

Response to Reviewer 2

We thank the reviewer for their helpful comments.

Romer et al. disentangles the impact of different processes affecting the O₃-T relationship in South Eastern US. The hypothesis and the arguments in the manuscript are well presented and provide robust evidence of the importance of soil-NO_x for continental O₃ production. Discussion of the results and their implications is scientifically sound. The manuscript should be published in ACP. I only have two minor comments that I would like the authors to address.

Minor comments

1. At page 9 lines 3-4 the loss of NO_x due to NO₂ + O₃ reaction is taken into account to extract the increase in NO_x due to soil emissions. I wonder how much of a change would accounting for the NO₂ + NO₃ reaction which has a five order of magnitude higher rate constant. I expect no NO₃ measurements for the CTR SEARCH network but for the SOAS measurements (Ayres et al. 2015) it should be possible.

Ayres et al. 2015 found that concentrations of NO₃ were extremely low during SOAS and that N₂O₅ chemistry was a negligible contributor to NO_x loss (Ayres et al., 2015, Fig. 4). Therefore, the NO₂+O₃ reaction rate is equal to the total nighttime NO_x loss. We have revised the section to explain this reasoning:

"To account for the chemical removal of NO_x, the cumulative loss of NO_x during the night was added to the observations. During SOAS, the nighttime loss of NO_x occurred almost exclusively through the reaction of NO₂ with O₃ to form NO₃, which then reacted with a VOC to form an organic nitrate (Ayres et al., 2015). N₂O₅ chemistry made a negligible contribution to total NO_x loss. The loss rate of NO_x during the night was therefore calculated as the rate of reaction of NO₂ with O₃. "

2. The authors are only concerned with soil-NO_x emissions although it is now known that soil bacteria are a comparable source of HONO (Oswald et al. 2013). HONO was measured during SOAS (<https://data.eol.ucar.edu/dataset/373.037>) and its impact on PO₃-T is likely convoluted in the 60% contribution of PHO_x shown in Fig. 6. In the manuscript it is stated that PHO_x is mainly driven by increased solar radiation without showing (or explicitly pointing to) relevant data. However, soil-HONO emissions might also contribute to the PHO_x category in Fig. 6. Could the authors attempt a sensitivity analysis or at least discussion of the soil-HONO impact on the results?

Oswald et al. 2013 found that soil HONO emissions required dry soils, and were enhanced by alkali environments. Neither of these conditions were true during SOAS, and therefore soil HONO emissions are likely negligible at this location. However, when considering ozone-temperature relationships in other locations, the effects of soil HONO emissions should definitely be considered. We have added a discussion of this effect, as well as further explanation of how we concluded that PHO_x was driven by increased solar radiation.

"In very wet environments, soil microbes typically emit N_2O or N_2 instead of NO_x , and in arid environments soil emissions of HONO can be equal to or larger than soil NO_x emissions (Oswald et al., 2013). Although conditions at the CTR site are too wet and acidic for soil HONO emissions to be significant, in environments where soil HONO emissions are large, they would likely have an even greater effect on ozone production by acting as a source of both NO_x and HO_x radicals."

"The increase in PHO_x with temperature is most likely caused by changes in solar radiation, which is well correlated with the total PHO_x rate (Fig. S7a) and increases strongly with temperature. In contrast, water vapor is not correlated with total PHO_x (Fig. S7b). "