Response to Anonymous Referee #1 comment

Received and published: 19 December 2019

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. More information is presented at the end of this document, after responses to reviewer's suggestions.*

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:

This manuscript by Marquetto et al presents a 47 yr black carbon record from a 20- m firn core recovered from the Pine Island Glacier in West Antarctica. The authors measured rBC using the SP2 method, and dated the core primarily using the seasonal cycle of rBC. Potential southern hemisphere rBC source regions to this site were explored by correlating the seasonal cycle of rBC to Australian and South American fire spot data and GFED biomass burning emissions estimates as well as by comparing the power spectrum of rBC to large-scale atmospheric patterns (ENSO, AAO, and ASL) and fire spot data.
While Antarctic rBC records are important for understanding changes in southern hemisphere biomass burning as well as radiative forcing, and undoubtedly an immense amount of work went into developing this highly-resolved dataset, the interpretation and discussion are not thorough or novel enough to add significantly to the understanding of rBC deposition in West Antarctica. The discussion focused on three analyses: SNICAR snow albedo modeling, identifying continental emissions sources, and linking rBC to atmospheric circulation using spectral analysis. The SNICAR modeling showed that rBC deposition at this site has little to no effect on snow albedo, which is not unexpected given the extremely low rBC concentrations and (as noted by the authors) has already been shown for clean Antarctic snow (Casey et al., 2017). The identification of source regions by comparing seasonal cycles in observed rBC and GFED fire emissions is purely correlativeâ˘Aˇ TI do not think is a strong enough approach, especially with my concerns about dating, to draw conclusions about source regions without a more robust approach that would consider atmospheric transport and magnitude of biomass burning emissions. Finally, the spectral analysis was similar to that conducted by Bisiaux et al. (2012) for the WAIS Divide and Law Dome rBC records and does not provide any concrete new links between atmospheric circulation and Antarctic rBC. Furthermore, I have concerns about the factor of 2-3 lower rBC concentrations compared to other West Antarctic ice cores (see comments below).

Overall, since the conclusions do not add substantial new insight or understanding to rBC deposition and mainly confirm what is already known, I do not think this manuscript is suitable for the scope of The Cryosphere.
I have two major concerns about the methods: 1.) rBC measurements, and 2.) dating.

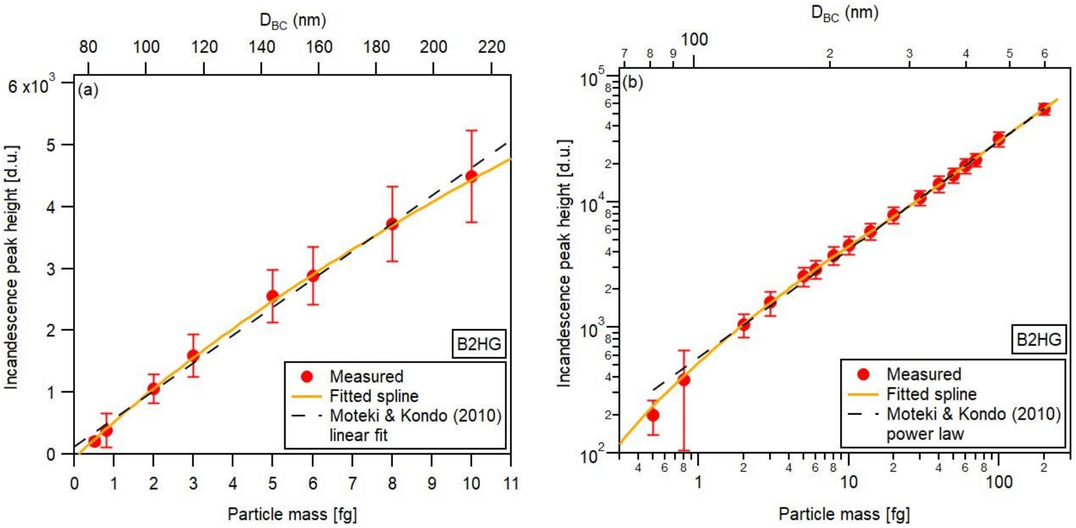
> *We respond to the two major concerns below.*

1. rBC measurements: The rBC concentrations presented in this study are a factor of 2-3 lower than rBC concentrations in other West Antarctic cores, including the WAIS Divide ice core as well as other measurements from Pine Island Glacier (Pasteris et al., 2014). Note that rBC measurements from early 1900s to 2006 from Pine Island Glacier (as well as Thwaites Glacier and the divide between Pine Island and Thwaites Glaciers) were previously published in Pasteris et al. (2014) and are also 2-3x higher than the concentrations presented in this manuscript- it would be worth including this citation and even comparing to this published dataset to see how the magnitude and temporal variability of the records compare.

> *We included* Pasteris et al. (2014) *as a citation along our manuscript and added their data as comparison in section 4.4. Thank you for pointing this out.*

The authors need to provide more information to determine if this offset is real or a result of the SP2 calibration/analytical system. Since the referenced Marquetto et al. (2019) manuscript does not appear to be published at the time of this review, please include details on the SP2 internal/external calibration in this manuscript.

> *We believe the lower rBC concentrations found by this work are not due to less-than-optimal SP2/analytical setting, and are, in fact, a reflection of the true rBC concentration in the samples. We were diligent in maintaining low background concentrations for this study, and detailed specifics on the methodology are provided in this response and in the revised manuscript.*
>
> *We attached Marquetto et al. (2020) so you can check the internal and external calibration thoroughly. In short, internal calibration was carried out using a Centrifugal Particle Mass Analyzer (CPMA) to select 23 particle masses from 0.5 fg to 800 fg from an Aquadag solution. Each selected mass ran for 30 min to 6 h to provide statistically significant particle triggers to calibrate the SP2, and calibration curves were then generated for all SP2 channels. The data presented in the manuscript in review is from the duplicated extended range broadband detector (B2HG), as this channel gave the best-fit calibration curve (spline) of all channels in the 0.5 – 200 fg range, but with an upper detection limit at ~ D_{BC} = 600 nm. The external calibration was carried out daily using five fresh Aquadag standards ranging from 0.01 to 1.0 μg L^{-1} and one environmental standard (diluted meltwater of a snow sample from Table Mountain, WY, USA) of known concentration (0.18 ± 0.04 μg L^{-1}).*



Calibration curves for the (a) B2HG channel (0.5 < M < 10 fg – linear scale), (b) B2HG channel (0.5 < M < 200 fg – logarithmic scale) obtained from the internal calibration carried out using the SP2+CPMA.

> *Please note that Marquetto et al. (2020) present a comparison between two subsampling methods (solid state cutting and continuous melter system) – both resulted in statistically similar rBC concentrations.*

How were the ice samples melted (room temperature?), and how soon before SP2 analysis were they melted (lines 96-97)? Wendl et al. (2014) show how rBC is lost after melting during sample storage in polypropylene vials over just a few days (with proportionally greater losses for low-concentration samples).

> *We added in section 3.3 the sentence: "Samples were melted at room temperature or in a tepid bath not exceeding 25°C, sonicated for 15 min, and then analyzed (in less than 1 h after melting)." This is also described in* (Marquetto et al., 2020)*.*

How often was 5% HNO3 used to clean the system (line 118), and how did you determine when the acid was flushed from the system? Wendl et al. (2014) also discuss how acidification can result in significant loss of rBC.

> *The results regarding acidification presented in Wendl et al. 2014 were conducted at Central Washington University, where we conducted this research, so we are well aware of these findings. Rather, we were very diligent to maintain the system at the MQ background level (at 0-0.5 particles cm^{-3}, translating to less than 0.01 µg L^{-1} rBC concentration). When MQ levels increased to 0.5 particles cm^{-3}, the HNO3 was used to clean the TruFlo liquid flow monitor and the nebulized. The HNO3 was only used on three days during the 43 working days, sometimes more than once a day. We ran the solution for 10 to 30 min from the peristaltic pump to the nebulizer outlet tubing (disconnected from the SP2). After using the acid we ran MQ water (MilliQ-Element, Millipore, 18.2 MΩ cm) for at least double the time we ran the acid. After connecting the SP2 back, we ran MQ water for at least 15 min. These are the procedures used in the Ice Core Laboratory (CWU), where the samples were analyzed.*

What kind of tubing was used to transport the aerosol from the Marin 5 nebulizer to the SP2?

> *We used black conductive tubing from Simolex Rubber Corporation (Plymouth, USA) - http://simolex.com/product/conductive-silicone-extrusion-products-tubing/. This is the standard tubing that comes with the SP2.*

2. The description of the dating, namely the role of the datasets from the Schwanck et al. (2017) study, is vague. Since much of the analysis, including the seasonal cycle correlations and spectral analysis, require precise dating, more explanation of the dating must be given. The annual picks in Fig. 1 are consistently on the austral summer (January) decrease of rBC concentration, but appear to be inconsistently placed across the S, Sr, and Na records.

> *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., 2012a; 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)*.*

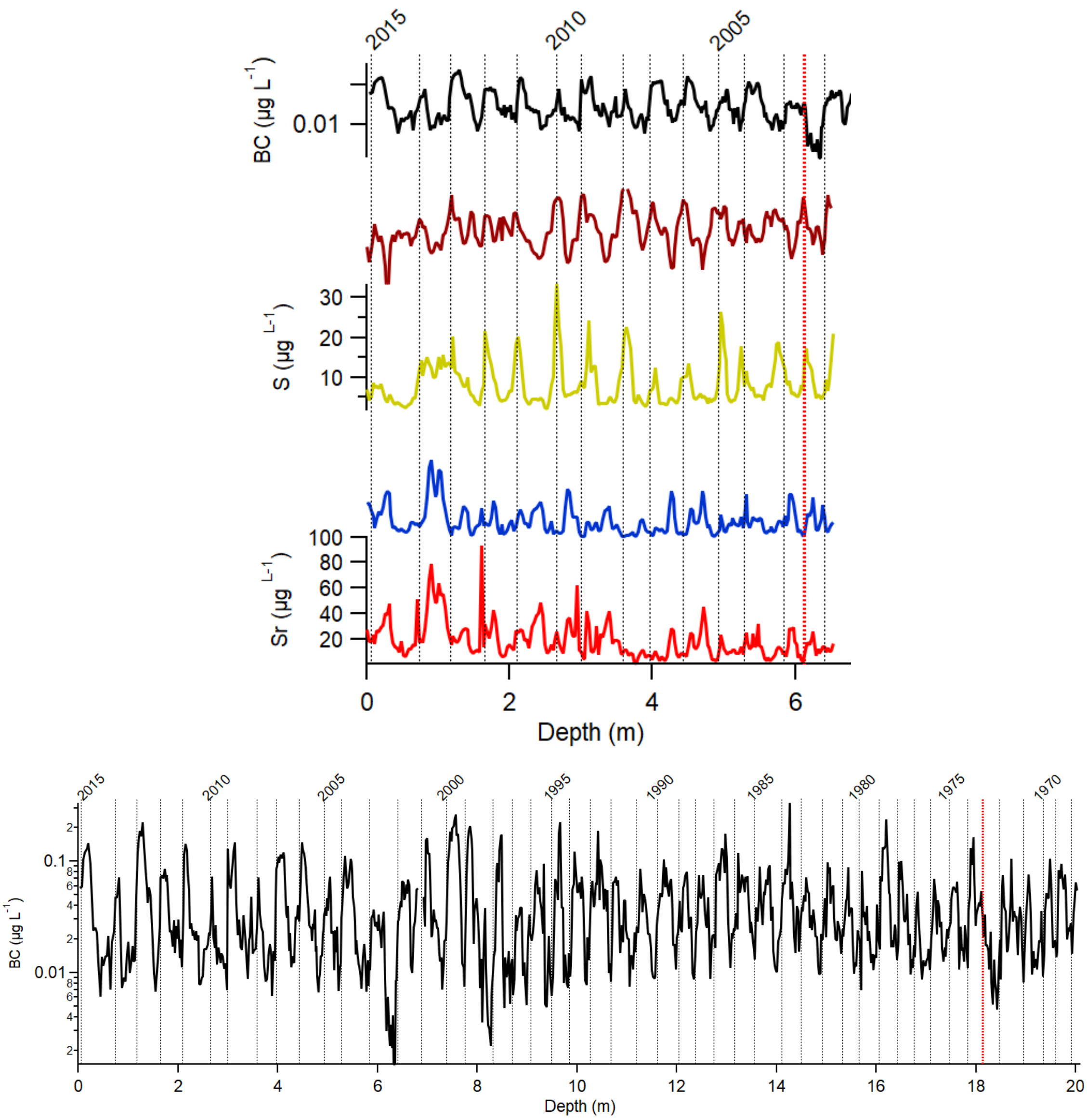


Figure 2. (a) Dating of the snow and firn core based on rBC and using S, Sr, Na and nssS/Na records from nearby core (see section 3.6) as support for the first 6.5 meters. Dashed lines indicate estimated New Year and red dotted line indicate uncertainty in dating, explained in the text. (b) Dating for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

Is the S, Sr, and Na data shown in Fig. 2 all from the Schwanck et al. core from _1 km away?

> *We opted to not use the 2008 record, which means the S, Sr and Na records in the next revision will be from the 2015 core collected a meter apart from the rBC record, down only to 6.5 m.*

I would expect cores 1 km apart to have a depth offset, so I would not be confident in correlating the picks from TT07 to the Schwanck et al. core without a common dataset to both cores to linking the cores in depth. Without showing that the chemical species from the TT07 core and Schwanck core are not offset in depth, it is not advisable to guide the dating of the TT07 core with data from the Schwanck et al. core, and likewise not justified to apply annual picks based on rBC in TT07 core to the Schwanck core Na record (I can't tell if the Na spectral analysis was conducted on the TT07 age scale or the original Schwanck et al. age scale). How does the Schwanck et al. data compare to the 5.6 m of S, Sr, and Na data for the core taken immediately adjacent to the rBC core (mentioned on lines 154-155)? There should be a few years of overlapping data to compare and that would at least be a start to justify comparing the two cores in depth.

> *There is an overlap in the two cores, from 2008-2002, although a perfect match in peaks means a 20% distortion in the records (~0.2 water equivalent meters difference between the 2008 and 2015 cores). Due to this we decided not to use data from Schwanck et al. (2017) for dating anymore.*

Furthermore, there appears to be circular logic between the dating and seasonal comparison to GFED. If indeed the drop in rBC concentration at the end of the austral summer was used as the primary annual pick, and this pick was justified by GFED/fire spot seasonality as stated in lines 161-163, how can you then draw meaningful conclusions about the timing of rBC vs. GFED (in Fig. 6a) to identify source regions? The rBC and GFED are already inherently linked based on how you defined the dating of the core. Based on GFED seasonality in Fig. 6a, BC emissions drop in November for Asia/Africa/S. America, two months before the January drop for Australia/NZ. Even a month or two difference on where the rBC drop is assigned could have significant implications for the correlations used in section 4.6 (based on n=12 months) which are used to underpin the conclusion on lines 286-289. It would be much more appropriate to date the TT07 core using an independent chemical species, and then use the independent dating to examine the rBC seasonality.

> *We do not see the dating as circular logic due to the use of GFED4s/fire spots data. We changed the text in section 3.6 to clarify this point:*
>
> *"We considered the new year to match the end of what we define as the austral dry season, as this is a reliable tie point in the record due to the abrupt drop in rBC concentrations. Previous studies have demonstrated that rBC deposition occurs in winter/spring, mostly September to December. For example: Arienzo et al. (2017) observed rBC concentrations to peak in September in the WAIS Divide ice core; Winstrup et al., (2017) used annual variations in rBC as the most reliable annual tracer for the Roosevelt Island Climate Evolution (RICE) ice core, stating that rBC tends to peak earlier in the year than January 1st. Pasteris et al. (2014) also corroborates rBC to peak in October and drop after for the Pine Island and Thwaites Glaciers, with lowest values from February to June. Bisiaux et al. (2012a) state that sub-annual rBC concentrations are highly seasonal in the WAIS Divide ice core for the period spanning 1850-2000 - low austral wet season and high austral dry season concentrations - and presented annual picks in the drop in rBC concentrations, as in this work. This is also consistent with the BC emission estimates from GFED4s and the fire spot databases from Australia and South America."*
>
> *As for dating the core using independent chemical species, we used the trace element data as a secondary parameter to dating due to the offset that could exist between it and the rBC record. Samples are not co registered,*

they were not analyzed from the same core and same vial, so their resolution will be different. Using the independent chemical species to date the rBC record in monthly resolution would lead to greater uncertainty than using the rBC dating, especially considering rBC seasonality is already well defined in West Antarctica (Arienzo et al. 2017; Bisiaux et al., 2012; Winstrup et al., 2017; Pasteris et al. 2014).

Other comments

Lines 14-15: Please specify what you mean by wet and dry season? I assume southern hemisphere wet/dry season, not at the ice core site (also on lines 145-146).

We mean austral dry and wet season. Added the word "austral" in all dry/wet season citations.

Lines 87 and 82: Is Marquetto et al. (2019) published and available?

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.

Lines 83 and 90: Same heading title for sections 3.2 and 3.3.

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

Line 116: Did you average the rBC data for the full 5 minutes that it was run? Can you quantify the stability of the measurement with a standard deviation over the time period averaged?

The total rBC mass during the period of measurement, along with liquid and air flows, is used to calculate the rBC concentration for each sample using the Paul Scherrer Institute SP2 toolkit 4.200f. During sample analysis we monitored the samples to ensure that the incandescent particle concentration was stable. Liquid and air flows are also stable during analysis, as described in lines 108-115, leading to a robust measurement. We don't see the utility in in applying a standard deviation to the time period averaged, as the liquid concentration is based on the entire sampled period. The total mass of the measured particles is what is used in the calculation. For Antarctic samples, particularly the low concentration wet season (austral summer/fall), the concentrations are very low (0.015 ug/L). At these low concentrations particles are crossing the SP2 laser at a regular, but intermittent rate (i.e., the SP2 incandescent concentration can be stable, maintaining incandescent particle concentrations at 0-2 particles/cc during the analysis period).

We observed, though, that stability of the measurements depends more on sample rBC concentration than analysis time. Marquetto et al. (2020) presents this reproducibility test. We analyzed samples more than once and *for different times (5, 20 and 40 minutes), and our coefficient of variation (mean of all measurements of the sample × standard deviation) did not vary with different measurement times, but was much higher for concentrations lower than 0.03 µg* L^{-1} *(25.7 ± 16.9%) than for higher concentrations (for concentrations between 0.03 and 0.07 µg* L^{-1}*, it was 10.4 ± 6.6%, and for concentrations higher than 0.07 µg* L^{-1} *it was 7.3 ± 4.4%.*

Lines 135-143: The Brazilian hotspot data is defined here, but never mentioned in the results and discussion section. Can it be omitted? Or did it result in a null finding?

It resulted in a null finding. Line 318 briefly talks about that: "All other spectra showed only well-marked annual periodicities and intra annual periodicities of 2 and 3 cycles per year (0.5 and 0.3-year bands, not shown)."

We changed the sentence to: "All other spectra (including Programa Queimadas satellite data) showed only well-marked annual periodicities and intra annual periodicities of 2 and 3 cycles per year (0.5 and 0.3-year bands, not shown)."

Line 140: How do the timeseries of the fire hotspot data compare to the rBC data? You only compare the power spectrums in this study. Do years with more hotspots correspond to years with more rBC deposition?

This comparison can be seen in the figure below (Fig. S3 in Marquetto et al. (2020)). Years with more hotspots do not necessarily correspond to years with more rBC deposition. This is the reason why we compared the power spectrums, to identify increases and decreases in common for the datasets without looking only at the absolute values. rBC concentrations in Antarctica are not only a result of BC emissions, but also of atmospheric transport, deposition during transport and physical processes in the drilling site (Bisiaux et al. 2012), so we do not expect hotspots intensity to necessarily match years with more rBC deposition.

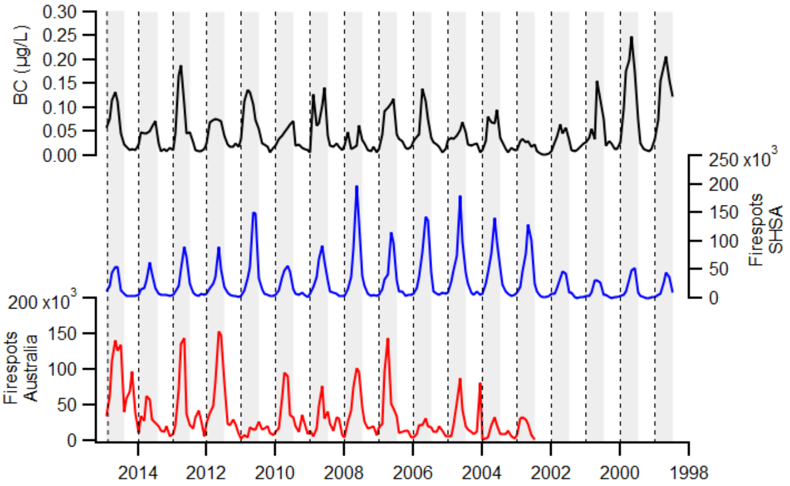


Fig. S3 from Marquetto et al. (2020). TT07 rBC record (rescaled to monthly resolution) compared to Southern Hemisphere South America (SHSA) and Australian/New Zealand firespot records.

Lines 147-148: Please define Na, Sr, and S before using abbreviations

Added definition in the abstract and in the manuscript (lines 14-15).

Lines 167-173: Please plot the average of the two density profiles in Fig. 3 that the quadratic equation was fit to.

Plotted. Density was averaged using curve fitting (polynomial terms = 3). Figure and caption are as follows:

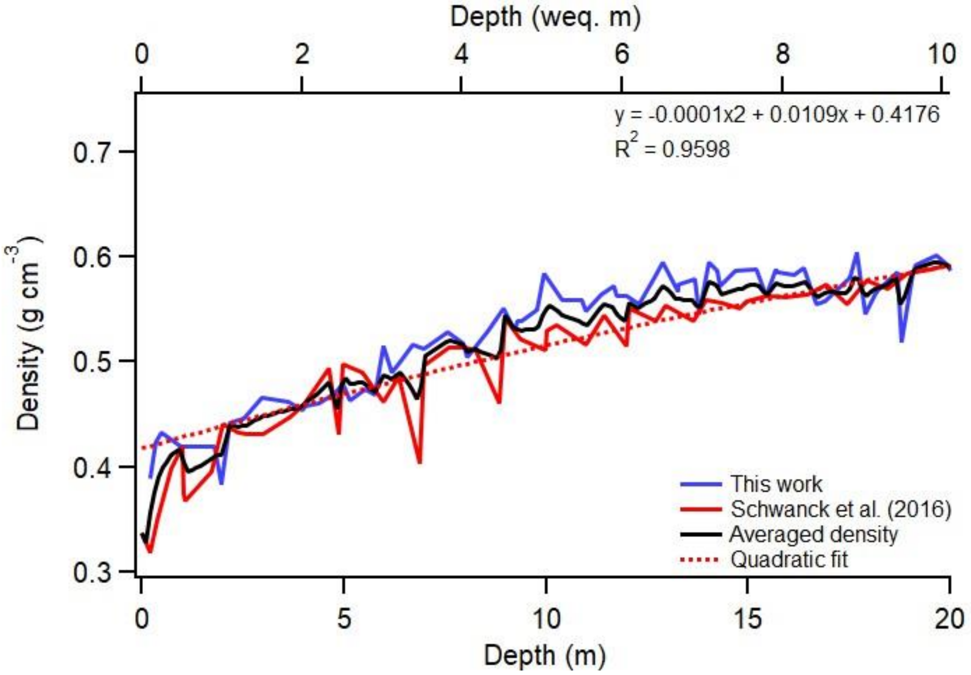


Figure 3. TT07 density profile (blue). Depth is presented in meters and water equivalent (weq) meters. The quadratic fit was calculated from the average density profile (black) from this work and from Schwanck et al. (2016).

Line 180: Please explicitly state which months went into the summer/fall and winter/spring averages.

Added to the text: summer/fall (wet season – January to June); winter/spring (dry season – July to December). We also corrected this specific line, where wet and dry season were attributed to the wrong annual seasons:

"We present our data as summer/fall (~~dry~~ wet season – January to June) concentrations and winter/spring (~~wet~~ dry season – July to December) concentrations."

Line 193 and 296: Is the Na record you use for spectral analysis from the TT07 core, or is it from Schwanck et al.? If it is from Schwanck et al., have you changed the dating?

We 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.

Lines 211-212: Per comments above, please clarify what dating is used for which chemical species.

> *We 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.*

Lines 244-246: Please include comparison to rBC data from Pasteris et al. (2014) from Pine Island Glacier.

> *We included Pasteris et al. (2014) as a citation along our manuscript and added their data as comparison in section 4.4. Thank you for pointing this out.*

Line 338: Abstract says record extends from 1968-2015.

> *Corrected the date to 1968-2015.*

Figure 1: Please check location of B40 ice core site.

> *Corrected B40 site and added locations from Pasteris et al. (2014) – THW, DIV and PIG (see below)*

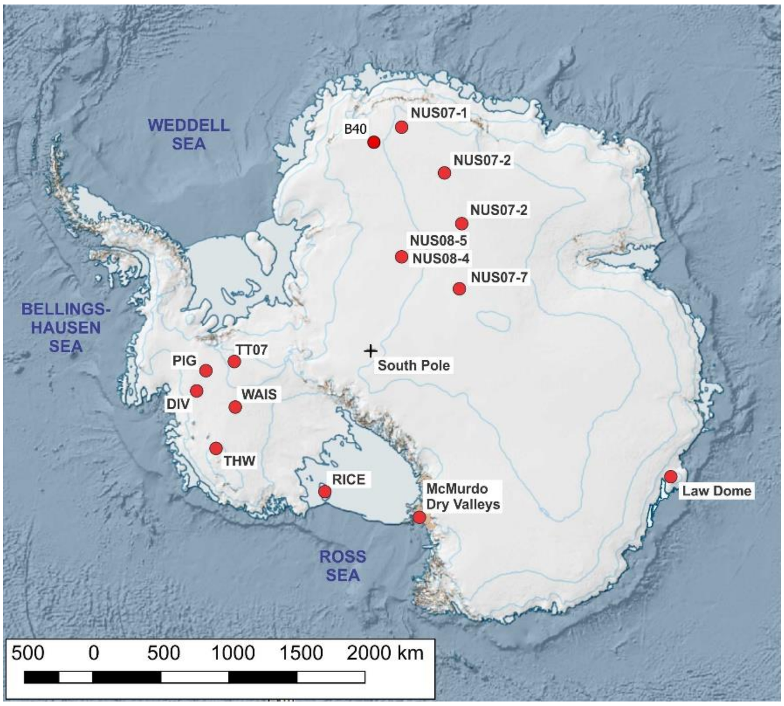


Figure 2: Please define which records are from the TT07 core and which are from Schwank et al.

> As commented above, we decided not to present the 2008 core from Schwanck et al. (2017) for the dating.

Figures 4, 5: It would be helpful to include the standard deviation (or standard error) of the measurements that went into the annual averages and wet/dry season plots as an error bar.

> *We added the geometric standard deviation (GSD) of annual and wet/dry rBC concentrations, as in the image and caption below. We did not include standard deviations in Figure 5 as the GSD for rBC annual average is already presented in Figure 4. We opted to present GSD in shaded area instead of error bars for readability, especially in the top graph.*

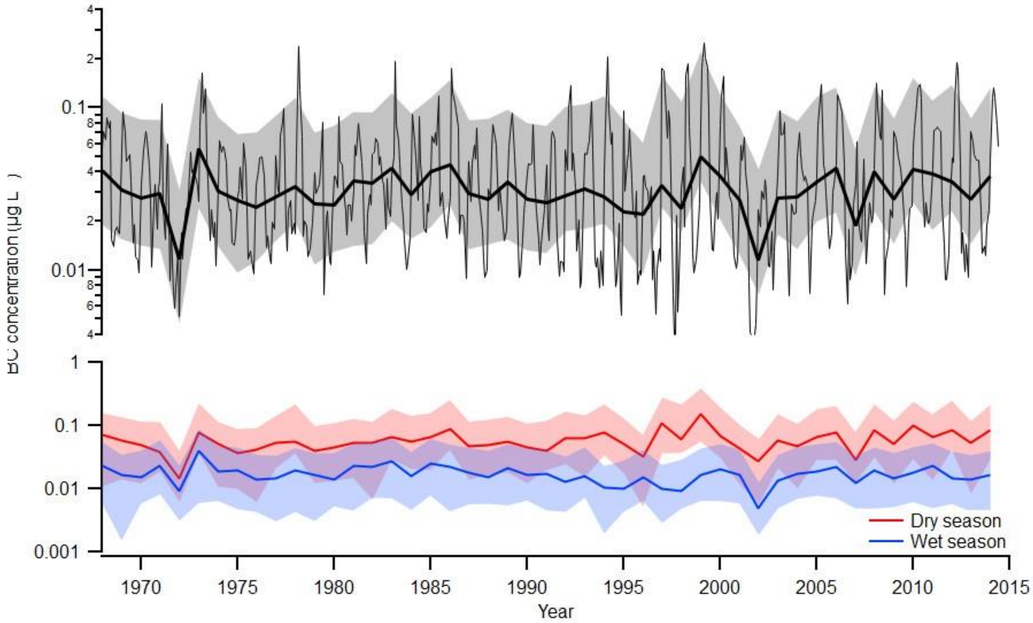


Figure 4. (top) rBC concentrations for the entire core. Black thick line represents annual averages, while black thin line represents monthly values. (bottom) Dry season and wet season average concentrations per year. Note the y axis scale is logarithmic. Shaded areas in both top and bottom represent one geometric standard deviation of the monthly values.

Figures 6, 8: Again, it would be helpful to have an estimate of uncertainty on the seasonal cycles.

> *Added the geometric standard deviation to Figure 6 (for rBC), as below. We decided not to present the Na and rBC seasonality comparison anymore, as there is no point relating Na and rBC now that we are not talking about cycle differences in the spectral analysis.*
>
> *Note that the geometric standard deviation is broad for rBC, which was expected considering concentrations are very low and seasonal peaks vary with time (e.g. the rBC dry season peak in 1999 is much higher than the 2002 dry season peak).*

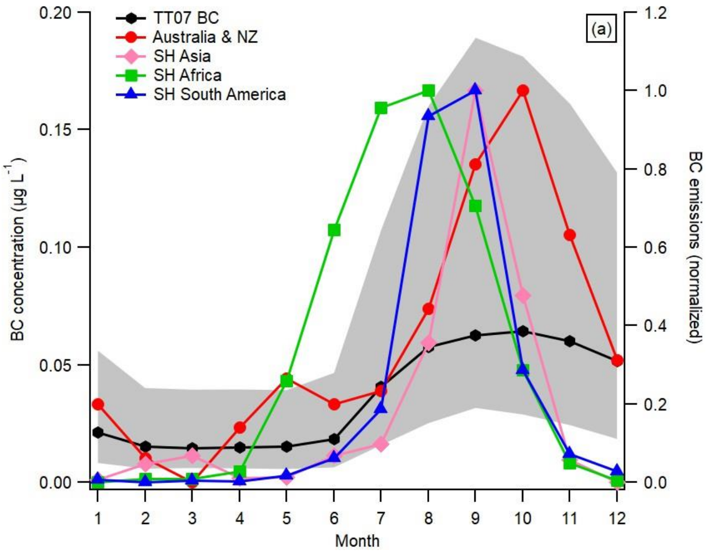


Figure 6(a). TT07 rBC (monthly averages, 1968-2014) and BC emissions estimated from GFED4s for the four SH regions (normalized, 1997-2014). Shaded area indicates rBC geometric standard deviation.

Figure 7: Did you use rBC flux or concentration for the spectral analysis?

> *We used rBC concentration. Added this information to the figure caption.*

Figure 8: Could the broad Na peak over 4 months be a result of mixing/matching Na data and depth picks between the TT07 and Schwanck et al. cores?

> *As mentioned above, we are not comparing Na and rBC cycles anymore.*

Table 4: Please include Pasteris et al. (2014) rBC data.

> *We included Pasteris et al. (2014) as a citation along our manuscript and added their data as comparison in section 4.4. Thank you for pointing this out.*

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.

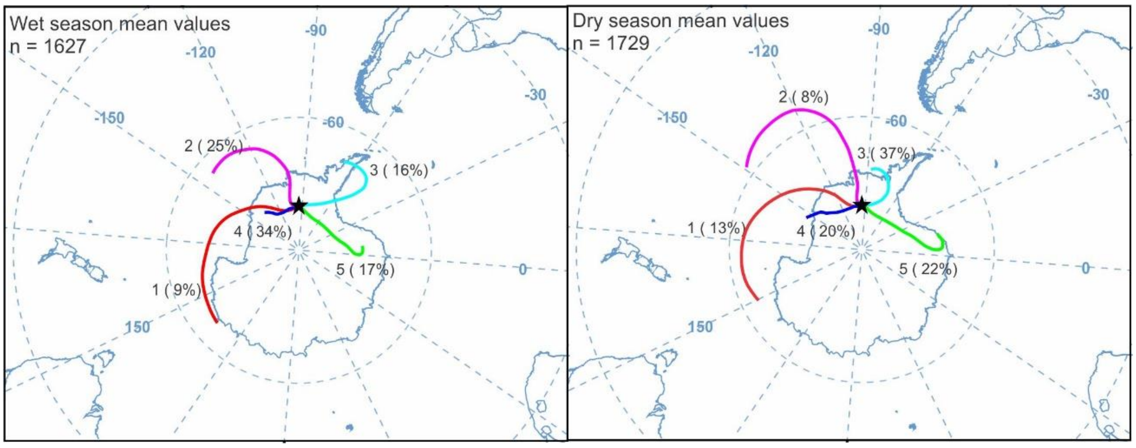


Figure 9. HYSPLIT clusters of 10-day back-trajectories ran every 5 days from 1968 to 2015 arriving at the TT07 drilling site. Results are separated by wet and dry season, and grouped in five clusters (percentage of trajectories for each cluster is shown in parenthesis). Number of trajectories (n) used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

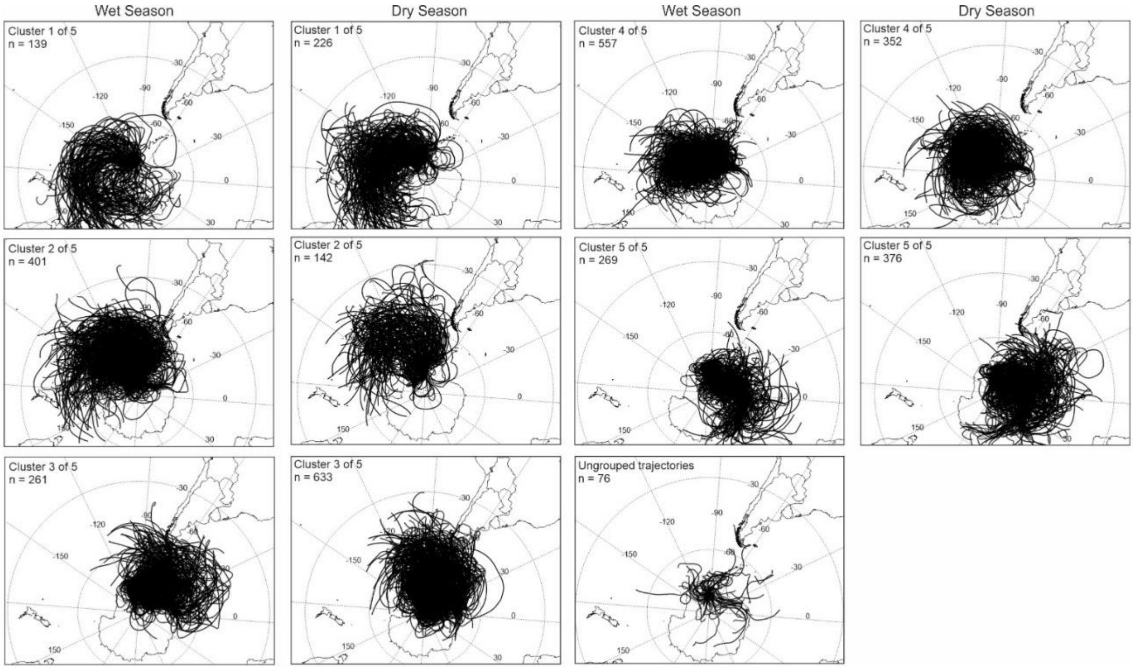


Figure 10. Individual trajectories used for the cluster analysis in figure 8. Number of trajectories (n) used for each cluster is shown at the top, on the left side. Clusters 1, 2 and 4 show air masses arriving from Australia and New Zealand to the TT07 drilling site, while clusters 2, 3 and 4 show the (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.

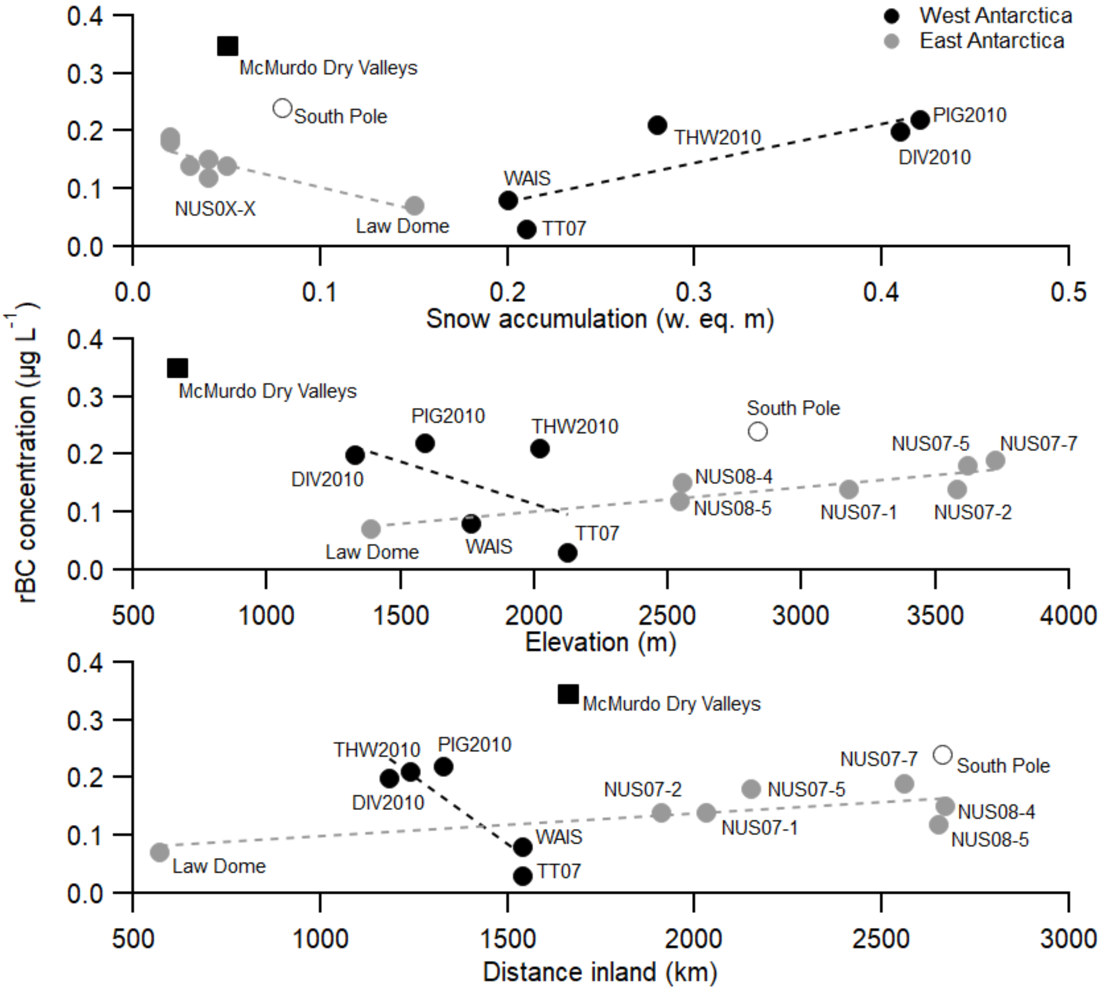


Figure 6. rBC records from Antarctica. rBC concentrations plotted against snow accumulation, elevation and distance from the sea. Solid lines indicate statistically significant correlations ($p < 0.05$), while dashed lines indicate not significant correlations ($p > 0.05$).