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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Effect of changing $NO_x$ lifetime on the seasonality and long-term trends of satellite-observed tropospheric $NO_2$ columns over China" *by* Viral Shah et al.

**Anonymous Referee #3**

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The study examines and compares the trends of spaceborne NO2 columns and bottom-up NOx emissions (MEIC) over Eastern China. The observed NO2 trends between 2012 and 2017 appear consistent with the emission trends in summer as well as in winter, when taking into account changes in the NOx lifetimes during that period. GEOS-Chem simulations for the years 2012 and 2017 indicate very little change in the summertime NOx lifetime, due to the compensation of higher daytime OH and lower nighttime aerosol loss; in winter, however, the increasing trend in nighttime ozone appears to drive a decreasing trend of the NOx lifetime, due to the large share of the overall NOx sink being due to N2O5 hydrolysis on aerosols. These model findings appear consistent with the comparison of MEIC emissions and OMI columns.



The topic of the paper is very significant since the community and policy-makers will be interested by further information on NOx emission trends and how they relate to NO2 data. The paper is clear and well written. However, I am not entirely convinced by the robustness of the conclusions.

Major comments

For one thing, uncertainties should be better acknowledged and (wherever possible) evaluated. Given the importance of the aerosol sink in the discussion of the trends, sensitivity studies are necessary to demonstrate that the conclusion holds despite uncertainties in aerosol surface densities and especially in the N2O5 uptake coefficient. I understand that the NO2+O3 reaction might be the main limiting factor to N2O5 loss, at least during the winter, but it might not be the case during summer, and in any case it requires more than just qualitative arguments. It is worth noting that campaign data (e.g. Brown et al., JGR 114, D00F10, doi:10.1029/2008JD011679, 2009) suggested much lower N2O5 uptake coefficients than those (Bertram and Thornton) used in GEOS-Chem.

A second major comment is related to the calculation of NOx lifetimes (Figure 3), which are averages for the bottom 0-2 km of the atmosphere. I assume that the values given (about 6 hours in the summer and 21-27 hours in winter) are 24-hour averages. Two issues arise: 1) in summer, the relevant lifetime for OMI NO2 columns is the average NOx lifetime during the few ($\sim$6) hours preceding the satellite overpass time (13:30 LT); and 2) the vertical profile of the OMI sensor sentivity should be taken into account. I know that at least 70% of the columns lies below 2 km altitude, but the sensitivity profile is very steep and very anti-correlated with the NO2 vmr profile, even below 2 km. Furthermore, the part of the column lying above 2 km altitude is non-negligible and might have very different trends from the lowermost part. I suspect that taking these effects into account will increase the relative importance of NO2+OH to the total loss, with possibly significant consequences for the trend and for the seasonal evolution.

Minor comments:

- line 112 how important is the correction using GEOS-Chem simulated concentrations of HNO3 and organic nitrates?

- why not use the ground-based NO2 measurements to evaluate the trends?

- l. 135 the NO2 hydrolysis coefficient is said to be decreased from $10^{-4}$ to $10^{-5}$. Relative to what study or what model version? Is this value used for all RH and all aerosol types? Jaeglé et al. (JGR 2018) use $10^{-4}$ and assume that the reaction makes only HONO (i.e. no HNO3), which would lead to very high HONO/NO2 ratios over China. A comment on this would be appreciated.

- what is the model performance for aerosol surface density over China?

- l. 136-138 This gives the impression that the well-known issue of the HONO missing sources has been solved, which is not the case, since many other sources have been proposed to explain the observations. What is the contribution of NO2 hydrolysis to the NOx sink in the simulations?

l. 172-174 The NO2 column measurement and the surface NO2 measurement represent different times, therefore the similar winter-summer ratios are not necessarily expected.

l. 186 Regarding the aerosol loss, a discussion of the uptake coefficient is required (see above, major comment)

l. 187-190 and Figure 4: not useful for the discussion, could possibly be dropped.

- Figure 1: I suggest to indicate on each plot, the domain-averaged NO2 column

Technical comments:

- the reference Manders et al. (2017) is missing

- the reference Bertram and Thornton (2009) is missing



- references: use journal abbreviations

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