Dear Dr. Fischer,

We want to thank you for your handling of our manuscript, and thank the reviewers for their comments. We have made the requested revisions and include a detailed response below. We include the responses for both reviewers in this document. Thank you for consideration of our manuscript.

Sincerely,

Maxence Guillermic and Aradhna Tripati on behalf of the co-authors

Response reviewer 1 starts page 2

Response reviewer 2 starts page 13
Reviewer 1

Guillermic et al. present boron isotope data from the Western Equatorial Pacific (WEP) at a variety of resolutions across the Neogene. Whilst new boron isotope data from this region is welcome, their results (and interpretations) are not consistent with major findings from many other studies, a point which is not expressed well in the submitted manuscript.

Response 1: We have added comparisons to additional datasets, including Sosdian et al., 2018 and Rae et al., 2021, as seen in Figure 6, which explores the use of different models for the second carbonate system parameter in seawater. This comparison shows that results are broadly consistent between these studies. In addition, we do a direct comparison with data for Site 872 as calculated in both studies, and then recalculated here, in Figure 7, and see that both pH and pCO2 are in strong agreement.

I find that the paper needs some significant reworking, as the message is not clear, it is a big topic to cover and many of the appropriate references are in, but not necessarily called out at the appropriate place.

My biggest concern is with the interpretation of the record. In many places there are large offsets between the WEP data and other published data. I do not suggest that this invalidates the data themselves, but I think a more in depth explanation is often required. The reason given is commonly air-sea offsets, but this cannot be relied upon for >150ppm changes in the mid Pliocene when all the sites are supposedly ‘in equilibrium’. Many of the sites used in the previous compilation of Sosdian et al 2018 are close by (particularly ODP 872, which agrees in absolute terms with ODP 871 in the Indian Ocean).

The comparison to ODP site 999 for the Plio-Pleistocene needs some more careful thought, as I do not follow the discussion on changes to air-sea equilibrium there.

Response 2: We agree that the variability in the pCO2 mostly comes from the assumptions made for the reconstructions in each of the studies. Therefore, we now explore systematically the impact of choice of carbonate system parameter, and methods used for calculations, for our data and those of Sosdian et al., 2018 and Rae et al., 2021, in Figures 6 and 7, and other studies. Figure 6 explores the use of different models for the second carbonate system parameter in seawater. This comparison shows that results are broadly consistent.

We do a direct comparison with data for western equatorial Pacific ODP Site 872 as calculated in both studies, and then recalculated here, in Figure 7, and see that δ11B, pH, and pCO2 are in strong agreement, that strengthens our confidence in the use of these sites.

We agree with the comment that ODP Site 806 should be more influenced by the constriction of the Indonesian through-flow but our reconstruction is consistent with Site 761 for the Miocene which suggests that the constriction does not support main changes in air-sea equilibrium. We have added this to the text in line 492: “The similarity between our reconstructed values and those published for ODP Site 871 in the Indian Ocean (Sosdian et al., 2018) suggests that changes in Indonesian through-flow do not induce substantial changes in air-sea exchange in the WEP.”.

Diagenesis has the potential to affect these records and this is not discussed at all, nor is the reader pointed to the supplement or data comparisons to compare the raw data, calculations, and calibrations used to gather the final data. Given the nature of this paper as a low resolution Neogene time-series, I think further validation and comparison are required.

Response 3: To further validate and compare, we now show the raw data in Figures 2, and point to this in the section 3.1, line 283 “Geochemical results”.

2
We have now added text on the potential influence of diagenesis to a section in the methods.

“2.2 Preservation

Microfossils in sediments at these sites, as with any sedimentary sequences, have the potential to be influenced by diagenesis. Despite evidence of authigenic carbonate formation, recent modeling work concluded the influence of dissolution and reprecipitation at Sites 806 and 807 was relatively minor (Mitnik et al., 2018). Prior work has also found minimal impacts on the B/Ca ratio of Pliocene foraminifera from Site 806 (White and Ravelo, 2020), and on the Mg/Ca ratio of Miocene *D. altispera* shells at Site 806 (Sosdian et al., 2020). The weight/shell ratio is commonly used to monitor dissolution, and the only published record at Site 806 for the Pliocene does not show a trend consistent with dissolution of *T. sacculifer* (Wara et al., 2005). We do note that while the “coccolith size-free dissolution” index reported in Si and Rosenthal (2019) indicates higher dissolution rates in the Miocene, their records were thought to be biased from changes in foraminifera assemblages as discussed in White and Ravelo (2020).

To further assess the potential impact of dissolution in our geochemical data, the weight/shell ratio was examined in our samples. The weight/shell index used to monitor dissolution does not present any trend within the interval studied consistent with dissolution. Absolute weights/shell are increasing in the Miocene, which is not consistent with dissolution influencing the record (Fig. 2E). Additionally, we note that reconstructed pH and pCO₂ values also exhibit reasonable correspondence with the Vostok ice core data. Downcore δ¹³B values from Sites 806 and 807 are similar, despite evidence for higher authigenic carbonate at Site 807 relative to Site 806 (Mitnik et al., 2018). Further, the consistency of our boron isotope and Mg/Ca results with at the two sites with each other, and to the published data from Site 872 (Sosdian et al., 2018), each with different sedimentation rates, are consistent with diagenesis not being a primary driver of the record. Comparison of raw data, and derived parameters, is shown in Figs. 2 and 7.”

Please note for the below:


Some additional references I have called out to:

Subâ€”Permil Interlaboratory Consistency for Solutionâ€”Based Boron Isotope Analyses on Marine Carbonates - Gutjahr - 2021 - Geostandards and Geoanalytical Research - Wiley Online Library

Robust Constraints on Past CO2 Climate Forcing From the Boron Isotope Proxy - Hain - 2018 - Paleoceanography and Paleoclimatology - Wiley Online Library

Decreasing Atmospheric CO2 During the Late Miocene Cooling - Tanner - 2020 - Paleoceanography and Paleoclimatology - Wiley Online Library

Upregulation of phytoplankton carbon concentrating mechanisms during low CO2 glacial periods and implications for the phytoplankton pCO2 proxy - ScienceDirect

North Atlantic temperature and pCO2 coupling in the early-middle Miocene | Geology | GeoScienceWorld

Miocene Evolution of North Atlantic Sea Surface Temperature - Super - 2020 - Paleoceanography and Paleoclimatology - Wiley Online Library
A diatom record of CO2 decline since the late Miocene - ScienceDirect

**Response 4:** We have added these references to the text except Meija et al. (2017) - a reconstruction from the EEP.

Specific points:

Line 26: What is the evidence that these sites are in equilibrium today and for the interval studied. To my knowledge the WEP is not considered to be a stable oceanic environment over time.

**Response 5:** At present, Sites 806 and 807 are in quasi-equilibrium with the atmosphere (annual average offset of +28 ppmv; Takahashi et al.; Tripati et al., 2009, 2011; Shankle et al., 2021). This region is considered to be warm and thermally stable. Our calculated pre-industrial pCO2 is 298 ppm (calculated using the GLODAP database and corrected for anthropogenic values) which when compared to the Vostock value of 282 ppm at 1.08ka to a value of +16ppm. This pCO2 difference is similar to our pCO2 uncertainty (~17ppm on average before taking into account the $\delta^{11}$Bsw). This is why we assumed the site was near air-sea equilibrium with the atmosphere.

A few lines of evidence suggest the region was in quasi-equilibrium in the past: 1) zonal temperatures are at a maximum in pre-industrial times and we are able to reconstruct atmospheric pCO2 values, 2) the compilation of temperature proxies shows a weak and stable zonal temperature gradient from ~12 Ma to early Pliocene which would support air-sea stable conditions and air-sea equilibrium (Nathan and Leckie, 2009; Zhang et al. 2014; Liu et al., 2019).

We have added discussion of this to the manuscript on Line 127. “These two sites have been examined in other boron-based studies (Wara et al., 2003; Tripati et al., 2009, 2011; Shankle et al., 2020), as has the region more broadly (Pearson and Palmer, 2000), because they are understood to be in equilibrium with the atmosphere and have relative stable hydrography. The region experiences equatorial divergence but is not strongly affected by upwelling and has a current estimated annual air-sea CO2 flux of +28 ppmv (Takahashi et al., 2014). The pre-industrial air-sea CO2 flux is calculated to be +16 ppm, (GLODAP database corrected from anthropogenic inputs), with a value of 298 ppm, compared to the Vostok ice core value of 282 ppm at 1.08 ka. This pCO2 difference is similar to our pCO2 uncertainty (an average of ~17 ppm for the youngest samples). If trade winds were much stronger, and equatorial divergence greater, in the past, than this could drive some disequilibrium. However, a few lines of evidence suggest the region was in quasi-equilibrium in the intervals we examine: 1) zonal temperatures are at a maximum in pre-industrial times and during the Pleistocene, and we are able to reconstruct atmospheric pCO2 values from the ice cores, 2) temperature proxies indicate the region is relatively stable with respect to temperature compared to other parts of the ocean, and also indicate a weak and stable zonal temperature gradient during the Miocene and Pliocene which would support air-sea stable conditions and air-sea equilibrium (e.g., Nathan and Leckie, 2009; Zhang et al. 2014; Liu et al., 2019).

This work builds on low-resolution prior reconstructions for these sites (Wara et al., 2003; Tripati et al., 2009, 2011; Shankle et al., 2020), Site 872 in the tropical Pacific (Sosdian et al., 2018), and other published boron isotope work, to provide additional data to constrain past seawater pH and pCO2 for the WEP using MC-ICPMS, thereby providing an invaluable new perspective on reconstructing past atmospheric CO2 via marine sediment archives.”

Line 29: ‘reproduce the ice core record’ is very strong language for the comparison which has been carried out. There are only 16 points and no comparative data is produced (e.g. numerical data or crossplotted data).
Response 6: We have modified the sentence, line 30 now reads “We use high-precision multi-collector inductively-coupled plasma mass spectrometry and show that data from these sites are consistent with ice core data and other boron-based studies.”

We also have added crossplots to Fig. 3, in order to quantitatively compare our data to ice core data. Fig. 3E does not show either a significant difference between the slope or intercept with a 1:1 line.

Line 308: “Crossplots comparing our data are presented in Figs. 3C, 3D, 3E; the slope and intercept are not statistically different from a 1:1 line (p=0.69 and p=0.48).”

Line 31: The Miocene data is higher than other published data, but so is the Pliocene, and arguably the latter is much more important as more data is available to facilitate the comparison.

Response 7: As show in Figures 6, 9, and 10, our data for the Pliocene overlap with published estimates from Sosdian et al. (2018) for the Pliocene and Miocene when uncertainties are considered, But Sosdian et al. (2018) do not use the same second carbonate parameter (e.g. DIC) and while Rae et al. (2021) data are higher in the Miocene due to the use of different scenarios of seawater $\delta^{11}$B and alkalinity. For the Pliocene, our reconstruction is marginally higher than these two studies, but when uncertainties are compared, they overlap.

We also compare our results in Figure 8 to recalculated values from Sosdian et al. (2018) using the same $\delta^{11}$Bsw scenario that we are using in this study. As observed in Fig. 6 or S2, the differences between reconstructions mostly arise in the Miocene, that are eliminated if the same methods are used (Figure 8).

Line 33: A 270 ppm transition during the Pliocene iNHG would be very interesting information. This is a huge change (effectively one halving of CO2). Please discuss this more in the context of the other records if you find your Pliocene data to be valid.

Response 8: We now write line 505: “The pCO2 concentrations that we calculate indicates a reduction to 350 ppm by 2.7 Ma, ~280 ppm by 2.6 Ma, and ~210 ppm by 2.4 Ma, in several steps. These results support roughly a halving of CO2 values when compared to values of ~530 ppm at 3.3 Ma. These values are consistent with the pCO2 thresholds proposed by both DeConto et al. (2008) and Koenig et al. (2011) for the intensification of Northern Hemisphere glaciation and the low atmospheric CO2 (280 ppmv) scenario from Lunt et al. (2008). Mg/Ca SST decline from 30°C to 26°C, supporting an Earth System sensitivity of ~4°C/doubling of CO2 over this range, although given uncertainties, higher values of ~6°C/doubling of CO2 that have recently been proposed (Tierney et al., 2020) can not be excluded.”

Line 68: There is no atmospheric CO2 data from the Pliocene available in the blue ice cores, although they did confirm the presence of ice which is of that age. Please correct this.

Response 9: Thank you. We have changed the “Pliocene period” to “the early Pleistocene period”.

Line 72: Foster 2008 is not a B/Ca to CO2 paper, and most recent studies have stopped plotting the B/Ca datasets as there were found to be too many divergent controls on the incorporation.

Response 10: We wanted to acknowledge the different proxies used for CO2 reconstruction. We agree that the reconstruction from Foster et al. (2008) is primary based on $\delta^{11}$B but that study does actually use B/Ca to constrain [CO$_3^{2-}$] as a second carbonate parameter. To acknowledge this we also added this reference when citing the boron isotopes based studies.

Lines 94-102: Use Hain et al. 2018 for the strongest case that $\delta^{11}$B can be a viable CO2 proxy, regardless of other uncertainties.
Response 11: Thank you! We have added the reference.

Lines 103: Here you refer to various $\delta^{11}B$ studies as ‘high resolution’ and yet earlier refer to the ice cores as ‘relatively high resolution’, I would suggest being more consistent within the manuscript regarding what is ‘high resolution’ given the timescales you are talking about. The ice cores, Martinez-Boti, Dyez, Chalk, de la Vega and Greenop studies are high resolution compared with your work here, but Foster, Hoenisch, Sosdian are more similar to your new records. Consistency with this will stop some of the false equivalency made about resolution in this paragraph.

Response 12: We removed the “Relatively” from “Relatively high-resolution” for the Vostok ice core. We have modified the statement in Line 106 to read “Given the evolution of the field, there are relatively few studies generating high-precision boron-based records over major climate transitions in the Cenozoic using recent analytical methods, that incorporate our current understanding of the proxy (e.g., Greenop et al., 2014; Martinez-Boti et al., 2015b; Chalk et al., 2017; Dyez et al., 2018; de la Vega et al., 2020).”.

Line 126: These studies are problematic in their interpretation of CO2 data and call into question the assurance that these sites have remained in equilibrium. This point is repeated on line 152 but without an explanation to the reasoning behind. Given that this is the key assumption in this manuscript I think it deserves more attention.

Response 13: Comparison with the recent boron isotope-based pCO2 studies including compilations from Sosdian et al. (2018) and Rae et al. (2021) shows that our data are in broad agreement, as discussed in our response above. We do acknowledge that the models used ($\delta^{11}B_{sw}$ and alkalinity) for each reconstruction induces important variability (Fig. S2), and we show that when using a similar set of assumptions, our results are similar to work from Sosdian et al. (2018) for Site 872 in the region. We also now show the raw data, as suggested, and the pH estimates, that further corroborates the fidelity of our reconstruction. With respect to air-sea equilibrium, we have expanded the discussion, as described in an earlier response. There is no evidence or physical oceanographic or biogeochemical explanation for why the region we are examining should have been in more disequilibrium than other regions examined, which is why it has been the target for other pCO2 work (Pearson and Palmer, 2000; Wara et al., 2003; Shankle et al., 2020). For example, the EEP/Carribbean did experience major gateway changes that are much more likely to have influenced the hydrography of the region over the timescales examined. Other areas may have experienced upwelling changes.

Line 155: How much could disequilibrium impact these data? No reference is made to preservation or potential diagenesis changes that may impact the data to a far larger extent.

Response 14: Please see our above response on disequilibrium, and the new text starting on Line 135. We have now added several sections that relate to preservation, including the section described above in the methods (section 2.2)

Line 160-166: The age models are not great for these cores, but I do not think that matters given the resolution of the data. It may be worth stating here that no direct comparison of ages between the cores is made

Response 15: We changed the age model of site 806 and used the polynomial regression developed in Lear et al. (2015) based on biostratigraphy.

Line 198: “The age model for Site 806 from 0-1.35 Ma is based on Medina-Elizalde and Lea (2005) which corresponds well with ages from the Lisiecki and Raymo LR04 stack (Fig. 2A). The fourth polynomial regression-based biostratigraphy from Lear et al. (2015) was used for the rest of the record, following
other work (Sosdian et al., 2020). Ages for Site 807 are based on published biostratigraphy (Berger et al., 1993) for 807 with additional constraints placed by Zhang et al., (2007) for the interval from 0-0.55 Ma.”

Line 170: Missing accent on Plouzané.

Response 16: This has been corrected.

Line 194: Gutjahr et al 2020 is the updated reference for this.

Response 17: This has been corrected.

Line 197: This is great, where is the data though? Can you add a supplementary table?

Response 18: We originally added the data as a supplementary table. You can find it in Table S4 and S5 in the supplemental information.

Line 210: you have ‘less than’ and ‘<’.

Response 19: We removed the “<”. This sentence now reads “contributed to less than 1% of the sample signal”.

Line 217: Why is the 2SD for the $\delta^{11}\text{B}_{\text{NEP}}$ so much larger than for the other data despite the increase in n?

Response 20: Good question, so the data come from independent microdistillations which took a small amount of powders each, we think the largest variability can be due to homogeneity in the NEP powder, as the same variability have been observed both at the University of Cambridge and at IUEM for various projects. Similar results were reported by Sutton et al. (2018). Note that the difference was not observed in McCulloch et al. (2014), Rapid, high-precision measurements of boron isotopic compositions in marine carbonates. The JCP-1 seems more homogeneous and is reported around 0.2 permill.

We now provide a table of those standards in a table in the supplementary materials (Table S4).

Section 3.1: This needs to be much more quantitative if you are going to use it to pin all of your results on.

Response 21: We now add a few crossplots in Fig. 3 in order to make it more quantitative, the slope and intercept of the linear regression between pCO₂ from ice cores and from boron isotopes (Fig. 3E) is not significantly different from the 1:1 line.

Line 309: These alkenone records are out of date, please refer to Tanner et al 2020 and Super et al 2018 for updated interpretations.

Response 22: We added those references in the text. We made a compilation of Mg/Ca based SST using our framework in Figure. 4, we also added a section to discuss changes in temperature where we added the work of Super et al. (2018). We have added Section 3.3: “3.3 Sea surface temperature in the WEP” . We also added the Tanner et al. (2020) pCO₂ reconstruction based on alkenones to the Figure 6.

Line 420: “However, reconstructions are still few and discrepancies between boron isotopes and alkenones based reconstructions lead to uncertain pCO₂ history. Nevertheless, recent literature was able to get coherent reconstructions from both proxies (Rae et al., 2021). To date, boron isotopes and alkenone-based pCO₂ reconstructions support higher pCO₂ during the MCO and a decrease over the MMCT (Sosdian et al. 2018; Tanner et al., 2020).”
Response 23: We have modified this section. It now reads line 407:

“However, reconstructions for the Miocene are still relatively limited (Sosdian et al., 2018; Rae et al., 2021). Current boron isotope and alkenone-based pCO2 reconstructions support higher pCO2 during the MCO and a decrease over the MMCT (Sosdian et al. 2018; Stoll et al., 2019; Tanner et al., 2020), consistent with what was previously inferred from B/Ca (Tripati et al., 2009, 2011).

We applied the same framework we used for calculations at Sites 806 and 807 to published boron isotope data from Site 872 (Sosdian et al., 2018) in order to extend the WEP record to the early Miocene (Figs. 7, 8). The Miocene data between Sites 806 and 872 do not overlap as both are low in resolution, but do show excellent correspondence in their trends in δ11B and reconstructed pH. The pH values we reconstruct are within error of published estimates from Site 872 (Sosdian et al. 2018, Figs. 7D and 8D). Collectively, these data suggest the early Miocene WEP was characterized by a mixed-layer pH of 8.1 ± 0.1 (2 SD, n=4) between 19.4 and 21.8 Ma, which decreased to reach a minimum during the MCO of 7.7 (± 0.11).

Given the sensitivity in absolute pCO2 to assumptions about the second carbonate system parameter, a few scenarios were explored for the combined 806/807/872 reconstructed pH values. For all alkalinity scenarios we used, reconstructed pCO2 shows an increase from the Early Miocene to the MCO, with the highest values in the MCO. Recalculated pCO2 for Site 872 between 19.4 and 21.8 Ma is 232 ± 92 ppm (2 SD, n=4), lower but within error of the ones presented in Sosdian et al. (2018) and also within error of a constant alkalinity scenario (Fig 8D). The main difference between reconstructions is when comparing the same data recalculated in Rae et al. (2021) that show higher pCO2 between 19.4 and 21.8 Ma, with an average value of 591 ± 238 ppm (2 SD, n=4) for Site 872, because of the different assumptions used in their study and ours. This difference is important because that would imply a relatively high and stable pCO2 from the early Miocene to MCO, which would imply a decoupling between pCO2 and temperature with no pCO2 change during an interval of decreasing benthic δ18O. However, our reconstructed pCO2 increase towards the MCO is in line with the observed benthic δ18O decrease and δ13C increase and suggest a coupling between temperature and pCO2 over this period. We note that overall, Mg/Ca-SSTs are warm (>32 °C), and there are relatively small changes in Mg/Ca-SST from the early Miocene into the MCO.”

Line 314: The S18 study is a reinterpretation of the same data from the other two, so this sentence either needs to explain that or just use the most recent iteration.

Response 24: Line 442, we changed for “Published δ11B-based reconstructions also support higher pCO2 for the MCO of ~350-400 ppm (Foster et al., 2012) or 300-500 ppm (Greenop et al., 2014) that was recalculated by Sosdian et al. (2018) to be ~470-630 ppm depending on the model of δ11Bseawater chosen.”

Line 321: This section is not a fair representation of the existing data. The δ11B presented here is ~13.8 % and the minimum in other records e.g. S18 is ~15 %, in addition, the data of S18 from ODP 806 is geographically very close to ODP 806. ODP 871 from G14 appears to match the data in S18 quite well, which would imply that another reason is responsible for the difference seen here between 872 and 806 (ocean frontier, preservation or analytical). In addition, G14’s raw values (for T. trilobus) are fairly similar to the data presented here. This would then suggest the reason for the difference is in interpretation and calculation. I think plotting the available data in raw δ11B and either recalculating or plotting calculated CO2 would really help with this point. There is not a huge amount of data for this period and it is worth discussing fully where it does and does fit.

Response 25: Raw data δ11B data are presented in Fig 2. We now provide a comparison of our calculated data with recent compilation of S18 and Rae21 in several places (see Figs. 5, 6, 7 and 8, S1, S2). We
believe that those figures show the variability of the pCO₂ values calculated depend on the assumptions made for alkalinity and δ¹¹Bsw.

Data from Site 872 were used to extend our record to the early Miocene and were reprocessed using our framework. The closest datapoint we have is Site 806 (15.6 Ma, δ¹¹B=14.47+/- 0.21 permill) and Site 872 (16.7 Ma, δ¹¹B=15.12+/- 0.25 permill) which have boron isotope signatures close to each other which support a minor effect of diagenesis and the variations a real environmental change which would be in line with the δ¹⁸O benthic slack.

Our δ¹¹B data are typically lower than published data from other ODP sites (926, 761, 1000) located in different oceans basins. While there is no data overlapping in time period between Sites 872 and 806, both sites are located in the WEP, and both records can be combined to provide a continuous record and the additional data provides further constraints to the early Miocene period. To robustly do this, we applied the same framework to the δ¹¹B data from S18 from Site 872 (Figs. 7 and 8). Those data are in absolute values, slightly lower than ours, but that is to be expected if CO₂ was changing in a manner similar to what is seen in the benthic δ¹⁸O stack. The combined records would suggest a larger CO₂ increase than recalculated from S18 alone, from the early Miocene into the MCO.

All three sites are located above the lysocline, and please see section 2.2 for the discussion about diagenesis.

Line 327: I’m not an expert on this topic but I think both of these ideas have been updated in Stoll et al 2019 and Tanner at al. 2020.

Response 26: Yes thank you for this remark, we have updated this and cited those references, as described in Response 23.

Line 328: these very warm temperatures also appear in the Atlantic TEX86 study of Super et al. 2020 which you could cite here.

Response 27: Thank you, we added the reference to section 3.3.

Line 363: Please given the ‘marginally consistent’ value here as well, rather than just the inconsistent. Also see S18 for reconciliation between the datasets of MB15 and B11.

Response 28: We now present comparisons with S18 and Rae201, and we removed this sentence.

Line 367: de la Vega capitalisation.

Response 29: We corrected it.

Line 371-373 : please expand on the good agreement here as above. Stap et al. 2016 does not agree particularly well with the other studies.

Response 30: The pCO₂ reconstructed from the original paper from Stap et al. (2016) appeared to match our reconstruction, however no overlapping data exist. Nevertheless, we note that the data reprocessed by Sosdian et al. (2018) lead to significant changes and lower pCO₂ values from Stap et al. (2016) compared to their original publication. We removed Stap et al. (2016) from the references of this sentence.

Lines 376-384: This section reads like it should be in the introduction rather than results.
**Response 31:** As with the other sections in the results and discussion, as we step through each time interval, we provide a broader perspective on the interval for the paleoceanographer/paleoclimatologist to contextualize the data.

Line 385: 150 ppm is a lot of disagreement, and as stated it is down to the raw $\delta^{11}$B values. I do not follow the vague argument about disequilibrium that has been made several times now.

As in theory, increased upwelling, increased respired carbon dissolved in surface water, reduce pH and increase CO2 estimate and vice versa.

One issue with 999 is the Panama Isthmus and potential influx of surface EEP water. Upwelling in EEP makes water more acidic therefore, if there were to be an increased influx into the Caribbean, this would reduce pH and increase CO2 estimates. For the inverse to happen, 999 would need to be a sink for CO2 so barring a huge change (e.g. reversal of AMOC), it is more likely that CO2 estimates at 999 will overestimate rather than underestimate CO2. With this in mind the difference between 806/7 and 999 would then require even more disequilibrium in the WEP.

I would favour that the Pliocene section here is more likely to have been impacted by diagenesis, or by the closure of the Indonesian through-flow, which is not discussed here.

**Response 32:** We agree that the variability in reconstructed pCO$_2$ mostly comes from the assumptions made for the reconstructions from the different studies. Our work on the EEP will help to shed light on the significance of differences in Site 999 compared to 806.

Please see Response 3 for the discussion of diagenesis, and Response 5 for discussions about air-sea equilibrium. We want to emphasize it is unlikely that the WEP is out of equilibrium with the atmosphere during the Pliocene, and the similarity in reconstructed values with Site 871 suggests that changes in Indonesian through-flow did not impact our site.

Line 389: I am not sure you have the resolution to say this, would suggest removing.

**Response 33:** We agree that we may not capture the minimum pCO$_2$ over those time intervals due to our resolution, so we removed this sentence and start the paragraph as followed:

Line 505: “The pCO$_2$ concentrations that we recalculated are consistent with the pCO$_2$ thresholds proposed by both DeConto et al. (2008) and Koening et al. (2011) for the intensification of Northern Hemisphere glaciation and the low atmospheric CO$_2$ (280 ppmv) scenario from Lunt et al. (2008).”

Line 401-405: Please reference this section. I would also incite the logarithmic nature of CO2 forcing here, see multiple papers e.g. MB15, C17, DIV20.

**Response 34:** We have added references to this section, including the three suggested references.

Line 415: All of these papers DO suggest a decline over the MPT, it is the main finding of all of them.

**Response 35:** We changed for Line 536: “Previous boron isotope studies for ODP Sites 668 and 999 in the tropical Atlantic Ocean have suggested that a decline in atmospheric CO$_2$ did occur during glacial periods in the MPT, but not during interglacials (Hönisch et al., 2009; Chalk et al., 2017; Dyez et al., 2018).”

Line 425: When is the end of the MPT, please define.
Response 36: We defined the MPT between 1.2 to 0.8 Ma and added this to the text.

Lien 427-437: This whole paragraph is extrapolated from one point. I think just show you agree with the data of H09 and move on.

Response 37: We have edited this section to say: “While data resolution are limited, we speculate this could explain why glacial/interglacial amplitudes in WEP pCO₂ values decrease from the MPWP towards the Pleistocene, whereas variations in δ¹⁸O are increasing – a speculation that could be tested with increased data resolution.”

Line 466: Mejia et al 2017 would be a useful reference here.

Response 38: We removed this section following comment from reviewer 2, we did not include Mejja et al. (2017) because reconstruction is based on Site 846 located in the EEP, influenced by upwelling.

Line 472: exchange ‘many others’ for an e.g. prior to the reference list, or give a few more.

Response 39: We removed this section following comment from reviewer 2.

Lines 497-500: This is precisely the point of H09, C17 and D18, I do not think that the addition of one data point allows the confirmation of these claims.

Response 40: Our data support this statement, but it is true that our resolution is low. We now state line 544: “Although our data are relatively limited, we note they have greater resolution for the middle and later part of the transition than prior publications that have drawn conclusions about the MPT (Hönisch et al., 2009; Chalk et al., 2017; Dyez et al., 2018) (Fig. 10D) and therefore we explore their implications.”

DIV20 study missing from Table 2, represents the key reference for the mid-Pliocene period. The ordering of this table is also confusingly out of temporal order.

Response 41: DIV20 is now included in Table 2 and this table is now in temporal order.

Figure 1: Watch the formatting on the scale axis for the map figure. Please add contours or change the colour scale to something more friendly for colour blindness. The x-axes are also cut off the other panels.

Response 42: We have slightly modified the colors.

Figure 2: Please add a legend to the plot to define the symbols and shades. It would be helpful to colourise your data and add raw data from other studies in grey behind to facilitate easier comparison. A d11Bborate plot would also be useful to account for the different calibrations between species.

Response 43: To allow comparison, we added the raw δ¹¹B data from other studies to Fig. 2 and have added color.

Figure 5: The updated ice core compilation of Bereiter et al. 2015 may facilitate a better comparison for the plot here, or at least including the WAIS divide data from Ahn et al. 2008. A cross plot or calculation of root mean squared error or similar would be a valuable addition.

Response 44: We updated the ice core compilation using the composite of Bereiter et al. (2015).

Figure 7: S18 data is missing from this plot.
Response 45: We added the S18 data to all graphs.

Figure 9: Much is made of the low CO2 found in MIS 30, but this figure shows lower(?) CO2 at 1-1.1 Ma which is not discussed.

Response 46: This decrease between 1-1.1 is MIS30 which we now explicitly mention in the text. Line 550: “We also find evidence that during the MPT, glacial pCO2 declined rapidly from 189 (±30) ppm at MIS 36 (Chalk et al., 2017) to reach a minimum of 170 (±52,24) ppm during MIS 30” and line 555: “In our record for the last 16 Myr, the lowest pCO2 is recorded at MIS 30 during the MPT, with values of 164 (±44,35) ppm, which supports an atmospheric CO2 threshold that leads to ice sheet stability. During this transition, the pCO2 threshold needed to build sufficiently large ice sheets that were able to survive the critical orbital phase of rising obliquity to ultimately switch to a 100 kyr world, was likely reached at MIS 30.”

Figure 10: Again S18 is missing from this figure, despite it being the most complete Neogene study to date (including this one!)

Response 47: We added the S18 data to all graphs.
Reviewer 2

The manuscript by Guillermic et al presents new Mg/Ca and d11B measurements of planktonic foraminifera spanning the last 17 million years from the Western Pacific Warm Pool at ODP 806 and 807 with the aim of estimating past evolution of Sea Surface temperatures and atmospheric pCO2. The most significant new contribution of this study is the addition of measurements between 5 and 17 Ma, as the majority of previous d11B measurements since the mid-Miocene are concentrated in the last 4 Ma whereas low resolution data illustrating long term trends in the 4 to 17 Ma time window are to date more limited. The estimation of pCO2 from d11B of foraminifera in this time period is sensitive to a number of uncertainties, including the assumptions of the evolution of d11B of seawater and alkalinity over time. This contribution employs the current best estimates for these parameters and illustrates the sensitivity of the pCO2 estimate to uncertainties in these parameter choices.

While the processing of the new data is clear and uses up to date alkalinity and d11B seawater estimates, the comparison with previous pCO2 estimates is not presented as clearly as it could be, and the discussion of phytoplankton (alkenone) pCO2 estimates for the Miocene is not up to date. I summarize the main content issues which I believe need to be addressed for this manuscript to provide a coherent step forward in understanding of pCO2 in this time interval. Subsequently, I have some suggestions on the organization and structure which I propose could improve transparency and clarity in the manuscript, as well as some more detailed comments.

Content and interpretation:

While the authors present for their own data an updated estimation of the CO2 considering recent proposals for d11B seawater (eg Greenop et al 2017) and alkalinity history derived from Caves 2016 or Zeebe 2005, they do not include in figures (and therefore not thoroughly consider) the published d11B-CO2 estimates which have been recalculated with these same alkalinity and Greenop et al 2017 d11B seawater parameters, namely those compiled and homogenized in Sosdian et al 2018. This is an essential update to make so that the new data from ODP 806 and 807 can be considered in context, and so that the new data contribute to an integrated better picture of the trends and absolute values. In particular, the latest Pliocene CO2 trend is quite clear in the Sosdian et al compilation. Additionally the early Pliocene and Miocene values in the Sosdian et al compilation are higher than in the original publications, and therefore are more consistent with the results as plotted here. Otherwise there is a disconnect between the figures (not homogenized parameters) and the textual mention of pCO2 values recalculated by Sosdian et al 2018. Once this is updated in figures, the discussion should also be updated and clarified.

Response 1: We have made these changes. We added the updated reconstruction from Sosdian et al. (2018) to our figures and compare with our data. We also added a comparison to the compilation published by Rae et al. (2021).

During the Pleistocene period, the reconstruction from Sosdian et al. (2018) does not reproduce the absolute CO2 values from the Vostok ice core. Their results can be offset by ~ +60 to +100 ppm. Their divergence for the Pleistocene is important because it means they have higher absolute pCO2 values for the early Miocene, after adjustment. However, their results are really similar to our reconstruction for the Pliocene and the Miocene periods.

We also present the recalculated data from Rae et al., 2021 for the Pleistocene and Pliocene periods which does accurately reconstruct ice core pCO2 values, and for this interval, choice of alkalinity and d11Bsw scenarios have a minimum effect.
The estimation of tropical SST over this time interval is not trivial given the uncertainty in seawater Mg/Ca, and I think this warrants further clarification and transparency. An inferred seawater Mg/Ca history is sketched in Figure 2 but the Figure caption does not specify the origin of this curve. The calculation equations are provided in supplement but the input data on seawater Mg/Ca should be illustrated along with the data forming the basis for its estimation (e.g. the basis from which it is derived). Figure 7 illustrates some uncertainty around the SST, but the Figure legend does not indicate what is represented by this uncertainty. To what extent does the uncertainty in calculated SST (e.g. choice of Mg/Ca seawater correction, and regression) affects the estimated pCO2 due to solubility? Also, have the authors considered the influence of a pH correction to the Mg/Ca SST, as conducted in recent study (Sosdian and Lear, 2020) and shown to be significant across the MCT (Leutert et al 2020)?

Response 2: The Mg/Casw were based on polynomial regression based on Gothmann et al. (2015) data, however, to be consistent with the literature and because variations were in line with our first regression we now utilized the fourth-order polynomial regression from Sosdian et al. (2020), data and figures are updated. However, the overall difference between the two data processing were <1°C. We applied a pH correction to G. ruber utilizing the sensitivity published in Gray and Evans, (2019). We used an iteration based on pH and temperature until difference was negligible. T. sacculifer did not yet proved to be influenced by pH so we did not apply a correction. Salinity correction was applied for both species. We now present the Mg/Ca data along the SST reconstruction in Fig. 2C and recalculated the temperature based on published Mg/Ca values at site 806 and 872 (Fig. 4). The temperature compilation was realized using Mg/Ca at Site 806 (Wara et al., 2005; Nathan and Leckie, 2009; Tripati et al., 2009) and Site 872 also located in the WEP (Sosdian et al., 2018).

The references on the alkenone pCO2 reconstruction are not up to date. Lines 308-310 refer only to older publications based on a theoretical diffusive model of CO2. Recent metaanalysis of culture carbon isotopic fractionation (epsilon p or ep) data suggested that due to the operation of carbon concentrating mechanisms, ep exhibits a much lower sensitivity to CO2 than originally inferred; application of the sensitivity observed in cultures to sedimentary ep measurements yields a significant pCO2 decline since the mid-Miocene (Stoll et al., 2019). This low ep sensitivity is supported by recent determinations over glacial cycles (e.g. Badger et al 2019) and further suggests significant pCO2 decline in the late Miocene (Tanner et al., 2020) which would be a relevant reference for comparison in section 3.4. A detailed updated discussion is provided in Rae et al, 2021.

Response 3: Thank you for this comment, we updated this paragraph with those references, and we also added Tanner et al. (2020) data to Figure 6.

Line 420: “However, reconstructions are still few and discrepancies between boron isotopes and alkenones based reconstructions lead to uncertain pCO2 history. Nevertheless, recent literature were able to get coherent reconstructions from both proxies (Rae et al., 2021). To date, boron isotopes and alkenone-based pCO2 reconstructions supports higher pCO2 during the MCO and a decrease over the MMCT (Sosdian et al. 2018; Stoll et al., 2019; Tanner et al., 2020).”

The discussion of previously published d11B records is in many places overly superficial. For example in lines 314-316, previously published d11B records of the MCO (Sosdian et al 2018, Greenop et al 2014) are diminished in importance by suggesting "it is unclear if these values accurately reflect the atmosphere given the sites may or may not have been in equilibrium with the atmosphere." The cited studies reflect multiple sites (ODP 926, 999, 668, 761..), all in comparably reasonable locations to be close to equilibrium with the atmosphere. Unless the authors would like to present clear new evidence that some of all of these sites are less likely to have remained in equilibrium with the atmosphere than ODP 806 or 807, the original interpretation that these sites (of preindustrial pCO2 disequilibrium <25 ppmv) remained close
to equilibrium should be respected, and other potential explanations for the differences should be explored.

**Response 4:** We now present a comparison with data from Sosdian et al. (2018) and Rae et al. (2021). It is clear that the choice of $\delta^{11}$Bsw and alkalinity scenarios is critical and lead to most of the variability we observed in pCO$_2$ reconstructions between studies. We then removed the disequilibrium argument from this paper as there is no strong evidence for it, especially since we can reconcile data from site 761 and our WEP data.

Site 806 and 807 are sites estimated to have fast rates of diagenetic recrystallization (Mitnik et al. 2018). For example, averaged over the upper 80 m of sediment (approx 3 million years given sedimentation rates), authigenic carbonate is estimated to comprise 19% of total carbonate at 806 and 36% at 807; in comparison other sites like ODP 999, the authigenic carbonate is <1% of total carbonate in the same depth and time interval. It might be helpful for the authors to acknowledge this and comment on evidence for how this may or may not affect the 11B and Mg/Ca results of the planktic foraminifera.

**Response 5:** We now present the raw boron data from this study along with other boron isotopes based studies (Figure 2). In our study, the boron isotopes data at sites 806 and 807 are lower than other sites. The samples picked were all visually preserved, however no SEM images were obtained during this study to assess potential recrystallization or dissolution, we only used the weigh/shell to monitor dissolution. This record is shown in Fig. 2 and present no decreasing trends toward the Miocene which is not in favor of sample dissolution over the time study. Previous literature observed a significant positive relationship with test size and the boron isotopes in coretop samples (Hönisch and Hemming, 2004; Ni et al., 2007), to overcome this problem we applied a correction based on empirical relationship between the weigh/shell and the boron isotopes (see supplement).

We acknowledge that the % of authigenic carbonate at sites 806 and 807 is important. However, the lack of porewater boron and boron isotopes data at those sites and the high variability in boron isotopes pore water profiles at other sites (Brumsack and Zuleger, 1992) do not permit to conclusively state a potential issue specifically at these sites. However, considering the proportion of authigenic carbonate, diagenetic processes are happening at those sites, which can effectively affect our data through partial dissolution or preferential dissolution of the test. Because our data are lower than other published data of ~2permill, and our samples going through rigorous cleaning, I would think that preferential dissolution of the heavy isotopes or preferential dissolution of onotogenetic calcite relative to the light gametogenic calcite could be relevant here. However, no trend is observed for the weigh/shell data and the empirical weigh/shell correction we are using is consistent for our pCO$_2$ reconstruction in comparison to the Vostok ice core (Fig. 5). Also, comparison between sites 806 and 807 shows that similar correction needs to be applied which is not going in the direction of a higher influence of authigenic carbonate at site 807. Moreover, recent study from White and Ravelo, (2020) tried to constrain the dissolution effect at site 806 by reconstructing deepwater saturation state through the Pliocene, no trend were observed in their record suggesting minor dissolution effect on the Mg/Ca. Despite evidence of diagenesis processes, the record suggests they have a minor effect on the $\delta^{11}$B of our shells.

We have added section 2.2 “Preservation” starting line 171 to address the preservation of the samples.

The coherency of the $d^{11}$B-pCO2 estimates with ice core pCO2 is always a useful comparison, but its effectiveness relies on the precision of the age model used for this portion of the sediment core (as well as the precision of the ice core age model, which cannot be investigated here). Particularly relevant to the last 800 ka, section 2.2. should detail on what the age models are based not just the publication source. From the reference cited for the last 1.35 Ma, it appears the age model is based on d18O of planktic G. ruber -
is it still tuned to SPECMAP chronology as in Lea et al 2000, or is it retuned to LR05? In Figure 5, I think it would be better to show the site 806 d18O G. ruber in the upper panel, eg the metric from the same site and age model as the d11B estimates, rather than the LR05. Then, I would suggest in addition to the time series, a scatterplot of the d18Oplanktic vs d11B-based pCO2 from 806 (assuming that d18O is available for the same core intervals - this gives an estimation of the coherency of pCO2 and glacial cycles in the same core without age uncertainties; were any d18O made on 807?), and also importantly a scatterplot of the d11B-based pCO2 vs ice core pCO2.

Response 6: The age model used between 0-1.35 Ma at site 806 is from Medina-Elizalde and Lea, (2005). We added the high resolution d18O G. ruber from the same study in Figure 3 alongside with the stack from Lisiecki and Raymo, (2005) for comparisons. The two records are showing good agreement with each other even if not tuning against each other but are resolving the IG/G cycles. The ages of the deeper samples were calculated based on the fourth-order polynomial regression based biostratigraphy events (Lear et al., 2015). Considering the low resolution in our samples and the availability of d18O data we still present our data with the Lisiecki and Raymo, (2005) and Zachos et al. (2008) stacks as observed in other studies (Sosdian et al., 2018, 2020).

For site 807, between 0-0.5 Ma, Zhang et al. (2007) derived an age model based on the correlation of the oxygen isotopic curve of C. wuellerstorfi to the SPECMAP stack (Imbrie et al., 1984). In addition, they used one planktonic foraminiferal δ18O event and two nannofossil datum levels (Prentice et al., 1993). Ages of the deeper samples were calculated based on linear interpolation of the biostratigraphy events.

We have added few crossplots in Fig. 3, in order to have a better quantitative comparison between our data, the ice core data and also the d18O. No linear regressions were significant (p<0.05) but the crossplot Fig. 3E does not show either a significant difference between the slope or intercept with the 1:1 line.

Line 308: “Crossplots comparing our data are presented in Figs. 3C, 3D, 3E; the slope and intercept are not statistically different from a 1:1 line (p=0.69 and p=0.48).”

Suggestions on organization:
I recognize the challenge of illustrating the effects of possible assumptions of d11B and alkalinity on the final CO2 calculation, but I am not convinced the current organization is the most effective and it leads to an unusual ordering of figures. The Methods heading "2.7 " effectively starts presenting results and sensitivity analysis.

The authors might consider if a more direct presentation of results and discussion could:

a) begin with section 3.1, and start with the current Figure 5 - the last 800 ka uses modern d11B sw and alkalinity so is not subject to the uncertainties/sensitivity analysis on both parameters.
b) continue with a section on the measured indices and summarizes the findings in of the current Figure 2 which presents the measured results.

Response 7: a) and b): We started with Figure 2 showing the raw results, like this we can introduce the comparison between raw published data and talk about potential impact of diagenesis. Then we are presenting the comparison with the ice core data.

c) comment on the inferred trends in SST and uncertainties in their calculations, and comparison with other SST histories both from Mg/Ca (Sosdian and Lear 2020) as well as TEX86 (Zhang et al. 2014, )

Lines 286-288 needs to clarify if the measured Mg/Ca is consistent with the other published Mg/Ca, or if the calculated temperatures are consistent with the published Mg/Ca calculated temperatures; and in the
latter case, have the temperatures for all these studies been recalculated using the same assumptions of Mg/Ca seawater and temperature regression as used for the new data here?

**Response 8:** We now made a compilation of available Mg/Ca at site 806 and 872, all data are on the same age model and were recalculated using the same framework which implies a pH correction for *G. ruber* (only few data), a salinity correction for *G. ruber* and *T. sacculifer* and taking into account the secular changes in seawater Mg/Ca using the fourth-order polynomial relationship derived in Sosdian et al. (2020). The raw Mg/Ca data are presented in Fig. 2 and the temperature reconstruction in Fig. 4.

d) discuss sensitivity of Neogene pCO2 estimations to assumptions of d11B seawater and alkalinity, which could introduce the current Figures 3 and 4 as the sensitivity of the results to d11Bseawater and alkalinity (and is there sensitivity to SST) and incorporate the introduction to this currently in the methods section

-continue with the discussion of temporal trends in calculated pCO2

**Response 9:** Section about sensitivities have been moved as suggested. We believe we present clearly the impact of the different scenarios line 323 in section “Scenarios of reconstructions”.

As the manuscript begins to go through the main pCO2 results, I am not fully convinced that the current organization of the discussion is the most straightforward and concise. In the current organization the authors new data seems like it gets buried within the discussion. If the current time interval based structure is used, I believe it would be useful if in each heading of the results/discussion section, the authors presented first the summary of their own new results, and followed it with comparison to other proxy pCO2 results and finally to climate.

Also, if organization based on time periods is used then clearer section headings are needed For example 3.3 is "Miocene" but 3.4 is "Late Miocene" which is a period nominally included within the Miocene heading. I am not sure if division of the Pliocene into the warmth then glacial intensification then Pleistocene (3 sections) is really needed to discuss the author's new data, as these are time periods with substantial previously published data and interpretations and the authors new data are largely consistent with and reproduce these earlier results. Overall, I believe the discussion section can be streamlined and made more concise. Some sections such as 3.9 seem very extraneous, as there is really limited new SST data and it is not coherently presented to evaluate east west gradients; I suggest this section be eliminated from the discussion. Section 3.8 is not really clear in advancing a mechanism for the CO2 variation, and I suggest the key points might be effectively commented within the context of the Miocene and Pliocene sections of the text.

**Response 10:** We changed the heading for “Miocene”, “Pliocene” and “Pleistocene”. We focused on the data comparison with Sosdian et al. 2018 and Rae et al. 2021, especially for the early Miocene period. We removed section 3.9 from the manuscript as we focus on the data.

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Detailed comments:

ALL of the figures should more accurately show the true data density, including Figures 3 and 4 - continuous fill patterns and no symbols is not a clear way to represent the data. At minimum, symbols are bars are needed to show where there are datapoints (alternatively rather than complete shading, points with error bars could be illustrated to show the sensitivity).
Response 11: We added the datapoints in Fig. 3. We kept the format of Fig. 3 but error bars are presented in Fig. 4. And in Figures 6-8 and 10, the broken shading at least alerts to the data gap, but I think it would be ideal to show no shading over the long intervals without datapoints.

Response 12: Figures now represent true data density, we left blank the periods between ~5-7, ~10-12 and ~14-15 Ma. Please clarify the basis of the age model (eg benthic d18O, biostratigraphy, etc), Section 2.2 is not sufficiently clear.

Response 13: We believe we clarified the age model in the main text and response above. Could a more direct heading for Methods section 2.6 be developed?

Response 14: We could reduce the method section but we think it is important to keep this section as it is for data validation from the community.

References cited (not cited in the manuscript):


