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Interactive comment on "Absorption and scattering properties of organic carbon vs. sulfate dominant aerosols at Gosan climate observatory in Northeast Asia" *by* S. Lim et al.

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Response to Referee (A. Jefferson) comments

1. Introduction: Page 33209 Line 5: remove the word "and" and make 2 sentences. "..directly and indirectly. The relative.." Line 13: remove the comma after the word "that"Line 27: replace word "particles" with "species" or "compounds".

The sentence in the line 5 was separated into two: "Atmospheric aerosols play an important role in climate change by altering the global radiation balance both directly and indirectly. The relative importance of these processes depends on the chemical

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composition and size distribution of aerosols (Forsteret al., 2009).”

Comma was removed in the line 13.

“particles” was replaced with “compounds”.

2. Page 33210 Line 1: remove “even though they theoretically should be the same”. Absorption measurements measure absorption and not BC. There never has been a theoretical basis for aerosol absorption being only attributed to BC. This has only been done for simplicity.

The phrase starting with “even though” was removed.

3. Page 33211 Line 16: Reword this sentence. I think you mean to say “ In the He/OÂËĞn2 Mode. . .”

This part was changed to “in an oxidizing atmosphere of 2% O2/He mode”.

4. Section 3.2.1 Part of your analysis of an enhanced absorption at the shorter wavelengths implies that you are using the correct mass absorption coefficient (MAC) for BC at these wavelengths. How accurate is the MAC that Magee Scientific uses to calculate EBC for EC at wavelengths other than 880? Bond et al. (2013) uses a value of 7.5 m2g-1 at 550 nm for freshly emitted BC. What value does Magee use at this wavelength? Since you have both the BC mass and the absorption coefficient it would be better to calculate the MAC from this study and compare it to MAC values from this region (ACE-Asia) or other regions. An MAC value at each wavelength is much more informative than the EBC from the aethelometer.

The aethalometer directly measures “optical attenuation (ATN)” of a beam of light transmitted through the sample, which is defined as ATN=100*ln(I0/I). The absorption of light by a broad band absorber is inversely proportional to the wavelength of the light used. Thus, for a given mass of black carbon [BC], the ATN at a fixed wavelength λ may be written as ATN(λ) = σ (1/λ) * [BC]. Here, [BC] is the mass of black carbon and σ (1/λ) is the optical absorption cross-section (‘sigma’) that is wavelength dependent, which is referred to as the ‘Specific Attenuation’ (Hansen, 2005). To derive EBC from absorption coefficient measured by the aethalometer, Hansen (2005) proposes two calibrations, one of which is “Classic Magee BC” calibration in terms of the ‘SIGMA’ specific attenuation parameters. Mie theory for small uniform spheres predicts that to first order, ‘SIGMA’ is inversely proportional to the optical wavelength λ. For “Magee BC” calibration, σ = 14625 / λ (nm). Thus, wavelength-dependent sigma values were applied to get EBC mass concentration from measured attenuation.

Because the SIGMA values were derived from empirical equation and inherent properties of the instrument, it is beyond our ability to tell or compare the accuracy of those values. The limitation of the aethalmeter for aerosol absorption and BC measurement are well stated in the manual and known to scientific community as well.

Because aethalometer measures attenuation, it should be corrected to get absorption coefficient. For this correction, we adopted the method of Arnot et al (2005). For MAC estimation, absorption coefficients at 7 wavelengths were divided by EC mass concentration. This is how we obtained MAC and there was no correction for MAC other than when getting absorption coefficient.

Our MAC values were 7.5ïĆś3.0 and 7.7ïĆś3.5m2g-1at 520 nm for 370 nm and 880 nm group, respectively. For 880 nm group, the mean EC mass concentration was higher by 35 % and MAC was also higher up to 13 % at near-IR than those of the 370 nm group. Bond et al. (2013) adopted a value of 7.5 m2g-1 at 550 nm for freshly emitted BC and Bond et al. (2006) suggested ambient MAC values in polluted regions as around 9-12 m2g-1. Recently, Lee et al. (2012) reported ambient MAC values of 7.4 ïĆś 2.0 m2g-1 and 4.2 ïĆś 1.1 m2g-1 at 520 nm during different types of pollution events, which we think reflects complex nature of aerosol compositions in the northeast Asia. Liu et al. (2008) claimed that the absorption cross section of BC particles changed by their morphology as they were being packed from chain-like shape, depending on the compactness of a cluster and the size and number of individual particles. BC particles undergo morphological change, accompanied by mixing, aggregating, or coating with hydrophilic during aging processes. In particular, the coating of scattering material is known to enhance MAC of carbonaceous core. Considering these complex facts, our MAC of 7.7 ïĆś 3.5 m2g-1 for 880 nm group likely represents the absorption characteristics of aged and sulfate dominant aerosols in the study region. On the other hand, the MAC of the 370 nm group was similar to those of the polluted aerosols in Lee et al. (2012). The OC-dominant aerosols of 370 nm group were less aged than those collected in other period of time, for which absorption was highly enhanced in UV region. These results suggest that the MAC of aerosol is greatly dependent on chemical compositions and atmospheric process during aging, which is the motivation of this study.

Liu, L., Mishchenko, M.I., Arnott, W. P., 2008, A study of radiative properties of fractal soot aggregates using the superposition T-matrix method, Journal of Quantitative Spectroscopy and Radiative Transfer, 109, 2656-2663, 2008.

As you recommended, the comparison of the results of previous studies was added to the revised manuscript.

5. Page 33216, line 12: There are several species that have similar or even much higher scattering efficiencies than (NH4)2SO4. Ammonium nitrate and oxidized organics have a similar scattering efficiency as ammonium sulfate. The MSE of organics is often higher than that of sulfate (Sciare et al., ACP, 2005). The Tang study is very limited.

As you pointed out, the sentence in line 10 page 10 was rephrased for clarification as follows.

In this group, OC and inorganic salts were considered as main contributors to light-scattering. Also, the chemical effect on light scattering was possibly outweighed by the size effect of the aerosols (Tang, 1996). Mishchenko et al. (2004) also reported when one of the aerosol components is much larger than the others, it dominates the total optical characteristics of the mixture, especially for semi-externally or externally mixed particles.

6. Section 3.2.2 Only a fraction of the scattering can be attributed to sulfate. OC and other ions may contribute as much if not more to the aerosol scattering than sulfate. Even though the relative scattering per sulfate mass was lower in the 880 group than in the 370 group, there could be other non-sulfate, highly scattering species in the880 group that contribute to the scattering. Also note that the samples taken for the370 group were early in the year when the ambient RH was low. The samples in the 880 group were taken in the spring when ambient dew point was quite high (seewww.wunderground.com for daily meteorological values from Cheju in 2008). What this means is that the aerosol in the 880 group may have taken up a significant fraction of water inside the instrument, which enhanced the scattering significantly compared to the 370 group. You need to check the differences in the internal nephelometer RH for these two sample times. It's best to either remove the discussion on BC enhanced absorption or explain the possibility of water uptake by the aerosol. Enhanced BC absorption with coatings may not be apparent in filter measurements of the aerosol may spread upon impaction on the filter.

As you pointed out, there could be other species contributing to scattering other than sulfate including OC and soluble ions. Although scattering coefficient was reasonably correlated with concentrations of aerosol mass and major chemical species at Gosan, it was correlated the best with PM1 sulfate for 880 nm group. In contrast, PM10 ions were not correlated with scattering coefficient at all for 880 nm group as shown in Figure 2a. For 880 nm group, sulfate was a predominant species and comprised about 40 % of total mass (Table 1). For comparison, sulfate as the second dominant species next to OC and took up 24 % of the mass for 370 nm group. If considering non-sulfate aerosols contributing to scattering, there would be more in 370 nm group than in 880 nm group. In addition, the less scattering coefficient per unit sulfate mass simply means that sulfate was less efficient in scattering for 880 nm aerosols (Figure 2b).

Sure enough, the aerosol scattering properties are subject to change, caused by hygroscopic growth under high relative humidity condition. It is one of the main factors for 880 nm aerosols to be formed and distinguished. However, it is nothing to do with artifact in instrument inlets. For the measurements of absorption, scattering, and particle size, air was drawn from the main aerosol inlet (∼ 20 m) and then split to each instrument. Please see other studies regarding optical properties of aerosols measured using the same instruments at Gosan (e.g., Kim et al., 2014, Lee et al., 2012,).

Kim, Y, Kim, S.-W., Yoon, S.-C., Kim, M.-H.Park, K.-H., Aerosol properties and associated regional meteorology during winter pollution event at Gosan climate observatory, Korea, Atmos. Environ., 85, 9-17, 2014. Lee, S., et al., Spectral dependency of light scattering/absorption and hygroscopicity of pollution and dust aerosols in Northeast Asia, Atmos. Environ., 50, 246–254,2012.

7. Section 3.3.1 Page 33220 An AAE of 1.0 is consistent with absorption of BC or soot. I don't see any correlation of AAE to an influence of sulfate on absorption as BC without sulfate has a similar AAE. You need to remove this sentence in lines 21-22.

As you recommended, this sentence was removed and the relevant discussion was modified in the revised manuscript as follows.

In a previous study performed at GCO, the AAEs of sulfate rich aerosols were estimated to be 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 ïĆś 1.1 m2g-1 for the Lee et al. (2012) and 7.7 ïĆś 3.5 m2g-1 for the 370 nm group in this study. Liu et al. (2008) showed that the absorption cross section of BC particles changed by their morphology and reduced or increased as they were being packed from chain-like shape, depending on the compactness of a cluster and the size and number of individual 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 SO2 availability likely to modify optical properties of carbonaceous aerosols. Considering these complex facts, our AAE of 1.0 for 880 nm group likely represents the absorption characteristics of aged and sulfate dominant aerosols in the study region. It is also noteworthy to mention that our AAEs were derived from regression method using seven wavelengths, whereas they were calculated using wavelength pairs in Lee et al. (2012).

8. Page 33221 This paragraph is very confusing as you contradict yourself quite a bit. You simply need to state that UV absorption was enhanced for 370 group and the 880 group had enhanced absorption in the IR. You mix up the terms former and latter, which is confusing. Try to state this in as few words as possible to make it clear.

As you pointed out, this part was reworded in the revised manuscript.

Considering the argument of Lack and Cappa et al. (2010), we compared AAEs calculated for four wavelength ranges: 370–520 nm, 450–660 nm, and 660–950 nm (Fig. 5c). For the 370 nm group, the AAE was increased only at UV region (370–520 nm) by 15 % relative to that of the visible range (470–660 nm) with no considerable difference in the other ranges. In contrast, the AAE of the 880nm group was decreased by 6 % at UV region (370–520 nm) but increased by 20 % at IR region (660–950 nm), compared to that of the visible range (470–660 nm). These results imply that the enhanced absorption of UV and longer visible light were responsible for the spectral dependence observed in the EBC of the 370nm and 880nm groups, respectively.

In addition, the last paragraph in section 3.3.1 was modified as follows.

When sulfate was predominant, particularly in aged air masses in Northeast Asia, sulfate likely contributed to light absorption at longer visible light probably as a coating on BC particles.

9. Throughout the paper you need to better define “enhanced absorption”. Enhanced relative to what, BC? A better term would be to use brown carbon absorption rather

than enhanced absorption.

It should be clarified as you pointed out. The “enhancement” was based on comparison between the EBC along 7 wavelengths and BC concentration shown in Figure 1a, which was stated in the revised manuscript (33213, line 18).

The following discussion regarding absorption enhancement along wavelength is based on this analysis.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C13164/2014/acpd-13-C13164-2014-supplement.pdf

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