Author’s response to referee comment (RC) 1

We would like to thank referee #1 (Alexandre Pohl) for his time to review our manuscript and for raising important questions that will help to improve the overall quality of our manuscript. We will address the main concerns below in a detailed point-to-point response. New Supplementary Figures 1-3 are referenced in this response and are included at the end of this document.

Paper summary:

Dummann et al. investigated the mechanisms driving organic carbon burial during the Aptian–Albian in the deep Cape Basin at Deep Sea Drilling Project (DSDP) Site 361. To that purpose, the authors produced abundant (bulk and molecular) geochemical data and conducted general circulation model runs using the ocean-atmosphere Kiel Climate Model. Dummann et al. identified two main temporal trends based on the sedimentary record: (1) a long-term trend toward more oxygenated conditions, as can be seen based on the changes in sedimentary facies from black shales to grey shales and red beds, and (2) a higher-frequency change, inside the Aptian black shale unit, between low-TOC black shales and high-TOC black shales (the latter comprising OAE1a, but also other layers). The authors explain the reported long-term change in deep-water redox conditions at DSDP Site 361 by a reorganization of the ocean circulation in response to the evolution of marine gateways, based on the findings of their previous study (Dummann et al., 2020). Regarding the high-frequency cycles, proxy data demonstrate a common geochemical signature of the high-TOC black shales, suggesting that the same mechanisms potentially drove the deposition of the successive layers. These data point toward an increasing contribution of marine organic matter during the deposition of these high-TOC shales. The systematic increase in the K/Al ratio during these periods suggests that the latter may correspond to periods of increased precipitation and runoff from the continent, bringing unweathered sediments and also nutrients to the ocean, thus explaining the increase in marine primary productivity and organic matter deposition under anoxic–euxinic conditions. In order to check this hypothesis, the authors conducted 2 climatic simulations at 600 ppm and 1200 ppm CO2, supposedly representative of Aptian background climate and OAE1a. Although total precipitation over the region of interest does not significantly increase in response to climate warming in the model, spatial contrasts strengthen and runoff during the austral winter doubles. From this point of view, simulations support the authors’ hypothesis.

This summary is mostly correct, but we want to highlight that our manuscript actually covers three different driving mechanisms of organic carbon burial in the Cape Basin. We want to briefly reiterate these processes to avoid any potential misunderstanding and to facilitate our response to the remaining reviewer comments:

1. **Long-term (>1 Myr) tectonic changes gradually increased the South Atlantic-Southern Ocean deep water mass exchange, which in turn enhanced bottom water oxygenation and reduced organic carbon burial in the deep Cape Basin.**
2. **Higher atmospheric pCO_2 during OAE 1a enhanced seasonal precipitation and surface runoff from the African continent. The associated increase in weathering-derived nutrient input into the South Atlantic temporarily promoted local organic carbon burial. This data-derived hypothesis is supported by our two high and low pCO_2 modelling experiments.**
3. **In contrast to OAE 1a, we have no evidence that other recurring carbon burial maxima during the Aptian were linked to global pCO_2 changes. We rather propose that these short-term fluctuations were caused by orbitally driven changes in the seasonal inland precipitation over South Africa that ultimately controlled riverine nutrient input and productivity in the Cape Basin.**

General comment:

As a paleoclimate modeler interested in proxy data but not expert in geochemistry, I really appreciated the manuscript. Geochemical analyses and results are exposed in a very clear and accessible way. The authors further use a climate model to validate the hypotheses established based on the analysis of the geological record. The manuscript is well organized and the statements are mostly very well supported by analyses/figures of high quality. I recommend rapid publication of this study after minor revisions.

We thank the reviewer for the positive feedback.

<u>Major comments:</u>

1. Model-data comparison and the change in sediment supply between low-TOC and high-TOC shales. On lines 414–416, the authors "*propose that the shift in provenance from low- to high-TOC black shales reflects enhanced moisture supply to the continent's interior, augmenting river run-off in the upstream regions of the paleo-Karoo River and/or increasing chemical weathering intensity of basaltic rocks*". However, results of the climate model suggest that coastal S and SW coastal regions became more humid during OAE1a-like warming events, while the continent interior became more arid. These two elements – the hypothesis derived from proxy data and the results of the GCM – thus seem in contradiction. This discrepancy should be resolved.

We argue that there is no discrepancy between the proxy-derived hypothesis and the modeling results but agree that the current presentation of the precipitation maps in Figure 10 can be improved. While the current annual mean map in Figure 10b indeed shows a more arid continental interior, our results reveal that it is actually the enhanced <u>seasonality</u> of precipitation over South Africa in the high CO_2 simulation (Figure 10c) that drives the overall increase in surface runoff (Figure 10d). Higher rainfall during austral winter (June to August) disproportionately increases surface run-off due to reduced evaporation and high soil moisture saturation. To better illustrate these seasonal fluctuations, we therefore decided to change the maps in Figure 10a-b and rather show averages for the months June to August instead of annual mean fields. The updated figure is attached as Supplementary Figure 1 and clearly shows that the increase in austral winter precipitation in response to a CO_2 doubling occurred over large areas in the South African hinterland. We conclude that these modeling results are in overall agreement with the data-derived hypothesis of enhanced river run-off during OAE 1a and should now be clearer to the reader. We will further emphasize the importance of the seasonal precipitation cycle for the surface run-off in the main text. Additionally, in response to a comment by reviewer #2, we will add seasonal and annual mean maps for both simulations to Supplementary Figure 2, to allow a more in-depth assessment of the modelling results.

2. The paleo-Karoo River. The existence of this river has a major role in the hypotheses drawn in this contribution. Are the arguments supporting (1) the existence of the river and (2) the location of the river mouth during the Aptian (especially lower Aptian) robust? Topography, in particular, is generally very difficult to reconstruct in deep-time periods, thus making the reconstruction of watersheds challenging, too. I suggest expanding the arguments supporting the existence of this river, of its watershed such as drawn in Fig. 3 and of the location of the river mouth. If arguments are solid, such discussion will strengthen the manuscript. If they are not, I still believe that the authors' arguments are very interesting but I think it would be good in that case to highlight these uncertainties as a limitation.

The evolution of the geomorphology and paleo-drainage systems in SW Africa has been studied by numerous authors, who generally acknowledge that two, SW–W-flowing river systems drained most of the interior of southern Africa since the Cretaceous (e.g. De Wit, 1999; Dingle and Hendry, 1984; Partridge and Maud, 1987). Recent apatite fission track data identified a paleo-river valley, dated at 120–110 Ma, which coincides spatially with the present course of the Krom/Sout River that enters

the S-Atlantic via the Olifants River near 32° S today (Kounov et al., 2008), roughly 450 km NE of the coring location of Site 361. Based on these results, Kounov et al. (2008) proposed that this deeply incised paleo-valley marks the river course of the paleo-Karoo River, which may have provided "a major outlet to the Atlantic Ocean for its inland drainage system". This interpretation is consistent with onshore geological and morphological evidence (De Wit, 1999). However, we agree that the exact routing of S-African river systems and associated catchment topography during the Early Aptian remains speculative, in particular in the widely eroded upstream regions. To this end, we note that our model results indicate an increase in seasonal precipitation over the entire southern tip of Africa (Supplementary Figure 1), which would cover the assumed catchment area of the paleo-Karoo River.

3. GCM simulation and OAE1a. The authors suggest that a pCO2 doubling from 600 ppm to 1200 ppm is well representative of the climate change that accompanied OAE1a. The choice of the pCO2 values is based on the argument that these values fall "*within the lower range of pCO2 estimates for OAE 1a (Naafs et al., 2016)"* (also lines 160–163). However, different climate models have different climatic sensitivities, i.e., a given change in pCO2 will not have the same effect depending on the climate model in use. Is the climate change simulated in the Kiel model between 600 and 1200 pm really representative of what we know of OAE1a? Interestingly, recent temperature estimates (Naafs and Pancost, 2016; O'Brien et al., 2017) provide an overview of the ocean warming that accompanied OAE1a. I think it would be instructive to provide a rapid comparison between the modeled temperatures and these recent proxy data. Such comparison would satisfactorily complement the interesting comparison of the modeled climatic belts with the lithological indicators of climate already proposed by the authors.

We agree that a model-data comparison of OAE 1a temperatures would be helpful for the reader to allow an independent validation of the model set-up and results. In fact, we recently published an extensive comparison of our simulated upper ocean temperatures and available proxy records (Steinig et al., 2020), including the data from Naafs and Pancost (2016) and O'Brien et al. (2017). Steinig et al. (2020) used the same model set-up, but with a slightly different South Atlantic and Southern Ocean bathymetry compared to our current study. While the relative temperature increase during OAE 1a is not well constrained due to stratigraphic limitations, we were able to show that absolute OAE 1a proxy temperatures are broadly consistent with our model results at CO_2 levels between 1200 to 2400 ppm. This model-data congruence can be achieved by assuming a regional warm bias in the Early Cretaceous TEX_{86} record in the young Atlantic Ocean for which we presented evidence in Steinig et al. (2020). High-resolution atmospheric CO_2 reconstructions estimate average levels across OAE 1a of around 1550 ppm (Naafs et al., 2016). We are therefore confident that our modelling approach does reasonably well represent an OAE 1a-like climate state at a CO_2 level of 1200 ppm, while the 600 ppm simulation rather reflects the lower Early Cretaceous background CO_2 concentrations (Jing and Bainian, 2018). We will add the Steinig et al. (2020) reference and the major results of this study to section 4.4 of the main text to validate our OAE 1a simulation and to provide a broader context on pCO_2-SST relationships.

Minor comments:

1. Lines 441–442: the authors state that "*The climate zonation generated by our model indicates that Site 361 and the southern African continent were located in a temperate humid climate belt*". How was this climatic belt identified? Was is through some Köppen classification or similar? Please support this statement.

We did not apply any quantitative climate zone classification. Instead, based on our modelling results, we located the region of the southern hemisphere westerlies with relatively high precipitation rates compared to the more arid regions in the northern part of the South Atlantic to

define the zone of principle change, both in precipitation and zonal atmospheric circulation. We will re-phrase this accordingly in the revised manuscript.

2. Lines 444–446: I suggest including a reference to the modeling study of Chaboureau et al. (2012), which specifically investigated the controlling mechanisms for the deposition of evaporites in the northern and central sections of the nascent South Atlantic Ocean.

We will reference this closely related study in the revised manuscript. It is important to note that Chaboureau et al. (2012) show a very similar shift towards a positive precipitation minus evaporation balance around the southern tip of Africa.

3. Lines 464–466: I suggest comparing the precipitation simulated at 600 ppm with the precipitation simulated in modern S-Africa in the present-day model control run. This might be more instructive (consistent) than comparing to an independent dataset.

This is a very helpful suggestion that we will implement in the revised manuscript. The simulated present-day simulation in South Africa amounts to ~2.0 mm/day, which is higher than the observed value of ~1.3 mm/day. The offset is comparable to the CMIP5 multi-model ensemble mean wet bias in this region of ~0.5 - 1 mm/day (Eyring et al., 2016).

4. Lines 496: "*no substantial ocean upwelling occurred along the SW African margin during the Aptian*". Here as well, I suggest approaching the question of the limitations. Indeed, the authors are here reaching the limit of the modeling setup. Coastal upwellings s.s. are 10s of km wide, not 100s. As a consequence, they cannot be captured by the ocean model, the mean horizontal resolution of which is ca. 200 km.

We fully agree that the model cannot reproduce small-scale local coastal upwelling. However, our results indicate that, unlike today, no large upwelling system (i.e. on the scale of the upwelling system along the west coast of South America; Figure 9a) existed along the SW African coast during the Aptian, as far as our modelling can resolve it. We will acknowledge these limitations and sub-grid-scale upwelling in the revised manuscript.

5. Figs. 9–10: Since the position of Site 361 is critical, please state how its paleocoordinates were calculated and how the point was subsequently located on the mixed Blakey-Sewall map.

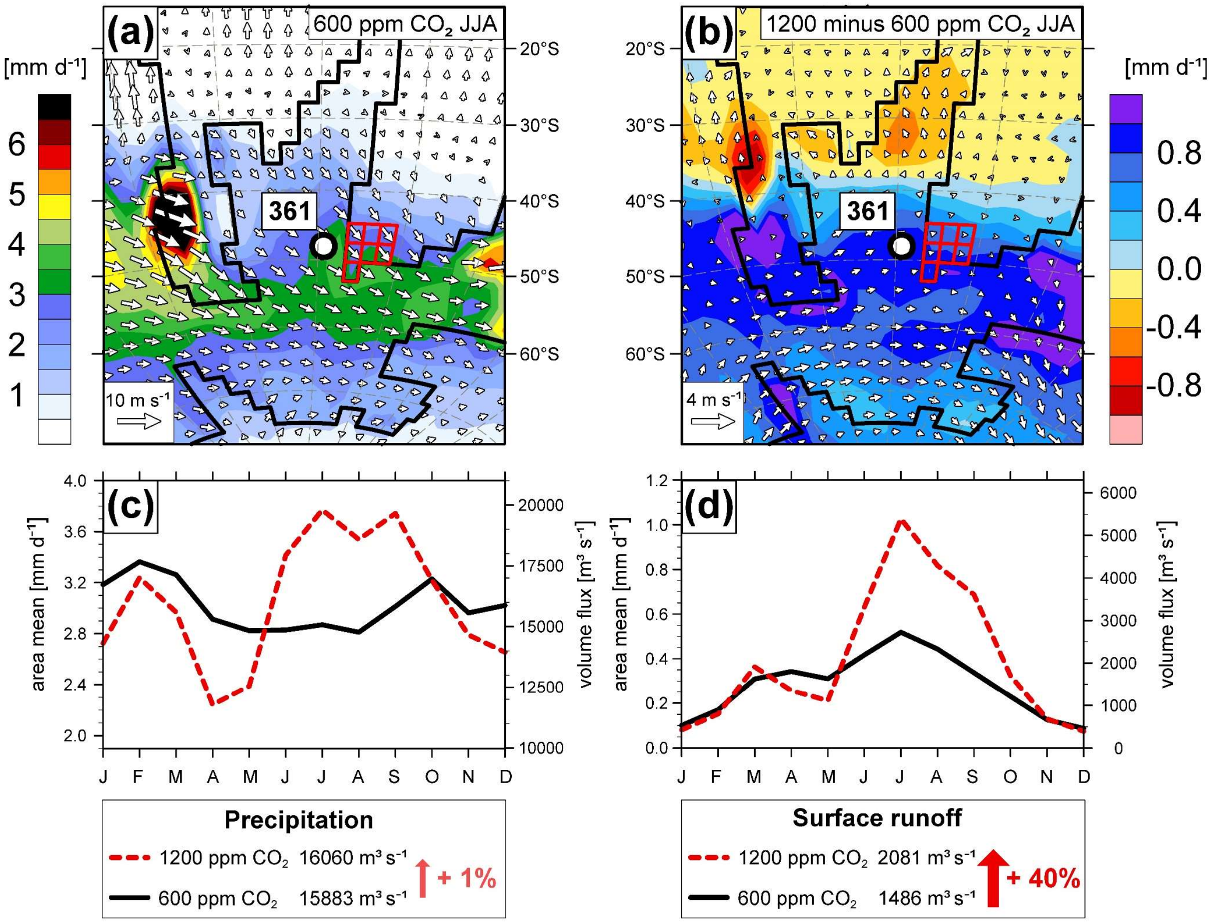
The paleo-coordinates for Site 361 shown in Figure 1 were quantitatively reconstructed using GPlates (Boyden et al., 2011) and the rotational poles published by Matthews et al. (2016). In our model, this paleo-location was qualitatively adapted relative to the model bathymetry. We located the site at the eastern slope of the Cape Basin just off the southern tip of the African continent, consistent with its position on the lower continental rise of South Africa (The Shipboard Scientific Party, 1978). In response to a comment raised by reviewer #2, we added Supplementary Figure 3 showing a detailed map of the implemented model bathymetry. We think this results in a reasonable, qualitative approximation of the 361 paleo-position, especially as we do not discuss any single grid point results for the ocean model.

<u>Technical comments:</u>

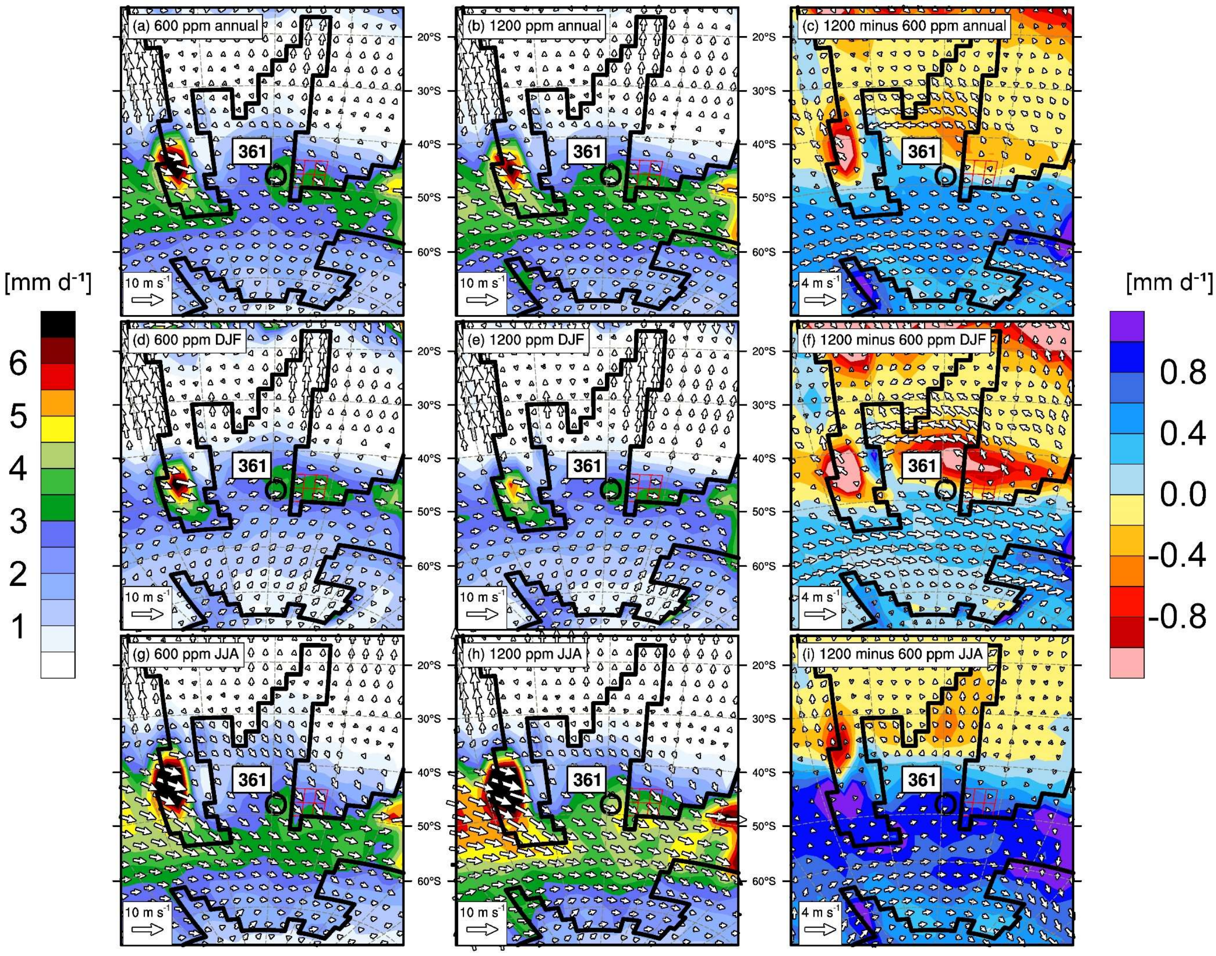
- Line 129: "*The distribution of acyclic hydrocarbons (i.e. n-alkanes, acyclic isoprenoids) were was analyzed*"
- Line 207: "*Distribution of the redox-sensitive trace metals (TMs) Ni, Cu, V, and Zn in black shales are is presented as*"
- Line 417: "*furthermore*" is misleading because the new idea is not used to support the previous paragraph.

• Fig. 2 caption: Missing general title for the Fig.
• Fig. 9 caption: “Positive contours” > positives values (there are no contours).

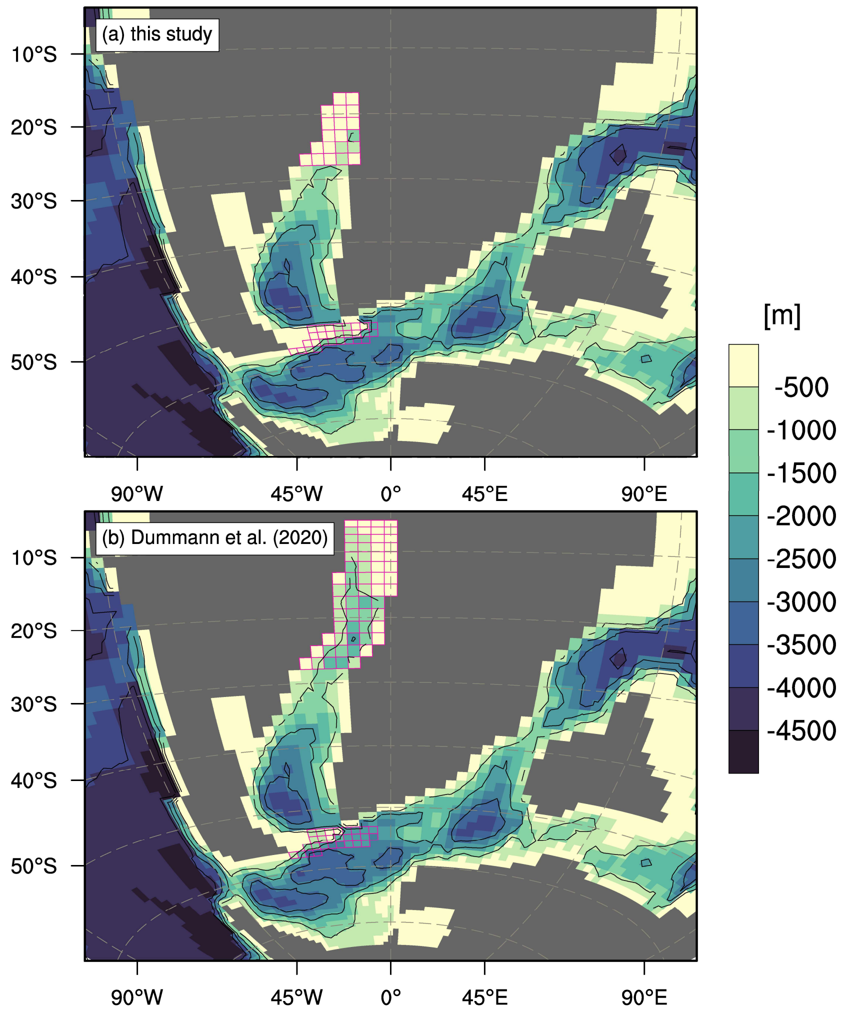
We thank reviewer #2 for his technical comments and will include all suggested changes in the revised manuscript.



Supplementary Figure 1: (a) Simulated June to August surface wind and total precipitation in the study area at 600 ppm pCO_2, (b) relative June to August surface wind and precipitation change due to a doubling of atmospheric pCO_2. (c) Monthly mean precipitation and (d) generated surface runoff averaged over the catchment area indicated by the highlighted grid points in (a) and (b). Volume fluxes are integrated over the whole catchment area (~450.000 km2) with annual mean values reported in the respective legends. This figure is similar to Figure 10 of the main text, but the annual mean fields in (a) and (b) are replaced by the respective June to August maps.



Supplementary Figure 2: 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).



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 in (a) was reduced and replaced by the early Aptian boundary conditions from Sewall et al. (2007) northward of ~30°S. 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).

References cited:

Boyden, J. A., Müller, R. D., Gurnis, M., Torsvik, T. H., Clark, J. A., Turner, M., Ivey-Law, H., Watson, R. J., and Cannon, J. S.: Next-generation plate-tectonic reconstructions using GPlates, in: Geoinformatics: Cyberinfrastructure for the Solid Earth Sciences, edited by: Keller, G. R., and Baru, C., Cambridge University Press, Cambridge, 95-113, 2011.

Chaboureau, A.-C., Donnadieu, Y., Sepulchre, P., Robin, C., Guillocheau, F., and Rohais, S.: The Aptian evaporites of the South Atlantic: a climatic paradox?, Clim Past, 9, 1047-1058, https://doi:10.5194/cp-8-1047-2012, 2012.

De Wit, M.: Post-Gondwana drainage and the development of diamond placers in western South Africa, Economic Geology, 94, 721-740, https://doi.org/10.2113/gsecongeo.94.5.721, 1999.

Dingle, R. V., and Hendry, Q. B.: Late Mesozoic and Tertiary sediment supply to the eastern Cape Basin (SE Atlantic) and palaeo-drainage systems in southwestern Africa, Marine Geology, 56, 13-26, 1984.

Dummann, W., Steinig, S., Hofmann, P., Flögel, S., Osborne, A. H., Frank, M., Herrle, J. O., Bretschneider, L., Sheward, R. M., and Wagner, T.: The impact of Early Cretaceous gateway evolution on ocean circulation and organic carbon burial in the emerging South Atlantic and Southern Ocean basins, Earth Planet Sc Lett, 530, 115890, https://doi.org/10.1016/j.epsl.2019.115890, 2020.

Eyring, V., Righi, M., Lauer, A., Evaldsson, M., Wenzel, S., Jones, C., Anav, A., Andrews, O., Cionni, I., and Davin, E. L.: ESMValTool (v1. 0)–a community diagnostic and performance metrics tool for routine evaluation of Earth system models in CMIP, Geoscientific Model Development, 9, 1747-1802, https://doi:10.5194/gmd-9-1747-2016, 2016.

Kounov, A., Viola, G., De Wit, M., and Andreoli, M.: A Mid Cretaceous paleo-Karoo River valley across the Knersvlakte plain (northwestern coast of South Africa): Evidence from apatite fission-track analysis, South African Journal of Geology, 111, 409-420, https://doi.org/10.2113/gssajg.111.4.409, 2008.

Matthews, K. J., Maloney, K. T., Zahirovic, S., Williams, S. E., Seton, M., and Müller, R. D.: Global plate boundary evolution and kinematics since the late Paleozoic, Global and Planetary Change, 146, 226-250, https://doi.org/10.1016/j.gloplacha.2016.10.002, 2016.

Naafs, B. D. A., Castro, J. M., De Gea, G. A., Quijano, M. L., Schmidt, D. N., and Pancost, R. D.: Gradual and sustained carbon dioxide release during Aptian Oceanic Anoxic Event 1a, Nature Geoscience, 9, 135-139, https://doi.org/10.1038/ngeo2627, 2016.

Naafs, B. D. A., and Pancost, R. D.: Sea-surface temperature evolution across Aptian Oceanic Anoxic Event 1a, Geology, 44, 959-962, https://doi.org/10.1130/g38575.1, 2016.

O'Brien, C. L., Robinson, S. A., Pancost, R. D., Sinninghe Damsté, J. S., Schouten, S., Lunt, D. J., Alsenz, H., Bornemann, A., Bottini, C., and Brassell, S. C.: Cretaceous sea-surface temperature evolution: Constraints from TEX86 and planktonic foraminiferal oxygen isotopes, Earth-Sci Rev, 172, 224-247, https://doi.org/10.1130/abs/2018am-323378, 2017.

Partridge, T. C., and Maud, R. R.: Geomorphic evolution of southern Africa since the Mesozoic, South African Journal of Geology, 90, 179-208, 1987.

Sewall, J. O., van de Wal, R. S. W., van der Zwan, K., van Oosterhout, C., Dijkstra, H. A., and Scotese, C. R.: Climate model boundary conditions for four Cretaceous time slices, Clim Past, 3, 647-657, https://doi.org/10.5194/cpd-3-791-2007, 2007.

Steinig, S., Dummann, W., Park, W., Latif, M., Kusch, S., Hofmann, P., and Flögel, S.: Evidence for a regional warm bias in the Early Cretaceous TEX_{86} record, Earth Planet Sc Lett, 539, 116184, https://doi.org/10.1016/j.epsl.2020.116184, 2020.

The Shipboard Scientific Party: Cape Basin continental rise; Sites 360 and 361, in: Initial Reports of the Deep Sea Drilling Project, Volume 40, edited by: Bolli, H. M., and Ryan, W. B. F., U.S. Government Printing Office, Washington DC (USA), 29-182, https://doi.org/10.2973/dsdp.proc.40.102.1978, 1978.