**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.

Anonymous Referee #3

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General Comments. The manuscript entitled, "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" presents a comprehensive set of chemical measurements of atmospheric aerosol. The rigorous chemical analyses provide use-

ful, accurate measurements of many aerosol components including WSOC, molecular tracers, and inorganic ions. The observed correlation between diacids and levoglucosan is an important finding, since many previous reports of diacids have not indicated a source so clearly. I do have two main concerns with the implications of this work, as presented by the authors. The first is that these results of size-resolved chemistry are presented as relevant for CCN prediction. However, all particles below 1 micron are grouped together meaning that over the 100nm-400 nm range, there is no observed difference in particle composition. More detail on this is presented below. The second concern pertains to the extremely limited sample size of this measurement set (6 samples). Although air was sampled from a variety of source regions, it seems like a stretch to say that the composition of 1 or 2 samples can represent "biomass burning" while just 1 other sample can represent "pollution."

The authors should consult a native English speaker for grammatical corrections. After making more than 11 grammatical corrections to the abstract alone, I have not attempted to provide further text editing. However, the authors do make clear points and do provide adequate motivation for their work. Once the many small mistakes are corrected, the paper will communicate their findings clearly.

I recommend this paper for publication with major grammatical correction and the following itemized corrections.

Specific Comments.

1. Pg. 6715, lines 16-21: add standard deviations to mean values to indicate the range of observed variability.

2. Pg. 6716, lines 4-6: This sentence is awkward, and it's unclear whether the authors intend to state that primary sources outweigh secondary sources, or whether they simply wanted to say that both primary and secondary sources exist. Not all sources can be "major" sources.

3. Pg. 6716, line 27: Please add a reference for the uptake of acidic gases onto alkaline dust components. Krueger et al. 2004 can be used.

4. Pg. 6718, line 17: The measurements of size-segregated chemical composition are relevant to understanding chemical transformation pathways (e.g. photochemical and cloud processing). However, with respect to cloud condensation nuclei activity, it is necessary have size resolved composition below 1 micron since particles larger than 1 micron are likely to activate with only a small amount of soluble material. This point is particularly important since this paper is reporting measurements of aged aerosol that is more likely to contain some soluble material. Therefore, the implications of this work for future CCN studies are quite limited.

5. Pg. 6718, line 20: The small number of samples collected means that sampling statistics are poor, so the authors should be careful about drawing too many conclusions from the differences in the composition related to difference back trajectories. Wherever possible, the authors should add error or variability bars to help readers understand that the measured differences are real, and not within the noise. This comment applies to the text in Section 3.1 as well, where the composition of each type of aerosol is discussed.

6. Pg. 6721, line 22: Add appropriate month after "8 August and 8-9" or, since both are in August, rewrite it as "8 and 8-9 August." This pattern should be applied to all instances where dates are listed.

7. Pg. 6724, lines 12-13: The authors state that diacids are higher in samples 1-3 than in 4-6. Perhaps a better measure would be diacid fraction of OC. It's difficult to argue that biomass burning contributes significantly more diacid in absolute terms.

8. Pg. 6724, lines 15-18: What are the correlation coefficients between sulfate and oxalic acid (and sulfate with total diacids)? To show that biomass burning is a stronger source of diacids than fossil fuel combustion, the authors must provide a comparison of the correlation coefficients. It may be that many components increase their concen-

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tration simultaneously, but this does not mean they are from the same source.

9. Pg. 6728: Could some of the sugars be attributed to marine aerosols? There has been recent work (Russell et al., 2010) showing the prevalence of sugar compounds in primary marine aerosol. Either way, the authors should address this issue, as other readers will be likely to have a similar thought.

References listed:

Krueger, B.J., V.H. Grassian, J.P. Cowin, and A. Laskin (2004), Heterogeneous chemistry of individual mineral dust particles from different dust source regions: the importance of particle mineralogy, Atmospheric Environment, 38, 6253-6261.

Russell L.M., L. N. Hawkins, A. A. Frossard, P. K. Quinn, and T. S. Bates (2010), Carbohydrate-Like Composition Submicron Atmospheric Particles and their Production from Ocean Bubble Bursting. Proceedings of the National Academy of Sciences, doi:10.1073/pnas.0908905107

Technical Comments. 1. Pg. 6715, line 4: insert "an" before "Anderson"

2. Pg. 6715, line 8: pluralize "trajectory" to "trajectories"

3. Pg. 6715, line 9: insert "the" before "campaign"

4. Pg. 6715, line 9: omit "the" before "air masses" and omit "were"

5. Pg. 6715, line 10: insert "a" within parentheses before "biomass burning region" and insert "from" before "China" and insert "an" within parentheses before "anthropogenic"

6. Pg. 6715, line 11: insert a comma after "9-10 August"

7. Pg. 6715, line 13: put "i.e. SO4 [. . .]" in parentheses

8. Pg. 6715, line 18: replace "maximized" with "were highest"

9. Pg. 6715, line 22: replace "is abundant" with "are abundant" to match "biomass burning products"

10. Pg. 6715, line 22: replace "that" with "of"

11. Pg. 6715, line 23: replace "in Siberian region have" with "illustrating that the Siberian region has"

12. Figure 2 needs vertical error bars to clearly show that one point is statistically higher than another.

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