

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

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Received and published: 11 August 2014

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: In general large parts of the Results and Discussion section could be written more concisely. There are several repetitions, especially concerning chapters 3.4, 3.5 and 3.6, making the text somewhat exhausting and a challenge for the readers' endurance.

Reply from Authors: We attempt to arrange the statements in sections of 3.4–3.6. Sev-

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eral repetitions were removed from the text.

Referee's comment: Furthermore, potential consequences of single particle modification and fractionation after sampling (storage as well as high vacuum conditions under SEM-EDX analysis) should be discussed or an adequate reference should be provided.

Reply from Authors: We added some references in the text as follows;

After direct sampling, aerosol samples were kept in air tight boxes including desiccant until analysis in our laboratory in Japan to avoid humidification that can engender morphological change, modification (chemical reactions), and fractionation, as described by Hara et al. (2002, 2005, 2013). Therefore, aerosol modification and fractionation might be negligible, although aerosol constituents can be localized in each aerosol particle because of deliquescence of deliquescent aerosol particles under dry conditions.

Referee's comment: Chapter 2-3-1, Page 11398, lines 21-22: Please clarify: Is 23-25 s the temporal resolution of the measurement? How long was the typical over-all sampling period at each site?

Reply from Authors: Yes. Time of 23-25 s is measuring resolution. Aerosol number concentrations were measured during direct aerosol sampling at camp sites. Because sampling time was shown in Section of 2-3-2, we modified the description here as follows;

Aerosol number concentrations were recorded in resolution of every 23–25 s (corresponding to 1 liter air sucking) during direct aerosol sampling.

Referee's comment: Chapter 2-3-2, Page 11399, lines 18-22: Did you analyze all sampled particles? If not, please specify the percentage/fraction of the analyzed fine and coarse mode particles.

Reply from Authors: Because of low number density on the sampling substrates in coarse mode, most aerosol-sampled area on the substrates was analyzed in this study. In some samples, we analyzed all coarse particles on the substrates. On the other

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hand, 20 – 30 % of aerosol-sampled area on the substrates was analyzed in aerosol samples in fine mode.

The description is modified in the text as follows;

In aerosol samples in coarse mode, most aerosol-sampled area on the substrates was analyzed in this study. Although we attempted to analyze as many coarse particles as possible, the lower aerosol number concentrations in coarse mode limit the number of the analyzed aerosol particles in this study. On the other hand, 20 – 30 % of aerosol-sampled area on the substrates was analyzed in aerosol samples in fine mode.

Referee's comment: Page 11400, lines 19-20: What is the meaning of “: :the diurnal maxima were mutually synchronized: :”?

Reply from Authors: Features of wind speed was synchronized to those of air temperature. The description is modified in the text as follows;

However, the diurnal maxima of wind speed were mutually synchronized well to those of air temperature at latitudes higher than 74 °S, when their diurnal features occur.

Referee's comment: Chapter 3-1-2, Page 11401: Please specify the method of trajectory calculation (isentropic, 3-dimensional, : :) and provide a description of the colour code for Fig. 3 (what is meant with “latitude”? Is it the starting point of the respective trajectory?).

Reply from Authors: Methods of trajectory calculation were described already in the text as follows;

“the 5-day backward trajectory was computed using the NCEP reanalysis dataset and kinematic mode in this study (Draxler and Rolph, 2013).”

Because words of “vertical motion mode” were used in HYSPLIT web page, the description in text was modified to

“the 5-day backward trajectory was computed using the NCEP reanalysis dataset and

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vertical motion mode in this study (Draxler and Rolph, 2013).”

Color code in Fig.3 indicates latitude at starting points of trajectory. The following explanation was added into caption in Fig.3.

Color code corresponds to latitude at starting points of trajectory.

Referee's comment: It should also be noted that especially the reliability of vertical (but also lateral) movement of a given trajectory may be very low due to the lack of appropriate meteorological data from continental Antarctica. This has to be considered when interpreting the data.

Reply from Authors: The following description about reliability (uncertainty) of trajectory analysis was added into the text in the revised manuscript.

Uncertainty of trajectory analysis is derived from resolution of meteorological data (wind field), calculation scheme, and trajectory model. Kahl et al. (1989) and Stohl et al. (1995) presented that error reached to ~1000 km after trajectory calculation for 5 days. Therefore, the 5-day backward trajectory was analyzed in this study. Indeed, previous works (Reijmer et al., 2001, 2002; Hara et al., 2004, 2013; Suzuki et al., 2008) used 5-day backward trajectory analysis to discuss origins of moisture and aerosols. In general, uncertainty can be larger when starting altitude of trajectories is located in boundary layer. The 5-day backward trajectories from altitudes in boundary layer – free troposphere, however, were consistent well with vertical profiles of aerosol constituents over Syowa Station, Antarctica (Hara et al., 2013). In this study, we calculated the trajectories from 200, 500, and 1000 m above ground level. The trajectories showed similar vertical features and transport pathway in most cases, so that 5-day backward trajectory might be applied to discuss air mass history even in the Antarctic continent.

Referee's comment: Chapter 3-3, Page 11405, lines 6-8: Did you consider the “satellites” in your analyses?

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Reply from Authors: Yes. But most signals may be derived from center nuclei of the “satellite”.

Referee's comment: Chapter 3-3, Page 11405, lines 13-14: Please specify the term “stain or staining”!

Reply from Authors: “Stain” is correct. We changed to “stain” in the text.

Referee's comment: In addition, the particles presented in Fig. 6 are in both cases clearly smaller than 2 micrometer, but 6b (upper picture?) was denoted by coarse mode particle. Was there some kind of shrinking process during SEM analysis under high vacuum conditions?

Reply from Authors: We use “aerodynamic diameter” in the cut-off diameter. Aerodynamic diameter was estimated in particles density of 1 g cm⁻³. Density of aerosol particles is larger, for instance, NaCl (2.2 g cm⁻³), and H₂SO₄ (1.84 g cm⁻³). Therefore, aerosol particles slightly smaller than the cut-off diameter can be collected on the sample substrates, depending on particle density.

These explanations were added into the text in Section 2-3-2.

Furthermore, most aerosol particles in atmosphere contain water in this study. The stain around particles was direct evidence of presence of liquid phase. After sampling, aerosol samples were kept under dry conditions. Aerosol particles were exposed in high vacuum condition in SEM-EDX analysis. Thus, dry and high vacuum conditions can cause water evaporation from aerosol particles. Therefore, small residual of water soluble and insoluble matters remained inside of the stain or satellite structure on the substrates, as shown in Fig.6.

Referee's comment: Chapter 3-4, Page 11405-11406: Being no expert in EDX analysis like probably most of the potential readers, some words about the reliability of this method would be helpful, especially in terms of the specific problem, that for large coarse mode particles potentially mainly the surface composition is probed, which may

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not be representative for the whole particle.

Reply from Authors: Analytical depth in SEM-EDX analysis depends on accelerating voltage, chemical compositions, density, and others. Although secondary electron can be emitted from depth of ~10 nm (e.g., Ding et al., 2009), characteristic X-ray can be emitted from depth of a few micrometer (e.g., Goldstein et al. 2003). Most of coarse particles were smaller than 3 micrometer in diameter in this study. Thus, characteristic X-rays were obtained from whole particle in fine and coarse mode in this study. When thickness of particles is larger than ca. 5 micrometer, compositions near surface (thickness of a few micrometer) may be detected. However, only a few particles larger than 5 micrometer were present in one sample.

These descriptions were added into the text.

Referee's comment: Page 11410, lines 4-6: The conclusion that differences of the relative abundance between incoming and outgoing traverse were caused by seasonal features appears arguable. I suggest that different general weather situation and meteorological effects could have played a significant role.

Reply from Authors: Although blizzard by cyclone approach occurs until early December at Syowa Station, few blizzards occur in mid-December – January (Sato and Hirasawa, 2007). The seasonal feature of cyclone approach relate closely to origins of air masses over the Antarctic continent as suggested by Suzuki et al. (2013). Therefore, the difference of the relative abundance between in the incoming traverse and in the outgoing traverse might correspond to seasonal features of sea-salt particles, air mass origins, and transport pathway from the end of spring toward summer on the Antarctic continent.

The descriptions in the text were changed to above sentences.

Referee's comment: Pages 11411-11413, lines 14-15, 3-4, 19-21, and 5-8: Please specify the corresponding detection limits.

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Reply from Authors: Mean analyzed particles in each sample were 58 particles in coarse modes and 963 particles in fine mode. Detection limit of relative abundance was ca. 0.1 % in fine mode and 1-2 % in coarse mode except the samples with lower aerosol density on the substrate in coarse mode.

These descriptions were added into the text.

Referee's comment: Page 11411, lines 9-10: "Figure 5 shows [: :] lower near the surface on the Antarctic continent" – for me it is not obvious that Figure 5 really shows this! Please clarify.

Reply from Authors: The description about Figure 5 seemed to give you confusion, and was necessary for what authors want to discuss. Therefore, the description was removed from the text.

Referee's comment: Page 11411, lines 11-13: "sulphate particles can be grown to coarse mode through coagulation and condensation: : : " – To me this sounds rather unrealistic! Most probably sulphate in super-micron aerosol has been formed by heterogeneous chemical processes on the surface of pre-existing coarse mode sea salt or mineral dust particles or by cloud processes (see chapter 3.5.3).

Reply from Authors: Coarse sulfate particles did not contain Na, Al, or Si, so that these particles might be composed of SO₄²⁻ and CH₃SO₃⁻, as suggested previous works by Hara et al., 1995. Therefore, coarse 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 continent.

The sentences were added into the text.

Referee's comment: Page 11411, lines 19-21: Here again: seasonal features causing the differences in K-rich particles appear arguable.

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Reply from Authors: Similar to discussion about features of sea-salt particles, this difference implies seasonal features of K-rich sulfate particles, air mass origins, and transport pathway in the inland area during late-spring – summer.

The descriptions in the text were changed to above sentences.

Referee's comment: Page 11416, lines 6-11: I do not really understand why high Cl/Na ratios (and strong winds) indicate surface generated sea-salt particles modified by HNO₃ (I would suppose low Cl/Na ratios in this case).

Reply from Authors: We attempt modify sentences easy to explain as follows,

As discussed in sections of 3-2 and 3-5-1, therefore, most coarse sea-salt particles on the Antarctic plateau were likely to have originated from surface snow on the Antarctic continents. Figures 11 and 12 show that high Cl/Na ratios on the Antarctic plateau often corresponded to conditions with strong winds and drifting snow. Thus, sea-salt particles immediately after release from snow surface may have high Cl/Na ratios. Then, sea-salt particles in coarse mode might be modified gradually with reactive nitrogen oxides such as HNO₃ in the continental atmosphere during transport over the Antarctic continent.

Referee's comment: Page 11416, lines 25-29: This section appears diffuse and I do not understand what the authors try to tell us with this statement.

Reply from Authors: To simplify the statement, some sentences were removed from the text. The sentences were changed to

By contrast, S/Na ratios in fine sea-salt particles exceeded mostly 0.5 during the traverse. The high S/Na ratios in each sea-salt particle in fine mode imply that sulfates were formed on the fine sea-salt particles through heterogeneous reactions with gaseous sulfur species such as H₂SO₄ and SO₂.

Referee's comment: Page 11418, lines 20-21: I can see no reason for a "Supplementary" for just one figure, which can readily be presented in the main text!

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Reply from Authors: Plots of Mg/Na ratio in fine mode were added to Figure 14. Some explanation was added into the main text.

Referee's comment: Concluding remarks, Page 11422, line 9: sea salt modification (not fractionation) is meant here.

Referee's comment: Exactly, "sea salt modification" is corrected here.

Other points such as typos were corrected on 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/>).

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 11393, 2014.