

We thank reviewer #2 for the helpful comments. In the following, we respond (*in italics*) to each point raised in the review

Specific Questions: 1. Page 6, line 180. In photolysis experiments, authors allowed the signal to return to baseline for 60 min. Highlighting that gas phase NO₂ decrease as a function of RH, have authors considered any secondary reactions of HONO or NO₂, i.e. acid hydrolysis of NO₂, during the 60min dark periods of the experiment?

The drop off in detected HONO is quite rapid, with time taken to reach baseline thought to be due to the lifetime of the carrier gas pass through the apparatus rather than significant secondary chemistry. Since the source of HONO/NO₂ is turned off in the light, even if secondary chemistry is happening, the products would quickly decrease in concentration because the primary source has been removed. Secondary chemistry is possible, but with the data we currently have, it would be difficult to say with any certainty.

2. Page 7, line 234. Since the inorganic nitrate does not show a direct link to the source of gas-phase nitrogen species and authors speculate that organo-nitrate may have a role, have authors considered investigating the change in the total nitrogen in grime samples before and after photochemical experiments.

This is an interesting suggestion for further study.

3. Page 8, line 267. The work cited here by Grassian and co-workers have reported decrease in %nitrate loss between %RH 20 and 80, due to increase in re-adsorption of gas phase NO₂ at higher RHs. Did authors notice similar observation(s) in the current study? Can this be one of the dark reaction occurring during the above mentioned 60 min dark period?

We only studied nitrate loss from samples illuminated at an RH of 35%, however, the gas phase products do not suggest that this re-adsorption is as important, as it was in the Grassian study. We do not see the decrease in products at higher relative humidities, only a leveling off. However, such a process cannot be ruled out as possibly contributing to this leveling off. We have included this possibility in the manuscript in an expanded discussion of the product formation above 35%.

4. Page 7, line 215. The water uptake on grime seems to follow more like a condensation isotherm rather than a typical water adsorption isotherm with a monolayer formation. If this is the case, does the multilayers formation of water block surface sites or limit photolysis products after %RH 35?

We do not see a decrease in products, only a leveling off. This suggests that the water is not playing a significant role in hindering the photolysis products. Further study looking at relative humidities beyond 60% could help determine this aspect in the role of water for the chemistry.