Stratospheric and anthropogenic contributions to enhanced O_3 in the tropical western Pacific

21 Oct. 2014

Dan Anderson

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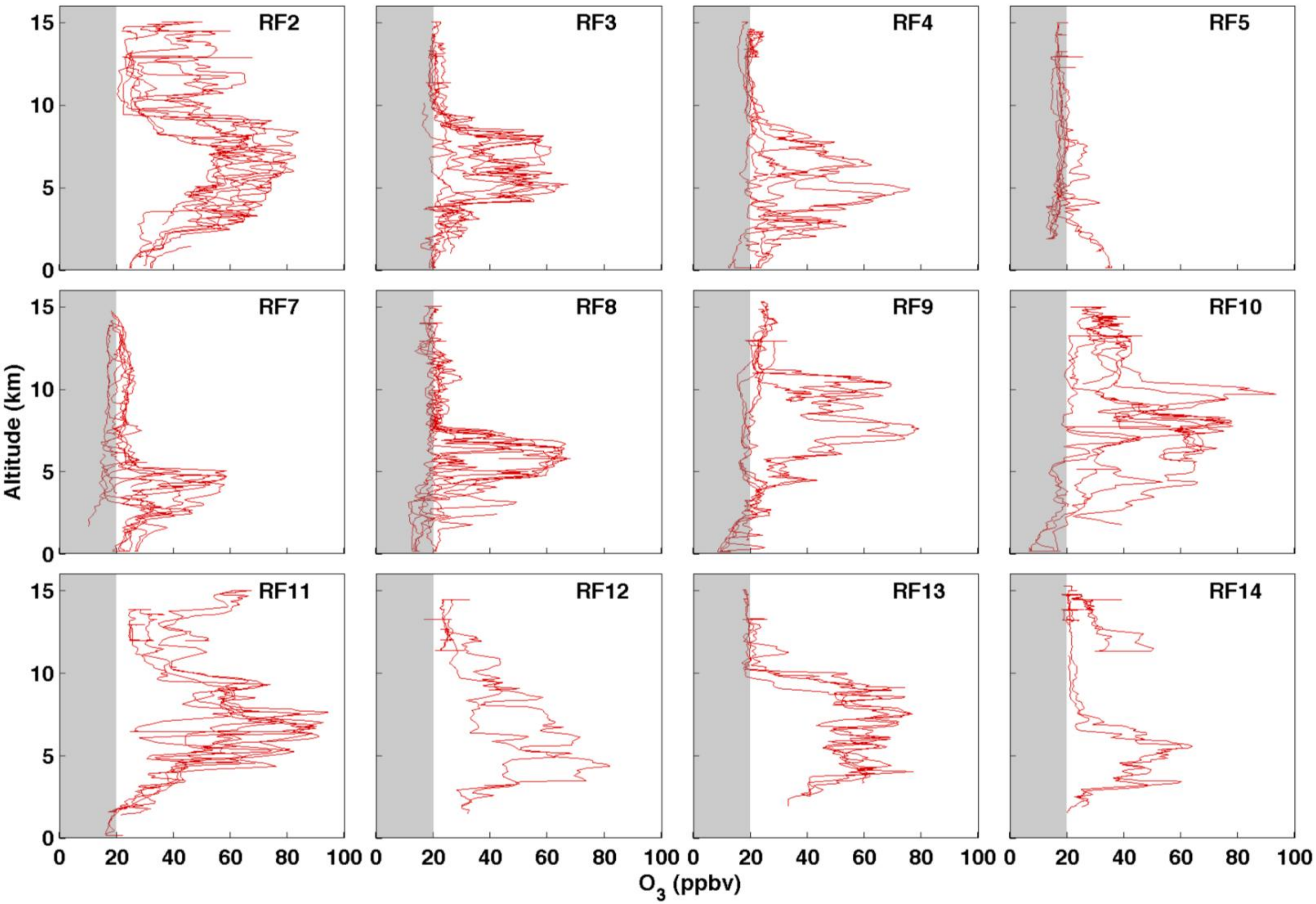
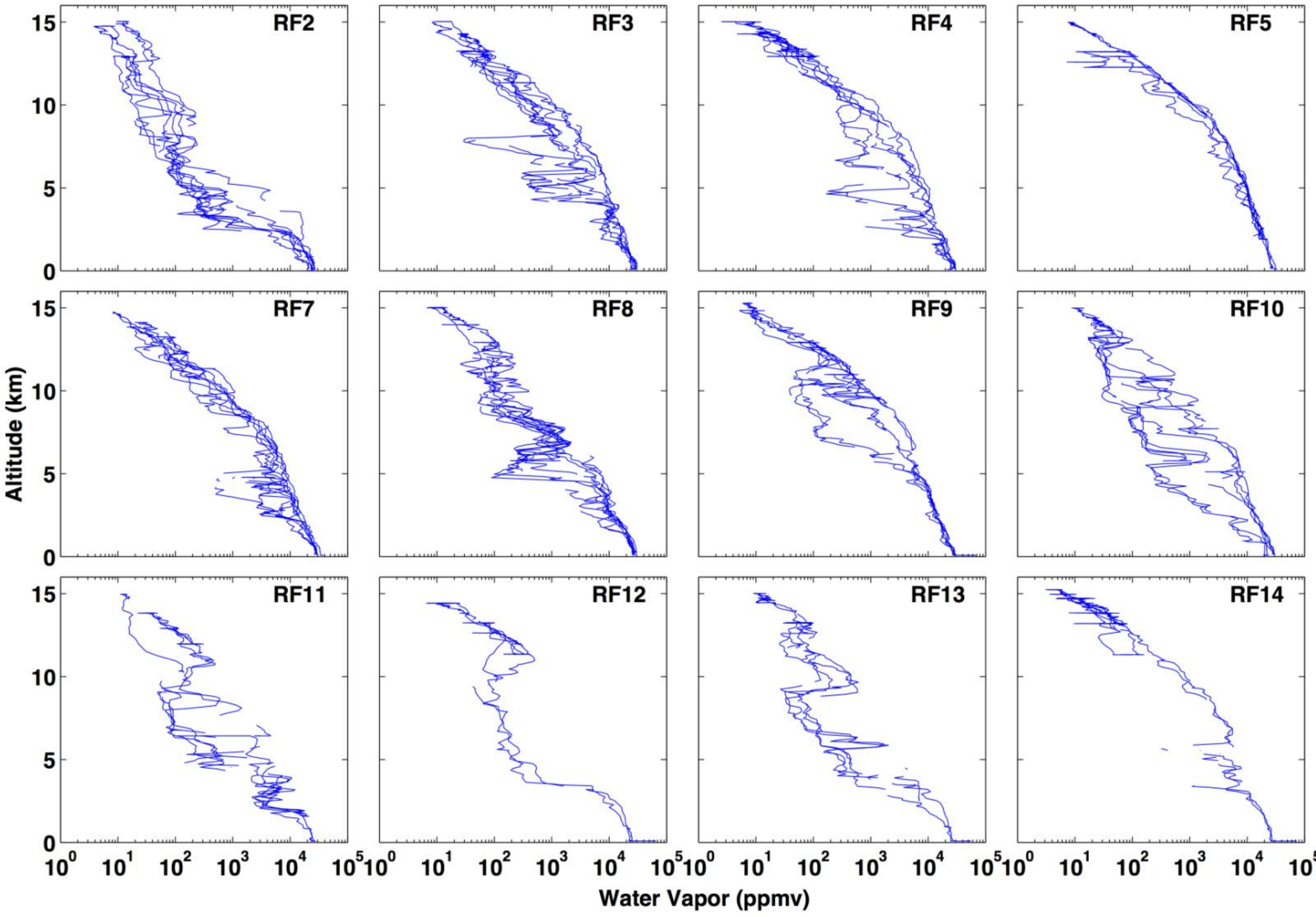
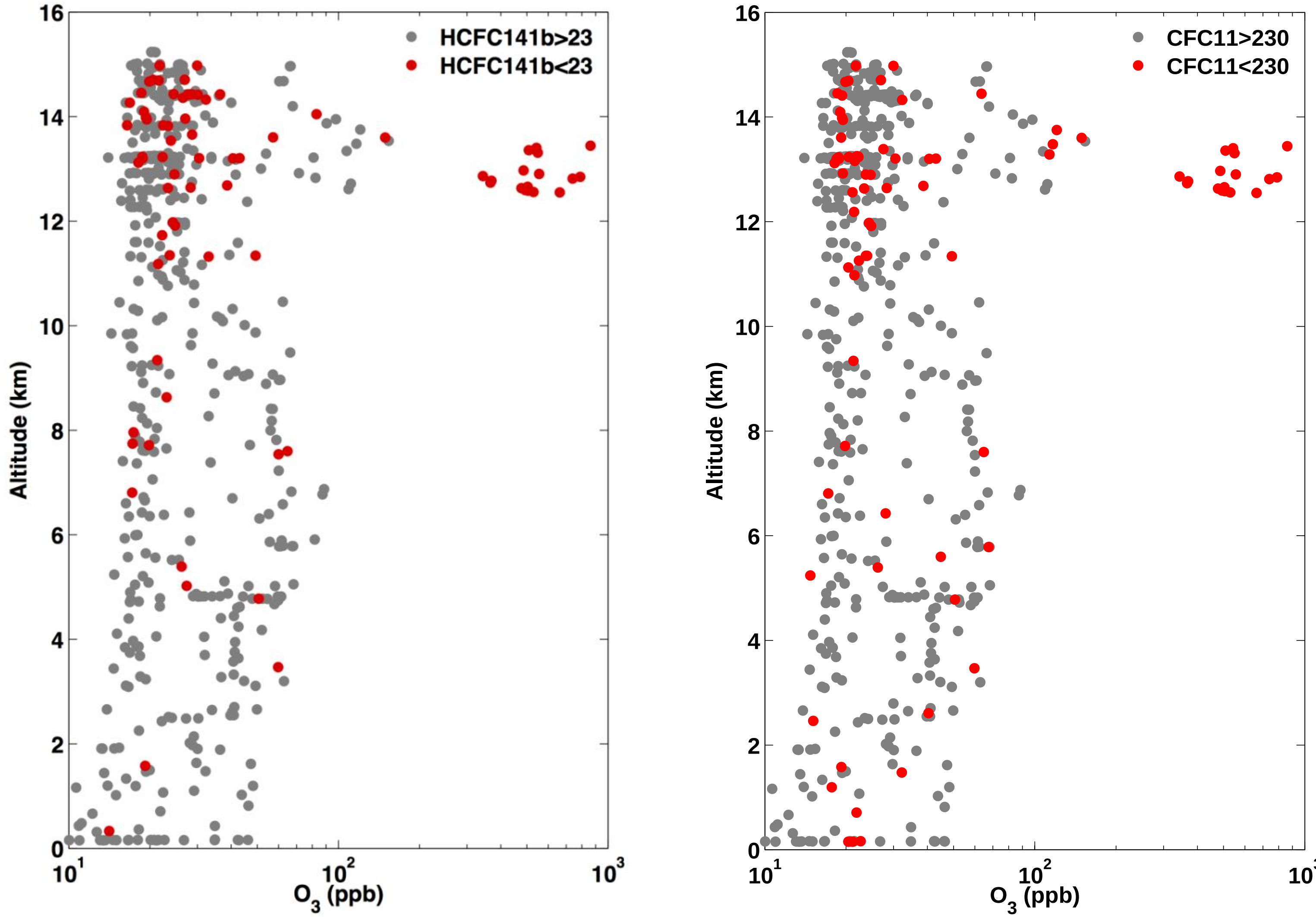
Photo: Becky Hornbrook

Objectives:

- Determine the source of enhanced mid-tropospheric O_3 in the tropical western Pacific.
  - Photochemical production/transport from regions with biomass burning emissions
  - Injection of stratospheric air
- Why is the enhanced O_3 frequently anti-correlated with water vapor?


RF2
RF3
RF4
RF5
RF7
RF8
RF9
RF10
RF11
RF12
RF13
RF14
Altitude (km)
O3 (ppbv)
0
5
10
15
0 20 40 60 80 100

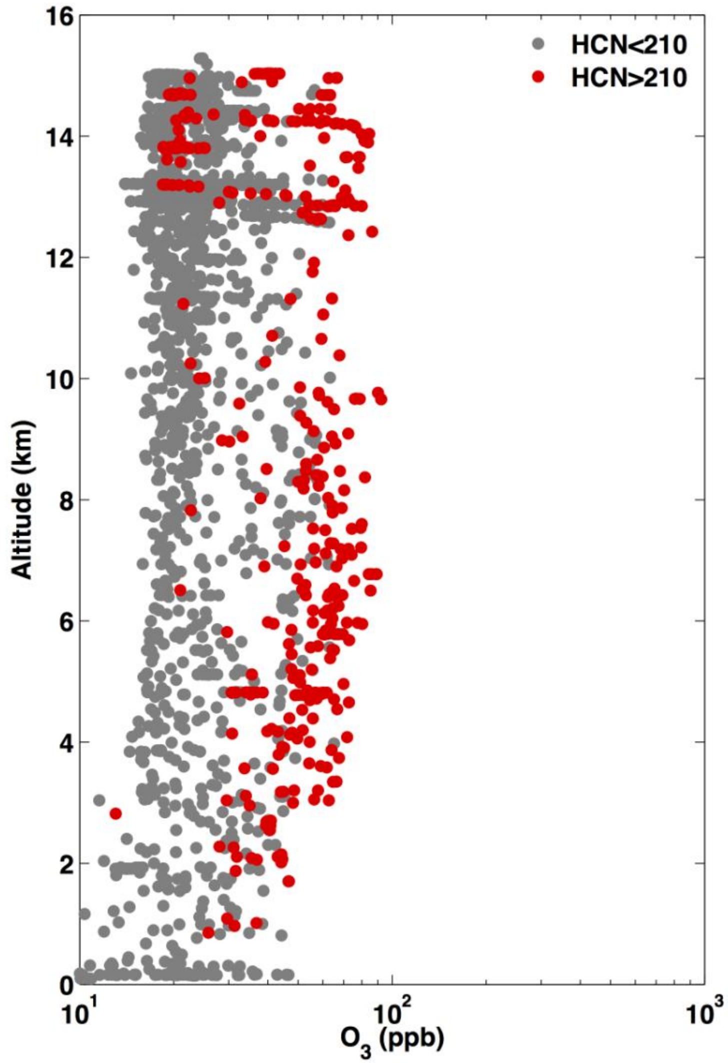

RF2
RF3
RF4
RF5
RF7
RF8
RF9
RF10
RF11
RF12
RF13
RF14
Altitude (km)
Water Vapor (ppmv)
0
5
10
15
10^0 10^1 10^2 10^3 10^4 10^5

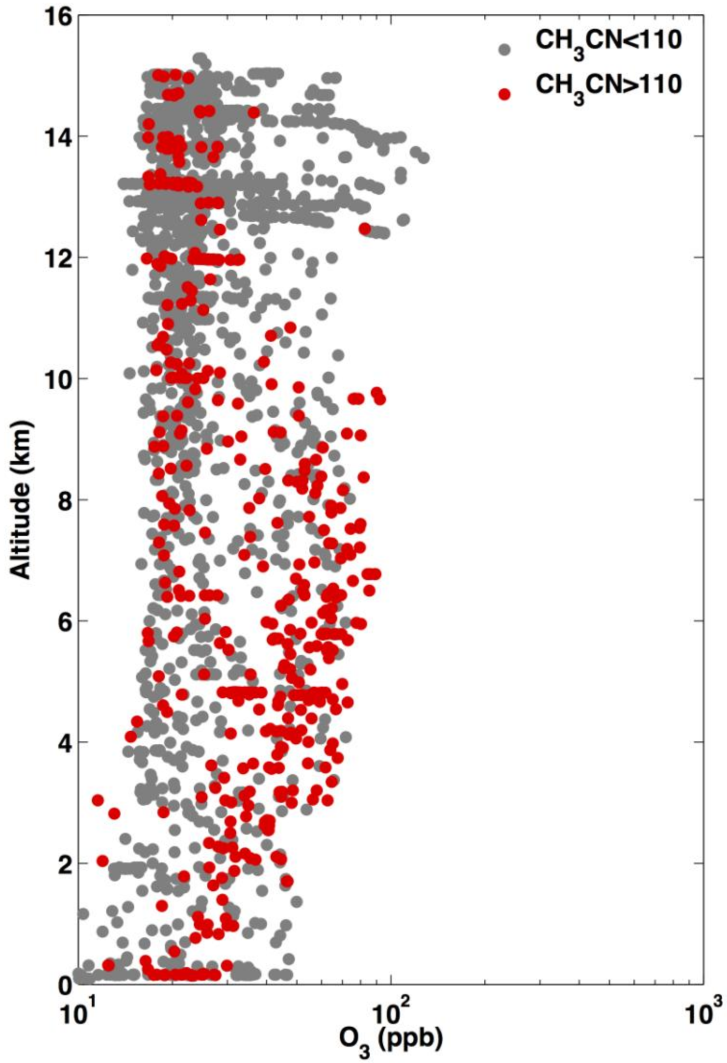


(H)CFC levels in high O_3 filaments consistent with other tropospheric values.

Implies **limited stratospheric** influence in filaments.

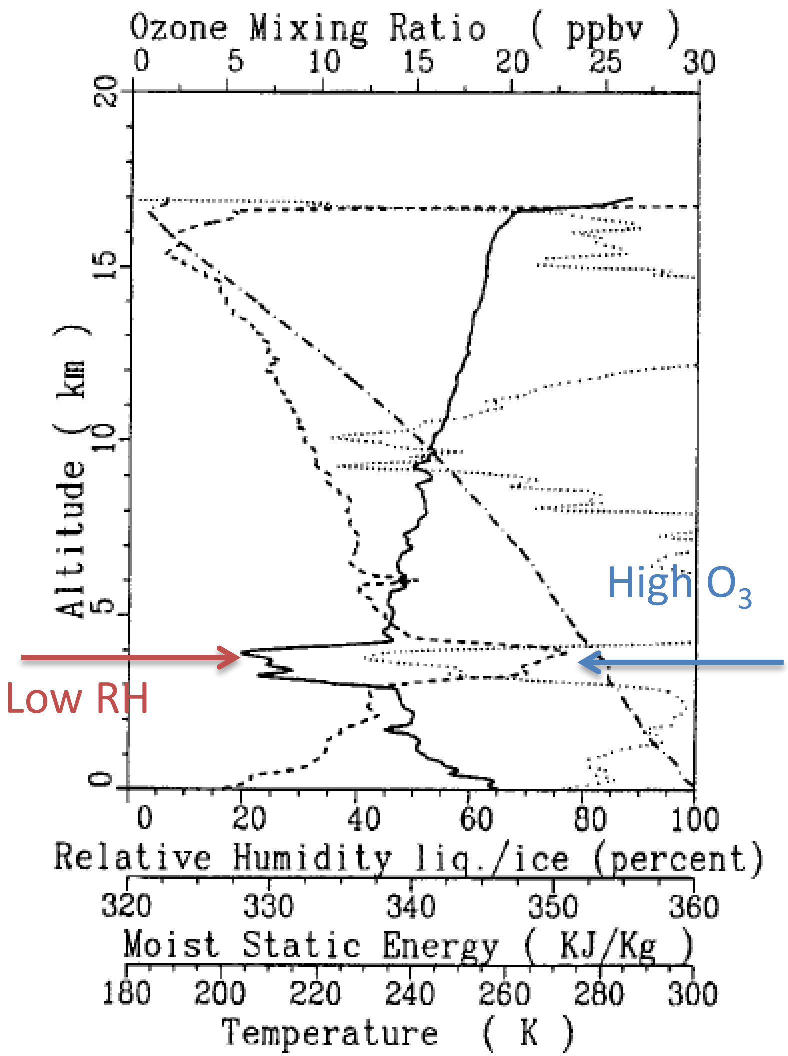
AWAS observations





O_3 filaments enhanced in cyanides.
Implies **strong biomass burning** influence in filaments.

TOGA observations

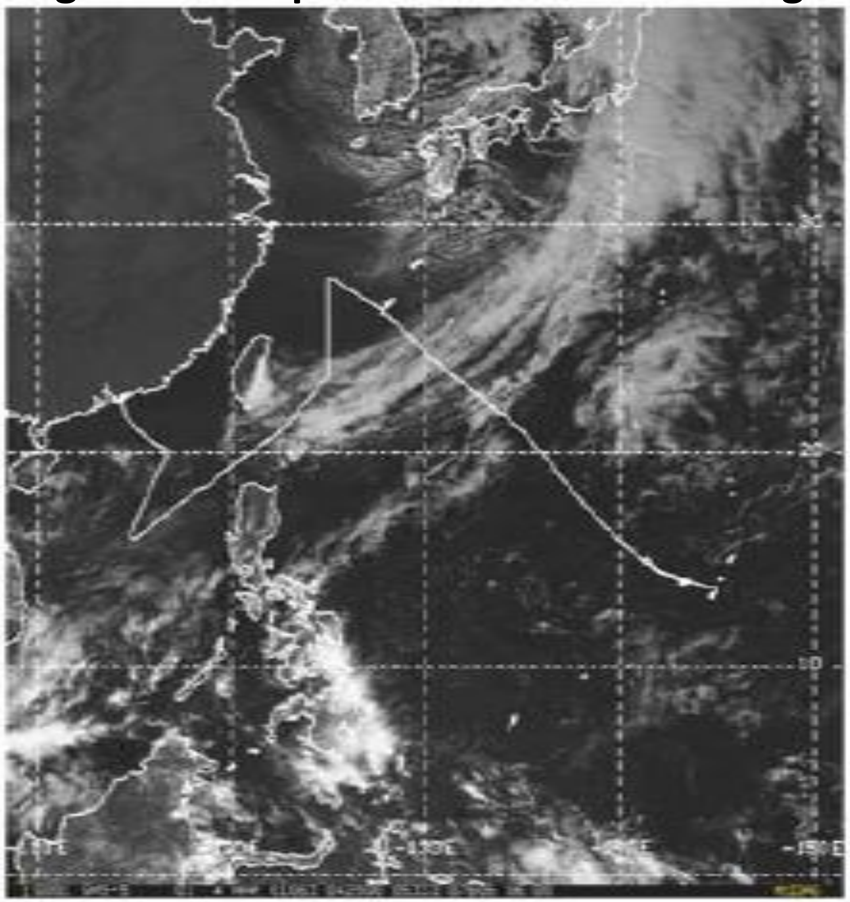


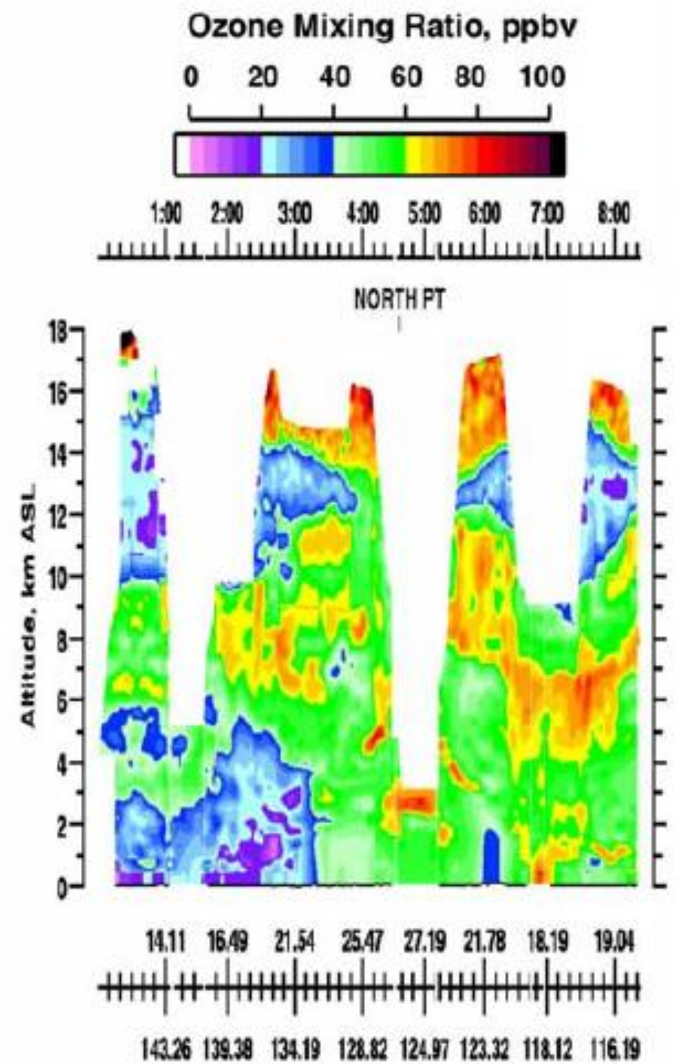
Kley et al. (1997)

CEPEX campaign, March 1993

Conclude that stratospheric intrusions are source of anomalous O_3.

TRACE-P campaign, March 2001
Flight track imposed on IR Satellite image

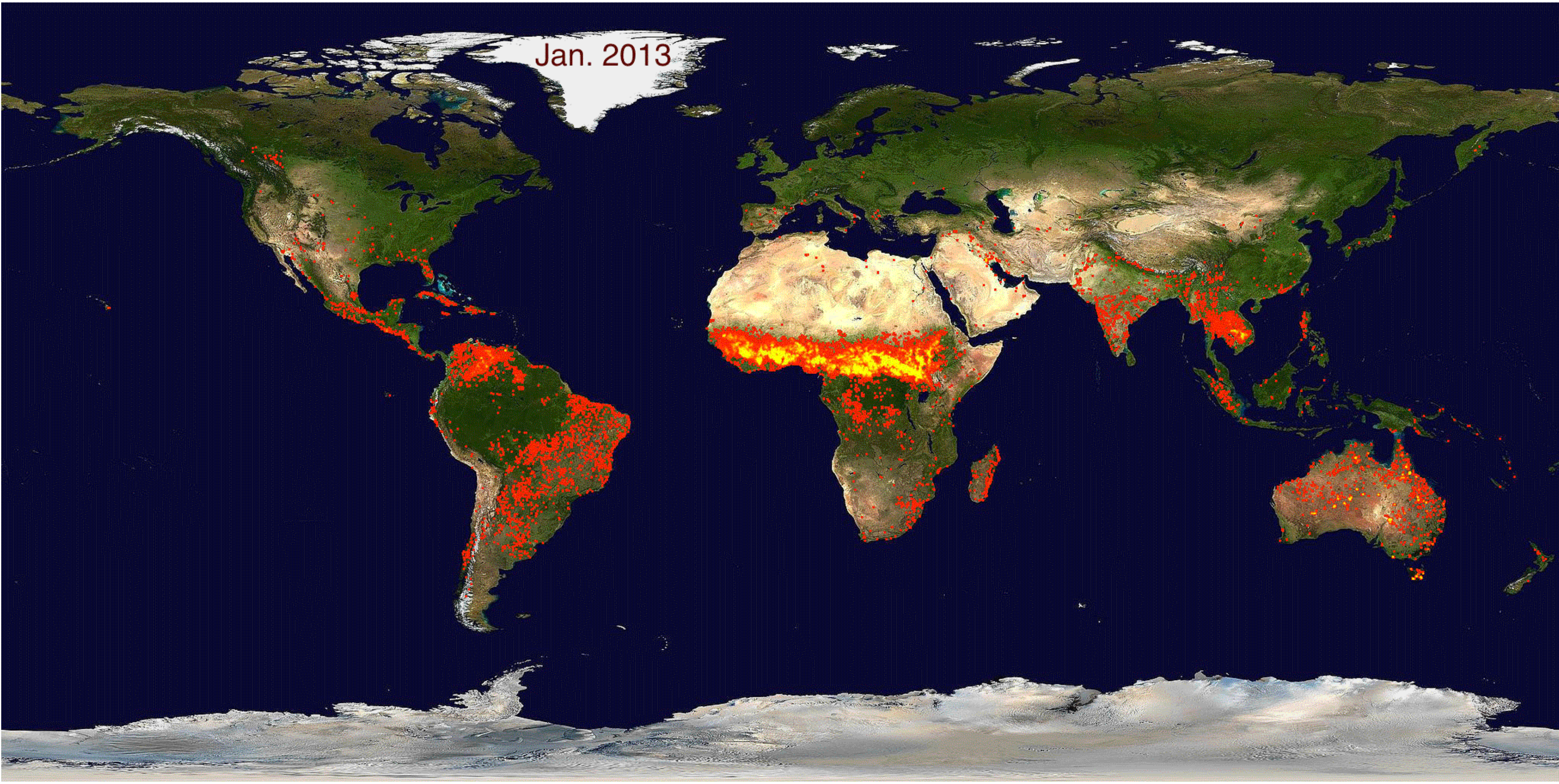




Browell et al. (2003)

Conclude that biomass burning is source of anomalous O_3.

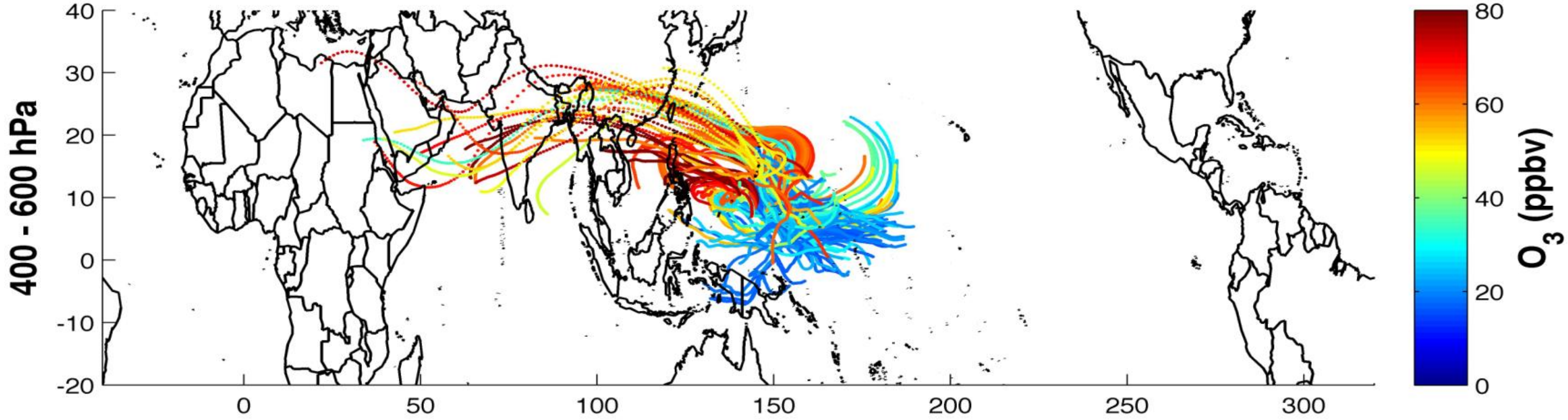
MODIS Fire Counts for 2013



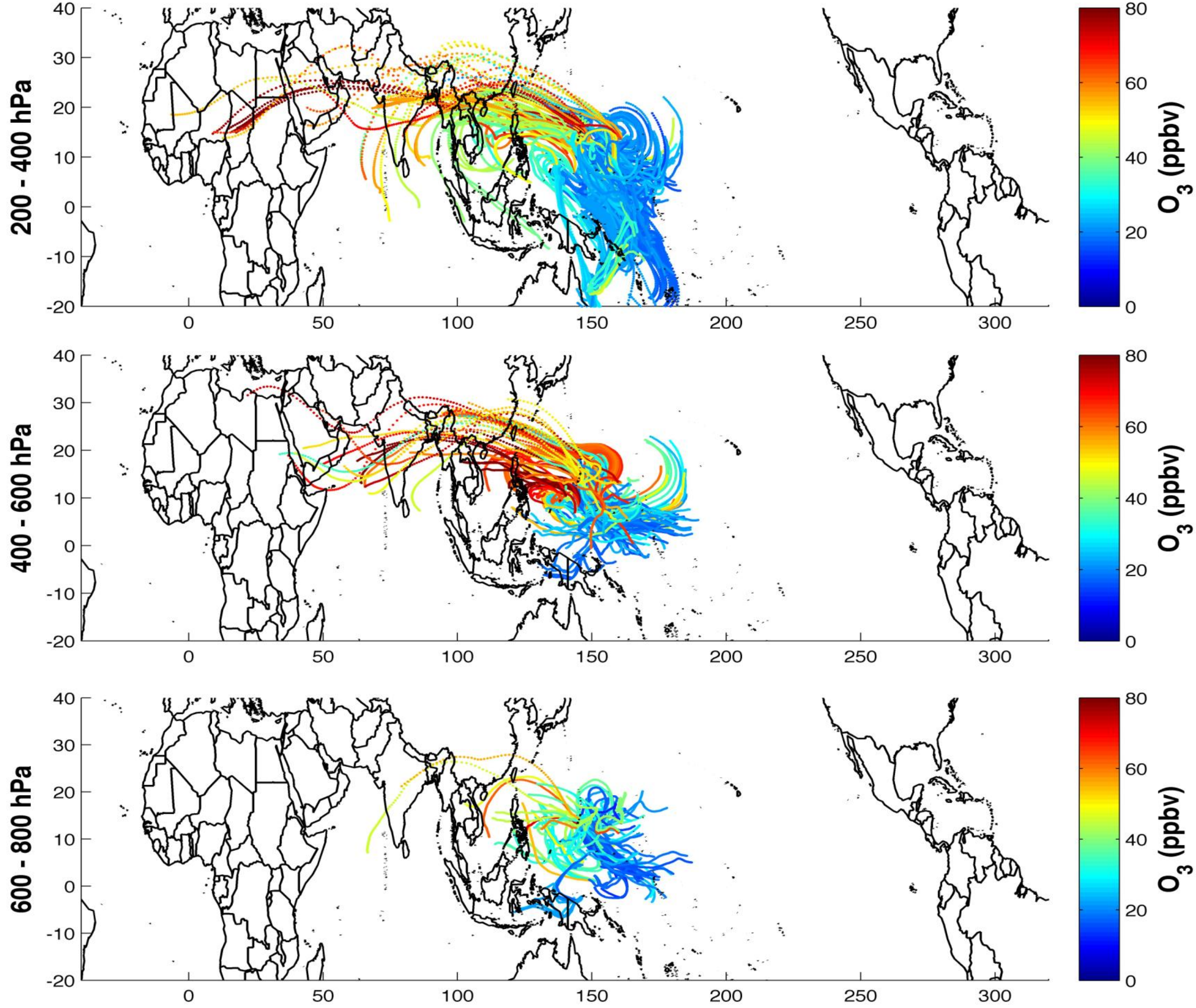
http://rapidfire.sci.gsfc.nasa.gov/cgi-bin/imagery/firemaps.cgi

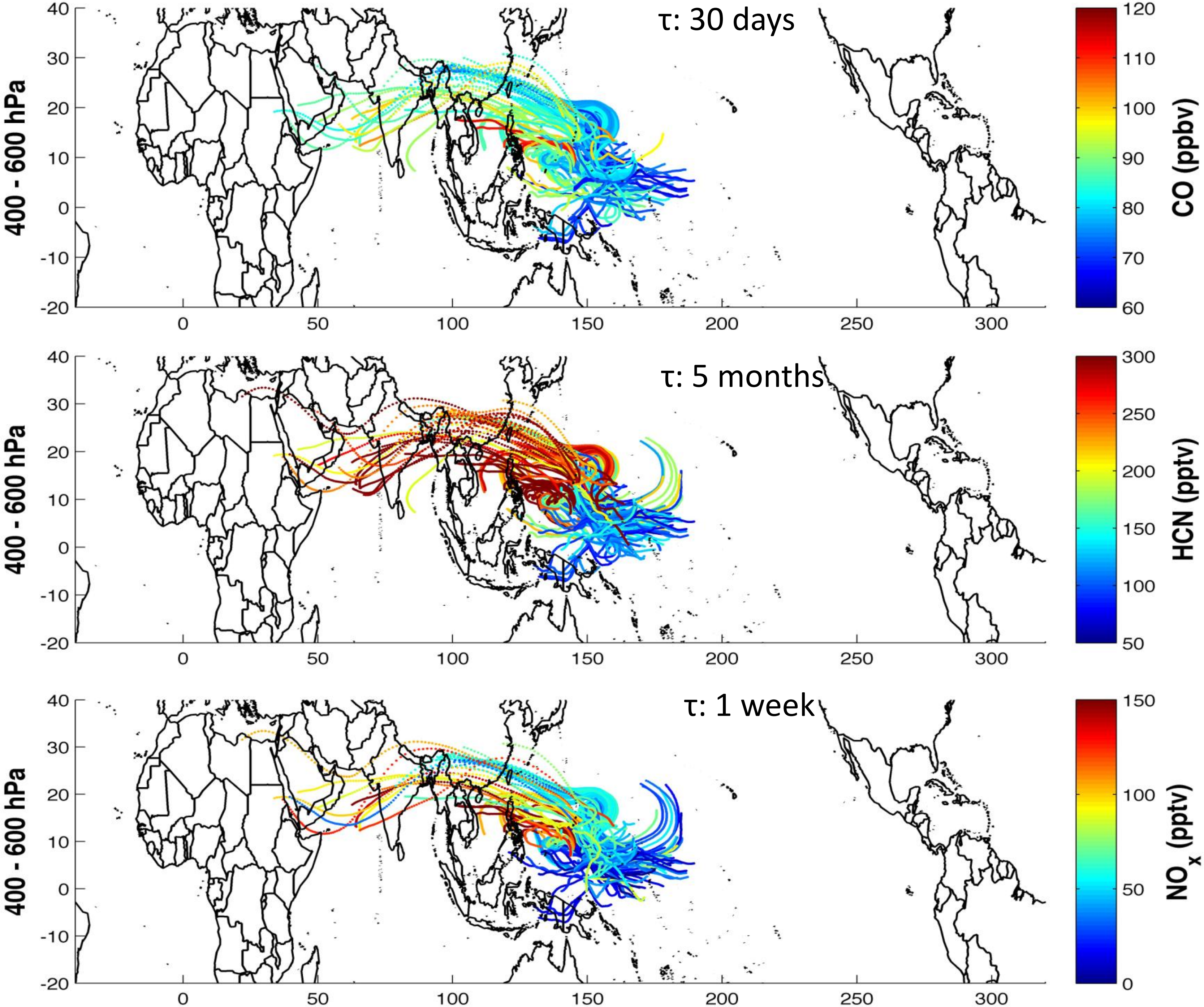
Biomass burning in southeast Asia peaks in late winter/early spring.

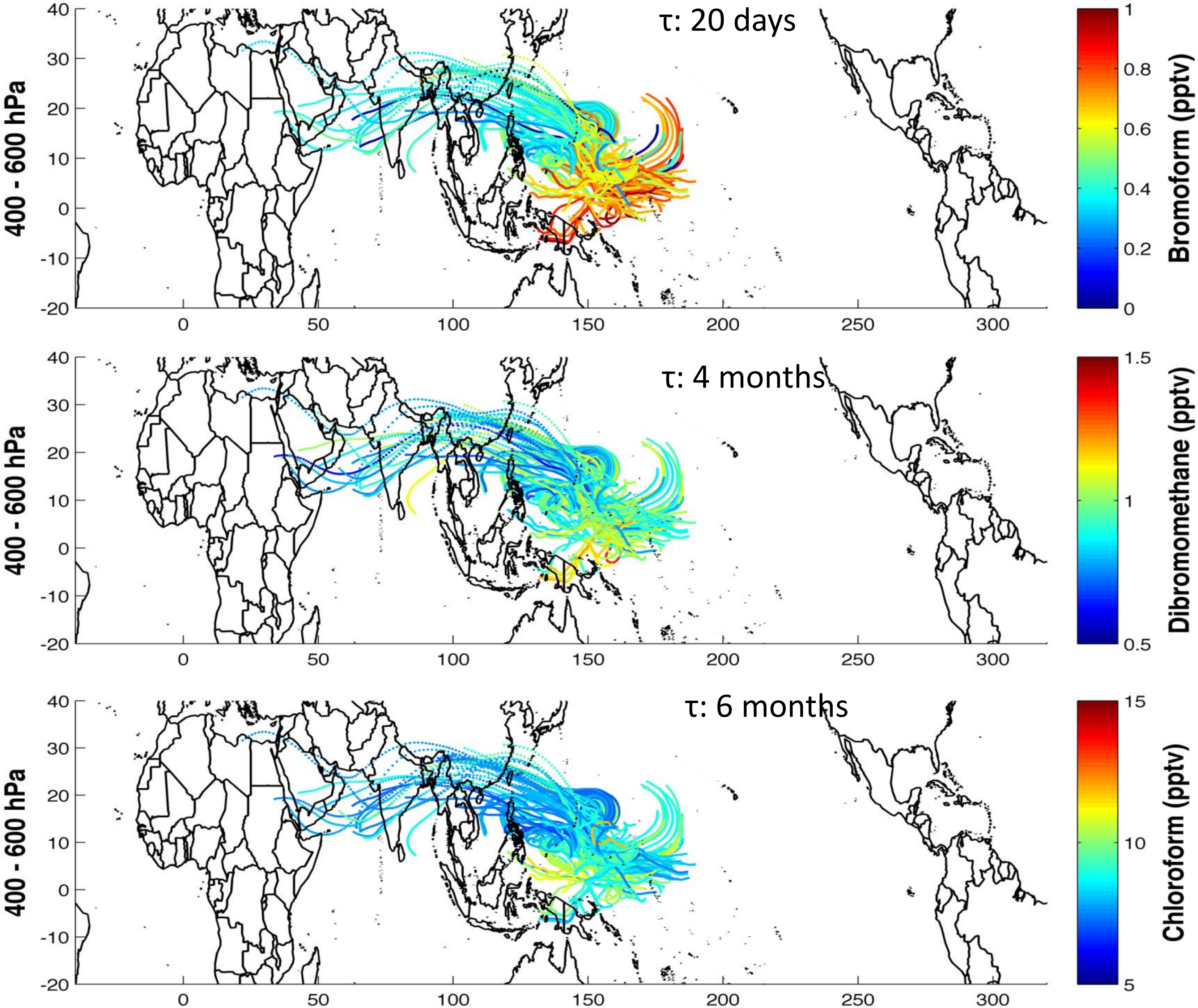
HYSPLIT Back Trajectory Analysis



- 5 day, isentropic back trajectories at TOGA observation time
- GDAS meteorology
- Excludes transit flights (1, 2, 16) and Japan flights (6, 15).
- Colored by O_3 at point of observation

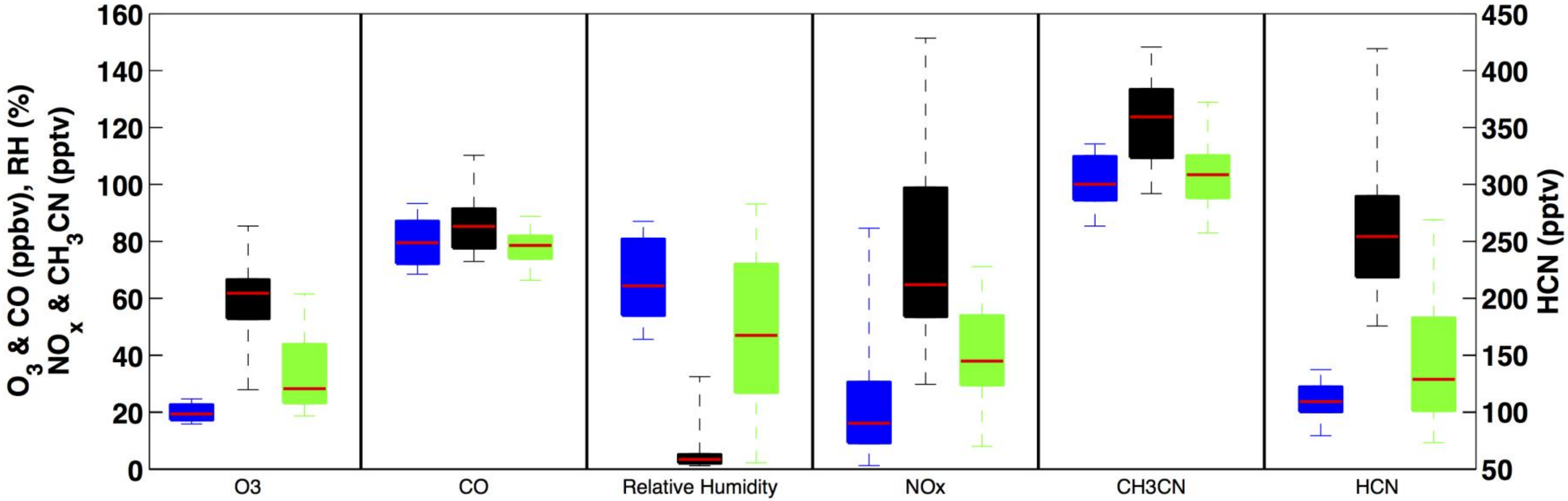

200 - 400 hPa
400 - 600 hPa
600 - 800 hPa
O_3 (ppbv)
O_3 (ppbv)
O_3 (ppbv)
40
30
20
10
0
-10
-20
0
50
100
150
200
250
300
80
60
40
20
0


τ: 30 days
400 - 600 hPa
CO (ppbv)
120
110
100
90
80
70
60
40
30
20
10
0
-10
-20
0
50
100
150
200
250
300
τ: 5 months
400 - 600 hPa
HCN (pptv)
300
250
200
150
100
50
τ: 1 week
400 - 600 hPa
NOx (pptv)
150
100
50
0

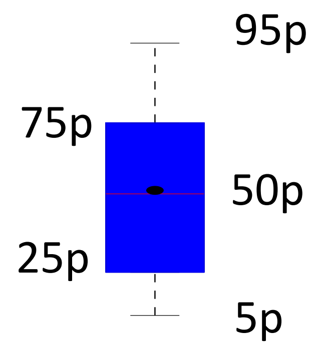

τ: 20 days
400 - 600 hPa
Bromoform (pptv)
τ: 4 months
400 - 600 hPa
Dibromomethane (pptv)
τ: 6 months
400 - 600 hPa
Chloroform (pptv)

Southern Hemisphere
Southeast Asia/China
Eastern Pacific

Species by air parcel origin (400-600 hPa)

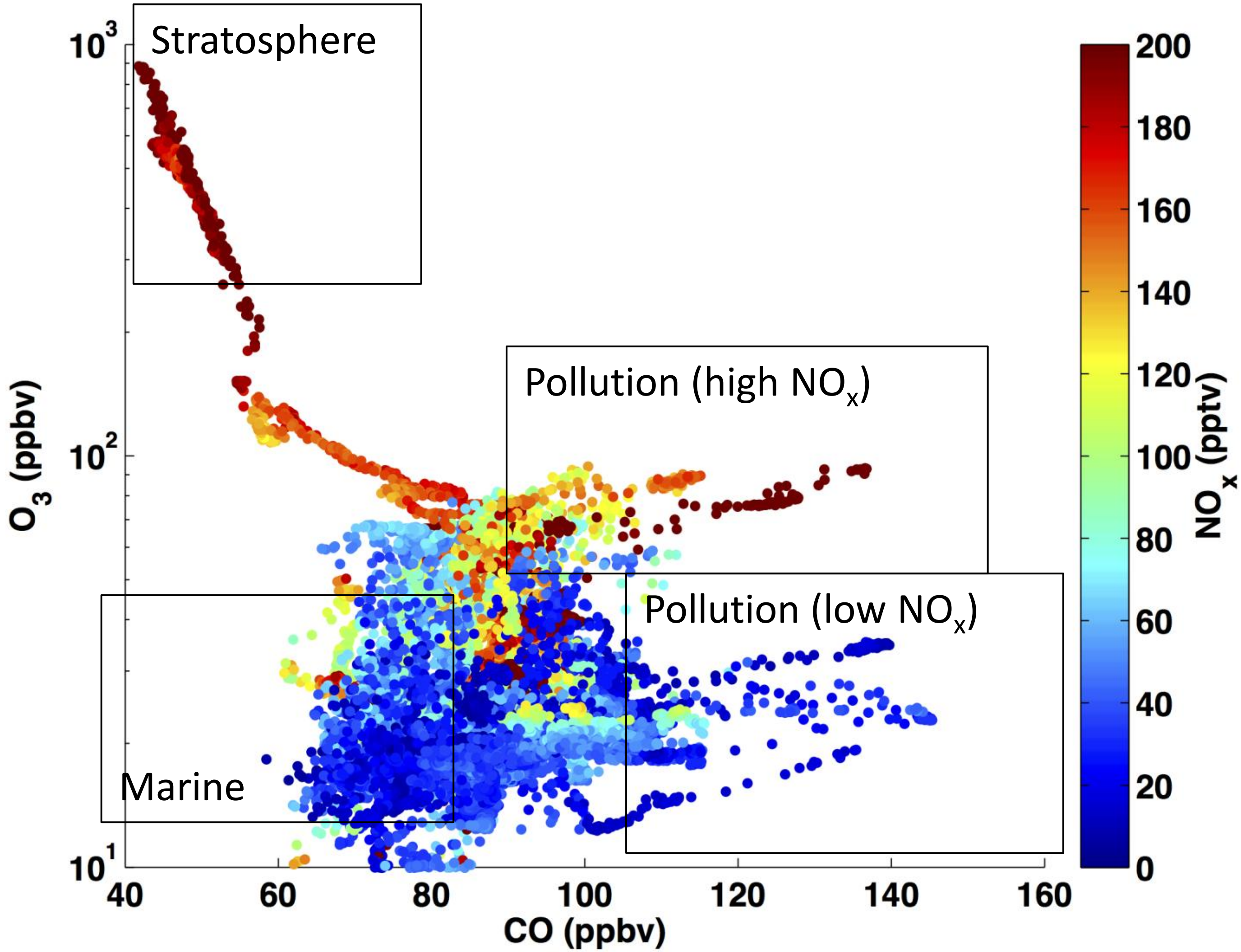


τ: 1 mo τ: 5 mo τ: 6.5 mo τ: 10 d

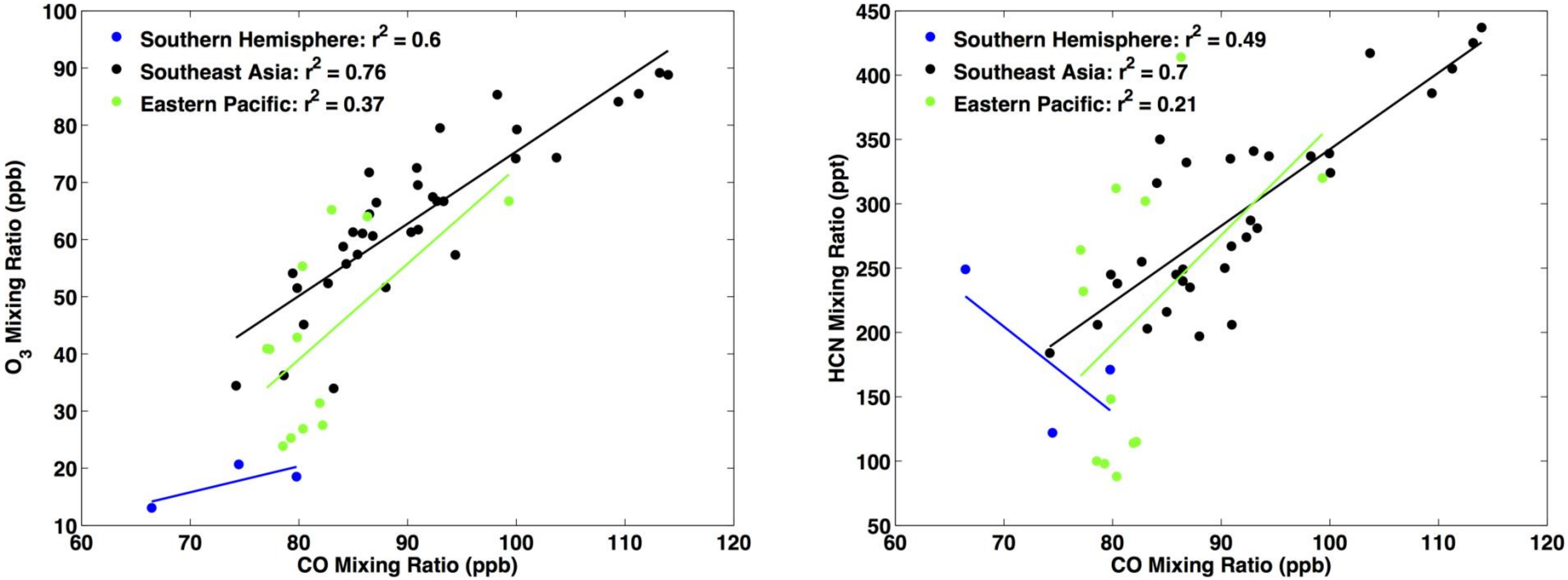


For air masses from southeast Asia:

- Enhanced O_3, HCN, CH_3CN, NO_x
- Depleted H_2O
- Slightly enhanced CO.



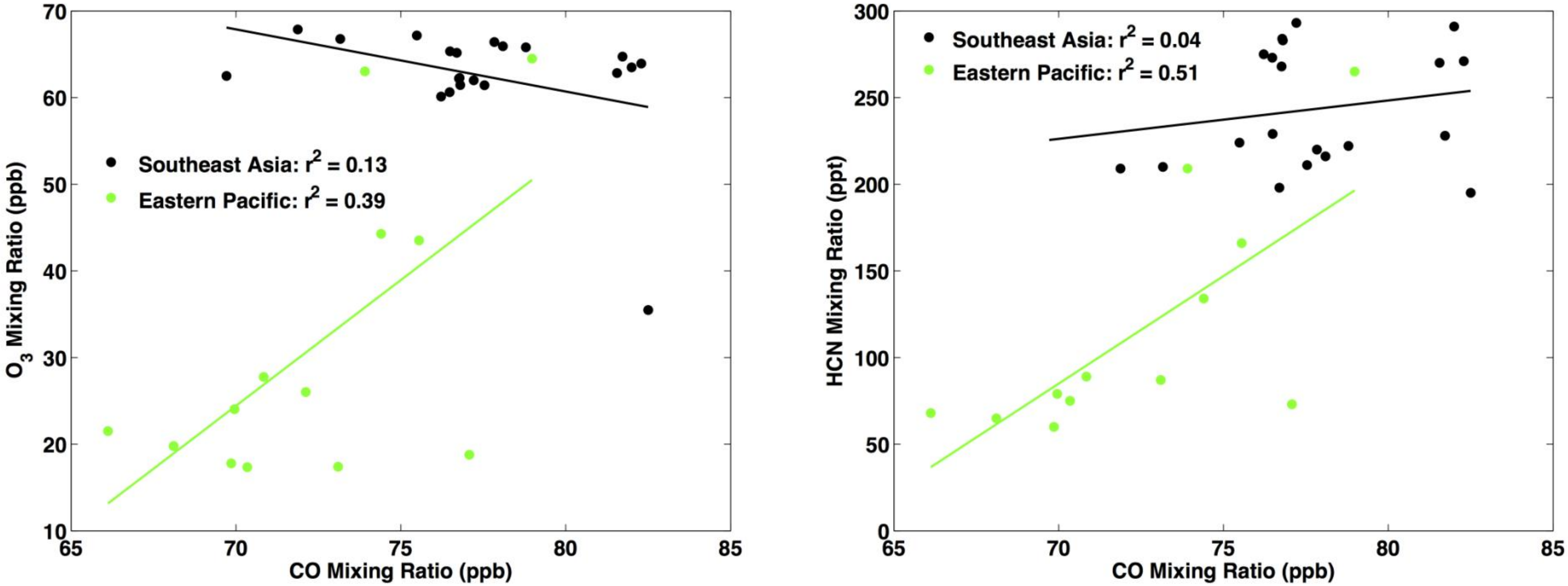
10s averaged data.



Note: For NO_x greater than 40 pptv, pressures between 400 & 800 hPa, and all flights with backtrajectories except RF08.

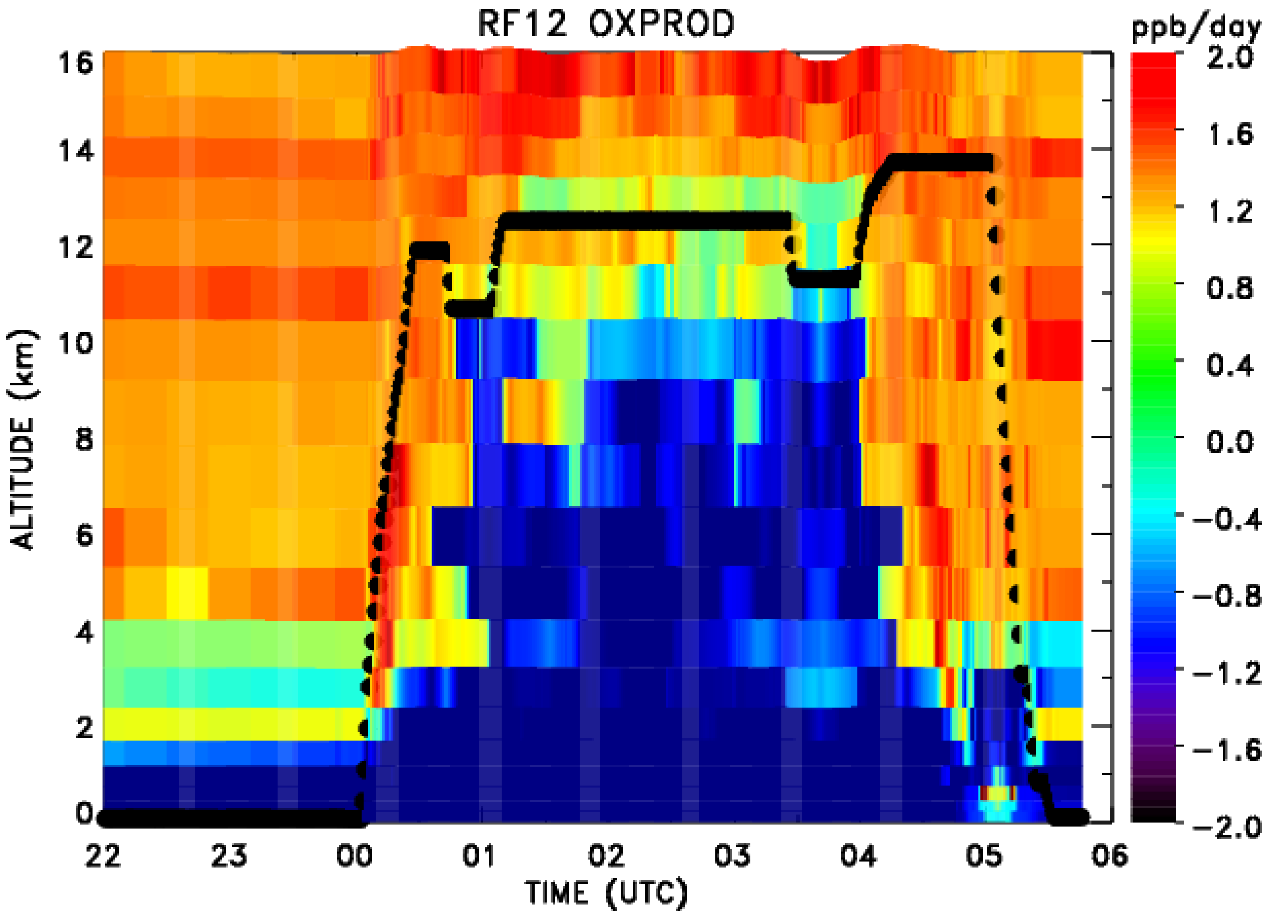
- Strong correlation between CO/O_3 and CO/HCN for air from southeast Asia implies a biomass burning origin.

CO Regressions for RF08



- Mean CO significantly lower (15 ppb) than for other flights
- Slight negative correlation for CO/O_3

O_3 Production along the flight track from RAQMS

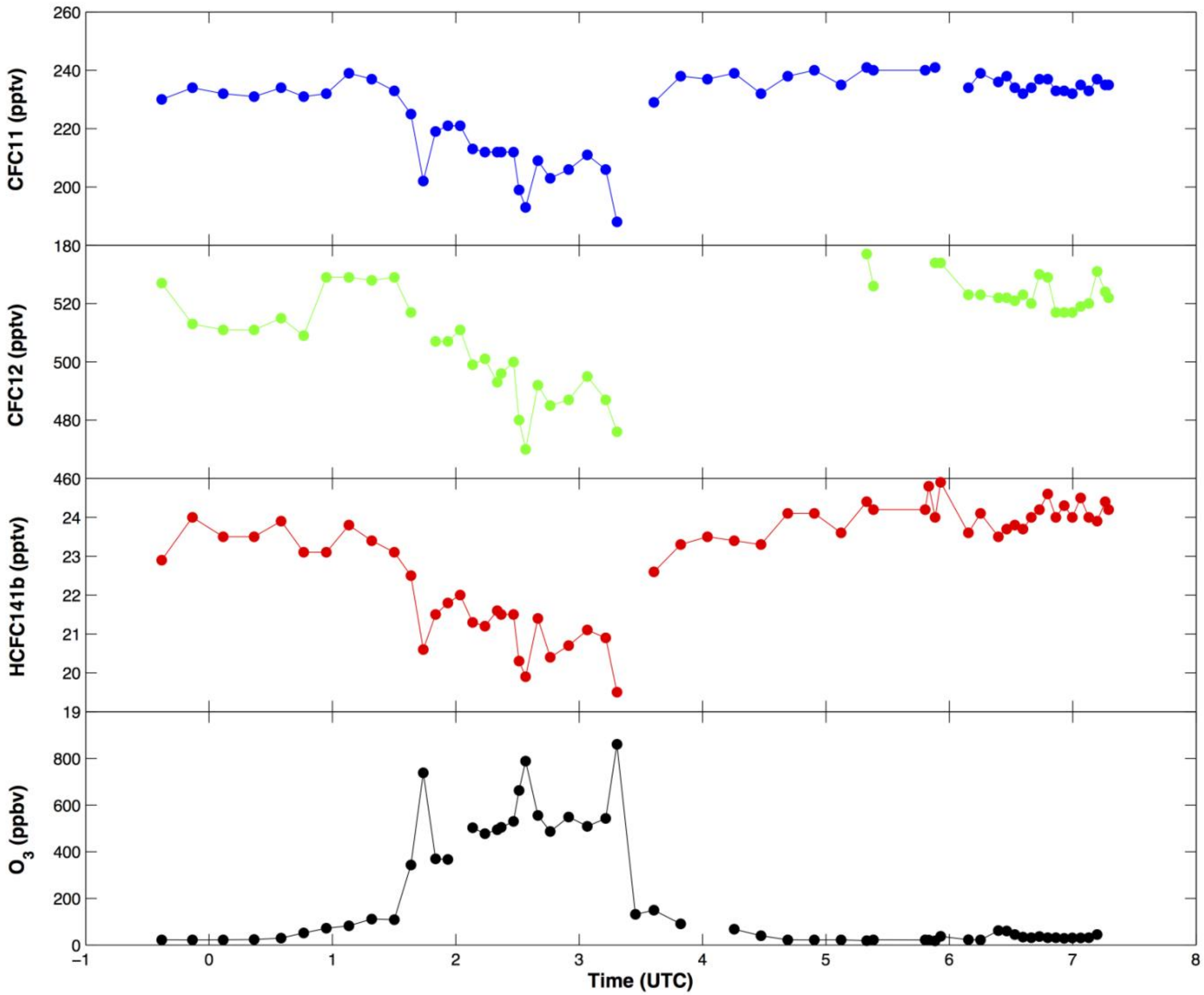


- Suggests can produce 10 ppb of O_3 over 5 days.

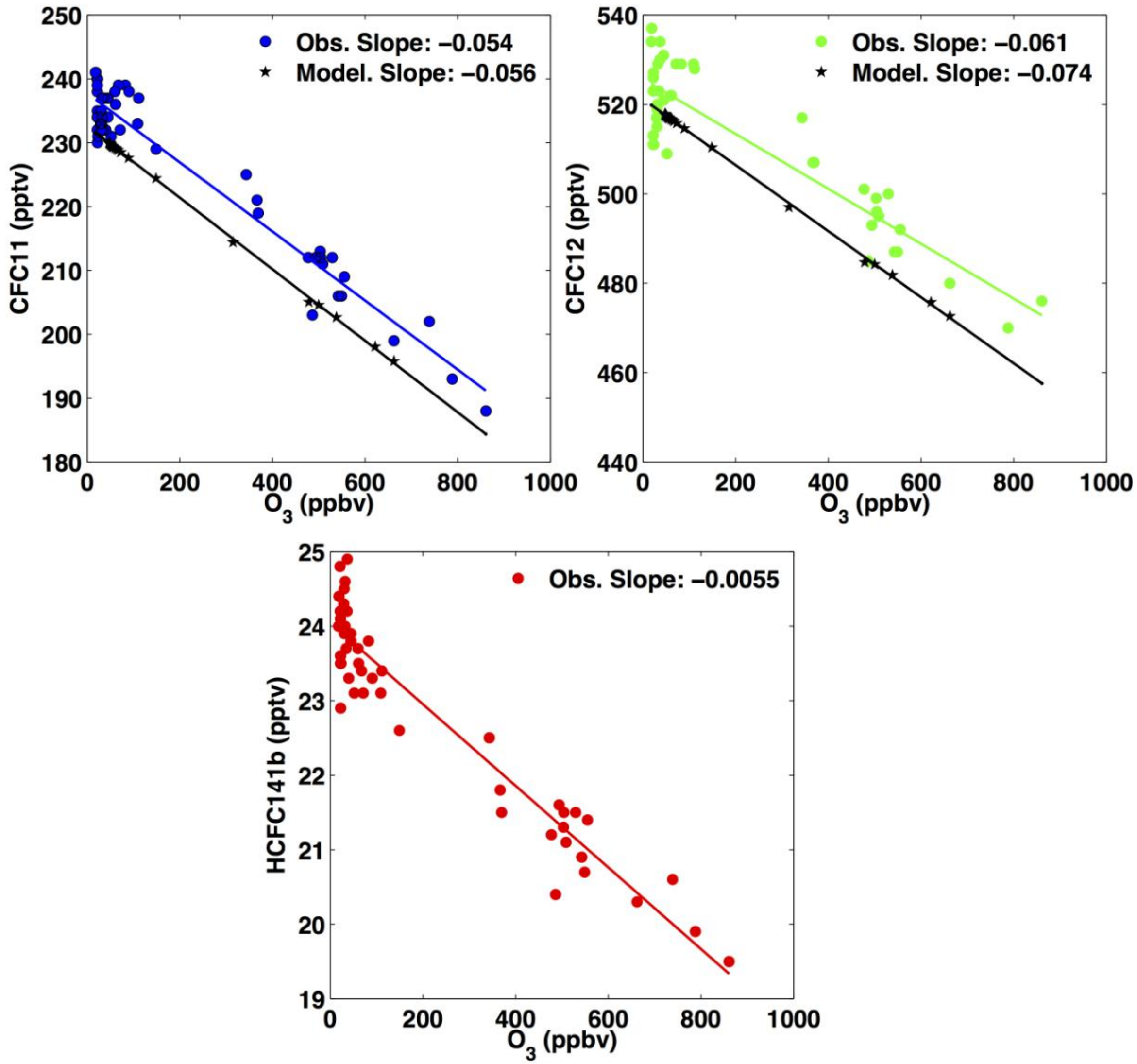
Figure courtesy of Brad Pierce

Time Series of Stratospheric Tracers for RF15

*Data from AWAS

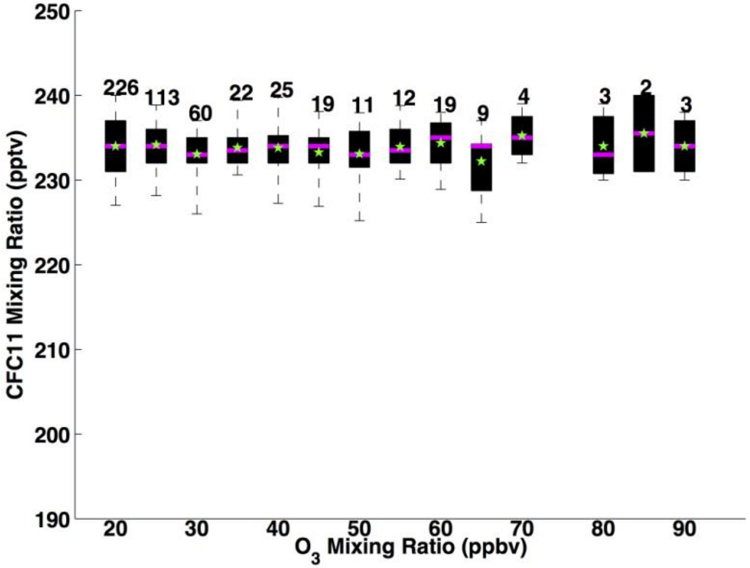


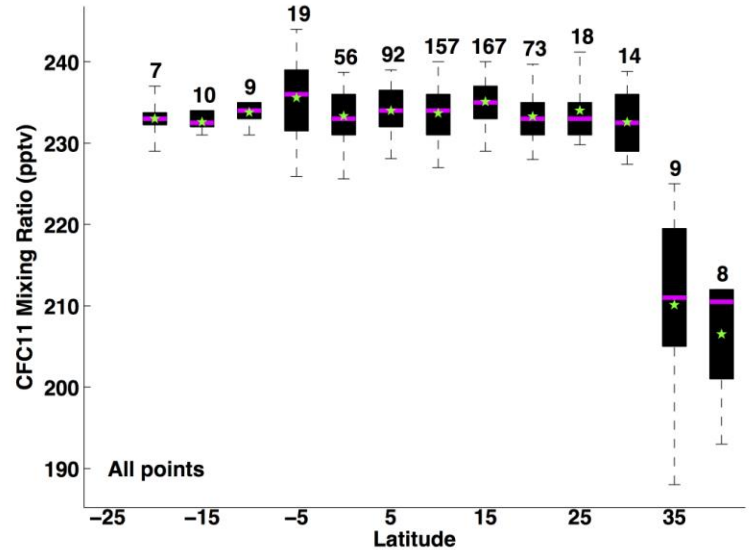
Strong anti-corrleation between O_3 and (H)CFC's in the stratosphere.

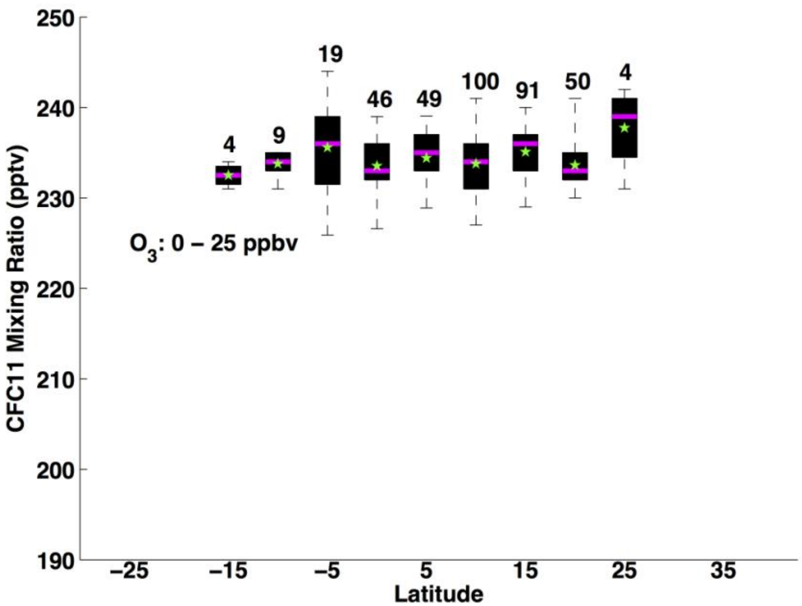


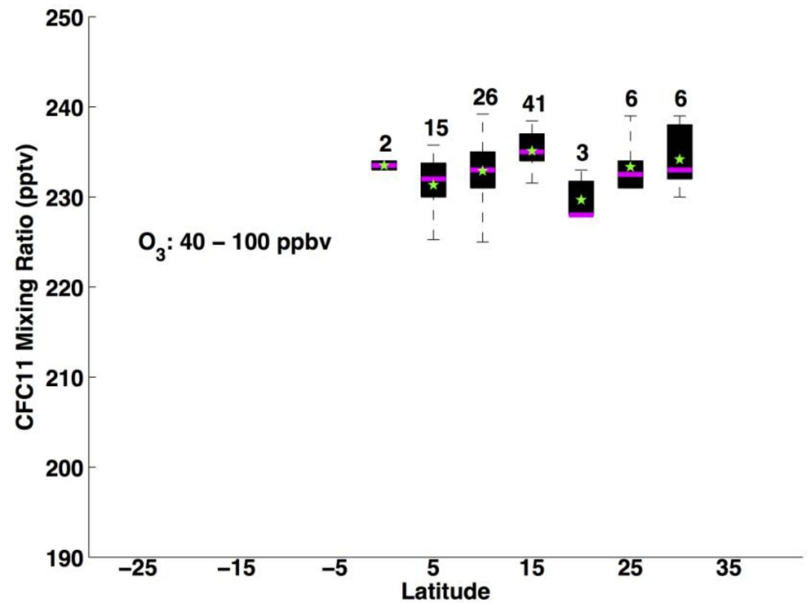
Strong anti-corrleation between O_3 and (H)CFC's in the stratosphere.

CFC11 (CCl_3F) **τ** = 45 yrs $\overline{[CFC11]}_{Bgrnd} = 235 \pm 4.5$





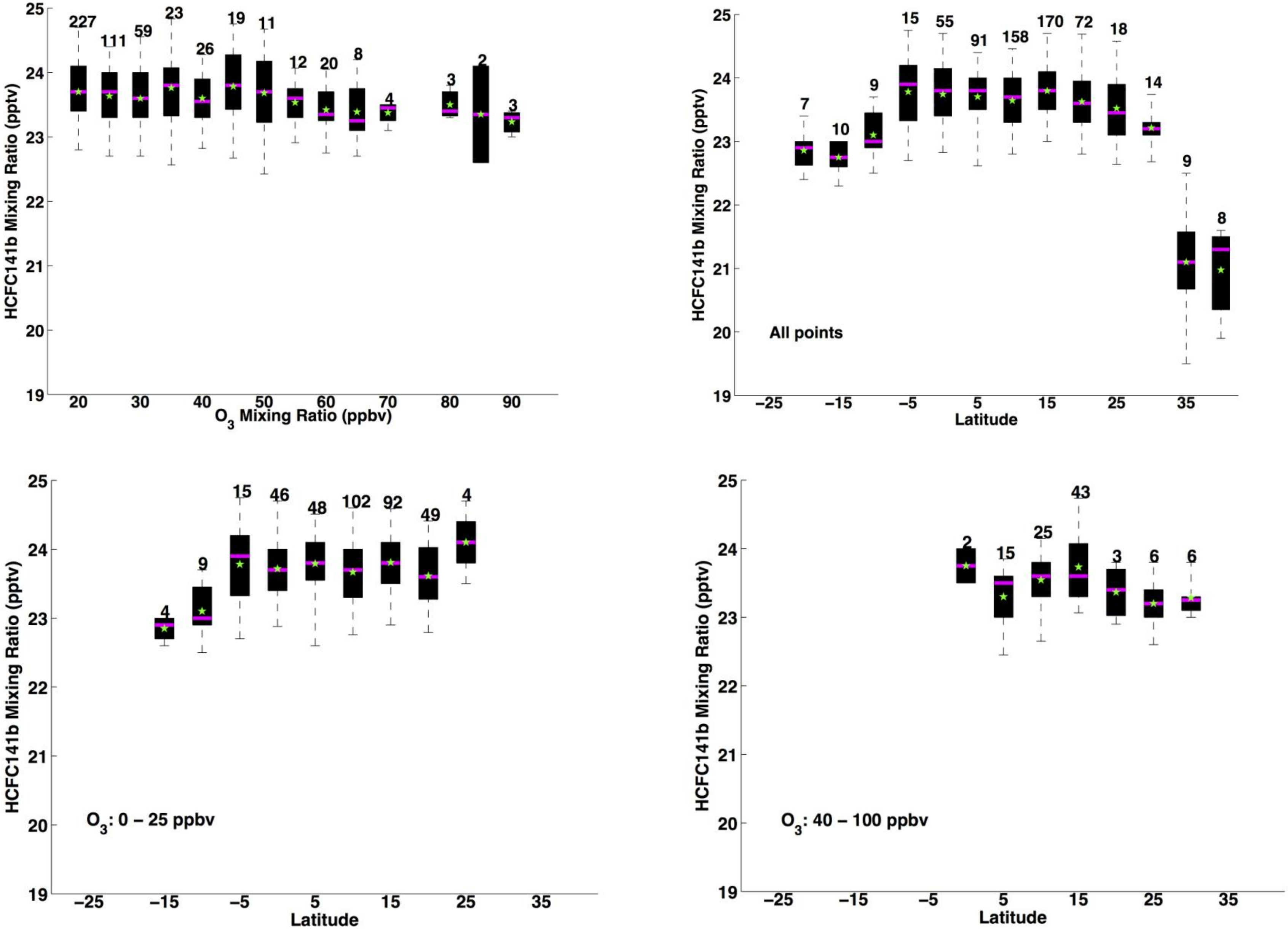




AWAS: flights 5-16

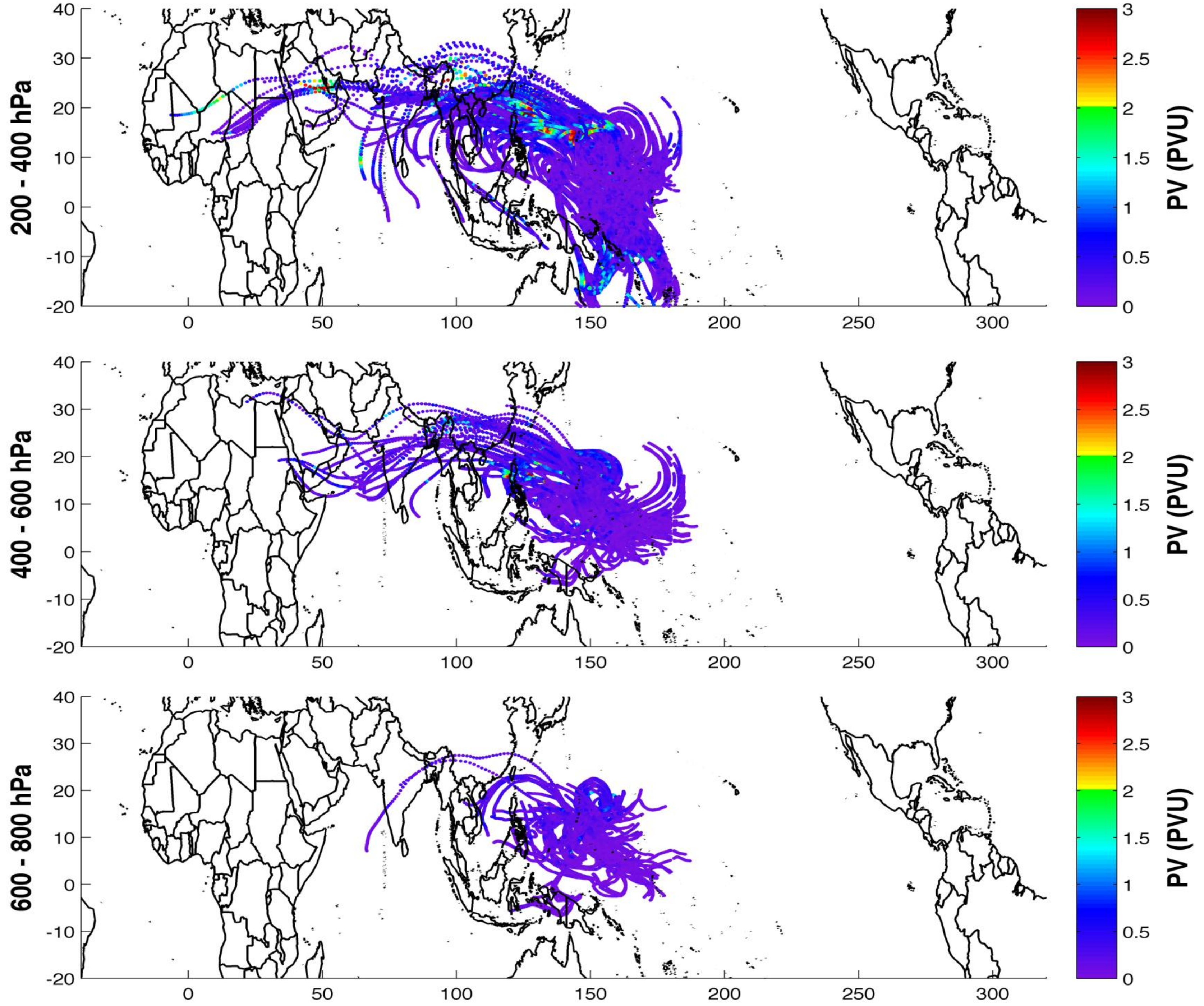
Little variation in CFC11 with latitude or O_3 concentration.

HCFC141b ($CH_2Cl_2CH_3F$) **τ = 11 yrs** $\overline{[CFC141b]}_{Bgrnd} = 23.88 \pm 0.62$



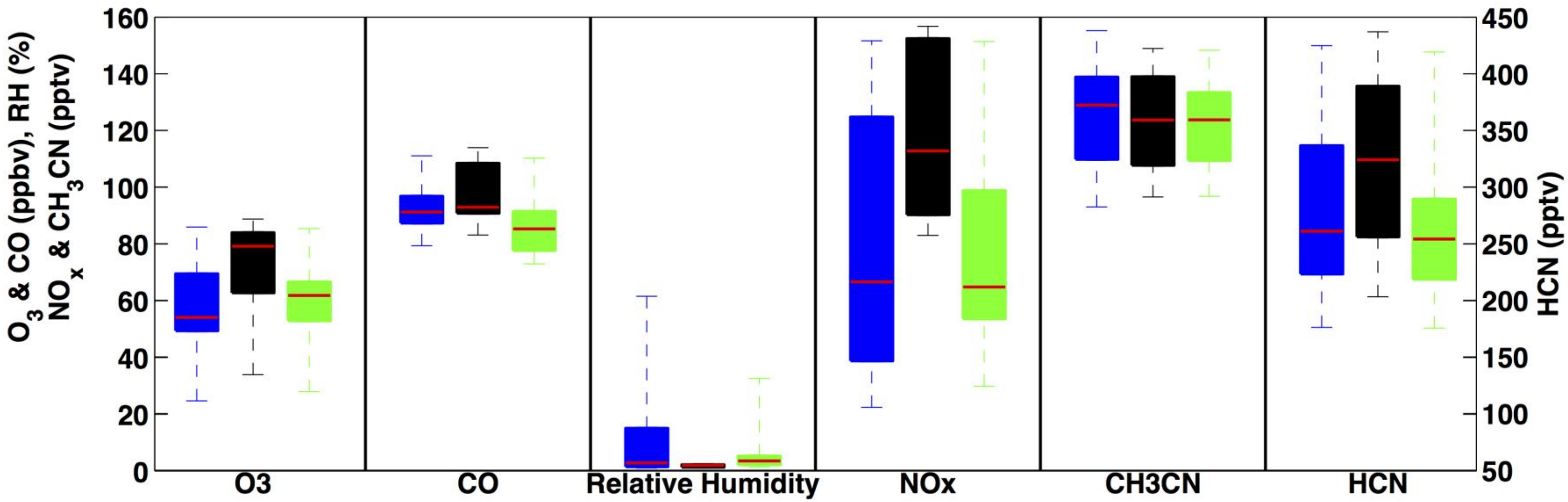
AWAS: flights 5-16

Little variation O_3 concentration. Hemispheric Gradient.

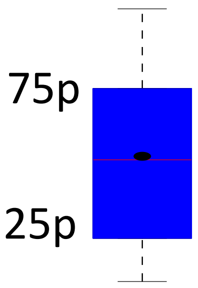

200 - 400 hPa
400 - 600 hPa
600 - 800 hPa
PV (PVU)
PV (PVU)
PV (PVU)
40
30
20
10
0
-10
-20
0
50
100
150
200
250
300
0
0.5
1
1.5
2
2.5
3

PV<2
PV>2
All

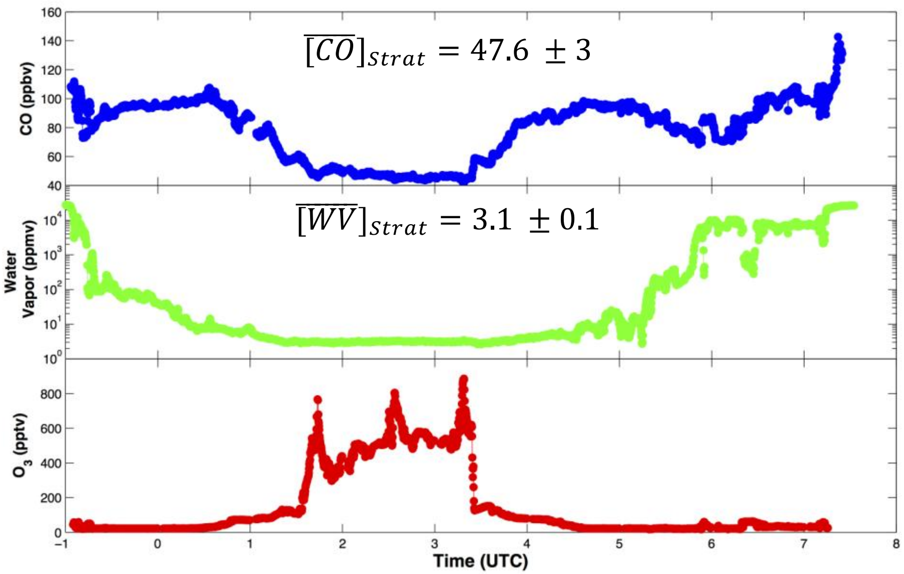
Species by stratospheric exposure (400-600 hPa)



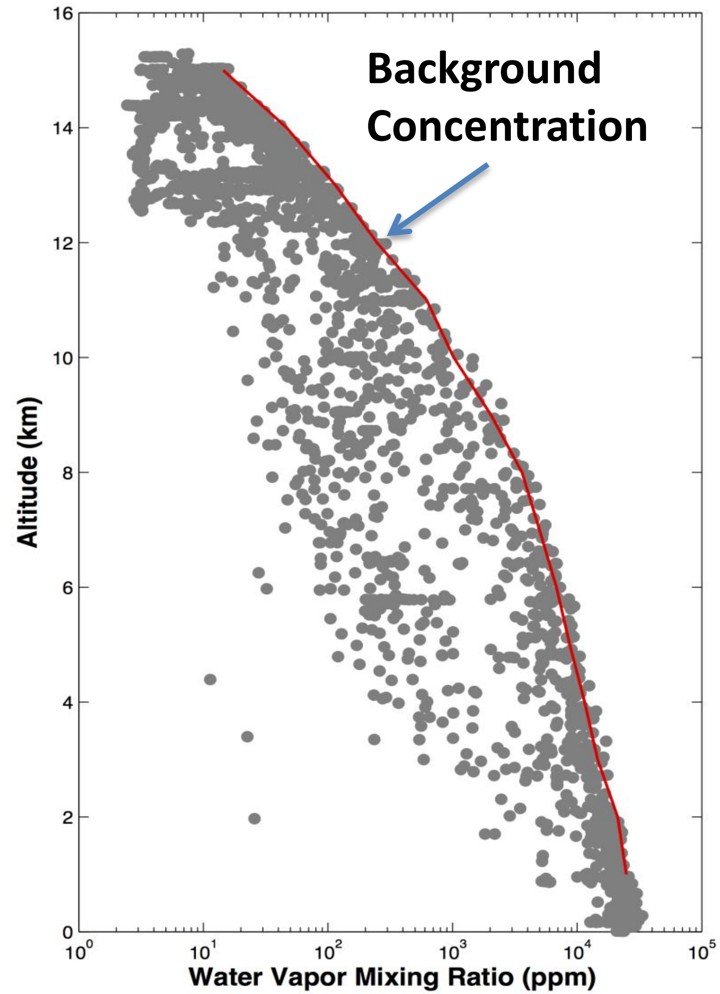
- Only parcels from southeast Asia
- PV>2 parcels have enhanced O_3, CO, HCN, and NO_x.
- If parcels have been stratospherically influenced, they've also mixed with air influenced by biomass burning.



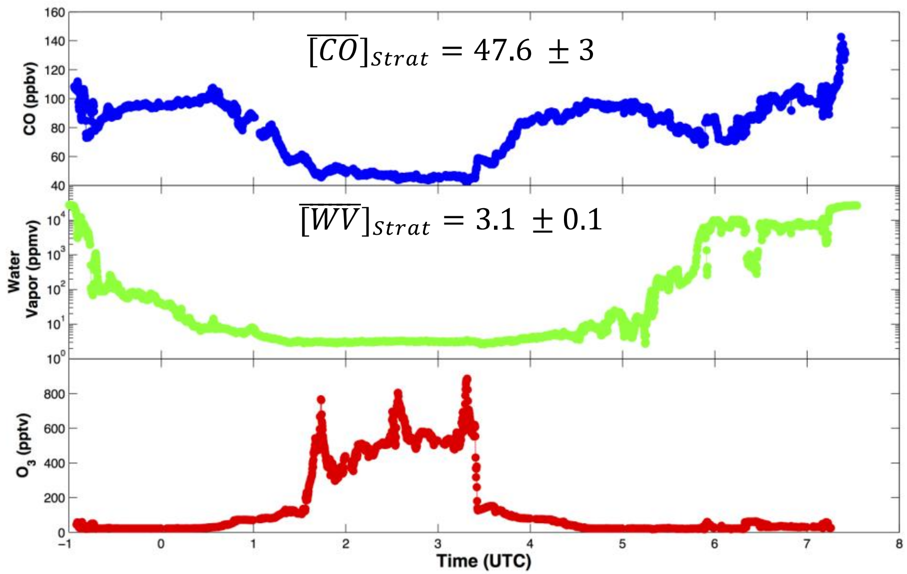
Time Series of RF15

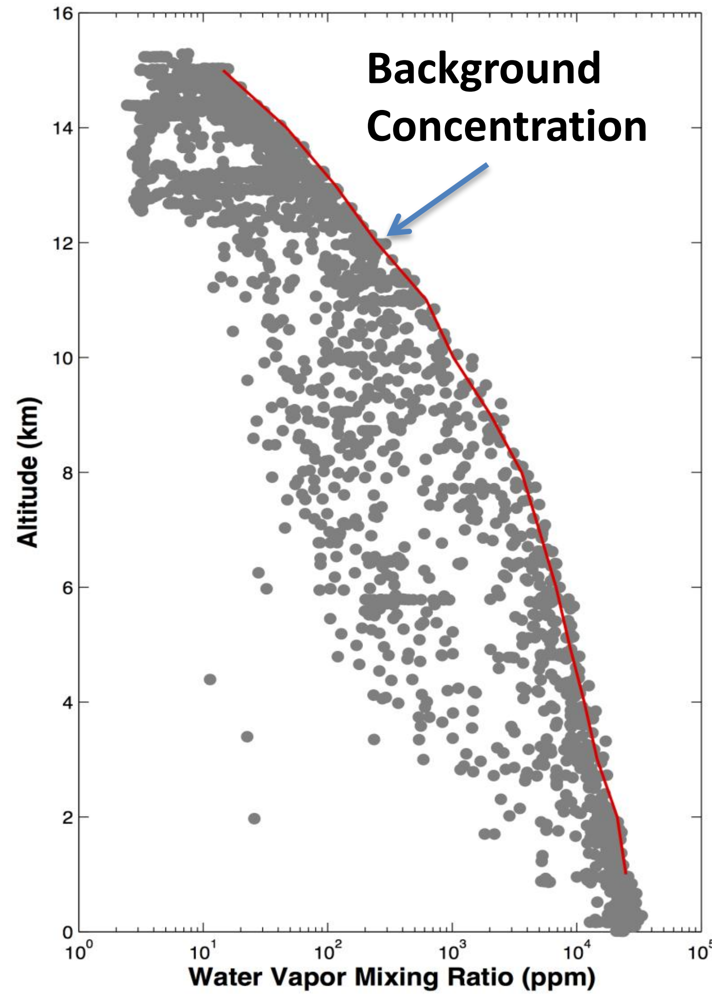


$H_2O_{obs} = f_{strat} * H_2O_{strat} + (1 - f_{strat}) * H_2O_{Trop}(z)$



Time Series of RF15

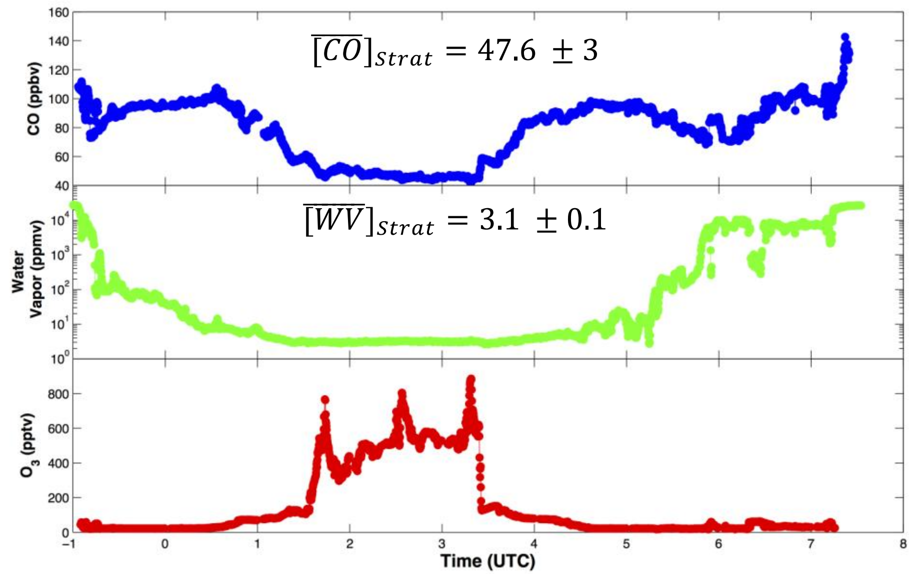


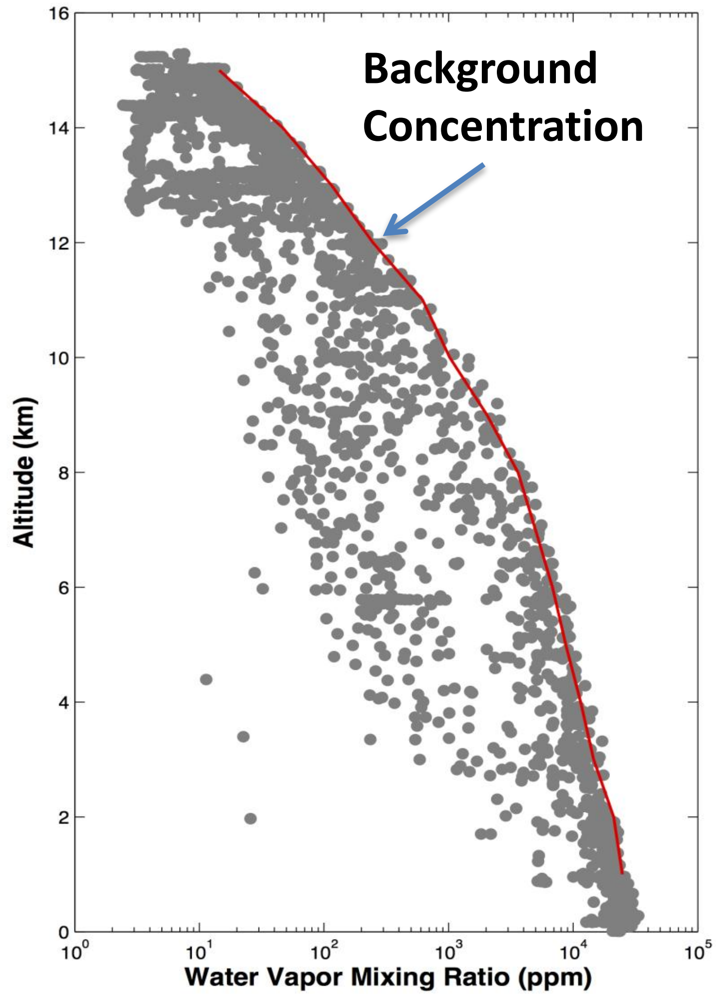


$H_2O_{obs} = f_{strat}{*}H_2O_{strat} + (1\text{-}f_{strat}){*}H_2O_{Trop}(z)$

$H_2O_{obs} = f_{strat}{*}3\text{ ppm} + (1\text{-}f_{strat}){*}H_2O_{Trop}(z)$

Time Series of RF15



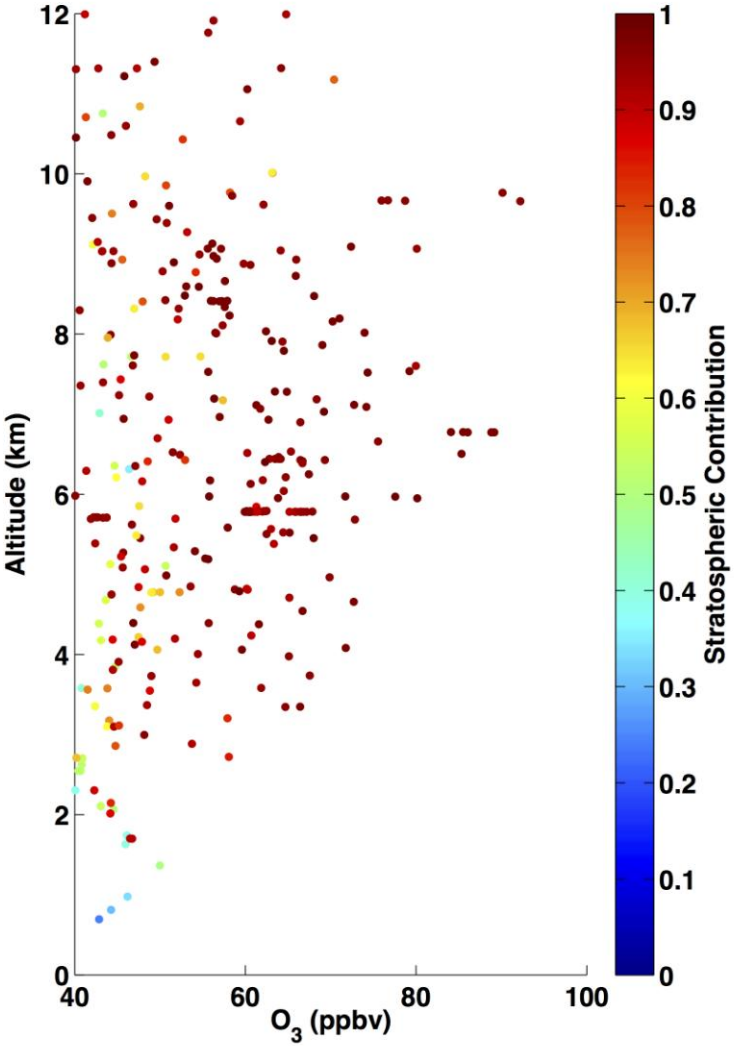


$H_2O_{obs} = f_{strat} * H_2O_{strat} + (1 - f_{strat}) * H_2O_{Trop}(z)$

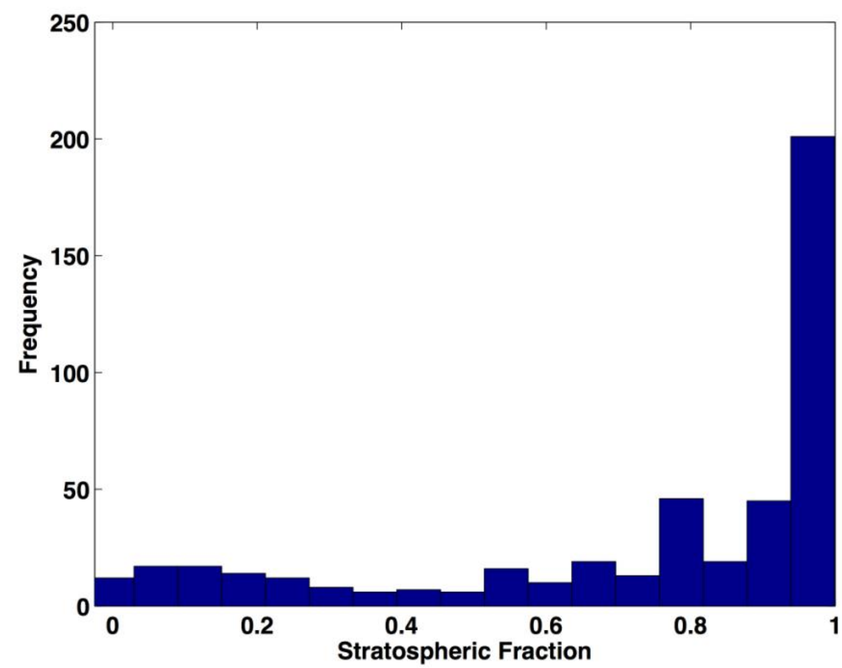
$H_2O_{obs} = f_{strat} * 3 \text{ ppm} + (1 - f_{strat}) * H_2O_{Trop}(z)$

$$f_{Strat} = \frac{H_2O_{Obs} - H_2O(z)_{Trop}}{3 - H_2O(z)_{Trop}}$$

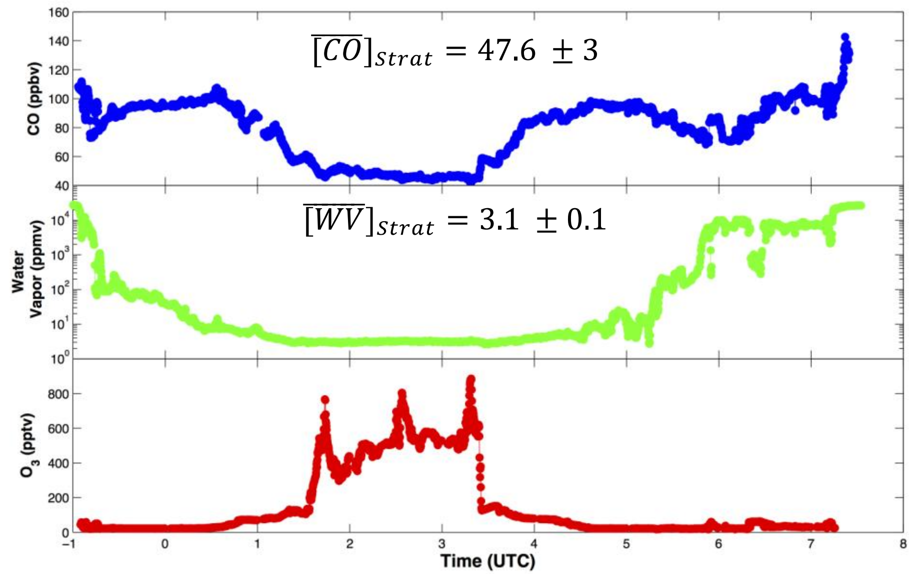
$$f_{Strat} = \frac{H_2O_{Obs} - H_2O(z)_{Trop}}{3 - H_2O(z)_{Trop}}$$



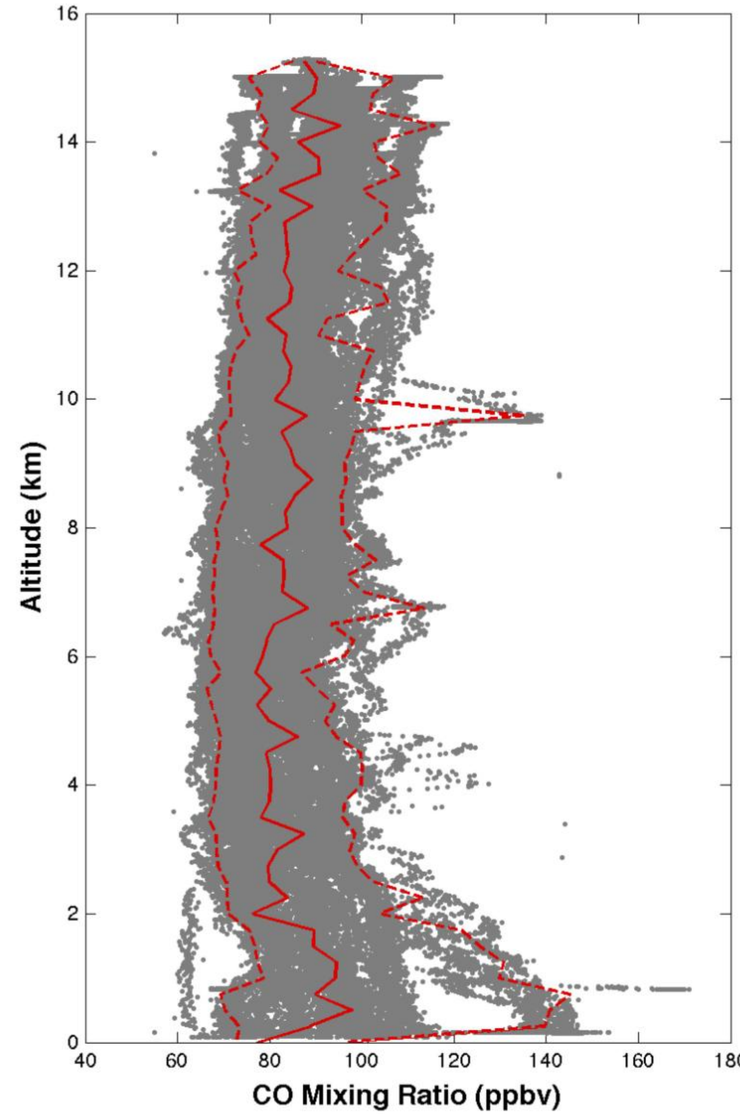
If low water vapor is from stratospheric mixing, ~90% of air must be stratospheric to match observations.



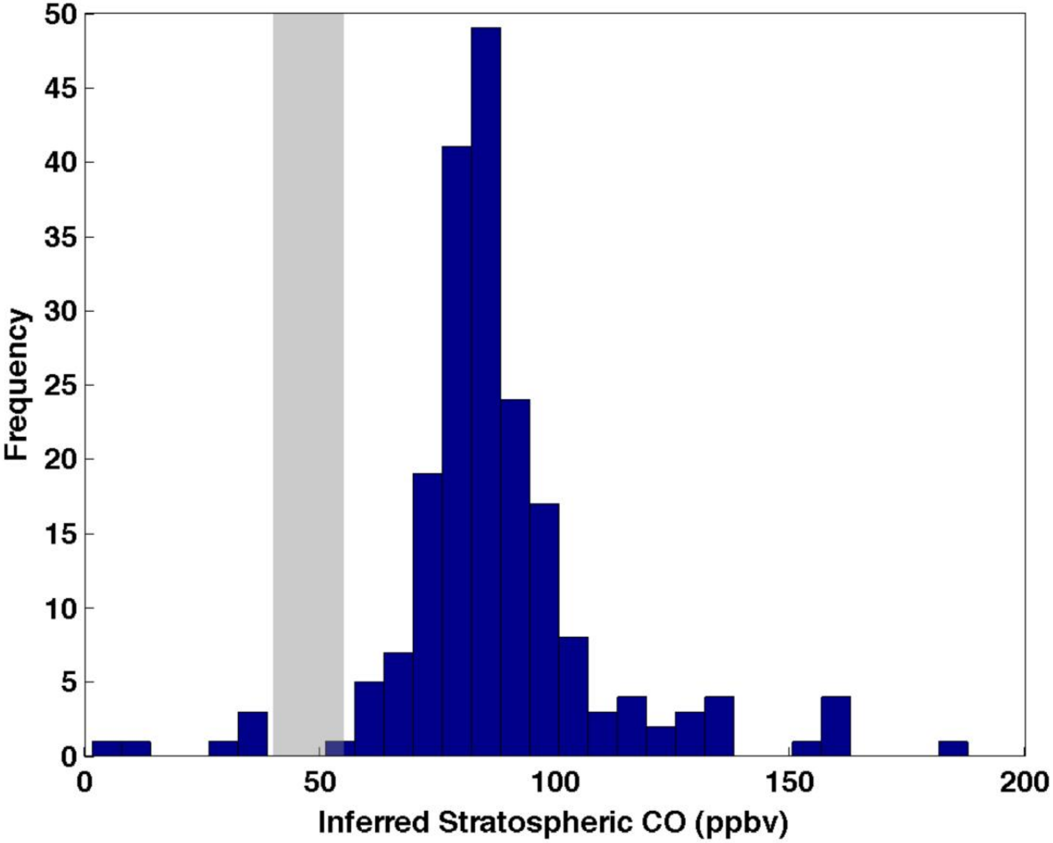
Time Series of RF15



$$CO_{Strat\ inferred} = \frac{CO_{Obs} - (1 - f_{Strat}) * CO_{Trop}}{f_{Strat}}$$



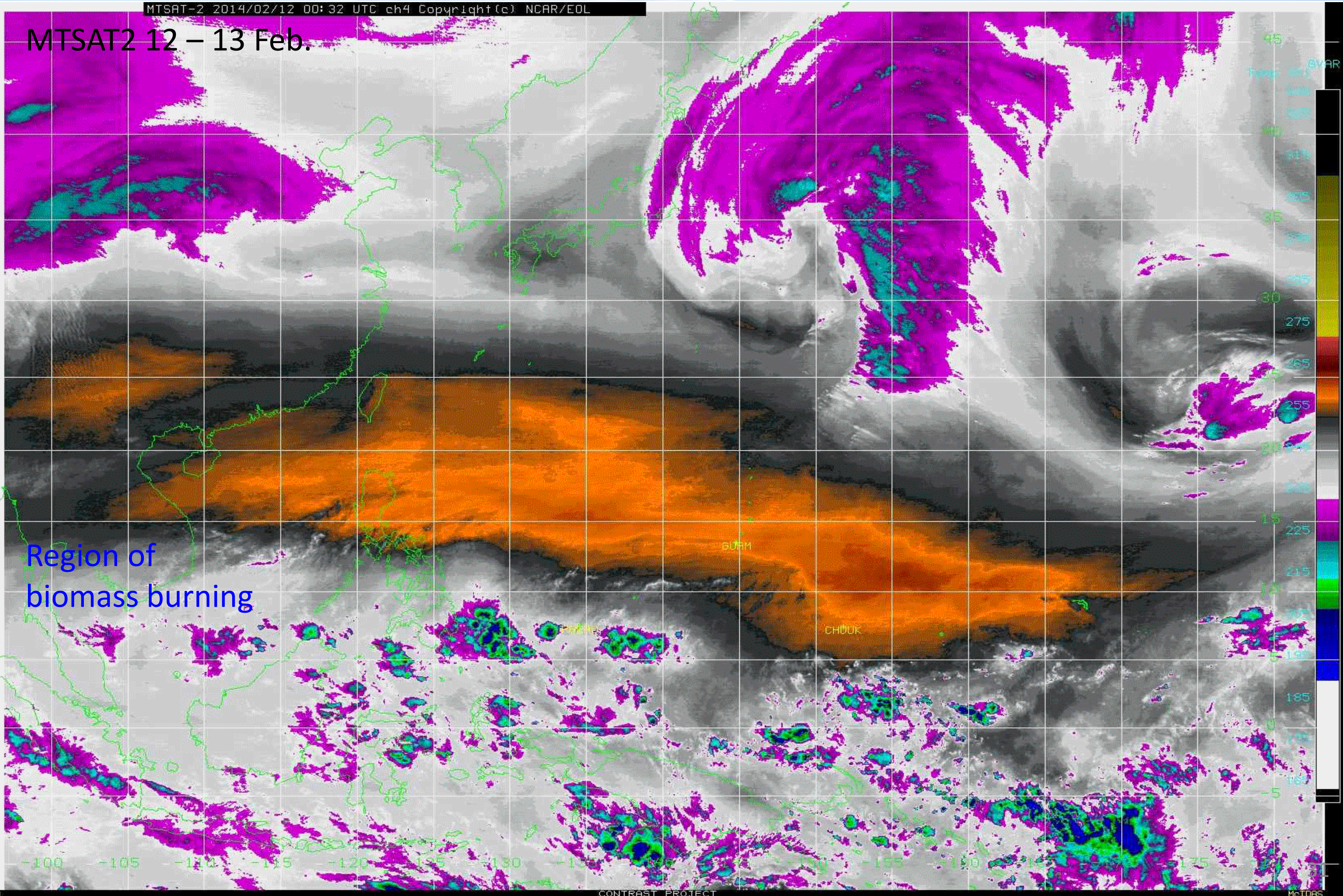
$$CO_{Strat\ inferred} = \frac{CO_{Obs} - (1 - f_{Strat}) * CO_{Trop}}{f_{Strat}}$$

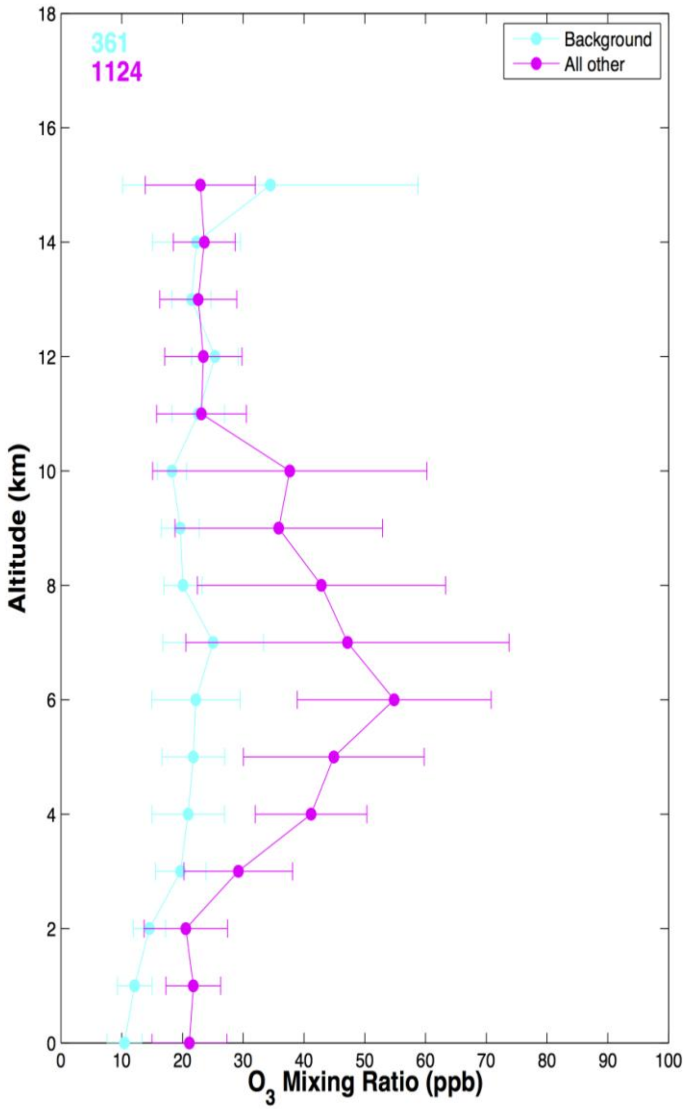


Calculated f_{strat} from water vapor requires stratospheric CO concentrations that are significantly higher than observations.

CONTRAST

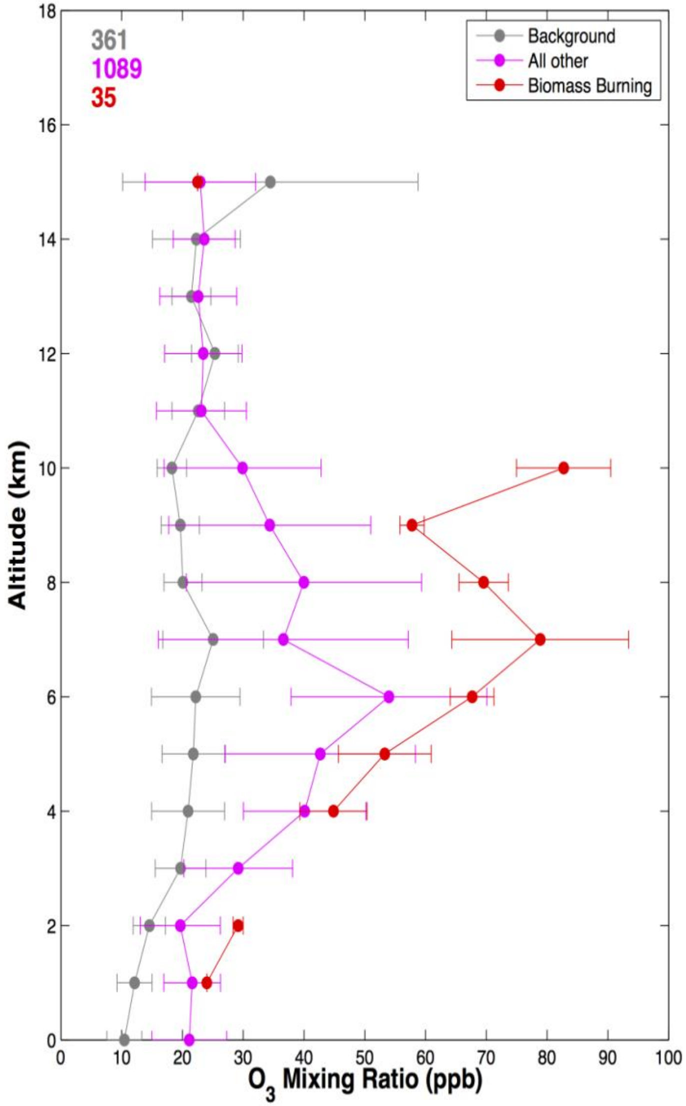
Guam, Jan–Feb 2014





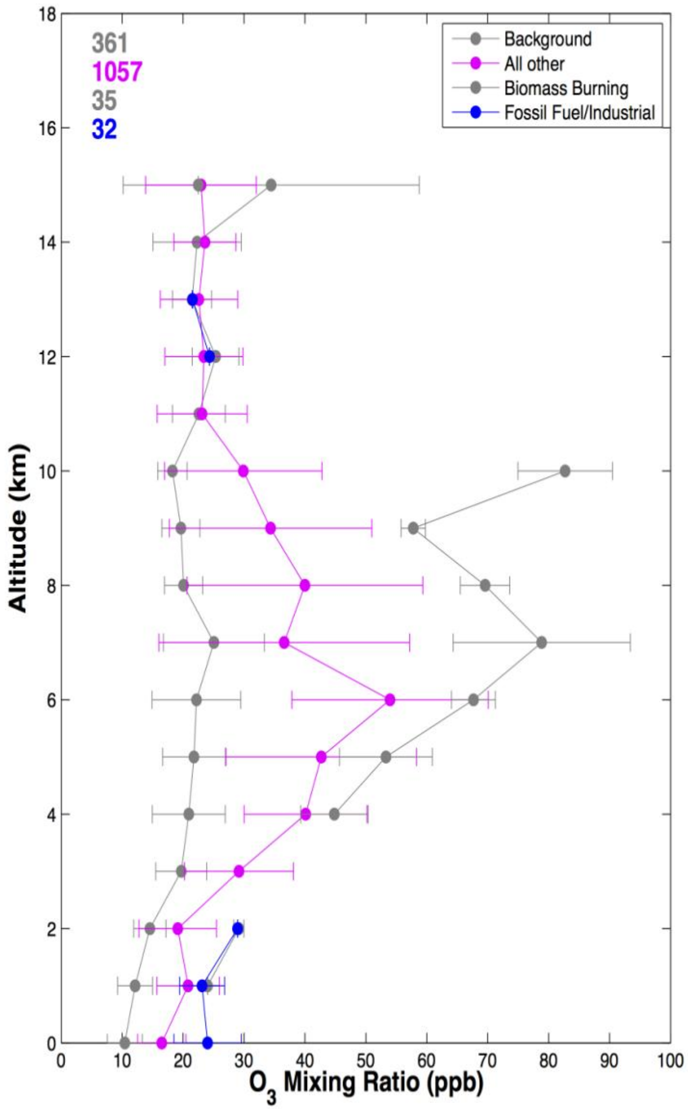
- Background: RH>45%, PV<0.5, & CO<90 ppbv

Data on TOGA time; abs(Latitude)<25



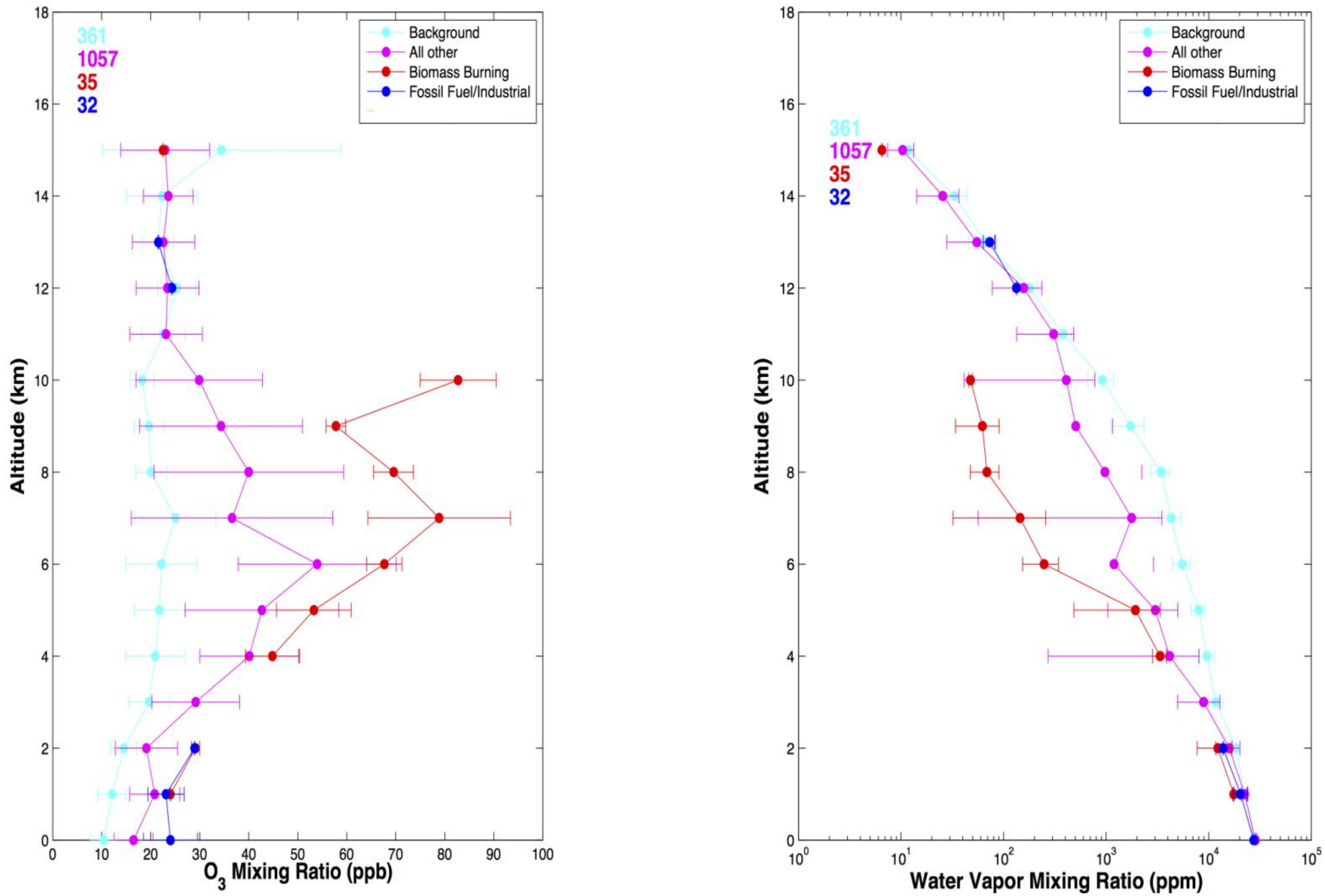
- Background: RH>45%, PV<0.5, & CO<90 ppbv
- BB: CO>90 ppbv, HCN>200 pptv, CH_3CN>100 pptv, PV<0.5

Data on TOGA time; abs(Latitude)<25



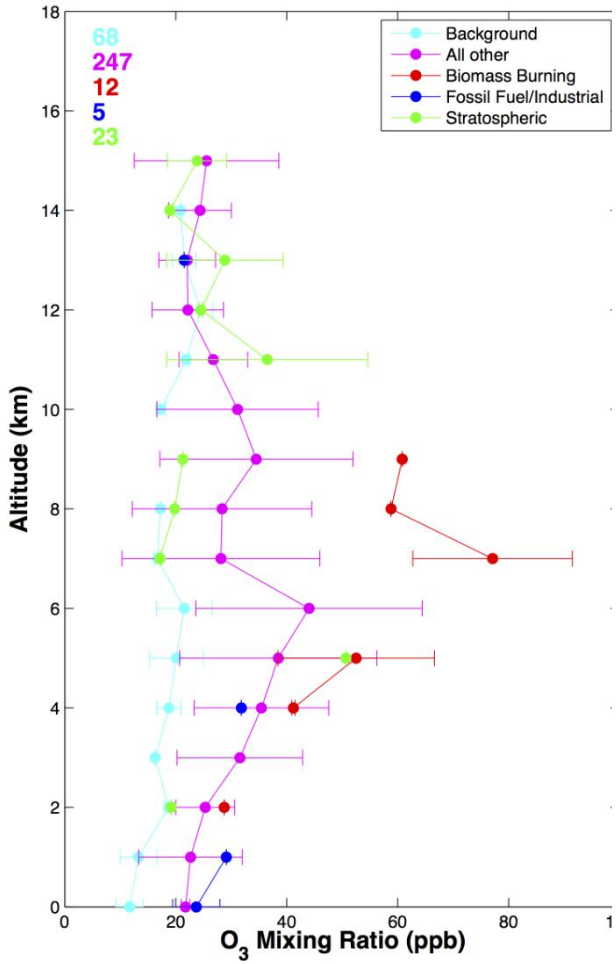
- Background: RH>45%, PV<0.5, & CO<90 ppbv
- BB: CO>90 ppbv, HCN>200 pptv, CH_3CN>100 pptv, PV<0.5
- FFI: CO>90 ppbv, HCN<200 pptv, C_2Cl_4> 1.75 pptv, C_3H_8>100 pptv, PV<0.5

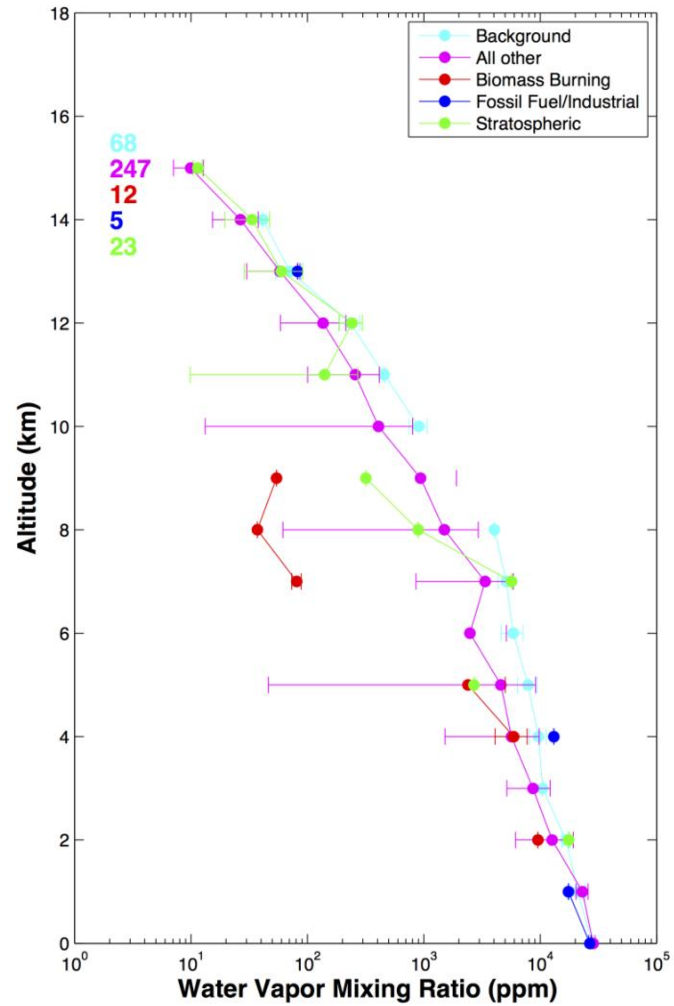
Data on TOGA time; abs(Latitude)<25



Departures from background evident in both O_3 and water vapor.

- Strat: CFC11<230, HCFC141b<23





Departures from background evident in both O_3 and water vapor.

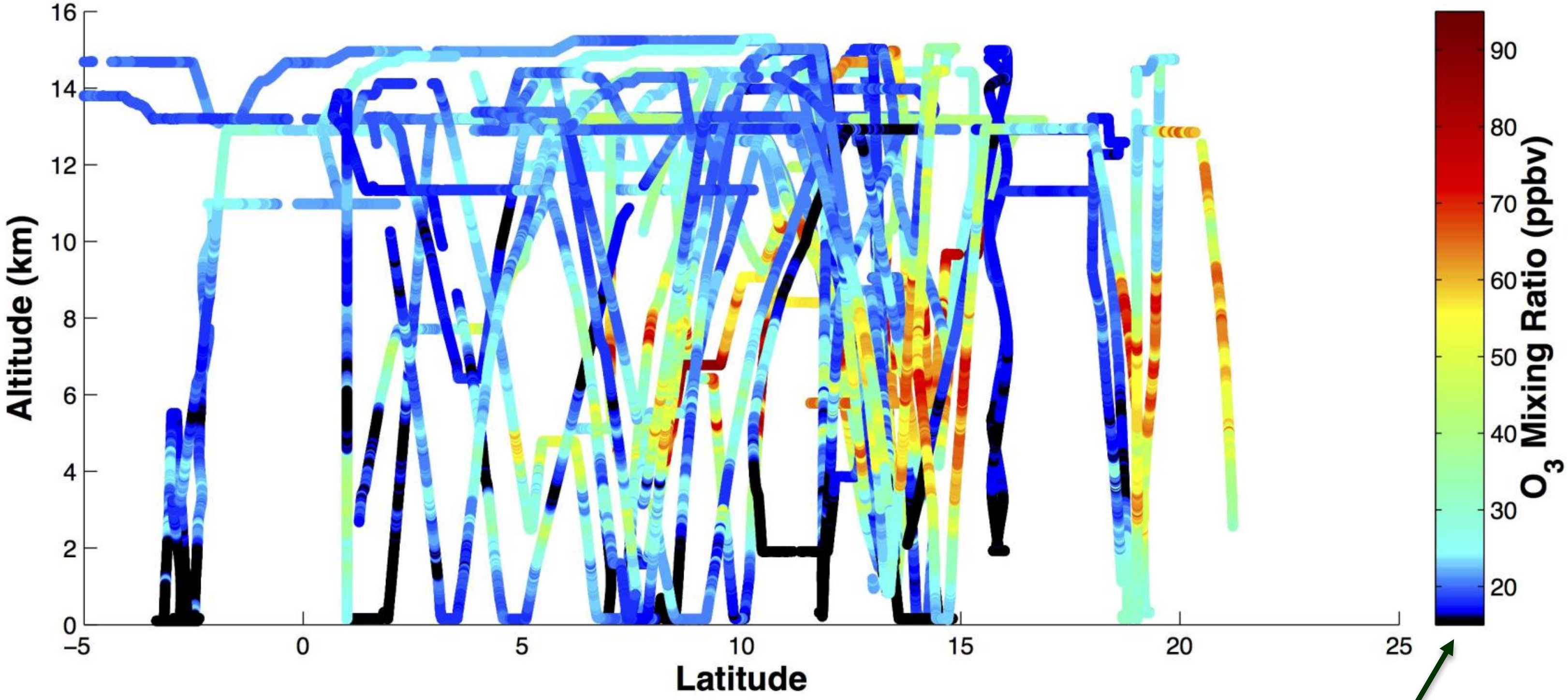
Conclusions:

- Biomass burning a large contributor to enhanced, mid-tropospheric O_3.
- Limited stratospheric influence based on CO, CFC's, etc.
- Low water vapor possibly results from dry/pyro-convective processes.

Future Work:

- Compare CAM-Chem/RAQMS output from baseline runs and those without biomass burning emissions.
- Determine convective influence along the back trajectories.

BACKUP SLIDES

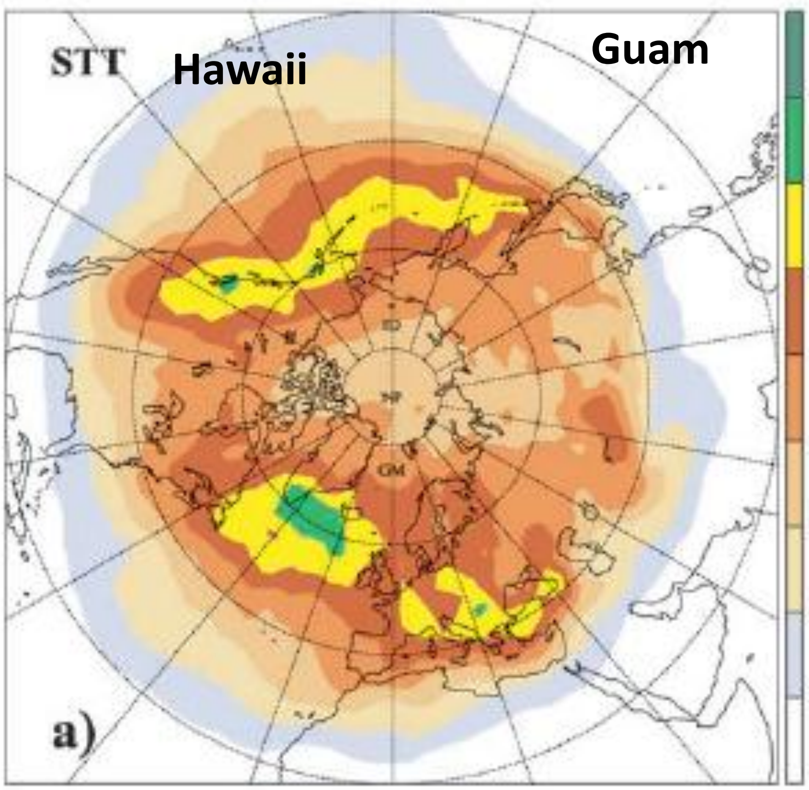


Note elevated O_3 is higher, and spread over a larger altitude range, in the northern part of the tropics

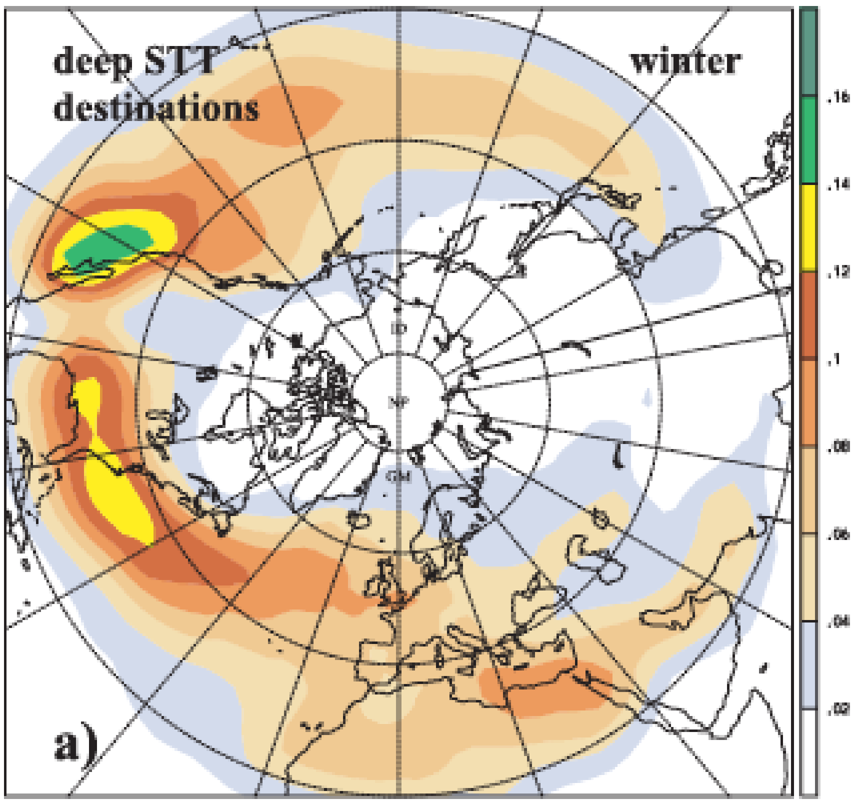
Black denotes O_3 below 15 ppb

Preliminary data: please do not cite or distribute

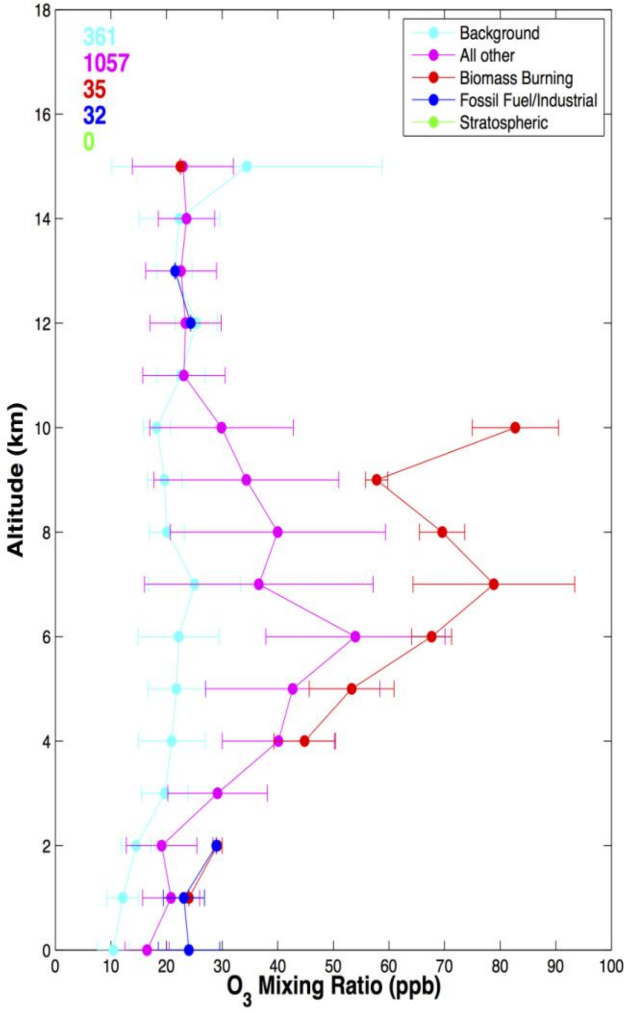
Distribution of mean stratosphere to troposphere (STT) exchange

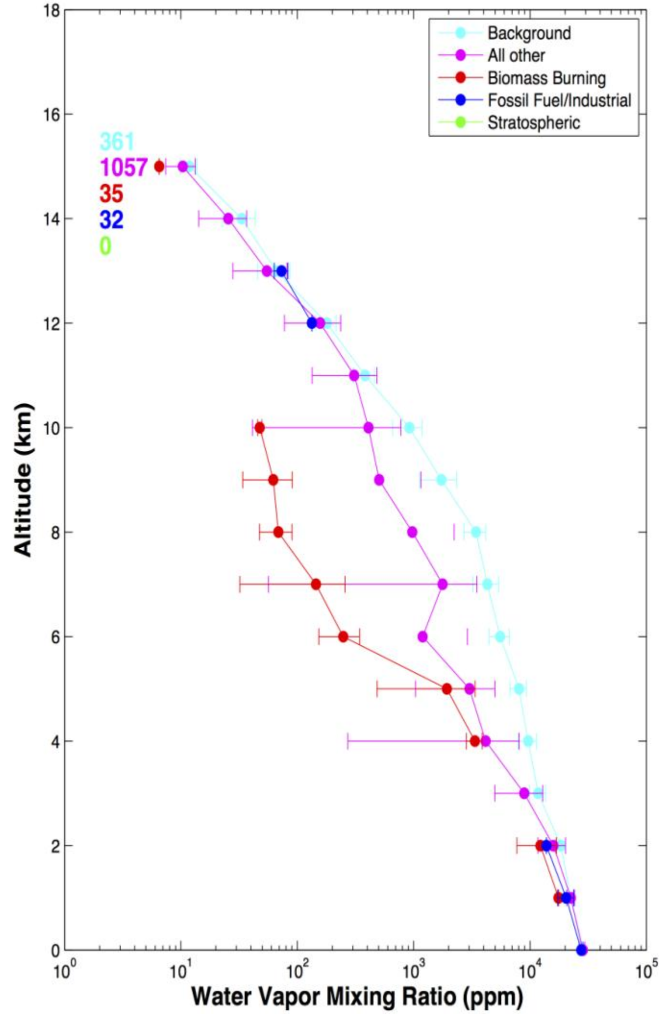


Destination of STT exchange below 700 hPa



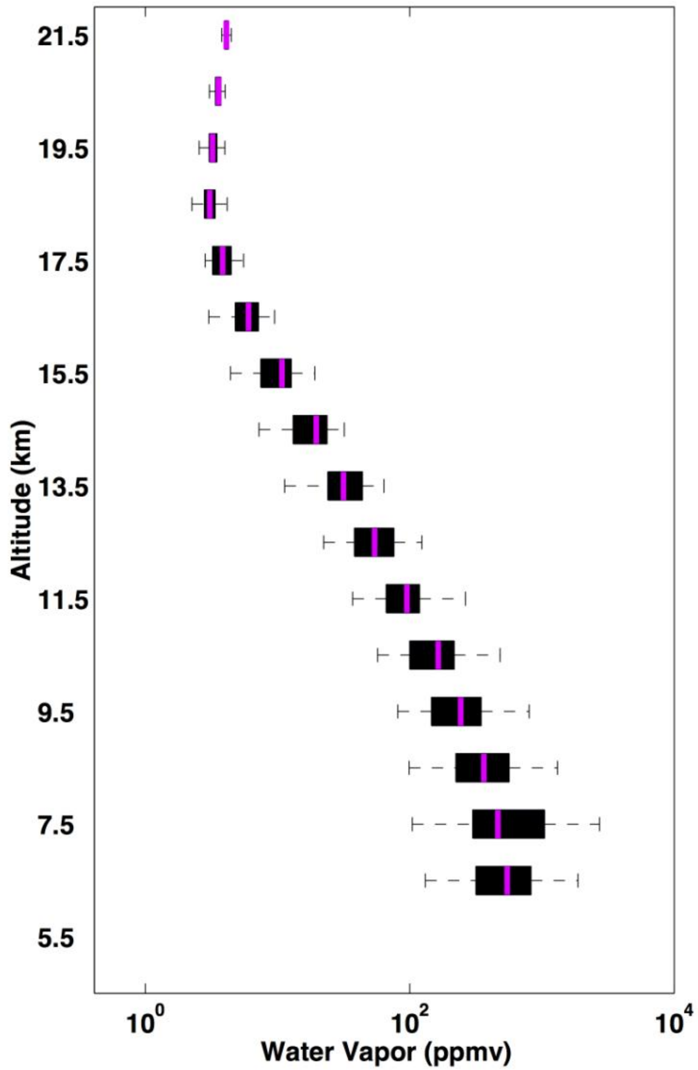
Sprenger et al. (2003)

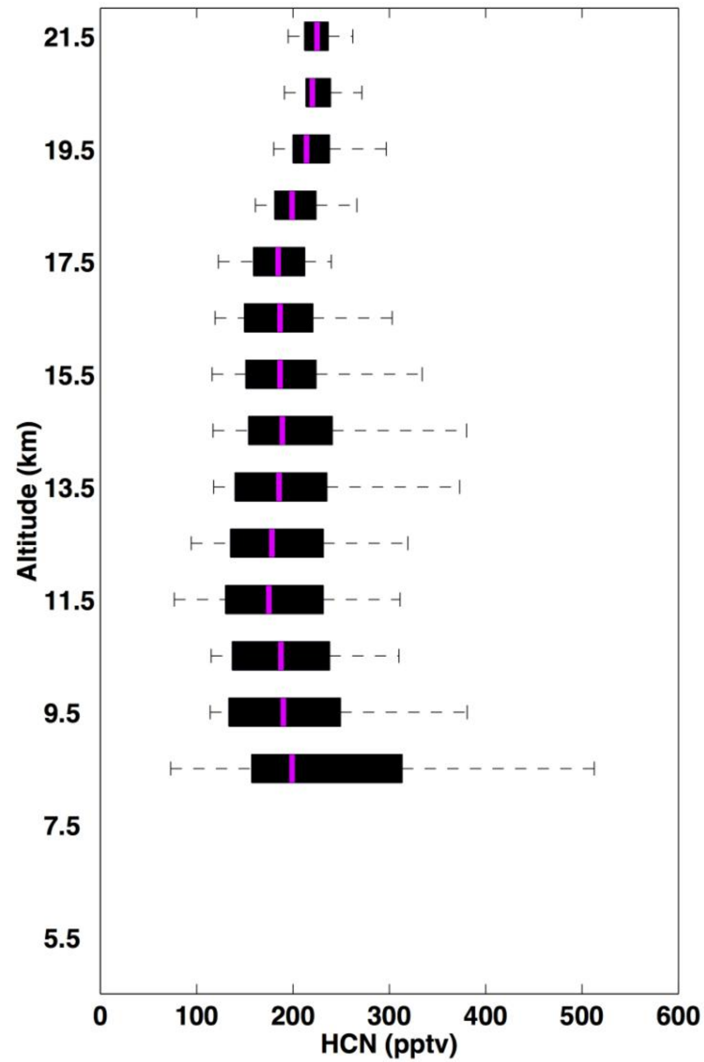




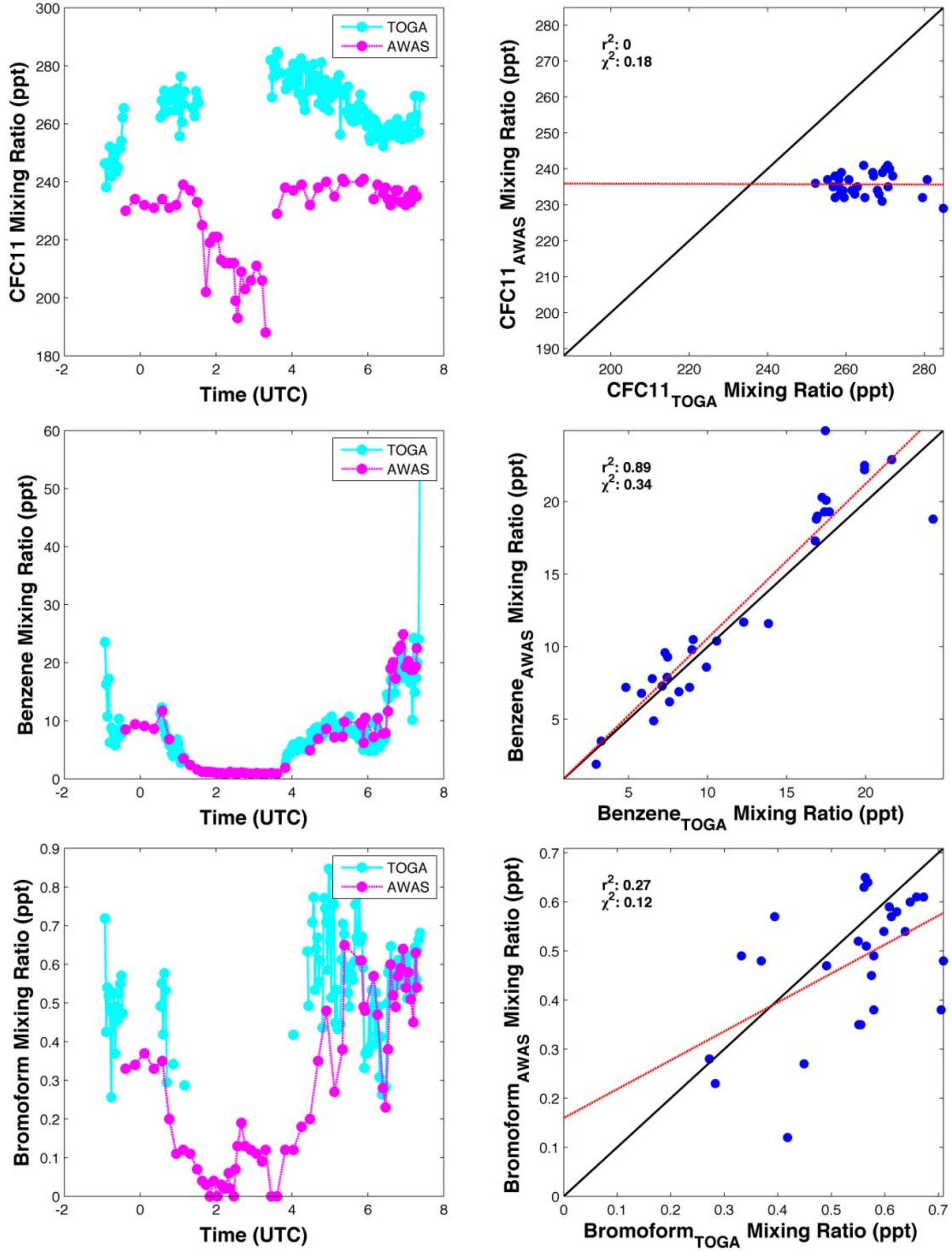
Departures from background evident in both O_3 and water vapor.

ACE-FTS Observations in Jan-Feb between 25N and 25S from 2005-2010





RF15



Uncertainties

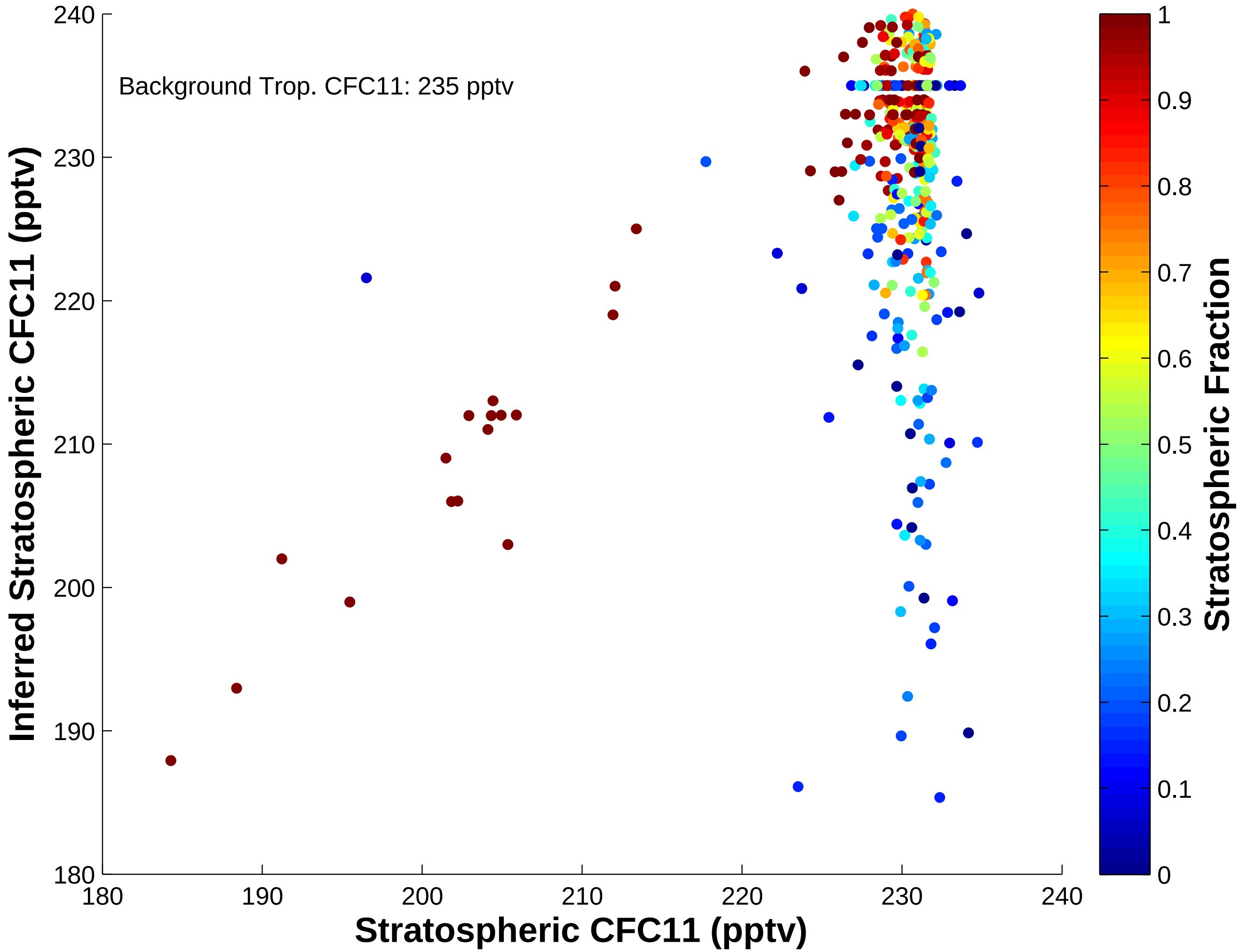
AWAS: 2-20%

TOGA:

CFC114 – 30% or 10

C6H6 – 15% or 2

CHBr3 – 30% or 0.4

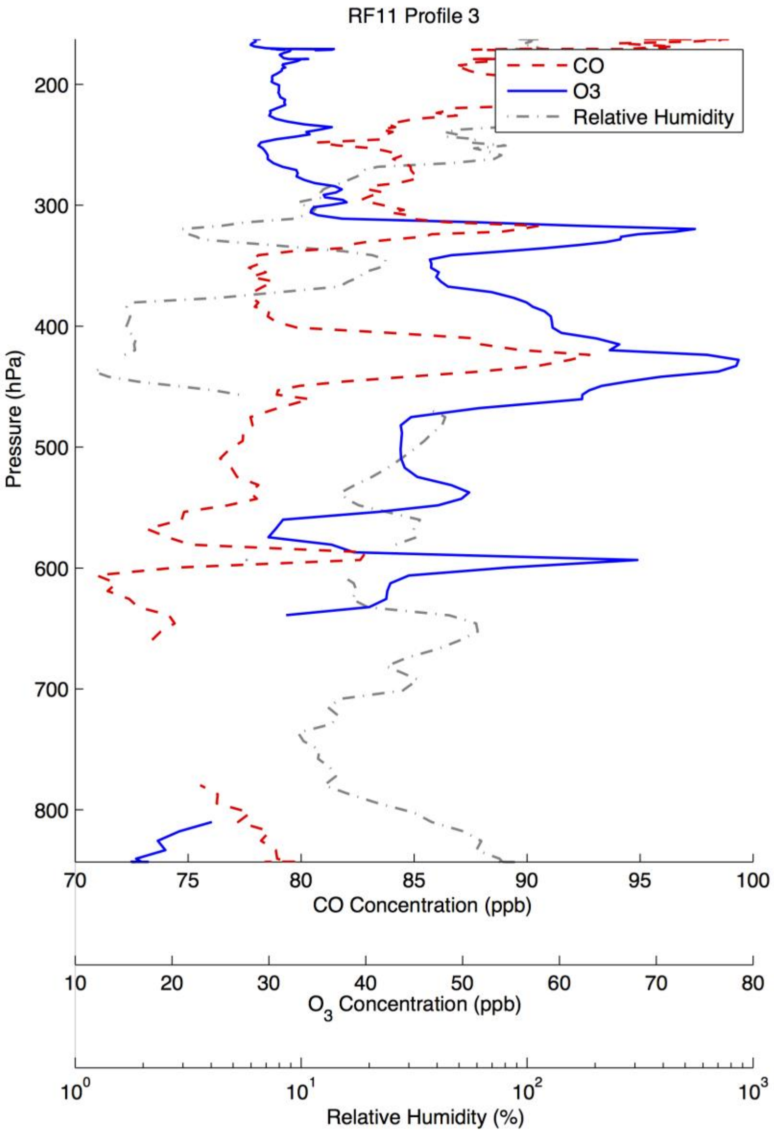


$C_f = f_{strat} * C_{strat} + f_{trop} * C_{trop}$

-Calculate stratospheric O_3 assuming C_{trop} = 20 ppbv & f_{strat} from water vapor calculation.

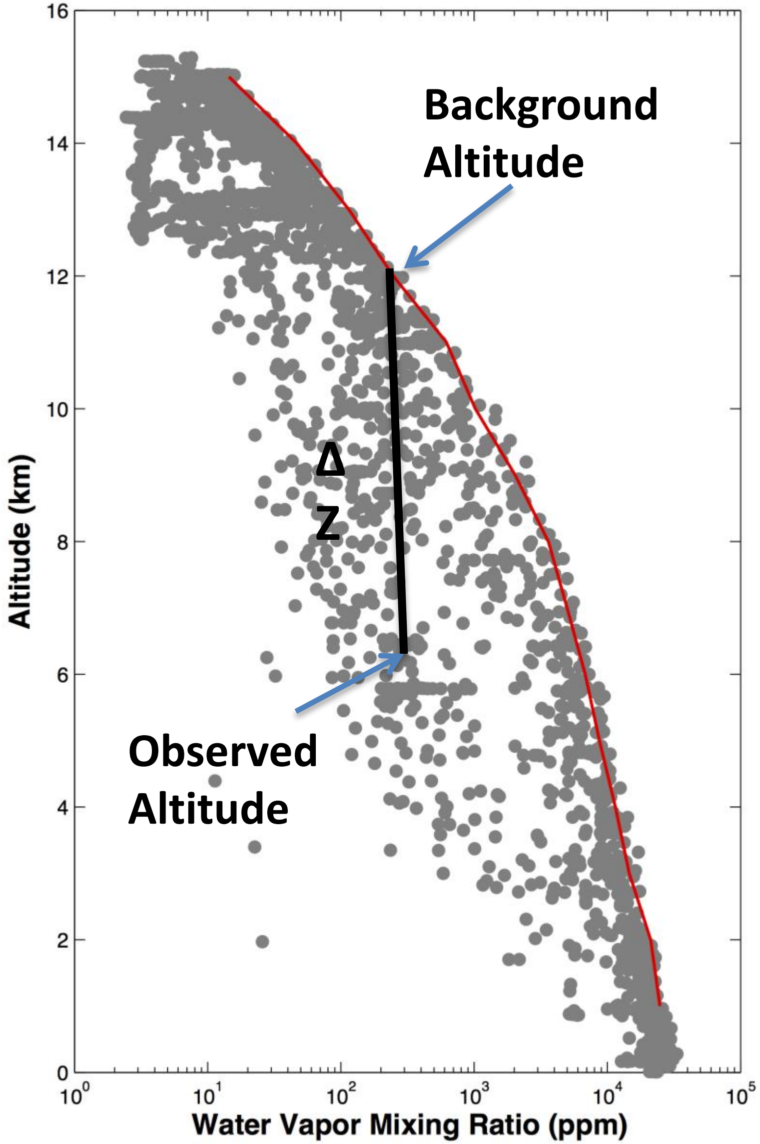
- Calculate stratospheric CFC11 from equation: $[CFC11] = -0.056[O_3] + 233$

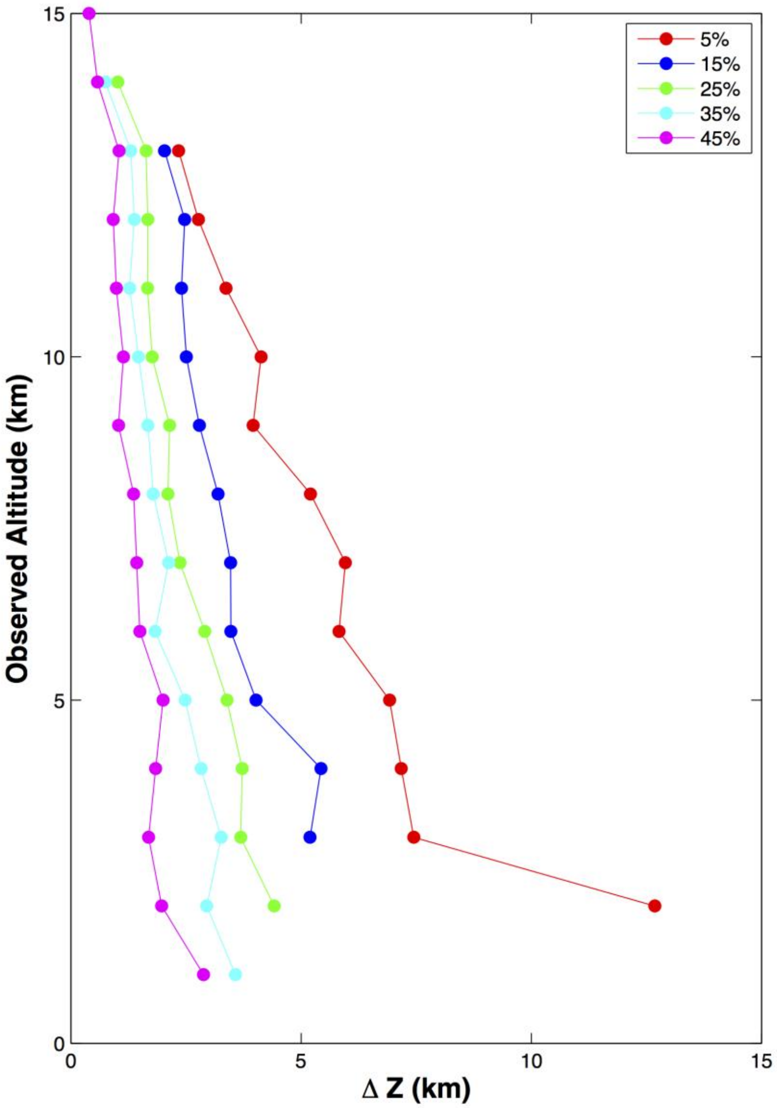
Remove pts w/ O3 less than 25



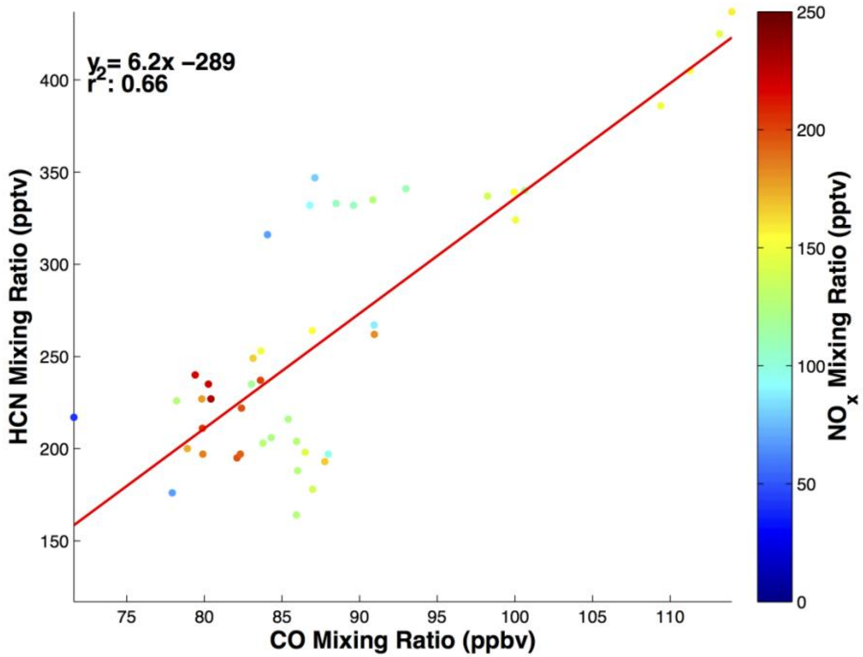
RF11 Flight Track

O_3 and CO correlated between 600 and 300 hPa; anticorrelated with relative humidity.

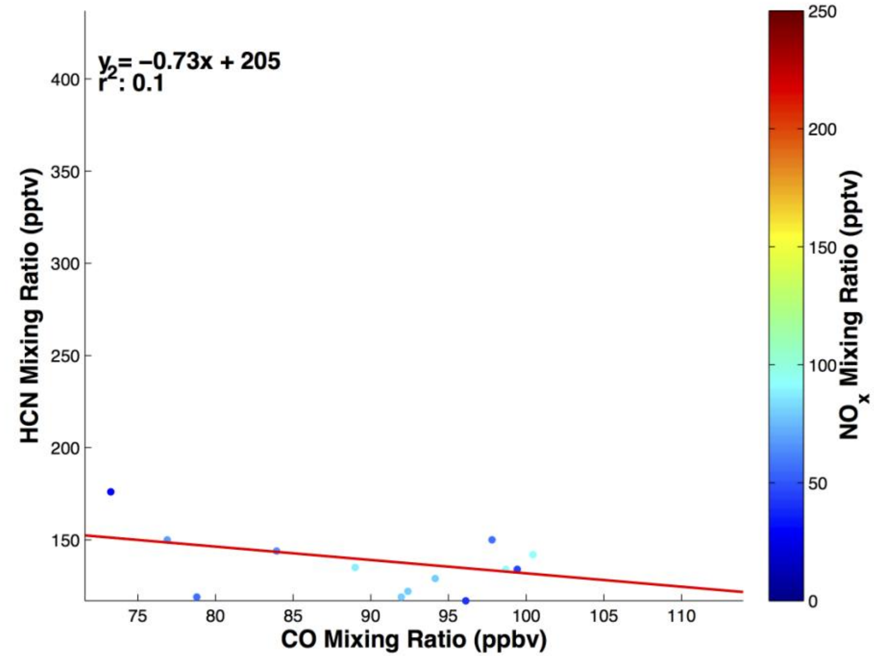

Background Altitude
Observed Altitude
Δ Z
Altitude (km)
Water Vapor Mixing Ratio (ppm)
0
2
4
6
8
10
12
14
16
10^0
10^1
10^2
10^3
10^4
10^5


5%
15%
25%
35%
45%
Observed Altitude (km)
Δ Z (km)
0
5
10
15

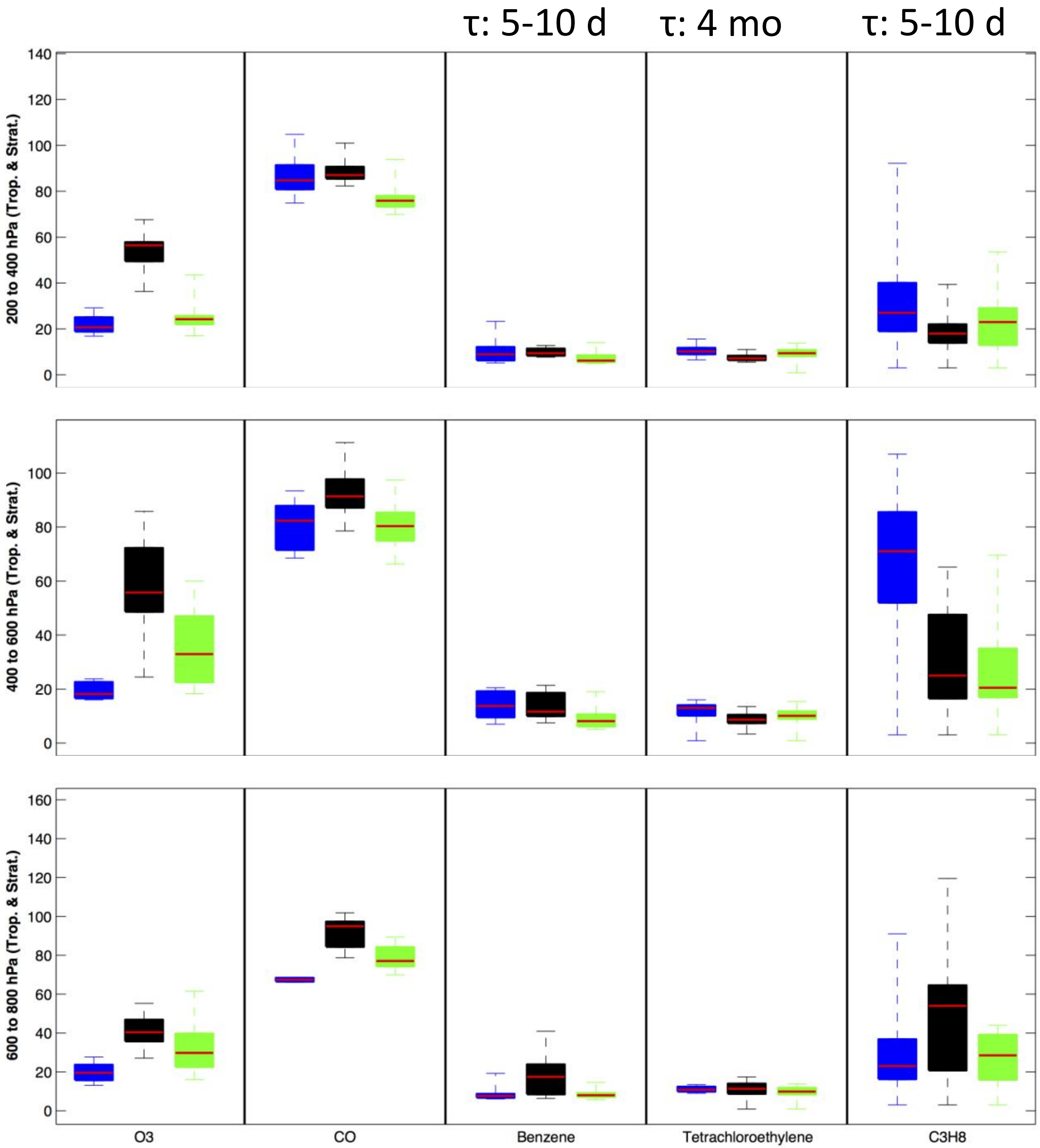
O_3 > 40 ppbv



O_3 < 25 ppbv



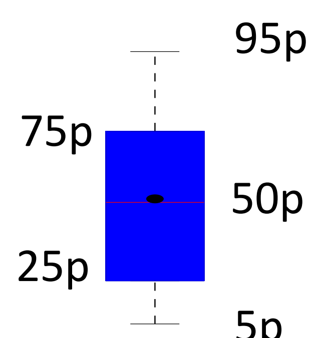
- Elevated O_3 air parcels correlate well with biomass burning tracers.
- Parcels depleted in O_3 correlate with fossil fuel/industrial emissions.



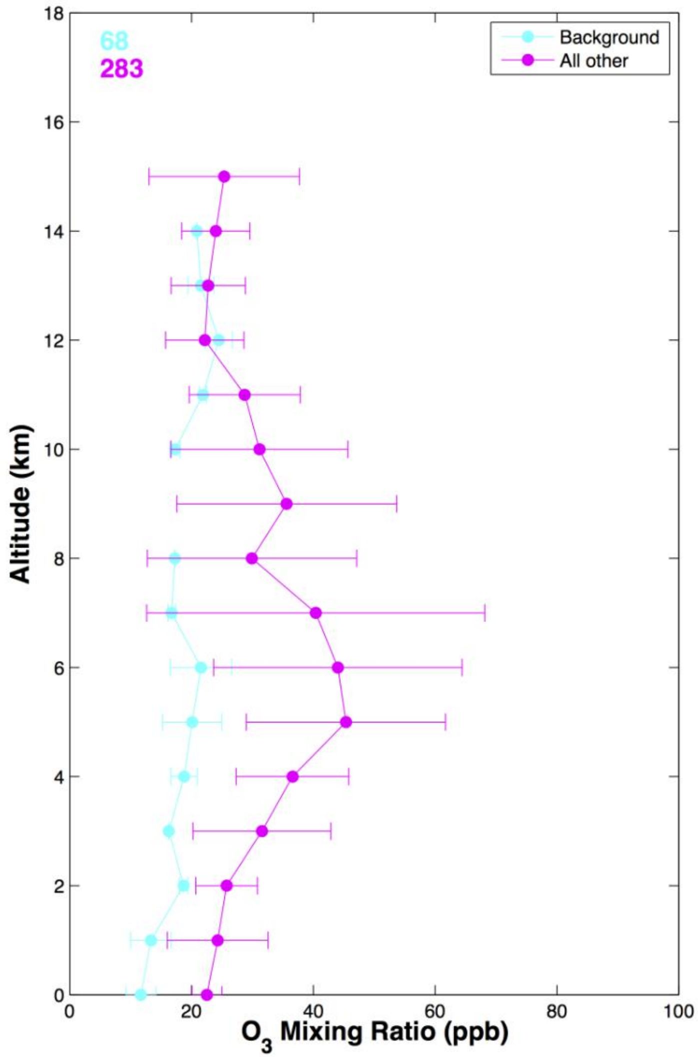
Southern Hemisphere
Southeast Asia/China
Eastern Pacific

For southeast Asian air masses:

- Little evidence of significant influence from fossil fuel combustion & industrial emissions.

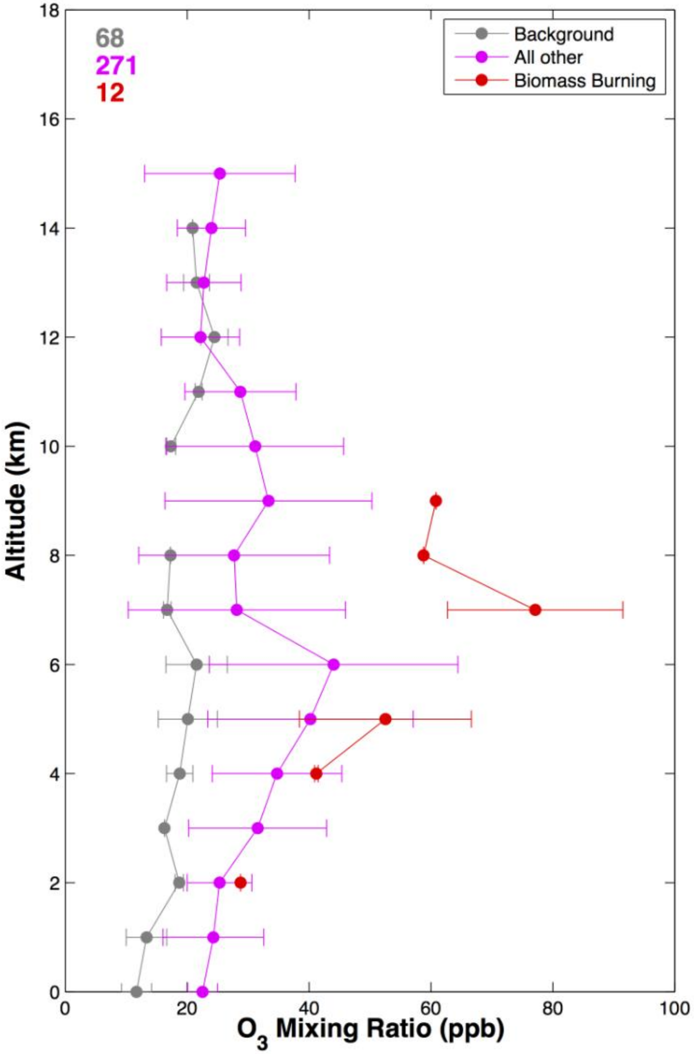


C_2Cl_4 values have been multiplied by 10 to fit on the axis.



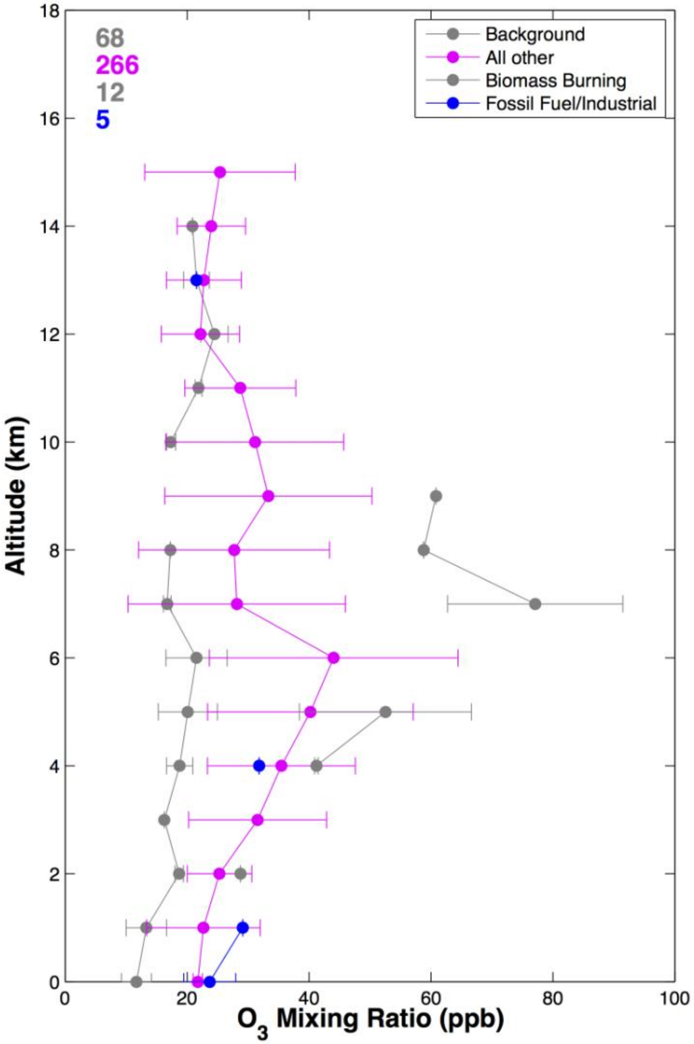
- Background: RH>45%, PV<0.5, & CO<90 ppbv

Data on AWAS time; Flights 5-16; abs(Latitude)<25



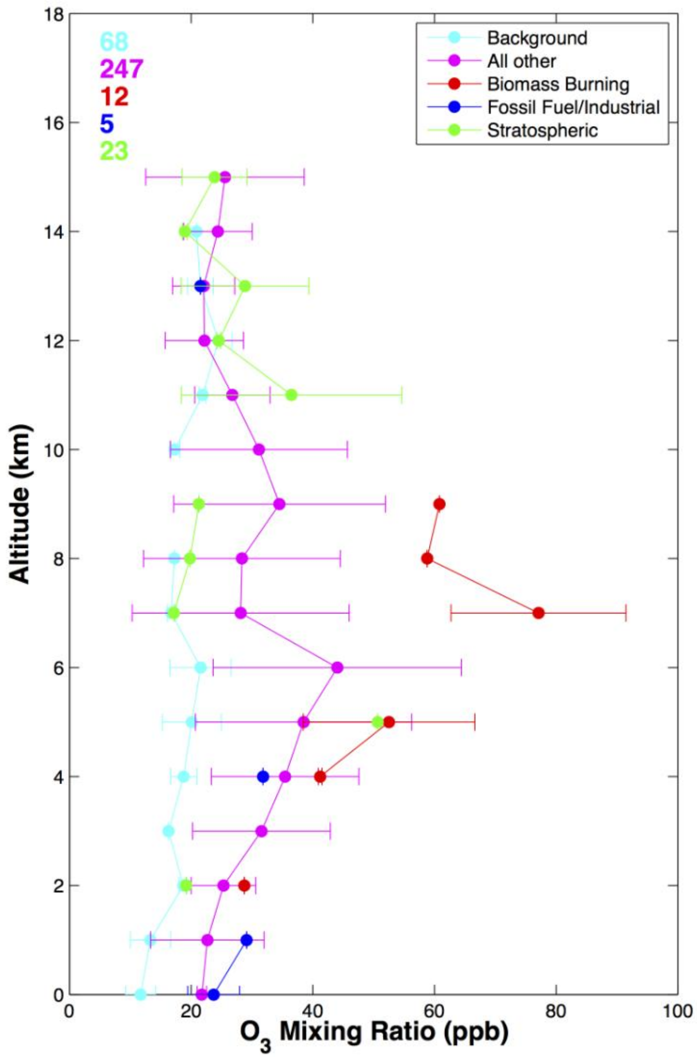
- Background: RH>45%, PV<0.5, & CO<90 ppbv
- BB: CO>90 ppbv, HCN>200 pptv, CH_3CN>100 pptv, PV<0.5

Data on AWAS time; Flights 5-16; abs(Latitude)<25



- Background: RH>45%, PV<0.5, & CO<90 ppbv
- BB: CO>90 ppbv, HCN>200 pptv, CH_3CN>100 pptv, PV<0.5
- FFI: CO>90 ppbv, HCN<200 pptv, C_2Cl_4> 1.75 pptv, C_3H_8>100 pptv, PV<0.5

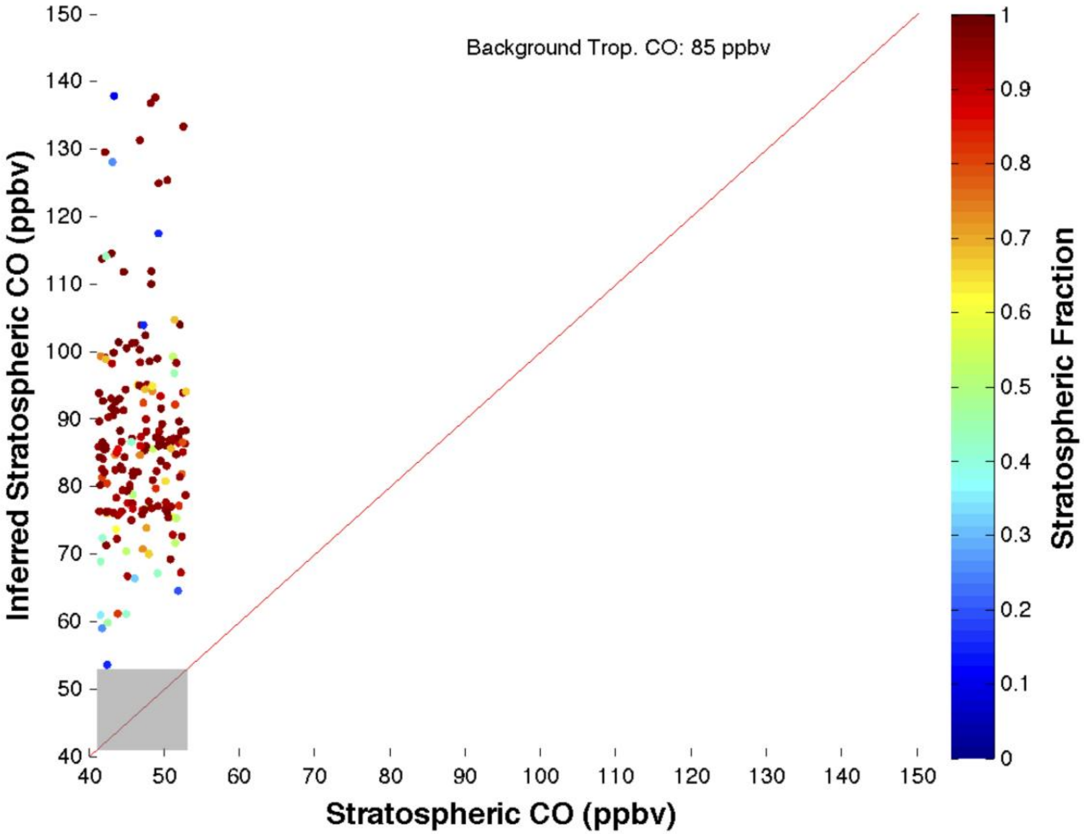
Data on AWAS time; Flights 5-16; abs(Latitude)<25



- Background: RH>45%, PV<0.5, & CO<90 ppbv
- BB: CO>90 ppbv, HCN>200 pptv, CH_3CN>100 pptv, PV<0.5
- FFI: CO>90 ppbv, HCN<200 pptv, C_2Cl_4> 1.75 pptv, C_3H_8>100 pptv, PV<0.5
- Strat: CFC11<230 pptv, HCFC141b<23

Data on AWAS time; Flights 5-16; abs(Latitude)<25

$$CO_{Strat\ inferred} = \frac{CO_{Obs} - (1 - f_{Strat}) * CO_{Trop}}{f_{Strat}}$$



Calculated f_{strat} from water vapor requires stratospheric CO concentrations that are significantly higher than observations.