Response to reviewers of the manuscript entitled "Sources, Reactivity and Burial of Organic Matter in East China Sea Sediments, as Indicated by a Multi-geochemical Proxy Approach" authored by X. Yu, S. Liang, G. Zhang, S. Li, H. Huang and H. Ma.

Below, the reviewer comments are included in blue, and responses in black font. All page/line numbers referenced below refer to the preprint.

Reviewer #2

Comment 1: In this study a set of surface samples, taken along a transect from the Changjiang outer estuary into the Okinawa Trough, were investigated with the main focus on the contribution of terrestrial (plant + non-plant) and marine organic matter. In addition to $\delta^{13}C$ and lignin which are used for end-member mixing, e.g. quantification of terrestrial vs. marine organic matter contribution, $\delta^{15}N$, TOC, TN, grain sizes, neutral sugar and amino acid content and spectral distributions are measured. This study is thus interesting due to its multi-proxy approach. A similar approach was used by Cowie, G., et al. (2014) (Comparative organic geochemistry of Indian margin (Arabian Sea) sediments: estuary to continental slope. Biogeosciences, 11(23), 6683–6696. doi:10.5194/bg-11-6683-2014.), and could thus be an important paper to compare the results with. It is interesting that despite a different major question the results are somewhat similar and stress the importance of grain size for organic matter accumulation. There may of course be more studies of a similar kind in the literature.

Response: Thank you for your comment and for providing the relevant reference. We have carefully read the suggested paper by Cowie et al. (2014) and agree that it provides a meaningful comparison in terms of methodological approach and findings. We will cite this reference in our manuscript (Part 4.1, Page 14, Line 322).

Comment 2: In the data presentation many of the results are shown but I really miss a Figure of the % terr as in Figure S2. This is a very informative Figure as it relates different variables such as clay content and % terr as well as lignin-phenol. Plant % would also be a good variable to be included in an additional Figure. The authors should include a Figure of the results of the end-member model with some of the relevant other variables.

Response: We will add a new Figure 10. Figure 10(b) further illustrates the outcomes of the end-member model alongside Clay (%), THAA (%TOC), and DI, thus addressing the reviewer's comments. In addition, Figure S2 was renamed as Figure 10(a). This figure is as follows:

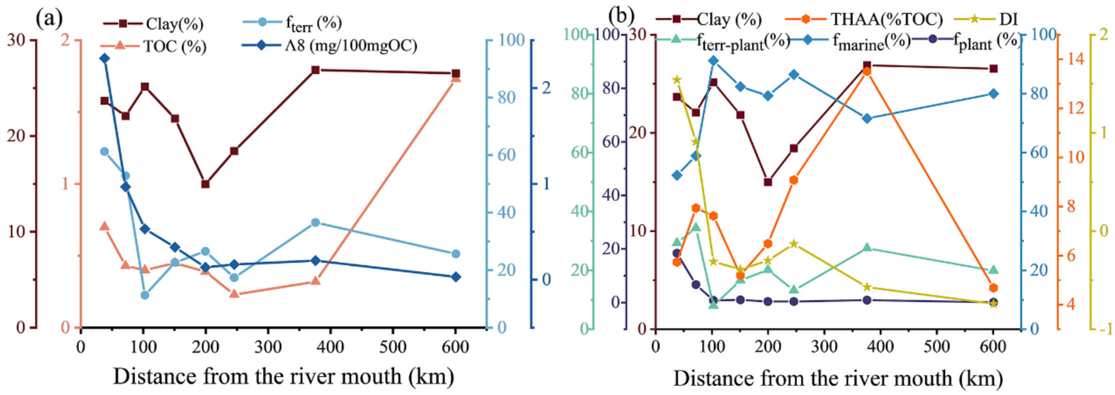

Figure 10. (a) Total organic carbon (TOC) content, percent (%) clay, % terrestrial organic matter (f_{terr}) and OC-normalized total lignin-phenol (Λ8) concentration; (b) Carbon-normalized amino acid yields (THAA(%TOC)), % clay, % plant organic matter (f_{plant}), % non-plant organic matter ($f_{terr-plant}$), and % marine organic matter (f_{marine}) content, and degradation rate (DI) in sediments along the East China Sea study transect.

Comment 3: The introduction to neutral sugars and amino acid is quite short and should be enlarged with some reference to the use of these indicators (see comment to Lines 364-365).

Response: Following the reviewer's valuable suggestion, we will expand the introduction to include additional information on neutral sugars and amino acids, with particular emphasis on the use of Asp/Gly versus Ser+Thr as indicators. The relevant references will be added in Part 1, Page 3, Line 90, as follows:

"In addition, the aspartic acid (Asp)/glycine (Gly) ratio and serine (Ser) + threonine (Thr) (mol%) have been employed to differentiate between diatomaceous and calcareous sources of OM (Gupta and Kawahata, 2003; Ittekkot et al., 1984). Diatomaceous materials exhibit relatively lower Asp/Gly ratios (≈0.62%) than calcareous materials (≈1.88%), whereas calcareous materials are distinguished by relatively lower Ser + Thr (mol%) values (≈9%) than diatomaceous materials (≈16.7%) (Wei et al., 2022; Müller et al., 1986). Carbohydrates generally account for 3%-10% of TOC in marine sediments. NS are a class of monosaccharides within saccharides (i.e., carbohydrates), serving as important carbon and energy sources for microorganisms, with their yields typically declining during OM decomposition. The phytoplankton primary production is the main process controlling the distribution of carbohydrates on the shelf, while the high value of the relative percentage of fucose and rhamnose to total neutral sugars (mol% (Fuc+Rha)) indicates intensive diagenetic alteration (He et al., 2010)."

Comment 4: Lines 73-75: In contrast to the previous introduction to $\delta^{13}C$, the introduction to $\delta^{15}N$ is rather short. In addition, the range of terrestrial $\delta^{15}N$ values is much wider and a $\delta^{15}N$ of 0 ‰ is certainly not representative. A very comprehensive overview is available by "Kendall, C., Elliott, E. M. & Wankel, S. D. (2007). Tracing anthropogenic inputs of nitrogen to ecosystems, in *Stable Isotopes in Ecology and Environmental Science*. edited by R. H. Michener and K. Lajtha, pp. 375–449, Blackwell Publishing." But there are many more studies which suggest that less polluted and large rivers would have a $\delta^{15}N$ signal of about 4 ‰, whereas polluted rivers can have a much higher $\delta^{15}N$ and pristine mountain rivers a lower $\delta^{15}N$ value than 4 ‰ (Voss, M., Deutsch, B., Elmgren, R., Humburg, C., Kuuppoo, P., Pastuszak, M., Rolff, C. & Schulte, U. (2006). River biogeochemistry and source identification of nitrate by means of isotopic tracers in the Baltic Sea catchments. Biogeosciences Discussions, 3, 475–511.).

Response: We have carefully considered your suggestions and incorporated relevant information accordingly. Specifically, we will cite the two referenced articles and expanded the discussion on $\delta^{15}N$ in the Introduction section (Part 1, Page 3, Line 74) as follows:

"The range of $\delta^{15}N$ also suffers from the issue of overlapping isotopic signals, as values of algae and other aquatic plants range from -15‰ to 20‰, a range that encompasses those of terrestrial plants (-5‰ to 2‰), animal excreta (5‰), and soils (0.65‰ and 2.73‰ for cultivated and uncultivated soils, respectively) (Kendall et al., 2007). Isotopic $\delta^{15}N$ values are influenced by multiple processes. Biologically mediated reactions, including nitrogen fixation, assimilation, mineralization, nitrification, and denitrification, commonly result in an increase in the $\delta^{15}N$ values of substrates and a decrease in those of corresponding products (Kendall et al., 2007). Meanwhile, human activities also markedly influence the magnitude of $\delta^{15}N$ values. As noted by Voss et al. (2006), the $\delta^{15}N$ signal of large rivers with relatively little pollution is ca. 4‰, while that of polluted rivers may be much higher than this value, and the $\delta^{15}N$ values of pristine mountain rivers may be lower than 4‰."

Comment 5: Line 90: AA are not really molecular biomarkers as they are not very specific indicators of certain organisms but are ubiquitous in living organisms. They have, therefore, often been called biogeochemical indicators as they delineate degradation pathways (as the authors also describe).

Response: In accordance with the comment, we have revised the manuscript to reflect that amino acids (AAs) are better described as biogeochemical indicators rather than molecular biomarkers, given their ubiquity across living organisms and lack of specificity. Specifically, the following changes have been made:

Part 1, Page 3, Line 90: “molecular biomarkers” has been replaced with “biogeochemical indicators”.

Comment 6: Lines 118-120: a short description of the impact of currents on the study area is missing rather than just reporting their presence.

Response: A brief description of the impact of currents on the study area will be added in the Introduction section, Part 2.1, Page 4, Line 120, as follows:

“The YSCC flows southward throughout the year but is unable to penetrate the ECS during summer. In winter, however, it extends deeply into the northern ECS (Hwang et al., 2014). Together with the Yellow Sea Warm Current (YSWC), the YSCC forms a circulation system that induces upwelling, thus transporting nutrient-rich waters from deeper layers to the surface and enhancing primary productivity (Wang et al., 2019). The CDW contributes significantly to the nutrient pool of the ECS. During summer, most river-borne sediments are temporarily deposited in the subaqueous delta and estuarine zones. In winter, CDW flows southwestward along the coast toward the Taiwan Strait, and, in conjunction with the southward-flowing ZFCC, facilitates the transport of resuspended sediments (Hwang et al., 2014; Hu et al., 2014). The TWC flows northward year-round along the 50–100 m isobaths. In summer, the TWC originates from warm water in the Taiwan Strait and Kuroshio subsurface water, whereas in winter, it is primarily sustained by the Kuroshio subsurface intrusion, which reaches as far north as the subaqueous valley off the Changjiang Estuary (CJE) (Lian et al., 2016). Both the OKBC and the nearshore Kuroshio Branch Current (NKBC) deliver nutrient-enriched waters, particularly in phosphorus (P), thereby promoting primary productivity (Yang et al., 2012; Yang et al., 2013). The OKBC, flowing at depths of 60–120 m, moves northwestward along the 100 m isobath and partially rejoins the Kuroshio Main Stream near 28°N. The NKBC, originating at depths of 120–250 m, turns upward at approximately 27.5°N, 122°E, and then flows northeastward along the 60 m isobath until it reaches 30.5°N before veering eastward (Yang et al., 2012; Yang et al., 2013) (Fig. 1).”

Comment 7: Line 178: C/N instead of N/C

Response: We have corrected the expression from "N/C" to "C/N".

Comment 8: Line 300: Glu is missing in the Figure caption of Figure 5.

Response: We have revised the order of amino acids in the Figure 5 caption, placing Glu at the end to make it more prominent.

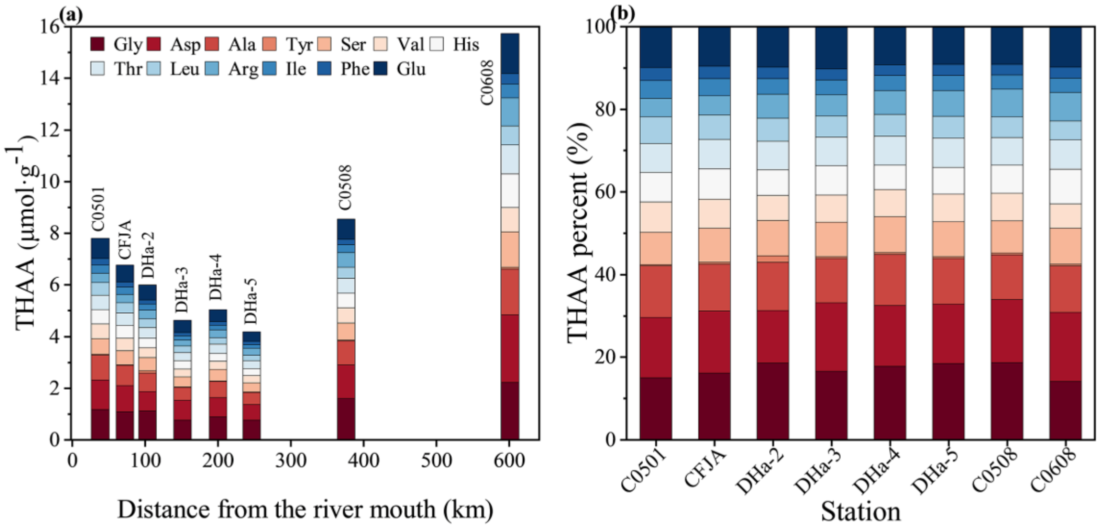

Figure 5. Concentration of each individual hydrolysable amino acid (HAA) (a) and the molar percentages of individual HAAs out of the total (THAA) (b) in surface sediments at sampling stations in an East China Sea transect (see Table S2). Gly: glycine, Asp: aspartic acid, Ala: alanine, Ser: serine, Val: valine, His: histidine, Thr: threonine, Leu: leucine, Arg: arginine, Ile: isoleucine, Phe: phenylalanine, Tyr: tyrosine and Glu: glutamic acid. Tyr concentration was too low to be visible in the plot.

Comment 9: Lines 323-324: imprecise: clay minerals are possibly not the sinks for AA but clay minerals may have organic coatings or fine organic matter may be transported with the clay fraction?

Response: We agree that the original statement was imprecise and will revise it accordingly. The sentence in Part 4.1, Page 14, Line 323: "clay may be the main sink for THAA and NS." will be replaced with a more accurate expression: "clay minerals may have organic coatings or fine organic matter (THAA and NS) may be transported with the clay fraction."

Comment 10: Line 331-333: The increase of $\delta^{15}N$ from the prodelta in offshore direction could be due to a reduced contribution of terrestrial material (see remark above).

Response: We thank the reviewer for this valuable comment. In response, we will revise the discussion accordingly and added relevant references. Specifically, we will supplement the discussion in Part 4.1, Page 14, Line 332 as follows:

"The increase of $\delta^{15}N$ from the prodelta in offshore direction could be due to a reduced contribution of terrestrial material (Kendall et al., 2007; Voss et al., 2006)."

Comment 11: Lines 364-365: the use of Asp/Gly vs. Ser+Thr as a source indicator should be explained with reference in the methods section or introduction.

Response: The rationale for using Asp/Gly versus Ser+Thr as a source indicator will be explained and supported with references in the Introduction (Part 1, Page 3, Line 90). We will supplement the Introduction as follows:

"In addition, the aspartic acid (Asp)/glycine (Gly) ratio and serine (Ser) + threonine (Thr) (mol%) of THAA have been employed to differentiate between diatomaceous and calcareous sources of OM (Gupta and Kawahata, 2003; Ittekkot et al., 1984). Diatomaceous materials exhibit relatively lower Asp/Gly ratios (≈0.62%) than calcareous materials (≈1.88%), whereas calcareous materials are distinguished by relatively lower Ser + Thr (mol%) values (≈9%) than diatomaceous materials (≈16.7%) (Wei et al., 2022; Müller et al., 1986)."

Comment 12: Lines 421ff: When the TOC, TN and THAA contents are related to grain size and sorting rather than degradation processes it is feasible that THAA% and DI are not related as the material degrades as it is transported offshore or to greater depths. Further, the much lower SR and the decrease in offshore direction match the degradation as reflected in the DI. These aspects are discussed later but some of the discussion can be deleted by combining these aspects.

Response: Thank you for your insightful comment. In response, we will revise the original sentence "This is likely because each index is most sensitive to a particular diagenetic stage. DI_{AA} was most effective in reflecting diagenetic alterations during intermediate stages of decomposition over timescales of years to decades, whereas THAA(%TOC) and THAA(%TN) were more sensitive indicators of early stages of OM degradation stages (Davis et al., 2009; Chen et al., 2018)." to "This is likely because when the TOC, TN, and THAA contents are related to grain size and sorting rather than degradation processes, it is feasible that THAA(%TOC), THAA(%TN), and DI are not correlated as the material degrades during offshore transport or deposition at greater depths (Fig. 10(b)). Further, the much lower SR and the decrease in offshore direction match the degradation as reflected in the DI. ".

We have corrected the sentence "and the percentage of Gly increased (Fig. 5(b)) while that of Glc decreased" to "and the percentage of Gly (Fig. 5(b)) and Fuc+Rha (Fig. 4(b)) increased" in Line 418 of page 17 in part 4.2. The sentence "The mol% Glc concentration decreased seaward, except at DHa-5 (Fig. 4(b))." also was corrected to "The mol% (Fuc+Rha) increased seaward (Fig. 4(b))." in Line 277 of page 12 in part 3.3.2.

Comment 13: Line 465: see earlier comments on "molecular biomarkers" and change the term here too.

Response: The term “molecular biomarkers” has been revised accordingly throughout the manuscript.

Reference

Chen, Y., Yang, G. P., Ji, C. X., Zhang, H. H., and Zhang, P. Y.: Sources and degradation of sedimentary organic matter in the mud belt of the East China Sea: Implications from the enantiomers of amino acids, Org. Geochem., 116, 51-61, https://doi.org/10.1016/j.orggeochem.2017.11.011, 2018.

Cowie, G., Mowbray, S., Kurian, S., Sarkar, A., White, C., Anderson, A., Vergnaud, B., Johnstone, G., Brear, S., Woulds, C., Naqvi, S. W. A., and Kitazato, H.: Comparative organic geochemistry of Indian margin (Arabian Sea) sediments: estuary to continental slope, Biogeosci., 11, 6683-6696, https://doi.org/10.5194/bg-11-6683-2014, 2014.

Davis, J., Kaiser, K., and Benner, R.: Amino acids and amino sugar yields and compositions as indicators of dissolved organic matter diagenesis, Org. Geochem., 40, 343-352, https://doi.org/10.1016/j.orggeochem.2008.12.003, 2009.

Gupta, L. P. and Kawahata, H.: Amino acids and hexosamines in the Hess Rise core during the past 220,000 years, Quat. Res., 60, 394-403, https://doi.org/10.1016/j.yqres.2003.07.012, 2003.

He, B., Dai, M., Huang, W., Liu, Q., Chen, H., and Xu, L.: Sources and accumulation of organic carbon in the Pearl River Estuary surface sediment as indicated by elemental, stable carbon isotopic, and carbohydrate compositions, Biogeosci., 7, 3343-3362, https://doi.org/10.5194/bg-7-3343-2010, 2010.

Hu, B., Li, J., Zhao, J., Wei, H., Yin, X., Li, G., Liu, Y., Sun, Z., Zou, L., Bai, F., Dou, Y., Wang, L., and Sun, R.: Late Holocene elemental and isotopic carbon and nitrogen records from the East China Sea inner shelf: Implications for monsoon and upwelling, Mar. Chem., 162, 60-70, https://doi.org/10.1016/j.marchem.2014.03.008, 2014.

Hwang, J. H., Van, S. P., Choi, B.-J., Chang, Y. S., and Kim, Y. H.: The physical processes in the Yellow Sea, Ocean Coastal Manage., 102, 449-457, https://doi.org/10.1016/j.ocecoaman.2014.03.026, 2014.

Ittekkot, V., Deuser, W. G., and Degens, E. T.: Seasonality in the fluxes of sugars, amino acids, and amino sugars to the deep ocean: Sargasso sea, Deep-Sea Res. Part I-Oceanogr. Res. Pap., 31, 1057-1069, https://doi.org/10.1016/0198-0149(84)90012-8, 1984.

Kendall, C., Elliott, E. M., and Wankel, S. D.: Tracing Anthropogenic Inputs of Nitrogen to Ecosystems, in: Stable Isotopes in Ecology and Environmental Science, edited by: Michener, R. H., and Lajtha, K., Blackwell Publishing, Oxford, 375-449, https://doi.org/abs/10.1002/9780470691854.ch12, 2007.

Lian, E., Yang, S., Wu, H., Yang, C., Li, C., and Liu, J. T.: Kuroshio subsurface water feeds the wintertime Taiwan Warm Current on the inner East China Sea shelf, J. Geophys. Res.-Oceans, 121, 4790-4803, https://doi.org/10.1002/2016JC011869, 2016.

Müller, P. J., Suess, E., and AndréUngerer, C.: Amino acids and amino sugars of surface particulate and sediment trap material from waters of the Scotia Sea, Deep-Sea Res. Part I-Oceanogr. Res. Pap., 33, 819-838, https://doi.org/10.1016/0198-0149(86)90090-7 1986.

Voss, M., Deutsch, B., Elmgren, R., Humborg, C., Kuuppo, P., Pastuszak, M., Rolff, C., and Schulte, U.: Source identification of nitrate by means of isotopic tracers in the Baltic Sea catchments, Biogeosci., 3, 663-676, https://doi.org/10.5194/bg-3-663-2006, 2006.

Wang, Z., Xiao, X., Yuan, Z., Wang, F., Xing, L., Gong, X., Kubota, Y., Uchida, M., and Zhao, M.: Air-sea interactive forcing on phytoplankton productivity and community structure changes in the East China Sea during the Holocene, Glob. Planet. Change, 179, 80-91, https://doi.org/10.1016/j.gloplacha.2019.05.008, 2019.

Wei, J. E., Chen, Y., Zhang, N., Yang, J. Q., Chen, R., Zhang, H. H., and Yang, G. P.: Variability and composition of amino acids and amino sugars in sediment cores of the Changjiang Estuary, Org. Geochem., 163, 104330, https://doi.org/10.1016/j.orggeochem.2021.104330, 2022.

Yang, D., Yin, B., Sun, J., and Zhang, Y.: Numerical study on the origins and the forcing mechanism of the phosphate in upwelling areas off the coast of Zhejiang province, China in summer, J. Mar. Syst., 123, 1-18, https://doi.org/10.1016/j.jmarsys.2013.04.002, 2013.

Yang, D. Z., Yin, B. S., Liu, Z. L., Bai, T., Qi, J. F., and Chen, H. Y.: Numerical study on the pattern and origins of Kuroshio branches in the bottom water of southern East China Sea in summer, J. Geophys. Res.-Oceans, 117, c02014, https://doi.org/10.1029/2011jc007528, 2012.