<u>Author's response to referee comment (RC) 2</u>

We would like to thank referee #2 for his/her comments, which we think will significantly improve the quality of our manuscript. Below, we provide a detailed point-to-point response addressing the reviewer's main concerns. New Supplementary Figures 1-6 and a Supplementary Table are referenced in this response and are included at the end of this document.

This manuscript argues that variation between high TOC and modest TOC Aptian black shales at DSDP 361 in the Cape Basin was forced by changes in precipitation patterns over southern Africa. These relatively short-term climatic variations are proposed to be separate from a longer-term tectonic shifts that led organic carbon-rich deposits to be replaced organic carbon-poor deposits from the Aptian through the Albian at this site. Primary evidence cited in support of the climatic interpretation is biomarker analyses, REE distributions, and modelling results. In addition, elemental abundances and other bulk geochemical results are discussed relative to potential diagenetic overprints and redox conditions in the water and sediment column. Data are largely limited to a portion of the lithologies present at one site that seemingly had quite incomplete recovery.

The interpretation is interesting and consistent with the data presented, but to fit the observations, the interpretation does invoke a rather specific set of conditions (e.g., changes in seasonality and geographic focus of precipitation leading to more precipitation and nutrient input from chemical weathering in the interior but more physical weathering along the coast) that are difficult to test. The study also leans heavily on Dummann et al., 2020, EPSL, for data and documentation to the point that I wonder if the last sentence of the first paragraph of the discussion of the EPSL paper refers to the data split off and presented in this manuscript. Regardless of decisions made by authors about whether and how to partition findings among publications, this manuscript would be stronger if the unique contributions of this study were featured and less prominence was given to the broader and longer-term topics addressed in the EPSL paper.

We acknowledge that there is a degree of overlap in content between the present study and our previous study published in EPSL. However, we would like to stress that long-term changes in bottom water redox state related to the opening of South Atlantic-Southern Ocean gateways, targeted in the EPSL publication, played a pivotal role in preconditioning the Early Cretaceous Cape Basin for enhanced organic carbon burial. Different to that, this study focusses on the internal dynamics of organic carbon burial in the Cape Basin, based on diverse and new geochemical data combined with regional climate modeling. Climatic forcing mechanisms acting on shorter time-scales modulated the magnitude of organic carbon burial (i.e. changes between high-TOC and low-TOC black shales), but without an appropriate basin configuration, they were insufficient to generate enhanced organic carbon burial, as stated in e.g. lines 546-547. As such, we consider it important to recapitulate trends in long-term basin evolution to provide a wider context for the fluctuations in organic carbon burial presented and discussed in this study. We are therefore confident that this study presents genuine and new data with a distinct interpretation, which is enriched by reference to other relevant studies.

Emphasizing and expanding presentation of sedimentological data would be one route to this end especially given the importance of deposition patterns and sediment sources in the conclusions. Currently the lithologic data presented are relatively sparse. The paper proposes 3 (or 4) lithostratigraphic units are present that can be differentiated based on color and TOC of the finest grained lithologies. Is there any information on mineralogy, bedding characteristics, or sedimentary structures (physical or trace fossils)?

We fully agree that sedimentology is a central element to understand carbon preservation and we were fortunate to have a large body of sedimentological work already covered in the public literature with a focus on our study site. We consequently decided not to include further

sedimentological analyses into the workflow but rather ensure full coverage of the literature data and interpretation in the revised manuscript. We are confident that the sedimentology presented balances the distinct focus of this study on geochemistry and modeling. Our choice of lithostratigraphic units largely conforms with the units defined by the shipboard scientific party of DSDP Leg 40 (The Shipboard Scientific Party, 1978; Natland, 1978). They placed the boundary between their lithologic units 6 and 7 atop core 28 (The Shipboard Scientific Party, 1978), consistent with the transition from black shale to gray shale deposition proposed in the present study (Figure 2). This transition marks a profound change in the sedimentary environment, as indicated by the appearance of cross-laminated siltstone beds (Natland, 1978; Kagami, 1978) and burrowing structures (The Shipboard Scientific Party, 1978). The shipboard scientists interpreted these lithological changes to reflect a strengthening of deep water circulation and ventilation, respectively, which is consistent with Nd isotope evidence presented in our EPSL paper (Dummann et al., 2020) and a shift towards more oxygenated conditions evident in our new data (cf. lines 312–317, Figure 4, Figure 5). In contrast, black shales in cores 28–48 below (lithostratigraphic unit 7 as defined by the The Shipboard Scientific Party (1978)) are mostly fissile, partly laminated, and lack bioturbation, consistent with a suboxic–anoxic environment (cf. lines 291–311 and Fig. 4). However, it is important to note that gray shales and red beds share similar sedimentary characteristics and thus have been assigned to the same lithological unit 6 by the shipboard scientists. We distinguish them based on their distinct color, differences in TOC content (Figure 2c), S content (Figure 4b), redox-sensitive trace metal enrichment (Figure 4c–f), and Fe-S-TOC relationships (Figure 5), which indicate a shift to fully oxic conditions during the deposition of red beds.

Description of the samples analyzed, their context, and why they were chosen is effectively missing (both for the 'complete set' and the presumably subsets thereof in plots with < than 131 data points).

The criteria for sample selection are detailed below and will be included in the revised manuscript.

What about the coarser grained lithologies? It seems these lithologies are an important part of the depositional system, but they are mentioned mostly for having been excluded from consideration. The graphic column, on the other hand, seems to show changes in the proportion of lithologies through time (as well as substantial coring gaps). How does the stratigraphic pattern of different lithologies fit into the provenance/sediment routing interpretation proposed? Petrographic and geochemical examination of the coarser lithologies could provide information about provenance and transport. Do K/Al and Si/Al measures of sedimentologically identified distal turbidites support the assertion that hemipelagic deposition and turbidites can be separated by the metrics used at this site.

Sandstone and sandy mudstones indeed form a substantial part of the sedimentary record, accounting for ~55% of recovered sediment (Figure 2a). These lithologies are interpreted to represent turbidites, dense traction or debris flows, and potentially bed loads, which were deposited in a fan to fan-valley environment (cf. lines 380-381). Detailed information on sandstone petrography, geochemistry, and provenance is provided by Kagami (1978) and Natland (1978). Provenance analyses suggest that sandstones were derived from granitic weathering sources located along the SW African coast (Natland, 1978), indicating a similar weathering source region for sandstones and low-TOC black shales (cf. lines 403 to 410, Figure 3). Long-term changes in relative proportions of the different lithologies have also been discussed by Natland (1978), who proposed that the decreasing abundance of sandstones throughout the Aptian (Figure 2a) was related to the effects of subsidence and erosion of sediment source regions that may have diminished massive coarse-grained turbidites, which characterize the lower part of the black shale interval. Carbon burial in coarse grained sediments is well known to be highly limited and therefore only a secondary focus of our study. At Site 361, TOC contents of sandstones and sandy mudstone beds range from 0.3 – 3% (Erdman and Schorno, 1978) and predominantly comprise terrigenous organic matter, as indicated by Rock-Eval data (Van der Spuy, 2003 and references therein) and the frequent occurrence of wood

debris (The Shipboard Scientific Party, 1978). To avoid any bias related to changes in grain-size and to ensure that turbiditic coarse-grained lithologies were omitted, we carefully screened our sample set using geochemical grain-size proxies (i.e. Si/Al and Zr/Al ratios shown in Figure 2e,f; e.g. Croudace and Rothwell (2015) and references therein). These ratios are consistently below or close to average shale values in all chosen samples (Figure 2e,f), and distinct from interbedded sandstones/siltstones, which have been analyzed for reference (Supplementary Figure 1).

However, Si/Al and Zr/Al ratios probably do not allow to readily discriminate between hemipelagic and fine-grained distal turbidites, as they may share similar grain size characteristics. Previous studies support the existence of two genetically distinct (turbiditic and hemipelagic) types of shale (Kagami, 1978; Natland, 1978). Natland (1978), noted that shales frequently comprise finely laminated, varve-like alternations of nannofossil layers and mudstone/shale (Noël and Melguen, 1978). Scanning electron microscopy revealed similar lamination features containing dissolution imprints of coccoliths also in shales in which calcium carbonate has been removed diagenetically (Noël and Melguen, 1978). These shale intervals are up to several meters thick and have been interpreted to be deposited under tranquil hemipelagic conditions (Arthur and Natland, 1979; Natland, 1978). In contrast, grain-size and bedding thickness analyses led Kagami (1978) to conclude that shales associated with sandstone and sandy mudstones beds were probably "deposited by the same sedimentary process" and presumably represent fine-grained turbiditic material, which settled out of suspension. Based on these results, we omitted shale intervals directly on top of sandstone and sandy mudstone during sampling. We further note that $\delta^{13}C_{org}$ patterns at Site 361 closely track variations in the global ocean-atmosphere reservoir, as discussed in detail in Dummann et al. (2020). This precise reproducibility of $\delta^{13}C$ trends further supports that only hemipelagic shales were sampled.

How was the 5% organic carbon threshold chosen? Are there really two different lithologies (<5% and >5% Corg) within fine-grained portions of the "black shales" or is there a continuum? What is the stratigraphic character of the most TOC-rich intervals– discrete beds (how thick?) or trends over some thickness (cm? dm? m?). One way to illustrate/test this classification would be a plot of K/Al vs. %organic carbon (cf. Fig. 3b). Statistical tests of apparent geochemical patterns would enhance confidence in the relationships proposed.

Alternations of "high-TOC black shales" (TOC >5%) and "low-TOC black shales" (TOC <5%) appear to be unrelated to changes in lithology and bedding characteristics (i.e. high-TOC intervals are not confined to discrete beds), as illustrated in Supplementary Figure 2a. Instead, increases in TOC content occur over several dm- to m-intervals (considering the relatively low sampling resolution) suggesting transient and gradual shifts in carbon burial, favoring a paleoclimatic/paleoceanographic forcing mechanism. As such, transitions from low-TOC to high-TOC black shale deposition may indeed represent a continuum. This is further supported by steady increases in S/Fe and K/Al ratios with TOC content (Supplementary Figure 2b,c), indicating gradual changes in porewater (and bottom water) oxygenation and sediment (and nutrient) influx from the S-African continent, respectively. However, we argue that the relatively low sampling resolution, which is due to large coring gaps and the scattered occurrence of hemipelagic shales (Figure 2a), does not allow to fully resolve such a continuum. Hence, we make a general distinction between two end member states in terms of redox conditions and burial of organic carbon by using the terms "high TOC black shale" and "low TOC black shale" to convey to the reader the principle differences between these two interrelated units. The exact placement of the TOC threshold along this continuum is debatable. We chose a TOC threshold of 5% based on our paleo-redox analysis, as it appears to mark the transition from suboxic to anoxic–euxinic conditions (Figure 5 and Figure 6). We will discuss these limitations and our choice of TOC threshold in the revised manuscript.

Were there efforts to separate lithogenic from hydrogenic contributions to the REE profiles measured? Could the differences in 3c-d between bulk analyses reflect oceanographic/diagenetic conditions correlated with organic carbon content rather than changes in sediment provenance causing changing in oceanographic/depositional conditions? Some of this information is in the EPSL manuscript and I assume some is in the DSDP volumes cited, but this study would be stronger if more supporting sedimentological and stratigraphic information was summarized and synthesized in the manuscript.

The REE and immobile trace element data presented were obtained by total digestion (cf. lines 102–112) and thus reflect trends in whole-rock composition. Although scavenging by organic carbon has been shown to contribute substantial amounts of REE to the sediment (up to 20% of the total sedimentary REE content; Abanda and Hannigan (2006)), this process probably did not contribute significantly to the differences in REE distribution observed in this study (Figure 3). To this end, we note that "high-TOC black shales" show overall lower REE concentrations than "low-TOC black shales", which is inconsistent with an enhanced scavenging of REEs by organic carbon. Furthermore, studies from various modern sedimentary environments indicate that REE distributions in sedimentary organic matter are characterized by strong enrichments of MREEs (Freslon et al., 2014). "High-TOC black shales", however, lack a similar MREE enrichment relative to "low-TOC black shales", further supporting that scavenging of REEs by organic carbon had a negligible impact on the bulk sedimentary REE distribution.

Along similar lines, the presentation of the modelling and modelling results is limited although those results arguably provide the only independent assessment of the model proposed. Description of the model (2.4) is brief and the changes in bathymetry between the Dummann et al. 2020 EPSL paper's and the bathymetry used in this paper is not illustrated. The paleogeographic figures in the EPSL paper are superior to the rather simple Fig. 1 in this manuscript. In figures 9 and 10 it would be nice to see a panel that has the 1200 ppm CO2 results shown rather than simply showing 600 ppm results (9b, 10a) and a difference (9c, 10b).

We agree with the reviewer that more space could be given to the presentation of our modeling results. Hence, we will add several supplementary figures to the revised manuscript (Supplementary Figures 3–6), which illustrate the implemented bathymetry, the model spin-up, and the results of individual modelling runs (600 ppm and 1200 ppm CO_2). Firstly, a detailed map of the implemented regional model bathymetry, along with a comparison to the Dummann et al. (2020) study, will help to illustrate and justify the small bathymetry differences between both studies (Supplementary Figure 3). Secondly, preempting the next reviewer comment, we will also add a time series of simulated upper and deep ocean temperatures, both globally and for the study area during model spin-up (Supplementary Figure 4). These data clearly show that the model reached a quasi-steady state after about 2000 yr during the 3000 yr long model spin-up. Thirdly, in response to the comments made by Alexandre Pohl (reviewer 1), we will add a short reference to a recently published study demonstrating the capability of the KCM to reproduce available Aptian SST proxy data (Steinig et al., 2020). References to these additional figures and studies will be included in section 2.4 of the revised manuscript, which will allow a more in-depth assessment of the model results by the reader. Finally, we agree that some readers might be interested in a more detailed presentation of the modeling results. However, we argue that the current versions of Figure 9 and 10 provide a balance between simplicity and the main results of the modeling. Hence, we prefer to include the current version of Figure 9 and, in response to the comments made by reviewer 1, we will include a modified version of Figure 10 in the main text, which shows precipitation pattern during austral summer (cf. Supplementary Figure 6g,i). Detailed results of the individual simulations (Supplementary Figures 5 and 6) will, however, be provided in the supplement, using the same quantities and contour intervals as in Figures 9 and 10. All supplemental figures will be referenced in section 4.4 of the main text.

The idea that an intermediate water mass isolates the deep Cape Basin promoting organic carbon accumulation is an interesting model-based suggestion. Could cross sections across the basin showing model predicted water properties that indication a saline layer divides the water column be provided as well as sensitivity tests of the persistence and extent of such a layer under the two climatic modes proposed to explain the distribution of TOC in the black shales? Could discussion and supporting graphics demonstrating the models have stabilized be provided especially relative to the deep basin?

We agree that the impact of deep and intermediate water mass circulation on organic carbon burial is an interesting and relevant question. Changes in ocean circulation are, however, difficult to assess based on the available data, which renders it difficult to validate circulation model results. We therefore think that such a discussion is beyond the scope of the present proxy-based study. However, a follow-up modeling study with a distinct focus on the sensitivity of the South Atlantic circulation system towards climatic (i.e. changes pCO_2) and tectonic forcing (i.e. opening of gateways) processes is currently in preparation. The model equilibrium is illustrated in Supplementary Figure 4 as described in our response to the previous comment.

Finally, the manuscript suggests that increased CO2 might be the cause of the proposed changes in precipitation leading to nutrient input and enhance physical weathering proposed to explain organic carbon enrichment associated with OAE1a. The manuscript also suggests orbital variations might be the cause of the other organic carbon rich black shale intervals. Are the authors proposing that orbital variations cause a change in CO2? If so, how? If they instead are suggesting both orbital variations and CO2 increase cause the same climatic changes, this would be an idea that seems worth including in section 4.6. Would the authors hypothesize what orbital configuration would be expected to mimic high CO2 precipitation patterns and why? Such a prediction could be tested when/if an orbital timescale is generated for the Aptian Cape Basin.

In line with previous studies (e.g. Naafs et al., 2016), we invoke an increase in pCO_2 as a primary forcing mechanism to explain changes in continental hydrology and marine organic carbon burial during OAE 1a, while orbital forcing is considered as an additional modulating mechanism, with different mechanisms and impacts on OC production and burial depending on the study region. We do not claim that orbital scale fluctuations directly impact atmospheric pCO_2, however, over longer time scales orbital forcing responds to fluctuations in pCO_2. Wagner et al. (2013) addressed these relationships by presenting a conceptual model that links marine organic carbon burial dynamics to changes in atmospheric circulation and continental hydrology under the descending/ascending limbs of the atmospheric Hadley Cell. They proposed that orbitally driven contraction/expansion of the Hadley Cell caused latitudinal shifts of climate belts, inducing cyclic fluctuations in continental river run-off and oceanic upwelling and thereby variations in marine organic carbon burial. Given the position of Site 361 close to the descending limb of the southern hemisphere Hadley Cell (Figure 9 and 10), we speculate that the episodic occurrence of high-TOC black shales in the Cape Basin may have been related to a recurrent northward migration of the austral westerlies due to contraction of the Hadley Cell. This may have increased humidity and precipitation in the S-African hinterland, augmenting continent-ocean nutrient transfer and ultimately organic carbon burial in the Cape Basin. However, we are currently limited in providing reliable estimates of orbital cyclicity forcing high-TOC black shale formation due to the overall low stratigraphic coverage at Site 361 (Fig. 2a).

55- does the mid-ocean ridge represent a barrier to circulation?

The role of the mid-ocean ridge for bottom water circulation (i.e. >1000 m water depth) is currently unclear, as no Early Cretaceous sediment records are available from the deepest part of the Argentine Basin west of the ridge. However, it probably did not represent a barrier to intermediate water mass circulation (i.e. <1000 m water depth), as detailed in Dummann et al. (2020).

57- how is global excess OC burial defined/calculated

McAnena et al. (2013) used biogeochemical modeling to calculate the amount of excess organic carbon buried that is required to explain a positive $\delta^{13}C$ excursion during the Late Aptian, which was associated with a cooling event (the Late Aptian Cold Snap; LACS). The modeling results suggest that an excess of ~812,000 gigatons of carbon may have been buried over a time span of 2.5 million years, about 16% of which may have buried in the emerging South Atlantic. Technical details on the modeling approach can be found in McAnena et al. (2013).

59- Angola Basin not shown in Figure

We will add the names of the different basins to Figure 1.

63- which data- TOC percentages?

The study of Behrooz et al. (2018) is primarily based on biomarker data.

87- beyond TOC (and color?) are there sedimentological differences among lithostratigraphic divisions, gray shales and low-TOC black shales do not seem different based on the criteria used
95- describe sample set

Our sampling strategy and choice of lithostratigraphic units is outlined above and will be detailed in section 2.1 (cf. line 80) of our revised manuscript.

126, 235- don't start sentence with lowercase letter, write around

We will re-phrase these sentences accordingly.

144- not sure this is a method, odd to exclude sandstones for provenance work

As mentioned above, detailed information on the provenance of sandstones can be found in the literature (e.g. Natland, 1978).

149- These data

Will be amended as suggested.

177- very different precision suggested for characteristic AS Si/Al and Zr/Al ratios

Average shale values are taken from the literature (Wedepohl, 2004, 1971). The different precisions reported for major (Si) and minor/trace elements (Zr) most likely depend on the analytical method chosen by these authors.

180- difference seems small given variance- is it statistically supported. What does a plot of TOC vs. K/Al look like?

See Supplementary Figure 1b.

254- is proposed difference statistically supported?

To evaluate if the differences in the fractional abundances of desmethylsteranes are statistically significant, we performed a t-test (p=0.05). The results, summarized in Supplementary Table 1, indicate that high-TOC black shales contain significantly higher mean abundances of C_{27}- and C_{28}-desmethylsteranes, reflecting inputs from marine sources (Huang and Meinschein, 1979), and lower

abundances of C_{29}-desmethylsteranes, commonly attributed to higher land plant inputs (Huang and Meinschein, 1979), compared to low-TOC black shales.

Figure 2- using red shading for organic carbon rich black shales and orange shading for red beds seems like an odd choice; panels d-f are labeled grain size/mineralogy, but the plots show elemental ratios.

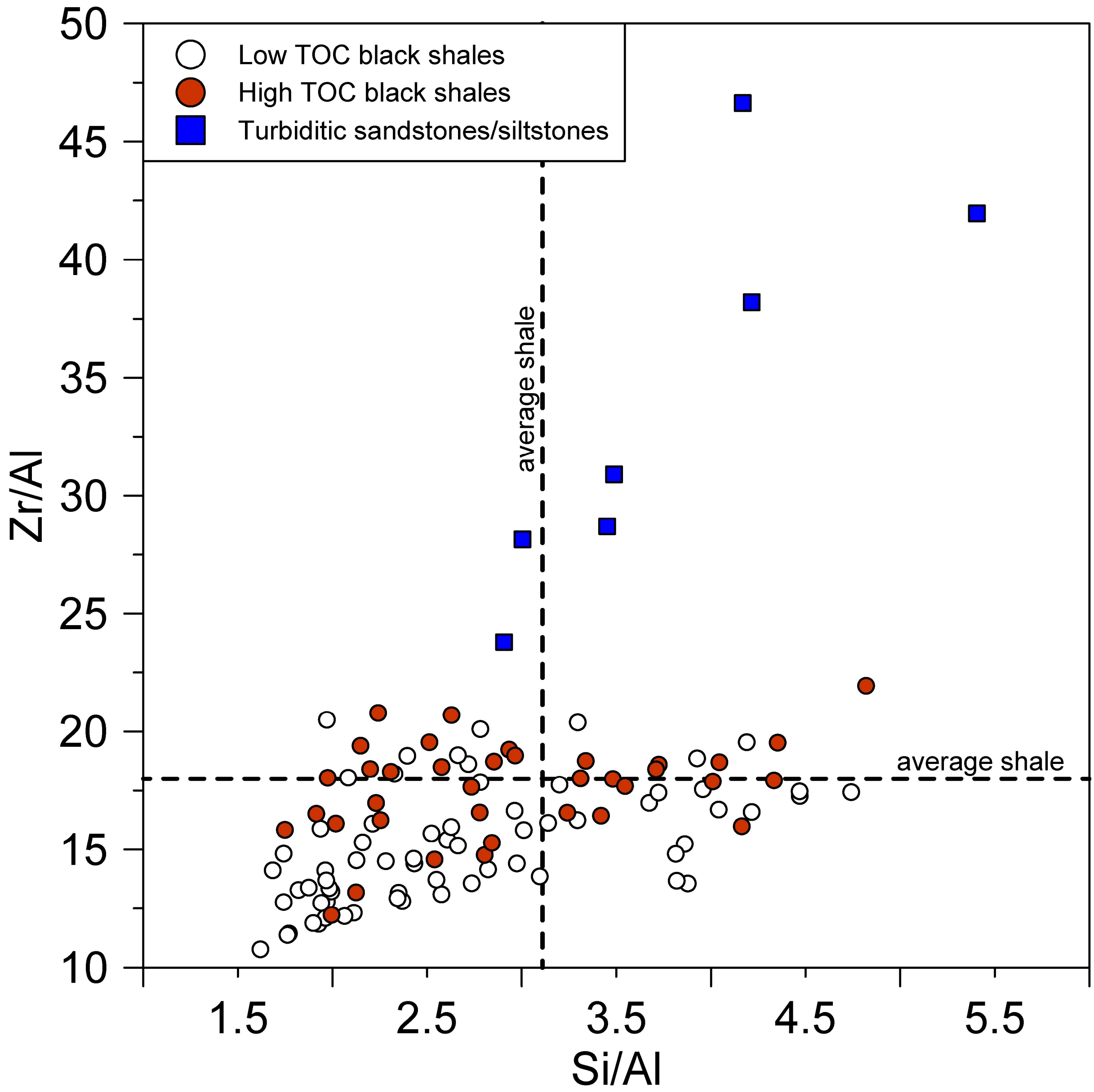
We will change the color shading for "high-TOC black shales". The labels in the different panels of Figure 2d–f intend to clarify our interpretation of the individual geochemical proxies (i.e. Zr/Al and Si/Al are used as proxies for grain-size, K/Al is used as proxy for changes in clay mineralogy).

Supplementary information could include some description of the plots.

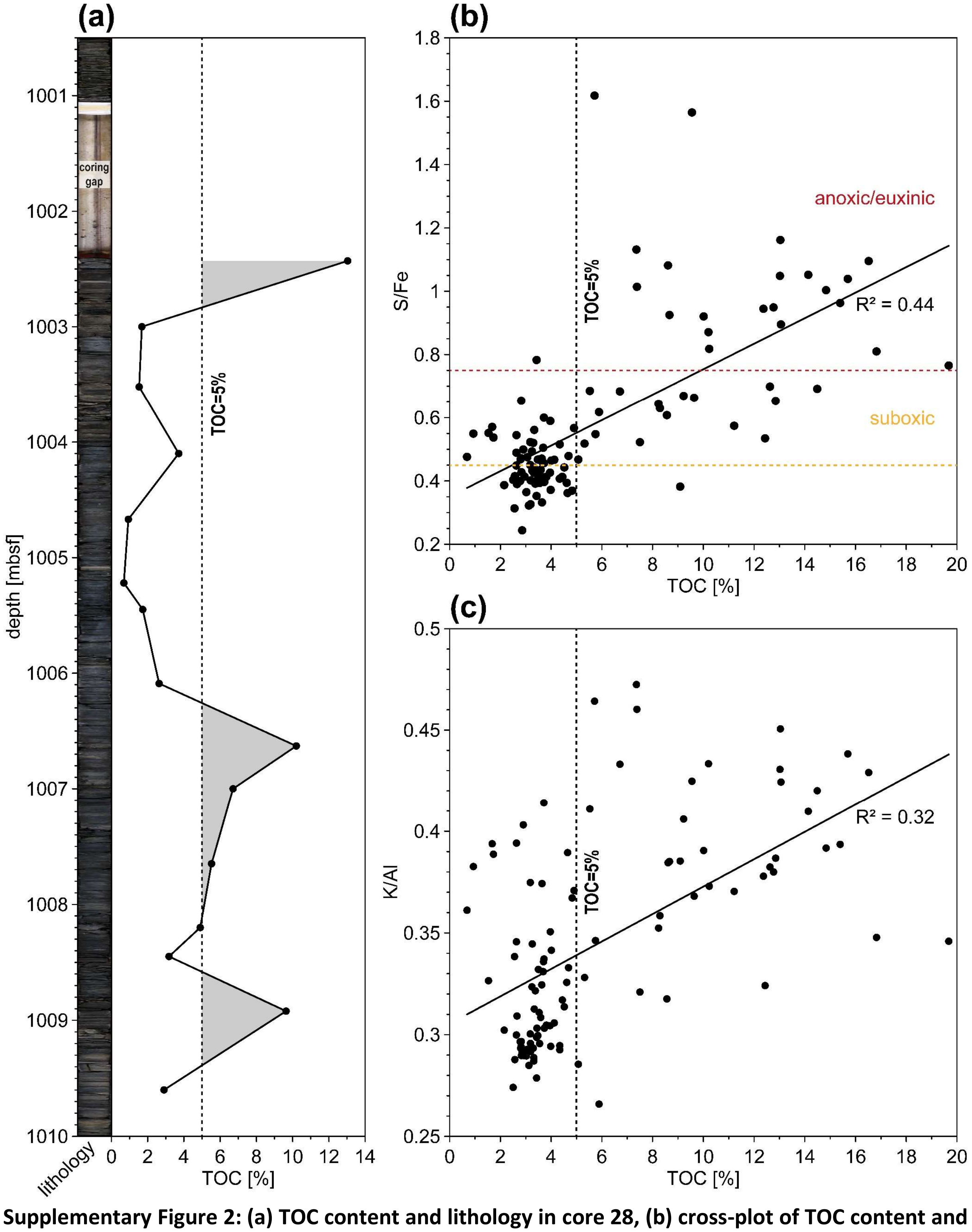
Descriptions will be added alongside supplementary information on our modeling approach.

Supplementary Table 1: T-test results (p=0.05) for the fractional abundances of C_{27}–C_{29}-desmethylsteranes (LTBS=low-TOC black shales, HTBS=high-TOC black shales)

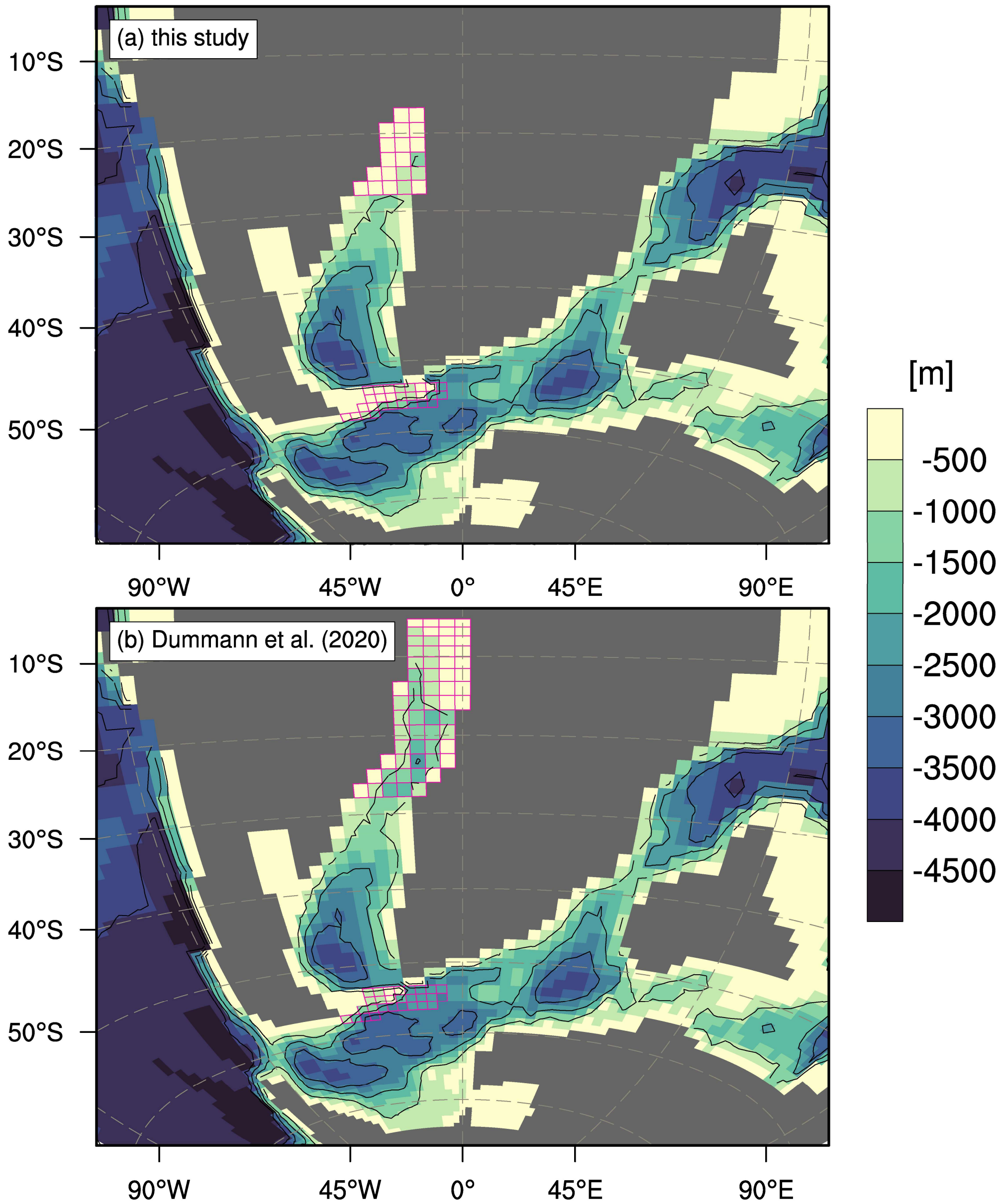
	C_{27}-desmethylsteranes [%]	C_{28}-desmethylsteranes [%]	C_{29}-desmethylsteranes [%]
$\bar{x}$ LTBS	25.4	33.5	41.1
$\bar{x}$ HTBS	28.6	39.1	32.3
σ^2 LTBS	11.9	26.2	44.5
σ^2 HTBS	22.2	30.2	32.1
Hypothetical difference of means	0	0	0
T-statistics	-3.1	-4.1	5.8
Critical t-value (one-tailed t-test)	1.7	1.7	1.7
Critical t-value (two-tailed t-test)	2.0	2.0	2.0



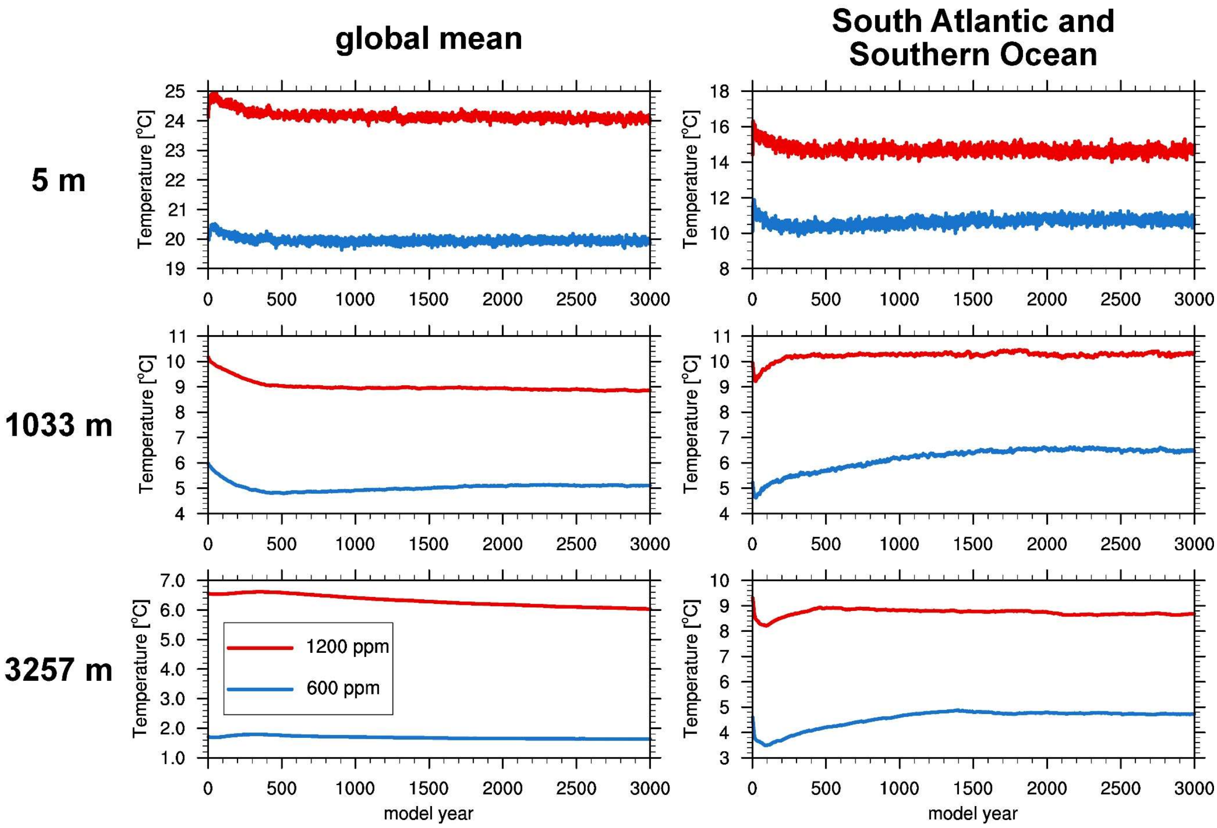
Supplementary Figure 1: Cross-plot showing Si/Al and Zr/Al ratios for high-TOC black shales (TOC>5%), low-TOC black shales (TOC<5%), and interbedded sand- and siltstones.



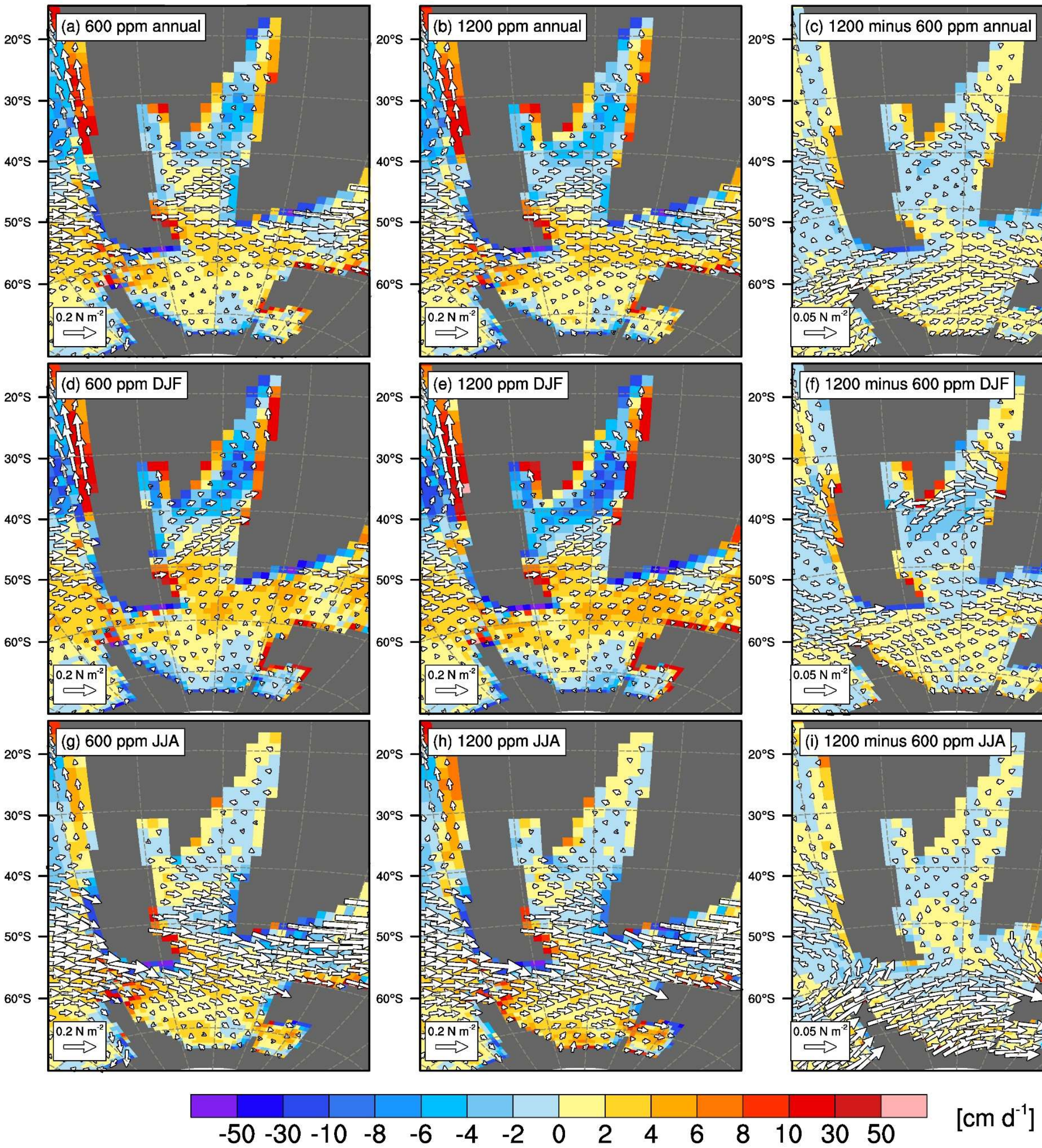
Supplementary Figure 2: (a) TOC content and lithology in core 28, (b) cross-plot of TOC content and S/Fe ratios, and (c) cross-plot of TOC content and K/Al ratios containing all (black shale) samples from cores 28–48.



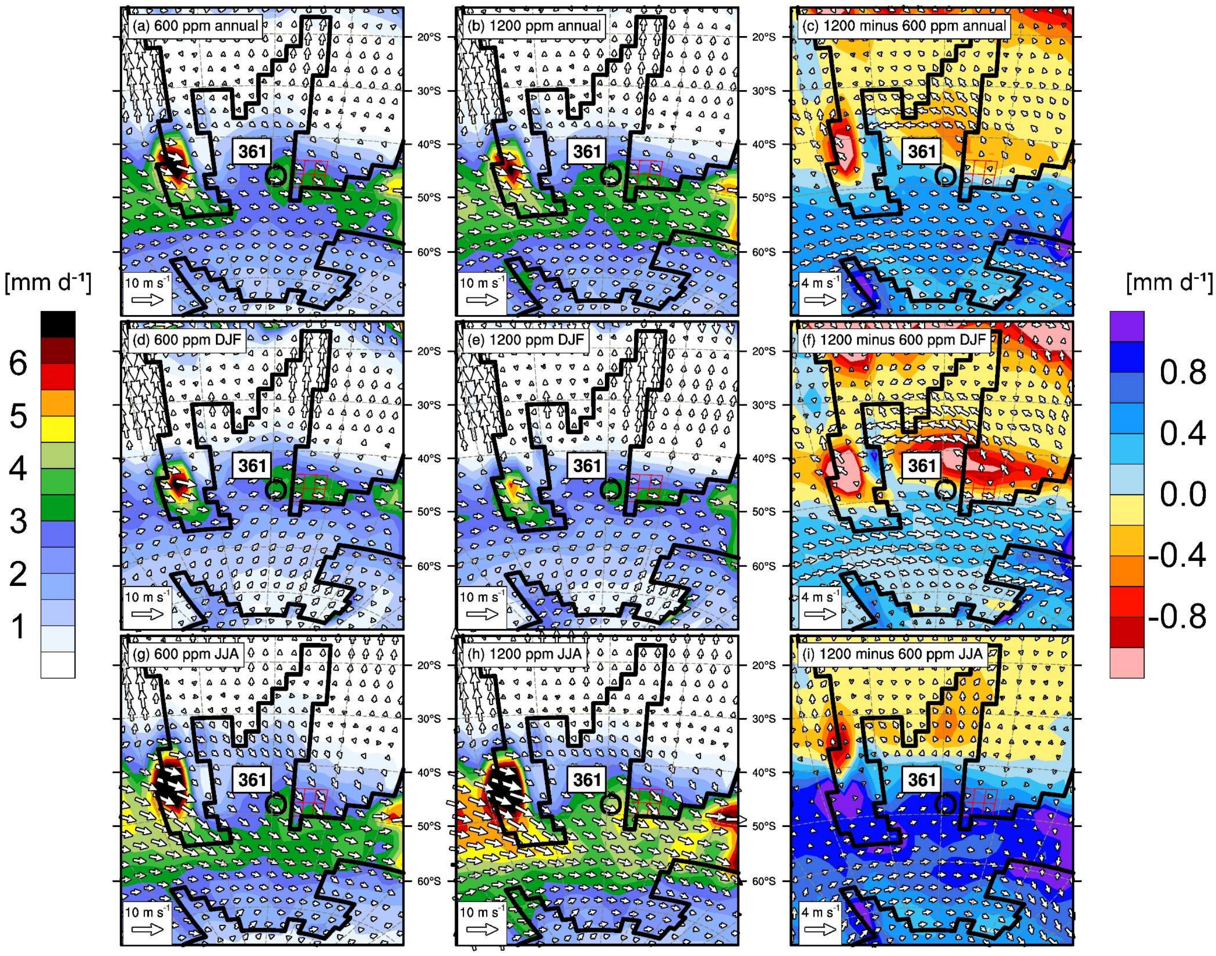
Supplementary Figure 3: Regional ocean model bathymetry used in (a) this study and (b) in the Late Aptian/Early Albian simulations of Dummann et al. (2020). Changed grid points are highlighted in pink. Both studies use the same Kiel Climate Model. To account for the slightly older Aptian time interval investigated in this study, the northward extent of the South Atlantic (b) was reduced and replaced by the early Aptian boundary conditions from Sewall et al. (2007) northward of 30°S (a). Furthermore, the Falkland Plateau (~55°S) is moved to a more eastward position to limit intermediate water exchange between the South Atlantic and Southern Ocean during the Early Aptian, consistent with the results of Dummann et al. (2020).



Supplementary Figure 4: Time series of simulated annual mean ocean temperatures averaged globally (left-hand panels) and over the South Atlantic and Southern Ocean study region (right-hand panels; 0°–90°S, 40°W–20°E) shown at the 5 m, 1033 m and 3257 m model levels, respectively. Simulations were initialized with the final state of the 6,000 year long integrations described in Steinig et al. (2020) using the same model with a slightly different paleobathymetry (Supplementary Figure 3).



Supplementary Figure 5: Surface wind stress and ocean vertical velocity at 10 m water depth simulated by the KCM at 600 ppm CO_2 (left-hand side panels), 1200 ppm CO_2 (panels in the center), and their respective differences (right-hand side panels). Results are averaged across the whole year (top panels), the months December to February (middle panels) and the months June to August (bottom panels). Positive values indicate upwelling.



Supplementary Figure 6: Surface wind and total precipitation simulated by the KCM at 600 ppm CO_2 (left-hand side panels), 1200 ppm CO_2 (panels in the center) and their respective differences (right-hand side panels). Results are averaged over the whole year (top panels), the months December to February (middle panels) and the months June to August (bottom panels).

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