NO_2 VERTICAL PROFILES OVER SOUTH KOREA AND THEIR RELATION TO OXIDANT CHEMISTRY:

IMPLICATIONS FOR GEOSTATIONARY SATELLITE RETRIEVALS

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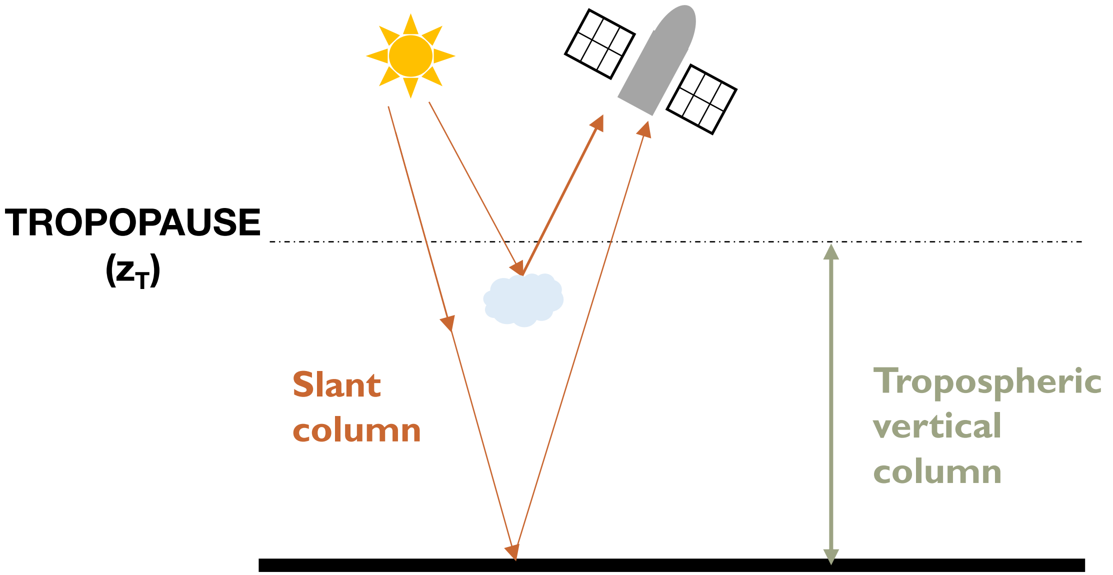
Overview of monitoring NO_2 from space (Solar backscatter retrieval)

1. Convert radiance to slant column (SC)
2. Remove stratospheric portion from SC
3. Convert tropospheric SC to vertical column (VC)



$$VC = \frac{SC}{AMF}$$

AMF = Air Mass Factor

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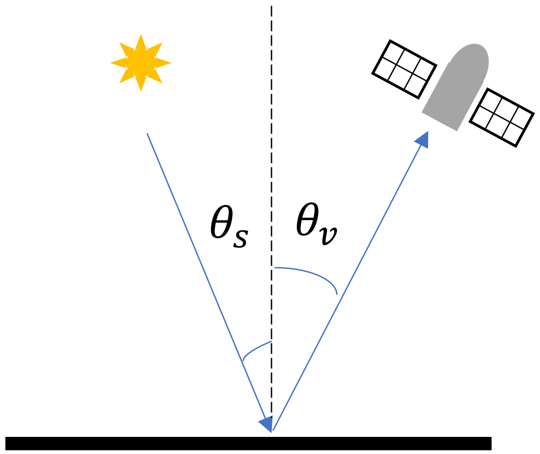
Air Mass Factor (AMF) depends on 3 quantities

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

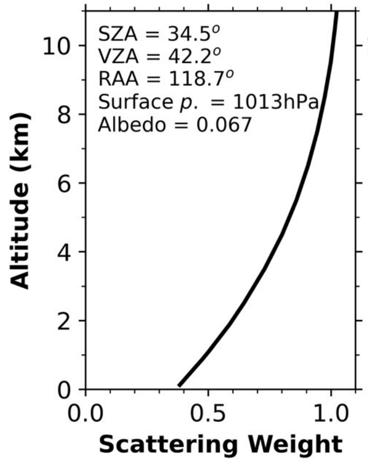
Viewing Geometry

Solar zenith angle (SZA; θ_s)
Satellite viewing angle (VZA; θ_v)



Scattering Weight

Captures where the satellite is sensitive to (from RTM)

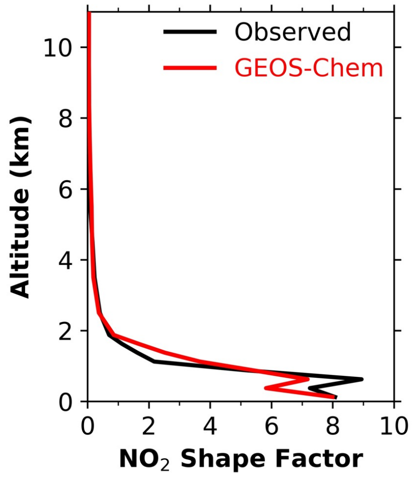


Scattering weight vs. altitude

RTM: Radiative Transfer Model
RAA: Relative Azimuth Angle

Shape Factor

Vertical distribution of NO_2 (from CTM like GEOS-Chem)



NO_2 shape factor vs. altitude

CTM: Chemical Transport Model

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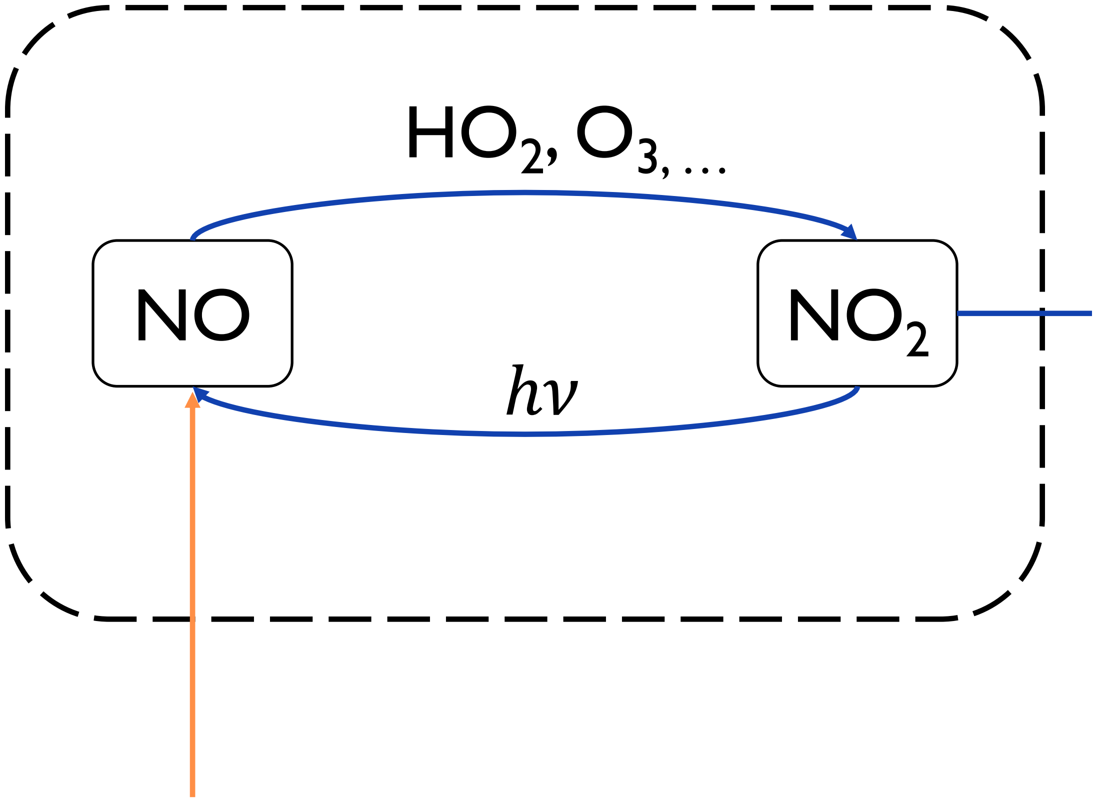
NO_2 concentrations are controlled by oxidant chemistry



$$PSS = \frac{[NO]}{[NO_2]} = f([O_3],[HO_2],\ldots)$$

PSS: Photostationary Steady State

Emission

Deposition

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KORUS-AQ campaign offers observational constraint for chemical species

GEOS-Chem

Standard Model

v13.3.4

0.25° × 0.3215°

No nitrate aerosol photolysis

No HNO_3 uptake by PMC

No VCP emission

CO boundary condition not scaled up

γ_{HO_2} = 0.2

GEOS-Chem

Modified Model

GEOS-Chem

0.25° × 0.3215°

With nitrate aerosol photolysis

With HNO_3 uptake by PMC

With VCP emission

CO boundary condition ×1.5

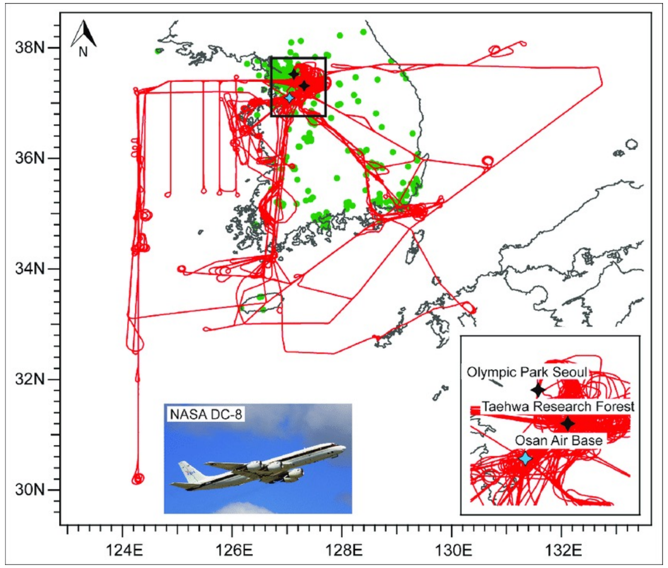
γ_{HO_2} = 0.1

PMC: Coarse PM
VCP: Volatile Chemical Product
γ_{HO_2}: HO_2 uptake coefficient

KORUS-AQ

May – June 2016

Aircraft Observation



Source: Peterson et al. 2019
[Crawford et al. 2021]

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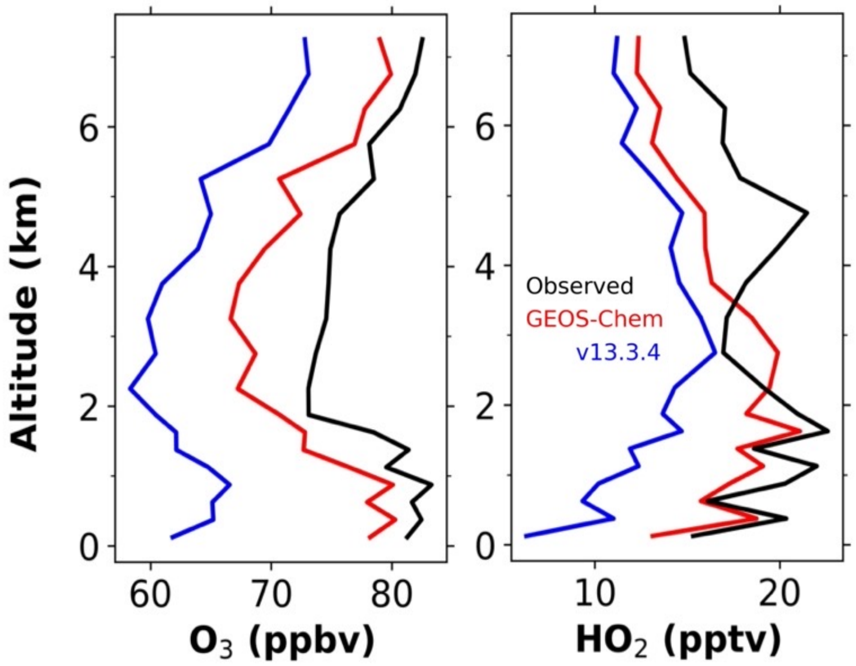
GEOS-Chem is successful in simulating key species that drives NO_2 formation & oxidant chemistry



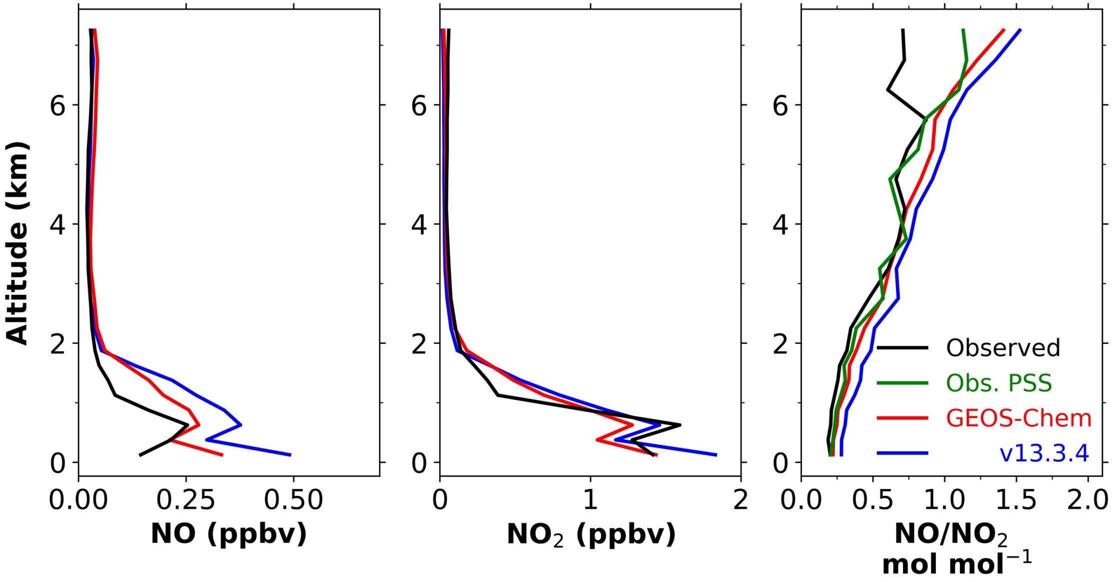
Median vertical profiles

Instruments/PIs	
Chemiluminescence:	A. Weinheimer
ATHOS:	W. Brune

O_3 and HO_2 are key driver species for forming NO_2

O_3 underestimation was significant issue in standard GEOS-Chem (Park et al., 2021)

GEOS-Chem successfully simulates NO, NO_2, and NO/NO_2



Median vertical profiles

Instruments/PIs	
Chemiluminescence:	A. Weinheimer
TD-LIF:	R. Cohen

$$\left(\frac{[NO]}{[NO_2]}\right)_{PSS} \textbf{ Equation}$$

$$\frac{j_{NO_2}}{k_{O_3+NO}[O_3] + k_{HO_2+NO}[HO_2] + k_{BrO+NO}[BrO] + k_{RO_2+NO}[RO_2]}$$

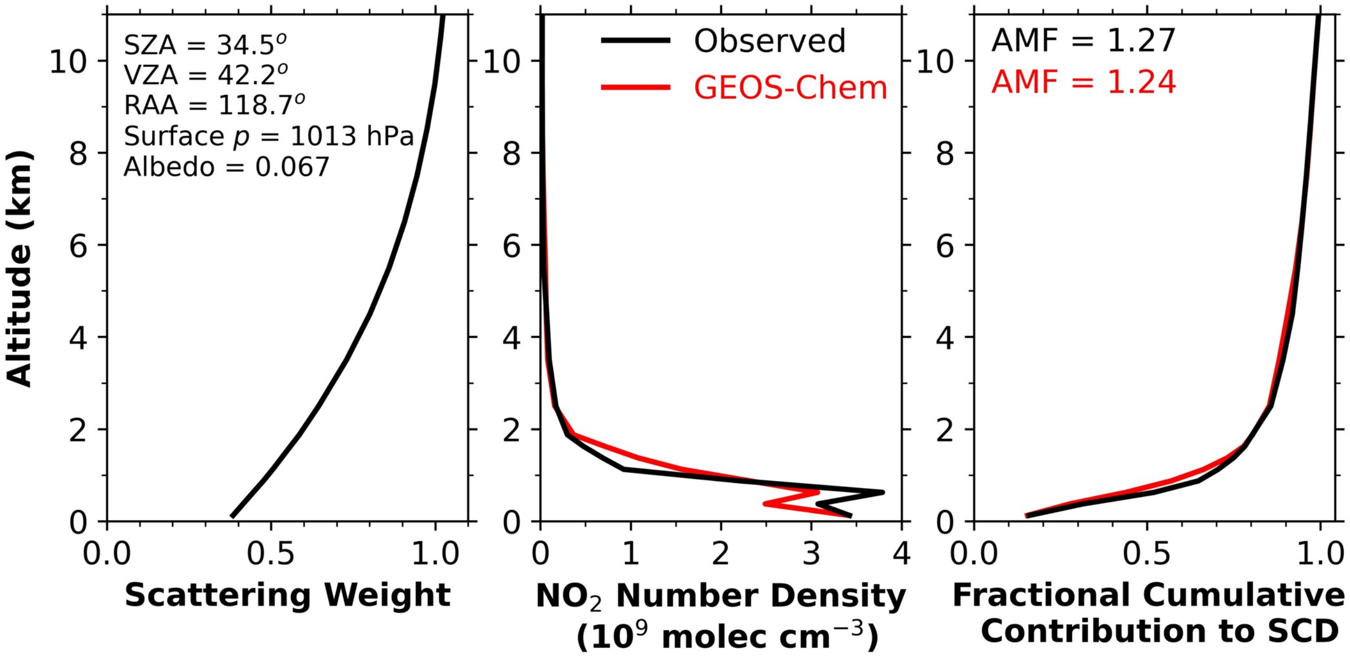
Observed

GEOS-Chem (<4%)

NO/NO_2 observation departs from the model above 5km (TD-LIF NO_2 positive interference)

Photostationary Steady State (PSS) is more reliable & updated model is in closer agreement with PSS

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

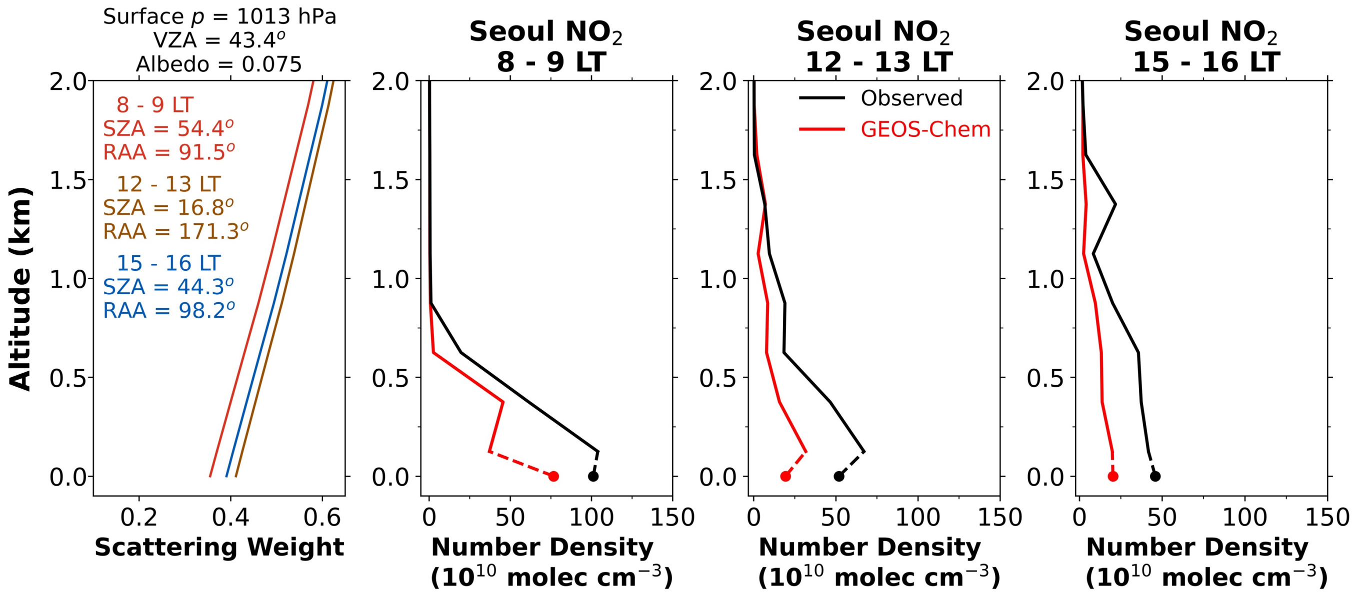


Reflects highly polluted condition

Over the U.S., only 20 – 35% of the column is contained within PBL (Travis et al. 2016)

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

Accounting for diurnal variation of scattering correction factor is critical



Column's diurnal variation (22%) is much smaller than that of surface (87%) [Crawford et al. 2021]

Solar zenith effect (24%) and scattering correction factor (18%) offset each other

Time of day	AMF_G	$\int_0^{z_T} w(z)S(z)dz$	AMF
8-9 AM	3.09	0.38 (0.39)	1.19 (1.20)
12-1 PM	2.42	0.46 (0.47)	1.11 (1.14)
3-4 PM	2.77	0.46 (0.46)	1.26 (1.27)

Diurnal variation in AMF (14%) is comparable to that of column (~25%)

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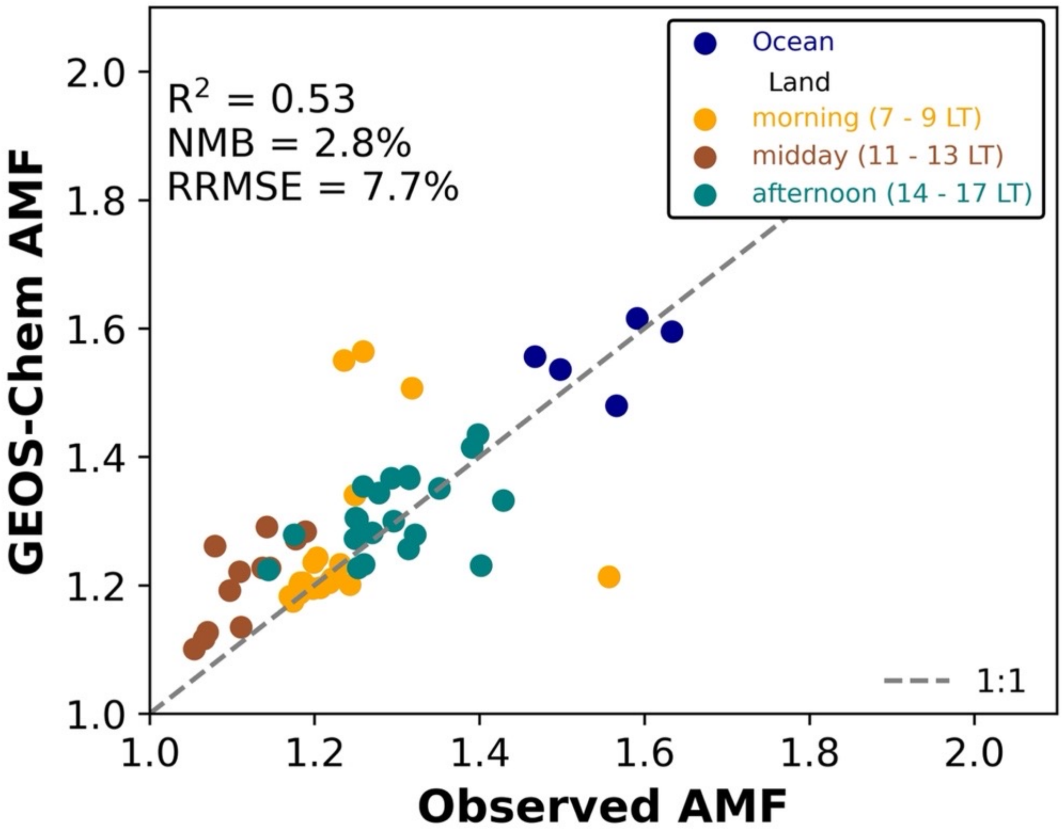
GEOS-Chem can capture the variability of observed AMF



Observed AMF shows high variability (1.05 – 1.63)

Ocean vs. land, and the time-of-day drive observed variability

Timing of the mixed layer growth in the morning is the largest contributor to the model error

Takeaways

Accurate accounting of oxidant chemistry is important for modeling the shape factor that is used in the GEMS NO_2 retrieval

Accurate accounting for the diurnal variation in AMF is critical in interpretating the diurnal variation in NO_2 columns

GEOS-Chem can provide AMFs for GEMS retrieval with relatively low error (NMB: 2.8%, RRMSE = 7.7%)