Effects of satellite clouds-corrected photolysis rates on ozone over CONUS

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Motivation

- Relatively less attention in air quality community since cloud-free skies are conducive to high ozone levels
- However, clouds play a critical role in UV radiation for photochemistry and so need to be well predicted.

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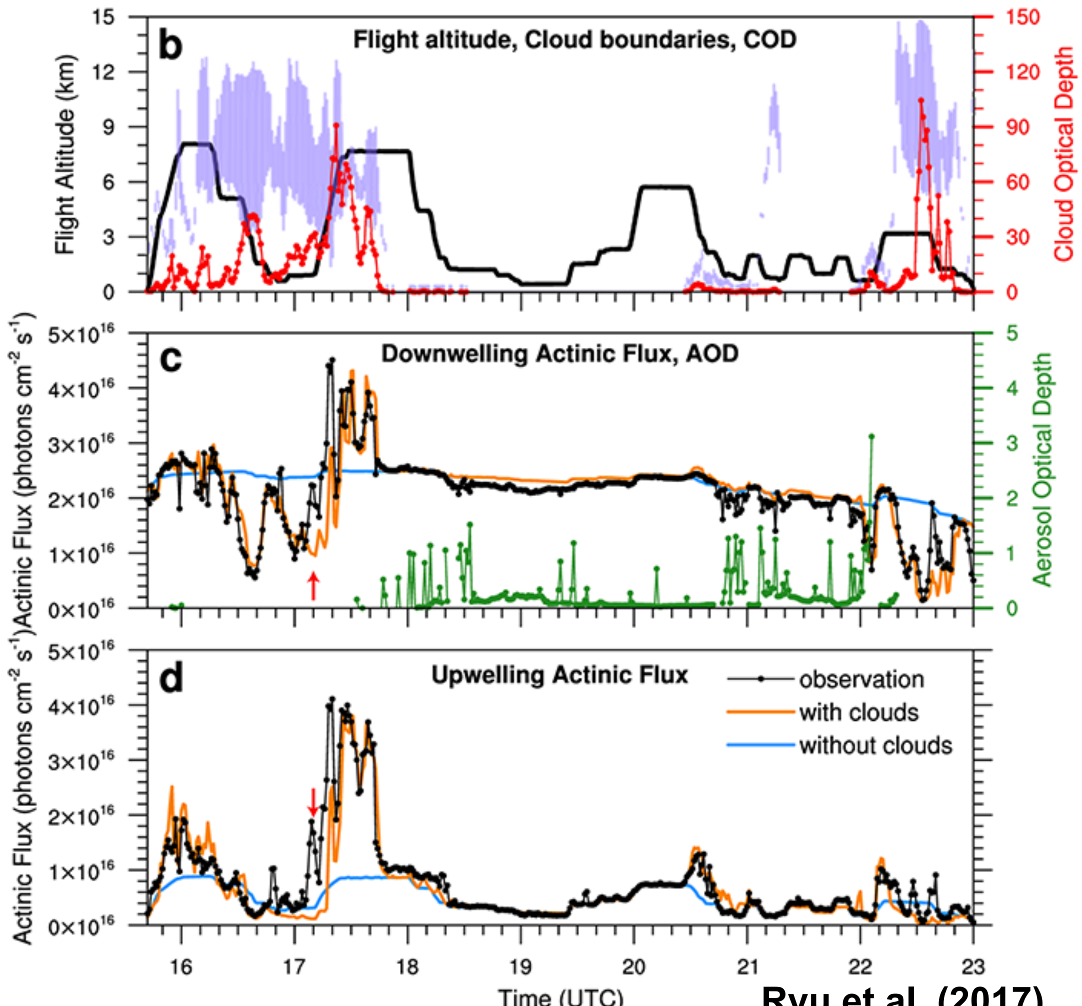
- Recent studies using satellite cloud retrievals to quantify the effects of clouds on actinic fluxes/ photolysis rates (e.g., Ryu et al. 2017)



Ryu et al. (2017)

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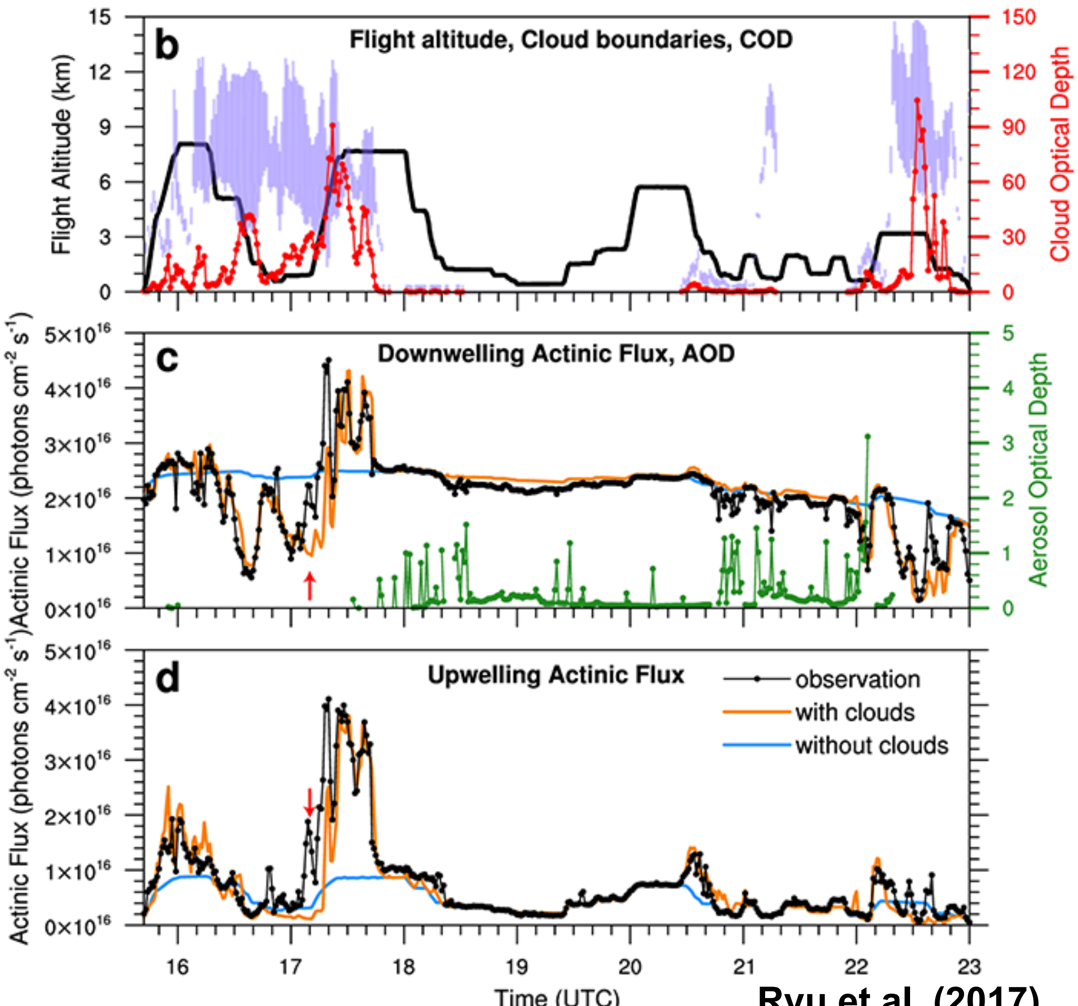
- However, clouds play a critical role in UV radiation for photochemistry and so need to be well predicted.
- Recent studies using satellite cloud retrievals to quantify the effects of clouds on actinic fluxes/ photolysis rates (e.g., Ryu et al. 2017)
- A few studies on effects of satellite-constrained (corrected) photolysis rates on ozone formation (Pour-Biazar et al. 2007; Tang et al. 2015)



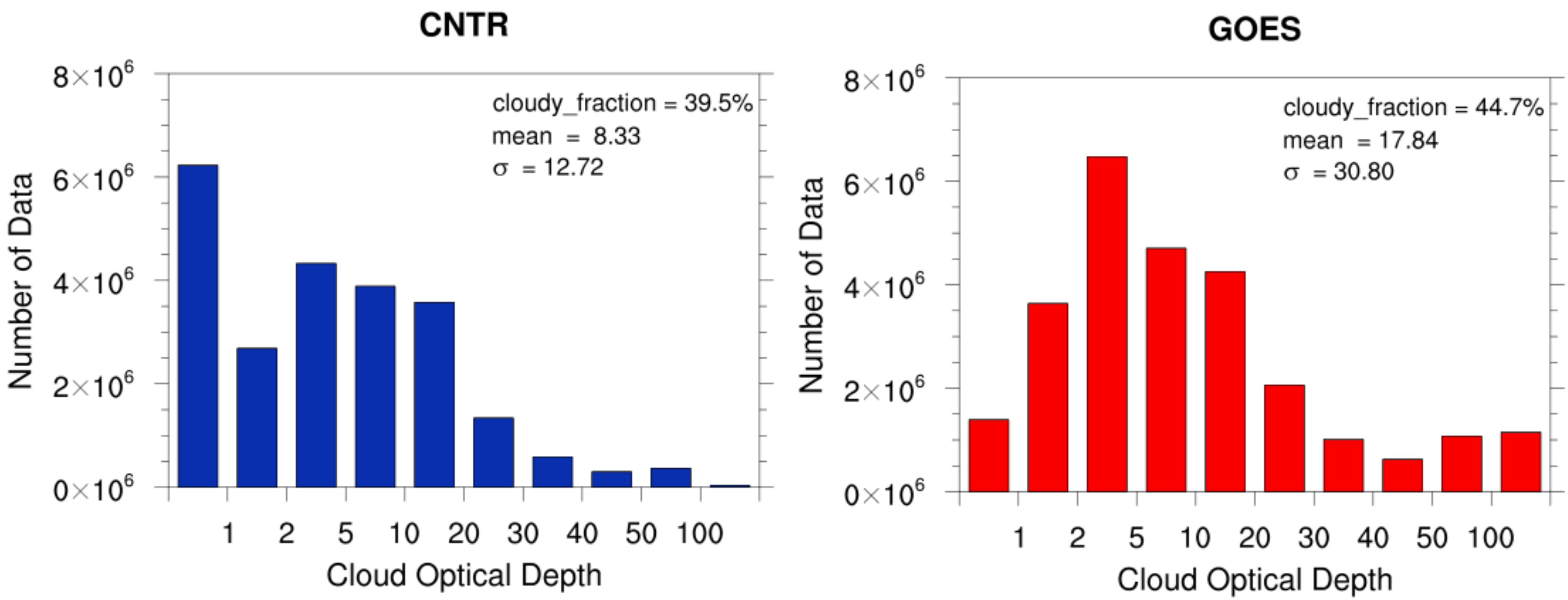
Ryu et al. (2017)

Goals and methods

- Employ satellite-derived (GOES; Geostationary Operational Environmental Satellite) cloud optical depth and cloud boundaries in the WRF-Chem over CONUS for 2013 Summer
- **Goals**
  - Evaluate vertical profiles of photolysis rates in the presence of clouds using aircraft data ($SEAC^4RS$)
  - Evaluate ground-level ozone biases statistically for a long term period (4 months in 2013)

- **12-km WRFchem simulations**
- Simulation period: June–September 2013, Horizontal grid size: 12 km
- Chemistry mechanism: MOZART_MOSAIC_KPP
- New (updated) TUV for photolysis rate computations
- Meteorology: reinitialized every 2 days using FNL reanalysis (6-hr spin up is allowed in each 2-day simulation) & No nudging
- Control (CNTR) simulation: WRF-generated clouds
- GOES simulation: satellite-derived clouds only in the photolysis computations (TUV in WRF-Chem).

Comparison of WRF and satellite clouds



- Daytime (16–23 UTC) hourly Cloud Optical Depth (COD) over the WRF domain (land only) for the period of 11 June 2013 through 30 September 2013

Cloud Evaluation — Contingency

		GOES satellite	
		Cloudy	Clear
WRF	Cloudy	A (hit)	B (false alarm)
	Clear	C (miss)	D (correct negative)

4-month domain averages

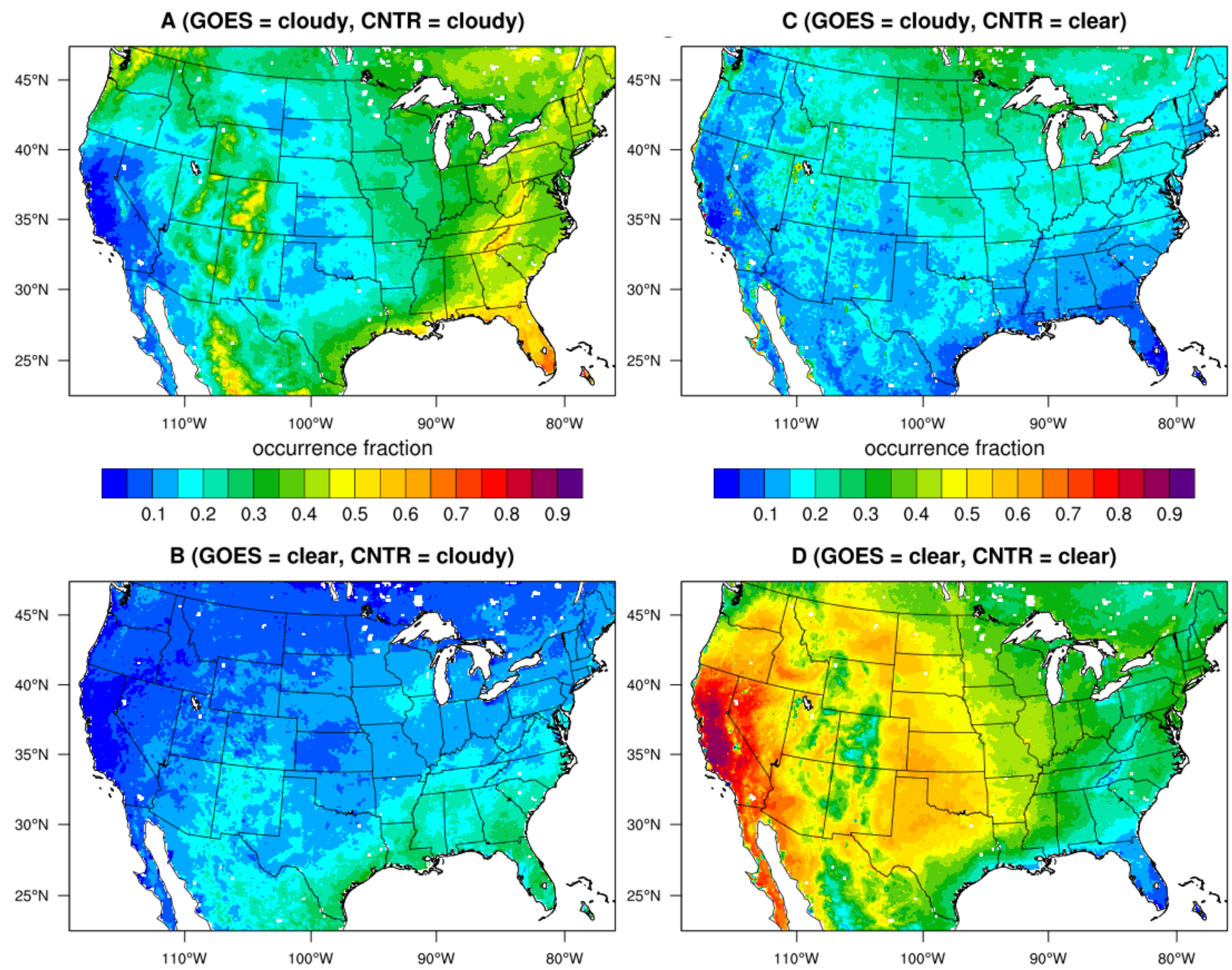
A = 27.1%

B = 12.4%

C = 17.5%

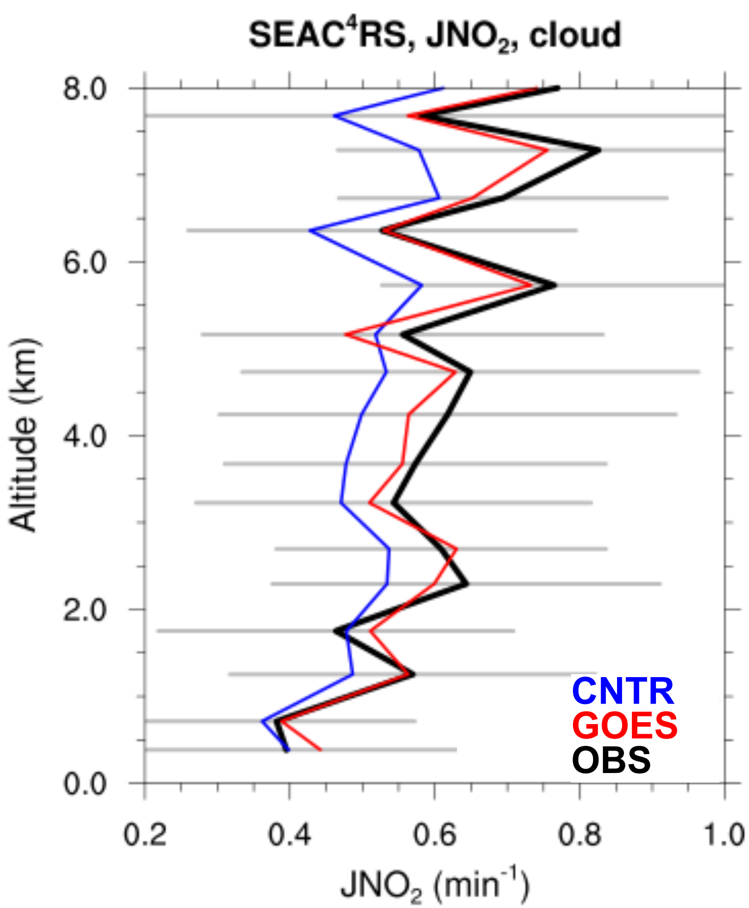
D = 43.0%

Probability of detection (POD), A/(A+C), = 60.8%



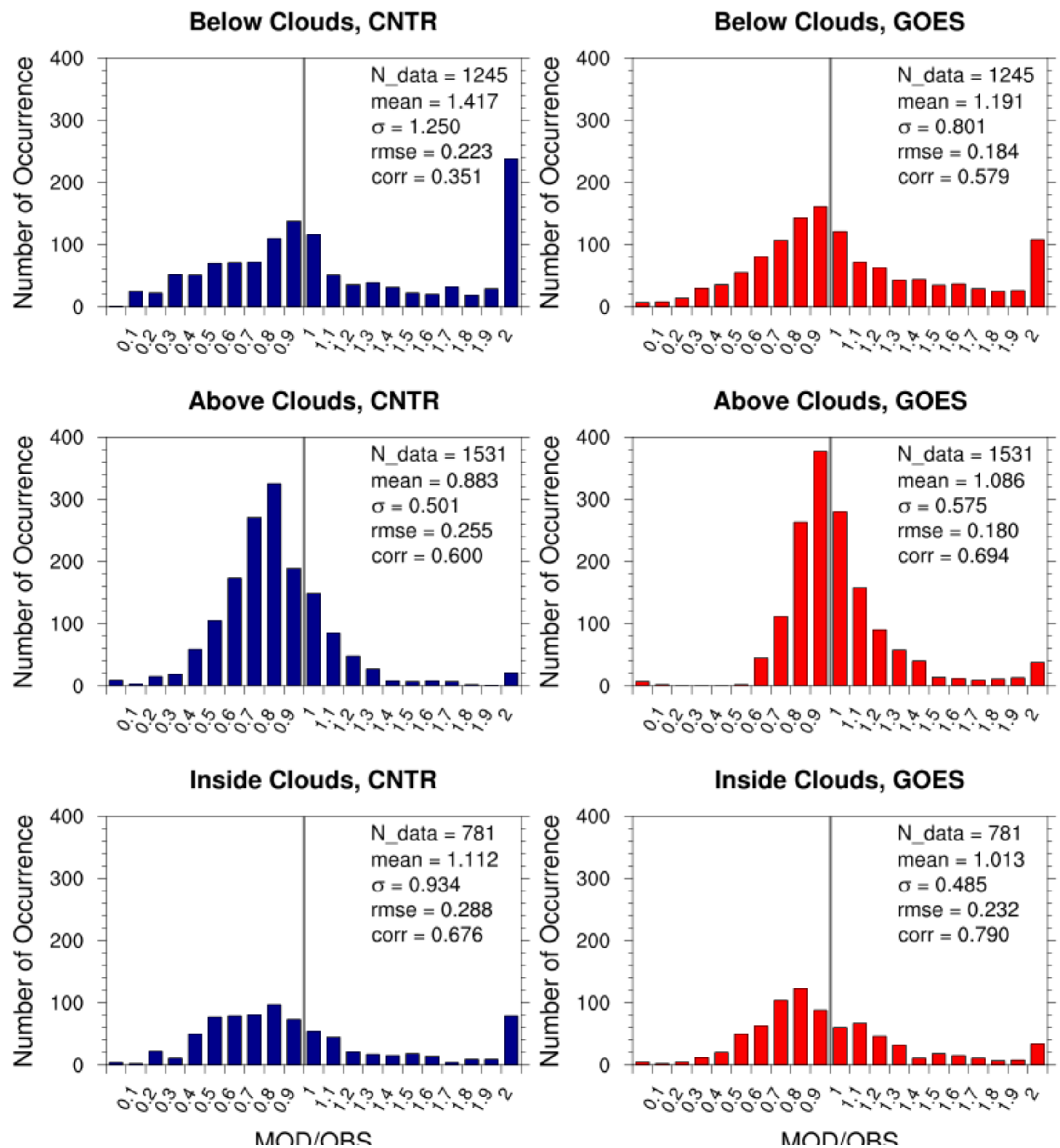
JNO_2 comparison with $SEAC^4RS$ data

Cloudy-sky average



Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys ($SEAC^4RS$)

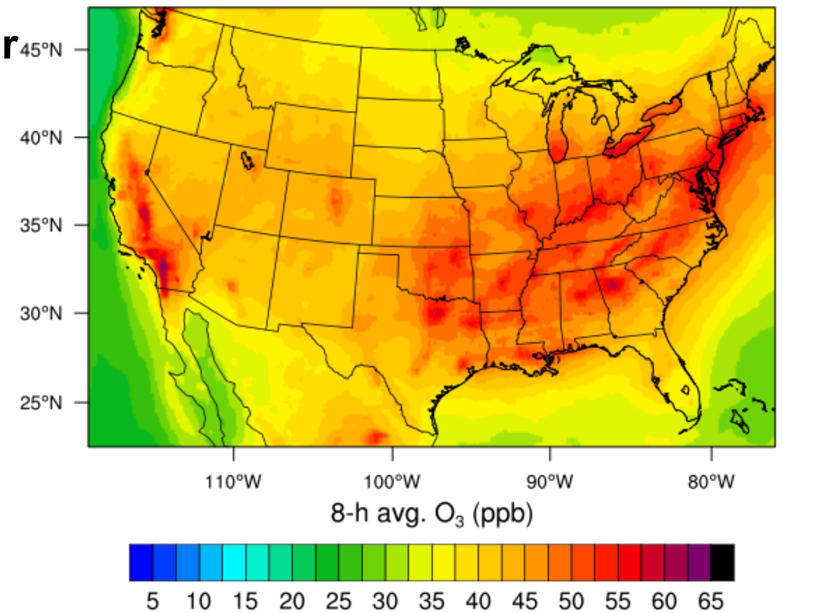
Ratio of Model JNO_2 to OBS JNO_2



Effects of cloud correction on O_3

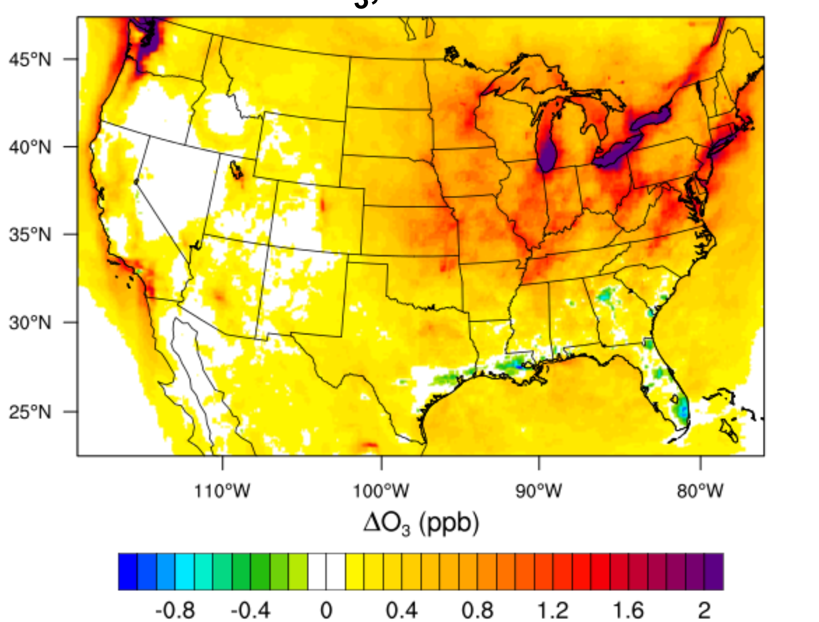
8-h Ozone (O_3) average, CNTR (Control)

average over 4 months



Difference in O_3, CNTR minus GOES

Max. diff. ~ 5 ppb



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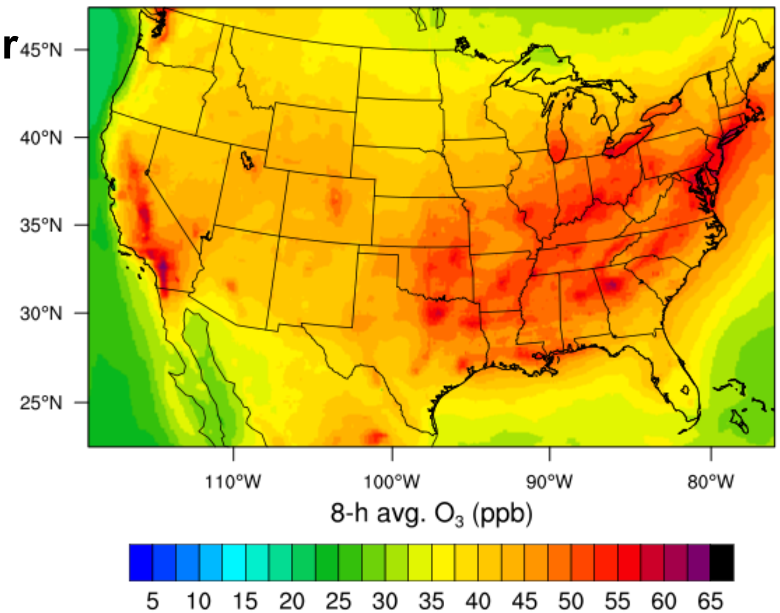
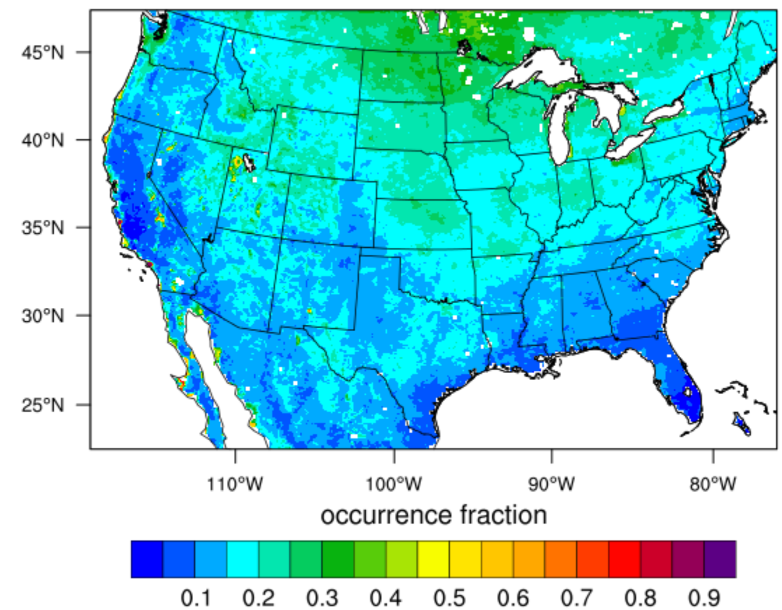


C (GOES = cloudy, CNTR = clear)



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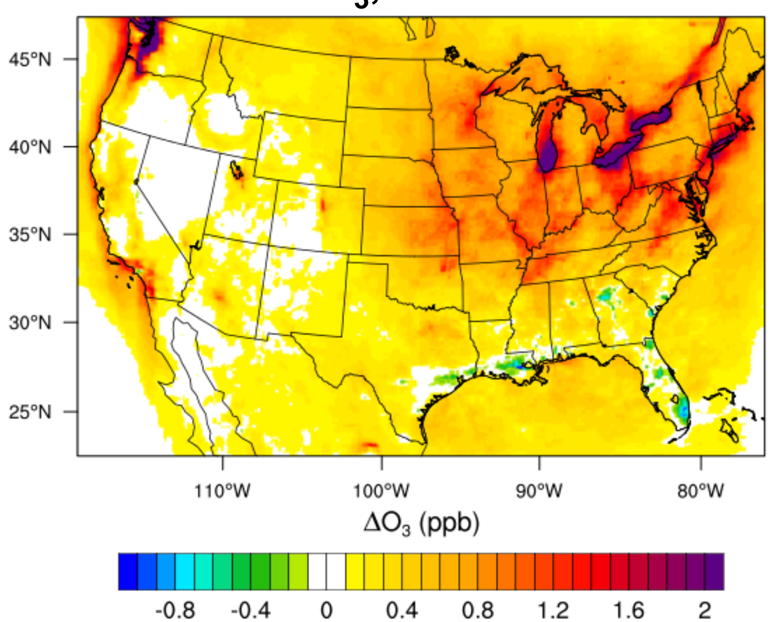
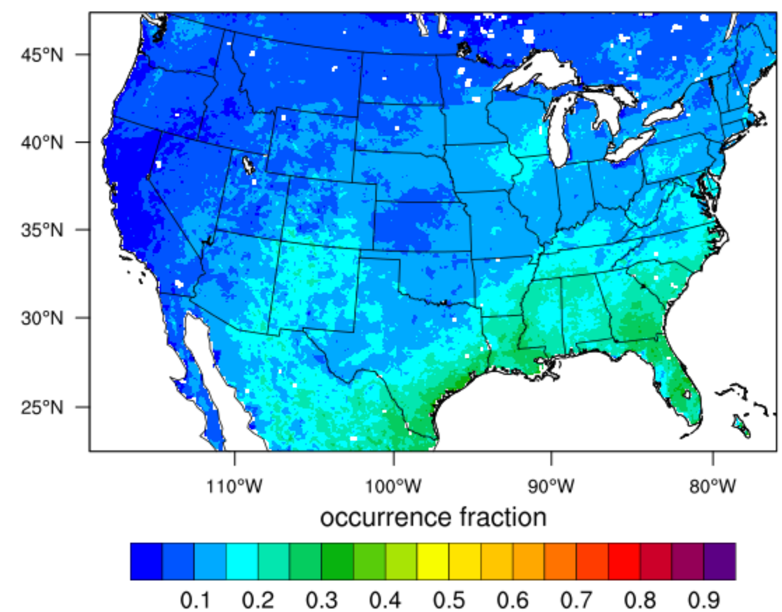


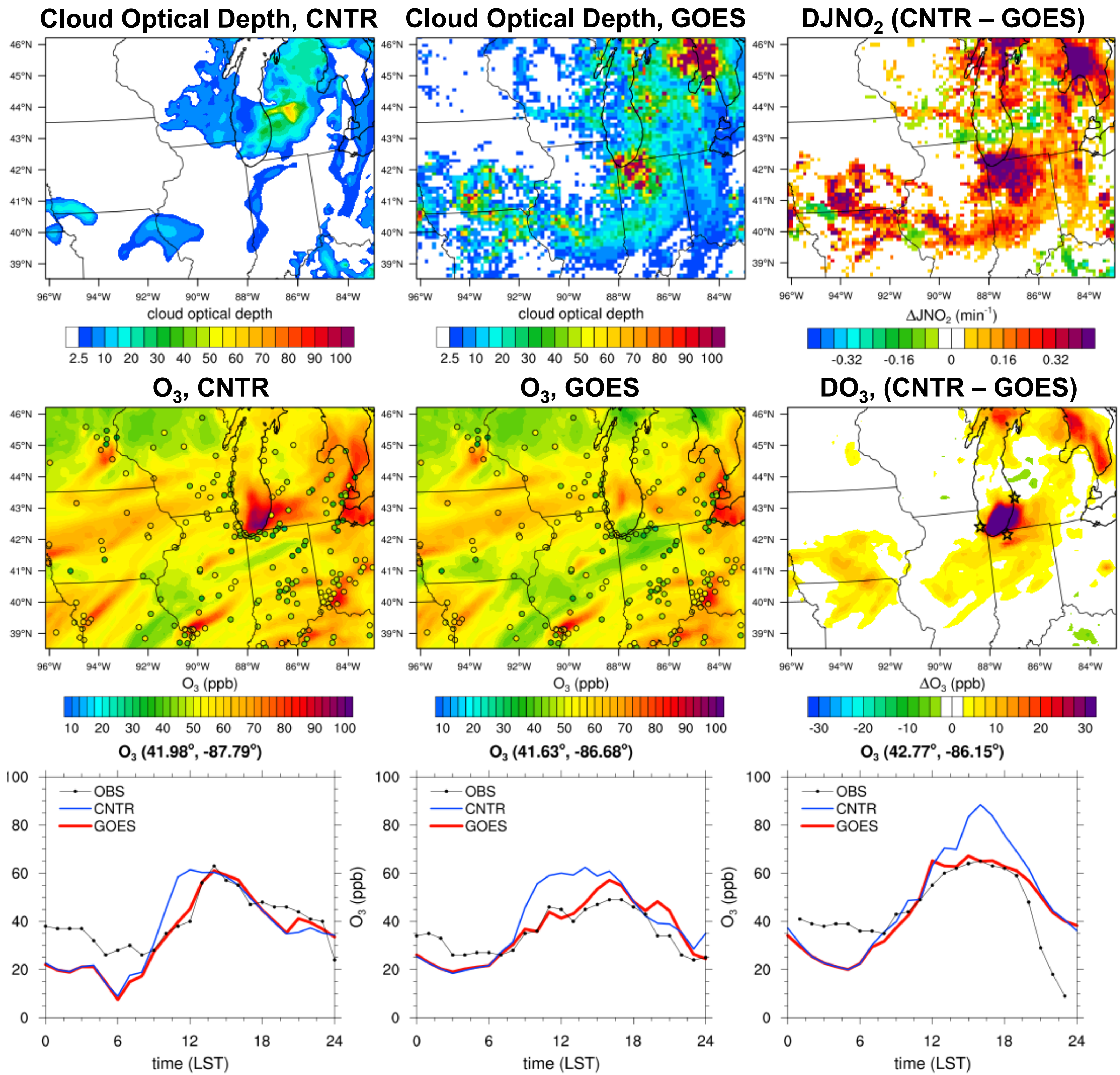
B (GOES = clear, CNTR = cloudy)



Example in Midwestern US

13 LST 8 July 2013

Maximum O_3 difference ~60 ppb

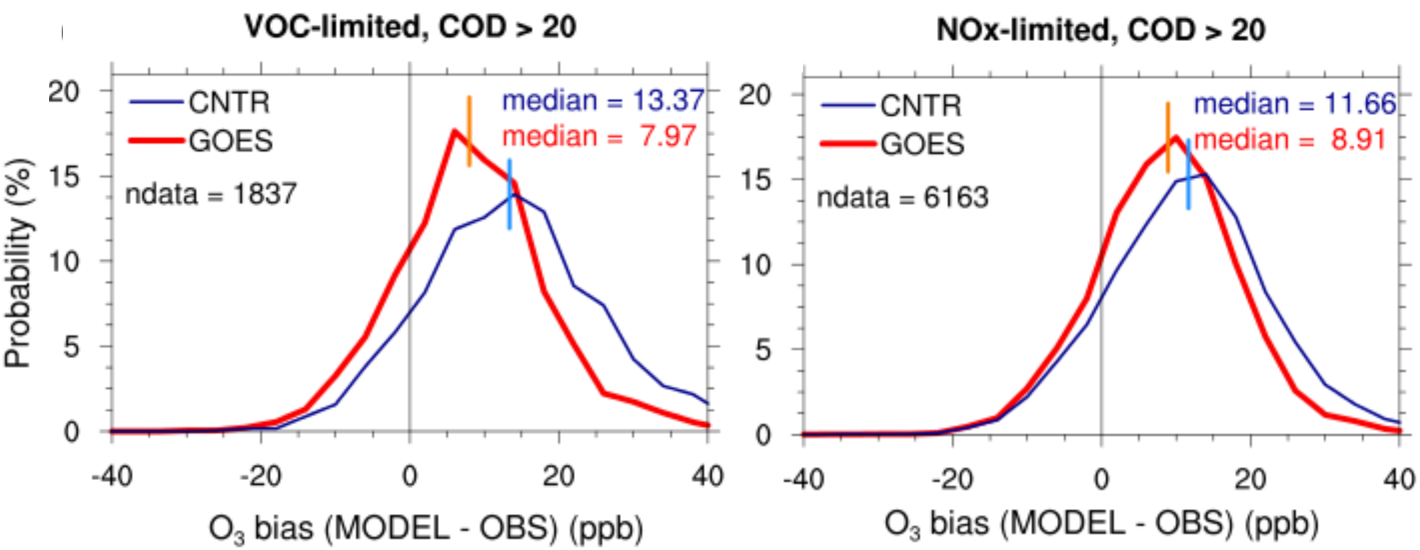


Reduction in 8-h O_3 bias due to clouds

- 8-h average O_3 bias is evaluated using EPA ground measurements for VOC-limited regimes and NOx-limited regimes.

$$\sum_{10\ LST}^{17\ LST} 1[COD > COD\ threshold] \geq 4\ (hour)$$

Cloudy sky conditions



- O_3 bias due to clouds reduced by 5.4 ppb in VOC-limited regimes, 2.75 ppb in NOx-limited regimes under cloudy sky conditions with COD threshold of 20
- Larger bias reduction in VOC-limited regimes

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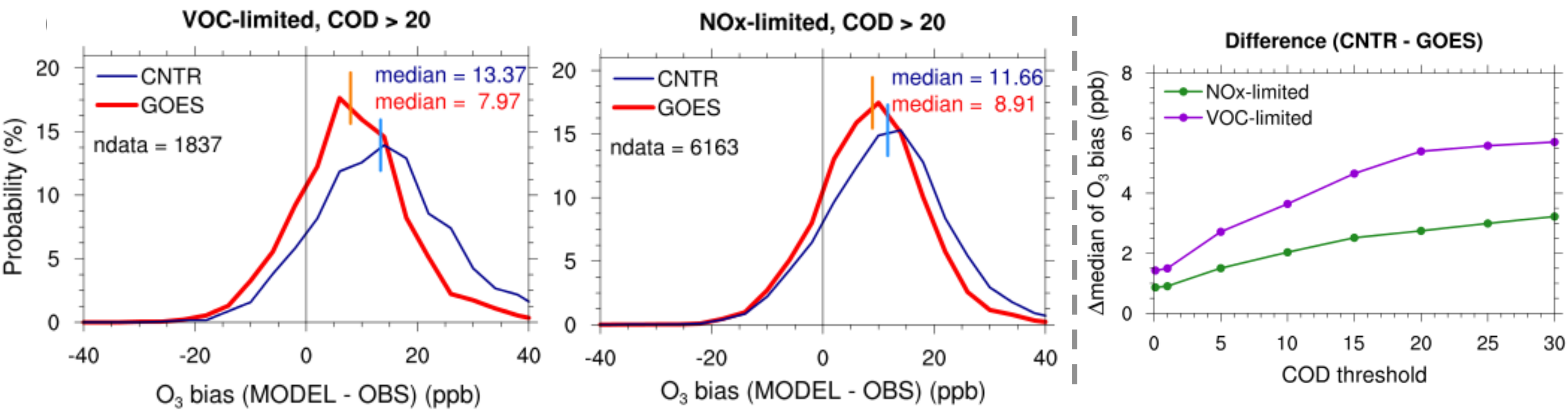

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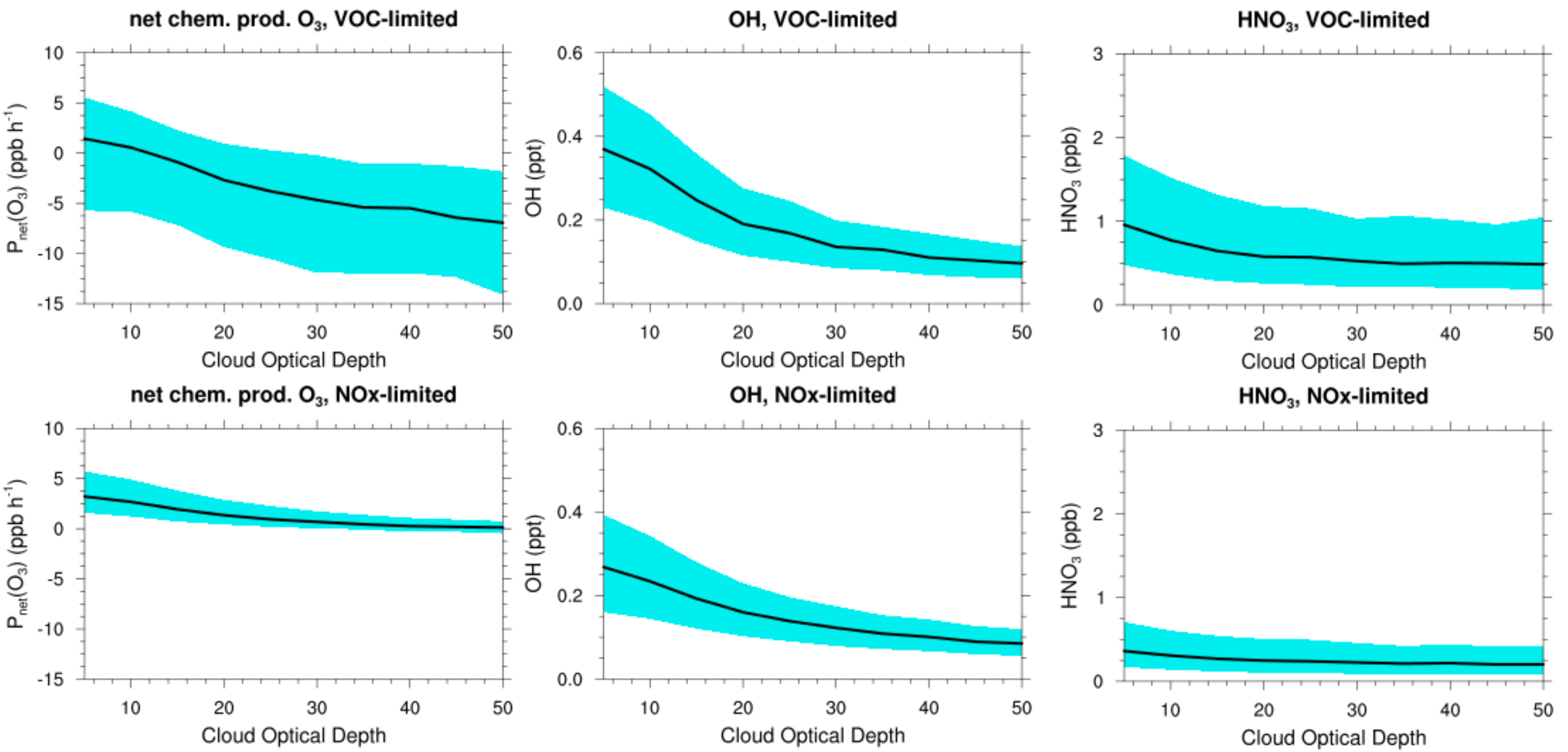
O_3 formation, OH, and HNO_3 with COD

Black line: median, cyan shading: 25 & 75 quartiles



- Primary OH formation decreases as COD increases due to decreasing JO1D.

VOC-limited (high NOx)

- OH reacts with NO_2 and is removed.
- Low HO_2 (& RO_2)
- Low O_3 formation

NOx-limited (low NOx)

- OH mainly reacts with VOCs producing HO_2/RO_2
- Considerable secondary OH sources ($HO_2 + NO \rightarrow OH + NO_2$), which reaches a maximum at NOx ~ 1ppb (Weinstock et al. 1981)

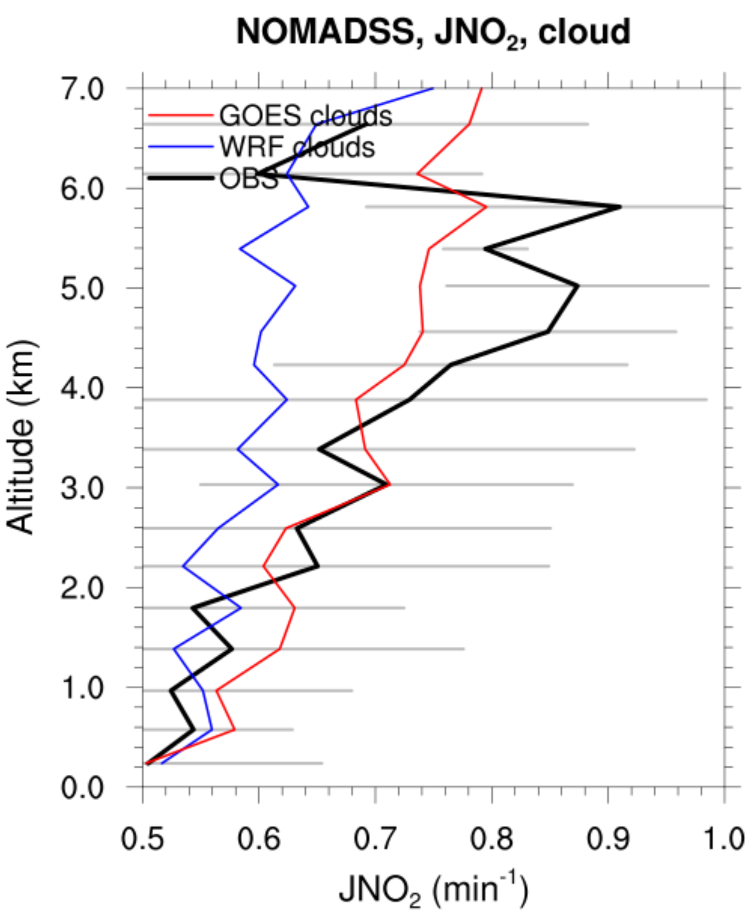
Summary and next step

- Satellite corrected photolysis rates and their vertical profiles show better agreements with airborne data.
- Satellite corrected photolysis rates are found to reduce O_3 biases by 1–6 ppb on average at the ground level.
- The reduction in O_3 (and bias) due to reduced radiation (due to clouds) is greater in VOC-limited regimes than NOx-limited regimes.
- In other words, more accurate cloud predictions would benefit more accurate O_3 predictions in VOC-limited regimes (polluted regions).
- Evaluate the effects and benefits of assimilation of satellite-derived clouds on O_3 predictions

Thank you!

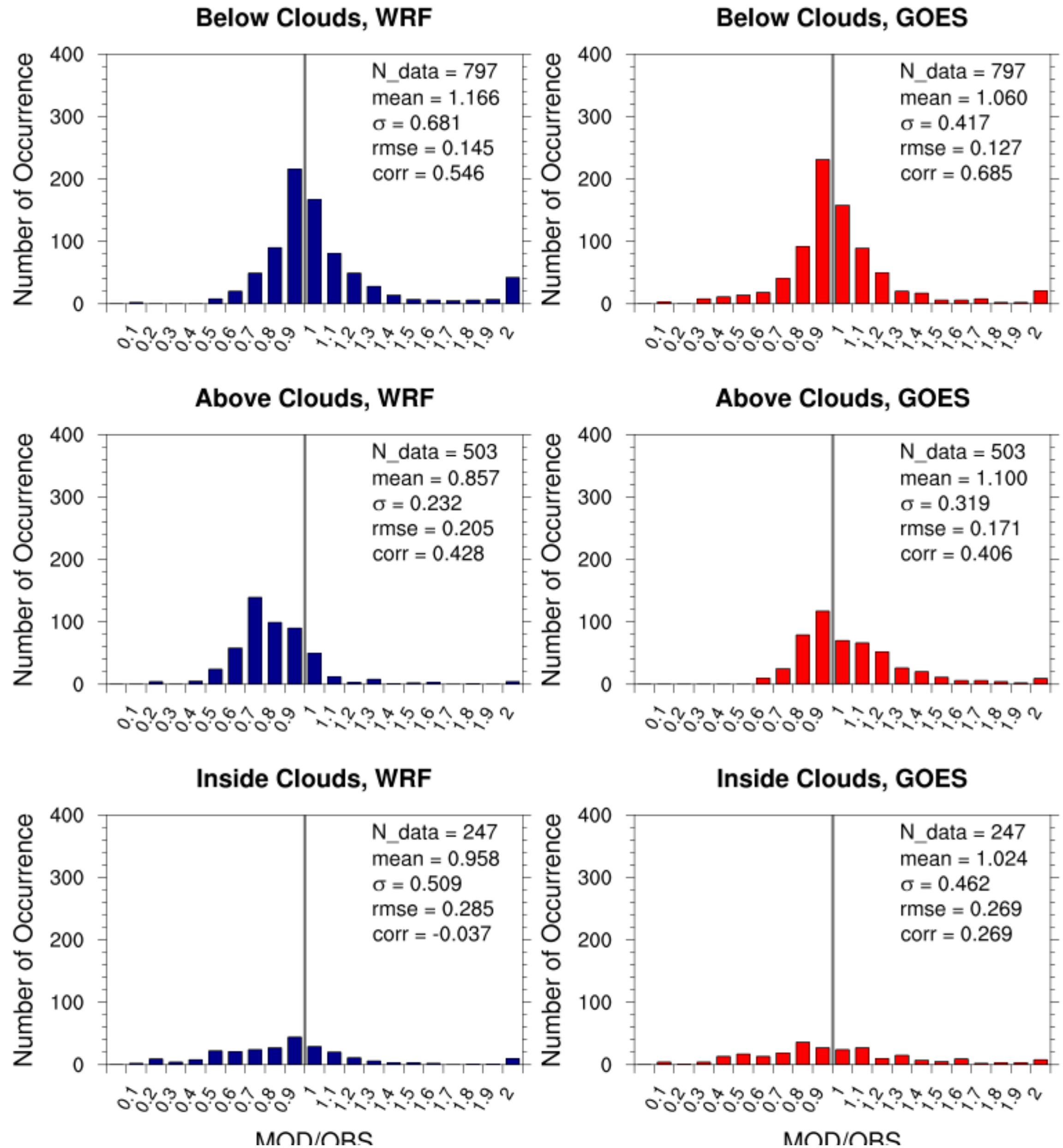
Comparison with NOMADSS campaign

Cloudy-sky average



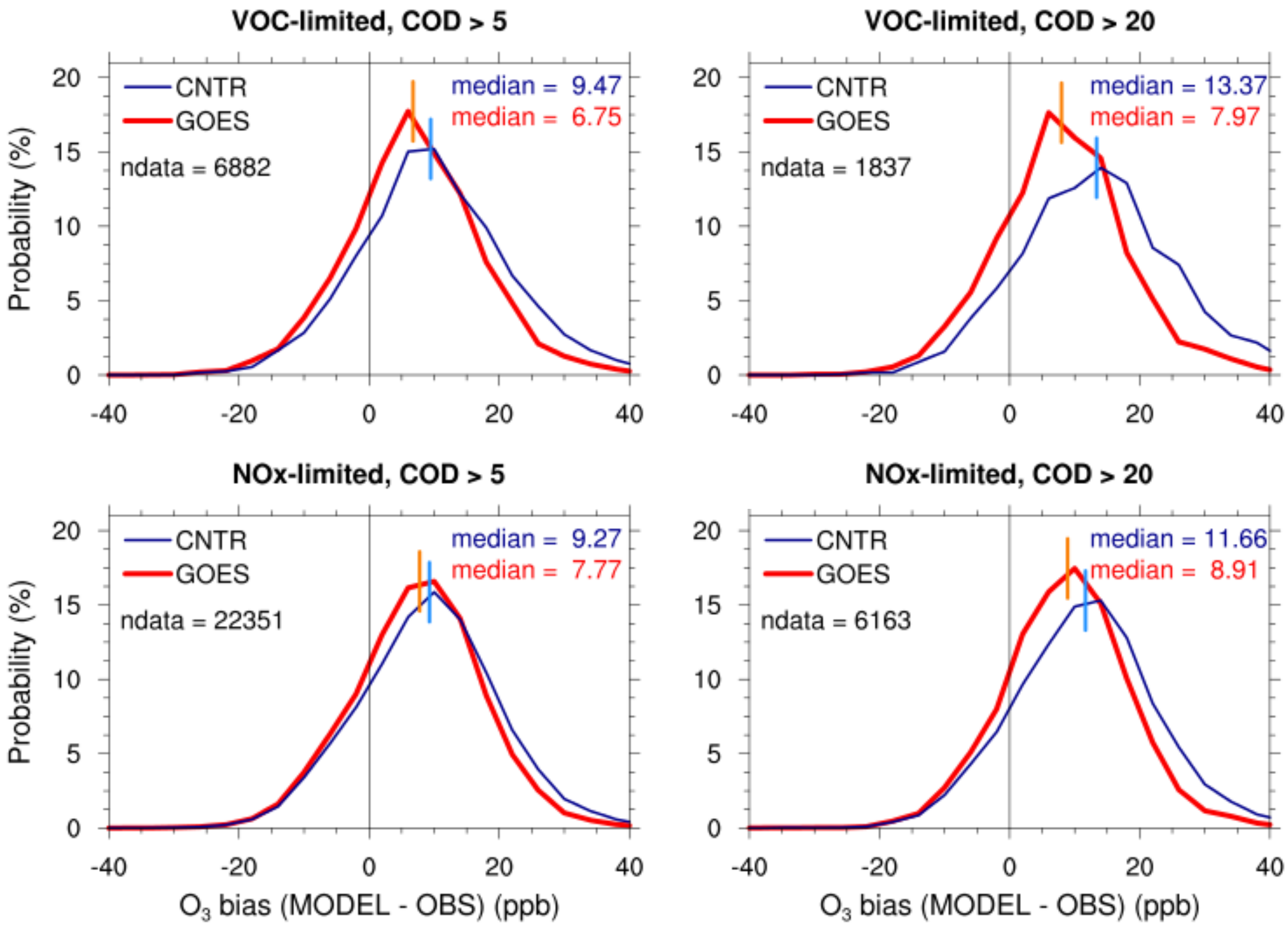
Nitrogen, Oxidants, Mercury and Aerosol Distributions, Sources and Sinks (NOMADSS)

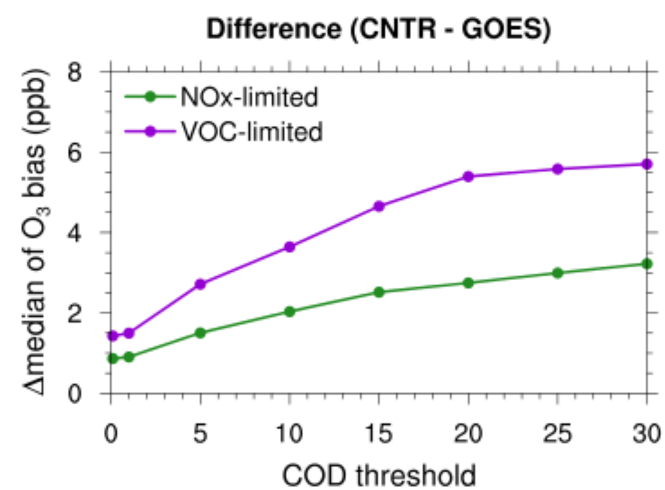
Ratio of Model JNO_2 to OBS JNO_2



Effects on 8-h O_3 bias

Cloudy sky conditions $\sum_{10}^{17} 1[COD>COD\ threshold]/8 \geq 0.5$

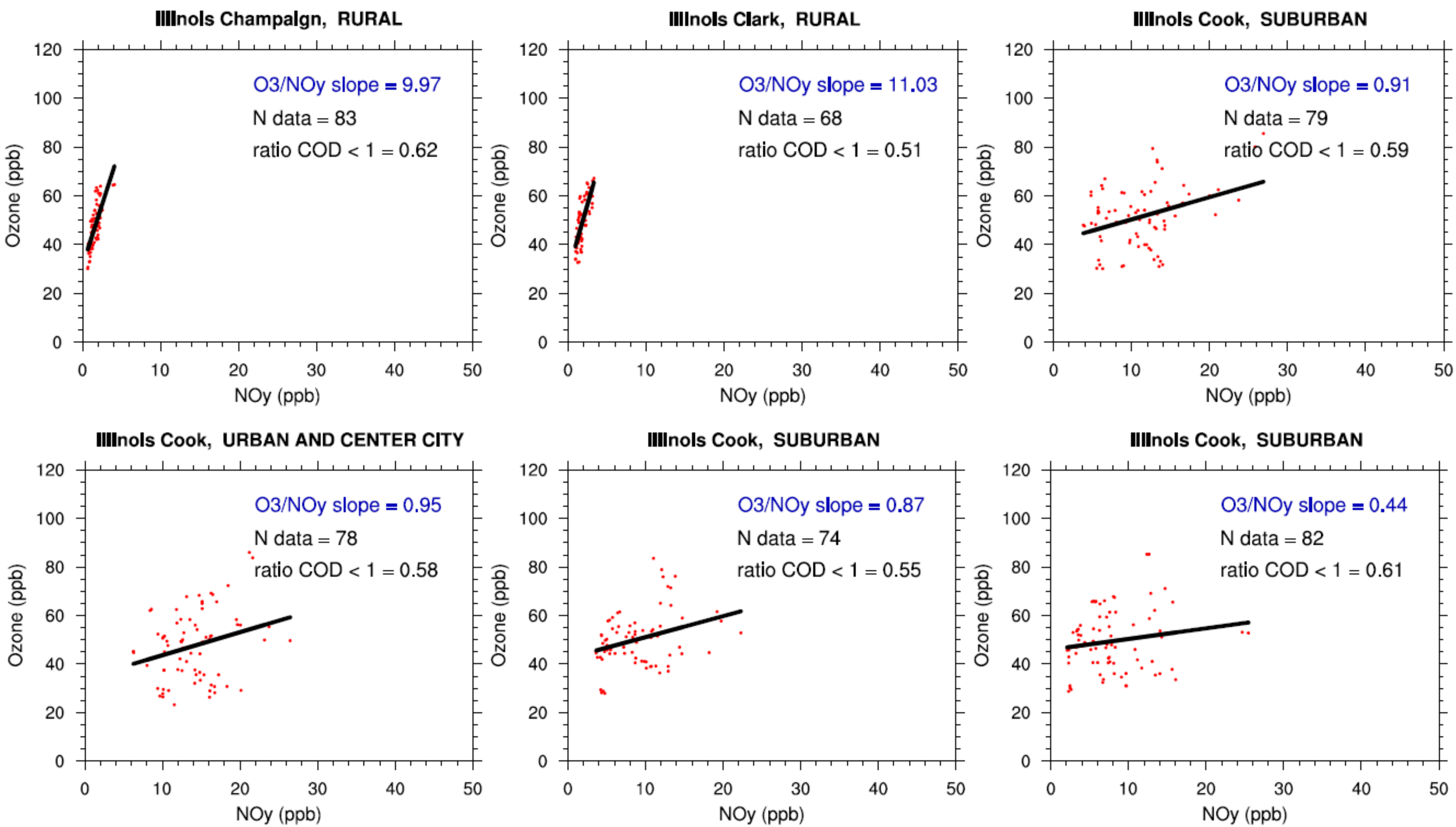




- O_3 bias due to clouds reduced by 2.7 ppb in VOC-limited regimes, 1.5 ppb in NOx-limited regimes under cloudy sky conditions with COD threshold of 5
- Larger bias reduction in VOC-limited regimes

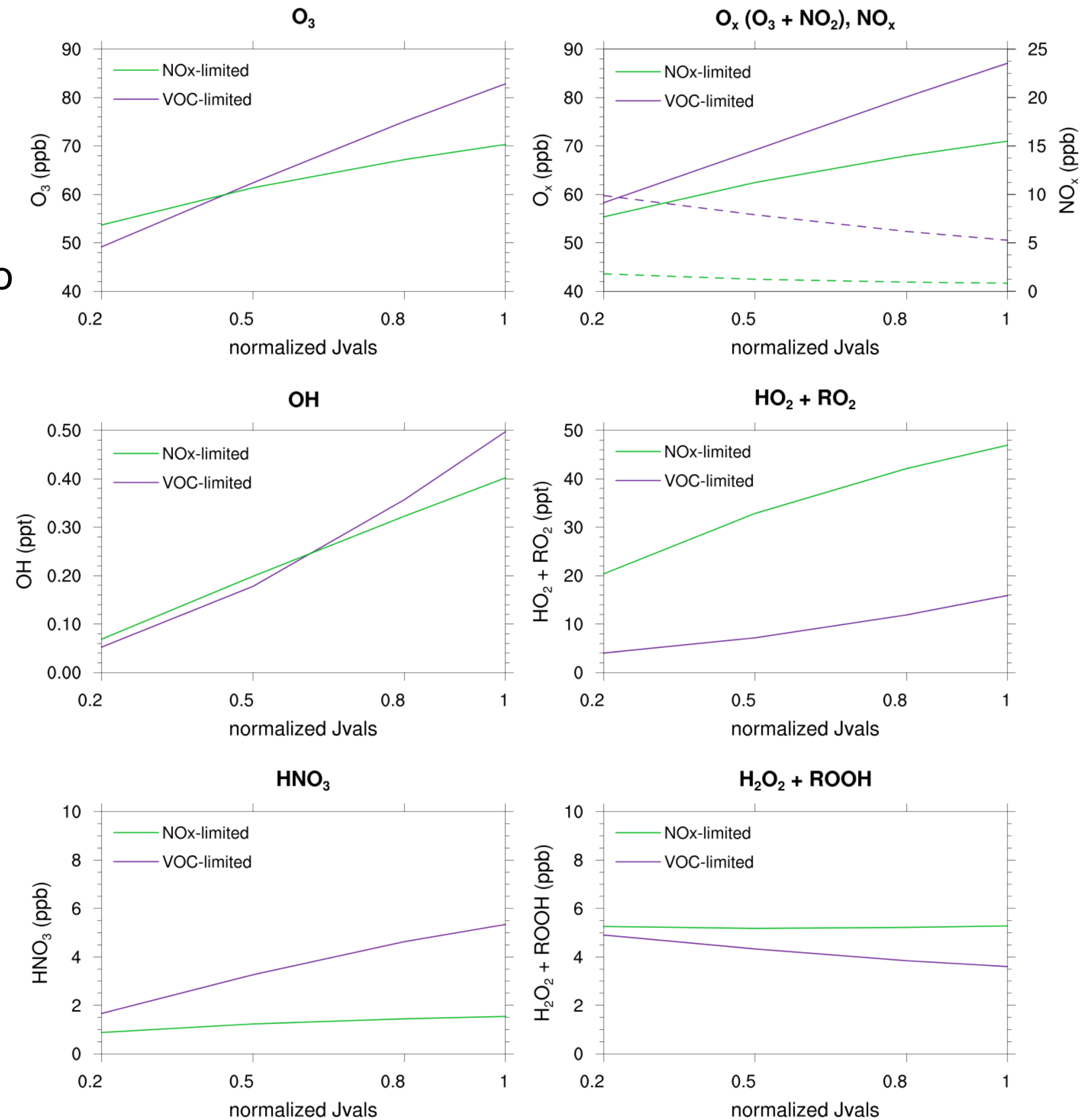
VOC-limited/NOX-limited regimes

- For EPA ground stations, ratio $\Delta O_3/\Delta NO_y$ is used to identify the sensitivity regime.
- NOx-VOC transition: $\Delta O_3/\Delta NO_y$ = 4–6 (Sillman and He, 2002)

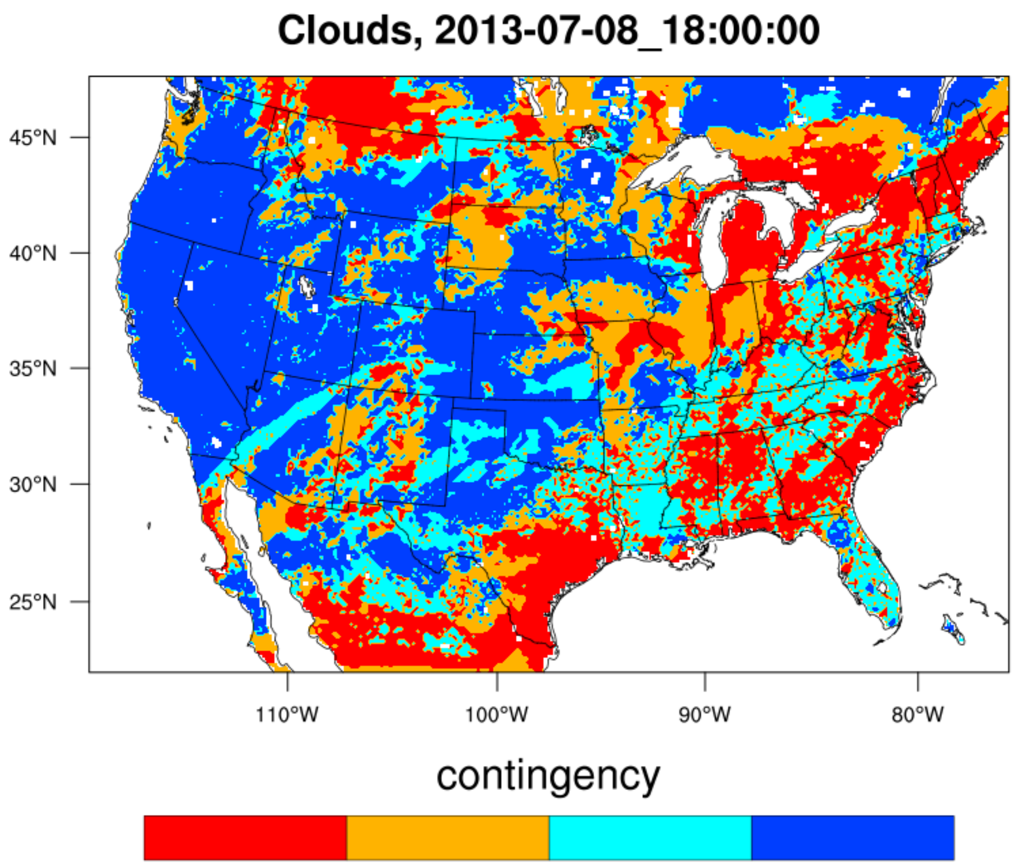


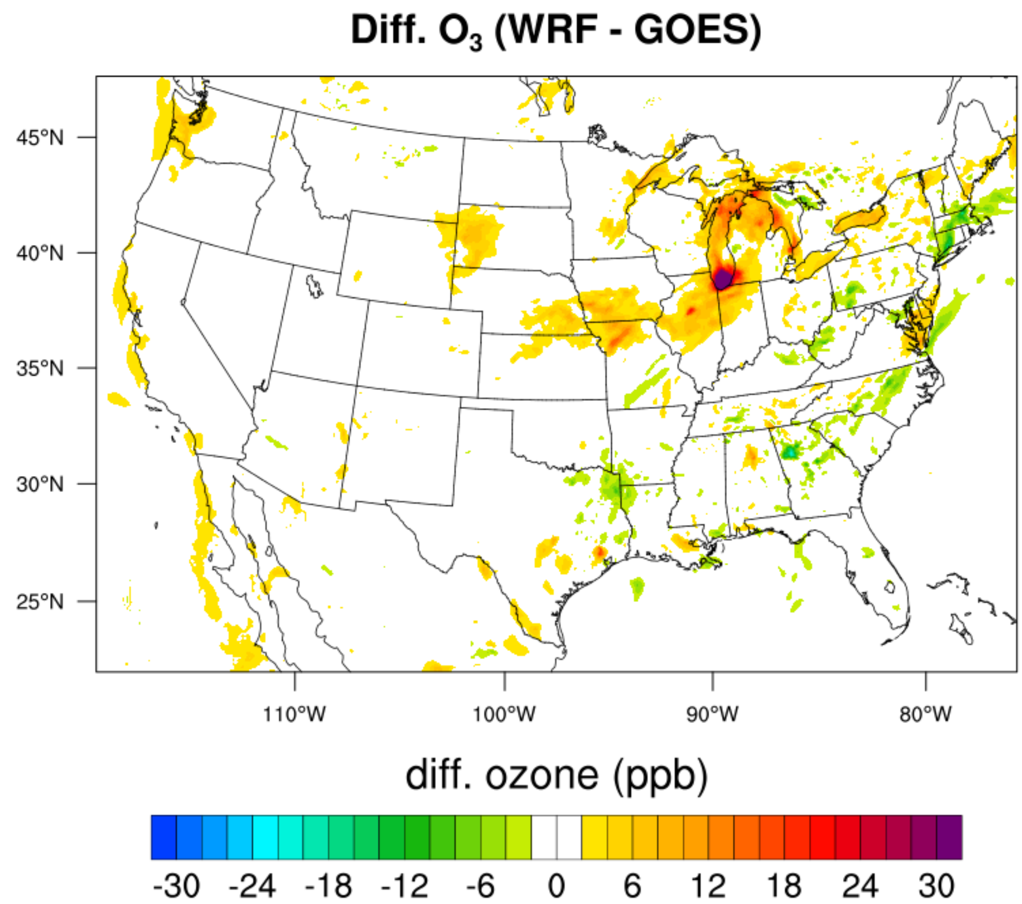
Test with a box model (BOXMOX)

- Same chemical mechanism (MOZART-4)
- Initial conditions/Emissions from WRFChem
- VOC-limited: Chicago urban area
- NOx-limited: Subruban of Chicago
- 12-hour runs



Effects on ground-level ozone

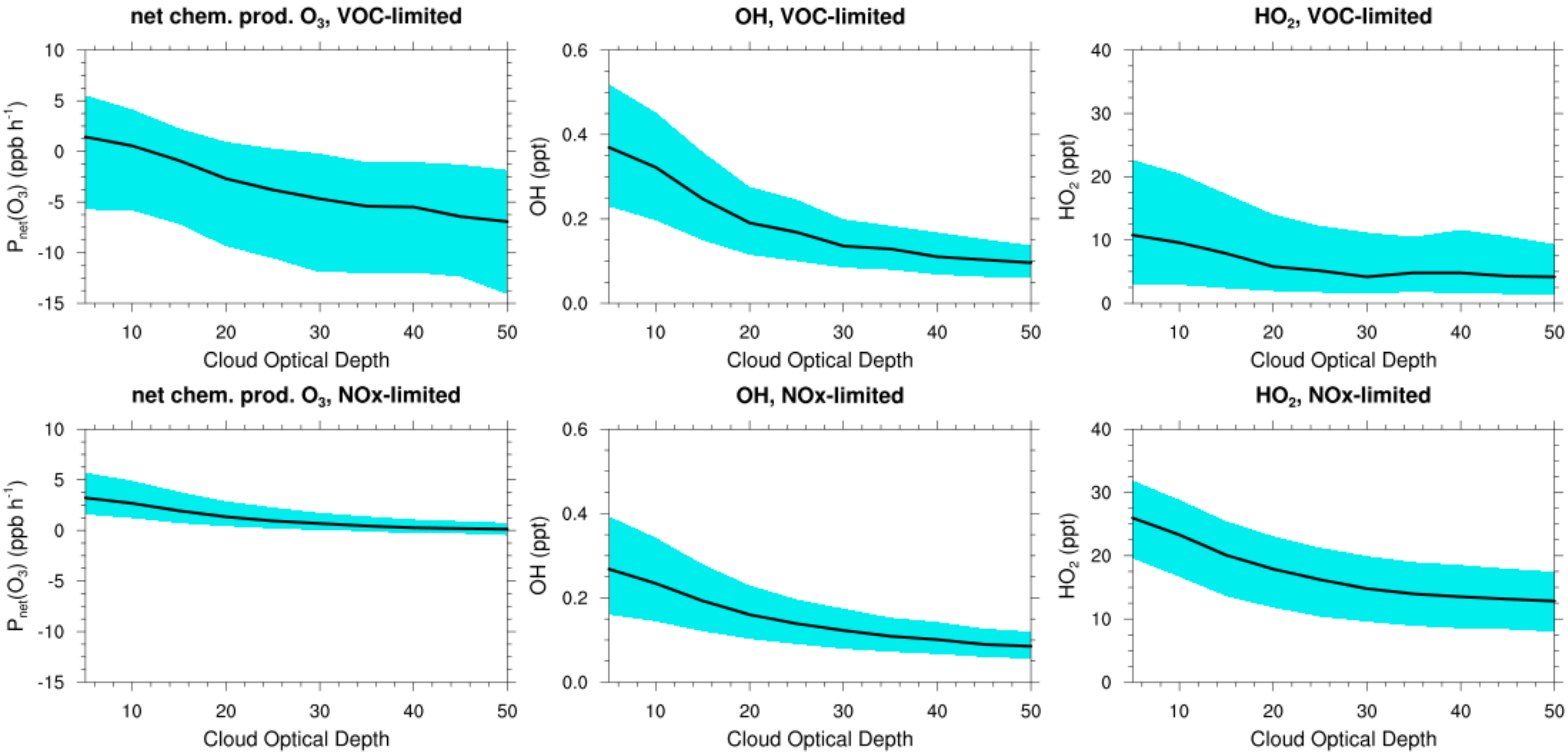




		WRF	
		cloudy	clear
GOES	cloudy	A	B
	clear	C	D

Effects on O_3 formation and radicals

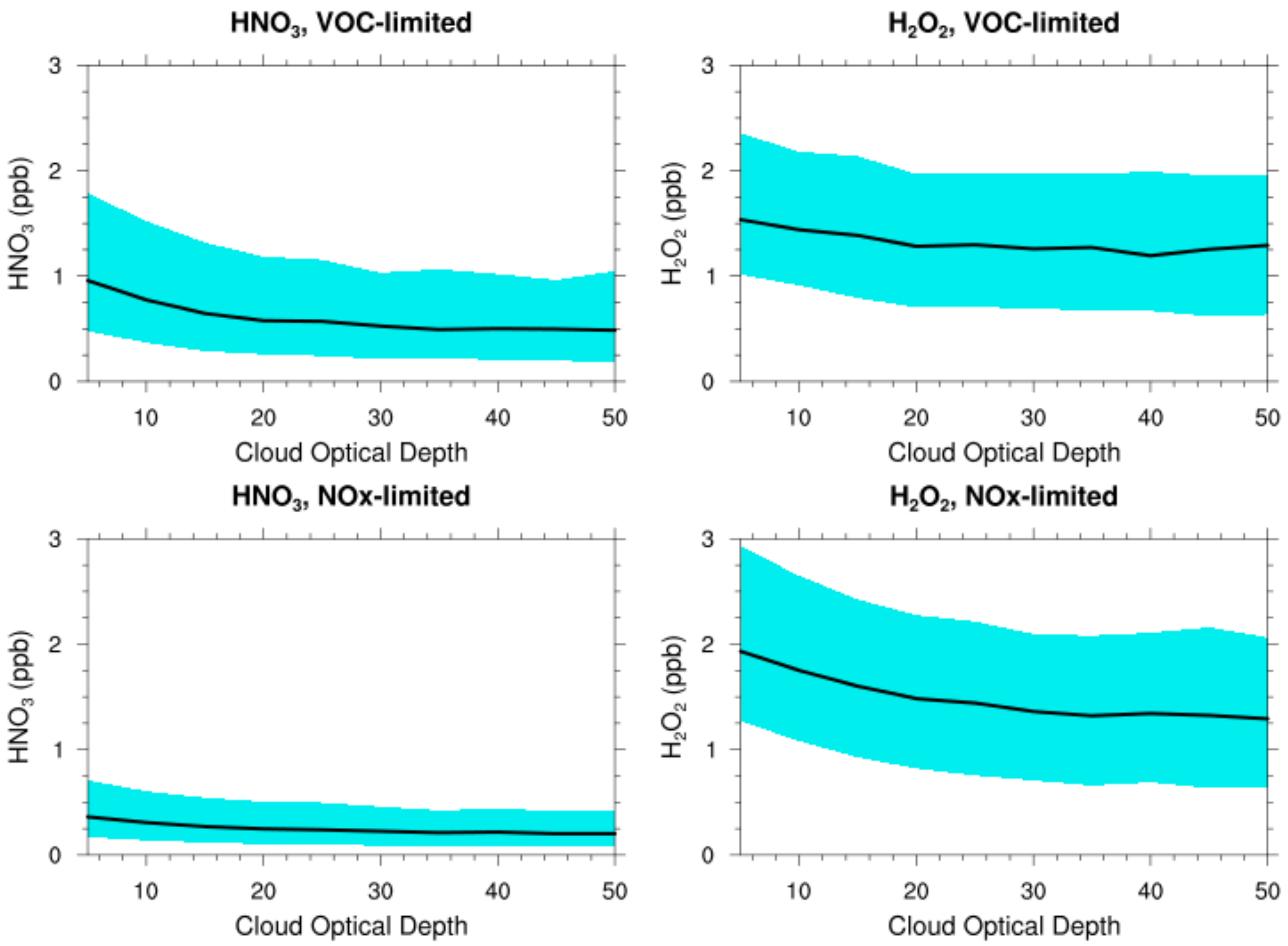
Black line: median, cyan shading: 25 & 75 quartiles



- Primary OH formation from O1D + H2O is linearly dependent on JO1D.
- Larger sensitivity of O_3 formation and OH radical to changes in COD in VOC-limited regimes

Effects on radical sinks

Black line: median, cyan shading: 25 & 75 quartiles



- VOC-limited regime: formation of HNO_3 is the major sink of HOx radicals
- NOx-limited regime: formation of H_2O_2 (& ROOH) is the major sink of HOx radicals

12-km WRFChem simulations

- WRFChem V3.6.1, a single domain over CONUS
- Simulation period: June–September 2013, Horizontal grid size: 12 km
- Chemistry mechanism: MOZART_MOSAIC_KPP
- Meteorology: reinitialized every 2 days using FNL reanalysis (6-hr spin up is allowed in each 2-day simulation) & No nudging
- Chemistry: cycled/continuous

- Control (CNTR) simulation uses WRF-generated clouds.
- GOES simulation uses GOES (Geostationary Operational Environmental Satellite) clouds
    - Available originally 8-km horizontal resolution every hour from 11 June 2013
    - In the GOES simulation, GOES cloud bottom/top height and cloud optical depth retrievals are used and updated every hour.
    - The GOES clouds are used instead of WRF-generated clouds **only in the photolysis computations (TUV in WRF-Chem)**.
    - The GOES clouds are not used in other dynamics and physical processes such as atmospheric radiation and microphysics.