Authors' Response

We are grateful to both of the reviewers and the associate editor for their constructive comments on our manuscript. We have revised the manuscript according to the points raised by both reviewers, and have also lightly edited the text to further improve the language and to shorten the text where possible. Our response to each of the reviewers is listed below.

Response to Reviewer 1

We thank Reviewer 1 for their time in reviewing our manuscript, and for providing constructive criticism. We have made a number of changes based on these comments, as detailed below (the reviewer's comments are quoted in blue italic font).

Martin et al. present a concise empirical study on the spatial and temporal cycling of DOC in peatland draining rivers, Sarawak. Of particular interest are the results of the photo-degradation experiments and the fate of the riverine DOC component. The manuscript is well written and the study makes a valuable contribution to the scientific knowledge database. I have a few minor comments:

Page 5 Ln 30: acidification was chosen to preserve the DOC samples, however, this has been shown to reduce DOC concentrations (Kaplan.1994: https://doi.org/10.4319/lo.1994.39.6.1470) as well as alter spectral properties (Tfaily et al. 2011: doi:10.1016/j.aca.2011.08.037). I just wondered why cold storage was not considered?

Appropriate sample storage is a problematic subject, to say the least, and neither for DOC nor for CDOM has any one protocol really become established above all others. In acidifying our DOC samples immediately upon collection and then storing them cold, we followed a method that is very commonly used in the oceanographic community, with acidification inhibiting microbial DOC metabolism. The Kaplan paper shows interesting results, although we note that it does not show a decrease in DOC upon acidification in all samples. Their analysis was also done with persulfate oxidation, which is typically less efficient than high-temperature combustion systems, so some of this apparent DOC loss could involve modification of DOC, not necessarily only mineralisation to CO_2. Given that we needed to store our samples for up to 1.5 months before they could be analysed, we decided that acid-preservation seemed like a safer approach than simple cold-storage; unfortunately, we didn't have the capacity to collect and store extra samples to test this.

We certainly agree that spectral properties of DOM would be altered upon acidification. All of our CDOM samples were therefore not acidified, but preserved with sodium azide to inhibit microbial activity, and stored cold alongside our DOC samples. As also explained in our response to Reviewer 2, we collected our CDOM data in part also to develop a remote sensing

algorithm, and this preservation method is the recommended protocol in the ocean colour community – although measurements are then generally made at wavelengths >300 nm. Given our experience with the absorbance blanks from NaN_3 up to around 270 nm, we agree with the reviewer than simple unpreserved cold storage is perhaps preferable if CDOM analysis in the UV range is planned (that said, as explained in the manuscript text and also in response to Reviewer 2, we are very confident that our blank correction was sufficiently accurate that this did not compromise our data). We have also added a new supplementary figure (now Supplementary Fig. 1) to show the absorption spectrum of the sodium azide blanks.

Page 6 Ln 2: Mentions that freezing did not affect the DOC results, how as this assessed? Is there any evidence that could be added (maybe to supplementary material to support?)

This was based on comparing the unfrozen samples to frozen samples from adjacent stations in the Maludam. Since all of the September Maludam samples follow very clear conservative estuarine mixing, with only a narrow range of DOC and CDOM parameters in freshwaters, frozen and unfrozen samples clearly had very similar results. Some protocols for DOC and for CDOM or FDOM samples in fact even recommend preservation by freezing, and while we think it is better not to alter samples in such a drastic way, major impacts from sample freezing are probably not common. We have added a brief explanation to this section to explain why we think that freezing did not impact these results.

Page 8 Ln 9: were the quartz bottles overfilled with the water and then capped to eliminate headspace and therefore eliminate atmospheric exchange during the photo- degradation experiment? If so it would be good to include this information in the meth- ods and also how the quartz bottles were prepared e.g. combusted? I wondered if there was any sign of bacterial growth in the water samples during the experiment (just out of curiosity).

The bottles were actually filled with a headspace of around 20–40 mL air, which was done to ensure that the samples would remain oxygenated throughout the duration of the experiment. If samples are filled without a headspace, DOC degradation can in principle become oxygen-limited, depending on the degradation rate. The quartz bottles were not combusted, but were acid-rinsed instead. This information has been added to the methods. We did not enumerate microbial cells in these samples, but there was no visible evidence of growth (no cloudiness or particulate matter visible).

Page 9 Ln 14: is there any way to include some information about the total distances travelled during the different campaigns (maybe on figure 1, by including a scale bar) or the distance between sampling stations to be added into the supplementary material? How was the distance between the sampling points decided?

We have added a scale bar to Figure 1a to give a quick overview of the distance. The other panels already contain a lot of information so we do not want to clutter them with further scale bars, but distances can be estimated easily from the latitude axes on these panels. Distances between river stations are partly shown in Figure 3 for those rivers where multiple freshwater samples were taken.

Distances between sampling points were inevitably decided according to multiple factors: during estuarine sampling we chose stations mostly according to salinity, but distance between stations was considered as well. Logistical considerations were naturally important, e.g. the total distance that could be covered in one day. Many of the marine sampling stations were chosen according to optical water types, because of the need to use our data for remote sensing purposes – in this case we selected stations according to distance to the coast, the presence of distinct fronts, and differences in water colour. Logistical considerations were of course again

important, e.g. sea conditions and prevailing currents, the total distance that could be covered given fuel, water, and time constraints, and the location of sand banks that had to be avoided for safety reasons.

Page 9 Ln 16: Was there any seasonal variability in salinity i.e. did you see a drop during the wet season due to greater freshwater input? Did you observe any salinity induced flocculation in the DOC samples which may have complicated analysis?

There was some seasonality in salinity in the Western Region, as mentioned at the end of Section 3.1 (this is also visible in Figures 2 and 4). We did not specifically test for salinity-induced flocculation of DOC owing to logistical constraints; we hope to test for this during future fieldwork in the region. However, none of the estuaries showed evidence of non-conservative removal of DOC, so we suspect that any salinity-induced flocculation probably does not have a significant impact on DOC concentrations in this region.

Page 11 Ln 26: Was there any evidence of photo-bleaching?

Yes. As also requested by Reviewer 2, we have now added additional panels to Figure 6 and Table 1 to show the decrease in CDOM concentration as a_{350}.

Page 13 Ln 14: Was any POC data taken? Would have been interesting to see if there was any change in the DOC:POC ratio during the photo-degradation experiment, but as stated POC to DOC turnover is unlikely to have been captured within the experiments time frame.

We did not measure POC either in our environmental samples or in the photodegradation samples. POC data for many of the stations will be presented in another manuscript in this special issue, but unfortunately the volume of water in the photodegradation incubations was insufficient anyway to filter for POC at each time-point. However, we are planning to do more follow-up work on photo-degradation of SE Asian peatland DOC, and we will certainly attempt to measure POC as part of that. Note that the water for the photo-degradation experiments was already filtered at the start of the experiment, and the incubation bottles were then sub-sampled at each time-point. These sub-samples were not re-filtered, so any DOC that was transformed to suspended POC would still be quantified as DOC with our measurements.

Page 13 Ln 24: Do you have any information on the extent of oil palm plantation coverage in this region/ the % of land likely to be covered by disturbed peatlands?

At this point we do not have updated estimates for plantation coverage and land disturbance for the region, but these questions are being addressed by other manuscripts that are in preparation for this special issue. Unfortunately, land-use data are difficult to obtain in this region on a catchment-basis. Essentially, practically all of the peatlands need to be considered as at least partly disturbed, in the sense that they have a history of logging, and even the Maludam National Park peat forest is classed as a secondary forest. In the Rajang delta in particular, the majority of peatlands has been converted to oil palm plantations in the past years. However, the objective of this paper is not specifically to analyse effects of land-use change on DOC and CDOM concentrations, and our sampling scheme was not designed specifically to address this question.

Page 13 Ln 27: Another reference that might be good to add is Materic et al. (2017) (https://doi.org/10.1038/s41598-017-16256-x) who observed differences in the composition of organic compounds between forest and disturbed peatlands (some data is from Sarawak).

Thanks, we had not yet come across this paper. We have added the citation.

Page 14 Ln 9: I wonder if the lower DOC concentrations observed at the end of the drier season could be a reflection of the interaction between the hydrology and photo- degradation i.e. during the dry season flow conditions will be low which could lead to longer residence times leading to greater UV exposure and thus DOC degradation. Again, just a thought.

We certainly cannot rule out this possibility, but we suspect that photo-degradation in the rivers is probably not so pronounced. All rivers showed significant flow in all seasons, and because of the extremely high light attenuation in all rivers (due to CDOM and sediments) a large change in water residence time would be needed to allow for significant photo-degradation within the rivers. Moreover, if photo-degradation was significant in the rivers, we would probably see more consistent seasonal variation in DOC across the rivers. Given that we can really only speculate about this, we have not made any changes to the manuscript in this regard.

Page 14 Ln 20: considering how photo liable the DOC is could this not have produced molecular level changes complicating its identification with respect to terrestrial sources, especially in samples collected further downstream which have been exposed to greater periods of light exposure? So maybe the terrigenous signal identified in the samples is even stronger? Just a thought.

Yes, this is a very valid point: we agree that any estimate of terrigenous contribution in marine samples based on $S_{275\text{-}295}$ would, if anything, be an under-estimate. As also mentioned above, we suspect that photo-degradation is not really at play within the rivers themselves, because of the short water residence times and the extremely high light attenuation. Instead, photo-degradation of tDOM most likely only becomes significant in coastal waters once sediments have partly settled and the euphotic depth is greater. We have not modified the manuscript further in this regard.

Page 16 Ln 16: If the majority of the landscape is oil palm plantation I guess the drainage channels/ continuous yearly peat drainage could be ensuring that there is a continuous and direct flow of water (and thus DOC) into the river system, topping up the supply. Maybe this could also explain why there are higher DOC concentrations above the mixing line?

This is certainly a possibility, although given the generally high year-round precipitation, it is likely that there is also simply a high amount of natural input year-round. The peatlands in the Rajang delta are drained naturally by a network of small rivers and streams that discharge into the Rajang distributary branches. We cannot say whether conversion to oil palm plantations may have increased this DOC input, although we cite the literature from site-specific studies elsewhere on Borneo that have shown increased DOC losses due to land-use. We have therefore decided not to make any changes to the manuscript in this regard, given that our focus in this paper is not to discuss the effects of land-use. Any discussion of natural *versus* plantation-induced DOC fluxes here would necessarily be entirely speculative.

Page 16 Ln 19: As the majority of Rajang catchment is draining disturbed landcover (i.e. oil palm plantation) this could be contributing to an increased nutrient input from the pesticides and fertilisers and thus cause eutrophication to come degree and contribute to the DOC pool (even though I see that the chlorophyll is low). However, the sampling campaigns are designed to show us a 'snap-shot' of the spatial variability of the stream network across different seasons, so perhaps it is hard to rule out autochthonous DOC sources completely? Maybe there could be a lag response?

Trying to understand the impact of fertiliser input from the plantations would be an interesting and important study. Depending on the magnitude of this nutrient input, it certainly might contribute to eutrophication. Again, however, we strongly suspect that primary production within the rivers is too strongly light-limited for there to be a response to nutrient input within

the rivers and estuaries, so most likely there would be a down-stream effect in coastal waters, once the water clarity is higher. The question of how the underwater light level is controlled in this region will be addressed in a separate manuscript that is currently in preparation for this special issue. We would point out that although our sampling is indeed a snap-shot, any nutrient input from the plantations should have been happening also well in advance of our sampling campaign, so if there really was a downstream increase in chlorophyll due to nutrient inputs we would expect to see elevated chlorophyll concentrations within the rivers and estuaries, but the chlorophyll concentrations there are mostly very low. However, a more thorough discussion of these patterns will be the subject of the bio-optical manuscript that is currently in preparation.

Response to Reviewer 2

We thank Reviewer 2 for their time in reviewing our manuscript, and for providing constructive criticism. We have made a number of changes to the manuscript based on these comments, and our point-by-point responses are detailed below (the reviewer's comments are quoted in blue italic font).

Martin et al. explore the spatiotemporal variations of dissolved organic carbon (DOC) and colored dissolved organic matter (CDOM) using in-situ data obtained from a total of six peatland draining rivers and coastal zones in Sarawak. The photo-liability and the cycling of these riverine DOM are also further investigated and discussed by conducting field photo-degradation experiments. Although some of phenomena and conclusions presented in this work are not new in this region or elsewhere, it's valuable to have seasonally-resolved DOC measurements in this important tropical marine biodiversity hotspot area. Overall, the data obtained in "black waters" are very interesting and the overall quality of the study is positive and contributes to a better understanding of the DOM properties in a region that accounts for a large fraction of DOC export to the global ocean while also facing strong anthropogenic influences; however, the manuscript needs some improvements in the text and figures.

Specific comments and suggestions for further improvements: The authors spent 5- pages to describe the Materials and Methods part, which is much longer than the results. Some of the methods described are not new (e.g., section 2.2.1 and 2.2.2 for measuring DOC concentration and CDOM absorbance), and can be properly shortened. Regarding the precipitation data in Fig. 1a, you may consider to highlight the locations of meteorological stations using corresponding colors in Fig. 1b-1d.

We agree that the methods were quite long. We have shortened the relevant sections, although because we need to describe numerous different analyses and the photo-degradation experiment, the Methods section is inevitably somewhat long.

The meteorological stations were previously highlighted with pink arrows in each of the panels. We have now changed the arrow colour to match the colour of the bars in panel (e).

Regarding the section 2.2.3 of conservative mixing model, you may consider giving more details about the procedures using a table or other means.

It's not clear to us exactly what additional details the reviewer is requesting. We have decided to add an additional column to our supplementary data table that indicates which stations were used as end-members for the mixing models. We have also changed the wording in this section a little to make it clearer. We had already referenced the Stedmon & Markager paper that explains how CDOM mixing models should be calculated, and we now emphasise this as a methods paper more clearly. The actual calculation of two-endmember conservative mixing models is simple and this is done very commonly in studies of estuarine gradients, so we do not want to expand the methods section even further by explaining this in detail.

Page 6, line 30, can you add a reference here and describe a bit the advantages of adding NaN3 to DI water as blank?

There is no specific reference for this, but maybe the reviewer misunderstood this slightly: we did not add NaN_3 to the DI water in the reference cuvette of the spectrophotometer, we made proper reagent blanks with the NaN_3 that were measured against the DI reference and then subtracted from the samples. We necessarily had to do this, because NaN_3 does have some absorbance at wavelengths >250 nm, so we needed to correct for this. We have tried to clarify this as best as possible in Section 2.2.2 without adding excessive length.

Page 7, line14, absorbance should have no unit. Also, page 7, line 1 – NaN3 absorbances around 26 m-1 at 230 nm, 4 m-1 at 254 nm: shouldn't these be absorption coefficients? These are very large values and likely to influence SUVA values.

We apologise for this oversight, these values are decadic absorption coefficients, not absorbances. We have corrected the text. We report these as decadic rather than Napierian absorption coefficients here because the blank is most relevant for calculating $SUVA_{254}$, which is done using decadic instead of Napierian coefficients. For the most part, the blanks at 254 nm were still small relative to the CDOM absorbance, given the high CDOM concentrations in most of our samples. Because the NaN_3 concentration was also identical across all samples, this blank could be subtracted very accurately. On Page 11, we show that $SUVA_{254}$ is extremely closely related with SUVA at 280 nm, a wavelength at which NaN_3 has no significant absorbance, across our dataset (r^2 = 0.990). This indicates that our $SUVA_{254}$ estimates were not affected by the NaN_3 blank. We have now also added a new supplementary figure (now Supplementary Fig. 1) to show the spectrum of a sodium azide blank.

We used NaN_3 because this is the recommended preservation protocol for CDOM samples in the ocean colour / remote sensing community (as in Tilstone et al. 2001, cited on Page 5), and we are currently using our CDOM data for a satellite remote sensing analysis. However, because CDOM measurements for remote sensing purposes are usually made at wavelengths above 300 nm, we were unaware of this blank issue in advance. We then continued with this protocol for the sake of consistency across our dataset. With hindsight, it is maybe better not to use NaN_3 for CDOM analysis at wavelengths below 300 nm, but we were clearly able to correct for this blank without significantly compromising our data.

Page 11, line 21-25, can you describe more about the highly scattered data in range of 2.5-3.5 of log DOC (Fig. 5b) and 1.0-2.0 of log S275-295 (Fig 5c) (e.g., geolocation of the scattered data) since you

mentioned that no strong seasonal changes in DOM composition within your study region in discussions; Page 16, line 17-18), which contradict a bit with the results here. In addition, you may consider to keep same scale of these parameters in Fig. 5 instead of using log scale for some parameters.

The scatter in these relationships at the high DOC concentrations (>2.5 log(DOC)) is largely due to inherent variability between the rivers: the Rajang, Sematan, and Simunjan have somewhat higher $S_{275–295}$ at a given DOC or CDOM concentration than the Maludam or Samunsam. The lack of seasonal variation in these relationships is most obvious when comparing the March and September data: both datasets cover nearly the full range of data and both follow the same trajectory (including the scatter at higher DOC concentrations). The reason why it looks like there is some seasonality is two-fold: first, because we were unable to sample all sites in all seasons, and thus the June data (only Rajang and marine samples) only cover a smaller range of values than the other seasons. Secondly, in June and September, we were able to sample marine waters with lower DOC concentrations, which could not be sampled because of weather conditions in March. Therefore, the March data do not extend to such low DOC / high $S_{275–295}$ values as the other two seasons. However, our point here is that all of the data essentially follow the same trajectory on these plots, rather than clustering into two parallel relationships by season (as found in some studies of other regions).

We have expanded this section slightly to explain some of this in more detail, and hopefully this is enough to clarify our point sufficiently.

We did try plotting these data using linear scales instead of log-scales. Unfortunately, because our data span such a large range in DOC and in CDOM concentration, plotting on a linear scale makes it very hard to properly see most of the data, because it is not possible to distinguish properly any samples with less than about 250 µM DOC. We agree that log-scales make it harder to directly compare these data to our other figures, but otherwise the relationships we are trying to visualise are simply impossible to see across the full dataset.

Regarding Fig. 6, can you specify the black and yellow symbols in figure caption? Also, page 11, line 27, it is written that both ". . .DOC and CDOM decreasing after sunlight exposure" however, Figure 6 and Table 1 do not show CDOM absorption values. It would be important to also show the relative decrease in CDOM at 350 nm with light exposure. Figure 6 is also repeated in supplementary S3.

We have specified the symbol colours in the figure caption now. CDOM did indeed decrease, and we have now added one more row of panels to Figure 6 to show the decrease in CDOM, as a_{350}. We agree with the reviewer that showing the a_{350} data for the photodegradation experiments is important, and we have therefore also decided to show the a_{350} data in Table 1 instead of showing the $S_{350–400}$ data, given than $S_{350–400}$ mostly showed little change relative to the controls, as is obvious in Figure 6. Unfortunately, the table is too small to show all these parameters.

Figure 6 is not exactly repeated in the supplementary information: our SI Fig. 3 shows the photo-degradation data plotted against the number of days of sunlight exposure, while Figure 6 shows the data plotted against our estimated cumulative irradiance. We think it is important to show both, because the different days do have differences in irradiance. Conversely, because we could not measure the cumulative irradiance exactly, we think it is important to also show the data plotted against time for comparison.

Page 13, line 10-15, it might not accurate to rule out a major autochthonous source of DOC just according to low surface Chl a concentration since the DOC fluxes from benthic flora to the overlying water column might also be another possible reason for high DOC in shallow estuary.

This is a good point. Essentially, this would require significant stocks of benthic macrophytes in the river. However, although we have no systematic data of macrophyte cover, we believe that it is very unlikely that macrophytes are present in significant quantities in any of our rivers. First, in the Rajang and Sematan rivers, the suspended sediment concentrations are very high and prevent deep light penetration; the same is true for all of the blackwater rivers due to the high CDOM concentrations. Secchi depths were measured at some stations in the Rajang and were always less than 30 cm relative to a river depth of often ≥10 m. Moreover, although moderate to large amounts of terrestrial plant debris (branches and leaves of trees, tree trunks, entire clusters of palm trees) were always seen floating at the surface of all rivers and out at sea, we never observed debris of aquatic macrophytes. Exposed river banks at low tide also never showed evidence of aquatic macrophytes. In the blackwater rivers we often had the opportunity to see the upper 10–30 cm of the river bank below the water line, but also never saw any macrophytes, only the sediment. We have now slightly expanded this passage to state that we never saw evidence of aquatic macrophytes, and that benthic primary production is likely to be at most minimal owing to the low light penetration in all rivers.

Page 14, line 10-14 "the high precipitation in Maludam in September ... . .. DOC concentrations" which is not clear to me and I am wondering if there is previous study that reported this phenomenon; if yes, you may want to add a reference here. In addition, the lower DOC concentrations mentioned there could probably be associated with other environmental factors. The hydrological and meteorological conditions during wet and dry season could be different, which will change the residence time of waters and solar radiation, further affect DOC and CDOM properties there.

We agree that this was not very clearly phrased. We have expanded this section slightly to clarify our meaning: especially in peatlands, it has been noted that high precipitation can lower DOC concentrations in rivers by essentially creating a dilution effect, as described in the Clark et al. (2007) paper that we cited a few sentences previously. We now refer to the Clark paper again in this sentence and explain that this could be a dilution effect. Of course, the reviewer is quite correct in pointing out that numerous other environmental factors could influence the DOC seasonality in each river, and our intention was not to try and argue for one factor over another (hopefully this is now clearer in the manuscript). However, because the Maludam catchment did appear to experience particularly high precipitation shortly before we sampled (as is apparent in Fig. 1), we feel that it is appropriate to point out that this might have influenced the apparent seasonality we recorded in this particular river.

The authors may consider mentioning the conservative mixing model in the abstract and conclusions since the model was used to "validate" their DOC and CDOM measurements in the result for several times. In addition, it's better to describe the advantages or reasons to include this model in this work, so far, it looks weakly linked to other parts.

We already implicitly refer to the mixing models in the abstract when we state that “DOC and CDOM showed conservative mixing with seawater”. Because the abstract is already around 275 words, we have decided not to include more information here.

Calculating conservative mixing models is basically a standard practice when studying biogeochemical fluxes from rivers into seawater across the estuarine mixing zone. We never actually use the result of our mixing models to validate our results, we use them instead to show whether or not our results are consistent with DOC and CDOM mixing conservatively, or whether non-conservative addition/removal is happening in the estuaries. This can’t be done properly without actually calculating the mixing models. Given that it is an important objective of our study to determine whether or not the DOC and CDOM are mixing conservatively in these estuaries, the mixing models are actually integral to our analysis, which is why we show the curves of all our mixing models in Figures 2 and 4. These models essentially show us the theoretically expected changes in DOC and CDOM with salinity if all of the DOC and CDOM are mixing fully conservatively. In most cases we find that our data are consistent with these theoretical predictions, except in the Rajang. These conclusions cannot be properly supported without showing the theoretical mixing lines from the mixing models.

Regarding Table 1, please keep the font size and typeface consistent and change “” to “×”.*

Done

It would also be valuable to provide more information on the six rivers regarding their size, length, drainage basin, discharge, etc.

We have added some additional information about the rivers to Section 2.1, especially the approximate lengths of the rivers. Other manuscripts that are currently in preparation for this special issue will present more detailed information about the catchments, including estimates of the extent of peat soils and plantations in the catchments. Unfortunately, there are no readily available datasets for the areal extent of most of the drainage basins, the exact proportion of peat soils in each basin, or river discharge. These estimates are still being put together by other groups, so the information will ultimately be available within the special issue, but the data are not yet finalised enough to be summarised here. Since the objective of our manuscript is mostly to understand the distribution of DOC and CDOM across the river-to-seawater gradients and examine the biogeochemical processing of this DOM, this information is clearly not critical for an understanding of our paper.

Distribution and cycling of terrigenous dissolved organic carbon in peatland-draining rivers and coastal waters of Sarawak, Borneo

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Abstract. South-East Asia is home to one of the world's largest stores of tropical peatland, and accounts for roughly 10% of the global land-to-sea dissolved organic carbon (DOC) flux. We present the first-ever seasonally-resolved measurements of DOC concentration and chromophoric dissolved organic matter (CDOM) spectra for six peatland-draining rivers and coastal waters in Sarawak, north-western Borneo. The rivers differed substantially in DOC concentration, ranging from 120–250 μmol L^{-1} (Rajang river) to 3,100–4,400 μmol L^{-1} (Maludam river). All rivers carried high CDOM concentrations, with a_{350} in the four blackwater rivers between 70–210 m^{-1}, and 4–12 m^{-1} in the other two rivers. DOC and CDOM showed conservative mixing with seawater except in the largest river (the Rajang), where DOC concentrations in the estuary were elevated, most likely due to inputs from the extensive peatlands within the Rajang delta. Seasonal variation was moderate and inconsistent between rivers. However, during the rainier north-east monsoon, all marine stations in the western part of our study area had higher DOC concentrations and lower CDOM spectral slopes, indicating a greater proportion of terrigenous DOM in coastal waters. Photo-degradation experiments revealed that riverine DOC and CDOM in Sarawak are photo-labile: up to 25% of riverine DOC was lost within five days of exposure to natural sunlight, and the spectral slopes of photo-bleached CDOM resembled those of our marine samples. We conclude that coastal waters of Sarawak receive large inputs of terrigenous DOC that is only minimally altered during estuarine transport, and that any biogeochemical processing must therefore occur mostly at sea. It is likely that photo-degradation plays an important role in the degradation of terrigenous DOC in these waters.

1 Introduction

The annual flux of terrigenous dissolved organic carbon (tDOC) from rivers into the sea is an important part of the global carbon cycle, estimated as around 0.2 Pg C y^{-1} (Dai et al., 2012). South-East Asian rivers contribute roughly 10% of this flux (Baum et al., 2007; Huang et al., 2017; Moore et al., 2011), chiefly owing to the extensive peat deposits along the coasts of Borneo and Sumatra (Dommain et al., 2014; Page et al., 2011). The rivers draining these peatlands typically carry milli-molar concentrations of DOC, and are often called "blackwater" rivers (Alkhatib et al., 2007; Baum et al., 2007; Cook et al., 2017; Moore et al., 2011; Rixen et al., 2008).

However, our understanding of the fate of tDOC in rivers, estuaries, and in the ocean, is still limited. Most tDOC is derived from soils, from which it is leached by rainwater, and it is thus rich in lignin and humic substances. Classically, these high-molecular weight, highly aromatic molecules have been assumed to be inherently refractory to degradation (Bianchi, 2011), which would imply that they should accumulate in the ocean. However, dissolved organic matter (DOM) in the open ocean does not show clear chemical signatures of terrigenous origin, which indicates that terrigenous DOM (tDOM) must be partly remineralised and chemically altered before reaching the open ocean (Bianchi, 2011; Cai, 2011). Although it is now established that tDOM is indeed partly labile to both photo-oxidation (Helms et al., 2014; Miller and Zepp, 1995; Moran et al., 2000; Spencer et al., 2009; White et al., 2010) and to microbial degradation (Fasching et al., 2014; Leff and Meyer, 1991; Moran and Hodson, 1990; Obernosterer and Benner, 2004; Stutter and Cains, 2016; Ward et al., 2013), we are still far from having a quantitative understanding of tDOM processing in estuaries and shelf seas globally. For example, some studies have reported major losses of tDOM, with 40–50% of the tDOC flux being remineralised on the Louisiana Shelf and in the Eurasian Arctic shelf sea (Fichot and Benner, 2014; Kaiser et al., 2017). Yet in contrast, recent analysis of carbon isotopes in different molecular weight fractions in the open ocean suggests that a larger proportion of oceanic DOM may have a terrigenous origin than currently thought (Zigah et al., 2017). High-resolution mass spectrometry has also identified new terrigenous biomarkers and shown that they are widely distributed throughout the oceans (Medeiros et al., 2016). Moreover, experimental work has clearly shown that some tDOM fractions are resistant to photo-degradation (Stubbins et al., 2017). Clearly, more work is needed to trace tDOM fluxes through estuaries, and to determine where, how, and to what degree tDOM is biogeochemically processed.

Because tDOM is rich in chromophoric dissolved organic matter (CDOM), optical measurements are commonly used as proxies to trace tDOM fluxes into the ocean (Chen et al., 2015; Fichot and Benner, 2012; Fichot et al., 2016; Kowalczuk et al., 2003; Osburn et al., 2016; Yamashita et al., 2011). The last decade in particular has seen significant advances in our understanding of how CDOM spectral characteristics vary between tDOM and marine DOM, and how they are affected by different biogeochemical processes (Hansen et al., 2016; Helms et al., 2008; Helms et al., 2013; Helms et al., 2014; Shank et al., 2005). As a result, CDOM spectral slope coefficients in the ultraviolet (UV) part of the spectrum have emerged as a robust way to trace tDOM fluxes across salinity gradients and to infer biogeochemical transformations of tDOM (Chen et al., 2015; Fichot et al., 2014; Fichot et al., 2016; Helms et al., 2008; Osburn et al., 2016; Stedmon and Markager, 2003).

So far, however, most studies of tDOM fluxes to the sea have focused on North America (Chen et al., 2015; Durako et al., 2010; Fichot and Benner, 2014; Fichot et al., 2014; Leech et al., 2016; Medeiros et al., 2017), Europe (Painter et al., 2018; Rathgeb et al., 2017; Stedmon et al., 2000; Yamashita et al., 2011), and the Arctic (Benner et al., 2005; Dittmar, 2004; Kaiser et al., 2017; Semiletov et al., 2016). Much less work has been conducted in South-East Asia, despite the disproportionately large fluxes of tDOC through South-East Asia's peatland-draining rivers. Most research in South-East Asia has focused on the peatlands themselves to quantify their extent, carbon stocks, and biogeochemistry (Cobb et al., 2017; Dommain et al., 2014; Gandois et al., 2013; Gandois et al., 2014; Gastaldo, 2010; Page et al., 2011), or has examined just rivers and estuaries, but not traced tDOM further beyond the coast (Alkhatib et al., 2007; Baum et al., 2007; Cook et al., 2017; Harun et al., 2016; Müller et al., 2015; Rixen et al., 2008; Wit et al., 2015). Moreover, most studies of rivers focused either on the total DOC concentration (Alkhatib et al., 2007; Baum et al., 2007; Cook et al., 2017; Rixen et al., 2008) or on water–air CO_2 fluxes (Müller et al., 2015; Müller et al., 2016; Müller-Dum et al., 2018; Wit et al., 2015). These studies have shown clearly that peatland-draining rivers in Sumatra and Borneo have amongst the highest-reported DOC concentrations from any rivers globally (up to 3,000–5,500 µmol L^{-1}, or 36–66 mg C L^{-1}). Yet surprisingly, the CO_2 fluxes out of these rivers were found to be quite low relative to the extremely high DOC concentrations, implying that most of the tDOC they carry is delivered to the sea (Wit et al., 2015). To understand the biogeochemical processing of South-East Asian tDOC, more work clearly needs to be done in coastal waters. This need is particularly urgent because most peatlands in South-East Asia have been converted to agricultural use over the past two decades (Miettinen et al., 2016), and such conversion appears to enhance riverine tDOC fluxes by destabilising the peatland C pool (Moore et al., 2013).

Here, we present what is to our knowledge the first analysis of DOC concentrations and CDOM spectral properties in six rivers and the surrounding coastal sea in the western part of Sarawak, Malaysian Borneo. Samples were collected at three different times of the year to constrain seasonal variability, and photo-degradation experiments were conducted to determine tDOM photo-lability.

2 Materials and Methods

2.1 Study region and sample collection

Three field expeditions to Sarawak were undertaken in March, June, and September 2017. Six rivers were sampled in March and September: the Rajang (~550 km length), the Maludam (~33 km length), the Sebuyau (~58 km length), the Simunjan (~54 km length), the Sematan (~15 km length), and the Samunsam (~34 km length) (Fig. 1). The June expedition sampled only the Rajang river. On all expeditions, the river estuaries and open coastal waters were sampled (Fig. 1). In September, one sample was also taken in the estuary of a seventh river, the Lundu river (94 km length). All station locations, sampling dates, and measured data are shown in Supplementary Table 1. Four of the rivers (Maludam, Simunjan, Sebuyau, and Samunsam) are blackwater rivers that drain catchments with high peatland coverage, while the Sematan and Lundu rivers

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drain catchments with limited peatland cover. The Rajang river drains mineral soils until the town of Sibu, from where it branches into multiple distributary channels (Fig. 1). The distributaries each have unique names; the main ones (Rajang, Serendeng, and Igan) are identified in Fig. 1. These distributaries are surrounded by extensive peatlands that drain directly into the distributary channels (Staub et al., 2000). Mangroves grow along the estuaries of all of the rivers. All river samples are distinguished below by river name, while marine samples are distinguished by whether they were collected in the region east of Kuching ("Eastern Region", influenced strongly by the Rajang river outflow), or in the region west of Kuching ("Western Region", influenced by the Samunsam and Sematan rivers). The Talang Islands in the Western Region (Fig. 1) are surrounded by coral reefs.

The three sampling periods corresponded to the end of the north-east monsoon (March, end of the wettest season of the year), the south-west monsoon (June, lower precipitation), and shortly before the beginning of the north-east monsoon (September, end of the drier season). Monthly precipitation across Sarawak can vary several-fold across the year, but is mostly ≥100 mm per month (Sa'adi et al., 2017). Precipitation data were obtained from weather stations in Sibu, Maludam, and Sematan. Monthly averages were calculated for the period 1999–2017, omitting the few months for Maludam and Sematan for which there were days with missing data (there were no missing data in 2017). Precipitation in 2017 was mostly within 1 standard deviation of the 1999–2017 means (Fig. 1e). It should be noted that precipitation in this region is strongly driven by small-scale convective systems; however, 2017 was overall not an unusual year in terms of precipitation. Water temperatures in Sarawak show essentially no seasonal variation (average water temperatures during all expeditions fell within 28.5–29.5° C).

To collect samples in the Rajang river and the Eastern Region, a liveaboard fishing boat was chartered for 4–7 day cruises, and all samples were filtered and preserved upon collection. All other stations were sampled from small outboard-powered boats, in which case samples were stored dark at ambient temperature in insulated boxes on board, and filtered back on land each afternoon/evening. All samples were collected within the upper 1 m using either a bucket or a hand-held jug; sampling devices were rinsed thoroughly with sample water before sampling. Sample water was decanted into either amber borosilicate glass bottles (DOC and CDOM) or HDPE bottles (chlorophyll and total suspended solids).

DOC and CDOM samples were filtered through 0.2-µm pore-size Anodisc filters (47-mm diameter) using an all-glass filtration system that was rinsed with 1 M HCl and ultrapure deionised water (18.2 MΩ cm^{-1}, referred to as "DI water" below) in between each sample. Each Anodisc filter was rinsed by filtering 100–150 mL of DI water and then 50–100 mL of sample water, before a further 100–150 mL of sample water were filtered and taken as the sample. DOC samples (30 mL) were immediately acidified with 100 µL of either 25% H_3PO_4 (March expedition) or 50% H_2SO_4 (all other samples) to pH <2.0. CDOM samples (30 mL) were preserved with 150 µL of 10 g L^{-1} NaN_3, following Tilstone et al. (2001). DOC and CDOM samples were stored in amber borosilicate vials with PTFE-lined septa at +4° C until analysis (within 1.5 months of collection), although some river samples in September froze for 1-2 days due to a refrigerator malfunction in the field. However, freezing did not appear to affect the DOC or CDOM results, as seen from comparing DOC and CDOM data for samples from adjacent stations in the Maludam river that did and did not freeze.

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Samples (50–1000 mL) for chlorophyll-*a* were filtered onto pre-ashed (450º C, 4 h) 25-mm diameter Whatman GF/F filters, wrapped in aluminium foil, and immediately frozen in a liquid nitrogen dry shipper. They were stored in the dry shipper until analysis within 6 months of collection.

Samples for total suspended solids (TSS, 50–1000 mL) were filtered onto pre-ashed (450º C, 4 h), pre-rinsed, pre-weighed 25-mm diameter Whatman GF/F filters. Filters were rinsed three times with DI water and stored at -20º C in Petri dishes. Procedural blanks for all parameters were prepared in the field using DI water.

2.2 Chemical analyses

2.2.1 Dissolved organic carbon analysis

Dissolved organic carbon was analysed as non-purgeable organic carbon on a Shimadzu TOC-L system with a high-salt combustion kit after a 5-min sparge, using potassium hydrogen phthalate for calibration. Instrument performance was monitored using certified Deep-Sea Water from the Hansell Laboratory, University of Miami (42–45 µmol L^{-1}). Our analyses consistently yielded slightly higher values for the reference water, with a long-term mean ± 1 SD of 47 ± 2.0 µmol L^{-1} (n = 51). Procedural blanks prepared in the field almost all contained <10 µmol L^{-1}, except for those prepared in between blackwater river samples, which contained 13–27 µmol L^{-1}; a correction for these procedural blanks was not applied.

2.2.2 Chromophoric dissolved organic matter analysis

CDOM samples were warmed to room temperature and their absorbance measured from 230–900 nm against a DI water reference, using a Thermo Evolution 300 dual-beam spectrophotometer. Samples from March were measured in either a 10-cm or a 1-cm pathlength quartz cuvette, or in a 1-cm quartz cuvette after 10-fold dilution with DI water (for blackwaters). Samples from June and September were measured undiluted in either 10-cm, 1-cm, or 0.2-cm pathlength cuvettes. Instrument performance was checked according to Mitchell et al. (2000). Reagent blanks of NaN_3 in DI water were measured and subtracted from all samples. NaN_3 was found to absorb significantly from 230–265 nm, with decadic absorption coefficients of ~26 m^{-1} at 230 nm, ~4 m^{-1} at 254 nm, but ≤0.1 m^{-1} at wavelengths ≥275 nm (Supplementary Fig. 1). Blank absorbances at wavelengths ≥275 nm were nearly always <10% of sample absorbances, and mostly around 1% or less. CDOM spectra were baseline-corrected (Green and Blough, 1994), and converted to Napierian absorption coefficients following Eq. (1):

$$a_\lambda = 2.303 \times \frac{A_\lambda}{l}, \quad (1)$$

where a_λ and A_λ are, respectively, the absorption coefficient and the absorbance at wavelength λ, and l is the cuvette pathlength in m. These calculations were performed using the R package hyperSpec (Beleites and Sergo, 2018). Our raw CDOM spectra (as decadic absorption coefficients) are shown in Supplementary Table 1, and representative spectra are shown in Supplementary Fig. 2. CDOM spectral slope coefficients were calculated for the intervals 275–295 nm and 350–

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400 nm using linear regression of log-transformed data as in Helms et al. (2008). Specific UV Absorbance at 254 nm ($SUVA_{254}$) was calculated from the decadic absorption coefficient at 254 nm and DOC concentration in mg L^{-1}. Dissolved inorganic constituents such as bromide, sulphide, nitrate, iodide, and molecular oxygen have negligible absorbance at wavelengths ≥250 nm compared to CDOM (Fally et al., 2000; Guenther et al., 2001).

2.2.3 Conservative mixing models for DOC and CDOM

Two-endmember mixing models for DOC and CDOM were calculated for the Rajang, Samunsam, and Sematan rivers, and in September also for the Maludam river. For the other rivers/seasons, there was either insufficient variation in salinity (Maludam and Simunjan), or the salinity was influenced strongly by adjacent rivers that were not sampled (Sebuyau, which drains into the Lupar river estuary). Linear mixing models were calculated from the end-member DOC concentrations and CDOM spectra at salinity intervals of 1.0 from salinity 0 (river water) to the salinity of the marine end-member station (29–32.5). For CDOM, we calculated the full absorption spectrum at each interval and then re-calculated the spectral slopes and $SUVA_{254}$ values, following Stedmon and Markager (2003). It should be noted that conservative mixing of CDOM results in non-linear changes in spectral slopes and SUVA with salinity. Appropriate end-member stations were identified from salinity, DOC, and geographical location (Supplementary Table 1). In March, rough seas prevented us from sampling fully marine waters in the Eastern Region, so the marine end-member station from the June expedition was used instead.

2.2.4 Chlorophyll-*a* and total suspended solids analysis

Chlorophyll samples were extracted in 10 mL 90% acetone at -20º C in the dark for 24 h, and fluorescence measured at excitation 436 nm / emission 680 nm (both with 5 nm bandpass) on a Horiba Fluoromax 4 spectrofluorometer (Welschmeyer, 1994). The fluorescence signal was normalised to the excitation lamp reference intensity and calibrated against a chlorophyll-*a* standard from spinach (Sigma-Aldrich, C5753). The limit of detection (3 SD of the blank) was <2 ng chlorophyll per filter.

TSS samples were dried at 75º C for 24 h before re-weighing. In March and September, they were then ashed at 450º C for 1 hour to remove organic matter and weighed again to determine inorganic weight. All weighing was performed on a Mettler-Toledo microbalance with ±1 µg accuracy.

2.3 Photo-degradation experiments

Four short-term photo-degradation experiments were conducted in the field (one in June, three in September). For each, 1 L of sample water was filtered as for DOC and CDOM samples (using multiple Anodiscs if necessary), and filled into 150 mL acid-washed quartz bottles with ground quartz stoppers, leaving a headspace to prevent O_2 limitation. Dark bottles were wrapped in aluminium foil and black plastic sheets. One dark and one light control bottle were filled with DI water and treated the same way as sample bottles to check for any systematic contamination (which was not found). All bottles were secured inside a clear, open, plastic food storage container and exposed to natural sunlight on the roof of the boat (Rajang

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and seawater experiments) or in an open clearing on land (Samunsam experiment; incubations were completely unaffected by shade between at least the hours of 08:00 to 16:00). To moderate the temperature during sun exposure, the container was filled with clear seawater to the same level as the samples in the bottles.

Experiments were run for 3–6 days. Solar radiation was monitored at varying intervals throughout each day with the reference irradiance sensor of a Trios RAMSES hyperspectral radiometer (318–800 nm at 2-nm resolution). We integrated the measured irradiance from 318–450 nm (*i.e.* the portion of the spectrum with highest CDOM absorption), and then integrated this irradiance over time for each day, using exponential averaging to interpolate across measurement gaps, to estimate the cumulative irradiance from 318–450 nm, in J m^{-2}, that samples were exposed to throughout each experiment. This allowed us to account for differences in light intensity between days and between experiments by plotting the observed changes in DOM against actual irradiance, instead of just as a function of time. However, owing to the large diameter and complex geometry of our quartz bottles, the diurnal change in sunlight angle, and the presence of the plastic container, we could not estimate absorbed light doses reliably enough to calculate apparent quantum yields of DOM photo-degradation. We therefore used the estimated irradiances to help us understand qualitatively how CDOM and DOC changed during sunlight exposure.

During the first three experiments, the radiometer was installed adjacent to the exposed samples and run throughout the experiments. During the experiment with Samunsam river water, the radiometer was in use on board the sampling boat while the samples were being exposed on land; however, there was only little cloud cover during those days, and this was evenly distributed across land and sea. The Samunsam photo-degradation experiment was then continued for an additional three days, during which no radiometer measurements could be taken. To estimate the approximate irradiances for these days, the amount of cloud cover on each day was noted, and radiometer measurements were taken from previous days that approximately matched the cloud conditions. The integrated irradiance for the Samunsam experiment is therefore less well constrained than for the other experiments, but since we are not attempting to quantitatively relate DOC degradation to absorbed photon dose, these uncertainties do not compromise our conclusions about the photo-lability of tDOM in Sarawak.

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3 Results

3.1 Concentrations of DOC

Concentrations of DOC differed significantly between rivers, with highest freshwater concentrations (salinity = 0) in the Maludam (3,100–4,400 µmol L^{-1}), and lowest concentrations in the Rajang (120–250 µmol L^{-1}) (Fig. 2a–e). Seasonal differences were clearly apparent in the river samples, but did not show a consistent direction: the Sematan and Samunsam rivers carried ≥50% higher DOC concentrations in September than in March, the Sebuyau river had marginally higher concentrations in September than in March, while the Maludam and Simunjan rivers had 20%–40% lower DOC

concentrations in September than in March. Seasonality in the Rajang river is not apparent in Fig. 2a, possibly because of variability between distributary channels.

In March, DOC concentrations at freshwater stations in the Maludam and Sebuyau increased with distance downstream, but decreased slightly in the Simunjan (Fig. 3a–c). In September, DOC concentrations varied little with distance downstream in these three blackwater rivers (Fig. 3a–c). In the Rajang river, DOC concentrations at salinity 0 in each individual channel were somewhat higher in March than in September (Fig. 3g–i; June data are not shown because only one station had salinity 0 in June). Notably, DOC concentrations in the Rajang delta increased substantially with distance downstream in each of the distributary channels (Fig. 3g–i), with concentrations doubling to around 240 μmol L^{-1} during passage through the northern-most distributary, the Igan.

DOC concentrations decreased with increasing salinity in all river estuaries (Fig. 2a–e). In the Samunsam, Sematan, and Maludam rivers this decrease closely followed the predictions from a two-end-member mixing model, with the single sample from the Lundu river plotting very close to the Sematan mixing model. For the Sebuyau river, a conservative mixing model could not be constructed, because it discharges into the estuary of the larger Lupar river (Fig. 1), for which the freshwater end-member DOC concentration was not measured. For the Simunjan river, and for the Maludam river in March, too few data from brackish waters were available to construct a reliable mixing model.

In contrast, the DOC concentrations in the Rajang delta do not fit the conservative mixing models well: most of the March and June data from brackish stations have higher DOC concentrations than expected, and in fact are closer to predictions from the September mixing model (Fig. 2a). The September mixing model was calculated using the DOC concentration of the two northern-most stations in the Igan distributary as the freshwater end-member, which had the highest DOC concentrations of any of the Rajang river stations.

Some seasonality in DOC concentration was seen at the stations furthest off-shore, which had the highest salinities (29.0–32.5). This was most evident in the Western Region: in March, the stations with highest salinities (28.9 and 29.0) contained 93 and 87 μmol L^{-1} DOC, while in September the same stations had 76 and 78 μmol L^{-1} DOC, and salinities >32.0. In the Eastern Region, the highest off-shore salinities in June were 30.2–32.1 with DOC concentrations of 81–99 μmol L^{-1}, while in September the DOC concentrations were lower at 76–83 μmol L^{-1} with salinities of 29.7–31.6 (except for one station with 88 μmol L^{-1} DOC).

3.2 Spectral characteristics of CDOM

3.2.1 CDOM absorption coefficient

CDOM concentrations, quantified as a_{350}, were high throughout our study region. Nearly all samples in the blackwater rivers had a_{350} values >50 m^{-1}, with samples from the Maludam reaching 210 m^{-1} (Fig. 2f–j). Lower values were found in the Rajang and Sematan rivers, between 3–11 m^{-1}. The lowest a_{350} value (0.23 m^{-1}) was found in the furthest off-shore station in the Western region in September. The mixing behaviour of a_{350} closely mirrored that of DOC in each of the rivers (Fig. 2).

3.2.1 Spectral slopes

The CDOM spectral slope from 275–295 nm ($S_{275–295}$) was low in all freshwater samples, ranging from 0.0102–0.0144, and increased with salinity to a maximum of 0.0254 (Fig. 4a–e). $S_{275–295}$ was somewhat lower in most of the blackwater samples (Samunsam, Maludam, Sebuyau, and Simunjan) than in the Rajang and Sematan. Seasonal differences were clearly seen in the marine samples in the Western Region, with $S_{275–295}$ in March always below 0.0200, but up to 0.0254 in September (Fig. 4b). In the Rajang distributaries, $S_{275-295}$ was lower in March than in September (Fig. 3j–l), but no clear seasonality was seen in the Eastern Region marine samples (Fig. 4a), although we were unable to collect many marine samples in March due to rough seas.

In the Samunsam and Maludam rivers, $S_{275–295}$ closely followed the conservative mixing models, but this was not the case in the Rajang and the Sematan rivers (Fig. 4a–c). In the Rajang, samples at salinities 3–20 typically had higher $S_{275–295}$ than predicted by the mixing models (except in June), while many samples at salinities >20 had $S_{275–295}$ values that were lower than predicted by the mixing models. Similarly, in the Sematan river in March, samples in brackish water up to salinity 20 showed higher $S_{275–295}$ than expected from conservative mixing. In September, we were unable to sample fully freshwater in the Sematan river owing to the timing of the tides, so the freshest sample still had a salinity of 3.4, and may therefore have already been affected by any non-conservative processes in the estuary. However, all samples in the Western Region with salinities >25 fell very closely between the conservative mixing lines for the Sematan and Samunsam rivers.

The spectral slope from 350–400 nm ($S_{350–400}$) showed more complex trends: freshwater samples had values mostly between 0.014 and 0.018, while brackish and fully marine waters spanned a greater range of 0.0076–0.0206 (Fig. 4f–j). The marine end-member stations in the Eastern and Western Regions both had lower $S_{350–400}$ than the river end-members in September, but had higher values (Western Region) or nearly identical values (Eastern Region) in March and June. In the Rajang and Sematan rivers, $S_{350–400}$ showed conservative mixing up to salinities of 20–25, but was lower than predicted by conservative mixing in the blackwater Samunsam and Maludam rivers (Fig. 4f–h). At salinities >20–25, many samples clearly depart from the conservative mixing models, except for samples in March in both regions.

However, the spectral slope ratio, S_R (the ratio of $S_{275–295}$ to $S_{350–400}$), showed trends very similar to $S_{275–295}$, *i.e.* low values in river waters (0.601–0.867) and higher values in marine waters with salinity >25 (0.786–2.33, Fig. 4k–o). In brackish waters, S_R was typically slightly greater than predicted by the conservative mixing models, especially in March in the Rajang, Sematan, and Samunsam rivers, and in September in the Rajang, Samunsam, and Maludam rivers (Fig. 4k–m).

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3.2.2 Specific UV absorbance

The specific UV absorbance at 254 nm ($SUVA_{254}$) was higher in river samples (3.08–6.89 at salinity 0) than in marine samples (0.81–5.00 at salinity >25), and decreased with salinity for all rivers and seasons (Fig. 4p–t). $SUVA_{254}$ was somewhat higher in the Rajang and in the Simunjan in March than in June or September, but otherwise seasonal differences in the rivers were not pronounced. However, as for the other CDOM parameters, there was a clear difference between March

and September in the marine samples from the Western Region (Fig. 4q). The data from the Maludam, Sematan, and Samunsam rivers closely followed the conservative mixing lines, while in the Rajang, the majority of brackish samples with salinity >20 showed somewhat greater $SUVA_{254}$ than expected from conservative mixing (Fig. 4p–r). Because sodium azide contributes a relatively high blank absorbance at 254 nm but not beyond 270 nm, we compared our $SUVA_{254}$ values to the specific UV absorbance at 280 nm, $SUVA_{280}$, for each sample. We found a very strong, linear relationship between $SUVA_{280}$ and $SUVA_{254}$ for the entire dataset across rivers and seasons, with $SUVA_{280} = 0.792 \times SUVA_{254} - 0.0141$ ($r^2 = 0.990$, $p < 0.001$, $n = 154$), suggesting that our $SUVA_{254}$ measurements were not compromised by the NaN_3 blank (NaN_3 has no absorbance at 280 nm, Supplementary Fig. 1).

3.2.3 Relationships between DOC and CDOM

The CDOM concentration, as a_{350}, was closely related to the DOC concentration for the entire dataset (Fig. 5a). $S_{275\text{-}295}$ was also strongly related to DOC, though with somewhat greater scatter at DOC concentrations greater than about 150 µmol L^{-1} (Fig. 5b). Consequently, there was also a strong relationship between $S_{275\text{-}295}$ and a_{350}, although also with more scatter wherever $a_{350} > 10$ m^{-1} (Fig. 5c). The increased scatter in Figs. 5b,c at high DOC and CDOM concentrations is due to the fact that the rivers differed somewhat in $S_{275–295}$: in particular, the Rajang, Sematan, and Simunjan had higher $S_{275–295}$ for a given DOC or CDOM concentration than the Samunsam and Maludam. There was no seasonal variation in any of these relationships, inasmuch as the datasets from all three seasons plot along a single trajectory in all three plots, rather than segregating into parallel trajectories by season.

3.3 Photo-degradation of DOC and CDOM

DOM from the Rajang and Samunsam rivers was photo-labile, with DOC and CDOM decreasing after sunlight exposure. In contrast, marine water collected in the Eastern Region only showed some changes in CDOM, but no decrease in DOC (Fig. 6, Table 1). Daily irradiances, integrated from 318–450 nm, ranged from 0.92 to 3.00 MJ m^{-2}, with cumulative irradiances for each experiment ranging from 5–11 MJ m^{-2}. Irradiance data for each day are shown in Supplementary Fig. 3. In practice, plotting our data against estimated cumulative irradiance showed the same trends as plotting simply against time of exposure (Supplementary Fig. 4), although we estimate that the Samunsam water received a slightly higher irradiance over five days than the marine water over six days, and that the two Rajang experiments differed by about 20% in irradiance despite both lasting three days.

The Rajang water in June lost 16.1 ± 0.5 µmol L^{-1} DOC by the end of the experiment (mean ± 1 SD, representing 8.8%–9.4% of the starting DOC), with a_{350} decreasing as well. $S_{275–295}$ and S_R both increased, while $S_{350–400}$ remained essentially unchanged, and $SUVA_{254}$ decreased slightly (Fig. 6). In September, we found very similar changes in the Rajang water after sunlight exposure: 18.9 ± 6.1 µmol L^{-1} DOC were lost (mean ± 1 SD, representing 5.6%–10.7% of starting DOC), a_{350} decreased, and $S_{275–295}$ and S_R increased by amounts similar to June. Although $S_{350–400}$ decreased slightly relative to the initial sample, there was no difference between light and dark bottles in this parameter. $SUVA_{254}$ decreased slightly in the light

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bottles, and actually increased somewhat in the dark bottles. Marine water showed no change in DOC upon light exposure, although light bottles had very slightly higher DOC concentrations than dark bottles at the end of the experiment (by 4.5–6.1 µmol L^{-1}). However, light bottles had lower a_{350} than dark bottles at the end of the experiment, and $S_{275–295}$ increased strongly due to light exposure, reaching values higher than seen in any of our environmental samples (>0.030). $S_{350–400}$ increased both in light and dark bottles relative to the initial sample, and S_R consequently dropped in the dark bottles but remained relatively constant in the light bottles. $SUVA_{254}$ decreased slightly after light exposure. The greatest effects of photo-degradation were seen in the Samunsam river blackwater, with a decrease in DOC by 432 ± 42 µmol L^{-1} (mean ± 1 SD, representing 21%–26% of initial DOC) and a large reduction in a_{350}. $S_{275–295}$ and S_R both increased, $S_{350–400}$ decreased, but $SUVA_{254}$ remained essentially unchanged (Fig. 6). Notably, DOC and a_{350} showed a linear decrease with cumulative irradiance in all three river water experiments, suggesting that more DOC could have been mineralised (and more CDOM lost) if sunlight exposure had continued.

3.4 Distributions of chlorophyll-*a* and suspended sediments

Chlorophyll-*a* concentrations were mostly <3 µg L^{-1} throughout the region, and never exceeded 5.5 µg L^{-1}, indicating quite oligotrophic conditions (Fig. 7a–e). Concentrations in the rivers at salinity 0 were always <1 µg L^{-1} except in the Simunjan (up to 3.8 µg L^{-1}), and higher values were generally found in the estuaries at salinities between 10 and 25.

Total suspended solids in the Rajang reached values up to nearly 400 mg L^{-1}, with values in the brackish waters of the Rajang delta varying mostly between 10–70 mg L^{-1} (Fig. 7f–i). More than 90% by weight of this material was inorganic. The other rivers, and the most distant marine samples, all contained far lower TSS concentrations, but the estuaries always had >10 mg L^{-1}.

4 Discussion

4.1 Distribution of DOM within and between rivers

Rivers in Sarawak clearly differ substantially in their DOM concentrations. All of the blackwater rivers had DOC concentrations above 1,200 µmol L^{-1}, with highest values in the Maludam river. These results are consistent with previous measurements in the Maludam (Müller et al., 2015), and in other blackwater rivers in South-East Asia (Alkhatib et al., 2007; Cook et al., 2017; Harun et al., 2016; Moore et al., 2011; Rixen et al., 2008; Wit et al., 2015), but they are high compared to DOC measurements in blackwaters from other continents, which are typically below 2,000 µmol L^{-1} (Lawrenz et al., 2010; Leech et al., 2016). The Maludam, Sebuyau, and Simunjan river drain peatlands along most of their catchments (Müller et al., 2016), while the Samunsam river drains an extensive area of peatland in its upper reaches. The lower DOC concentrations in the Rajang, Sematan, and Lundu rivers are closer to concentrations reported from the Lupar and Saribas (mostly <500 µmol L^{-1}), the two larger rivers that flank the Maludam peat dome (Fig. 1) (Müller et al., 2016). The Rajang

river drains mineral soils along most of its catchment, and peatlands (up to several metres thick) are only found in the delta surrounding the distributaries (Gastaldo, 2010; Staub et al., 2000). The Sematan and Lundu river catchments also have at most limited peat deposits. The pronounced increase in DOC concentration with distance downstream in the three main Rajang distributary channels clearly shows that there are large organic matter inputs into the distributaries. Chlorophyll-*a* concentrations in the Rajang were very low, and no traces of aquatic macrophytes were seen in any of the rivers; given the very low light penetration due to suspended sediments and CDOM (see Section 4.3.2), benthic primary production is likely to be at most minimal. We therefore rule out a significant autochthonous DOC source. Bacterial solubilisation of particulate organic carbon (POC) is a possible *in-situ* source of DOC, but our CDOM data did not indicate a substantial bacterial DOM source (see Section 4.4). Photochemical POC solubilisation could also produce DOC *in situ* (Kieber et al., 2006; Mayer et al., 2006). However, estimated rates of photochemical POC solubilisation under realistic conditions of light penetration are only around 4–6 mmol m^{-2} d^{-1} (Kieber et al., 2006; Riggsbee et al., 2008), which is too low to explain the DOC increase we observed, given the likely short transit time of water through the Rajang delta (see Section 4.3.2). Instead, the DOC input most likely originates from the peatlands in the delta. Peatlands are found throughout the delta, but are most extensive and deep along the Igan distributary (Gastaldo, 2010; Staub et al., 2000), and the Igan also showed the greatest increase in DOC with distance downstream of all the Rajang distributaries, consistent with our hypothesis of a peatland DOM source to the Rajang delta. However, future work should explicitly address the possibility of POM solubilisation to DOM in South-East Asian rivers.

None of the river catchments consist of genuinely pristine peat swamps: much of the peatland surrounding the Simunjan, Sebuyau, and Rajang rivers has been converted to oil palm plantations, and even the less impacted Maludam and Samunsam catchments have been disturbed to some degree by logging. Human disturbance has been shown to increase the loss of DOC from peatlands at field sites in central Borneo (Moore et al., 2013), and depending on water table height, peatland oil palm plantations in Sarawak can also export old DOC (Cook et al., 2018). Differences in DOM quality between agricultural and natural peatland sites on Borneo have also been noted (Harun et al., 2016; Materić et al., 2017). While it is thus clear that land-use can impact DOC export from South-East Asian peatlands, our study was not designed specifically to determine the effect of land-use on DOM concentrations or quality in the rivers; more field work would be needed to do so.

Our CDOM data show that all rivers were characterised by high levels of tDOM, with the blackwater rivers in particular having extremely high absorption coefficients, very low $S_{275-295}$, and high $SUVA_{254}$. $S_{275-295}$ is now well established as an optical tracer for tDOC in estuarine and marine waters (Fichot and Benner, 2011, 2012; Helms et al., 2008), and is inversely related to the mean molecular weight of DOM in a sample (Helms et al., 2008). $SUVA_{254}$ is positively related to the aromaticity of DOM (Traina et al., 1990; Weishaar et al., 2003). Our values for $S_{275-295}$ and $SUVA_{254}$ in the rivers are, respectively, on the low and the high end of values reported from other freshwaters, especially in our blackwater rivers (Fichot and Benner, 2014; Helms et al., 2008; Leech et al., 2016; Massicotte et al., 2017). These river systems in Sarawak are therefore characterised by DOM with high average molecular weight and high aromaticity, consistent with a terrigenous

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rather than aquatic source. Moreover, even the clearest marine waters that we sampled had $S_{275–295}$ no greater than about 0.025, which is consistent with some of the DOM in these waters having a terrigenous origin.

Riverine DOC concentrations do not always show a simple relationship with precipitation and river discharge, especially in peatland-draining rivers (Clark et al., 2007). Previous DOC measurements in the Saribas and Lupar rivers in Sarawak did not show a consistent seasonality (Müller et al., 2016). Our data indicate that DOC concentrations at the end of the wettest season (March) were higher than at the end of the drier season (September) in the Rajang, Maludam, and Simunjan rivers, but by different amounts. In contrast, the Samunsam, Sematan, and Sebuyau showed lower concentrations in March than in September, but again by different amounts between the rivers. These differences between rivers probably reflect differences in catchment hydrology rather than variation in weather conditions, given that precipitation in 2017 was mostly close to the 18-year mean. However, the Maludam experienced unusually high precipitation in September, with nearly all of the excess precipitation relative to the mean falling during two consecutive days and less than one week before our sampling. It is possible that this high precipitation could have had a dilution effect (Clark et al., 2007) and contributed to the lower September DOC concentrations in this river. Because the Maludam is a very small river, the precipitation record from Maludam village is likely to reflect rainfall across the Maludam catchment. This is unlikely to be the case for the Rajang river; therefore, the elevated February precipitation in Sibu would probably not have affected our March data for the Rajang.

Overall, our data clearly show that there is inconsistent seasonality in riverine DOC concentrations across Sarawak, while precipitation is relatively high year-round. Sarawak is thus clearly characterised by high tDOC fluxes to sea in all seasons, as reflected by the low $S_{275-295}$ values at all of our marine stations. Nevertheless, we found a stronger terrigenous signal during the north-east monsoon in the Western region marine samples, although we cannot say whether this reflects a seasonal change in the magnitude of the tDOC flux to sea, or perhaps a seasonal difference in the degradation rate of tDOC at sea. For example, rougher, more turbid seas and greater cloud cover during the north-east monsoon might reduce the solar irradiance underwater and thus reduce the rate of photo-degradation.

Despite the fact that there are large differences in CDOM concentration and some differences in CDOM spectral properties between rivers, we found very strong relationships between DOC concentration, a_{350}, and $S_{275–295}$ across our entire dataset. Unlike, for example, in the northern Gulf of Mexico (Fichot and Benner, 2011), there was no seasonal variability in these relationships. This suggests that there are no strong seasonal changes in tDOM composition within our study region, despite the seasonal concentration differences discussed above.

4.2 Photo-lability of riverine DOM

Our experiments clearly showed that DOM in Sarawak rivers is photo-labile, with both DOC mineralisation and substantial changes to the CDOM absorption spectrum occurring within days of sunlight exposure. The linearity of DOC loss with cumulative irradiation in our river samples suggests that our experiments were too short for all photo-labile DOC to be lost; our results are therefore an under-estimate of the total proportion of tDOC in Sarawak rivers that can potentially be photo-mineralised (of course, it must be noted that the *rate* of tDOC photo-oxidation in our incubations was likely much higher

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than the true *in-situ* rate of photo-oxidation, at least in the highly light absorbent rivers). Only the marine sample did not lose DOC upon exposure to sunlight, although this sample did show a significant increase in $S_{275-295}$ to higher values than found at any of our stations. This increase in spectral slope for marine CDOM further suggests that there was a terrigenous fraction of DOM even at those stations farthest off-shore.

High photo-lability of tDOM has been shown in many cases, with freshwater samples from North America (Gao and Zepp, 1998; Helms et al., 2008; Helms et al., 2014; Miller and Zepp, 1995; White et al., 2010), Africa (Spencer et al., 2009), and the Arctic (Stubbins et al., 2017) showing loss of DOC and changes in absorption spectra upon solar irradiation. In some cases, however, riverine tDOC was found to be resistant to photo-degradation, possibly because the photo-labile fraction had already degraded upstream of the sampling site (Chupakova et al., 2018). Phytoplankton-produced DOC and marine surface-water DOC are typically not photo-mineralised (Obernosterer and Benner, 2004; Ziegler and Benner, 2000), although DOC from aphotic deep-sea samples has been shown to be photo-labile (Helms et al., 2013). The changes that we observed in all our samples in $S_{275-295}$ (pronounced increase) and in $S_{350-400}$ (decrease or no change relative to dark bottles) upon sunlight exposure, and the resulting increases in S_R, are also consistent with photo-degradation studies of tDOM in other regions (Helms et al., 2013; Spencer et al., 2009). Our data thus validate these optical measures as indicators of photo-degradation also in South-East Asia. Interestingly, the Samunsam blackwater sample did not show a change in $SUVA_{254}$ upon solar irradiation. Given the very high $SUVA_{254}$ of this sample, it is possible that the contribution of aromatic molecules to the total DOC was so high that the degradation of aromatic moieties was proportional to the overall loss of DOC during photo-exposure.

Our experimental results thus indicate that tDOM in rivers in Sarawak contains a significant photo-labile fraction that is partly photo-modified and partly photo-mineralised. Because South-East Asian peatlands are predominantly found in coastal lowlands (Dommain et al., 2014), peatland-derived DOM probably has too short a residence time in rivers for significant photo-degradation to occur in the rivers before it reaches the sea, unlike in some Arctic rivers (Chupakova et al., 2018). We therefore suggest that most photo-chemical transformations of tDOC in Sarawak likely take place after tDOC reaches the sea, rather than inside the rivers and estuaries.

4.3 Mixing of riverine DOM with marine water

4.3.1 Conservative mixing

In the Maludam, Samunsam, and Sematan estuaries, DOC and most CDOM parameters showed conservative mixing between river water and seawater. Conservative behaviour of tDOM is often reported from estuaries elsewhere (Chen et al., 2015; Kowalczuk et al., 2003; Rochelle-Newall and Fisher, 2002; Yamashita et al., 2011), including the few South-East Asian rivers that have been studied to date (Alkhatib et al., 2007; Baum et al., 2007; Rixen et al., 2008). Even though tDOM is now recognised as being less refractory than previously assumed (Bauer et al., 2013; Bianchi, 2011; Cai, 2011), as also

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shown by our photo-degradation experiments, the removal time-scales of tDOM by photochemical, biological, and other processes are clearly longer than the transit times through these river systems.

The marine waters in the Western Region showed a clear seasonal difference in CDOM spectral characteristics and salinity. The only significant rivers in this region are the Samunsam, Sematan, and Lundu. In spite of their small size, these rivers clearly deliver enough tDOM to measurably impact the optical characteristics of the DOM pool in coastal waters 10–20 km beyond the river mouths, including on the coral reefs surrounding the Talang Islands. Even our highest values of $S_{275-295}$ are actually on the low end of values reported from marine samples (usually 0.020–0.050, Stedmon and Nelson (2015)), suggesting that tDOM contributes significantly to the total DOM pool in coastal waters in Sarawak. Beyond the immediate river plumes, these coastal waters have low concentrations of suspended sediments and of chlorophyll, which means that tDOM delivery might act as an important control over the underwater light availability in this region, and thus over coastal productivity (Cherukuru et al., 2014; Durako et al., 2010; Lawrenz et al., 2010).

4.3.2 Possible non-conservative mixing in the Rajang

In the Rajang delta, we found evidence of non-conservative mixing, with most of the brackish waters having higher DOC concentrations than expected. We attribute this to continued input of tDOC from the extensive peatlands surrounding all of the distributaries (Gastaldo, 2010; Staub et al., 2000), since we observed increasing DOC concentration with distance downstream in each distributary. Moreover, several of the Eastern Region marine samples, with salinities >30, had higher DOC concentrations than predicted by the mixing models, possibly indicating that DOC-rich run-off from peatlands also flows directly into the coastal sea (*e.g.* from peatlands on Pulau Bruit, Staub et al. (2000)). Higher DOC concentrations than predicted by conservative mixing have sometimes been noted, for example, in the Chesapeake Bay, and attributed to *in situ* production of DOC by phytoplankton (Rochelle-Newall and Fisher, 2002). However, chlorophyll-*a* concentrations in our study region very rarely exceeded 2.5 µg L^{-1}, and were always below 1 µg L^{-1} in the Rajang river, which rules out phytoplankton production as a major source of riverine DOC.

Given this large input of likely peat-derived DOC into the Rajang Delta, one would also expect non-conservative behaviour of CDOM. Surprisingly, however, the majority of brackish samples actually had higher $S_{275-295}$ values than predicted, contrary to what one would expect from the addition of peatland-derived DOM (indeed, all other rivers had lower $S_{275-295}$ than our Rajang samples). These results suggest that there is selective removal of a high-molecular weight CDOM fraction within the Rajang delta, despite the continued input of tDOM in the distributaries. Our data from the Sematan river in March actually hint at a similar increase in $S_{275–295}$ than expected from conservative mixing, although our dataset from this river is too limited to conclude this with confidence.

Non-conservative removal of tDOM in estuaries can occur due to photo-degradation and microbial degradation, flocculation due to rising salinity, or adsorption to sediments. We hypothesise that adsorption of tDOM to suspended inorganic sediments is the most likely explanation for the non-conservative increase in $S_{275–295}$. We measured extremely high suspended inorganic sediment in all of the Rajang distributaries on all expeditions (100–360 mg L^{-1}), which is consistent with previous data

(Staub et al., 2000). Such concentrations of sediments have been shown to lower CDOM absorption coefficients and to increase the CDOM spectral slope of estuarine tDOM samples in laboratory incubations (Shank et al., 2005; Uher et al., 2001). While flocculation of tDOM due to rising salinity can occur (Sholkovitz et al., 1978; Uher et al., 2001), we would also expect this process to affect all of the other, less sediment-rich, rivers similarly, but this was not the case (except, possibly, in the Sematan estuary). Photo-degradation is unlikely to account for CDOM removal in the Rajang delta, because the high sediment loads attenuate light very strongly; Secchi depths, measured on two of our expeditions in the Rajang, were typically in the range of 10–30 cm. Although bacterial degradation of tDOM almost certainly does take place in the Rajang, the transit time of river water through the distributaries is probably too short for biological degradation to account for the observed changes: the total distance through the distributaries to the coast is around 80–120 km, and current speeds on the order of 1-2 knots were typical, implying a total transit time to sea of just a few days. Sediment adsorption, however, has been shown to alter CDOM spectra within hours to days (Shank et al., 2005; Uher et al., 2001). Müller-Dum et al. (2018) reported relatively low concentrations of CO_2 in the Rajang river and along the distributaries, with ^{13}C content of dissolved inorganic carbon indicating little contribution of peatland DOC to the dissolved CO_2 pool, which supports our interpretation of the DOC and CDOM data in this river.

4.4 Biogeochemical processing of tDOM

Both photo-degradation and microbial degradation typically cause increases in $S_{275-295}$ (Helms et al., 2014; Lu et al., 2016; Spencer et al., 2009). However, photo-degradation concomitantly reduces $S_{350-400}$, and thus increases S_R (Helms et al., 2013; Helms et al., 2014; Spencer et al., 2009), as seen in all of our photodegradation experiments. In contrast, incubation experiments suggest that bacterial processing of tDOM either has limited impact on, or actually increases, $S_{350-400}$ (Hansen et al., 2016; Lu et al., 2016). Similarly, Moran et al. (2000) found that CDOM spectral slopes over a larger wavelength range are increased by photo-degradation, but reduced by microbial degradation. The fact that S_R showed a very clear increase with salinity in all rivers, with marine samples mostly showing values >1.0, suggests that photo-degradation plays an important role in processing tDOM in this region. However, the erratic variation seen in $S_{350-400}$, with marine samples having either higher or lower values than river water, may be indicative of a role for microbial tDOM degradation as well. Typically, photo-degradation makes tDOM more labile to microbial remineralisation (Kieber et al., 1989; Miller and Moran, 1997; Obernosterer and Benner, 2004), which makes it hard to disentangle the importance of the two processes based only on CDOM measurements. While our CDOM data are consistent with an important role for photo-degradation, estimating the amount of tDOC that is actually photo-oxidised to inorganic carbon would require estimates of the water residence time, solar irradiation, and light attenuation coefficients, which are not available for Sarawak at the present time. Based on such a calculation, Fichot and Benner (2014) estimated that the majority of tDOC delivered by the Mississippi river to the Gulf of Mexico is actually remineralised by bacteria, even though the tDOC in their study region was found to be photo-labile in irradiation experiments.

Importantly, our data suggest that most of the tDOM that is lost from South-East Asia's coastal peatlands is transferred to the seas on the Sunda Shelf, where it becomes part of the global ocean circulation *via* the different branches of the Indonesian Throughflow. However, owing to the large geographical extent of the Sunda Shelf, the residence time of peatland tDOM on the shelf is likely to be at least several months to a year. During this time, the tDOM always experiences temperatures of 25–30ºC (promoting rapid microbial metabolism), and is also likely exposed to relatively high doses of solar irradiation, because these oligotrophic, tropical waters are relatively clear and shallow (mostly <100 m deep). The geographical extent and oceanography of the Sunda Shelf seas are therefore likely to strongly promote the remineralisation of tDOM before this material enters the open ocean. Remineralisation of tDOC would explain the high pCO_2 over-saturation reported by Kartadikaria et al. (2015) across the Java Sea, which is thus consistent with our results.

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5 Conclusions

We have undertaken the first seasonally-resolved study of DOC and CDOM for South-East Asia that includes peatland-draining rivers, estuaries, and coastal waters. Most of the rivers we sampled carried very high concentrations of tDOC and CDOM that showed conservative mixing with seawater in the estuaries. Non-conservative mixing was only found in the Rajang river delta, where our data point towards increasing inputs of tDOM from peatlands along the delta, but also to the removal of a high-molecular weight fraction, probably due to adsorption to sediments. Seasonality in tDOM concentrations differed between rivers, but our CDOM data showed that tDOM concentrations were higher at all marine stations in the western part of our study region during the north-east monsoon. Overall, our CDOM spectral slope coefficients are consistent with a significant contribution by tDOM to the total DOM pool even at our marine end-member stations, but also suggest that photo-degradation plays an important role in the biogeochemical processing of tDOM in Sarawak's coastal waters. This is also supported by our direct experimental evidence showing that tDOM from rivers in Sarawak is both remineralised and altered upon solar irradiation. Our results therefore suggest that much of the biogeochemical processing of peatland-derived tDOM in South-East Asia may take place in shelf seas rather than rivers.

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Table 1: Summary of results from photo-degradation experiments for DOC, $S_{275-295}$, and CDOM concentration (a_{350}) . All values are mean ± SD for each treatment. Day 0 data are the values measured at the corresponding station from which water for each experiment was taken. All irradiance data are in Joules m^{-2}, integrated from 318–450 nm.

Rajang experiment June							
	Cumulative irradiance	DOC, µmol L^{-1}		$S_{275-295}$		a_{350}	
Day		Light	Dark	Light	Dark	Light	Dark
0	0	178		0.0132		5.89	
1	1.93×10^{6}	176 ± 0.31	178 ± 1.34	$0.0155 \pm 6.4\times10^{-5}$	$0.0131 \pm 3.5\times10^{-5}$	4.88 ± 0.06	6.03 ± 0.02
3	6.53×10^{6}	162 ± 0.51	177 ± 0.07	$0.0183 \pm 1.6\times10^{-4}$	$0.0132 \pm 6.3\times10^{-6}$	3.79 ± 0.11	6.05 ± 0.02
Rajang experiment September							
	Cumulative irradiance	DOC, µmol L^{-1}		$S_{275-295}$		a_{350}	
Day		Light	Dark	Light	Dark	Light	Dark
0	0	238		0.0130		7.91	
1	1.10×10^{6}	235 ± 1.4	246 ± 4.8	$0.0138 \pm 2.0\times10^{-4}$	$0.0128 \pm 1.3\times10^{-4}$	8.00 ± 0.08	8.89 ± 0.67
3	4.72×10^{6}	219 ± 6.1	238 ± 4.6	$0.0166 \pm 3.7\times10^{-4}$	$0.0124 \pm 5.8\times10^{-4}$	6.04 ± 0.25	9.24 ± 0.64
Marine experiment September							
	Cumulative irradiance	DOC, µmol L^{-1}		$S_{275-295}$		a_{350}	
Day		Light	Dark	Light	Dark	Light	Dark
0	0	83		0.0228		0.55	
4	6.08×10^{6}	85 ± 0.8	79 ± 1.6	$0.0299 \pm 5.2\times10^{-4}$	$0.0242 \pm 9.1\times10^{-4}$	0.261 ± 0.022	0.402 ± 0.045
6	9.71×10^{6}	86 ± 1.2	81 ± 1.6	$0.0306 \pm 2.5\times10^{-4}$	$0.0245 \pm 2.3\times10^{-4}$	0.247 ± 0.006	0.385 ± 0.012
Samunsam experiment September							
	Cumulative irradiance	DOC, µmol L^{-1}		$S_{275-295}$		a_{350}	
Day		Light	Dark	Light	Dark	Light	Dark
0	0	1799		0.0109		97.6	
1	2.99×10^{6}	1640 ± 50	1730 ± 11	$0.0119 \pm 1.3\times10^{-4}$	$0.0108 \pm 3.2\times10^{-5}$	85.0 ± 2.6	93.5 ± 1.5
3	7.13×10^{6}	1535 ± 30	1781 ± 38	$0.0126 \pm 7.0\times10^{-5}$	$0.0112 \pm 5.6\times10^{-4}$	79.9 ± 2.3	89.2 ± 5.9
5	11.2×10^{6}	1366 ± 42	1779 ± 17	$0.0133 \pm 1.2\times10^{-4}$	$0.0109 \pm 7.1\times10^{-5}$	70.0 ± 3.2	96.4 ± 1.2

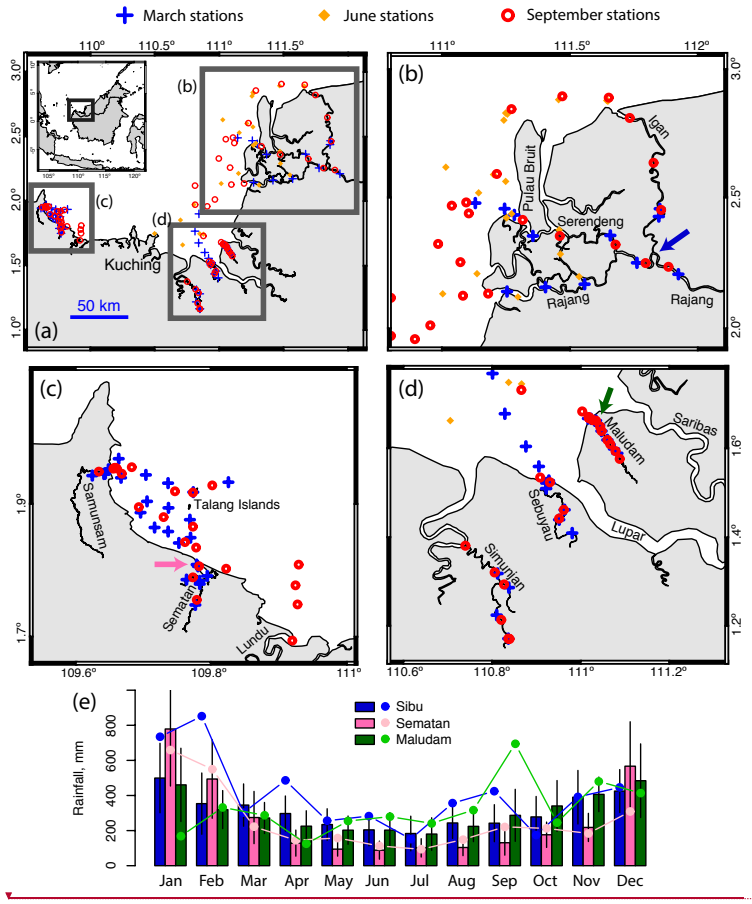

Figure 1: (a) Map of the study region showing station locations for each of the three expeditions. Thick grey boxes with letters indicate the areas shown in panels (b–d). (e) Monthly mean precipitation for the towns of Sibu, Sematan, and Maludam (locations of the rain gauges are marked with arrows in panels b, c, and d, arrow colours correspond to the bar colours in panel e). Bars show mean ± 1 SD for 1999–2017, while points show values for 2017. Bars and points for the three locations in each month are separated horizontally for better readability, but correspond to the same time periods.

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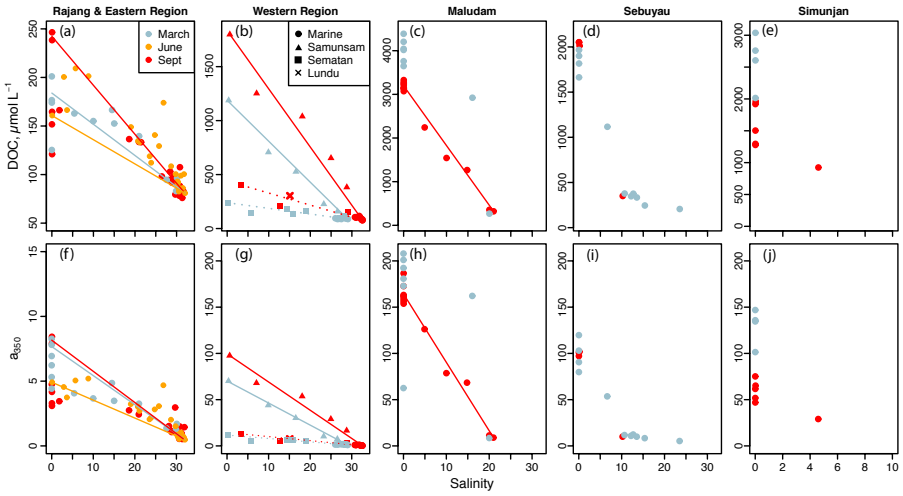

Figure 2: Changes in (a–e) dissolved organic carbon concentration, and (f–j) a_{350} from rivers to coastal seawater. Coloured lines show conservative mixing models for the data from the corresponding season. In (b) and (c), solid *versus* dashed lines distinguish the mixing models for the Sematan and Samunsam rivers in the Western Region. Data are separated by sampling region in columns, indicated by the column titles. Colours of plotting symbols are used to distinguish sampling seasons in all panels as per the legend in panel (a).

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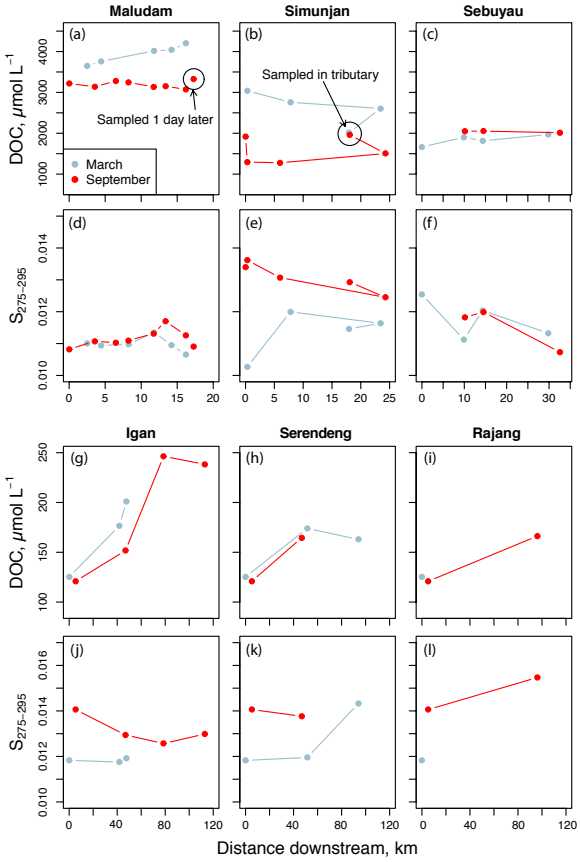

Figure 3: Changes in (a–c and g–i) dissolved organic carbon concentration and (d–f and j–l) $S_{275\text{-}295}$ with distance downstream for all stations with salinity of 0. Data in (a–f) are for the Maludam, Simunjan, and Sebuyau rivers, while (g–l) show data for the three main Rajang distributaries (named in Figure 1b): panels (h,k) show data for the Serendeng branch (includes the Lebaan and Paloh sections), while panels (i,l) show data for the Rajang branch (includes the Payang section).

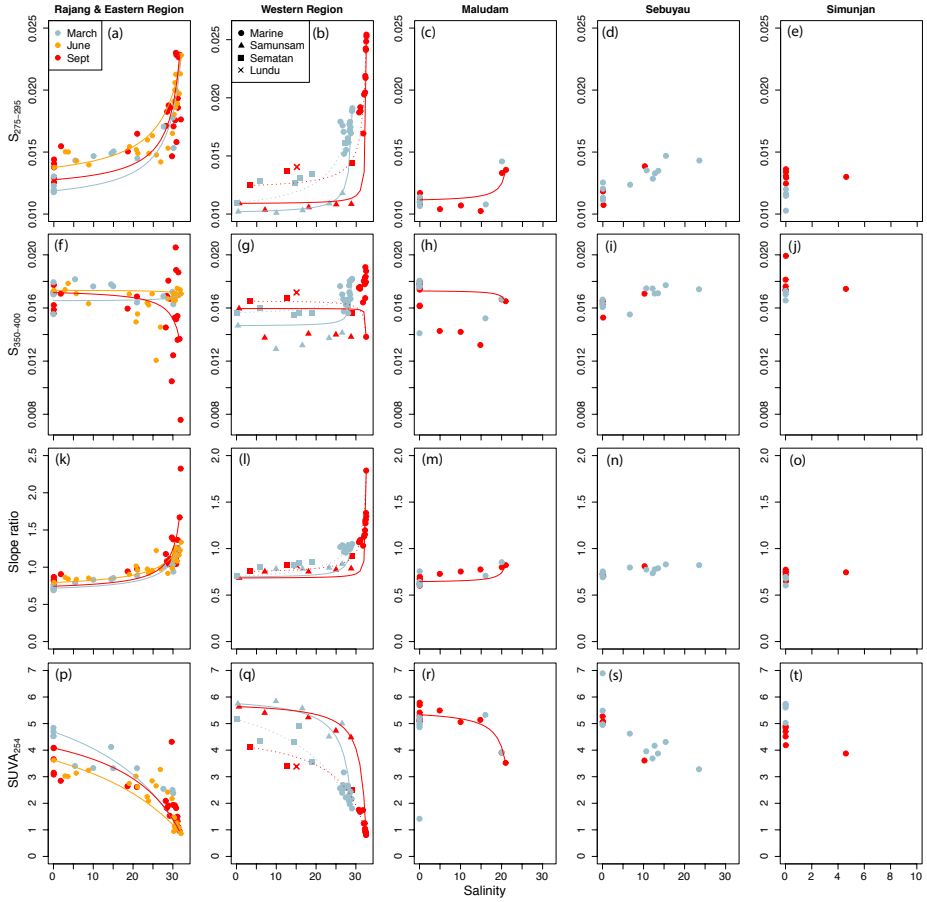

Figure 4: Changes in (a–e) $S_{275-295}$, (f–j) $S_{350-400}$, (k–o) CDOM spectral slope ratio, and (p–t) $SUVA_{254}$ from rivers to coastal seawater. Conservative mixing lines are shown as in Figure 2 (note that conservative mixing of CDOM properties is non-linear). Data are shown separately for each sampling region as indicated by column titles.

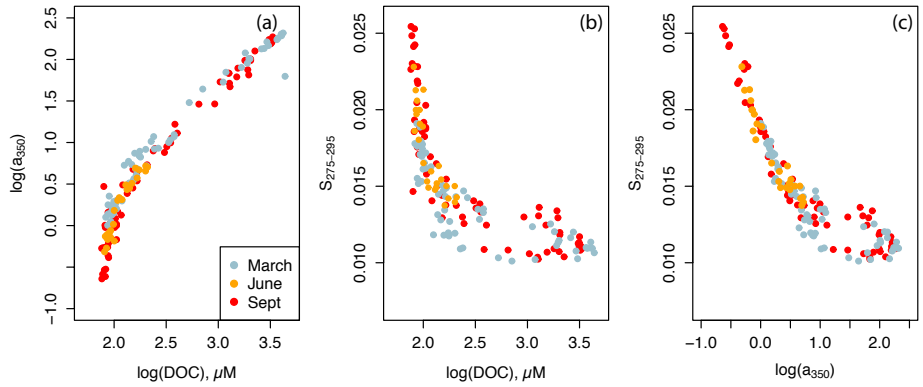

Figure 5: Scatter plots of (a) CDOM absorption *versus* DOC concentration, (b) $S_{275–295}$ *versus* DOC concentration, and (c) $S_{275-295}$ *versus* CDOM absorption for the entire dataset. Strong relationships were found between these parameters, but without seasonal variation.

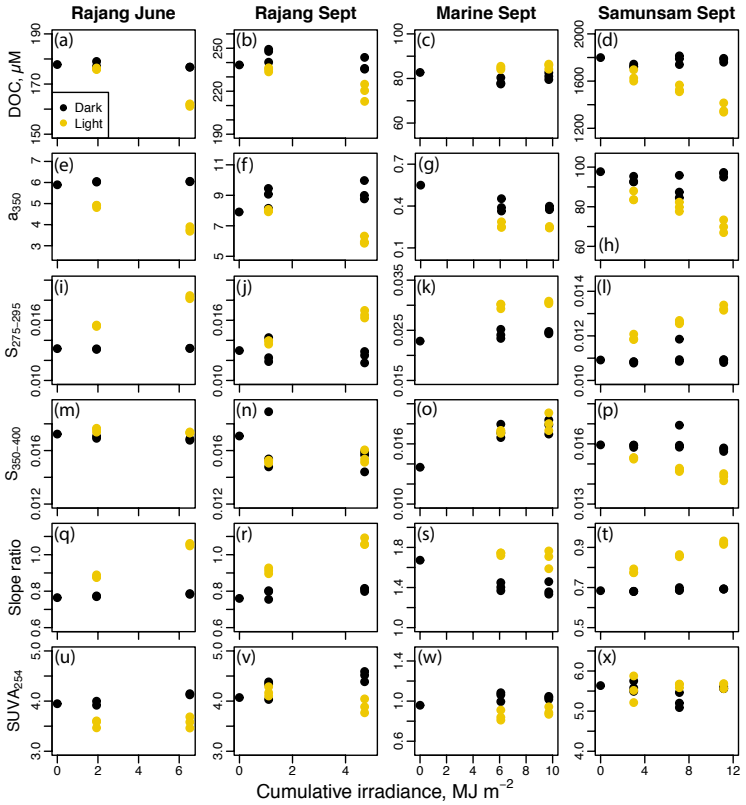

Figure 6: Results from photo-degradation experiments showing the decrease in DOC (top row), CDOM concentration (second row), $S_{275\text{-}295}$ (third row), $S_{350\text{-}400}$ (fourth row), CDOM spectral slope ratio (fifth row), and $SUVA_{254}$ (bottom row) with cumulative irradiance from 318–450 nm wavelength. Each column corresponds to one degradation experiment, as indicated in the column titles. Black symbols indicate dark control bottles, yellow symbols indicate light-exposed bottles.

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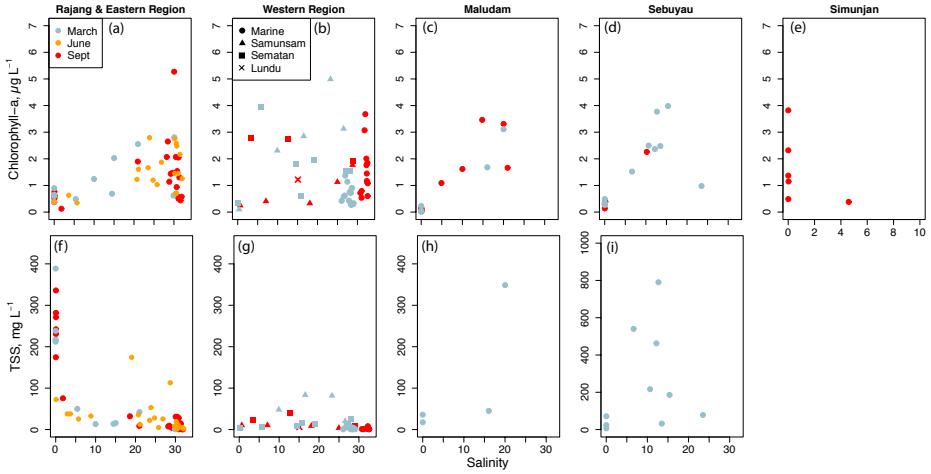

Figure 7: Distribution of (a–e) chlorophyll-*a*, and (f–i) total suspended solids from rivers to coastal seawater for each study region. TSS was not measured in the Simunjan.