

## ***Interactive comment on “Long-term trends of black carbon and sulphate aerosol in the Arctic: changes in atmospheric transport and source region emissions” by D. Hirdman et al.***

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Dear Anonymous Referee #2, Thank you for your positive comments on our paper.

We have taken all your comments into consideration and hope that you will find our response and changes to the manuscript satisfying.

Introduction: “It is mentioned that Zeppelin has two EBC stations with contradictory results about recent trends. It is not really clear which one is chosen and why are results different?”

We have now rewritten section 2.1.2 in order to clarify which EBC data set from Zep-

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pelin has been used. “At Zeppelin, the measurements which are analyzed and presented in this study have been performed since 2002 using a custom built PSAP that is based on the same measurement principle. The BC study by Eleftheriadis et al. (2009) used data from an aethalometer running in parallel with the PSAP.” We thought it more suitable to specify this in the data description section rather than to raise this issue in the introduction. A few of the reasons why the results are different might be that the instrumentation used are different as well as the fact that the time period over which the trends have been calculated differ in the two studies.

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Section 2: “is very complete except may be a better readability of section 2.3 by using examples as explained before. (The methodology based on a cluster analysis is one of the major achievement and this is explained in section 2.3. It is however the most difficult part to read mainly because a lack of some examples to illustrate the meaning of the 4 clusters which are identified in section 3.2. I am wondering if a (l,n) plot for a given example would help to clarify how the cluster analysis works, l being the geographical region of fig.1 and n a set of measurement times.)”

We agree that section 2.3 is rather difficult to read as it contains many technical details of the method. However, we have chosen not to include more example figures of the different steps of the cluster analysis. The paper is already very long and adding figures (and corresponding text) regarding these rather technical features would make the paper probably more difficult to read and would extend the long paper even more. The attached figure below is an example of a figure which we have chosen to exclude from the submitted version of the manuscript to condense the paper and avoid too much technicalities. It displays a step in the analysis where the four different transport clusters just have been identified as composed by the average influence from each of the predefined geographical regions. On the x-axis are the 9 different geographical regions as defined in Figure 1 in the manuscript (1=EU; 2=SNE; 3=EA; 4=CGA; 5=US; 6=TR; 7=AO; 8=NAO; 9=NBO). The y-axis displays the influence of the different

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regions relative to the mean atmospheric transport.

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"The bias in the Barrow record due to data filtering is mentioned, but how will this change the results? The instrumentation change is pointed out in the difficulty to identify trends but not the influence of this data filtering."

We have added the following explanations to the paper: "This screening effectively excludes direct transport from most North American source regions. Thus, the statistical analysis of North American source regions of BC for Barrow relies on indirect transport events, which are likely associated with larger errors in the simulated transport. In addition, signals of BC from source regions both in North America and on other continents are likely often mixed during such indirect transport events, which make a clear identification of source regions in North America more difficult."

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"In section 3.2 line 6 better define what you mean by "number of cases" (measurement times)?"

A case is represented by a model calculation with an associated measurement where both components are averaged over the same time span. We have now clarified this even more in section 2.3 line 16-17: "Here,  $i$  and  $j$  are the indices of the latitude/longitude grid and  $n$  runs over the total number of cases  $N$ , where each case resembles a model calculation linked with a corresponding measurement and where both are averaged over the same time interval."

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Section 3.1. "This a very interesting section. The large number of cases for AO means that local processes (removal, aerosol transformation, local sources) are very important and that measured BC aerosol could be quite old since their emission from midlatitudes. This could be somewhat more recognized. Information about season dependent

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processes are given but not derived from given figures (line 24 p. 12146, line 4-6 p. 12147, line 23 p. 12147, : : :)"

We have discussed the content of the lines above, but we are uncertain of what you are trying to point out. Section 3.1 and figures 2-4 discuss the transport climatologies for the three different stations in this study, the seasonal differences in this transport as well as tendencies in this transport when comparing one five year interval in the beginning of the studied time period with a five year period from the end of the time period. We do not try to identify any source regions in this part and therefore are no measurement data used in this analysis. In the updated version of the manuscript we have added a discussion concerning the influence from removal processes in section 2.3: "However, trends in precipitation or other scavenging processes along the major transport pathways the stations may also influence the trends of the measured species concentrations for particular clusters. Such trends (New et al., 2001; Adam and Lettenmaier, 2008) are not homogeneous across the Arctic and their overall impact on aerosol wet scavenging has not been studied. Furthermore, the trends of acidifying species in Arctic precipitation agree well with reported emission trends (Hole et al., 2009), whereas changes in removal efficiency seem to have a smaller influence on the reported deposition values. In this study we therefore assume that the influence of temporal trends in scavenging processes is not significant allowing us to separate the effects of changes in atmospheric transport to the Arctic from effects of emission changes in a few important source regions, on the Arctic EBC and sulphate concentrations."

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Section 3.2 From this section it is clear that the season is an important parameter.

Yes, season of the year is an important parameter. However, since the seasonality of the transport of different short-lived species was presented in great detail in our last paper (Hirdman et al. 2010) we felt that we can keep the discussion on seasonal

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differences at a minimum here. Trends for individual seasons are very interesting and we have done such a seasonal analysis. We considered including these results in the paper but finally decided against it as consistently including seasonal trends for all species and all stations would have made the paper much too long. Reporting only certain results would have been very confusing for the reader, so we removed all seasonal trend results from this paper. We consider reporting the seasonal trends in a future paper.

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Section 3.3 "The trend of the cluster frequency is indeed a good way to discuss the long term transport variability. It is surprising that the Barrow and Alert trends are not more similar considering their common sensitivity to the NAO. Influence of the data filtering?"

Yes, the data filtering might have an influence here. However, we have not tried and therefore do not know if it is possible to quantify the magnitude of this influence.

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"The fact that no trend in Zeppelin is related to the shorter period is not so clear because it is only shorter by 5 years compared to the longest Alert record. Is there a trend for the summer period considering the correlation with NAO in summer?"

The great interannual variability in the frequency in all four transport clusters identified at Zeppelin is seen not only for the yearly mean but also during all four seasons. None of the clusters display a statistically significant trend in any of the seasons.

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Section 3.4 and 3.5 "In these sections, yearly trends of EBC and sulphate are recalculated using the cluster analysis to compute an annual mean. The aim is I believe to identify regional source changes responsible of the observed yearly trends. In fact some information about the expected results of this methodology should be given at

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the beginning of section 3.4 ( see for example the introduction of section 3.6 which nicely explains the methodology used in this section). Fig. 12 is very illustrative of the importance of the NE sources in the EBC variability in Alert and ENE in Zeppelin."

We have added a new introduction part to section 3.4 where we briefly describe the aim of the analysis that follows: "Identifying the different clusters representing distinct regimes of atmospheric transport arriving at the three stations was a crucial step in our analysis to investigate the trends of EBC and sulphate. In this section we calculate the annual mean concentrations associated with each of the clusters to reveal differences in measured concentration levels for the various source regions. We then determine the trends in the measured concentrations for each of the clusters in order to relate these trends to emission changes in the respective source regions."

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"Sulphate variability at these stations (Fig. 14) exhibits the same kind of results. But the trends are quite different for both species in table 3. In fact I am confused by the differences between table 3 trends and trend lines shown in fig. 13 and 15. I do not see positive sulphate trends in fig. 13 and 15 for Alert."

Thank you for pointing out this error. The sulphate trends at Alert should be negative as seen in figure 15. This has now been corrected in Table 3.

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"Even though there is a short record for EBC in Zeppelin, sulphate and EBC interannual variability are quite different for this time period, could it be related to differences in the ENE cluster frequency?"

The relative cluster frequency is the same independent of the species considered. The difference in interannual variability seen in EBC and sulphate at Zeppelin must therefore have another explanation such as the difference in emission sources and their interannual variability.

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Section 3.6 “In this section the cluster frequency interannual variability and the cluster annual means are combined to derive more meaningful calculated trends which can be compared with the observed trends. The most important result is the large influence of the AO cluster because of its high frequency. It again indicates the importance of the local source and sink trends. I am confused by the author’s statement (line 13 p. 12155) that circulation do not drive the trend. In fact trends could be quite different if there were not the compensation between the AO and NA frequency for Alert. So the measured trend in this station is not just a mirror of the source trend.”

We do recognize the importance of changes in atmospheric transport to the stations as explained in section 3.1 as well as in the previous paper (Hirdman et al. 2010) where we focused on the seasonal variability. However, from our analysis we come to the conclusion that long-term trends in the atmospheric circulation are not large enough to have a major impact on the measured species and may therefore only explain a minor fraction of the observed trends. If we assume that the long-term trends in the scavenging processes such as precipitations are negligible (see added information on this issue in section 2.3), we find that observed trends mainly represent changes in emissions from the dominant source regions.

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It may be also interesting to show the Zeppelin sulphate trend for the shorter time period corresponding to the EBC record for the purpose of comparison. In the same way, the Zeppelin cluster frequency for the EBC shorter record might be also useful to show (Fig. 11).

In order to have as statistically robust results as possible we choose to calculate the trends for the complete data set available for each of the species individually.

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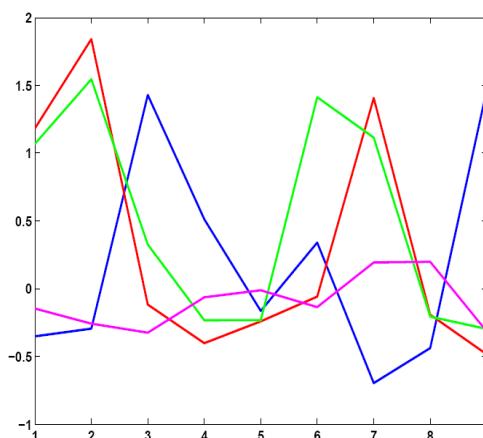


Figure XX. Geographical influence on the four transport clusters from the 9 predefined geographical regions relative to the mean atmospheric transport. On the x-axis are the 9 different geographical regions defined as shown in Fig. 1 in the manuscript: Europe (1); Siberia and North-East Asia (2); Eastern Asia (3); Canada, Greenland and Alaska (4); USA (except Alaska and Hawaii) and Central America (5); land masses in the tropics and in the southern hemisphere (6); Arctic Ocean (7); North Atlantic Ocean (8); and North Pacific Ocean (9). The y-axis display the relative influence of transport from these regions compared to the mean atmospheric transport.

**Fig. 1.**

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