# Interactive comments on "Preservation of terrestrial organic carbon in marine sediments offshore Taiwan: mountain building and atmospheric carbon dioxide sequestration" by S.J. Kao et al.

Referee comments in *Italics*. Underlined text indicates changes made in the revision manuscript.

## Anonymous Referee #3

*Referee 3: This manuscript reports organic carbon characteristics in surface sediments offshore of Taiwan that are influenced by hyper- and hypo-pycnal deposition of terrestrial materials emanating from fluvial systems on the island. The study builds on several prior observations that have revealed extensive export of carbon from Taiwan and other islands of Oceania, particularly when tropical cyclones make landfall and strip vegetation and friable bedrock from the steep catchments. The quantities of organic carbon (OC) discharged to the ocean by these processes, and the nature of their dispersal, via hyperpycnal flow during the most intense events when the bloated rivers are loaded with suspended sediment and debris, suggest dramatic and efficient transfer of terrestrial OC to the surrounding ocean. What has remained less clear is the fate of this material upon entering the ocean. The fate of terrestrial OC discharged under hyper- versus hypo-pycnal also remains particularly uncertain.*

*The authors show data from sediment traps and cores which provide strong evidence for efficient transfer of terrestrial OC, of both fossil and non-fossil (biospheric OC) origin, to marine sediments in locations adjacent to Taiwan that are impacted by hyperpycnal sediment delivery. Stable carbon and radiocarbon characteristics are used to distinguish between inputs of OC from the terrestrial biosphere and fossil components from erosion of underlying bedrock, as well as to account for marine organic matter contributions to the sediments. The measurements indicate highly efficient transfer and burial of both fossil and non-fossil (i.e., derived from recent vegetation) carbon under hyperpycnal flow. The results also suggest that preservation of terrestrial carbon delivered via more diffuse hypopycnal flow is also efficient (> 70%).*

*Overall, I think this is an interesting and important contribution, with solid interpretations and significant implications for our understanding of the carbon cycle and geologic controls on terrestrial carbon export and burial in ocean sediments. The results and relationships that are observed strongly support the authors' conclusions of high terrestrial OC burial and high overall OC preservation efficiency in regions influenced by both hyper- and hypo-pycnal sediment supply from Taiwan Island. I hope the following comments and questions will help to further improve what in my view is already a strong contribution.*

Reply: We thank the referee for their positive comments on our manuscript, its data and our interpretations. We are grateful for their thorough review which has allowed us to further improve the manuscript.

*Referee 3: Units: The paragraph beginning on line 14 of Page 180 ("The Himalayan orogeny . . .") contains a range of values for the size and flux of materials. The units used to describe these values vary (Tg, Mg, Pg). While I can appreciate that the values change dramatically depending on whether these values relate to reservoir sizes or fluxes of sediment or, I find it a bit confusing to switch between the units. I would propose sticking with one or maximum so the reader can more readily inter-compare between values.*

Reply: In the revised version we do not use Pg and only use Tg for clarity when referring to sediment (e.g. Tg $yr^{-1}$) and carbon transfers (e.g. TgC $yr^{-1}$). However, when reporting carbon yields we refer to Mg $km^{-2}$ $yr^{-1}$ (i.e. t $km^{-2}$) because this has been the standard notation and avoids unnecessary decimal points in the text.

*Referee 3: Gaoping Canyon sediment trap: - With respect to the 600m sediment trap deployed in the Gaoping Canyon, is it known whether the trap is subject to any trapping biases in particle types as a function of hydrodynamic processes over the trap funnel (e.g. Buesseler et al 2007 J. Mar. Res. 65 345-416)? Specifically, if there are changes in current velocity (e.g., as a consequence of hyperpycnal flow), this might potentially change trapping efficiency, and could give rise to compositional biases in the materials collected depending on trap geometry and flow velocity.*

Reply: The discussion in Buesseler et al., (2007) is relevant to our methods and so we thank the referee for pointing us towards this paper. As Buesseler et al., (2007) note, '*trap-particle interactions have remained virtually unstudied because of the difficulty in quantifying such changes in situ*' and so we are not aware of any compositional biases produced by varying the current velocity over the trap funnel. However, the conical trap used here (Liu et al., 2012) may result in a lower net accumulation in the trap due to some recirculation and 'swirls' which may remove trapped settled sediment before it enters the cylindrical trap. While we do not use the accumulation rate data in this study, previous work has established that mean sedimentation rates from the traps are up 190 times greater during the typhoon-affected period, suggesting the high turbidity and sediment throughput is the dominant record from the trap (Huh et al., 2009).

The discussion by Buesseler et al., (2007) imply that conical traps may experience some loss of finer, more easily re-suspended particles during rapid current velocities by the mechanisms explained above, although this is not stated explicitly nor demonstrated by data. The grain size data from the trap suggest this is not significant, with >60% of the sediment deposited during the typhoon comprised of clay and silt particles, with lighter macro-particles of woody debris also accumulated in the trap (Liu et al., 2012). While we cannot be definitive, and more research is needed on this methodological issue, our data suggest no systematic bias has been introduced.

To clarify some of these methodological details to the reader we have added text to the revised version:

> "Conical sediment traps may result in conservative estimates of accumulation rate due to potential re-suspension of sediment in the funnel (Buesseler et al., 2007). Here we do not rely on accumulation rate data, but note that hydrodynamic sorting may result in a lower percentage of smaller, more buoyant particles present in the trap than the sediment plume during high current velocities." (Section 2.2, Paragraph 1)

*Referee 3: - The observation that relatively large, mineral-free organic detrital material (woody debris) is included in material intercepted by the trap deployed in the submarine canyon (page 190, line 6) is intriguing given expectations that might be expected to be have a higher buoyancy. It would be helpful to know whether this woody debris was visible to the naked eye or was microscopic in size (i.e., approximate dimensions). The fact that this debris sank to the sediment trap depth implies it must have been water-logged. Given the short residence and transit times of terrestrial materials in the Taiwan watersheds, where might such material reside for sufficient time to become water logged?*

Reply: We agree with the reviewer that the mineral-free organic material in the trap is a very interesting observation given its expected buoyancy. Given the comments by Referee #1, we have significantly expanded the discussion on this point, including as Referee #3 requests, confirmation of the approximate size of the material. We also refer to Figure 4 of Liu et al., 2012 (cited in the manuscript) which shows this debris:

> "The trapped sediment included a 'young' organic rich sub-sample ($C_{org}$ = 1.6%, $\Delta^{14}C_{org}$ = –112‰, (Fig. 3b) with some visible shredded woody debris visible to the naked eye and up to ~1cm in size (see also Figure 4 in Liu et al., (2012))." (Section 4.2, Paragraph 2)

> "Moreover, it appears that the natural buoyancy of some macro-particles of $OC_{non\text{-}fossil}$ can be overcome during flood discharges, as observed in modern source-to-sink settings elsewhere (Leithold and Hope, 1999) and in the geological record (e.g. Saller et al., 2006). This suggests that the density of the turbid river plume may be high enough to effectively sequester woody debris carried in the sand fraction, while coarser woody material (e.g. logs) float upon discharge to the ocean (West et al., 2011). In addition, water logging of sand sized woody debris may occur prior to entrainment or during transport, as observed in the sand-sized bedload of larger fluvial systems (Bianchi et al., 2007). In the short mountain rivers of Taiwan, it is unclear whether this mechanism operates and the observation warrents further investigation of the transport of macro-particles of $OC_{non\text{-}fossil}$ in mountain rivers." (Section 4.2, Paragraph 3).

*Referee 3: - In addition to the water depths of the sediment trap moorings, it would be useful to know the distance above the seafloor that the sediment traps were placed in order to gain an appreciate thick the sediment plume.*

Reply: The sediment trap was installed at 608 m water depths, which is 42 m above the sea floor (Figure 2). A Nortek current meter was also placed ~12 m above the sediment trap. As we outline in the manuscript, the details of this mooring can be found in Liu et al., (2012). We have added this detail to the revised manuscript:

> "sediment trap mooring was deployed at 608 m water depth, 42 m above the seafloor, in the submarine Gaoping Canyon" (Section 2.2. Paragraph 1)

*Referee 3: POC (and DOC) export & burial: In examining the methods followed, I note that sediments were rinsed to remove salts, and also centrifuged after acidification (a typical procedure for sediments). Both of these procedures will remove readily desorbable organic matter. While this may reflect a minor component of the total organic carbon, this is not always necessarily the case (Keil et al 1994 Nature 370 549-552). Given the very rapid transfer of young, fresh carbon from the terrestrial biosphere associated with storm events, it would be interesting to speculate whether this entrains a significant proportion of labile organic matter sorbed to the mineral load, and what the fate of this might be under hyperpycnal and hypopycnal flow?*

Reply: We note that the references Galy et al., (2007b) and Komada et al., (2008) cited in the manuscript also discusses the potential loss of a fraction of labile organic carbon during pre-treatment of samples to remove carbonate prior to isotopic analyses. To clarify this to the reader we have added the following statement, to make it clear we are examining acid-insoluble POC:

> "As such, following previous work, all OC isotope measurements refer to the acid-insoluble OC (Galy et al., 2007b; Hilton et al., 2010)." (Section 2.3)

However, we are reluctant to speculate on whether the methods pick up different labile OC pools sorbed to the mineral load during different river flows, since these changes are likely to be of the order ~10-20‰ to bulk $\Delta^{14}C_{org}$ (Komada et al., 2008). Instead, we focus on explaining $\Delta^{14}C_{org}$ values which vary over ~800‰ in the terrestrial samples, and we can explain by the variable contribution of fossil OC to the OC load.

*Referee 3: - In the context of the question of loss of, versus replacement of OC, it might have been useful to examine whether there are changes in the organic carbon loadings normalized to mineral surface area (i.e., work by L. Mayer, as well as Hedges & Keil) for sediments deposited under different flow regimes. Did the authors make such measurements?*

Reply: While we acknowledge this previous work on organic carbon-loadings in the manuscript (Hedges and Keil, 1995; Burdige, 2005), the normalisation of OC content to mineral surface area may be misleading offshore Taiwan. First is the presence of macro-organic particles which are not associated with mineral surfaces (Hilton et al., 2010; Liu et al., 2012). Second, fossil OC is common (Kao et al., 2008; Hilton et al., 2010) and while it is associated with clastic sediment, is not bound to mineral surfaces (e.g. Galy et al., 2008). Instead, we prefer to focus on weight % OC which makes no assumption about the distribution of OC in the sample.

*Referee 3: - In addition, I am curious why nitrogen-to-carbon ratios were not examined given that a sharp contrast is likely to exist between fresh (protein-rich, and nucleic acid-rich) marine organic matter and terrestrial organic matter. Of course, selective degradation processes may modify the N/C ratios of the source materials but given the short transit times and rapid burial processes that appear to be occurring these effects may be minimal and such measurements may have provided additional constraints on inputs.*

Reply: Previous work by some of the authors has examined the N/C ratios of terrestrial samples from Taiwan in detail (e.g. Kao and Liu, 2000; Hilton et al., 2010). N/C ratios in Taiwan's rivers are dominantly controlled by the N/C values of fossil OC (Hilton et al., 2010). Some fossil OC has N/C values which overlap the range expected for fresh marine organic matter (N/C~0.16). The same is true for $\delta^{13}C_{org}$, with some fossil OC from Taiwan and marine OC having overlapping values ($\delta^{13}C_{org}$ ~-21‰) (see Hilton et al., 2010 for detailed discussion). In stark contrast, the fossil OC is radiocarbon-dead, meaning that marine organic matter ($\Delta^{14}C_{org}$ ~0‰) is compositional distinct from fossil OC ($\Delta^{14}C_{org}$ ~-1000‰). Therefore, we focus on the availability of $\Delta^{14}C_{org}$ data to better understand the source, transfer and fate of terrestrial OC offshore Taiwan.

*Referee 3: Carbon dynamics during storm events: Although this is mostly beyond the scope of this manuscript, the authors to bring up the link between warming (i.e., CO2-driven) increases in tropical cyclone activity and the enhanced burial of terrestrial OC as a factor that provides a negative feedback (page 194, line 24). However, enhanced storm activity also likely significantly increases air sea gas exchange as a consequence of increased wave energy/activity and bubble formation, resulting of enhanced transfer of CO2 from surface waters to the atmosphere. It is not clear to me which of these effects - as well as those associated with controls of CO2 on terrestrial productivity, and hydrological processes on export – may be most important. This is clearly a complicated system characterized by both positive and negative feedbacks.*

Reply: As the reviewer is correct to point out, on short timescales there may be positive feedbacks between tropical cyclone activity and $CO_2$ levels through air-sea gas exchange as a consequence of increased wave energy and bubble formation. However, our paper focuses on long-term geological sequestration of atmospheric $CO_2$. In this regard, it is the burial of recent OC in long-lived sediments which can remove $CO_2$ from the atmosphere-ocean system. By tracing OC from source to sink on a mountain island, we are able to comment on the longer-term geological $CO_2$ sequestration. On shorter timescales, the system may be more complicated. In order to keep the discussion focused, we stay focused on the 'geological timescales' which are explicitly mentioned at several points in the final paragraph, the Abstract and Introduction.

*Referee 3: How does sediment and carbon discharge vary across the duration of a tropical storm event? I would assume that initially the river behaves hypopycnally before transitioning to hyperpycnal flow (when suspended sediment concentrations are > 40 g per liter), and then as the storm subsides it could return to hypopycnal conditions? Is this the case, and does the nature of the organic matter (fossil vs non-fossil) change? Over such a storm, what fraction of the discharge is hyperpycnal and what fraction is hypopycnal?*

Reply: Available data where we have high-temporal suspended load samples where we have quantified $OC_{fossil}$ and $OC_{non\text{-}fossil}$, suggests that during the rising limb (<3 hours) and flood peak at hyperpycnal conditions (~12 hours) $OC_{non\text{-}fossil}$ will be enriched compared to background flow (Hilton et al., 2008;

Hilton et al., 2012). This is linked to the geomorphic processes which mobilise $OC_{non\text{-}fossil}$ from the mountain forest which are active during heavy precipitation. We discuss these previous findings in detail in Section 4.1.

Unfortunately, at present it is not possible to quantify the fraction of OC discharge achieved during hyperpycnal and hypopycnal conditions across Taiwan (also see next comment). This is because OC samples have only been collected from a handful of typhoon-induced floods (~3 events) (Kao and Liu, 1996; Hilton et al., 2008, Hilton et a., 2012) and it is difficult to generalise these observations to a single percentage. However, we have much better constraint on the suspended sediment from >30 years of river gauging data. These show 30-40% of the suspended load is discharged under hyperpycnal conditions (Dadson et al., 2005). Because $OC_{non\text{-}fossil}$ is relatively enriched in the river load at these times (Hilton et al., 2008; Hilton et al., 2012) it is likely that a similar of $OC_{non\text{-}fossil}$ is transferred under hyperpycnal conditions.

*Referee 3: While most sediment (~60-70%) is discharged under hypopycnal conditions (page 181, line 27), what are proportions of organic carbon discharged under hyper- vs hypopycnal discharge?*

Reply: The 60-70% estimate for clastic sediment is derived from 30 years of sediment discharge measurements from Taiwan across >20 river basins, including samples from many tropical-cyclone induced floods (Dadson et al., 2003). Unfortunately, because of the need for carefully designed water sample and filtration methods, in addition to intensive geochemical methods, the datasets on OC transfer are more recent and limited to <2 years in length (Hilton et al., 2008; Hilton et al., 2012). Therefore, it is not possible to directly compute this long-term mean for OC. However, we do note in the manuscript (Section 4.1) that a large proportion of the OC transfer is likely to be achieved during typhoon-triggered floods when hyperpycnal discharges are likely, based on previous work (Hilton et al., 2008).

*Referee 3: There appears to be only one sample - a box core – that was collected from a water depth of only 160 m from the thalweg of the Gaoping Canyon. This leaves open the question concerning the fate of OC transported and deposited further downslope as a consequence of hyperpycnal discharge? Are the authors able to comment on this?*

Reply: The referee is correct regarding the number of cores collected from the thalweg of the Gaoping Canyon. Our data allows us to confirm that terrestrial OC is transferred and accumulates in the canyon with high preservation efficiencies. However, we do not have definitive observations as to what happens to terrestrial OC deposited further downslope and on the abyssal plain. To clarify this point to the reader, we have added the following sentence to the revised version:

> "While the fate of terrestrial OC transported deeper down the canyon (e.g. Carter et al., 2012) remains to be assessed, the low $O_2$ levels (Garcia et al., 2009) and high accumulation rates (Huh et al., 2009) are likely to promote longer-term OC burial." (Section 4.2, Paragraph 2)

*Referee 3: Specific points: Page 180, Line 12: ". . . which are not represented in current models of the carbon cycle"*

Reply: Corrected.

*Referee 3: Pg 189 Lines 18-24. It is mentioned that the under-sampled region off the east coast of Taiwan exhibits low O2 concentrations (leading to conservative estimates of terrestrial carbon burial). To what water depth do these low O2 levels persist (i.e., is this an oxygen minimum zone?)? Is low oxidation expected to extend to the very deep regions (e.g., > 5000m)?*

Reply: Based on the $O_2$ maps of Garcia et al., (2009), the O2 levels in deepwaters offshore Taiwan are similar to the Bay of Bengal at 2500 m. Low $O_2$ conditions (<3.5mg/L) persist down to >4000 m. To clarify this point we have added the text:

> "However, $O_2$ concentrations in the un-sampled region which reach water depths >4000 m are low and comparable to those in the Bay of Bengal at 2000 m water depth (Garcia et al., 2010). OC preservation may be higher in these un-sampled areas due to the lower oxidation potential of these deep waters (e.g. Cai and Sayles, 1996; Galy et al., 2007a)" (Section 4.2., Paragraph 1)

*Referee 3: Page 190, Line 5. ~600m does not really constitute "deep waters", but rather mesopelagic depths.*

Reply: We have changed 'deep waters' to 'mesopelagic depths' and removed references to 'deep waters' elsewhere in the manuscript where it was potentially misleading.

*Referee 3: Page 191, Lines 23,24: I would re-phrase ". . .and so the patterns in the data are not consistent with terrestrial OC loss, nor selective (OCnon-fossil, Fig. 4b) or pervasive (OCnon-fossil and OCfossil Fig. 5b)" to "and so the patterns in the data are not consistent with either selective (OCnon-fossil, Fig. 4b) or pervasive (OCnon-fossil and OCfossil Fig. 5b) loss of terrestrial OC.*

Reply: Corrected.

*Referee 3: Figure 1: while the dots are a guide, it would be useful to indicate (a) the location of specific rivers, (b) the main trajectories for hyperpycnal flow, and (c) the general trajectories for hypopycnal flow.*

Reply: As suggested, we have added labels to Figure 1 to highlight the major rivers for which we have sampled suspended load. However, given the harsh conditions and torrential rain associated with storm events, it is very difficult to constrain the trajectory information of hypo- and hyper-pycnal flows for rivers around Taiwan and so we cannot provide a schematic of this information.

*Referee 3: In Figures 2, 3 & 4, it would be helpful to keep the x-axis scale the same (for one it is -19 to -27 permil (Fig 4) and others it is -19.5 to -27 permil (fig 2 & 3).*

Reply: As suggested by the reviewer, we have modified the axis in Figure 4 to align it with Figures 2 and 3.

*Referee 3: Figure 5. Instead of "Radioactive isotopic compositions of organic carbon (D14Corg, permil)" it would be more straightforward to say "Radiocarbon contents of organic carbon (expressed as D14Corg, permil)".*

Reply: Corrected.

*Referee 3: Table S3. It would be useful in this table to list the water depth at which sediment cores were collected.*

Reply: We have included the water depths of sediment cores investigated in our study.

Additional references added to the revised version:

Aller, R.C.: Mobile deltaic and continental shelf muds as suboxic, fluidized bed reactors, Mar. Chem., 61, 143–155, 1998.

Aller, R.C., and Blair, N.E.: Carbon remineralization in the Amazon-Guianas tropical mobile mudbelt: A sedimentary incinerator, Continental Shelf Res., 26, 2241–2259, 2006.

Aller, R.C., Blair, N.E., Xia, Q., and Rude, P.D.: Remineralization rates, recycling and storage of carbon in Amazon shelf sediments. Continental Shelf Res., 16, 753–786, 1996.

Aller, R. C., Blair, N. E., and Brunskill, G.J.: Early diagenetic cycling, incineration, and burial of sedimentary organic carbon in the central Gulf of Papua (Papua New Guinea). J. Geophys. Res., 113, F01S09, 2008.

Bianchi, T.S., Allison, M.A., Canuel, E.A., Corbett, D.R., McKee, B.A., Sampere, T.P.,Wakeham, S.G., and Waterson, E.: Rapid export of organic matter to the Mississippi Canyon. EOS 87 (50), 565, 572–573, 2006.

Bianchi, T.S., Galler, J.J., and Allison, M.A.: Hydrodynamic sorting and transport of terrestrially-derived organic carbon in sediments of the Mississippi and Atchafalaya Rivers. Estuarine Coastal Shelf Sci., 73, 211–222, 2007.

Bianchi, T.S.: The role of terrestrially derived organic carbon in the coastal ocean: A changing paradigm and the priming effect. Proc. Nat. Acad. Sci., 108, 19,473-19,481, 2011.

Buesseler, K.O., Antia, A.N., Chen, M., Fowler, S.W., Gardner, W.D., Gustafsson, O., Harada, K., Michaels, A.F., Rutgers van der Loeff, M., Sarin, M., Steinberg, D.K., and Trull, T.: An assessment of the use of sediment traps for estimating upper ocean particle fluxes. J. Marine Res., 65, 345–416, 2007.

Carter, L., Milliman, J.D., Talling, P.J., Gavey, R. and Wynn R.B.: Near synchronous and delayed initiation of long run-out submarine sediment flows from a record-breaking river flood, offshore Taiwan. Geophys. Res. Lett., 39, L12603, 2012.

Sampere, T.P., Bianchi, T.S., Wakeham, S.G., and Allison, M.A.: Sources of organic matter in surface sediments of the Louisiana Continental Margin: Effects of primary depositional/transport pathways and Hurricane Ivan. Cont. Shelf Res., 28, 2472–2487, 2008.