Reply to reviewer #1

I found the paper entitled "The dispersal of fluvially-discharged and marine, shelf-produced particulate organic matter in the northern Gulf of Mexico" by Yedema et al. (2022) interesting, well-written, and analytically sound in terms of the biomarker work. However, there are a number of shortcomings that need to be addressed with the overall interpretation of the data. This is a complex dynamical region and the much of the prior OC and biomarker work, which could really help here, seems to be largely ignored. Similarly, the hypoxia "issue" in this region need to be better incorporated since there has been considerable controversy about the sources of OM in fueling it (see Bianchi et al., 2010, citation in minor comments below). Nevertheless, I do like the addition of select biomarkers that have to date, not be measured and add some interesting parallels to OC cycling in the northern GOM that if build from the data of previous studies, could be impactful. This data also supports the idea that hypoxia is not only driven by phytoplankton produced from the river plume, but also by inputs of terrestrial OM that is either directly consumed by microbes and/or via priming of coastal plankton.

Reply: We thank the reviewer for the positive evaluation of our contributions presented in this manuscript and for their constructive suggestions to improve the parts on priming and hypoxia.

Major comments:

1) Since surface sediment composition could vary through seasons, timing of field sampling and oxygen availability are critical for interpreting OC-sources and preservation. However, this manuscript neglected the significant impact of hypoxia by providing the reason that water may become reoxygenated in the next season. In fact, the systems may not be completely reset, and this seasonal redox oscillation itself could also enhance or retard OC degradation by various mechanisms.

Reply: Surface sediments are here represented by the upper 2 cm of multicore sediments. We do have ^{210}Pb dates for several of these samples. The highest sedimentation rates found in these 2 cm is ~1cm/year in the proximity of the Mississippi river. Consequently, the surface sediments presented in this manuscript represent at least one year of organic matter deposition, therefore integrating seasonally varying oxygen conditions. Moreover, sampling was carried out in February 2020, and all sampled locations had oxic water conditions at that time. We will include this information in the revised version of our manuscript.

2) The discussion of priming mechanism is still obscure. There was no explicit evidence to suggest that decomposition of soil-derived OC was boosted by addition of labile materials. Moreover, the authors should provide more evidences to support why priming of OC decomposition selectively affected soil-derived OC, but not influenced plant-derived OM.

Reply: We agree with the reviewer that we have no direct evidence of priming in the coastal zone. Priming is hard to directly measure in natural systems, especially without using isotope labeling. Hence, we here infer priming from our observation that offshore TerrOM transport is limited and coincides with a high amount of heterotrophic dinocysts close to shore, suggesting an enhanced activity of secondary producers, as described in the manuscript (lines 549-566).
We also do not have direct evidence that soil-OM is preferentially targeted by priming per se. However, based on the distinct dispersal patterns of the TerrOM types, we hypothesize that certain pools of OM (such as plant-derived OM) may be protected by mineral associations (see Lines 511-513), consistent with previous observations (e.g. Repasch et al., 2022, Geophys. Res. Lett; Kirkels et al., 2022 Biogeosciences). We will extent the discussion of the priming process in the revised manuscript with an emphasize on the difference between soil and plant TerrOM.

Line 44-46: "initial composition of this particulate OM influences the burial efficiency of TerrOM" The discussion on "burial efficiency" requires incorporation of other data that used cores and biomarkers in this region and the issues of hypoxia. Please look at the following papers, and references therein, that I think should prove useful: Bianchi et al., 2002 Mar. Chem. 77: 211-223; Chen et al. 2003 GCA.: 67: 2027-2042; Chen et al. 2003 Mar. Chem.,: 81: 37-55; Bianchi et al., 2006 Eos: 87 (50): 565, 572-573; Bianchi et al., 2007 Estuar. Cast Shelf Sci., 73: 211-222; Bianchi et al., 2007 GCA: 71: 4425-4437; Sampere et al., 2008 Cont Shelf Res. 28: 2472-2487; Bianchi et al., 2010: Sci. Total Env. 408: 1471-1484; Sampere et al 2011 Estuar. Coastal Shelf Sci. 95: 232-244.

Reply: We thank the reviewer for these suggestions. We will take a careful look at the above-mentioned papers and include references in the revised manuscript where relevant.

Line 115-117:

1) Is "ammonium oxidizer" more commonly used than "ammonia oxidizer"?

Reply: Ammonia oxidizer is indeed the more commonly used term. We will change this in the revised version of the manuscript.

2) Since Thaemarchaeota is an ammonia oxidizer, are their other papers form this region on their abundance as related to oxygen availability, not just ammonia concentration?

Reply: Previous studies that monitored Thaumarchaeota populations in the northern Gulf of Mexico (e.g. Tolar et al., 2013; Front. Microbiol.) show that while Thaumarchaeota are present across the northern GoM, there is no clear relation with oxygen availability. Since the northern GoM is characterized by high primary productivity and subsequent high degradation of OC releasing ammonia, we believe that ammonia availability is a more important factor that influences the abundance of Thaumarchaeota than oxygen concentrations.

Line 133-134: Seems like the n-alkanes data set would be more comprehensive if the authors added short-chained (C17-C19, C21) and mid-chained (C23, C25) n-alkanes as proxies of marine algae and aquatic macrophytes, respectively. This could be linked to some of the papers cited above that use algal biomarkers in this region.

Reply: We chose to focus on the concentrations of n-C29-C35 to specifically target the terrestrial plant material as part of the TerrOM that is discharged by the Mississippi River. The short- and mid-chain n-alkanes occur in substantially lower concentrations than the long-chain n-alkanes, which is why we decided not to include the data in the manuscript. Regardless, Paq (Ficken et al., 2000) values are ≤0.3 for almost all samples, reflecting the dominance of long-chain n-alkanes and suggesting a predominant higher plant source over that of submerged/floating aquatic plants. Moreover, the short-chain alkanes represent only a very minor (<3%) portion of the total n-alkane pool at most sample sites but reach highest relative abundances close to the Atchafalaya River mouth (9-11% at site A15-A50), consistent with the high concentrations of the marine biomarkers. This shows that these short-chain n-alkanes indeed correlate with the marine algae here. As indicated above, we will take a careful look at the above-mentioned papers and include references in the revised manuscript where appropriate.

Line 180-182: How might loop current seasonal variation matter? Check papers by Doug Biggs…

Reply: In general, the Loop Current extents further north in the GoM during spring-summer months. However, the Loop Current is usually affecting water properties to the east of the Mississippi river, and rarely reaches as far north as our study sites (i.e., the Louisiana shelf), except in unusual conditions *via detached warm water eddies. We can thus assume that the Mississippi-Atchafalaya River is by far the dominant factor at our site, but before resubmission we will check recent observations of the Loop Current to assess its potential influence on our sedimentary components. Earlier palynological studies have also indicated little influence of the Loop Current in our region (Limoges et al., 2013 Mar. Micropaleontol.; 2014 Palaeogeogr. Palaeoclimatol. Palaeoecol.).*

Line 185-188: The discussion could use more perspective on the differences in slope and particle export rates between MR and AR. See McKee et al. 2004 Cont. Shelf Res. 24: 899-926.

Reply: We believe that this section already touches upon the difference in slopes between the Mississippi River and Atchafalaya River, as we already explain that the particle export rate of the Mississippi River is higher compared to that of the Atchafalaya River (lines 184-189). However, we will add further details on the export towards the Mississippi canyon and the impact of hypoxia on our study sites based on the suggested literature.

Line 199: Surface sediments (0-2 cm) should be discussed in the context of known sedimentation and burial rates and periods of export (see citations above).

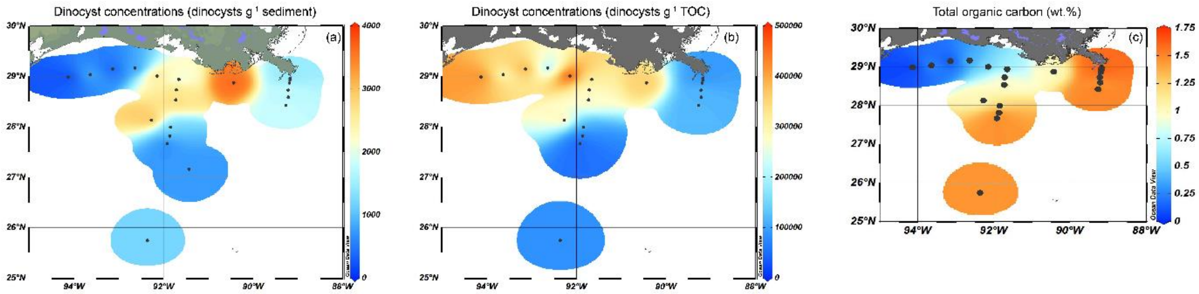
Reply: We will do this. See our reply on comment 1.

Line 207-208: Please look at Bianchi et al 2010 paper on hypoxia that cites relevant physical mixing and hypoxia seasonality papers to better interpret the context of these biomarkers. For example, if sediment discharge and OC input is extremely high during summer hypoxia, rapid burial rate may push fresh OC deep down into sediments.

Reply: We agree with this comment. Indeed, if discharge is high, hypoxia will facilitate the rapid burial of fresh OM. However, as we stated above, the ^{210}Pb dating of our sediment cores indicates that at the time of sampling, the surface sediments are younger than the underlying sediment at our sampling locations. The upper 2 cm we selected contain at the least the last 2 years.
Regardless, we will add relevant references, including Bianchi et al., 2010 and Hetland and DiMarco (2008; J Mar Syst.) to our text to better outline the potential effects of hypoxia on OM burial rates close to both river mouths in the revised manuscript.

Line 266: for the whole palynological processing paragraph: From figure 7, the authors state that dinocyst counts were normalized to TOC., are pollen counts also normalized to TOC as well? For comparisons, it might be interesting to normalize pollen and dinocyst count by weight (or volume) of sediments, since they are part of the less reactive sedimentary OC pool, similar to what is done with sigma lignin.

Reply: Generally pollen grains and dinocysts concentrations are indeed calculated and reported per gram sediment (weight). Nevertheless, we here chose to report the concentrations per gram TOC to enable a more direct comparison with biomarker data, as we explained in lines 161-169 of the original manuscript. The pollen/dinocyst concentrations per gram sediment and per gram TOC are both part of the datafile we submitted to the PANGAEA open-access database. Regardless of the normalization considered (per gram sediment or per gram TOC), the spatial trends are comparable. Only samples located on the western shelf (especially site 20f), deviate from the trend when using the two normalizations. As these sites are characterized by very low TOC values (0.05-0.5 wt.%) and larger grain size (sand) compared to the others, we think that normalization to TOC may even be a better way to present palynological data. In the following figure (not in the manuscript) we compare the dinocyst concentrations per gram sediment and per gram TOC, which indicates that the difference is solely *caused by the low TOC values of the western shelf. This has not changed our interpretation of the dinocyst trends. As mentioned above, we use the concentrations per gram TOC here to enable comparison with our biomarkers.*



Line 305: Figure 4: I personally agree with the ideas that the authors classified proxies into 4 figures including soil-derived, fluvial-derived, marine-derived OC, and plant-derived OC. However, according to Line 137-144, the authors mentioned that most of these sterols (especially, β-sitosterol, stigmasterol, and sitosterol) can be derived from terrestrial sources as well. Moreover, "total sterols" do not really reflect specific terrestrial and/or marine sources, since it commonly includes a mixture of both. In order to avoid misconception, the authors could remove these "sterol proxies" from figure 4, and be added to another figure specifically for sterol proxies.   Once again, using 2 and 3 end-member stable isotopic mixing models previous published for this region should help ground the interpretations here.

Reply: We thank the reviewer for their advice on the placement of the sterol proxies. We will make a separate figure with the sterol data for the revised manuscript and add clarifications to the text where appropriate.

Line 310: Figure 5: It appears that the C32, 1-15 diol was transported west in the Louisiana Current with very little export off shore, look at physical oceanography paper in this region by Steve DiMarco, Ron Hetland etc. This is different from the other biomarkers that show strong export trend both along shelf and across shelf (e.g., n-alkanes). Is there any difference in hydrodynamics between these biomarkers?

Reply: This is indeed curious. We surmise that differences in transport pathways between biomarkers resulting from e.g., mineral sorption, plays a role here. Alternatively, the C32 1,15 diols (together with brGDGTs) are less resistant to oxidation compared to n-alkanes (see Hoefs et al., 200 GCA), which can limit their transport further offshore. In addition, the C32 1,15-diols plot in between the terrestrial and marine proxies in our PCA results, implying that it might be possible that in situ production of C32 1,15 diols takes place on the shelf, near sites A15 and 20b. On the other hand, the FC32 clearly shows high relative abundances of the C32 1,15-diol near the Mississippi river, indicating a mainly riverine source of these diols. We will include this discussion in the revised manuscript.

Line 315: Add "total" to The highest "total" sterol concentrations… (The phrase "The highest sterol concentrations" alone may be misinterpreted that the concentrations of each individual sterol are all highest between MR and AR).

Reply: Thanks for spotting this, we will add the word 'total' to this sentence for clarification.

Line 347-350:

"Almost all variables plot positively on PC1, together with shallow shelf (<20 m water depth) sediments. The only exception is the concentration of alkenones, which plot negatively on PC1, with sediments at intermediate water depth (<80 m) on the Atchafalaya transect. Sediments from the deeper parts of the Mississippi (>50 m) and Atchafalaya (>200 m) transects also plot negative on PC1." Sediments were separated… "sediments at intermediate water depth (<80 m) on the Atchafalaya transect" from "sediments from deeper Atchafalaya transect", why? Were they both plotted negatively on PC1?

Reply: In hindsight, this part of the text was rather confusing and we will clarify this in a revised version. But indeed, sediments from both parts of the Atchafalaya transect plotted negatively on PC1. They were first separated since the alkenones plotted close to site 80b, which we called intermediate depth here, while the remainder of the transect also plotted negatively, but not close to the alkenones. In the revised version we will abstain from using 'intermediate' and 'deeper part as a way of describing the transect in this section.

Line 369: Since δ13C are all in negative range, the authors may want to use the term "less negative" or "more enriched" rather than "more positive"

Reply: We will revert to the consequent use of the conventional indications: (relatively) ^{13}C-depleted vs ^{13}C-enriched or higher vs lower $\delta^{13}C$.

Line 394-395: The plume of high concentration of C32 1, 15 diol is correlated with zone of δ13Corg enrichment. Is this evidence for enhanced marine productivity via fluvial export?

Reply: We think that the less negative δ13Corg in this area is mostly caused by the high marine productivity at this site as revealed by high concentrations of marine markers (crenarchaeol, alkenones, 1,14 diols) and dinoflagellates. The increased marine productivity is probably triggered by nutrients supplied by the Mississippi. We interpret the presence of TerrOM at the same site as an indicator that this OM may contribute to marine productivity (see lines 525 -529), as we later describe in the priming section.

Line 459-463: What's about fluvial OM, can sorption on mineral surface be important?

Reply: This is a very interesting point. This dataset indeed raises questions on mineral protection but the present data does not allow for a detailed analysis on this aspect. We are currently investigating the sorption to minerals of soil-, fluvial- and plant derived OM on a land-sea transect in the GoM to follow up on such observations.

Line 468: Can we use brGDGTs as a representative of soil-derived OM in term of sorption mechanism? brGDGTs may represent a small fraction of total soil-derived OM. Does the rest of soil-derived OM (e.g., humic substances which enriched in polar functional groups) share the same sedimentation pattern with brGDGTs?

Reply: In this paper, we present brGDGTs as representation of soil-derived OM, but the long-chain n-alkanes, while derived from higher plant, can of course also be stored in soils prior to mobilization and be transported to the coastal zone. As brGDGTs and n-alkanes show different dispersal patterns, this suggests that the OM source (in this case soil microbial vs higher plant) is more important than the specific compartment (e.g. soil, vegetation, aquatic) of the river system that the OM is derived from.

Line 500-502: Alternatively, is it possible that the distribution of n-alkanes and pollen greatly represented terrestrial input because they were more resistant toward degradation. However, sterols are more enriched in reactive functional groups; thus, their spatial patterns were more irregular due to heterogenous conditions for degradation (e.g., oxygen availability, the presence of microbes etc.). As discussed in previous comment (Line 468), n-alkanes represent only one fraction of total plant-derived OM. Can we assume that the rest of plant-derived OM share the same behavior with n-alkanes?

Reply: Other plant material might behave differently compared to n-alkanes. However, previous studies from the Gulf of Mexico show that lignin concentrations decrease further offshore (Bianchi et al., 2002 Mar. Chem; Sampere et al., 2008 Cont. Shelf Res; Sampere et al., 2011) or remain constant in waters >100 m deep (Goñi et al., 1998 GCA). Another study that compared offshore trends of n-alkanes and fatty acids found similarly decreasing trends of both biomarkers (Hou et al., 2020 J. Geophys. Res. Biogeosci.). Nevertheless, lignin and n-alkanes both represent a resistant part of the plant OM-pool and might therefore be transported further than less resistant plant material (Hoefs et al., 2002 GCA). Therefore, it is possible that different types of plant-derived OM have different dispersal patterns. Comparison of trends in n-alkane concentrations with those of sterols and pollen might not be totally fair due to the mixed sources of sterols in the GoM and the likely different transport mechanism of pollen, respectively. Regardless, we will add this discussion to section 5.2.3 of our manuscript.

Line 505-507: Is there any difference in sorption mechanism of soil-derived, fluvial-derived, and plant-derived OM on mineral surface? (For example, type of minerals, particle size, and etc.), see paper by Mayer et al., 2009 Mar Chem.

Reply: We indeed think that this may be the case, and will be the focus of a follow-up study.

Line 568-569: For the discussion on priming mechanism:

1) Is there any more detailed evidence of priming, which I do believe is happening in this system. Wysocki et al., 2006 made reference to this which may be useful. I do like the notion of algal-drive material being linked in this ad these materials get processed along the way as they move west. Also, why would priming could enhance the decomposition of soil-derived OM, but not plant-derived OM? This needs some further justification with refs.

Reply: We thank the reviewer for this suggestion and will add citations to the work of Wysocki et al., to the manuscript. As mentioned earlier, we cannot provide direct evidence that soil-OM is preferentially targeted by priming in this study. However, several previous studies on river transport of brGDGTs (Li et al., 2015 Org. Geochem; Freymond et al., 2017, Org. Geochem.; Kirkels et al., 2022, Biogeosciences) have shown that the proxy signal derived from soil-derived brGDGT represents local environmental conditions and therefore seems to be continuously renewed during transport. Furthermore, these studies have suggested that brGDGTs are not transported in association with mineral surfaces, due to the dissimilar trends in concentration of brGDGTs, bulk OM and elemental compositions of the catchment soils. On the other hand, several studies also report that the brGDGT signal that is discharged is overprinted by brGDGTs that are produced in the coastal marine environment (De Jonge et al., 2014 GCA; Zell et al., 2013 Limnol. Oceanogr; Warden et al., 2016 Biogeosciences; Sinninghe Damsté, 2016, GCA), although we find no evidence for that in our surface sediments.
Overall, the composition and transport mechanism of brGDGTs would make these compounds more sensitive to degradation upon discharge. In contrast, a study focusing on the fluvial transport of n-alkanes found that n-alkane concentrations were correlated to fine grain size fractions and aged accordingly with prolonged river transport (Repasch et al., 2022, Geophys. Res. Lett.), indicating that n-alkanes represent a more resistant pool of OM that is likely transported as mineral associated OM. These differences in composition and transport mechanism can possibly result in the preferential

targeting of soil OM during priming. As indicated above, a discussion on this topic will be included in the manuscript.

Line 584-585: The authors need to better state whether the trends they observed in OM cycling were only controlled by source-differentiation, hydrodynamic transport, and/or hot spots of decomposition.

1) Any proxies here to confirm that the residual of soil-derived OM is more "transformed" than plant-derived OM? Perhaps comparing the concentration of each soil-derived OM biomarker in GoM sediments vs. in riverine sediments. Again, why priming mechanism can facilitate decomposition of soil-derived OM, but not plant-derived OM?

Reply: With this study, we mainly conclude that source-differentiation causes the differences in OM dispersal patterns as that is what we can firmly conclude from our data set. Several follow-up studies are being undertaken to assess possible variations in hydrodynamic sorting, transport mechanisms and loss upon discharge (see previous comments). In the summer of 2022 we have collected soil, vegetation material, and riverbed sediments from the Mississippi delta to obtain biomarker and palynological end-members for the terrestrial realm (yes, it was hot). We will use this material to better connect the terrestrial and marine environments.

3) What's about non-point source input of plant-derived OM (e.g., marshes) vs. soil-derived OM?

Reply: Non-point material could be derived from aeolian input and/or erosion of sediments from coastal areas. Such input could indeed contribute to the total TerrOM pool in our samples. However, given the amount of water and sediment transported to the coast and offshore by such a big river as the Mississippi, we tend to think that non-point sources are a minor contributor.
We do not have a direct way to quantify the total contribution of – for instance - marsh input in our samples, but we can make an estimation by using pollen from typical marsh plants. Our pollen data indicate that marsh taxa occur in higher relative abundances close to shore (~20%), while their total concentration remains overall low. Notably, marsh plant pollen in the shelf sediments can still (also) be introduced by the Mississippi river, as its plume extends westwards onto the shelf. Literature also suggests that the primary source of pollen (and possibly OM) on the Louisiana shelf is the Mississippi river (e.g. Chmura et al., 1999, Paleogeogr.). Furthermore, the average chain length (ACL) of long-chain n-alkanes does not show a spatial trend that would reveal a change in plant type source. The input of e.g. n-alkanes by aeolian transport has been discussed in the manuscript. Therefore, we can conclude that if there is a non-point source of plant input, it is likely neglectable in comparison with the point source represented by the rivers. This will also be clarified in the revised manuscript.