X5.107 ( ITS1.8/CL0.2)

# Creating Global, Gap-Free Stratospheric Datasets for Montreal Protocol Assessments

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Aim: *To construct good quality observation-based data for the envi. monitoring & chemical model evaluations*

- *CFC11 V2 : https://doi.org/10.5281/zenodo.18145730*
- *CFC12 V2 : https://doi.org/10.5281/zenodo. 18147392*
- *CH4 V2 : https://doi.org/10.5281/zenodo.18197333*
- *N2O V2 : https://doi.org/10.5281/zenodo.18197444*
- *HCl V2 : https://doi.org/10.5281/zenodo.18184430*
- *HF V2 : https://doi.org/10.5281/zenodo.18184779*
- *HNO3 V2 : https://doi.org/10.5281/zenodo.18199002*
- *O3 V2 : https://doi.org/10.5281/zenodo.18199586*
- *H2O V2 : https://doi.org/10.5281/zenodo. 18199962*
- *COF2 V2 : https://doi.org/10.5281/zenodo. 18201786*

**Status:** this preprint is currently under review for the journal ESSD.

TCOM-CFC11 and TCOM-CFC12: A Gap-Free, Observationally Constrained Global Dataset of Stratospheric CFC-11 and CFC-12 Profiles (v2.0)

https://tomcat.leeds.ac.uk/tomcat/tcom/

occultation measurement-based stratospheric methane (TCOM-CH4) and nitrous oxide (TCOM-N2O) profile data sets

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from a chemical transport model

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NATURAL ENVIRONMENT RESEARCH COUNCIL

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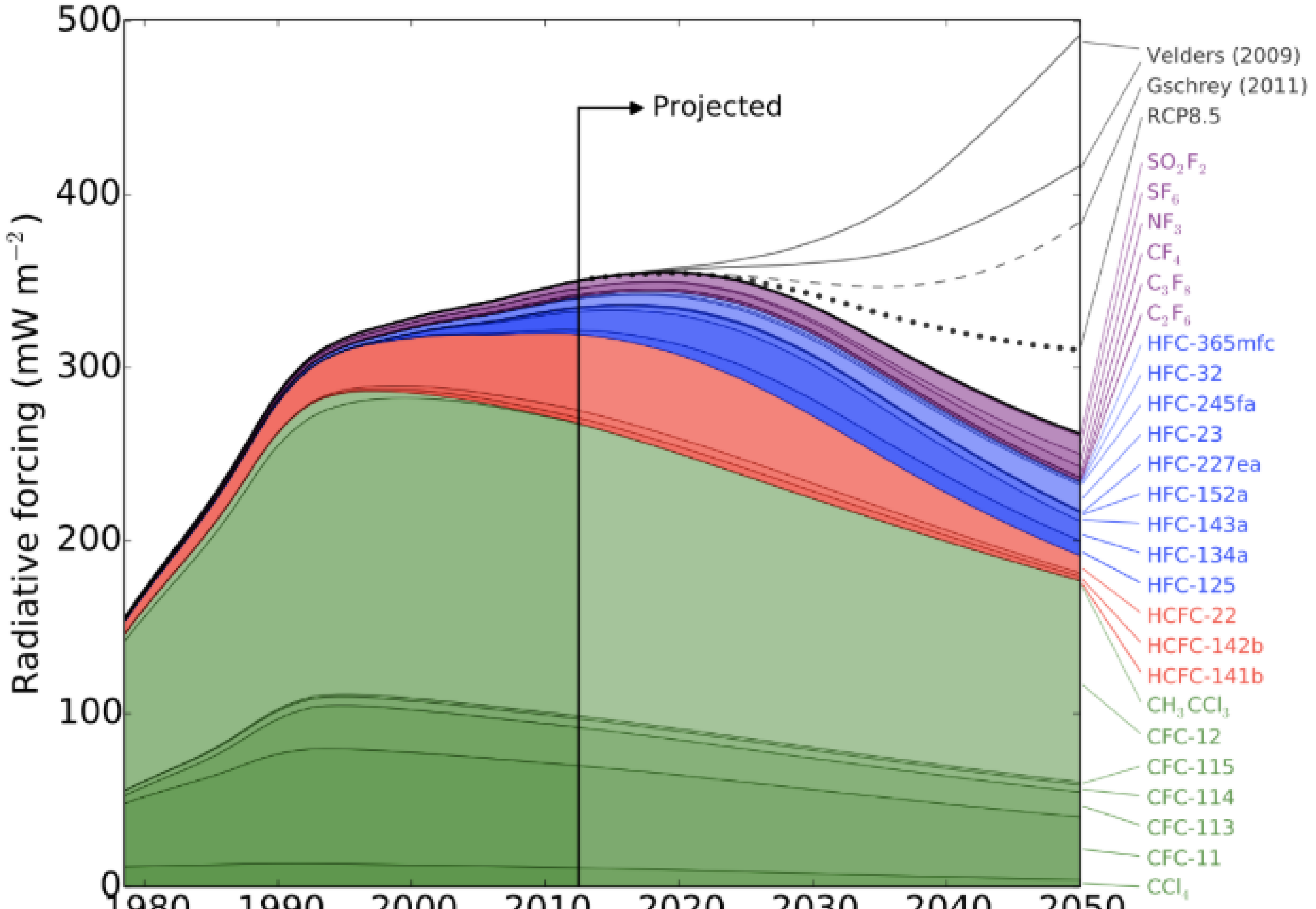
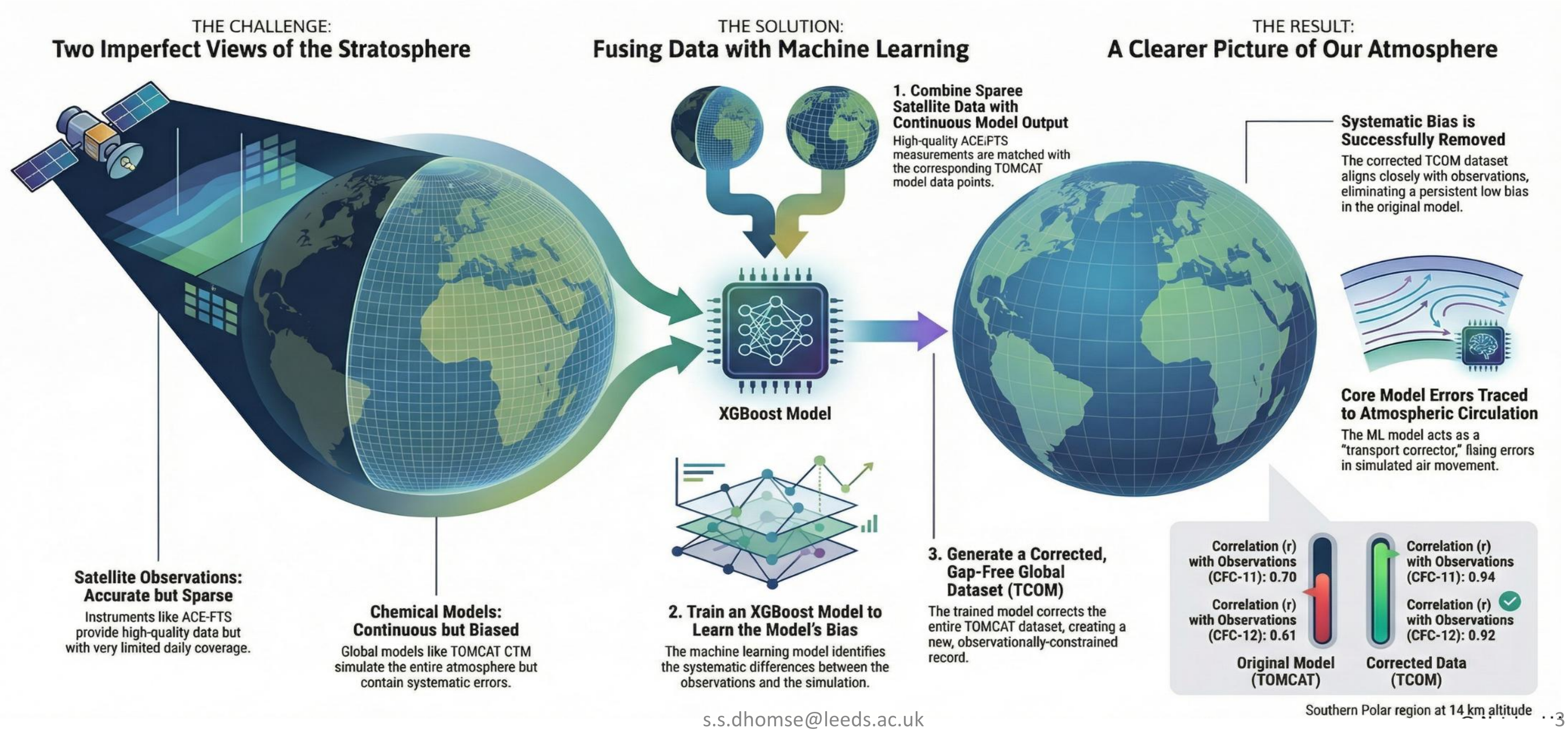


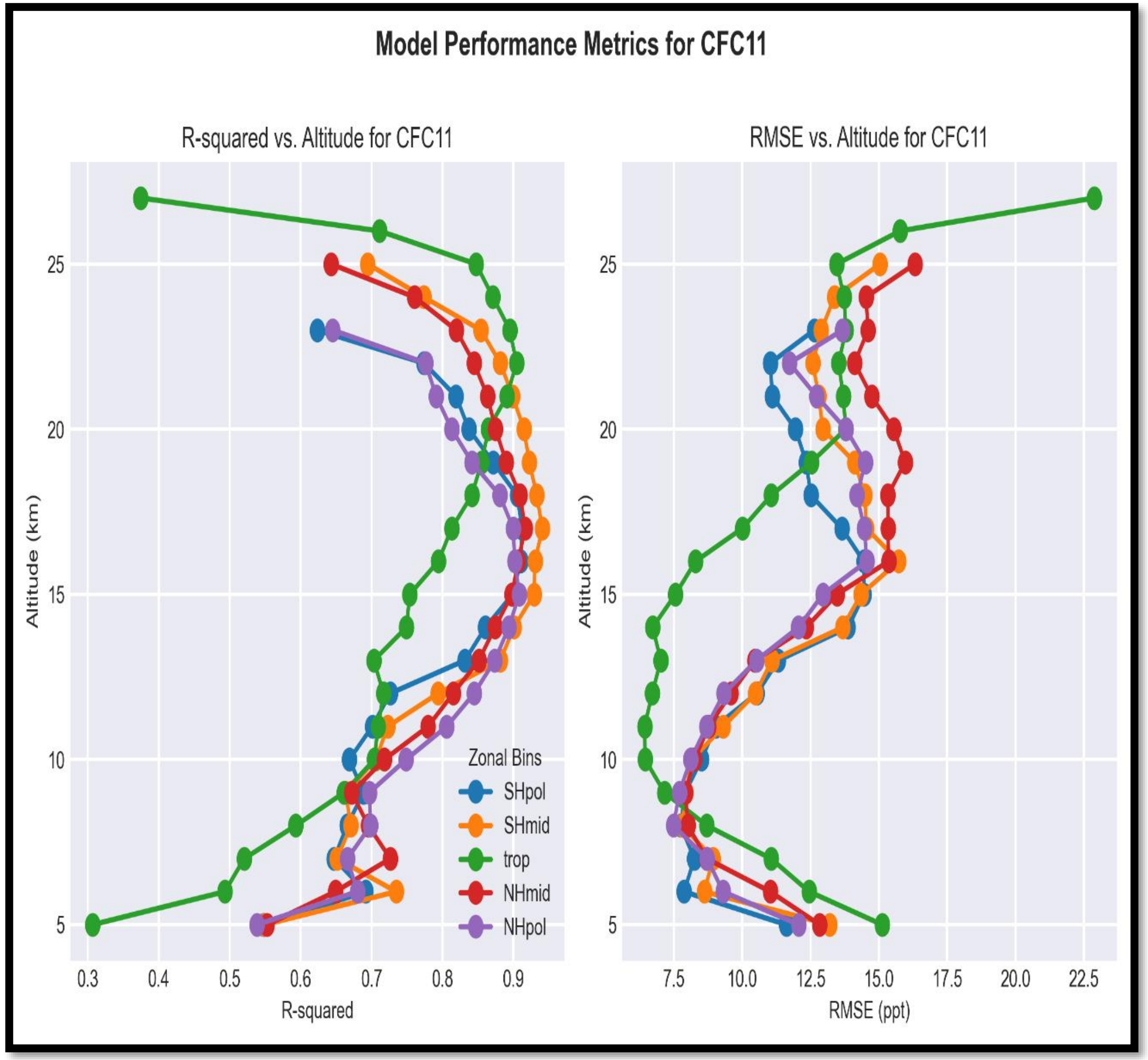

Predicted Radiative forcing from ozone depleting substances and greenhouse gases. Rigby et al., 2014

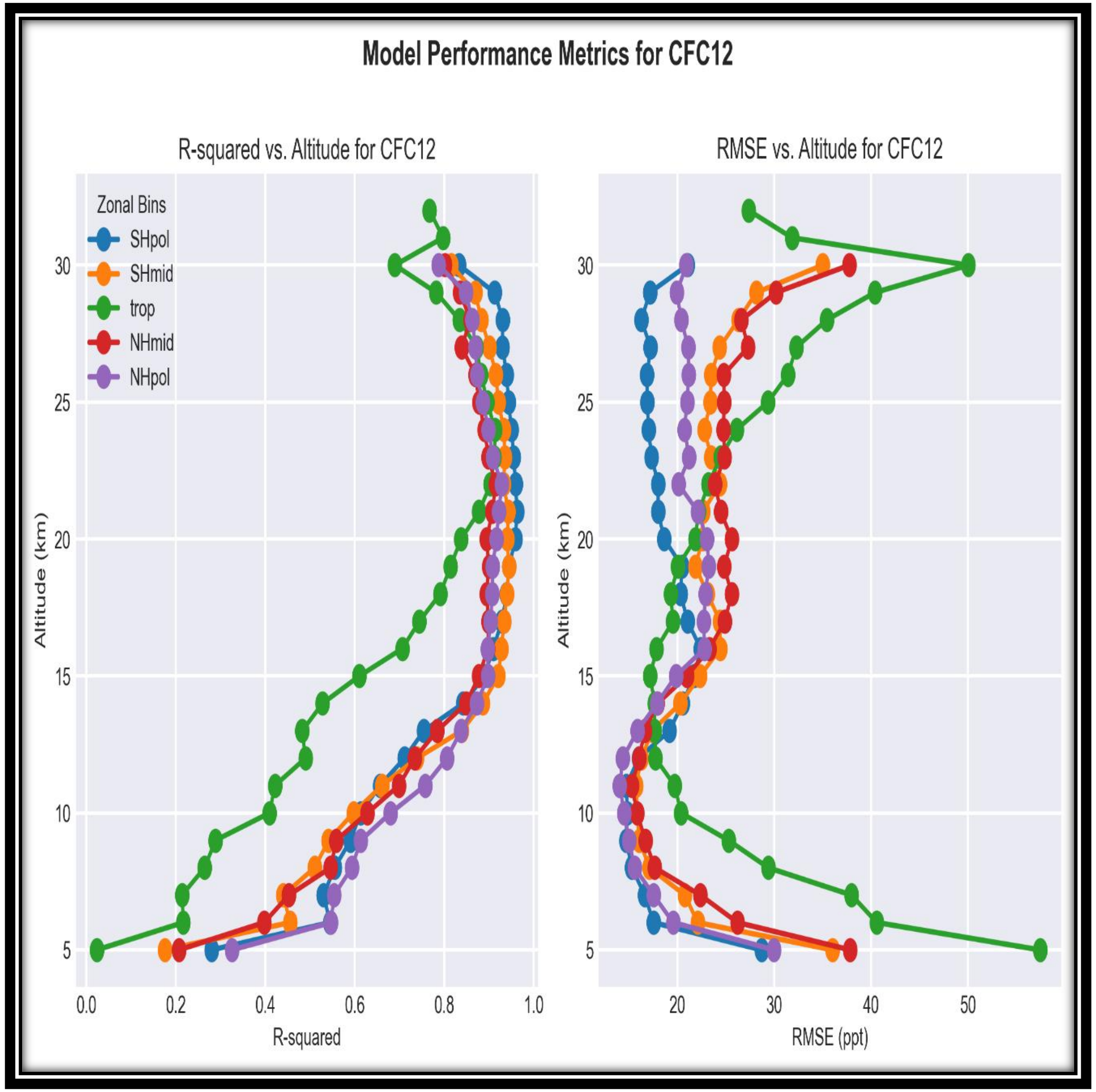
- Main objective is to improve our understanding about sources/sinks/radiative forcing from Halocarbons and GHGs
- We need good quality profile data to improve those estimates

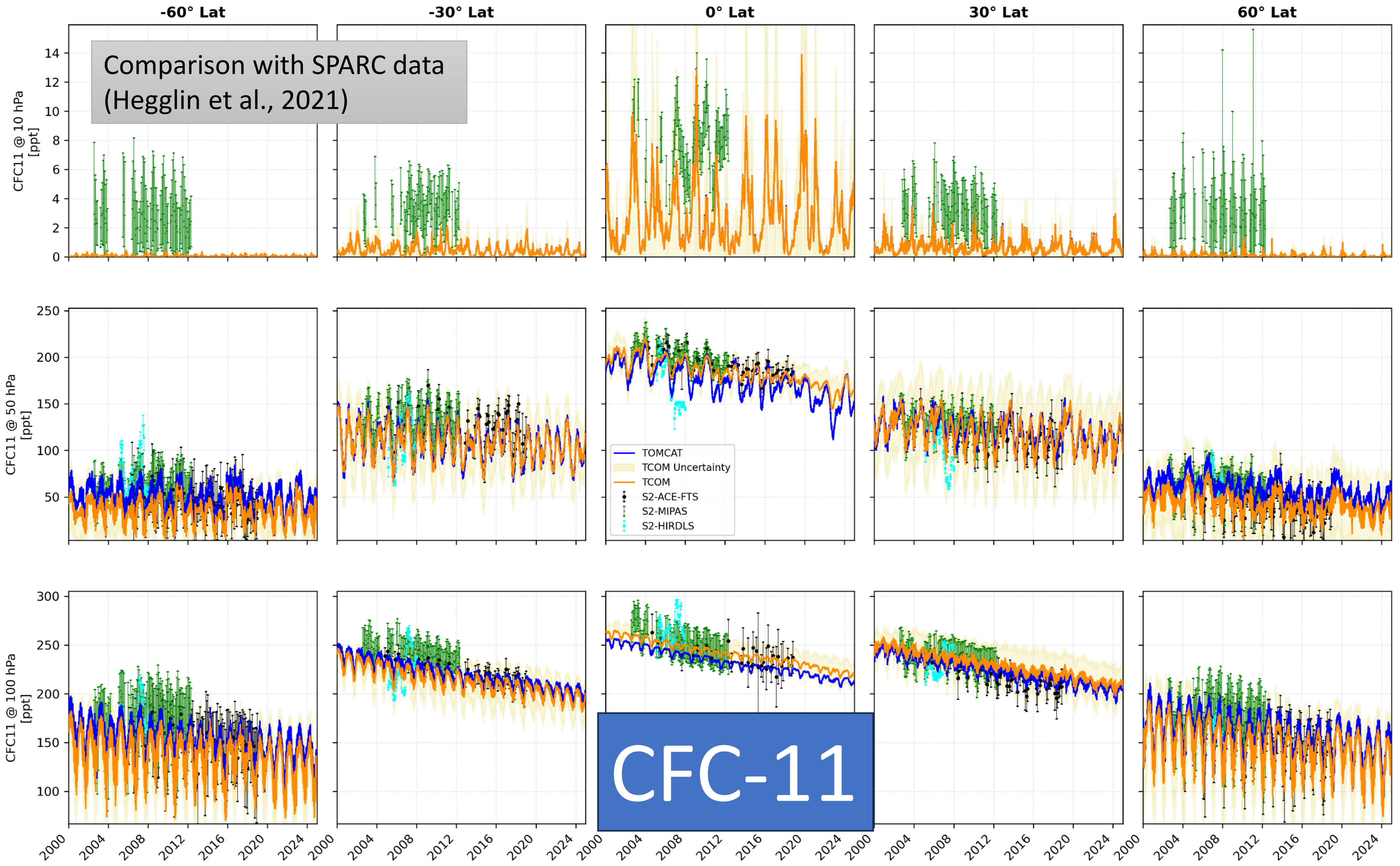
# TCOM- TCOMCAT CTM and Occultation Measurement Based Data



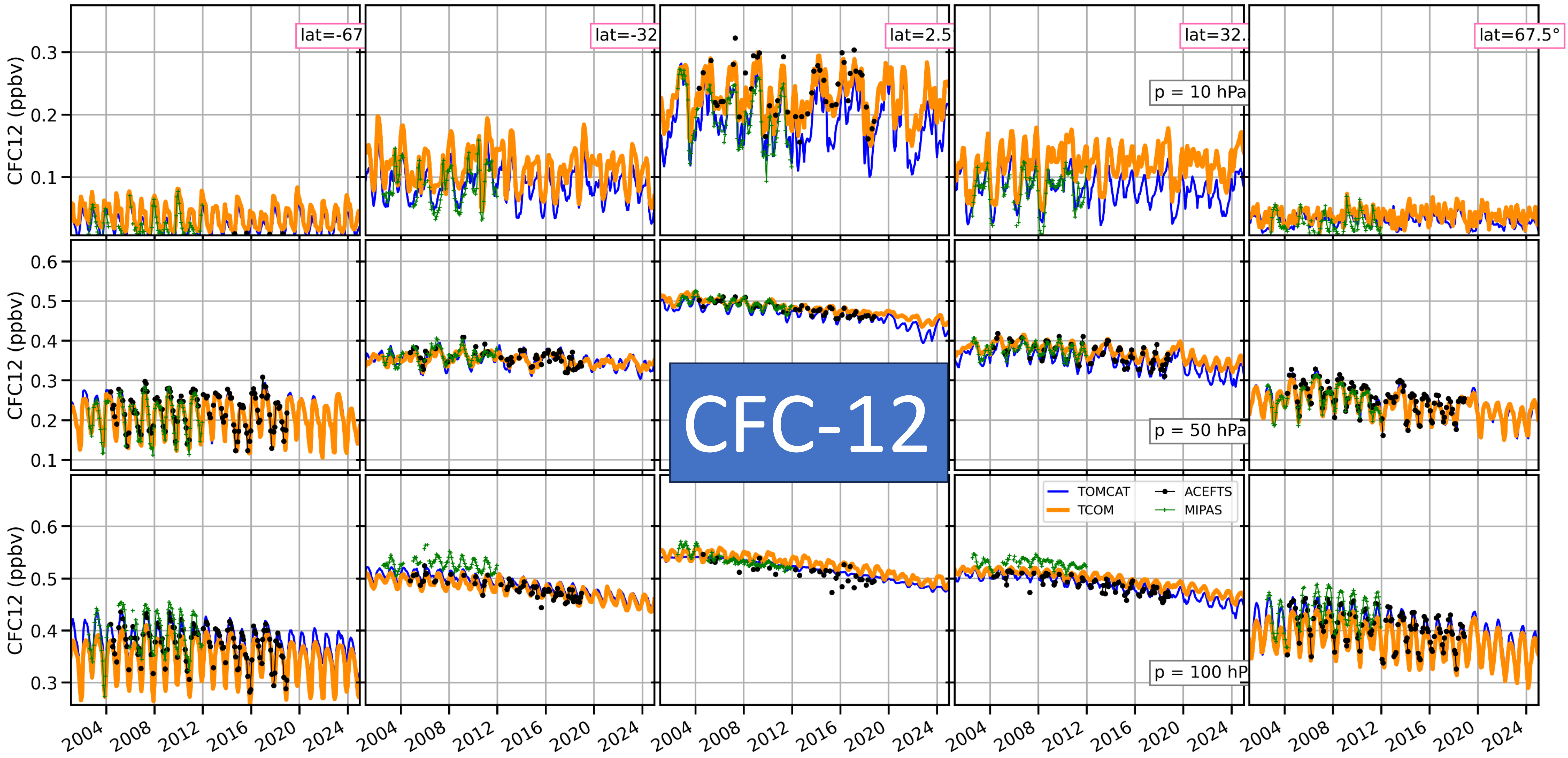
# CFC11 - XGBoost Performance for all 5 latitude bins





Comparison with SPARC data
(Hegglin et al., 2021)
CFC-11
-60° Lat
-30° Lat
0° Lat
30° Lat
60° Lat
CFC11 @ 10 hPa [ppt]
CFC11 @ 50 hPa [ppt]
CFC11 @ 100 hPa [ppt]
TOMCAT
TCOM Uncertainty
TCOM
S2-ACE-FTS
S2-MIPAS
S2-HIRDLS
2000
2004
2008
2012
2016
2020
2024

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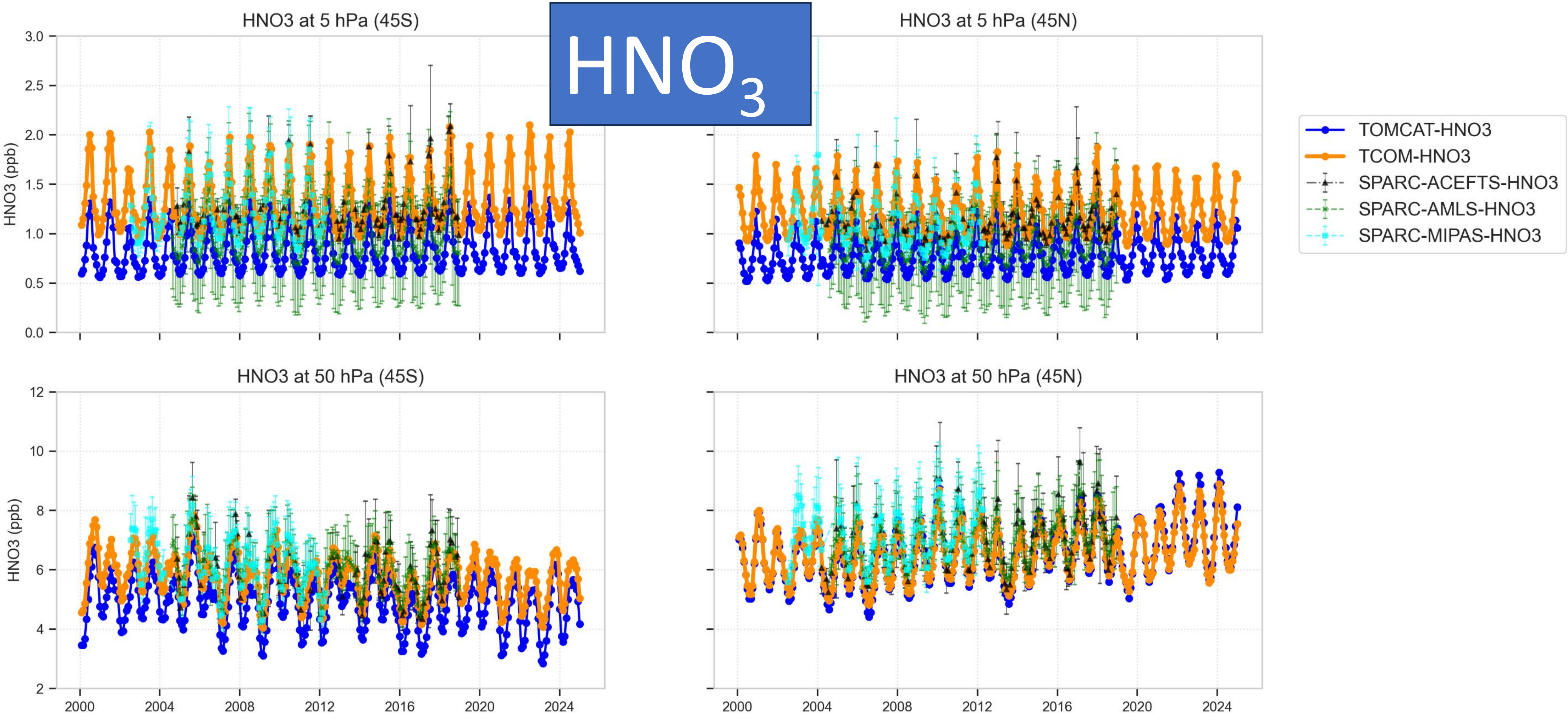
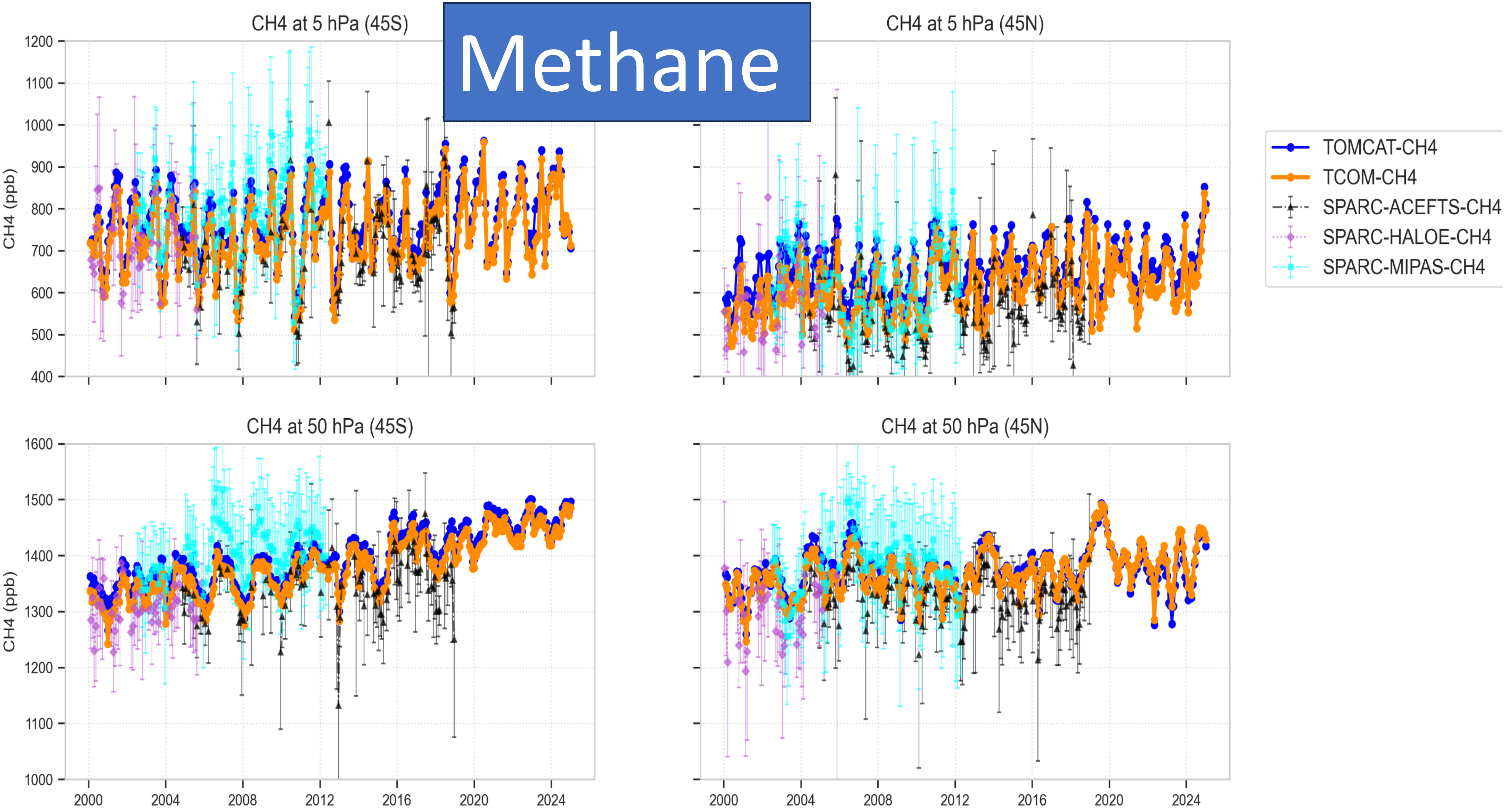
# $HNO_3$



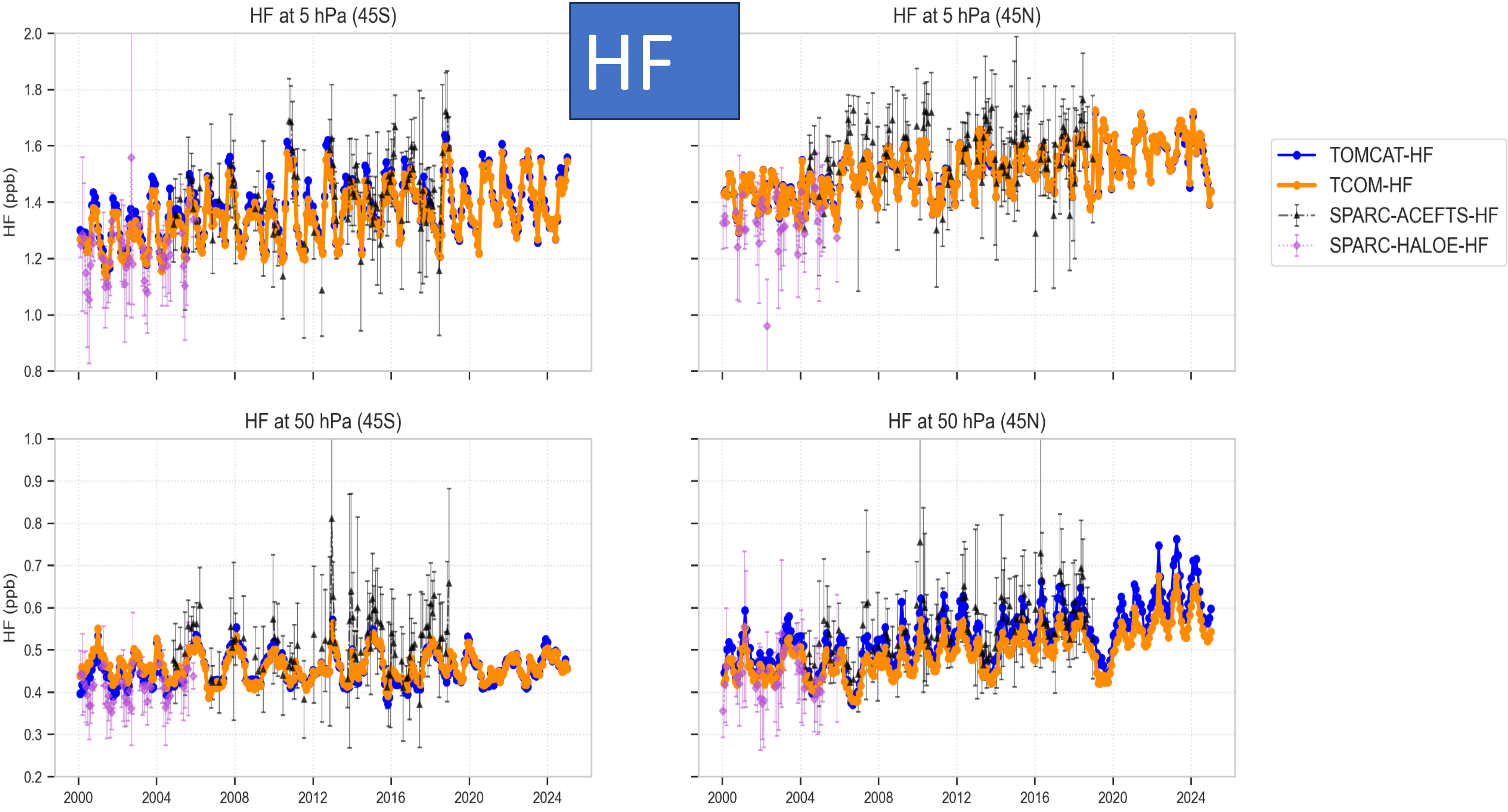
HNO3 at 5 hPa (45S)
HNO3 at 5 hPa (45N)
HNO3 at 50 hPa (45S)
HNO3 at 50 hPa (45N)
HNO3 (ppb)
TOMCAT-HNO3
TCOM-HNO3
SPARC-ACEFTS-HNO3
SPARC-AMLS-HNO3
SPARC-MIPAS-HNO3
2000
2004
2008
2012
2016
2020
2024

# Methane



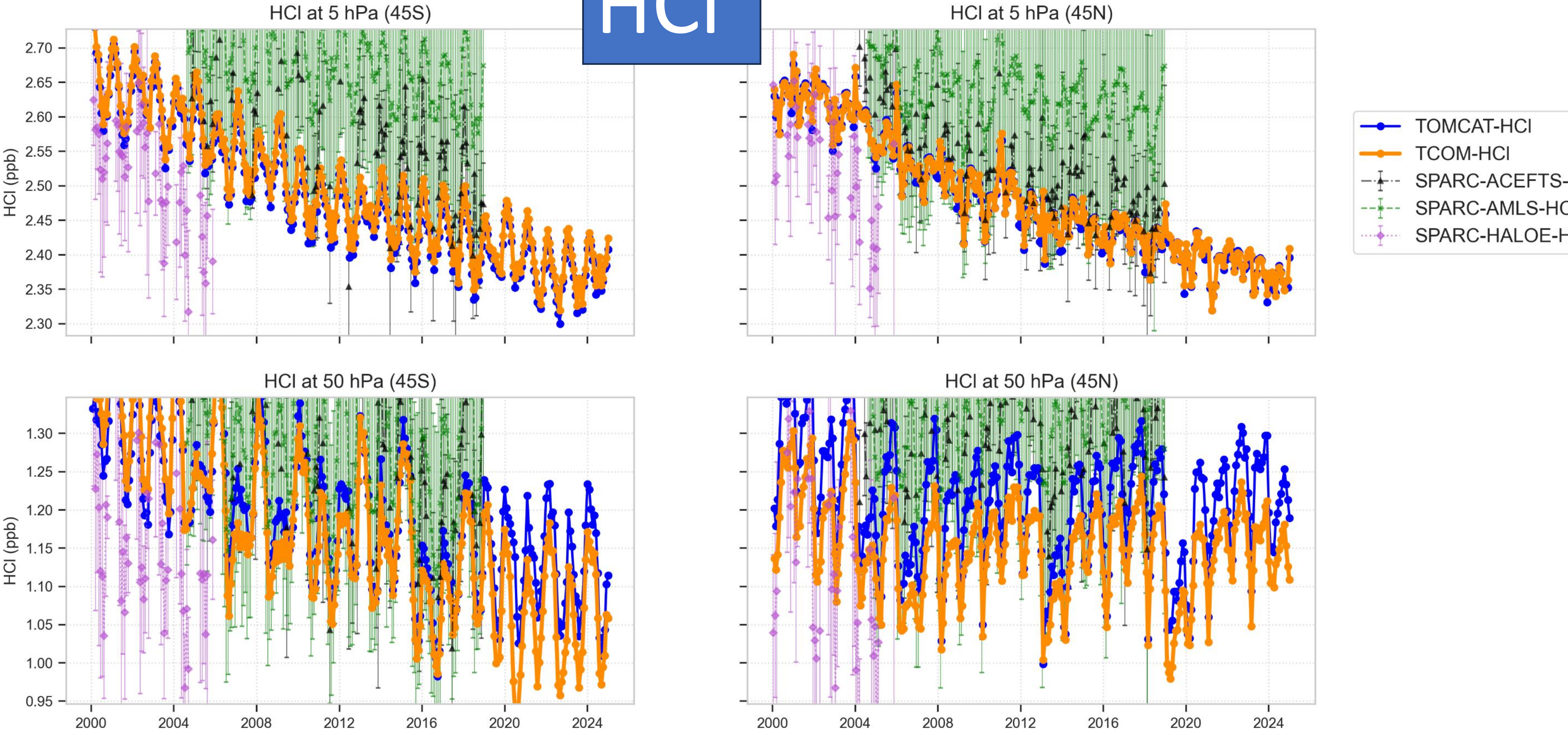
CH4 at 5 hPa (45S)
CH4 at 5 hPa (45N)
CH4 at 50 hPa (45S)
CH4 at 50 hPa (45N)
CH4 (ppb)
CH4 (ppb)
1200
1100
1000
900
800
700
600
500
400
1600
1500
1400
1300
1200
1100
1000
2000
2004
2008
2012
2016
2020
2024
TOMCAT-CH4
TCOM-CH4
SPARC-ACEFTS-CH4
SPARC-HALOE-CH4
SPARC-MIPAS-CH4

# HF



HF at 5 hPa (45S)
HF at 5 hPa (45N)
HF at 50 hPa (45S)
HF at 50 hPa (45N)
HF (ppb)
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.9
0.7
0.6
0.5
0.4
0.3
0.2
2000
2004
2008
2012
2016
2020
2024
TOMCAT-HF
TCOM-HF
SPARC-ACEFTS-HF
SPARC-HALOE-HF

# HCl



HCl at 5 hPa (45S)
HCl at 5 hPa (45N)
HCl at 50 hPa (45S)
HCl at 50 hPa (45N)
HCl (ppb)
2.70
2.65
2.60
2.55
2.50
2.45
2.40
2.35
2.30
1.30
1.25
1.20
1.15
1.10
1.05
1.00
0.95
2000
2004
2008
2012
2016
2020
2024
TOMCAT-HCl
TCOM-HCl
SPARC-ACEFTS-
SPARC-AMLS-HC
SPARC-HALOE-H

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# Methodology

- ACE observations are divided in 5 latitude bins (two polar, two mid-lats and tropics)

- Model output is sampled at ACE-measurement collocations
- Model output fields are interpolated at ACE vertical grids (1 km resolution)

- Target (y) – ACE CFC11 or CFC12 measurements at given level and latitude bins

- Feature Matrix (or X) – TOMCAT tracers such as measurement errors, $O_3$, $CH_4$, $HNO_3$, $N_2O$, $NO_2$, CFC11, CFC12, CFC113, ….., PV, temp, latitude, measurement time (total 26)

# Micro-windows used to retrieve CFC11 & CFC12

**Table 28: Microwindow list for $CCl_3F$ (CFC-11)**

| Centre Frequency (cm⁻¹) | Microwindow Width (cm⁻¹) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 829.03 [1] | 0.50 | 5-6 | 23-28 |
| 832.50* | 5.00 | 5 | 23-28 |
| 837.00* | 1.80 | 5 | 23-28 |
| 838.55* | 0.70 | 5 | 23-28 |
| 845.25* | 5.50 | 5 | 23-28 |
| 849.50* | 3.00 | 8-11 | 23-28 |
| 851.50* | 1.00 | 7-10 | 23-28 |
| 854.50* | 3.00 | 11-14 | 23-28 |
| 857.00* | 2.00 | 5 | 23-28 |
| 861.50* | 3.00 | 5 | 23-28 |
| 1950.10 [2] | 0.35 | 5-8 | 21 |
| 2976.50 [3] | 2.00 | 7 | 20 |

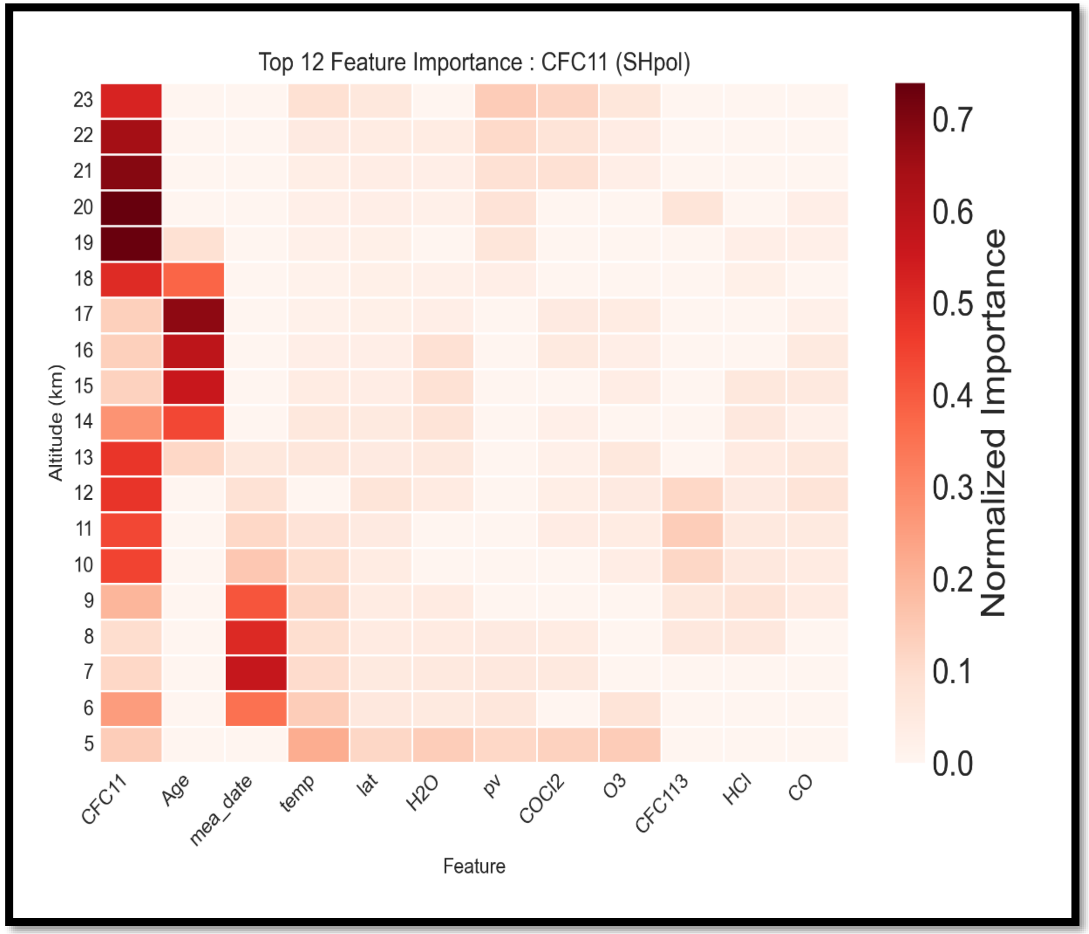
**Microwindows employ the same baseline parameters (scale and slope)*
*[1] Included to improve results for interferer $CHF_2Cl$ (HCFC-22)*
*[2] Included to improve results for interferer $H_2O$*
*[3] Included to improve results for interferer $C_2H_6$*

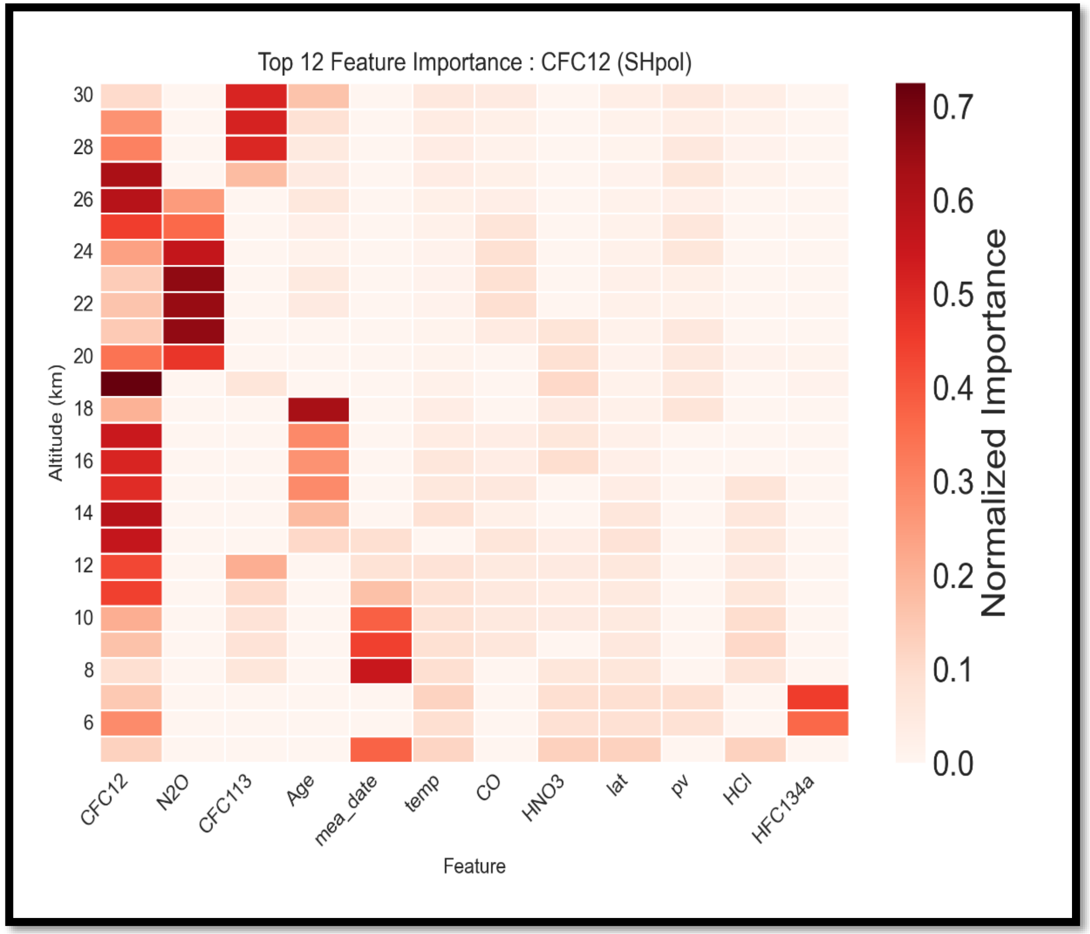
**Table 30: Microwindow list for $CCl_2F_2$ (CFC-12)**

| Centre Frequency (cm⁻¹) | Microwindow Width (cm⁻¹) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 919.70* | 1.60 | 5 | 23-28 |
| 919.70 | 1.60 | 23-28 | 26-30 |
| 921.10* | 1.64 | 5-10 | 23-28 |
| 921.10 | 1.64 | 23-28 | 30-35 |
| 921.70* | 0.24 | 5-6 | 23-28 |
| 923.28 | 0.80 | 23-28 | 30-35 |
| 923.54* | 1.32 | 5 | 23-28 |
| 926.30* | 1.00 | 5 | 23-28 |
| 927.89* | 0.98 | 5 | 23-28 |
| 930.00* | 1.20 | 5 | 23-28 |
| 931.70* | 1.00 | 5 | 23-28 |
| 933.90* | 1.00 | 5 | 23-28 |
| 935.90* | 0.70 | 5 | 23-28 |
| 939.50* | 0.60 | 5 | 23-28 |
| 941.66* | 0.40 | 5 | 23-28 |
| 1158.15 | 1.10 | 15 | 26-30 |
| 1161.07 | 1.85 | 15 | 30-35 |
| 1950.10 [1] | 0.35 | 5-8 | 21 |
| 2521.22 [2] | 0.35 | 8-10 | 20 |

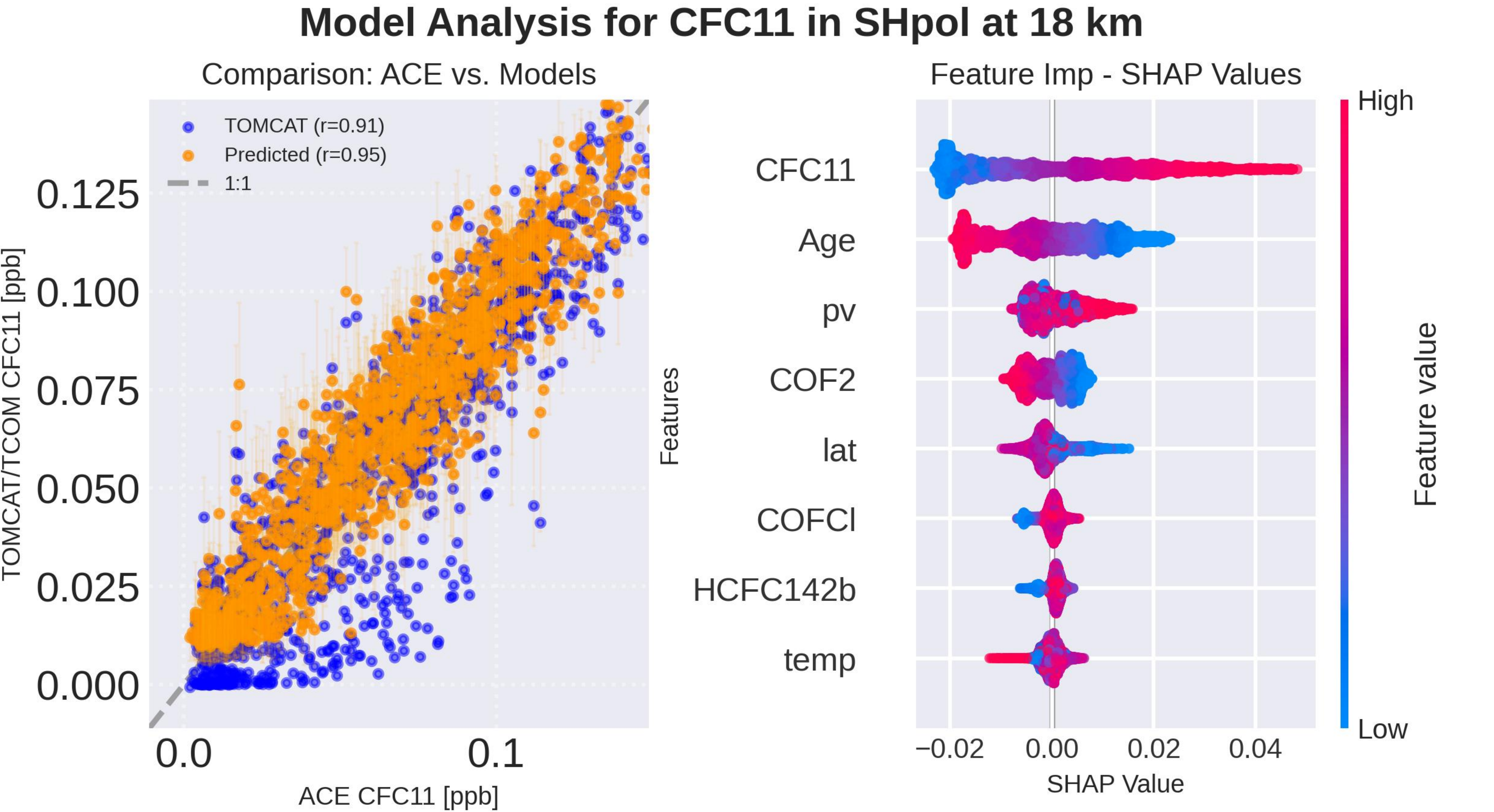
**Microwindows employ the same baseline parameters (scale and slope)*
*[1] Included to improve results for interferer $H_2O$*
*[2] Included to improve results for interferer $N_2O$*

# Feature Importances - SHpol




- Basic chemistry/transport is good
- Some transport-related variables are also important


Model Analysis for CFC11 in SHpol at 18 km
Comparison: ACE vs. Models
TOMCAT (r=0.91)
Predicted (r=0.95)
1:1
TOMCAT/TCOM CFC11 [ppb]
0.125
0.100
0.075
0.050
0.025
0.000
0.0
0.1
ACE CFC11 [ppb]
Feature Imp - SHAP Values
Features
CFC11
Age
pv
COF2
lat
COFCl
HCFC142b
temp
−0.02
0.00
0.02
0.04
SHAP Value
High
Low
Feature value

# Summary and Outlook

- We use machine learning algorithm to estimate biases in chemical model output w.r.t. occultation instrument measurements.
- Bias-corrected, daily, global zonal-mean gap-free data at 1 km vertical resolution (5-50 km) are publicly available for 2000-2023.
- XGBoost (and other regression model) performance or correction estimates are better for high latitudes but not significant in the tropics (ACE viewing geometry means fewer measurements in the tropics)
- Updates of following data sets are released (V2.0)
  - $O_3$ (v2.0) https://zenodo.org/records/18199586
  - $H_2O$ (v2.0) https://zenodo.org/records/18199962
  - $CH_4$ (v2.0) https://zenodo.org/records/18197333
  - $N_2O$ (v2.0) https://zenodo.org/records/18197444
  - CFC-11 (v2.0) https://zenodo.org/records/18145730
  - CFC-12 (v2.0) https://zenodo.org/records/18147392
  - HF (v2.0) https://zenodo.org/records/18184779
  - HCl (v2.0) https://zenodo.org/records/18184430
  - $HNO_3$ (v2.0) https://zenodo.org/records/18199002
  - $COF_2$ (v2.0) https://zenodo.org/records/18201786

**https://tomcat.leeds.ac.uk/tomcat/tcom/**

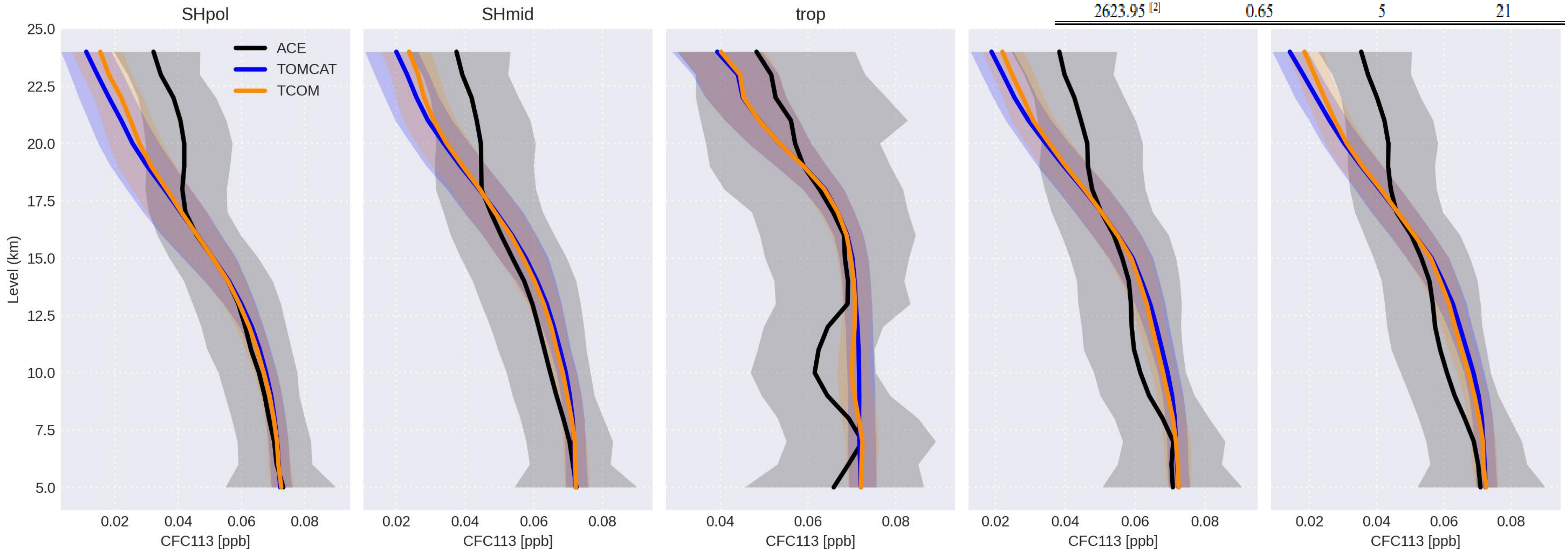
# Extra slides

# CFC113

Table 90: Microwindow list for $C_2Cl_3F_3$ (CFC-113)

| Centre Frequency ($cm^{-1}$) | Microwindow Width ($cm^{-1}$) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 1094.10* | 3.80 | 5 | 25 |
| 1104.34* | 2.60 | 5 | 25 |
| 1109.51* | 1.10 | 5 | 25 |
| 1113.52* | 1.80 | 5 | 25 |
| 1115.63* | 1.50 | 5 | 25 |
| 1118.99* | 1.54 | 5 | 25 |
| 1122.75* | 1.10 | 5 | 25 |
| 1950.10 [1] | 0.35 | 6-7 | 21 |
| 2623.95 [2] | 0.65 | 5 | 21 |

**Median Vertical Profiles by Zonal Bin for CFC113**

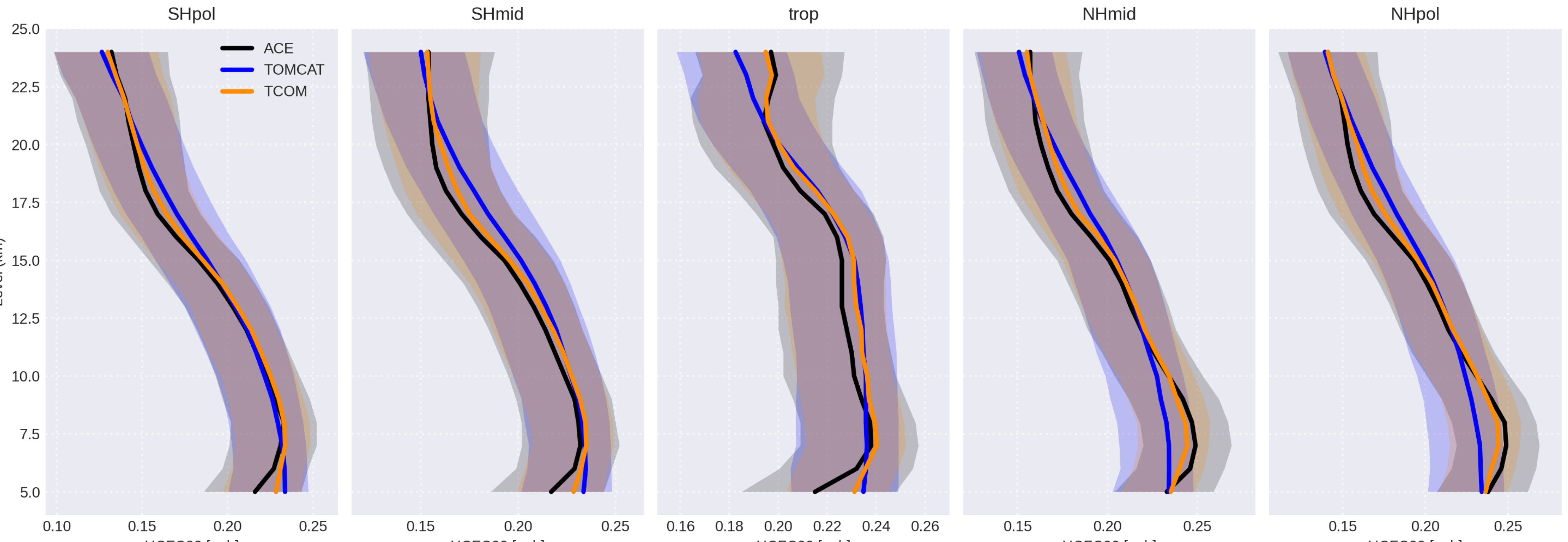


# COF2

**Table 88: Microwindow list for $CHF_2Cl$ (HCFC-22)**

| Centre Frequency (cm$^{-1}$) | Microwindow Width (cm$^{-1}$) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 802.89 [1] | 2.08 | 10 | 25 |
| 804.70* | 1.20 | 5 | 25 |
| 809.26* | 1.20 | 5 | 25 |
| 818.00* | 3.00 | 5 | 25 |
| 820.85* | 0.50 | 5 | 25 |
| 829.00* | 0.40 | 5 | 25 |
| 1950.10 [2] | 0.35 | 6-7 | 20 |
| 2004.10 [3] | 0.60 | 7 | 18 |
| 2013.55 [3] | 0.40 | 12 | 22 |
| 2620.81 [4] | 0.45 | 8 | 22 |
| 2976.80 [5] | 0.40 | 7-10 | 20 |

**Median Vertical Profiles by Zonal Bin for HCFC22**

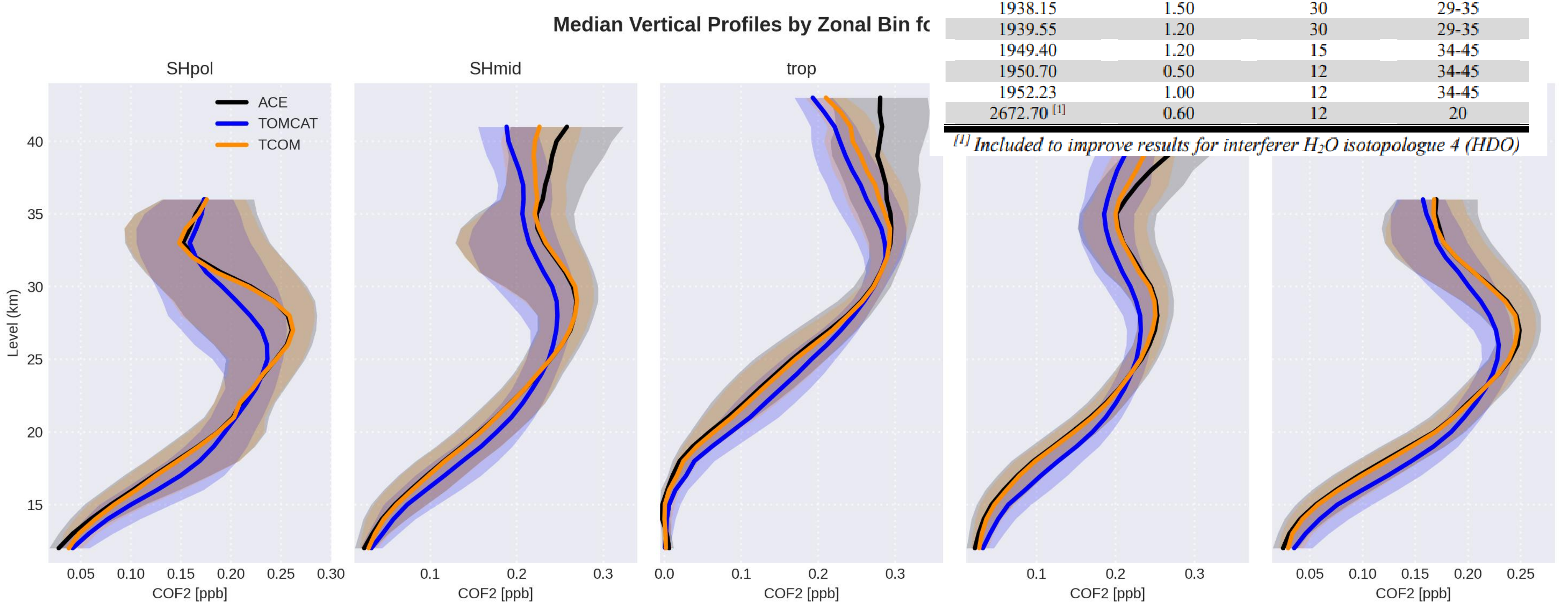


# COF2

**Table 36: Microwindow list for $COF_2$**

| Centre Frequency ($cm^{-1}$) | Microwindow Width ($cm^{-1}$) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 1234.70 | 1.40 | 12 | 34-45 |
| 1236.90 | 1.40 | 25 | 34-45 |
| 1238.00 | 0.80 | 15 | 34-45 |
| 1239.90 | 1.00 | 15 | 34-45 |
| 1930.60 | 1.40 | 12-15 | 34-45 |
| 1936.48 | 0.65 | 12 | 34-45 |
| 1938.15 | 1.50 | 30 | 29-35 |
| 1939.55 | 1.20 | 30 | 29-35 |
| 1949.40 | 1.20 | 15 | 34-45 |
| 1950.70 | 0.50 | 12 | 34-45 |
| 1952.23 | 1.00 | 12 | 34-45 |
| 2672.70 [1] | 0.60 | 12 | 20 |

*[1] Included to improve results for interferer $H_2O$ isotopologue 4 (HDO)*

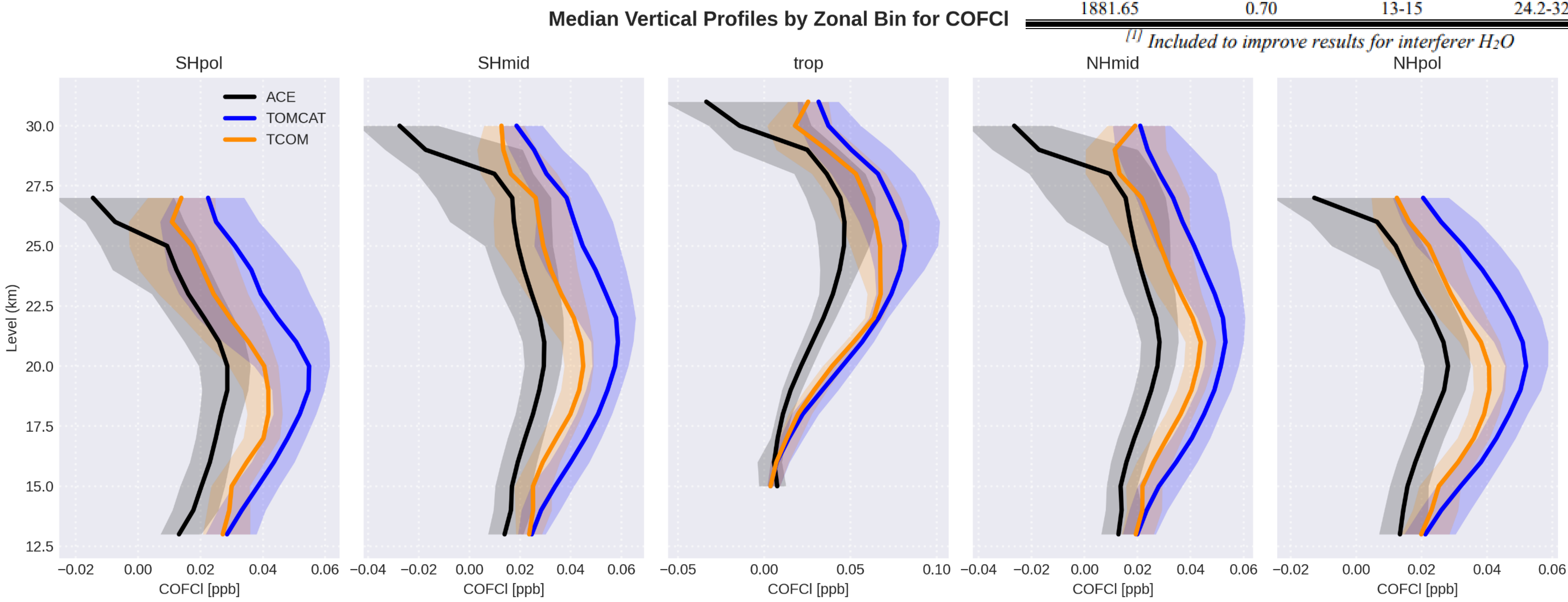


# COClF

**Table 66: Microwindow list for COClF**

| Centre Frequency ($cm^{-1}$) | Microwindow Width ($cm^{-1}$) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 1553.00 [1] | 0.35 | 13-15 | 24.2-32 |
| 1862.60 | 1.00 | 13-15 | 24.2-32 |
| 1864.40 | 0.60 | 13-15 | 24.2-32 |
| 1865.40 | 0.85 | 13-15 | 24.2-32 |
| 1866.82 | 0.35 | 13-15 | 24.2-32 |
| 1867.38 | 0.30 | 13-15 | 24.2-32 |
| 1869.98 | 0.35 | 13-15 | 24.2-32 |
| 1870.50 | 0.30 | 13-15 | 24.2-32 |
| 1881.65 | 0.70 | 13-15 | 24.2-32 |

*[1] Included to improve results for interferer $H_2O$*

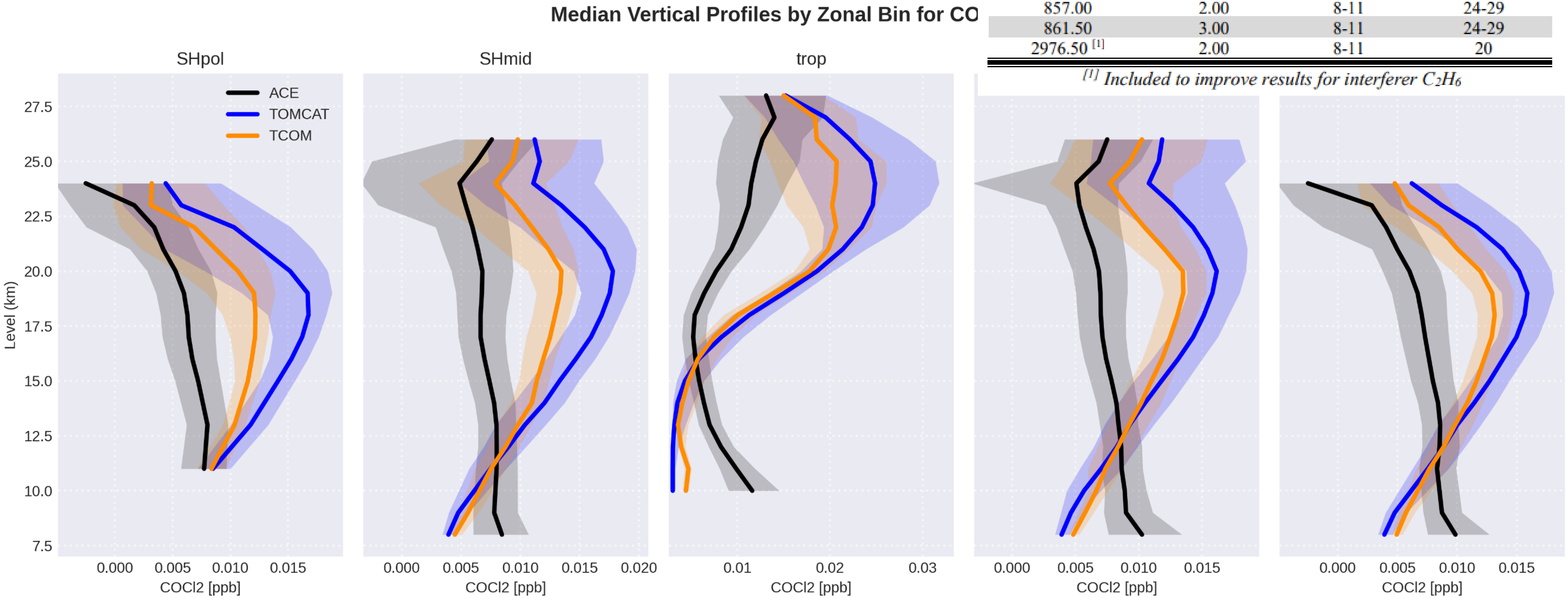


# COCl2

**Table 60: Microwindow list for $COCl_2$**

| Centre Frequency ($cm^{-1}$) | Microwindow Width ($cm^{-1}$) | Lower Altitude (km) | Upper Altitude (km) |
|---|---|---|---|
| 829.03 | 0.50 | 8-11 | 24-29 |
| 832.50 | 0.50 | 8-11 | 24-29 |
| 837.00 | 1.80 | 8-11 | 24-29 |
| 838.55 | 0.70 | 8-11 | 24-29 |
| 845.25 | 5.50 | 8-11 | 24-29 |
| 849.00 | 2.00 | 8-11 | 24-29 |
| 851.00 | 2.00 | 8-11 | 24-49 |
| 854.50 | 3.00 | 11-14 | 24-29 |
| 857.00 | 2.00 | 8-11 | 24-29 |
| 861.50 | 3.00 | 8-11 | 24-29 |
| 2976.50 [1] | 2.00 | 8-11 | 20 |

*[1] Included to improve results for interferer $C_2H_6$*



Dependent Variable
(Response Variable)

Independent Variables
(Predictors)

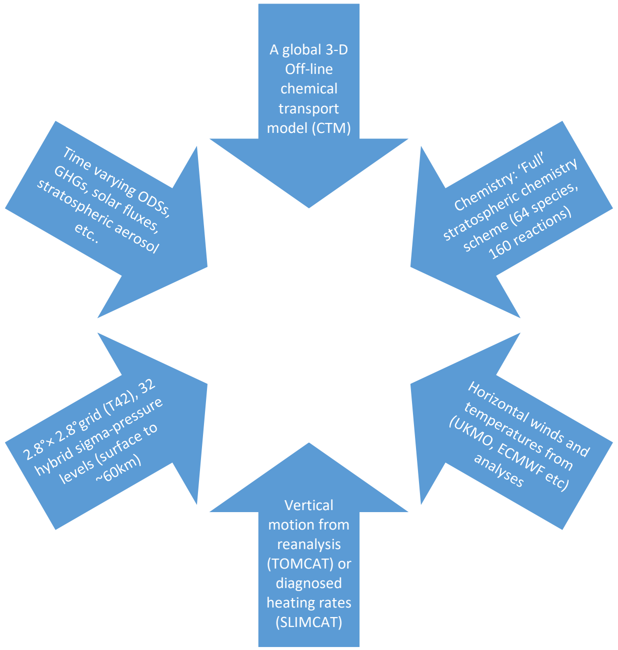
$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \cdots + \varepsilon$$

# Multivariate Regression

- Y - response → ***Target*** – difference between TOMCAT and ACE-FTS measurements
- X1, X2, X3 - predictors → ***Features*** → Variables that might explain observed biases
- Find optimum ***betas*** so that we can estimate differences for all the gridpoints in TOMCAT CTM

# TOMCAT – Chemical Transport Model

tomcat.leeds.ac.uk/tomcat




- **Model simulation (ERA5)** forced with ECMWF ERA5 reanalysis (2000 - 2025)

e.g. Chipperfield, 2006; Dhomse et al., 2015; 2016; 2019; Feng et al., 2011; 2021; Li et al., 2020; 2022…