Atmos. Chem. Phys. Discuss., 11, C14455–C14470, 2012
www.atmos-chem-phys-discuss.net/11/C14455/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Large scale changes in 20th century black carbon deposition to Antarctica" *by* M. M. Bisiaux et al.

M. M. Bisiaux et al.

marion.ma.bisiaux@gmail.com

Received and published: 18 January 2012

Referee 1 comments are answered point by point below. A pdf combining all responses and figures is also attached.

1- A more accurate title would be: Investigations of past changes in black carbon deposition in Antarctica using two ice core records.

Response: We agree to change the title. This could be: "Changes in black carbon deposition to Antarctica from two ice core records, A.D. 1850-2000". Or "Changes in black carbon deposition to West Antarctica and Coastal East Antarctica from two ice core records, A.D. 1850-2000"

2- The abstract should highlight the differences between the two ice core records (or

quantify their coherence), and mention the changes from 1950 to 1990 in the perspective of earlier changes, showing large decadal variability.

Response: We added correlation coefficients to the abstract, and modified the text:

"Concentrations of rBC in the ice cores displayed significant variability at annual to decadal time scales, notably in ENSO – QBO and AAO frequency bands. The records were uncorrelated from 1850 to 1950, but were highly correlated from 1950 to 2002 (cross-correlation coefficient at annual resolution: $r=0.54$, $p<0.01$) due to a common decrease in rBC variability. The decrease in ice-core rBC displays similarities with inventories of SH rBC grass fires and biofuel emissions, which show reduced emission estimates from the 1950's to late 1980's."

We also added this sentence to section 3.1, to highlight again the coherence: "cross correlation coefficient for annual data: $r=0.54$, $p<0.01$; for monthly data: 0.20, $p<0.01$."

3- Some parts of the introduction should be revised. Text in Page 27817, lines 7 to 11 does not read easily. I would suggest to first mention findings from northern hemisphere (Greenland, Tibet ice cores), and then differences expected in the southern hemisphere regarding the sources of BC.

Response: We moved the sentence at the end of the introductory paragraph as suggested and modified the phrasing at the end of the paragraph: "In the Southern Hemisphere (SH), rBC emissions are primarily from dry-season biomass burning in Australia, southern Africa and South America (Mouillot and Field, 2005). However, while a number of paleo-biomass burning records have shown centennial scale variability in biomass burning, high temporal resolution rBC records have not been reported (Marlon et al., 2008; Wang et al., 2010; Falk et al., 2010; Whitlock and Tinner, 2010). Here we show the first high resolution rBC records from ice cores of two disparate regions of Antarctica, covering the period 1850-2001, and investigate the connexions between rBC deposition and SH biomass burning and climate."

4- There is a missing section on the investigations of back trajectories of air masses transported to Law Dome or WAIS sites, which could be useful when discussing causes for differences (and the importance of air masses from different ocean basins) (e.g. Reijmer et al, J. Clim., 2002 albeit not for the two sites investigated here).

Response: Added the following paragraph to section three:

"Ultimately variability in the ice core records reflects variability in rBC emissions, atmospheric transport, 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 further general circulation modelling studies are needed."

5- In Sections 1 or 2, the reader should be guided to understand the choice of the two investigated ice cores. While the WAIS ice core offers seasonal resolution and accurate dating, the choice of DSSW19K remains more difficult to understand, as dating is more uncertain, and post deposition effects limit the temporal resolution of the

record. Section 2.1 should introduce the other DSS ice cores used to guide the dating of DSSW19K. Section 2.2 should summarize the information from the appendix, and particularly quantify the uncertainty associated with the analytical method.

Response: The two sites were chosen as two high resolution sites, located in two disparate regions of Antarctica and presenting different characteristics. In section 2, we modified the text as follows: "Two high temporal resolution ice cores were chosen for this study and included: the core WDC06A from West Antarctica (referenced as "WAIS" core further in text) and DSSW19K from Law Dome in East Antarctica (referenced as "Law Dome" core further in text), Table 1." Further down we also characterize Law Dome as a more "coastal site". Even if the temporal resolution at Law Dome is not as high as at WAIS, it does remain a very accurately dated core, with regard to other ice cores drilled in East Antarctica (cf, Figure 1 and SI-1, with annual cycles discernible visually almost all years). Thus, we modified the "dating" paragraph, to stress this accuracy. We also added precisions on the other DSS core in this section:

"Although annual cycles were extremely well preserved in the WAIS record, the measurements at Law Dome lacked unambiguous sub-annual markers that could have been tied to specific calendar dates, in part because the net snow accumulation rate approximately equalled the height of the local surface roughness (Figs. 1 and SI-1). However, cross comparisons of continuous high resolution S, Na, and Cl measurements from another Law Dome ice core (DSS0506), were used to confirm the annual layer counting. This DSS0506 core, drilled near the Law Dome summit (DSS) in 2005 in a higher snow accumulation zone and analysed in a similar fashion but not for rBC, contained distinct annual cycles in many chemical species, and dating was unambiguous. Over the 150yrs period, we estimate the dating uncertainty to be less than a year for the WAIS core and to be about a year for the Law Dome core."

All dating information was moved from the appendix to main text.

The analytical average uncertainty associated with the method is 15%. This was added to the rBC ice core analysis section and to the captions of Figures 1 and 2.

5.b- Aerosol records from Antarctica are known to be characterized by a significant deposition noise. Is there any information available, related to the signal to noise level of rBC records between nearby ice cores at the same location?

Response: To investigate the signal to noise level in these records in systematic way, we would need an array of ice cores in nearby sites. Overlapping sections of ice from the same ice core were determined on a regular basis and show that the small-scale variability (cm) is within $\sim$ 15%. A systematic study of rBC deposition noise is planned for Law Dome sites and will allow a robust determination of the signal to noise ratio, with respect to accumulation rate and surface processes. However, we added a third pane to the Figure 2, showing a stacked rBC record constructed from the two Z-scores of WAIS and Law Dome ice core records. We also added the standard error to give a sense of the common variability between the two signals. This text was added to the manuscript: "Figure 2c shows the single stacked rBC record reconstructed from Z-scores of WAIS and Law Dome ice core records. Both sites displayed significant annual to decadal scale variability prior to 1950, and common variability with low standard error from 1950-onwards." See figure 1 (figure 2 in manuscript), now with three panes.

6- Section 3.1 discusses mean concentrations and fluxes in the two ice core records. Some sentences are difficult to understand, such as "The DSSW19K rBC concentrations were less variable. . ." and "The DSSW19K rBC concentrations were more variable. . .". Please explain what is compared to what. How are results from two ice cores integrated over the whole continent?

Response: We re-wrote this part of section 3.1 as: "The seasonal variability (summer/fall and winter/spring) of rBC concentrations at Law Dome was less than at WAIS, with an average intra-annual range of 0.08 μg/kg at Law Dome and 0.12 μg/kg at WAIS. In contrast, year-to-year variability of annual averaged (geometric) concentrations was greater for Law Dome than WAIS, despite a comparable annual geometric mean of

$\sim$0.08 μg/kg for the entire record (Table 1)." (see geometric standard deviations in Table1).

The integration calculation was based on too many assumptions and we removed it from the text.

7- Please show estimates of rBC fluxes for the two records (only concentrations are shown).

Response: Should these be to the figures? Or in the Table? The fluxes have same variability as annual concentrations (see Figure 2-supp), we decided not to show the fluxes on the graphs, finding them quite busy already.

8- Section 3 should be reorganized with 3.1) mean concentration and fluxes, 3.2) temporal variability and comparison with the variability of accumulation and Na fluxes, 3.3) relationships with ENSO, and 3.4) comparison with SH rBC emission inventories.

Response: We agree and made changes accordingly.

9- The comparison between Na and rBC records needs to be written more clearly. One may first compare the mean seasonal cycles, and then the temporal variations for annual mean values. Investigations of coherency at the inter-annual or decadal scales are not discussed.

Response: We added this sentence to the text: "Na is primarily delivered to the Antarctic in the winter/spring {Sneed, 2011}, with a strong seasonality. The records of rBC and Na at Law Dome and WAIS were found to be autocorrelated due to the presence of an annual cycle in both species." The coherence of Na with rBC at interannual time scale is discussed in the ENSO paragraph. However, further discussion on the variability of Na and rBC at inter-decadal time scale wasn't included in the manuscript because we didn't find common periodicities for periodicities of such time periods.

10- Regarding the inter-annual variability, systematic comparisons with SAM and ENSO power spectrum, and coherency and phase analyses should be conducted each rBC record and indices of modes of variability. Why didn't the authors also investigate the relationships with regional sea ice information, from 1979 to 2001?

Response: We've now added power spectrum from SOI (ENSO), AAO (SAM) and QBO to the figure 3.

The text was also modified to discuss all the multi-annual periodicities:

"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 (AR1, >95%) ENSO and AAO periodicities 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 Amer-

ica 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)."

Regarding sea-ice: No significant correlation was found between the Law Dome MSA proxy record of sea-ice extent from Curran et al. (2003) and the Law Dome rBC record. Other studies have shown that the correlation between MSA and sea-ice extent is only valid for certain regions (Abrams et al., 2007). The relationship to sea-ice extent was therefore not investigated. Instead the study focused on Na as a co-registered proxy for changes in meridional transport.

11- What are the analyses supporting statements such as "The time lags between the ice core records and rising emissions in the inventories. . . suggest that these records may be insensitive to BC emissions transported across the Atlantic sector. . ." ?

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

12- Causes for differences in the two ice core records prior to 1950 should be discussed.

Response: We added a paragraph discussing rBC variability prior to 1950: “On the other hand, during the earlier period of the records (1850-1950), the two series don’t share the same variability, and little evidence of anthropogenic disturbance of rBC emissions in the SH has been documented (Mouillot and Field, 2005; Lamarque et al., 2010). Even if coal mining and burning had started to grow at the end of the 19th century in Australia and South Africa (Vallelonga et al., 2002 and references therein), it was shown that this was probably not a pollution source for Lead in Antarctica (Vallelonga et al., 2002), and therefore not likely to be a source of rBC either. We thus suggest that the rBC signal for this period is closer to that of natural variability, maybe influenced by ENSO, and which anthropogenic fire suppression and fossil fuel combustion have overwhelmed since the 1950’s.” Also, see response to major comment 4.

13- In the discussion of ENSO impacts on ice core rBC records, a further discussion of transport versus source effects would be appreciated.

Response: We added a paragraph to the text for this purpose, which is included in the response to major comment 10 (please, see above).

Minor comments: 1- Table 1 should be reorganized to place all Antarctic information together, for an easier comparison.

Response: Changes made accordingly.

2- Figure 1 should show the accuracy of measurements. Figures 1 and 2 may also show estimates of fluxes (not only concentrations).

Response: See response to major comment 7: Since fluxes have same variability as annual concentrations, we decided not to show the fluxes on the graphs, finding them quite busy already. We could have added the overall flux values to the table 1, but we don’t have any other flux to compare them with, so we choose to rather keep it in the text only. Accuracy was added to the caption and in the text.

3- How much of the power spectrum of concentrations is related to that of accumulation? Figure 3 should also include the power spectrum of ENSO and QBO

Response: See the Figure 4 below and response to major comment 10. We calculated MTM for annual accumulation records at both WAIS and Law Dome. Most of the variability was in the 2 to 3 years band (we don’t sub-annual accumulation). For WAIS accumulation MTM was strongly different than rBC MTM. For Law Dome, there was some similarity in the 2.3yrs band (0.45), which may have suggested an effect of QBO on accumulation. However, the dating at LD is not a reliable as at WAIS, since it relies on cross-comparison with another ice-core. Overall, it is very difficult to link accumulation and rBC, in part the deposition processes of rBC (wet/dry) are not well known yet and thus, we prefer to leave this discussion out of the paper.

4- Figure 4 shows emissions from fossil fuels and grass fires on different vertical scales. Do I understand correctly that Australian biofuel emissions are two orders of magnitude smaller than the other rBC emission sources? What is then the relevance of showing them? Could the authors include a more quantitative discussion of the coherency between the Antarctic ice core rBC decadal variations, and the rBC inventories, given transport aspects?

Response: The atmospheric transport study of Stohl and Soderman (2010) showed that Antarctic rBC may be extremely sensitive to Australian emissions even thou current emissions are orders of magnitude less than those of Africa etc. Also see response to major comment 4.

5- Appendix: some statements are wrong; such as “the two ice core records have monthly to seasonal resolution” (this is not the case for the DSS record as discussed in the main text).

Response: Text modified accordingly, and all information on dating was moved to main text.

6- Appendix, Dating: what is the uncertainty on the DSS ice core accumulation?

Response: The uncertainty at both Law Dome and WAIS ice cores is estimated to be 3.1 cm wep per year. This was determined by several studies notably at South Pole and at WAIS. In the studies, measurements with multiples shallow cores and hundreds of snow stakes revealed that uncertainty in a single year's net snow accumulation was ∼3.1 cm weq per year over a wide range of annual net snowfall amounts and temperatures. This is the result from spatial variability in accumulation. We modified the SI text to precise that this estimation is for both Law Dome and WAIS cores. Paper references: âĂć McConnell, J. R., Bales, R. C., and Davis, D. R.: Recent intra-annual snow accumulation at South Pole: Implications for ice core interpretation, J. Geophys. Res., 102, 21947-21954, 10.1029/97jd00848, 1997. âĂć McConnell, J. R., Mosley-Thompson, E., Bromwich, D. H., Bales, R. C., and Kyne, J. D.: Interannual variations of snow accumulation on the Greenland Ice Sheet (1985-1996): new observations versus model predictions, J. Geophys. Res., 105, 4039-4046, 10.1029/1999jd901049, 2000 âĂć Banta, J. R., McConnell, J. R., Frey, M. M., Bales, R. C., and Taylor, K.: Spatial and temporal variability in snow accumulation at the West Antarctic Ice Sheet Divide over recent centuries, Journal of Geophysical Research, 113, D23102, 10.1029/2008jd010235, 2008.

7- Appendix, Spectral Analysis: none of this is new, I would suggest to remove this. However, the authors should justify the choice of 21 year running analyses.

Response: We simplified the text, but kept some precision on the parameters we used for SSA. Some of the spectral analysis work is now described in the main text under "data analysis", to respond to the referee#2 comments. We choose 21yrs because it was the longest average we could use (reproduced at least 5 times in the 150yrs period), and still capturing the drop in the 50's and increase at the end of record.This sentence was added to text: "Smoothing was estimated with an R implementation of Nadaraya-Watson kernel regression for a period of 21yrs, long enough to be repeated at least five times in the 150yrs period, but still capturing main variability, notably the in the top part of record."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C14455/2012/acpd-11-C14455-2012-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 27815, 2011.

Figure 1 (Figure 2 manuscript)

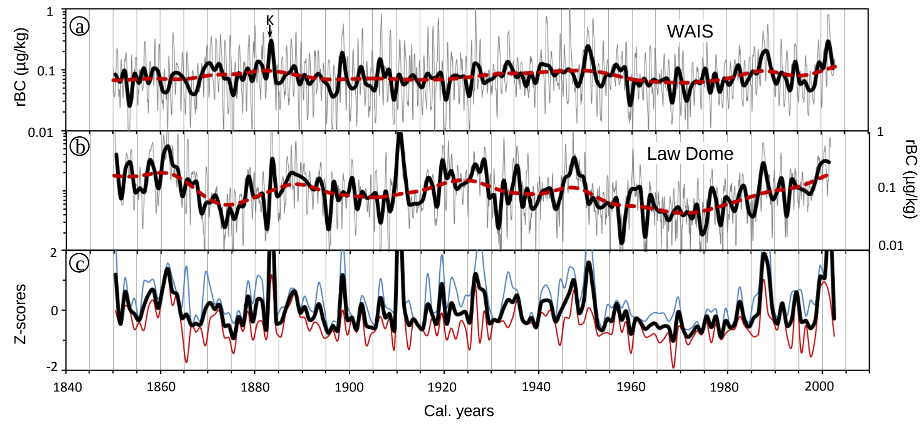

Fig. 1. rBC concentrations in monthly resolution (dots and thin line), and resampled to annual (thick line) for WAIS (a) and Law Dome (b). Red dash line is 21yrs smoothing. K marks the Krakatoa volcanic erupt

Figure 2-Supp

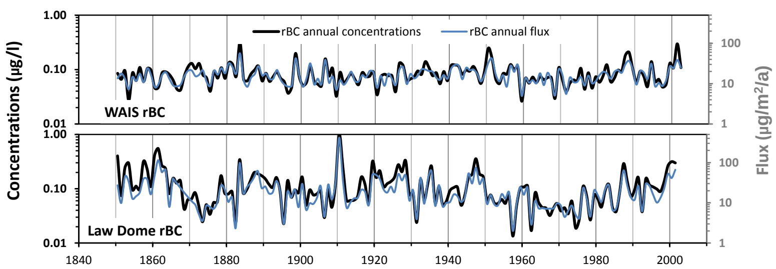

Fig. 2. Concentrations (black) versus fluxes (blue) of rBC at Law Dome and WAIS

Figure 3

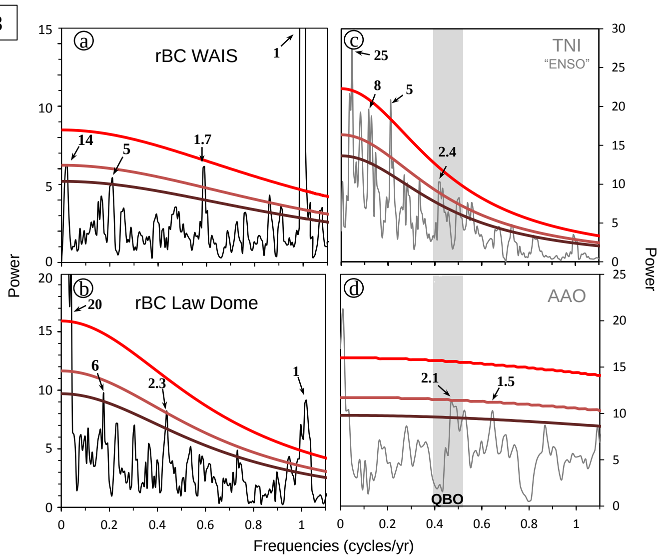

Fig. 3. Spectrums obtained by multitaper method, for WAIS (a) and Law Dome (b) monthly rBC records for 1850-2001 period. For reference, spectrums for ENSO (c) and AAO (d) are also represented. The QBO band (2

Figure 4 Supp

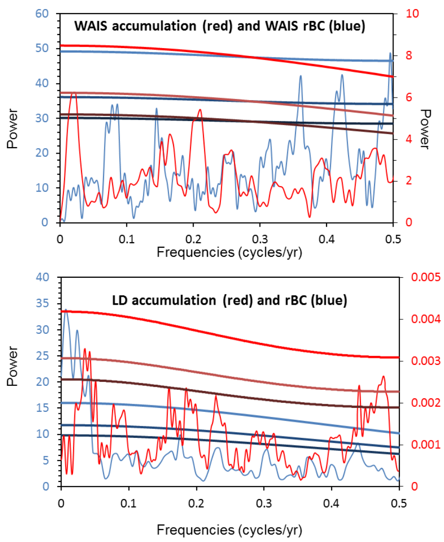

Fig. 4. Comparisons of Multi Tapers of accumulation (red) and rBC concentrations (blue) at Law Dome and WAIS