

General: I am receiving the revised manuscript without having reviewed it before. I think the paper is generally ok and the authors have addressed the concerns of the reviewers.

The authors tend to see the observed oxygen-dependence as a support for the direct single electron transfer between HSO_3^- and NO_2 which directly leads to SO_3^- and $\text{NO}_2^- / \text{HONO}$. The finding of the oxygen effect is interesting but possible more measured points would have been helpful – that could be included into an outlook. Please also note that the NO_2 and SO_2 mixing ratios applied are gigantic.

Regarding the mechanism of the reaction between S(IV) and N(IV) thermochemistry should be considered. Such treatment shows that the reaction is only exothermic by a very small extend and it should be slow. The complex formation mechanism appears much more feasible to me because of this. Maybe oxygen can promote the decomposition of the adduct by scavenging the formed SO_3^- , if there is not only a unimolecular decomposition of the complex but an equilibrium. Then oxygen would be able to promote scavenging of SO_3^- to SO_5^- thus shifting a decomposition equilibrium on the product side. Maybe this could be discussed and included into the discussion / outlook.

Anyway, whether there is a concerted single electron transfer or whether the reaction runs through a complex, at the end sulfur-oxy radical anions will be produced the role of which is S(IV) oxidation has been studied extensively in the late 1980s and early 1990 both in the US and in Europe. These reactions are included into state-of-the art aqueous chemical mechanism and a chemistry such as suggested here would add a source of SO_3^- . One thought which is important for the whole scope of the paper: Such an additional SO_3^- radical anion source will not lead to a much increased S(VI) production as the sulfur radical chain will not establish but the reactive chain carriers will be scavenged and reduce the effectivity of the potential radical reaction chain. This has been demonstrate in many studies and , to some extent, it is surprising that this is not being discussed adequately in a study motivated to explain particle sulfate formation correlating to NO_2 in the gas phase. Especially in China, the sulfur-oxy radical chemistry will be scavenged by the abundance of organics in particles – this occurs via the reaction of SO_4^- with organics where many kinetic data are available but also by such reactions of SO_5^- where the data basis is more sparse. This overall thought should surely be considered in the study.

Details

- Line 53: The postulation of complexes involving more than one NO_2 unit probably comes from pulse radiolysis experiments with quite high NO_2 concentrations. It should be considered that not only the complex with three NO_2 unit can decompose but also the others, especially the most simple one being formed in R3.
- L 205-208: See above remarks, I would be a bit more cautious here.
- L 209-257: All of this must be put into context with the sensitivity of the sulfur-oxy-radical reaction chain towards organics under real environmental conditions.
- L 286 ff: Conclusions section: I feel this should be modified somewhat in view of the above.