

The questions and comments from reviewers are in black, the responses are in black italics and the lines included in the main text are in blue.

Reviewer 2

General comment This study explores the diurnal variation of total NO₂ columns and surface NO₂ concentrations using GCHP model simulations with independent NO₂ measurements from DISCOVER-AQ campaign and Pandonia Global Network. Two corrections are applied on PGN/Pandora total NO₂ columns to better represent the dependence of NO₂ cross section on the temperature, and different local solar time along the PGN/Pandora line-of-sight. Besides, the authors also test the influences of horizontal resolution and planetary boundary layer height (PBLH) modification on the model performance against aircraft and ground-based observations. It is demonstrated that compared with the other two sensitivity runs, fine scale (~12km) modelled NO₂ columns with PBLH modification show smaller bias to independent measurements and better agreement in terms of NO₂ diurnal variation. Based on model simulations, the authors find that NO₂ columns below 500m show much stronger diurnal variation that that of total columns, which is dampened by residual columns above with much weaker variability. The findings of this study are important for understanding the relationship between NO₂ columns and surface concentrations, and I recommend it to be published after addressing following issues.

Response: We thank the reviewer for acknowledging the importance of this work. Specific responses are listed below:

Specific comments

Line 39: what is “connected layers”?

Ans: We have modified it to –

“the differences in hourly variation of atmospheric layers”

Line 200-203: please re-write this sentence and explain the meaning of each term in this equation.

Ans: Corrected. Lines 198-202:

To account for the hourly changes in vertical variation of column temperature, we calculate simulated NO₂ effective temperatures T_{eff} using the site-specific hourly GEOS-FP temperature profiles $(T)_i$, NO₂ cross section $\sigma(NO_2)_i$, and GCHP NO₂ vertical profiles $VC(NO_2)_i$ following equation (1) of Herman et al. (2009):

$$T_{eff} = \frac{\sum_i^N(\sigma(NO_2)_i \cdot VC(NO_2)_i \cdot (T)_i)}{\sum_i^N(\sigma(NO_2)_i \cdot VC(NO_2)_i)} \quad (1)$$

Line 241: what do you mean by “left panel” and in which figure?

Ans: Noted and corrected. We deleted the redundant "(left panel)" in Line 241.

Line 254-255: why the simulated effective temperature is lowest in the early afternoon? Line 258-260: please explain the scientific meaning of “0.2” and “(294-220)” in the equation.

Ans: We explain the effective temperature as a function of hourly variation of GHCP simulated total NO₂ columns, which show lower concentrations during the early afternoon and increases during the evening. We modified the main text for better clarification.

Lines 262 – 265:

The GHCP simulated effective temperature is also warmer for Asian sites, however the effective temperature is lower during the early afternoon when near-surface NO₂ concentrations tend to be minimum such that the stratospheric NO₂ that makes a larger fractional contribution to the total column.

We clarified the scaling factors in lines 267-274:

The corresponding correction factor (CF) for hourly variation in the effective temperature is calculated as:

$$CF = 1 + \left(\frac{1}{0.8} - 1 \right) \times \frac{(T_{eff}(GHCP(hour)) - T_{eff}(Pandora(hour)))}{294 - 220} \quad (3)$$

The factor of $\left(\frac{1}{0.8} - 1 \right)$ reflects the difference between the NO₂ columns fitted with a 220 K NO₂ spectrum that are about 80% of those fitted with a 294 K NO₂ spectrum.

Line 289: it seems that PBLH modification has larger impact on simulated NO₂ columns in the morning and evening than midday. What is the reason for this?

Ans: The PBLH modification reduces the NB by 3-4% for total NO₂ vertical columns averaged between 9 AM- 6 PM local solar time across all sites. The difference between the impact of PBLH modification on morning/evening columns as compared to midday columns is about 2-3%. This small difference is primarily driven by the increment in NO₂/NO_x ratios in the total column is stronger during the morning and evening, which is further enhanced by the PBLH modification as observed in Figure A6.

Line 292: in Figure 4 and 5, both corrected PGN/Pandora NO₂ columns and aircraft partial NO₂ columns present a distinct increase in early morning, which is also found in GEMS NO₂ observations. However, this feature is not reproduced in modelled NO₂ columns even though NO_x emissions have a morning peak around 9:00 a.m. local time. What is the explanation for this discrepancy?

Ans: The simulated NO₂ columns show a peak at 9 AM local time (capturing the morning peak in NO_x emissions) and then consistently reduce till noon. The Pandora and aircraft columns show a late increase (between 10-11 AM local time) that is not well captured by the model. This difference is most likely driven by two factors –

1. *Even at 12 km, the representation of NO_x emissions peak in the morning could be diluted especially in developed regions where NO_x has been massively reduced so that background NO₂ is significant. In Asian sites the model capture the increase because the NO_x emissions there are still very strong and distinguished from background NO₂.*
2. *The use of coarse meteorological fields.*

Although we use fine scale emissions and meteorology, we recognize the importance of more detailed emissions and metrological fields. We add lines 423 -438 for better clarification –

“Despite the skill of the 12 km simulations in representing the Pandora column measurements, there appears to be greater hourly variation in the simulation, the aircraft measurements, and the surface measurements than in the Pandora observations. Future work should continue to understand this relationship. Future work should also leverage the information developed here to test the performance of surface NO₂ concentrations inferred from the geostationary constellation against ground-based measurements.”

Technical comments

Line 31-35: please simplify this sentence.

Ans: Done.

Line 36: change “column” to “columns”.

Ans: Corrected.

Line 70: please expand the abbreviation “KORUS-AQ” when it appears for the first time. Line 85: please expand the abbreviation “CTMs” when it appears for the first time.

Ans: Corrected.

Line 97-102: please combine these two sentences to make it less redundant.

Ans: Corrected.

Line 108: please expand the abbreviation “CONUS” when it appears for the first time. Line 136-137: please re-write this sentence.

Ans: Corrected.

Line 143: please keep consistent expression of longitude (sign) in Tables A1 and A2.

Ans: Corrected.

Line 198: change “identifies” to “identified”.

Ans: Corrected.

Line 315: change “3.3” to “3.4”.

Ans: Corrected.

Line 340: change “3.4” to “3.5”.

Ans: Corrected.

Line 355: change “3.5” to “3.6”.

Ans: Corrected.

Line 397: remove the comma.

Ans: Corrected.