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Interactive comment on “Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols via analysis of organosulfates and related oxidation products” by Q. T. Nguyen et al.

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Responses to reviewers' comments “Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols via analysis of organosulfates and related oxidation products” by Q. T. Nguyen et al.

We kindly thank the reviewers for their thorough reviews and suggestions, and also for the time that they have spent reviewing our paper. We feel that we have been able to improve the paper a great deal trying to address your concerns and questions. Please

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find our responses to the comments below. Quynh T. Nguyen, on behalf of all authors.

Anonymous Referee #2 Received and published: 29 March 2014

This paper reports on a field study conducted at two sampling sites in Denmark with an overall goal to characterize and quantify organo-sulfate (OS) constituents of atmospheric organic aerosols in specific geographic area of the study. Samples of the field collected particulate matter were systematically analyzed using HPLC-ESI-TOFMS and ion chromatography. A number of OS compound were detected and quantified in the samples. Variability of OS concentrations between two sites, during day and night periods, and as an overall function of time was examined. Correlations between OS species, complementary real time measurements, and meteorology records were analyzed using statistical methods. The authors present and discuss their analysis results in a context of the plausibility of regional versus local sources of VOC and atmospheric chemistry leading to formation of OS. The paper is a logical continuation of authors' previous work and reports an unpublished data set from a new geographic location. Overall, the OS measurements are of good quality and relevant to the scope of the ACP journal. However, I think the paper would benefit from significant revisions and shortening before its final publication in ACP.

My major reservation is that the manuscript in its present form is very descriptive of all the aspects of analytical chemistry and details of statistical analysis, but its scientific discussion and data interpretation is fairly ambiguous.

1) I find it misleading that Figs 4, 5 and associated discussion present meteorology and emissions data on the scale of a few thousand kilometers while the field measurements were limited to two field sites 30 km apart. Furthermore, a single pixel size of Fig 5 is about half of the entire Denmark. Drawing conclusions on the 'regional impact' from this type of data is not very convincing and need to be revised. For instance, Fig 4 shows lower VOC emissions in Denmark, but no clear arguments are presented that would rule out impact of the local VOC sources with lower emission rates.

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Reply: We have combined Figure 4 and 5 into the current Figure 4, with improved resolution and same spatial coverage to facilitate the discussion in order to meet the comments. Specific discussions of local and regional impacts were included in the text, as further discussed below.

As can be seen in the original figures, local VOC emissions were very similar on the 30 May (westerly air mass) and 31 May (southeasterly air mass) and 1 June (westerly air mass), while 31 May showed very high concentrations. We consider it sensible to think that VOC emissions do not directly govern the overall high concentrations of the species.

2) Figs 3 and 6 show time resolved records of total concentrations of organic acids, OS, NOS, and of selected individual OS and NOS species that show close correlation between measurements at two sites. Total PM mass measurements at two sites are not presented, but I suspect they might be correlated too. Then, the overall conclusion that both sites were engulfed by the same air mass is logical, but I see no reason to believe that OS compounds were necessarily transported from a long distance. Again, why local sources are ruled out?

Reply: We have tried to put in some additional arguments to elucidate our argument.

In the beginning of section 3.4 (Regional impacts), we added: ... It is apparent that all three classes of compounds showed similar temporal patterns, and the two sites do not differ substantially, which were attributed to the large contribution of compounds belonging to the correlating group affecting both sites. The observations strongly indicated that the major sources or chemistry governing the total concentration levels of the compounds must occur at a spatial location affecting both the urban curbside and semi-rural background sites, which represent quite different environments (Figure 1).

In the end of section 3.4 (Regional impacts), we added: ... It is difficult to determine the exact spatial location of the major sources or chemistry governing the total concentrations of the compounds at the two sites, which is partly due to the coarse resolution

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of the SO₂ emission map. However, it is unlikely that the major source location would be in the local proximity of the two sites due to the highly different local conditions between the urban curbside site HCAB and the semi-rural background site Risø (Figure 1). Such a local source, if present, would be subject to immediate different local background conditions and also varying local wind conditions, while in contrast, a highly similar temporal variation pattern of total observed concentrations was found at the sites during the whole campaign period (Figure 3). Furthermore, any possible point source located southeasterly from HCAB and Risø, which could affect both sampling sites would unlikely be in Denmark, as both sites are located only 10 - 20 km from Baltic sea in the southeasterly direction (Figure 1). The source region therefore is possibly located at the broader regional scale across the Baltic sea extending to the southerly neighboring countries.

We also argue that regional scale could be understood from 10 to hundreds of km, which really should be the case with our observation. We have inserted here Figure 1.4 from Seinfeld and Pandis (2006) to illustrate our point on extension of the “regional scale”.

3) Clarity of the manuscript can be improved by substantial shortening of the descriptive text and by placing some excessive Figures and Tables into supplemental file. For instance, Figs. 4, 5, can be either simplified to show smaller areas or moved to the SI file. Table 5, Figs 7 and 8 can be moved to the SI file.

Reply: Thank you for your suggestions, we have made the following changes: Figure 4 and 5 (ACPD version) have been combined to the current Figure 4 (revised version), which is simplified and shows “zoomed-in” area. Table 5 has now been moved to the Supplementary Information (it is now Supplementary Table 4). We have also moved Figure 7 and 8 to the SI section, while extra information has been added to Figure 3 and 5. We have also shortened some unclear sentences and redundant descriptive text, as can be seen in our responses to specific comments from Reviewer 1 and Reviewer 3. In addition, we have also added more depth to the discussions in general.

4) Perhaps a bar chart plot would better present data of Table 4.

Reply: We have tried to add a vertical line to Table 4 to separate the 2 sites according to suggestion from Reviewer 1. Adding a bar chart plot would not be difficult; however it would probably be too much to add to a manuscript already full with complex plots. In addition, we also find it beneficial to keep the current exact number concentrations format, so that other researchers could have the exact values to compare to.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 2449, 2014.

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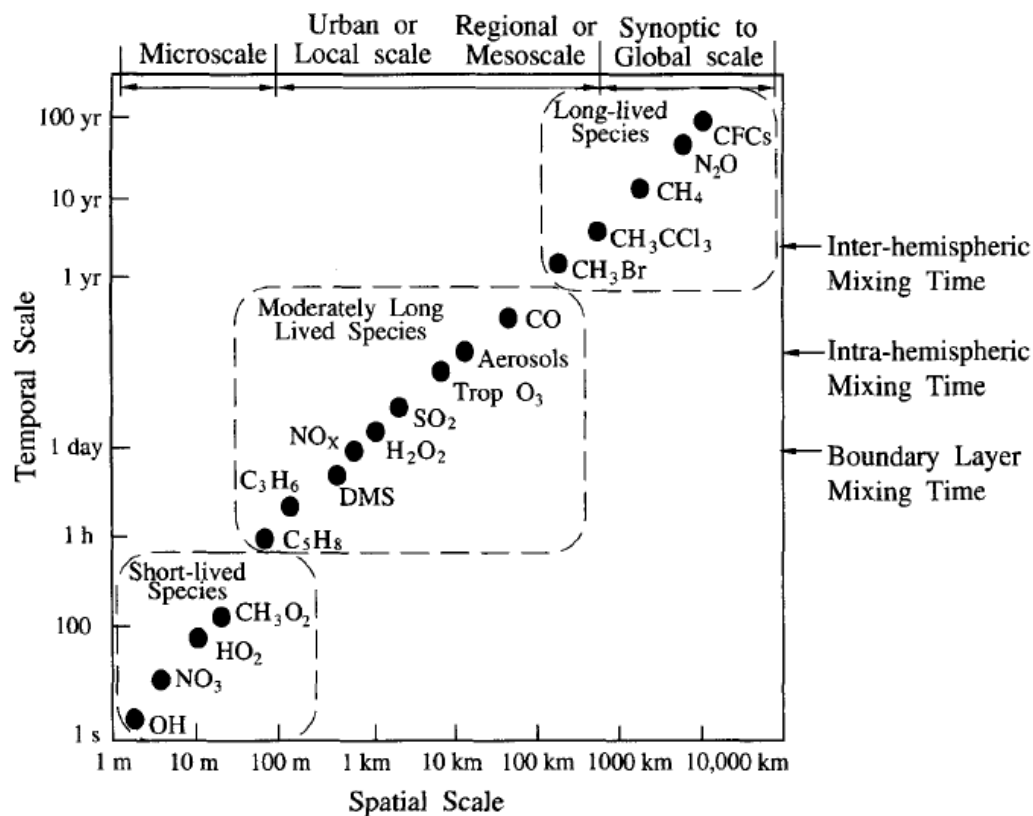


FIGURE 1.4 Spatial and temporal scales of variability for atmospheric constituents.

Fig. 1.

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