Interpreting GEMS geostationary satellite observations of the diurnal cycle of nitrogen dioxide (NO_2) over East Asia

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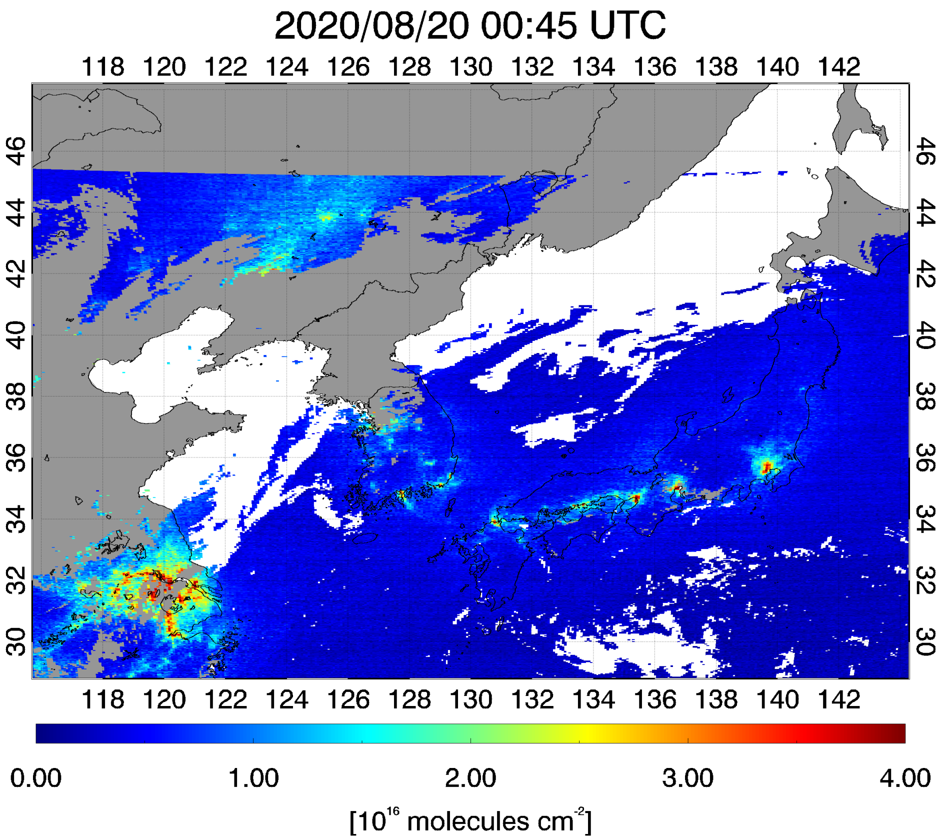
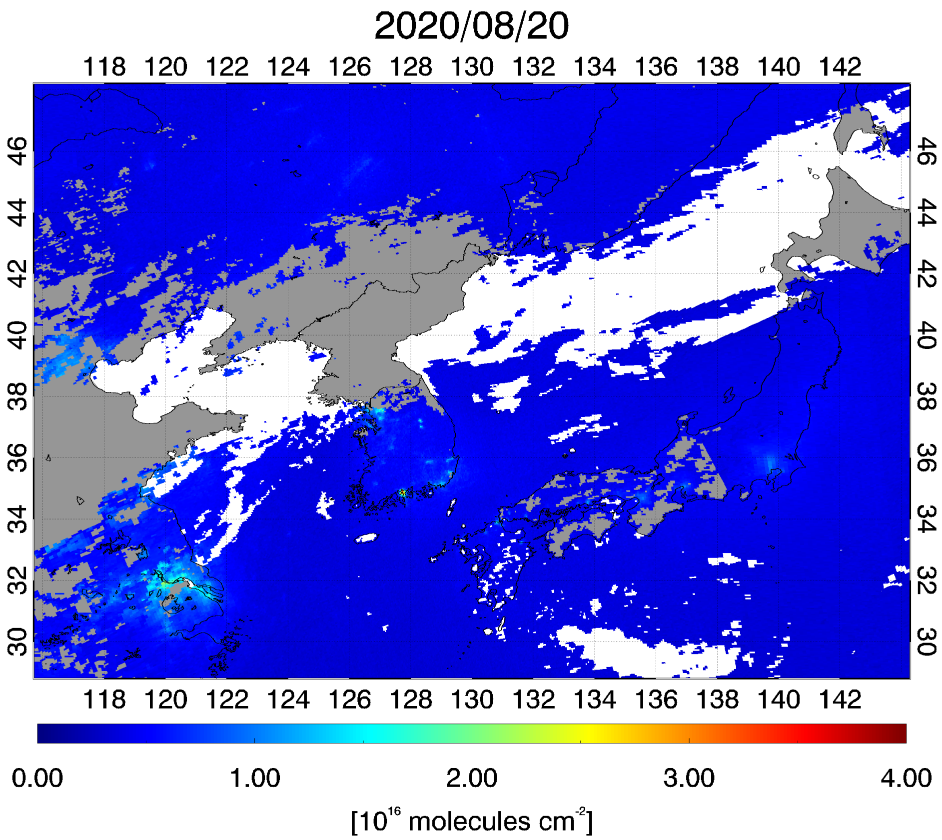
December 5th, 2023

NO_x emissions, transport, and chemistry can be better characterized with hourly geostationary observations



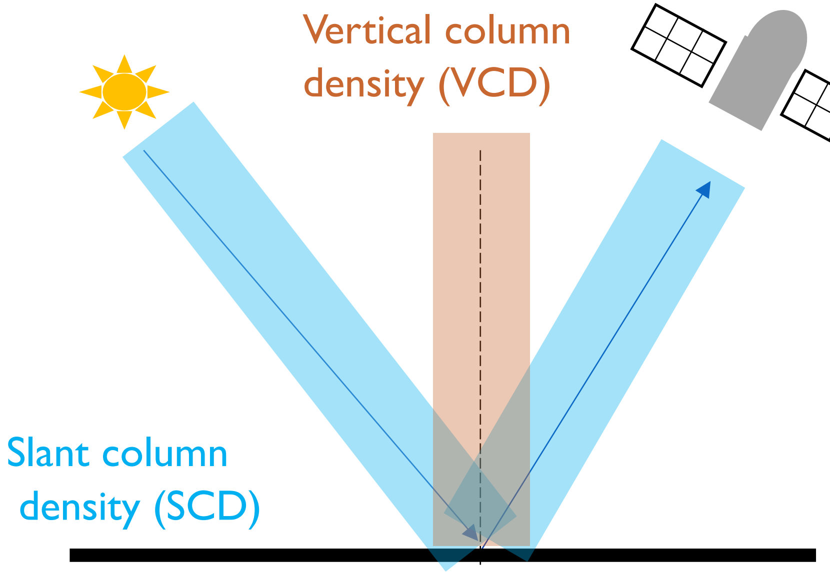
Geostationary satellite
GEMS



Low-earth orbiting satellite
TROPOMI

[Courtesy of Hanlim Lee (PKNU)]

Air mass factor allows us to convert from slant column density to vertical column density and varies diurnally



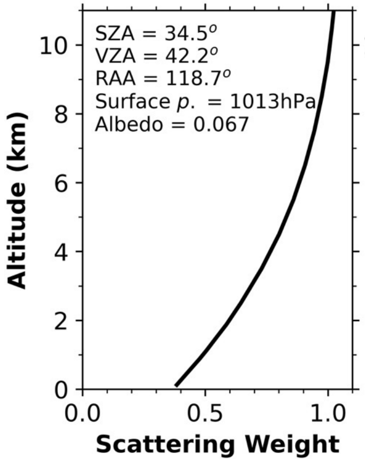
$$\mathrm{VCD} = \frac{\mathrm{SCD}}{\mathrm{AMF}}$$

$$\mathrm{AMF} = \mathrm{AMF_G} \int_0^{z_T} w(z) S(z)\, dz$$

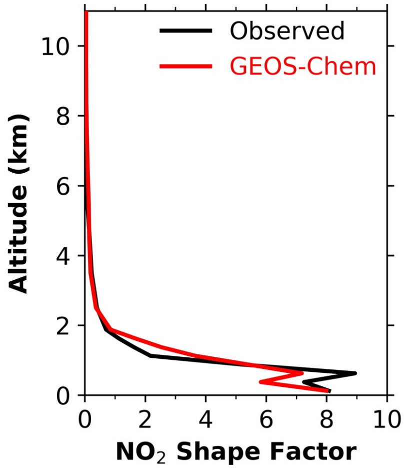
Geometric correction

Scattering weight

Vertical shape factor



From radiative transfer model



From chemical transport model

Yang et al. (2023) used KORUS-AQ aircraft campaign to evaluate GEOS-Chem's ability to provide accurate shape factor and thus AMF

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Tropospheric NO_2 vertical profiles over South Korea and their relation to oxidant chemistry: implications for geostationary satellite retrievals and the observation of NO_2 diurnal variation from space

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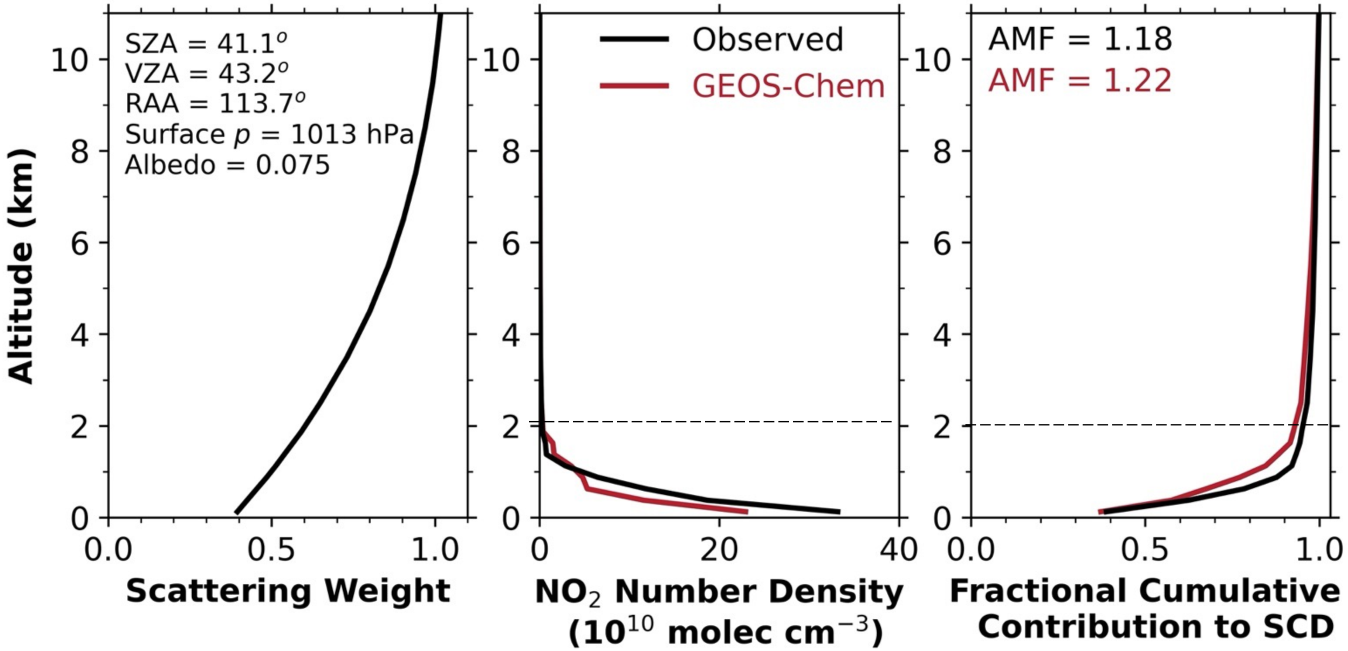
Over South Korea, NO_2 columns are mainly (80%) contained within the planetary boundary layer (PBL; $z \leq 2$ km)

In the Seoul Metropolitan Area, 95% of NO_2 column is contained within the PBL

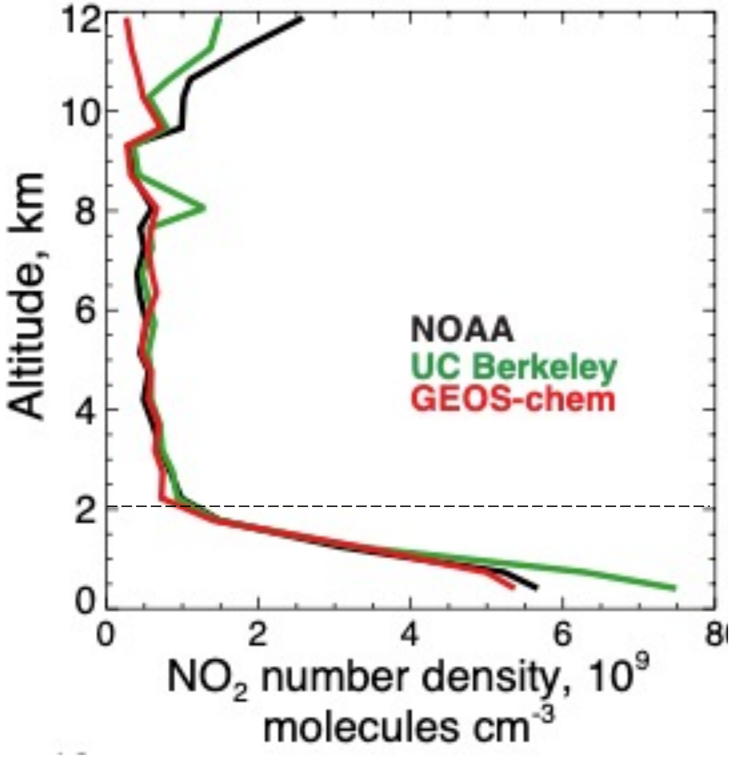


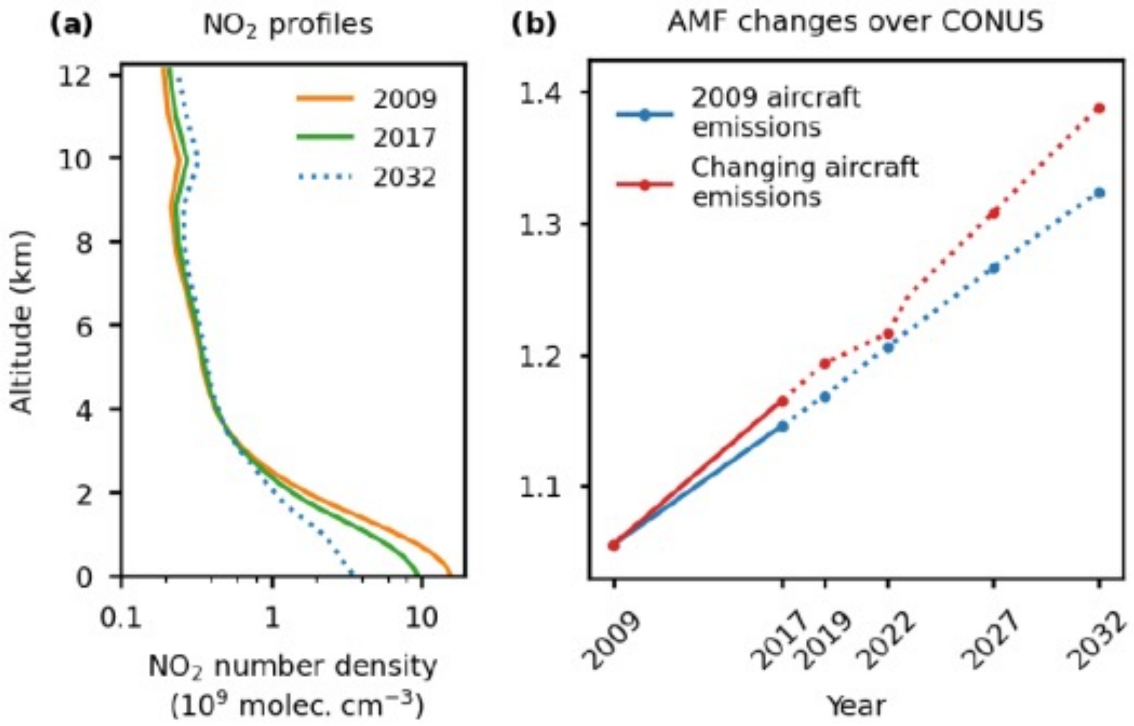
Reflects highly polluted condition

SZA: Solar Zenith Angle
VZA: Viewing Zenith Angle
RAA: Relative Azimuth Angle
SCD: Slant Column Density (Same as SC)

Over the US, only 20-35% of NO_2 column is contained with the PBL

Southeast US
SEAC[4]RS NO_2 vertical profile



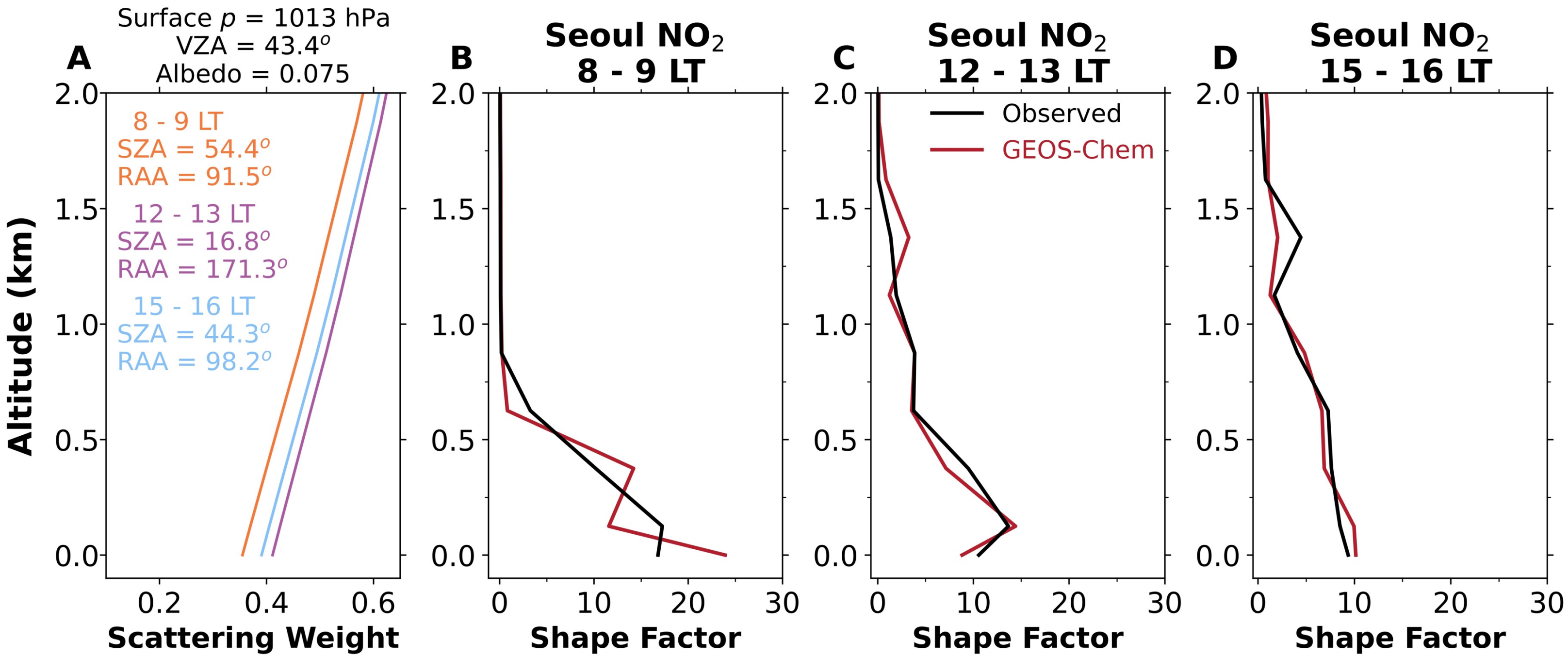


FT NO_x from lightning, fire, aircraft, organic nitrates, particulate nitrate

FT tropospheric background is rising in the US due to ↑ NO_x emission from aircraft and fires leading to ↑ in AMF

[Left figure from Travis et al., 2016; Right figure from Dang et al. 2023]

Diurnal variation in AMF is driven by mixed layer growth offset by the length of the light path

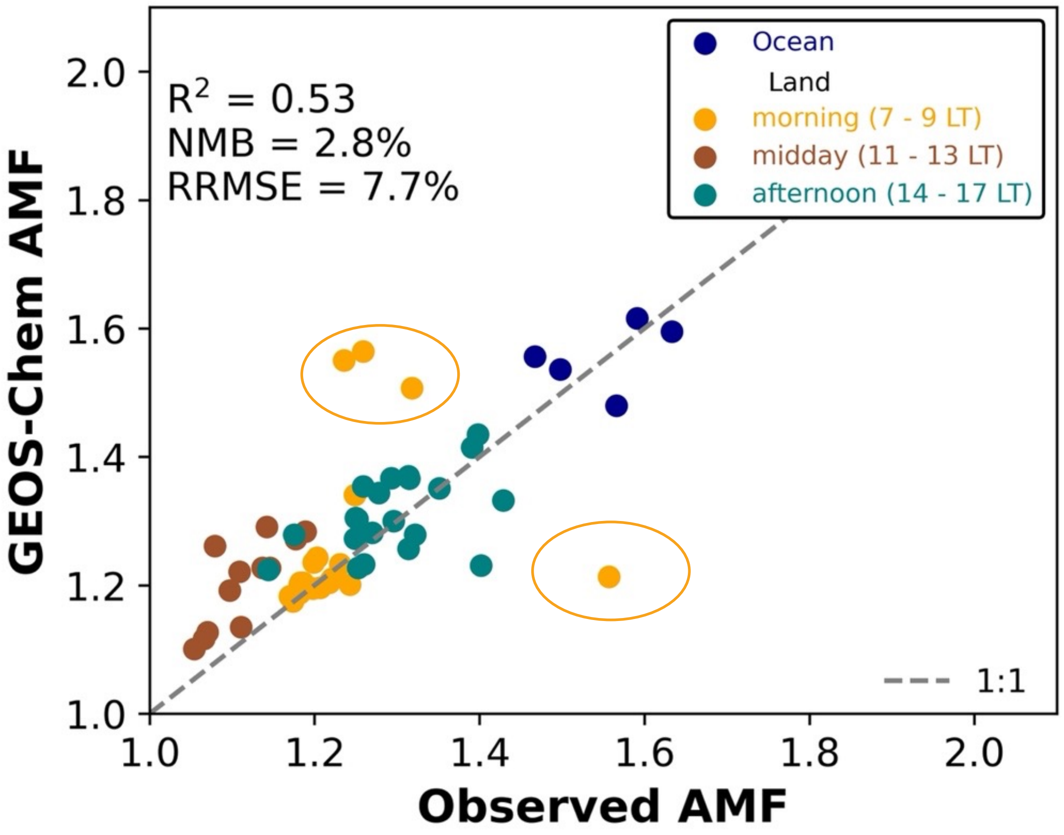


LT (hours)	AMF_G	$\int_0^{z_T} w(z)S(z)dz$	AMF
8-9	3.09	0.38	1.19
12-13	2.42	0.46	1.11
15-16	2.77	0.46	1.27

AMF varies 13% diurnally

- 18% dirunal variation in scattering correction factor driven by mixed layer growth
- Solar zenith effect (24%) offsets the scattering correction factor variation

GEOS-Chem can capture the variability of observed AMF



- 50% of variability in AMF mainly driven by ocean vs. land and time of the day over land

- Largest model errors in the time of morning mixed layer growth → difficult for the model to get it right

Diurnal variation of NO_2 column is driven by emission, chemistry and transport of NO_x column

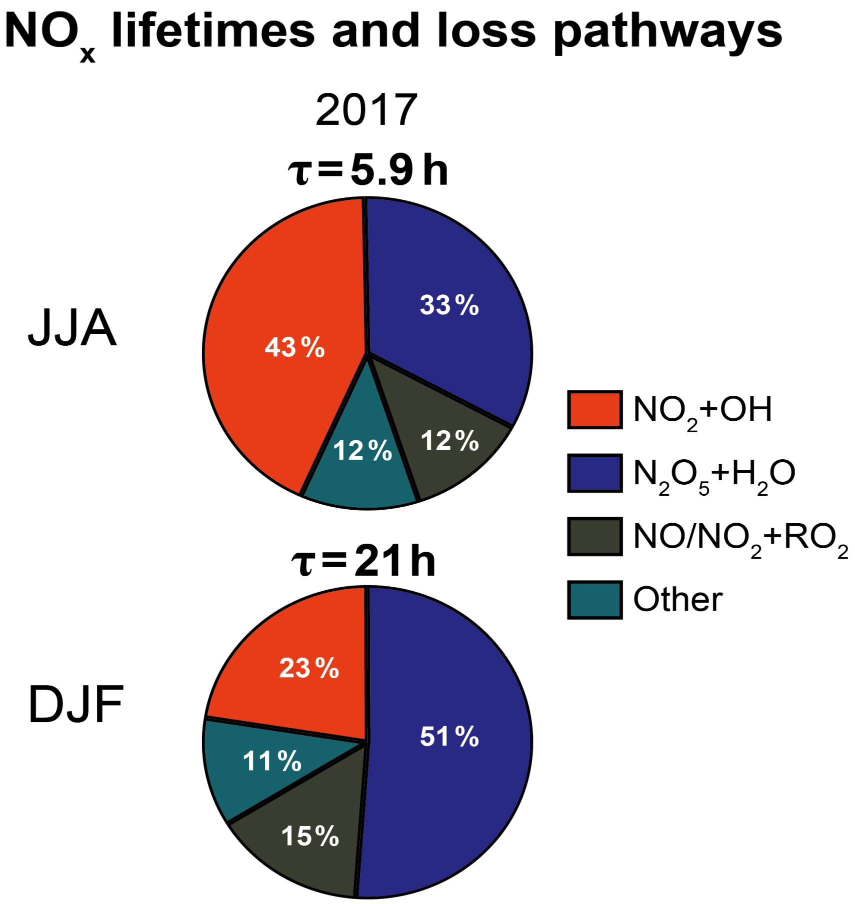
$$\boxed{\frac{\partial \Omega_{\mathrm{NO_2}}}{\partial t} = \alpha(t)\left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{net}}}$$

where $\alpha(t) = \frac{\mathrm{NO_2}}{\mathrm{NO_x}}$

$$\left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{net}} = \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{emission}} + \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{chemistry}} + \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{transport}}$$

$NO_x = NO + NO_2 + NO_3 + 2N_2O_5 + HONO + HNO_4 + ClNO_2$

Differences in NO_x lifetime between different seasons can result in distinct diurnal variations in NO_2 column



- In winter, [OH] and [RO_2] radicals are lower leading to a longer lifetime of NO_x

- Using NO_2 diurnal variation:
  - Can we constrain chemistry in the summer?
  - Can we constrain emission in the winter?

Figure from Shah et al. (2020)

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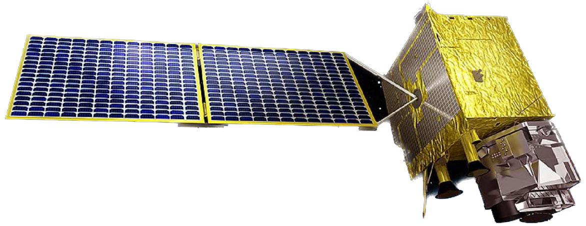
A chemical transport model (CTM) can elucidate the factors driving diurnal variation in retrieved and measured NO_2

GEMS
NO_2 column



Pandora
NO_2 column

Drivers of
diurnal variation in NO_2

Emission?
Chemistry?
Transport?

GEOS-Chem

Diurnal variation in
simulated NO_2

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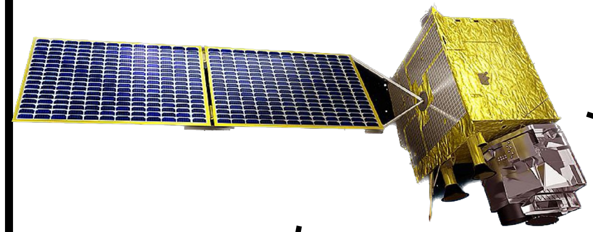
We recomputed our total column of GEMS using AMF computed from GEOS-Chem

Total slant column converted to total vertical column using GEOS-Chem AMF (GEMS L2 NO_2 v2 data)



Modified v13.3.4 (Yang et al. 2023) to better represent oxidant chemistry over East Asia

Pandora total NO_2 column with NO2 data with flag = 12 excluded (recommended by Crawford's group)

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Why did we use the total NO_2 column instead of the tropospheric NO_2 column?

- > 80% of NO_2 resides within the PBL over East Asia (Shah et al., 2020; Yang et al., 2023)
- Stratospheric NO_2 column has a minor diurnal variation (background) that can be well characterized by a CTM like GEOS-Chem

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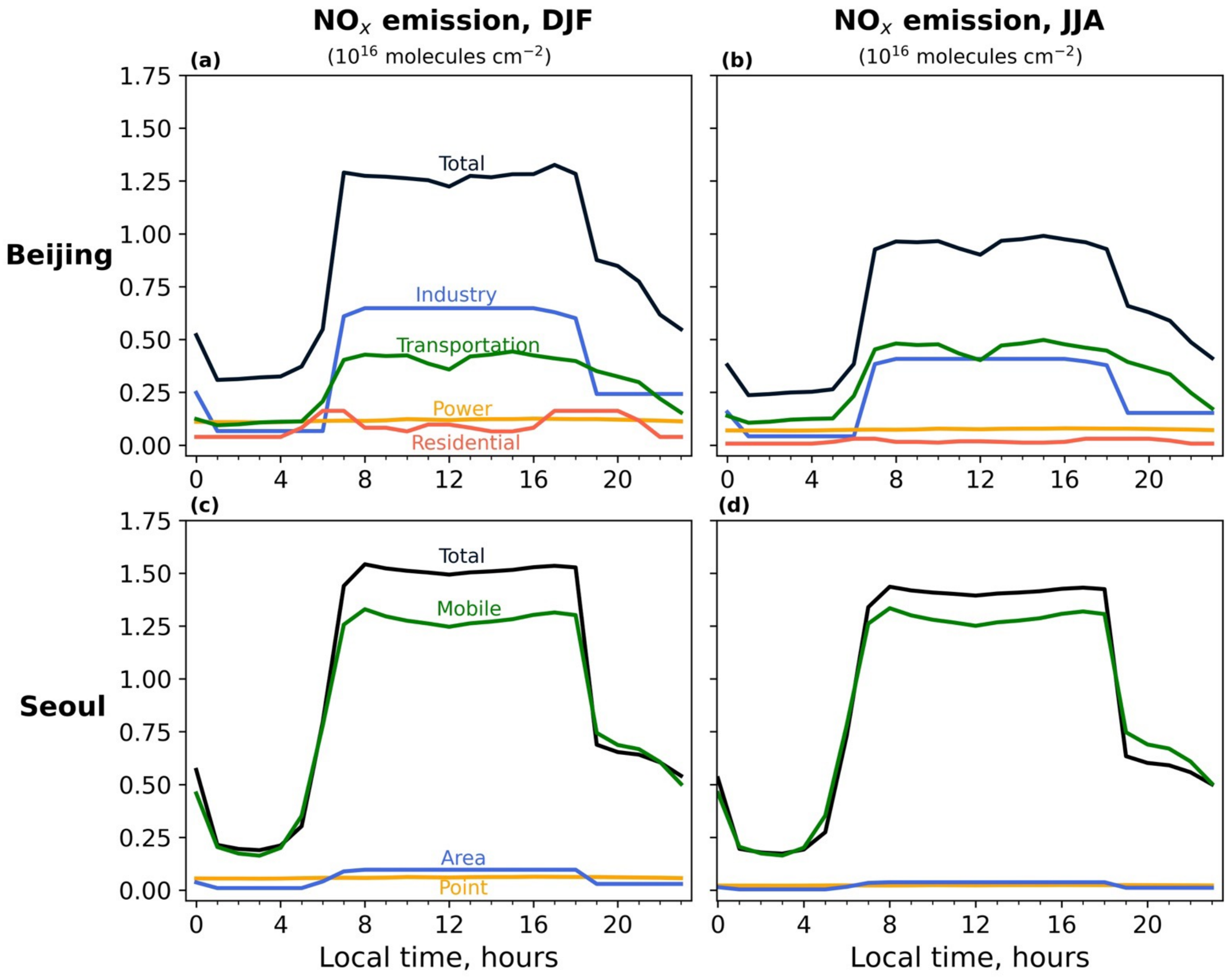
Can the same logic apply to the TEMPO NO_2 data?

- 25-35% NO_2 resides within the PBL over the U.S. (Travis et al., 2016)
- Background FT NO_x emission from aircraft, lightning, and fire plays an important role in NO_2 column in the U.S. (Dang et al., 2023)
- Artifact from incorrect strat.-trop. separation can induce an error

The main difference in diurnal variation of NO_x emissions occurs between day and night, which impacts the morning behavior of NO_2



- Beijing (MEIC): Liu et al. (2019) – power sector; Miao et al. (2020) for other sectors
- Seoul: (KORUSv5): Liu et al. (2019) – point source; adopted industrial sector's profile for area source; TOPIS transportation counts for mobile source

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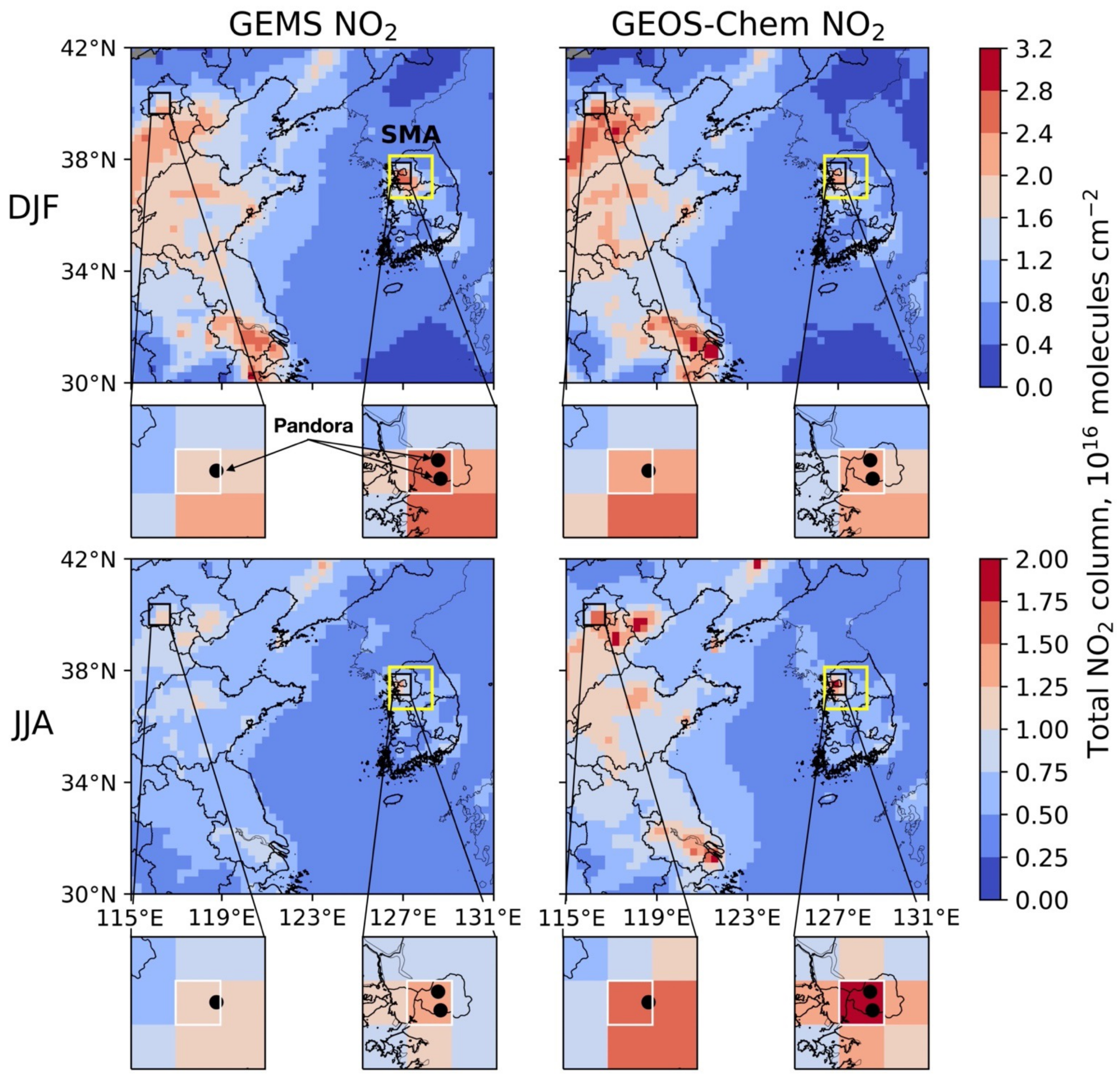
The transport effect is prominent during the wintertime, causing the NO_2 plume to shift to the south of the urban core



* GEMS data is regridded to GEOS-Chem 0.25° × 0.3125° grid boxes

- Yellow box: Regional analysis
- Black box: Beijing and Seoul
- White box: Urban analysis

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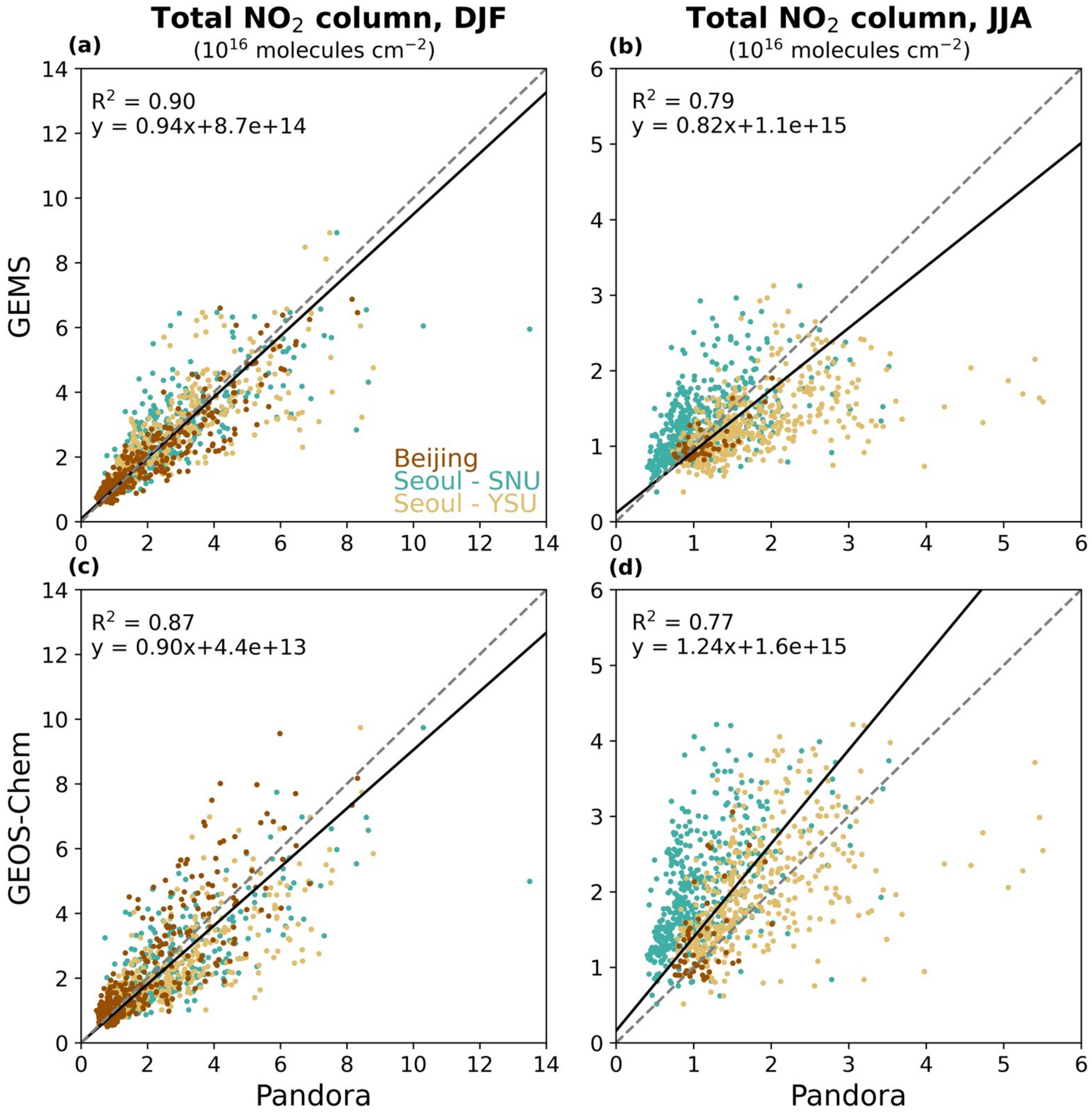
GEMS and GEOS-Chem reproduce the diurnal and day-to-day variability and magnitude observed by Pandora in winter and summer



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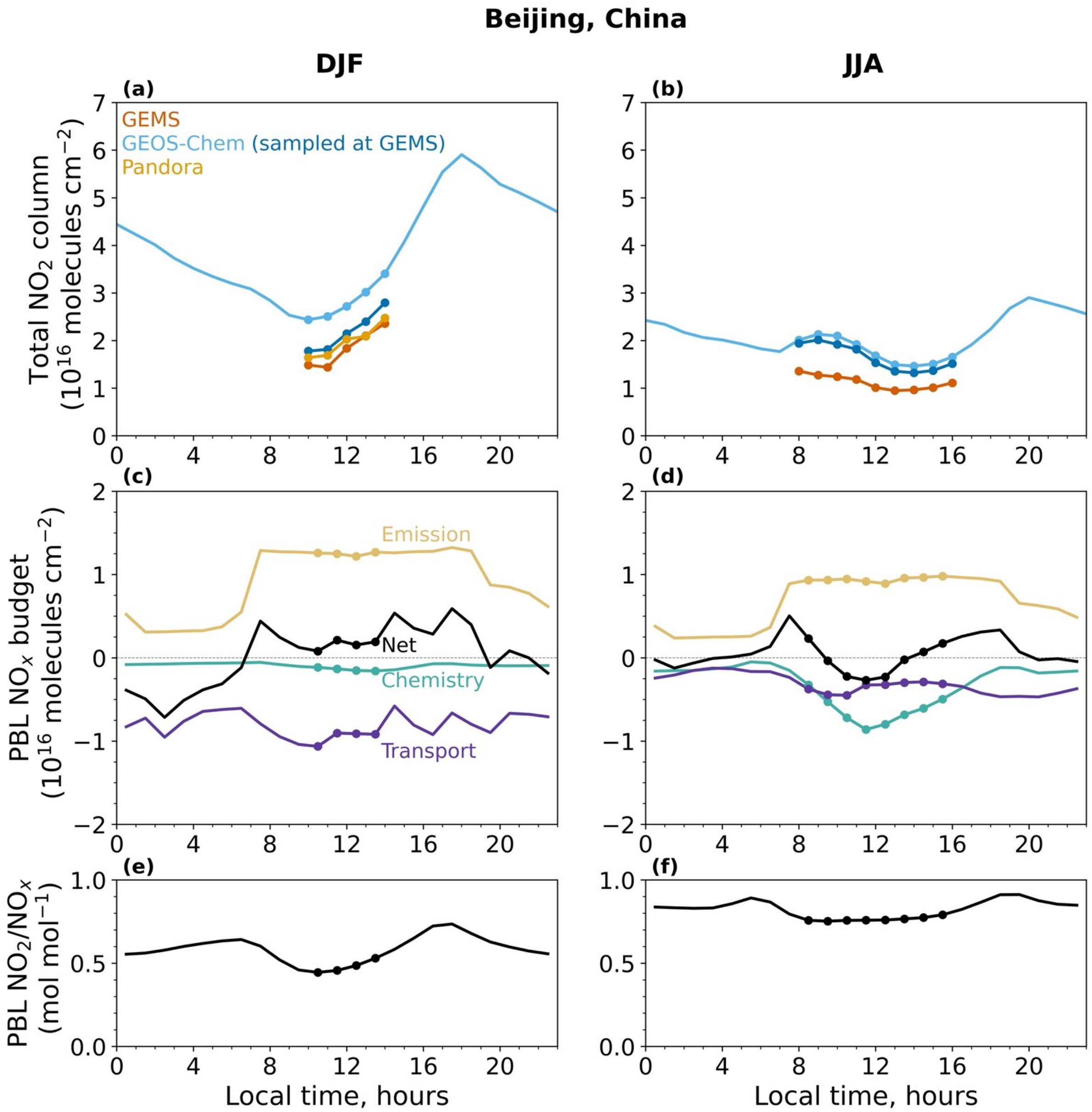
Emission dominates in winter, while the combined effects of chemical and transport loss terms dominate in summer in Beijing



$$\boxed{\frac{\partial \Omega_{\mathrm{NO_2}}}{\partial t} = \alpha(t)\left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{net}}}$$

where $\alpha(t) = \frac{\mathrm{NO_2}}{\mathrm{NO_x}}$

$$\left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{net}} = \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{emission}} + \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{chemistry}} + \left[\frac{\partial \Omega_{\mathrm{NO_x}}}{\partial t}\right]_{\mathrm{transport}}$$

NO_x = $\underbrace{\mathbf{NO + NO_2}}$ + NO_3 + $2N_2O_5$ + HONO + HNO_4 + $ClNO_2$

These two species dominate NO_x, and the interconversion reactions define $\alpha(t)$

$$\mathrm{NO + O_3 \rightarrow NO_2 + O_2} \quad \text{(R1)}$$

$$\mathrm{NO + HO_2 \rightarrow NO_2 + OH} \quad \text{(R2)}$$

$$\mathrm{NO_2} + h\nu \xrightarrow{+\mathrm{O_2}} \mathrm{NO + O_3} \quad \text{(R3)}$$

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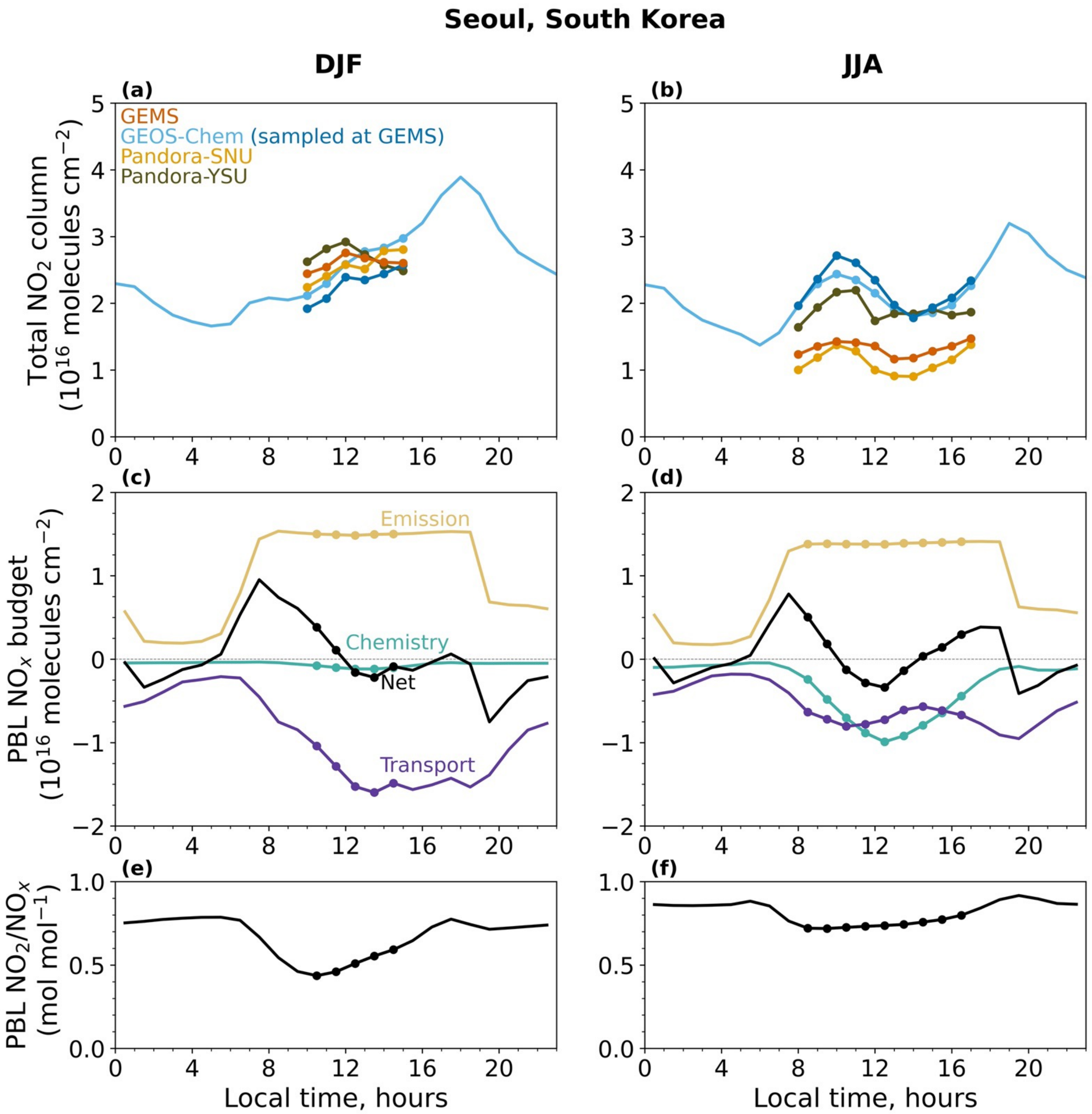
The diurnal variation of NO_2 columns in winter are inconsistent across datasets, and higher emissions affect the early morning in summer



- The NO_x budget in GEOS-Chem turns negative after noon due to stronger winds ventilation
  - This effect is compensated by increase in NO_2/NO_x ratio
- Seoul has higher summertime emissions than Beijing, leading to a net accumulation in NO_2 column in the early morning

Emission and chemical loss can be independently inferred from the GEMS observations if the role of transport can be quantified (Simple mass balance)

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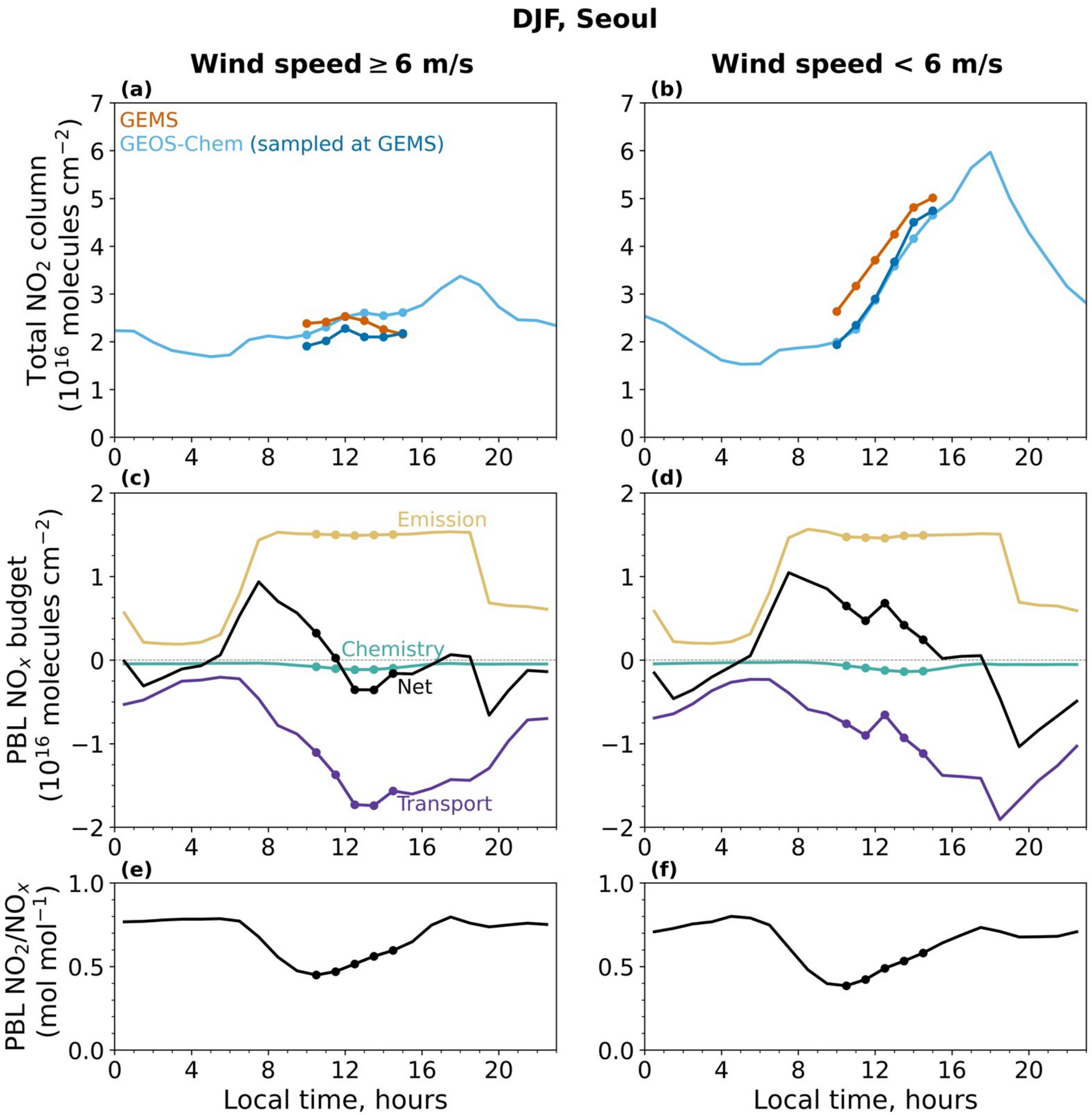
Segregating by data by hourly wind speed cannot fully eliminate the transport effect in winter



- **At high wind speed**:
  - Emission is balanced by transport leading to weak diurnal variation
- **At low wind speed:**
  - NO_2 accumulates as transport cannot keep up with emissions

Transport is not negligible even at low wind speed (3 m/s) since ventilation timescale for 25-km domain is only 2 hours

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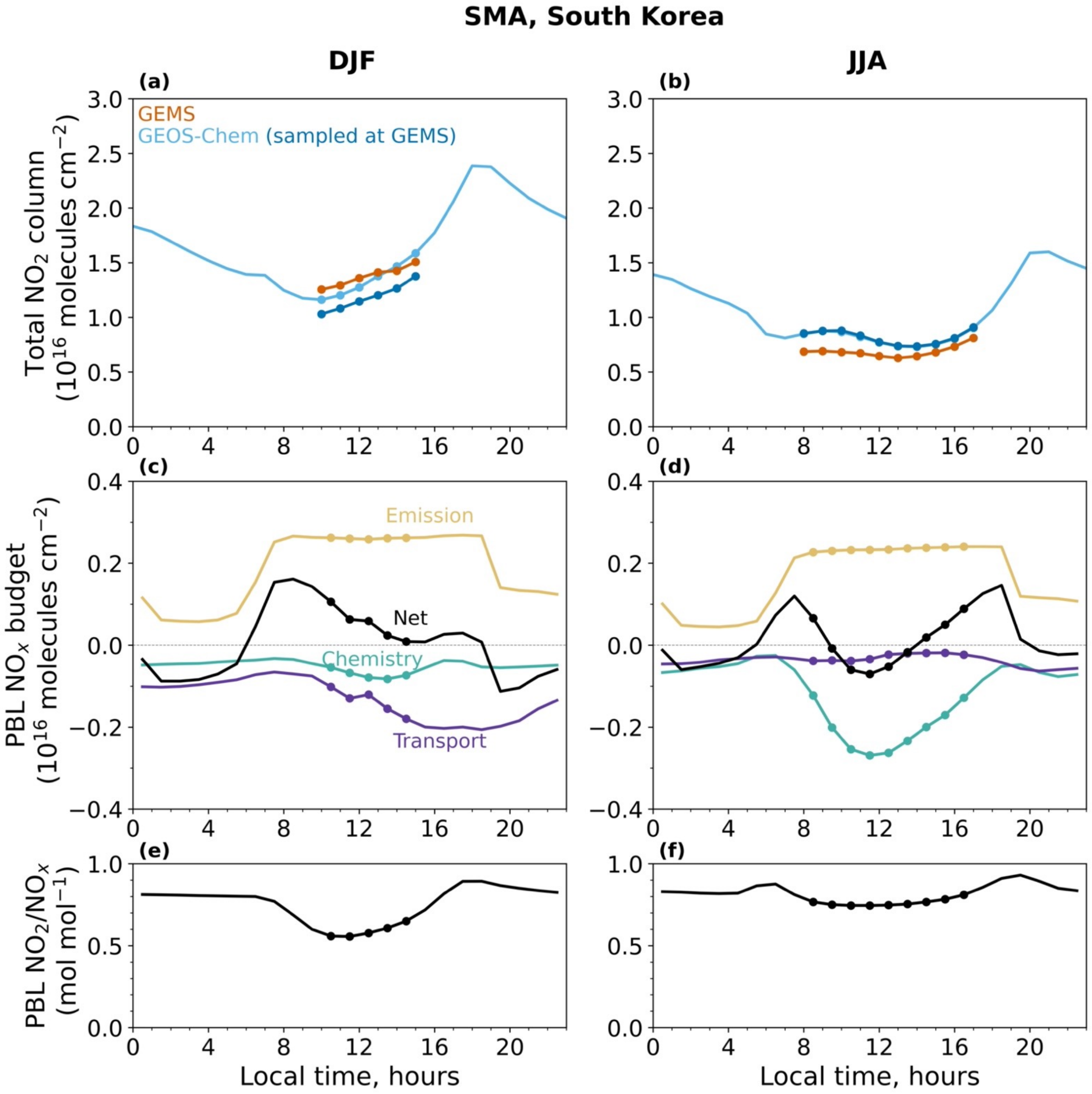
Broadening to a regional scale (150-km) successfully eliminates the effect of transport in summer



- The wintertime transport effect cannot be minimized even by going on a regional scale
- The inversion using CTM is needed to constrain emission during the winter
- By having information on emission, chemical loss can be constrained in the summer

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How would this work's results apply to interpreting NO_2 diurnal variation using TEMPO?

- Is the diurnal variation of NO_x emissions and surface NO_2 similar in the US and East Asia?
- Will a larger contribution of the FT background affect the interpretation of the TEMPO diurnal variation?

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Takeaways

NO_x emissions are four times higher in the daytime than at night, driving an accumulation of NO_2 over the course of the day offset by losses

NO_2 in winter increases throughout the day due to high daytime emissions and increasing NO_2/NO_x ratio balanced by loss from transport

In summer, chemical loss combined with transport drives minimum in NO_2 column at 13-14 local time and spatial averaging can minimize the effect of transport

We can independently infer the emission in the winter and chemistry in the summer by quantifying the transport term using an inversion with a CTM

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