OBSERVATIONS (& MODELING) OF GAS-PHASE ORGANIC SPECIES IN THE ASM

TOGA-TOF VOCs on the NSF NCAR GV during ACCLIP

Eric Apel (PI), Rebecca Hornbrook, Alan Hills, Barbara Barletta (UC Irvine), Behrooz Roozitalab, Daun Jeong, Doug Kinnison, Louisa Emmons
(NSF NCAR/ACOM)

April 30, 2024

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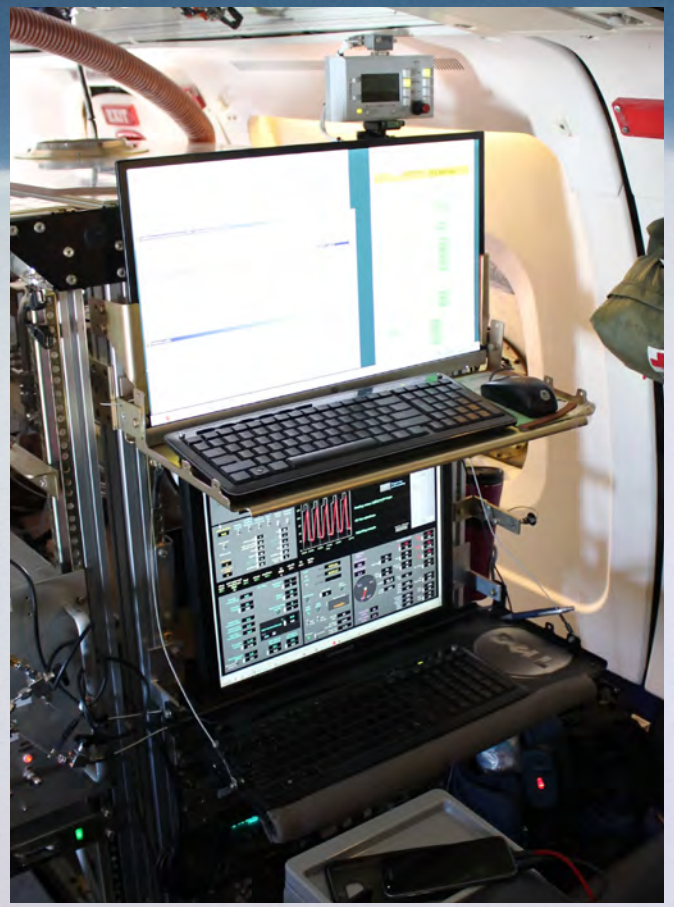
Measurements: TOGA-TOF

Fast online GC-MS on the GV



Modeling – CESM2 simulation

Emissions:

- Anthropogenic: CAMSv5.3 and CMIP6-SSP.
- Biomass Burning: QFED2.6_FINN
- SLH:
  - CH_2Cl_2 and C_2Cl_4 (Claxton et al.)
  - $CHCl_3$ and 1,2-DCA (lower boundary condition)
  - $CHBr_3$, CH_2Br_2, CH_3I (Ordonez et al.)
- Chemical mechanism: MOZART + SLH chemistry (provided by Rafael Fernandez – Alfonso Saiz Lopez)
- Resolution: 1 degree
- Meteorology: MERRA2
- Dynamics: CAM6 (WACCM 70 layers)

1st part of talk

Major VSLS distributions and modeling

2nd part

Minor VSLS distributions

Recent published work relevant to this study – NASA ATom + other results

Measurements and Modeling of the Interhemispheric Differences of Atmospheric Chlorinated Very Short-Lived Substances

Behrooz Roozitalab[1], Louisa K. Emmons[1], Rebecca S. Hornbrook[1], Douglas E. Kinnison[1], Rafael P. Fernandez[2], Qinyi Li[2,3,4], Alfonso Saiz-Lopez[2], Ryan Hossaini[5], Carlos A. Cuevas[2], Alan J. Hills[1], Stephen A. Montzka[6], Donald R. Blake[7], William H. Brune[8], Patrick R. Veres[9], and Eric C. Apel[1]

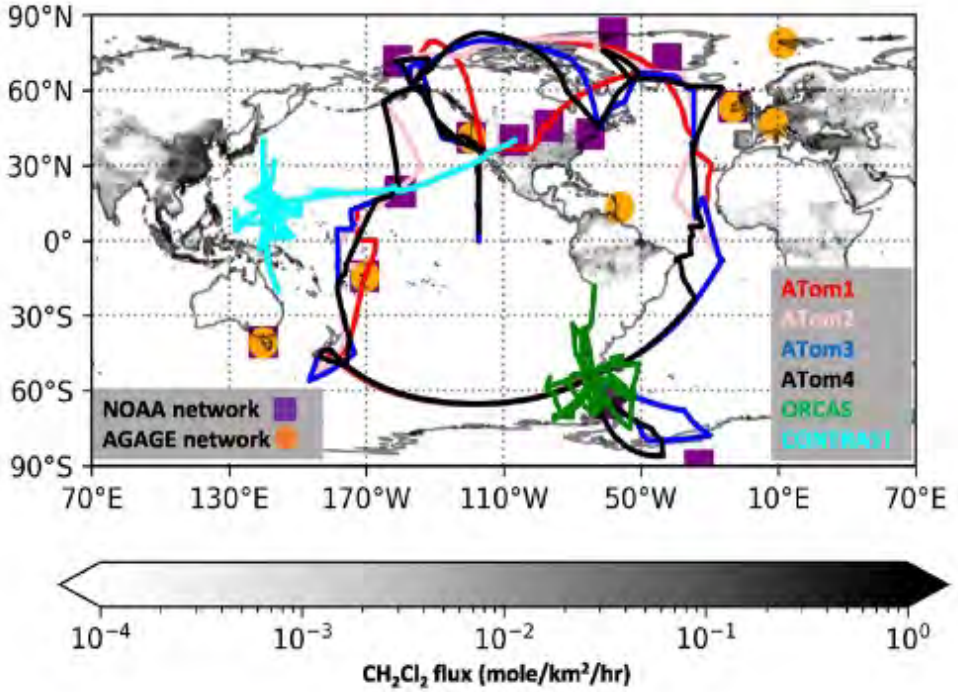


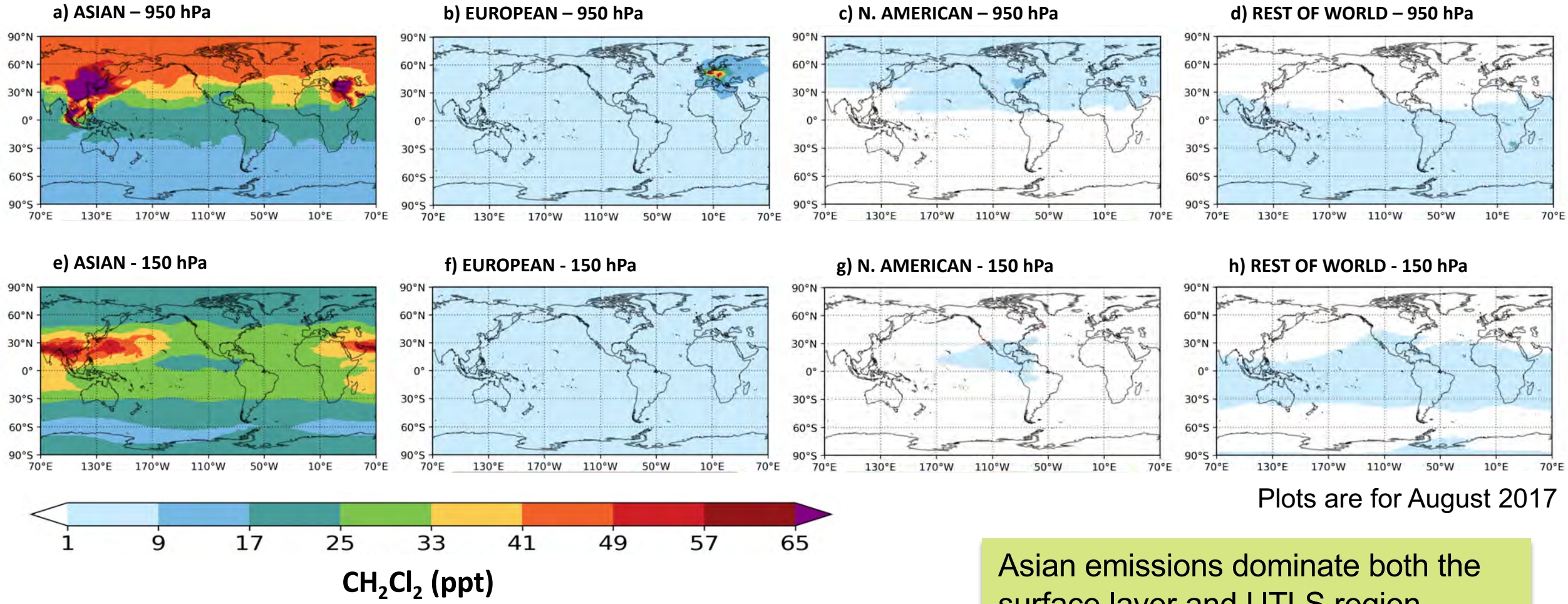
Figure 1. Map of the flight tracks for individual ATom deployments, and ORCAS and CONTRAST missions. The ground measurement sites in the NOAA and AGAGE networks used in this study are also shown. The map is shaded by the 2016 annual average CH_2Cl_2 emissions developed by Claxton et al. (2020).

- Global observations of **Cl-VSLS** (CH_2Cl_2, $CHCl_3$, C_2Cl_4, and 1,2-dichloroethane) were made during ATom (4 seasons, BL to ~12 km)
- Comparisons to updated CESM global chemical transport model show reasonable agreement at simulating VSLS distribution
- Disproportionately high impact of Asian Cl-VSLS emissions regionally, and globally throughout the troposphere.

Roozitalab et al., 2024

Tagged CH_2Cl_2 Distribution

Figure 7 from Roozitalab et al., 2024

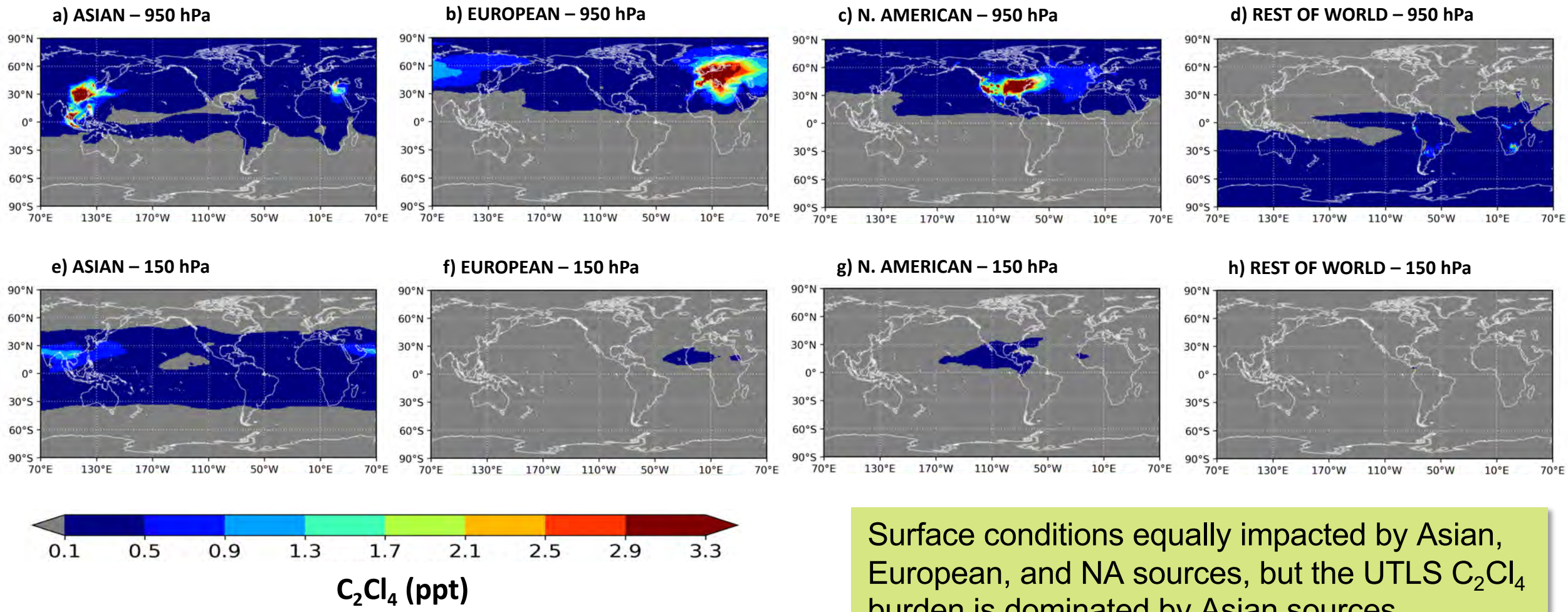


Plots are for August 2017

Asian emissions dominate both the surface layer and UTLS region.

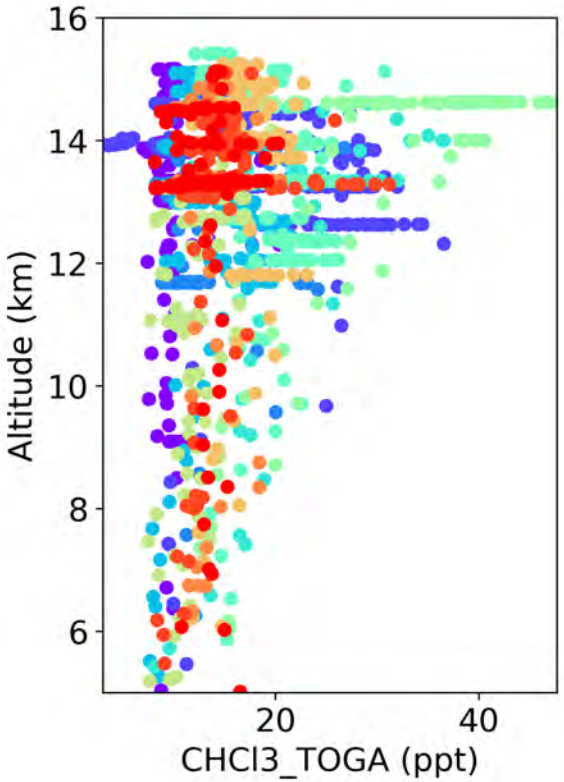
Tagged C_2Cl_4 Distribution

Figure 8 from Roozitalab et al., 2024

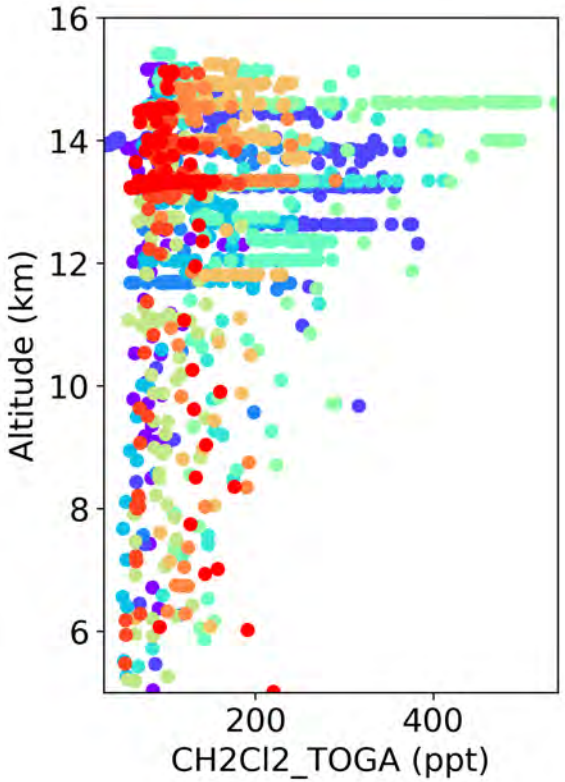


Surface conditions equally impacted by Asian, European, and NA sources, but the UTLS C_2Cl_4 burden is dominated by Asian sources.

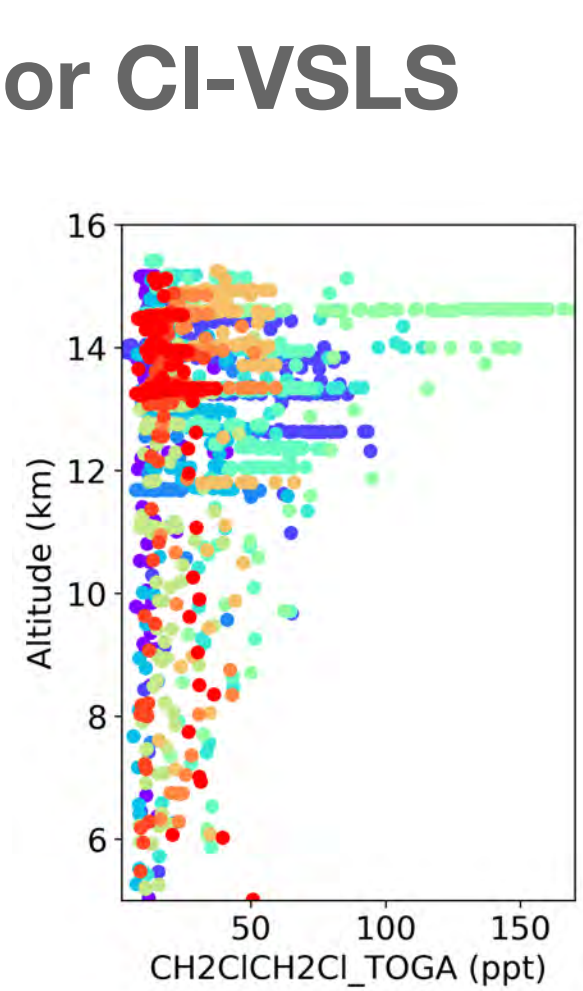
ACCLIP Observations of Major Cl-VSLS



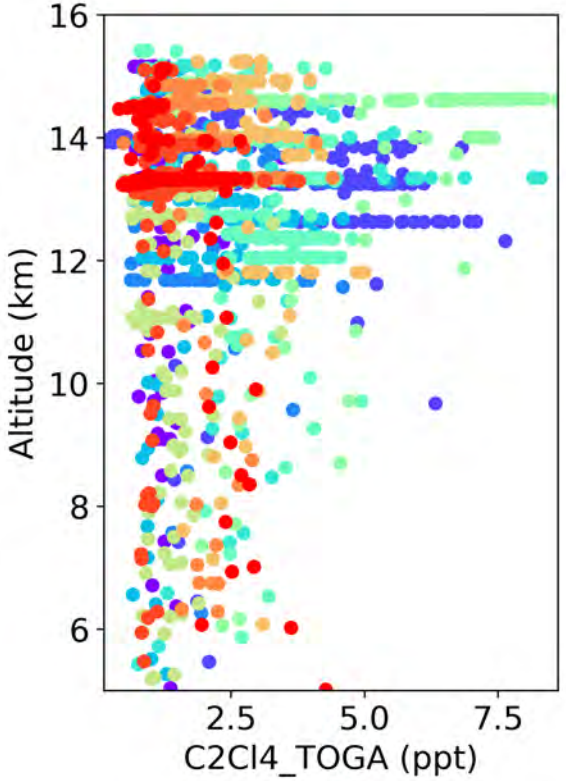
Chloroform
$CHCl_3$
lifetime ~ 5-6 months



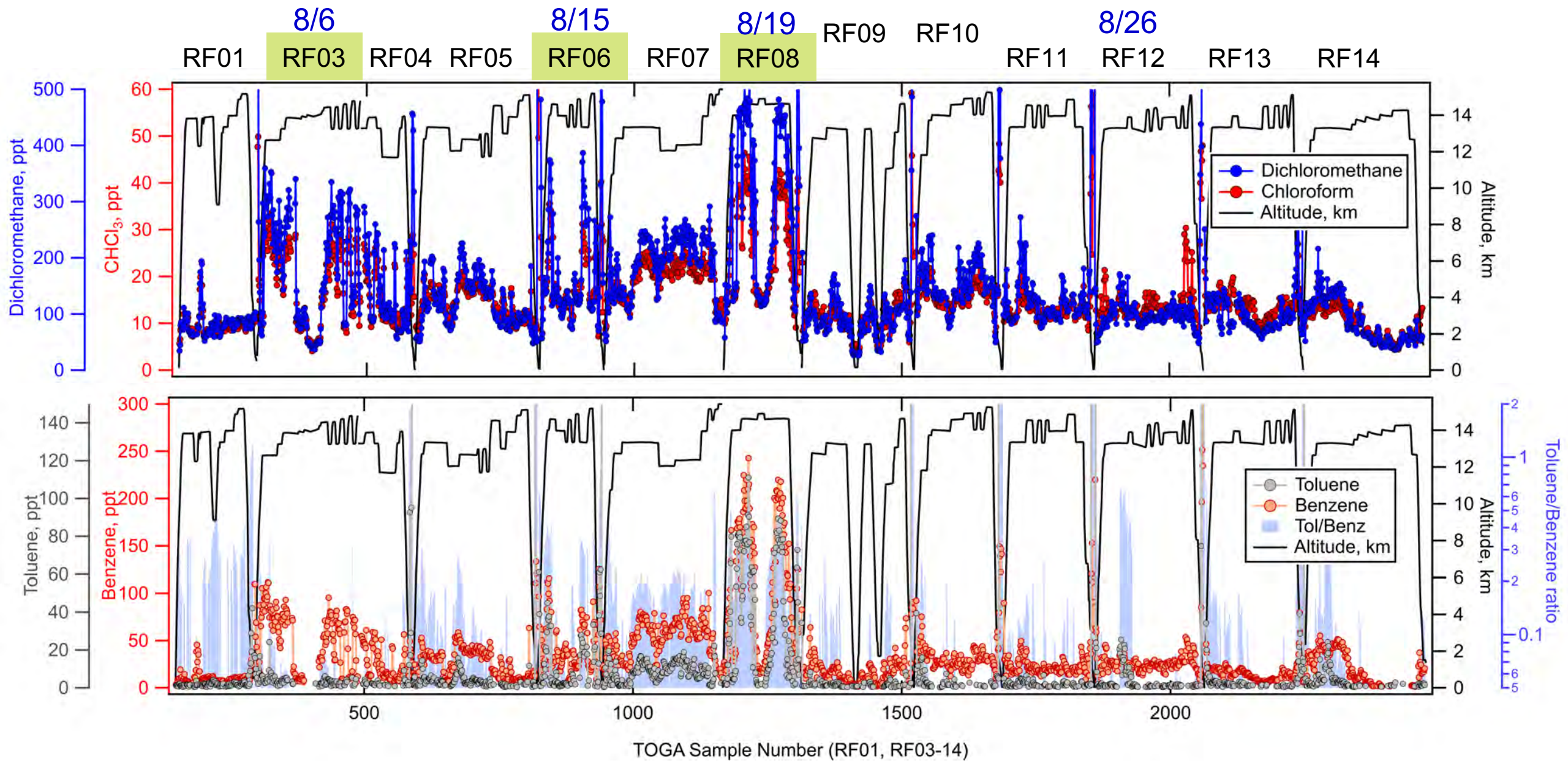
Dichloromethane
CH_2Cl_2
lifetime ~ 5-6 months



1,2-Dichloroethane
CH_2Cl-CH_2Cl
lifetime ~ 2 months



Tetrachloroethene
C_2Cl_4
lifetime ~ 2-3 months



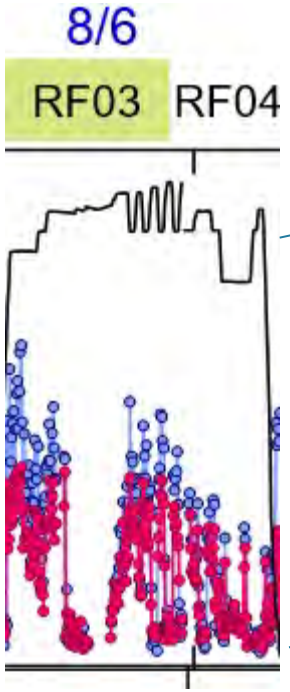
TOGA-TOF; gas-phase VOCs during ACCLIP

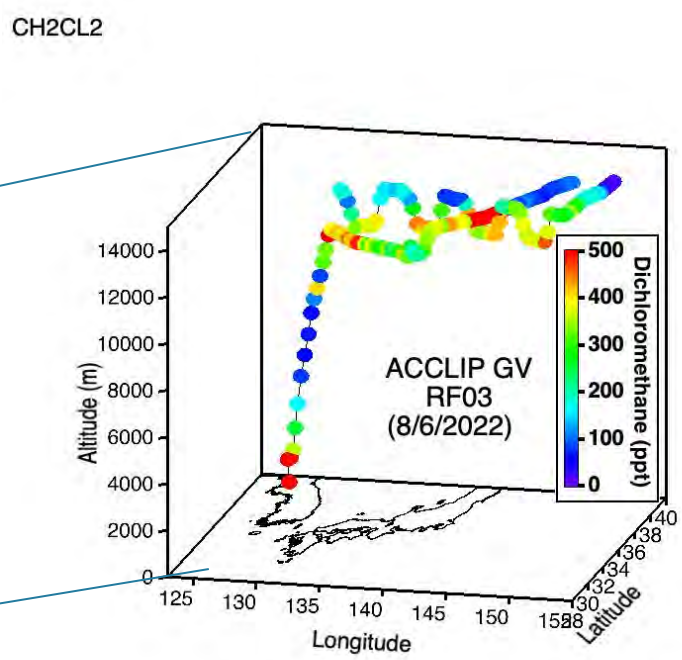
3D views of flight tracks (dichloroethane) RF03, RF06, RF08

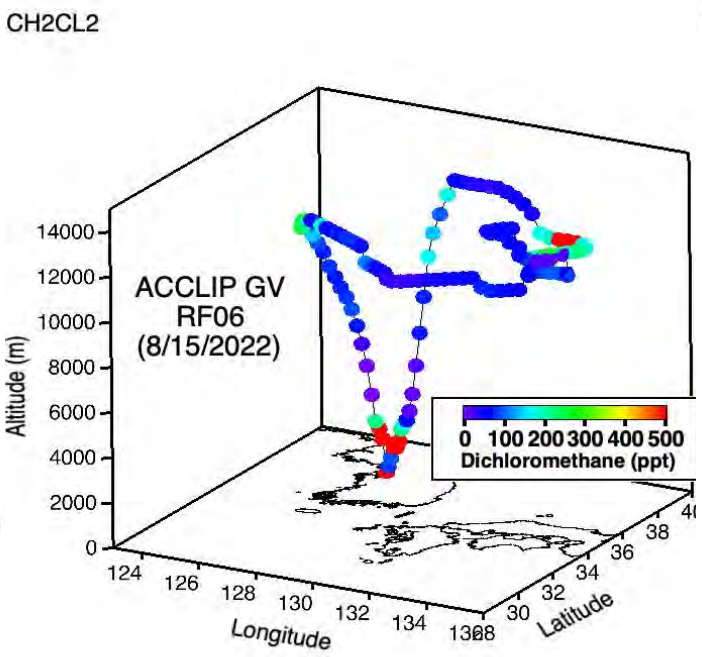
RF03 8/6

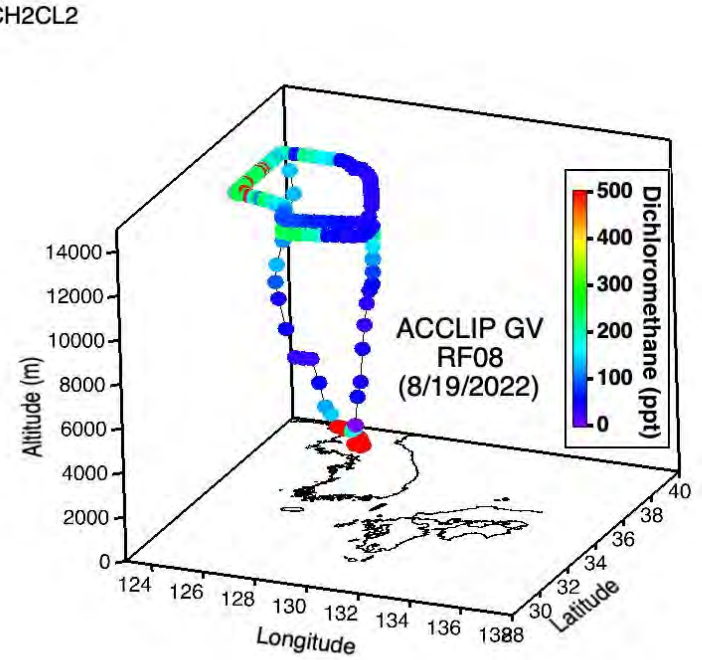
RF06 8/15

RF08 8/19





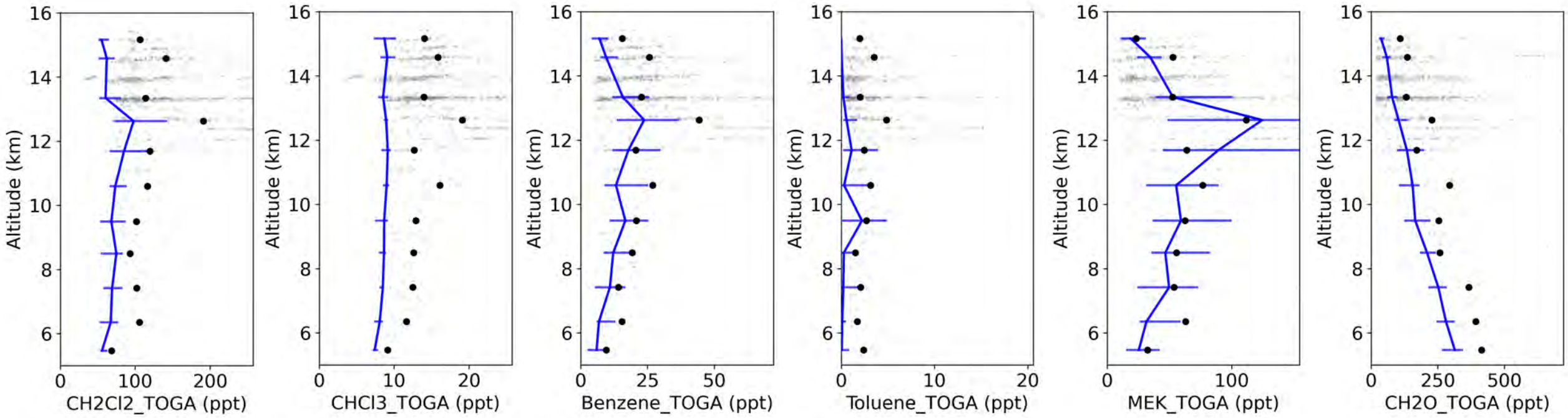




Comparisons of ACCLIP VOC/VSLS observations to CESM2 Model Output (all flights + select flights)

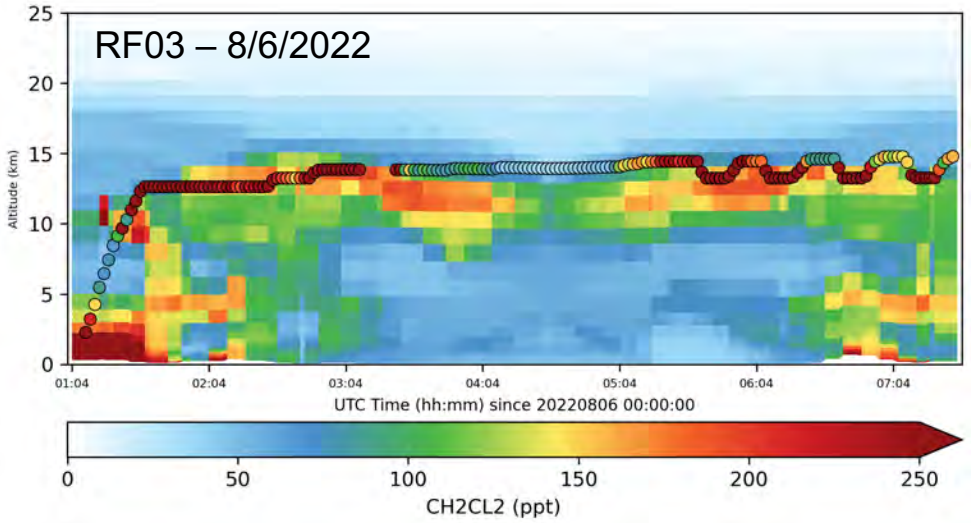
ALL GV flights

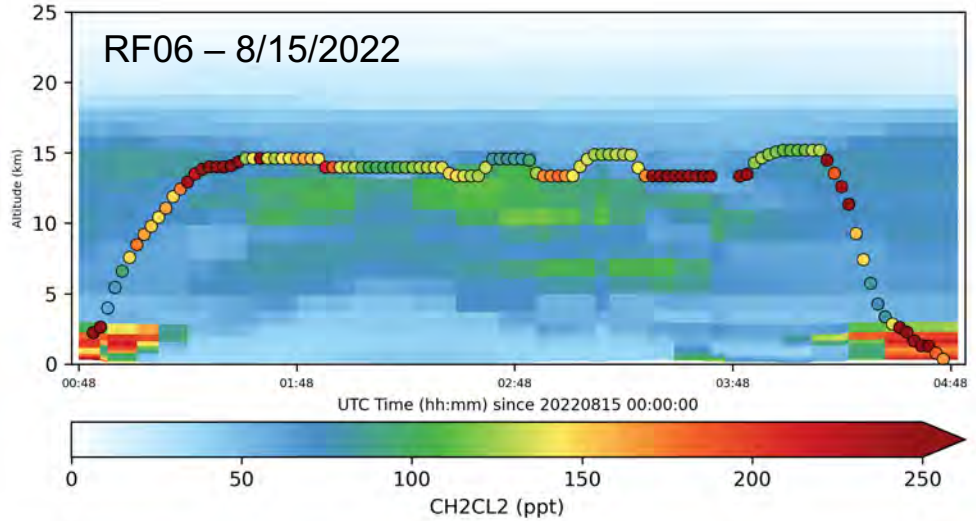
Comparisons of ACCLIP individual species observations to CESM2 Model Output

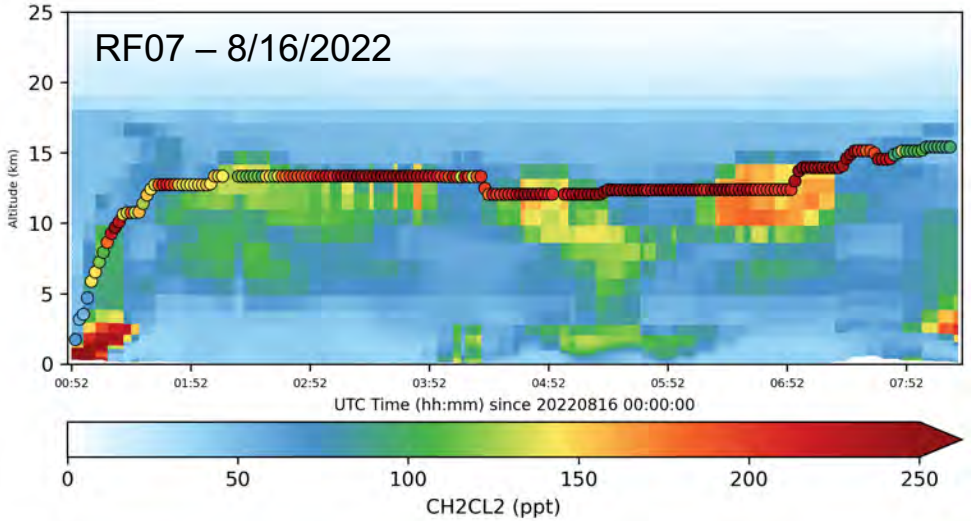


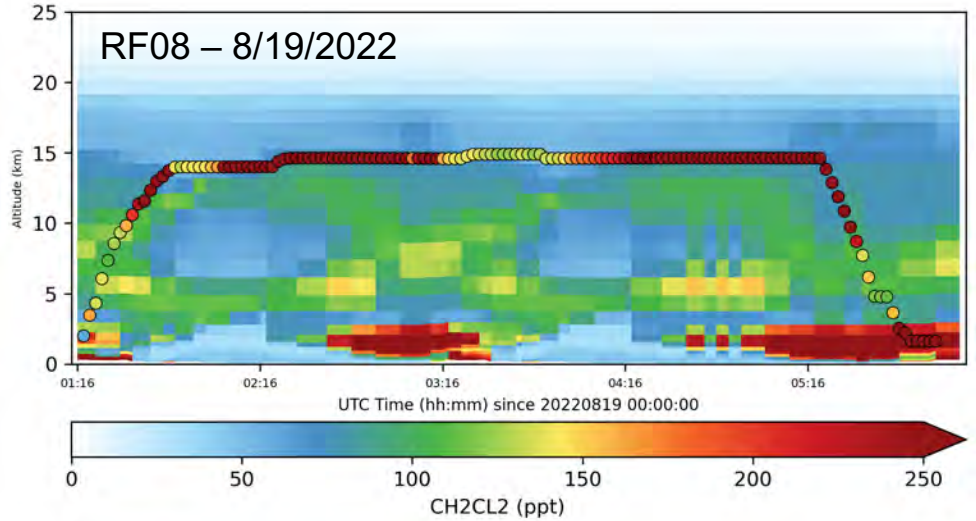
Select VOC species

Curtain plots: Comparisons of ACCLIP observations to CESM2 Model Output: CH_2Cl_2



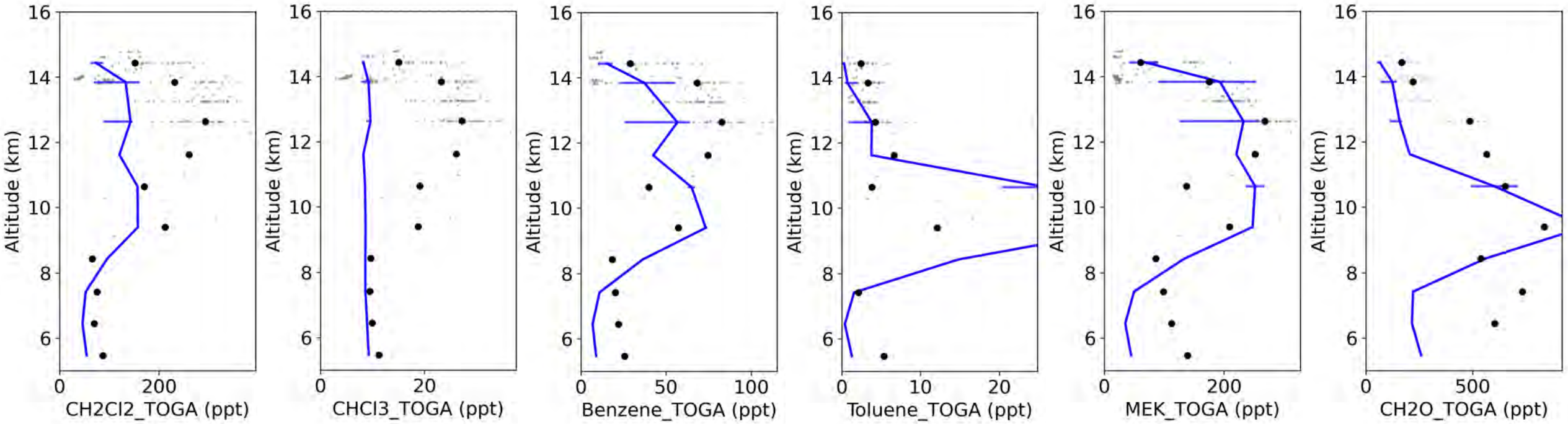






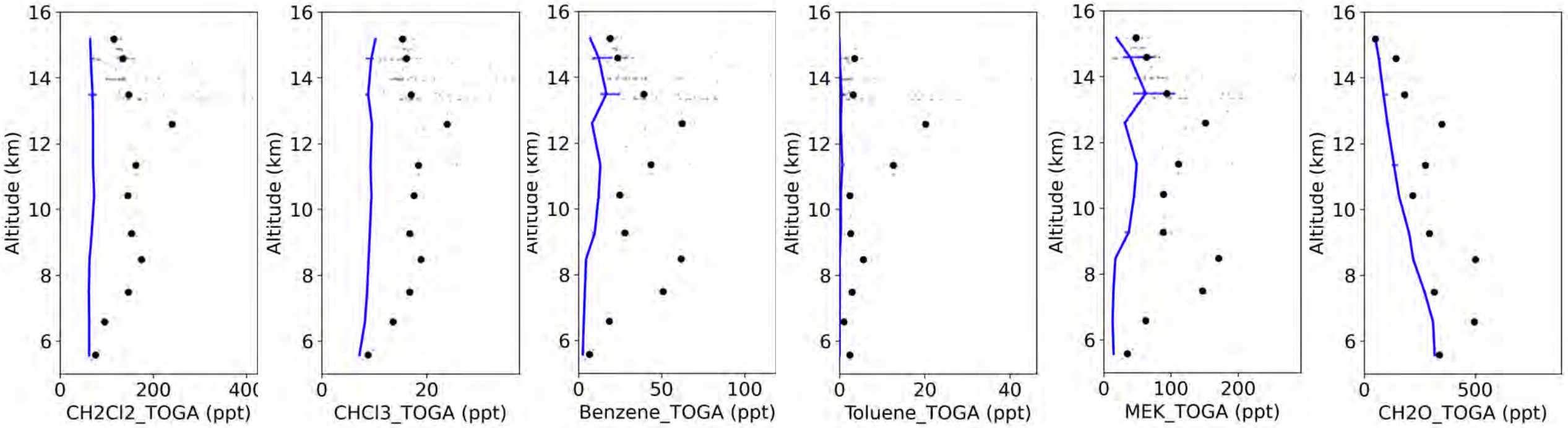
RF03
8/6

Comparisons of ACCLIP individual species observations to CESM2 Model Output

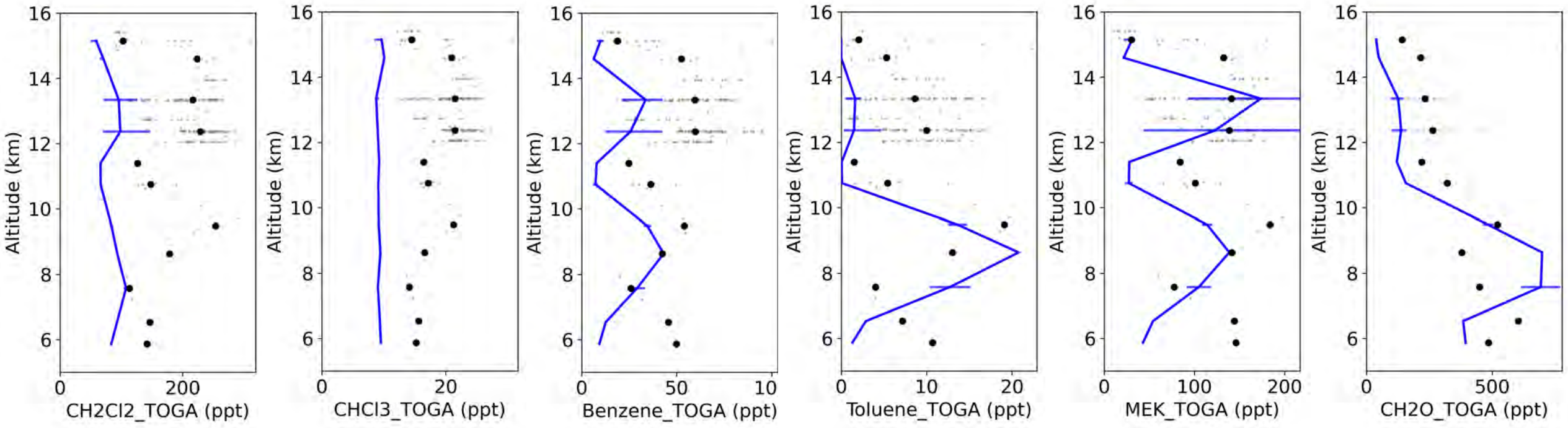


RF06
8/15

Comparisons of ACCLIP observations to CESM2 Model Output

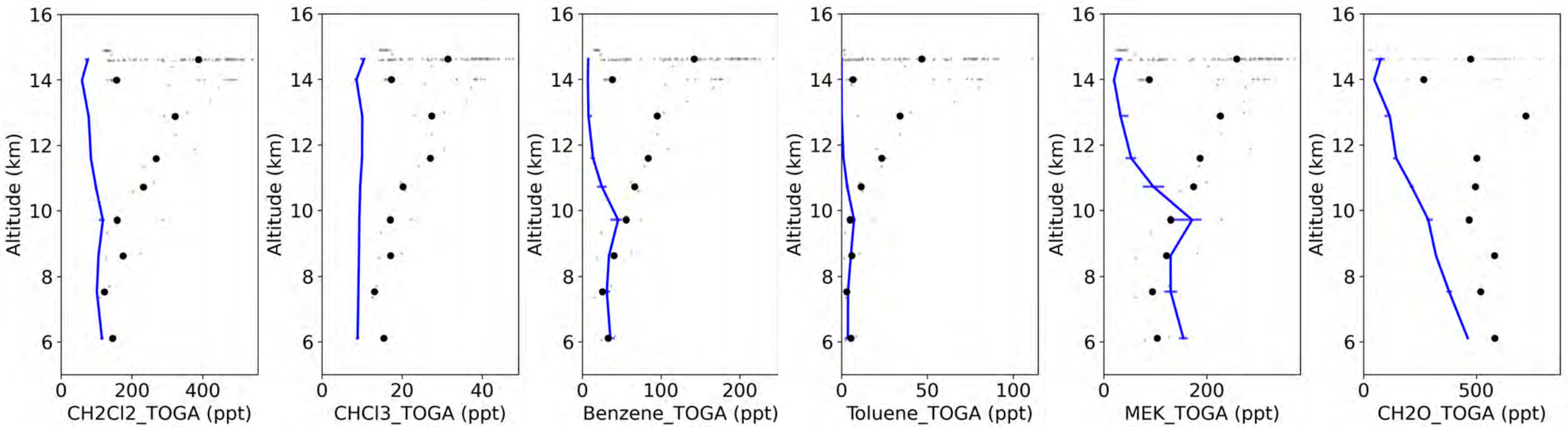


Comparisons of ACCLIP observations to CESM2 Model Output

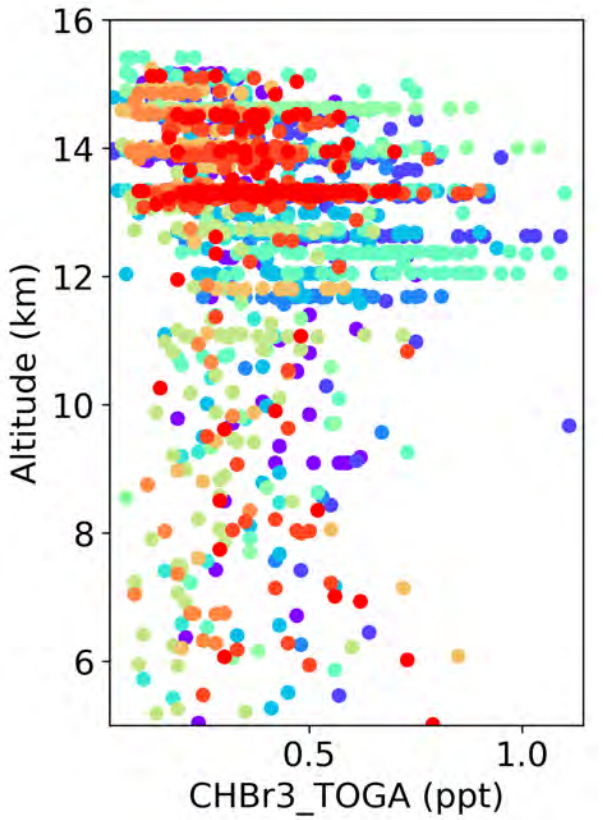


RF08
8/19

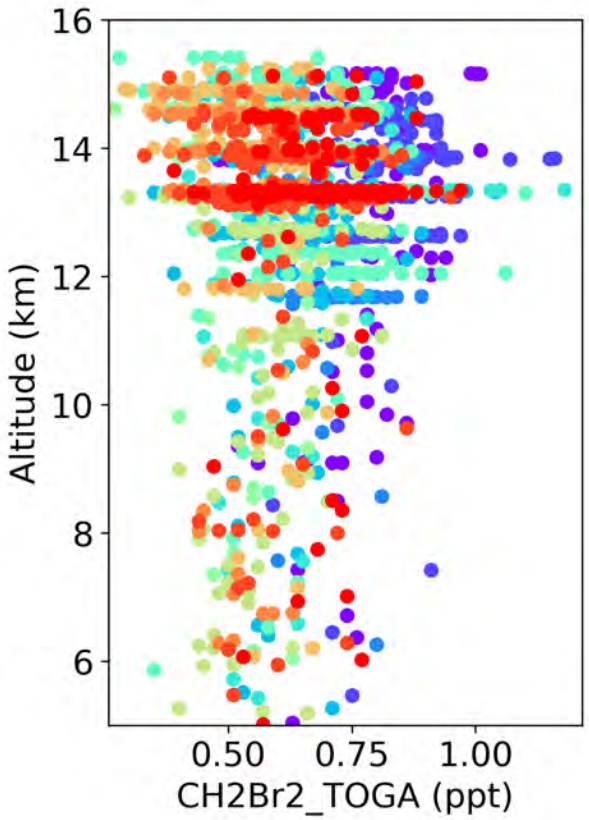
Comparisons of ACCLIP observations to CESM2 Model Output



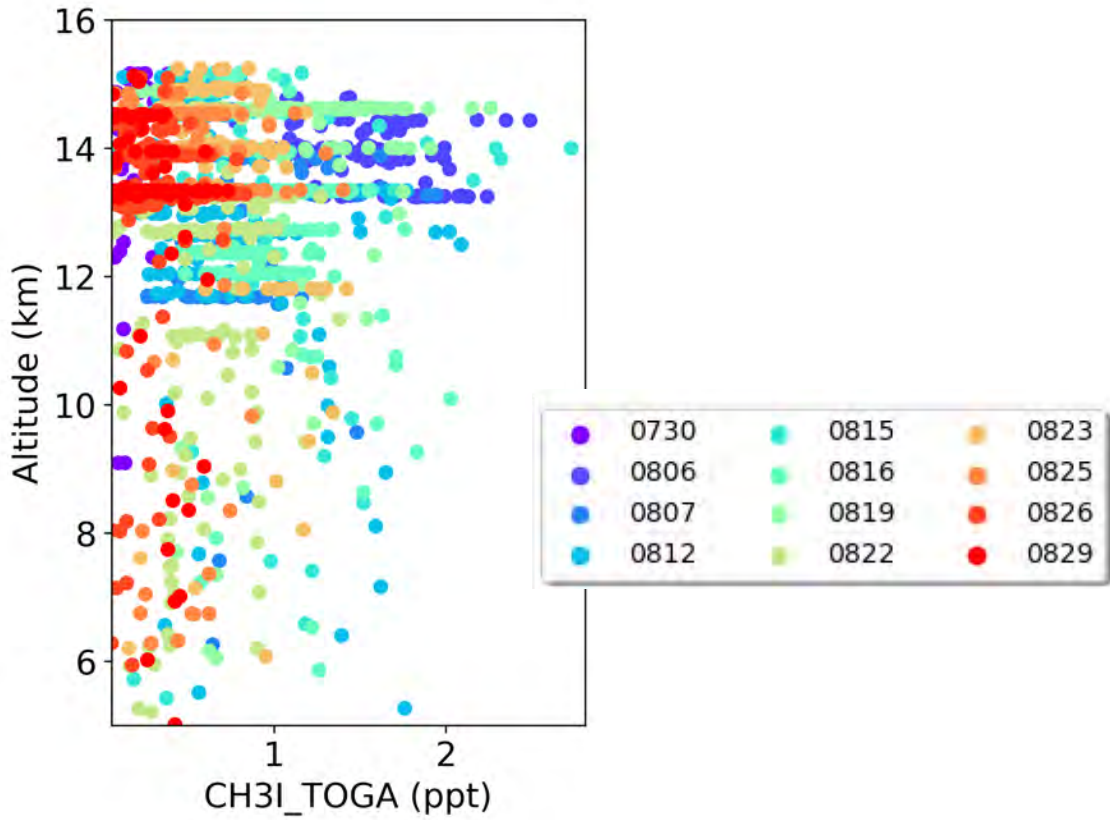
TOGA ACCLIP Observations of Other “Major” VSLS



Bromoform
$CHBr_3$
Lifetime ~ 1 month



Dibromomethane
CH_2Br_2
Lifetime ~ 3-4 months

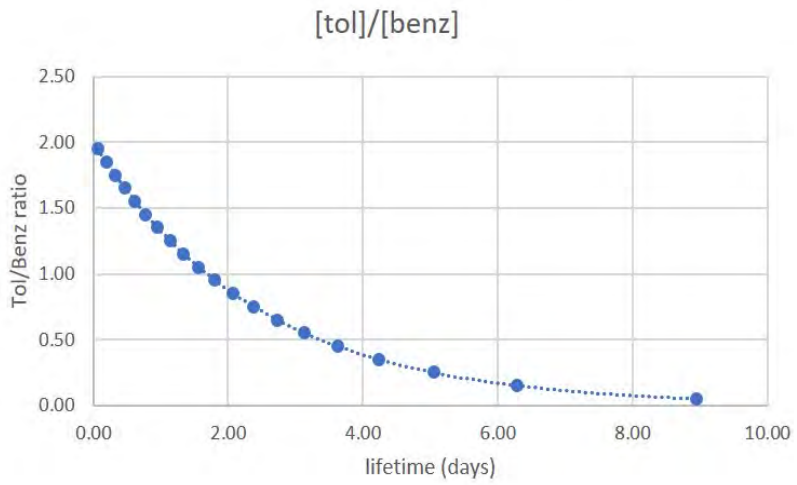


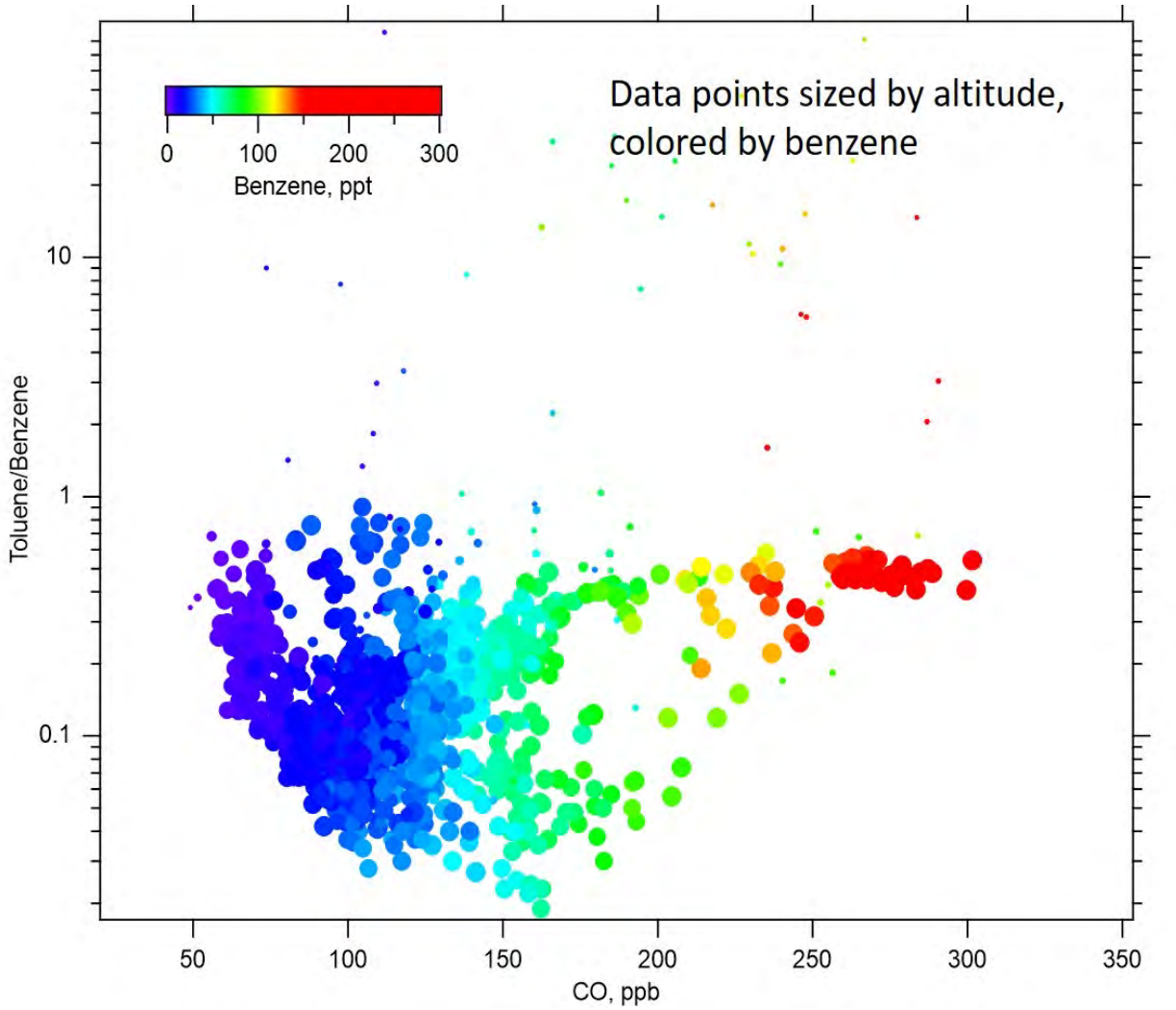
Methyl Iodide
CH_3I
Lifetime ~ < 1 week

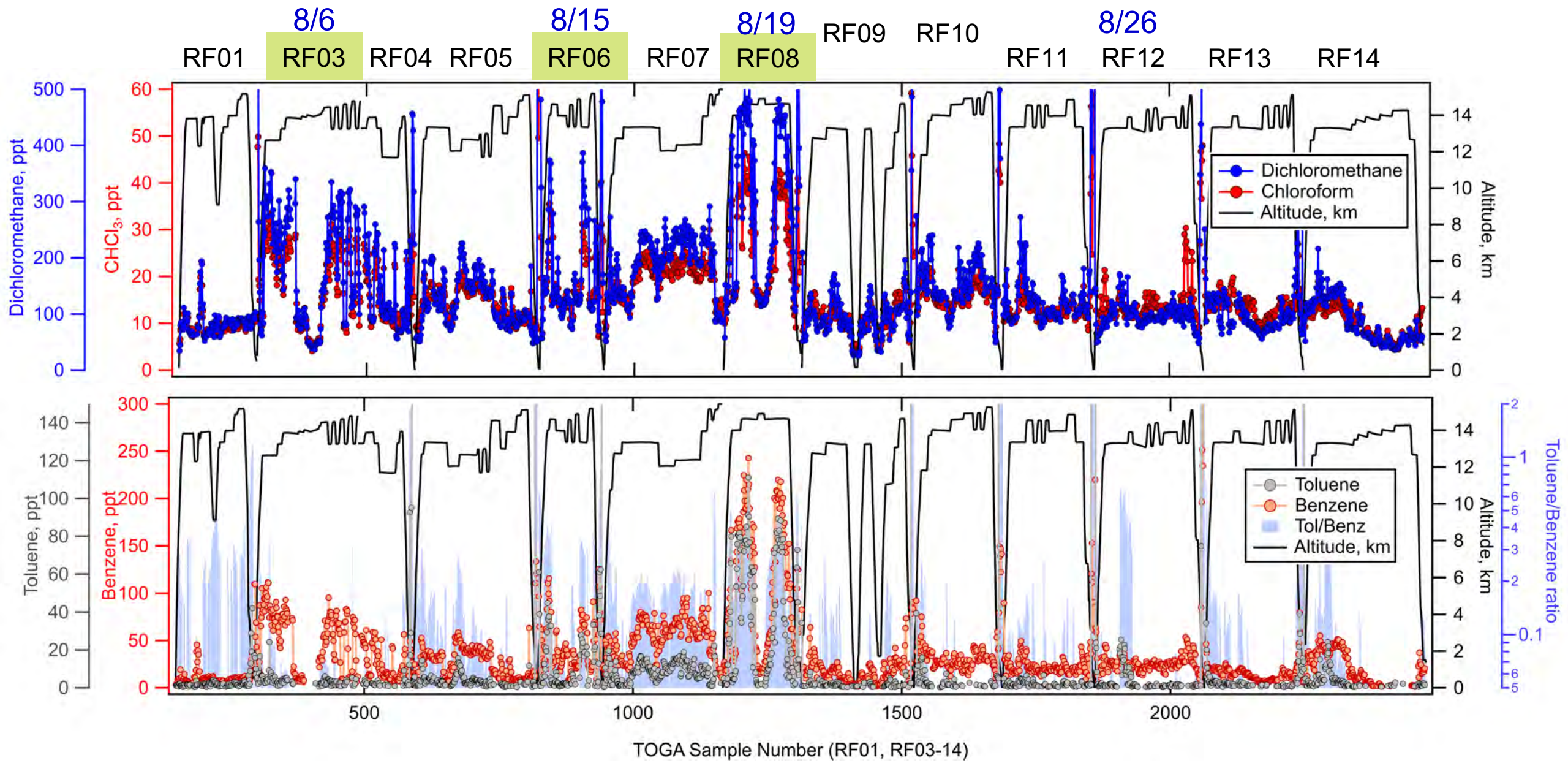
“Minor” VSLS observations

Lifetime considerations – benzene/toluene ratios

@ [OH] = 1E-06 and an assumed emission ratio tol/benz = 2

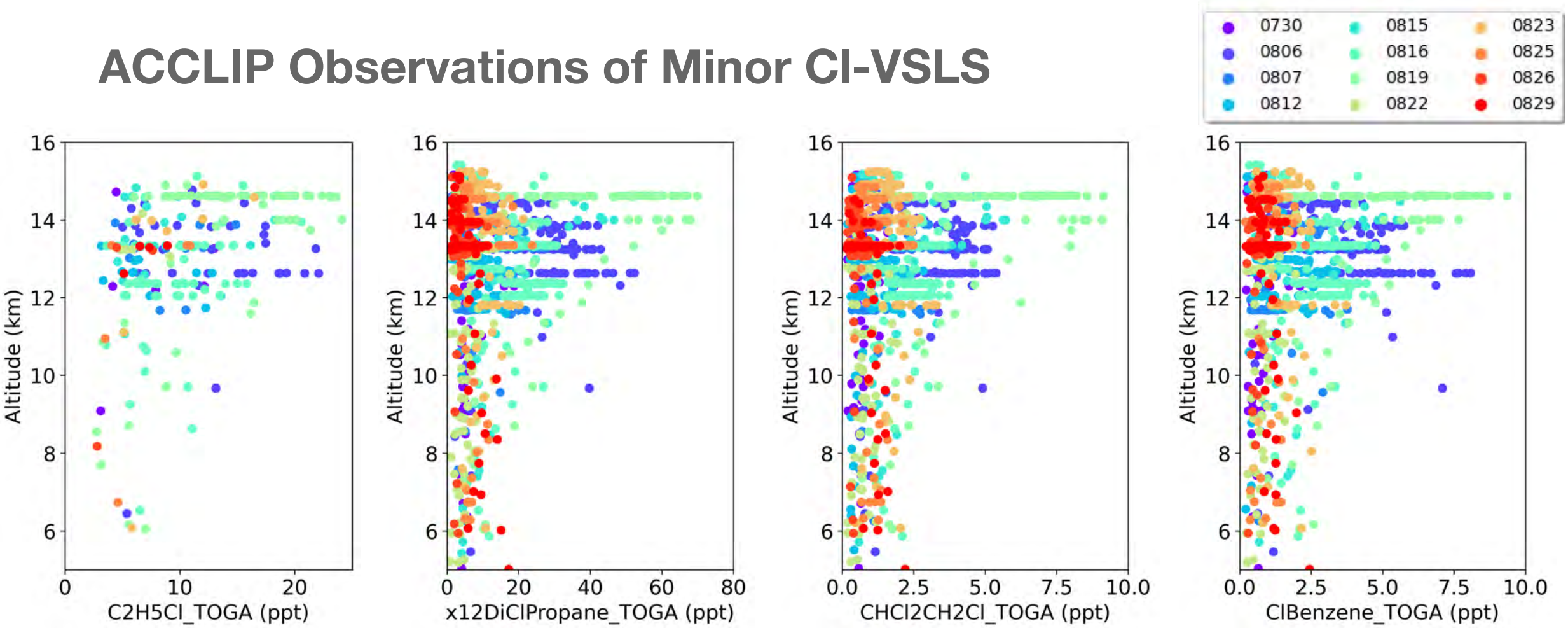






TOGA-TOF; gas-phase VOCs during ACCLIP

ACCLIP Observations of Minor Cl-VSLS



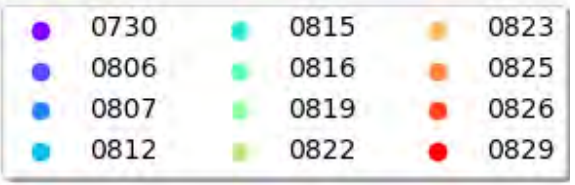
Chloroethane
C_2H_5Cl
Lifetime ~ 1 month

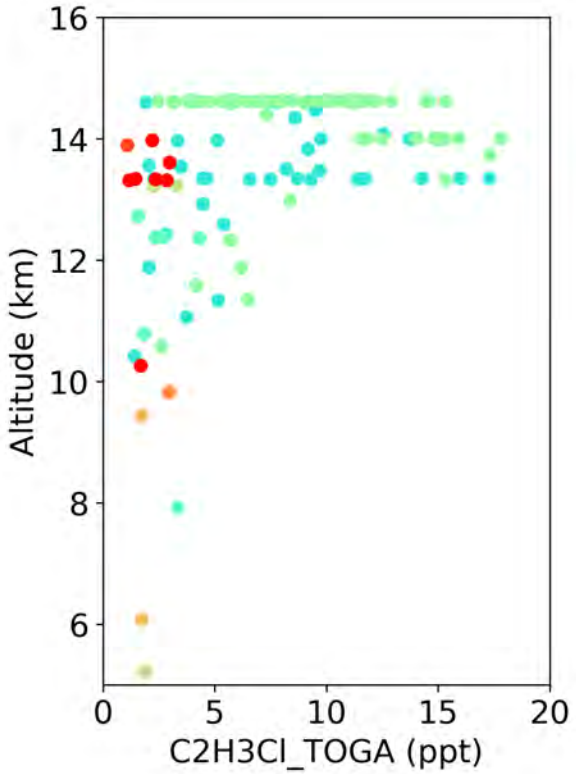
1,2-Dichloropropane
$C_3H_6Cl_2$
Lifetime ~ < 2 weeks

1,1,2-Trichloroethane
$C_2H_3Cl_3$
Lifetime ~ 1-2 months

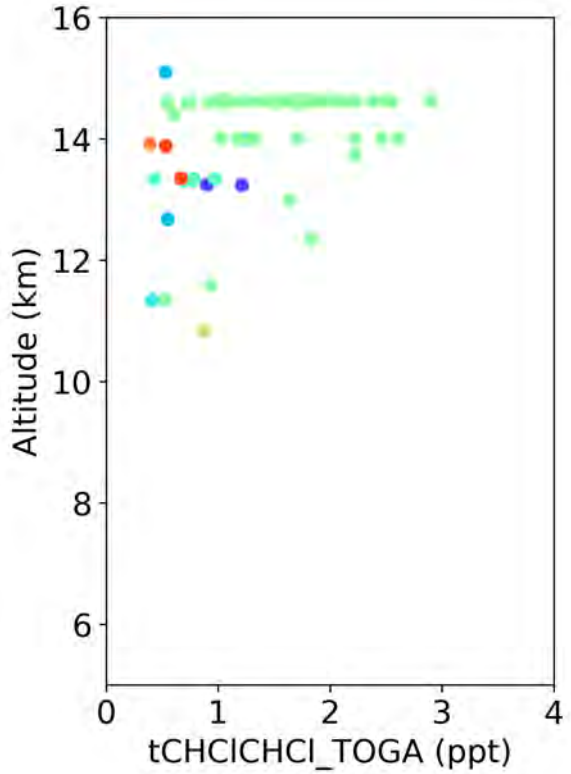
Chlorobenzene
C_6H_5Cl
Lifetime ~ 3 weeks

ACCLIP Observations of Minor Cl-VSLS

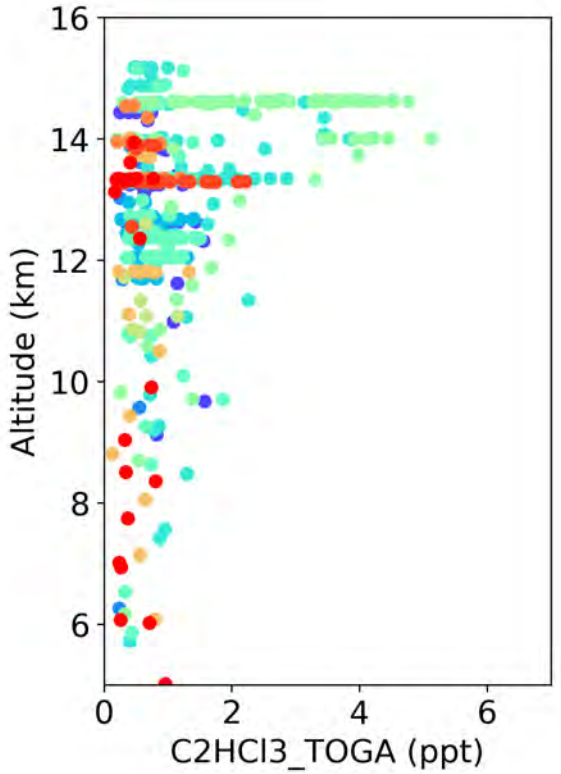




Chloroethene
C_2H_3Cl
Lifetime ~ 2 days

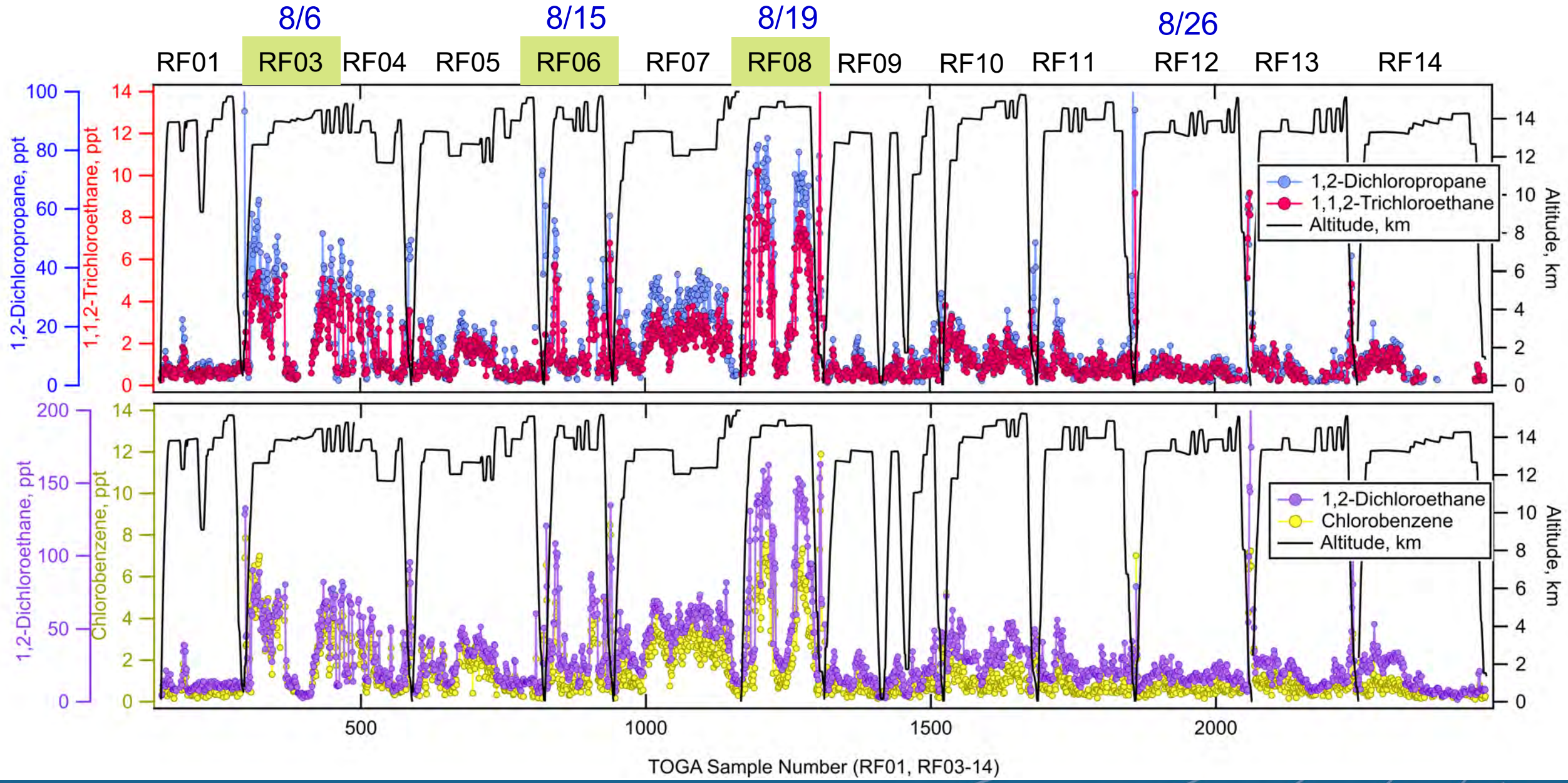


trans-1,2-Dichloroethene
$C_2H_2Cl_2$
Lifetime ~ 5 days

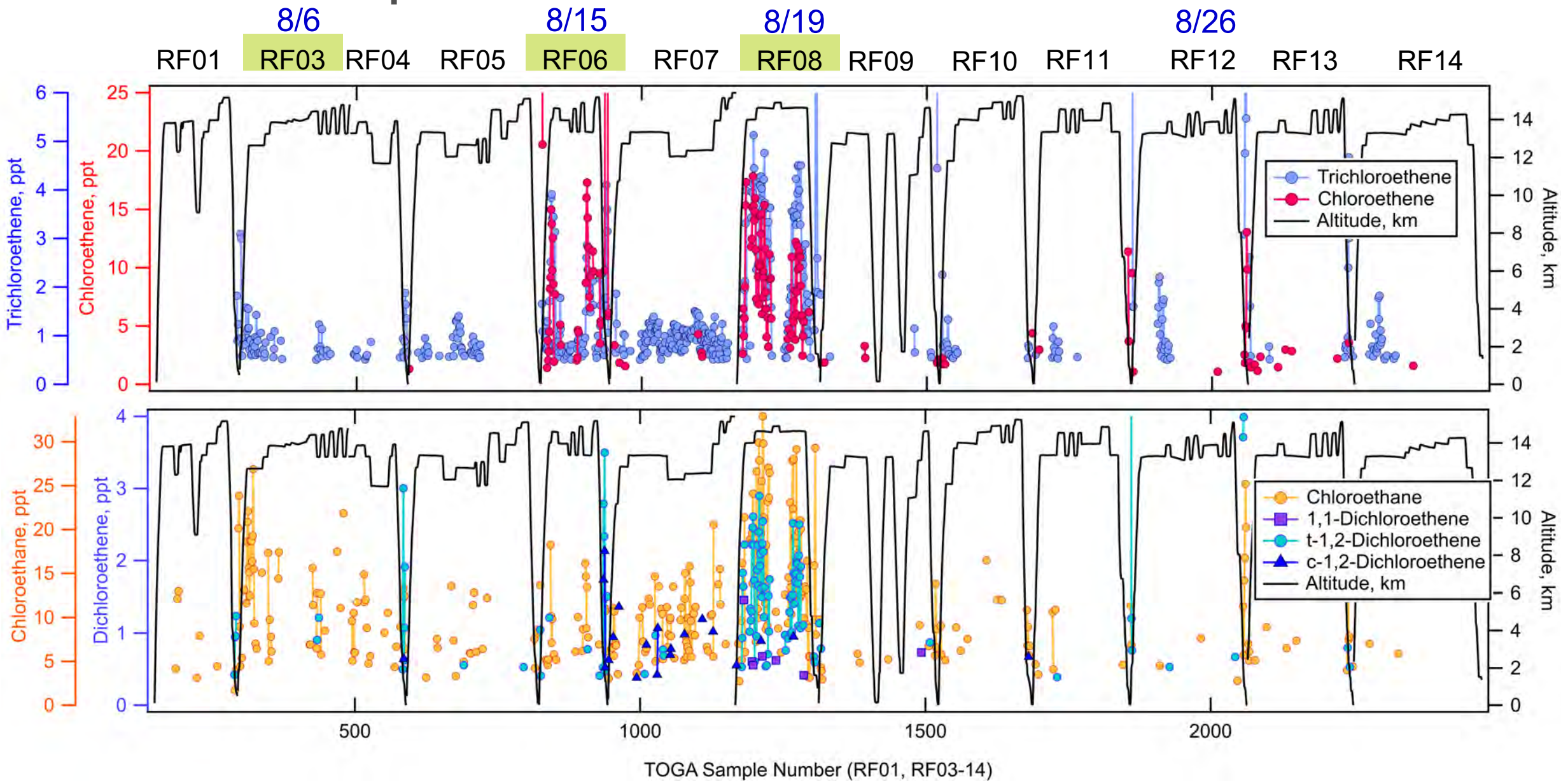


Trichloroethene
C_2HCl_3
Lifetime ~ 1 week

Minor longer-lived species

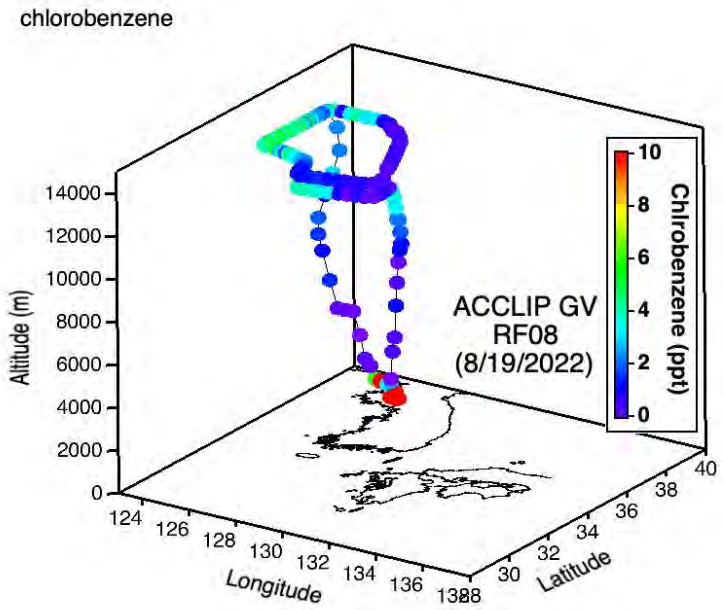


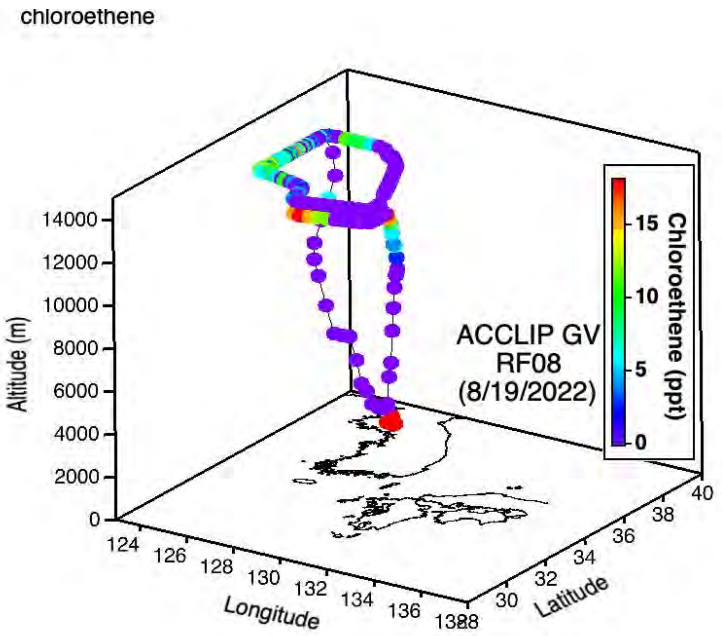
Minor shorter-lived species

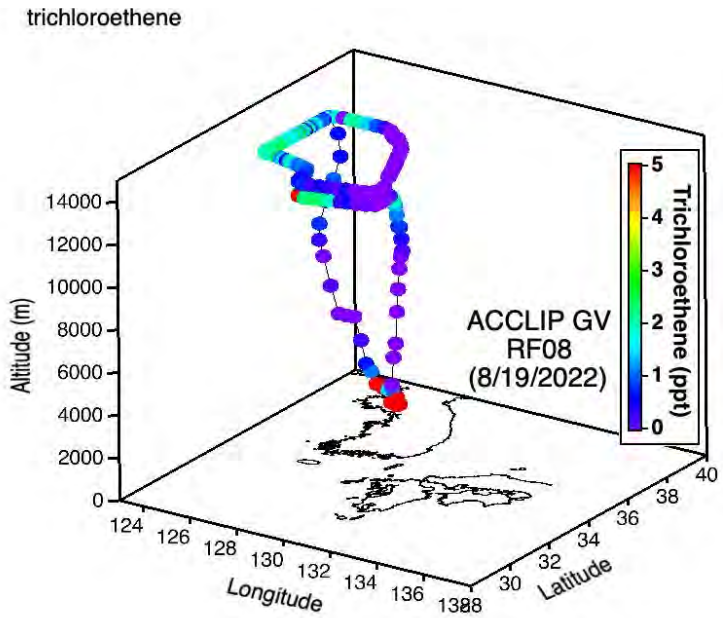


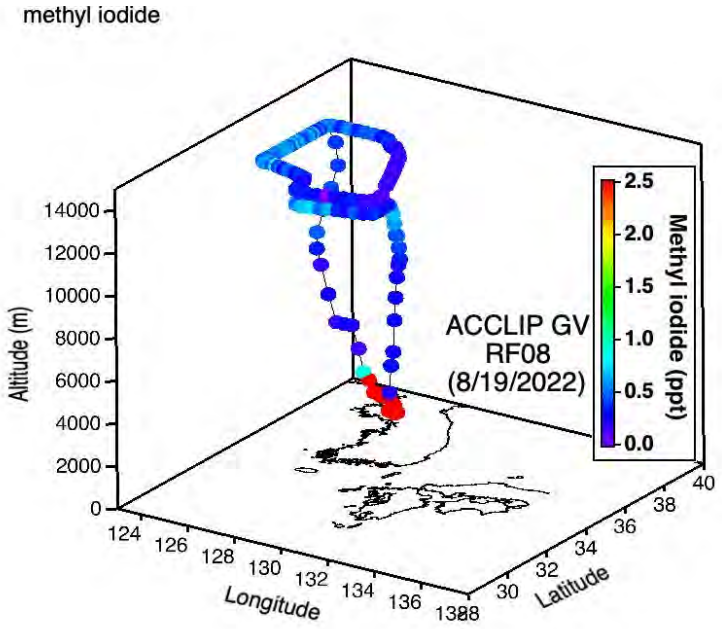
RF08
8/19

3D Plots of some minor Cl-VSLS
+
CH3I



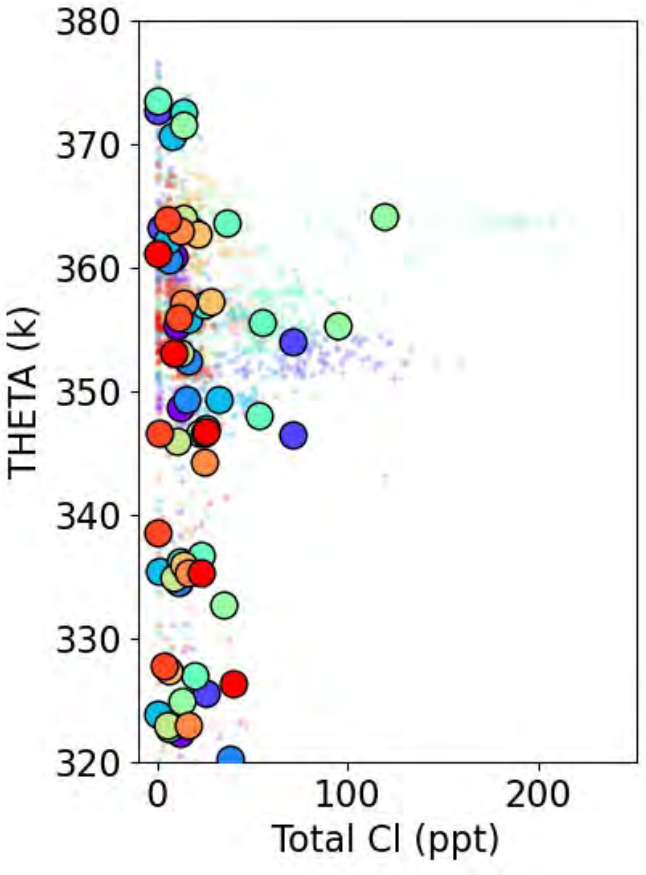




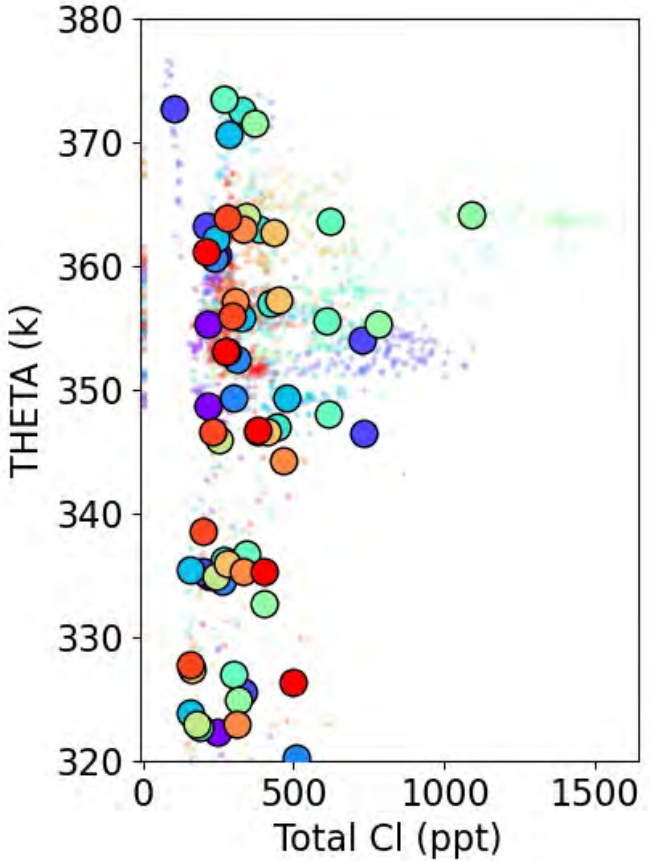


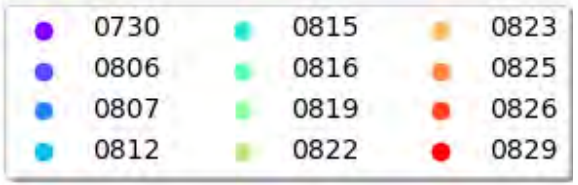
Total Cl from Cl-VSLS species

Minor Species (7 compounds)



Major Species (4 compounds)





Summary

Major VSLS species were observed and modeled

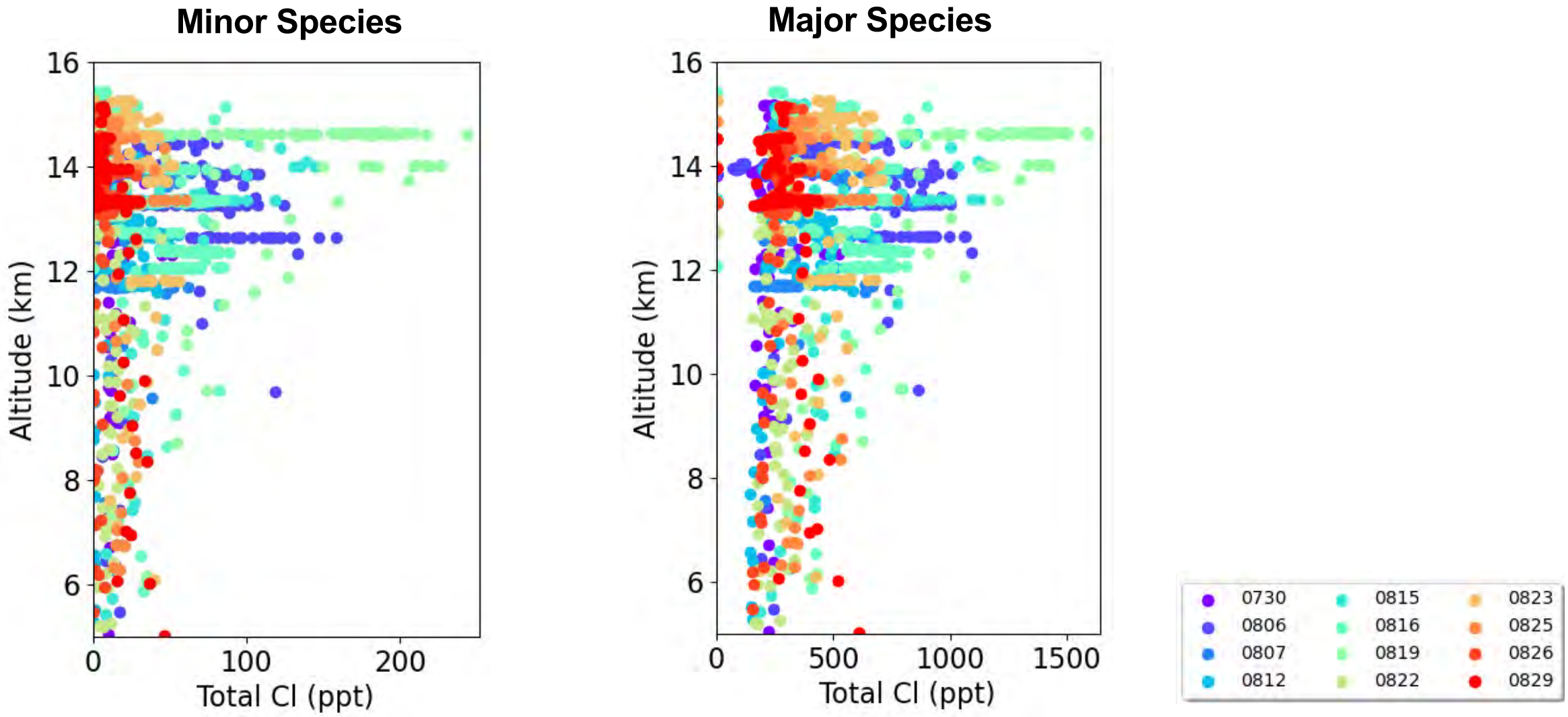
- Cl species generally underrepresented in the model

Minor VSLS species were observed as well in the convective outflow in cases of fresh convection with their impact still under study

Thank you and entire ACCLIP science team!

Extra

Total Cl from Cl-VSLS species



CH_2Cl_2 Emissions From Different Regions of Globe