

Response to Anonymous Referee #1

We appreciate Referee #1's critical and detailed assessment of our manuscript and we are grateful for his/her constructive comments which helped us to greatly refine our paper. Here we provide a point-to-point response to all the issues raised by the referee. We hope our replies and revisions will satisfy all the requests.

General Comments:

Comment 1:

The manuscript of Liu et al presents very interesting information regarding the triggering of soil carbon losses via respiration and leaching by extreme precipitation events. The results of the soil column experiments illustrate that leaching losses of carbon from soils as consequence of extreme precipitation events may well exceed carbon losses due to enhanced respiration. However, the overall relevance of dissolved organic and dissolved inorganic carbon leaching losses will ultimately depend on the fate of the leached carbon on its way to groundwater and further through rivers into the ocean. If the dissolved organic carbon and inorganic carbon are retained in subsoils, then the leached C might well be finally emitted from the soil to the atmosphere in the form of CO₂, if the dissolved organic carbon (DOC) is mineralized or if the soil water is lost via evapotranspiration, thus releasing the dissolved inorganic carbon (DIC)...This aspect of the importance of the downstream fate of leached carbon for the overall relevance of the leaching pathway for the carbon balance is missing in the manuscript.

Response:

This is a very good point! The downstream fate of the leached carbon is indeed very important as it will ultimately determine the relevance of leaching processes to the overall carbon budget or balance along the "soil-river-ocean" continuum. If part of the leached carbon is retained in the deeper soils or transformed and carried along from the river to the ocean in the form of DIC, it will not constitute a source of atmospheric CO₂ on a relatively short term (over years or decades). However, compared to DOC and DIC in the soil solution, the leached carbon is more likely to be subject to more intensified mineralization and outgassing during the land-ocean transfer, given more intensified mixing processes, oxygen exposure and photo-oxidation of terrestrial carbon upon releasing into the river (Hedges et al., 1997; Battin et al., 2009). Hence, we postulate that carbon leached from soils is more vulnerable to decomposition and/or release compared to that retained in the soil. That being said, it will be necessary to confirm our results and hypothesis using field-based leaching experiments to better understand the ultimate fate of leached soil carbon: whether it will be retained in the deeper soil or show a higher degradability upon leaving the soil matrix. Such information will be complementary to our study and further elucidate the importance of leaching processes in terms of ecosystem carbon budget.

The above considerations and discussions are now added as a separate paragraph in Section 3.4 in the revised paper:

"An uncertainty related to the importance of leaching processes in the overall carbon budget along the "soil-river-ocean" continuum lies in the ultimate downstream fate of the leached carbon. If part of this carbon is retained in the surrounding soils or carried along from the river to the ocean in the form of DIC without outgassing into the air, it will not constitute a source of atmospheric CO₂ on a relatively short term (over years or decades). However, soil columns used in our study has a depth (60 cm) typical of or even deeper than the average soil depth in the alpine grasslands of Qinghai-Tibetan Plateau

(Wang et al., 2001). Hence, we assume that carbon leached in our experiments will have minimum retention in the soil. Furthermore, compared to DOC and DIC in the soil solution, the leached carbon is more likely to be subject to more intensified mineralization and outgassing during the land-ocean transfer, given more intensified mixing processes, oxygen exposure and photo-oxidation of terrestrial carbon upon releasing into the river (Hedges et al., 1997; Battin et al., 2009). Hence, we postulate that carbon leached from soils is more vulnerable to decomposition and/or release compared to that retained in the soil. That being said, it will be necessary to confirm our results and hypothesis using field-based leaching experiments to better understand the ultimate fate of leached soil carbon: whether it will be retained in the deeper soil or show a higher degradability upon leaving the soil matrix. Such information will be complementary to our study and further elucidate the importance of leaching processes in terms of ecosystem carbon budget.”

References:

- Battin, T. J., Luysaert, S., Kaplan, L. A., Aufdenkampe, A. K., Richter, A., and Tranvik, L. J.: The boundless carbon cycle, *Nature Geoscience*, 2, 598-600, 2009.
- Hedges, J. I., Keil, R. G., and Benner, R.: What happens to terrestrial organic matter in the ocean?, *Org. Geochem.*, 27, 195-212, 1997.
- Wang, S.Q., Zhu, S.L., and Zhou, C.H.: Characteristics of spatial variability of soil thickness in China, *Geographical research*, 20, 161-169, 2001.

Comment 2:

When judging the relevance of dissolved inorganic carbon leaching for the carbon balance, it is also crucial to differentiate between the biogenic fraction of DIC and its lithogenic (carbonate-rock derived) fraction. In my opinion, it is much more straight forward to compare the biogenic leaching losses of DIC with the NEP, than total DIC leaching losses. The authors might want to consider this in their discussion of their results in lines 25ff on page 9. In this context the application of the isotopic mass balance model is important. The results of this model depend strongly on the delta 13C values of the end-members carbonate and CO₂ from soil respiration. The authors decided to use the delta 13C of the soil organic matter of -24 per-mille to calculate the biogenic fraction of dissolved inorganic carbon. Because isotopic fractionation occurs during the mineralization of soil organic matter, the authors might additionally use their delta 13C value of -23.1 per-mille as end member in order to assess the uncertainty that is associated with potential isotopic fractionation during mineralization and diffusive CO₂ transport in soil (Cerling et al., 1991. On the isotopic composition of carbon in soil carbon dioxide. *Geochim. Cosmochim. Acta* 55, 3403-3405). Quantitatively more important than the isotopic fractionation during mineralization and diffusion of CO₂ for the delta 13C value used as end member for the soil organic carbon derived fraction of DIC is the isotopic fractionation between CO₂ in the gas phase and bicarbonate (Zhang et al., 1995. Carbon isotope fractionation during gas-water exchange and dissolution of CO₂. *Geochim. Cosmochim. Acta* 59, 107-114). In the pH range of the investigated soils, the vast majority of the DIC will be present as bicarbonate (HCO₃⁻). According to Zhang et al. (1995), isotope fractionation between the gas phase and the aqueous phase will shift the delta 13C of bicarbonate in equilibrium with gaseous CO₂ by some 10-11 per-mille. Hence, the end member delta 13C of DIC in equilibrium with CO₂, which has a delta 13C value of -24 per-mille, can be around -14 to -13 per-mille. Considering this isotopic fractionation between gaseous CO₂ and bicarbonate will greatly increase the calculated fractions of biogenic (soil organic carbon-derived) DIC.

Response:

We must thank the reviewer for pointing out an excellent point that we overlooked. The dissolution of CO₂ produced by SOC degradation does cause large isotope fractionation on the biogenic carbonate/bicarbonate. Taking this into account, we have revised our endmember values ($\delta^{13}\text{C}_{\text{biogenic-DIC}}$ is estimated to be -16‰), and revised the following parts:

Section 2.4:

“The relative contribution of lithogenic carbonate and biogenic DIC derived from SOC degradation to leached DIC was assessed according to the following isotopic mass balance model:

$$f_{\text{carbonate}} + f_{\text{biogenic-DIC}} = 1 \quad (1)$$

$$f_{\text{carbonate}} \times \delta^{13}\text{C}_{\text{carbonate}} + f_{\text{biogenic-DIC}} \times \delta^{13}\text{C}_{\text{biogenic-DIC}} = \delta^{13}\text{C}_{\text{DIC}} \quad (2)$$

where $f_{\text{carbonate}}$ and $f_{\text{biogenic-DIC}}$ are proportion of carbonate- and biogenic DIC in total DIC; $\delta^{13}\text{C}_{\text{carbonate}}$ is the $\delta^{13}\text{C}$ value of soil carbonate, equivalent to 0‰ (Edwards and Saltzman, 2016); and $\delta^{13}\text{C}_{\text{biogenic-DIC}}$ is the $\delta^{13}\text{C}$ value of biogenic carbonate/bicarbonate derived from the dissolution of CO₂ produced by SOC degradation, which is estimated to shift by approximately 8‰ compared with the $\delta^{13}\text{C}$ value of soil-respired CO₂ (-24‰ here) due to isotope fractionation during CO₂ dissolution (Zhang et al., 1995). Hence, $\delta^{13}\text{C}_{\text{biogenic-DIC}}$ is estimated to be -16‰ . $\delta^{13}\text{C}_{\text{DIC}}$ is the measured $\delta^{13}\text{C}$ signature of leached DIC. According to Hendy (1971) and Doctor et al. (2008), isotopic fractionation of leached DIC due to CO₂ loss in an open system is insignificant when the partial pressure of CO₂ ($p\text{CO}_2$) in the solution is lower than twice that of the surrounding atmosphere. Therefore, due to the much lower $p\text{CO}_2$ in the XLHT leachates ($\sim 200 \mu\text{atm}$; Table S2) compared to that in the ambient atmosphere ($> 400 \mu\text{atm}$), the influence of CO₂ outgassing on the $\delta^{13}\text{C}$ of leached DIC was not considered in the present study.”

Section 3.3:

“Based on the isotopic mass balance Eq. (1) and (2), lithogenic carbonate (with a $\delta^{13}\text{C}$ value of 0‰) contributed 51.2% to the leached DIC while biogenic DIC produced by SOC degradation contributed 48.4% (Fig. 5). The $\delta^{13}\text{C}$ value of leached DIC decreased to -12.3‰ and -13.5‰ during the second and third EPEs, corresponding to a contribution of 77.0% and 84.4% by biogenic sources in the total DIC, respectively (Fig. 5). These results confirm our previous hypothesis that SOC decomposition contributed significantly to soil DIC fluxes. Combined with the total flux rate, we calculated that both lithogenic and biogenic DIC fluxes were $\sim 2.1 \text{ g C m}^{-2}$ in the first EPE. Subsequently, lithogenic DIC flux decreased to $\sim 1.3 \text{ g C m}^{-2}$ while biogenic DIC flux increased to 7.6 g C m^{-2} in the third EPE. This demonstrates that increased SOC degradation mainly contributed to the increased DIC fluxes with repeated EPEs.”

Section 3.4:

“It is worth mentioning that biogenic DIC loss ($16.0 \pm 3.4 \text{ g C m}^{-2}$) caused by SOC degradation accounted for 184% of NEP at XLHT, indicating the importance of biogenic DIC to SIC loss during EPEs.”

Reference:

Zhang, J., Quay, P. D., and Wilbur, D. O.: Carbon isotope fractionation during gas-water exchange and dissolution of CO₂, *Geochim. Cosmochim. Acta*, 59, 107-114, [http://dx.doi.org/10.1016/0016-7037\(95\)91550-D](http://dx.doi.org/10.1016/0016-7037(95)91550-D), 1995.

Specific Comments:

Comment 3:

On page 3, line 19, the soil is classified as “chestnut soil”. This classification is not in line with the international soil classification system of the World Reference Base for Soil Resources (WRB, 2015). Please classify your soils also according to the WRB system (<http://www.fao.org/3/a-i3794e.pdf>).

Response:

Revised. Soils in Xilinhot and Keqi are classified as Calcic Chernozems according to the World Reference Base for Soil Resources (Steffens et al., 2008; IUSS working group WRB, 2015) while soils in Gangcha are mainly Gelic Cambisol (IUSS working group WRB, 2015).

Comment 4:

On page 6, lines 20ff, the authors argue that the variation in SIC contents between sites are caused by the variation of pH values, suggesting a causality between pH (independent variable) and SIC content (dependent variable). The question is, whether the pH is really controlling SIC contents or vice versa. . .

Response:

SIC content is related to parent materials as well as soil pH. However, in the present study regions, soil pH is the key factor controlling SIC variation across the Mongolian and Tibetan grasslands according to the results of Shi et al. (2012). We have added one sentence to clarify this:

“This dependence of SIC content on soil pH is consistent with the results of Shi et al. (2012), showing that pH is the most important factor controlling SIC variation across the Mongolian and Tibetan grasslands.”

Comment 5:

On page 9, lines 29ff, the authors cite Kindler et al. (2011) for numbers of DIC leaching losses equaling 12% of NEP and DOC leaching losses equaling 2% of NEP. I do not understand how the authors extracted these numbers from the Kindler et al. (2011) publication.

Response:

This is clarified now in the revised paper:

According to the report of Kirschbaum et al. (2001), both Net Ecosystem Exchange (NEE) and NEP refer to net primary production minus carbon loss from heterotrophic respiration, R_h :

$$NEE = NEP = NPP - R_h = GPP - R_{\text{autotrophic respiration}} - R_h$$

These terms are used somewhat interchangeably, with NEE used more often when they are addressed based on measurements of gas exchange rates using atmospheric measurements over the time scales of hours, whereas NEP is more often used if measurements are based on ecosystem-carbon stock changes, typically measured over a minimal period of one year.

The NEE in Kindler et al. (2011) equals to gross primary productivity minus ecosystem respiration, excluding C deprivation with harvest, fires, etc. Therefore, we extracted NEE in Table 5 of Kindler et al. (2011) as NEP for calculating the proportion of leaching DOC and DIC in NEP. Relevant description has been added in Section 3.4 as follows:

“Nonetheless, the EPE-induced soil carbon loss relative to NEP was higher in this study than that

estimated for grassland topsoil across Europe (12% for DIC loss, 2% for DOC loss; Kindler et al., 2011) where Net Ecosystem Exchange (NEE) reported by Kindler et al. was used as NEP according to the report of Kirschbaum et al. (2001)."

Comment 6:

Starting on page 9, line 31, the authors argue that the carbon loss due to extreme precipitation events was much greater than carbon losses through warming-enhanced respiration. This comparison is perhaps misleading, because it implies that extreme precipitation events occur only as consequence of climate change. More correct would be the comparison of carbon losses due to warming-enhanced respiration with carbon losses due to "climate change-enhanced" extreme precipitation events.

Response:

The reviewer raised a good point. It is not fair to compare carbon leaching through all annual EPEs with warming-induced respiration increase. To be more consistent and robust, we decide to delete the discussion on the comparison of carbon losses due to warming-induced respiration with carbon losses due to annual EPEs.