Response to Editor and Reviewers

Dear Professor Shen,

Thank you for the thoughtful review of our manuscript (No. bg-2022-217) and the opportunity to submit a revision. We also greatly appreciate the very helpful review from all reviewers. We have revised the manuscript, taking into account all the comments we got.

All changes have also been highlighted in yellow in the revised version of the manuscript and the Supplementary Information. We hope the revised version of the manuscript will be acceptable for publication in the journal Biogeosciences. We look forward to hearing from you.

Thank you very much for your kind consideration.

Sincerely,
Lishan Ran
On behalf of all the authors

Response to Anonymous Referee #1

- There are some contradicting statements of how DOM chemistry change with elevation between lines 59-62 and lines 66-69. Do you use these examples to show that there are differing results in past studies?

Our response: Thanks. We have revised the first part (lines 59-62) in the revised manuscript to avoid misleading: "*A recent global study on lakes and rivers found that increasing elevation is associated with greater protein-like fluorescent DOM and lower specific ultraviolet absorbance at 254 nm ($SUVA_{254}$), which indicates the effect of enhanced UV radiation and accumulation of autochthonous DOM in higher elevation areas (Zhou et al., 2018)*." (P2 Line 62-64)

- Line 86: There are other papers you can cite here as well on DOM lability in agricultural streams and under high nutrient loads

Our response: We have added another paper here as you suggested: "*In addition, DOM tends to have a more reduced redox state and is likely more labile and accessible to the microbial community in agricultural streams when compared to the DOM found in natural streams (Fasching et al., 2019; Williams et al., 2010)*." (P3 Line 87-89)

- Line 417: I'm guessing this should say DOM characteristics

Our response: Thanks. We have replaced it with "*DOM characteristics" in the revised manuscript: "Anthropogenic impacts on DOM characteristics and age have been widely proposed in the last two decades (Butman et al., 2014; Coble et al., 2022; Vidon et al., 2008; Zhou et al., 2021)*." (P18 Line 423)

Response to Anonymous Referee #4

This manuscript describes relationships between riverine organic carbon concentration and quality in three mountainous catchments. The authors highlight the importance of catchment slope and anthropogenic land use as important factors affecting these organic carbon properties, and cite the literature extensively to support their hypotheses. There is a lot of interesting data collected and summarized, which is useful for comparisons with mountain stream and river ecosystems globally.

Our response: Thank you very much for your valuable opinions and suggestions. Details are provided in the specific response below.

Three major issues with the manuscript in its current form:

1) Even after several comments from previous reviewers, there remains some concerns about the methods employed and a lack of description in some cases. Additionally, with such multivariate data, it is concerning that the authors rely solely on univariate statistics. The complicated and sometimes overwhelming interpretation of linear regressions could be simplified with multivariate approaches.

Our response: Thanks. We have performed a stepwise multiple linear regression (MLR) modeling and the partial least squares path model (PLS-PM) as multivariate approaches to simplify the previous interpretation of linear regressions. Details are provided in the specific response below and the revised manuscript.

Some specific issues in the Methods:

Line 131: "Annual air temperature…[was] determined using ArcGIS." Perhaps this is in reference to a specific data layer, but as written it is unclear.

Our response: Thanks. We have showed the related information in the manuscript that the data on annual air temperature were obtained from a cloud platform: "*Data on land use types and air temperature in 2015, as well as a 90 m digital elevation model (Shuttle Radar Topography Mission, SRTM) were obtained from the Resource and Environment Data Cloud Platform of the Chinese Academy of Sciences (http://www.resdc.cn/).*" (P4 Line 112-114)

Line 136: Description of carbonate rocks suggests weathering rates could be high, but the impact of this on carbon ages is not thoroughly discussed.

Our response: Thanks. Yes, we have discussed the role of carbonate rocks in controlling chemical weathering and carbon isotopes of DIC in previous manuscript: "As discussed above, lithology is the underlying factor controlling the spatial distribution of DIC in these rivers (Fig. 3a). However, the $\delta^{13}C_{DIC}$ and $\Delta^{14}C_{DIC}$ did not show significant variations with increasing carbonate weathering intensity (Fig. 3b), implying that riverine carbon isotopes of DIC could be influenced by multiple biogeochemical processes. (please see (Chen et al., 2021))". This study was mainly focused on DOM and DOC dynamics. Thus, we did not include the above discussion in the revised manuscript. Further details are provided in (Chen et al., 2021).

Line 139: What is the spatial distribution of soil organic carbon or soil depth? No data on soil properties is described despite likely being the most important source of organic carbon to the streams.

Our response: Thanks. We have added spatial distribution of soil organic carbon as you suggested in the revised manuscript: "*The topsoil SOC exhibited a spatial distribution that resembled elevation, as areas with greater elevation displayed higher SOC content (Fig. S2).*" (P5 Line 128-129) We also added the figure in the Supplement, as shown below. Additionally, the role of SOC in controlling DOC dynamics was considered in the revised manuscript. For instance, SOC was included in the MLR model and the PLS-PM model, as shown in Table 4 and Figure 6 shown below.

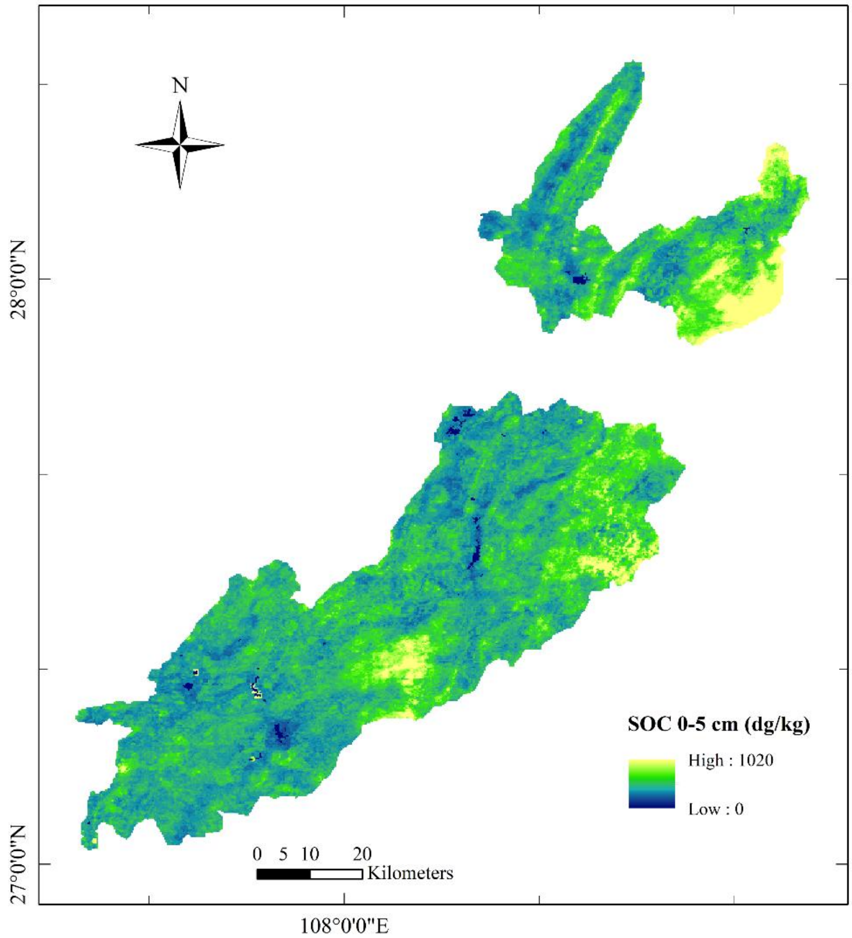


Figure S2 Spatial distribution in SOC content in the surface layer (0–5 cm).

Table 4. Multiple stepwise linear regression models of catchment attributes and water chemistry on DOC concentrations and DOM properties.

Dependent variables	Predictors	Model equation	*n*	Adj R^2	Significance level
DOC[a]	slope, NH_4^+-N	= –0.109*slope + 4.295*NH_4^+-N+ 3.375	28	0.50	$p < 0.001$
DOC	SOC, POC	= –0.006*SOC + 0.384*POC + 4.145	28	0.59	$p < 0.001$
$SUVA_{254}$	urban and agricultural land use, slope	= –5.461*urban and agricultural land use + 0.145*slope+1.318	26	0.77	$p < 0.001$
HIX	urban and agricultural land use	= 0.433*urban and agricultural land use + 0.438	27	0.34	$p < 0.001$
FI		No variables were entered into the equation.	27		
β/α	pH	= –0.195*pH + 2.476	27	0.41	$p < 0.001$
C1	DO, TP, urban and agricultural land use	= 7.713*DO – 220.846*TP +90.905*urban and agricultural land use – 36.005	27	0.46	$p < 0.001$
C2	urban and agricultural land use, DO	= –48.748*urban and agricultural land use – 2.515*DO + 58.255	27	0.36	$p = 0.002$
C3	NO_3^--N, POC	= 4.181*NO_3^--N + 3.738*POC + 3.826	27	0.34	$p = 0.003$

[a] SOC was not included as predictors in this model to examine the impacts of human activities and geomorphology, rather than the direct influence of SOC on DOC concentrations.

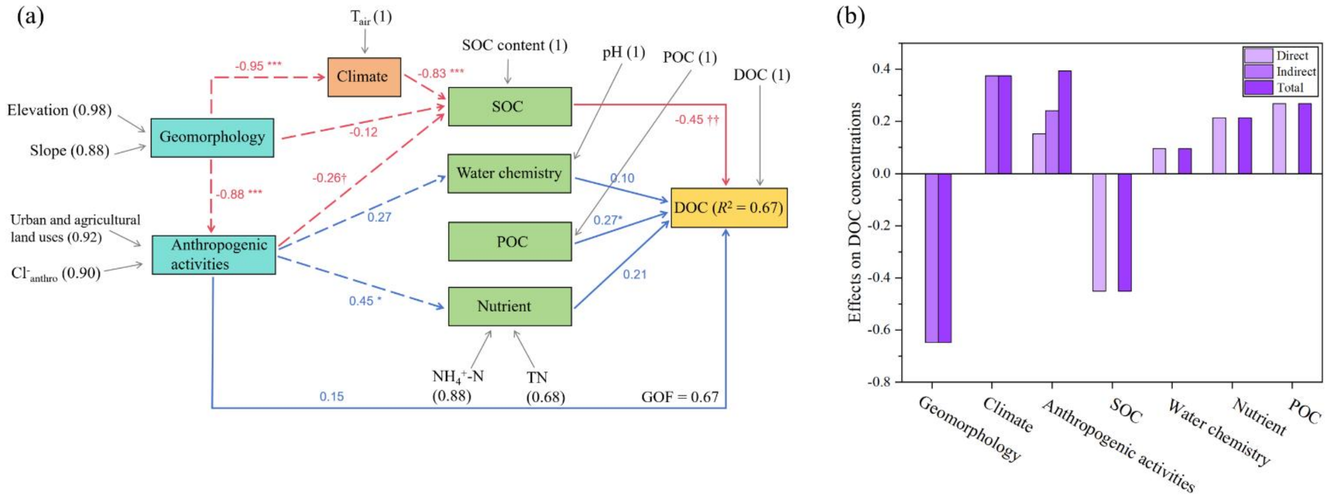


Figure 6. The most parsimonious PLS-PM model showing the direct and indirect effects of geomorphology and anthropogenic activities on DOC concentrations. (a) Path coefficients are shown as arrows with blue and red to represent positive and negative effects, respectively. The solid and dotted lines indicate the direct and indirect influence pathways of environmental drivers on DOC concentrations, respectively. The indicators (e.g., TN) of latent variables (e.g., nutrient) are shown at the beginning of the grey arrows. The numbers in the parentheses are the loading scores. GOF denotes the goodness of fit of the entire model. R^2 indicates the amount of variance in DOC concentrations explained by its independent latent variables. The standardized path coefficients that are significantly different from zero are indicated by *p = < 0.05, ** p = < 0.01, *** p = < 0.001, † p = 0.06, †† p = 0.07. (b) Standardized direct and indirect mean effects of environmental drivers on DOC concentrations derived from the PLS-PM analysis.

Line 147: Runoff units would be helpful, either instead or additionally.

Our response: Thanks. We have added runoff in the revised manuscript as an additional information: "*This study area is highly affected by monsoon-influenced humid subtropical climate with April to October being the rainy season, and the average annual precipitation, runoff, and discharge are 1100 mm, 1004 mm/yr and 14.4 m³/s, respectively, in the Yinjiang River catchment.*" (P6 Line 141-143)

Line 152: Time of year of sampling is likely very important, but is not discussed later.

Our response: Thanks. Our sampling time (September 2018) is during the wet season (April–September). We have added it in the discussion as you suggested: "*Moreover, the groundwater contribution was probably much less significant in the wet season (e.g., September in the study area), even in catchments where DOC is mainly derived from groundwater (Lloret et al., 2016)*." (P17 Line 393-394)

Line 161: How long were samples stored?

Our response: Thanks. The period for sample storage was previously reported in 2.3 Laboratory analysis as "Refrigerated water samples for DOM absorbance and fluorescence were analyzed within one week after sampling." We now add this information earlier in the revised manuscript: "The filtered water was stored in a Milli-Q water and sampling water pre-washed brand-new low-density polyethylene container at low temperature (4°C) in the dark within one week before optical properties analysis and acidified by phosphoric acid to pH = 2 for DOC analysis." (P6 Line 156-158)

Line 170: Is this true for all samples?

Our response: Thanks. Yes, it is true. The normalized inorganic charge balance ranged between 0.02 % to 4.52 % for all samples. (P Line)

Line 172: Relative deviation of what? Did you take replicate field samples? Lab samples? This a concern for several analyses, i.e., if duplicates were taken, and if so, what kind of duplicate was measured.

Our response: Thanks. Here, relative deviation means the relative deviation of lab samples. Replicate field samples were taken for several analyses (e.g., DOC, stable and radiocarbon isotopes) and measured when necessary.

Line 178: Which 9 samples? Is that all of the samples from the Yinjiang River? If not, why were they chosen?

Our response: Thanks. Yes, all of the ^{14}C samples are from the Yinjiang River. "*The Yinjiang River catchment has the greatest change in geomorphologic characteristics (i.e., elevation and channel slope) and the highest proportion of agricultural and urban land uses among the three catchments*" is one of the reasons we only collect 14C-DOC data in the Yinjiang River. The other reason is the expensive analytical cost (McNichol and Aluwihare, 2007), which is about $500 per sample.

Reference: *McNichol A. P. and Aluwihare, L. I.: The power of radiocarbon in biogeochemical studies of the marine carbon cycle:Insights from studies of dissolved and particulate organic carbon (DOC and POC), Chem. rev., 107, 443-466, doi:10.1002/chin.200724246, 2007.*

Line 186: At what interval?

Our response: Thanks. We have added this information in the revised manuscript as: "*DOM absorbance of river water samples was measured from 250 to 750 nm at 1 nm intervals using a UV (ultraviolet)-visible spectrophotometer (UV-2700, Shimadzu) with a 1 cm quartz cuvette.*" (P7 Line 181-183)

Line 196: Is there any concern about DOM properties changing in 1 week?

Our response: Thanks. DOM absorbance and fluorescence analyzed within one week was a suggested period for DOM optical measurement (Coble et al., 2014).

Reference: *Coble P. G., Lead, J., Baker, A., Reynolds, D. M. and Spencer, R. G.: Aquatic organic matter fluorescence, Cambridge University Press, 2014.*

Line 215: If you are doing all of these correlations, how do you determine what is "predominant?" Anything significant? Do you have concerns about doing so many univariate analyses?

Our response: Thanks. Following your suggestions above, we have performed stepwise multiple linear regression (MLR) modeling and the partial least squares path model (PLS-PM) as multivariate approaches to identify the dominant drivers of DOC dynamics. Details are provided in "2.4. Statistical analysis", "Results" and "Discussion" of the revised manuscript.

Line 220: A mean is not necessarily representative if you are working with small sample sizes and non-normal data.

Our response: Thanks. We have compared the mean, median and their differences (i.e., (Mean-Median)/Mean) of the major dataset we reported in the table below. The results showed that the differences are mostly lower than 10%, which indicated the mean is also an appropriate value to represent our data. In addition, for the data we reported mean value in the manuscript, we also provided box plots (e.g., Figs. 2 and 3) for these data, which is useful for the reader to have a better understanding of the distribution of data.

	DOC	$\delta^{13}C_{DOC}$	$\Delta^{14}C_{DOC}$	$SUVA_{254}$	FI	HIX	β/α	Cl	NH4-N	NO3-N	pH
Mean	1.5	-26.8	-54.7	3.2	1.78	0.55	0.78	3.1	0.057	1.1	8.29
Median	1.33	-25.7	-52.2	3.0	1.76	0.59	0.74	3.3	0.06	0.83	8.21
(Mean-Median)/Mean	11.3	4.2	4.6	6.8	1.1	-7.3	5.1	-5.1	-5.3	24.5	1.0

Line 221: Sometimes removing and sometimes including a data point seems difficult to defend without a specific metric.

Our response: Thanks. The reason why we did not remove Y12 in the whole study was because radiocarbon isotopes of the Y12 were measured, and these data can provide additional information on carbon dynamics under the influence of heavy rainfall events. In addition, we only exclude Y12 in Figures 4 and S4, where we add details on the data selection in figure captions. So the reader can easily distinguish the impact of an outlier on the research results and the reason why we did not include outlier in the result analysis.

Line 302: Anthropogenically derived chloride is not defined in the text.

Our response: Thanks. Anthropogenically derived chloride was discussed in the caption of Fig. 5 in the previous manuscript. We have moved it earlier in the revised manuscript as: "The environmental factors used in the model were categorized into seven latent variables, including geomorphology (elevation and slope), anthropogenic activities (e.g., urban and agricultural land uses and anthropogenically derived Cl^- (Cl^-_{anthro}, calculated as the total Cl^- concentration minus atmospheric contributed Cl^- concentration, which is the lowest Cl^- concentration at site Y5 in the Yinjiang River; Gaillardet et al., 1997; Meybeck, 1983))". (P Line)

2) The Results are exhaustive, and as a result, quite unfocused. It is unclear which results are important to the central message of the manuscript. This is indicative of a lack of clear focus within the entire manuscript. As previous reviews have pointed out, there are a lot of figures and tables. I would agree and suggest that most of these figures are relatively simple and do little else than to show correlation. Some higher-level analysis or results would be greatly beneficial, including some multivariate approaches.

Our response: Thanks. We have deleted many figures to avoid exhaustive, especially those figures showing the regression correlation. For example, previous Figs. 4-7 were removed in the revised manuscript. Then, we applied multivariate approaches in this study following your suggestions, such as stepwise multiple linear regression (MLR) modeling and the partial least squares path model (PLS-PM). Details are provided in the "2.4. Statistical analysis" of the revised manuscript: "*We performed a stepwise multiple linear regression (MLR) modeling to identify significant environmental factors of DOC concentrations and DOM properties using SPSS 26. All environmental factors were included in the models except for SOC, because we aim at examining the impacts of human activities and geomorphology rather than the direct influence of SOC on DOC concentrations and DOM properties. The objective model with the highest adjusted R^2 value was used to infer the DOC concentrations and DOM properties. In addition to the MLR and Pearson correlation analyses to explore the relationships between environmental factors and DOC, we further performed the partial least squares path model (PLS-PM) to infer direct and indirect effects of multiple factors (e.g., geomorphologic and anthropogenic impacts) on DOC concentrations. The PLS-PM analysis was performed using the R package "plspm" (Sanchez, 2013). Because PLS-PM offers the advantage of not imposing any distributional assumptions on the data, which enhances its broad applicability (Sanchez, 2013), and allows for the exploration of complex cause-effect relationships involving latent variables, it is a suitable technique for multivariate analyses . Each latent variable consists of one or more manifest variables (e.g., geomorphology, including elevation and slope). The environmental factors used in the model were categorized into seven latent variables, including geomorphology (elevation and slope), anthropogenic activities (e.g., urban and agricultural land uses and anthropogenically derived Cl^- (Cl^-_{anthro}, calculated as the total Cl^- concentration minus atmospheric contributed Cl^- concentration, which is the lowest Cl^- concentration at site Y5 in the Yinjiang River; (Gaillardet et al., 1997; Meybeck, 1983))), climate (T_{air}), SOC (SOC content), water chemistry (pH), POC (POC concentrations) and nutrient (NH_4^+-N and TN). The environmental factors and their manifest variables included in the model were the most critical variables identified based on the Pearson correlation results. These variables were selected after reducing the full models (initial models with more variables) to meet the requirements of the PLS-PM analysis (Du et al., 2023; Sanchez, 2013; Tian et al., 2019). In addition, the structure of the model was simplified to focus on the major effect of environmental factors on DOC concentrations rather than to explore the effects on other factors (e.g., the geomorphologic controls on POC were ignored). The significance of the path coefficients was determined through a nonparametric bootstrap resampling of 1000 times.*" (P8-9 Line 216-239)

Further results of the MLR and PLS-PM are shown in Table 4 and Figs. 6, S4, and S5. Please see the revised manuscript for more details.

Table 4. Multiple stepwise linear regression models of catchment attributes and water chemistry on DOC concentrations and DOM properties.

Dependent variables	Predictors	Model equation	n	Adj R^2	Significance level
DOC[a]	slope, NH_4^+-N	= –0.109*slope + 4.295*NH_4^+-N+ 3.375	28	0.50	$p < 0.001$
DOC	SOC, POC	= –0.006*SOC + 0.384*POC + 4.145	28	0.59	$p < 0.001$
$SUVA_{254}$	urban and agricultural land use, slope	= –5.461*urban and agricultural land use + 0.145*slope+1.318	26	0.77	$p < 0.001$
HIX	urban and agricultural land use	= 0.433*urban and agricultural land use + 0.438	27	0.34	$p < 0.001$
FI		No variables were entered into the equation.	27		
β/α	pH	= –0.195*pH + 2.476	27	0.41	$p < 0.001$
C1	DO, TP, urban and agricultural land use	= 7.713*DO – 220.846*TP +90.905*urban and agricultural land use – 36.005	27	0.46	$p < 0.001$
C2	urban and agricultural land use, DO	= –48.748*urban and agricultural land use – 2.515*DO + 58.255	27	0.36	$p = 0.002$
C3	NO_3^--N, POC	= 4.181*NO_3^--N + 3.738*POC + 3.826	27	0.34	$p = 0.003$

[a] SOC was not included as predictors in this model to examine the impacts of human activities and geomorphology, rather than the direct influence of SOC on DOC concentrations.

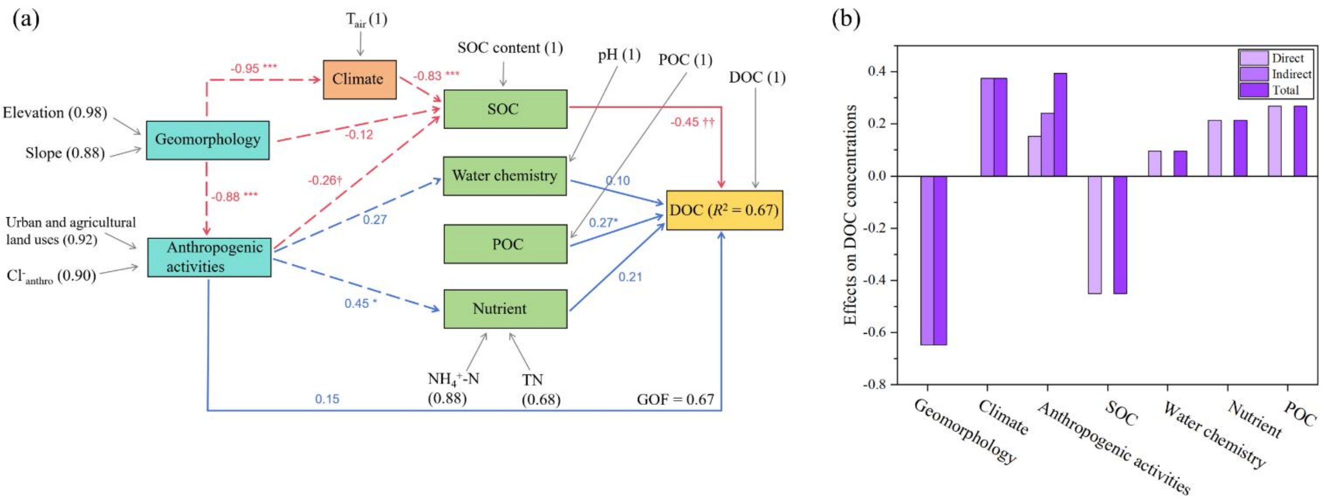


Figure 6. The most parsimonious PLS-PM model showing the direct and indirect effects of geomorphology and anthropogenic activities on DOC concentrations. (a) Path coefficients are shown as arrows with blue and red to represent positive and negative effects, respectively. The solid and dotted lines indicate the direct and indirect influence pathways of environmental drivers on DOC concentrations, respectively. The indicators (e.g., TN) of latent variables (e.g., nutrient) are shown at the beginning of the grey arrows. The numbers in the parentheses are the loading scores. GOF denotes the goodness of fit of the entire model. R^2 indicates the amount of variance in DOC concentrations explained by its independent latent variables. The standardized path coefficients that are significantly different from zero are indicated by *p = < 0.05, ** p = < 0.01, *** p = < 0.001, † p = 0.06, †† p = 0.07. (b) Standardized direct and indirect mean effects of environmental drivers on DOC concentrations derived from the PLS-PM analysis.

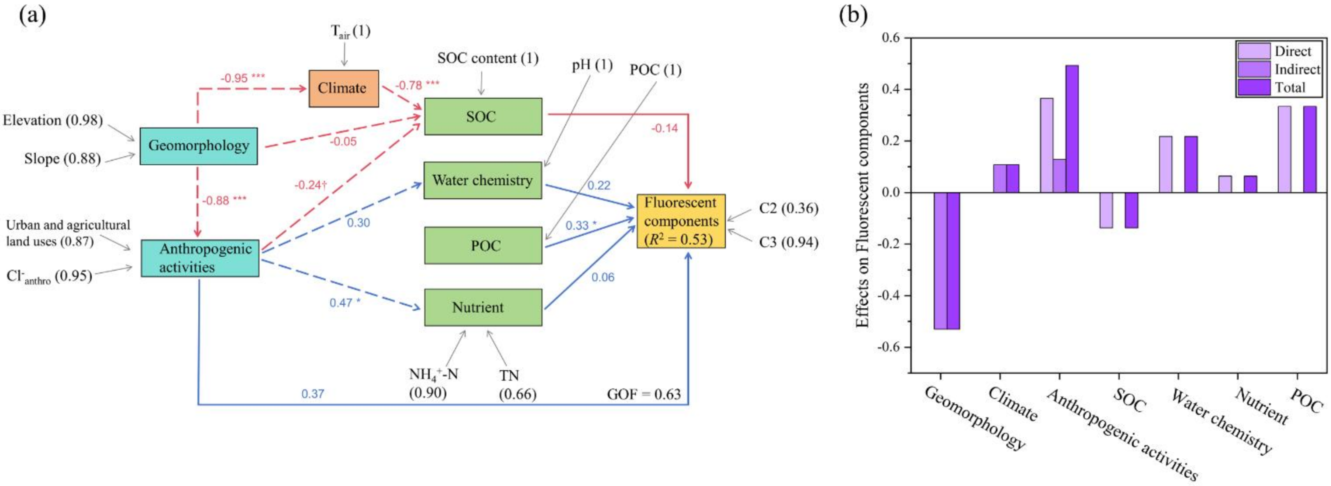


Figure S4 The most parsimonious PLS-PM showing the direct and indirect effects of geomorphology and anthropogenic activities on fluorescent components. (a) Path coefficients are shown as arrows with blue and red to represent positive and negative effects, respectively. The solid and dotted lines indicate the direct and indirect influence pathways of environmental drivers on fluorescent components, respectively. The indicators (e.g., TN) of latent variables (e.g., nutrient) are shown at the beginning of the grey arrows. The numbers in the parentheses are the loading scores. GOF denotes the goodness of fit of the entire model. R^2 indicates the amount of variance in fluorescent components explained by its independent latent variables. The standardized path coefficients that are significantly different from zero are indicated by *p = < 0.05, ** p = < 0.01, *** p = < 0.001. (b) Standardized direct and indirect mean effects of environmental drivers on fluorescent components derived from the PLS-PM analysis. C1 was initially included in the model but had to be removed to fulfill the requirements of the model analysis.

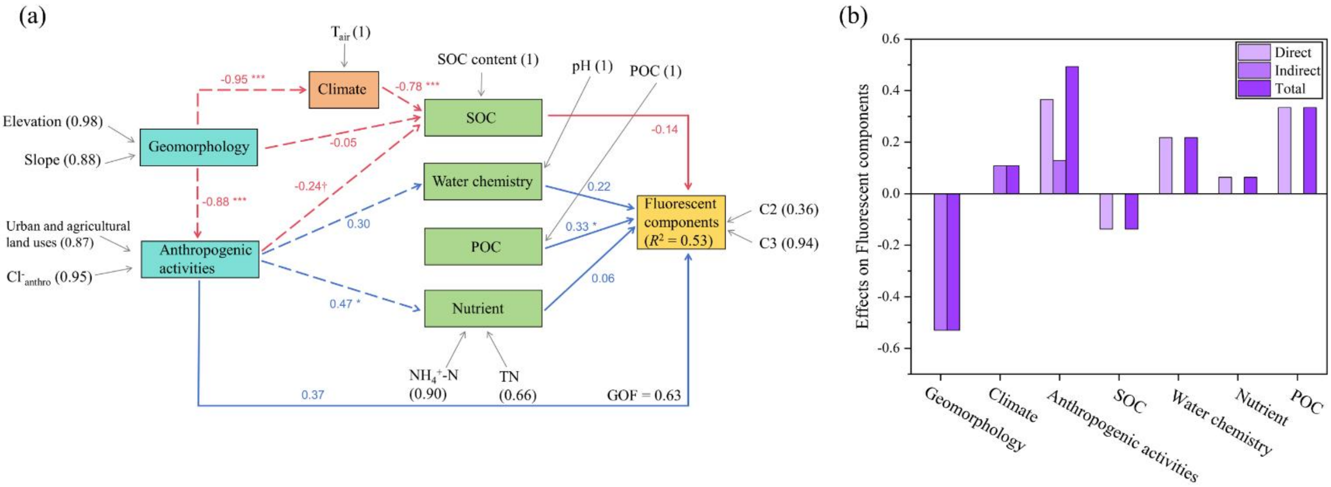


Figure S5 The most parsimonious PLS-PM showing the direct and indirect effects of geomorphology and anthropogenic activities on DOM optical parameters. (a) Path coefficients are shown as arrows with blue and red to represent positive and negative effects, respectively. The solid and dotted lines indicate the direct and indirect influence pathways of environmental drivers on DOM optical parameters, respectively. The indicators (e.g., TN) of latent variables (e.g., nutrient) are shown at the beginning of the grey arrows. The numbers in the parentheses are the loading scores. GOF denotes the goodness of fit of the entire model. R^2 indicates the amount of variance in DOM optical parameters explained by its independent latent variables. The standardized path coefficients that are significantly different from zero are indicated by *p = < 0.05, ** p = < 0.01, *** p = < 0.001. (b) Standardized direct and indirect mean effects of environmental drivers on DOM optical parameters derived from the PLS-PM analysis. FI was initially included in the model but had to be removed to meet the requirements of the model analysis.

3) The Discussion section is written in a way that does not clearly differentiate what are interpretations of this study and what are supporting ideas from other studies. Perhaps this is because the interpretation is complex, but the authors do not clearly establish what are their main conclusions or interpretations. Line 474 is telling, "disentangling the dual influences…is challenging." The results do not accomplish this, the discussion does not do much to clarify either. But the analysis is perhaps what is most limiting, in which univariate statistics are repeatedly used to approach a complex and multivariate problem. The repeated interpretation of correlation as causation is also problematic. Additionally, while the authors mention how land use and slope are not independent variables towards the end of the discussion, this should be a major caveat and approached more thoroughly early in the manuscript.

Our response: Thanks. We have restructured the discussion by removing many paragraphs/sentences that are not so clear or insignificant based on our new analysis. As showed in the last question, we have performed a stepwise multiple linear regression (MLR) modeling and the partial least squares path model (PLS-PM) to deal with the complex and multivariate problem. Using these tools, we made significant changes to the manuscript, especially the results and discussion. We believe that our new results and discussion are clear enough to demonstrate the dual influences of geomorphologic characteristics and anthropogenic activities on DOM. For example, PLS-PM analysis has clearly indicated the direct and indirect effects of environmental factors on DOC concentrations: "*The PLS-PM analysis showed that 67% of the variance in DOC concentrations could be explained by our constructed seven environmental factors (R^2 = 0.67, Fig. 6a). The total effect on DOC concentrations is strongest from geomorphology (-0.65), followed by SOC (-0.45), anthropogenic activities (0.39), climate (0.38), POC (0.27), nutrient (0.21), and water chemistry (0.10) (Fig. 6b). The results indicated that geomorphology was the most significant factor in controlling DOC concentrations, primarily through indirect regulation on SOC content, which was directly influenced by annual catchment temperature and anthropogenic activities (Figs. 6a and b). In comparison, anthropogenic activities not only indirectly regulated riverine DOC concentrations through SOC, but also had a significant indirect impact through the regulation of nutrient levels. Similar to DOC concentrations, geomorphology (-0.53) exhibited the most pronounced effects on fluorescent components (Fig. S4). However, anthropogenic activities (0.49) demonstrated a comparable effect on fluorescent components, primarily through a direct pathway (0.37; Fig. S4b). Anthropogenic activities (-0.84) were the strongest driver for DOM optical parameters, although geomorphology (0.59) played a significant role in indirectly influencing DOM optical parameters (Fig. S5).*" The PLS-PM analysis also clearly shows us the correlation between land use and slope (Fig. 6). (P14 Line 332-342)

Response to Anonymous Referee #5

The manuscript here explores how geographical and anthropogenic factors impact DOM abundance and composition. This topic is important in global carbon cycling, especially under the scenario of climate change and intensified anthropogenic activities.

Our response: Thank you for your very helpful review of our manuscript once again and for giving us very useful comments. We have already depleted some of the unnecessary figure panels and previously published data in the modified manuscript, such as Figs. 1 and 4. We also streamlined the discussion text, such as the discussion on groundwater contribution on riverine DOC. Details are provided in the specific response below.

However, here are some specific comments which should be addressed before acceptance.

Specific comments:

L56-57 The authors should check this sentence for grammar and logic issues: "Recent studies have shown that geographical (e.g., elevation and catchment slope) controls on DOC export may also be important for riverine carbon cycling (Connolly et al., 2018; Li Yung Lung et al., 2018).", and also the reference format here "Li Yung Lung et al., 2018".

Our response: Thanks. We have rewritten this sentence as: "*Recent studies have indicated the significance of geomorphologic factors, such as elevation and catchment slope, in influencing the export of DOC and riverine carbon cycling (Connolly et al., 2018; Li Yung Lung et al., 2018)*." The reference format here, "Li Yung Lung et al., 2018" is correct as the full name of the first author is "Joanna Y. S. Li Yung Lung" (P2 Line 58-59). The official citation is "Li Yung Lung, J. Y. S., Tank, S. E., Spence, C., Yang, D., Bonsal, B., McClelland, J. W., & Holmes, R. M. (2018). Seasonal and geographic variation in dissolved carbon biogeochemistry of rivers draining to the Canadian Arctic Ocean and Hudson Bay. Journal of Geophysical Research: Biogeosciences, 123, 3371–3386. https://doi.org/10.1029/2018JG004659"

L59: "characterized by greater releases of DOC", what does this mean? "Release of DOC", from where or to where? "Greater releases", the quantity or the flux or the rate?

Our response: Thanks. We have rewritten it to make it clear: "*Compared with high-relief catchments, low-relief regions with longer water residence time, stronger hydrologic connectivity to rivers, and greater development of wetlands are typically characterized by increased concentration of riverine DOC*" (P2 Line 59-62)

L60: "protein-like fluorescence DOM" should be "protein-like fluorescent DOM".

Our response: Thanks. We have replaced it with "*protein-like fluorescent DOM*". (P3 Line 63)

L134: The authors should check the value of the slopes ("mean catchment slope (from 14.3° to 25.5°)". Is the magnitude right? Not one order less? I'm not familiar with this, however, it should be one order less considering the spatial scale (30-60 km) and the altitude difference (about 2 km).

Our response: Thanks for your concerns. We have checked the degree of slope. The study area is mountainous area, which leads to the high catchment slope. This magnitude is reasonable, as evidenced by other studies (e.g., (Harms et al., 2016)).

L189-190: The authors should check the definition of a254 and A254, and also the unit of absorption coefficients. The definition and calculation of absorbance (A254) and the absorption coefficient (a254) could refer Hu et al. (2002) and Li et al. (2017). Meanwhile, commonly the unit of a254 is m-1, not cm-1.

Our response: Thanks. We have checked the definition of a254 and A254 as you suggested and changed the unit of a254 to m^{-1}. (P7 Line 186)

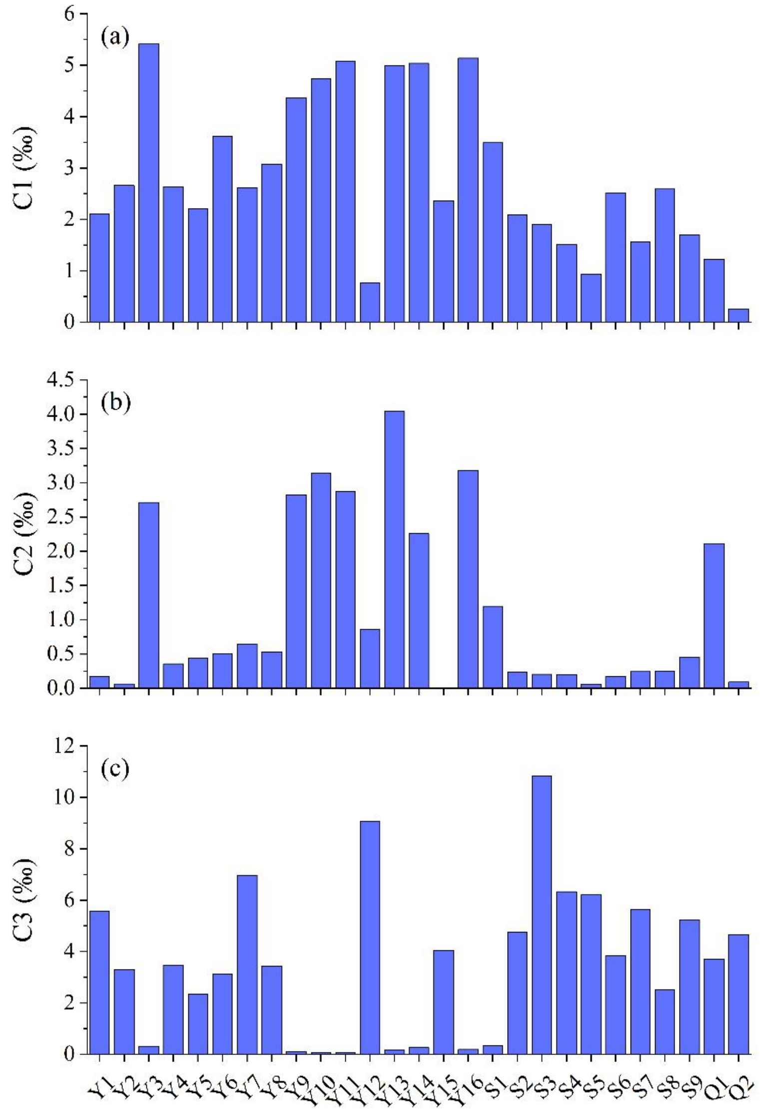
Ref: Hu C, Muller-Karger F E, Zepp R G. Absorbance, absorption coefficient, and apparent quantum yield: A comment on common ambiguity in the use of these optical concepts[J]. Limnology and Oceanography, 2002, 47(4): 1261-1267.

Li P, Hur J. Utilization of UV-Vis spectroscopy and related data analyses for dissolved organic matter (DOM) studies: A

review[J]. Critical Reviews in Environmental Science and Technology, 2017, 47(3): 131-154.

L194-206: Have the authors considered the inner filter correction for fluorescence data?

Our response: Thanks. As we discussed earlier with another reviewer, we did not perform inner-filter correction because the inner filter effects could be ignored as the absorbance at 254 nm was lower than 0.3 m-1 (Ohno, 2002). According to Ohno (2002), "An exact correction using explicit correction factors for both primary and secondary inner filtration effects was shown to give humification index values that are concentration invariant when absorbance of the solution at 254 nm was less than approximately 0.3 unit." All the absorbance at 254 nm of the water samples in this study was lower than 0.3 m-1. We also tried to perform innerfilter correction to see the differences if inner-filter effect was not corrected. The differences (Absolute value of (uncorrected value - corrected value)/ corrected value *1000‰) were shown in the figure below. Most of the differences are lower than 10‰, indicating inner filtration effects can be ignored in this study.



Figures show the differences (Absolute value of (uncorrected value - corrected value)/ corrected value *1000‰) between inner-filter effect uncorrected and corrected values.

Reference:

Ohno T.: Fluorescence inner-filtering correction for determining the humification index of dissolved organic matter, Environ. Sci. Technol., 36, 742-746, doi:10.1021/es0155276, 2002.

L225: The authors should check the definition and calculation of SUVA254 in Table 2. In Weishaar et al. (2003), they use A254, not a254.

Our response: Thanks. According to (Weishaar et al., 2003) et al. (2003), "SUVA254 is defined as the UV absorbance at 254 nanometers measured in inverse meters (m^{-1}) divided by the DOC concentration measured in milligrams per liter (mg

L^{-1}).” In application, $SUVA_{254}$ was calculated by dividing the decadic absorption coefficient at 254 nm by DOC concentration (Luzius et al., 2018; Poulin et al., 2014; Weishaar et al., 2003). Thus, the $SUVA_{254}$ we provided in this study was correct.

L225: Will the authors consider the updated calculation and interpretation of fluorescence index (FI)? FI = Em470/Em520, at Ex 370 nm. The authors could refer to the updated reference (Cory R M, Miller M P, McKnight D M, et al. Effect of instrument-specific response on the analysis of fulvic acid fluorescence spectra[J]. Limnology and Oceanography: Methods, 2010, 8(2): 67-78.).

Our response: Thanks for your suggestions. I would like to update the calculation of fluorescence index as you suggested. However, we measured the emission wavelength from 280 to 500 nm at 2 nm increments, which makes the update impossible. We will measure the emission wavelength in a wider range in future research.

L229: “between” or “among”?

Our response: Thanks. The original sentence was removed in the revised manuscript as we have replaced DO with the pH.

L278-279: The term used here should be consistent with Figure 3. What does FI mean here? Fluorescence index? Then it should be related to Figure 3b according to the Y label of Figure 3b. But the authors related FI here with Figure 3d (“FI of DOM ranged from 1.66 to 1.94, averaging 1.78 (Fig. 3d)”), while the Y label for Figure 3d is freshness index.

Our response: Thanks. We are sorry for the misleading. We have updated this figure, and figure caption to show the DOM property correctly.

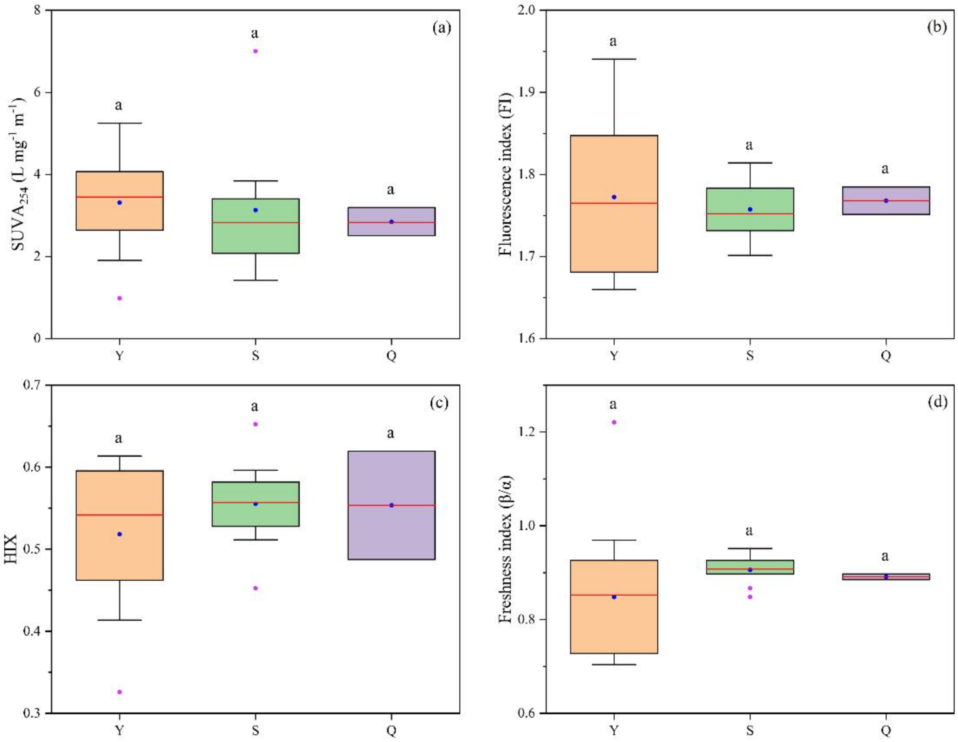


Figure 3 Spatial variations in DOM property in the Yinjiang (Y), Shiqian (S), and Yuqing (Q) catchments. (a) $SUVA_{254}$, (b) fluorescence index (FI), (c) HIX, and (d) freshness index (β/α). In each box plot, the end of the box represents the 25th and 75th percentiles, the blue solid dot represents the average, the horizontal red line represents the median, and whiskers represent 1.5 IQR. The magenta solid dot represents the outlier, which is outside of the 1.5 interquartile ranges. Different lowercase letters above the boxes denote significant differences across rivers based on statistical analysis with $p < 0.05$.

L294: The authors should provide the references to support/indicate why Cl- is anthropogenically derived here.

Our response: Thanks. We have provided the references in the revised manuscript: “*The environmental factors used in the* *model were categorized into seven latent variables, including geomorphology (elevation and slope), anthropogenic activities (e.g., urban and agricultural land uses and anthropogenically derived Cl^- (Cl^-_{anthro}, calculated as the total Cl^- concentration minus atmospheric contributed Cl^- concentration, which is the lowest Cl^- concentration at site Y5 in the Yinjiang River; Gaillardet et al., 1997; Meybeck, 1983))*" (P9 Line 227-230)

Figure 5: The box for legend did not display well.

Our response: Thanks. We have deleted this figure as we added a correlation plot and PLS-PM model in the revised manuscript.

Figure 5: As stated in the introduction part (L80-89) and L393-394, agricultural and urban land use exerted different impacts on DOM abundance and composition, why the authors combined these two landuse patterns to demonstrate the anthropogenic impacts, rather than separating these two to provide detailed explanation?

Our response: Thanks. We combined these two landuse patterns is mainly due to two reasons. On the one hand, urban land use only accounts for a small proportion of the total land use area (Fig. 1c). On the other hand, urban rivers shares some similarities with agricultural rivers in many studies, so it is common to combine them as an indicate of human disturbance (e.g., (Butman et al., 2014)). To avoid misunderstanding on the difference between urban and agricultural rivers, we have deleted the following content in the revised manuscript: "*Although the DOM in urban rivers shares some similarities with agricultural rivers (such as microbial origins), the sources of DOM in urban rivers are much more complex, which may originate from urban point-source inputs (e.g., wastewater treatment facilities) and nonpoint source inputs (e.g., household sewage and petroleum-based hydrocarbons) (Hosen et al., 2014).*"

L352-354: Both geographical and anthropogenic factors influence the DOM abundance and composition. Here, the export of DOC with higher aromaticity may be due to the landuse pattern (Fig 7(a), and steeper catchments could be coincident. How to differentiate these factors here?

Our response: Thanks. We performed a stepwise multiple linear regression (MLR) modeling to identify significant environmental factors of DOM properties in the revised manuscript. Additionally, we performed the partial least squares path model (PLS-PM) to infer the direct and indirect effects of multiple factors (e.g., geomorphologic and anthropogenic impacts) on DOC concentrations. These analyses enable us to differentiate these factors. As shown in the revised manuscript: "*$SUVA_{254}$ showed an increasing trend with increasing mean catchment slope ($p < 0.001$; Fig. 4). Furthermore, there was a significant negative correlation between $SUVA_{254}$ and the proportion of urban and agricultural land uses ($p < 0.001$; Fig. 4). This is consistent with the constructed stepwise MLR models that urban and agricultural land uses and catchment slope were the best predictors of $SUVA_{254}$ (Table 4)*" (P13 Line 312-315); "*However, anthropogenic activities (0.49) demonstrated a comparable effect on fluorescent components, primarily through a direct pathway (0.37; Fig. S4b). Anthropogenic activities (-0.84) were the strongest driver for DOM optical parameters, although geomorphology (0.59) played a significant role in indirectly influencing DOM optical parameters (Fig. S5)*" (P14 Line 340-342). We can conclude that anthropogenic activities are more important in controlling the aromaticity of DOM than geomorphology.

L366: "increasing DOC concentrations … due to microbial degradation"? Please check the logic.

Our response: Thanks. We have revised it as: "*Previous studies have reported a decreasing $\delta^{13}C_{DOC}$ with increasing DOC concentrations (Fig. 4) in spring water (Nkoue Ndondo et al., 2020) and for TOC in soil profiles*" (P16 Line 376-377)

L401-402: The authors should provide more details for this statement "anthropogenic impacts can also decrease DOC concentrations". How?

Our response: Thanks. We have added details on the reasons for the decrease of DOC in the revised manuscript: "*Yet, anthropogenic impacts can also result in decreased DOC concentrations globally due to reduced organic carbon inputs into soils and enhanced SOC decomposition induced by warmer temperatures (Nagy et al., 2018; Spencer et al., 2019) or lead to undetectable changes in DOC concentrations (Veum et al., 2009).*" (P17 Line 404-407)

L428: "consistent with previous studies", how to explain this pattern?

Our response: Thanks. We have added more details to explain this pattern: "*Lower DOM aromaticity in the urban and agricultural streams and rivers was consistent with previous studies (Hosen et al., 2014; Kadjeski et al., 2020), which suggested a microbial origin for the DOM.*" (P18 Line 434-435)

L451-453: If "the weak positive correlation …indicated that DOC and POC may have been derived from the same source", what does the strong positive correlation indicate? The authors could rephrase this sentence to avoid ambiguity.

Our response: Thanks. We have deleted this sentence in the revised manuscript to avoid ambiguity. The correlation between DOC and POC is partly explained by: "Geomorphology is also associated with the reduction in water retention time due to rapid flows, leading to a lower input of terrestrially-derived DOC into rivers as discussed earlier. It is worth noting that the conversion of POC to DOC through dissolution and desorption (He et al., 2016) is also an important source of riverine DOC (Fig. 6)". (P19 Line 449-452)