Atmos. Chem. Phys. Discuss., 10, C7560–C7563, 2010
www.atmos-chem-phys-discuss.net/10/C7560/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size-resolved particulate water-soluble organic compounds in the urban, mountain and marine atmosphere" *by* G. Wang et al.

Anonymous Referee #2

Received and published: 15 September 2010

The manuscript is based on a total of 14 sets of size-segregated samples, 6 sets collected in an urban location in China, 3 sets on the mountaintop of Mt. Tai, and 5 sets in a remote marine location. The samples at different locations were collected on different days and in different seasons. The work focuses on the size distribution characteristics and relative abundance of sugar compounds (include levoglucosan) and dicarboxylic acids. While levoglucosan and the sugar compounds are mainly derived from primary emission sources, the dicarboxylic compounds are of mainly secondary origin. The site contrast offers an opportunity to compare size distribution characteristics of the same class compounds in different atmospheric environments and to explore source information of the measured WSOC species. However, wording of a few major findings

presented in the abstract and in the summary/conclusion sections could be misleading. A more serious problem is lack of rigor in data interpretation. Specific comments are given below.

Specific comments

1. The description of size distribution characteristics for the secondary WSOCs is not accurate. The authors state that "...all the secondary WSOCs, except for benzoic acid and azelaic acids, showed a unimodal size distribution with a peak at 0.7-1.1 μm" (Lines 15-16 in abstract). The size distribution of succinic acid in Figure 5 clearly shows a dominant coarse mode in the marine samples and a minor but nevertheless noticeable coarse mode in both the urban and the mountaintop samples.

2. Related to the previous comment, there are more examples in which the authors describe the size distributions of a species to be unimodal while data in Table 2 show significant presences of the species in both fine and coarse mode. For example, the authors reported that phthalic acid showed a unimodal size distribution in samples at all three sites (page 17475). However, the phathalic data in fine and coarse modes (using 2.1 μm as the cut point) show that there were comparable abundances in the fine and coarse modes (e.g., 102 ng/m3 in the fine mode vs. 47 ng/m3 in the coarse mode in the urban winter samples).

3. The authors reported a larger geometric mean diameter (GMD) of the WSOCs in the fine mode in the winter samples than in the spring samples at the urban site. They attributed this to an enhanced coagulation effect under the development of an inversion layer (Lines 16-19, abstract). However, they did not present an analysis to support this speculation. While it is possible that coagulation plays a role, it is also possible that this might be caused by more condensation of secondarily-formed species due to more abundant precursors in the winter. A more quantitative analysis is needed to ascertain the relative importance of condensation vs. coagulation before such a speculation can be included in the abstract or the summary section.

4. In the abstract and summary sections, the authors state that “levoglucosan is the most abundant WSOCs in the urban and mountain atmosphere”. This statement is misleading. Oxalic acid/oxalate is typically more abundant than levoglucosan. Oxalic acid is not among list of the analytes that can be reliably quantified using the method employed here by the authors (presumably due to low recovery as a result of the TMS-derivative of oxalic acid being relatively too volatile). A more accurate description is: “levoglucosan is the most abundant compound among the quantified WSOCs in the urban and mountain atmosphere”. Similarly, the statement in the summary section “. . . WSOCs in the marine air are dominated by malic and succinic acids” is misleading for the same reason. The range of dicarboxylic acids and aromatic acids quantified by the analytical method should be clearly described in the sample analysis section. I note this information was given in section 3.2. It is more suitable to place it in the experimental section, where readers expect to find such information.

5. Page 17474, line 1: It should be “Among the measured secondary WSOCs in the urban and mountain top air, . . .”. Line 7: it should be “However, the composition of the measured secondary WSOCs is different. . .”

6. p17476, lines 8-9: The authors state “. . .azelaic acid is much more abundant on coarse mode in the marine samples compared to the urban and mountain samples (Fig. 5d-f)”. While the separation between the fine and coarse modes in the urban spring samples (Fig. 5d) was less distinct than that in the marine samples (Fig. 5f), the abundance of the coarse mode relative to the fine mode in the urban spring samples (15 vs. 26 ng/m3) was similar to that in the marine sample (0.5 vs. 0.9 ng/m3) (Table 2). Therefore, the authors’ statement is not correct.

7. Work from Schauer and coworkers indicate that pathalic acid was likely a secondary product of vehicular emissions. It will be useful if the authors show the size distributions, along with that of benzoic acid in Figure 6.

8. Glucose has a more prominent fine mode than its coarse mode in the urban winter

samples (Figure 2). Other sugars also have significant presence in the fine mode in the urban winter samples (Table 2). The authors attribute the fine-mode glucose to the hydrolysis of levoglucosan. The literature cited by the authors (Seinfeld and Pandis, 1998; Helle et al., 2007) could not support the authors’ argument as the conditions in the literature studies do not reflect typical tropospheric ambient conditions. In addition, such a hydrolysis mechanism could not explain the significant presence of fine-mode sugar alcohols (e.g., arabitol, mannitol, inositol, and glycerol).

9. Page 17472, line 7: levoglucosan was in the order of a few hundreds of ng/m3 in the urban environment and in the order of a few ng/m3. Therefore, the concentration of levoglucosan in the urban atmosphere was two orders of magnitude higher than that in the marine atmosphere, not “three orders of magnitude”, as the authors have stated.

10. Table 3: The GMD for coarse-mode fructose and sucrose in the marine samples are larger than 14 μm (14.2 and 14.7 μm, respectively). How is this possible, considering the highest cut size provided by their sampler is 11.3 μm?

11. The authors attribute the larger GMAs of levoglucosan and the secondary WSOCs in the coarse mode in the marine air to enhanced hygroscopic growth of the marine particles related to higher humidity in the marine atmosphere. Again, there no analysis/evidence presented to

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 17467, 2010.