Global intercomparison of tropospheric oxidant chemistry in a common Earth system model environment using GEOS-Chem (v14.1.1) and CAM-chem chemistry within the Community Earth System Model version 2 (CESM2)

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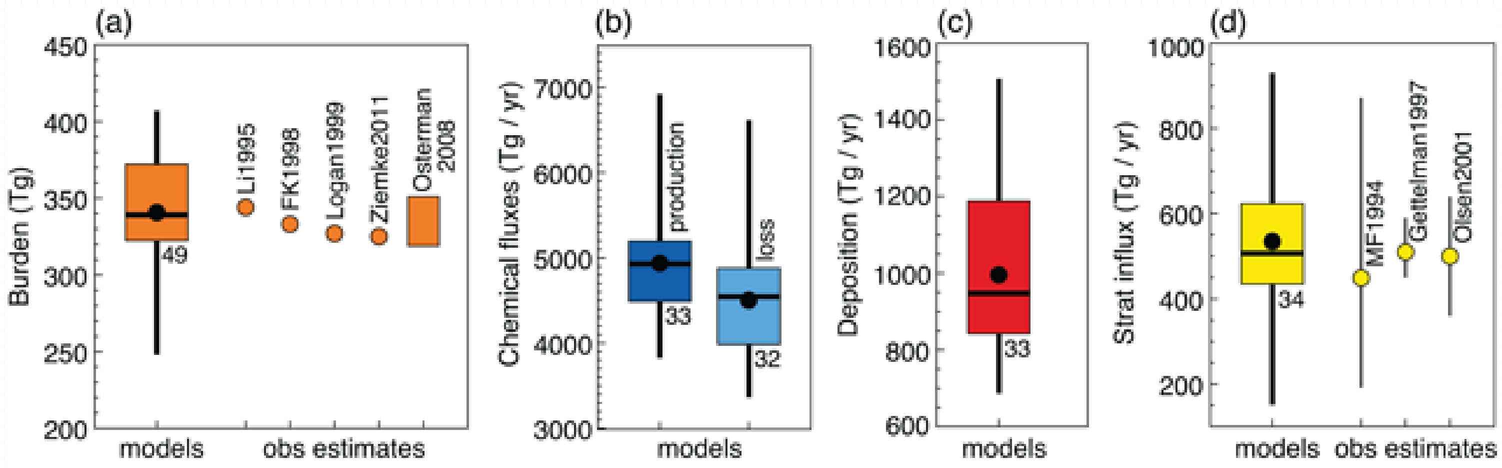
v2

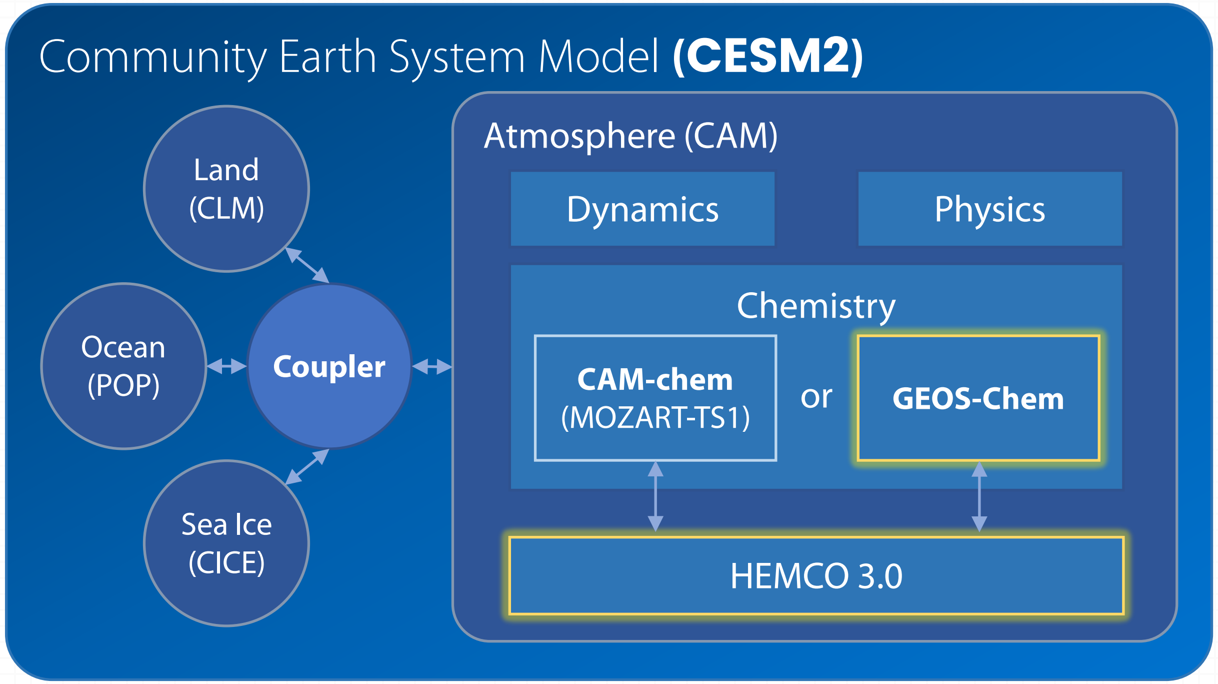
Ozone is a central species in tropospheric chemistry and an important indicator of model skill, but current models show large differences in individual processes controlling it



***TOAR:** Young et al., 2018*

Large differences in process magnitudes imply large differences in sensitivity to perturbations, which pose difficulty for chemistry-climate models aiming to quantify chemical feedbacks to climate change

Implementation of the GEOS-Chem chemical option in the Community Earth System Model (CESM2) allows for direct comparison between two state-of-the-art chemical modules



Previous model intercomparisons generally compared entire modeling systems.
Implementation of GEOS-Chem within CESM2 allows for detailed, process-based comparison to CAM-chem

GEOS-Chem within CESM: *Fritz et al., 2022*
HEMCO 3.0: *Lin et al., 2021*

Our work identifies and evaluates major differences between GEOS-Chem and CAM-chem chemistry and their effect on reproducing features in observations

	Chemistry Mechanism	Aerosol composition/ Microphysics	Photolysis scheme
GEOS Chem	**GEOS-Chem v14.1.1** 286 species, 914 reactions O_x-NO_x-VOC-**halogen**-aerosol • Aerosol nitrate photolysis • N_2O_5 uptake in clouds	Bulk aerosols mapped to MAM4 modes for ARI/ACI effects • Explicitly represents nitrate aerosol	Fast-JX • Aerosol extinction effects
CAM-chem (within CESM®)	**MOZART-TS1** 229 species, 541 reactions O_x-NO_x-VOC-aerosol	MAM4 modal aerosols	TUV lookup table

Both models use meteorology from CESM2.3 (cam6_3_095) **nudged to MERRA2 and emissions from HEMCO (CEDSv2+KORUSv5)**

Both models show **similar global burden** of tropospheric ozone and OH **but large differences in budget terms**

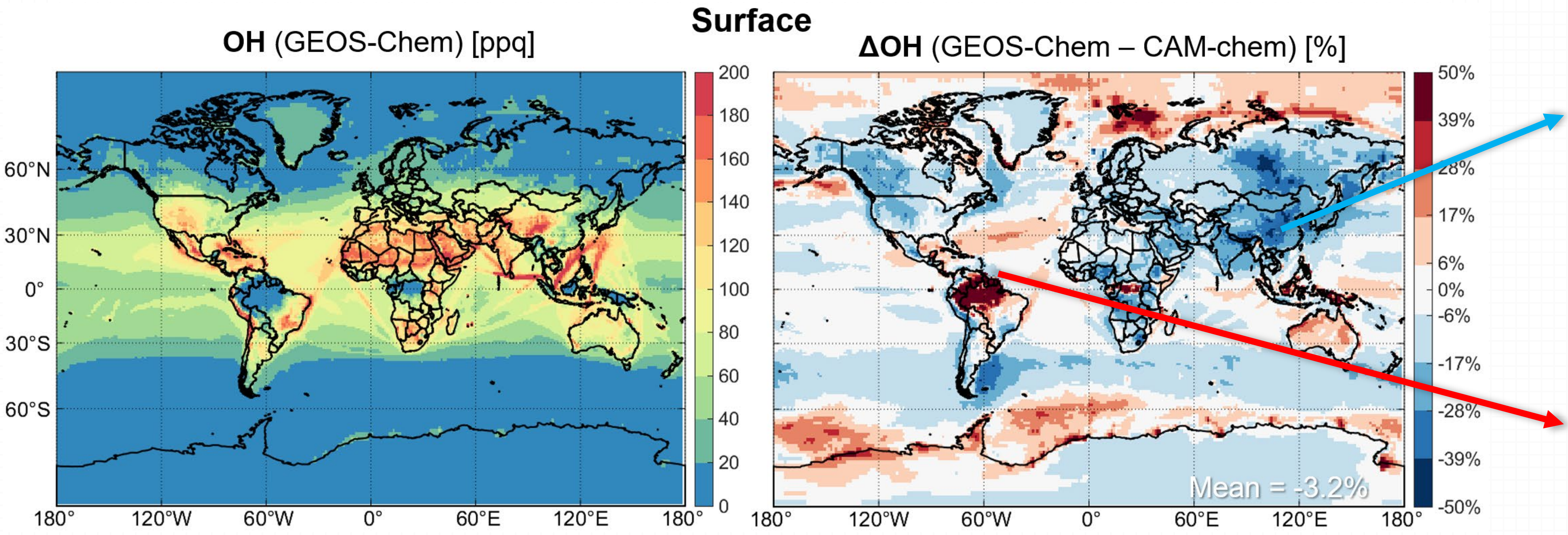
Budget terms	**GEOS-Chem**	**CAM-chem**	**Model ranges from literature** (Young et al., 2018, Naik et al., 2013)
Tropospheric ozone **burden** (Tg)	350	342	340 (250–410)
O_x chemical **production** (Tg a^{-1})	5395	5052	4900 (3800–6900)
O_x chemical **loss** (Tg a^{-1})	4813	4465	4600 (3300–6600)
O_x **deposition** (Tg a^{-1})	878	967	
Ozone **dry deposition** (Tg a^{-1})	749	826	1000 (700–1500)
O_x **STE** (Tg a^{-1})	341	380	500 (180–920)
O_x **Lifetime** (days)	23.0	23.7	22.3 (19.9–25.5)
Global **OH** (10^6 molecule cm^{-3})	1.21	1.22	1.11 ± 0.16
Stratospheric ozone **burden** (Tg)	2743.7	2744.4	

Driven by:
Aerosol nitrate photolysis
& Halogen chemistry

Slower deposition velocities over the ocean from **GEOS-Chem**
(~20 days)

Both models show similar global burden of tropospheric ozone and **OH but large regional differences**

2016 annual mean surface OH from GEOS-Chem and differences with CAM-chem



GEOS-Chem has

Lower OH over polluted regions

- More complex representation of VOC chemistry
- Higher OH reactivity

Higher OH over Amazon/Congo basin

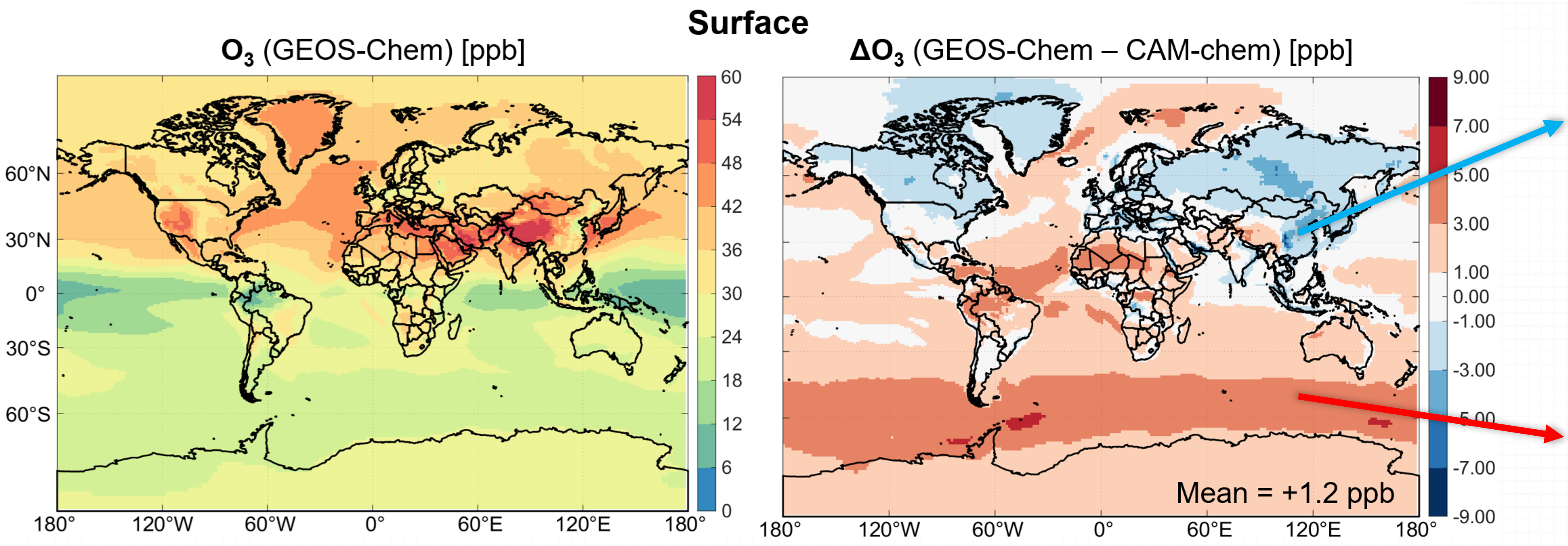
- Updated isoprene chemistry recycling OH in low-NO_x conditions

(Bates & Jacob, 2019)

Leads to lower CO in CAM-chem

Both models show similar global burden of tropospheric **ozone** and OH **but large regional differences**

2016 annual mean surface ozone from GEOS-Chem and differences with CAM-chem



GEOS-Chem has

Lower ozone in the NH

- Loss to halogen chemistry and cloud N_2O_5 uptake *(Wang et al., 2021; Holmes et al., 2019)*

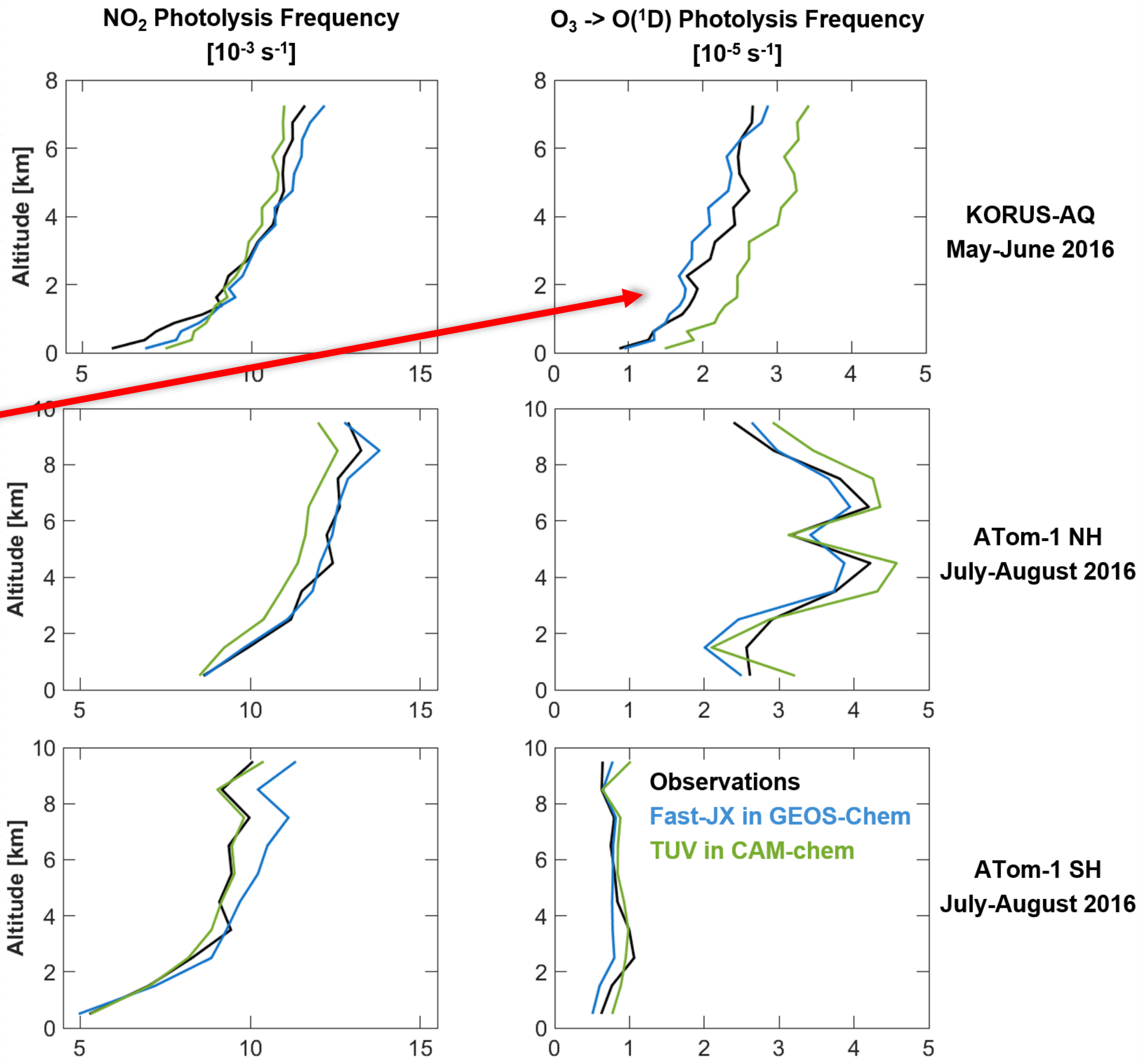
Higher ozone in the oceans and SH

- Slower ozone deposition over ocean due to using GEOS-Chem velocities *(Pound et al., 2020)*

Fast-JX (in GEOS-Chem) and TUV (in CAM-chem) photolysis schemes generally agree on $J(NO_2)$ but **differ in $J(O^1D)$ over polluted regions**

***J*(O^1D) is overestimated by TUV (CAM-chem)**

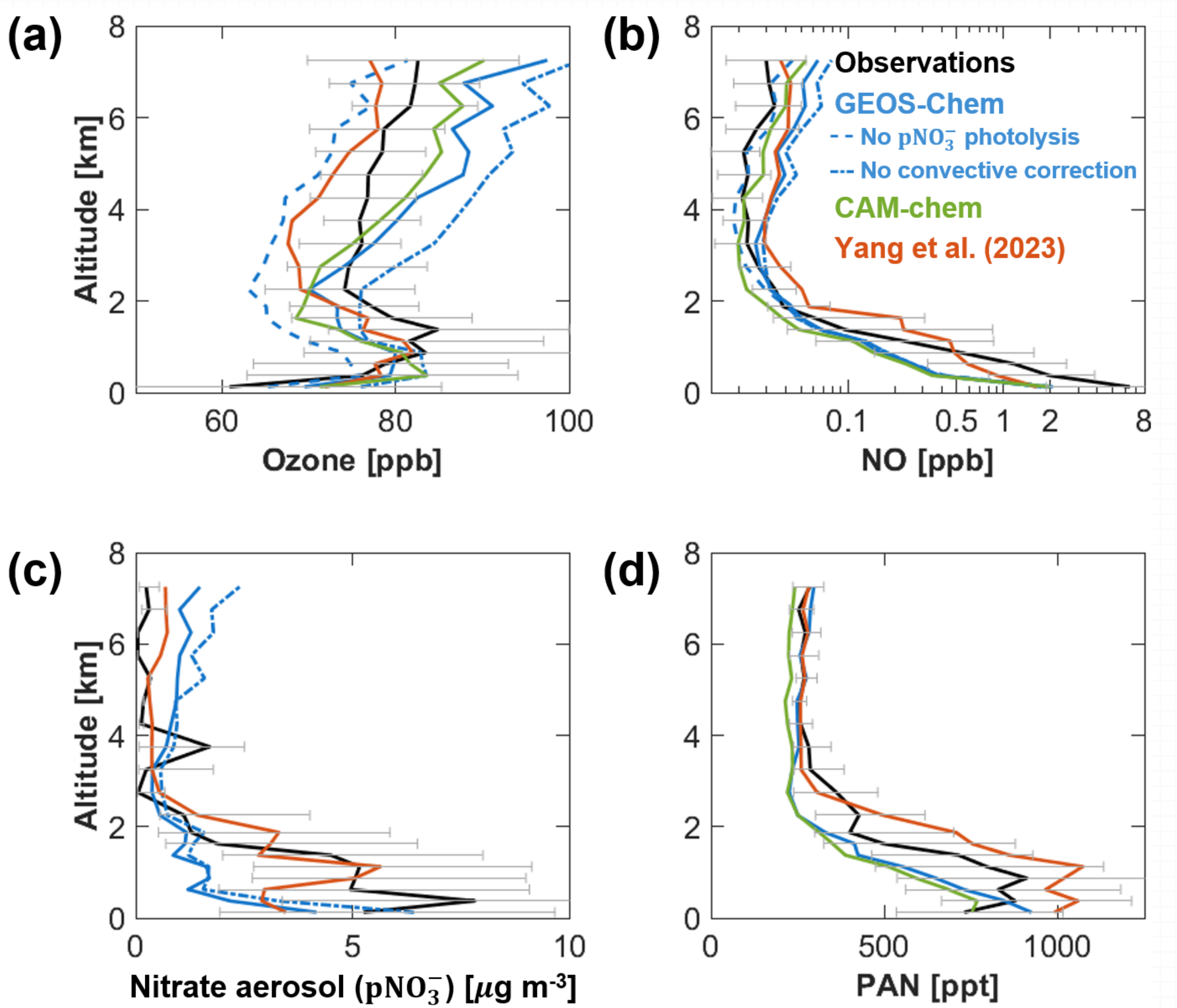
- *Not* aerosol extinction (or clouds), as difference persists in clear-sky *J*-values
- *Not* overhead ozone column, as **difference disappears by using Fast-JX in CAM-chem**
- Most noticeable over polluted regions. Why?



We can attribute particular features of GEOS-Chem chemistry to differences against CAM-chem in the comparison to KORUS-AQ

- **CAM-chem** simulates ozone well but **GEOS-Chem** can only do so with aerosol nitrate (pNO_3^-) photolysis
- Effect of pNO_3^- photolysis in GEOS-Chem has a strong dependence on pNO_3^- which is **not wet scavenged in convective updrafts** in the CESM2 environment
- **GEOS-Chem (offline)** can simulate pNO_3^- well below 2km but not **GEOS-Chem (CESM).** This may be due to boundary layer dynamics in CESM

Tropospheric vertical profiles, KORUS-AQ (May-June 2016) Over the Seoul Metropolitan Area (SMA)



Take home messages

- Implementation of **GEOS-Chem** as an **alternative chemistry option to CAM-chem** in CESM2 allows for side-by-side intercomparison of two state-of-the-art chemistry representations
- Major differences between GEOS-Chem and CAM-chem are driven by: (1) the photolysis scheme, (2) aerosol nitrate photolysis, (3) N_2O_5 uptake in clouds, (4) tropospheric halogen chemistry, and (5) ozone deposition to oceans.
- While GEOS-Chem and CAM-chem have similar ozone and OH budgets, **there are important differences in the underlying processes** and **major regional differences**, which imply differences in sensitivity to perturbations.

GEOS-Chem within CESM2 is available for testing and will be merged into mainline CESM in the near future.

HEMCO emissions for CAM-chem are available in beta versions of CESM (cam6_3_118+)