

Interactive comment on “Atmospheric chemistry of carboxylic acids: microbial implication versus photochemistry” by M. Väitingom et al.

B. Ervens (Editor)

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Thank you for submitting your manuscript ‘Atmospheric chemistry of carboxylic acids: microbial implication versus photochemistry’ to Atmospheric Chemistry Discussions

After the discussion stage has been closed, I would like to give you my summary of the major concerns raised by the reviewers to be considered for further proceeding of your manuscript : Prior to submission of a revised manuscript for possible publication, please decide whether the relevance, and accuracy of your results warrant publication in ACP. If so, of course, please, go through all individual reviewer comments and address them point-by-point as well.

1) Atmospheric relevance

Reviewer 4 doubts the atmospheric relevance of your study since the lifetime of cloud droplets is on the order of minutes, and not hours as assumed in your experiments. While cloud droplet residuals (hygroscopic aerosols), certainly still contain some water, the aqueous phase on such particles is definitely very different in terms of ionic strength, pH etc as in relatively dilute cloud droplets.

While your results are definitely interesting in terms of kinetics, product and loss studies, I agree with the reviewer that they might not be atmospherically relevant (and thus warrant publication in ACP), unless you can show their validity under more realistic atmospheric conditions,

- Are the results independent of time? I.e. do you derive the same rates (rate constants) also for shorter time scales (minutes)?
- how realistic is the oxidant-free environment in your experiments solutions for microorganisms in real cloud droplets? Are microorganisms impacted by the presence of OH, H₂O₂, pH etc?
- what is the effect of additional products, such as antioxidants (e.g., catalase – cf Reviewer 1) for feedbacks on photodegradation rates?
- are the microorganism concentrations and number distributions similar to those in clouds?

2) Context of the role of carboxylic acids

All reviewers agree that the abstract and introduction contains vague if not even wrong statements about the atmospheric abundance and chemical (production and loss) pathways. The fraction of carboxylic acids to total organic aerosol mass usually does not exceed a few %. Also in cloud water, their mass contribution to the solute mass is i) strongly dependent on location (ii) usually no more than a few%. Note that common analytical techniques cannot resolve the total organic mass but only a small fraction of it.

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3) Methodology, and derivation of results

a) since readers of ACP are not necessarily familiar with microorganisms, you need to add more information on these biological systems and the analytical methods applied.

b) Even though you have performed similar experiments in a previous study, it is desirable to give more detail on the methodology of the experiments. In order to be more confident of your experimental method, results of quality assurance experiments should be presented

c) how applicable are Equations 1-3 to derive meaningful results?

- do the rates change over time? These could be illustrated by a figure.

- assuming an OH concentration from model studies in the literature definitely introduces a huge uncertainty in the interpretation of results. [OH] should be calculated based on the light intensity and H₂O₂ concentration. In addition, it is likely that OH reacts over the course of the experiment. Instead of assuming a constant concentration, I suggest to set up a box model to simulate the experimental conditions.

- It would be more useful to present data in terms of rate constants (k) that are independent of time and [OH] and not as reaction rates in order to facilitate a more general comparison.

4) Structure of manuscript

I agree with the reviewers that several sections of the paper are hard to read:

The abstract should only state your main findings: Under what conditions is microbial degradation of carboxylic more, as important as, or less important than photochemistry? Details on methodology do not need to appear there.

Make sure that 'Methods' and 'Discussion' are clearly separated and only refer to the results of the experiments presented here. E.g., information on T dependence of photo- vs microbial degradation are not a direct conclusion from the present study.

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If relevant, you can add an extra section 'Atmospheric implications' that may include a discussion of your results in the context of atmospheric conditions (air mass types (anthrop., marine), OH scenarios etc.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 4881, 2011.

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