Review of "Night-time NO emissions strongly suppress chlorine and nitrate radical formation during the winter in Delhi"

This manuscript presents an atmospheric chemistry story from 25 days of data (11 January – 5 February) from a FIGAERO-CIMS complemented with AMS, NOx, and O3 measurements in Delhi, India. The authors use these measurements with a 0-D chemical box model to understand N2O5 sources and sinks. The study finds that the high night-time NO depletes O3, NO3, and N2O5 (a precursor to ClNO2 and thus Cl). The authors finally suggest that decreasing NO emissions will result in NO3 becoming an important nocturnal oxidant during the night and Cl during the day. The manuscript provides interesting insights into the "unusual" atmospheric chemistry, especially the role of various atmospheric oxidants, in Delhi which may be relevant for other polluted cities in the Indo-Gangetic Plain where there is similar combination of large emissions (including NOx) and unfavorable (nighttime) meteorology.

We thank the reviewer for these comments, which have improved our manuscript. Please find our responses below in blue font. Extracts from the manuscript are presented in pink, with changes from the original underlined.

Some comments:

1. The authors should be careful in the distinction between "emissions" and "levels". These are not always interchangeable. As the authors themselves discuss in the excellent discussion in the last paragraph of page-9, boundary layer dynamics (unfavorable nighttime meteorology) plays an important role in Delhi. As such, it may not be simply the "nighttime NO emissions", but the "nighttime NO levels" that are of relevant to the discussion. While this may seem trivial, this is extremely relevant from a policy perspective and specifically how the authors discuss policy implications. I suggest that the authors carefully go through the manuscript and check if the use of "emissions" and "levels" is correct and intentional.

We agree with the reviewer that this detail is important and thank them for highlighting it. We have made changes in the manuscript in the following places:

Line 256: 'These night-time NO levels deplete O_3 to extremely low concentrations…'

Line 372: 'These results suggest that, if night-time NO concentrations were to be reduced in line with WHO guidance…'

We have chosen to retain the word 'emissions' in the manuscript title. Although we appreciate that boundary layer dynamics will contribute towards the high concentrations, we still feel it is important to highlight the importance of the high night-time emissions.

Our second reviewer also suggested that our original manuscript did not sufficiently explore the role of boundary layer dynamics on the night-time NO concentration in Delhi. As such, we have made a number of changes to the manuscript in order to give this phenomenon sufficient weight. Details of these changes can be found in our responses to reviewer 2.

2. Delhi has large seasonal variations in pollution loadings (including aerosol and gas) because of a combination of changing sources (especially heating in winter) and meteorology. As a result, a 25-day study from January (2019) should be careful in generalizing the loadings (including chloride) and chemistry for the entire year. I am not saying that the findings are not important, but that they should be put in context of the study period. For example, the authors could use previously published year-round data for aerosol composition and NOx (even if no FIGAERO-CIMS) to put the study period in context.

We agree with the reviewer that the larger-scale annual context is extremely important for a study such as this. We have been careful to note in the title that we are discussing a wintertime phenomenon, as we feel that this pattern is unlikely to hold true during the summer.

We have now included a more explicit literature analysis of NOx, O_3 and particulate matter concentrations and patterns throughout the year, to seat our own results in context. The following text has been added to Section 3.1:

Lines 230-39: 'Similarly high concentrations of NO and low concentrations of O_3 at night have been observed in previous wintertime studies in Delhi. For example, Nelson et al., (2021, 2023) observed a very similar diel NO and O_3 pattern to that displayed here in October-November 2018. In a year-long study, Sharma et al., (2021) demonstrated that these high night-time NOx concentrations last from September until May, while night-time O_3 was found to reach a minimum during November and December. Previous observations of aerosol concentrations are similarly consistent with our observations: during October and November 2018, Gunthe et al., (2021) observed aerosol concentrations with a strong nocturnal increase. Gani et al., (2019), in a long-term study, showed that this pattern holds throughout the winter (December – mid-February) and, more weakly, during the spring (February – March). The same study indicates that particulate chloride concentrations are highest during the winter, and extremely low during the summer. We therefore anticipate that conclusions from our own study are likely to be most relevant in Delhi from October until March.'

3. The authors should also include the study-period in the figure captions. For example, instead of "The diel cycles of key oxidants and oxidation products in Delhi" it should be "The diel cycles of key oxidants and oxidation products in Delhi during the study period" (even better if you include the study dates)

The captions have been updated for Figs. 3, 4 and 5.

4. How are “night” and “day” defined for the analysis? I could not find this in the methods.

In Sections 3.1 and 3.2, where the overall campaign results are discussed, we used the average sunrise and sunset times to split the dataset into ‘daytime’ and ‘night-time’ values. We have now added the following text to clarify this:

Lines 215-6: ‘For these calculations, an average campaign sunrise time of 07:12 and sunset of 17:53 were used to split the data into daytime and night-time values.’

However, for the average night-time datapoints presented in Fig. 5, we include a buffer of 2 hrs at each side of the night to ensure there are no residual daytime influences on the average. In the caption for Fig. 5, we have already included the following text:

Line 393: ‘Data points from between 20:00 and 05:00 the following morning are included.’

We now feel this is also important to highlight in the main text, and have therefore included the following:

Lines 339-41: ‘(In this analysis, night-time averages are taken between 20:00 and 05:00 the following morning, in order to prevent residual influence from daytime processes.)’

5. In Section 2.1, the name of the centre is incorrectly written. I believe the correct name of the centre is the same as the affiliation of one of the authors of the study. Also, are the authors sure that the distance from the major roadway is just 80m? Please also include which floor the measurements were conducted in.

We thank the reviewer for spotting this. We have now corrected the name of the centre in line 79.

The IIT-Delhi campus lies on the outer ring-road (Gamal Abdel Nasser Marg, here), so it is immediately north of the measurement site – about 80 m according to Google maps. The slightly larger inner ring-road (Mahatma Gandhi Rd) is 2.4 km to the north. We have chosen to reference the slightly smaller, outer ring-road, here, which is close to the measurement site. It is still a large, busy road and, due to its proximity, we consider that it is more likely to have direct impacts on our measurements in this case.

Measurements were conducted in the fourth floor; this is already stated in line 81.

6. In Section 2.3, line 151, the measurement location for meteorological data should be included.

Meteorological parameters were supplied from the measurement station at the airport, 8 km to the west of the measurement site. This information has now been included in lines 153-4.

7. In Figure 2, adding MLH from reanalysis dataset such as ERA5 or MERRA2 may provide interesting insights.

The planetary layer boundary height has now been included in Fig. 2, and a description of the origin of the data added to the method section.

Lines 186-8: 'The planetary boundary layer height (PBLH) displayed in Fig. 2 and the friction velocity (*U**) displayed in Fig. S5 were obtained from the Real-time Environmental Applications and Display sYstem (READY; Rolph et al., 2017) website, and were available at 3 h resolution.'

Thanks to the authors for writing an interesting atmospheric chemistry manuscript. I hope that the comments above help improve the article.

References

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