

Interactive comment on “Thermodynamic properties and cloud droplet activation of a series of oxo-acids” by M. Frosch et al.

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We thank the Referee for the useful remarks which induced us to make the changes listed below:

Comment: 1) Section 1: For a publication in ACP the atmospheric relevance of the investigated oxo-acids could be elaborated in more detail, or at least that of the parent carboxylic acids. Are there any numbers given in the literature related to their atmospheric abundance in the particulate and the gas phase phase? What are potential sources of the investigated compounds?

Response: We have added elaboration on the sources and atmospheric abundances (page 3757, line 17 to page 3758, line 2):

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Old version: “4-oxopimelic acid has been detected in particles in the polar atmosphere (Kawamura et al., 1995, 1996), in marine environments (Sakaguchi and Kawamura, 1994; Kawamura and Sakaguchi, 1999; Wang et al., 2006), and suburban locations (Yokouchi and Ambe, 1986). 2-oxoglutaric acid and 4-oxopimelic acid have structural features similar to other compounds frequently detected in secondary organic aerosols and are thus considered as standards for organic dicarboxylic acids (Gao et al., 2004). Oxosuccinic acid has been studied previously and is also considered a standard for dicarboxylic acids (Rissman et al., 2007). It has been demonstrated that oxosuccinic acid particles can act as cloud condensation nuclei (Rissman et al., 2007), but so far the effects of enolization and decarboxylation on cloud droplet formation have not been considered. Such effects will be investigated in this work. Neither oxosuccinic acid nor 3-oxoglutaric acid have been detected in atmospheric aerosols. However, both compounds can react or decompose in the aqueous phase under ambient conditions to products that have been detected, e.g. pyruvic acid (Saxena and Hildemann, 1996).”

New version: “4-oxopimelic acid has been detected in particles in the polar atmosphere in concentrations up to 0.53 ng/m³ (Kawamura et al., 1995, 1996), in marine environments in concentrations up to 2.5 ng/m³ (Sakaguchi and Kawamura, 1994; Kawamura and Sakaguchi, 1999; Wang et al., 2006), and in trace amounts at suburban locations (Yokouchi and Ambe, 1986). 2 oxoglutaric acid and 4-oxopimelic acid have structural features similar to other compounds frequently detected in secondary organic aerosols and are thus considered as standards for organic dicarboxylic acids (Gao et al., 2004). Oxosuccinic acid has been studied previously and is also considered a surrogate standard for dicarboxylic acids (Rissman et al., 2007). Rissman et al. (2007) demonstrated that oxosuccinic acid particles can act as cloud condensation nuclei, but did not consider the effects of enolization and decarboxylation on cloud droplet formation. Such effects will be investigated in this work. Neither oxosuccinic acid nor 3-oxoglutaric acid have been detected in atmospheric aerosols. However, both compounds can react or decompose in the aqueous phase under ambient conditions to products that have been detected, e.g. pyruvic acid (Saxena and Hildemann, 1996).”

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Furthermore, we add the following text (page 3758, line 2): "The main source of oxo-acids in the atmosphere is photochemical reactions from the parent dicarboxylic acid or from monovalent oxo-acids (Sakaguchi and Kawamura, 1994; Kawamura et al., 1996; Wang et al., 2006). Several sources have been suggested for dicarboxylic acid, mainly anthropogenic, but also biogenic sources and secondary production can be relevant (e.g. Kawamura and Kaplan, 1987, Rogge et al., 1993; Kawamura et al., 1995)."

Comment: 2) Section 4.1: Where does the relatively large range of 1-5 seconds for the calculated evaporation time inside the DMA and/or the tubing comes from? Different tubing length for different experiments or a variability in the flow rates? Please explain.

Response: The components of evaporation time have been specified in the text as suggested. The following changes have been made (Page 3769, lines 20-22): Old version: "Times (t_{evap}) were in the range 1–5 s. The corrected dry particle diameters were determined using the vapor pressure of glutaric acid reported by Bilde et al. (2003)."

New version. " t_{tubing} was estimated to be approximately 0.3 s. In some experiments, an additional 2.7 meters of tubing was inserted between the DMA and the CCNC to check for potential evaporation, increasing t_{tubing} to 3 s. Since the aerosol flow rate was not changed significantly between different experiments, the dependence on the flow rate was neglected for t_{tubing} . t_{DMA} was estimated to be in the range 1.0-1.2 s, depending on aerosol and sheath flows in the DMA. Times (t_{evap}) were of the order of 1 s in experiments without the additional tubing and of the order of 4 s in experiments with additional tubing."

Comment: 3) Section 4.1: In the paper is mentioned that it has been suggested that glutaric acid particles might be unstable and that fissioning or restructuring to a smaller size can happen between the DMA and the CCNC (Kumar et al., 2003). When taking the calculated evaporation times inside the DMA and the tubing into consideration it puts your results in agreement with Köhler theory, and, therefore, it is concluded that glutaric acid activated according to Köhler theory. However, do your findings rule out

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the above mentioned possibility of a restructuring of the particles being responsible for a smaller size instead of assuming evaporation before entering the CCNC? Please clarify.

Response: When evaporation is taken quantitatively into account (which, to our knowledge, has not been done in any previous study), the CCN activity of glutaric acid agrees well with Köhler theory. This, of course, does not rule out that glutaric acid particles due to instability may fission or restructure to a smaller size. However, the very good agreement between Köhler theory and corrected CCN measurements indicates that if such processes occur, they are not significant enough to influence the CCN activity. Additional text, clarifying the point, has been added to the last paragraph of Section 4.1, page 3769, line 28: "No other effect is necessary to explain the initially observed deviations."

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