

Interactive comment on “On the abundance and source contributions of dicarboxylic acids in size-resolved aerosol particles at continental sites in Central Europe” by D. van Pinxteren et al.

Anonymous Referee #3

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General comments

This paper reports a data set of size-resolved dicarboxylic acids (DCAs) obtained at several continental sites in Germany. Based on these datasets together with back-trajectory analysis and principal component analysis, the authors suggest that low-molecular DCAs formed via gas-phase reactions can be important as well as those formed via aqueous-phase reactions. They also suggested that the formation of DCAs is significant in anthropogenically-influenced air masses under high photochemical conditions. The present work may provide valuable data sets in our understanding on formation processes of DCAs, which is an important component of water-soluble

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organic aerosols. The manuscript likely fits with the scientific scope of ACP. However, I have many concerns on the manuscript. Overall, the manuscript lacks quantitative discussion (see comments below), which makes discussion rather weak throughout the manuscript. Although a large data set presented is valuable, there are a number of important issues that need to be worked out. I recommend its publication in ACP after some major revisions.

Specific comments

(1) The authors suggest that formation of DCAs in the gas phase occurs on “short” timescales, whereas the formation in the aqueous phase occurs on “longer” timescales. However, the terms “short” or “longer” are rather qualitative. The authors should discuss the timescales for each process.

(2) The analysis presented in this manuscript much relies on trajectories. According to “mean trajectory length” in Fig.2, the horizontal scale which the authors discuss seems to be a few thousand km. How about the influence of local (or urban-scale) emissions on source apportionment especially at urban sites? What is horizontal resolution of meteorological data used in the trajectory calculation? Is the resolution high enough to resolve the effect of local-scale emissions of DCA precursors?

(3) Based on size-resolved DCA concentrations and its correlations with concentrations of sulfate and other tracers, the authors defined PC2 as anthropogenically-influenced DCAs possibly formed via aqueous-phase reactions. What are the RH values along the trajectories and at the sampling site to support aqueous-phase reactions? The authors mentioned RH for PC1 which is suggested to represent gas-phase formation of DCAs, but not for PC2. The discussion should be more systematic.

(4) The authors have combined all the data for the PCA calculation. However, relative importance of sources and formation pathways may be different in different seasons, at different sites. Is the authors’ major conclusion (importance of gas-phase formation of DCAs) representative for any seasons at any sites in central Europe?

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(5) If the gas-phase formation in 0.05-0.14 μm is really important for the abundance of DCAs (Table 2), then, why the size distributions show that DCAs showed the peak in accumulation mode (0.14-1.2 μm) (Fig. 1)? Doesn't this mean that the aqueous-phase formation of DCAs is the most important to control the abundance of DCAs?

(6) If the photochemistry and anthropogenic sources are important factors to determine the amount of gas-phase DCAs, the authors should show concentrations of gas species (O_3 , NO_x , CO , SO_2) in the text and Table 1. In addition, I suggest the authors to use some indicators of photochemical aging (e.g., the observed ratios of oxalate/DCAs, sulfate/(sulfate+ SO_2), etc.) and discuss these indicators in comparison with the trajectory analysis to show their consistency.

(7) P. 32098, L. 22-28: The authors should address the reproducibility of this analytical method for individual DCAs. The authors should also show more QA/QC data to provide more information on their method for DCAs analysis. P. 32099, L. 7-10: Please add more information on the gas measurement, such as instruments/methods used, measurement uncertainty, etc.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 32093, 2013.

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