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

Anonymous Referee #1

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General comments:

Aerosol measurements addressing chemical composition and particularly mixing states derived from single particle analyses are certainly rare for the Antarctic continent. The submitted manuscript by Hara and colleagues presents such results conducted during the Japanese-Swedish joint Antarctic expedition from November 2007 through January 2008. To my knowledge the paper presents original and new results leading to the following crucial conclusions: (i) Measurements indicate aerosol production caused by drifting snow; (ii) significant aerosol modification (Cl-depletion caused by biogenic sulphur aerosol and probably HNO₃) as well as fractionation (especially

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concerning Mg/Na ratios); (iii) there is indication of sea salt fractionation occurring on snow surfaces on the plateau; (iv) high sulphate enrichment in coarse mode particles was detected, probably by sulphate formation from gases precursors by heterogeneous processes on sea salt particles.

Even though such measurements are spatially and temporally restricted and thus parts of the conclusions (role of HNO₃, suggested Mg fractionation mechanism) appear speculative, the outcome of this study has some relevant implications for understanding geochemical and atmospheric processes in Antarctica. Nevertheless, there are several points which require careful revision/supplements which will be detailed below. 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. 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 (this point is not explicitly addressed in the here cited papers by Hara et al., 2005 and 2013). Finally, the language often reads not fluently and is partly circuitous. I recommend proof read of the text by a native speaker! Notwithstanding, I am confident that the data presented here are of high quality and on the whole, the subject is appropriate to ACP. Hence, the paper can be accepted after revisions according to my specified suggestions from above and listed below.

Specific comments:

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?

Chapter 2-3-2, Page 11399, lines 18-22: Did you analyse all sampled particles? If not, please specify the percentage/fraction of the analysed fine and coarse mode particles.

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

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?). 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.

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

Chapter 3-3, Page 11405, lines 13-14: Please specify the term “stain or staining”! In addition, the particles presented in Fig. 6 are in both cases clearly smaller than $2 \mu\text{m}$, but 6b (upper picture?) was denoted by coarse mode particle. Was there some kind of shrinking process during SEM analysis under high vacuum conditions?

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 not be representative for the whole particle.

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.

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

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.

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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).

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

Page 11416, lines 6-11: Maybe I am a bit slow-witted, but 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).

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

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!

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

Some typos etc.:

Page 114395, line 25: "...during summer in Antarctica..." (not "during the summer on the Antarctica").

Page 11401, line 21: Fig. 3d is missing!

Page 11406, line 26: "... Geilfus et al." (not "Gelifus).

Page 11414, line 5-6: maybe a better phrasing is "...internally mixed sea-salt and mineral particles..."

Page 11415, line 16: "...approximately around 0.2..." (not. "...in 0.2...").

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Supplementary-Fig.01, caption: Atomic ratio of Mg/Na in fine mode (NOT coarse mode!) is shown.

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

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Interactive
Comment

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Interactive Discussion

Discussion Paper

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