Authors' response to the reviewers' comments

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Title: Spectral optical properties of long-range transport Asian dust and pollution aerosols over Northeast Asia in 2007 and 2008

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Reviewer #1 (Comments):

Authors appreciate the reviewer's constructive comments and suggestions. The manuscript has been revised to accommodate the reviewer's comments.

Answer to the reviewer's specific comments and questions;

Q: *My major concern is that the manuscript is written not like a research article but like a report. The authors just report the data with little data interpretation or discussion.*

A: The originality of the manuscript includes not only improvement of analytic technique but also scientific data analysis. Multiple techniques were combined to develop a spectral signature of Asian dust and long-range transport aerosol in the region, which will contribute to reduction of uncertainties associated with direct climate forcing of these aerosols. There are many artifacts present in filter-based absorption measurement (e.g., aethalometer and PSAP), causing considerable uncertainties in aerosol absorption coefficient. Reliable correction schemes are suggested by many researchers to derive "true" values (Weingartner et al., 2003; Arnott et al., 2005; Schmid et al., 2006). However, these schemes need to assume the mixing state of EC. Thus, this study suggests a novel approach to estimate the mixing state of EC using BC/EC ratio. In addition, we combine multiple techniques to develop a spectral signature of Asian dust and pollution aerosols.

We have added more data interpretation and discussion in Chapter 4 as suggested by reviewer. Volume size distribution of atmospheric aerosols has been separated from Chapter 4.4 and reorganized in Chapter 4.5 with more data interpretations.

Following paragraphs have been inserted in page 21, line 11 as;

"4.5 Volume size distribution of long-range transport aerosols

Log volume distributions of ambient and volatile aerosols during the AD and LTP periods are seen in Figs. 17−18. Volatile aerosols in this study represent the aerosols, which are volatilized under 110 °C. Tri-modal distributions with the predominance at coarse mode (4−5 μm) were observed during the Asian dust periods. However, four mode distributions peaked at 0.4, 0.6, 2, and 4−5μm corresponding to the condensation mode, droplet mode-1, droplet mode-2, and coarse mode, respectively, were observed during the LTP periods. Interestingly, it was clearly observed the droplet mode-2 during not only the LTP but also the AD periods.

When air mass was originated from China continent, tri-modal mass distribution with additional droplet mode peaked at 1.0-1.8 μm was observed at downwind site in northeast China (Liu et al., 2008). Guo et al (2010) also observed additional droplet modes peaked at 0.8 and 1.4 μm based on size-segregate aerosol measurement and PMF peak separation. From the chemical analyses, they found that the additional droplet modes were mainly consisted of sulfate, nitrate, and ammonium formed by in-cloud or aerosol droplet process. Guo et al (2010) argued that the droplet mode-2 was mainly from regional transport. As a result, it can be estimated that the droplet mode-2 during the AD and LTP periods is probably due to secondary aerosols formed by in-cloud and/or aerosol droplet process during the long-transport.

Condensation mode is the result of growth of ultrafine particles by coagulation and vapor condensation while coarse mode is mainly from nitrate, sea salt, and soil (Liu et al., 2008; Guo et al., 2010). However, droplet modes are from particle growth by in-cloud or aerosol droplet process. Volume distributions of volatile aerosols during the LTP periods are similar to those of ambient aerosols. However, volatile fraction to ambient aerosols showed different volume size distributions with the predominance at droplet mode-1, followed by droplet mode-2. It is well known that most of ammonium nitrate is volatilized under 110 °C but only certain fractions of ammonium sulfate and organic aerosols are volatilized. Thus, it can be estimated that higher fractions of droplet mode-1 and droplet mode-2 were consisted of ammonium nitrate and/or high volatile organic aerosols."

Q: *Another concern is that the manuscript is rather lengthy and hard to read.*

A: We have shortened the manuscript by simplifying Chapters 2 and 3. The manuscript has been revised by reorganizing Chapter 4.

Q: *In my opinion, Chapters 2 and 3 can be shortened significantly and Sections 4.1 and 4.2 can be described in more concise way.*

A: Chapters 2 and 3 have been revised by shortening contents significantly. However, on the request of another reviewer, explanations about measurement techniques have been added in

Chapter 2. Chapters 4.1 and 4.2 have been revised in more concise way.

Q: *Also, there are a couple of loose points: 1. P. 13, b_{scat} by the nephelometer is a point measurement, but the Ångström exponent of aerosol light extinction data were from column integrated data from the Skyradiometer to calculate spectral dependence of b_{ext}. I think, it is inevitable but still the authors should provide justification on that or possible error bound.*

A: This study assumes vertically well mixed aerosols inside boundary layer in calculating the spectral dependence of light extinction coefficient (b_{ext}) from Ångström exponent obtained by the Skyradiometer. Also it can be assumed that aerosol optical depth is dominated by these particles within the boundary layer at urban sites with high aerosol loading.

Following sentences have been inserted in page 13, line 16 as;

"This study assumes vertically well mixed aerosols inside boundary layer in calculating the spectral dependence of b_{ext} from Ångström exponent obtained by the Skyradiometer. Also it can be assumed that aerosol optical depth is dominated by these particles within the boundary layer at urban sites with high aerosol loading."

Q: 2. *P. 14, the authors suggest that the low Angstrom exponent of b_{scat} in summer is due to the increase of particle size through water uptake. Is there supporting evidence for that suggestion?*

A: It has been well known that hygroscopic urban particles grow in summer as RH increases. During the entire measurement period, b_{scat} of dry particle was also measured. From the b_{scat} of dry and ambient particles, light scattering enhancement factor, *f*(RH) (= $b_{scat}(RH)$ / $b_{scat}(dry)$) was calculated as a function of RH and newly presented in Fig. 7. Average RH in summer and winter were 74.7±13.6% and 52.0±18.2%, respectively. Average *f*(RH=75%) and *f*(RH=52%) were obtained to be 1.7±0.4 and 1.2±0.2, respectively. Higher average *f*(RH) in summer indicate increase of particle size by water uptake. Thus, lowest α_s in summer is believed to be due to the increase of particle size through water uptake.

Above paragraph has been inserted in page 14, line 18 as;

"It has been well known that hygroscopic urban particles grow in summer as RH increases. During the entire measurement period, b_{scat} of dry particle was also measured. From the b_{scat} of dry and ambient particles, light scattering enhancement factor, *f*(RH) (= $b_{scat}(RH)$ / $b_{scat}(dry)$) was calculated as a function of RH and shown in Fig. 7. Average RH in summer and winter were 74.7±13.6% and 52.0±18.2%, respectively. Average *f*(RH=75%) and *f*(RH=52%) were obtained to be 1.7±0.4 and 1.2±0.2, respectively. Higher average *f*(RH) in

summer indicate increase of particle size by water uptake. Thus, lowest α_s in summer is believed to be due to the increase of particle size through water uptake."

Below figure and figure caption have been inserted in the figure section as;

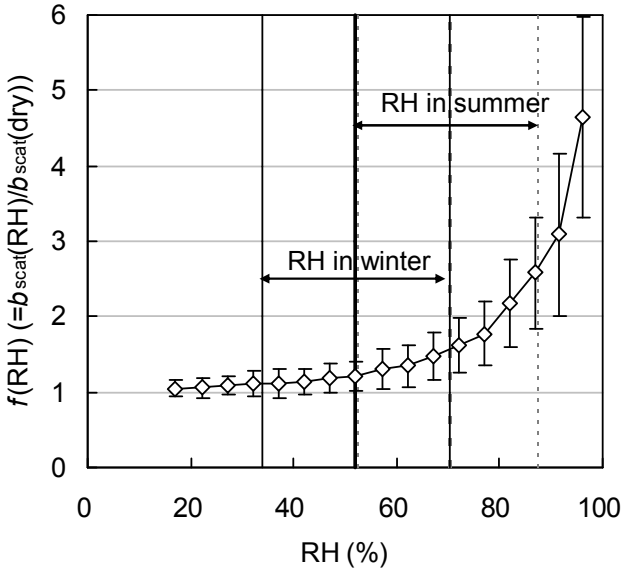

Figure 7. Humidity dependence light scattering enhancement factor, *f*(RH) ($=b_{scat}(RH)/b_{scat}(dry)$). Error bars represent 1σ. Bold vertical solid and dotted lines represent average RH in winter and summer, respectively. Arrows indicate 1σ.

Q: *3. The starting point of 200 m high seems too low for the backward trajectory analysis. Do the authors have justification on it or some kind of error analysis?*

A: Air mass backward trajectories starting at 200 m high have been removed from the trajectory plots. Backward trajectory plots in Figs. 12 and 14 have been revised accordingly.