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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size distributions of dicarboxylic acids, ketoacids, α-dicarbonyls, sugars, WSOC, OC, EC and inorganic ions in atmospheric particles over Northern Japan: implication for long-range transport of Siberian biomass burning and East Asian polluted aerosols" *by* S. Agarwal et al.

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Response on the comments from Referee #1

(Note: The revised version of the MS is attached as supplement document)

General comments:

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Interactive Discussion

Discussion Paper

This manuscript deals with a comprehensive characterization of the organic and inorganic composition of size-segregated aerosols that were collected in Northern Japan during a 2005 summer episode and are affected by air masses from different origins, including Siberia, the Asian continent, and seas surrounding Japan. An important result is that the organic chemical composition is heavily affected by biomass burning that is going on in the Siberian boreal forest region. Detailed and valuable information is provided on the organic acid composition of the carbonaceous aerosols, allowing one to draw important conclusions about the aging of the aerosols. It is clearly shown that short-chain dicarboxylic acids such as oxalic acid have high loadings in the fine size mode and that their contribution to the OC mass is enhanced in the biomass burning-affected samples, due to chemical processing of their organic precursors during long-range transport. The manuscript needs to be improved with regard to clarity, and English grammar and style.

Specific comments:

6717 – l. 7: with regard to the sources of saccharidic compounds, I suggest to mention in a first instance that they originate from fungal spores and plant material; it is well known that arabitol, mannitol and trehalose are characteristic for fungal spores (Lewis and Smith, 1967; Bieleski, 1982), while monosaccharides such as fructose, glucose and inositol are characteristic for plant material such as pollen, fruit, and fragments (Bartolozzi et al., 1997; Baker et al., 1998; Pacini, 2000). In a second instance, soil dust (as an aerosol source) and biomass burning should be mentioned. Soil dust probably contains fungal spores, and plant pollen and debris, while biomass burning of plant material may also release substantial amounts of sugar compounds.

Refs:

Baker, H. G., Baker, I., and Hodges, S. A.: Sugar composition of nectars and fruits consumed by birds and bats in the tropics and subtropics, Biotropica, 30, 559–586, 1998.

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Bartolozzi, F., Bertazza, G., Bassi, D., and Cristofori, G.: Simultaneous determination of soluble sugars and organic acids as their trimethylsilyl derivatives in apricot fruits by gas-liquid chromatography, J. Chromatogr. A, 758, 99–107, 1997.

Bieleski, R. L.: Sugar alcohols, in: Encyclopedia of Plant Physiology, volume 13A, Plant Carbohydrates, volume I, Intracellular Carbohydrates, Loewis, A. and Tanner, W. (Eds), Springer-Verlag, Berlin, 158–170, 1982.

Lewis, D. H. and Smith, D. C.: Sugar alcohols (polyols) in fungi and green plants: 1. Distribution, physiology and metabolism, New Phytol., 66, 143–184, 1967. Pacini, E.: From anther and pollen ripening to pollen presentation, Plant Sys. Evol., 222, 19–43, 2000.

Response: Following the comments, we have modified the text as:

"In the ambient aerosols, dicarboxylic acids are mainly produced by photochemical processes (Kawamura and Yasui, 2005) of the precursor species from different sources, such as biomass burning, vehicular exhaust, etc., while saccharidic compounds are originated from fungal spores and plant material. More precisely, arabitol, mannitol and trehalose are characteristic for fungal spores (Lewis and Smith, 1967; Bieleski, 1982), while monosaccharides such as fructose, glucose and inositol are characteristic for plant material such as pollen, fruit, and fragments (Bartolozzi et al., 1997; Baker et al., 1998; Pacini, 2000). Soil dust probably contains fungal spores, and plant pollen and debris, while biomass burning of plant material may also release substantial amounts of sugar compounds (such as levoglucosan) in aerosol particles (Simoneit et al., 2004a, 2004b)." Please see Lines 78 to 87 in the revised MS.

We have also incorporated the references as suggested by the reviewer in the revised version. Please see Lines 604 to 608, 614 to 616 and 707 to 708 in the revised MS.

6719 – l. 13: essential details need to be provided about the trimethylsilylation procedure. Was pure BSTFA used or did it contain 1% trimethylchlorosilane as a catalyst?

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Was the reagent diluted with pyridine? It is known that the use of BSTFA alone without catalyst results in an incomplete derivatization of levoglucosan.

Ref.: Zdráhal, Z., Oliveira, J., Vermeylen, R., Claeys, M., and Maenhaut, W.: Improved method for quantifying levoglucosan and related monosaccharide anhydrides in atmospheric aerosols and application to samples from urban and tropical locations, Environ. Sci. Technol., 36, 747–753, 2002.

Response: As suggested, the information about BSTFA derivatization has been incorporated in the revised manuscript as follows.

"On the other hand, sugars were extracted from another aliquot of filter cut with a dichloromethane and methanol mixture (2:1), derivatized with N,O-bis-(trimethylsilyl) trifluoroacetamide (BSTFA) (containing 1% trimethylsilyl chloride) and pyridine as a catalyst, and determined using a GC/mass spectrometry (GC/MS) (Wang et al., 2006b)." Please see Lines 146 to 149 in the revised MS.

6719 – l. 8: The GC detection technique should be mentioned. I assume it was flame ionization detection.

Response: The details have been provided in the revised manuscript as follows.

"The derivatives were determined using a capillary gas chromatograph (GC) with a flame ionization detector (FID)." Please see Lines 141 to 142 in the revised MS.

6722 – l. 10: earlier in the paper (page 6721 – l. 13) it was already mentioned that sulfate can have a marine or anthropogenic origin. I suggest to simply write here: "Moderate concentrations of sulfate were found in the 10 and 10-11 August samples (Fig. 2), suggesting an influence of marine and local anthropogenic origin."

Response: As suggested, the sentence has been corrected. Please see Lines 225 to 226 in the revised MS.

6726 – l. 24: There is recent literature data indicating that levoglucosan is not as stable

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as originally thought (Hoffmann et al., 2010). I am also aware of on-going studies on the photochemical degradation of levoglucosan (poster by A. Robinson from Carnegie-Mellon, Pittsburg, at the 2009 AGU Fall Meeting).

Ref.: Hoffmann, D., Tilgner, A., Iinuma, Y., and Herrmann, H.: Atmospheric stability of levoglucosan: A detailed laboratory and modeling study, Environ. Sci. Technol., in press, 2010; doi:10.1021/es902476f.

Response: As suggested by the reviewer, we have modified the sentence giving the references as follows.

"Levoglucosan (1,6-anhydro-β-D-glucopyranose), anhydrosaccharide, is a major pyrolysis product of cellulose, and is recognised as a tracer of biomass burning (Simoneit et al., 1999). However, it can also be generated by non-combustive processes, e.g., hydrolysis or microbial degradation of carbohydrates (Simoneit et al., 1999; 2000). Although it is believed that levoglucosan is a non-degradable species in the atmosphere, it is important to note that recent studies show that levoglucosan is not as stable as originally thought (Hoffman et al., 2010)." Please see Lines 364 to 370 in the revised MS.

6727 – l. 15: Here again, I would in a first instance mention that the primary saccharides are derived from fungal spores (which are airborne or become airborne due to biomass burning) and plant material, and only in a second instance by surface soil and unpaved road dust.

Response: As suggested by the reviewer, we have modified the text as:

"The primary saccharides are derived from fungal spores (Lewis and Smith, 1967; Bieleski, 1982) and plant materials (Bartolozzi et al., 1997; Baker et al., 1998; Pacini, 2000) (which are airborne or become airborne due to biomass burning), and by resuspension of surface soil and unpaved road dust (Simoneit et al., 2004c). Recently, Russell et al. (2010) reported the prevalence of sugar compounds in ocean aerosols

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via bubble bursting processes" Please see Lines 385 to 390 in the revised MS.

Technical corrections:

The manuscript needs to be carefully checked for English grammar and style (relating to the correct use of articles, plural/singular, tenses, etc. . .). The list below is far from complete (except for the abstract):

Response: We have carefully checked the English grammar and style of the manuscript.

6715 – l. 4: . . .. an Andersen impactor . . ...

Response: corrected. Please see Line 27 in the revised MS.

6715 – l. 6: . . ... molecular composition of . . .. . .

Response: corrected. Please see Line 28 in the revised MS.

6715 – l. 8: . . .. . . backward trajectories . . .. . .

Response: corrected. Please see Line 31 in the revised MS.

6715 – l. 9: . . .. . . during the campaign, the air masses arrived from . . ...

Response: corrected. Please see Line 32 in the revised MS.

6715 – l. 16: . . .. aerosols transported from Siberia . . ..

Response: corrected. Please see Line 38 in the revised MS.

6715 – l. 17: . . ... aerosols originating from China . . ..

Response: corrected. Please see Line 39 in the revised MS.

6715 – l. 21: . . ... in the fine mode,. . ..

Response: corrected. Please see Line 43 in the revised MS.

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6716 – l. 10: The changes in the chemical composition can alter the optical and microphysical . . ...

Response: corrected. Please see Line 56 in the revised MS.

6716 – l. 17: The western North Pacific rim is a receptor region for anthropogenic and mineral dust aerosols originating from the Asian continent. (too much repetition of "Pacific region")

Response: corrected. Please see Line 63 in the revised MS.

C938 6717 – l. 13: However, the size distributions . . .. (Mochida et al., 2007). This information is important because it allows one to gain important information on the sources and . . ..

Response: corrected. Please see Line 93 in the revised MS.

6719 – l. 4: Briefly, a part of the filter . . .. (instead of "aliquot")

Response: corrected. Please see Line 138 in the revised MS.

6719 – l. 23: 2 M HCl

Response: corrected. Please see Line 156 in the revised MS.

6720 – l. 2: . . .. eluent of 2 mM Na2CO3 + 1.7 mM NaHCO3 ????

Response: corrected. Please see Line 162 in the revised MS.

6721 – l. 6: The trajectories showed that air masses arriving at Sapporo originated from . . ...

Response: corrected. Please see Line 195 in the revised MS.

6721 – l. 10: . . ... aerosols originating from . . ...

Response: corrected. Please see Line 198 in the revised MS.

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6724 – l. 17: This suggests that biomass burning . . ...

Response: corrected. Please see Line 305 in the revised MS.

6725 – l. 1: . . .. in the coarse mode . . ...

Response: corrected. Please see Line 324 in the revised MS.

6725 – l. 13: . . ... aerosol particles have a marine influence.

Response: corrected. Please see Line 328 in the revised MS.

6728 – l. 14: Bauer et al. (2002)

Response: corrected. Please see Line 414 in the revised MS.

6735 – l. 13: Bauer, H., ...

Response: corrected. Please see Line 609 in the revised MS.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C3287/2010/acpd-10-C3287-2010-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6713, 2010.