Review replies for the manuscript entitled "Representing improved tropospheric ozone distribution over the Northern Hemisphere by including lightning NOx emissions in CHIMERE" (egusphere-2024-3087) by Sanhita Ghosh et al.

We sincerely thank the editor and the reviewer for the constructive feedback and insightful comments on our manuscript. Below, we address each comment in detail and outline the corresponding revisions made to improve the manuscript.

Reviewer 1

Summary

The authors implement a cloud-height based flash rate scheme and an ice-flux based scheme into the regional model CHIMERE and then evaluate the impact of lightning-NOx production on ozone profiles, the tropospheric OH burden, the methane lifetime, and ambient ozone and NO_2 concentrations. This version of the paper is much improved but remains unpublishable until the authors explain how the addition of LNOx leads to a decrease in NO_2 columns over much of the domain.

Reply: We thank the reviewer for the valuable feedback. In response to the concern regarding the decrease in the NO_2 column due to inclusion of LNOx, we have provided a detailed explanation of how the inclusion of LNOx emissions in our model influences the NO_2 levels. We have also updated the manuscript with additional plots to better illustrate and clarify this effect. We also have thoroughly responded to each comment and highlighted the revisions made to enhance the manuscript.

Major Comments:

1. L281: Be specific as to what is improved in this simulation over water versus the original PR92. My hunch is that you obtained a better ocean-simulation because the ratio of a/b you use is smaller than that used in PR92, thus after scaling a higher fraction of flashes are over water lessening the low-bias others have observed with PR92 over water. – Do you agree?

Reply: We have now specified the improvement made in our simulation with the scheme based on cloud top height (CTH) in respect to the original scheme by Price and Rind (1992), due to which we have obtained an improved flash rate over the ocean. Yes, our study demonstrates an improved flash rates over tropical oceans, using the CTH scheme with a correction factor of 0.5 applied to the constant 'b' in Equation 1 (please see Section 2.2.1), for the oceanic grids. A simulation with the original scheme by Price and Rind (1992) showed an overestimated flash rates over the tropical ocean by a factor $\approx$2, which is lowered to 1.3–1.4, after application of the correction factor in the present study.

Changes in the manuscript: Please see the lines 293–296, "However, our study demonstrates an improved flash rate distribution over tropical oceans, using the CTH scheme with a correction factor of 0.5 applied to constant 'b' in Equation 1, for the oceanic grids. A simulation with the original scheme by Price and Rind (1992) showed an overestimated flash rates over the tropical ocean by a factor $\approx$2, which is lowered to 1.3–1.4, after application of the correction factor in the present study."

2. L289-296: I don't follow your rationale here. Are you saying that more realistic midlatitude flash rates could be produced if cloud heights were allowed to extend above the tropopause? Shouldn't cloud heights be limited by the tropopause? Yes,

overshooting storms exist and do transport trace gases into the stratosphere but aren't anvils typically limited by the tropopause. Perhaps the heights of tropical storms are too high.

Reply: Here we intend to highlight the limitation of using cloud top height as the sole parameter for estimating lightning flash rates. While CTH scheme provides an useful approximation, since deeper convection generally correlates with higher lightning activity, it likely doesn't capture the full complexity of the processes driving lightning generation. Overshooting tops are a clear example of where relying solely on cloud top height can fall short, as these strong convective bursts can extend above the tropopause and influence lightning rates, even if the average cloud top height remains lower. Moreover, factors, such as updraft strength, cloud depth, ice water content and mixed-phase regions play critical roles in charge separation and lightning production. By strictly capping cloud heights at the tropopause in the CTH scheme, the model may indeed underestimate flash rates in the mid-latitudes. This highlights the need to consider a multi-parameter approach for estimating flash rates, incorporating updraft dynamics, cloud microphysics and ice-phase processes alongside cloud top height.

We have modified the lines for more clarity.

Changes in the manuscript: Please see the modified lines 304–310, "While CTH scheme provides an useful approximation, since deeper convection generally correlates with higher lightning activity, it likely doesn't capture the full complexity of the processes driving lightning generation. Factors, such as updraft strength, cloud depth, ice water content and mixed-phase regions play critical roles in charge separation and lightning production. By strictly capping cloud heights at the tropopause in the CTH scheme, the model may indeed underestimate flash rates in the mid-latitudes. This highlights the need to consider a multi-parameter approach for estimating flash rates, incorporating updraft dynamics, cloud microphysics and ice-phase processes alongside cloud top height."

3. Figure 5: Your scale does not include negatives or is there room for negatives in the point of the arrow? I'm surprised that O_3 from LNOx-ICEFLUX exceeds ozone from LNOX-CTH everywhere given the similarity in their total flash rates. Please double check your calculations and also verify that the legend matches what is plotted.

Reply: We have now modified the Figure 5 by changing the scale including negative values. The O_3 mixing ratio from LNOx-ICEFLUX is higher than that obtained from LNOx-CTH over most of the NH except few regions over the ocean. This is probably because of the higher LNOx emissions obtained in LNOx-ICEFLUX than the LNOx-CTH simulation. The O_3 mixing ratio from LNOx-ICEFLUX shows the maximum increase over the central Africa, followed by other tropical region for all the altitude bands. However, the increase in LNOx-ICEFLUX in respect to LNOx-CTH is smaller over most of the NH. Overall ther O_3 burden over NH from LNOx-ICEFLUX is 3% higher than that estimated from LNOx-CTH, while the increase in O_3 burden is 7%–11% with respect to the simulation without LNOx (noLNOx).

Changes in the manuscript: Please see the Figure 5 with modified scale.

4. L475-479: I don't understand how the inclusion of LNOx can lead to a decrease in column NO_2 over most of your non-tropical domain. This seems wrong. Please double check your calculations. How do the changes vary seasonally? Do you see increases during the spring and summer? Yes, you can see local decreases especially in the boundary layer over polluted regions but total columns must increase.

Reply: In our study, the decrease in NO_2 column density (in molecules cm^{-2}) due to inclusion of NOx from lightning, is primarily observed over the regions with high NO_2 pollution, such as southern and eastern Asia (India and eastern China), north-west Europe and eastern part of USA. The inclusion of LNOx in model increases large-scale ozone and OH concentrations, therefore reducing the lifetime of NOx through oxidation reactions with HOx including OH (Labrador et al., 2005; Schumann and Huntrieser, 2007). Figure S5 in supplementary material depicts the increase in HNO_3 column density over the above mentioned region, supporting the fact that NO_2 is oxidized and converted to the HNO_3, increasing the column density of HNO_3. Hence, rapid conversion of NO_2 into other compounds, such as HNO_3 leads to its subsequent removal and a net decrease in NO_2 column density over the regions with high anthropogenic pollution. A decrease in NO_2 column density over the above mentioned regions, is also observed during the late spring and summer season (May–August).

The Figure S6 in supplementary material, showing changes in annual mean NO_2 mixing ratio (in ppbv) from experiment LNOx-CTH with respect to noLNOx, demonstrates a decrease in NO_2 by 0.1–0.3 ppbv over the regions with higher anthropogenic NO_2 pollution as mentioned above, at the altitude band 998–900 hPa, i.e., mostly near surface, followed by the altitude band 900–750 hPa. A very small increase (0.05 ppbv) is observed over most of the NH at the higher altitude bands (750–500 and 500–200 hPa), due to inclusion of LNOx emissions. Overall the NO_2 column density decreases over the regions with high anthropogenic pollution.

Changes in the manuscript: Please see the lines 512–527, "A decrease in NO_2 column density (0.2–0.6 $\times$ 10^{15} molecules cm^{-2}) due to inclusion of LNOx emissions, is primarily observed over the above mentioned regions with high NO_2 pollution (Figure 9b). The inclusion of LNOx in model increases large-scale O_3 and OH concentrations, therefore reducing the lifetime of NOx through oxidation reactions with HOx including OH (Labrador et al., 2005; Schumann and Huntrieser, 2007). Figure S5 in supplementary material depicts the increase in HNO_3 column density over the above mentioned region, supporting the fact that NO_2 is oxidized and converted to the HNO_3, increasing the column density of HNO_3. Hence, rapid conversion of NO_2 into other compounds, such as HNO_3, leads to its subsequent removal and a net decrease in NO_2 column density over the regions with high anthropogenic pollution. The Figure S6 in supplementary material, showing changes in annual mean NO_2 mixing ratio (in ppbv) from experiment LNOx-CTH with respect to noLNOx, demonstrates a decrease in NO_2 by 0.1–0.3 ppbv over the regions with higher anthropogenic NO_2 pollution as mentioned above, at the altitude band 998–900 hPa, i.e., mostly near surface followed by the altitude band 900–750 hPa. A very small increase (0.05 ppbv) is observed over most part of NH at the higher altitude bands (750–500 and 500–200 hPa), due to inclusion of LNOx emissions. Overall the NO_2 column density decreases over the regions with high anthropogenic pollution."

Minor Comments:

5. L88: 20 vertical levels is quite coarse and a 200 hPa model top Is very low.

Reply: Currently the simulations are conducted with twenty vertical levels in σ-pressure coordinates ranging from surface to 200 hPa. Yes, the vertical resolution is coarse and model top is low. Although, boundary and initial conditions (derived from Copernicus Atmosphere Monitoring Service (CAMS) reanalysis dataset of atmospheric compositions produced by ECMWF for this study) takes the stratospheric chemistry into account.

6. L125-130: As equation (2) is not used, to avoid confusion, this section could be shorted with the equation removed

Reply: The Equation 2 has been used to estimate the scaling factor (C), which is further used in Equation 3 to estimate the total flash rate (F_{CTH}). The scaling factor is used to adapt the Equation 1 to various model resolutions.

Please see the Section 2.2.1 and Equations 1, 2 and 3.

$$\begin{aligned} F_l &= a \times H_{top}^{4.9} \\ F_o &= b \times H_{top}^{1.73} \end{aligned} \quad (1)$$

Price and Rind (1994) formulated an equation to adapt the above equations to various model resolutions. Here, the product of longitude and latitude resolution, denoted as $\Delta x \times \Delta y$, is measured in degrees^2.

$$C = 0.97241e^{0.048203 \times \Delta x \times \Delta y} \quad (2)$$

The total flash rate (F_{CTH}) is then calculated as follows:

$$F_{CTH} = \frac{C \times (x_{\text{sea}} \times F_o + (1 - x_{\text{sea}}) \times F_l)}{25} \quad (3)$$

7. L156-157: Why is this distinction made? Why is it important that flashes above the freezing level are considered IC and flashes below it CG?

Reply: Cloud to ground (CG) flashes are typically associated with strong charge separation within the cloud, often involving a negative charge at the base of the cloud and a positive charge on the ground or vice versa (Dwyer and Uman, 2014). This type of lightning requires a large electric potential difference between the cloud and the ground, and this process is generally more common below the freezing level. In cloud (IC) flashes occur within the cloud are often associated with the discharge of charge between different regions inside the cloud. Above the freezing level, the presence of ice particles contributes to the charge separation mechanism. The interaction between ice and liquid water are more prominent in the upper troposphere where the temperature is below freezing. The freezing level acts as a natural boundary between the upper and lower parts of the cloud. Above the freezing level, ice particles contribute to the development of IC lightning, while below it, the atmosphere is typically in a liquid state, with the warmer environment aiding in the development of CG lightning.

We have added these details in the manuscript.

Changes in the manuscript: Please see the line 163–166, "The freezing level acts as a natural boundary between the upper and lower parts of the cloud. Above the freezing level, ice particles contribute to the development of IC lightning, while below it, the atmosphere is typically in a liquid state, with the warmer environment aiding in the development of CG lightning (Dwyer and Uman, 2014)."

8. L170: "Beta" = IC/CG = "formula"

Reply: This is done.

Changes in the manuscript: Please see the line 178, "β = IC/CG (Equation 7), obtained in our study again shows consistency with the above mentioned results."

9. L201-202: Justify why you also include NO_2 emissions from lightning.

Reply: Lightning is one of the largest natural sources of NOx, including both NO and NO_2 in the atmosphere. During a lightning strike, the extremely high temperature causes the dissociation of nitrogen (N_2) and oxygen (O_2) molecules in the air into atomic nitrogen (N) and oxygen (O). These free radicals then recombine to form NO, which can be converted to NO_2 in the presence of oxygen (Murray et al., 2012; Murray, 2016; Finney et al., 2014). Lightning generate NO_2 with NO_2/NOx ratio $\approx$0.1 to 0.5 (Schumann and Huntrieser, 2007). Therefore, it is important to include NO_2 emissions also.

Changes in the manuscript: Please see the line 210–212, "Lightning generate NO_2 with NO_2/NOx ratio varying from 0.1 to 0.5 (Schumann and Huntrieser, 2007) Therefore, it is important to include NO_2 emissions also."

10. L260-262 and L341-344: Do you also divide by 5 in the tables? Assuming yes, introduce the factor of 5 in section 2.2.2. Move discussion in 341-344 2.2.2 too. You do apply scaling factors to LNOx-CTH but before the model was run.

Reply: Yes, all the calculations and statistical analyses in tables are conducted for the flash rates estimated with ICEFLUX scheme, divided by the factor 5. We have now included this information in Section 2.2.2.

Yes, we have applied scaling factors to LNOx-CTH before conducting the simulation as described in Section 2.2.1, and no scaling factor is applied in the simulated flash rates from LNOx-CTH. This is mentioned in the lines 361–362 "While no scaling factor is applied in the simulated flash rates from LNOx-CTH, the flash frequency from LNOx-ICEFLUX is divided by a factor 5 to reconcile with the satellite-observed frequency.".

Changes in the manuscript: Please see the lines 155–156 added to Section 2.2.2, "The estimated flash frequency from LNOx-ICEFLUX has been scaled down by a factor of 5 to align with satellite-observed frequencies. Consequently, the evaluation of LNOx-ICEFLUX results has been carried out using these adjusted flash rates."

11. L274: where in the United States are you referring to?

Reply: We mean the central and south-eastern part of the United States. This is now added in the manuscript.

Changes in the manuscript: Please see the lines 286–288, "However, patches of high flash rate observed in satellite data over central Canada, central and south-eastern part of the United States, central European countries, and northern Russia are not reflected in the modelled flash rates from either experiment."

12. L288: "explaining the effectiveness of ICEFLUX scheme over CTH, in capturing flashes over the midlatitudes". Yes, the mean flash rates from the ICEFLUX scheme appear to be more realistic in the midlatitudes. However, the error statistics you show in Table 3 still favor the CTH scheme. Why is that? Is there a metric for which the ICEFLUX scheme does better in the midlatitudes?

Reply: As presented in Table 2 and discussed in the manuscript, the mean flash rate from LNOx-ICEFLUX over the mid-latitudes closely aligns with satellite observations and is nearly twice that estimated from LNOx-CTH. This highlights the effectiveness of the ICEFLUX scheme in capturing a more realistic magnitude of flash rates over the mid-latitudes.

However, Table 3 presents the statistical evaluation of the spatial variability of annual flash rates, comparing modelled flash rates with ISS-LIS and LIS/OTD satellite observations on a grid scale. This statistical scores represent the efficiency of the CTH scheme in reproducing the spatial variation of lightning flashes reasonably well than that observed for ICEFLUX scheme. While LNOx-ICEFLUX provides a reasonable estimate of flash rate magnitudes over both the tropics and mid-latitudes, it struggles to accurately capture the observed spatial pattern of lightning activity.

The ICEFLUX scheme offers significant advantages over CTH for estimating lightning flash rates as it directly accounts for the microphysical and thermodynamic processes that drive charge separation in convective storms. While CTH is often used as a proxy for convective intensity, it does not explicitly capture the ice-phase interactions crucial for electrification, such as graupel formation, supercooled liquid water content and ice crystal collisions. ICEFLUX scheme, by explicitly modelling ice fluxes, provides a more realistic approach in predicting charge separation and lightning activity (Finney et al., 2014). This leads to improved flash rate estimations, particularly in midlatitude storms where vertical motion, ice microphysics and latent heat fluxes play a complex role in thunderstorm electrification.

Changes in the manuscript: Please see the modified lines 331–332, "While LNOx-ICEFLUX provides a reasonable estimate of flash rate magnitudes over both the tropics and mid-latitudes, it struggles to accurately capture the observed spatial pattern of lightning flashes, emphasizing the need for further improvement."

Also see the lines 310–313 added newly, "ICEFLUX scheme, by explicitly modelling ice fluxes, provides a more realistic approach in predicting charge separation and lightning activity (Finney et al., 2014). This leads to improved flash rate estimations, particularly in midlatitude storms where vertical motion, ice microphysics and latent heat fluxes play a complex role in thunderstorm electrification."

13. Table 2: Add rows showing corresponding values for ISS-LIS and LIS/OTD.

Reply: This is done.

Changes in the manuscript: Rows showing flash rate values from ISS-LIS and LIS/OTD are added in Table 2.

14. L304-308: The accuracy of the lightning data is also lower at high latitudes. As the data sets contain fewer years. Also, what fraction of flashes does ISS-LIS capture? Does it only view a given area for a few minutes each day? Data are also lower quality over the oceans due to sampling limitations.

Reply: Lightning Imaging Sensor (LIS) is mounted on the International Space Station (ISS) (domain: ±55° latitudes). ISS operates in low Earth orbit (LEO) and overpasses one region on the earth surface up to three times a day and up to two times in the tropics. Lightning observation of a specific point lasts up to 90 seconds per overpass (Erdmann et al., 2020). The flash detection efficiency of ISS-LIS is around 60% with diurnal variability of 51%–75% (Blakeslee et al., 2020), as already mentioned in the manuscript (line number 236–237). Despite of its lower quality over the oceans, the lightning data provided by ISS-LIS is available for the year 2018, which is our study period, giving the opportunity to evaluate the simulated flash rates with respect to the ISS-LIS observations. We also have included comparison with respect to combined climatology product of satellite observations from the Optical Transient Detector (OTD) and the LIS, for the period of May, 1995 to December, 2014 (please see the lines 237–241).

Changes in the manuscript: We have included more details about ISS-LIS data. Please see the lines 234–236, "ISS operates in low Earth orbit (LEO) and overpasses one region on the earth surface up to three times a day and up to two times in the tropics. Lightning observation of a specific point lasts up to 90 seconds per overpass (Erdmann et al., 2020)."

15. L330: Are these correlations temporal or spatial? Based on the context and values, it appears they are temporal – correlations between monthly average flash rates for each latitude band?

Reply: The correlations are temporal. The statistical analysis presented in Table 4 are conducted for monthly mean flash rates from simulations in comparison to the monthly mean flash rates from satellite observations, for each latitude band. This is mentioned in lines 347–348, "The modelled monthly mean flash rates from the simulations exhibit a strong positive correlation with satellite observations (ISS-LIS and LIS/OTD), with correlation coefficients ranging from 0.85 to 0.97 (Table 4)." The word 'temporal' is now included in lines 351–352.

Changes in the manuscript: Please see the lines 351–352, "In contrast, a weaker negative temporal correlation is observed over tropical and mid-latitudinal oceans, indicating an inverse relationship between simulated and observed seasonality in flash rates in these regions."

16. L348: You hint at factors of 2-3 magnitudes but only show factors varying by factors of 2-4. Rephrase or give more details.

Reply: As per Tost et al. (2007), the scaling factors may vary up to 2–3 orders of magnitude to match observations, based on the lightning parameterization used and the results obtained. Tost et al. (2007) reported scaling factors as large as 435 and as low as 0.74 to match observations. A recent study by Finney et al. (2016) determined that the global flash rate scaling factors required for the UKCA model are 1.44 and 1.12 for the CTH and ICEFLUX lightning parameterizations, respectively. Another study by Gordillo-Vázquez et al. (2019) produce scaling factors 2.05 and 4, respectively for the CTH and ICEFLUX lightning schemes in Community Atmosphere Model (CAM5). In our study, the flash rates from LNOx-ICEFLUX simulation, are adjusted by a factor 5 to reconcile with the satellite observations.

Changes in the manuscript: Please see the rephrased lines 367–369, "A study by Tost et al. (2007) reported that scaling factors may vary by up to 2–3 orders of magnitude, depending on the lightning parameterization used and the resulting flash rate, to better match the observations."

17. L350: Given the 4.9th power of cloud height variation, varying the minimum depth would have little effect.

Reply: A study by Luhar et al. (2021) has shown that increasing or decreasing the minimum cloud thickness value by 1 km from 5 km result in a change of -3.2% and 1.7%, respectively, in the modelled global flash rate, estimated using the CTH scheme. The effect may be lesser but still contributes to the uncertainty in estimating flash rates. Therefore, we mention that, it would be worthwhile to study the sensitivity of the modelled flash rates to the minimum cloud depth. We have moved these lines to the Section 2.2.1, where we introduced the minimum required cloud depth for the first time.

Changes in the manuscript: Please see the lines 125–128, "The assumption of minimum required cloud depth of 5 km, may introduce uncertainty in estimating lightning flashes, as it inherently assumes that every convective cloud with depth of 5 km corresponds to a thunderstorm (Luhar et al., 2021). It would be worthwhile to investigate the sensitivity of the modelled flash rates to the minimum cloud depth by varying this arbitrary threshold, either increasing or decreasing it."

18. L354: I'm not sure why you use "but" here. How did Finney determine "better"?

Reply: The study by Finney et al. (2014) showed that the correlations of the monthly average of upward ice flux at 340, 440 and 540 hPa, formed from ERA-Interim reanalysis for the grid cells over land, against LIS flash density were found stronger for lower pressure levels (340 and 440 hPa) in comparison to the higher pressure level (540 hPa), specifically over land. As per Finney et al. (2014), further investigation at that point was not necessary since only slight gains could be made by arbitrarily optimising the pressure level. We have now removed this line to avoid confusion.

Changes in the manuscript: We have removed the line.

19. L378: Remind the reader how you determined the vertical partitioning of LNOx emissions. Do these plots sum the contributions from IC and CG flashes? Do you assume only IC flashes for some altitudes and only CG for others etc.

Reply: We have added few lines reminding how we determined the vertical partitioning of LNOx emissions. The emissions from CG and IC flashes are calculated separately considering CG flashes only below the freezing level and the IC flashes only above the freezing level and below the cloud top. A simple vertical structure of the emissions is adopted in this study, considering the emissions to be evenly distributed over an altitude range.

Changes in the manuscript: Please see the lines 391–394, "The emissions from CG and IC flashes are calculated separately considering CG flashes only below the freezing level and the IC flashes only above the freezing level and below the cloud top. A simple vertical structure of the emissions is adopted in this study, considering the emissions to be evenly distributed over an altitude range."

20. L385: What previous studies are you referring to and are you certain they are plotting mass and not mixing ratio.

Reply: We have added the references of the previous studies (Pickering et al., 1998; Ott et al., 2010; Luhar et al., 2021). These studies plotted the LNOx emissions as percentage of total LNOx mass per km.

Changes in the manuscript: Please see the lines 401–403, "The vertical profiles available from previous studies, e.g., Pickering et al. (1998); Ott et al. (2010); Luhar et al. (2021), reveal a similar shape of all the profiles but contributing maximum at upper tropospheric region (within 2–4 km of the tropopause) rather than mid-troposphere."

21. L393: "evenly distributed over an altitude range". This information should be given earlier in the text.

Reply: The line has been shifted to provide the information earlier in the text.

Changes in the manuscript: Please see the lines 393–394, "A simple vertical structure of the emissions is adopted in this study, considering the emissions to be evenly distributed over an altitude range."

22. L417-420: Midlatitude and polar region changes vary greatly by season. Thus, it is not surprising that annual mean changes are relatively small. What percent changes are seen in the late spring/summer.

Reply: Yes, it is true that large seasonal changes are observed at mid-latitudes and polar region, lowering the annual mean increase in the O_3 mixing ratio due to inclusion of LNOx emissions. As mentioned in the manuscript, a moderate (3%–5%) to low (1%–2%) increase in annual mean of mid and upper tropospheric O_3 is observed over mid-latitudes followed by the polar region. The increase is comparatively higher during late spring and early summer (May–August) being 6%–15% over mid-latitudes and 2%–4% over polar regions.

Changes in the manuscript: Please see the newly added line 433–434, "The increase is comparatively higher during late spring and early summer (May–August) being 6%–15% over mid-latitudes and 2%–4% over polar regions."

23. L438: Your model top may be too low to adequately resolve cross-tropopause transport.

Reply: We acknowledge that the current model top (200 hPa) may limit the ability to fully capture cross-tropopause transport, especially in the upper troposphere and lower stratosphere. Future model improvements will focus on extending the model top to higher altitudes to better resolve these processes and improve the modelling of stratosphere-troposphere exchange.

Changes in the manuscript: Please see the modified line 454–456, "The underestimation suggests that the modeled stratosphere-troposphere exchange still requires significant refinement, and the cross-tropopause transport may not be adequately resolved due to the low model top."

24. L485: What is causing that unobserved peak?

Reply: The peak as observed at the latitude band 20°–30°N, of 1.75×10^{15} molecules cm^{-2}, is due to the high NO_2 column density estimated from simulations over the southern and south-east Asia due to high NO_2 emissions from larger industrial activities, than that obtained from OMI observations. As we mentioned in the manuscript, this peak is not observed from OMI. On the other hand, a study by Luhar et al. (2021) has depicted that the NO_2 column density obtained from Copernicus Atmosphere Monitoring Service (CAMS) reanalysis data, shows a peak of 1.5×10^{15} molecules cm^{-2} at this latitude band (20°–30°N), where OMI underestimates the NO_2 column density. The higher uncertainty in OMI retrieved NO_2 columns, as compared with available satellite observations (GOME-2, SCIAMACHY and TROPOMI) is considerable in this regards. The uncertainties are primarily due to instrumental errors, limitations of the OMI sensor in capturing the NO_2 below the cloud level, vertical profile assumptions and surface reflectivity (Bucsela et al., 2013; Boersma et al., 2018).

Changes in the manuscript: Please see the modified lines 511–519, "The peak at 20°–30°N, of 1.75×10^{15} molecules cm^{-2}, is due to the high NO_2 column density estimated from simulations over the southern and south-east Asia due to high NO_2 emissions from larger industrial activities. This peak is however not observed in OMI observations. On the other hand, a study by Luhar et al. (2021) has depicted that the NO_2 column density obtained from Copernicus Atmosphere Monitoring Service (CAMS) reanalysis data, shows a peak of 1.5×10^{15} molecules cm^{-2} at this latitude band (20°–30°N), where OMI underestimates the NO_2 column density. The higher uncertainty in OMI retrieved NO_2 columns, as compared with available satellite observations (GOME-2, SCIAMACHY and TROPOMI) is considerable in this regards. The uncertainties are primarily due to instrumental errors, limitations of the OMI sensor in capturing the NO_2 below the cloud level, vertical profile assumptions and surface reflectivity (Bucsela et al., 2013; Boersma et al., 2018)."

25. L486: Higher uncertainty in OMI observations relative to what?

Reply: The higher uncertainty in OMI retrieved NO_2 columns, as compared with available satellite observations (GOME-2, SCIAMACHY and TROPOMI), is primarily due to instrumental errors, limitations of the OMI sensor in capturing the NO_2 below the cloud level, vertical profile assumptions and surface reflectivity (Bucsela et al., 2013; Boersma et al., 2018).

Changes in the manuscript: Please see the modified lines 516–519, "The higher uncertainty in OMI retrieved NO_2 columns, as compared with available satellite observations (GOME-2, SCIAMACHY and TROPOMI) is considerable in this regards. The uncertainties are primarily due to instrumental errors, limitations of the OMI sensor in capturing the NO_2 below the cloud level, vertical profile assumptions and surface reflectivity (Bucsela et al., 2013; Boersma et al., 2018)."

26. L553: How do you determine the CH_4 lifetime, i.e., what CH_4 distribution do you assume. Yes, a CH_4 lifetime of 4.5–4.8 years is quite short and yes your OH must have a considerable high-bias. Your mean concentrations of $\approx 15 \times 10^5$ are approximately 50% greater than the canonical value of 10×10^5 and as you note higher than the multi-model mean of 11×10^5.

Reply: The CH_4 concentration is considered from chemical boundary conditions from CAMS reanalysis dataset of atmospheric compositions produced by ECMWF, as in our study, CH_4 anthropogenic emissions are not taken into account. This is mentioned in the lines 218–219, "[OH] is taken from the simulations in CHIMERE, whereas [CH_4] is from chemical boundary conditions derived from CAMS reanalysis dataset of atmospheric compositions, as CH_4 anthropogenic emissions are not taken into account in the model". The annual mean CH_4 burden (1930–1933 Tg) estimated in this study over the NH is ≈20% lower than the multi-model mean CH_4 burden, obtained from ACCMIP simulations (Naik et al., 2013), considering half of the global CH_4 burden over the NH (≈2406 Tg). The annual mean OH concentration is also overestimated by 30%–38%, in comparison to the multi-model mean obtained from ACCMIP simulations (Naik et al., 2013) promoting the increased chemical loss of CH_4. Therefore, the lower CH_4 burden and higher chemical loss due to reaction with OH cause the underestimated lifetime of CH_4 in this study, even in the absence of LNOx. Therefore, the underestimated CH_4 lifetime in the present study is not attributed to LNOx but likely stems from other factors including issues related to deficiencies in CH_4 burden, the chemistry or photolysis schemes.

We have included these details in the manuscript.

Changes in the manuscript: Please see the lines 591–599, "The CH_4 concentration is considered from chemical boundary conditions from CAMS reanalysis dataset of atmospheric compositions produced by ECMWF, as in our study, CH_4 anthropogenic emissions are not taken into account. The annual mean CH_4 burden (1930–1933 Tg) estimated in this study over the NH is ≈20% lower than the multi-model mean CH_4 burden, obtained from ACCMIP simulations (Naik et al., 2013), considering half of the global CH_4 burden over the NH (≈2406 Tg). As mentioned above, the OH concentration is also overestimated in our study. Therefore, the lower CH_4 burden and higher chemical loss due to reaction with OH, cause the underestimated lifetime of CH_4 in this study, even in the absence of LNOx. Therefore, the underestimated CH_4 lifetime in the present study is not attributed to LNOx but likely stems from other factors including issues related to deficiencies in CH_4 burden, the chemistry or photolysis schemes. Addressing and resolving these concerns will require further investigation in future studies."

Grammatical Comments:

27. L29: uncertainties — uncertainty

Reply: This is done.

28. L98-99: and fire emissions are taken — and fire emissions and are taken

Reply: The sentance is modified.

Changes in the manuscript: Please see the lines 98–100, "Anthropogenic emissions and fire emissions in the model are incorporated respectively from CAMS-global and CAMS Global Fire Assimilation System (GFAS, https://atmosphere.copernicus.eu/global-fire-last access: 16 May, 2024)."

29. L193: have reported — used

Reply: This is done.

30. L200: is close to — are close to

Reply: This is done.

31. L218: Flash rate — Flash data

Reply: This is done.

32. L245: observation data — observations

Reply: This is done.

33. Table 3: parenthesis — parentheses

Reply: This is done.

34. L296: showed — shown

Reply: This is done.

35. L335: specially while — especially when

Reply: This is done.

36. L405: maximum — maximum ozone

Reply: The sentence is modified.

Changes in the manuscript: Please see the lines 420–422, "The increase in O_3 is maximum over the tropical region of America, central Africa, southern Asia and the Maritime continents in south-east Asia (Indonesia, Philippines and Malaysia)."

37. L410: even higher increase

Reply: This is done.

38. L547: usually effected — usually affected

Reply: This is done.

39. L596: which have — which

Reply: This is done.

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