Referee#2

General issues:

1) My primary recommendation for improvement is to elaborate on the linkage of BC variability to ENSO. The authors showed that the variance in BC concentration peaks with frequencies similar to those of ENSO, but did not go much further than this. Similar frequencies of variability do not necessarily imply causation, and the authors seem to acknowledge this, i.e., with their statement in conclusions: "These records appear to be influenced by variability similar to tropical Pacific climate variability (ENSO)." The paper would be stronger if something more concrete could be said about this, and a more thorough analysis may produce a clearer picture. For example, there are publicly available ENSO phase/index data back until at least 1950, and likely earlier. Is there any coherence between the BC deposition and the ENSO phase? (i.e., does deposition tend to be greater during El Nino phases?) If so, can these observations be related to a specific ENSO-related emission pattern or transport pathway?

Response

No direct link between ENSO index and rBC records were found in these records, probably because ENSO has by nature, a dual effect on fire potential, by inducing droughts on one side of the Pacific and floods on the other side. Moreover, the delay between ENSO and fire occurrence may be different considering different biome (forest versus grass). We added a sentence to make this point clearer (**see response to minor comment 8**). However, it is very difficult to draw a direct line between ENSO and BC. We investigated the phasing between the ENSO time series and the rBC at WAIS and Law Dome and didn't find any clear trend at such a specific scale.

For better exploration of the link between climatic oscillation we added other panes to the figure1 (figure 3 in manuscript), showing the spectrum analysis of ENSO, AAO and QBO. You can find the figure below:

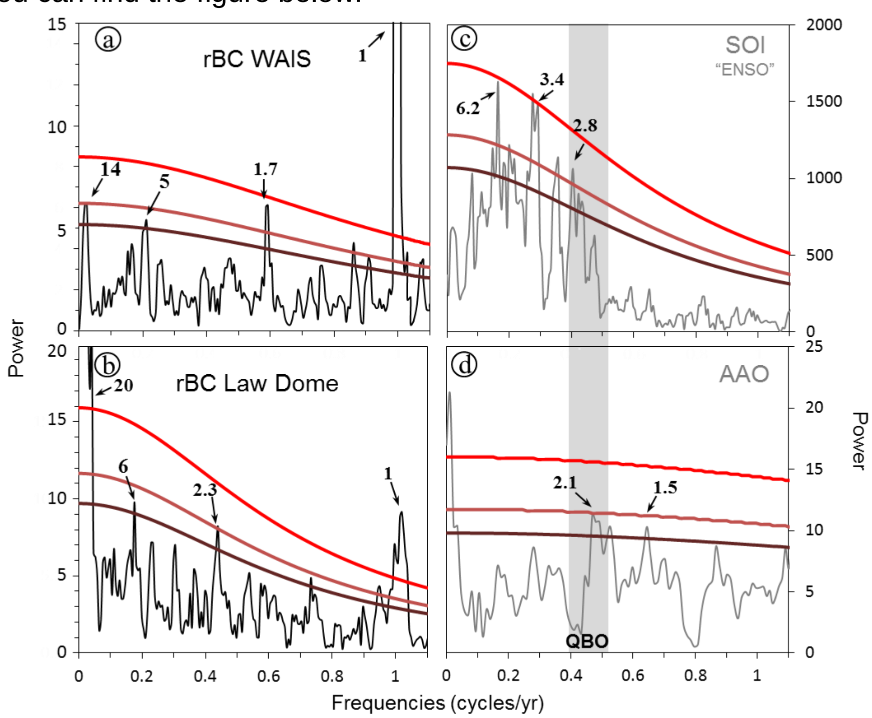


Figure 3: Spectrums obtained by multitaper method, for WAIS (a) and Law Dome (b) monthly rBC records for 1850-2001 period. For reference, spectrums for ENSO (c) and AAO (d) are also represented. The QBO

band (2.3-2.6 yrs band) is indicated as a shaded area. Confidence levels (AR1) are indicated as red lines (90, 95, 99%). Significant periodicities are indicated with an arrow and corresponding values in years are noted. For ENSO, we used the monthly Southern Oscillation Index from 1877 to 2002 from the Bureau of Meteorology, National Climate Centre Climate Analysis Section downloaded at http://www.bom.gov.au/climate/current/soihtm1.shtml. For AAO, we used the BAS NERC dataset from 1957 to 2007 downloaded at http://www.nerc-bas.ac.uk/public/icd/gjma/newsam.1957.2007.txt. For QBO, we used the NOAA/ESRL PSD dataset from 1948 to 2011 (30mb zonal wind at the equator, zonal average), downloaded at http://www.esrl.noaa.gov/psd/data/correlation/qbo.data on Jan/2012

2) Related to (1), the discussion on coherence between the two sites also needs some more detail. Specifically:

- p.27821,23: "These periodicities were coherent..." - Please explain the coherence coefficient and calculation in a bit more detail. What is the meaning of a coherence coefficient > 0.38, and why does this threshold define coherence?

Response

On Analyseries software (Paillard 1996), we used the Blackman-Tukey method and two different filters to get the best spectral coherence for any given frequency. This coherence is a function of frequency with values between 0 and 1, and is a fraction of a common variance between two time series x and y through a linear relation (in other words, a correlation coefficient). Coherence coefficients are given with 3 levels of confidence. Coherence is considered non-zero when coefficients are > 0.38 by the software developer (Didier Paillard, 1996), recommendation that we choose to follow.

In fact, coherence is given at three levels of significance (min, med, max), based on white noise hypothesis and analytically given by the normal shape of the hyperbolic tangent function (tanh). The formula used is dependent upon the determination of a coefficient (0.38 in our case), which is a function of size and shape of the window used, time the inverse function the error function (erf) at desired confidence level (which we choose as medium). The coherence is thus determined as non-zero when this coefficient is > Tanh. The reference used for this is "*Blackman, R.B., and Tukey, J.,W., 1958: The measurement of power spectra from the point of view of communication engineering. Dover Publications, 190 pp*."

- Are the periodicities coherent over 1850-1970? Why was coherence only calculated over 1970-2001? If the records are not coherent prior to 1970, what are some possible explanations for why the level of coherence changed?

Response

We choose a short period of time for coherence analysis for two reasons. First, coherence readability decreases with increased time period (because our records have high temporal resolution), in particular for periodicities > 2yrs. However, note that the delay between annual max of Na and rBC remains similar to that of the 1970-2000 period when looking at the 1850-2000 period. Second, we think that the two records have best chances to be well dated on the top part of the record, and are more reliable. This is why we choose a period where annual cycles were well marked in both cores, for both Na and rBC (e.g. 1970-2000). We thus added this sentence to the text: "*For coherence and phasing, we used a short period of 30 years for better readability of coherence. The period 1970-2001, estimated as the best dated (marked annual cycles), was chosen.*"

- Significant periodicities of 1.7 and 5 years were found at one site, and 2.3 and 6 years at the other site. Given that the temporal resolution of the ice measurements was limited to about 1 year at Law Dome (p.27818,14 and section 2.3), is it possible that these periods are the same in both cores? Can this likelihood be described statistically? (Does the temporal resolution / dating uncertainty factor into the coherence calculation? If not, should it?)

Response

Since these periodicities represent the major oscillations for the whole period 1850-2000, and knowing that the period of return of ENSO oscillates between 2 and 8 yrs, it is possible that those periods are the same. This was actually implicitly assumed. We modified the text, and separated the interpretations on QBO and ENSO to make clearer the discussion:

"Spectral analysis of the rBC records over the 1850 to 2001 period revealed significant periodicities in the 5yrs band at WAIS (AR1 CI = 90%) and 6yrs band at Law Dome (AR1 CI = 95%), Fig. 3 a,b. This suggests that El Niño Southern Oscillation (ENSO) related climate variability may be responsible for some of the intra-annual variability in the records (Li et al., 2011), Fig. 3c. Moreover, the two rBC records were found to be coherent in the ENSO band (average coherence coefficient >0.38 for 1970-2001 period, Fig. SI-5a) confirming a common modulation by ENSO. No ENSO periodicities were found in the WAIS Na record (Fig. SI-3a), suggesting that the ENSO signal found in the WAIS rBC record is likely to be linked to a variability of source emission rather than transport. On the contrary, at Law Dome significant ENSO and AAO periodicities (AR1, >95%) were found in the Na record (Fig. SI-3b), which suggests ENSO and AAO affect atmospheric transport of sea salt to Law Dome (Morgan et al., 1997). Goodwin et al. (2004) also report ENSO and AAO related variability in the Law Dome Na record over the past 700 yr. The study found that early winter Na concentrations (May to July) were highly correlated with mean sea level pressure (MSLP) in the South Indian and southwest Pacific Oceans, and southern Australian regions. Furthermore, Na was found to be anti-correlated with AAO variability and associated with enhanced meridional atmospheric transport. Compared to the Law Dome Na record, the rBC-ENSO periodicities were found to be systematically delayed by 0.3 to 2.2yrs (Fig. SI-4d). The delay suggests that, at Law Dome, ENSO influences the rBC record in a differently than the Na record. This is coherent with the current understanding of fire occurrence in response to changes in rainfall, which is also modulated by ENSO (Chen et al., 2011). For instance, an El Niño event may induce exceptional moisture in South America and prevent fires from occurring notably in forests. On the contrary, an increase in rainfall during La Niña may accelerate vegetation growth in Australian savannahs, increasing fire emissions for several years after the La Niña (Krawchuk and Moritz, 2011). Thus, the link between rBC emissions and ENSO may be related to changes in SH rainfall rather than atmospheric transport. This may explain the delay found between the Law Dome ENSO rBC and Na.

Other significant periodicities were found in the rBC records. At Law Dome, a 2.3yrs oscillation (AR1 CI = 95%) may correspond to the Quasi-biennial Oscillation band (QBO, Fig.3 a,c,d). Since this periodicity wasn't observed in the Na record, we suggest that the QBO is likely to affect rBC emissions in a similar fashion as ENSO through hydroclimate modification (Baldwin et al., 2001). At WAIS, a 1.7yrs periodicity in the rBC record (AR1 CI = 99%, Fig. 3a) and in the Na record (AR1 CI = 90%, Fig. SI-3a), may be associated with the Antarctic Oscillation (AAO, Fig. 3d). This relationship may reflect an influence from atmospheric transport in the mid to high-southern latitudes (Gong and Wang, 1999)."

However, according to a paper by Rhines and Huybers (2011), dating uncertainty shouldn't influence spectrum analysis results. Moreover, for coherence analysis, the calculations are made on Na and rBC which are co-registered (measured in line as the same time in the lab). Thus, dating uncertainty shouldn't influence the comparison between the two records.

(Citation: A. Rhines and P. Huybers (2011), Estimation of spectral power laws in time uncertain series of data with application to the Greenland Ice Sheet Project 2 $\delta^{18}O$ record, J. Geophys. Res., 116, D01103, doi:10.1029/2010JD014764.)

3) It would be helpful to see some more detail/discussion on transport pathways and potential dominant source regions of particles for these two sites. Such detail could come from back-trajectory analysis or reference to other publications that have explored atmospheric transport to Antarctica. A more detailed back trajectory analysis of source regions could incorporate BC emission inventories (such as that used from Lamarque et al, 2010), whereas a simple analysis or discussion of air parcel trajectories would also be helpful.

Response

We've added a paragraph on this topic to the section 3.2:

"Ultimately variability in the ice core records reflects variability in rBC emissions, atmospheric transport, and deposition during transport and physical processes at the ice core site. Stohl and Soderman (2010) developed a 5.5-year climatology (1999 to 2005) for atmospheric transport into the Antarctic troposphere using a Lagrangian particle dispersion model (FLEXPART). The study used rBC emissions described in Bond et al. (2007) and Schultz et al. (2008) and did not include depositional processes. The results of the study suggest that the rBC in the Antarctic troposphere is most sensitive to austral-winter Australian and South American fire emissions as well as South American anthropogenic emissions. Surprisingly, Southern Africa, which, has the largest rBC emissions, had the least potential to influence Antarctic rBC. De Dekker et al. (2010) investigated dust transport from Australia using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT, R. R. Draxler and G. D. Rolph, Hybrid Single-Particle Lagrangian Integrated Trajectory model, 2003). The back trajectory analysis showed that aerosols (rBC and dust), from central Australia may perturb the aerosol mass loading over West Antarctica before circumnavigating Antarctica. By virtue of its location, the Law Dome site should be sensitive to changes in atmospheric transport from South Eastern Africa. We speculate that enhanced meridional transport of African rBC prior to the 1950's may account for the lack of correlation between the records, but more atmospheric further general circulation modelling studies are needed."

4) Related to this: p.27822,25: "... these records may be insensitive to BC emissions transported across the Atlantic sector of the Southern Ocean." This hints that these two sites may not be (frequently) exposed to Atlantic air masses (or that deposition occurs before air masses reach these sites). There must be references or meteorological data showing dominant wind directions or transport pathways to these two sites which could be used to evaluate this idea.

Response

The sentence was removed from text as it no strong bibliographic references were found to support the statement.

Minor comments:

1. p.27816,23: What is the reference for 1.2 W/m2 forcing?

Response

The range of BC forcing was estimated by Ramanathan and Carmichael, 2008 to be from 0.4 to 1.2 W/m2, averaging around 0.9 W/m2. In text, we precise “as high as” to show the top end of the range. However, since Ramanathan and Carmichael, 2008 used Chung et al., 2005’s estimation to evaluate this forcing, we added this reference to the text.

2. section 2.1: What is the context or protocol for the ice core labels (WDC06A and DSSW19K)? After introducing these tags, it would be helpful to simply refer to the two cores/sites with more common names (e.g., "WAIS divide" and "Law Dome").

Response

What do you mean by context and protocol? The core WDC06A, stands for WAIS Divide Core, number 06A. The core DSSW19K stands for 19Km to the West of Law Dome Summit.
We replaced the name tags by WAIS and Law Dome in the manuscript.

3. section 2.1: What are the altitudes of the two sites?

Response

That was in Table 1 but we added the elevations (1766 m for WAIS and 1230 m for Law Dome) to the text.

4. section 3.1: "Concentrations of rBC in both records were lognormally distributed." - This is interesting. What is the geometric standard deviation of these lognormal distributions?

Response

Geometric standard deviation for annual WAIS = 0.42 → 0.05<0.08<0.12 ug/l
Geometric standard deviation for annual Law Dome = 0.73 → 0.05<0.09<0.2 ug/l
These values were inserted into Table 1.

5. section 3.1 : "Geometric means of 0.8 and 0.9 ug/kg." - Are these values mis-quoted by an order of magnitude? They are inconsistent with subsequent text and the means listed in Table 1.

Response

Yes these values were misquoted, we corrected them.

6. section 3.1: "After 1950, concentrations decreased until (about) 1980 and then rose to pre-1950 concentrations." - Looking at Figure 2, the inflection points in both red curves are prior to 1980. (I would say closer to 1970 or 1975).

Response

Text modified accordingly

7. p.27820,25-30: The wording is a bit unclear here.

Response

We modified the text and inserted missing words: *"The DSS record shows an unusual increase in snow accumulation after 1975 associated with changes in zonal atmospheric circulation. However, no significant correlation was found between the DSS snow accumulation record and DSSWK19 rBC (R2 = 0.07, p = 0.41 for annual records and R2 = -0.14, p = 0.08 for 5yrs smoothed records) and the two records appear to be unrelated."*

8. p.27822,2: "However, the rBC signal was found to be systematically delayed from Na by 0.3 to 2.2 yrs." - Why? Do you propose any mechanism to explain this?

Response

The sources of Na are thought to be largely dominated by the open ocean and sea ice. Higher Na concentrations occur when storms lift up salt from the sea, and thus depend on direct modification of climate/weather.

On the contrary, natural rBC is emitted through fires and is thus tightly linked to vegetation. The link between rBC emissions and climate/weather is thus more complicated. A period of increased moisture may accelerate grass growth, which may burn the following year and thus increase rBC emissions with a delay. On the contrary, in the case of rain forest, increase precipitations in the wet season may decrease fires in the dry season, and again, affecting rBC emission with a delay.

The fact that rBC signal is delayed from Na suggests they are not affected by the same processes, although the cause of these processes may be the same (for instance, modification of storm tracks and intensity as a function of ENSO phase).

We modified the text to make clearer the idea behind this sentence: "*Compared to the Law Dome Na record, the rBC-ENSO periodicities were found to be systematically delayed by 0.3 to 2.2yrs (Fig. SI-4d). The delay suggests that, at Law Dome, ENSO influences the rBC record in a differently than the Na record. This is coherent with the current understanding of fire occurrence in response to changes in rainfall, which is also modulated by ENSO (Chen et al., 2011). For instance, an El Niño event may induce exceptional moisture in South America and prevent fires from occurring notably in forests. On the contrary, an increase in rainfall during La Niña may accelerate vegetation growth in Australian savannahs, increasing fire emissions for several years after the La Niña (Krawchuk and Moritz, 2011). Thus, the link between rBC emissions and ENSO may be related to changes in SH rainfall rather than atmospheric transport. This may explain the delay found between the Law Dome ENSO rBC and Na."*

9. p.27822,14: "Similar temporal variability does not occur in the emission inventory of SH forest fires." - Please elaborate on this. Is the variability in emissions less than that seen in the ice core?

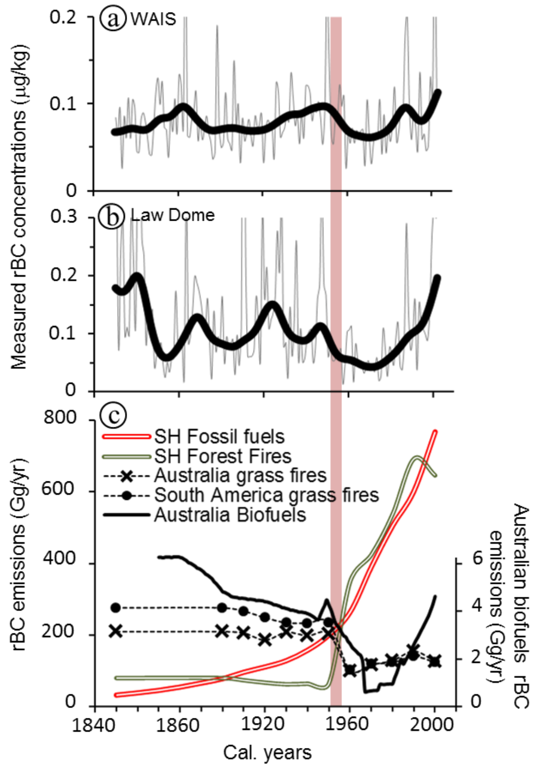
Response

We changed the text to add the precision requested (increased SH forest fire emissions since 1950): *"Similar temporal variability does not occur in the emission inventory for SH forest fires which show an opposite increasing trend from the 1950's (Mouillot and Field, 2005). However, we note that recent estimates of SH rBC emissions (GFED 3, van der Werf et al., 2010) attribute ~41% of SH rBC to grass fires (61% if woodlands are included), ~ 9% from forest fires and 26% from deforestation (primarily in South America). Thus, changes in the SH grassland (and woodland) fire regime from human activity and climate (hydroclimate) could dominate the SH rBC distribution."*

10. p.27822,21: "Emissions from SH deforestation, forest fires, and fossil fuel combustion increased markedly after 1950 (..., fig 4)." - Fig 4 actually only shows the increase associated with SH fossil fuels. It would be helpful to also show SH forest fire / deforestation emissions on this plot.

Response

Figure modified accordingly, see below:



11. Table 1 caption: "... annual concentrations are calculated from the log values of monthly data..." - Why are log values used to calculate the mean?

Response

Since the distribution of concentrations is log-normal, a mean value has to be estimated from log values, in order to represent all values (and not being biased by higher values). In other words, this is a “geometric mean”.

12. Table 1 caption: "out-layer" -> outlier ?

text modified accordingly.

13. Figure 1 caption: Annual smoothing is shown in the thick line

text modified accordingly.

14. Figure 2 caption: Maybe reword "decimal"?

decimal replaced by “monthly”