Peer Review File

Manuscript Title: Hemispheric black carbon increase after 13th C Māori arrival in New Zealand

Editorial Notes:

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

I was asked to review this paper specifically from the viewpoint of an expert in Polynesian archaeology, and my comments are limited to that perspective. I do not see anything in the paper that contradicts current views on the chronology of Polynesian settlement of New Zealand; the authors have cited appropriate current literature on that topic. Their conclusions regarding anthropogenic burning in New Zealand following Polynesian arrival in the 13th century are entirely consistent with recent archaeological and paleoecological studies. Indeed, this paper adds significant new evidence for the large-scale regional impact of such burning.

Referee #2 (Remarks to the Author):

This article shows ice core evidence for an increase in BC deposition following the Maori settlement of New Zealand. The authors use a combination of models and ice core data to show that the most likely hypothesis is that the change is from biomass burning in New Zealand.

Overall, I think this is an excellent paper, that is really important, and thus worthy of being published in Nature because of how interesting and synthetic it is...

I do have a couple minor points:
1. using the authors methodology, they should be able to provide statistical analysis of the odds that it is from the biomass burning in New Zealand.
2. the authors should put this paper in context: there are other papers arguing that European expansion changed biomass burning, and this paper supports that idea: people change the fire regimes! these arguments are really important for understanding what the fires were in preindustrial times. Please cite and put into the context of: Hamilton etal., 2018: Nature Communications.

Referee #3 (Remarks to the Author):

Ice cores drilled from ice sheets and glaciers are ideal material to reconstruct the emissions from natural wildfires and human activities, as they directly preserve the information of atmospheric aerosol transport and deposition. In this study, McConnell and his colleagues used a broad Antarctic array of well-dated, 2000-year records from ice cores and modeling to investigate atmospheric black carbon concentrations over the northern Antarctic Peninsula (nAP) and much of continental Antarctica. The results showed the rBC concentrations and deposition rates were approximately stable over the continental Antarctica during the Common Era, while they presented a 2- to 3-fold increase over the nAP during the past 700 years. Meanwhile, by using FLEXPART simulations and sediment-charcoal records, McConnell and his colleagues demonstrated the main source of the rBC increase over nAP was from New Zealand after the Māori settlement. The BC emissions from biomass burning by Māori was quantified.

New Zealand was the last major landmass to be settled before the industrial age. The precise date of first colonization has been discussed. Previous studies use several methods to determine the date of Māori migration, including the radiocarbon dating of short live samples (e.g., Wilmshurst et al., 2008, 2011), archaeological signature (e.g., Higham et al. 1999; Walter et al., 2017), charcoal records (e.g., McWethy et al., 2010), the extinction of moa (e.g., Holdaway et al., 2014). This study provides a new method to study the precise time of Māori settlement. McConnell and his colleagues suggest

that the date of Māori settlement is 1297 (±30), which is consistent with previous studies.

In my opinion the results presented are of immediate interest to many people in study areas of environmental chemistry, climatology, and paleoclimatology.

Most of my research is about the transport and deposition of anthropogenic aerosols using ice core records and in situ observations. In this review, I will focus on the biomass burning records in the Antarctic ice core array.

The timing and magnitude of the biomass burning increase of rBC are very likely robust. However, the following issues may need being considered.
1. BC emissions from Māori BB were estimated to be 38 (±19) GgrBC/y. This value was used in the forward simulations. The results showed that the BC from New Zealand, as shown in Figure 3, deposited largely over the Pacific Ocean, and substantial over the Atlantic and Indian Ocean sectors of the Southern Ocean. This is very likely true. However, what should be noticed is the depositions over the Patagonia region are considerably large as well. As shown in Figure 3, the deposition value in the Patagonia region was even higher than nAP. If Māori settlement and consequently widespread biomass burning since 1297 (±30) resulted in the rBC deposition 2- to 3-fold higher at nAP, then the charcoal records in the Patagonia should have increased during the past 700 years as well. However, the Patagonia charcoal records did not show this pattern, but presented an evident decrease. Reduced natural biomass burning by wet climate cannot fully explain the decrease.
2. All the charcoal records, including New Zealand charcoal (Figure 1 e and f in the manuscript), showed low values during the 16th C. This pattern also can be seen in the ice records from the continental Antarctica. But the rBC depositions over the nAP showed a linear increase during the 14th C to 16th C.
3. A simple question: When using the FLEXPART atmospheric aerosol and deposition simulation, the rBC aerosol was set to be released over a grid from 37 to 45°S and 170 to 180°E, while the most west of New Zealand is 166°E. Why not cover all land of New Zealand?

Reference:
Higham, T., Anderson, A. & Jacomb, C. Dating the first New Zealanders: the chronology of Wairau Bar. Antiquity 73, 420–427 (1999).
Holdaway, R. N., Allentoft, M. E., Jacomb, C., Oskam, C. L., Beavan, N. R., & Bunce, M. An extremely low-density human population exterminated New Zealand moa. Nature Communications, 5(1), 1-8 (2014).
McWethy, D. B. et al. Rapid landscape transformation in South Island, New Zealand, following initial Polynesian settlement. Proceedings of the National Academy of Sciences 107, 21343, doi:10.1073/pnas.1011801107 (2010).
Walter, R., Buckley, H., Jacomb, C. & Matisoo-Smith, E. Mass Migration and the Polynesian Settlement of New Zealand. Journal of World Prehistory 30, 351-376, doi:10.1007/s10963-017-9110-y (2017).
Wilmshurst, J. M., Anderson, A. J., Higham, T. F., & Worthy, T. H. Dating the late prehistoric dispersal of Polynesians to New Zealand using the commensal Pacific rat. Proceedings of the National Academy of Sciences,105(22), 7676-7680 (2008).
Wilmshurst, J. M., Hunt, T. L., Lipo, C. P. & Anderson, A. J. High-precision radiocarbon dating shows recent and rapid initial human colonization of East Polynesia. Proceedings of the National Academy of Sciences 108, 1815, doi:10.1073/pnas.1015876108 (2011).

Referee #4 (Remarks to the Author):

Overview:

This manuscript presents a new, 2000-year long ice-core record of refractory black carbon deposition (rBC, i.e. "soot") from the atmosphere in snow on James Ross Island (JRI) at the tip on the Antarctic Peninsula. The authors use this record, in combination with atmospheric aerosol transport simulations (FLEXPART model) to offer an interpretation of the most probable geographical sources

of rBC deposited on JRI in past centuries. One of the main inferences made (reflected in the manuscript title) is that at least from the 13th to the 16th century, biomass burning (BB) emissions associated with prehistorical deforestation in New Zealand caused widespread hemispheric BC dispersion in the circum-Antarctic atmosphere and were chiefly responsible for the rBC deposition trends documented in the JRI ice core. Estimates of the possible magnitude of these BC emissions are presented, based also on FLEXPART simulations, constrained by the ice-core data.

Relevance and significance:

A recent assessment of the uncertainties in estimating aerosol radiative forcing of climate pointed out that "A major component of the variance in preindustrial natural aerosol burden is due to uncertainties in the spatial distribution and total magnitude of fire emissions. A lack of understanding about human influence on fires over the historical period results in a large range of possible land use and land cover change scenarios associated with preindustrial fire estimates." (Wan et al., 2021)

The present manuscript is timely and of great interest precisely because it provides new information on the preindustrial (and more recent) burden of BC aerosols in the southern hemisphere, and of the possible role played by past land use changes (deforestation) and fires in BC emissions. Therefore I think this paper is certainly of great interest. Furthermore it brudges across several disciplines (pyrogeography, biogeography, climate and atmospheric science, glaciology), making it interesting to a broad audience. Much of the work presented in the paper is based on state-of-the-art methods (not truly novel, but sound) and seems very robust. However, I think the authors over-reach their interpretation and are more confident in some of their main findings than is warranted. Specifically, I think that the following inferences in the paper are well-supported:

-There is clear evidence of an abrupt increase in rBC deposition at JRI starting in the 13th century, and sustained thereafter. This contrasts with ice-core records of rBC from the East Antarctic interior, which show no such increases.

- FLEXPART model simulation results show that unlike the interior regions of Antarctica, the northern part of the Peninsula is much more sensitive to BC emissions from low to mid- latitude southern hemisphere (SH) regions (especially those in South America, New Zealand, Tasmania, Australia).

-It therefore seeks likely that the increase in rBC deposition recorded in the JRI core after the 13th century reflects enhanced BC emissions from one or more of these SH regions, and at least some of these emissions were likely due to human deforestation in the said regions. This would be consistent with wide-scale dispersion of BC by the circum-Antarctic westerlies.

However, I believe that the assignment of Maori-induced fires from New Zealand as the main BC emission source, at least in the 13-16th part of the JRI record, is much more dubious, and I am skeptical of this claim, for reasons explained below. Chiefly, I think the authors are far too confident on the inferences based on FLEXPART simulations, and over-interpret their results while underestimating the uncertainties. I am also unconvinced by their arguments based on the charcoal records from source regions, which seem oddly selective (see comment **7** below). I can not comment, however, on the reliability of the inferred pyrogeography implications (e.g., prehistorical Maori deforestation activities).Based on those aspects of the paper I examined closely, I recommend that the authors either provide more convincing evidence that their rBC geographical source identification is sound. At the very least, a more realistic and explicit presentation of uncertainties needs to be offered. Failing that, the main conclusions need to be "toned-down" and be much more conservative.

Specific criticisms:

1) Concerning ice core analyses: The ice-core analysis methods appear to be sound, and were based on state-of-the-art techniques developed and applied in similar studies before by the lead author (McConnell). The depth-age model used to establish the ice-core chronology is explained in detail and is well-constrained by many time markers (such as volcanic tephra). The JRI site experiences modest amounts of summertime surface melt, which can lead to post-depositional effects (e.g., reduced sensitivity in rBC detection) but these are unlikely to have had a major effect of the decadal- to centennial-trends in rBC concentrations (or fluxes) discussed in this paper. Therefore I regard the JRI ice-core record of rBC deposition as likely reliable for documenting past trends in atmospheric rBC deposition at these time scales. However the possible effects of changing snow accumulation rates at this site are not discussed, and should be.

The JRI ice-core record of rBC deposition is presented in terms of depositional fluxes (□g m^{-2} y^{-1}), obtained by multiplying the measured rBC concentrations in the ice core by the estimated annual ice accumulation (L394-396 in the paper). This implies that the temporal variability of rBC flux

depends both on past temporal variations in rBC concentrations in snowfall, and in the snow accumulation rates. The authors should show, either in the manuscript itself or in the supporting material, the records of rBC concentrations and flux *separately*, so that the reader can judge to what extent the observed flux variations could have been caused by changes in snow accumulation. The plot of the modeled depth-age relationship (Extended Data Figure 6) suggests that there were no major shifts in snow accumulation during the period of interest, but it is difficult to be sure at the scale of this figure. The JRI ice-core □D (deuterium) paleo-temperature record of the past ~500 years actually shows considerable century-scale variability (Mulvaney et al., 2012; their Fig. 4a) and a paleo-accumulation reconstruction based on it suggests that a steep, but non-monotonic rise in snow accumulation occurred at JRI over the past few centuries (Martín et al., 2015). Is there any evidence for this in the JRI accumulation record used to compute rBC fluxes ? If so, how would it affect the interpretation of the rBC flux trends with respect to BC emissions ?

2) The "emission sensitivities" derived from the FLEXPART backward simulations express the source-receptor relationships calculated by the model between gridded historical BC emissions on the one hand (GFED and CEDS inventories) and the histories of rBC deposition recorded in Antarctic ice cores on the other hand. These emission sensitivities, and how they differ between sectors of the Antarctic (e.g., Peninsula vs. East Antarctic Plateau) are used by the authors to support their thesis that New Zealand BB dominated the BC emissions that reached JRI, and also to support their estimates of rBC contributions from New Zeeland BB emissions. Because they are so central to the argumentation, it is especially important to explain clearly how these BC emission sensitivities are derived, and how uncertain they may be. In this respect, I find the manuscript deficient.

It is of course legitimate to employ a model such as FLEXPART to explore potential source- area emission sensitivities for long-range aerosol transport to a receptor site. However I question whether emission sensitivities obtained from backward transport simulations are truly reliable for estimating BC deposition at a receptor site in Antarctica when the transport distances between emission sources and receptor may be >7000 km (New Zealand to the Antarctic Peninsula). Eckhardt et al. (2017), compared backward and forward FLEXPART simulations of BC transport(deposition) from mainland Europe to Svalbard. Results showed that in ~60 % of cases, the difference between forward and backward simulations of BC dry deposition in Svalbard exceeded 20 %, and for wet deposition, the difference was >20 % in ~40 % of cases (Table 3 in Eckhardt et al., 2017). I would expect the uncertainties to be much larger for simulations in the circum-Antarctic region, where source to receptor transport distances are even larger, and the reliability of gridded BC emission data and meteorological fields used in model simulations are considerably lesser than for the North Atlantic region. An experiment comparable to that in Eckhardt et al. (2017) needs first to be performed for the long-range transport of BC to Antarctica, and the findings of that experiment could then be used to better constrain the uncertainties in the present paper. So far as I can tell from the description of methods, this has not yet been done (the forward simulations were only used to simulate expected rBC deposition, but were not compared with backward simulations).

3) The period for which emission sensitivity for *fire-emitted* BC could be evaluated using FLEXPART must be limited to the period during which gridded historical emissions from fires are available, i.e. 1998-2007 in GFED, since the longer CDES emission database does not include BB emissions (Hoesli et al., 2018, p. 371). So the *fire* emission sensitivities are really based on backward simulations for a 9-year period only. I question whether the geographic distribution of fire emissions during this short period was sufficiently similar to that which might have occurred in pre-historical times to assume that the inferred emission sensitivities for the recent period are a valid proxy for earlier, historical to prehistorical periods. Are emission sensitivities for other, pre-1998 anthropogenic BC emissions assumed to be the same as for BB-emitted BC ? Is that a valid assumption, given that these are quite different emission processes ? Can the authors elaborate on this ?

4) The FLEXPART-inferred emission sensitivities basically express linear relationships between BC emissions in source regions and rBC fluxes at the receptor point (JRI). If atmospheric transport conditions and precipitation rates are assumed to have remain largely unchanged, then the emission sensitivities inferred from backward simulations in the recent past should (as their name implies) largely reflect the influence of BC emissions on rBC deposition, and can be applied or extrapolated back in time, as was done here.

However it is known that the Antarctic Peninsula has been experiencing rapid and steep increases in snow accumulation in the recent decades during which emission sensitivities to fire-emitted BC were evaluated (positive snow accumulation trend > 15 mm y^{-1} per decade since 1957; Medley and Thomas, 2019). The source-receptor relationships obtained by FLEXPART therefore represent some mixed function of both BC emissions and of precipitation variations (which controls wet deposition) during the period of evaluation, and are in fact *emission-precipitation* sensitivities. Given that, can these sensitivities truly be applied to infer past BC emission trends in purported BC source regions, as was done in this paper ? Would the sensitivities have been the same at times in the past when snow accumulation trends on JRI were different (or nil, i.e., no trends) ? And if the source-receptor relationships depend in a complex way on both BC source emissions and precipitation, is

the assumption of linearity supported (a prerequisite for using sensitivities for source contribution calculations) ? These points need to be discussed and clarified.

5) It is unclear to me how the errors (± 1□) on the regional source-specific BB emissions of BC (or the errors on contributions of these emissions to rBC deposition on JRI) were computed. There are multiple sources of uncertainties involved. In the GFED, van der Werf et al. (2017) ascribe a "general" "best-guess uncertainty" of 50 % for total BB carbon emissions (irrespective of emission type, e.g., BC vs CO) but other studies (Randerson et al., 2012; van Vees and van der Werf, 2019) suggest that BB carbon emissions in some regions (e.g., south America) may be severely underestimated due to poor representation of small fire contributions in the relatively coarse (0.5 x 0.5° grid) of GFED.

There are also uncertainties in the regional emission sensitives obtained using FLEXPART. On L121-124, the authors say: "To identify possible source regions (...) to allow for ~50% uncertainty in the simulations." The source of the cited 50 % figure is a previous paper by McConnell et al. (2018) on the prehistorical record of atmospheric Pb deposition in Greenland ice. The 50 % figure was obtained "based on differences between (...) different model tracers evaluated in FLEXPART" (McConnell et al., 2018, p. 5730). The "model tracers" used in that paper were other aerosol species measured in the Greenland ice core, which were correlated with Pb. In the present study, however, no corresponding evaluations of model uncertainties were performed (so far as I can tell from the description of methods), and it is questionable whether the uncertainty estimate from McConnell et al. (2018) can be applied to aerosol transport around the circum-Antarctic region. The authors acknowledged this (L445-446) but still used the 50 % figure.

When calculating contributions of BB emissions from any given fire region to rBC deposition at the receptor point (in this case: JRI), the gridded emissions of BC are multiplied by the corresponding emission sensitivity coefficients, so the errors on both terms should be compounded. i.e., the total uncertainty on the estimated deposition should reflect the uncertainties in both the BB emissions and the emission sensitivity coefficients. Was this, in fact, accounted for ? The text is unclear in that regard. Also, do the emission sensitivity coefficients *themselves* account for the uncertainty in the BB emissions. One way to find out would be to perform a Monte Carlo simulation in which historical BB emissions from a specific fire region (e.g., New Zealand) over a period of a few years would be allowed to vary within the range of uncertainties of the GFED database (**±** 50 %), and then carry out forward dispersion simulations with FLEXPART to find out how much the inferred emission sensitivity at the receptor point (JRI) varies for each iteration. I don't believe that this has been done, such that at present, the uncertainties of the regional BB emission sensitivities may in fact be seriously underestimated. I recognize, however, that this is computationally much more intensive to carry out than a receptor-based backward simulation (which is what makes the latter method appealing).

Comments 2 to 5 above are not meant to criticise the use of FLEXPART (or in fact any other atmospheric transport model) in supporting ice-core interpretation. This is indeed a powerful tool for this line of research. However based on my own reading, the uncertainties associated with the method when applied to this region (circum-Antarctic) are probably much larger than the authors acknowledge (or at least they are insufficiently explicit), and consequently I remain very skeptical of their assignment of New Zealand BB emissions as the predominant source of the rBC deposited on JRI between the 13th and 16th centuries (and possibly later). I think the authors express an exaggerated confidence in this inference, given the many large source if uncertainties involved. More work is needed to make the case more convincing.

6) I find it disturbing that no mention at all is made of the work of Pereira and others, who, starting in the 1990s, documented on many occasions the atmospheric transport of gases and aerosols from Patagonia to the northern Antarctic Peninsula (^{222}Rn, mineral dust, BC; Pereira et al., (2006, and references therein). These studies provide very strong evidence that the Peninsula is affected by emissions from low latitudes in South America. In contrast, not a single case study, to my knowledge, has shown direct evidence of atmospheric aerosol transport from New Zealand to the Antarctic Peninsula (in fact, none has shown evidence of transport from New Zealand to the Patagonia either). The authors' contention that New Zealand (>7000 km away) is the predominant source of pre-historical rBC deposition on JRI therefore rests largely on the FLEXPART simulations. To a lesser extent, it rests also on comparisons of the JRI ice-core rBC record with charcoal records in fire source regions, but here also, there are issues, as I explain below.

7) L151 and Figure 1: *"Paleofire records from Patagonia and Tasmania (Fig. 1) ...*". Panel f on Figure 1 presents charcoal records from two sites, one from Tasmania (Lake Vera; Fletcher et al. 2018), and one from Patagonia (Lago Cipreses; Moreno et al., 2014). Both of these records show a conspicuous "dip" in charcoal abundance after ~1300-1500 CE, which is the opposite of what the JRI record or BC deposition shows, and this is one of the lines of evidence used by the authors to argue that New Zealand, rather than these other regions (Patagonia or Tasmania), was the primary source of BC emissions at least until the 16th century (L208-211). But why were these two records (Lake Vera and Lago Cipreses) selected ? Are the trends in these records truly representative of paleofire histories for the whole of Tasmania and Patagonia ? Considering the large spatial noise that exist in lake sediment charcoal records within any given region, it would make more sense to present standardized, composite records of charcoal abundance from multiple sites in each region, as was done from New Zealand (panel e on Fig. 1). Consider for example the composite, charcoal-based paleofire record for Tasmania shown on Fig. 4a of Mariani and Fletcher (2016). This shows a very different picture than the single Lake Vera record: It shows a sharp increase in paleofire activity starting 500 years ago, and a continued rise thereafter.
Based on that composite record, Tasmania could have just as easily been the main source of BC emissions in the 16th century. One gets the unfortunate (and hopefully unintended) impression that the Lake Vera and Lago Cipreses records shown on Fig. 1 on the present paper were cherry-picked to support the authors' thesis. Some justification must be provided as to why these two records were the only one selected for comparison. Even better would to present *all* available charcoal records from each potential source region, and their standardized composite, as was done on panel e of Figure 1 for the New Zealand fire histories.

8) L212-215: *"The evolution of natural and anthropogenic BB poleward of 40°S suggested by charcoal records is substantially different than the rBC concentration and deposition history recorded in the Antarctic ice cores, illustrating the challenges of inferring atmospheric BB emissions from local paleofire records."*

I recommend removing this statement entirely. Given the uncertainties in the rBC source- region assignment methods used in the paper (cf my earlier comments), one could just as well conclude that the study illustrates the challenges of inferring regional atmospheric BB emissions from a single ice core depositional record which may receive BC from multiple regional sources.

References cited *(other than those cited in the ms itself)*

Martín et al. (2015) *Clim. Past* 11, 547–557, doi:10.5194/cp-11-547-2015.
McConnell et al. (2018) *PNAS* 115, doi:10.1073/pnas.1721818115
Mariani and Fletcher (2016) doi:10.1002/2016GL068082
Medley and Thomas (2019) *Geophys. Res. Lett.* 43, doi:10.1002/2016GL068082.
Pereira, E. et al. (2006). *J. Geophys. Res.*, 111, doi:10.1029/2005JD006086.

Randerson et al. (2012) *J. Geophys. Res.* 117, G04012, doi:10.1029/2012JG002128
van der Werf et al. (2017) *Earth Syst. Sci. Data* 9; doi:10.5194/essd-9-697-2017

van Vees & van der Werf (2019) *Geosci. Model Dev.*, 12, doi:10.5194/gmd-12-4681-2019
Wan et al. (2021). *Geophys. Res. Lett.* 48, e2020GL089758; doi:10.1029/2020GL089758

Author Rebuttals to Initial Comments:

Referees' comments (Initial peer review):

Referee #1 (Initial peer review):

I was asked to review this paper specifically from the viewpoint of an expert in Polynesian archaeology, and my comments are limited to that perspective. I do not see anything in the paper that contradicts current views on the chronology of Polynesian settlement of New Zealand; the authors have cited appropriate current literature on that topic. Their conclusions regarding anthropogenic burning in New Zealand following Polynesian arrival in the 13th century are entirely consistent with recent archaeological and paleoecological studies. Indeed, this paper adds significant new evidence for the large-scale regional impact of such burning.

We thank the Referee for the positive and supportive comments. We very much hope that the well-constrained timing and evolution of large-scale Māori burning provided by our ice core records and interpretation will be of use to the archaeological community.

Referee #2 (Initial peer review):

This article shows ice core evidence for an increase in BC deposition following the Māori settlement of New Zealand. The authors use a combination of models and ice core data to show that the most likely hypothesis is that the change is from biomass burning in New Zealand.

Overall, I think this is an excellent paper, that is really important, and thus worthy of being published in Nature because of how interesting and synthetic it is...

We thank the Referee for the positive and supportive comments and address their specific points below.

I do have a couple minor points:

1. using the authors methodology, they should be able to provide statistical analysis of the odds that it is from the biomass burning in New Zealand.

We are hesitant to provide numerical odds since there are two fundamental steps in our attribution of the enhanced rBC fallout in the northern Antarctic Peninsula (nAP) after the late 13th C to Māori-related burning in New Zealand. Only the first – identifying rBC emissions in regions poleward of 40^{o}S as the source of enhanced burning after the late 13th C – is based entirely on objective ice-core measurements and FLEXPART modeling. We are quite confident in the conclusions from this step as discussed in the manuscript text. The second identifying anthropogenic burning in New Zealand rather than climate-modulated burning in Tasmania or southern Patagonia as the source is more subjective since it relies on local biomass burning proxies that are less precisely dated and not as spatially representative. We emphasize, however, that our interpretation is entirely consistent with a substantial body of published paleofire and paleoclimate research on the histories and drivers of biomass burning in Tasmania, New Zealand, and Patagonia during the Common Era.

2. the authors should put this paper in context: there are other papers arguing that European expansion changed biomass burning, and this paper supports that idea: people change the fire regimes! these arguments are really important for understanding what the fires were in preindustrial times. Please cite and put into the context of: Hamilton et al., 2018: Nature Communications.

While we agree that European expansion in the higher mid-latitude Southern Hemisphere very likely changed biomass burning patterns, the focus of our manuscript is primarily on the Common Era prior to European arrival and the effects of Māori burning in New Zealand starting in the late 13th C. Similarities in relative transport and deposition of rBC aerosols from Tasmania, New Zealand, and Patagonia (poleward of ~40^{o}S) across the Southern Ocean to Antarctica mean that it is difficult to distinguish between burning emissions from these three regions using just the array of ice core records and transport modeling. This is why we included local lake-sediment records (and references to published studies of climate and drivers of biomass burning prior to European arrival) from all three regions. These show lower climate-modulated burning in Tasmania and Patagonia after the 13th century and higher anthropogenic burning in New Zealand. Although we are unable to attribute increased burning emissions specifically to one of the three locations, we are confident that changes in burning after European arrival poleward of ~40^{o}S included substantial emissions from Tasmania,

New Zealand, and/or Patagonia based on the spatial pattern of rBC deposition and associated aerosol transport modeling.

We added the Hamilton et al., 2018 citation but have no available space within the length limits (the Editor asked us to reduce the original manuscript by >300 words) to discuss in any detail changes or uncertainties in preindustrial biomass burning after European colonization.

Referee #3 (Initial peer review):

Ice cores drilled from ice sheets and glaciers are ideal material to reconstruct the emissions from natural wildfires and human activities, as they directly preserve the information of atmospheric aerosol transport and deposition. In this study, McConnell and his colleagues used a broad Antarctic array of well-dated, 2000-year records from ice cores and modeling to investigate atmospheric black carbon concentrations over the northern Antarctic Peninsula (nAP) and much of continental Antarctica. The results showed the rBC concentrations and deposition rates were approximately stable over the continental Antarctica during the Common Era, while they presented a 2- to 3-fold increase over the nAP during the past 700 years. Meanwhile, by using FLEXPART simulations and sediment-charcoal records, McConnell and his colleagues demonstrated the main source of the rBC increase over nAP was from New Zealand after the Māori settlement. The BC emissions from biomass burning by Māori was quantified.

New Zealand was the last major landmass to be settled before the industrial age. The precise date of first colonization has been discussed. Previous studies use several methods to determine the date of Māori migration, including the radiocarbon dating of short live samples (e.g., Wilmshurst et al., 2008, 2011), archaeological signature (e.g., Higham et al. 1999; Walter et al., 2017), charcoal records (e.g., McWethy et al., 2010), the extinction of moa (e.g., Holdaway et al., 2014). This study provides a new method to study the precise time of Māori settlement. McConnell and his colleagues suggest that the date of Māori settlement is 1297 (Å}30), which is consistent with previous studies.

In my opinion the results presented are of immediate interest to many people in study areas of environmental chemistry, climatology, and paleoclimatology.

We thank the Referee for the positive and supportive comments and address their specific points below.

Most of my research is about the transport and deposition of anthropogenic aerosols using ice core records and in situ observations. In this review, I will focus on the biomass burning records in the Antarctic ice core array.

The timing and magnitude of the biomass burning increase of rBC are very likely robust. However, the following issues may need being considered.

1. BC emissions from Māori BB were estimated to be 38 (±19) GgrBC/y. This value was used in the forward simulations. The results showed that the BC from New Zealand, as shown in Figure 3, deposited largely over the Pacific Ocean, and substantial over the Atlantic and Indian Ocean sectors of the Southern Ocean. This is very likely true. However, what should be noticed is the depositions over the Patagonia region are considerably large as well. As shown in Figure 3, the deposition value in the Patagonia region was even higher than nAP. If Māori settlement and consequently widespread biomass burning since 1297 (±30) resulted in the rBC deposition 2- to 3-fold higher at nAP, then the charcoal records in the Patagonia should have increased during the past 700 years as well. However, the Patagonia charcoal records did not show this pattern, but presented an evident decrease. Reduced natural biomass burning by wet climate cannot fully explain the decrease.

Charcoal in lake sediments is an indicator of fire occurrence within the watershed or perhaps a nearby watershed and, because of its relatively large size (see Adolf et al., Global Ecol Biogeogr,

2018), cannot be transported across 7000 km of the South Pacific from New Zealand. rBC aerosols are orders of magnitude smaller than charcoal and so can be transported much farther through the air. In theory it would be possible to detect the Māori-related changes in rBC in ice-core records from this region of southern Patagonia, although emission sensitivity to nearby burning would be many orders of magnitude greater than to emissions from New Zealand 7000 km away, so even relatively small emissions from southern Patagonia probably would dominate any records from southern Patagonia. In addition, to our knowledge there are no ice core records spanning the past 1000+ years from southern Patagonia and the well-known Sajama and Illimani ice cores were collected at drilling sites in Bolivia that are well north of the simulated Māori burning plume and heavily influenced by Amazon burning emissions.

Most or all recent publications of the paleo-climate proxies (tree-ring, lake-sediment pollen), paleo-fire proxies (lake-sediment charcoal, fire-scarred trees) (e.g., Moreno et al., 2014; Fletcher et al., 2018; Mariani and Fletcher, 2016; Mundo et al., 2017) and related modeling for Patagonia and Tasmania (e.g., Quintana and Aceituno, 2012; Mundo et al, 2017) attribute reduced biomass burning after the ~14th C. to wetter conditions associated with weaker Southern Westerly Winds (SWW) characterized by a negative Southern Annular Mode (SAM). Because we are not aware of any publications supporting the statement that "Reduced natural biomass burning by wet climate cannot fully explain the decrease," we are unable to address this comment.

2. All the charcoal records, including New Zealand charcoal (Figure 1 e and f in the manuscript), showed low values during the 16th C. This pattern also can be seen in the ice records from the continental Antarctica. But the rBC depositions over the nAP showed a linear increase during the 14th C to 16th C.

One of the larger conclusions of our study (see the final paragraph of the main text) is that deriving atmospheric rBC emissions from lake-sediment charcoal records alone is challenging and highly uncertain. Lake-sediment records provide reliable information on the occurrence and timing of burning but not the magnitude since they reflect only local burning within the watershed (or perhaps nearby watersheds) and the amount of charcoal deposited depends heavily on where burning occurred relative to the lake and coring site. Conversely, the Antarctic ice-core records directly document changes in atmospheric rBC near the drilling sites. Since our coring sites are located 1000s of kilometers from potential sources, the records reflect large-scale rBC emissions, and therefore are representative of atmospheric concentrations over vast regions of the South Pacific, Southern Ocean, and Antarctica.

3. A simple question: When using the FLEXPART atmospheric aerosol and deposition simulation, the rBC aerosol was set to be released over a grid from 37 to 45°S and 170 to 180°E, while the most west of New Zealand is 166°E. Why not cover all land of New Zealand?

Many thanks for catching this. We agree that the original definition of the release over New Zealand was not chosen ideally, so we redid the forward FLEXPART simulation using releases from three boxes instead of one (Fig. R1). This captures the shape of New Zealand much better (upper panel in the figure below) than the original one-box release (lower panel in figure below). The simulation results are quite similar, so there are no changes in our overall conclusions. Fig. 3 has been replaced using results of the new simulation and Methods related to the forward modelling updated accordingly.

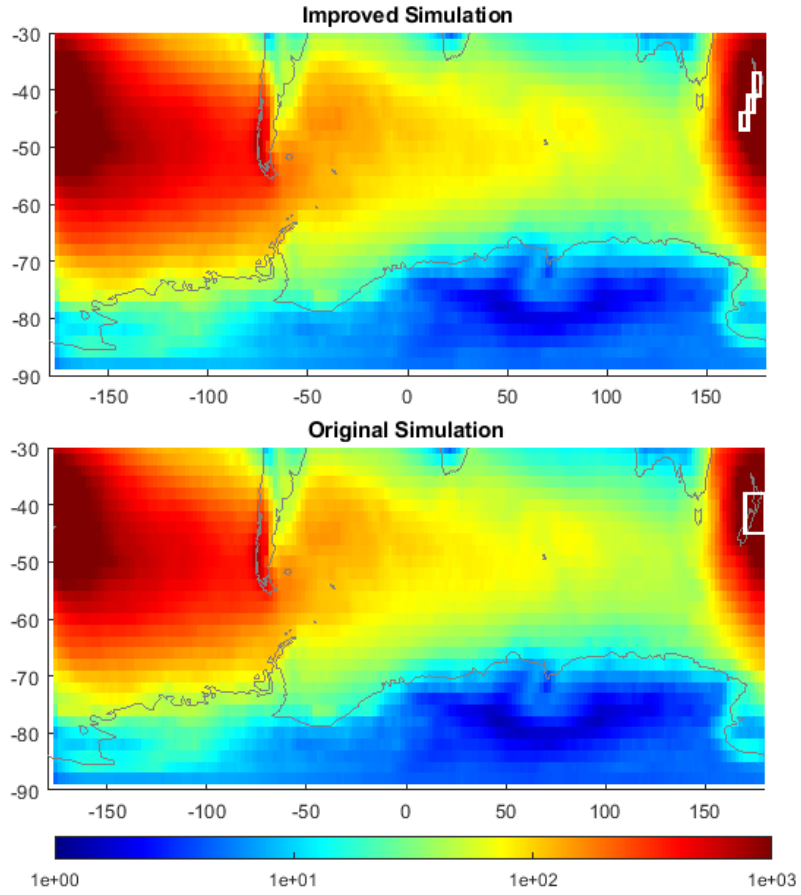

Fig. R1: FLEXPART forward simulations of total annual deposition (µg/m2/y) with a constant release rate for the revised release region (upper panel) and the original release region (lower panel). The total annual amount of rBC released was the same in both simulations.

Referee #4 (Initial peer review):

Overview:

This manuscript presents a new, 2000-year long ice-core record of refractory black carbon deposition (rBC, i.e. "soot") from the atmosphere in snow on James Ross Island (JRI) at the tip on the Antarctic Peninsula. The authors use this record, in combination with atmospheric aerosol transport simulations (FLEXPART model) to offer an interpretation of the most probable geographical sources of rBC deposited on JRI in past centuries. One of the main inferences made (reflected in the manuscript title) is that at least from the 13th to the 16th century, biomass burning (BB) emissions associated with prehistorical deforestation in New Zealand caused widespread hemispheric BC dispersion in the circum-Antarctic atmosphere and were chiefly responsible for the rBC deposition trends documented in the JRI ice core.

Estimates of the possible magnitude of these BC emissions are presented, based also on FLEXPART simulations, constrained by the ice-core data.

Relevance and significance:

A recent assessment of the uncertainties in estimating aerosol radiative forcing of climate pointed out that "A major component of the variance in preindustrial natural aerosol burden is due to uncertainties in the spatial distribution and total magnitude of fire emissions. A lack of understanding about human influence on fires over the historical period results in a large range of possible land use and land cover change scenarios associated with preindustrial fire estimates." (Wan et al., 2021)

The present manuscript is timely and of great interest precisely because it provides new information on the preindustrial (and more recent) burden of BC aerosols in the southern hemisphere, and of the possible role played by past land use changes (deforestation) and fires in BC emissions. Therefore I think this paper is certainly of great interest. Furthermore it bridges across several disciplines (pyrogeography, biogeography, climate and atmospheric science, glaciology), making it interesting to a broad audience. Much of the work presented in the paper is based on state-of-the-art methods (not truly novel, but sound) and seems very robust.

We thank the Referee for the positive and supportive comments and address their specific points below.

However, I think the authors over-reach their interpretation and are more confident in some of their main findings than is warranted. Specifically, I think that the following inferences in the paper are well-supported:

-There is clear evidence of an abrupt increase in rBC deposition at JRI starting in the 13th century, and sustained thereafter. This contrasts with ice-core records of rBC from the East Antarctic interior, which show no such increases.

- FLEXPART model simulation results show that unlike the interior regions of Antarctica, the northern part of the Peninsula is much more sensitive to BC emissions from low to midlatitude southern hemisphere (SH) regions (especially those in South America, New Zealand, Tasmania, Australia).

-It therefore seeks likely that the increase in rBC deposition recorded in the JRI core after the 13th century reflects enhanced BC emissions from one or more of these SH regions, and at least some of these emissions were likely due to human deforestation in the said regions. This would be consistent with wide-scale dispersion of BC by the circum-Antarctic westerlies.

However, I believe that the assignment of Māori-induced fires from New Zealand as the main BC emission source, at least in the 13-16th part of the JRI record, is much more dubious, and I am skeptical of this claim, for reasons explained below. Chiefly, I think the authors are far too confident on the inferences based on FLEXPART simulations, and over-interpret their results while underestimating the uncertainties. I am also unconvinced by their arguments based on the charcoal records from source regions, which seem oddly selective (see comment 7 below).

I cannot comment, however, on the reliability of the inferred pyrogeography implications (e.g., prehistorical Māori deforestation activities). Based on those aspects of the paper I examined closely, I recommend that the authors either provide more convincing evidence that their rBC geographical source identification is sound. At the very least, a more realistic and explicit presentation of uncertainties needs to be offered. Failing that, the main conclusions need to be "toned-down" and be much more conservative.

Please see below. We emphasize that our findings are based on wide array of well-dated and synchronized ice core records spanning nearly 2000 kms in the north-south direction from 64 ^{o}S to 82 ^{o}S. As the Referee points out, aerosol transport and deposition is very different in the northern Antarctic Peninsula (nAP) and continental Antarctica leading to very different source regions and it is these differences that underpin our findings. Our identification of potential source regions (i.e., Tasmania, New Zealand, southern Patagonia) is based largely on how and when very significant changes in relative rBC deposition (i.e., ratios) occurred between the nAP and continental Antarctic sites as documented in the ice core array. We also emphasize that our characterization of substantially lower biomass burning in Tasmania and southern Patagonia associated with changes in the Southern Annual Mode after the late 13th C (illustrated with local fire proxies from the three regions in Fig. 1) is entirely consistent with prevailing views and underpinned by a number of published paleofire and paleoclimate studies.

Specific criticisms:

1) Concerning ice core analyses: The ice-core analysis methods appear to be sound, and were based on state-of-the-art techniques developed and applied in similar studies before by the lead author (McConnell). The depth-age model used to establish the ice-core chronology is explained in detail and is well-constrained by many time markers (such as volcanic tephra).

The JRI site experiences modest amounts of summertime surface melt, which can lead to post-depositional effects (e.g., reduced sensitivity in rBC detection) but these are unlikely to have had a major effect of the decadal- to centennial-trends in rBC concentrations (or fluxes) discussed in this paper. Therefore I regard the JRI ice-core record of rBC deposition as likely reliable for documenting past trends in atmospheric rBC deposition at these time scales. However the possible effects of changing snow accumulation rates at this site are not discussed, and should be.

The JRI ice-core record of rBC deposition is presented in terms of depositional fluxes (µg m-2 y-1), obtained by multiplying the measured rBC concentrations in the ice core by the estimated annual ice accumulation (L394-396 in the paper). This implies that the temporal variability of rBC flux depends both on past temporal variations in rBC concentrations in snowfall, and in the snow accumulation rates. The authors should show, either in the manuscript itself or in the supporting material, the records of rBC concentrations and flux separately, so that the reader can judge to what extent the observed flux variations could have been caused by changes in snow accumulation. The plot of the modeled depth-age relationship (Extended Data Figure 6) suggests that there were no major shifts in snow accumulation during the period of interest, but it is difficult to be sure at the scale of this figure. The JRI ice-core δD (deuterium) paleo-temperature record of the past ~500 years actually shows considerable century-scale variability (Mulvaney et al., 2012; their Fig. 4a) and a paleo-accumulation reconstruction based on it suggests that a steep, but non-monotonic rise in snow accumulation occurred at JRI over the past few centuries (Martín et al., 2015). Is there any evidence for this in the JRI accumulation record used to compute rBC fluxes ? If so, how would it affect the interpretation of the rBC flux trends with respect to BC emissions ?

We converted from measured rBC concentrations to rBC fluxes using well-established methods and feel that our discussion in Methods of (1) the potential effects of changing snow accumulation rates including a detailed discussion of the net accumulation record at James Ross Island and (2) how we converted from rBC concentration to rBC depositional flux is sufficient. To accommodate the Referee's request we have added a panel to Extended Data Figure 1 showing rBC concentrations at all the ice cores sites. Comparison of the concentration and flux panels demonstrates that the late 13th century rBC increase in the northern Antarctic Peninsula is robust (i.e., it is clear in both the concentration and the flux records and so definitely is not the result of changes in snow accumulation rate at James Ross Island). We also have added a plot of the net accumulation rates at James Ross Island from the constrained ice flow modeling to Extended Data Figure 6. A discussion of the net accumulation rates and the less than ±10% variation over the Common Era in James Ross Island record already is included in Methods but we have added additional text to Methods discussing accumulation rate variability at the continental sites (also generally less than ±10%) to further accommodate the Referee's concerns about the effects of changing accumulation rates.

Please also note that the JRI1 age scale used in Mulvaney et al., 2012 has been superseded by our new age scale that is based on (1) new, far more extensive continuous chemical and elemental measurements of the same ice core that enabled annual layer counting throughout the 2nd millennium and (2) a new definitive tephra chronological marker at ~3568 yBP that constrains flow modeling in the deeper record far better than previously. This already is described in Methods and illustrated in the Extended Data figures.

2) The "emission sensitivities" derived from the FLEXPART backward simulations express the source-receptor relationships calculated by the model between gridded historical BC emissions on the one hand (GFED and CEDS inventories) and the histories of rBC deposition recorded in Antarctic ice cores on the other hand. These emission sensitivities, and how they differ between sectors of the Antarctic (e.g., Peninsula vs. East Antarctic Plateau) are used by the authors to support their thesis that New Zealand BB dominated the BC emissions that reached JRI, and also to support their estimates of rBC contributions from New Zeeland BB emissions. Because they are so central to the argumentation, it

is especially important to explain clearly how these BC emission sensitivities are derived, and how uncertain they may be. In this respect, I find the manuscript deficient.

It is of course legitimate to employ a model such as FLEXPART to explore potential source area emission sensitivities for long-range aerosol transport to a receptor site. However I question whether emission sensitivities obtained from backward transport simulations are truly reliable for estimating BC deposition at a receptor site in Antarctica when the transport distances between emission sources and receptor may be >7000 km (New Zealand to the Antarctic Peninsula). Eckhardt et al. (2017), compared backward and forward FLEXPART simulations of BC transport(deposition) from mainland Europe to Svalbard. Results showed that in ~60% of cases, the difference between forward and backward simulations of BC dry deposition in Svalbard exceeded 20%, and for wet deposition, the difference was >20 % in ~40% of cases (Table 3 in Eckhardt et al., 2017). I would expect the uncertainties to be much larger for simulations in the circum-Antarctic region, where source to receptor transport distances are even larger, and the reliability of gridded BC emission data and meteorological fields used in model simulations are considerably lesser than for the North Atlantic region. An experiment comparable to that in Eckhardt et al. (2017) needs first to be performed for the long-range transport of BC to Antarctica, and the findings of that experiment could then be used to better constrain the uncertainties in the present paper. So far as I can tell from the description of methods, this has not yet been done (the forward simulations were only used to simulate expected rBC deposition, but were not compared with backward simulations).

FLEXPART is a very widely used aerosol transport and deposition model; describing the underlying mechanics is beyond the scope of this manuscript and the necessary citations already have been provided in the text. We emphasize that gridded historical or other emissions fields are not used in FLEXPART to calculate the emission sensitivities presented in this study.

The differences in Eckhardt et al. (2017) are quite typical for backward-forward comparison when considering short-time averages on the order of a few hours (see also Seibert and Frank, 2004). They result from small numerical differences in the interpolation of meteorological data (especially the wind) and the numerical integration of the trajectory equation. One needs to consider two important factors in the context of our current study: Firstly, the values reported in Eckhardt et al. (2017) are for hourly and 3-hourly deposition values. These short time intervals were chosen to highlight and study the backward-forward differences in detail. However, these differences are random and there is no obvious bias and little time-correlation between forward and backward simulations. For instance, Fig. 5 in Eckhardt et al. (2017) shows the 3-hourly differences for one full month. While the individual 3-hourly values deviate substantially, the averages for the full month from backward and forward simulations were already much smaller (~13%). In the present study, we calculated monthly values but these were further averaged over an 80-year period. Our interpretation is only based on these 80-year averages, and for these the errors should, assuming purely random errors, only be ~0.4%. Secondly, the differences shown in Eckhardt et al. (2017) are absolutely no indication that backward calculations are less accurate than forward calculations. They show that both forward and backward calculations of atmospheric transport (for short-time averages) are very sensitive to integrating the trajectory equation, and small errors in interpolated winds and numerical integration can lead to substantial random errors. This is long known and true for all Lagrangian models. We have also no indication that these relative differences grow with

transport distance (see also Stohl et al., 1995, who showed relative trajectory errors to be rather constant over a large range of transport time scales).

It is also important to notice that numerical diffusion in Eulerian models leads to much larger errors for long-range atmospheric transport simulations, including systematic biases (see, e.g., Rastigejev et al., 2010, and several follow-up papers)! Thus, our Lagrangian approach is particularly sensible for the very long transport distances considered in our study and should be superior to Eulerian simulations of atmospheric transport.

The referee is correct in pointing out that source attribution is less specific when involving very long transport distances. It is, however, important to note that numerical diffusion in Eulerian models leads to much larger errors for long-range atmospheric transport simulations, including systematic biases (see, e.g., Rastigejev et al., 2010, and several follow-up papers). Thus, our Lagrangian approach is particularly sensible for the very long transport distances considered in our study and should perform better than Eulerian simulations of atmospheric transport.

We also emphasize that we are careful not to over-interpret our model results. Our results indicate that rBC emissions from southern Patagonia, Tasmania and New Zealand could plausibly explain the observed increase in rBC deposition at JRI while maintaining the observed depositional ratios between JRI, nAP, and iEAP. We then discriminate between these potential regions by using charcoal records, which clearly suggest that New Zealand is the most likely source region of rBC..

In summary, previous work to explore the backward-forward FLEXPART model differences in great detail confirms that our application of FLEXPART is appropriate and has negligible errors for this study.

Rastigejev, Y., R. Park, M. P. Brenner, and D. J. Jacob (2010), Resolving intercontinental pollution plumes in global models of atmospheric transport, J. Geophys. Res., 115, D02302, doi:10.1029/2009JD012568.

Stohl, A., G. Wotawa, P. Seibert, and H. Kromp-Kolb (1995): Interpolation errors in wind fields as a function of spatial and temporal resolution and their impact on different types of kinematic trajectories. J. Appl. Meteor. 34, 2149-2165.

3) The period for which emission sensitivity for fire-emitted BC could be evaluated using FLEXPART must be limited to the period during which gridded historical emissions from fires are available, i.e. 1998-2007 in GFED, since the longer CDES emission database does not include BB emissions (Hoesli et al., 2018, p. 371). So the fire emission sensitivities are really based on backward simulations for a 9-year period only. I question whether the geographic distribution of fire emissions during this short period was sufficiently similar to that which might have occurred in pre-historical times to assume that the inferred emission sensitivities for the recent period are a valid proxy for earlier, historical to prehistorical periods. Are emission sensitivities for other, pre-1998 anthropogenic BC emissions assumed to be the same as for BB-emitted BC? Is that a valid assumption, given that these are quite different emission processes ? Can the authors elaborate on this?

Again, we reiterate that gridded historical or other emissions fields are not used in the emission sensitivity calculations so past to present changes in spatial patterns of biomass burning do not impact the FLEXPART emissions sensitivities used in this study. However, we did use gridded rBC emissions from the GFED and CEDS datasets to evaluate FLEXPART performance at many of our Antarctic coring sites and that evaluation might be marginally effected by a difference in the spatial pattern of burning.

As described in the manuscript, we used modern gridded GFED (biomass burning) and CEDS (anthropogenic) emissions inventories (in units of kg/s) in conjunction with the gridded FLEXPART emission sensitivities (in units of (ug/m^2/y)/(kg/s)) to evaluate how well FLEXPART predicted deposition (ug/m^2/y) at our ice cores sites. We focused on this period because recent gridded emissions estimates are based on satellite data and other modern measurements, and therefore are the most accurate emissions inventories available even though the recent spatial pattern in burning may be somewhat different than in the distant past.

As discussed in Methods and illustrated in Extended Data Figure 8, the simulated emissions were ~2.6 larger than the observed. However, as the Referee points out in their point 5) and we discuss in Methods, even these modern, satellite-observation-based estimates of biomass burning and BC emissions are highly uncertain (+/-50% or more) so it is impossible to quantify how much of the ~2.6 fold difference between simulated and observed BC deposition is caused by the emission sensitivity errors and how much by GFED and CEDS gridded emissions errors. Using highly uncertain gridded reconstructions of rBC emissions during earlier periods (e.g., van Marle et al., 2017) that might better match the preindustrial burning patterns would result in even more ambiguous comparisons between simulations and observations.

We also emphasize that our source constraint is based on the ratio of emission sensitivities for different ice core sites. While our model, as any other model, does contain biases (e.g., lifetime of BC too short or too long), such systematic differences partly cancel out when taking the ratio of emission sensitivity maps for the different ice cores, as both sites would be affected similarly. Thus, the rBC deposition ratio time series in Fig. 1 and the emission sensitivity ratio plots in Fig. 2 and Extended Data Fig. 2 that were used to constrain potential increased emissions after the late 13th century to Tasmania, New Zealand, and/or Patagonia suffer much less from systematic biases/uncertainties in the model than the emission sensitivities for a specific single site. To reinforce this, we have modified Extended Data Fig. 8 to include a comparison of simulated and observed rBC deposition ratio relative to B40 at the different Antarctic sites to match the ratios presented in Figs. 1 and 2. Results show that the simulated deposition ratios around Antarctica are similar to the observed. Methods has been updated accordingly.

Because we used the magnitude of the FLEXPART emission sensitivity to estimate the New Zealand emissions required to create the observed changes in Antarctic rBC deposition after ~1297, we acknowledge the large uncertainties associated with long range transport to Antarctica in Methods and stipulated a ±60% uncertainty in the required emissions. Note that this has been increased from ±50% to better reflect the evaluation results (factor of ~2.6 overestimation) shown in Extended Data Fig. 8.

4) The FLEXPART-inferred emission sensitivities basically express linear relationships between BC emissions in source regions and rBC fluxes at the receptor point (JRI). If atmospheric transport

conditions and precipitation rates are assumed to have remain largely unchanged, then the emission sensitivities inferred from backward simulations in the recent past should (as their name implies) largely reflect the influence of BC emissions on rBC deposition, and can be applied or extrapolated back in time, as was done here.

However it is known that the Antarctic Peninsula has been experiencing rapid and steep increases in snow accumulation in the recent decades during which emission sensitivities to fire-emitted BC were evaluated (positive snow accumulation trend > 15 mm y-1 per decade since 1957; Medley and Thomas, 2019). The source-receptor relationships obtained by FLEXPART therefore represent some mixed function of both BC emissions and of precipitation variations (which controls wet deposition) during the period of evaluation, and are in fact emission-precipitation sensitivities. Given that, can these sensitivities truly be applied to infer past BC emission trends in purported BC source regions, as was done in this paper ? Would the sensitivities have been the same at times in the past when snow accumulation trends on JRI were different (or nil, i.e., no trends) ? And if the source-receptor relationships depend in a complex way on both BC source emissions and precipitation, is the assumption of linearity supported (a prerequisite for using sensitivities for source contribution calculations) ? These points need to be discussed and clarified.

This is a very good point. If the precipitation (and also circulation) patterns a few hundred years ago were vastly different from what they are today, this could lead to substantial deviations. Unfortunately, the state of the atmosphere a few hundred years ago is unknown and, thus, no re-analyses extend so far back. Precipitation changes at single sites cannot really be used to bracket these uncertainties. However, we do not believe that circulation and precipitation patterns changed so much that the emission sensitivity fields would change as much as to drastically alter our conclusions. To support these assumptions, we have split our re-analysis period into three 25-year sub-periods and calculated emission sensitivities separately for these periods for the JRI ice core (Fig. R2). The emission sensitivity fields look very similar and the differences are everywhere within 50% and smaller than 20% over New Zealand. Some of these differences also are due to the relatively short 25-year periods, where inter-decadal variability is still very important. The differences would thus likely be smaller when comparing longer-term averages such as 80-year periods.

It is also important to note that the source-receptor relationships are totally independent of the emissions. Thus, they do not depend in a complex way on both BC source emissions and precipitation. Changes in source-receptor relationships (e.g. due to precipitation changes) would affect our source estimates linearly.

5) It is unclear to me how the errors (± 1σ) on the regional source-specific BB emissions of BC (or the errors on contributions of these emissions to rBC deposition on JRI) were computed. There are multiple sources of uncertainties involved. In the GFED, van der Werf et al. (2017) ascribe a "general" "best-guess uncertainty" of 50 % for total BB carbon emissions (irrespective of emission

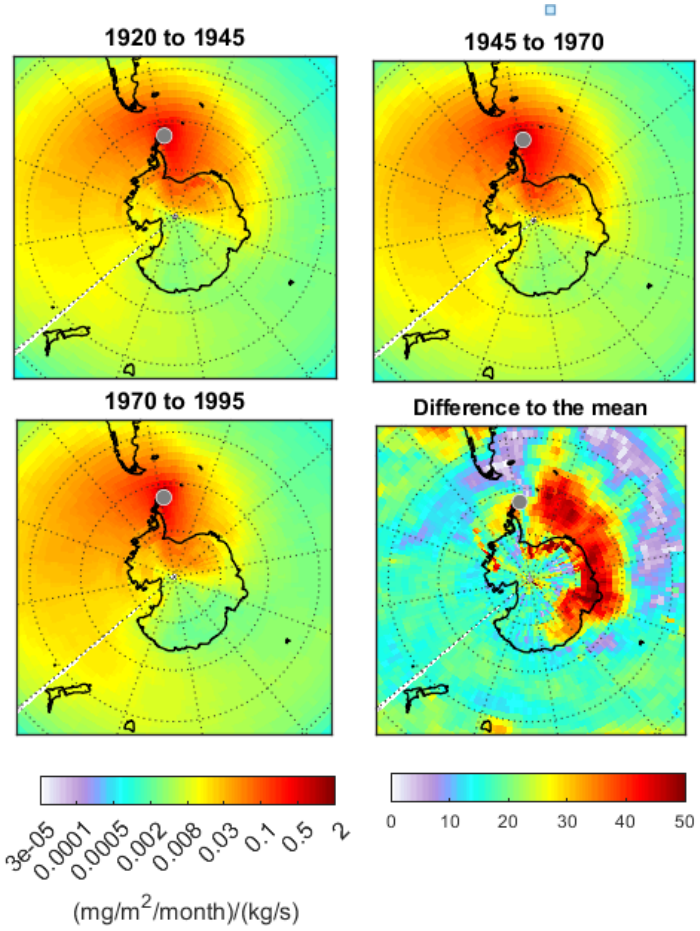

Fig. R2. Emission sensitivities for the JRI ice core for three different periods of 25 years. The last panel shows the maximum percentage difference of the three periods relative to the mean of all footprints.

type, e.g., BC vs CO) but other studies (Randerson et al., 2012; van Vees and van der Werf, 2019) suggest that BB carbon emissions in some regions (e.g., south America) may be severely underestimated due to poor representation of small fire contributions in the relatively coarse (0.5 x 0.5° grid) of GFED.

There are also uncertainties in the regional emission sensitives obtained using FLEXPART. On L121-124, the authors say: "To identify possible source regions (...) to allow for ~50% uncertainty in the simulations." The source of the cited 50 % figure is a previous paper by McConnell et al. (2018) on the prehistorical record of atmospheric Pb deposition in Greenland ice. The 50 % figure was obtained "based on differences between (...) different model tracers evaluated in FLEXPART" (McConnell et al., 2018, p. 5730). The "model tracers" used in that paper were other aerosol species measured in the Greenland ice core, which were correlated with Pb. In the present study, however, no corresponding evaluations of model uncertainties were performed (so far as I can tell from the description of methods), and it is questionable whether the uncertainty estimate from McConnell et al. (2018) can be applied to aerosol transport around the circum-Antarctic region. The authors acknowledged this (L445-446) but still used the 50 % figure.

When calculating contributions of BB emissions from any given fire region to rBC deposition at the receptor point (in this case: JRI), the gridded emissions of BC are multiplied by the corresponding emission sensitivity coefficients, so the errors on both terms should be compounded. i.e., the total uncertainty on the estimated deposition should reflect the uncertainties in both the BB emissions

and the emission sensitivity coefficients. Was this, in fact, accounted for ? The text is unclear in that regard. Also, do the emission sensitivity coefficients themselves account for the uncertainty in the BB emissions. One way to find out would be to perform a Monte Carlo simulation in which historical BB emissions from a specific fire region (e.g., New Zealand) over a period of a few years would be allowed to vary within the range of uncertainties of the GFED database (± 50 %), and then carry out forward dispersion simulations with FLEXPART to find out how much the inferred emission sensitivity at the receptor point (JRI) varies for each iteration. I don't believe that this has been done, such that at present, the uncertainties of the regional BB emission sensitivities may in fact be seriously underestimated. I recognize, however, that this is computationally much more intensive to carry out than a receptor-based backward simulation (which is what makes the latter method appealing).

Comments 2 to 5 above are not meant to criticize the use of FLEXPART (or in fact any other atmospheric transport model) in supporting ice-core interpretation. This is indeed a powerful tool for this line of research. However based on my own reading, the uncertainties associated with the method when applied to this region (circum-Antarctic) are probably much larger than the authors acknowledge (or at least they are insufficiently explicit), and consequently I remain very skeptical of their assignment of New Zealand BB emissions as the predominant source of the rBC deposited on JRI between the 13th and 16th centuries (and possibly later). I think the authors express an exaggerated confidence in this inference, given the many large source if uncertainties involved. More work is needed to make the case more convincing.

We agree with the Referee that uncertainties of the model simulations (which are very hard to quantify for any model) are important to take into account, and that care must be taken when using the model results to interpret the ice core data. It is important not to overinterpret the model results, and therefore we base our arguments on several lines of evidence (e.g., also on charcoal data from the possible source regions). We do not think that the model results are so uncertain that entirely different BC sources must be accounted for. It is important to point out again that no emission information is needed for determining the source-receptor relationships, thus there is no compounding effect where uncertainties in both terms affect our results. It is furthermore of crucial importance to notice that our source constraint is based on the ratio of emission sensitivities for different ice core sites. While our model, as any other model, does contain biases (e.g., lifetime of BC too short or too long), such systematic differences partly cancel out when taking the ratio of emission sensitivity maps for the different ice cores, as both sites would be affected similarly. Thus, the emission sensitivity ratio plots, which were used to attribute the emissions to New Zealand, suffer much less from systematic biases/uncertainties in the model than the emission sensitivities for a specific single site.

As in our response to point 3), we have modified Extended Data Fig. 8 to include a comparison of simulated and observed rBC deposition ratio relative to B40 at the different Antarctic sites to match the ratios presented in Figs. 1 and 2. Results show that the simulated deposition ratios around Antarctica are similar to the observed providing support for the reliability of FLEXPART simulations of rBC transport and deposition over Antarctica. Methods has been updated accordingly.

6) I find it disturbing that no mention at all is made of the work of Pereira and others, who, starting in the 1990s, documented on many occasions the atmospheric transport of gases and aerosols from

Patagonia to the northern Antarctic Peninsula (222Rn, mineral dust, BC; Pereira et al., (2006, and references therein). These studies provide very strong evidence that the Peninsula is affected by emissions from low latitudes in South America. In contrast, not a single case study, to my knowledge, has shown direct evidence of atmospheric aerosol transport from New Zealand to the Antarctic Peninsula (in fact, none has shown evidence of transport from New Zealand to the Patagonia either). The authors' contention that New Zealand (>7000 km away) is the predominant source of pre-historical rBC deposition on JRI therefore rests largely on the FLEXPART simulations. To a lesser extent, it rests also on comparisons of the JRI ice-core rBC record with charcoal records in fire source regions, but here also, there are issues, as I explain below.

We do not posit that transport of gases and aerosols from southern South America to James Ross Island are/were not significant and/or important. In fact, the gridded FLEXPART emission sensitivity map for James Ross Island shown in Fig. 2a and Extended Data Fig. 2a clearly demonstrates that rBC deposition at the site is most sensitive to biomass burning emissions from southern-most Patagonia.

However, our findings pointing to Māori-related burning as the primary source of enhanced rBC emissions after the late 13th century are based on the entire Antarctic ice core array (not just on the James Ross Island records), as well as published burning histories from lake-sediment charcoal and fire-scar tree records (and associated climate modeling studies and understanding). If burning emissions were substantially larger in southern Patagonia, then it would indeed be a major source of rBC deposited over the Peninsula and also would show up in the DML records from continental Antarctic - but rBC deposition declined over DML during this period suggesting that southern Patagonia was not the source of enhanced burning starting in the late 13th C. Note also that this is consistent with preindustrial open burning emission reconstructions (e.g., van Marle, 2017) that show very little burning in southern Patagonia.

To our knowledge, this is the first study to report changes in biomass burning aerosol deposition in the northern Antarctic Peninsula prior to European expansion poleward of 40^{o}S so it is not surprising that transport from New Zealand has not been identified previously. Dust aerosol deposition during the past century at James Ross Island has been attributed to Patagonia (McConnell et al., PNAS, 2007) , but 20th C sources of dust there were much larger than in New Zealand probably as a result of European settlement and sheep grazing in southern-most Patagonia.

7) L151 and Figure 1: "Paleofire records from Patagonia and Tasmania (Fig. 1) ...". Panel f on Figure 1 presents charcoal records from two sites, one from Tasmania (Lake Vera; Fletcher et al. 2018), and one from Patagonia (Lago Cipreses; Moreno et al., 2014). Both of these records show a conspicuous "dip" in charcoal abundance after ~1300-1500 CE, which is the opposite of what the JRI record or BC deposition shows, and this is one of the lines of evidence used by the authors to argue that New Zealand, rather than these other regions (Patagonia or Tasmania), was the primary source of BC emissions at least until the 16th century (L208-211). But why were these two records (Lake Vera and Lago Cipreses) selected ?

Are the trends in these records truly representative of paleofire histories for the whole of Tasmania and Patagonia ? Considering the large spatial noise that exist in lake sediment charcoal records within any given region, it would make more sense to present standardized, composite records of charcoal abundance from multiple sites in each region, as was done from New Zealand (panel e on Fig. 1). Consider for example the composite, charcoal-based paleofire record for Tasmania shown on

Fig. 4a of Mariani and Fletcher (2016). This shows a very different picture than the single Lake Vera record: It shows a sharp increase in paleofire activity starting 500 years ago, and a continued rise thereafter.

Based on that composite record, Tasmania could have just as easily been the main source of BC emissions in the 16th century. One gets the unfortunate (and hopefully unintended) impression that the Lake Vera and Lago Cipreses records shown on Fig. 1 on the present paper were cherry-picked to support the authors' thesis. Some justification must be provided as to why these two records were the only one selected for comparison. Even better would to present all available charcoal records from each potential source region, and their standardized composite, as was done on panel e of Figure 1 for the New Zealand fire histories.

The published Lake Vera and Lago Cipreses charcoal records in Fig. 1 are intended only to visually illustrate the overall pattern of burning in Tasmania and Patagonia described in a number of peer-reviewed publications by paleofire and paleoclimate researchers in the lake sediment, tree ring, and climate modeling communities. These studies argue that pre-European burning in Tasmania and Patagonia was largely climate modulated and linked to large-scale circulation changes in the southern westerly winds (SWW) characterized by the Southern Annular Mode (SAM). From a review of the literature, it is clear that our contention that biomass burning in Tasmania and southern Patagonia is/was modulated by climate and large scale circulation is not controversial. The overall assumption that biomass burning in these regions was much lower after the late 13th C when SAM shifted from strongly positive to strongly negative and remained low through at least the 17th C also is well supported in the literature. In fact, the main conclusion from the Mariani and Fletcher (2016) article referred to by the Referee is exactly that and uses the charcoal record in Fig. 4a and comparisons to tree-based and multiparameter SAM reconstructions to support exactly that contention.

We reiterate that we chose the Lake Vera and Lago Cipreses records only because they are among the most robustly dated, recently published lake sediment records available from these regions that extend over the Common Era and so are comparable to our ice core records and period of study.

To directly address the Referee's comment, we plotted the 1000-year charcoal record referred to from Fig. 4a of Mariani and Fletcher (2016) along with the rBC deposition ratio between the northern Antarctic Peninsula and continental Antarctic sites (Fig. R3). Consistent with our interpretation of the lake-sediment and ice-core records, there is little or no correspondence between the ice-core deposition ratios presented in Fig. 1 of our manuscript and the 1000-year Tasmanian composite charcoal record from Mariani and Fletcher, 2016.

We have modified the Fig. 1 caption to emphasize that these two records are used only to illustrate the general history of climate modulated burning in Tasmania and southern Patagonia documented in many proxy records and in a number of peer-reviewed publications.

8) L212-215: "The evolution of natural and anthropogenic BB poleward of 40°S suggested by charcoal records is substantially different than the rBC concentration and deposition history recorded in the Antarctic ice cores, illustrating the challenges of inferring atmospheric BB emissions from local paleofire records."

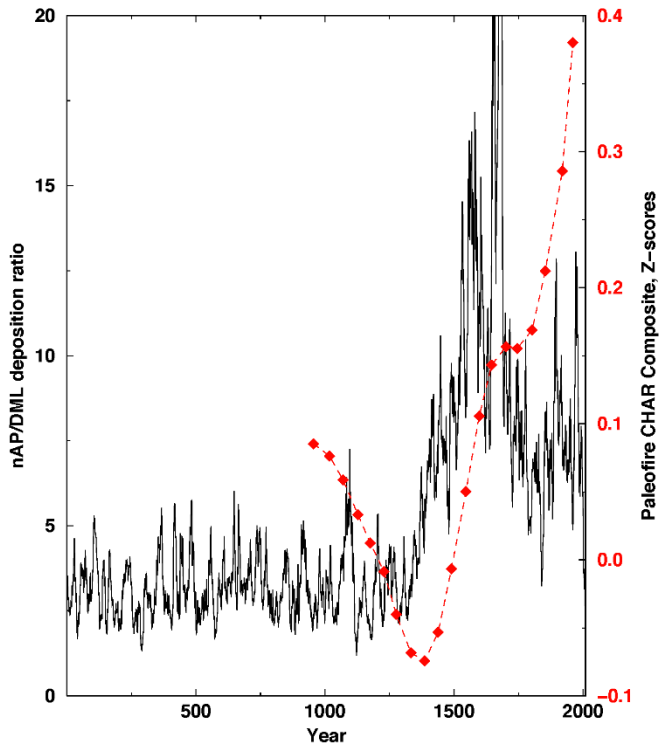

Fig. R3. Comparison of the nAP/DML rBC deposition ratio (from Fig. 1d of our manuscript) compared to the CHAR Composite from Mariani and Fletcher, 2016 referred to by the referee. Consistent with our interpretation, there is little or no correspondence between them indicating that Tasmania probably was not the source of increased rBC emissions after the late 13th C. .

I recommend removing this statement entirely. Given the uncertainties in the rBC source region assignment methods used in the paper (cf my earlier comments), one could just as well conclude that the study illustrates the challenges of inferring regional atmospheric BB emissions from a single ice core depositional record which may receive BC from multiple regional sources.

This is an important finding and we prefer to retain it.

We disagree with the Referee's statements on two points. First, we emphasize that lake-sediment charcoal records and fire-scar records in trees say little or nothing about the magnitude of burning emissions. They reflect only the incidence of local burning (e.g., for lake sediments, only in the watershed or perhaps nearby watersheds). As we discuss in the manuscript, the Antarctic ice core records presented here are located 1000s of kilometers from potential sources and document atmospheric concentrations over the coring site and along transport pathways. Therefore, they reflect regional-scale emissions and document large-scale atmospheric rBC concentrations which was the primary objective of our study. The ice records are direct, not indirect, indicators of atmospheric concentrations and so very different than lake-sediment charcoal or fire-scar tree records.

Second, although we agree that there would be significantly greater uncertainties associated with interpreting a single ice-core rBC record, we again reiterate that our findings are based on spatial and temporal patterns captured in a wide array of well-dated and synchronized ice-core records – not a single ice core.

Reviewer Reports on the First Revision:

Referee #3 (Remarks to the Author):

The authors have addressed the points raised in my previous review.

Referee #4 (Remarks to the Author):

My comments (below) are related to the rebuttal and the revisions made to the first version of this manuscript, so I will not go through the entire "checklist" of aspects of the study.

My criticisms of the initial paper concerned chiefly (1) the possible effects of historical changes in snow accumulation on the rBC fluxes recorded in the James Ross Island ice-core; (2) the uncertainties in the FLEXPART-based rBC emission sensitivities, and how these might affect the ice-core record interpretation; and (3) the apparently rather selective choice of paleofire charcoal records from regions other than New Zealand used in Fig. 1.

Overall, I think that the authors have done a very decent job at addressing my criticisms and answering my questions on points (1) and (2) above. Their response and the changes made to the paper (or the supplementary material) have contributed to clarifying many points and dispelling ambiguities. I still deplore the fact that no clear justification is offered for only showing some selected paleofire records from Tasmania and South America in Figure 1. This could easily have been remedied but was not. In their response to my criticisms (and those of other reviewers) the authors stress that the final "attribution" of Maori settlement as the chief cause of enhanced rBC emissions (the main conclusion of their study) depends strongly on a comparison of paleofire histories in the different possible source regions. Considering this, it seems to me all the more important to show how reliable or uncertain those histories are, and the fact that only some are selected in Fig. 1 still conveys (at least to me) the impression of selective biases in the data interpretation.

Altogether, I do not think this is "fatal" to the authors' main thesis, and I am (while not truly satisfied) content to leave it at that. In many other respects, the study is very thorough and worthy of praise, and shows a very interesting blend of methods and approaches applied for paleoclimate and paleo-atmospheric reconstruction. It also brings some much needed nuance in the discourse on historical human pollution impacts on the global atmosphere, suggesting that even (pre-European colonial) indigenous people were capable of having such impacts.

I am thankful to the editors to have allowed me to see how the authors replied to to other reviewers' criticism, which was very helpful in assessing the overall value and significance of the paper.