

Interactive comment on “Sources and characteristics of size-resolved particulate organic acids and methanesulfonate in a coastal megacity: Manila, Philippines” by Connor Stahl et al.

Anonymous Referee #2

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This manuscript reports the analytical results of five organic acids and MSA in the size-resolved aerosols from Manila and discusses their sources and characteristics based on the correlation analyses with a 16-month dataset of inorganic ions and trace metals. Because of no studies on organic acids in the aerosols from Manila areas, authors' dataset may be of interest for the community of atmospheric chemistry. However, this manuscript is very descriptive and sometimes redundant. As authors pointed, the concentration levels of organic acids are surprisingly low compared with other polluted areas (lines 22-23, 704-706) although authors did not provide a reasonable explanation. My major concern is that the concentrations of organic acids and MSA could be seriously underestimated due to biodegradation because there is no description in

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the text to avoid possible decomposition of organic acids and other chemical species during sample storage and analytical procedure. Another concern is that, although authors mentioned “Little is reported in terms of the size-resolved nature of organic acids and MSA ..” (line 92-93), there are several studies that reported size distributions of organic acids (see below for details). The above points should be clarified in the revision before the consideration for the decision on a possible acceptance to ACP. More specific comments are followed:

1. Lines 31-33. Oxalate was approximately an order of magnitude more abundant. . . Which size fraction are you talking about? Total (<0.056 to >18 μm)?
2. Line 51 and others. Some of the references are not properly cited. For example, in line 51, authors cited Kondo et al. (2011) in the discussion of organic acids. However, Kondo et al. (2011) focused on black carbon but not for organic acids. Please check the possible mistakes in referring previous citations.
3. Lines 92-93. There are several studies on the size-segregated dicarboxylic acids from different regions in the world. For example:

Mochida, M., N. Umemoto, K. Kawamura, H. Lim, and B. J. Turpin (2007), Bimodal size distributions of various organic acids and fatty acids in the marine atmosphere: Influence of anthropogenic aerosols, Asian dusts, and sea spray off the coast of East Asia, *J. Geophys. Res.*, 112, D15209, doi:10.1029/2006JD007773.

Kawamura K., M. Narukawa, S.-M. Li and L. A. Barrie, Size distributions of dicarboxylic acids and inorganic ions in atmospheric aerosols collected during polar sunrise in the Canadian High Arctic. *J. Geophys. Res.*, 112, D10307, doi:10.1029/2006JD008244, 2007.

Gehui Wang, Kimitaka Kawamura, Mingjie Xie, Shuyuan Hu, Junji Cao, Zhisheng An, John G. Waston and Judith C. Chow, Organic Molecular Compositions and Size Distributions of Chinese Summer and Autumn Aerosols from Nanjing: Characteristic Haze

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Event Caused by Wheat Straw Burning, *Environ. Sci. Technol.*, 43 (17), 6493–6499, 2009.

Gehui Wang, Kimitaka Kawamura, Mingjie Xie, Shuyuan Hu, and Zifa Wang, Water-soluble organic compounds in PM_{2.5} and size-segregated aerosols over Mt. Tai in North China Plain, *J. Geophys. Res.*, 114, D19208, doi:10.1029/2008JD011390, 2009.

Smita Agarwal, Shankar Gopala Aggarwal, Kazuhiro Okuzawa, and Kimitaka Kawamura, 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, *Atmos. Chem. Phys.*, 10, 5839-5858, 2010.

Gehui Wang, Kimitaka Kawamura, Mingjie Xie, Shuyuan Hu, Jianjun Li, Bianhong Zhou, Junji Cao, Zhisheng An, Selected water-soluble organic compounds found in sized-resolved aerosols collected from urban, mountain, and marine atmospheres over East Asia, *Tellus*, 63B, 371-381, 2011.

Miyazaki, Y., K. Kawamura, and M. Sawano (2010), Size distributions and chemical characterization of water-soluble organic aerosols over the western North Pacific in summer, *J. Geophys. Res.*, 115, D23210, doi:10.1029/2010JD014439.

Dhananjay Kumar Deshmukh, Kimitaka Kawamura and Manas Kanti Deb, Dicarboxylic acids, ω -oxocarboxylic acids, α -dicarbonyls, WSOC, OC, EC and inorganic ions in wintertime size-segregated aerosols from central India: Sources and formation processes, *Chemosphere*, 161, 27-42, 2016.

Those references should be cited.

4. Method section. There is no description on the sample storage from sampling to chemical analysis. Did you store the samples at room temperature or at -20°C in a freezer? After the water extraction of the sample, how long did you store the water extracts at room temperature? Storage of filter samples and water extracts at

room temperature may be subject to biodegradation of organic acids. In Manila, high humidity conditions may provide more moistures to the aerosols. Please provide the information of storage of samples and extracts.

5. Results section. Some paragraphs are too long and redundant (e.g., 31 lines in the paragraph starting at line 253). By reorganizing the paragraphs and rephrasing the sentences, authors could improve them to become more readable.

6. Lines 340-344. Authors mentioned lower concentrations of oxalate and MSA than expected for a megacity Manila. This point should be critically verified including the discussion of methods used; potential biodegradation of organic acids and MSA during sample storage and analytical protocol.

7. Discussion. What is the reason to start the discussion with phthalate that is not the major organic acid? Why do you discuss oxalate at the end that is the most abundant organic acid? I wondered if you could improve the discussion section by reorganizing the order of the compounds.

8. Lines 514-515. How do you explain the crustal sources (35.9%) for adipate? In other places in the text, authors discuss the combustion sources of adipic acid (lines 261-262).

9. Lines 621-622 and 704-706. Again how do you explain the “surprisingly low concentrations of oxalate” ? This reviewer is skeptical about data quality; i.e., a potential loss of organic acids and MSA due to biodegradation during the sample storage and analytical procedure used in this study.

10. Lines 695-698. Is it possible to discuss the fraction of organic acids and MSA on a carbon basis? Did you measure TOC or WSOC?

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