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Cloud Impacts on Photochemistry: Statistical Analysis of Global Chemistry Models and Measurements from the Atmospheric Tomography Mission

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Abstract

The influence of clouds on photochemistry remains a significant uncertainty in global chemistry models. Variability in cloud fraction, morphology, phase and optical properties provides significant challenges to models with horizontal resolutions that far exceed the scale of most clouds. Measured photolysis frequencies derived from the Charged-coupled device Actinic Flux Spectroradiometers (CAFS) on board the NASA DC-8 during the Atmospheric Tomography (ATom) mission in 2016 provide an extensive set of statistics on how clouds alter the photolytic rates throughout remote ocean basins. Here we focus on north and tropical pacific transects during the first deployment (ATom-1) in August 2016 including regular profiles through cloudy, partly cloudy and clear conditions. Nine global chemistry–climate or –transport models provide similar statistics on J-values for regional domains encompassing the measured flight path. The statistical picture of the impact of clouds on J-values emerges through the distribution of the ratio of the cloud influenced models and measurement to corresponding cloud free model runs (J-cloudy/J-clear). The models all reproduce general patterns of enhancement above and shading below cloud, but diverge in distribution patterns and clear sky prevalence.

Cloud impacts on photochemistry

Statistical analysis of global chemistry models and measurements from the NASA Atmospheric Tomography (ATom) mission

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Introduction

The influence of clouds on photochemistry remains a significant uncertainty in global chemistry models. Variability in cloud fraction, morphology, phase and optical properties challenges the model determinations of photolysis rates (j-values) as horizontal resolutions far exceed the scale of most clouds.

The airborne NASA ATom mission provided an extensive set of measurements to statistically examine how clouds alter j-values throughout remote ocean basins.

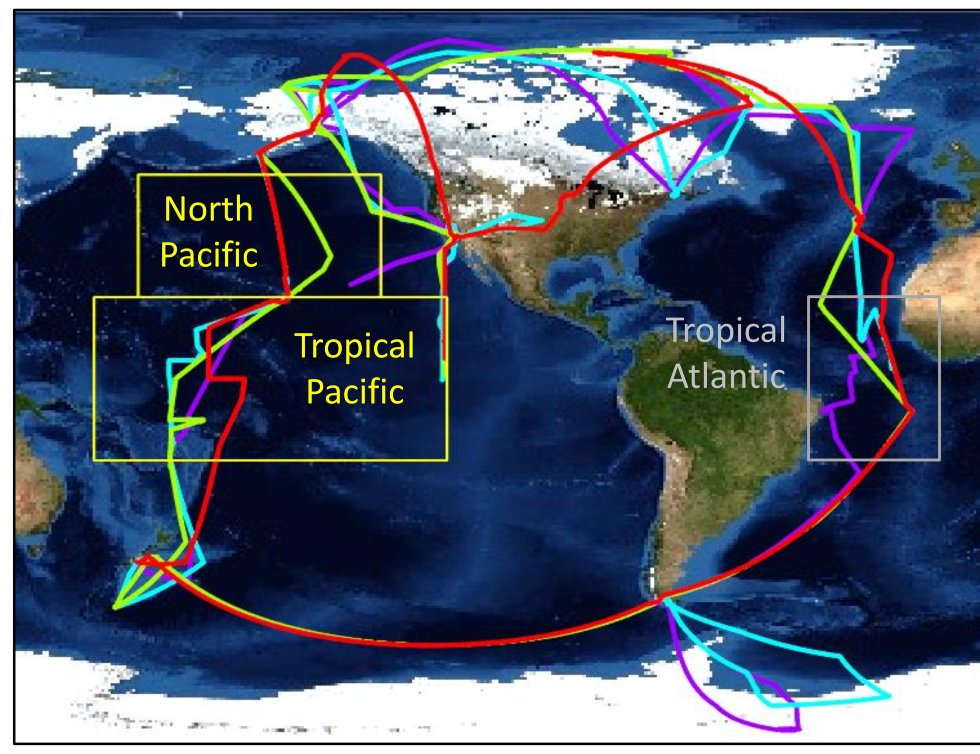
The J-values were calculated from

- 9 global chemistry – climate / transport models
- Actinic flux measurements along the NASA DC-8 flight track

We focus on model and measurement comparisons during the north and tropical Pacific transects during the first deployment (ATom-1) including regular profiles through cloudy, partly cloudy and clear conditions. The statistical cloud impact on J-values emerges through the distribution of the ratio of cloud influenced models and measurement to corresponding cloud-free model runs (J-cloud/J-clear).

For comparison, we also show CAFS observations for the complete mission (ATom 1-4) including the tropical Atlantic and global datasets to assess the representativeness and variability of the dataset.

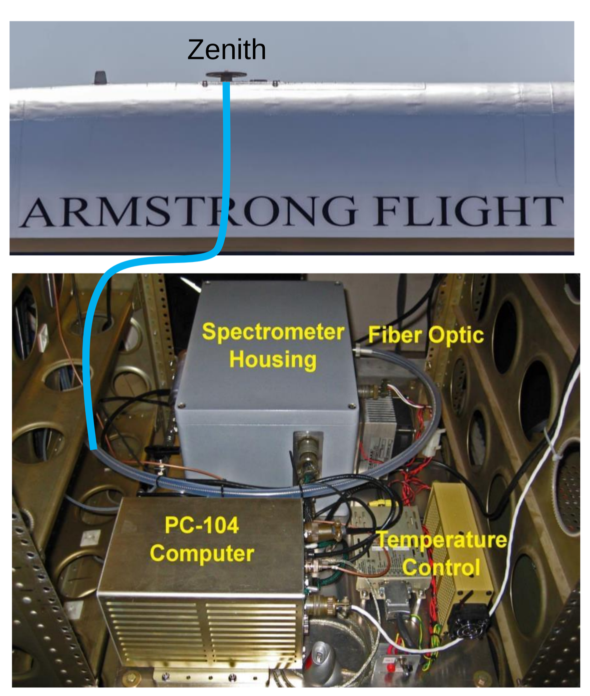
NASA Atmospheric Tomography (ATom)

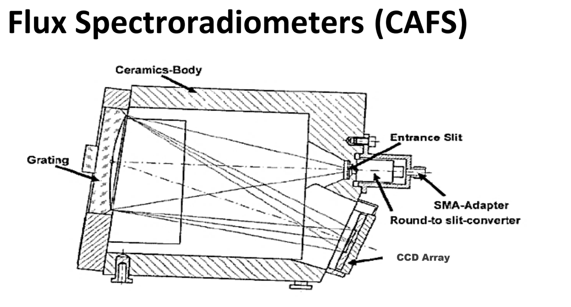


ATom 1	Aug 4 – Aug 31 2016
ATom 2	Jan 26 – Feb 22 2017
ATom 3	Sep 28 – Oct 25 2017
ATom 4	Apr 26 – May 23 2018

- PI: Steven C. Wofsy, Harvard
- 4 DC-8 aircraft campaigns in 4 seasons
- Systematic, global-scale profile sampling
- 20 instrument payload: trace gases, aerosols, actinic flux
- Anthropogenic impacts on remote trace gases and aerosols
- Focus on CH_4, O_3 and aerosols for climate change influences
- Improve GCM chemistry and prediction
- Satellite validation

Charged-coupled device Actinic Flux Spectroradiometers (CAFS)





Actinic flux

Photolysis $AB + h\nu \rightarrow A + B$

Photolysis frequency from actinic flux

$$j[AB] = \int F(\lambda)\sigma(\lambda,T,p)\phi(\lambda,T,p)d\lambda$$

Tropospheric Ultraviolet and Visible (TUV)

Radiative transfer model to determine cloud and aerosol-free actinic flux and photolysis frequencies along the flight track (version 5.3)
Madronich and Flocke, 1999

Discussion

ATom 1: CAFS observations guide the accuracy of J-value calculations in global models

- Models reproduce general enhancement above and shading below cloud
- Models vary in distribution patterns
- Models diverge into two distinct classes of higher and lower clear-sky prevalence
  - CAFS data supports lower prevalence but not robustly enough to be conclusive
  - CAFS/TUV rlnJ exhibits broader features due to nearby cloud influences or incomplete albedo and aerosol parameterizations in the cloud-free model

ATom 1-4: CAFS full data set now available for additional analysis

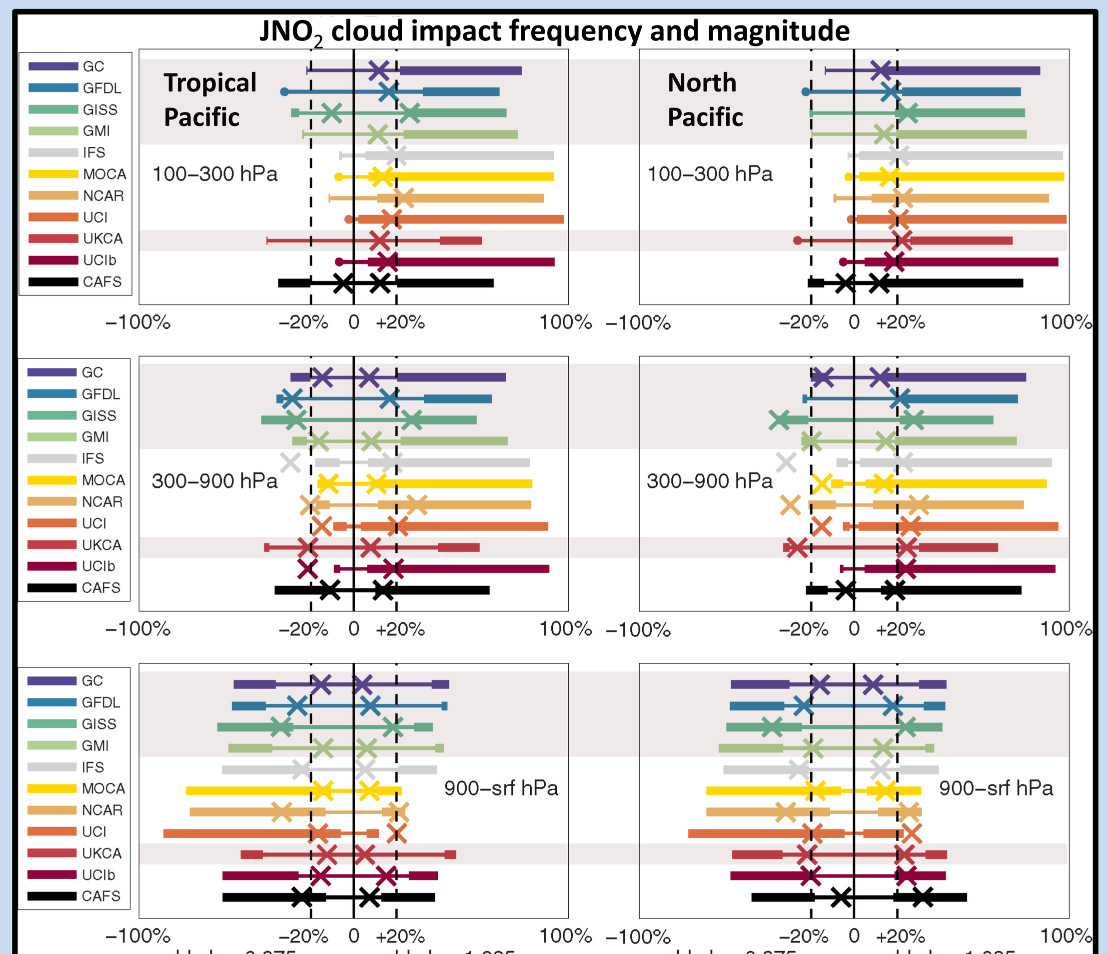
- Tropical Pacific region is relatively most consistent across seasons
- Tropical Atlantic is more variable, likely due to aerosols and greater cloud dynamics
- CAFS enhancements above boundary layer consistent with UCI and NCAR models
- Global analysis biased by flights with high polar albedos, particularly ATom-4

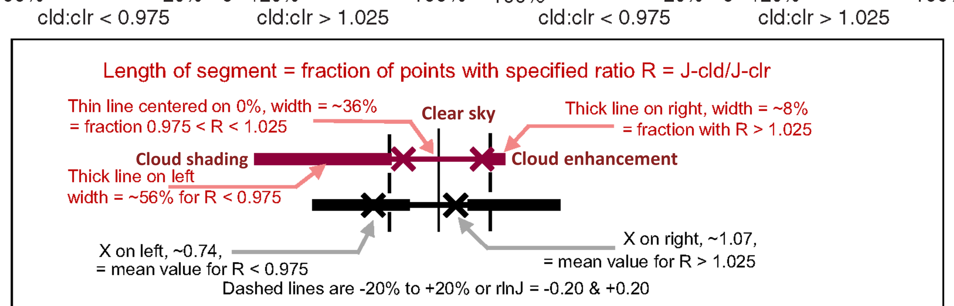
Modeled photolysis and cloud fields

Model (abbrev)	Cloud data (resolution) and date	J-values and cloud fraction treatment	Model references including J-values
GEOSChem (GC)	Cloud CF+OD from MERRA-2; GC v11_01 (2.5°x2.0°) 2013 Aug 16	Fast-J* v7.0, single column Briegleb averaging**	Gelaro et al., 2017 Liu et al., 2006, 2009
GFDL AM3 (GFDL)	0.5° AM3 using 1.4° NCEP (u,v) (0.5°x0.5°) 2013 Aug 16	Fast-J v6.4, liquid cloud C1 (12 µm) and ice clouds per Fast-J Briegleb averaging	Donner et al., 2011 Naik et al., 2013; Mao et al., 2013 Li et al., 2018; Lin et al., 2012
GISS Model E2 (GISS)	Nudged to MERRA fields (2.5°x2.0°) 2013 Aug 16	Fast-J2	Schmidt et al., 2014 Shindell et al., 2012 Rienecker et al., 2011
GSFC GMI (GMI)	Cloud CF+OD from MERRA-2 (1.3°x1.0°)2016 Aug 16	Fast-J v6.5, liquid cloud C1 (6 µm) and ice cloud hexagonal (50 µm) Briegleb averaging	Strahan et al., 2013 Duncan et al., 2007
ECMWF IFS (IFS)	IFS (0.7°x0.7°) 2016 Aug 15	Williams et al. (2012) Liquid cloud (4-16 µm, using CCN) ice clouds, random overlap	Flemming et al., 2015 Sun and Rikus, 1999; Sun, 2001
MOCAGE (MOCA)	ARPEGE operational analysis, 3h (1.0°x1.0°) 2017 Aug 16	From Brasseur et al, (1998), using CF and liquid water (10 µm) Briegleb averaging	Guth et al., 2016 Arteta & Flemming, 2015
CESM (NCAR)	CAM5 physics on MERRA (u,v,T, …) (0.6°x0.5°) 2008 Aug 16	TUV lookup J-tables, scaled using CF and liquid water content Briegleb averaging	Tilmes et al., 2016 Madronich, 1987
UCI CTM (UCI)	IFS T159L60N160 forecasts by U Oslo (1.1°x1.1°) 2005 Aug 16	Cloud-J v7.3, quadrature column atmospheres from decorrelation length Liquid & ice clouds per Fast-J	Neu et al., 2007 Holmes et al, 2013 Prather 2015; Prather et al., 2017
UCI CTM (UCIb)	same as UCI	Cloud-J v7.3, single column Briegleb averaging	Same as UCI
UKCA (UKCA)	UK Unified Model (1.9°x1.3°) 2008 Aug 17	Fast-J v6.4 cloud optical depths per Telford et al (2013) Briegleb averaging	Morgenstern et al 2009 O'Connor et al 2014 Walters et al 2017

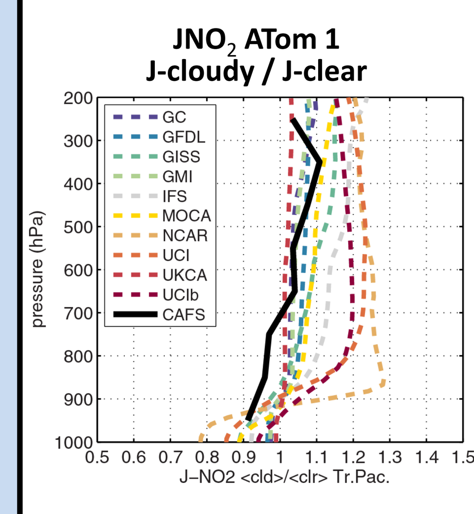
Cloud data includes: cloud fraction (CF), in-cloud ice/liquid water path and effective radius, or in-cloud ice/liquid optical depth (OD in the visible).
*Fast-J versions based on Bian and Prather (2002) with updates, including standard tables for cloud optical properties and simplified estimate of effective radius. Cloud C1 refers to Deirmendjian liquid cloud size distribution from the Fast-J data tables (Wild et al., 2000).
**Briegleb's (1992) method approximates max-random overlap with a single column atmosphere and adjusted effective CF such that the COD in the grid cell is COD(in-cell) = COD(in-cloud) x $CF^{3/2}$.

JNO_2 cloud impact frequency and magnitude





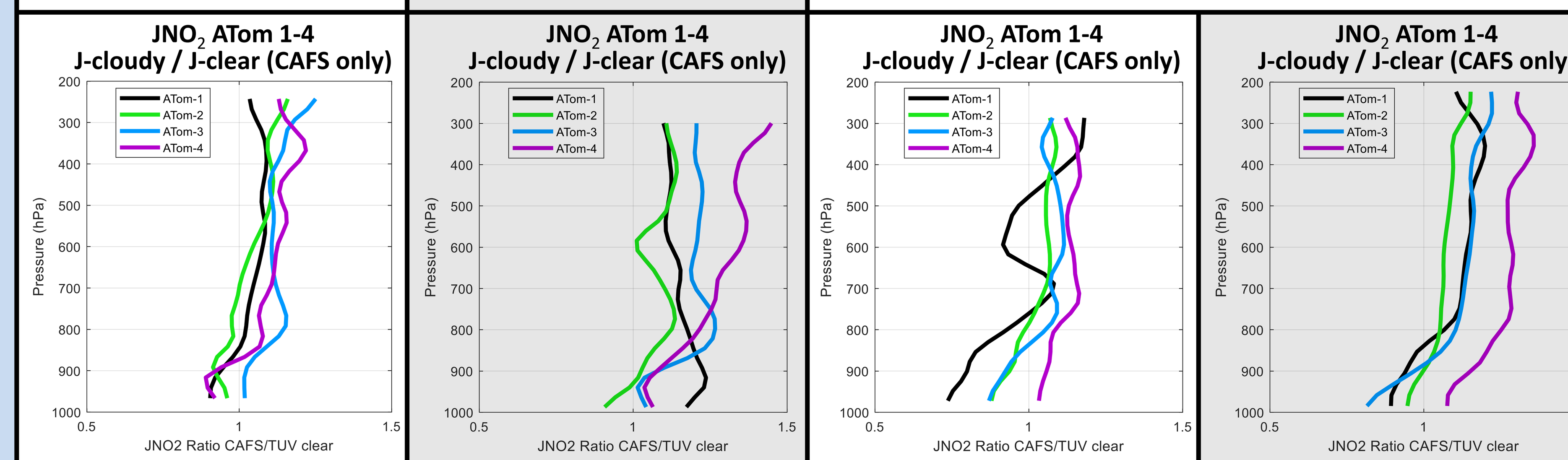
Tropical Pacific	North Pacific	Tropical Atlantic	Global
20°S-20°N 160°E-120°E	20°N-50°N 170°E-135°W	20°S-20°N 40°W-10°W	80.6°S-82.9°N 164.6°E-14.3°W

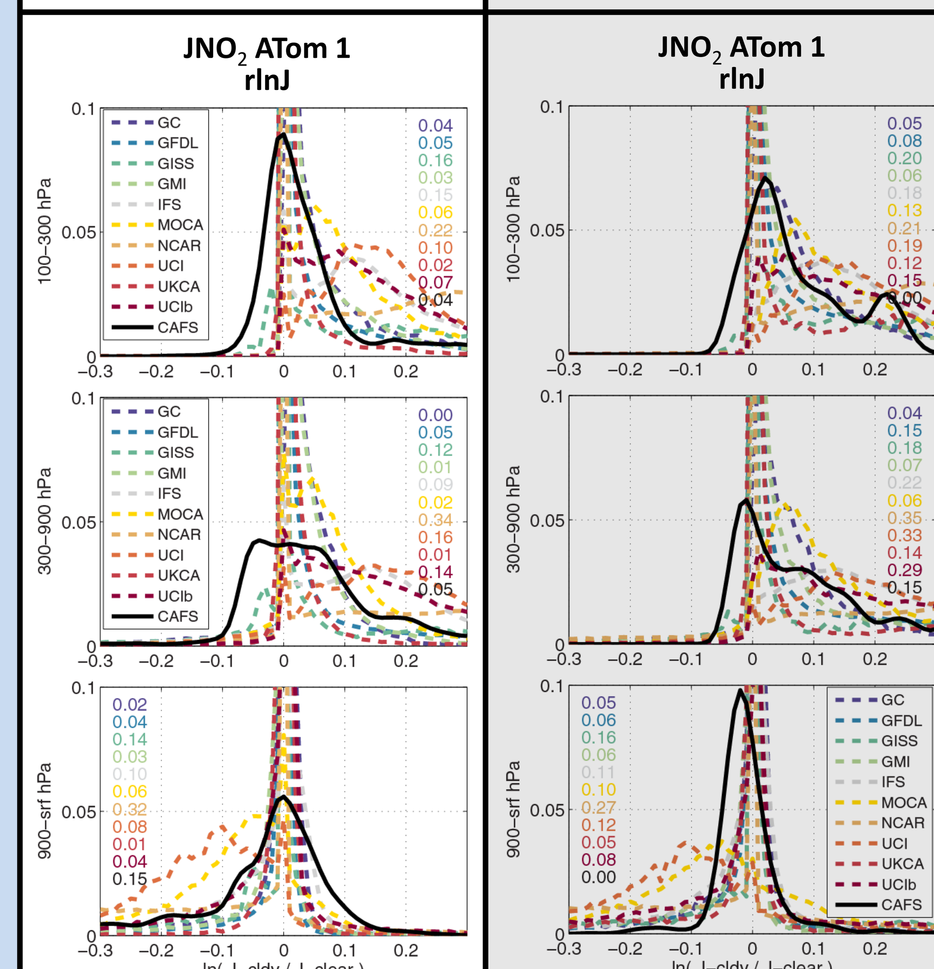


Vertical profiles of JNO_2 (J-cloudy / J-clear)

for the Tropical and North Pacific blocks for ATom-1 (left). CAFS/TUV is determined from the measurements and position data along the flight path. The 10 models are sampled for cloudy and clear over 24 hours from a day in mid-August. Data is selected for SZA < 36°. Some of the spread in JNO_2 is likely due to different choices for interpolating cross sections and quantum yields.

CAFS/TUV for ATom 1-4 (below) for each region but adjusted for SZA < 60° to allow for significant data/bin.





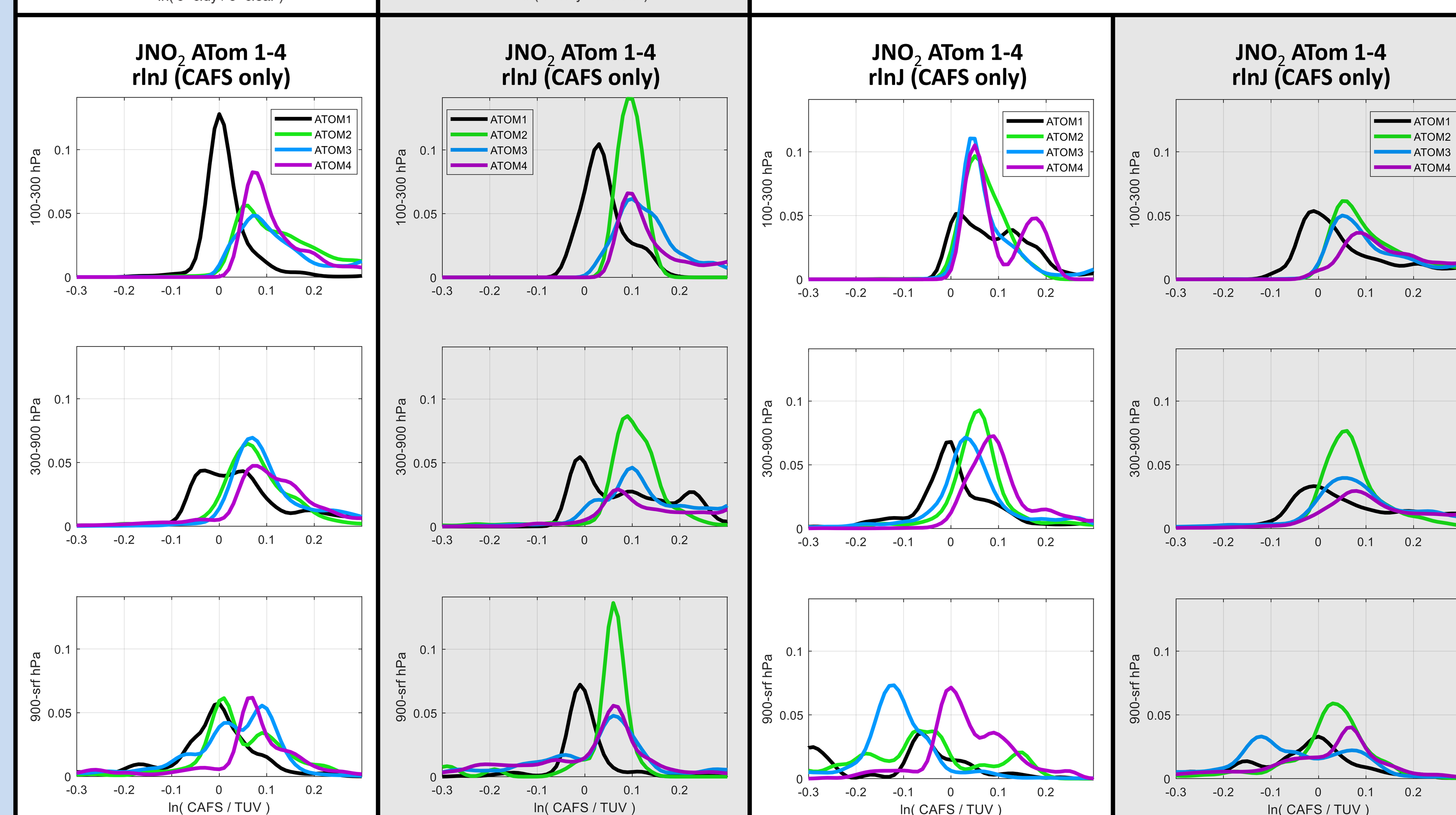
Probability Distributions

of the natural log of the ratio of cloudy-to-clear JNO_2 (rlnJ) for the Tropical and North Pacific blocks for ATom-1 from 10 models and CAFS/TUV (left).

rlnJ = ln (J-cloudy / J-clear)

The 3 rows represent pressure layers (100 – 300, 300 – 900, 900 – surf hPa). All data is selected for SZA < 36°. All histograms sum to 1, but for many models the peak values about rlnJ = 0, corresponding to cloud-free skies, are truncated. Where a significant fraction of events does not fit within the ± 0.3 range (on the high side for 100 – 900 hPa and low side for 900 – surf hPa) the column of numbers, placed on the appropriate side and color coded to the legend, gives the fraction of occurrences outside the range.

CAFS/TUV for ATom 1-4 (below) for each region but adjusted for SZA < 60° to allow for significant data/bin.



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