Peer Review File

Manuscript Title: Neogene burial of organic carbon in the global ocean

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

****

Referee #1:

The carbon isotope mass balance approach to reconstructing past variations in organic carbon burial has become the norm in these types of studies. So, it is very refreshing to see someone take a new perspective on the topic and (potentially) develop new insights as a result.

Splitting the global ocean into different regions is obviously an approximation, but we have a limited number of sites so some assumptions have to be made. This seemed to be well justified as shown by comparison between the three different models of this aspect.

The key aspect of this paper is the method. Once the method is shown to be robust, then I am reasonably relaxed about the authors coming up with their own interpretation. If others disagree with that interpretation, they are free to come up with their own hypotheses. However, the underlying dataset must be shown to be reliable and adequate for addressing this important problem.

If you are going to take a radically new approach to a problem, then there is a heavy burden to make sure that you can justify your approach. In this context, the figure that worried me most was Figure 2. In this diagram, different areas of the ocean are shown as hot spots of OC burial that appear to switch off and on. For example, the east coast of the US is shown as a hot spot in the Miocene, but switches off in the Pliocene. The China Sea is a dark area for the early and middle Miocene, switches on in the late Miocene and then goes dark again in the Pliocene. The Namibian shelf is dark for the early and middle Miocene, starts to appear in the Late Miocene, and is then the brightest spot in the Pliocene.

How consistent is the OC concentration data? The DSDP/IODP program has been running for several decades and the analytical procedures on the ship (where many of the OC concentrations are measured) have changed during this time and there have also been changes in on-shore laboratory measurements (e.g. regarding pyrolysis temperatures and pre-treatment of samples). One check might be to look at old DSDP sites that are in close proximity to more recent IODP sites. Do they yield broadly similar OC results?

There are varying estimates of modern OC burial rates. Burdige (Chem. Rev. 2007, 107, 467-485) estimates ~0.3 Gt/yr – i.e. ~double that termed modern in this study. The authors say in the manuscript that they have “modern” (0.5 – 2.5 Ma) values of OC burial. I would like to see this map and how it compares with conventional core top measurements of OC burial. We would expect that there would be less OC burial during 0.5 – 2.5 Ma than modern, but we might expect the geographical patterns to be similar. If it looks a lot different, the authors will need to explain why.

In terms of the statistics, how do the number of sites used to compile the age variations in OC burial vary as a function of age? If the number of sites used within each time interval varies, it would be nice to see a plot of this in the supplementary information. In addition, are all the regions represented in each time slice? Again, if not then we need to see how this varies.

****

Referee #2:

Review of Li et al. for Nature

The authors seek to address the fact that global rates of OC burial are typically reconstructed using the mass balance and isotopic composition of inorganic and organic carbon, an approach that depends on a set of assumptions about carbon sources and isotopic averages, as well as major fluxes of the carbon cycle. Naturally direct observations would be better. In this study, the authors employed data from ocean drilling studies to directly calculate mass accumulation rates of OC, an effort made possible by the standardized collection of TOC, sedimentation rate, and bulk density data from DSDP, ODP and IODP cores. Their temporal focus was the Miocene and their objective was to address the so-called Monterey Hypothesis, which posits a global increase in OC burial to explain the positive d13C excursion of the mid-Miocene, and the decline in pCO2 following this event as a trigger for the MCT.

As someone who has tackled the problem of MAR reconstruction in ancient hemipelagic facies, I was impressed by many elements of this study and it strikes me as a perfectly logical step to take. I commend the authors on their exhaustive data compilation effort and their inclusion of various methods to assess the uncertainties in their results. Finally, if the results hold up over time, this work will have done great service as it turns the interpretation of OC burial during the Miocene on its head and demands a reconsideration of the Monterey Hypothesis. Such results seem very worthy of publication in Nature.

My criticisms of the paper are modest and may be easily addressed in a revision. The first issue with the paper is English grammar and syntax. I provide an anonymous track changes version of the ms. in which I have corrected these issues.

The next question I have concerns the implications of a Nature paper in which the methods section is almost as long as the text of the paper – this naturally makes me wonder if the study might not be better suited for a longer format journal (but I understand that everybody wants a Nature paper if the significance of their results can make the cut). This issue is obviously the purview of the editors and I assume they are generally OK with it or they would not have sent the ms. out for review.

Lastly, I have two specific questions about methodology:

(1) How are MAR values from one or two sites used to extrapolate to vastly larger areas? I understand that an algorithm is used – it is referenced, but not presented or fully explained. One

might assume that the selection of 'biogeochemical provinces' has already done part of the work by identifying regions with similar conditions of OC production and preservation, but those unfamiliar with the literature on more recent marine biogeochemistry may be left wondering about the details.

(2) The authors argue that %TOC is the dominant control on MAR-OC. Measured %TOC must be a consequence of both the OC flux AND the diluent flux and by itself, is essentially a resultant observable, not a process. This issue may be simply semantic – after the efforts to constrain the Sadler Effect, I believe the authors are arguing that changes in bulk sediment accumulation alone cannot account for the pattern revealed in the time series of OC burial observed in Figures 2 and 4 (i.e., after the artifactual effects of differences in sampling resolution are accounted for). What I believe this means is that patterns of OC flux and preservation (which produce the resultant %TOC values) are more important. My problem is one of circular reasoning – isn't the preservation factor related to the bulk sedimentation rate? I would hope the authors can clarify this issue in their revision.

*************************END*************************

Author Rebuttals to Initial Comments:

Response to Reviews

We thank both reviewers for their thoughtful comments, and the editor for handling o ur manuscript. All issues raised have been either addressed or rebutted as detailed below.

Reviewer #1

1. The carbon isotope mass balance approach to reconstructing past variations in organic carbon burial has become the norm in these types of studies. So, it is very refreshing to see someone take a new perspective on the topic and (potentially) develop new insights as a result.

Splitting the global ocean into different regions is obviously an approximation, but we have a limited number of sites so some assumptions have to be made. This seemed to be well justified as shown by comparison between the three different models of this aspect.

The key aspect of this paper is the method. Once the method is shown to be robust, then I am reasonably relaxed about the authors coming up with their own interpretation. If others disagree with that interpretation, they are free to come up with their own hypotheses. However, the underlying dataset must be shown to be reliable and adequate for addressing this important problem.

If you are going to take a radically new approach to a problem, then there is a heavy burden to make sure that you can justify your approach. In this context, the figure that worried me most was Figure 2. In this diagram, different areas of the ocean are shown as hot spots of OC burial that appear to switch off and on. For example, the east coast of the US is shown as a hot spot in the Miocene, but switches off in the

Pliocene. The China Sea is a dark area for the early and middle Miocene, switches on in the late Miocene and then goes dark again in the Pliocene. The Namibian shelf is dark for the early and middle Miocene, starts to appear in the Late Miocene, and is then the brightest spot in the Pliocene.

We agree with the referee that demonstrating that the methodology we used here is sound and reliable is a critical component of this work. We took this opportunity to further examine and explain all aspects of the approach that we used, from the raw data (e.g., TOC% data) all the way to global OC burial rates. This has led to expansion of discussions in the main text, new sections in ‘Methods’, and new extended data figures and tables. <u>These efforts further demonstrate the robustness of this “bottom up” approach for constraining OC burial changes in the global ocean over time.</u>

We understand the concern of the Referee regarding the original Figure 2 which plots TOC MAR from individual sites on a global map. The hotspots shown here are clearly limited spatially and temporally by the available IODP records with high quality and continuous TOC MAR records. However, for this exact reason, instead of solely using these individual records, we used them (or their mean value if more than one records exist for any province) to scale for OC burial variability in the provinces and then to compute global OC burial rates based on these. This approach reduces the sensitivity of calculated global OC burial to any individual TOC MAR record.

To improve clarity, we have moved the original Fig. 2 to the extended data (Extended Data Fig. 4) and instead, our new Fig. 2 shows relative burial rate changes of each province (a) and their percent contribution to global burial at any particular time (b). With these new illustrations we hope to provide a better presentation of the regional or provincial data which are more directly linked to our calculated global OC burial, and also make it easier for the reader to perceive how the final product is produced.

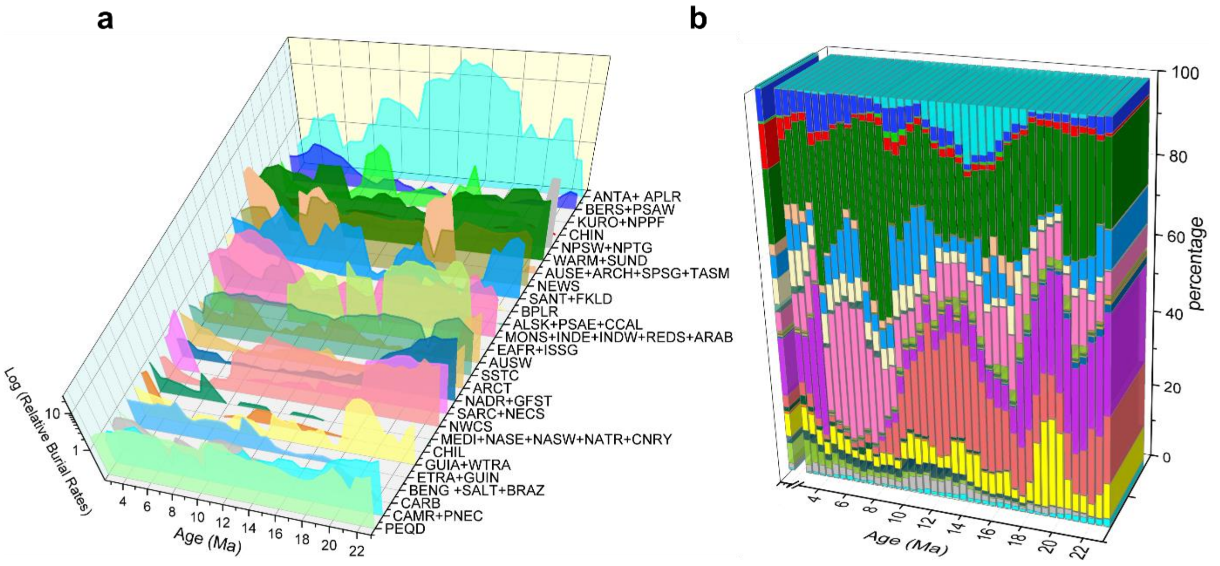


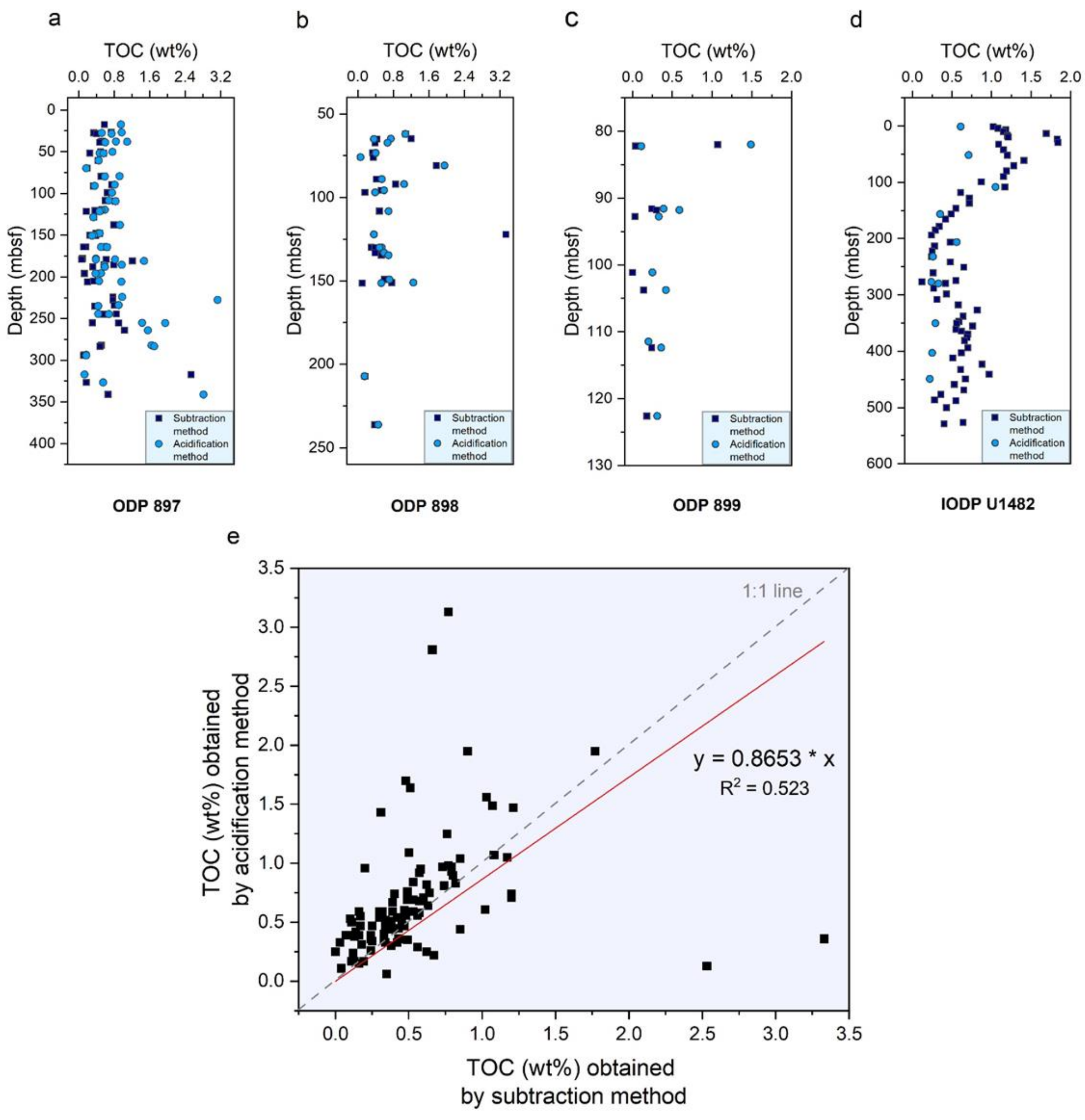
Fig. 2 | Provincial OC burial changes and their contribution to the global OC burial. a. Relative changes of provincial OC burial rates over time, with the “modern” (Pleistocene) value defined as 1; **b**. Provincial contribution to the global OC burial rates during the Neogene (total = 100%). The provinces presented here are identical to those shown in Fig. 1. Refer to Supplemental Data Table 1 for the details of each province and the IODP sites used to construct provincial records.

2. How consistent is the OC concentration data? The DSDP/IODP program has been running for several decades and the analytical procedures on the ship (where many of the OC concentrations are measured) have changed during this time and there have also been changes in on-shore laboratory measurements (e.g. regarding pyrolysis temperatures and pre-treatment of samples). One check might be to look at old DSDP sites that are in close proximity to more recent IODP sites. Do they yield broadly similar OC results?

The Referee raises an important point. Below we briefly describe the common methods used for determining TOC% of marine sediments, summarize previous views on the accuracy of IODP-derived TOC% data, and then explore the consistency of OC data following the suggested "tests" by the Referee. The figures and tables shown here are also included in the revised submission. These tests confirm that the OC concentration data is sufficiently consistent for use in this study.

DSDP/ODP/IODP (hereafter referred to as IODP) use a "subtraction" method onboard to determine TOC%, which is based on the differences between the total carbon measured by a CHNS elemental analyzer (EA) and the inorganic carbon measured by a coulometer. This method is generally considered to be accurate. For example, Meyers and Silliman (1996) replicated the ship-board TOC% measurements by performing shore-based experiments that removed the carbonate fraction of samples through acid digestion. The residues were then washed, dried, and measured on an EA. After comparing the results, their shore-based results (0.06 -3 wt% TOC) were positively correlated with the shipboard values, from which they concluded that "little error was introduced into TOC determinations over a wide range of concentrations" when using standard shipboard by-difference procedures (Meyers and Silliman, 1996). However, Olivarez Lyle and Lyle (2005) argued that the subtraction method introduces large errors when TOC% is low (<0.3%). By using carefully designed acid digestion method, Olivarez Lyle and Lyle (2006) were able to report very low mean TOC concentrations of about 0.03 wt% at Sites 1218 and 1219, levels that are "below detection" using the standard subtraction method (Shipboard Scienfitic Party, 2002). In this case, the acidification method clearly showed its advantage over the standard subtraction method when sediment samples bear very low TOC%. However, samples which have "0%" or "below detection" levels of TOC constitute only 2.5% of our database.

Several IODP publications also attempted to evaluate of the accuracy of the standard IODP method for determining TOC% (e.g., Expedition 306 Scientists, 2006; Expedition 317 Scientists, 2011; Rosenthal et al., 2018). For example, shipboard organic geochemists M. Yamamoto and Y.G. Zhang (co-author of this MS) of Expedition 363 went beyond their regular job duties onboard of the *JOIDES Resolution* and applied acid digestion to Site U1482 samples to verify the TOC% data obtained by the subtraction method (Rosenthal et al., 2018). Here, we compiled all available TOC% data that have been constructed from both the "subtraction" and "acidification" methods and compare them downcore as well as scatter plot on the absolute abundance scale. Some offsets between the two datasets were observed. Nevertheless, the overall relationship suggests that the two independent methods yield comparable results (p-value = 1.38^{e-17}), supporting the robustness of the standard subtraction method for TOC% used by IODP. Assuming that the "acidification" method yields more accurate results, statistics show a general uncertainty of the "subtraction" method of ~22%.

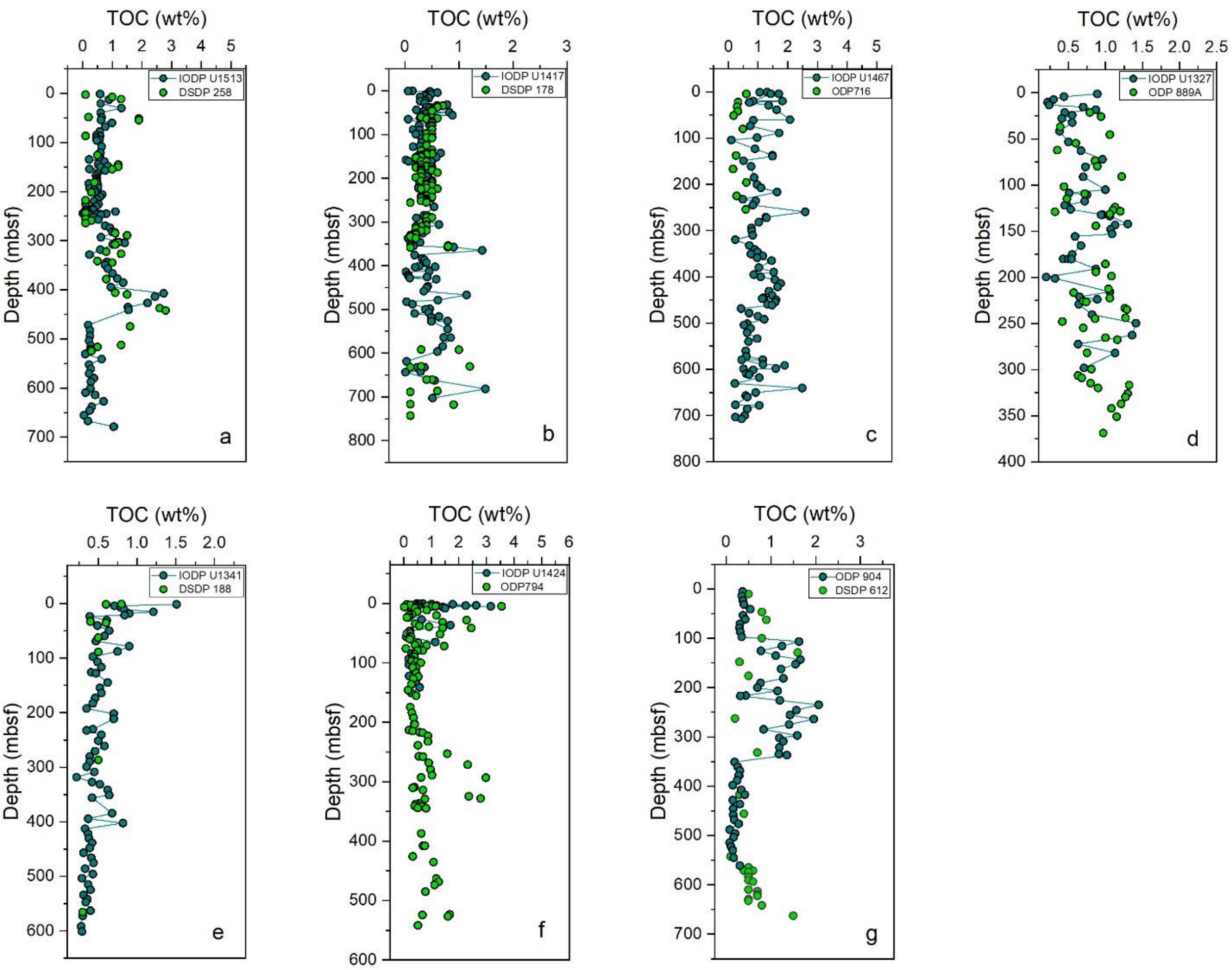


Extended Data Fig. 1 | Comparisons of TOC% obtained by "subtraction" and "acidification" methods. a-c. ODP Sites 897-899. d. IODP Site U1482. Navy blue squares are the measurements on board by subtraction between total and carbonate carbon contents, while blue dots are from the acidification method direct measured from carbonate-free samples. **e. Linear regression of TOC contents determined by the two independent methods.** The regression equation is expressed as $y = 0.8653*x$, with $R^2 = 0.523$ and p-value = 1.38^{e-17}, suggesting a significant linear relationship at the 0.05 level of significance. The dashed grey line indicated the one-to-one correspondence of the two variables.

Also, we followed the Referee's suggestion to verify the reproducibility of TOC data reported by DSDP, ODP or IODP expeditions from nearby locations. This type of comparison is enabled by IODP expeditions that revisited earlier DSDP or ODP site locations. Since coordinates and water depth are almost identical for the original and revisited sites, a direct comparison of TOC% on the depth scale (y-axis) was conducted. This type of "revisit" includes ODP Site 794 and IODP Site U1424 (see figure and table below).

When the sites were not directly "revisited" but still drilled within a short distance and similar water depth, we used a criterion of 10 km and 200 m to identify sites that qualify for comparisons, with one exception of IODP U1341 and DSDP 188 that are slightly further apart but can be correlated by physical property data. For sites that are beyond this range, substantially different sedimentation rates would make TOC% comparison on the depth scale much more difficult.

DSDP, ODP and IODP-derived TOC% results show broad agreement, except for the comparison of ODP Site 716 and IODP U1467 where the ODP results constantly represent the lower end of the IODP measurements. The resemblance could improve further if age instead of depth was used for such comparisons. However, this would require precise age models for all sites examined here which is beyond the scope of the current study.



Extended Data Fig. 2 | Comparison of TOC% reported by DSDP, ODP or IODP expeditions from nearby locations. a. IODP U1513 vs DSDP 258. **b**. IODP U1417 vs DSDP 178. **c**. IODP U1467 vs ODP 716. **d**. IODP U1327 vs ODP 889A. **e**. IODP U1341 vs DSDP 188. **f**. IODP U1424 vs ODP 794. **g**. ODP 904 vs DSDP 612. Sites details are shown in Extended Data Table 2.

Extended Data Table 2 | Location, water depth and distance of sites compared in Extended Data Fig. 2.

	Exp/Leg	Site	Longitude	Latitude	Water depth (m)	Distance (km)	notes
a	Exp 369	IODP U1513	112.486	-33.794	2800m	1.1km	
	Leg 26	DSDP 258	112.474	-33.795	2793m		
b	Exp 341	IODP U1417	-147.110	56.960	4188m	1.338km	
	Leg 18	DSDP 178	-147.131	56.956	4218m		
c	Exp 359	IODP U1467	73.284	4.850	487m	9.206km	
	Leg 115	ODP 716	73.283	4.933	544.3m		
d	Exp 311	IODP U1327	-126.865	48.698	1322.1m	0.257km	
	Leg 146	ODP 889A	-126.868	48.699	1311.2m		
e	Exp 323	IODP U1341	179.008	54.033	2140m	38.562km	
	Leg 19	DSDP 188	178.659	53.754	2649m		

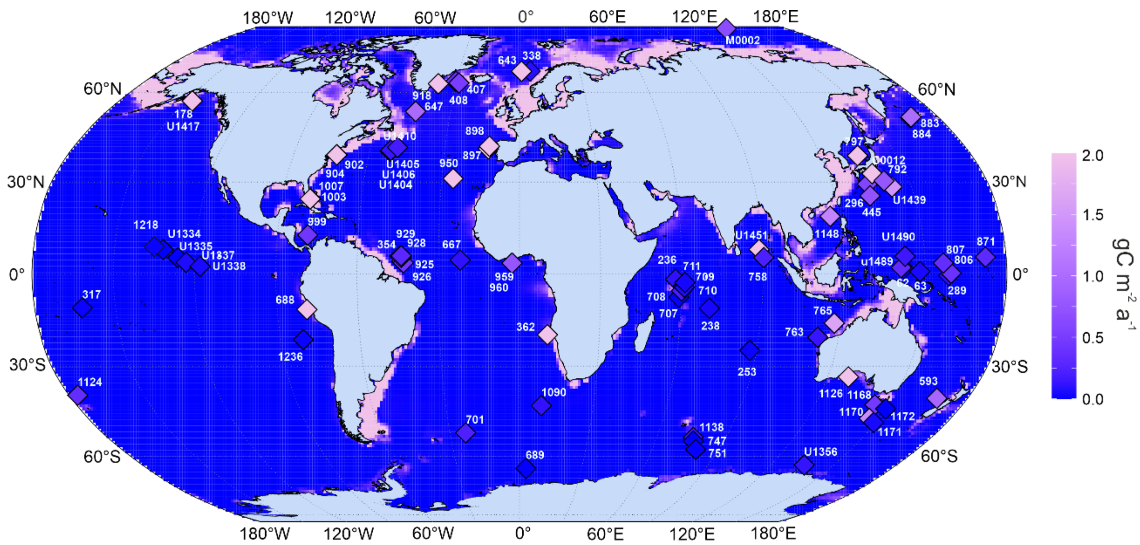
f	Exp 346 Leg 127	IODP U1424 ODP 794	138.232 138.231	40.190 40.190	2808m 2820m	0km	revisit
g	Leg 150 Leg 95	ODP 904 DSDP 612	-72.768 -72.774	38.863 38.820	1123m 1386m	4.826km	

3. There are varying estimates of modern OC burial rates. Burdige (Chem. Rev. 2007, 107, 467-485) estimates ~0.3 Gt/yr – i.e. ~double that termed modern in this study. The authors say in the manuscript that they have "modern" (0.5 – 2.5 Ma) values of OC burial. I would like to see this map and how it compares with conventional core top measurements of OC burial. We would expect that there would be less OC burial during 0.5 – 2.5 Ma than modern, but we might expect the geographical patterns to be similar. If it looks a lot different, the authors will need to explain why.

We thank the Referee for pointing us to the Burdige paper, which is now cited in the manuscript. Different estimates of modern OC burial rates in the global ocean are now acknowledged in "Calculation of regional and global OC burial" section. There is a range of estimates for modern OC burial rates, and Burdige (2007) of 309 Tg C/yr is close to the higher end, which was acknowledged in that exact paper: "In particular, many of these burial estimates are significantly larger than the commonly cited whole ocean OC burial rate of 120-160 Tg C yr^{-1}". Because this estimate is dependent on the calculated remineralization rates, Burdige (2007) discussed the possibility that this OC budget could have integrated results over different timescales due to the fact that even "core-top" sediments could carry signals that are tens of thousands of years old. Our cited modern OC burial is from Dunne et al. (2012) which was based on a series of data and algorithms considering satellite-based primary production, particulate OM generation, transportation and burial in the sediments. Their estimate of 146 Tg or 0.146 Gt C/yr is confirmed by a recent global study (Hayes et al., 2021) that <u>utilized ^{230}Th-normalized fluxes to minimize the age model uncertainty – the exact problem pointed out by Burdige (2007)</u>. This study compiled results from 12,000 globally distributed marine cores and reported <u>"deep water" (>2,000 m)</u> annual OC burial of 17±5 Tg C/yr that is much more in line with Dunne et al. (2007, 2012) deep water rate of 12 Tg C/yr. Also, the essence of our approach is to use the Neogene TOC MAR records to scale for global OC burial changes (modern burial x variability), such that the results can easily be modified if our understanding of the modern global burial further improves in the future.

Modifications have been made to "Calculations of regional and global OC burial" section, which now reads: "*Modern OC burial in the global ocean with a 1×1° spatial resolution of Dunne et al.*[27,60] *is the basis for our modern provincial OC burial rates, obtained from areal integrals of all data points within that province (Extended Data Fig. 6). This modern OC burial map is produced by a series of data and algorithms considering satellite-based primary production, particulate organic matter generation, transportation and burial in the sediments, which leads to a global burial of ~0.146 Gt* *yr^{-1} (Ref [27,60]). However, different methods have yielded different estimates of modern burial, possibly due to the fact that core-top samples could carry signals that are tens of thousands of years old but the sedimentation/remineralization rates were poorly constrained[61]. Nonetheless, recent studies utilized ^{230}Th-normalized fluxes to tackle this issue by minimizing age model uncertainties. For example, Hayes et al.[62] compiled results from 12,000 globally distributed marine cores and 1,068 flux estimates of the deep ocean. Their reported "deep water" (>2,000 m) annual OC burial of 0.017±0.005 Gt yr^{-1} is much in line with Dunne et al. (2007, 2012) deep water rate of 0.012 Gt yr^{-1}, supporting the robustness of the modern OC burial map used in this study*". Also, after discussing how to use modern OC burial and provincial variability based on TOC MARs, we mentioned that "*This approach also allows for easy adjustments if our understanding of the modern global burial further improves in the future*".

Indeed, we used the 81 TOC MAR records plus the modern provincial burial rates to define the Pleistocene (2.5-0.5 Ma) or "modern" burial. This approach intends to leverage the relatively sparse data while reducing the error introduced by any individual record, and is comparable with recent studies on the burial fluxes on late Pleistocene (Cartapanis et al., 2016) and modern (Hayes et al., 2021) timescales. We noted that "core-top" type of studies that define the actual modern burial utilize hundreds, thousands, or tens of thousands of sediment core datasets (cf., Dunne et al., 2012; Dunne et al., 2007; Hayes et al., 2021; Jahnke, 1996), vs 81 IODP sites used here. Also, as we acknowledged in the MS, IODP typically operates in relatively deeper water (>500 m), potentially missing the modern day "hotspots" of OC burial on continental shelves (L153-166). Consequently, instead of a direct comparison of OC burial rates between our "modern" burial with core-top results, we plotted the color-coded "modern" (Pleistocene) OC burial rates of the 81 individual sites on the Dunne et al. (2007, 2012) map, which show broad agreement.



Extended Data Fig. 6 | "Modern" OC burial rates of the global ocean. The map was generated by data from Dunne et al. (2007, 2012). Also shown are "modern" (Pleistocene) burial rates of our 81 IODP sites (in diamond squares) color coded with the same scheme of the Dunne map.

Overall, we believe that this work further demonstrates the robustness of the TOC% measurements carried out by IODP (see answers to Q#2). Additionally, IODP represents a rare, if not the only opportunity to broadly examine long-term global carbon cycle from sedimentary records.

4. In terms of the statistics, how do the number of sites used to compile the age variations in OC burial vary as a function of age? If the number of sites used within each time interval varies, it would be nice to see a plot of this in the supplementary information. In addition, are all the regions represented in each time slice? Again, if not then we need to see how this varies.

We have now added a supplemental data table to detail the sites used for each province during any of the 0.5 Myr time windows from 23 to 5 Ma. These results are also visualized in the new Fig. 2. When we produced the data compilation, we exhausted all possible ways to ensure that each province is represented by at least one site over any particular time interval. Where data from our primary sites are unavailable, we supplemented the results by using nearby sites typically drilled by the same expedition. On average, any particular site used in this study has data to cover 76 ± 7.9% (1σ) of the Neogene (23-2.5 Ma segmented into 0.5 Myr time windows). The temporal coverage for any particular province is 95.3% ± 4.1% (1σ). For the remaining 4.7%, data were interpolated to ensure that the contribution of any province does not drop to 0 simply because of unavailable TOC MAR data. Interpolated data are clearly labelled in the data table.

Reviewer #2

1. The authors seek to address the fact that global rates of OC burial are typically reconstructed using the mass balance and isotopic composition of inorganic and organic carbon, an approach that depends on a set of assumptions about carbon sources and isotopic averages, as well as major fluxes of the carbon cycle. Naturally direct observations would be better. In this study, the authors employed data from ocean drilling studies to directly calculate mass accumulation rates of OC, an effort made possible by the standardized collection of TOC, sedimentation rate, and bulk density data from DSDP, ODP and IODP cores. Their temporal focus was the Miocene and their objective was to address the so-called Monterey Hypothesis, which posits a global increase in OC burial to explain the positive d13C excursion of the mid-Miocene, and the decline in pCO2 following this event as a trigger for the MCT.

As someone who has tackled the problem of MAR reconstruction in ancient hemipelagic facies, I was impressed by many elements of this study and it strikes me as a perfectly logical step to take. I commend the authors on their exhaustive data compilation effort and their inclusion of various methods to assess the uncertainties in their results. Finally, if the results hold up over time, this work will have done great service as it turns the interpretation of OC burial during the Miocene on its head and demands a reconsideration of the Monterey Hypothesis. Such results seem very worthy of publication in Nature.

My criticisms of the paper are modest and may be easily addressed in a revision. The first issue with the paper is English grammar and syntax. I provide an anonymous track changes version of the ms. in which I have corrected these issues.

We are very grateful to the Referee for these corrections. All edits have been accepted and the presentation of our work is much improved. Thank you!

2. The next question I have concerns the implications of a Nature paper in which the methods section is almost as long as the text of the paper – this naturally makes me wonder if the study might not be better suited for a longer format journal (but I understand that everybody wants a Nature paper if the significance of their results can make the cut). This issue is obviously the purview of the editors and I assume they are generally OK with it or they would not have sent the ms. out for review.

To ensure all necessary information is provided, our method section is indeed lengthy, but on par with typical Nature articles published recently. Considering the Referee's comment, we took this opportunity of revision to streamline the method section and to ensure it is both informative and succinct.

3. Lastly, I have two specific questions about methodology:

(1) How are MAR values from one or two sites used to extrapolate to vastly larger areas? I understand that an algorithm is used – it is referenced, but not presented or fully explained. One might assume that the selection of 'biogeochemical provinces' has already done part of the work by identifying regions with similar conditions of OC production and preservation, but those unfamiliar with the literature on more recent marine biogeochemistry may be left wondering about the details.

We thank the Referee for pointing this out and have improved our explanation of the algorithm in this revision.

The new Fig. 2 and Extended Data Table 1 now provide key components used to calculate the global OC burial with the goal of helping our readers to better understand the workflow of generating the final product of global OC burial. We also added the equation (Equation 6) used to calculate global OC burial from provincial OC MARs. Lastly, the definition and necessity of Longhurst biogeochemical provinces was better elaborated in the main text –L151-154 now reads "*The definition of Longhurst provinces was based on atmospheric circulation, light,* *coastlines, water column stratification, chlorophyll content and other environmental factors*[16] *and has been widely applied in marine biogeochemistry studies*".

4. (2) The authors argue that %TOC is the dominant control on MAR-OC. Measured %TOC must be a consequence of both the OC flux AND the diluent flux and by itself, is essentially a resultant observable, not a process. This issue may be simply semantic – after the efforts to constrain the Sadler Effect, I believe the authors are arguing that changes in bulk sediment accumulation alone cannot account for the pattern revealed in the time series of OC burial observed in Figures 2 and 4 (i.e., after the artifactual effects of differences in sampling resolution are accounted for). What I believe this means is that patterns of OC flux and preservation (which produce the resultant %TOC values) are more important. My problem is one of circular reasoning – isn't the preservation factor related to the bulk sedimentation rate? I would hope the authors can clarify this issue in their revision.

This is a very well-taken point - we totally agree with the referee that the measured TOC% must be the result of OC flux and diluent flux, which is essentially a resultant observation in itself, rather than a process. In the original MS method section we stated, "although these three factors (referring to SR, TOC%& and DBD) are not independent of each other" to acknowledge this point. However, we realized that this point has not been fully elaborated and therefore made entirely clear to our readers. We therefore expanded this section to discuss the nature of interdependence of TOC% and sedimentation rate and provided ample warning that these are simple fitting analyses. The following is now added to the "Apparent controls of OC MARs" section: "*To evaluate the apparent controlling factor of OC burial, we performed simple liner regression fittings between MAR and SR, TOC%, and DBD of each site. Importantly, SR and TOC% are interdependent – for example, rapid sedimentation dilutes OC flux, whereas quick burial potentially reduces the exposure time of OC to oxygen and therefore enhances the preservation of organic matter. Our analysis does not distinguish between these interconnections".* And the concluding sentence now reads: "*Our results show that TOC content appears to exert the dominant control of OC burial in a large number of sites, although TOC% itself is impacted by SR (Extended Data Fig. 10)*".

5. Line 41-42 "Our results demonstrate the necessity to consider a larger role for the OC cycle over geologic timescales." Don't you mean a larger role for changes in the source and isotopic composition of carbon inputs to the oceans? The prominent d13C perturbations associated with OAE's are largely driven by changes in the OC cycle, right?

Both. Since our global OC burial shows much larger variability than shown by more traditional global carbon cycle models, we argue that it should play a larger role in our perception of the global carbon cycle than previously assumed (e.g. for controlling long-term CO_2 levels). Our results also show a larger role of changing carbon source and isotopic composition, which is discussed in L260-290. Although OAEs are not a topic of this MS, it is within our interest and we speculate that both OC burial and changing carbon input due to intense volcanism (e.g., Ontong Java Plateau) might have played a role in driving marine $\delta^{13}C$ changes.

6. Line 129-130 “enhanced production and preservation at these places contribute to high OC accumulation rate” But they are all regions of overall higher bulk sediment accumulation, which is essentially part of the preservation process – i.e., the Hartnett et al. (1998) work on oxygen exposure time. Obviously primary organic matter flux and oxygen concentration and renewal rate play a role, but I am struggling with your later assertions that %TOC is a dominant controlling factor on MAR’s. The ultimate measured %TOC must be a consequence of both the OC flux AND the diluent flux – it is a resultant observable, not a process in and of itself.

Agreed. Hartnett et al. (1998) is now cited, and the controlling factors for OC MARs are discussed in the response to Q4 above.

7. Line 241 “e. Our estimates of OC burial in the global ocean” Does the uncertainty envelop correspond to the dark purple shading in Figure 3?

Yes – thanks for pointing this out. This has been clarified in the revised version, which now reads: “*Neogene OC burial in the global ocean with ± 1σ uncertainty envelope*”.

Cited References:

Burdige D. J., 2007. Preservation of organic matter in marine sediments: controls, mechanisms, and an imbalance in sediment organic carbon budgets? *Chemical Reviews* **107**, 467-485.

Cartapanis O., Bianchi D., Jaccard S. L., and Galbraith E. D., 2016. Global pulses of organic carbon burial in dee-sea sediments during glacial maxima. *Nat. Commun.* **7**, 10796.

Dunne J. P., Hales B., and Toggweiler J. R., 2012. Global calcite cycling constrained by sediment preservation controls. *Global Biogeochem. Cyc.* **26**, GB3023, doi:10.1029/2010GB003935.

Dunne J. P., Sarmiento J. L., and Gnanadesikan A., 2007. A synthesis of global particle export from the surface ocean and cycling through the ocean interior and on the seafloor. *Global Biogeochem. Cycles* **21**, GB4006, doi:10.1029/2006GB002907.

Expedition 306 Scientists, 2006. Site U1312-U1315 methods. In: Channell J. E. T., T. Kanamatsu, T. Sato, C. A. Alvarez Zarikian, M. J. Malone, and the Expedition 303/306 Scientists Eds.), *Proceedings of the Integrated Ocean Drilling Program*. IODP, College Station, TX.

Expedition 317 Scientists, 2011. Methods. In: Fulthorpe C. S., K. Hoyanagi, P. Blum, and the Expedition 317 Scientists Eds.), *Proceedings of the Integrated Ocean Drilling Program*. IODP, College Station, TX.

Hayes C. T., Costa K. M., Anderson R. F., Calvo E., Chase Z., Demina L. L., Dutay J.-C., German G. R., Kohfeld K. E., Kravchishina M. D., Lippold J., Mekik F., Missiaen L., Pavia F. J., Paytan A., Pedrosa-Pamies R., Petrova M. V., Rahman S., Robinson L. F., Roy-Barman M., Sanchez-Vidal A., Shiller A., Tagliabue A., Tessin A. C., van Hulten M., and Zhang J., 2021. Global Ocean sediment composition and burial flux in the deep sea. *Global Biogeochem. Cyc.* **35**, e2020GB006769.

Jahnke R. A., 1996. The global ocean flux of particulate organic carbon: Areal distribution and magnitude. *Global Biogeochem. Cyc.* **10**, 71-88.

Meyers P. A. and Silliman J. E., 1996. Organic matter in Pleistocene to Quaternary turbidites from Sites 897, 898, 899 and 900, Iberia Abyssal Plain. In: Whitmarsch R. B., D. S. Sawyers, A. Klaus, and D. G. Masson Eds.), *Proceedings of the Ocean Drilling Program, Scientific Results*. Ocean Drilling Program, College Station, TX.

Olivarez Lyle A. and Lyle M., 2005. 20. Organic carbon and barium in Eocene sediments: Possible controls on nutrient recycling in the Eocene equatorial Pacific Ocean. In: Wilson P. A., M. Lyle, and J. V. Firth Eds.), *Proceedings of the Ocean Drilling Program, Scientific Results*. Ocean Drilling Program, College Station, TX.

Olivarez Lyle A. and Lyle M., 2006. Missing organic carbon in Eocene marine sediments: Is metabolism the biological feedback that maintains end-member climates. *Paleoceanography* **21**, 10.1029/2005PA001230.

Rosenthal Y., Holbourn A., Kulhanek D. K., Aiello I. W., Baliba T. L., Bayon G., Beaufort L., Bova S. C., Chun J.-H., Dang H., Drury A. J., Dunkley Jones T., Eichler P. P. B., Fernando A. G. S., Gibson K. A., Hatfield R. G., Johnson D. L., Kumagai Y., Li T., Linsley B. K., Meinicke N., Mountain G. S., Opdyke B. N., Pearson P. N., Poole C. R., Ravelo A. C., Sagawa T., Schmitt A., Wurtzel J. B., Xu J., Yamamoto M., and Zhang Y. G., 2018. Site U1482. In: Rosenthal Y., A. Holbourn, D. K. Kulhanek, and the Expedition 363 Scientists Eds.), *Proceedings of the International Ocean Discovery Program*. IODP, College Station, TX.

Shipboard Scienfitic Party, 2002. 12. Site 1219. In: Lyle M., P. A. Wilson, and T. R. Janecek Eds.), *Proceeding of the Ocean Drilling Program, Initial Reports*. Ocean Drilling Program, College Station, TX.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I really like the concept of this paper. For too long, studies of organic carbon burial have gone round in circles with “tweaks” to isotope fractionation factors and ever more sub-divisions of input and output fluxes to balance models. In contrast, this paper takes what might be considered the very obvious approach of actually measuring burial rates. Of course, it is not as simple as that – we have a limited number of suitable sites to study and the lack of data around the Antarctic and in the centres of the North and South Pacific gyres is of particular concern. In addition, there are questions around age models, sedimentation rates, ocean provinces, etc. However, none of these issues are within the control of the authors and I believe they have done as good a job as is possible with the data that are available. They have engaged constructively with my original queries (reviewer #1) and I am happy to see the present paper published.
This paper is not the last word on the subject, but the authors have done as well as is possible in acknowledging and addressing the many uncertainties, while still proposing some testable hypotheses. And this final point is important. While I have no doubt that this paper will attract considerable comment (and possibly criticism), the hypotheses presented here are testable with targeted data collection in future expeditions. The same cannot be said for many of the more esoteric isotope balancing models.

Minor queries
Why is Figure 2b presented in 3-dimensions? There is no label on the z-axis and no indication of any changes along this axis. Why not just use a simple x-y plot?
Line 176 – exemplified not “examplified”

Martin Palmer

Referee #2 (Remarks to the Author):

I really liked this paper the first time I reviewed it based on the conceptual approach and the implications of the results. I believe that I said these things qualified it for publication in Nature. I then made a series of comments about methodology, etc. in addition to text editing for grammar and sentence construction. The other reviewer of the paper made similar comments about methodology, data quality, etc. In their revision, the authors have done an excellent job of addressing the comments of both reviewers and I found the ms. text to be much improved. I think it reads quite well, and at this point the authors have done a very fine job of making every aspect of the work transparent so that the community can evaluate and independently verify the results. I think this paper should be published as is.

Author Rebuttals to First Revision:

Response to Reviews

Reviewer #1

1. Minor queries

Why is Figure 2b presented in 3-dimensions? There is no label on the z-axis and no indication of any changes along this axis. Why not just use a simple x-y plot?

We felt that a 3D plot of Fig. 2b is more consistent with Fig. 2a and together they present the absolute and relative contribution of each province to global OC burial.

2. Line 176 – exemplified not "examplified"

Corrected. Thank you!