Response to Anonymous Referee #2 comment
Received and published: 1 January 2020

Dear Anonymous Referee,

We thank you for your time, expertise, and helpful suggestions.

We apologize for the inconvenience that Marquetto et al. (2019) was not available during the review process. Marquetto et al. (2019) was accepted for publication in October 2019 and will be available online in April 2020 in "Advances in Atmospheric Sciences" as Marquetto et al. (2020). We have added the accepted manuscript as supplementary material for your review. Considering our system setup and calibrations carried out for the SP2, we believe the rBC concentrations found in this work are solid results, as described with more details along the text.

We made several improvements to the manuscript based on the reviewer's suggestions. First, we opted to remove the 2008 trace element records from the dating section. Although the 2015 and 2008 cores presented an overlap of 7 years (2002-2008), there is a 20% distortion between them in order to match both. This raised an issue about using a core 850 m away, with topographical differences, to date the rBC core. We decided this brings more uncertainty to our work than using the 2015 trace element record to constrain the dating down to 2002, and then base the rest of the dating on the relations of rBC, Na, Sr and S observed for the 2015-2002 period.

We also removed the sodium record from the spectral analysis and comparison of rBC and Na transport, as the 2015-2002 Na record is too short to show cycles in the spectrum.

On the other hand, we added atmospheric transport simulations using the HYSPLIT model to identify BC source areas. Results corroborated our initial conclusions of Australia and New Zealand as the most probable sources of BC to the TT07 drilling site, and indicated limited influence of South American air masses. More information is presented at the end of this document, after responses to reviewer's suggestions.

We also added a comparison of the Antarctic rBC records with snow accumulation, elevation and distance from open sea. In East Antarctica, rBC concentrations have a negative correlation with snow accumulation and positive correlation with elevation and distance to the sea, whereas in West Antarctica rBC concentrations present a positive correlation with snow accumulation and a negative correlation with elevation and distance to the sea.

These opposite trends may indicate differences in rBC transport to East and West Antarctica. While for East Antarctica upper tropospheric transport and dry deposition may be the main controllers of rBC concentrations (Bisiaux et al., 2012b), for West Antarctica rBC concentrations may be modulated by intrusion of air masses from the marine boundary layer, contrary to what *was previously suggested (Bisiaux et al., 2012a). Low elevations in West Antarctica facilitates the intrusion of moisture-rich cyclones and the transport of aerosols inland (Neff and Bertler, 2015; Nicolas and Bromwich, 2011), while the positive relationship between West Antarctica rBC concentrations and snow accumulation may indicate rBC to be primarily deposited through wet deposition, being scavenged along the coastal regions were snow accumulation is higher.*

The rBC record we present in this work is from an unique area – the Pine Island/Institute Glacier Divide, where air masses from the Weddel and Bellingshausen Seas converge (Parish and Bromwich, 2007), and is the highest altitude rBC core collected in West Antarctica. Considering all these improvements and new findings, we believe our manuscript is suitable for The Cryosphere.

Please find our responses in italic, while we kept your original comments in normal text.

Original referee comment:

The paper provides a 47-year ice core record of refractory black carbon (rBC) from West Antarctica, specifically Pine Island Glacier. rBC was analyzed by a Single Particle Soot Photometer. The core was dated to 1968, primarily using seasonality of rBC. BC impacts on snow albedo were modeled using the Snow, Ice, Aerosol, Radiation (SNICAR) model. BC emissions were explored with fire spot inventories and spectral analysis was conducted by the REDFIT method.
With respect to the TC guidelines: 1. The paper provides additional field observations of rBC in snow and ice in a data-sparse region of the cryosphere. Making field observations like this available to the community is important for refining our understanding of impurities in the cryosphere and their impact on surface albedo of the Antarctic ice sheet. 2. While the paper provides an additional valuable dataset, it is unclear to me whether the record interpretation is particularly novel. 3. The main finding appears to be that BC transport to the site is not related to marine air masses, which has previously been shown in other ice core records in Antarctica (i.e. Bisiaux et al., 2012). 4. The analytical details appear to be outline in Marquetto et al., 2019, however, I am having trouble locating the manuscript. 5. Thus, I have some remaining analytical questions outlined below. 6. Having access to Marquetto et al., 2019 would assist with reproducibility. 7. The authors provide credit to related work, but I think they should further identify/emphasize the novelty of their contribution. 8. The title clearly reflects the content of the paper. 9. The abstract provides a concise and complete summary of the existing manuscript. 10. The paper could have benefited from more thorough proofreading before submission; there are some typos. 11. Please refer to 10. 12. Black carbon and refractory black carbon are abbreviated at times and then spelled out at others (i.e. Lines 36, 67, 338). 13. Current figures and tables seem to appropriately support the text. 14. The number of references seems appropriate. 15. The Marquetto et al., 2019 paper would have been useful supplementary information.

Responses

4: Marquetto et al. (2019) was accepted for publication in October 2019 and will be available online in April 2020 in "Advances in Atmospheric Sciences" as Marquetto et al. (2020). The accepted manuscript has been added as supplementary material.
5: Along with providing the Marquetto et al. (2020) file, we added additional analytical information in the manuscript based on your suggestions, as is detailed below.
6: Marquetto et al. (2020) is available as supplementary information.
7: We improved our manuscript, as already mentioned above. Additional information is given at the end of this document, after responses to the reviewer.
12: We are now consistent with using abbreviated versions.
15: Marquetto et al. (2020) is available as supplementary information.

Specific Suggestions:

Line 29: Typo: 'while there they change'

Changed the sentence to: "BC particles stay in the atmosphere for just one week to 10 days (Bond et al., 2013; Ni et al., 2014), but ~~while there~~ during that time they change the direct radiative forcing..."

Line 45 – 48: Sentence could be restructured for clarity.

Changed the sentence to: "More ice core records are needed to understand the spatial variability of BC transport and deposition to Antarctica, as well as to improve general circulation models (Bisiaux et al., 2012b). In this work we add another high-temporal-resolution rBC record from a West Antarctic snow and firn core to the existing literature, in order to contribute to the understanding of BC temporal and spatial variability in Antarctica.

Line 58 – 61: This section could be expanded, including more specific references.

Added more information in this section, see below in red color:

The core (TT07) was drilled in the 2014-2015 austral summer on the Pine Island Glacier (West Antarctica) at 79°55'34.6"S, 94°21'13.3"W (elevation 2122 m above sea level – a.s.l.), near the Mount Johns Nunatak (located 70 km NE of the drilling site) (Fig. 1) and close to the Institute/Pine Island ice divide. The drilling site was chosen due to its relatively high accumulation rate, that ensures a well preserved seasonal stratigraphic record (Schwanck et al., 2016; Thoen et al., 2018), as well as the region being influenced by air masses from the Weddel, Amundsen and Belingshausen seas (Parish and Bromwich, 2007; Thoen et al., 2018)*.*
The West Antarctic Ice Sheet (WAIS) presents lower elevation and lower coastal slopes than the East Antarctic Ice Sheet (EAIS), which facilitates the intrusion of moisture-rich cyclones to the interior of the continent and the transport of aerosols inland (Neff and Bertler, 2015; Nicolas and Bromwich, 2011)*. Katabatic winds at the drill site are not as strong as they are in most*

of West Antarctica, due to the higher site elevation compared to the surrounding region (Parish and Bromwich, 2007). *Seasonal differences in atmospheric transport have been reported for the TT07 drilling site, with particle trajectories during the austral summer being slow moving and more locally influenced, while during the winter, air trajectories are influenced by oceanic air masses due to strong westerlies. The majority of air masses arrive from the Amundsen Sea and, secondarily, from across the Antarctic Peninsula and Weddell Sea* (Schwanck et al., 2017)*. These are also the preferred pathway for dust particles* (Neff and Bertler, 2015)*.*

Line 77: I cannot find Marquetto et al., 2019 online. Thus, a lot of important analytical details seem to be missing from this manuscript. For example, how long before analysis were the samples melted?

Marquetto et al. (2020) has been added as a supplement. We added melt time information in the first paragraph of section 3.3: "Samples were melted at room temperature or in a tepid bath not exceeding 25°C, sonicated for 15 min, and then analyzed with one hour of melting."

Lines 83 and 90: Duplicate sub-section titles.

Corrected. Section 3.3 should be entitled: "Laboratory and vial cleaning".

Line 87: Why were polypropylene vials used instead of glass vials? Was particle loss explored with leaching on the vials? Additionally, how long did the samples sit in the vials before analysis?

Polypropylene vials are widely used for rBC analysis in the SP2. Previous work has tested polypropylene vs glass, and found that polypropylene vials are as suitable to glass unless the sample is left in the liquid state for an extended period of time (Wendl et al., 2014)*, which was not the case in this study. Samples were melted shortly after analysis.*

Line 146: I don't think Sr is mentioned in Legand and Mayewski, 1997.

Corrected and added bibliography for Sr, as Legrand and Mayewski (1997) cite only Na. The full sentence is now: "Na and Sr also peak in the dry season (during winter) due to intense atmospheric circulation and transport (<u>Legrand and Mayewski, 1997; Schwanck et al., 2017</u>). Increased marine biogenic activity reflects an increase in S in late austral summer (<u>Schwanck et al., 2017; Sigl et al., 2016</u>) .

Line 197: Suggest 'fit' as opposed to 'fitted'.

Suggestion accepted, thank you.

Line 199: Suggest using the same past tense, 'chose' as opposed to 'choose'.

Suggestion accepted, thank you.

Section 4.1 Dating: Given that the main findings of the paper rely on dating based on seasonality of the rBC record, I think this section could be expanded. For example, the authors could add more discussion as to why the authors think the addition of the rBC record to the layer counting would lead to a dating difference of one year or more, with respect to the core collected nearby that was analyzed for trace elements.

> *We opted to use only the 2015 trace element record for dating and constrain the rest of the dating based on the rBC well defined seasonality for West Antarctica (Arienzo et al. 2017; Bisiaux et al., 2012; Winstrup et al., 2017) and for the Pine Island Glacier (Pasteris et al. 2014).*
>
> *To improve our dating for these first meters we reviewed sample resolution for the rBC and trace element cores, and added an additional parameter to dating: the maxima in the non-sea-salt sulfur to sodium (nssS/Na) ratio, a robust seasonal indicator that peaks around the new year (Arienzo et al. 2017). This parameter helps in the identification of the annual layers more than the Na and S records alone. Non-sea-salt sulfur was calculated using Eq. 3 to 6 from Schwanck et al. (2017) and references therein.*
>
> *We consider this dating to have ±2 years uncertainty. The first uncertain year is located at 6.18 m (between 2003 and 2002, figure 2a), where S and nssS/Na peak but no full cycle is observed in the rBC record. We did not consider this to be a year, as rBC does not present a full cycle. The second uncertain year is located at 18.14 m (year 1973, figure 2b) where there is no clear rBC peak but snow accumulation would be anomalously high if considered to be only a year instead of two. We consider this to be an annual pick and consequently two years, as there is no evidence of higher-than-normal snow accumulation in the region for this period* (Kaspari et al., 2004).

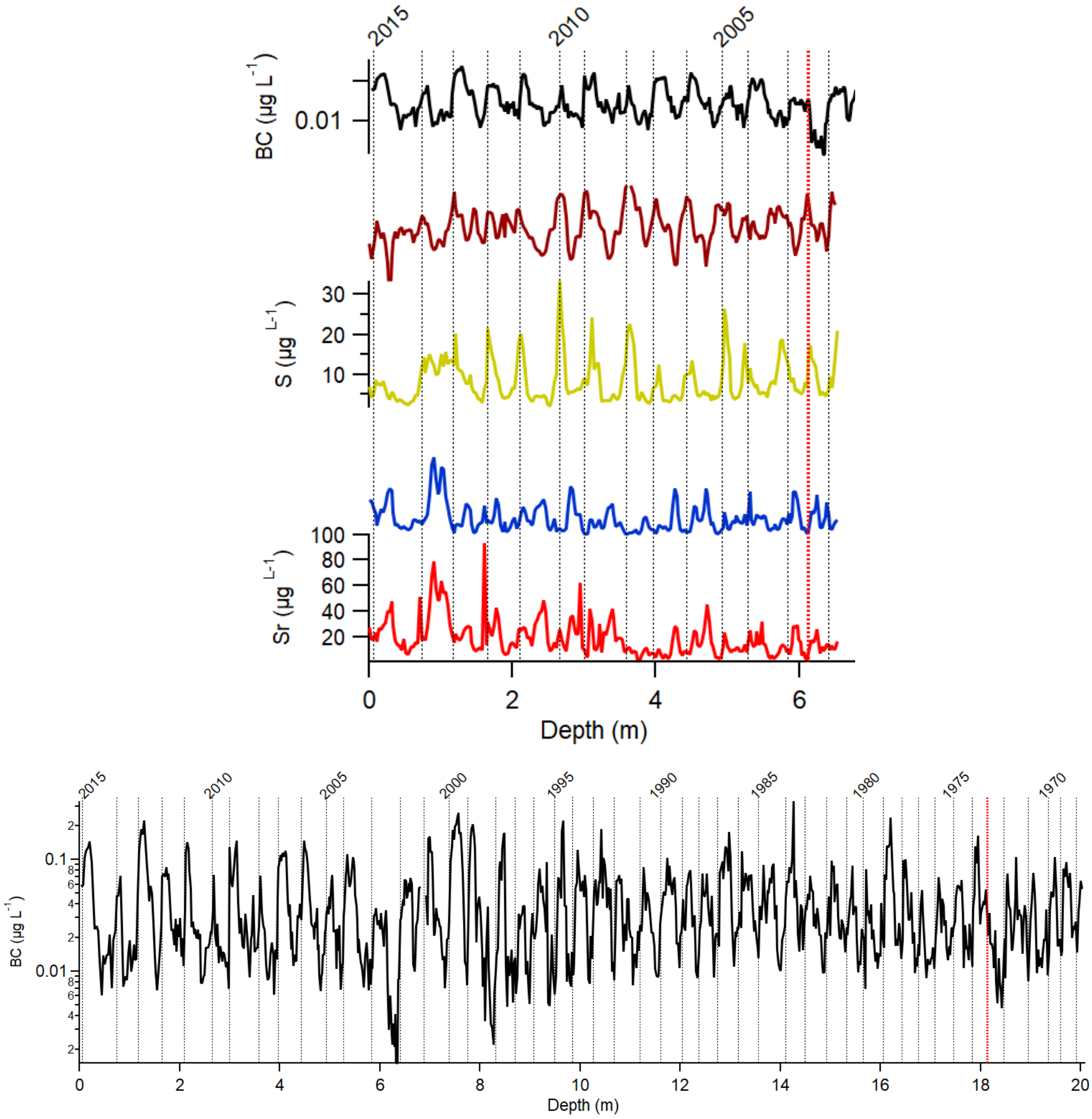

Figure 2. (a) Dating of the snow and firn core based on rBC and using S, Sr, Na and nssS/Na records from nearby core (see section 3.6) as support for the first 6.5 meters. Dashed lines indicate estimated New Year and red dotted line indicate uncertainty in dating, explained in the text. (b) Dating for the full core (y axis logarithmic). Red dotted line indicates uncertainty in dating, as explaned in the text.

Line 338: The starting date here (1969 – 2015) does not match the abstract or Table 4 (1968 – 2015).

Corrected the date to 1968-2015.

Figure 4 Legend: Suggest (bottom) instead of (base).

Suggestion accepted, thank you.

Additional information regarding HYSPLIT model:

We added atmospheric transport simulations from using the HYSPLIT model to identify BC source areas. We added two additional sections, one in methodology and another in results. They are as follows:

3. Methodology

3.10 Particle trajectory simulations

In order to simulate rBC particle trajectories from source areas to the TT07 drilling site, we used the Hybrid Single-Particle Lagrangian Integrated Trajectory v4 model (HYSPLIT - Draxler and Rolph, 2003; Stein et al., 2015), from NOAA. Hysplit is a complete system for computing simple or complex transport and deposition simulations (Stein et al., 2015) that has been used in Antarctica by several authors (Dixon et al., 2011; Markle et al., 2012; Marquetto et al., 2015; Schwanck et al., 2016a, 2017; Sinclair et al., 2010).

We used global reanalysis data from the National Centers for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR) – the NCEP/NCAR data set – and ran 10-day (240 h) back-trajectories, every 5 days, from 1968 to 2015, at an initial height of 1000 m. We consider 10 days to be an appropriate simulation time as this is the estimated maximum lifetime of BC in the troposphere (IPCC et al., 2013). An initial height of 1000 m was used in order to minimize disturbance from the underlying terrain, but still maintaining a link with the surface wind field (Sinclair et al., 2010). To identify main airflow patterns at the TT07 drilling site, the individual trajectories were separated into dry and wet seasons (depending on day and month of each run) and simulations from each season were grouped into five clusters using the HYSPLIT model's cluster analysis algorithm.

4. Results

4.8 Particle trajectory simulations using HYSPLIT

We simulated particle transport during the austral wet and dry seasons as another mean of addressing rBC source areas. We ran the HYSPLIT back-trajectory model every five days from 1968 to 2015, for 10 days each (estimated maximum BC lifetime in the troposphere) and clustered the results in five groups for the wet and dry seasons (Fig. 9).

The majority of simulated air parcels arriving at the drilling site presented a slow-moving trajectory (speed is proportional to trajectory length), reflecting a local/regional influence more than long-range transport from other continents (clusters 3 and 4 in Figure 9). This local/regional influence is observed both in the wet and dry seasons, although during the former the contribution of air masses from the Antarctic Peninsula and across

(Weddell Sea) are higher than during the latter. A fast-moving, year-round, continental group is also present (cluster 5), and may partly represent katabatic winds flowing from the continent's higher altitudes (East Antarctica) towards lower-altitude West Antarctica. The strongest contribution of long-range air parcels is from the South Pacific (clusters 1 and 2). These air masses are also fast-moving and present slight seasonal variations, shifting pole wards during the wet season, when they represent 34% of all air parcels, and away from Antarctica during the dry season, when they respond for 22% of all air parcels modelled.

Results from clusters 1 and 2, along with individual trajectories of each cluster (Fig. 10) support our conclusion that Australia and New Zealand are the most probable sources of rBC to the drilling site, considering tropospheric transport. The most visible influence of air parcels from these two countries to the drilling site can be seen in the individual trajectories of cluster 1 (Fig. 10) for both dry and wet season, while for cluster 2 and 4 there are trajectory variations from one season to another. The poleward shift of cluster 1 trajectories in the wet season (Fig. 9) may be a reason why the Australian emissions earlier in the year (May) are not visible in the TT07 rBC record. South American influence to the TT07 drilling site, on the other hand, is restricted to the higher latitude countries (Chile, Argentina), as shown in the individual trajectories of clusters 2, 3 and 5 (Fig. 10). This suggests that South American fires are not significant contributors to the rBC concentrations observed at the TT07 site when considering only tropospheric transport.

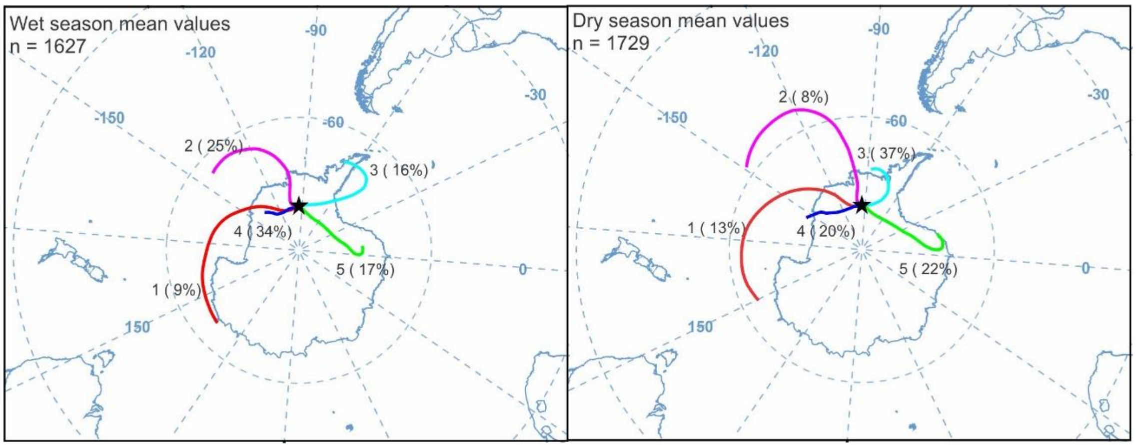

Figure 9. HYSPLIT clusters of 10-day back-trajectories ran every 5 days from 1968 to 2015 arriving at the TT07 drilling site. Results are separated by wet and dry season, and grouped in five clusters (percentage of trajectories for each cluster is shown in parenthesis). Number of trajectories (n) used for the cluster algorithm is shown at the top, on the left side.

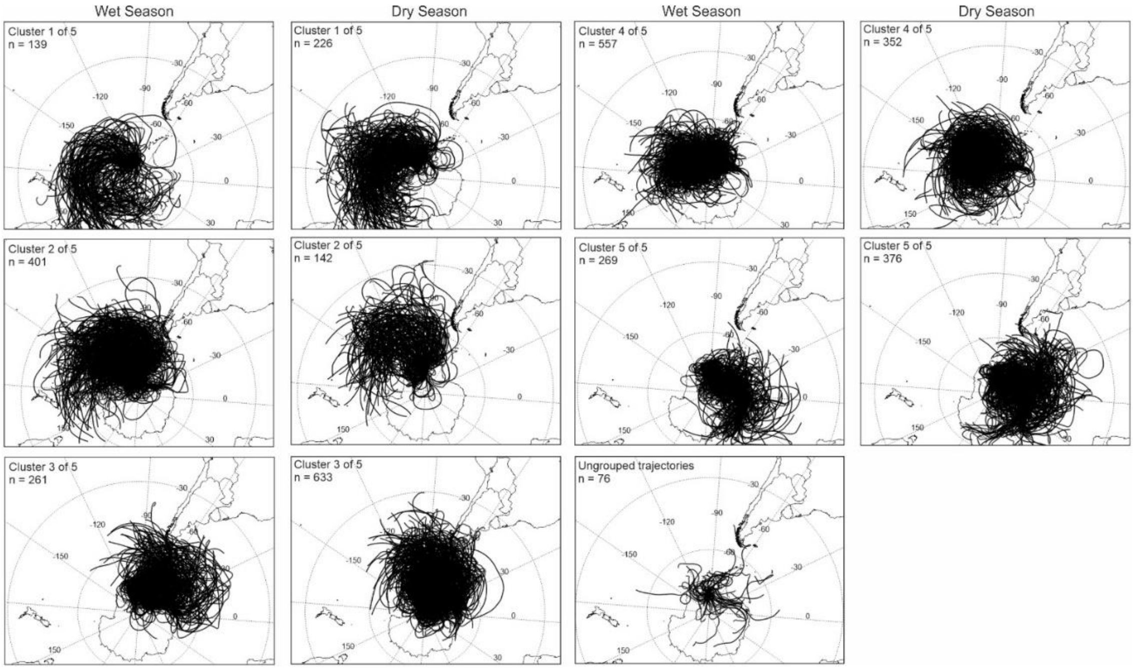

Figure 10. Individual trajectories used for the cluster analysis in figure 8. Number of trajectories (n) used for each cluster is shown at the top, on the left side. Clusters 1, 2 and 4 show air masses arriving from Australia and New Zealand to the TT07 drilling site, while clusters 2, 3 and 4 show the (limited) contribution of South American air parcels to the site. Similar clusters from wet and dry season are side by side for comparison. Wet season presented 76 ungrouped trajectories, while dry season presented none.

Additional information in section 4.4 of the manuscript:

> Figure 6 shows a comparison of the above mentioned rBC records with snow accumulation, elevation and distance from open sea. Distance from the sea influences rBC fluxes in West Antarctica (Arienzo et al., 2017), and was calculated considering the median sea ice extent from 1981 to 2010 for September (Matsuoka et al., 2018), when rBC emissions start to rise in South America/Australia/New Zealand and rBC concentrations begin to rise in West Antarctica (Arienzo et al., 2017; Bisiaux et al., 2012b; Pasteris et al., 2014). We measured the distance from the rBC records to the closest open sea source (Amundsen sea for West Antarctic records, Lazarev to Cosmonauts sea for NUS0X-X and Mawson sea for Law Dome). We acknowledge this is a simplistic approximation and that the preferred air mass pathways from the sea to the points are not as straightforward, but for the scope of this work we consider this approximation sufficient.
>
> No patterns are clear for both East and West Antarctica, whereas when considering the data from East and West Antarctica separately, opposite trends are observed. In East Antarctica, rBC concentrations have a negative correlation with snow accumulation and positive correlation with elevation and distance to the sea, whereas in West Antarctica rBC

concentrations present a positive correlation with snow accumulation and a negative correlation with elevation and distance to the sea.

We observed that for East Antarctica, rBC x snow accumulation and rBC x elevation presented statistically significant correlations ($r^2 = 0.78$, $p < 0.01$ for the former and $r^2 = 0.79$, $p < 0.01$ for the latter). On the other hand, distance from the sea does not seem to correlate with rBC ($r^2 = 0.52$, $p = 0.06$).

For West Antarctica, relationships are the opposite: positive correlation between rBC concentrations and snow accumulation ($r^2 = 0.69$, $p = 0.08$) and negative correlations between rBC concentrations and elevation/distance from the sea ($r^2 = 0.30$, $p < 0.33$ for the former and $r^2 = 0.79$, $p < 0.05$ for the latter). McMurdo and South Pole points are not considered in this calculation as they likely reflect local contamination instead of long-range transport (Casey et al., 2017; Khan et al., 2018). Bisiaux et al. (2012b) have also observed negative (positive) relationships between rBC concentrations and snow accumulation (elevation) for East Antarctica, although their comparison also included the WAIS Divide point to the dataset.

These opposite trends may indicate differences in rBC transport to East and West Antarctica. While for East Antarctica upper tropospheric transport and dry deposition may be the main controllers of rBC concentrations (Bisiaux et al., 2012b), for West Antarctica rBC concentrations may be modulated by intrusion of air masses from the marine boundary layer. Low elevations in West Antarctica facilitates the intrusion of moisture-rich cyclones and the transport of aerosols inland (Neff and Bertler, 2015; Nicolas and Bromwich, 2011), while the positive relationship between West Antarctica rBC concentrations and snow accumulation may indicate rBC to be primarily deposited through wet deposition, being scavenged along the coastal regions were snow accumulation is higher.

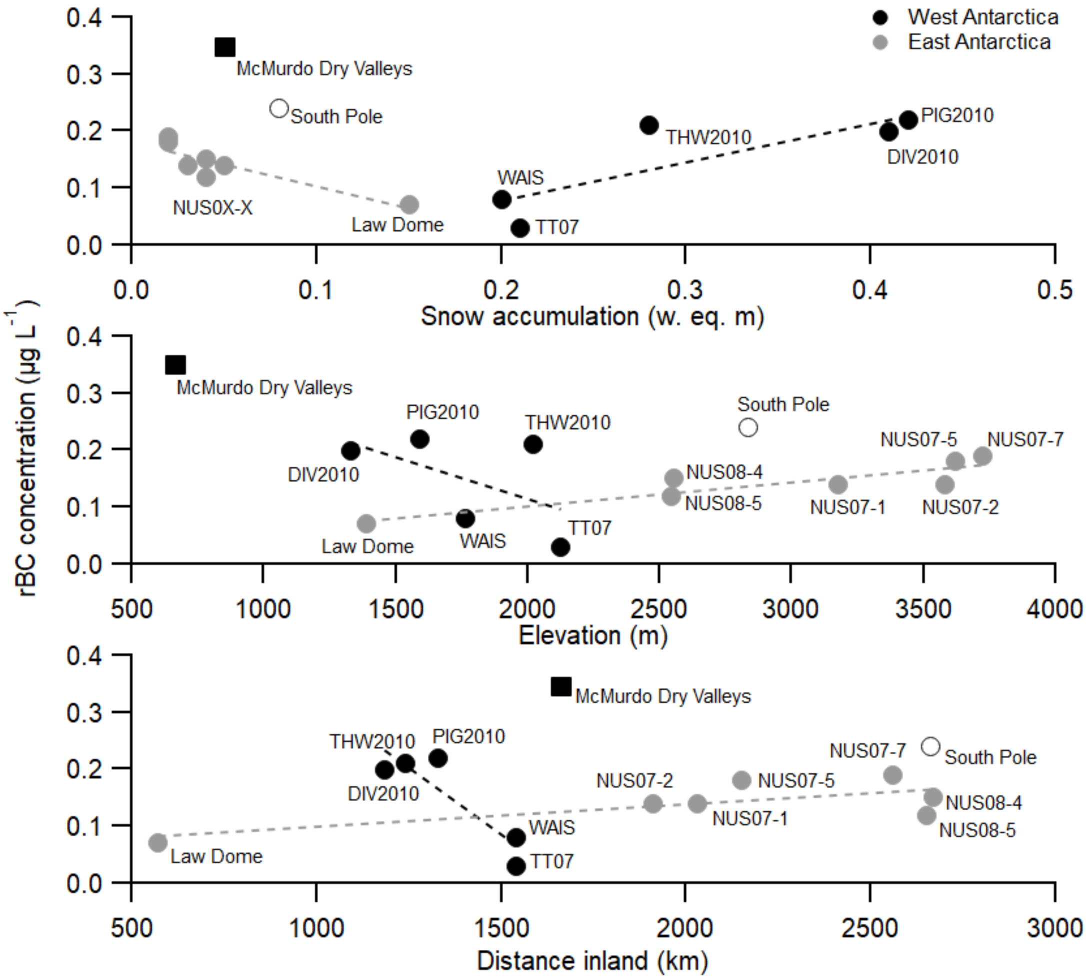

Figure 6. rBC records from Antarctica. rBC concentrations plotted against snow accumulation, elevation and distance from the sea. Solid lines indicate statistically significant correlations ($p < 0.05$), while dashed lines indicate not significant correlations ($p > 0.05$).