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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Horizontal distributions of aerosol constituents and their mixing states in Antarctica during the JASE traverse" *by* K. Hara et al.

K. Hara et al.

harakei@fukuoka-u.ac.jp

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We would like to thank useful comments for improvement of our manuscript. Statements were corrected on the basis of referee's comments as follows.

Referee's comment: Page 11403: The authors discuss the Junge-slope without i) explaining how this quantity should be interpreted in practice, ii) or performing a proper interpretation of the observed slopes. Very few people are familiar with Junge-slopes or their meaning, so this whole discussion is worth little without modifying the text to better consider the two issues mentioned above.

Author's reply: The Junge slope is often used as index of shape of aerosol size distribution in fine – coarse modes. For instance, higher (lower) Junge-slope values can be observed in cases of high (low) number concentrations in fine mode and/or low (high) concentrations in coarse mode, respectively.

Referee's comment: Section 3.5.2 contains a few errors that need to be corrected. First, particle growth rates rarely exceed 10 nm/hour outside urban areas, being rather closer to 1 nm/hour in remote areas. As a result, it practically impossible that particles grow into the coarse mode by condensation and coagulation in the continental atmosphere (lines 10-12 on page 11411). For the same reason, it seems very unlikely that new particle formation could explain larger fraction of even .ne (>200 nm) particles (lines 22-23 on page 11410). A much more plausible reason in both cases is that these particle very originally primary particles (e.g. sea salt, dust or other) which had accumulated enough sulfate by condensation or cloud-processing to be classified as sulfate particles.

Author's reply: Based on comments from Referees #1 and #2, the statements were changed in the text as follows.

Higher relative abundance of sulfate particles in the outgoing traverse is likely to result from (1) increase of the number concentrations of sulfate particles in fine mode by particle growth of ultrafine sulfate particles by hygroscopicity and cloud processes, (2) cloud processes, and (3) decrease of the number concentration of sea-salt particles and the modified sea-salt particles during the summer.

Such a high relative abundance of coarse sulfate particles was not observed in the boundary layer but often in the lower free troposphere over Syowa Station (Hara et al., 2013). Consequently, sulfate particles can be grown to coarse mode through (1) hygroscopic growth, (2) cloud processes, (3) heterogeneous sulfate formation and (4) coagulation and condensation of condensable vapors (e.g., H2SO4 gas) under conditions with the low number concentrations of pre-existing particles on the Antarctic

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continent.

Referee's comment: Third, it is incorrect to state that sulfate particles were dominant in coarse mode (line 13 on page 11410), as in many case sea salt particle accounted for more than half of the particles.

Author's reply: Description was changed to

"sulfate particles were one of major aerosol particles in coarse mode and dominant in fine modes."

Referee's comment: The authors need to be more careful when discussing the reaction of nitrogen com-pounds with sea salt during their measurements. While it is highly likely that nitrate explains most of the Cl loss not accounted by sulfate in these measurements, there are no data to support this statement. The authors should more explicitly bring up that Cl loss due to HNO3 or other nitrogen reactions is a plausible explanation rather than a fact (pages 11415-16, abstract). Especially, when one reads the abstract only, one easily gets the impression that also nitrogen compounds were measured in sampled particles.

Author's reply: Ternary plots of Na-S-Cl in sea-salt particles strongly suggested high contribution of acidic species other than SO42-and CH3SO3- to sea-salt modification (Cl-loss). Plausible acidic species other than SO42-and CH3SO3- for Cl loss from sea-salt particles are reactive nitrogen oxides such as HNO3, N2O5, and NO3 (e.g.., Hara et al., 1999), and organic acids (Kerminen et al., 2000; Laskin et al., 2012). Previous studies of aerosol chemistry in the Antarctic regions indicated that NO3- concentrations were higher than organic acids (oxalate, formate, and acetate) (Jourdain and Legrand, 2002; Rankin and Wolff, 2003; Legrand et al., 2004; Hara et al., 2010, Weller et al., 2002, 2007; Weller et al., 2011). Furthermore, higher concentrations of reactive nitrogen oxides (e.g., HNO3 and NO) were observed on the Antarctic continent and coasts during summer (Davis et al., 2004; Dibb et al., 2004; Jones et al., 2011). Therefore, heterogeneous NO3- formation on sea-salt particles might make an important contri-

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bution to sea-salt modification in inland areas during summer.

These descriptions were added to the main text. Also, statements in abstract were changed to

Horizontal features of Cl/Na ratios imply that sea-salt modification (i.e. Cl loss) occurred on the Antarctic continent during summer. Most sea-salt particles in the continental region near the coast were modified with acidic sulfur species such as SO42- and CH3SO3-. By contrast, acidic species other than the acidic sulfur species (likely NO3-) contributed markedly to sea-salt modification in inland area during the traverse.

Minor issues Referee's comment: Page 11400, line 13: The cause-effect relationship does not sound correct here. I seems more plausible that wind causes turbulence than other way around.

Author's reply: The statement was changed to

In addition to diurnal features of wind speed, the wind speed increased because of cyclone approach

Referee's comment: Page 11404: The authors discuss possible errors in estimating aerosol mass concentration from OPC measurements. Two other issues should be mentioned here. First, the association of water also dilutes salts, so the assumed density of the particles is overestimated at high relative humidity. Second, how accurately the diameter obtained from OPC measurement correspond to that needed to calculate particle mass (particle mass equivalent diameter)?

Author's reply: Plausible errors in the estimation of aerosol mass concentrations were added in the text as follows;

In section 2-3-1: OPC was calibrated using polystyrene latex particles with spherical shape and refractive index of 1.59–0i. Thus, size provided from OPC is "optically PSL-equivalent size".

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In section 3-2: Spherical shape and density were assumed in the estimation of aerosol mass concentration. As mentioned above, diameters in OPC are "optically PSL-equivalent size", so that the estimated aerosol volume means "PSL-equivalent volume" in this study.

Therefore, the estimated mass concentrations included masses of water in aerosol particles in this study. The presence of water in aerosol particles can cause overestimation of aerosol density and mass concentrations because of salt dilution by water.

Referee's comment: Page 11412, line 18: Hara et al (2014) is not in the reference list.

Author's reply: Paper of Hara et al (2014) is preparing for publication. Therefore, statement of "Hara et al (2014)" was removed from the text.

Referee's comment: Page 11413, lines 5 and 7: I do not understand the notation M % (mean, N %).

Author's reply: Descriptions of range and mean value of relative abundance were changed as follows;

The relative abundance of mineral particles ranged in n.d. – 14.6 % in coarse mode and n.d. – 0.4 % in fine mode during the incoming traverse, although the relative abundance ranged in n.d. – 5.7% in coarse mode and n.d. – 0.1 % in fine mode during the outgoing traverse. Mean relative abundance in coarse mode was 3.8 and 1.8 % in incoming and outgoing traverses, respectively, and that in fine mode was 0.1 % and 0.01 % in incoming and outgoing traverses.

Other points were corrected on the basis of the comments.

All modified sentences and words were marked in text of the revised manuscript.

The manuscript was checked by native English speaker again (FASTEKJAPAN: http://www.fastekjapan.com/).

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