

The authors would like to thank the reviewers for their positive and constructive comments on this manuscript. Reviewer comments are in **bold**, followed by the author response and resulting text amendments.

### **Reviewer 2: Comments**

**As the observed NO<sub>x</sub> and VOCs is very high in Delhi (much higher than previously published conditions in urban areas since 1990s), it is very interesting why the observed O<sub>3</sub> concentrations were actually not high in Delhi. I compared the conditions of Delhi with that of Los Angeles, and it is found that the current condition of Delhi is similar to that of 1970s in Los Angeles (cf. Pollack et al., JGR, 2013). Nevertheless, in 1970s, the ozone concentrations were about 400 ppbv in Los Angeles. So, it is actually quite useful for the authors to compare the observed conditions of air masses and the diagnosed ozone production rates and their controlling factors to other urban areas of the world (e.g. US, EU and China).**

*Response:* This is a very interesting point. Additional text has been added with respect to discussion on why ozone concentrations in Delhi are lower than Los Angeles in the 1970s, and more like observations in Beijing, Shanghai and Guangzhou, despite comparable VOC and NO<sub>x</sub> concentrations to Los Angeles.

*Text change:* Average mixing ratios of NO<sub>x</sub> and VOCs observed during the post-monsoon Delhi campaign are comparable to those observed in 1970s Los Angeles. High concentrations of NO<sub>x</sub> (≈ 200 ppbV) and VOCs (e.g. benzene ≈ 10 ppbV; toluene ≈ 30 ppbV) led to large concentrations of O<sub>3</sub> in the US city, averaging at around 400 ppbV (Pollack et al., 2013). Despite the high O<sub>3</sub> concentrations observed in Delhi, O<sub>3</sub> concentrations in post-monsoon Delhi did not exceed 150 ppbV and are of similar magnitude to observed O<sub>3</sub> in Beijing, Shanghai and Guangzhou despite much higher observed VOC and NO<sub>x</sub> concentrations (Tan et al., 2019). The lower O<sub>3</sub> observed in Delhi compared to 1970s Los Angeles can be attributed to differences in both topography and meteorology. The isolated coastal city of Los Angeles is surrounded by mountains to the North and East, with the prevailing wind dominating from the coast. Owing to its basin-like topography, and with a cool on-shore sea breeze often creating a temperature inversion, the air mass circulates within the city and the transport of emissions out of the basin is impeded. Although landlocked Delhi lies to the southwest of the Himalayas, the city is very flat and resides far enough away from the mountain range to allow for the efficient transportation of air masses from the city. It is also important to consider that the very high concentrations of NO<sub>x</sub> and VOCs observed peak to comparable concentrations to Los Angeles during the evening and at night, where they are trapped owing to a shallow, stagnant boundary layer, and there is little to no photochemical activity. O<sub>3</sub> production rates in Delhi peaks in the morning, when concentrations of pollutants, though still high, are much lower than at night due to rapid boundary layer expansion (see section 3.4).

**Followed by comment 1, it would be useful to present the diurnal variations of the diagnosed P(O<sub>3</sub>) in Section 3.4.**

*Response:* The diel profile of ozone production has been added to the supplementary and referenced in the text.

*Text change:* The modelled VOC-NO<sub>x</sub> P(O<sub>3</sub>) isopleth supports the assignment of Delhi being, on average, in a VOC-sensitive photochemical regime (Sillman et al., 1990), with the diel profile of O<sub>3</sub> production peaking at 09:00 (see supplementary, figure S3).

**Section 3.5 and 3.6, the study of the VOCs sensitivities by class may be represented by a well established metrics in the study of ozone photochemistry - relative incremental reactivity (C.A. Cardelino and W.L. Chameides, J. Air & Waste Manage. Assoc. 1995).**

*Response:* Relative incremental reactivity has previously been considered by the authors as a metric to assess the O<sub>3</sub> production potential of VOCs. However, it was decided that changes in in situ O<sub>3</sub> production rate on incremental changes of individual VOCs was a better metric to use in this work, rather than comparing changes in O<sub>3</sub> concentrations as they provide a more meaningful quantification of the impact of changing O<sub>3</sub> production. The discussion of O<sub>3</sub> production, rather than O<sub>3</sub> concentration, is kept consistent throughout the text.

**Section 3.7, the study on the impact from the aerosol uptake and radiative forcing is very useful. Nevertheless, the study needs to be projected with more reality. With respect to the aerosol uptake effect, it can change the HO<sub>2</sub> uptake rates when NO is small which is not the case for Delhi; but it can also change the HONO production rate which might be more important for Delhi in this case (high NO<sub>2</sub> and high ASA). With respect to the change of the photolysis rates, the impact of aerosol could be complicated, the photolysis rates may be reduced in the near surface but enhanced in the higher place in the boundary layer when the aerosol SSA is high. The box model to diagnose the photochemistry processes is normally running with an assumption that the air mass is well mixed for the planetary boundary layer. Thus, the photolysis rates used in the model may be slightly different from the surface observations especially for the high aerosol atmosphere (i.e. Castro et al, AE, 2001). In short, the change of photolysis rates has to be much smaller than the current range and the discussion with that of aerosol shall be improved in this direction.**

*Response:* The text has been changed in line with the reviewers suggestions for clarifying the real impact of aerosol on photolysis rates and ultimately ozone production.

*Text change:* However, the impact of reducing aerosol on photolysis rates is complex. Reductions in aerosol at ground level may lead to either increased or decreased photolysis rates near the surface, depending on the scattering properties of the aerosol. Changes in photolysis rates from increased or

decreased aerosol loading may also vary throughout the depth of the boundary layer. The box model assumes photolysis rates are uniform throughout the boundary layer and that aerosols are well mixed. A more detailed study into the temporal and spatial patterns of aerosol in Delhi and its impact on photolysis rates is required to fully assess the aerosol impact on in situ  $O_3$  production (Castro et al., 2001).

It is important to note that this study focuses on the sensitivity of  $P(O_3)$  to ASA through  $HO_2$  uptake only. Additional chemical consequences and feedbacks of decreasing aerosol, such as changes to HONO concentrations, have not been accounted for here. With the high levels of  $NO_2$  observed in Delhi, the heterogeneous conversion of  $NO_2$  to HONO on particle surfaces may be an important mechanism (Liu et al., 2014; Lee et al., 2016; Tong et al., 2016; Lu et al., 2018b). HONO reductions from decreased ASA may lead to reduced OH radical formation in Delhi, thus the impact of ASA reduction on  $P(O_3)$  may be underestimated in this work.

**The emission of monoterpene from anthropogenic sources is a new point worth to be highlighted. Even a paper Cash et al., 2021, in preparation is mentioned in the text some more description will be valuable also for this paper. The emissions from the process emission sector needs some more explains (e.g. which processes?).**

*Response:* A more detailed description of the potential sources of monoterpenes in Delhi has been added to the discussion of anthropogenic monoterpenes. The full breakdown of what constitutes as “process emissions” in the EDGAR inventory is detailed in the supplementary, and a reference to this has been added to the text. A more detailed analysis of the monoterpene sources in Delhi is beyond the scope of this work, and will be included in Cash et al., in preparation.

*Text change:* However, it is worth noting that the effectiveness of reducing the RT + PE source on modelled  $P(O_3)$  is dependent on the proportion of anthropogenic monoterpene emissions in Delhi. According to the EDGAR emission inventory, 98.1% of anthropogenic monoterpenes in Delhi are attributed to process emissions (PE) (Table 2). These emissions include sources such as emissions from chemical industry, and other industrial processes, and include solvent emissions and emissions from product use (see Table S3 in supplementary). As emissions from these sources are grouped together in EDGAR inventory, the exact sources from which monoterpenes are attributed to cannot be identified. The sensitivity of  $\Delta P(O_3)$  from reducing process emission sources (PE, RT+PE, and RT+PE+CM) is shown by the shaded regions in Figure 12, where the dashed lines represent the sensitivity limits where the observed monoterpenes are between 0% and 100% anthropogenic (as opposed to biogenic). There is relatively little impact on  $P(O_3)$  on reducing RT+PE when monoterpenes are assumed to have an entirely biogenic source. However, it is clear that although the degree to which reducing process emissions along with road transport in this study impacts  $P(O_3)$  cannot be accurately determined, even if no monoterpenes are reduced within this source, reducing it does not negatively impact  $P(O_3)$ . It is also important to consider possible under-estimations for the monoterpene contribution to RT. Monoterpene observations in Delhi were strongly correlated with CO emissions, suggesting anthropogenic sources (Stewart et al., 2021a). The EDGAR emissions inventory assigns 0% of the anthropogenic monoterpenes in the inventory to the RT source sector (table 2). An analysis of the PTR-QiTOF flux data, obtained at the IGDTUW measurement site directly after the concentration measurement period ended, suggests ~ 60% of the monoterpenes observed

could be attributed to traffic factors (Cash et al., 2021). A study by Wang et al, 2020 suggested vehicular and burning sources may contribute to the anthropogenic emissions of biogenic molecules. Other possible sources of anthropogenic monoterpenes in Delhi are emissions from cooking herbs and spices, and from fragrances and personal care products (Klein et al., 2016, McDonald et al., 2018).

## **Reviewer 2: Technical comments**

**Indian megacity of Delhi, may be better writes like "the megacity Delhi, India", I think Delhi is a megacity also world wide.**

*Response:* Title has been changed.

*Text change:* In situ ozone production is highly sensitive to volatile organic compounds in Delhi, India.

**Cash et al., 2021, in preparation. should not be included in the reference list, it may be simply wrote in the line of 393 as (Cash et al., 2021, in preparation)**

*Response:* Reference removed from reference list.

**Figure 9, the isopleth can be improved if more sensitivity model runs are included**

*Response:* The authors are unsure on the level of detail the reviewer is requesting to improve the isopleth but have revised the text to provide the reader with more information using the isopleth data. A more detailed description on the isopleth has been added to the text. Suggestions for ways in which NO<sub>x</sub> and VOCs can be reduced to achieve the prescribed national ozone limits have been provided, making the analysis more useful for those interested in air quality management (see response to Reviewer 1). Using the isopleth data provided, the required reductions in VOCs to achieve ozone limits when NO<sub>x</sub> is reduced to 25%, 50% and 75% of observed values are detailed in the text.

*Text change:* As has been previously discussed, O<sub>3</sub> concentrations limits of 50 ppbV were regularly exceeding during the campaign, with the maximum daily 8-hour averages peaking at 88 ppbV (Figure 4). To successfully reduce O<sub>3</sub> to the limit of 50 ppbV, O<sub>3</sub> production must be reduced by 56%. This can be achieved by reducing NO<sub>x</sub> by 25%, 50% and 75% along with a concurrent reduction in VOCs of 48%, 61% and 78% respectively. To obtain a reduction in O<sub>3</sub> production without reducing VOCs would require a NO<sub>x</sub> reduction of at least 92%.