

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 1: Major comments

1) The background material in the Introduction is largely out of date and here I list some references that provide a current assessment of ozone's distribution and trends. When discussing the impacts of ozone on human health, Jerrett et al. is a good reference, but it's quite old. Fleming et al. 2018 from the Tropospheric Ozone Assessment Report (TOAR) provide an overview of ozone's health impacts. In terms of ozone increases since the mid- 20th century, the earlier findings of Parrish et al. 2014 have now been superseded by Tarasick et al. (2019) (from TOAR). Since the 1990s, surface ozone trends vary by region (Gaudel et al., 2018; Cooper et al., 2020; Lu et al., 2020), but in the free troposphere trends since the 1990s have been overwhelmingly positive (Gaudel et al., 2020; Liao et al., 2020; also see the review by Cooper et al., 2020). The paper by Ni et al. (2018) is not a good reference regarding ozone trends as it only focuses on a single year (2008). A good paper that shows the increases of ozone across China is Lu et al. 2020.

Response: The reviewer makes a good point that there are more up to date references than those used in the introduction section of the manuscript. To improve this, additional text has been added to this section, along with updated references as suggested by the reviewer. The authors would like to thank the reviewer for kindly providing some excellent references to improve this section.

Text change: Tropospheric O₃ is both an air pollutant and an important greenhouse gas throughout the troposphere (Skeie et al., 2020). High levels of O₃ can adversely affect vegetation, global crop yields (Avnery et al., 2011) and human health, with long-term exposure increasing the risk of death from cardiovascular and respiratory illnesses (Jerrett et al., 2009), and short-term exposure leading to the exacerbation of asthma in children (Thurston et al., 1997). O₃ exposure has been linked to both acute and chronic pulmonary and cardiovascular health outcomes through both animal toxicological and human clinical studies, with one study showing statistically significant decreases in the lung function of adults on an average exposure of 70 ppbV of O₃ across five 6.6-hour windows (WHO 2005 and 2013, Schelegle et al., 2009, EPA 2013, Fleming et al., 2018).

As a result of increased anthropogenic emissions, tropospheric O₃ increased globally during the 20th century, and has continued to rise regionally in Asia during the 21st century (Fleming et al., 2018; Royal Society, 2008, Lu et al., 2020). Background tropospheric O₃ has also continued to increase (Parrish et al., 2014, Tarasick et al., 2019). Since the 1990s, surface O₃ trends have varied by region (Gaudel et al., 2018, Cooper et al., 2020, Lu et al., 2020), but trends in the free troposphere have been overwhelmingly positive (Gaudel et al., 2020, Liao et al., 2020). Both satellite data and global chemical transport models have identified India and East Asia as the region with the greatest O₃ increases between 1980-2016 (Ziemke et al., 2019), with the rate of change per decade between 2005-2016 more than double that of the rate between 1979-2005 (Ziemke et al., 2019).

2) According to the ACP/Copernicus Data Policy, the paper needs to include a "Data availability" section, as follows: Authors are required to provide a statement on how their underlying research data can be accessed. This must be placed as the section "Data availability" at the end of the manuscript. Please see the manuscript preparation guidelines for authors for the correct sequence. If the data are not publicly accessible, a detailed explanation of why this is the case is required. The best way to provide access to data is by depositing them (as well as related metadata) in FAIR-aligned reliable public data repositories, assigning digital object identifiers, and properly citing data sets as individual contributions. The authors have not provided a "Data availability" section, which needs to be addressed before the paper can be published. Further details are available here: https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html

Response: A data availability section has been added, providing a catalogue link to the project data on the CEDA archive.

Text change: Data availability. Data used in this study can be accessed from the CEDA archive: Nemitz, E., Acton, W. J., Alam, M. S., Drysdale, W. S., Dunmore, R. E., Hamilton, J. F., Hopkins, J. R., Langford, B., Nelson, B. S., Stewart, G. S., Vaughan, A. R., Whalley, L. K., (APHH India) Megacity Delhi atmospheric emission quantification, assessment and impacts (DelhiFlux), <https://catalogue.ceda.ac.uk/uuid/ba27c1c6a03b450e9269f668566658ec>, 2020.

3) The authors conducted a range of sensitivity tests to understand the response of ozone production to changes in NO_x and VOCs. However, an air quality manager who is tasked with keeping ozone levels below the Indian ozone standard of 50 ppbv needs more information. They need to know how much they need to cut NO_x and VOCs in order to keep the maximum daily 8-hour average below 50 ppbv. To make the study more relevant to air quality management the authors should experiment with their box model to find a range of NO_x and VOC mixing ratios that will keep ozone below 50 ppbv.

Response: The authors recognise that more information on possible ways NO_x and VOCs can be reduced to achieve ozone levels of 50 ppbV would be useful to policy makers and those interested in air quality management. A more detailed description of the isopleth has been added to section 3.5, to provide information on how much VOCs would need to be reduced by to ensure the maximum 8-hour average observed during the campaign is reduced to 50 ppbV on reducing NO_x by 25%, 50% and 70%.

Text change: As has been previously discussed, O₃ 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 O₃ to the limit of 50 ppbV, O₃ production must be reduced by 56%. This can be achieved by reducing NO_x by 25%, 50% and 75% along with a concurrent reduction in VOCs of 48%, 61% and 78% respectively. Alternatively, if VOCs were halved, NO_x reductions could not exceed 29% to peak O₃ below 50 ppbV. To obtain a reduction in O₃ production without reducing VOCs would require a NO_x reduction of at least 92%. However, it is important to consider that reducing emissions of NO_x and VOCs by these values will only impact the in situ formation of O₃. Regional O₃ production

leading to the transportation of O₃ from outside of Delhi is beyond the scope of this analysis and must also be taken into consideration by policy makers.

Reviewer 1: Minor comments

Line 53

Here ozone is described as an important greenhouse gas in the mid-troposphere. However, ozone acts as a greenhouse gas throughout the depth of the troposphere, with a maximum radiative impact in the upper troposphere. See Figure 1 in the Supplement of Skeie et al., 2020.

Response: Text has been changed to address that ozone is an important greenhouse gas throughout the troposphere, as seen in Skeie et al., 2020.

Text change: Tropospheric O₃ is both an air pollutant and an important greenhouse gas throughout the troposphere (Skeie et al., 2020).

Line 64

“cocktail” is a fine analogy for conversational discussions, but not for a scientific paper. Use something like “range” instead.

Response: “Cocktail” changed to “range” in text.

Text change: Unlike other pollutants such as NO_x and SO₂, ground-level O₃ is not directly emitted but is formed in the atmosphere from the photochemical processing of a range of reactive precursor species (Calvert et al., 2015).

Line 75

The presentation of basic ozone photochemistry should include a reference

Response: Reference added.

Text change: In general, O₃ formation is mediated by the reactions of peroxy radicals, RO₂ and HO₂, formed in the OH-initiated oxidation of VOCs (R1), with NO to produce NO₂ (R2 and R4). NO₂ is then rapidly photolyzed back to NO, forming O(³P) (R5), which can rapidly react with O₂, leading to O₃ (R6). This recycling of NO to NO₂ leads to a net production of O₃ (Calvert et al., 2015).

Line 96

Shouldn't but-2-enes be 2-butenes? Here is the relevant passage from Ran et al., 2011: “The most reactive species responsible for ozone formation are mainly alkenes and aromatics such as 2-butenes (18 %), isoprene (15 %), trimethylbenzenes (11 %), xylenes (8.5 %) and toluene (4.5 %).”

Response: Although “2-butenes” is used in Ran et al., 2011, and elsewhere in the literature, the authors have kept the text as but-2-enes as this is the IUPAC preferred nomenclature. See

“Nomenclature of Organic Chemistry: IUPAC Recommendations and Preferred Names 2013, IUPAC Blue book, prepared for publication by Henri A Favre and Warren H Powell, by RSC Publishing, 2014 [ISBN 978-0-85404-182-4]; <https://doi.org/10.1039/9781849733069>”.

Line 104

A reference is needed for the statement on personal care products. McDonald et al. (2018) is a good one.

Response: Reference added.

Text change: Oxygenated VOCs from solvent consumption and personal care products dominate the VOC-OH reactivity in recent years, leading to sustained high O₃ concentrations in the MCMA (McDonald et al., 2018, Zavala et al., 2020).

Line 105

A reference is needed for this statement: “Understanding which precursor species are key to O₃ production in any given city allows governments to introduce measures to combat air quality problems.”

Response: Reference added.

Text change: Understanding which precursor species are key to O₃ production in any given city allows governments to introduce measures to combat air quality problems (Molina, 2021).

Line 135

“was attributed” should be “were attributed”

Response: Changed.

Text change: However, increased concentrations of ground-level O₃ (> 10 %) were also observed and were attributed to reductions of NO leading to reduced consumption of O₃.

Line 136

I don’t think I’ve ever heard of the term “deweathered”. Do you mean to say that meteorological biases were removed?

Response: Changed.

Text change: Another study found that, after removing meteorological biases, concentrations of NO₂ and PM_{2.5} at urban background sites in Delhi to have reduced by ~ 51 % and ~ 5 % respectively, with O₃ concentrations increasing by ~ 8 % (Shi et al., 2021).

Line 182

high should be height

Response: Changed.

Text change: IGDTUW facilitated the sampling of ambient air from a height of ~ 5 m and measurements were made of a large range of VOCs, o-VOCs, NO_x, CO, SO₂, HONO, photolysis rates and PM.

Line 367

Here you state that the observations “suggest” that the standard was exceeded on 16 days. But to say “suggest” implies that you aren’t really sure. However, your measurements show that the standard was definitely exceeded on 16 days, and you should rephrase the sentence so that it reflects your confidence in your observations.

Response: “suggest” has been changed to “show”.

Text change: Our observations show that the 8-hour O₃ prescribed national standard is exceeded on 16 days during our 24-day measurement period (67 % of days)

Line 369

Again, why use the word “suggest”? An official government document should clearly state the policy, with no ambiguity.

Response: “suggest” has been changed to “state that”

Text change: The published national standards state that any pollutant which exceeds the prescribed values for two consecutive days qualifies for regular and continuous monitoring.

Line 444

On line 348 the ozone peak is stated to occur at 13:00, but here the peak is stated to occur at 12:00. Please reconcile.

Response: Thank you for spotting this – the peak occurs at 13:00. The text has been changed to reflect this.

Text change: In Delhi, O₃ concentrations rapidly increase from ~ 08:00, peaking around ~ 13:00.

Line 495

There seems to be a typo in the following sentence in the caption to Figure 9: “The red diamond represents at point 1,1 represents modelled P(O₃) at observed VOC and NO_x concentrations.”

Response: Corrected.

Text change: The red diamond at point 1,1 represents modelled P(O₃) at observed VOC and NO_x concentrations.

Line 573

Delete “the” before prevalence

Response: “the” removed.

Text change: Urban NO_x emissions (at tailpipe) are likely to decline over time, as a result of improved exhaust gas treatments, the turnover of the fleet to newer, less polluting vehicles, and the increasing prevalence of electric vehicles (Molina, 2020).

Line 658

“represents and aggregate” should be “represents an aggregate”

Response: Changed.

Text change: Whilst the OH reactivity analysis provides an instantaneous assessment of a VOC’s reactivity, based on local observations with a small spatial footprint, the EDGAR Delhi-wide sectoral split represents an aggregate of a wider region and may under-represent the RT VOC contribution observed at the measurement site.

References:

Cooper, O. R., et al. (2020), Multi-decadal surface ozone trends at globally distributed remote locations, *Elem Sci Anth*, 8(1), p.23. DOI: <http://doi.org/10.1525/elementa.420>

Gaudel, A., et al. (2020), Aircraft observations since the 1990s reveal increases of tropospheric ozone at multiple locations across the Northern Hemisphere. *Sci. Adv.* 6, eaba8272, DOI: 10.1126/sciadv.aba8272

Liao, Z., Ling, Z., Gao, M., Sun, J., Zhao, W., Ma, P., Quan, J. and Fan, S., 2021. Tropospheric Ozone Variability Over Hong Kong Based on Recent 20 years (2000–2019) Ozonesonde Observation. *Journal of Geophysical Research: Atmospheres*, 126(3), p.e2020JD033054.

Lu, X., Zhang, L., Wang, X., Gao, M., Li, K., Zhang, Y., Yue, X. and Zhang, Y., 2020. Rapid increases in warm-season surface ozone and resulting health impact in China since 2013. *Environmental Science & Technology Letters*, 7(4), pp.240-247.

McDonald, B.C., De Gouw, J.A., Gilman, J.B., Jathar, S.H., Akherati, A., Cappa, C.D., Jimenez, J.L., Lee-Taylor, J., Hayes, P.L., McKeen, S.A. and Cui, Y.Y., 2018. Volatile chemical products emerging as largest petrochemical source of urban organic emissions. *Science*, 359(6377), pp.760-764.

Skeie, R.B., Myhre, G., Hodnebrog, Ø., Cameron-Smith, P.J., Deushi, M., Hegglin, M.I., Horowitz, L.W., Kramer, R.J., Michou, M., Mills, M.J. and Olivié, D.J., 2020. Historical total ozone radiative forcing derived from CMIP6 simulations. *npj Climate and Atmospheric Science*, 3(1), pp.1-10

Tarasick, D. W., et al. (2019), Tropospheric Ozone Assessment Report: Tropospheric ozone from 1877 to 2016, observed levels, trends and uncertainties. *Elem Sci Anth*, 7(1), DOI: <http://doi.org/10.1525/elementa.376>