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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tethered balloon-borne aerosol measurements: seasonal and vertical variations of aerosol constituents over Syowa Station, Antarctica" *by* K. Hara et al.

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The authors thank the anonymous reviewer for helpful comments.

Ref. #2 comment: This study totally relied on the SEM-EDX technique, which is basically limited to determine specific element ratios. Hence, it would be helpful if an intercomparison with available ground-based aerosol samples, analysed e.g. by ion chromatography, is available.

Reply from authors: Because we made ground-base aerosol sampling in 2004 – 2006 at Syowa Station for bulk analysis using ion chromatograph, the intercomparison is

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available. At the moment, we have plan for other papers about seasonal variations of aerosol constituents at Syowa Station. We are going to discussed them elsewhere.ãĂĂThus, the intercomparison was not added in the revised manuscript.

Ref. #2 comment: The possibility and consequences of aerosol modification/fractionation after sampling should be discussed. Given that single particles are probed, this may cause considerable artifacts (e.g. post-sampling fractionation may be responsible for the formation of MgCl2 particles or totally Mg-free sea salt particles).

Reply from authors: Because analyzed particles were almost covered with analytical spots, analytical artifact contributed insignificantly to aerosol modification/fractionation in this study. If the post-sampling processes proceeds as pointed out by referee, plausible processes are (1) phase transformation under storage (until analysis), (2) particle separation under lower relative humidity and high vacuum conditions, and (3) particles separation by radiation of electron beam. Aerosol samples were kept under conditions with airtight and relative humidity lower than 5 – 10 % until analysis. Because of air tight and dry conditions, sea-salt modification proceeds hardly during sample storage. Also, dry condition did not result in phase transformation, so that aerosol separation and fractionation cannot occur. In SEM-EDX analysis, aerosol particles were exposed to high vacuum condition (ultimately dry condition). Particle separation, however, occur hardly, though dry conditions can engender deliquescence and localization of aerosol constituents in each particle. Particle separation by radiation of electron beam was not observed in SEM-EDX analysis. In addition, these particles have stains or satellite structures around the particles. These structures may be formed by impaction on sample substrates during the aerosol sampling as shown in Fig.4. Hence, we concluded that modified sea-salt particles, Mg-rich sea-salt particles, Mg-free sea-salt particles, Mg-rich sulfates, and MgCl2 were not derived from post-sampling modification/fractionation processes and analytical bias, but present in the ambient atmosphere. These descriptions were added to text in revised manuscript.

Ref. #2 comment: Analytical accuracy, reproducibility and limits of detection (LOD)

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should be provided throughout.

Reply from authors: The following descriptions were added into text revised manuscript. Generally, the detection limit of EDX analysis is 0.1–1% depending on analytical conditions, elemental compositions, and particle morphology. We analyzed artificial particles (such as NaCl, Na2SO4 and their mixtures) with the size range of 0.2–6 micrometer to verify their analytical quality. In our analytical conditions (e.g., counting time of 30 s), relative error from counting statics was a few percent, even in sub-micrometer particles for all elements heavier than Na. Analytical deviation in elements heavier than Na was estimated as ïĆśseveral percent in the range of sub-micrometer – super-micrometer in this study. The deviation tended to be smaller for larger particles. These values were closely coincident with results reported by Laskin and Cowin (2001).

Ref. #2 comment: Large parts of the Results and Discussion section could be written more concisely (see also comment by referee #1!). In addition short sub-summaries for each sub section would be helpful and welcome!

Reply from authors: We attempted to address description in sections 3-4 - 3-7. Base on referee's comments, comparison between "summer" and "winter" was shown and discussed at first in each section. In addition, approximately 30% of text in the ACPD manuscript was removed from these sections. Also, typical figures are merged and shown in the main text. All plots are moved to 'Supplementary'. Because we changed last section to "summary", short sub-summaries for each sub section was not added in the revised manuscript.

Ref. #2 comment: Page 8157, lines 20-29: The term "upper atmosphere" should be avoided in this context, because it is usually reserved for the atmosphere above the mesopause!

Reply from authors: We changed the term "upper atmosphere" to "boundary layer – free troposphere" in the revised manuscript.

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Ref. #2 comment: Page 8185-8187, Summary and Conclusions: This chapter is, if anything, just a summary but concrete conclusions are unfortunately missing and should be provided.

Reply from authors: We changed last section to “summary”. Furthermore, amount of text in “summary” decreased by ca. 70 % relative to the text in ACPD manuscript.

Ref. #2 comment: Page 8186, lines 22-25: It should be noted that the impact of nitrate was merely deduced and not really measured.

Reply from authors: We addressed description in the text as follows; Furthermore, comparison with aerosol chemical data from bulk analysis implies that sea-salt particles were modified slightly with NO3- through the year.

Ref. #2 comment: Page 8187, lines 6-7: Does this mean that strong vertical gradients in relative humidity occurred above the well-mixed boundary layer (in a well-mixed layer, any gradients should be negligible)? Please clarify.

Reply from authors: We addressed description in the text as follows; The third type was often obtained under the conditions with strong vertical gradient of relative humidity in the well-mixed boundary layer.

Others: Some typos and grammatical errors were corrected in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 8153, 2013.