Assessing cloud radiative effects on photolysis rates and key oxidants during aircraft campaigns using satellite cloud observations and a global CTM

Bo Zhang[1], Hongyu Liu[1], James H. Crawford[2], Gao Chen[2], Apostolos Voulgarakis[3], T. Duncan Fairlie[2], Bryan Duncan[4], Seung-Hee Ham[5], Seiji Kato[2], Melissa P. Sulprizio[6], and Robert M. Yantosca[6]

[1] National Institute of Aerospace, Hampton, VA, USA
[2] NASA Langley Research Center, Hampton, VA, USA
[3] Imperial College, London, UK
[4] NASA Goddard Space Flight Center, Greenbelt, MD, USA
[5] Science Systems and Applications, Inc., Hampton, VA, USA
[6] Harvard University, Cambridge, MA, USA

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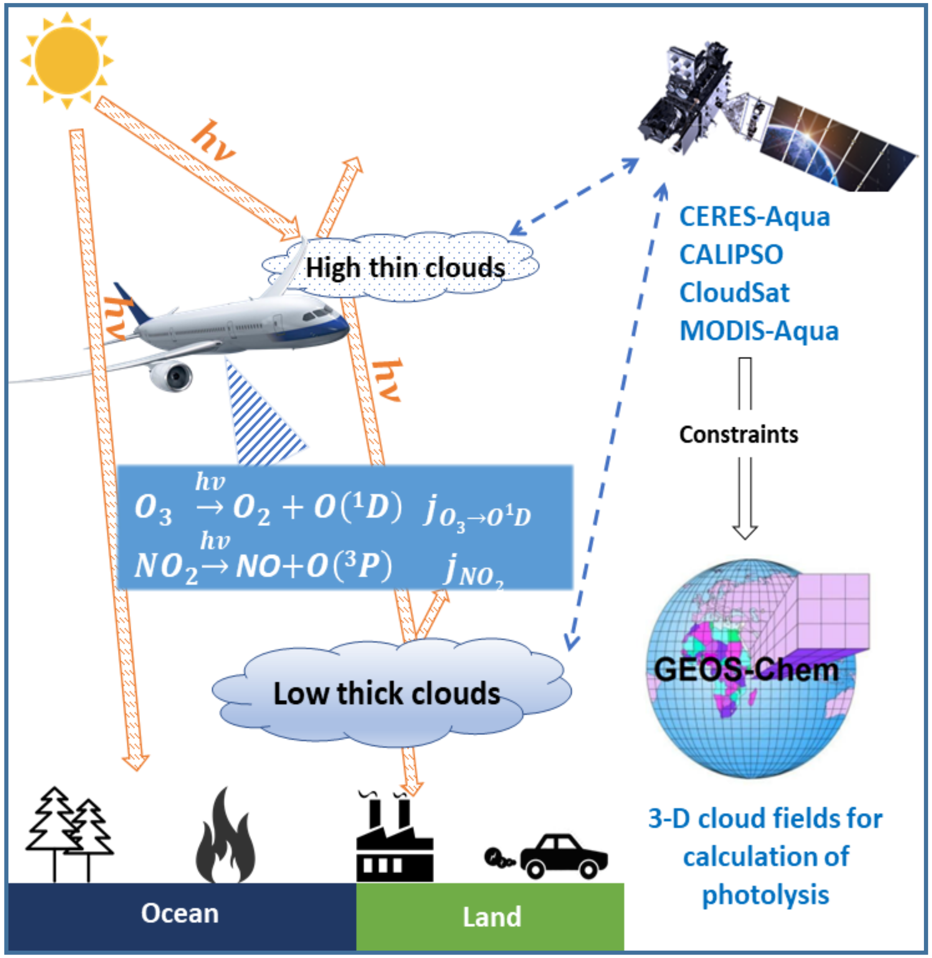
Cloud Radiative Effects (CRE) on Photolysis



- J[O^1D] (< 320 nm) and J[NO_2] (< 420 nm) are the most important photolysis rates in tropospheric ozone photochemistry.
- Clouds mainly scatter solar radiation. Actinic flux and photolysis rates are increased above and in the upper layers of clouds but reduced below thick clouds.
- Uncertainties/errors in model cloud distributions are substantial.
- Increasing availability of cloud observations from satellites provides strong constraints on cloud distributions and thus radiative effects in chemical transport models.

Focus of this work

1. Evaluate model simulated J[O^1D] and J[NO_2] with airborne measurements, and quantify the radiative effects due to clouds.
2. Examine the potential of using satellite (A-Train) 3D cloud observations to improve model calculations of photolysis rates.

Calculation of Photolysis in GEOS-Chem

GEOS-Chem CTM (v9.2, http://geos-chem.org/)

- Horizontal resolution 2°×2.5°, 47 levels in vertical.
- Driven by the MERRA reanalysis from NASA GMAO.
- Ozone-NOx-CO-VOC coupled to aerosol chemistry [e.g., Park et al., 2004].

Fast-J radiative transfer algorithm [Wild et al., 2000]

- Cross-sections and quantum yields for only 7 tropospherically-important wavelength bins. Accuracy of photolysis rates within 3%.
- Rayleigh scattering and Mie scattering by aerosols and clouds.
- Uses in cloud optical depth and cloud fraction from MERRA.
- monthly-mean TOMS ozone column.

Cloud vertical overlap

- Approximate Random Overlap (RAN) [Briegleb, 1992].

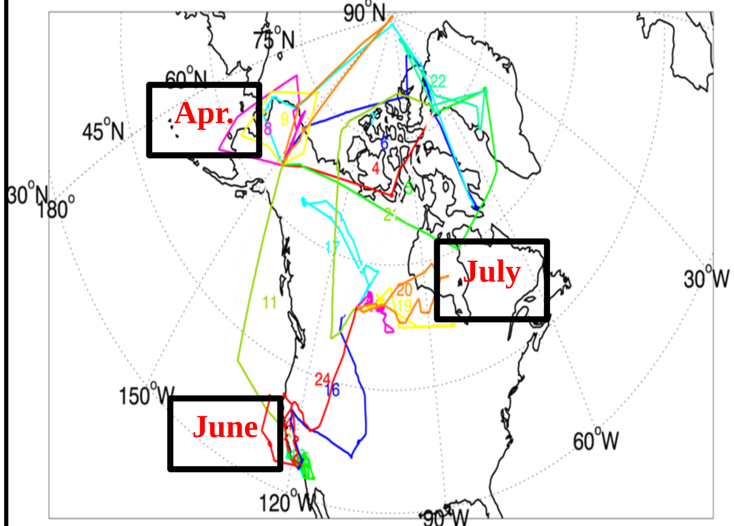
$$\tau_c' = \tau_c \times f^{1.5}$$

effective gridbox COD = in-cloud COD × cloud fraction$^{1.5}$

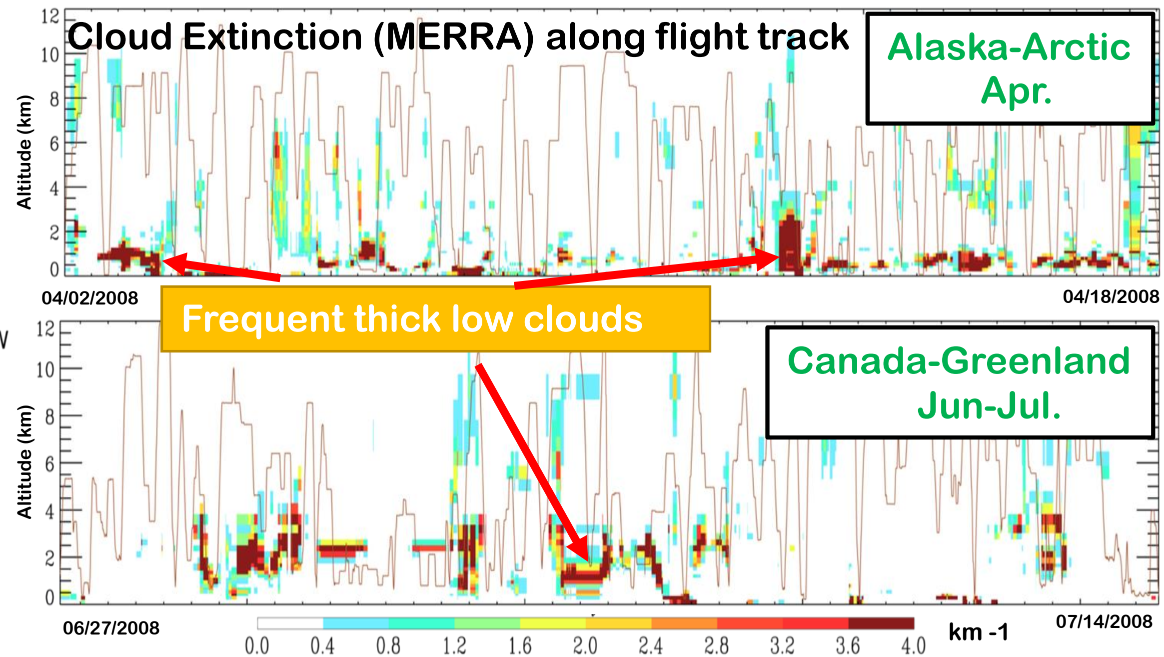
- RAN - default option in GEOS-Chem [Liu et al., 2006, 2009].

NASA Aircraft Campaigns: ARCTAS and SEAC4RS

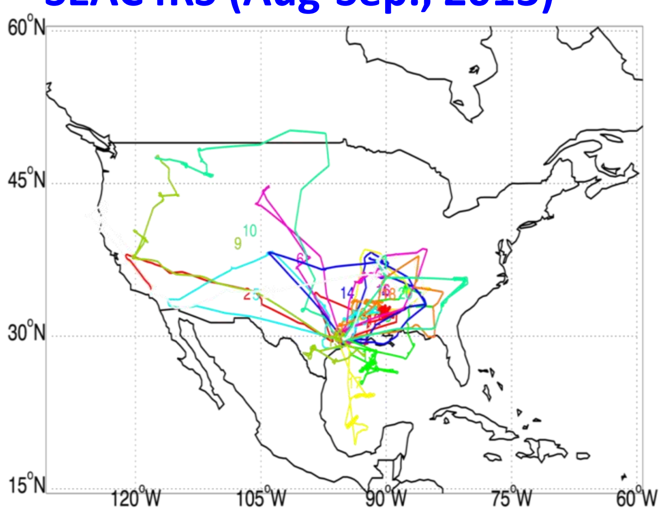
ARCTAS (Apr., Jun-Jul., 2008)



1-min data # 10,283



SEAC4RS (Aug-Sep., 2013)



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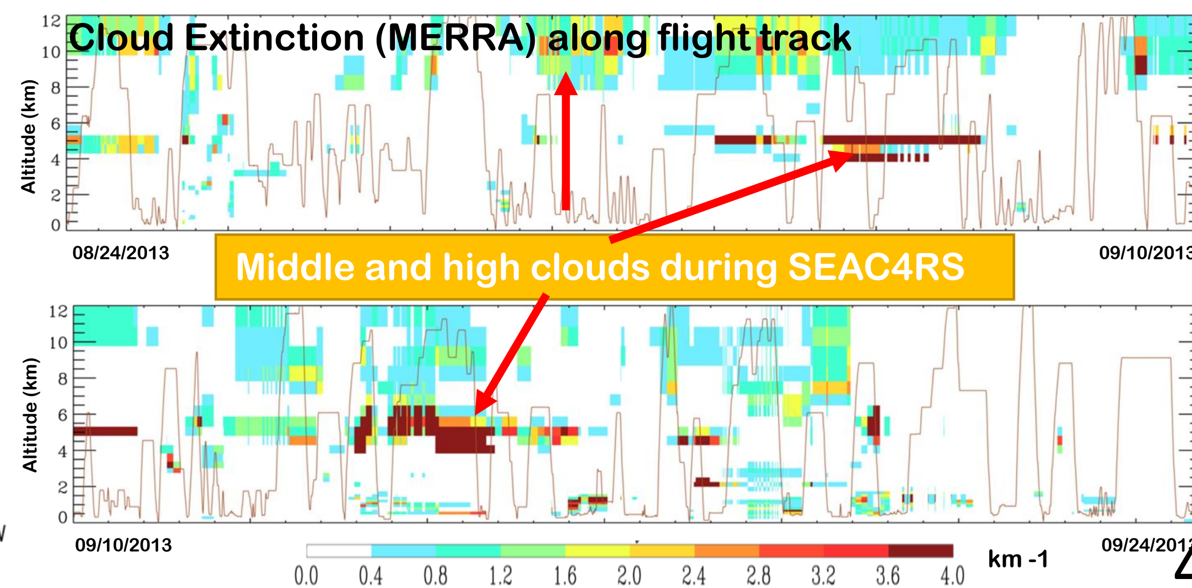
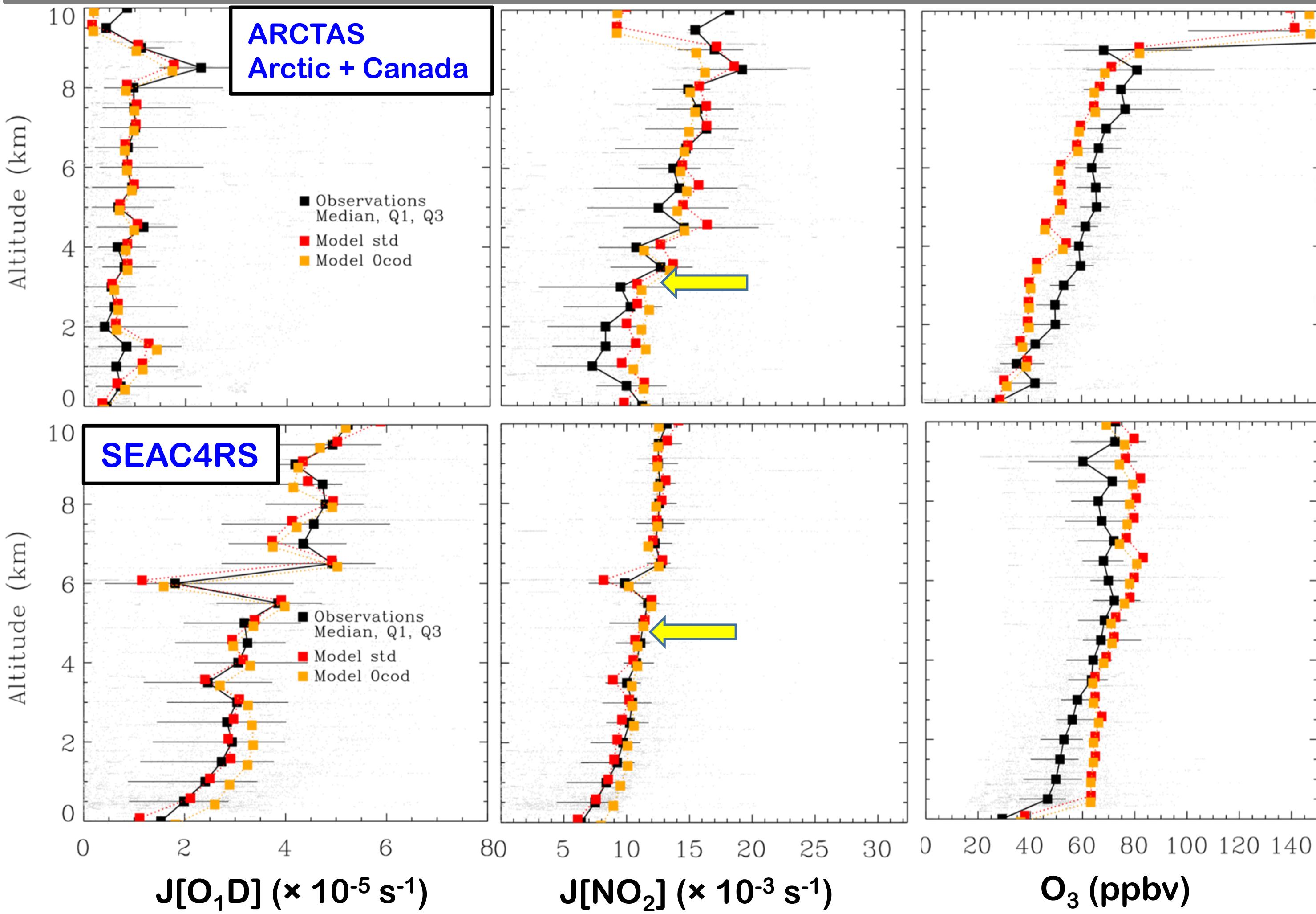
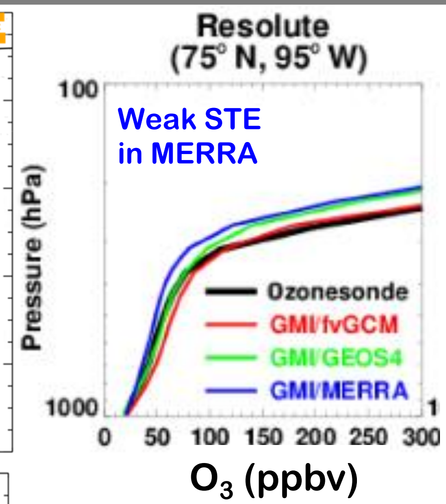


Cloud Radiative Effects on Photolysis during Campaigns

Observations | Model with MERRA COD (std) | Model with zero COD (0COD) | Median, Quartiles 1&3



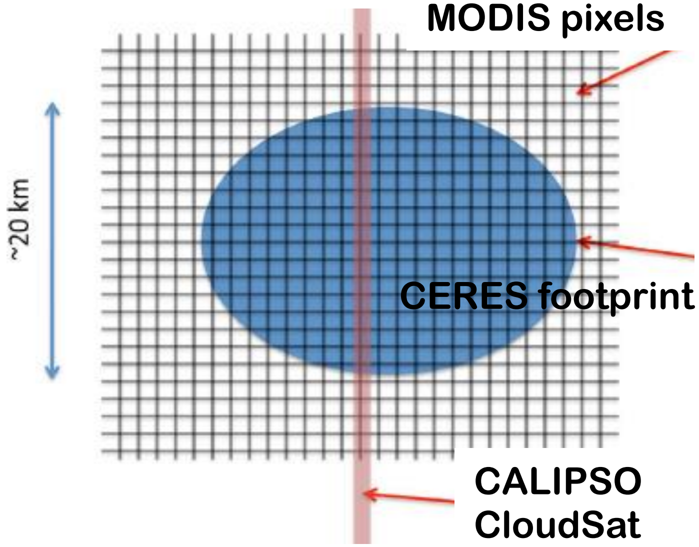


Choi, Liu et al., ACP 2017, Global O_3-CO

- Underestimated ozone at Arctic likely due to weak STE in MERRA.

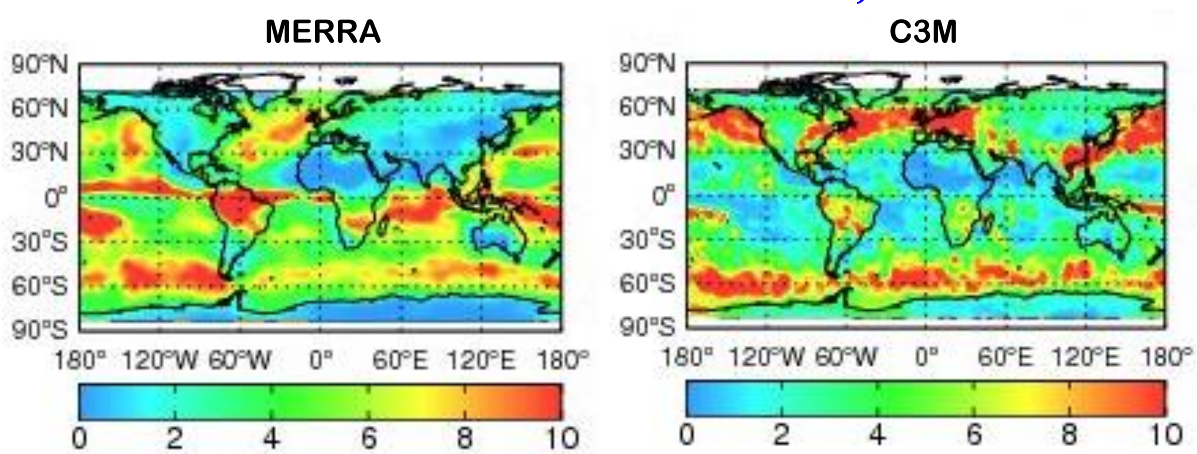
- CRE increase (reduce) photolysis rates above (below) 3 km during ARCTAS (vs. 5.5 km during SEAC4RS).

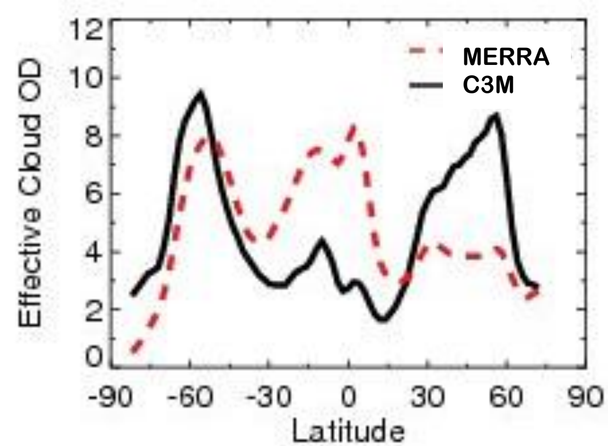
C3M – a Merged Satellite Cloud Data Product @ NASA LaRC



- Merged cloud vertical profiles from multiple A-Train satellite (CALIPSO, CloudSat, CERES, and MODIS) observations [Kato et al., JGR 2010, 2011].
- Collocation of 333-m CALIPSO and 1-km CloudSat mask profiles to 1-km MODIS pixel.
- The merged cloud profiles are further collocated & grouped within a 20-km CERES footprint.
- 3-D structures of cloud extinction & fraction.

Effective cloud OD, MERRA vs. C3M (Jan. 2008)

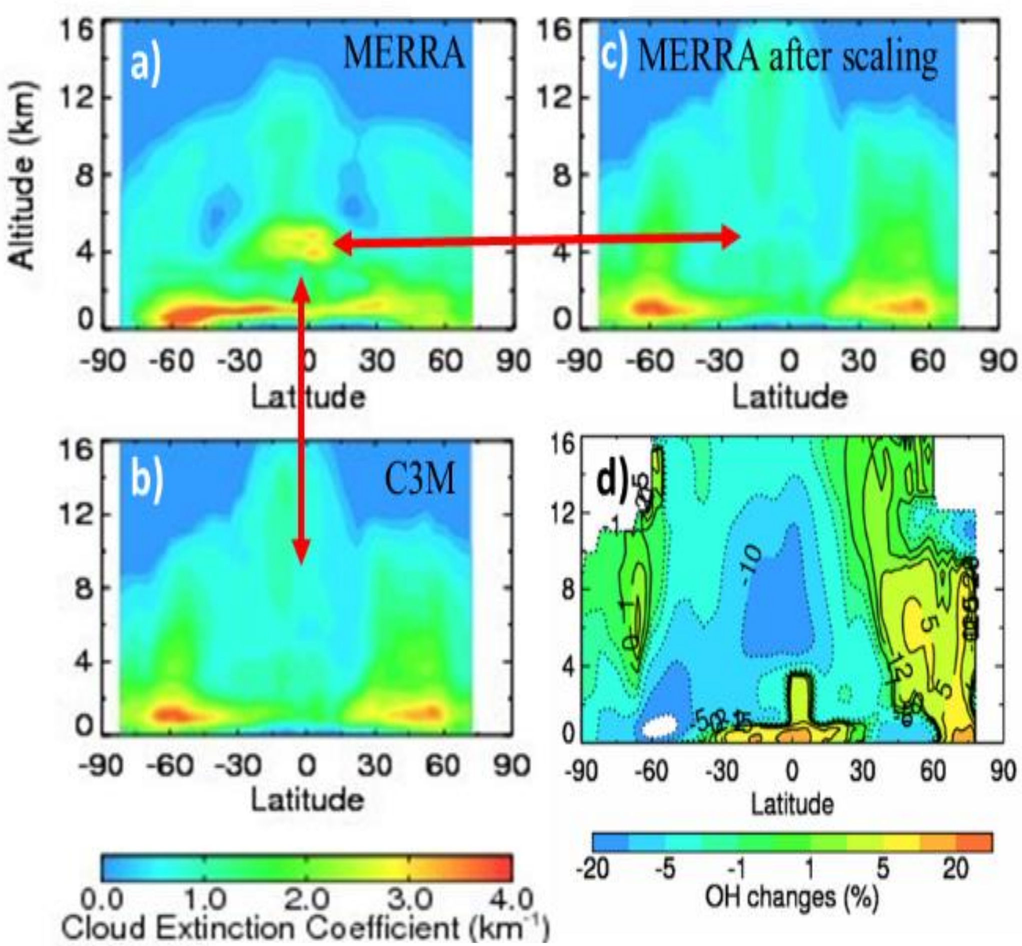




- MERRA clouds sampled along A-Train orbit track (daily 1:30pm LT).
- MERRA overestimates tropical cloud OD, but underestimates at NH mid-lat.

Changes in Global OH due to Constraints from C3M Cloud

Jan. 2008

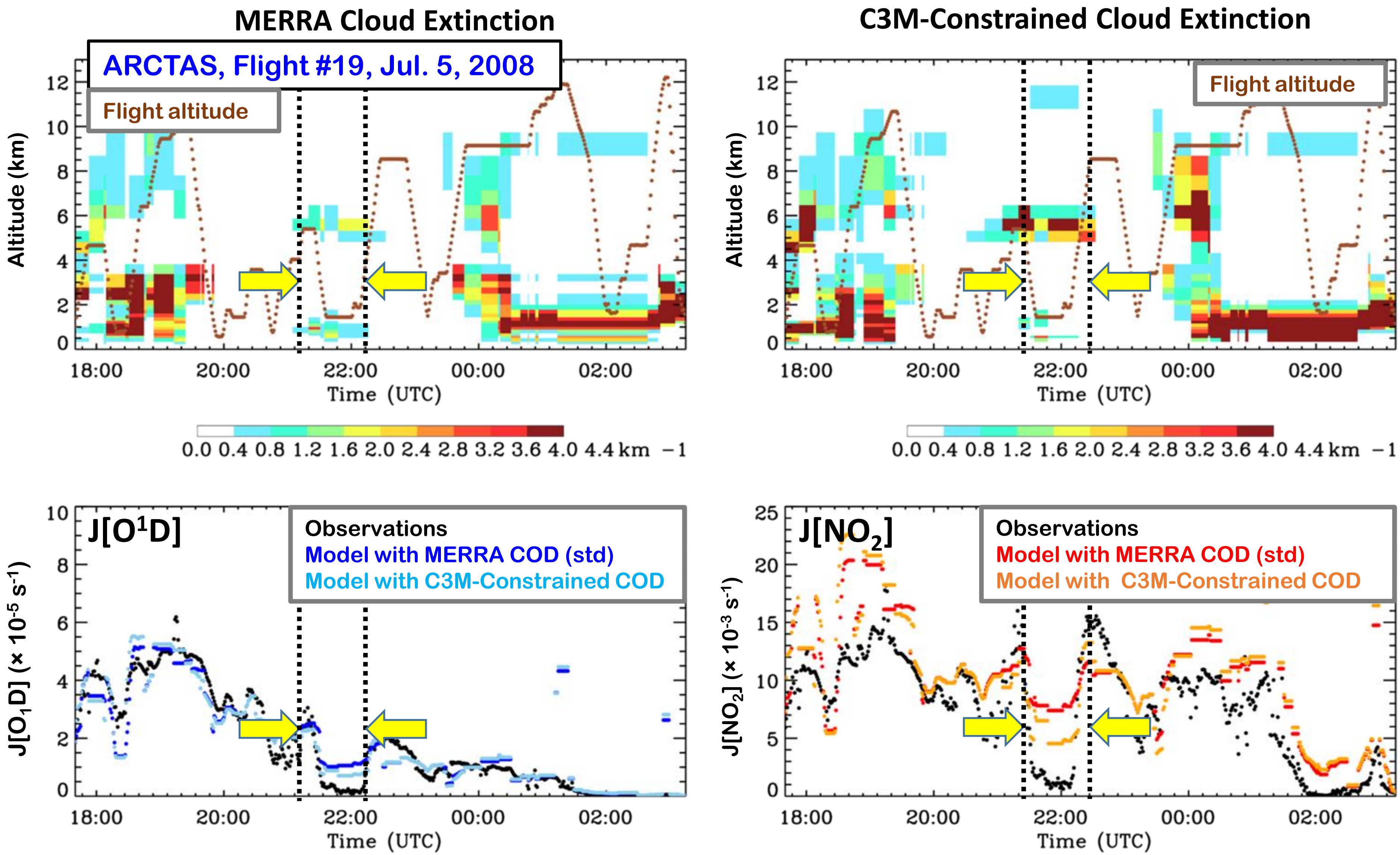


Liu et al., in prep., 2018

- 3D cloud extinctions in MERRA are scaled by C3M on a monthly basis. Constraints on monthly afternoon (~1:30pm) clouds.
- Model cloud extinctions in the tropical lower free troposphere are reduced.
- Global multi-model mean OH concentration is overestimated by 5-10% [Naik, V. et al., ACP 2013].
- Using C3M to constrain the model clouds reduces the global mean OH by ~4%.

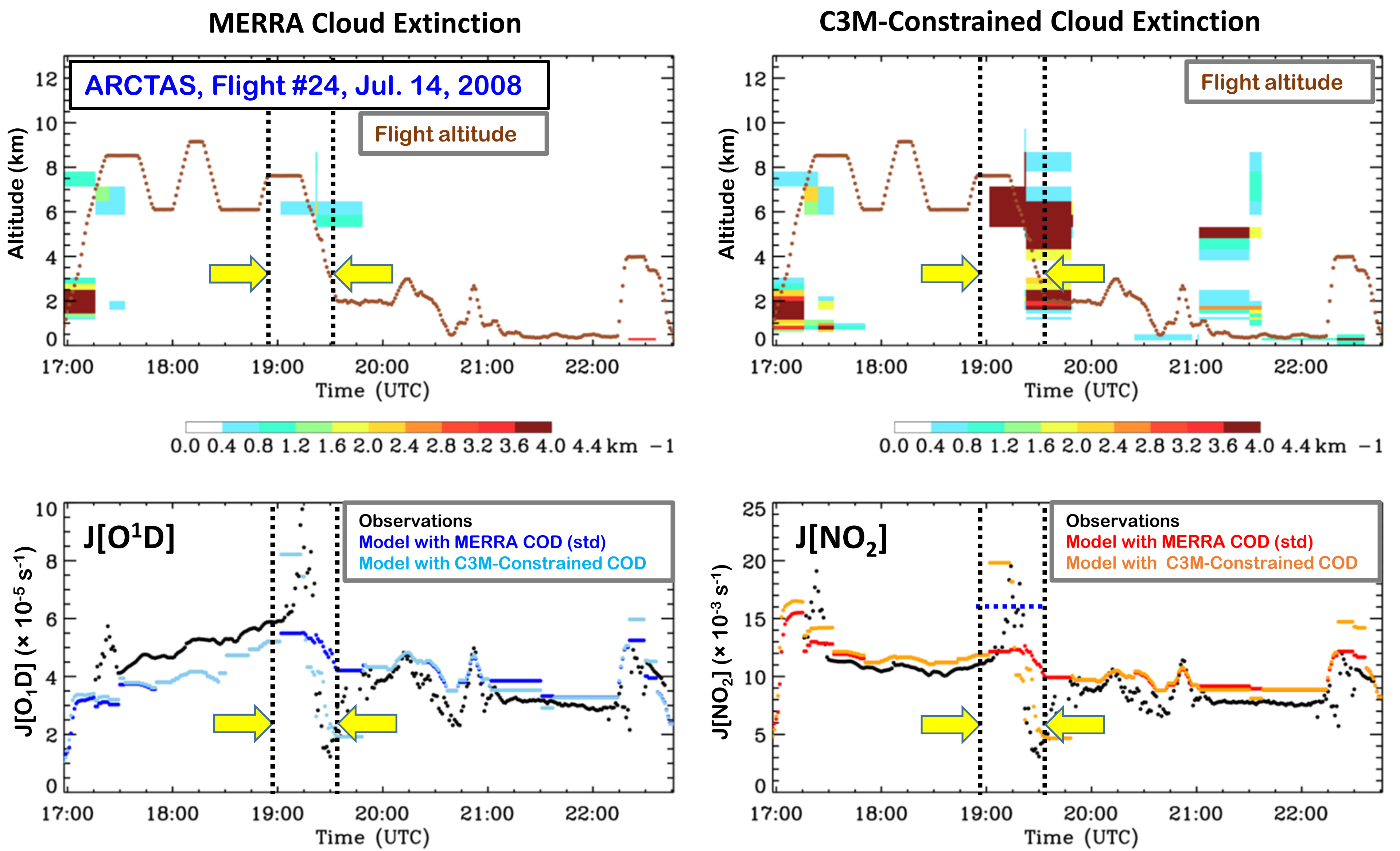
Will the C3M cloud constraints improve model calculations of photolysis rates as compared with aircraft measurements?

Improved Model Photolysis Rates with C3M-constrained Cloud: ARCTAS Flight #19



- **The correction by C3M increased (reduced) COD above (below) the points of measurement, leading to improved model photolysis rates.**

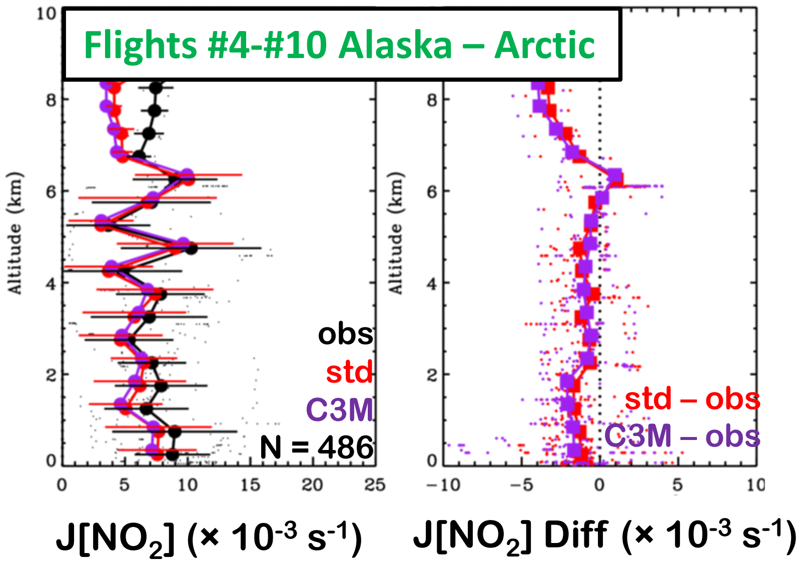
Improved Model Photolysis Rates with C3M-constrained Cloud: ARCTAS Flight #24

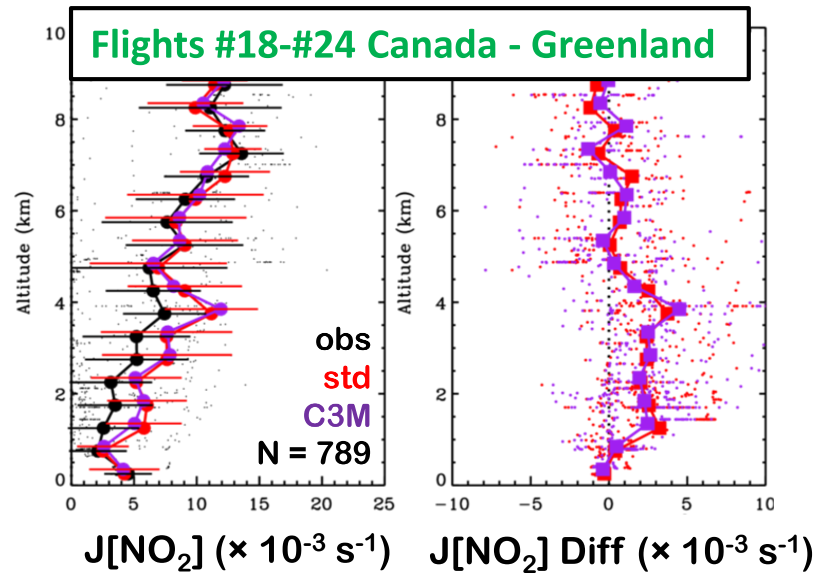


- **The correction by C3M increased COD throughout the tropospheric column, but appears to be a bit too large.**

Effect of C3M-constrained cloud on overall model performance during ARCTAS

Conditions: Column COD > 0.1, diff(std-obs)>5%, diff(c3m-std)>5%, and Altitude<10km





- **With C3M-constrained COD, model photolysis rates for ~50% of the samples are improved. ~10% over-corrected.**
- **Satellite 3-D cloud observations are needed with a higher spatial coverage & temporal resolution.**

Summary and Conclusions

- **Cloud radiative effect (CRE) - a major factor affecting tropospheric photolysis and oxidants. Using C3M (a 3-D cloud data product merged from multiple A-Train satellite observations) to constrain the GEOS-Chem / MERRA model clouds reduces the global mean OH concentration by ~4%.**

- **Model calculated photolysis rates reasonably agree with those from aircraft measurements during the ARCTAS (2008) and SEAC4RS (2013) campaigns.**

- **C3M-constrained cloud improves model photolysis rates in some cases, but does not significantly improve the overall model performance for the period of ARCTAS. Satellite 3-D cloud observations are needed with a higher spatial coverage & temporal resolution.**

- **Cloud vertical distribution is critical for the model calculated photolysis rate profiles. In the cases of improved model photolysis rates, C3M constraints result in the adjustments of both cloud column OD and vertical distribution.**

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