

***Interactive comment on “A trajectory analysis of atmospheric transport of black carbon aerosols to Canadian High Arctic in winter and spring (1990–2005)” by L. Huang et al.***

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Reply to Referee #3

The authors wish to thank the referee for the constructive, and thorough review, and suggestions that have led to improvements in this manuscript.

1) Abstract, line 1 -2: Change to: “Black carbon (BC) particles accumulated in the Arctic troposphere and deposited ON snow HAVE BEEN CALCULATED to have significant effects on radiative forcing of Arctic regional climate.” This wording should be used throughout the paper.

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This correction has been made.

2) Abstract and throughout: Emission intensity is never defined.

A statement has been added to the 2nd paragraph of the introduction to describe the BC emission dataset used in this study. "The annual BC emissions used in this study are based on the BC inventory prepared by Sharma et al. (2004;2009)."

Detailed distributions on BC emissions used in this study are provided in Section 2.3.

3) Abstract, line 6: Transport frequency relative to what? For different geographical regions? Please define this in the abstract.

Following the calculation of 10-day back trajectories arriving at Alert, the cluster analysis technique was applied to identify distinct clusters (or atmospheric transport pathways). The frequency of atmospheric transport to Alert taking a specific pathway in January (April) was estimated by the fraction of trajectories grouped into the relevant cluster relative to the total number of trajectories in January (April), 1990-2005.

The potential source regions affecting Alert are mentioned in the abstract, which include North America, Europe, and former USSR.

4) Abstract, line 9: Why only January and April? Have any trajectory analyses been done that demonstrate that these months are representative of winter and spring or just January and April?

The focus of the current study is to investigate the inter-annual variations of BC observed at Alert. The use of January and April data instead of DJF (for winter) and MAM (for spring) was to minimize the seasonal transformation of atmospheric transport patterns in the whole seasons of interest and to emphasize the inter-annual variations within each dataset.

Although no trajectory analyses have been done for the whole seasons, January and April trajectories are considered representative to winter and spring, respectively. This

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is because that long-range atmospheric transport to Alert investigated in this study is largely controlled by the near-surface circulation in the northern high latitudes. The seasonal characteristics of near-surface circulation have been revealed, for instance, by analyzing mean sea level pressure for the four mid-season months (i.e. January, April, July, and October) over the period 1970-1999 (Serreze and Barry, 2005).

5) Abstract, line 12 – 13: It would be useful to add a sentence saying “Other factors, such as deposition, could also contribute to the variability in BC concentrations but were not considered in this analysis.”

It has been added to the revised manuscript.

6) Abstract, line 22: “It is also found that the change in Eurasian contributions...” What change is being referred to here? Seasonal change? Decadal change?

The change referred to in this sentence is the inter-annual variation in Eurasian contributions estimated by the model in this study. The sentence has been modified to clarify this in the manuscript.

7) p. 2223, lines 1 – 2: Again, each of these references is a model study. Wording here should be changed to “have been calculated to have significant effects...”

Change has been made.

8) p. 2223, line 16: Name the two different sites, give latitude and longitude for both, and explain why it is hypothesized that they are impacted by different sources regions.

In the revised manuscript, the sentence has been changed to "Sharma et al. (2006), for example, showed that Alert (82.5°N, 62.5°W), Nunavut was about two times more frequently affected by the atmospheric transport of air mass from North America than Point Barrow (71°N, 156.6°W), Alaska from 1989 to 2003."

9) p. 2223, lines 20 – 22: Change sentence to “...a broad peak in BC concentration is observed from January to April...”

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Change has been made.

10) p. 2223, lines 22 – 24: State whether the observed decreasing trend in BC concentrations occurs for year-round data, Arctic Haze season data, and/or summer data.

The trend discussed in the manuscript refers to the trend obtained by Sharma et al. (2006) for January-April during the period 1989-2003. The sentence has been changed to "For the haze season (January to April), a marked monotonic decreasing trend of BC concentration at Alert during the 1990s, followed by signs of an increase in the early 2000s, was revealed using a geometric time variation model (Sharma et al., 2004; Sharma et al., 2006)."

11) p. 2223, line 26: Should be "associated with".

Change has been made.

12) p. 2224, line 10: which two sectors?

The two sectors are Eurasia and North America, which are now explicitly stated in the manuscript.

13) p. 2224, line 15: Explain how the "average length" of the 10-day back trajectories relates to the strength of transport. Is "average length" the distance covered in 10 days?

The average length in the original manuscript is the distance covered in 10 days, which indicates the average horizontal wind speed. Thus, the average length is only a rough indication of the speed of atmospheric transport to Alert. Therefore, the reason why January and April trajectories were used in this study is explained by the following two sentences. "The use of January and April data instead of DJF (for winter) and MAM (for spring) is to minimize the seasonal transformation of atmospheric transport patterns in the whole seasons of interest, and to emphasize the inter-annual variation in atmospheric transport. January and April were considered representative of winter and spring, respectively in characterizing the atmospheric circulation affecting the Arctic

(Serreze and Barry, 2005)."

14) p. 2224, line 18: Again, what is the definition of the BC emission intensity?

To be consistent, "BC emission intensity" has been changed to "BC surface flux".

15) p. 2224, line 23: Change to "Continuous hourly measurements of aerosol light absorption at Alert ..."

Change has been made.

16) p. 2226, lines 4 – 5: Change to "...black carbon in the northern mid-latitudes is predominantly..."

Change has been made.

17) p. 2226, line 5: should be "BC trend"

The sentence has been changed to "Analyzing the inter-annual variation in the Arctic BC required building annual BC emission inventories by country from 1990 through 2005."

18) p. 2226, line 7: what is meant by "transaction amounts"?

Transaction amounts include fuel production, imports and exports, bunkers, stock changes, and apparent consumption. The apparent consumption of fuel is derived from the formula "production + imports – exports – bunkers ± stock changes" (United Nations, 2007).

19) p. 2226, 1st paragraph: I found the discussion of emission inventories to be confusing. Did the authors use data from United Nations (2007) and the method of Cooke et al. (1999) to construct emission inventories for the different regions and years mentioned? What is the 50% reduction in BC emissions from the former USSR and the 10%/yr increase in South Asia based on? Why are the Bond et al. (2007) global emissions mentioned when regional emissions are used in this analysis? On lines 15- 16,

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it is stated that “only emissions every 10 years until 2000 are made available to the public on their web site...” Whose web site? Last line of the paragraph: were your emissions determined using the method of Cooke et al? In other words, it needs to be more clearly stated how regional surface fluxes were calculated. Also, this paragraph describes the development of regional emission inventories but not “BC emission intensity.” What is the BC emission intensity and how was it derived? It would be helpful if all of these terms (emission, emission intensity, surface flux) were defined.

Section 2.3 has been re-written to better present the preparation of BC regional emissions used in this study. The estimated global total BC emissions from fossil fuel combustion in this study were compared with Bond’s estimations for 1990 and 2000 to show that the difference in BC inventories from both studies is within the uncertainty of the current estimation of BC emissions. Then, the general characteristics of the obtained surface fluxes for European Union, North America, and former USSR were described in the last paragraph of the section.

Yes. Data from United Nations (2007) and the method of Cooke et al. (1999) were used in the current study to construct emission inventories for the regions and years mentioned.

The 50% reduction in BC emissions from the former USSR is based on its 1990’s level, which is now stated in the manuscript.

South Asian emissions are not used in this study, and the discussion on it has been removed from the text.

The global emissions by Bond et al. (2007) were used for the comparison purpose, which is now clarified in the text.

"Their web site" refers to Bond’s web site, which provides the global BC inventories.

The calculation of BC surface flux for a source region has been added to the manuscript. "BC emission intensity" has been replaced by "BC surface flux" in the

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manuscript, which were used to refer to the same thing in previous manuscript.

20) p. 2226, line 19 – 22: Sentence is poorly constructed. Change to “Based on the work of Stohl (2006), North America, the European Union, and the former USSR are the major BC source regions affecting Alert.”

Change has been made.

21) p. 2227, lines 21 - 22: As mentioned above, Section 2.1 – 2.3 needs a clearer description of the methodologies used to derive emission intensity.

Emission intensity has been replaced by BC surface flux, which is defined in Section 2.3.

22) p. 2228, line 2: Change to “...that BC emissions other than...”

Change has been made.

23) p. 2228, line 6: Presumably this should be “...for the case of a high LATITUDE Arctic surface site...”

The sentence has been removed from the manuscript.

24) Figure 2 caption: State in the caption that these are average 10 day backward trajectories.

Change has been made to Figure 2 caption.

25) p. 2228, line 26: Is Cluster 2 (20%) significantly different from Cluster 1 (17%) or Cluster 6 (18%)?

The fraction of trajectories in Cluster 2 (20%) is not significantly differently from that of Cluster 1 (17%) or Cluster 6 (18%). Therefore, the sentence has been replaced by the statement: "In terms of transport frequency, Clusters 1, 2, and 6 are among the most frequent transport pathways, which in total account for 55% of atmospheric transport in winter."

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26) p. 2229, lines 9 – 10: Based on the figure, it is not clear that trajectories in cluster 1 originated in Europe. The trajectory shows flow from the west and then northwest over Greenland.

This is because that the deviation of trajectory members from the cluster mean sometimes is not negligible due to the complexity of atmospheric transport. Therefore, cluster-member plots are shown in Figures S1-S2 in the Supporting information. The cluster-member plot of Cluster 1 for January, 1990-2005 shows that trajectories grouped into this cluster originated in the North Atlantic Ocean and the northern Europe. These trajectories, however, share a common pathway to the receptor, Alert by passing through Northern Europe.

27) p. 2229, line 10: Change to "...and Europe."

Change has been made.

28) p. 2229, line 15: Change to "...amount of time traveling..."

Change has been made.

29) p. 2229, lines 24 – 25: Change to "Such long range transport is found during the winter 18% of the time."

Change has been made.

30) p. 2229, lines 25 – 26: Clarify what is meant by the sentence that starts with "Transport from Eastern Siberia..."

The sentence has been replaced by "Cluster 4 (10%) represents atmospheric transport mainly from Eastern Siberia, but it contains few trajectories originated from Bering Sea and Alaska."

31) Tables 1 and 2: It would be easier to see the temporal trends and variability in the transport frequency if these data were presented as figures instead of tables. I would do this as individual panels in a figure where each panel has the winter and spring data

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for one cluster.

Values of transport frequency presented in Tables 1 and 2 were obtained based on the fraction of trajectories grouped into each distinct cluster (or pathway to Alert) for January and April, 1990-2005. Together with surface fluxes, these values were used to estimate b factors according to Equation 2. However, the figures with transport frequency of winter and spring for each individual cluster do not contain comparable information. Because the cluster analyses on the winter and spring trajectories were conducted separately. Therefore, one cluster from the wintertime transport pattern does not necessarily have its equivalent for springtime.

32) p. 2230, line 18: Change to “Given monthly average BC concentrations...”

Change has been made.

33) p. 2230, line 23: Change to “...monthly average BC concentrations...”

Change has been made.

34) p. 2230, line 26: Change to “...indicates the fraction of the inter-annual variations...”

Change has been made.

35) Figure 3 caption: squares should be squares.

The caption has been corrected.

36) p. 2231, lines 23 – 28: It would be more useful to put the degree of under- and overestimation in terms of percent difference between measured and modeled concentrations.

The difference between measured and reconstructed concentrations is also presented in percentage.

37) p. 2232, line 7: Omit “According to the model...”

Change has been made.

38) p. 2232 and Figure 4: Why are the contributions of BC transport from the North American and Eurasian sectors averaged over January and April? Why not consider the two months separately as BC deposited in winter vs. spring could have very different radiative impacts?

The contributions of BC transport from North America and Eurasia were averaged over January and April to compare with the estimations by Gong et al. (2010) based on January-April average and by Shindell et al. (2008) based on annual average.

39) p. 2232, lines 14 – 17: Sentence needs to be re-written for clarity.

The paragraph has been re-written.

40) p. 2232, line 23: Change to "...the first 8 – 10 years after 1990..."

Change has been made.

41) p. 2233, lines 3 – 6: It is stated that, for the North American source region, BC concentrations at Alert did not depend only on "regional BC emission, but also on other factors, especially atmospheric transport patterns." What were the other factors? Provide a discussion to support the phrase "especially atmospheric transport patterns".

For clarity purpose, the sentence has been modified as follows. "The North American contributions for the same period did not simply depend on regional BC emission, but also on other factors, especially atmospheric transport patterns, as indicated by the good agreement between measured and reconstructed BC surface concentration."

Here, other factors include the impacts of transport, deposition, mixing height at the source region, and so on. Atmospheric transport, however, is especially important among these factors. Because the model reconstructed BC concentrations agree fairly well with the observation, and about 80% of the inter-annual variation in BC concentration can be explained by including both transport and emission into the model.

42) p. 2233, lines 19 – 21: Again, the limitation of this method to account for the impact

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of the removal of BC from the atmosphere should be included in this discussion.

The limitation has been included in the discussion.

43) p. 2233: lines 22 – 23: Sentence needs to be re-written for clarity.

The sentence is now replaced by "The relative importance of North American and Eurasian emissions to BC concentration at Alert is also investigated."

References:

Gong, S. L., T. L. Zhao, S. Sharma, D. Toom-Sauntry, D. Lavoue, X. B. Zhang, W. R. Leaitch, and L. Barrie: Identification of trends and inter-annual variability of sulphate and black carbon in the Canadian High Arctic: 1981 to 2007. *Journal of Geophysical Research-Atmospheres*, doi:10.1029/2009JD012943, 2010. Serreze, M. C. and R. G. Barry: *The Arctic Climate System*. Cambridge Atmospheric and Space Science Series, Cambridge University Press, 412 pp, 2005. Sharma, S., D. Lavoue, H. Cachier, L. A. Barrie, and S. L. Gong: Long-term trends of the black carbon concentrations in the Canadian Arctic. *Journal of Geophysical Research-Atmospheres*, 109, 10, 2004. Sharma, S., E. Andrews, L. A. Barrie, J. A. Ogren, and D. Lavoue: Variations and sources of the equivalent black carbon in the high Arctic revealed by long-term observations at Alert and Barrow: 1989-2003. *Journal of Geophysical Research-Atmospheres*, 111, 15, 2006. Sharma, S., M. Ishizawa, D. Chan, D. Lavoué, R. Leaitch, D. Worthy, E. Andrews, K. Eleftheriadis, T. Mefford, and S. Maksyutov: Synoptic Transport of Anthropogenic BC to the Arctic. NOAA annual conference, Boulder, CO, 2009. Shindell, D. T., M. Chin, F. Dentener, R. M. Doherty, G. Faluvegi, A. M. Fiore, P. Hess, D. M. Koch, I. A. MacKenzie, M. G. Sanderson, M. G. Schultz, M. Schulz, D. S. Stevenson, H. Teich, C. Textor, O. Wild, D. J. Bergmann, I. Bey, H. Bian, C. Cuvelier, B. N. Duncan, G. Folberth, L. W. Horowitz, J. Jonson, J. W. Kaminski, E. Marmer, R. Park, K. J. Pringle, S. Schroeder, S. Szopa, T. Takemura, G. Zeng, T. J. Keating, and A. Zuber: A multi-model assessment of pollution transport to the Arctic. *Atmospheric Chemistry and Physics*, 8, 5353-5372, 2008. United Nations: *The United*

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Nations energy statistics database (2005), 5 pp, 2007.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/10/C1061/2010/acpd-10-C1061-2010-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 2221, 2010.

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10, C1061–C1072, 2010

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