

This review article written by Tilgner et al. nicely summarizes the fundamentals and recent literature on how acidity affects chemical partitioning and reactions occurring in the atmospheric aqueous phases. The topic is important and timely. The review covers all the important aspects of this topic, and the manuscript is extremely carefully written. I strongly recommend publication in ACP. I only have a number of minor comments listed below.

Minor comments

- A class of aqueous-phase reactions that are strongly pH-dependent is hydrolysis. Hydrolysis can be a major fate of many environmental pollutants in aquatic chemistry. While the manuscript contains a few discussions related to hydrolysis (e.g., sections 5.2.3 and 5.3), I suggest the authors consider having a section to systematically discuss hydrolysis. E.g., what functional groups can undergo hydrolysis and how; pH-dependence; the importance of hydrolysis in the atmosphere compared to other reactions already described in the manuscript.
- Figure 2-f. I suggest the authors added pKa2 for phthalic acid. I wonder why the aqueous fraction (XAaq) does not immediately increase at the pKa1 (~3) of phthalic acid? This trend seems to be different from the other monoacids.
- Figure 7a) and b) - the kinks on the total S(IV) oxidation rates at pH 5 appear very unnatural, and there are no explanations provided. My understanding is that the figure is based on the recommended S(IV) oxidation by Fe(III), as indicated in equation 11. I recommend that the authors explain why the Fe(III) oxidation drastically loses importance at pH > 5.
- Line 1207 "The deprotonation likely leads to a reduction in the electron density at the carbon-carbon double bond enabling an easier O₃ addition, i.e. a more rapid oxidation. From inductive effect theory, it is known that the COOH group is electron-withdrawing and COO⁻ is electron-donating. Thus, the obtained behavior is feasible." - This couple of statements seem to be conflicting with each other. Please clarify.
- Line 1246 "Please note that the overall second order reaction rate constants consider the dissociation speciation of the carboxylic acids but not their effective solubility. Thus, the overall chemical reaction rate will depend on both the aqueous oxidant concentration and on the total aqueous compounds concentration. The latter largely depends on the microphysical conditions present." - I agree with the author's statements. Can they comment on how important O₃ and NO₃ oxidation reactions are, considering their relatively small Henry's law constant? Or may their Henry's law constants are not so small?
-

Technical comment

- Figure 4. Can the authors consider adding pKa values of each compound, in a similar manner as in Figure 2?
- Figure 5 caption. The authors defined the red text and green text in the figure, leaving the blue ones unexplained. Maybe add that blue text describes physical changes of droplets?
- Line 358 - "sulfate oxidation" should be "S(IV) oxidation"

- Line 572 - "newly kinetically analyzed for the present review" sounds awkward. Please rephrase.
- Line 599 - Lee and Schwartz 1983 the citation format is inconsistent.
- Line 796 - extra "." at the end
- Line 802 - "substituted organic acid" - should it be "substituted aldehyde"?
- Line 820 - "hydrolysis" should be "hydration"
- Figure 12 caption - This caption uses "Fig. XX", while others all use "Figure XX"
- Line 886 - the phrase "which was discussed in more detail for acetaldehyde, glyoxal, as well as methylglyoxal" is repeated.
- Line 908 - "On the one hand"
- Line 917 - Li et al. 2019 - citation format is inconsistent.
- Line 1065 - "Overall, the different contribution of ETR and H-abstraction pathways modify also the product distribution as a function of pH." Does the author have any references for this statement?
- Line 1138 - "Therefore, the statements of Zhao et al. (2016) regarding the pH dependence in the reactivity of saturated carboxylic acids are by far too overgeneralized)" - the same authors have a recent paper showing that larger organic acids indeed do not exhibit much pH dependence. Please cite.
<https://doi-org.login.ezproxy.library.ualberta.ca/10.1021/acs.est.0c03331>
- Line 1190 - "due to of the"
- Line 1249 - "depend" should be "depends"
- Line 1386 - Just wanted to double check if the authors have defined "OM".