

Reply and Rebuttals

General Comments

I reviewed the original version of this manuscript and the authors' modifications/additions in response to those comments greatly improved the clarity and completeness of the methods section. In fact, the additional information provided in the Supplementary Information illustrated a considerably more thorough assessment of the modeling results, both the hydrology and water chemistry, than was evident in the original draft.

My previous summary of the research remains accurate in the updated version and I have copied it from the original review 'This research makes a useful contribution to the understanding of the hydrologic controls on DOC and DIC streamwater export in a humid tropical region, and by quantifying the annual fluxes and DIC/DOC ratios, define these systems as end-members or 'hot spots' within the context of global measurements. While there has been evidence that sub-tropical regions were important with respect to both DOC and DIC fluxes due to high productivity and rainfall, the detailed long-term measurements of concentrations and evaluation of relationships with flow had not been previously studied in any detail. By consistently sampling during a range of flow conditions (including frequently during typhoons) over a relatively long period (2 1/2 years) and using the data in conjunction with measurements of flow, flow simulation and end-member mixing analysis, they are able to evaluate the relationship between concentration and flow, estimate the relative contribution of different hydrologic flow paths to the DOC and DIC fluxes and quantify the importance of the typhoon events relative to non-event flow conditions.'

Although the work appears very thorough, and the majority of the figures present the data clearly and from a variety of perspectives, the grammar and scientific writing needs considerable improvement throughout the text. In many cases the errors in writing made the point of the sentence or paragraph difficult to understand or appear incorrect as written. I have indicated numerous examples within the specific comments. The dataset is unique and robust and the methods are very thorough, but the consistent grammatical/writing/wording errors throughout the manuscript and especially within the discussion, made it difficult to read and therefore difficult to critically evaluate the findings and discussion.

Reply:

We sincerely appreciate the time and careful review made by the kind reviewer. In response to the thorough review, we carefully replied to all the comments point-to-point. Also, we refined the manuscript for increasing the readability again. Below, we listed

all corrections and replies. All our corrections are checked in the revised manuscript.

Specific Comments

Introduction

(l. 49): delete second 'transport' in sentence

Reply: Removed.

(l. 53): correct grammar, 'DOC is mainly originated'

Reply: Corrected.

(l. 54/55): the phrase which includes '...organic matter is closely associated with different organic sources...' is redundant, I would delete the last half of the sentence or state the organic sources and describe more clearly how bacterial degradation and redox impact organic carbon.

Reply: We eliminated the last half of the sentence as suggested. [Line: 55]

(l. 61): list a few important 'direct effects' on downstream ecosystems.

Reply: We added some direct effects in this sentence. The phrase: '(i.e., organic matter burial, combination of auto- or hetero-trophic bacteria and CO₂ emission)' was added in Line: 60.

(l. 77-79): The citation Mei et al., 2014 relates to DOC, so I'm not sure why DIC transport is included in this sentence. The sentence needs clarification or DIC should not be included.

Reply: Corrected. We removed DIC in this sentence.

(l. 82-83): In the sentence 'This has become important', clarify what 'This' referring to. I would assume the understanding of shifts in the quantity and DIC:DOC ratio with different flow conditions.

Reply: The Reviewer is right. 'This' refers to 'Understanding of shifts in the quantity and DIC: DOC ratio'. We put it in the sentence instead of 'This'. [Line: 81]

(l. 92-93): correct sentence grammar/punctuation 1) comma after HBV, 2)'a' before 3 end-member, 3) correct 'during in different flow regimes'

Reply: Thanks for the corrections. We corrected the sentence as suggested [Line: 92].

Material and Methods

(l. 100-102): correct grammar

Reply: Corrected. Now, it is, 'The study was conducted in Tsengwen River watershed,

located in southwestern Taiwan.’ [Line: 100]

(l. 104): correct grammar, replace ‘ones’ with air temperatures

Reply: Corrected [Line: 104].

(l. 112-113): the discharge station at M3 doesn’t correspond with the map, which indicates a discharge station only further downstream, and not at M3.

Reply: There are two discharge stations. One is located at M3 and the other is downstream. We removed the discharge station downstream in Fig. 1 for avoiding misunderstandings.

(l. 115): correct grammar, should be ‘patterns’ not ‘pattern’

Reply: Corrected [Line: 116].

(l. 117-119): Not sure why only agricultural land is quantified in the text, perhaps because the rest is forest in those 3 watersheds. Please state the relative percent forest as well and indicate that the ‘bare land’ and ‘built up’ land use is a minor component (if that indeed is the case). Also, I’m am not familiar with the term ‘built-up’, please clarify. I assume ‘bare land’ refers to landslide scars, but please clarify in text, i.e. landslide scars are indicated by the ‘bare land’ land use category in Figure 1.

Reply: We added two sentences in this paragraph to describe the land use specifically. They are: ‘Forest is the main land use in the three catchments, accounting for 83.3, and 70.3, and 87.7% for T1, T2, and M3, respectively.’ and ‘Two other minor land uses are built-up and bareland. Built-up indicates the building, farmhouses, and roads. Bare land included the landslide scars, unplanted farms, or places under development/construction.’ [Line: 118-122]

(l. 126/127): correct grammar, ‘another bottle water samples’, perhaps ‘Water samples collected from the remaining 3 bottles’. Is this correct? Were the other 3 bottles filtered for major cations, anions and DOC?

Reply: It is another bottle of water sample [Line: 132]. The other two were shared with another collaborating lab.

(l. 134): Indicate that the water evaluated in this paper is in fact in the ‘neutral to alkaline’ range such that the ion balance method is appropriate. I don’t believe pH is presented in the paper for the reader to evaluate, though it was evaluated in-situ.

Reply: We added the pH measured in-situ for clarification. We inserted the sentence, ‘Note that the mean pH values of three sampled sites were 8.75, 9.0 and 8.57 for sites T1, T2 and M3, respectively. In this kind of neutral and weak alkaline water body, HCO_3^- , which is the main component (over 90%) of DIC, could be estimated by ion balance method.’ in Line: 138-140.

(l. 137/138): Indicate which samples were evaluated for DIC in the laboratory, how many, when and where were they collected, did they represent different concentrations and hydrologic conditions?

Reply: Twelve non-event samples (four samples for each site) were used for TOC analysis for determination of DIC, because we bought the TOC analyzer in the Nov. 2016.

(l. 139): correct grammar, suggestion along the lines of 'the strong relationship between calculated and measured DIC for the subset of samples (n=?) gives confidence in the accuracy in the values derived from the ion balance method'

Reply: Thank you. We rephrased the sentence as suggested [Line: 146].

(l. 147): 'F, Q, and dtime are the flux' doesn't make sense. Isn't Q discharge, and dtime change in time all of different units. Please clarify/correct.

Reply: Phrased to, 'where $\hat{F}$ indicates the estimated load ($\text{kg km}^{-2} \text{d}^{-1}$); Q represents stream discharge [mm d^{-1}] and $dtime$ denotes Julian day (in decimal form).' [Line: 155-156]

(l. 149): correct grammar, delete 'the' in 'the colinearity'.

Reply: Eliminated.

(l. 150): redundant/rewrite, 'The coefficient, a1 and a2, are coefficients'

Reply: Eliminated

(l. 151/152): the sentence 'the other coefficients which regulate the seasonal variation can mimic the seasonal change...' is awkward, coefficients themselves don't mimic change but are optimized so that the model can account for measured seasonal change. Rewrite.

Reply: Yes. We replaced the verb, 'mimic' by 'represent' [Line: 159]

(l. 153-156): multiple areas where grammar/punctuation needs to be corrected, change 'in LOADEST program' to 'into the LOADEST program', 'indicator' should be pluralized, comma after measure should be period, 'measure' should be pluralized.

Reply: Used plural nouns in the sentence.

(l. 160): correct grammar, 'The zero and the unity presents the performance is equivalent to expected value and perfectly matches between estimations and simulations' is very confusing. Is the author trying to state an NSE value equal to '0' or '1' indicates that the estimated values are equal to the observed values? Please clarify.

Reply: Rephrased to, 'The zero and the unity of NSE are equivalent to the expected value of observations and a perfect match between estimations and observations.' in

Line: 171-173.

(l. 161): similar to NSE, write out the Bp as an equation and define variables.

Reply: Added Equation 3 describing the indicator, Bp:

$$B_p = 100 * \frac{\sum_{k=1}^N (\hat{F} - F)}{\sum_{k=1}^N F} \quad \text{Eq. (3)}$$

where F is the observed load, $\hat{F}$ is the estimated load, and N is the number of observations during the period. [Line: 175-177]

(l. 162-164): I understand what the author is trying to communicate here, but improvement in clarity and grammar is needed. Add the word ‘the’ between ‘by’ and ‘flow weighted’. ‘The fluxes for typhoon events are estimated by the flow-weighted method directly’ please explain this method, how were fluxes between sampling periods determined? LOADEST can compute fluxes at an hourly timestep, so indicate why it was not used for event-based fluxes. Of note, the R LOADFLEX package (Appling et al., 2015 <http://dx.doi.org/10.1890/ES14-00517.1>) may be of interest to the authors in the future as it can incorporate high frequency event data into annual fluxes.

Reply: We appreciate the reviewer’s corrections and sharing of the LOADFLEX package in R. Although the LOADEST can be used for estimating the hourly fluxes as well, it would increase problematic estimations of parameters due to the inconsistent time step. Therefore, we did not used LOADEST for flux estimation during typhoon events. Our procedure for flux estimation was that we firstly applied LOADEST for daily flux estimation based on our non-event sampling. Secondly, we computed the hourly fluxes via the flow-weighted method during typhoon events based on the high-frequency sampling dataset. Later, we integrated the hourly fluxes into the corresponding day and replaced the original estimations.

We rewrote the following sentences to explain how we incorporated the event flux into daily flux. They are: ‘Note that LOADEST was only used for the estimation of daily flux based on the biweekly sampling. The event-based fluxes were directly estimated by the flow-weighted method based on the high-frequency sampling. The event-based fluxes were converted into daily fluxes, thus updating the original daily fluxes.’ [Line: 162-165]

(l. 172): after ‘its components’ list and define the three components, QRSR, QSSR, QDG.

Reply: Added the three components in the sentence, ‘(e.g., *RSR*: rapid surface runoff, *SSR*: subsurface runoff, and *DG*: deep groundwater)’ [Line: 185].

(l. 181): correct grammar, 'All the details and modeling works'

Reply: Replaced the word, 'and' to 'of'. [Line: 194]

(l. 192): After the equations, indicate which terms are known and which are being solved for since the application of EMMA here is not the typical one. I realize that the information requested here is clarified by reading the Supplementary Information II, but it should also should be articulated clearly, though briefly, within the manuscript as well. Simply, the Q terms have been determined previously from the HBV model and the subsequent use of EMMA was non-traditional, as in you already had the relative fractions of Q in each of the three components and are using that information, along with measured streamwater concentrations, to calculate the DOC and DIC concentrations in those end-members given.

Reply: Thanks for helping us to clarify this paragraph. As suggested, we marked the known and unknown terms clearly. The paragraph has been rewritten to: 'Note that the streamflow and the quantities of the three components have been determined by HBV model. Based on the known streamflow, runoff components and riverine DOC/DIC concentrations, the unknown end members can be estimated through comparing the observed and simulated riverine DOC/DIC concentrations. The details of the modeling procedure associated with: (1) accuracy of streamflow components, (2) accuracy of the estimated C sources and (3) time-invariant assumption for end-members are discussed in supplementary information II.' [Line: 210-215].

Results

(l. 207): correct grammar, suggested change 'concentrations varied widely from 1500 to 3500 uM presented the distinct seasonality' with 'concentrations varied widely from 1500 to 3500 uM during bi-weekly, non-typhoon events, illustrating a distinct seasonality.'

Reply: Thank you. We rephrased the sentence as suggested [Line: 224].

(l. 211/212/214/222/230): correct grammar for 'were satisfactorily', 'good performance in flux', 'concentrations...were not well', 'attributed by that the low rainfall', 'without typhoon invasion', 'as rainstorm begins'

Reply: Corrected [Line: 229]

(l. 223/224) incomplete sentence.

Reply: Corrected [Line: 230]

(l. 234/235) last sentence is a discussion point, does not belong in results.

Reply: Removed.

(l. 243/245) correct grammar, 'model accompanying with', 'details could be referred to'

Reply: Corrected [Line: 261].

Discussion

(l. 274-276) correct grammar/wording 'the high DIC concentration superimposing the high streamflow lead the extremely high DIC export...'

Reply: Corrected [Line: 285].

(l. 280-284) The introduction of new SOC data is a bit confusing, is the Schomakers et al. (2017) paper for the area studied in this paper? How does SOC data after a landslide relate to this paper? What is the SOC content of the region and Oceania, to which it is being compared?

Reply: Yes, Schomakers investigated the SOC content in this watershed. We attempt to justify that the SOC stock in this area is lower than those found in Oceania islands. Now, it became: 'Schomakers et al. (2017) reported that the SOC in shallow soils (< 100 cm) in Tsengwen watershed was only 2.9 ± 0.6 ton-C ha⁻¹ six years after a landslide and it increased to 75.7 ± 5.0 ton-C ha⁻¹ after 41 years, being still lower than those of the reference sites (117.9 ± 18.17 ton-C ha⁻¹), which are lower values than reported for other SMRs (100-300 ton-C ha⁻¹) (Scharlemann et al., 2014).' [Line:296-300]

Scharlemann, J. P. W., Tanner, E. V. J., Hiederer, R., and Kapos, V.: Global soil carbon: understanding and managing the largest terrestrial carbon pool, Carbon Management, 5, 81-91, 10.4155/cmt.13.77, 2014.

(l. 289/290) correct grammar/wording, not clear what is being communicated here 'The a little SOC, but high productivity could result in consistent DOC supply and high flow velocities leads to low productivity of lotic systems'

Reply: Rephrased to 'Although the high terrestrial productivity (owing to warm condition) could consistently supply DOC to rivers, the high flow velocities likely impair the productivity of lotic ecosystems.' [Line: 306-307]

(l. 316) Stated that 'main carbon export from Oceania and SMR's is from DIC and that is different than the global large rivers, but Table 4 indicates global large rivers are also dominated by DIC. Is the error in the sentence or the table?

Reply: Rephrased to 'DIC could account for, at least, 90% of the total dissolved carbon export from the studied SMRs, which is a much higher share than that observed for global large rivers (approximately 65%)' [Line:332-333]

(l. 319/321) Not clear how the prior discussion on DIC/DOC ratio's is related to the sensitivity of DOC export to environmental change. Correct grammar 'accounting for the small relative to global land mass'

Reply: After rethinking, the sentence should actually not appear here. We eliminated it.

(l. 334-335) It states the DOC concentrations in SSR and DG were 1-2 orders of magnitude lower than in RSR, does not appear to be true, concentration estimates based on figure 6 indicate RSR=108, SSR=206 and DG=86 microMole.

Reply: Sorry for the typo. It should be: 'The estimated DOC concentrations in *RSR* and *DG* were only **1/3 to 1/2** of magnitude lower than the DOC in *SSR*.' [Line: 348]

(l. 336-337) Where did Schomakers et al., (2018) measure SOC in top soil?

Reply: Upstream of M3. We inserted the locality in the sentence [Line: 351]

(l. 338-340) Unclear how measurement technique 'ultrasonic-induced soil breakdown method' related to lower estimated DOC concentrations? Assume wording is incorrect here.

Reply: Yes, the wording is quite confusing. We rephrased to, 'Schomakers et al. (2018) reported that the DOC concentrations in top-soils (0-10 cm) in the upstream area of M3 were $450 \pm 33 \mu\text{M}$ under simulating typhoon conditions by ultrasonic treatments.' [Line: 350-351]

(l. 356-357) several typos and/or grammatical errors

Reply: Rephrased. Now it became. 'Further studies are suggested to clarify the relative importance of riparian zones vs hillslopes on DOC export via using isotope techniques, for example, ^{13}C of DOM and ^{18}O of different runoffs at different locations along hillslopes' [Line: 369-371]

(l. 360-361) DOC quality introduced here, but not measured correct? The sentence makes it appear as if it was measured and changed with flow.

Reply: Following the context, this sentence is not necessary, because we demonstrated the DOC quality in the next sentence cited from Lloret et al. (2011). We eliminated the sentence.

(l. 372-280) Need to provide citations to back up hypothesis about future climate impacts including 'autotrophic production is favorable during longer dry seasons', 'DOC changes heterotrophic microbes and increases humic acid, decreasing pH' and the impact it has on dissolution of carbonate minerals. What is meant by 'reset the aquatic ecosystem'?

Reply: Yes, we added the paper from Huntington et al. (2016) for clarification. They

compiled a historical dataset and applied the regression LOADEST to fit the seasonally concentration discharge (C-Q) relation and demonstrated the response of riverine DOC export to global warming. They also revealed the change of DOC quality and the association with microbes.

Huntington, T.G., Balch, W.M., Aiken, G.R., Sheffield, J., Luo, L., Roesler, C.S., Camill, P.: Climate change and dissolved organic carbon export to the Gulf of Maine, J. of Geophys. Res. Biogeosci., 121, 2700-2716, 10.1002/2015JG003314, 2016

Table 2/Figure 3. Indicate fluxes are excluding typhoon events, if that is correct.

Reply: The fluxes include typhoon events. The hourly fluxes were integrated into the daily estimates. We addressed it in the caption of Fig. 3.

Figure 1. M3 is not denoted as a discharge site in the map

Reply: Corrected.

Figure 5. Define RSR, SSR and DG within caption.

Reply: Corrected

Figure 6. Does high flow represent mean typhoon metrics for the two events?

Reply: Yes.

Supplemental Information I

Figure S1 and S2. The (a) and (b) designation are in the incorrect location. Add r^2 value for relationships. Add the term 'hourly' in the caption for Figure S2.

Reply: We revised the two figures as suggested.

Supplemental Information II

(I. 2) Should the reference be equation 3 and 4, not 2 and 3?

Reply: Sorry for the typos. It has been corrected to Eq.4 and Eq. 5 in the main text, because we added Eq. 3 for Bp as suggested.

(I. 3-4) Please clarify, 'over 6 observations could be used to identify the end-members', do you mean 6 samples in time?

Reply: Yes. Replaced 'observation' by 'sample'.

(I 15) NSE was previously defined in the main text as it relates to discharge, but here it is relating to concentration of tracers. Is that correct? Please indicate the difference from the initial definition and what the NSE values represent with respect to confidence in the results.

Reply: It is correct. NSE is used to calculate the difference between observations and simulations in a time series. So, the indicator can be used for any kind of data.

(I 22) The phrase 'on the other hand' is not appropriate here.

Reply: Removed

(I 23) Table S5 is referenced after the presentation of the values for the three Q components, but that is not what is presented in the Table (only QSSR and QDG) so either present the estimated values for the 3 components in a separate table or do not reference a table at this location in the text.

Reply: We referred to this table in the next paragraph where it is suitable to demonstrate this table.

Table S5. Indicate the first row, hillslope/subsurface, is representing the QSSR component and the second and third are both representing the QDG component. Correct Toe with T1, T2, as illustrated in map. Indicate how many samples the observed values represent (1?).

Reply: There were only three samples for independent validation. We added a footnote, 'The three samples were collected in 2017-01-25', in Table S5.