

Interactive comment on “Composition and cycling of dissolved organic matter from tropical peatlands of coastal Sarawak, Borneo” by Zhou et al.

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We are very grateful for the reviewer's time and efforts spent on these helpful and constructive comments. Our responses to the reviewer comments are posted below, with the reviewer's comments quoted first in italics. We believe that we can address all of the reviewer's comments, and will revise our manuscript accordingly.

2. Comments to Author

Note: I also read the companion paper ‘Distribution and cycling of terrigenous dissolved organic carbon in peatland-draining rivers and coastal waters of Sarawak, Borneo’ and the relevant review comments and the authors’ answers to the comments.

This study (bg-2018-508) aimed to distinguish different fractions of dissolved organic matter (DOM) in peat-draining rivers, estuaries, and coastal waters of Sarawak, Borneo, using fluorescence spectroscopy and parallel factor (PARAFAC) analysis. The authors observed that the terrigenous fractions showed high concentrations at freshwater stations within the rivers, and conservative mixing with seawater across the estuaries, while the autochthonous DOM fraction showed low concentrations at all salinities. The authors claim that, based on the fluorescence data and little changes in optical properties of DOM, at least 20%–25% of the DOC at even the most marine stations (salinity >31) was terrestrial in origin. Although not all of the data provided is new to the relevant field, the content of this paper fulfills the requirements for the submission to Biogeosciences of which aims and scopes are to publish studies on all aspects of the interactions between the biological, chemical, and physical processes in terrestrial or extraterrestrial life with the geosphere, hydrosphere, and atmosphere. The title is representative of the article contents and the abstract summarize the contents clearly. Therefore, I recommend accepting this paper after the authors revise all the necessary points.

I have serious concerns about the use of sodium azide (NaN₃) as a preservative for samples analyzed by UV absorption and fluorescence spectroscopy. Also, the emission wavelengths used to calculate fluorescence

index (FI) seem inadequate. Finally, estimation of %tDOM by fluorescence is questionable.

Response:

We are glad that the reviewer appreciates our study. We have addressed all of the specific concerns raised by the reviewer in our responses below.

Comment 2.1.1 Estimate of terrestrial contribution

FDOM is only a small portion of the bulk DOM, and thus estimation of %tDOM by fluorescence is troublesome. PARAFAC components can be used to better understand biogeochemical processes that occur during the estuarine mixing, but PARAFAC components alone are not sufficient to estimate the tDOM contribution at given salinity. To make it possible, you must assume that all the rest of components in riverine DOM other than FDOM (PARAFAC component C1 in this case) behaves in the same way as C1 does during the estuarine mixing and that marine end-member has no C1. Please explicitly state your assumptions. It's not enough in the current form.

Response:

The reviewer correctly identifies the assumptions that underlie our estimate of %tDOC, *i.e.* that all the terrigenous DOM fractions, both fluorescent and non-fluorescent, behave in the same way during the river-coastal sea interactions as C1, and that C1 represents terrestrial humic-like fractions that only come from terrestrial sources, while the marine environment in the open ocean has no C1. We agree that these assumptions need to be made clearer than in our original submission, and we will explicitly state all these assumptions in the revised manuscript.

We believe that our assumptions are reasonable for the estimate of %tDOC within our relatively small study region because of the following three reasons.

(1) The predominantly conservative behavior of DOC concentration along the salinity gradient indicates that the distribution of DOC is mostly controlled by the mixing of freshwater and seawater, so our data do not suggest strong biogeochemical processing of the bulk DOC pool.

(2) Our C1 is very similar to terrigenous humic-like components identified in many other studies (Stedmon et al., 2003; Stubbins et al., 2014; Yamashita et al., 2015). Although we fully agree that fluorescent DOM only

accounts for a small fraction of the total DOM pool, it has already been shown elsewhere that FDOM components are appropriate proxies for both fluorescent and non-fluorescent terrigenous DOM in the coastal aquatic environment, with strong correlations noted between fluorescent DOM measurements (including PARAFAC analysis) and molecular-scale measurements by mass spectrometry (Wagner et al., 2015). This indicates that our assumption that C1 behaves in the same way as non-fluorescent terrigenous DOM fractions during the freshwater-seawater mixing is in principle plausible. A more likely source of error in our study might be the preferential loss relative to non-fluorescent DOM of C1 caused by photo-degradation, given the high photo-lability of C1 found in this study. Preferential loss of C1 would lead us to under-estimate %tDOC in our marine samples, although the exact degree of C1 photo-lability needs to be better constrained in future experiments with South-East Asian peatland samples. However, because our C1 showed predominantly conservative mixing behavior across our sample set, our data do not suggest that C1 was rapidly and preferentially removed within our study region.

(3) Other studies have found only very low concentrations of C1-like FDOM components in the open ocean environment. For example, Murphy et al. (2008) reported only ~0.006 R.U. of a terrestrial humic-like component in the tropical Atlantic, which is ten-times lower than the values we observed at our fully marine stations in Sarawak. Since, unfortunately, we do not have open-ocean samples as a pure marine endmember, we necessarily have to assume that our C1 is purely terrestrial in origin. While this assumption may lead us to slightly over-estimate %tDOC, existing open-ocean data do not suggest that this is a large source of error in our estimate (and, in fact, it would be counter-acted by the impacts of photo-degradation on C1).

We will add some additional discussion along these lines to justify our assumptions where appropriate in the manuscript.

Comment:

Also, how do you explain %tDOC of >100% in Samunsam and Western Region (in March) at salinity >10 under your assumption?

Response:

We agree that the few stations with %tDOC > 100% in the western region (mostly in March) are puzzling, and this clearly calls for further work to investigate the use of FDOM as a quantitative tracer of tDOC. One

possible reason is that the freshwater end-member value for C1/DOC ratio was underestimated for the Samunsam River. Because we could only collect a single freshwater sample in each season in the Western Region, the freshwater endmember might not be constrained sufficiently well. While the Samunsam does not have any large tributaries along the stretches we sampled, small channels from the surrounding mangroves do drain into the Samunsam estuary, so we cannot rule out additional inputs of C1-rich DOM at mid-salinities. We note also that the Samunsam estuary is shallow, and especially in March there was a lot of resuspension of sediments at the mid-salinity stations that we sampled. Because terrestrial DOM can flocculate and/or be sorbed and desorbed from sediments, it is possible that resuspended sediments at these few estuarine stations acted as an additional source of C1. More FDOM and DOC data from this river system would ideally be needed to determine why the C1/DOC ratios at mid-salinities were higher than in the freshwater endmember. We will add some additional discussion of this question to the appropriate part of the manuscript.

Comment:

In addition, F_{max}/DOC is known to be susceptible to errors caused by the fluorescence intensity and DOC measurements (Korak et al. 2014), and the authors should include an evaluation of such an uncertainty (error propagation analysis), since %tDOM estimation is I believe the most important part of this study.

Response:

This is an important point concerning the accuracy of our estimate of %tDOC. As suggested by the reviewer, we will add an evaluation of uncertainty to the revised manuscript. For uncertainty analysis, we adopted $\pm 4.2\%$ uncertainty for DOC, based on the percentage uncertainty of repeated DOC measurements of the certified deep-sea water reference material (data from Martin et al., 2018). We adopted $\pm 1\%$ as the estimated error of the F_{max} values of C1 (peak C) based on Korak et al., (2014). Formally propagating these uncertainties yields an uncertainty of around $\pm 6\%$ of the final tDOC estimate, so for a sample with 30% tDOC, the analytical uncertainty would amount to $\pm 2\%$ tDOC (so the sample would be estimated to have $30 \pm 2\%$ tDOC). Because this analytical error is very small compared to the range of %tDOC that we estimate for our marine samples (which ranges by a factor of around 2), the analytical uncertainties are not really relevant. We will add a short explanation of this in the revised version.

Comment:

Generally, in estuarine environments, contribution from estuarine vegetation (mangrove and marsh) is done by an end-member mixing model (0.1 salinity increment) using DOC concentrations of the fresh and marine end-member (Cawley et al. 2014). Because the main subject of this study is tropical peatlands, I feel that what the authors want to investigate is not riverine (derived from upper regions) inputs but inputs from the peatlands located in the estuary. The authors may reassess contributions from the peatlands using the method reported in, for example, Cawley et al. (2014).

Response:

We fully agree that a two-endmember mixing model using DOM concentrations of the freshwater and marine endmembers is an appropriate method for investigating DOM fluxes through estuaries to the sea. In coastal Sarawak, the companion paper (Martin et al., 2018) already conducted this analysis for DOC concentrations and in our study we use the same approach to study the distribution of FDOM components. We follow the same mixing model calculations as used by Cawley et al. (2014). Both in Martin et al. (2018) and the present study, we identified a conservative mixing pattern in the Western Region and additional input from the peatlands located in the delta of the Eastern Region, based on this mixing model approach. In this study, because we were able to decompose the FDOM as a mixture into multiple components representing different organic matter fractions from different sources, we could more confidently identify peatlands as the source of the additional DOC input along the Rajang Delta (as opposed to autochthonous production). However, the Rajang is the only one of our rivers in which the peatlands are exclusively located within the estuary, leading to the slightly non-conservative mixing pattern within the estuary. However, this could only be diagnosed because we used a mixing model based on a fully freshwater end-member station. Because we are already calculating mixing models as in Cawley et al. (2014) for all our rivers, we do not propose to make changes to these calculations.

Comment 2.1.2 NaN₃

Although you said 'NaN₃ did not contribute any blank fluorescence', it did contribute to sample absorbance, as you mentioned in the companion paper. Indeed sample preservation is still a major challenge, and I do use NaN₃ to preserve samples for DOC analysis. However, I never use NaN₃ to preserve samples for optical

analysis because of the strong UV absorbance by NaN₃ even at a low concentration (0.005% (w/v) in this study). I agree that if your samples have high absorbance, you could correct for the NaN₃ absorbance accurately. However, when measuring EEM for samples containing NaN₃, it seems that you failed to correct for the inner-filter effects (IFE) caused by NaN₃, because for the IFEs correction you used the absorbance of CDOM that were obtained by subtracting the absorbance of NaN₃ from that of samples containing NaN₃. In that way, you underestimated fluorescence in the EEM regions where NaN₃ absorbed light (Ex 250–280). This is very serious because you mentioned the protein-like component ‘showed consistently low values across the study region’, and this could be due to underestimation of the protein-like component. The relative degree of the underestimation will be larger with decreasing sample absorbance relative to that of NaN₃.

If you will correct (or may have corrected) for IFEs including NaN₃ absorbance, please explain the degree of uncertainty of the correction. Because, although you said all samples had the same NaN₃ concentration, there should be some variation in the concentration caused by, for example, repetitive volumetric measurements of samples (30 mL) and NaN₃ solution (150 µL).

Response:

The reviewer points out a critical aspect of EEM correction. We clearly did not explain the details of the inner filter effect corrections well enough. Indeed, we used the total absorbance of each sample (*i.e.*, absorbance of CDOM and NaN₃) for the inner filter effect correction. We then converted the fluorescence intensity to Raman Units, and then subtracted the fluorescence of our reagent blanks (DI water + NaN₃). Therefore, we do not underestimate the fluorescence intensity. This inner filter effect correction does not contribute any additional uncertainty from the presence of NaN₃, because the total absorbance of each individual sample was measured (we collected one single sample to measure both absorbance and fluorescence). Any variation in NaN₃ concentration between samples is therefore fully accounted for and included in the corrections. We will explain this important aspect more thoroughly in the methods section.

The issue of sample preservation with NaN₃ was already addressed in the discussion of the paper by Martin et al. (2018) in this issue: the reason we decided to try to use NaN₃ as a preservative was so we could follow the CDOM sampling protocols in use by the ocean remote sensing community, since our CDOM data are being used for remote sensing algorithm development. Given the problem with high blanks in the UV range, we would agree that this is not ideal for measurements below about 280 nm wavelength, but in our sample set we are very confident that we could correct for this blank with high accuracy, as discussed in Martin et al. and the

accompanying discussion.

Comment 2.1.3 FI

Did you apply instrument-specific correction for EEM? If so, the emission wavelength for FI must be 470/520 nm instead of 450/500 nm (Cory et al. 2010; Kida et al. 2018), because the emission peak often lies between 450 and 500 nm when the correction applied, which makes FI meaningless (FI must be calculated on the right side of the emission peak). If not, please write so in M&M section, because in that case your results are not directly comparable with other studies. It is often observed that if not corrected for the instrument-specific bias, the variability of FI between instruments is large for a given sample.

Response:

We are grateful to the reviewer for pointing out this issue. We did indeed apply an instrument-specific correction, so we have now re-calculated the FI using fluorescence intensities at 470/520 nm following Cory et al. (2010). This results in higher FI values for all samples, but the same distribution pattern along the salinity gradient, and the re-calculated FI values still show clear terrigenous signals for the blackwater rivers. The Eastern Region exhibits more mixed signals of terrestrial and microbial fulvic acids, but more towards the terrestrial endmember. We will interpret the new FI results accordingly in the revised manuscript. However, we also note that the ranges in FI of terrestrial *versus* microbial DOM endmembers are reported as quite variable in the literature, and the appropriate wavelength range to use for FI calculations is also still debated: even the paper mentioned by the reviewer (Kida et al. 2018) ultimately decided to calculate the FI at the traditional wavelengths of 450/500 nm because they judged the values at the longer wavelengths to be unreasonably high. We will include some extra discussion of this point in the revised manuscript.

2.2 Minor comments

Comment:

Table 2. Was the distribution of the PARAFAC components and chlorophyll-a normally distributed? If not, Spearman's rank correlation should be used instead. Note that strong parametric linear relationships between PARAFAC components are unlikely considering the theory of PARAFAC. If components have a strong linear correlation, PARAFAC cannot resolve these components and they appear as a single combined component. Correlations between PARAFAC components are generally expressed by a log-log plot or Fmax/DOC plot

(Stedmon and Markager 2005).

Response:

The PARAFAC components and chlorophyll-*a* were not normally distributed, so we will change our correlation analyses to use Spearman's rank correlation. Because the point of our correlation analyses is to show how our PARAFAC components co-vary with each other and with chlorophyll-*a* concentration across the salinity gradient, we decided not to normalize FDOM to DOC, because that would cancel out much of the variation that we are trying to analyze in this case. However, we agree of course with the point that the reviewer makes that very strong parametric linear relationships between PARAFAC components are ruled out by virtue of how PARAFAC models are calculated. However, this does not mean that PARAFAC components cannot be correlated with each other at all.

Comment:

P2L26 'extremely high DOC concentrations' Please specify the DOC range, as it depends on person when a DOC concentration is 'extremely' high.

Response:

The DOC range in the blackwater rivers in Sumatra and Borneo is up to 3000–5500 $\mu\text{mol L}^{-1}$ or 36–66 mg C L^{-1} , which lie on the highest DOC concentrations in the rivers reported globally. We will specify these numbers in the revised version.

Comment:

P3L11 Sampling

How was the weather on the sampling days? In addition to seasonal changes, daily changes in rainfall and water flow conditions would affect DOM concentrations and compositions. If you discuss seasonal changes, at least the weather should be the same.

Response:

During the sampling cruises, we did not encounter extreme weather events. Overall, during each expedition most days had part cloudy / part sunny weather conditions, and heavy rain showers of a few hours' duration

occurred on many days (usually in the afternoon), as is typical for this equatorial climate. Cloud-free days were rare. Because a lot of the rainfall in this region takes place across small spatial scales, the weather conditions during any one day at any one particular location are not necessarily indicative of the weather across an entire river catchment. Hence, it is unlikely that DOM concentrations and composition were affected in a significant way by day-to-day changes in weather conditions, and indeed we do not see any evidence of this in our dataset. We will include some additional description to this effect in the methods section of the revised manuscript.

Comment:

P3L30 Was the condition of the photodegradation experiment sterile (biodegradation-free)? If not, how about the effect of biodegradation? Please add some more details about the photodegradation experiment. For example, water inside the bottles was repetitively sub-sampled or you prepared many bottles and each bottle was collected as a sub-sample?

Response:

The photo-degradation samples were filtered by 0.2- μ m pore-size Anodisc filters to remove bacteria in order to rule out any effect of biodegradation or solubilization of particulate organic matter. Bottles were repetitively sub-sampled, and while this may have introduced some microbial contamination, this would have affected the dark control bottles to an equal extent. We will add these experimental details when we revise the manuscript.

Comment:

P4L3 'To minimize self-quenching of fluorescence intensity' Please add information on the maximum absorbance value of the measured samples, since IFE correction becomes invalid if sample absorbance is too high. Also, how you measured absorbance data is completely lacking. Please explain it in this section, and reference to the companion paper alone is not sufficient.

Response:

We will include a brief summary in the M&M section revised manuscript of how the absorbance measurements were conducted: we used a dual-beam Thermo Evolution 300 spectrophotometer with quartz cuvettes, and

selected a cuvette pathlength of either 100 mm, 10 mm, or 2 mm according to the sample absorbance (for the March data, high-absorbance samples were diluted with DI water because the 2-mm cuvette was unavailable).

For fluorescence measurements, we used a 1-cm cuvette for samples with low absorbance, while samples with high absorbance were either diluted 10-fold with DI water and then measured in a 1-cm cuvette (March samples), or measured undiluted in a 3-mm cuvette. For all samples, we used the A_{total} of the appropriate dilution and pathlength at which the fluorescence measurements were conducted.

Kothawala et al. (2013) proposed that the inner filter effect correction is invalid for EEM regions with $A_{\text{total}} > 1.5$ because of non-linearity between absorbance and fluorescence intensity. We have three samples for which $A_{\text{total}} > 1.5$ in part of the EEM spectrum, as shown in Figure 1 below. Therefore, the PARAFAC results of these three samples, especially the C5, should be treated with caution. The A_{total} values of all other samples are below 1.5 across the whole EEM, so the inner filter effect correction is fully valid for them. This information will be added to the M&M section and the Figure 1 below will be added to the Supplementary Information.

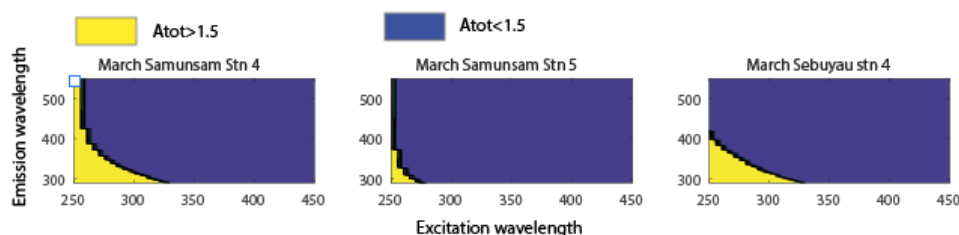


Figure 1. Samples with A_{total} above 1.5.

Comment:

P4L27 ‘chemical compound classes’ The authors need to be careful here. What PARAFAC can do is to statistically deconvolute EEMs into underlying building blocks, termed ‘components’, and these components are rarely related to specific chemical compounds. I think the authors understood that, but for those who are not familiar with PARAFAC, the author’s statement may be misleading.

Response:

We fully agree with the reviewer that caution is needed here, hence we referred to “compound classes”, not “compounds”. We will re-phrase this as “...which decomposes the variation between EEMs in a dataset into multiple mathematically independent components representing different organic compound classes, the aggregates/union of compounds with similar source, biogeochemical properties and behaviors.” to make this

clearer also to non-specialists and avoid misunderstanding each PARAFAC component as a specific chemical molecule.

Comment: P4L28 Specify how many samples were removed.

Response:

Four samples were removed. We will add this information to the revised manuscript.

Comment:

P4L29 Please add in Fig. 3 the excitation and emission loadings of the validated split dataset.

Response:

Response:

The excitation and emission loadings of the validated split dataset were saved during PARAFAC analysis and will be shown as an additional supplementary figure. It can provide further information about the validity of our five-component PARAFAC model for the readers.

Comment:

P5L1 Fmax is not just a score value. "Fmax is calculated by multiplying the maximum excitation loading and maximum emission loading for each component by its score, producing intensities in the same measurement scale as the original EEMs" (Murphy et al. 2013).

Also, Fmax cannot be a major of the concentration of each component in a sample, "because different fluorophores can have very different efficiencies at absorbing and converting incident radiation to fluorescence (Murphy et al. 2013)." Rather, "Quantitative and qualitative information may however be obtained from changes in the intensity of a given component, or in the ratios of any two components, between samples in the dataset (Murphy et al. 2013)."

Response:

We will correct our explanation of Fmax.

We fully agree that Fmax cannot indicate the absolute concentration of compounds, but instead indicates relative

changes in concentration of each component between samples, which is the way we interpret our PARAFAC results throughout the manuscript. We realise that our description here was perhaps slightly misleading, so we will re-phrase the section to read “which is taken as a measure of the relative concentration of each component in different samples of a dataset”.

Comment:

P5L8&L15 a350, S275–295, SR, and SUVA254 appeared for the first time here without explanations what they are. This is not kind for those who are not familiar with the optical indices. This is relevant to my comment on P4L3. Now I think that you need to make another section in M&M that explains the absorbance measurement and absorbance-based indices. However, personally I think that you can completely cut the sentences with respect to SR, a350 and SUVA254 since you mentioned about SR and a350 only once or twice and did not discuss SUVA results (just correlation with HIX).

Response:

We will review the need for mentioning each of these measurements, and will add a brief explanation of any CDOM terms that are used. We believe that it is useful to briefly summarise these CDOM results from the companion paper in order to provide the readers with additional background about the CDOM concentration, DOM molecular weight and source in these rivers so that readers can appreciate the FDOM analysis more easily.

Comment:

As for S275–295, you may want to use it to support your idea that an FDOM-based estimate of tDOM is OK. However, I am not totally convinced that being correlated with S275–295 supports the correctness of your fluorescence-based %tDOM (P11L24), because estimations of %tDOM based on S275–295 is non-linear (Fichot and Benner 2012).

Response:

We agree that a correlation between our %tDOC estimates and $S_{275-295}$ does not prove the correctness of our method of calculating the %tDOC, but we do believe that it adds additional support: as in Fichot & Benner

(2012), we find that there is an exponential relationship between %tDOC and $S_{275-295}$, as shown in Figure 2 below ($\%tDOC = \exp(\alpha + \beta S_{275-295})$, where $\alpha=1.48$, $\beta=-126.23$). We will add the exponential regression model of the relationship between %tDOC and $S_{275-295}$ to the revised manuscript and replace Figure S1(b) with Figure 2 below.

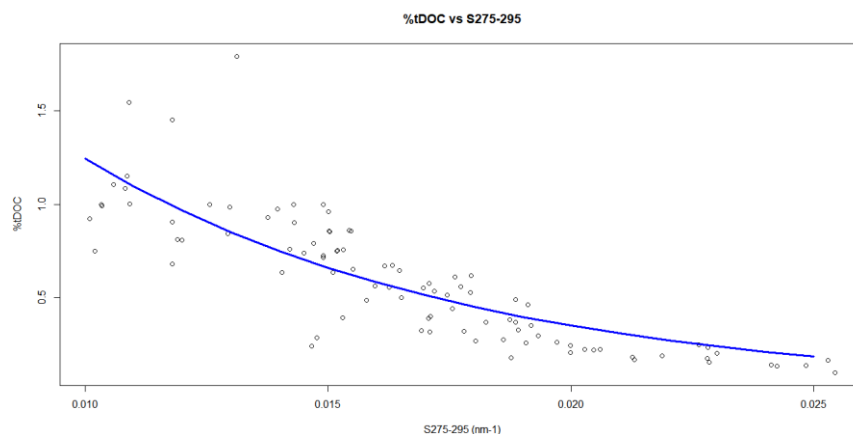


Figure 2. Relationship between estimate of %tDOC and $S_{275-295}$. The observation data is plotted using circles and the exponential regression model is presented by the blue solid line.

Comment:

P5L16 “SUVA254, 3.08–6.89” SUVA value of 6.89 is too high. Even the highest aromaticity sample ($Ar > 40\%$) in Weishaar et al. (2003) had the SUVA value of 5.3, and the possible maximum SUVA value (~ 5) has been recently suggested from a molecular analysis (Kellerman et al. 2018). Iron(III) is most probably interfering with SUVA determination in your sample dataset (Poulin et al. 2014; Kida et al. 2018). If the authors did not measure Fe(III) and also have no stored sample for Fe(III) measurement, please state in the manuscript that some of SUVA values in this study was overestimated by interferences from Fe(III) to an unknown degree. Note that, if Fe(III) contributes to SUVA254 to a similar degree for all the samples, SUVA254 and SUVA280 would still have a high correlation.

Another possibility is the interference by NaN3 even after the blank correction. This is possible when the sample CDOM absorbance was low. Please add the information on the NaN3 absorbance contribution to sample absorbance at 254 nm. According to Fig S1&2 of the companion paper, decadic absorption coefficient of the NaN3 solution was about 4 m^{-1} , which was about 10% - 30% of that of Rajang, Sematan, and Lundu samples and 50%–200% of marine samples. These values are not trivial.

Response:

Unfortunately we do not have Fe(III) measurements, so we cannot rule out that the very highest SUVA₂₅₄ values were impacted by iron, even though peat-draining blackwater rivers typically have very low dissolved mineral concentrations. While the Kellerman et al. paper is a very interesting study, we note that the authors only very tentatively propose an upper boundary of around 5.5 for SUVA₂₅₄, given their limited sample set, while other recent studies still use SUVA₂₅₄ up to 6.0 (e.g. Massicotte et al. (2017)). All but one of our samples had SUVA₂₅₄ below 6.0, and most samples were below 5.5, so even if our highest SUVA values are impacted by the presence of iron, this is unlikely to have affected our dataset to a very serious degree. The possibility that NaN₃ was responsible for the high SUVA₂₅₄ values was already ruled out in the paper by Martin et al. (2018), given the very strong and linear relationship between SUVA₂₅₄ and SUVA₂₈₀, because NaN₃ no longer has any significant absorbance at 280 nm (besides, while the NaN₃ absorbance at 254 nm was certainly high, the NaN₃ concentration was kept very consistent between samples and was thus corrected for accurately). Because SUVA₂₅₄ is only used in such a limited manner in the present study (indeed, the reviewer even suggested omitting SUVA₂₅₄ altogether) we think it is better not to side-track the paper by discussing these CDOM methodological aspects here, given that SUVA₂₅₄ and the problems with NaN₃ have already been covered in more detail in the paper by Martin et al. (2018) and the accompanying interactive discussion.

Comment:

P6L3 Please add seasonal climatic information (dry? rainy?) after months so that readers can easily understand climatic conditions, not only in the M&M section.

Response:

We will add this where appropriate. It is important to note that in this equatorial climate there are not very distinct wet and dry seasons, instead, there is high rainfall year-round. We will highlight this more clearly in the Methods section.

Comment:

P9L31 “correlating strongly with DOC-normalized amino acid yields” This is not a correct citation.

The correlation coefficient was $r = 0.62$ (Fig. 8b in Yamashita et al., 2015), at best moderate correlation.

Response:

This is a valid point. This sentence will be rephrased as “Furthermore, Yamashita et al. (2015) found that the DOC normalized protein-like component Fmax value was indicative of the amino acid content in DOM and thus the bioavailability of DOM.”

2.3 Technical corrections

Comment:

P2L5 & L7 ‘0.2-0.25 Pg C yr⁻¹’ and ‘40% - 50%’ should be 0.2–0.25 Pg C yr⁻¹ and 40–50% (or 40%–50%).

Please check the usage for minus (–), hyphen (-), en dash (–), and em dash (—). I did not correct for the rest of the manuscript.

Response:

We will check and correct the usage of these symbols.

Comment:

In Fig 2&4, it would be better to set the x axis to the same scale (maximum salinity of 35) except for the Simunjan River results so that comparisons between rivers become easier and more straightforward.

Response:

We will set the x axis to the same scale except the Simunjan river as suggested by the reviewer to make the figures easier for the readers.

Comment:

The caption of Fig. 4 says ‘while they distinguish samples from different regions in the panel (z)’, but I can’t find the panel (z).

Response:

The panel (z) was removed from the manuscript before submission, but we forgot to correct the caption. We will delete “panel (z)” from the caption. We apologize for the mistake.

Comment:

In Table 1, please add Tucker congruence coefficient (TCC) values so that readers can evaluate how much the comparisons are quantitative. Add the relevant explanations in M&M section as well.

Response:

The tucker congruence coefficients between our models and the models from the cited literatures were all above 0.95, which indicates strong correlations. The respective TCC values between our model and the cited models can be found in the OpenFluor report attached with this response. Specifically, both Coble et al. (1996) and Mcknight et al. (2001) did not run PARAFAC analysis so no TCC can be provided for them. We cited these two papers because the peak positions and spectra of our components are close to theirs identified by the peak-picking technique and they have been widely acknowledged as the nomenclature of FDOM EEM peaks. We are trying to keep the table concise and highlight the most critical information of the possible source and biogeochemical properties of the compound classes represented by our PARAFAC components so we are considering not add the respective TCC values for each pair of the models but this OpenFluor report will be uploaded as part of the Supplementary Information and we will add the relevant explanations of TCC in the M&M section.

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