Importance of boundary conditions for AQ modelling with CAM-Chem

Why is CO underestimated in global (and regional models for Asia)

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Atmospheric Chemistry Observations & Modeling Laboratory (ACOM)

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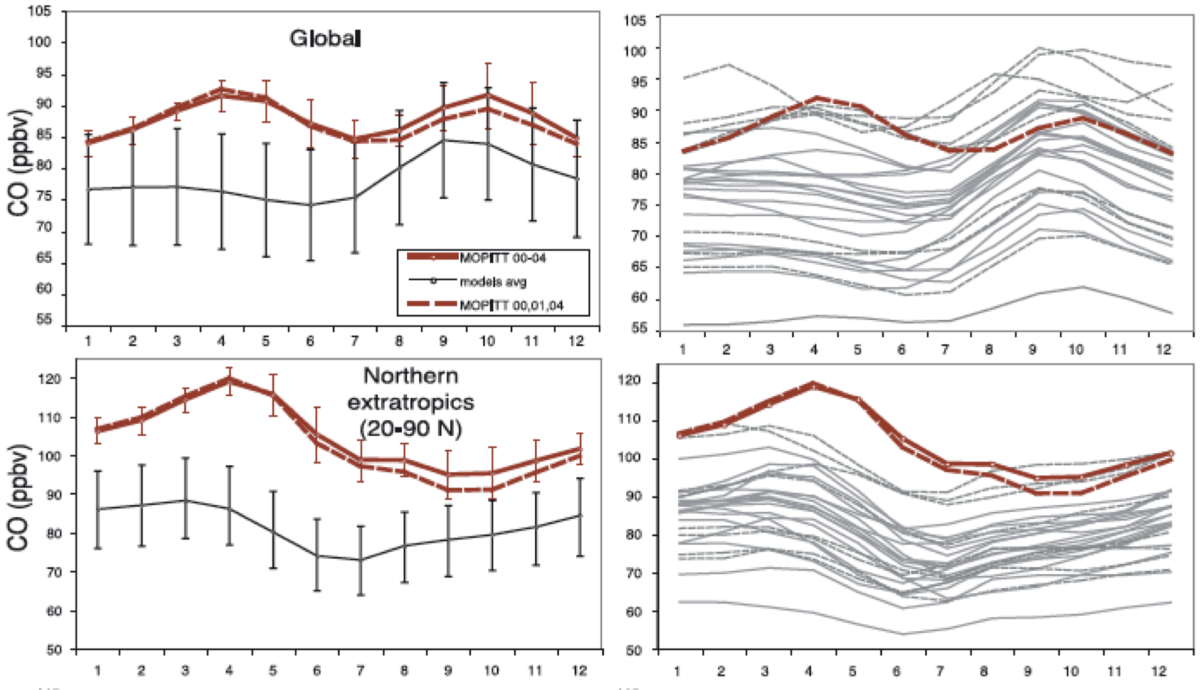
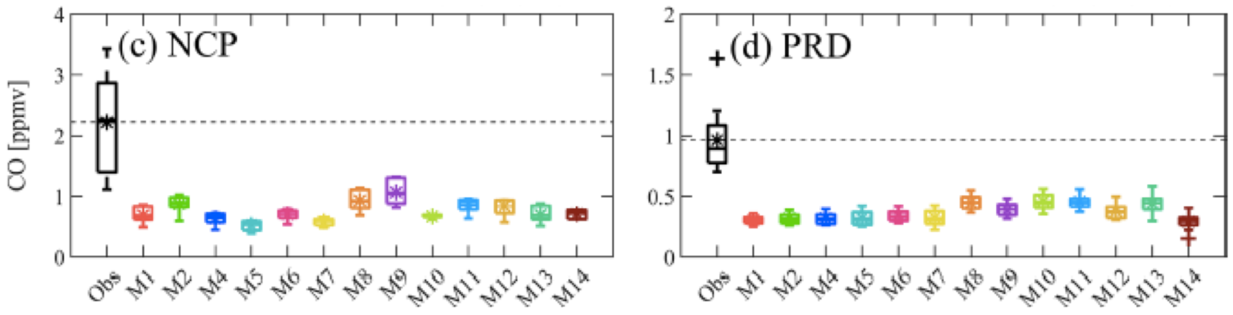
Understanding and modelling Carbon Monoxide CO

- CO bias in the northern hemisphere late winter and early spring
  - ✓ Year-round emissions, probably from fossil fuel burning in east Asia are greatly underestimated (Shindell et al., 2006).
  - ✓ Large intermodel variability in the regional distribution of OH and an overestimation of OH in the NH (Naik et al., 2013), overestimation of OH sources (water vapor, Ozone)
  - ✓ Stein et al. (2014) pointed out emissions from the traffic sector and suggests less deposition.
  - ✓ Kong et al. (2020), The Model Inter-Comparison Study for Asia III (MICS-Asia III); 13 state of the art models

Shindell et al., JGR, 2006



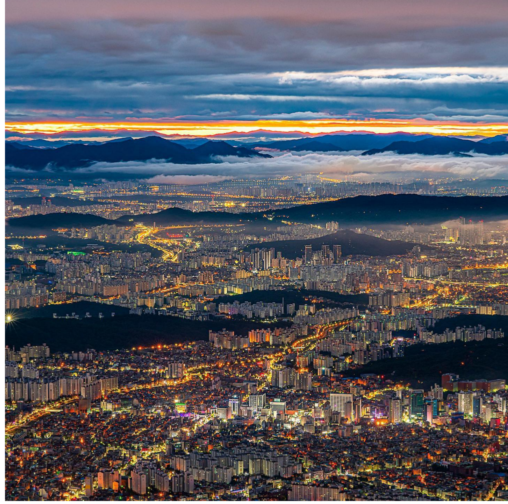


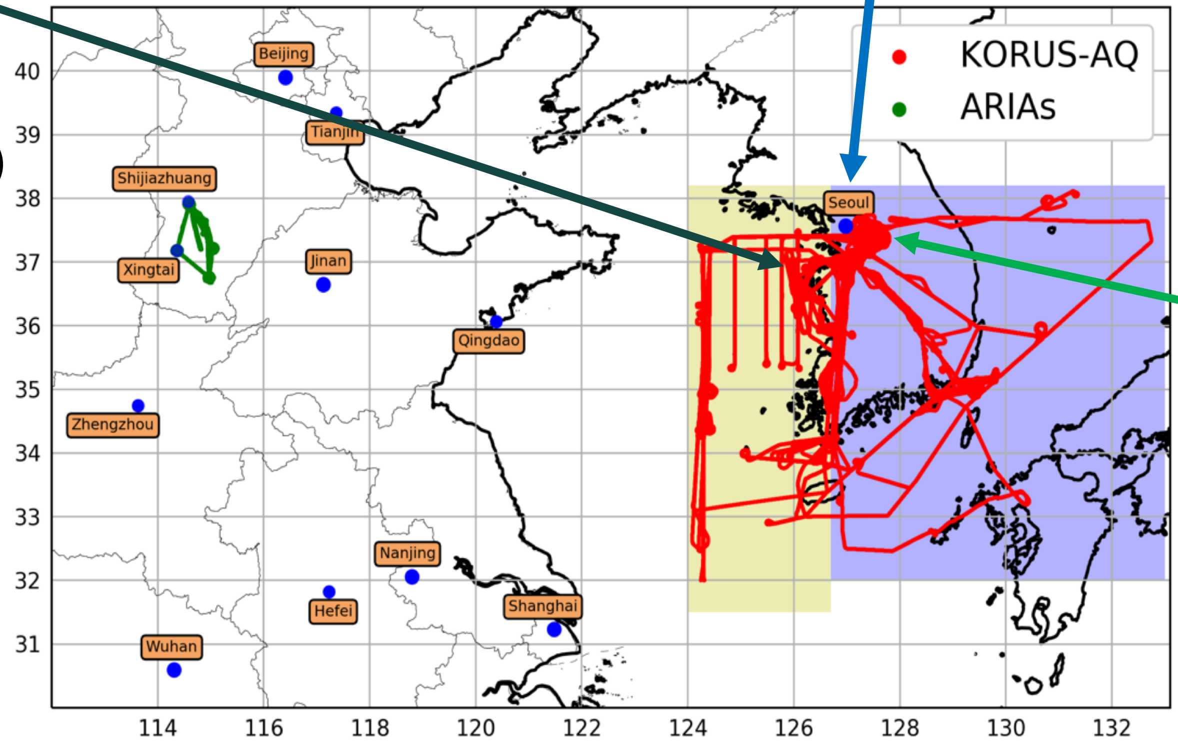
Kong et al., ACP, surface observations of CO for the North China Plain (NCP) and the Pearl River Delta (PRD) regions

Spatial gradients

Daesan petrochemical complex
(located 80 km southwest of Seoul)

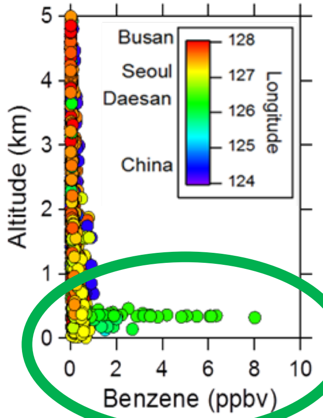
Seoul Metropolitan Area
25 Million inhabitants
(5th largest in the world)





Taehwa (forest) research site
(downwind of Seoul)

High Benzene concentrations were only measured over the Daesan petrochemical complex, filter when Benzene (PTRMS) > 1ppbv



➢ **20 science flights DC8 flight tracks**
(1 May 2016 and 9 June 2016)

Yellow area -> Yellow Sea

Violet area -> Korea (Korean peninsula)

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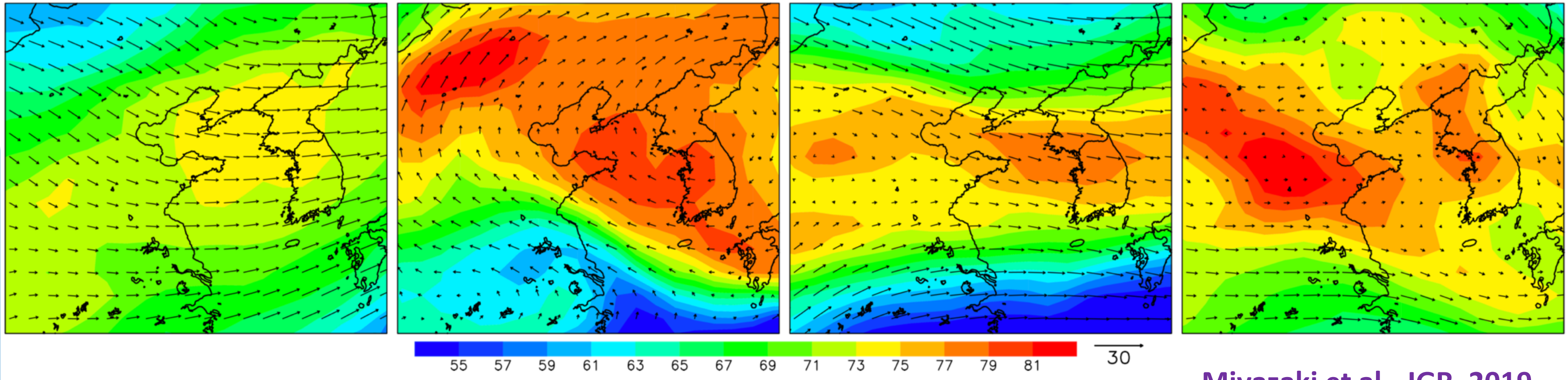
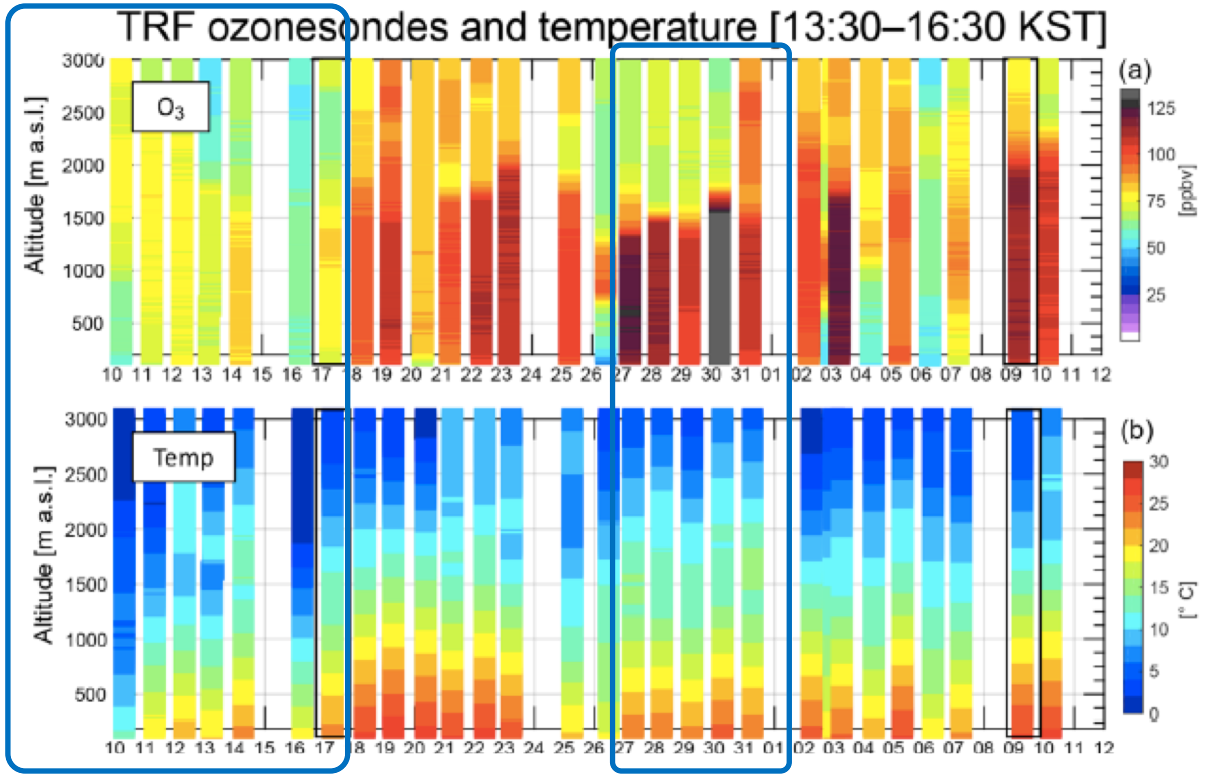
Temporal gradients
Peterson et al. (2019)

- Phase 1: (May 1st to May 16th) synoptic weather system dynamically changed
- Phase 2 (May 17th to May 25th) : Stagnant conditions.
- Phase 3 (25–31 May), Strong westerlies existed, Long Range Transport events.
- In phase 4 (1–6 June), a blocking pattern



Ozone at 700 hPa (ppbv)

Miyazaki et al., JGR, 2019



Sullivan et al., acp, 2019

NCAR UCAR CESM COMMUNITY EARTH SYSTEM MODEL

JAMES Journal of Advances in Modeling Earth Systems

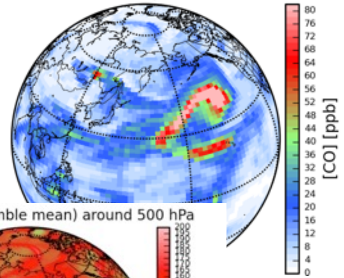
CESM2.1/CAM-CHEM

- CESM2.1 / CAM6 / 0.9°x1.25° / 32 layers
- TS1 chemistry, 221 species and 528 reaction (Emmons et al., 2020)
- MEGAN / FINN / CMIP6 emissions

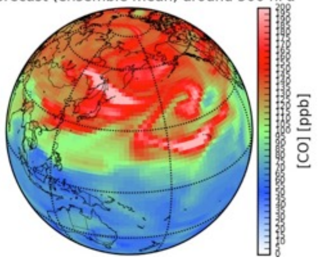
30 CAM-Chem forecasts

- Ensemble of emissions, different noise for sources
- Ensemble of transport
- Ensemble of deposition (land model)
- Initial conditions of CO

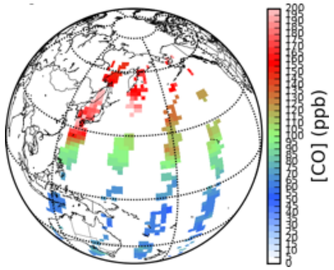
CO forecast (ensemble spread) around 500 hPa



CO forecast (ensemble mean) around 500 hPa

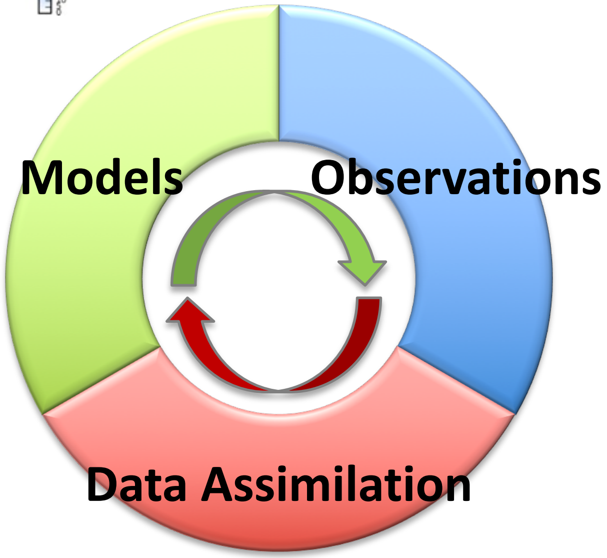


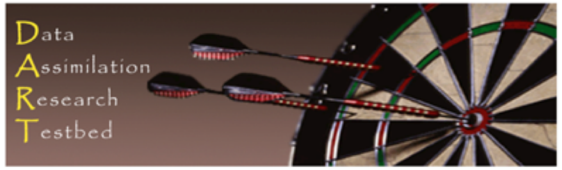
MOPITT obs around 550hPa



Observations

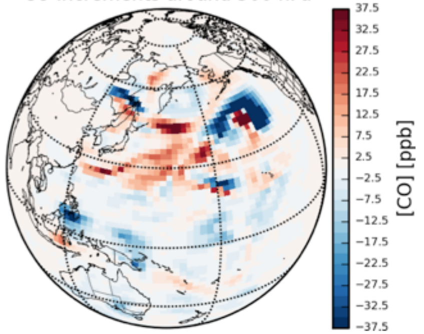
- Meteorological observations (Raeder et al. 2012)
- MOPITT V8J daytime retrieval





Ensemble of optimized initial conditions every 6 hours

CO increments around 500 hPa

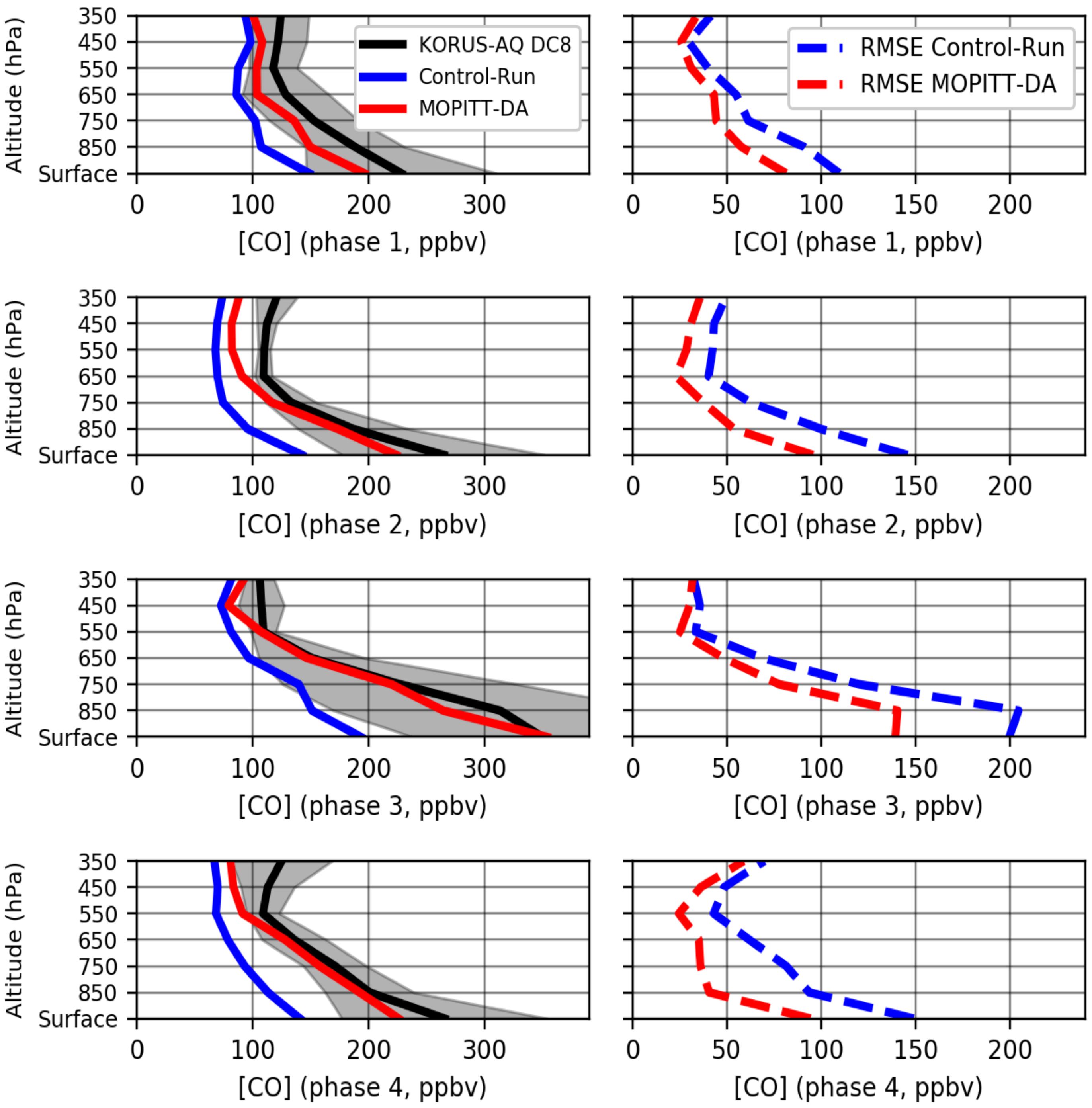


Data Assimilation Research Testbed (DART), Anderson et al. (2009)

- [CO] inferred by MOPITT, experiments with anthropogenic and BB CO emissions, [O3], and VOC’s
- P, T, U, V, Q inferred by Meteorological observations
- Space and time additive inflation (El Gharamti 2018) / Spatial localization

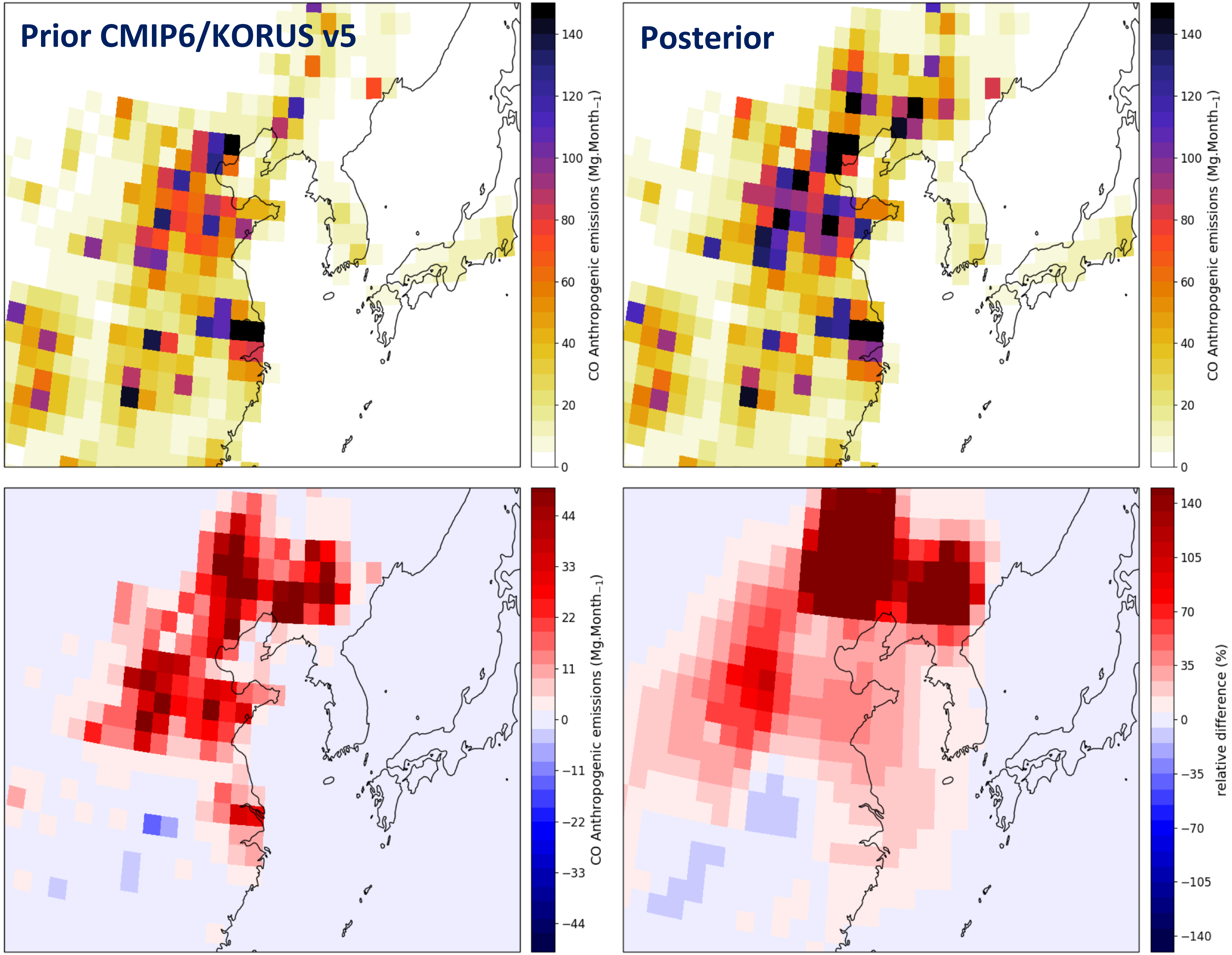
Results

- **Background CO (between 100 and 125 ppbv) is underestimated.**
- **Large bias and RMSE reduction after assimilation.**
- **Important enhancement during phase 3 quite well reproduced.**
- **On average, MOPITT-DA run underestimates the observed CO**



Emission inversion

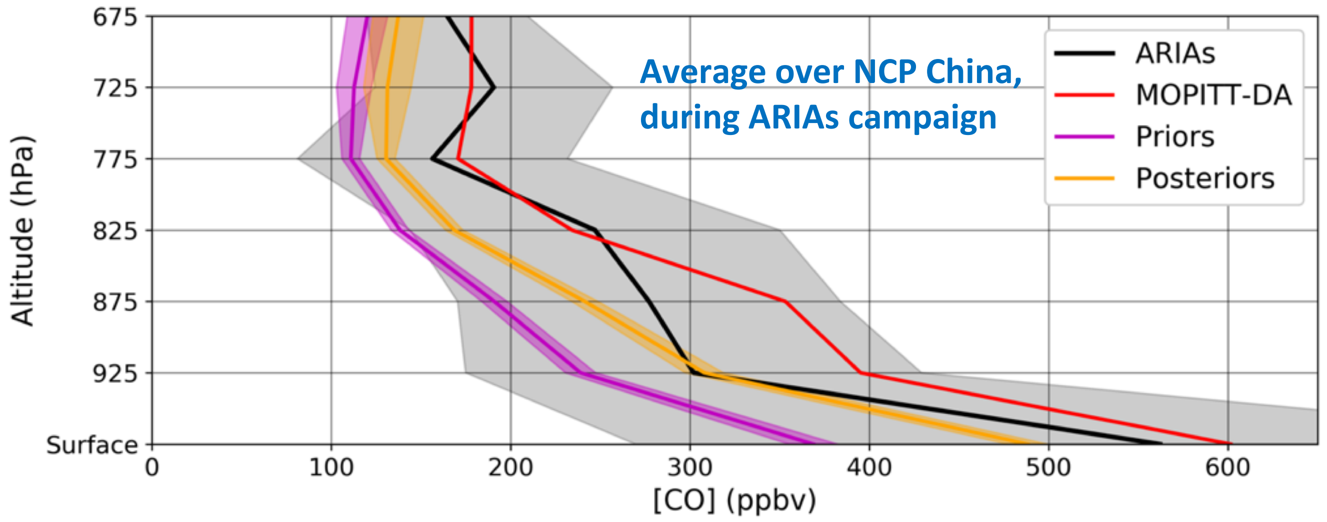
- Only small relative change over South Korea and Japan
- Large increase in Northern China
- Emission reduction west of Shanghai

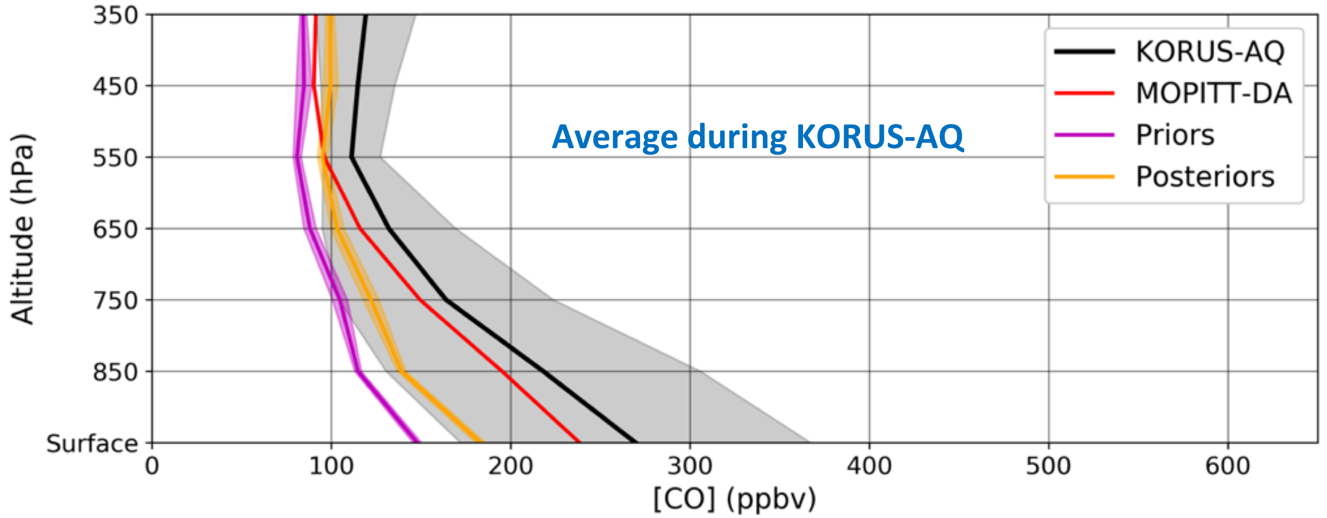


Emission inversion: sensitivity to transport

- **Perform nudging to MERRA-2 or DART deterministic run.**
- **Use of the posterior emissions clearly improves the CO vertical profile.**
  - **Probably overestimated posterior emissions for the ARIAs campaign.**
  - **Does not explain all the missing CO during KORUS-AQ.**

Simulation name	nudging	U, V, T (nudge)	CO emissions
g-prior-0.24	GEOS5	0.24, 0.24, 0.24	prior
d-prior-0.24	DART	0.24, 0.24, 0.24	prior
g-prior-0.48	GEOS5	0.48, 0.48, 0.48	prior
d-prior-0.48	DART	0.48, 0.48, 0.48	prior
g-prior-0.72	GEOS5	0.72, 0.72, 0.48	prior
d-prior-0.72	DART	0.72, 0.72, 0.72	prior
g-post-0.24	GEOS5	0.24, 0.24, 0.24	post
d-post-0.24	DART	0.24, 0.24, 0.24	post
g-post-0.48	GEOS5	0.48, 0.48, 0.48	post
d-post-0.48	DART	0.48, 0.48, 0.48	post
g-post-0.72	GEOS5	0.72, 0.72, 0.48	post
d-post-0.72	DART	0.72, 0.72, 0.48	post



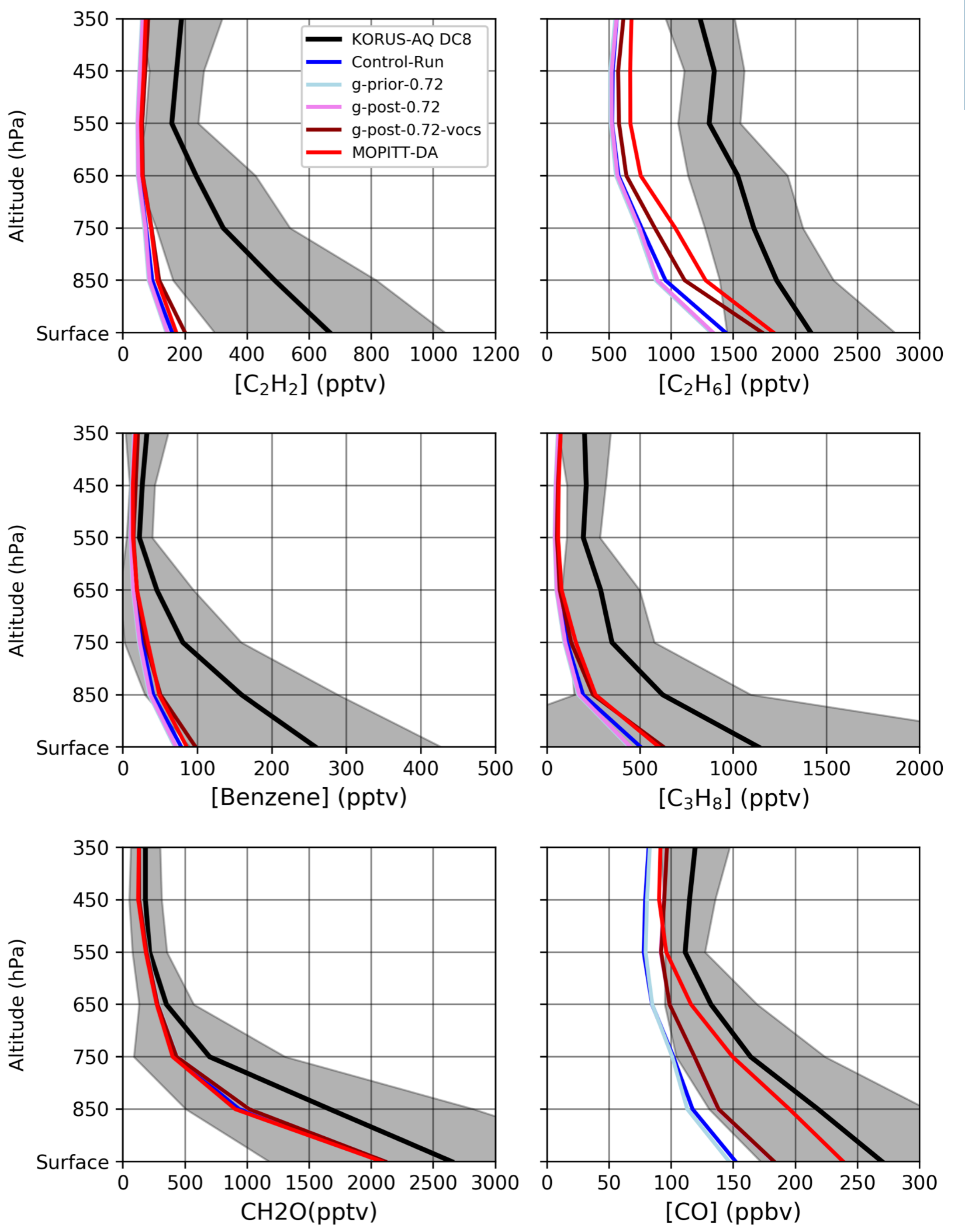


Anthropogenic VOCs

- **Sensitivity to anthropogenic VOCs emissions:** Scale VOCs emissions to the CO inversion for 4 VOCs

$$E_i^{post} = E_i^{prior} * \frac{E_{CO}^{post}}{E_{CO}^{prior}}$$

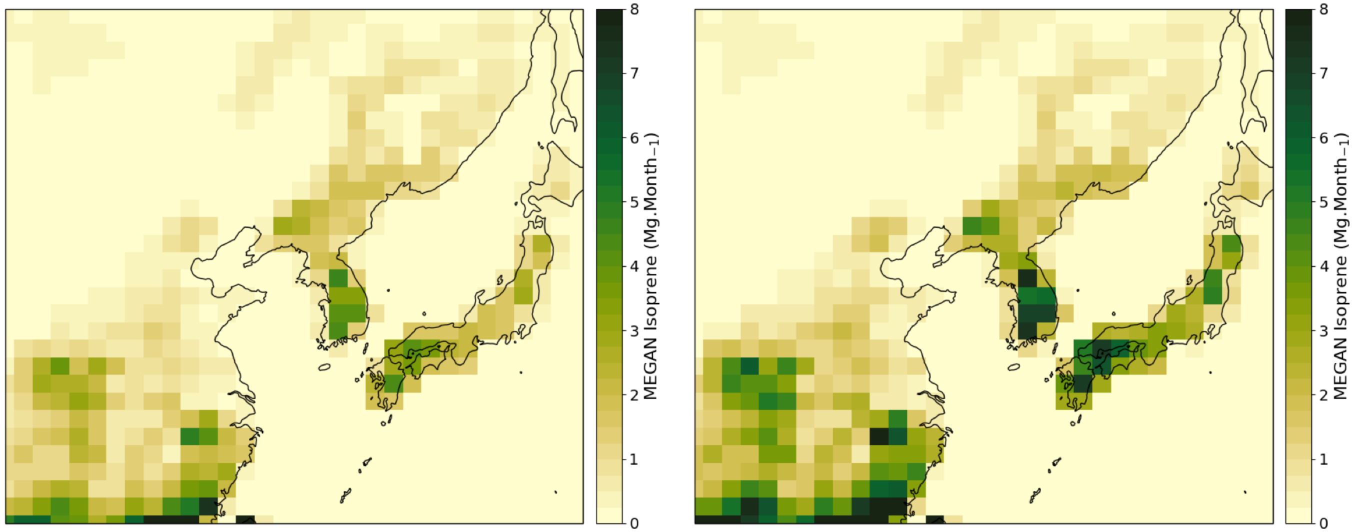
- Ethyne (C2H2), Ethane (C2H6), Benzene and Propane (C3H8) are primary VOCS, mostly from anthropogenic sources.
- Effect on CO and Formaldehyde are rather small.

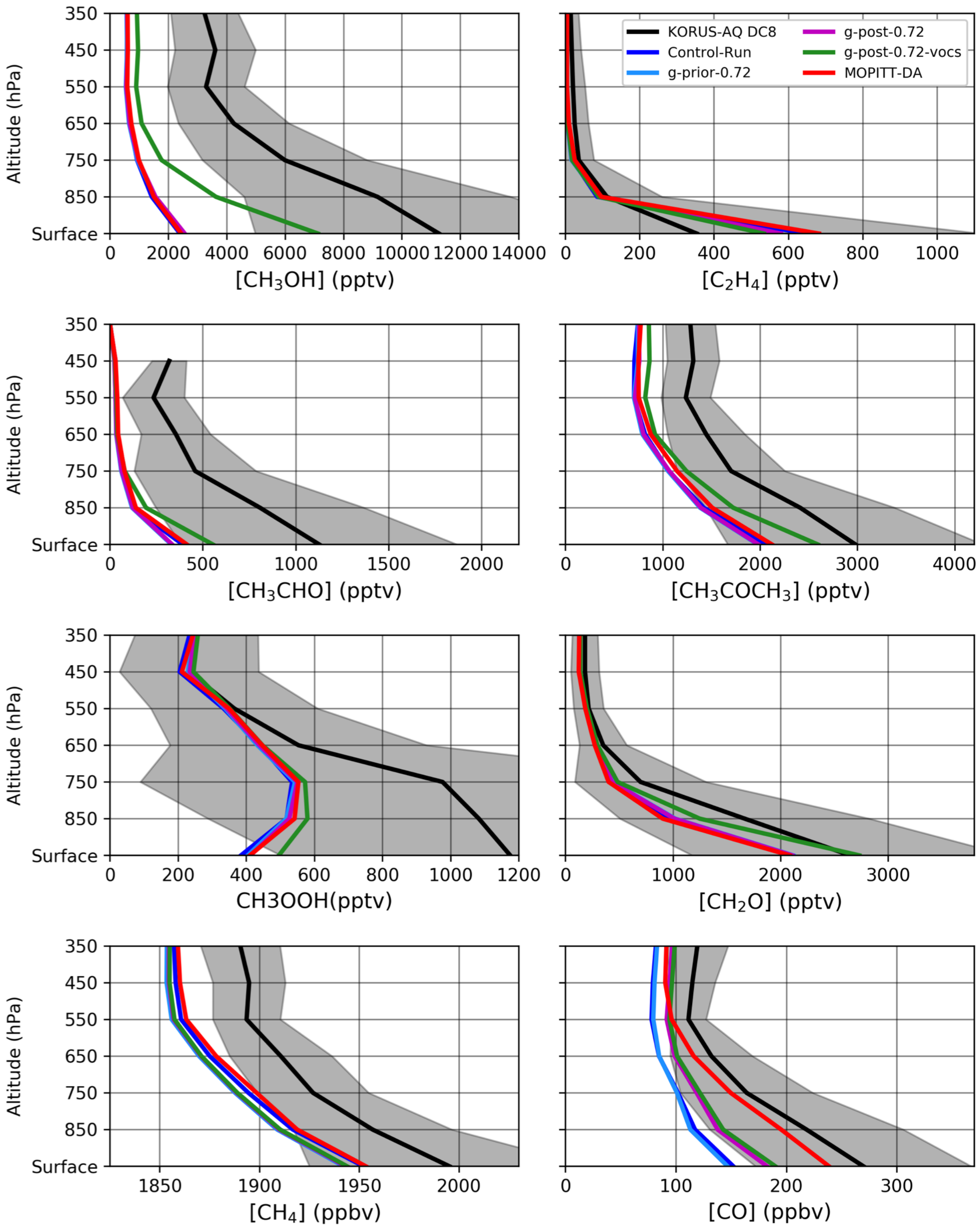


- **Sensitivity to biogenic emissions:** Increase emissions, for 3 given Plant Functional Types, “**Needleleaf Evergreen** Temperate Tree”, “**Broadleaf Evergreen** Temperate Tree”, “**Broadleaf Deciduous** Temperate Tree”

- Methanol (CH3OH), Ethylene (C2H4), Acetaldehyde (CH3CHO), Acetone (CH3COCH3) have large biogenic sources, CH3OOH and CH2O are mostly secondary.

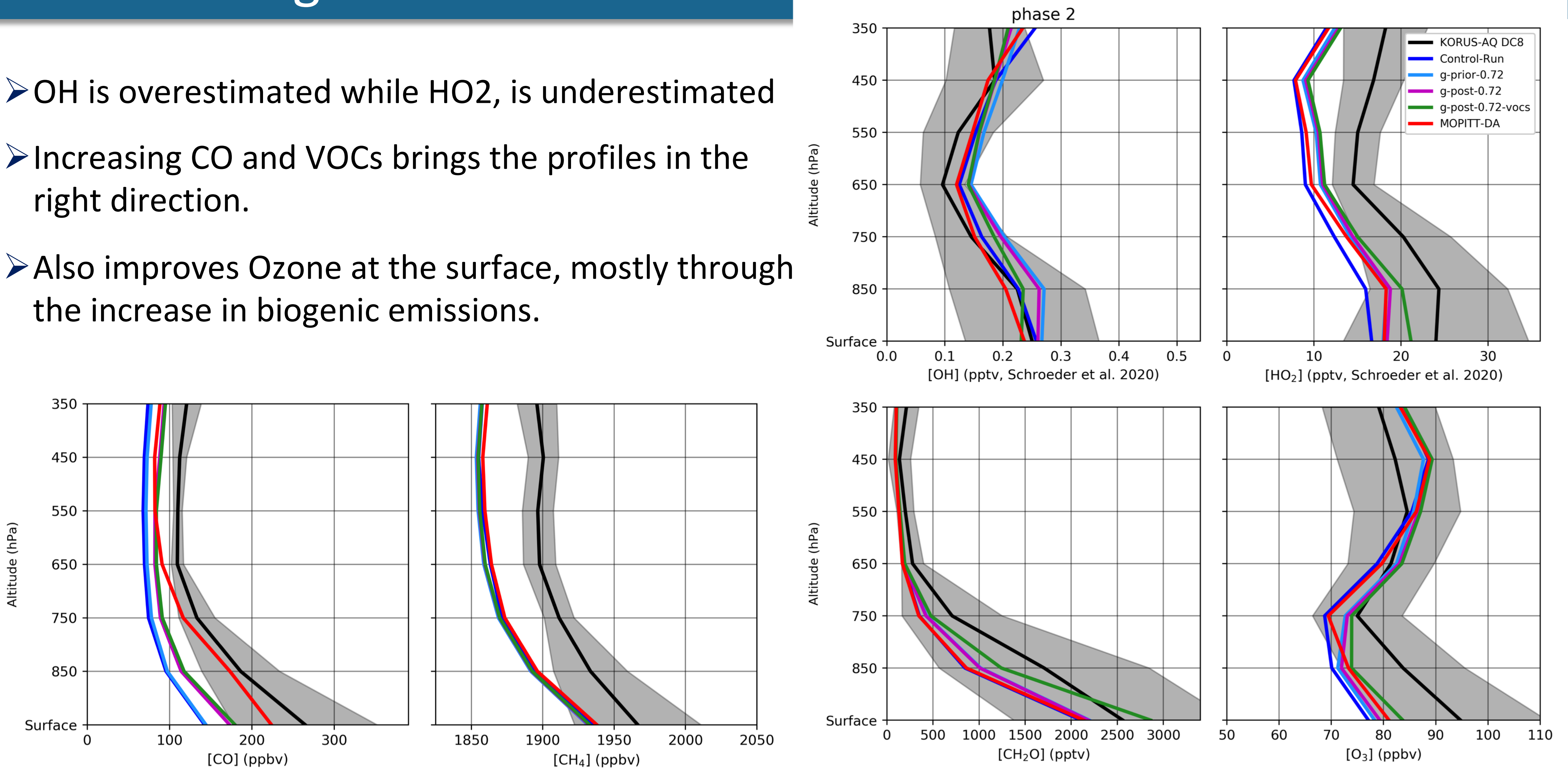
- Effect on CO is rather small, but bias on CH2O is significantly reduced at the surface.





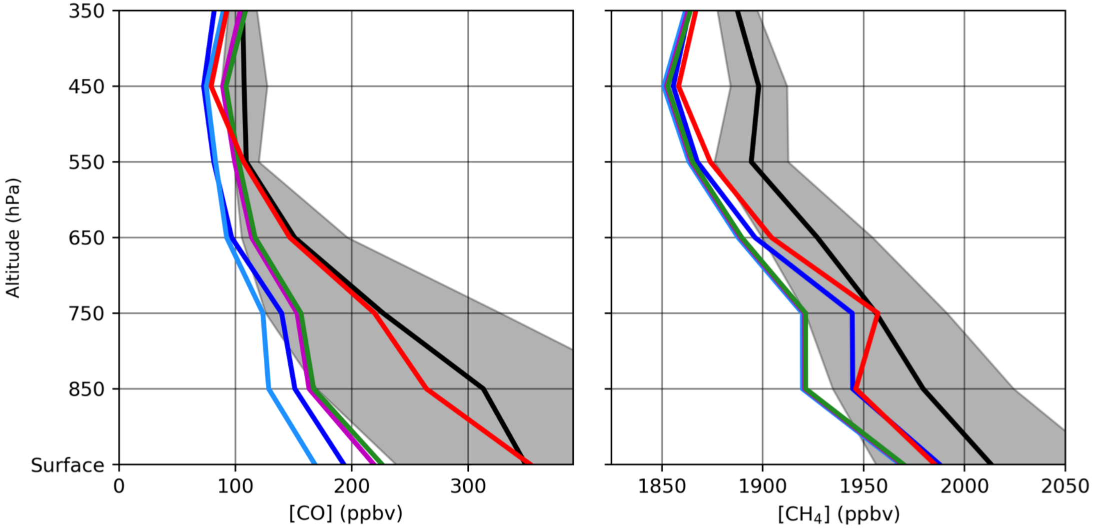
Phase 2: Stagnant conditions

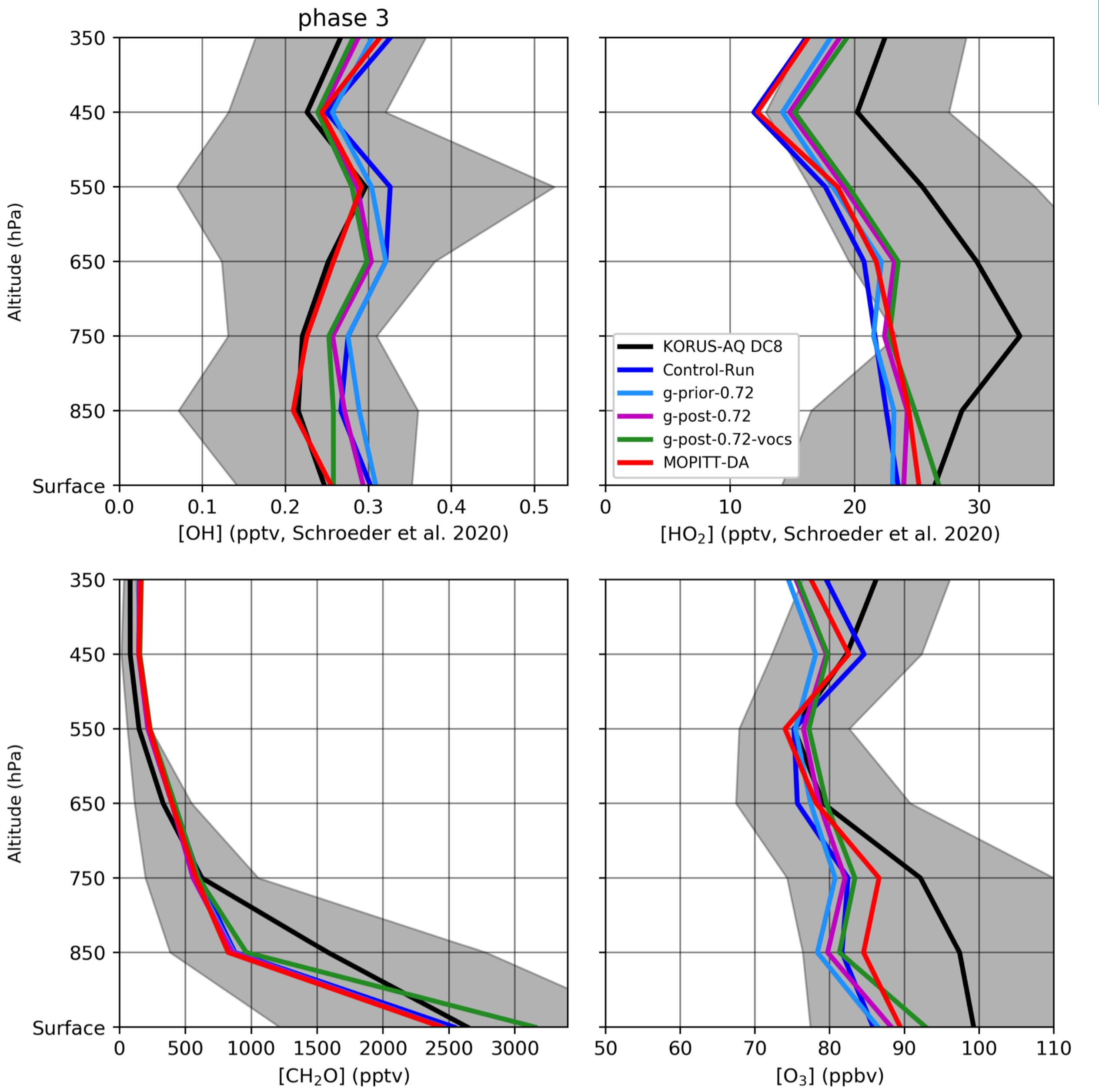
- OH is overestimated while HO2, is underestimated
- Increasing CO and VOCs brings the profiles in the right direction.
- Also improves Ozone at the surface, mostly through the increase in biogenic emissions.



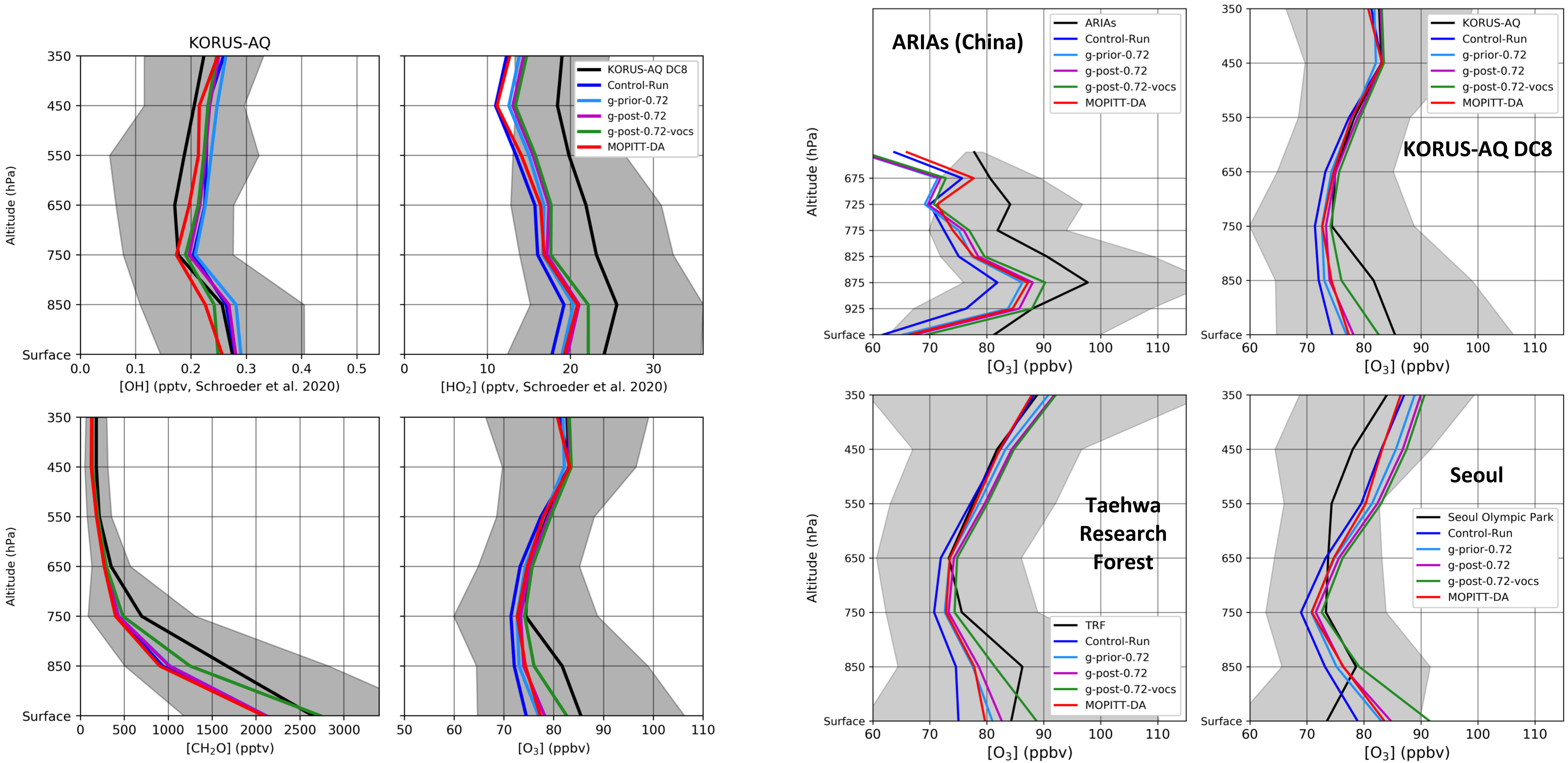
Phase 3: Long-range Transport

- OH is overestimated while HO2, is underestimated
- Higher HOx mostly through HO2 at higher altitude
- High Ozone formation





All KORUS-AQ: + Ozonesondes and ARIAs



Conclusions

- Updated Global Chemical Data Assimilation ensemble, that explicitly represents the non linear evolution of atmospheric chemistry and is designed to represents errors from transport, chemistry emissions.
- OH was pretty well modelled, while HO2 is underestimated. Underestimation of HOx points to missing emissions.
- Ozone is improve by correcting missing CO emissions and increasing VOCs, mainly from biogenic source
- Missing formaldehyde and OVOCs at the surface was corrected by increasing the MEGAN emissions but does not explain the missing CO.
- Remaining CO underestimation could be due to missing emissions, unseen by MOPITT from a direct anthropogenic CO or short lived VOCs.
- Assimilate the NOAA-19/CrIS CLIMCAPS CO retrievals.
- Work in progress on comparing with TM5-4D-Var inversion to assess impact of model error the assimilation algorithm, transport errors and chemistry.

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