Response to Anonymous Referee #2’s comments

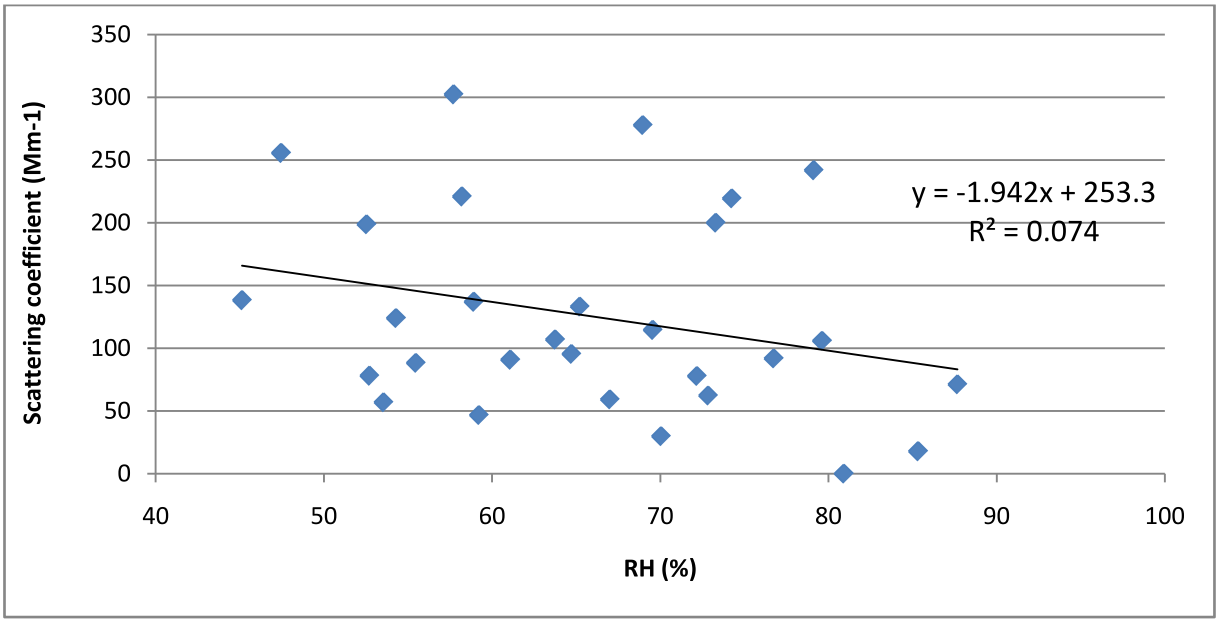
1. Page 7: Aerosol optical properties related to meteorological variables need to be addressed.

As shown in Table 1, RH at the 880 nm group is relatively high (58_79 %).In the humid condition, hygroscopic growth of water-soluble ions such as sulfate is concerned because the optical properties of inorganic aerosols could be changed.

The 880 nm group samples collected under high relative humidity condition showed high scattering coefficient at 550 nm and sulfate mass concentrations. In general, relative humidity (RH) influences the water content of aerosol particles through hygroscopic growth, which changes their optical properties. Not to mention, hygroscopic growth of sulfate aerosols must have increased scattering coefficients under humid condition. It is stated in the text “33218 line 8-10” in “3.2.2. Scattering in sulfate dominant regime”, as follows.

The high sulfate concentrations were coupled with stagnant conditions over the Yellow Sea, which supplies sufficient humidity and time for sulfate conversion in Chinese outflows (Lim et al., 2012) (Table 1).

It is also noteworthy that in this study, scattering coefficient at 550 nm tended to decrease with increase in relative humidity (shown below). The relative humidity was high up to 80~90 % in May, June, and September, when the scattering coefficient was the lowest. When relative humidity was comparable to that of the 880 nm group but inorganic salts contents were high, scattering coefficient was only half of that of 880 nm group. This result indicates that in conjunction with water vapor, the major composition played a significant role in determining scattering coefficient of aerosols, particularly in northeast Asia where SO_2 emission is the greatest and subject to change with season.



2. Page 9: I wonder if the inorganic ions of 370 nm group are derived solely from salts deposited in dry lakes in northeastern China. If any measurement data from a local source like Jeju island are used, they need to be specified in the study.

Gosan station is located on the cliff by the sea and thus falls under the local influence of sea salts. This fact was considered and stated in the text " 33216 line 3-5".

This type of soil particles was, however, not recognized until soil alkalization became an environmental issue in northeast China, which is pretty recent. We often observed that aerosol mass concentrations happened to be elevated with high inorganic salts and the low levels of pollutants at Gosan, particularly in spring and fall. Through this study, we were able to identify chemical and meteorological characteristics of this type of aerosols. When fast northerly winds developed under the influence of a strong high pressure system above the main land of China, the chemical composition of aerosols showed that the ratios of Cl/Na and Ca/Mg were closed to and higher than that of sea salts, respectively. We have worked on more data obtained at Gosan for four years and will hopefully be able to distinguish the salts from salt deposit from salts from nearby sea. There are measurements sets of aerosol composition from previous studies conducted at Gosan, which were mostly obtained during various kinds of high concentration events and thus not quite appropriate to be compared with our results to single out local influences of sea salts.

3. Page 12: The SMPS results in Figure 4 seem to be somewhat weak to claim that BC was internally mixed with sulfate. Can the authors distinguish the difference between externally mixed vs. internally mixed BC from the SMPS results? How?

Integrating all available measurements and analysis done in this study, we came to claim that the aerosols of 880 nm group were rather internally mixed. They were collected when wind speed was very low ~ 5 m/s and air mass moved very slowly from Shanghai region for a few days. In previous studies conducted nearby megacities in China such as Shanghai, aerosols were reported to be internally mixed during serious pollution events under prevailing stagnant condition (Fu et al., 2012; Tao et al., 2011; Yang et al., 2012).

Thus, the third paragraph (33218 line 25 ~ 33219 line 4) in section 3.2.2 was rephrased as follows.

However, the daily variation of the particle size distribution (top panels in Fig. 4) gives a hint of the mixing state of aerosols. For the 880 nm group, the mode and number concentration remained unchanged through the day (top panels in Fig. 4c and d). In conjunction with other measurements, it suggested that aerosols were internally mixed. In general, the atmospheric BC of remote areas is often internally mixed with other materials such as sulfate (Clarke et al., 1997; Pósfai et al., 1999) through intensive processing due to longer residence times (Hasegawa and Ohta, 2002). From observations nearby megacities in China including Shanghai, the results of single particle analysis indicate that aerosols were internally mixed during serious pollution events such as haze under prevailing stagnant condition (Fu et al., 2012; Tao et al., 2011; Yang et al., 2012).

4. Page 14: It seems the AAE estimate of the 880 group (_1.0) is too low. Such low AAE needs to be explained further.

In a previous study performed at GCO, the AAEs of sulfate rich aerosols were also estimated to be as low as 0.8 (0.8 –1.5) (Lee et al., 2012), which is in accordance with our result for the 880 nm group. However, MAC values were somewhat different between the two: 4.2 ± 1.1 m^2g^{-1} for the Lee et al. (2012) and 7.7 ± 3.5 m^2g^{-1} for the 880 nm group in this study. Liu et al. (2008) showed that the absorption cross section of BC particles changed by their morphology with reduced absorption or enhanced cross sections of packed particles compared to those of chain-like particles. This morphological change in BC particles is coupled with aggregating or internal mixing with hydrophilic particles and coating during aging processes. In particular, the coating of scattering material is known to enhance MAC of carbonaceous core. At GCO, sulfate rich aerosols are mostly found in aged air masses of Chinese outflows and therefore, the degree of aging associated with SO_2 availability was likely to modify optical properties of carbonaceous aerosols. Considering these complex facts, our AAE of 1.0 for 880 nm group probably represents the absorption characteristics of aged and sulfate dominant aerosols in the study region. Also, our results suggest that the absorption properties of aerosols such as absorption coefficient, MAC, and AAE are greatly dependent on chemical compositions and atmospheric process during aging, which is poorly understood at present.

5. Page 16: More discussion is needed with regard to the claim that the sulfate coating on BC particles contributed to the absorption of the longer visible light. In Figure 5(b),compared to the 370 nm group, MAC of the 880 nm group under sulfate dominant conditions is large as the wavelength is increased, contributing to the absorption of the longer visible light. However, it seems not reasonable that the 880 nm group SSA that was in the decreasing trend slightly increased at the end. For the 370 nm group, the SSA was the minimum when MAC was maximized at the UV region.

In the revised manuscript, more discussion was added to the text as you recommended.

In the present study, the SSAs of the 880 nm group were higher those of the 370 nm group in overall wavelengths. This is quite reasonable, considering much higher sulfate concentrations of the 880 nm group. As you pointed out, hygroscopic growth of aerosols also was likely to play a role in increasing SSA of the 880 nm group under high humidity condition.

In addition, SSAs for the two groups decreased with increase in wavelength with a greater spectral dependence of the 880 nm group. Bergstrom et al. (2002) discussed that the SSA of a mixture of BC and non-absorbing material decreased with wavelength in the solar spectrum, in contrast to most mineral dusts, of which SSA increased with wavelength increase. Thus, SSA can be used to distinguish aerosol types. For the 370 nm group, the steady SSA values around 350 – 500 nm could be associated with enhanced absorption by light-absorbing OC, in relation with MACs in Fig. 5. In the same context, the greater spectral dependence of the 880 nm group was likely due to absorption enhanced at longer wavelength.

6. Page 15: The climate effects of aerosol and/or radiative forcing are not fully addressed in Section 3.3. There is some related discussion in Section 3.3.2 but it is not enough.

The radiative forcing was not fully discussed in the preset study because the original motivation of this study was to understand the influence of aerosol composition on its optical properties. It is also beyond the scope of this study and will have to be investigated through further studies.

7. Page 28: I wonder if the authors can distinguish between external vs. internal mixture BCs from the Figures 3(a) and (b). More detailed description on the figures would be helpful, for example, distinction of fresh/aged particles by size, shape, or color. Also, elaborate further why Figures 4(c) and (d) are necessarily analyzed as internal mixture of BC, not external mixture of BC.

More explanation was given to and the relevant discussion was modified in the revised manuscript: section 3.2.1. 33216 line 17 and section 3.2.2. 33218 line 21 ~ 33219 line 1.

[section 3.2.1. 33216 line 17]

In the 370nm group, three types of particles recognized by their distinct shapes and sizes seemed to be mixed externally (Fig. 3a). One type consisted of agglomerates that were more irregular in size and shape and bigger than soot agglomerates shown in Figure 3b, and were thought to be organic particles coating soot (Adachi and Buseck, 2008; Adachi 20 et al., 2010).

[section 3.2.2. 33218 line 21 ~ 33219 line 1] as a separate paragraph

For the 880nm group, BC was likely to be internally mixed with sulfate during the aging processes, which modified the absorption properties of the aerosols and resulted in spectral dependence of EBC (Fig. 1a). The cluster of soot aggregates in Figure 3b was bigger and more compacted, compared to that in Figure 3a, which suggests the soot aggregates of the 880 nm group were more aged (Fu et al., 2012). In other studies, the more aged Chinese air masses were sampled, the more packed soot aggregates used to be observed (Kang et al., 2012). Although it was not determined if the BC core was surrounded by a well mixed shell including sulfate or simply incorporated into other components, the daily variation of the particle size distribution (to panels in Fig. 4) gives a hint of the mixing state of aerosols, in conjunction with other measurements (e.g., Figure 3). For the 880 nm group, the mode and number concentration remained unchanged through the day (top panels in Fig. 4c and d), implying that aerosols were rather internally mixed.

Fu, H., Zhang, M., Li, W., Chen, J., Wang, L., Quan, X., & Wang, W. (2012). Morphology, composition and mixing state of individual carbonaceous aerosol in urban Shanghai, *Atmospheric Chemistry and Physics*, *12* (2), 693–707. doi:10.5194/acp-12-693-2012.

Kang, E., Park, I., Lee, Y.-J., Lee, M., Characterization of atmospheric particles in Seoul, Korea using SEM-EDX, *Journal of Nanoscience and Journal of Nanotechnology*, 12, 6016-6021, 2012.