

The study by Chen et al. explored the influence of nitrate photolysis on sulfate formation from aqueous-phase bisulfite oxidation based on chamber experiments. The role of pH, halogen chemistry and O₂ are investigated. This is an interesting topic, and the experiment looks carefully conducted and calibrated. However, the results are poorly organized and reported, with ambiguous data interpretations and conclusions. I don't think the current manuscript meet the standard of ACP, unless it was largely rewritten. Moreover, the authors should treat publications more seriously. Typos are common, but it was very rare to be present at the first line of the abstract—where I believe the “exit” should be “exist”. I wonder whether the authors have carefully read the manuscript even once. If even the authors don't want to read the manuscript, neither do the readers.

Major concerns:

1. The study titled “Aqueous phase oxidation of bisulfite influenced by nitrate photolysis”, and one would expect the study to focus on reactions under light. In this context, the dark condition is expected to serve as a background. However, this study spends quite a lot of efforts emphasizing the importance of nitrate and its synergism with halogen chemistry in sulfate formations even under dark condition. Moreover, the dark condition is apparently not set as the background scenario, as few comparisons are conducted between dark and with light conditions when the other factors are the same. Either the title or the organization and analysis of this study should be modified.
2. Line 47-49 and 233-239: The review of aerosol pH is biased. The same research group of Guo et al. (2017) have reported much higher pH levels (even higher than 7) in Beijing in some later studies that they participated (e.g., Shi et al., 2017). In addition, some recent studies have further revealed the driving factors of aerosol pH (e.g., Pye et al., 2020; Zheng et al., 2020), which can explain the difference in reported pH levels. These advances should be included.
3. L147-150: The statements are confusing. Which one is insignificant?
4. Line 167-169: What's the point / conclusion of the whole part? Is NO₂ an important oxidant or not, based on your experiment results?
5. Line 176-178: The whole paragraph is repeating existing explanations of potential pathways. But what's the relationship with this study? Do the results support / disagree with any of the pathways? If not, this part should be simplified into one to two sentences. Discussion of existing studies that is not related to your results should not be part of a research article.
6. Line 187-188: if “nitrate itself can greatly promote the oxidation of bisulfite in the solution under dark condition” already, how important is the photolysis of nitrate? And what's their relative importance? More quantitative analysis should be conducted, as seems all required experiments are already conducted.
7. Line 213: the pH ranges that are achieved by addition of (NH₄)₂SO₄, NH₄HSO₄ and their mixtures should be stated here.
8. Line 9-10, 216-219, and 463-465: The behavior of pH in bulk solutions should not be mixed with that of aerosols. While in solutions, the pH can be sensitive to ammonia sulfate concentrations and the relative fractions of ammonia sulfate / ammonia bisulfate added, it was largely controlled by other factors for aerosols (Pye et al., 2020; Zheng et al., 2020).

The results should only be used to infer the dependence of reaction rate on pH, not to be interpreted as the dependence of aerosol pH on ammonia sulfates.

9. Line 223-224: within which pH range?
10. Line 225-227: This possible explanation could be checked with simple calculations. Just scale the formation rates to the pH dependence of bisulfite concentrations.
11. Line 243: What's the new aqueous phase oxidation pathways? And what's the relationship of the sunlight discussions with this section (i.e., the effect of pH on sulfate formation)?
12. Line 246-270: This part seems to argue that cation profile of nitrate is important due to two reasons: (1) NH_4NO_3 are with lower pH than NaNO_3 at the same concentrations, and (2) NH_4^+ can be oxidized by NO_2 to help generate $\cdot\text{OH}$. For the first explanation, as stated above, the influence of chemical compositions on solution pH should not be equaled to that on aerosol pH. Therefore, if the authors want to prove this explanation, the formation rates should be compared at the same pH, not the same concentrations. For the second explanation, not all spontaneous reactions can happen—the reaction rates must also be considered. Are there any experiments / references supporting that these reactions can happen at a reasonable rate under ambient conditions?
13. Line 273-284: What's the point of the long discussions of this part? Proving that 2-propanol can serve as $\cdot\text{OH}$ scavenger?
14. Line 294-297: If " O_2 has little effect on the generation of $\cdot\text{OH}$ by nitrate photolysis", how is it important in sulfate formation with nitrate photolysis? Any explanation of the potential pathways?
15. Line 317-326: Rewrite this paragraph to make the points clearer.
16. Line 336-342: Is the higher H_2O_2 under lower pH totally due to the higher $\cdot\text{OH}$ productions under lower pH, or is it due to that H_2O_2 formation is also favored under low pH even assuming same $\cdot\text{OH}$ concentrations?
17. Line 351: how high is the "higher" light intensity? And how sensitive is the formation rate on light intensities?
18. Line 367-391: which steps are proposed in this study? Judging from the manuscript, it seems like all the reactions are already proposed by others, and the study here is just trying to combine them in different ways. If not, clearly state the new pathways/steps proposed in this study and provide the experimental evidences.
19. Line 480-485: This implication part seems abrupt and over-interpreted.

Minor concerns:

There's a lot of grammar errors in this study. A thorough language editing is suggested. Some examples follow.

1. Line 1, "exit" should be "exist"
2. Line 154: ":" into ".".
3. Line 145-155: Missing reference of this statement.
4. Line 201: "then" should be "thus".
5. Line 219, 295, etc.: "on the one hand" should be "on one hand". In addition, after "on one hand" there should always be an "on the other hand", which is not seen in, e.g., line 295.
6. Line 221: "may because" is wrong in grammar.

References

- Pye, H. O. T., Nenes, A., Alexander, B., Ault, A. P., Barth, M. C., Clegg, S. L., Collett Jr, J. L., Fahey, K. M., Hennigan, C. J., Herrmann, H., Kanakidou, M., Kelly, J. T., Ku, I. T., McNeill, V. F., Riemer, N., Schaefer, T., Shi, G., Tilgner, A., Walker, J. T., Wang, T., Weber, R., Xing, J., Zaveri, R. A., and Zuend, A.: The acidity of atmospheric particles and clouds, *Atmos. Chem. Phys.*, 20, 4809-4888, 10.5194/acp-20-4809-2020, 2020.
- Shi, G., Xu, J., Peng, X., Xiao, Z., Chen, K., Tian, Y., Guan, X., Feng, Y., Yu, H., Nenes, A., and Russell, A. G.: pH of Aerosols in a Polluted Atmosphere: Source Contributions to Highly Acidic Aerosol, *Environmental Science & Technology*, 51, 4289-4296, 10.1021/acs.est.6b05736, 2017.
- Zheng, G., Su, H., Wang, S., Andreae, M. O., Pöschl, U., and Cheng, Y.: Multiphase buffer theory explains contrasts in atmospheric aerosol acidity, *Science*, 369, 1374-1377, 10.1126/science.aba3719, 2020.