Convective transport of chemical compounds from the surface to the UT/LS: The western Pacific vs. North America

Qing Liang[1]

&

Thomas Hanisco[2], **Steve Wofsy**[3], **Jasna Pittman**[3], **Elliot Atlas**[4], **Maria Navarro**[4], **Sue Schauffler**[5], **Donald R. Blake**[6], **Teresa Campos**[5], **Frank Flocke**[5]

[1] NASA Goddard Space Flight Center/ USRA GESTAR
[2] NASA Goddard Space Flight Center
[3] Harvard University
[4] University of Miami
[5] NCAR Atmospheric Chemistry Observations and Modeling Laboratory
[6] University of California, Irvine

Motivation and Goals

Surface to the UT/LS transport of trace gases can exert significant impacts on atmospheric abundances of ozone (O_3) and hydroxyl radical (OH), which will have chemical and radiative impacts on the atmosphere.

- Vertical transport of CO and NMHCs
- Vertical transport of very-short-lived (VSL) halogen compounds, i.e. $CHBr_3$ and CH_2Br_2.

Questions of interest

- Tropical and subtropical convective transport:
  - Frequency
  - The timescale it takes to ascent into the UT/LS
  - The regional differences in convective lofting: Western Pacific vs. North America
- What are transport efficiency of chemical compounds (the impact of tracer lifetime and solubility)?

What are our best choice of chemical tracers?

-- CO? VOCs and their ratios? Tracers of marine origin? CO_2, SF_6 …

NASA GEOS-5 Chemistry Climate Model (GEOSCCM)

- V2 - GEOS-5 with standard stratospheric chemistry and detailed VSL bromocarbons
- GCM simulations for the ATTREX/CONTRAST and SEAC^{4}RS period
  - Horizontal resolution is 2° latitude x 2.5° longitude
  - 72 vertical layers from surface to 0.01 hPa, with ~1 km/layer resolution at the UT/LS region

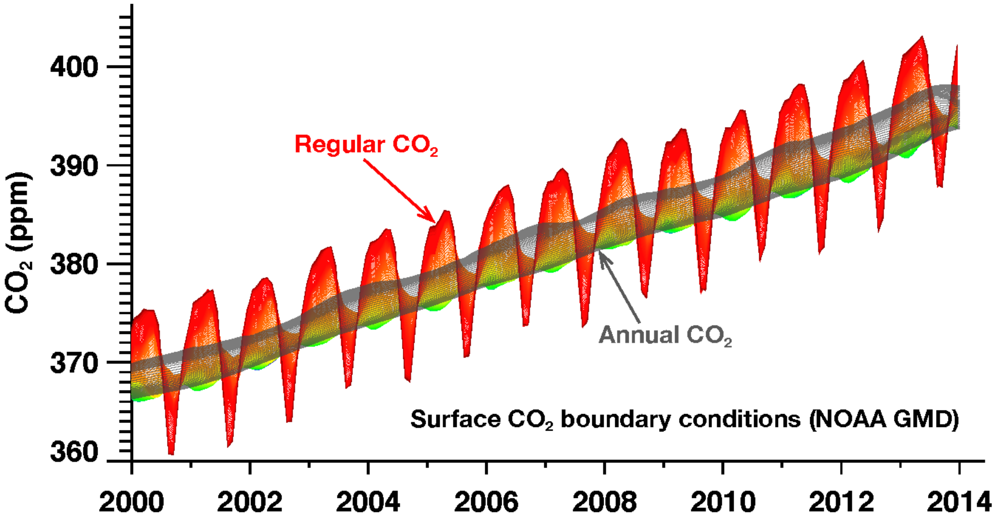
(will conduct a future simulation with MERRA meteorology further comparison)

Two new CO_2 tracers

– transport clock as well as model transport diagnostics

1) Regular CO_2 - with surface boundary conditions from the NOAA observations (latitude and monthly variable)

2) Annual CO_2 – same as regular CO_2 but with seasonal cycle removed at all latitude grids.

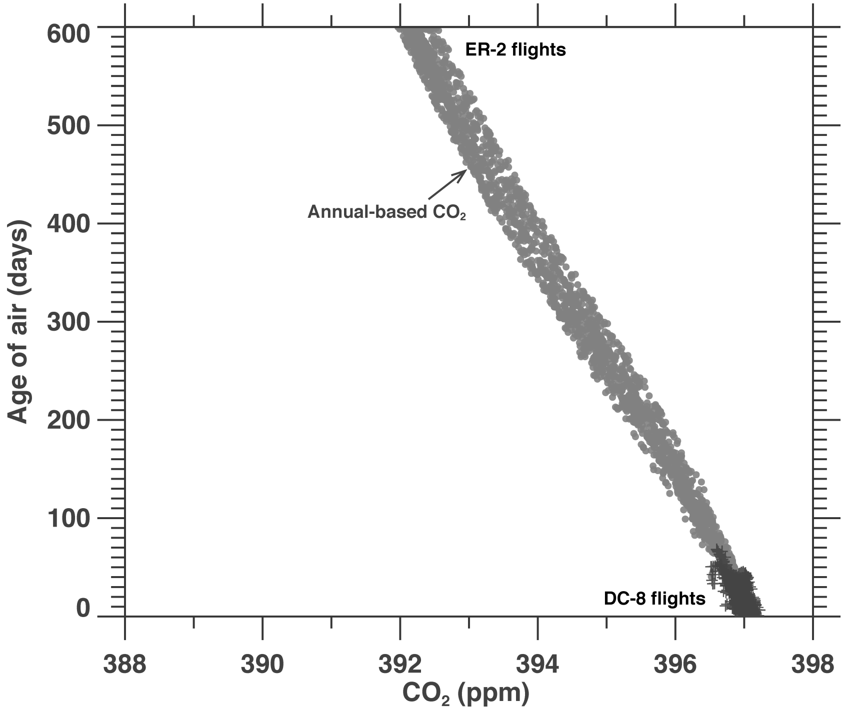


The model simulations are sampled at the same geographical and vertical location for the corresponding time as the observations for model-observation comparison.

CO_2 as a transport clock tracer

SEAC^{4}RS

(N. America, Aug-Sep 2013)



Model annual-based CO_2 shows a linear correlation with age of air, thus an accurate clock tracer for transport

CO_2 as a transport clock tracer

SEAC^{4}RS

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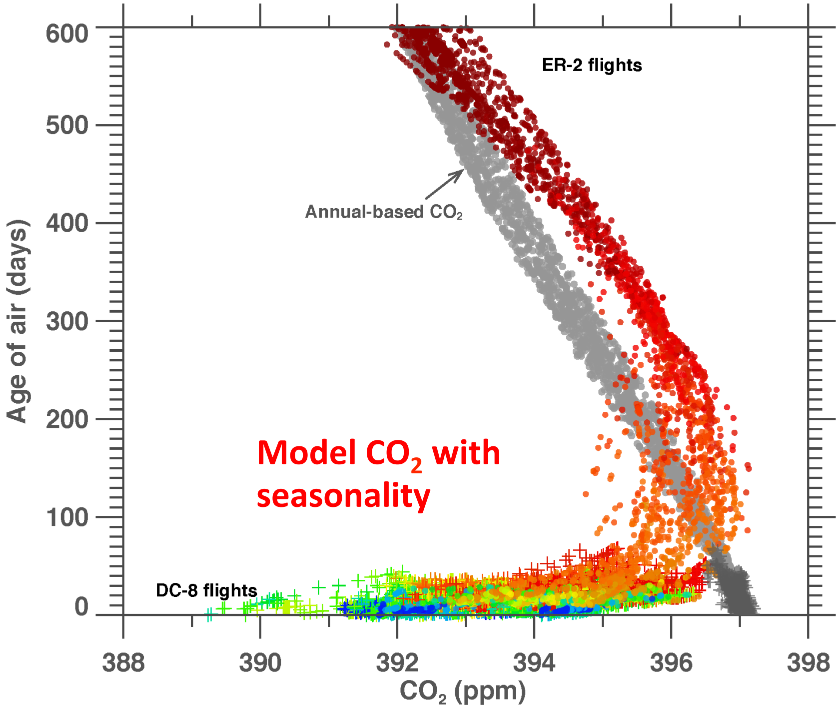
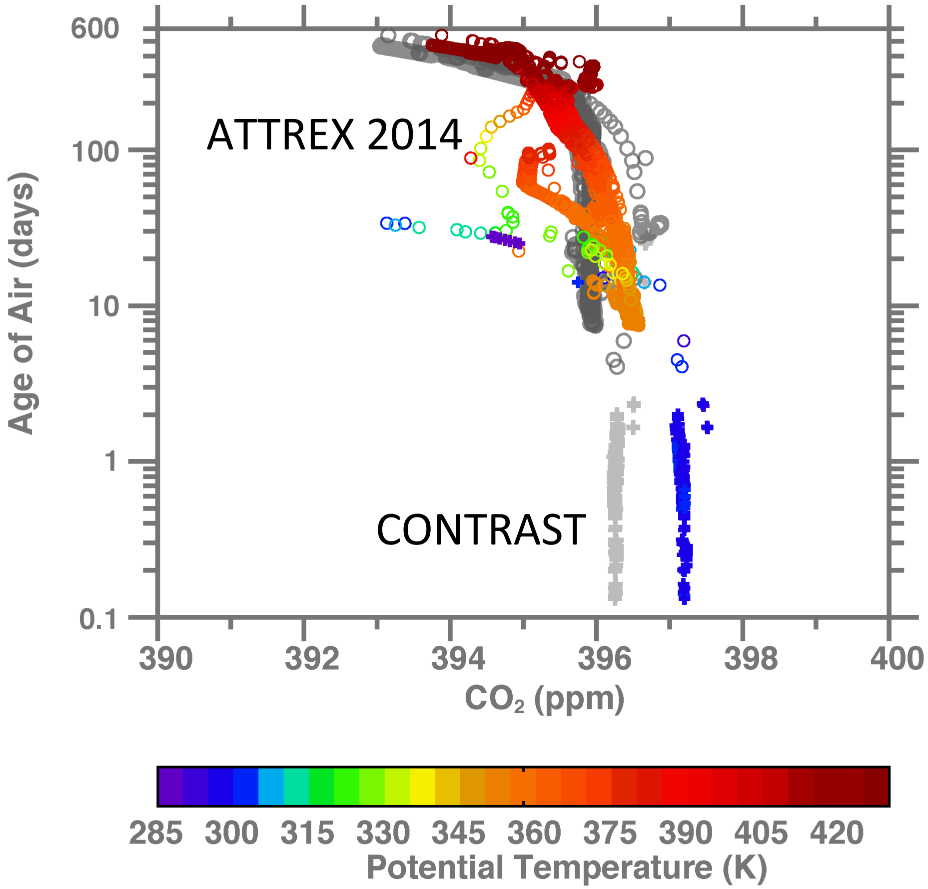


ATTREX-3/CONTRAST

(W. Pacific, Jan-Feb 2014)



However, the actual application as a clock tracer is complicated due to the surface seasonal variations in CO_2

Two-step approach on transport analysis

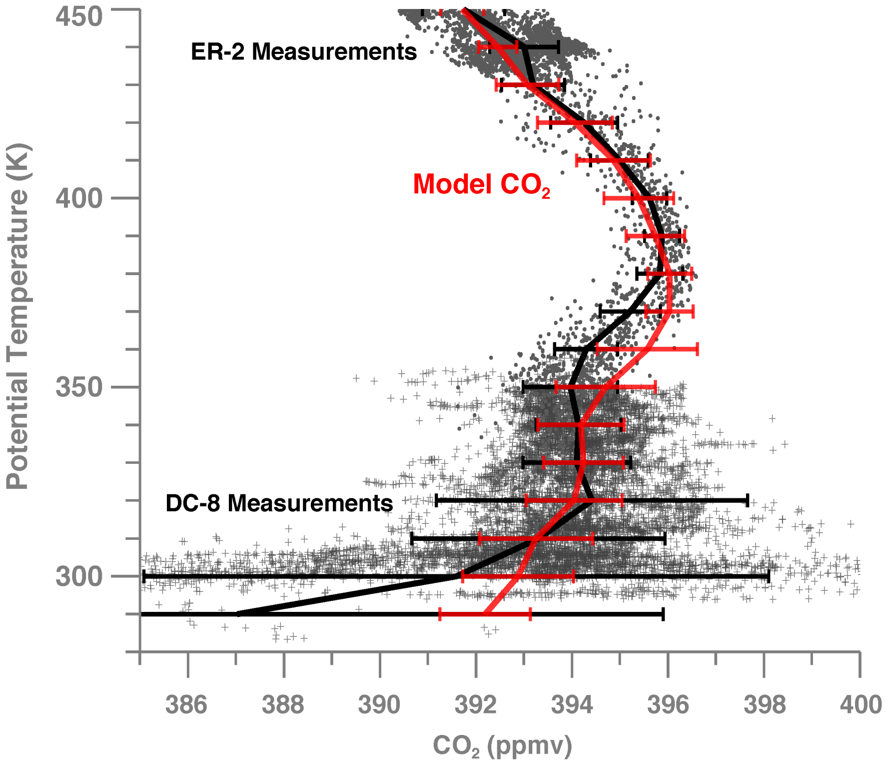
Our proposed 2-step approach:

Step 1. Use model CO_2 vs. observed CO_2 to evaluate model transport: convection and large scale ascent

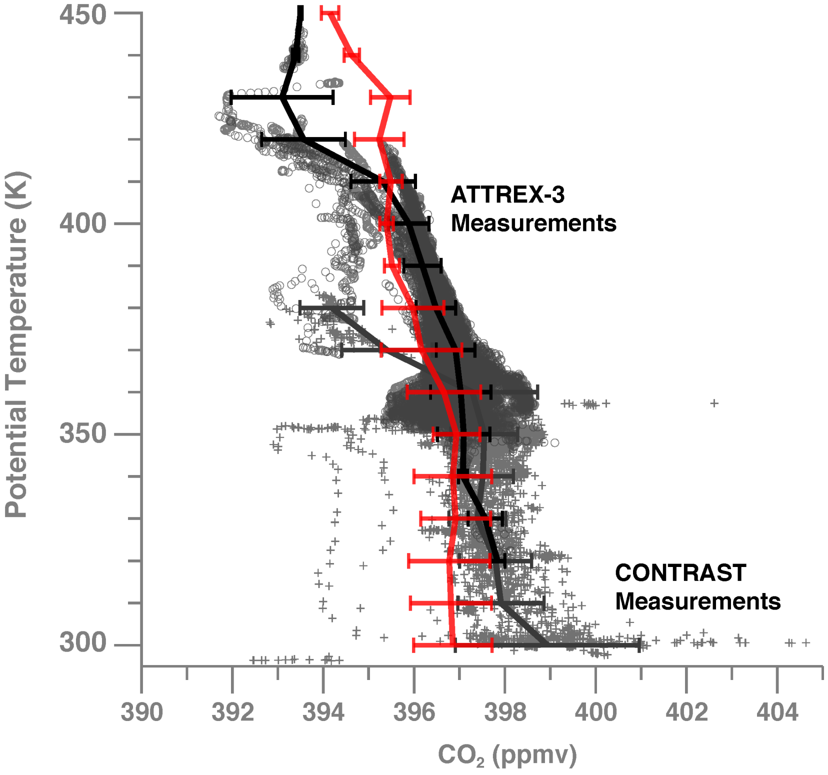
Step 2. Use the model age of air to quantify transport timescale

Step 1: Model transport evaluation using CO_2

SEAC^{4}RS
(N. America, Aug-Sep 2013)



ATTREX-3/CONTRAST
(W. Pacific, Jan-Feb 2014)

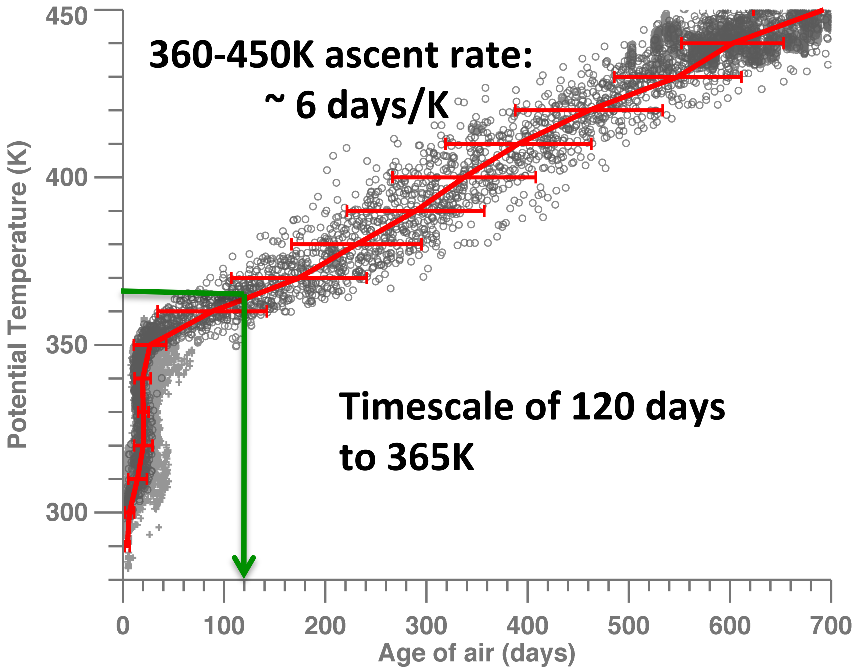


GEOSCCM modeled CO_2 agrees well with the SEAC^{4}RS and ATTREX-3/CONTRAST observations, indicating credible model transport in the vertical.

Step 2: Quantify vertical transport Timescale using model age of air

SEAC^{4}RS

(N. America, Aug-Sep 2013)

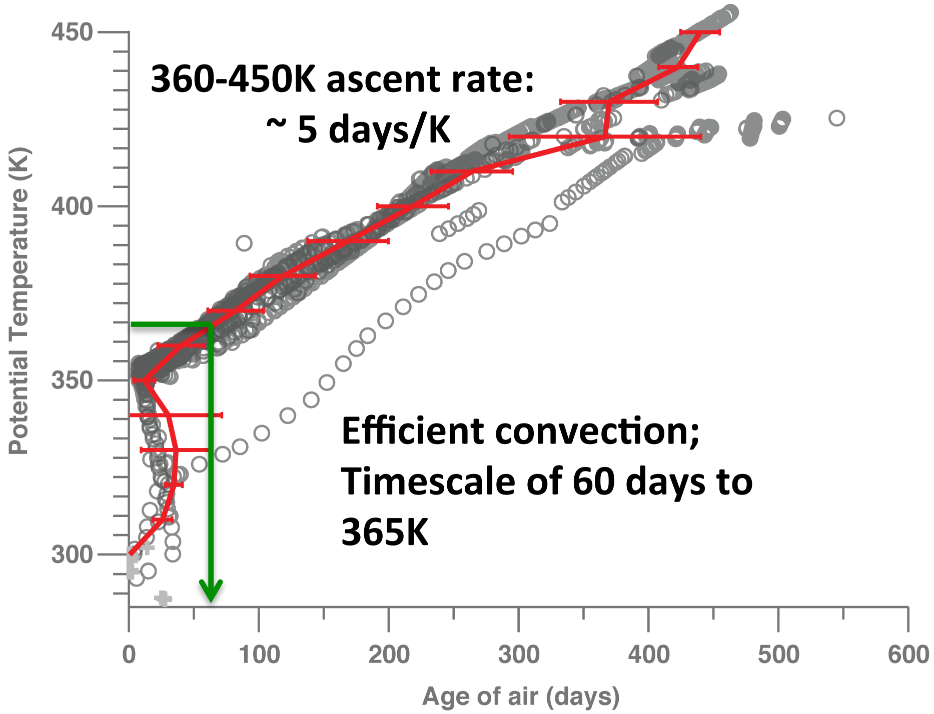


360-450K ascent rate:
~ 6 days/K

Timescale of 120 days
to 365K

ATTREX-3/CONTRAST

(W. Pacific, Jan-Feb 2014)

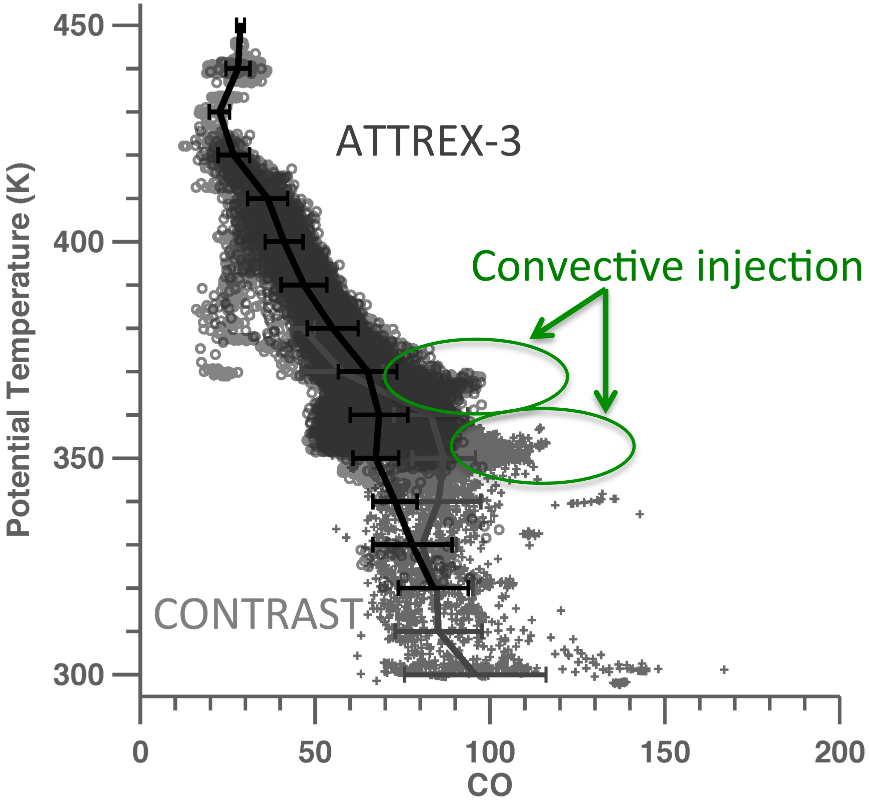


360-450K ascent rate:
~ 5 days/K

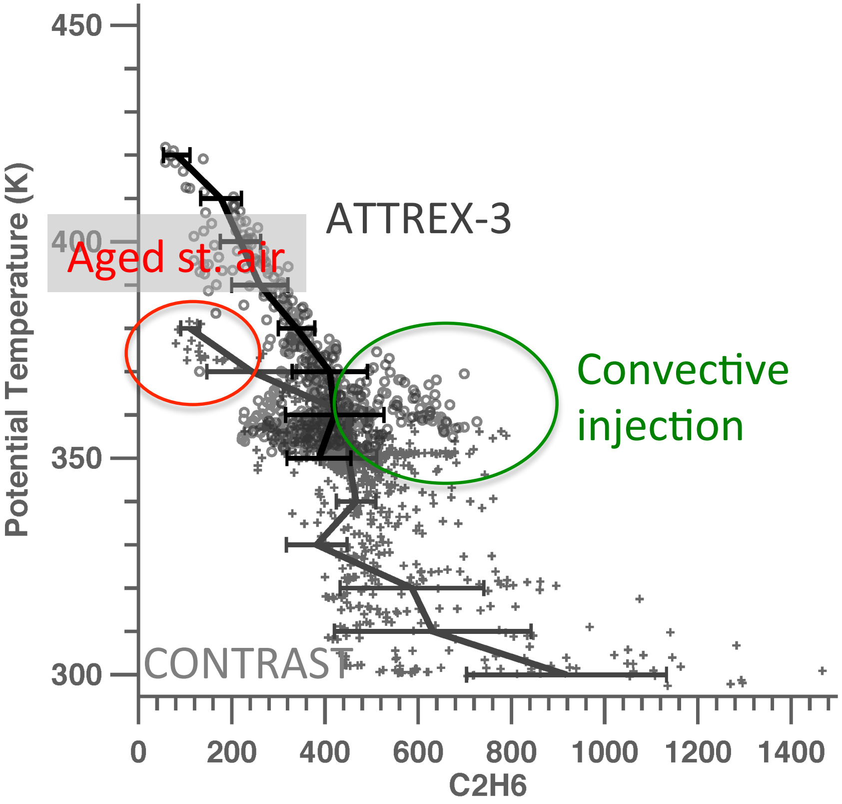
Efficient convection;
Timescale of 60 days to
365K

Surface to UT/LS transport of pollution tracers: ATTREX-3 and CONTRAST

CO



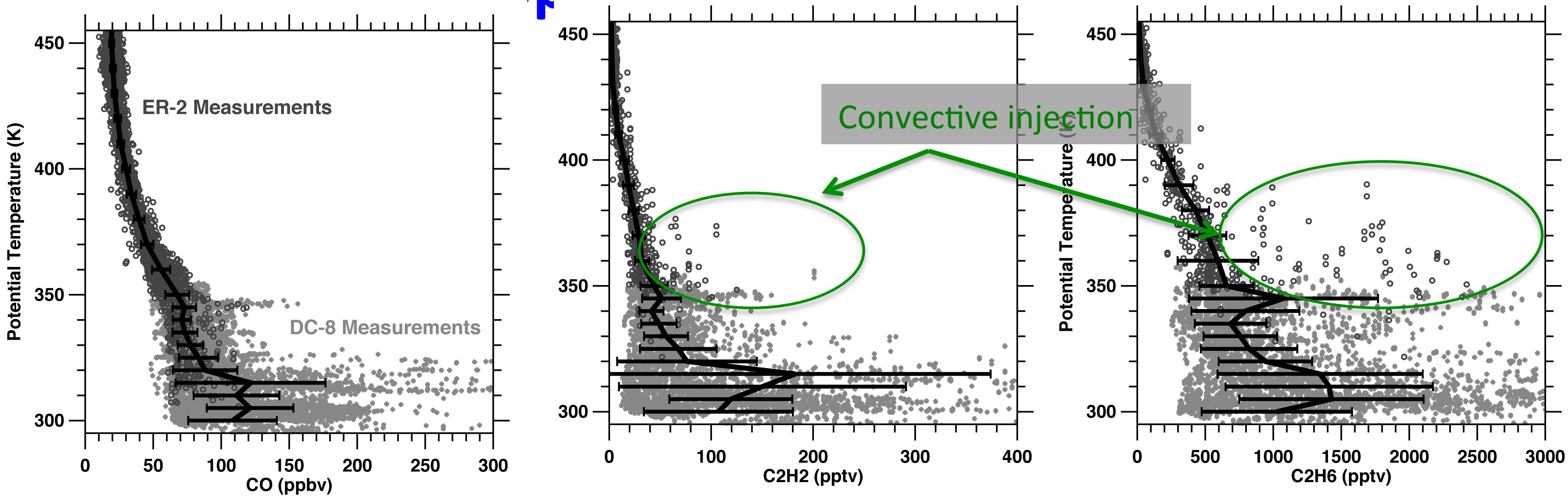
C_2H_6



For the deep convection active western Pacific, CO and C_2H_6 work well as convection tracers.

Surface to UT/LS transport of pollution tracers:

Subtropical N. America: SEAC^{4}RS



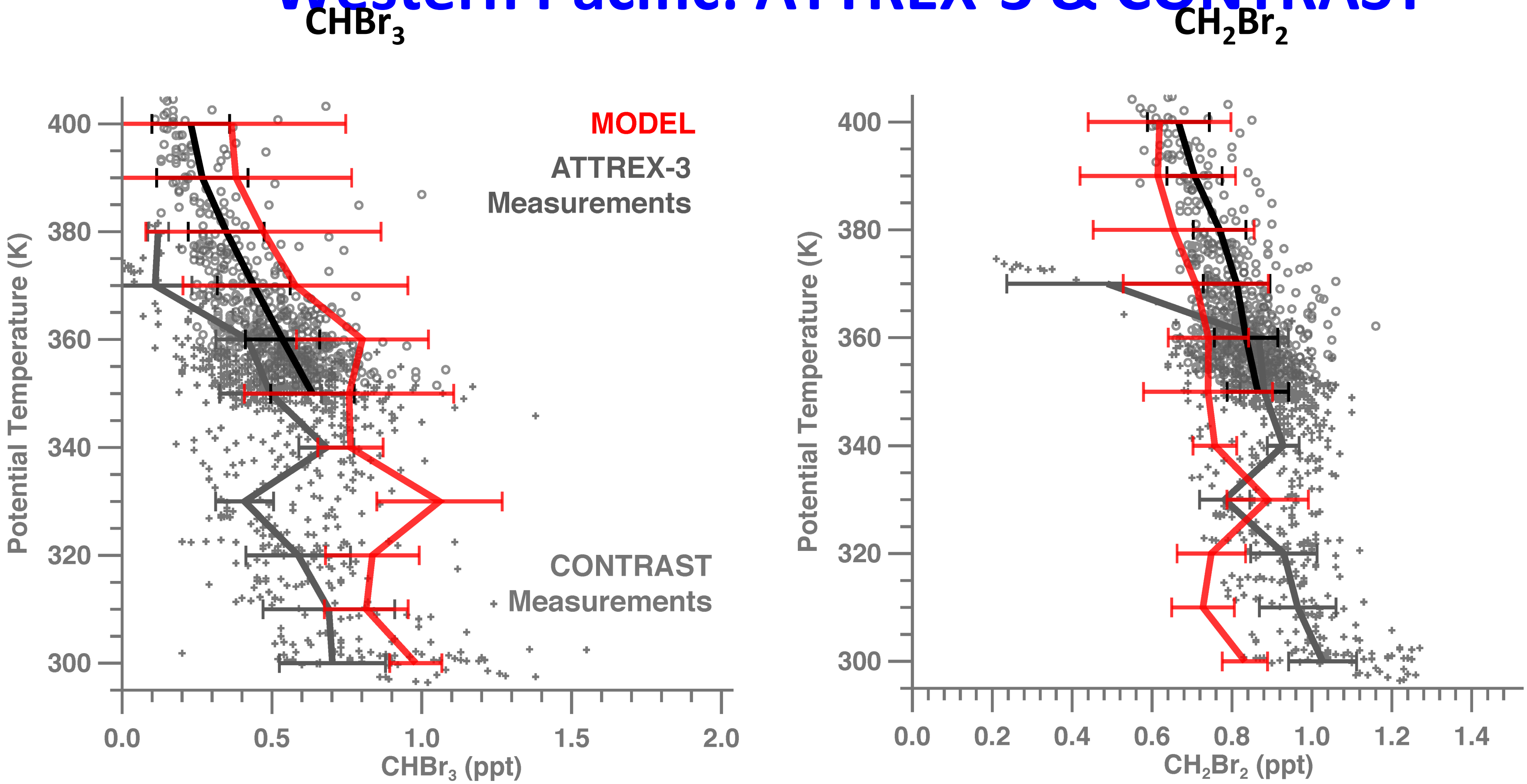
Tracer lifetime
At different
layers

	CO	C_2H_2	C_2H_6
Loss reactions	**CO + OH ->**	**C_2H_2 + OH ->** **C_2H_2 + O_3 ->**	**C_2H_6 + OH -> C_2H_5 --> CH_3CHO --> PAN** **C_2H_6 + Cl -> C_2H_5 --> CH_3CHO --> PAN**
400-450K	**~ 2 years**	**~ 6 months**	**~ 4 months**
350-400K	**~ 1 year**	**~ 10 months**	**~ 10 months**
Troposphere	**~ 1 month**	**~ 0.5 month**	**~ 3 months**

Efficient C_2H_6 loss via the C_2H_6 + Cl reaction creates a large vertical as well as tropical vs. high-latitude gradients in C_2H_6 -> making C_2H_6 an excellent transport tracer for tropical convective lofting.

Surface to UT/LS transport of $CHBr_3$ and CH_2Br_2

Western Pacific: ATTREX-3 & CONTRAST

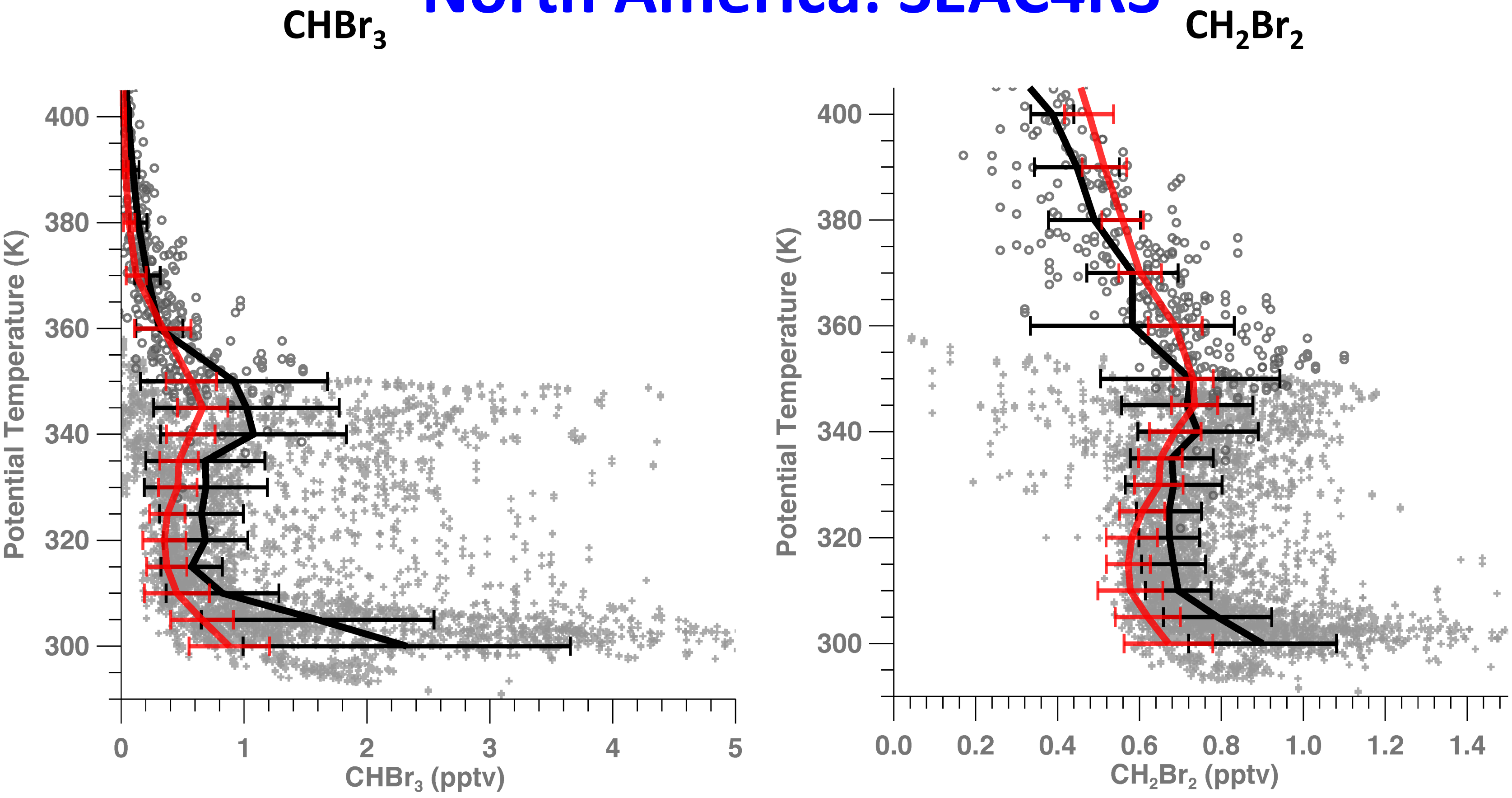


Model $CHBr_3$ are biased high wrt observations, but model CH_2Br_2 are biased low.

This is consistent with findings from the SHEVA measurements (Hossaini et al. 2013) (GEOSCCM emissions are fine, slightly high-based). However, why?

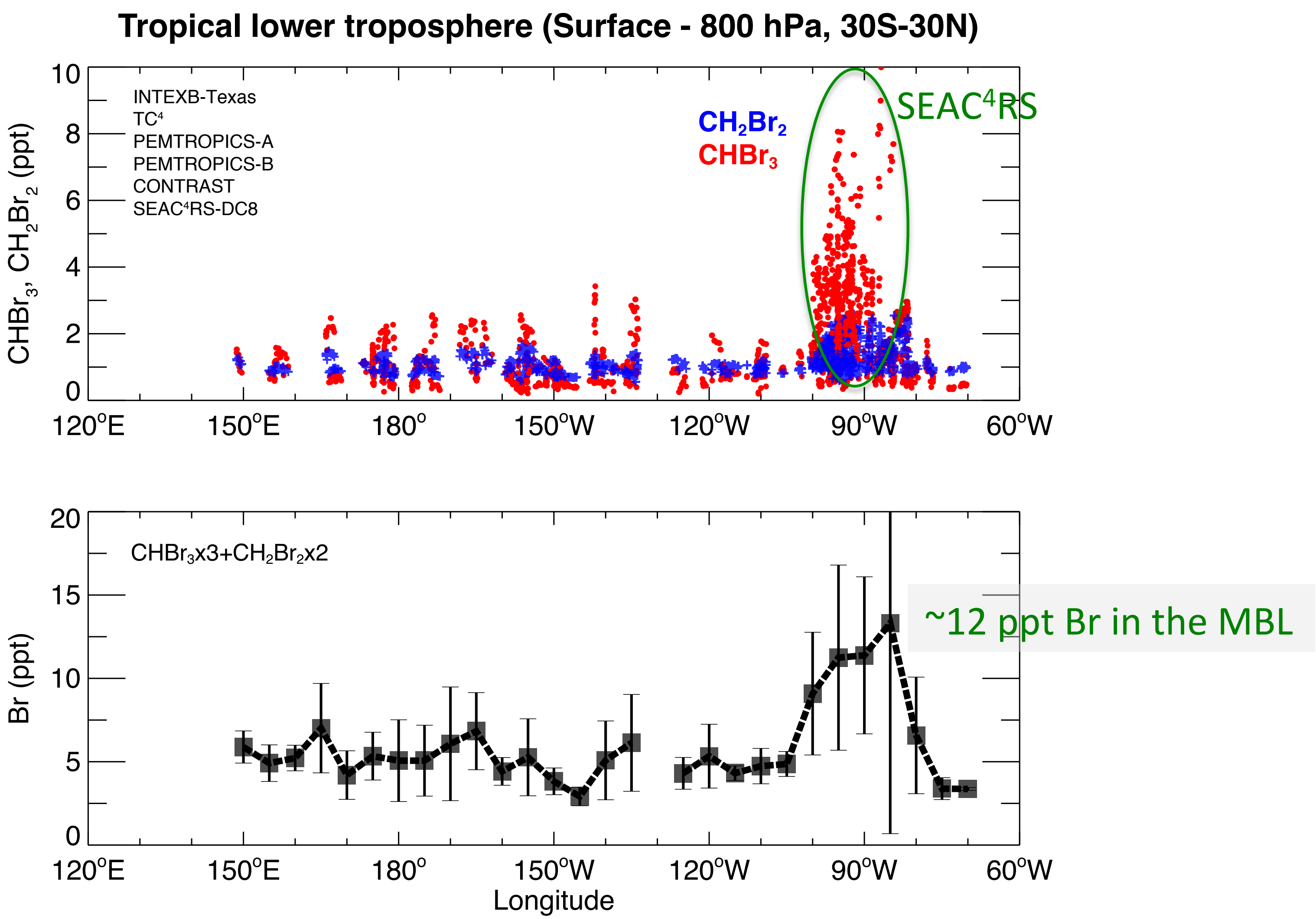
Surface to UT/LS transport of $CHBr_3$ and CH_2Br_2

North America: SEAC4RS



Tropospheric low biases -> Surface emissions of $CHBr_3$ and CH_2Br_2 are too small. This contradicts the findings from CONTRAST and Hossaini et al. (2013).

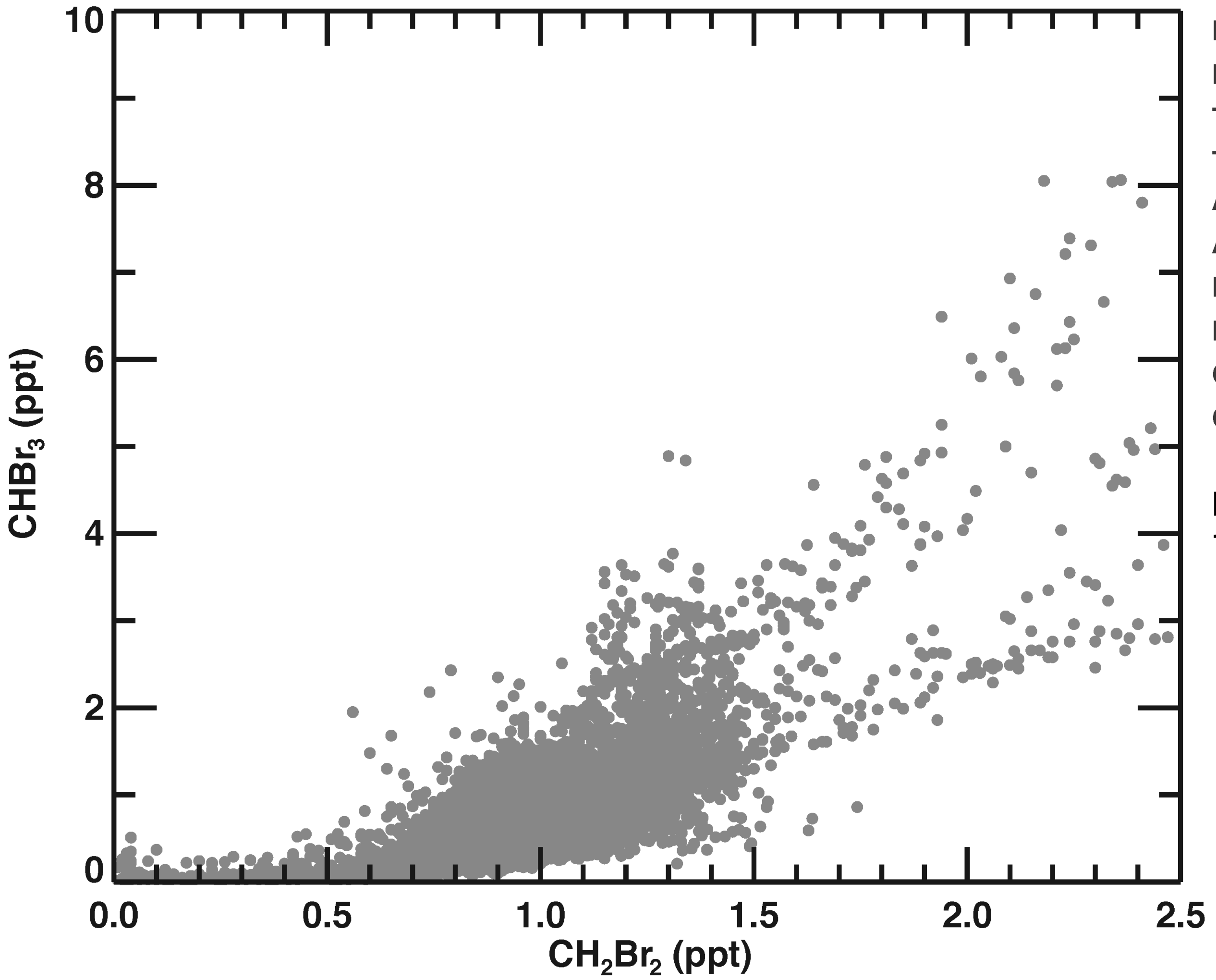
High levels of $CHBr_3$ and CH_2Br_2 during SEAC⁴RS


Tropical lower troposphere (Surface - 800 hPa, 30S-30N)

SEAC⁴RS

~12 ppt Br in the MBL

High $CHBr_3/CH_2Br_2$ ratios during SEAC⁴RS

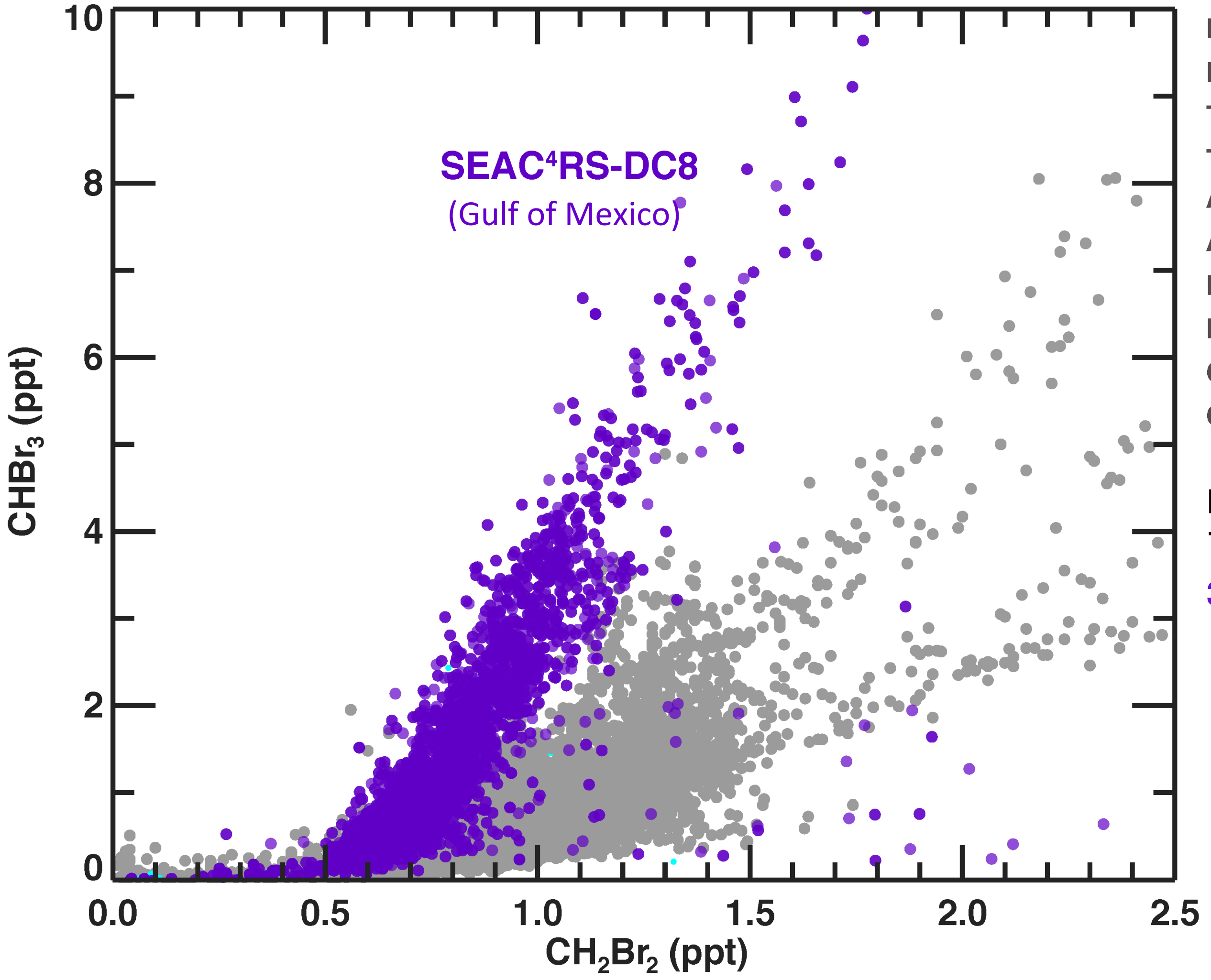


- INTEXB1
- INTEXB2
- TC4
- TRACEP
- ARCTASA
- ARCTASB
- PEMTROPICSA
- PEMTROPICSB
- CONTRAST-AWAS
- CONTRAST-TOGA

Pacific campaigns

Emission ratio:
1.3 ppt $CHBr_3$/1 ppt CH_2Br_2

High $CHBr_3/CH_2Br_2$ ratios during SEAC^{4}RS



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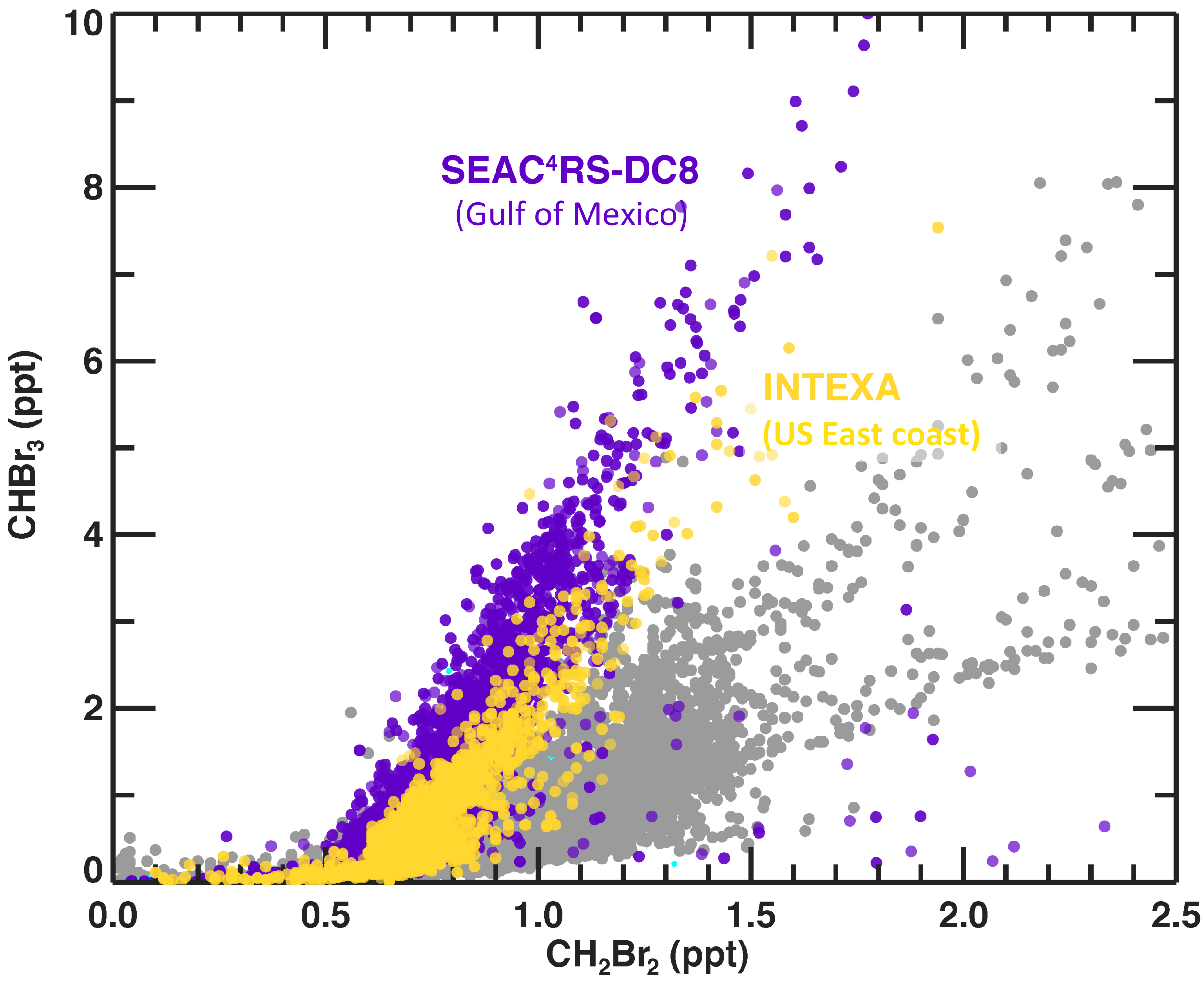
Pacific campaigns

Emission ratios:
1.3 ppt $CHBr_3$/1 ppt CH_2Br_2
3.0 ppt $CHBr_3$/1 ppt CH_2Br_2

High $CHBr_3/CH_2Br_2$ ratios during SEAC^{4}RS



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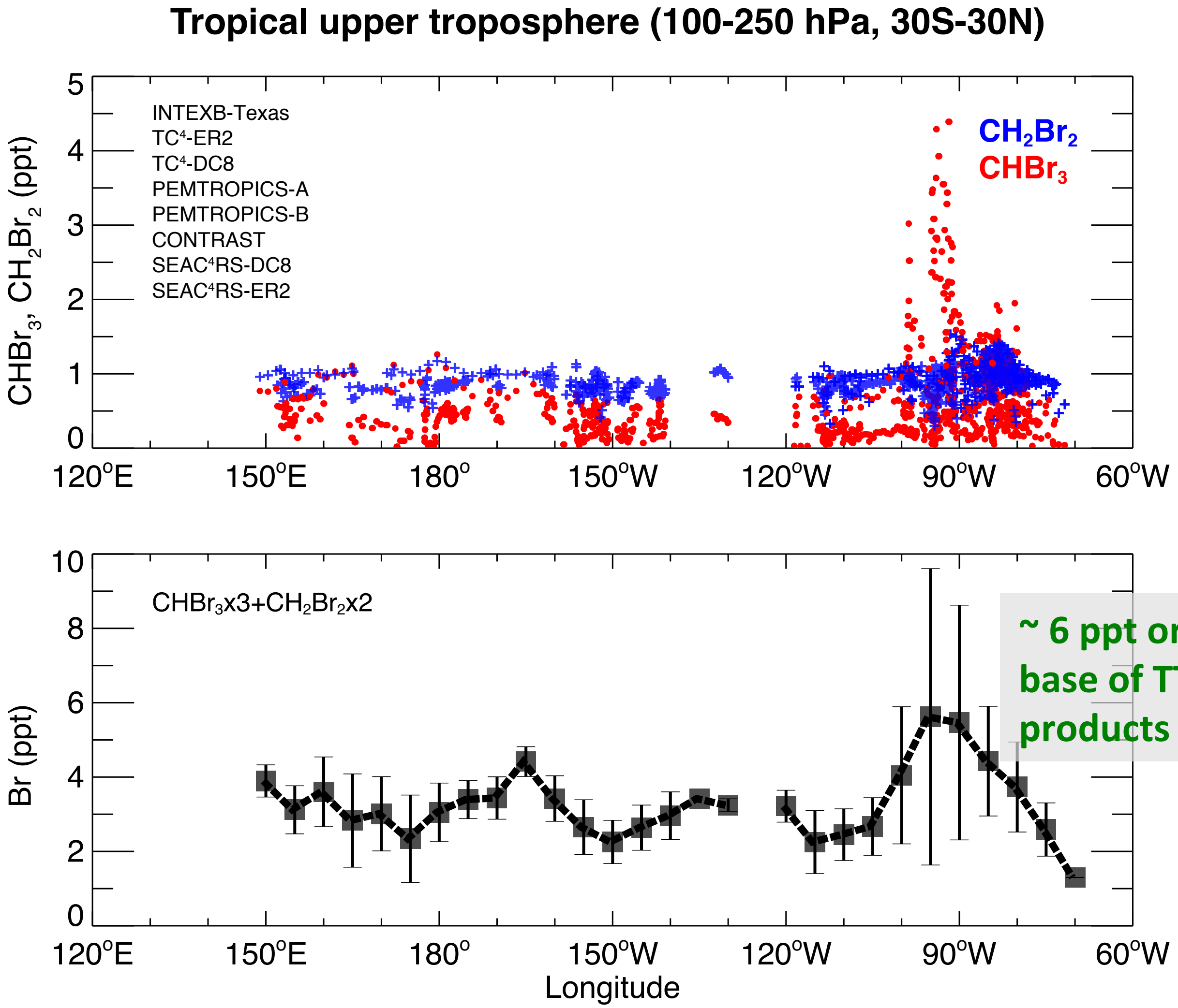
Pacific campaigns

Emission ratios:
1.3 ppt $CHBr_3$/1 ppt CH_2Br_2
3.0 ppt $CHBr_3$/1 ppt CH_2Br_2
2.0 ppt $CHBr_3$/1 ppt CH_2Br_2

The Atlantic Ocean features much higher biogenic bromocarbon emissions, in particular $CHBr_3$, than the Pacific Ocean

VSL bromine at the North American tropical UT


Tropical upper troposphere (100-250 hPa, 30S-30N)

~ 6 ppt organic Br at the base of TTL + inorganic products (ppt??)

Conclusions and future work

- An optimal approach is to use a combination of chemical tracers to examine UT/LS transport

  CO_2 and **Age of Air**:

  i) Rapid convective ventilation below 355K topped by slow ascent from 355K to 450K, ~5-6 days/K

  ii) Western Pacific is convectively efficient: 60 days from surface to 365K

  iii) The North Ameircan lower stratosphere is rather aged: 120 days from surface to 365K

  CO and **C_2H_6**:

  i) Active convective transport from surface to UT/LS in the western Pacific (350-360K)

  ii) Convective transport in continental N. America is sporadic, but can have injection as high as 400K

- The abundance of very-short-lived bromocarbons
  - About 5 ppt Br from $CHBr_3$ and CH_2Br_2 in the Western Pacific
  - The Golf of Mexico features very high $CHBr_3$ and CH_2Br_2 , ~ 12 ppt Br
  - The Golf of Mexico (and the Atlantic Ocean) features very high $CHBr_3/CH_2Br_2$ ratios, implying a need to revisit the $CHBr_3$ and CH_2Br_2 emissions inventory

Future work

- Extend the data analysis to more chemical tracers and additional campaigns (different seasons and different regions)