

We thank the reviewers for their thorough evaluation and suggestions. We have carefully addressed the comments below.

RC1

Major Comments:

1. The manuscript analyses trends in major air pollution in Korea using multi-source data, including satellites, ground-based observations, and emission inventories..., but there are significant differences in trends between the multi-source data that need to be clarified.

(a) Authors claim CO trend observed by MOPITT decreased slower than surface concentrations because of the background contribution to the CO VCD (Line155-156), why there is a consistent downward trend in MOPITT and surface concentrations in the period 2015-2018 and a huge difference in their downward trends in 2019-2023, it is clear that there is more than just the effect of background concentrations here.

We do not consider the difference to be huge, and its cause is not clear so we would rather not discuss it. We specified the background CO for clarity (Lines 170-172).

“MOPITT decreases at a rate of $-0.9 \pm 0.5\% a^{-1}$, slower than surface concentrations because of the background contribution to the VCDs ($\sim 2 \times 10^{18}$ molecules cm^{-2}).”

(b) Surface SO₂ concentrations and OMI VCDs have decreased at similar rates but there are differences (Line173), For example, SO₂ observed by OMI rises significantly in 2019-2020, and SO₂ observed by both OMI and GEMS rises in 2022-2023, whereas CAPSS and AirKorea only show a downward trend, and these details should be clarified.

The retrieval uncertainties of SO₂ from OMI and GEMS are large, therefore interannual variations appear to be associated with random or systematic errors during the retrieval. We clarified in the text as follows (Lines 188-189):

“There is large uncertainty in the satellite observations that likely contributes noise to the trend (J. Kim et al., 2020; C. Li et al., 2020).”

(c) I really don't understand why the diurnal variation of NO₂ VCD observed by GEMS in warm season (8-11:00 local time) and cold season (10-13:00 local time) is opposite to that of surface NO₂ (Figure 4e). The authors try to explain this phenomenon by using the variation of the mixed layer height, it is insufficient. Besides, the high NO₂ concentration in the morning and evening is affected by meteorological conditions. Vehicle emissions during the morning and evening rush hours are also an important factor. NO₂ is mainly concentrated near the surface and rapidly photolysis after sunrise, and the satellite and the surface observations should show similar diurnal trends, which can be confirmed by previous observations in some mega-cities (Tian et al., 2018) and background stations (Cheng et al., 2019). What's more, an observation from the GEMS also showed that NO₂ column concentrations began to decline at 10:00 (local time) (Xu et al., 2023). I recommend first comparing the GEMS and surface NO₂ concentrations on an hour-by-hour basis, and then carefully analysing the reasons for the opposite trend.

We mainly attribute the surface diurnal variability to the mixed layer height growth because traffic load in Seoul has weak daytime variability resulting in relatively flat emissions instead of a bimodal behavior (Yang et al., 2024). As pointed out, the photochemical sink plays a role in the daytime minimum, so we revised the explanation as follows (Lines 218-229):

“NO_x emissions in the SMA have small seasonal variations as they are dominated by mobile sources (Pandey et al., 2008; H. M. Lee and R. Park, 2022; Yang et al., 2024). The emissions are higher in the daytime (7–18 LT) than at night but do not show significant rush hour enhancements because traffic load is sustained with little variability throughout the daytime (Yang et al., 2024). Therefore, the peak in surface NO₂ concentrations at 8–9 LT is not due to the rush hour but to accumulation of daytime emissions in a shallow mixed layer (Moutinho et al., 2020). NO₂ then decreases in the morning by dilution as the mixed layer grows from solar heating, and increases again in the evening when the mixed layer collapses (J. Li et al., 2021). Increasing NO₂ photolysis as the morning progresses would also be expected to lower NO₂ concentrations but this is offset by entrainment of O₃ from aloft as the mixed layer grows, such that the NO₂/NO_x ratio increases during the morning hours (Yang et al., 2024).”

We find an 11 LT peak and subsequent decline in NO₂ VCDs during Apr-Sep, which is consistent with the 10-11 LT peak in Seoul during May-Sep from Xu et al. (2024). However, as pointed out by Xu et al. (2024) and shown in the comparison across major Chinese cities by Tian et al. (2018), diurnal patterns of VCDs vary depending on emission characteristics (energy consumption and transportation). There is also a clear difference in background regions as shown by Cheng et al. (2019), where the VCDs do not show morning accumulation of emissions but only a 11-14 LT minimum caused by the dominance of chemical loss and ventilation. We elaborated in more detail as follows (Lines 229-234):

“Geostationary satellite observations provide unique information on the diurnal variation of NO₂ VCDs (Tian et al., 2018; Cheng et al., 2019; Edwards et al., 2024; Xu et al., 2024). This is illustrated in Figure 4e for the SMA. A NO_x budget analysis by Yang et al. (2024) shows that NO₂ VCDs in Seoul increase steadily in the morning from accumulation of emissions as they are not affected by mixed layer growth, reaching a steady state in the afternoon due mostly to loss from ventilation.”

2. Line288-289 “Based on the criteria from Duncan et al. (2010) the positive trend in RFN implies that Korea is now mostly in the NO_x-sensitive regime (RFN > 2).” In order to avoid the misjudgment of O₃ formation sensitivity caused by arbitrary selection of FNR thresholds, I strongly suggest using a third-order polynomial model to investigate the empirical relationship between FNR and surface O₃ concentrations, which has been widely used in other studies (Ren et al., 2022; Jin et al., 2020). The criteria presented in Duncan et al. (2010) may not be applicable to the current diagnosis of O₃ formation sensitivity, the threshold is usually small (1 and 2), which causes the contribution of the NO_x limit regime to be overestimated.

We agree that the R_{FN} threshold may vary with time and region. Recent analyses by the authors of Jin et al. (2020) used GEOS-Chem to derive region-specific thresholds across the globe and applied the criteria to GEMS observations (Jin et al., 2024). Their results imply that $R_{FN} > 2\sim 3$ is a reasonable threshold applicable to South Korea, similar to the results over China (2.2-3.2) by Ren et al. (2022). We replotted Figure 7 using a threshold of 2.5 and revised the manuscript as follows (Lines 309-314):

“Recent studies over Northeast Asia suggest that NO_x-limited regimes are found where $R_{FN} > 2\sim 3$ (Ren et al., 2022; Jin et al., 2020, 2024). Here we use 2.5 as a threshold and find that Korea is now mostly in the NO_x-limited regime. Figure 7d–e shows May–June 2023 MDA8 O₃ and its sensitivity regimes inferred from GEMS R_{FN} . Most of the country is in a NO_x-limited and transition regimes while VOC-limited conditions are largely limited to the central SMA and Busan.”

Reference: Jin X., Yang, Y., and Wang, S.: Observing the diurnal cycle of ozone-NO_x-VOC sensitivity from geostationary satellite retrievals of ozone precursors, Abstract A21I-1872 presented at AGU24, Washington DC, 9-13 December, 2024

Minor Comments:

1. Line 57-58 “Synoptic meteorology and transport from China also contribute to seasonal and long-term variations of pollutants over Korea.” Missing relevant references.

Revised as follows:

“Synoptic meteorology and transport from China also contribute to seasonal and long-term variations of pollutants over Korea (H. M. Lee and R. Park, 2022; D. Park et al., 2021; J. Jeong et al., 2024).”

2. Line 204-205 “Both surface and column NO₂ are higher by a factor of two during the cold season, which can be explained by the longer NO_x lifetime (Shah et al., 2020).” Differences in warm- and cold-season emission patterns should have a greater impact.

Several studies confirm that NO_x emissions in the SMA are mainly from mobile sources which are persistent throughout the year, with small seasonal variations. We clarified this as follows:

“NO_x emissions in the SMA have small seasonal variations as they are dominated by mobile sources (Pandey et al., 2008; H. M. Lee and R. Park, 2022; Yang et al., 2024).”

3. Line 242-243 “but CHOCHO shows hotspots for manufacturing industries while HCHO shows hotspots for petrochemical facilities.” Unclear HCHO shows hotspots for petrochemical facilities, since HCHO observations are also more distributed, HCHO didn't just indicate petrochemical facilities.

We notice a distinct contrast in the CHOCHO and HCHO hotspots over industrial regions with different facilities and clarified as follows:

“CHOCHO shows hotspots for manufacturing industries (Incheon, Changwon) while HCHO shows hotspots for petrochemical facilities (Yeosu, Ulsan).”

4. Line 262-263 “has been previously reported as systematic low biases in satellite observations of CHOCHO and HCHO.” Please specify it.

Revised as follows:

“We find that the GEMS columns are lower than the aircraft columns, consistent with previously reported low biases in satellite retrievals of CHOCHO (−50%) and HCHO (−40% to −20%) (Chan Miller et al., 2017; Zhu et al., 2016; Zhu et al., 2020).”

5. GEMS is observed every hour during the day and the time should be clarified. For example, in Fig. 3d, does GEMS use all the observations during the day or just a certain hour of the mid-day.

We included detailed descriptions on satellite overpass times and how GEMS is sampled in the text (Lines 125-131) and figures.

“OMI and TROPOMI make afternoon overpasses at 13:30 local time (LT). We make use of morning overpasses for MOPITT (10:30 LT) and IASI (9:30 LT). We use hourly daytime observations from GEMS (7:45–16:45 LT), GOCI (9:30–16:30 LT), and GOCI-II (8:15–17:15 LT). For annual trend analyses we use GEMS observations made between 12–14 LT for consistency with the overpass time of OMI and TROPOMI measuring the same gases. We find no significant differences in observed trends when using surface observations sampled at satellite overpass times and therefore use all hours of the day.”

6. Figure 5g “OMI CHOCHO¹²⁰”, Does it mean 20 times magnification? This should be clarified in the legend.

We clarified this in the figure caption.

Suggestion:

Although well known, some instrument name abbreviations should indicate the full name when they first appear, i.e. OMI, TROPOMI, MOPITT...

We spelled out full names in the introduction as suggested.