of the rainfall. As shown in Figure 2, the DOC deposition fluxes were the predominant proportion and had higher concentrations in July. This might be explained by a higher contribution of anthropological emissions. A higher relative proportion of DOC deposition fluxes with a positive correlation observed between DOC and NO_3^--N concentrations may highlight a higher contribution of aerosols from anthropological activities, which was also reported by Santos et al. (2011). Furthermore, the NO_3^--N dissolved in rainwater that formed acidic conditions may reduce the disassociated carbonic acid, which shown lesser DIC concentration for the July rainfall events. Indeed, Santos et al. (2014) suggested that the acid neutralization was likely due to the presence of NH_4^+-N, which was beneficial for forming disassociated carbonic acid in the rainwater. This might be another aspect that is further supported by the positive relationship between DIC concentrations and pH and NH_4^+-N concentrations, which is showed in Figure 4-d & 4-e. Regarding the dissolved carbon source, rainfall characteristics, meteorological conditions and the interactions between ions in rainwater, the wet dissolved carbon deposition concentrations were varied with different rainfall events in the LPR.

Figure 2 shows the wet dissolved carbon concentration and deposition flux in July, August, and September. Therefore, it was estimated that the annual wet deposition of DOC and DIC were1.91 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 and 0.7 g C m^{-2} yr^{-1} in northern China (Pan et al., 2010), and the observed DOC flux was 0.63 g C m^{-2} yr^{-1} in the Tibetan Plateau (Li et al., 2016). With regard to worldwide sites, the estimated values in the present study were much lower than those 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-2.9 g C m^{-2} yr^{-1}) (Willey et al., 2000). Moreover, the values were comparable to the annual wet deposition flux of DOC in Seoul, South Korea (1.90 g C m^{-2} yr^{-1}), which mostly originated from emissions of fossil fuel combustion (Yan and Kim, 2012). In addition, the magnitude contributions

of DOC and DIC to wet dissolved carbon deposition were approximately equal. These results at this study site were within the range reported for other sites (Fahey et al., 2005; Liu et al., 2010) and was also comparable with values measured on Pennsylvania, USA, which reported that contributions of DOC and DIC were 53% and 47% , respectively (Iavorivska et al., 2017a). Therefore, atmospheric wet dissolved carbon deposition in the semi-arid catchment of the LPR may have considerable differences among domestic or worldwide regions. Even though the annual wet deposition flux of dissolved carbon was estimated and may have uncertainty due to limited samples, the flux value reflected the dissolved carbon deposition in the rainfall during the concentrated rainfall season and highlighted that the wet dissolved carbon deposition may be a substantial external input of carbon to a semi-arid catchment in the LPR. Hence, further investigations should be long-term undertaking to evaluate the concentration and flux of dissolved carbon deposition and its impact on terrestrial ecosystems in semi-arid regions.

Comment 10. The concentrations of DOC are sometimes very large. I have doubts that these concentrations can be supported by scavenging of aerosols (even dust). There may be a contribution of scavenging organic matter present in the gas phase, but this is never discussed or mentioned.

Response: Thanks for your suggestions. We have added some information in discussion part and details show in the following:

Line 166-179:

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).

Comment 11. I suggest to look at the air-mass back trajectories for the different sampling events. This could provide important information on source regions, and maybe explaining the different concentrations of DOC in July and September.

Response: Thanks for your suggestion. Back-trajectory analyses are an method to describe the origin of air masses contributing to the rainfall at the sampling site during the sampling period. Iavorivska et al. (2016) reported that atmospheric dissolved carbon deposition via rainfalls mainly through two general mechanisms:(1) aerosols or gas-phase organic compounds dissolved in raindrops; (2) washout processes by raindrops on their way to land surface. The source of dissolved carbon in these two mechanisms is distant and local emissions, respectively. In addition, chemical reactions mediated by sunlight or oxidants also impact the wet deposition of dissolved carbon. According to the processes included source emission, transport, rainfall characteristics, the wet dissolved carbon deposition is a complex process. Therefore, the back-trajectory analyses is not enough to give an explanation the different

concentrations of DOC and this is the reason that this manuscript didn't use the back-trajectory analyses method.

References:

Iavorivska, Boyer, Grimm et al. Variability of dissolved organic carbon in precipitation during storms at the Shale Hills Critical Zone Observatory. Hydrological Processes, 2017, 31:2935-2950.

Other changes:

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 a figure 3 shown the relationship between the rainfall amount and dissolved carbon concentration and flux. The details show in the following part:

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.