

Dear editor and referees

We thank you for the time and effort expended in reviewing our manuscript. We are pleased to learn that you find our work interesting and relevant to the Biogeosciences community. We are grateful for the issues you raised to help improve the quality of our work.

Following from your comments, we have tried to address all the issues raised and we have revised the manuscript in light of the comments and recommendations. The responses to your comments and the corresponding changes in the revised version of our manuscript are itemized below:

Reviewers' comments are shown in **red** and our responses are in **blue**. Changes in the revised manuscript are highlighted in **green**

Comments from Anonymous reviewer #1

First, several caveats— - I'm on extensive travel, and don't really have time to work my way through this quite dense manuscript. - I'm not a sedimentologist/mineralogist, so not well-qualified to speak to the technical details. That said, I thought this manuscript was well-done. It laid out the issues clearly, worked through the problem setup and execution, and did a good job of working towards answering the two main questions posed

We thank reviewer #1 for their comment and are pleased to know they find the manuscript acceptable as is.

Comments from Anonymous reviewer #2

A) Nature of river sediments samples: I'm not sure what was sampled. The authors explain bank and river bed materials, and seem to suggest that these are representative of the materials sourced by erosion upstream. But what grain size range were captured by these samples? and do they represent the grain sizes carried by the river throughout its length? If they are sand-silt-clay, are these the only sediment grain sizes present on the river bed? there is no gravel? In short, what they actually tell you?

The riverine sediments were classified into two categories namely riverbed and riverbank sediments based on the following scheme:

Riverbed sediments are generally bed loads that are perennially subaqueous. These sediments were sampled from the middle of the stream with a grab sampler.

Riverbank sediments are the deposits on the flank of the river that are prone to translocation during sustained period of higher discharge (i.e. the monsoon season), as well as sediment deposited on the river margin particularly shortly after high flow events such as episodic floods and storm. In contrast to riverbed sediments, the riverbank deposits are only intermittently subaqueous, and were sampled at the river flank using a hand shovel.

The particle size of both the bed and bank sediments range from clay ($<2\ \mu\text{m}$) to very coarse sand (2 mm) with small amount of debris ($>2\ \text{mm}$) such as wood chips and small rock fragments. This larger debris was removed during dry sieving using a 2 mm mesh sieve, as mentioned in the method section.

The grain size ranges of the riverbed sediments are represent the local and immediate surroundings of the sampling location but are fairly similar through the course of the river except in some locations in the middle reach of the river that were dominated by gravels. These set of samples were not analysed in the present study.

The above information is contained in line 253-268 in the original manuscript. The revised manuscript has been amended to include the grain size information from line 258-260 and now reads as follows:

“The sampling sites were selected as being representative of the local depositional settings of the rivers and its tributaries, and mostly comprise areas dominated by bedload sediments (channel thalweg) with particle sizes ranging from $<2\ \mu\text{m}$ (clay) to 2 mm (coarse sand) with minor proportions of pebbles and plant debris.”

B) MSA control on %OC: There are a few points in the manuscript which talk about the importance of mineral surface area for organic carbon loading (e.g around line 520). But the data really don't support those statements (see Figure 4, where there are no links between these measurements and %OC is pretty similar where MSA varies much more). In contrast, the ratio of these measurements (OC/MSA) does appear to be linked to the ^{14}C activity and stable isotope composition of the organic matter. Doesn't that suggest that the mineral surface control is linked to the residence time of organic matter and its processing in the catchments? Rather than its overall abundance in the sediments? This seems an apparent contradiction which is worth exploring.

To put it another way. . .

Figure 5 shows that generally, low OC/MSA is ^{14}C -depleted (older). But low %OC is not necessarily associated with low MSA (e.g. ranging from 5 to 70 m^2/g for %OC of $\sim 0.1\%$). Doesn't that suggest that the mineral surface area control is acting as protection (allowing organic matter to age), rather than promoting substrate for sorption (line 523)?

Take the other side of the story, Figure 5 suggests high OC/MSA is younger. But high %OC is not necessarily associated with high MSA (e.g. ranging from 10 to 80 m^2/g for %OC $\sim 0.8\%$). So could this be due to an entirely different reason, decoupled from the surface area? Another explanation for this material would be that it represents organic matter not associated with any mineral – i.e. is discrete particles of organic matter, which may be more likely to be younger.

It is indeed true that at first glance, the relationship between organic carbon and mineral surface area is not immediately obvious in Figure 4. However, by

subdividing the dataset into its different sample matrices (i.e. plotting the OC:MSA relations for soils and river sediments separately), it becomes apparent that surface area exerts a significant influence on OC loading in riverine sediments ($r^2 = 0.71$, p -value <0.0001) whereas this is not the case for soil samples ($r^2 = 0.02$, p -value = 0.002; See Figure C1 below). The addition, and the larger amount, of the soils dataset in Fig. 4 has masked the good relationship between OC and MSA for the river sediments and skewed the overall dataset towards a low r^2 value ($r^2 = 0.04$).

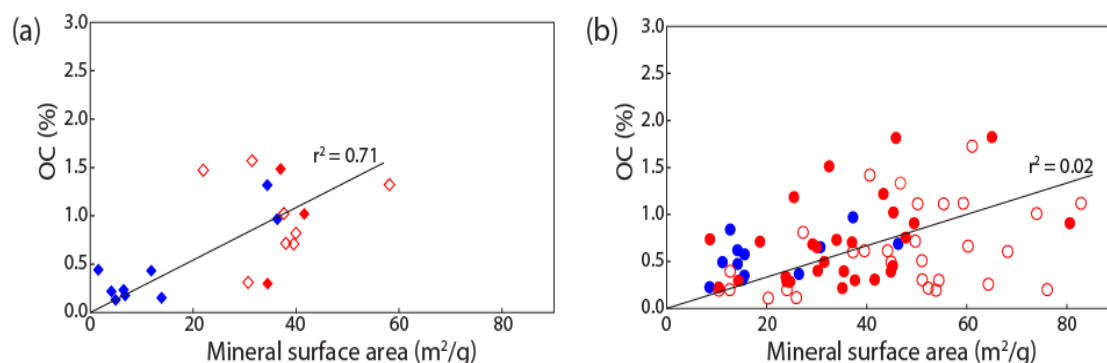


Fig. C1: OC versus MSA for riverine sediments (a) and soil samples (b) with their respective correlation coefficients

We agree with the reviewer that low OC is not necessarily associated with low MSA or vice versa. However, it should be noted that in addition to sediment mineralogy, the OC:MSA relation is also influenced by the type and reactivity of organic components that constitute the majority of the OM (e.g. Satterberg et al., 2003; Ding and Heinrich, 2002; Meyers and Quinn, 1971). Since soils represents the initial stages of OM-MSA interaction, it is possible that these initial organic-mineral associations are subject to evolution during fluvial transport. These would explain why soil samples exhibit a wider range of MSA, indicative of the contrasting geology in the Godavari basin.

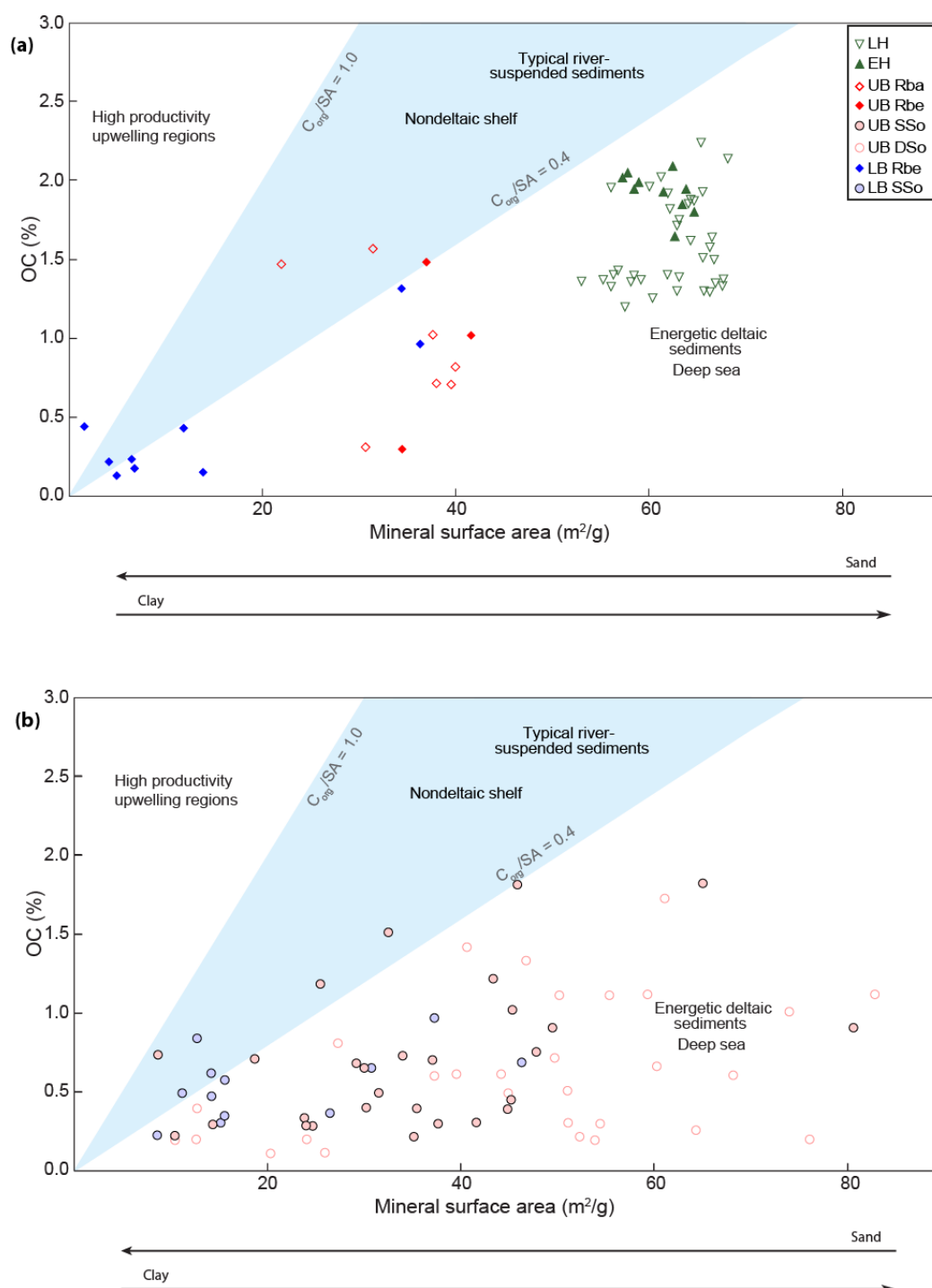
We do agree that discrete particles of (fresh) organic debris that are not associated with mineral phases likely play a role in the lack of correlation between OC and MSA in soils, and this may also contribute to the relationships between OC:MSA and organic matter $\Delta^{14}\text{C}$ (and $\delta^{13}\text{C}$) values. However, without in-depth studies of the spatial disposition organic matter within soil and sediment matrices, which vary from the nanoscale to those involving organo-mineral aggregates (Heister et al., 2012, Ransom et al., 1998), it is not possible to speculate further on this topic. Isolating the nature of organic matter-mineral interactions, and their role in organic matter stability was and remains beyond the scope of the present study.

The primary purpose of Figure 4 was to assess OC and MSA relationship in the context of parameter space described by Blair and Aller (2012) with respect to OC versus MSA relationship for sediments from other fluvial systems. We note that while Figure 2 in Blair and Aller (2012) presents conceptual framework

concerning the range of OC:MSA interactions, it is exclusively focused on fluvially-derived sediments (i.e no soils). Majority of our samples plot outside the typical range of river sediment (blue region in Fig. 4), suggesting disaggregation and degradation of OM related to prolong oxygen exposure during (re)mobilization, transportation, (re)deposition (Blair and Aller, 2012).

As a result, we have modified Figure 4 in order to more strongly emphasize data for riverine and marine sediment (Fig. 4a) and soils (Fig. 4b) in order to be more consistent with the convention that Blair and Aller (2012) originally described.

Fig. 4



Specific comments

30 – having read the paper, this final statement of the abstract seems to contradict those statements written on line 705 – does the abstract need some modification here?

We agree that the concluding sentence in the abstract was phrased incorrectly. The abstract section of revised manuscript has been amended accordingly. The concluding sentence now reads as follows:

While changes in water flow and sediment transport resulting from recent dam constructions have drastically impacted the flux, loci and composition of OC exported from the modern Godavari basin, complicating reconciliation of modern-day river basin geochemistry with that recorded in continental margin sediments, such investigations provide important insights into climatic and anthropogenic controls on OC cycling and burial.

255 – what grain size are the sediments on the river bed? What grain size was targeted? How do you know they are freshly deposited?

The grain size generally ranges from clay to very coarse sand, with minor contributions of loose pebbles and plant debris. The <2 mm size fraction was targeted for this investigation. “Freshly deposited” in this context refers to loose and unconsolidated sediments on the flank of the river that are not overgrown with vegetation (e.g. grass) and thus most likely deposited during recent flood events. The revised manuscript has been amended to include the grain size range. The relevant paragraph now reads as follows:

The sampling sites were selected as being representative of the local depositional settings of the rivers and its tributaries, and mostly comprise areas dominated by bedload sediments (channel thalweg) with particle sizes ranging from <2 μm (clay) to 2 mm (coarse sand) with minor proportions of pebbles and plant debris

345 – do you mean DELTA14C? this could be clearer, as I think this is slightly different as it is being used to compare the marine sediment core measurements and modern.

All ^{14}C values for the marine sediments have been age corrected (following Stuiver and Polach, 1977) to allow a direct comparison with modern river basin samples. This was highlighted in line 341 of the original manuscript (344 in revised version) and line 352 (355 in revised version) states that “henceforth all bulk ^{14}C values for offshore sediments core refer to the age corrected (Δ) value.”

420 – so are these river bed samples representative of the whole river material? If so this needs to be explained. Other studies have tended to focus on sampling the finer component of river flood deposits (sand and finer), as a way of linking (potentially) to the suspended sediments carried by the river. If this was the case here, then the grain size distributions could just reflect this sampling bias, and are not representative of parts of the basin as is suggested here.

The riverbed samples are considered representative of the local setting of the sampling area. We stated this explicitly in line 255-260. The particles size ranges (clay to coarse sand) are similar throughout the course of the river. As a result, we consider these samples to be representative of the section of the basin where they were collected.

520 – if the mineral surface area was important, why is the link between MSA and %OC so unclear when plotted? In figure 4 one could argue that MSA is not a control on %OC because there is no relationship across the sample set, nor in any one part of the sub samples.

As already noted above, yes there is indeed no correlation between OC and MSA when the full dataset (i.e. both sediments and soils) is considered. However, as Fig. C1 above clearly demonstrates, there is indeed MSA control on OC in riverine sediments, with a r^2 value that is similar to other river systems (e.g. Tao et al., 2015; Freymond et al., 2018). There have been fewer reported studies on OC-MSA relationships in soils (e.g. Kaiser and Guggenberger, 2003; Pennel et al., 1995), and the apparent non-correlation in our original Fig. 4, plotting the full dataset (i.e. soils and sediments) is mainly due to the soil samples that were measured.

540 - The bulk isotopic composition and radiocarbon activity are linked to the OC/MSA – this is interesting. The discussion was a little brief on this – particularly on the OC/MSA vs ^{14}C link. Is this degradation signal, or a OC loading signal? Or both? And where is this happening, presumably in the soil sections? Or is there a role for floodplain processes or processing within the river corridors?

Indeed, there appears to be a close coupling between OC loading and carbon isotopic composition (both stable and radiogenic). We suspect this may reflect a variety of processes including loss (preferential degradation) of fresh OC and/or preservation of old OC, replacement (loading) of OM during transport, as well as dilution of upstream signature in the lower basin. We hypothesized that majority of these processes are occurring during riverine transport, as argued from line 567 to 589 in the revised manuscript.

610 – “marked differences” – this wasn’t so clear looking at the figures

All figures have been modified to illustrate the different sample types.

Figures 2-5: the fact the stars are the marine sediment core samples could be much clearer. They don’t have the same label as Figure 1 and the caption doesn’t mention this explicitly. “SS” is used for suspended sediment in some studies so could be confusing.

Figures 2-5 have now been modified and new symbols were adopted to make the delineations clearer. Closed upright and open inverted triangles are now used in

the revised manuscript to denote early Holocene and late Holocene sediments, respectively (previously closed and open stars). In addition, the abbreviation “So” has been adopted instead of “S” to avoid confusing surface soil “SS” (now “SSo”) with the standard abbreviation for suspended sediments.

Figure 5 and Figure 7: for the marine core samples, its unclear in the caption or figure whether these are the bulk ^{14}C activity of OC, or a calculated D^{14}C at time of deposition using the ^{14}C -activity of the foraminifer (as per Giosan et al., 2017).

As stated above, all ^{14}C data for the marine sediments represents the age corrected value. We have now added this information to the figure captions to make this clearer.

Please clarify. Figure 5 – the text mentions some of these soils samples are soil depth profile from a single location. It would be interesting to consider how these look in this space (i.e. to what degree is the signal in the river set by carbon cycling in soils?).

Yes, in some selected locations we sampled the soil profile until the bedrock in addition to the surface soil. These depth profiles were explored for ^{13}C and ^{14}C variation in soil horizons. These plots are shown in the supplementary information that accompanied the original manuscript. We agree with the reviewer that it would be interesting to explore this facet of OC-soil dynamics on a spatial scale. However, we only have these depth profiles from the upper part of the basin and therefore we are unable to make a quantitative assessment of basin wide OC-soil interactions, as equivalent data from the lower basin is not available.

Figure 7 – the stars are hard to distinguish as filled (early Holocene) and open (late Holocene), please modify.

We have modified the symbols to make distinguishing early and late Holocene easier.

Figure 8 – the “soil” box doesn’t seem to correspond to the soil samples?

We think the reviewer meant Figure 7 rather than 8. In any case, the “soil box” was operationally defined by us as this represents the range of values that we have found in the literature. The range is by no means exhaustive as the soil box can theoretically be expanded on either axis. Thus, we have modified the box to envelope our dataset as well as other tropical systems.

Other minor typographical errors have also been corrected.

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