climate change initiative

LONG-LIVED GREENHOUSE GAS
PRODUCTS PERFORMANCES

LOLIPOP WP 3300: Monitoring of stratospheric chlorine levels and their impacts on ozone recovery

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MONTREAL PROTOCOL

The Montreal Protocol (in force since 1987) and its amendments control the long-lived halogenated source gases. **Total chlorine in the stratosphere has been decreasing since the late 1990s due to the Montreal Protocol.**

Hydrofluorocarbons (HFCs) do not contain chlorine, so they do not deplete ozone directly. However they have been included in the Montreal Protocol because they are powerful greenhouse gases that contribute to climate change. **HF** is their reservoir species.

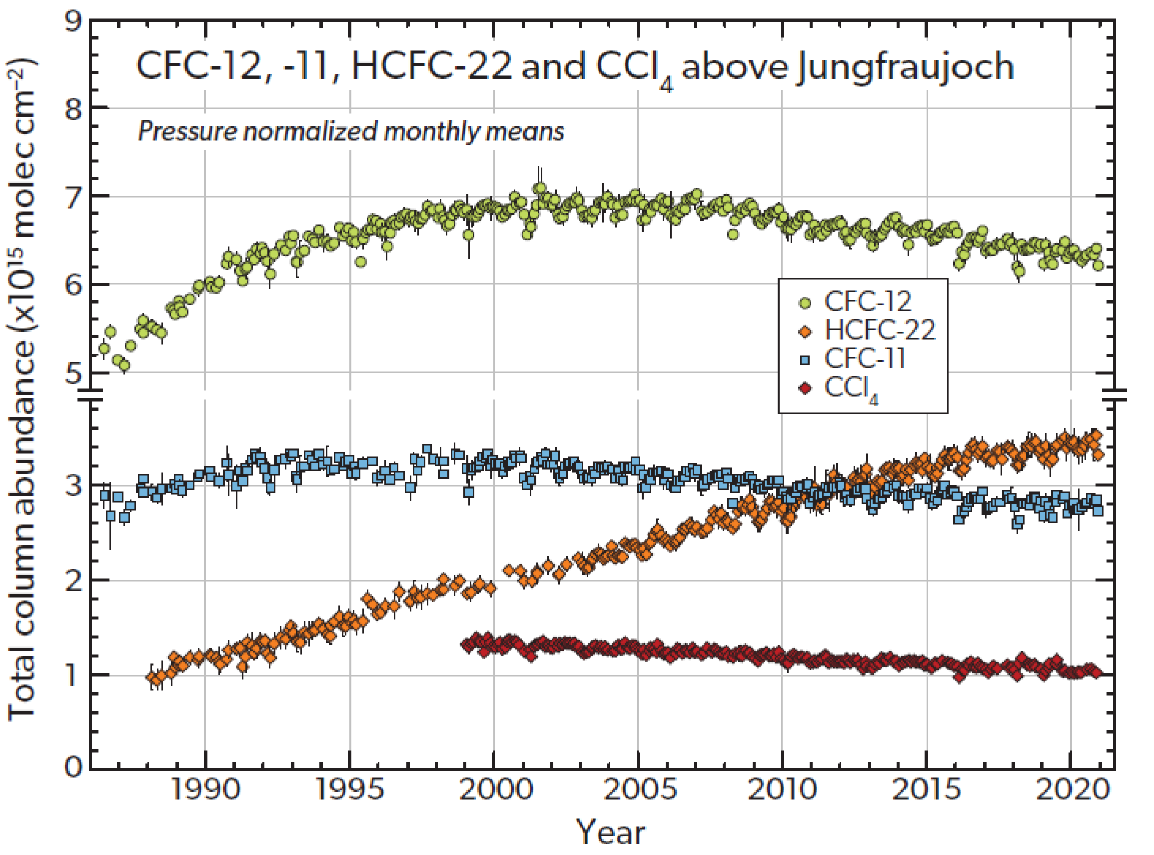


Figure. Monthly mean total vertical column abundance time series of CFC-12, CFC-11, HCFC-22, and CCl_4, derived from the long-term FTIR monitoring program conducted at the Jungfraujoch station, Switzerland (46.5° N), from 1986 to 2021 (updated from Zander et al., 2008; Gardiner et al., 2008; Rinsland et al., 2012; and Prignon et al., 2019). Note the discontinuity in the vertical scale.

WORK PACKAGE AIM

The aim of this work is to study over recent decades stratospheric chlorine levels linked to ozone depletion. To this end, we will use a combination of:

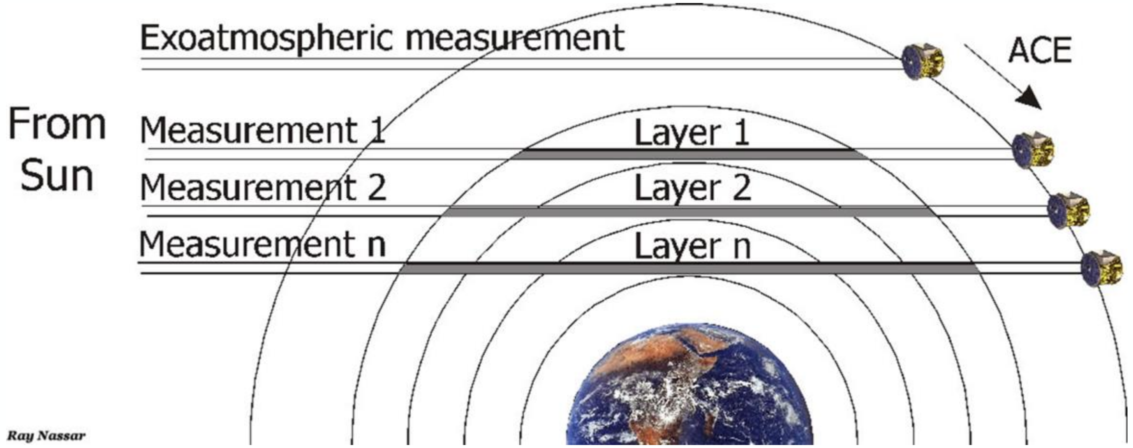
- Observational data, mainly ACE-FTS
- Chemical transport model (CTM) data (TOMCAT)
- A merged dataset using ML (TCOM)
    - Overcomes biases in CTMs (parameterisations) and gaps in ACE sampling

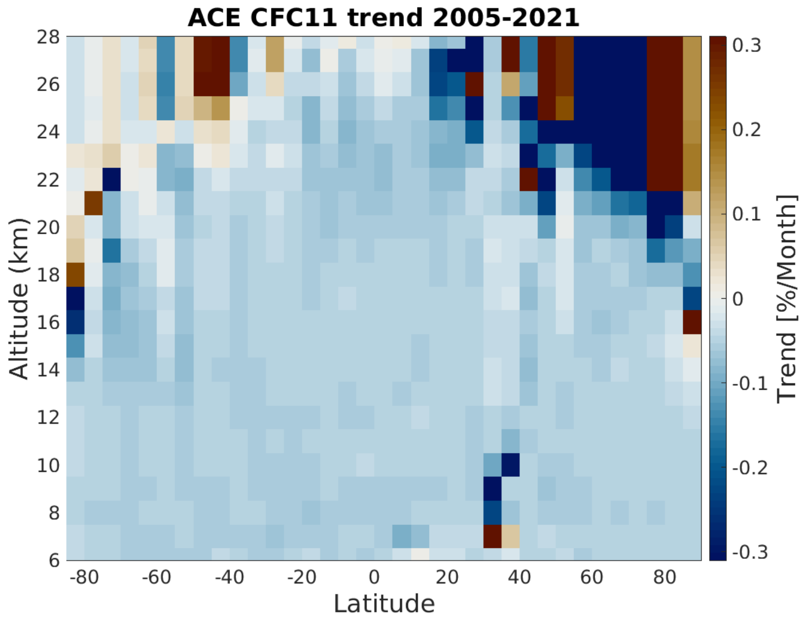
From these datasets we will:

- Determine **trends** for datasets relevant to the Montreal Protocol for different atmospheric regions over recent decades: *HCl and two source species CFC-11, CFC-12*.
- Estimate, through bespoke model simulation with constant Cl at peak values (the era of peak halogens in the mid-1990s), the changes in HCl trends.

ACE-FTS, **limb** instrument onboard the Canadian mission SCISAT:

1. Measuring since February 2004, over 40 atmospheric **trace gases**, particularly the regional ozone budget, as well as temperature and pressure.
2. Spectral resolution of 0.02 cm^{-1} in 750-4400 cm^{-1} region.
3. High vertical resolution of typically 3–4 km (vertical sampling of ~2-6 km).
4. Main mission aim: **monitor** and **analyse** the chemical processes that control the ozone distribution in the stratosphere.

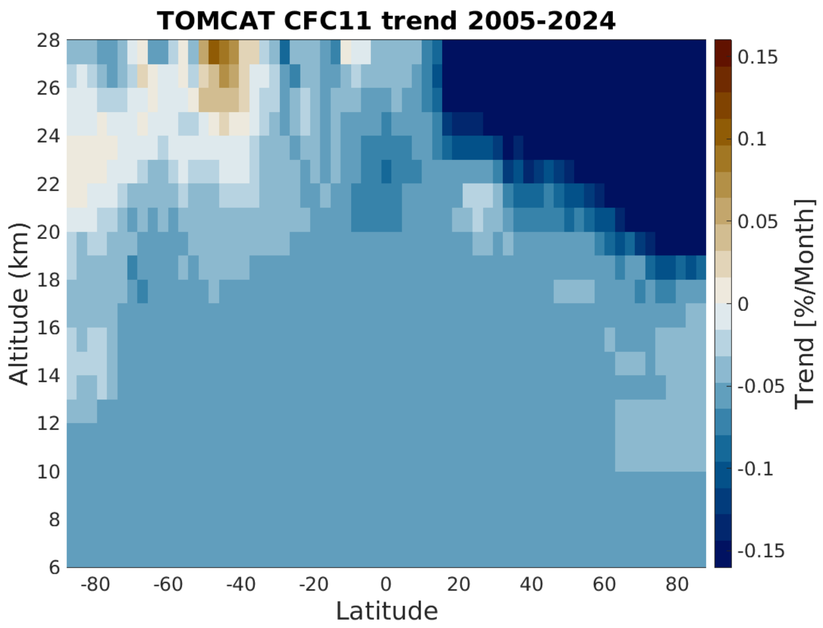




TOMCAT

Off-line 3-D chemical transport **model** (www.see.leeds.ac.uk/tomcat)

- Vertical coordinate (Hybrid σ-p levels). Variable resolution.
- Horizontal winds and temperatures specified from analyses (e.g. ERA 5.1).
- Vertical winds from analysed divergence.
- Advection: Prather [1986] second-order moments
- Physics: Tiedtke [1989] convection scheme.
  Holtslag and Boville [1993] *or* Louis [1979] PBL schemes.
- Chemistry: Stratosphere: O_x, NO_y, HO_x, Cl_y, Br_y, Fy, CHO_x
  Source: CH_4, N_2O, CFCs, HCFCs, HFCs etc.
- Aerosols: Specified sulfate surface area.
  Polar stratospheric clouds



TCOM

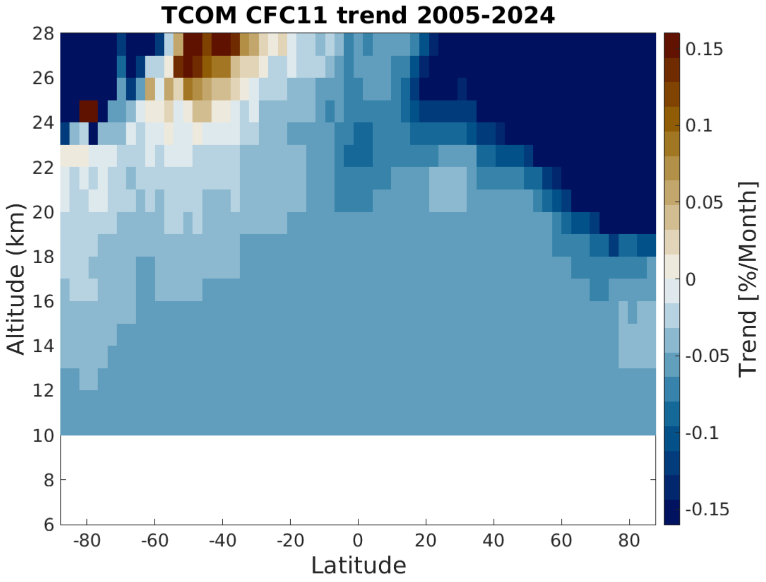
TCOM is a **gap-free dataset** derived from satellite data, e.g. ACE and HALOE, and TOMCAT CTM simulations using a **supervised machine learning (ML) scheme**.

- 70% training, 30% testing (2019-2021 – evaluation points).

XGBoost is a **decision-tree-based** ensemble ML algorithm that uses a gradient boosting framework. Evolution of XGBoost Algorithm from Decision Trees

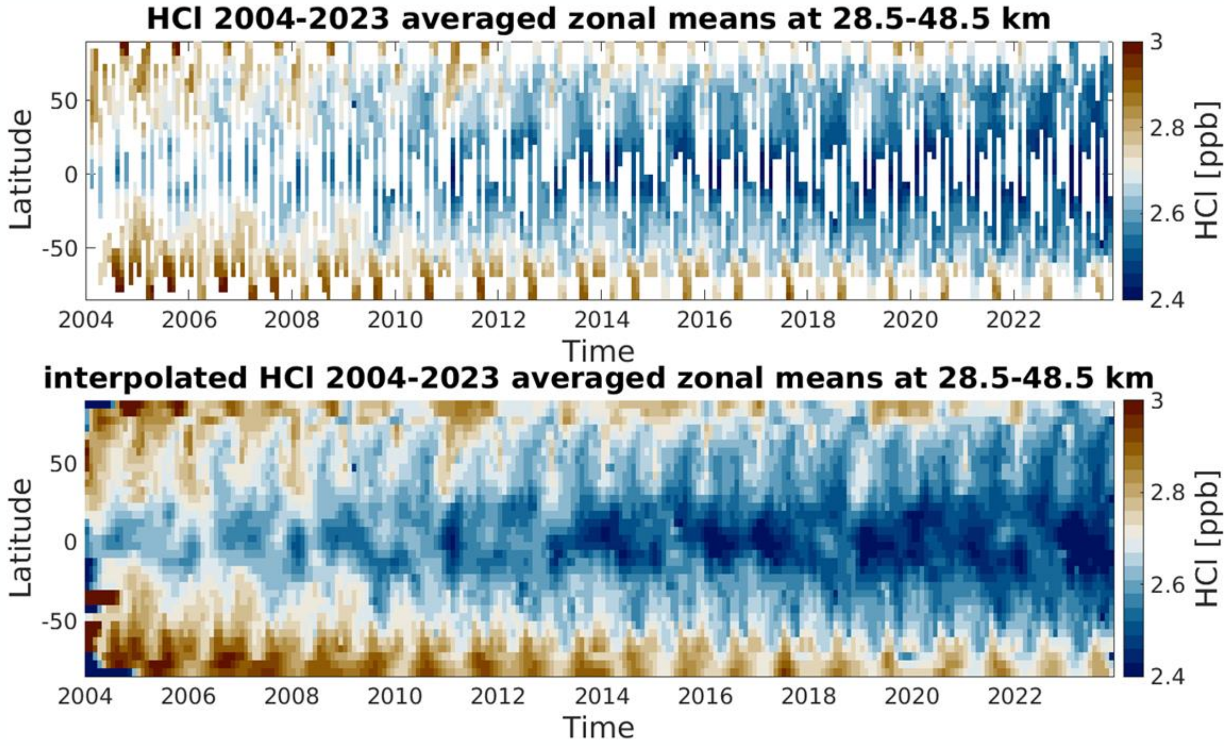
Grouping data → **Calculating difference** → **Model training** → **Daily biased profiles** → **CTM output**

- **Grouping data**: Group measurements into 5 latitude bins (poles, mid-latitudes and tropics)
- **Calculating difference**: Differences between model and satellite data at each 1 km vertical level
- **Model training**: Train XGBoost regression model for each level and latitude bin
- **Daily biased profiles**: Biases in daily profiles are estimated using XGBoost
- **CTM output**: XGBoost estimated biases are added back onto CTM output



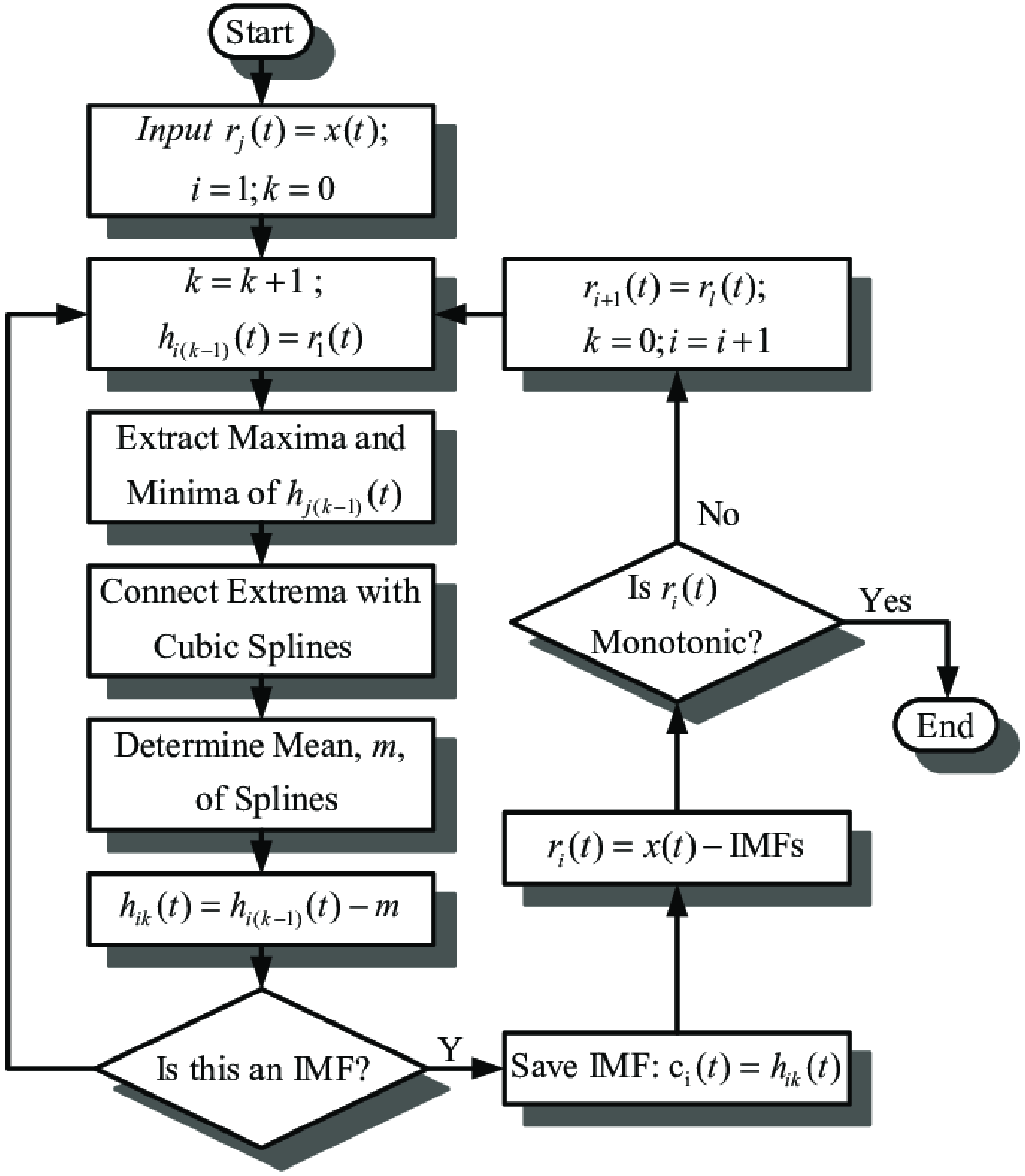
TREND CALCULATIONS

- **ACE** data have been **temporally interpolated** using a cubic spline to fill the gaps
- **Deseasonalization** of the signals using **empirical mode decomposition (EMD)**
  - EMD to decompose the signal into **intrinsic mode functions (IMFs)**
  - Select the IMFs with **seasonal** and **annual** period satisfying 3 months<period<14 months, $IMFs_{seasonal}$



- Trends calculated as the **linear regression** of deseasonalized signal (original signal-$IMFs_{seasonal}$)

EMPIRICAL MODE DECOMPOSITION (EMD)



Sifting Process

1. identify all extrema of x(t)
2. envelope of local minima/maxima $e_{min}(t)/e_{max}(t)$
3. compute the mean $m(t)=(e_{min}(t)+e_{max}(t))/2$
4. extract the detail d(t) = x(t) − m(t)
5. iterate on the residual m(t)

Intrinsic Mode Functions (IMFs)

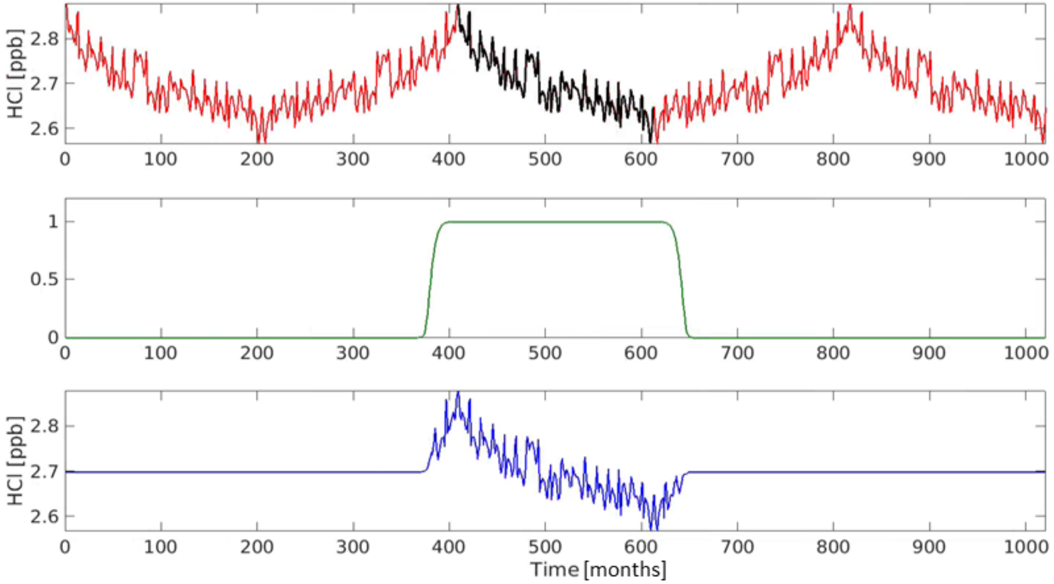
An IMF should satisfy two basic conditions:

- $N_{max}(N_{min}) = N_0(+1)$
- At any point, the mean value of the e_{max} and e_{min} should be zero

If not satisfied the shifting process (1-4) is repeated on $d_1(t)$.

BOUNDARIES

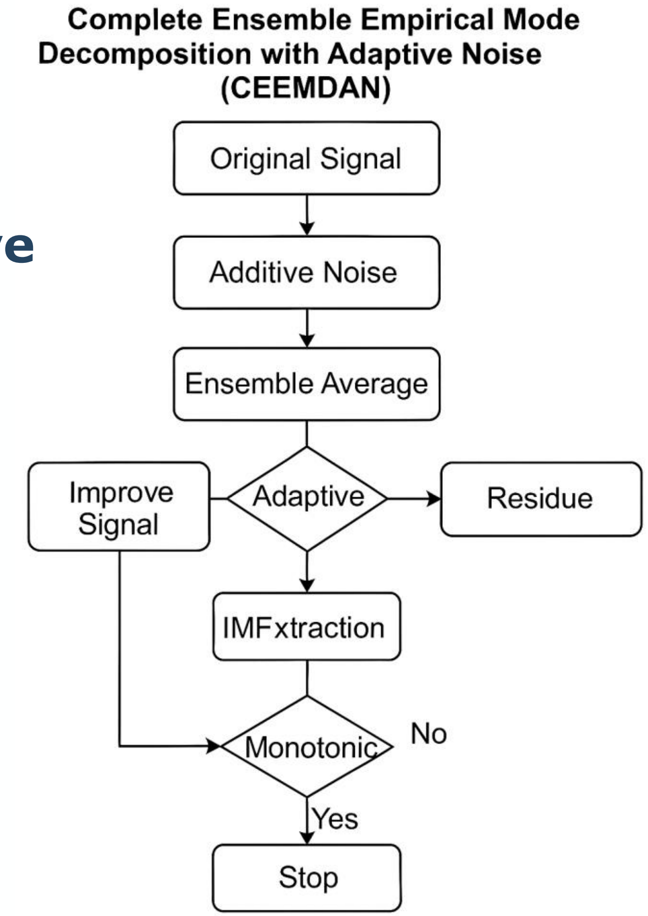
i. $x' = x(t) - x_m$
ii. Symmetric **extension** of x′ outside the boundaries (5 times)
iii. Super-Gaussian function for **windowing** the extended signal
iv. Addition of the mean to the result



MODE MIXING

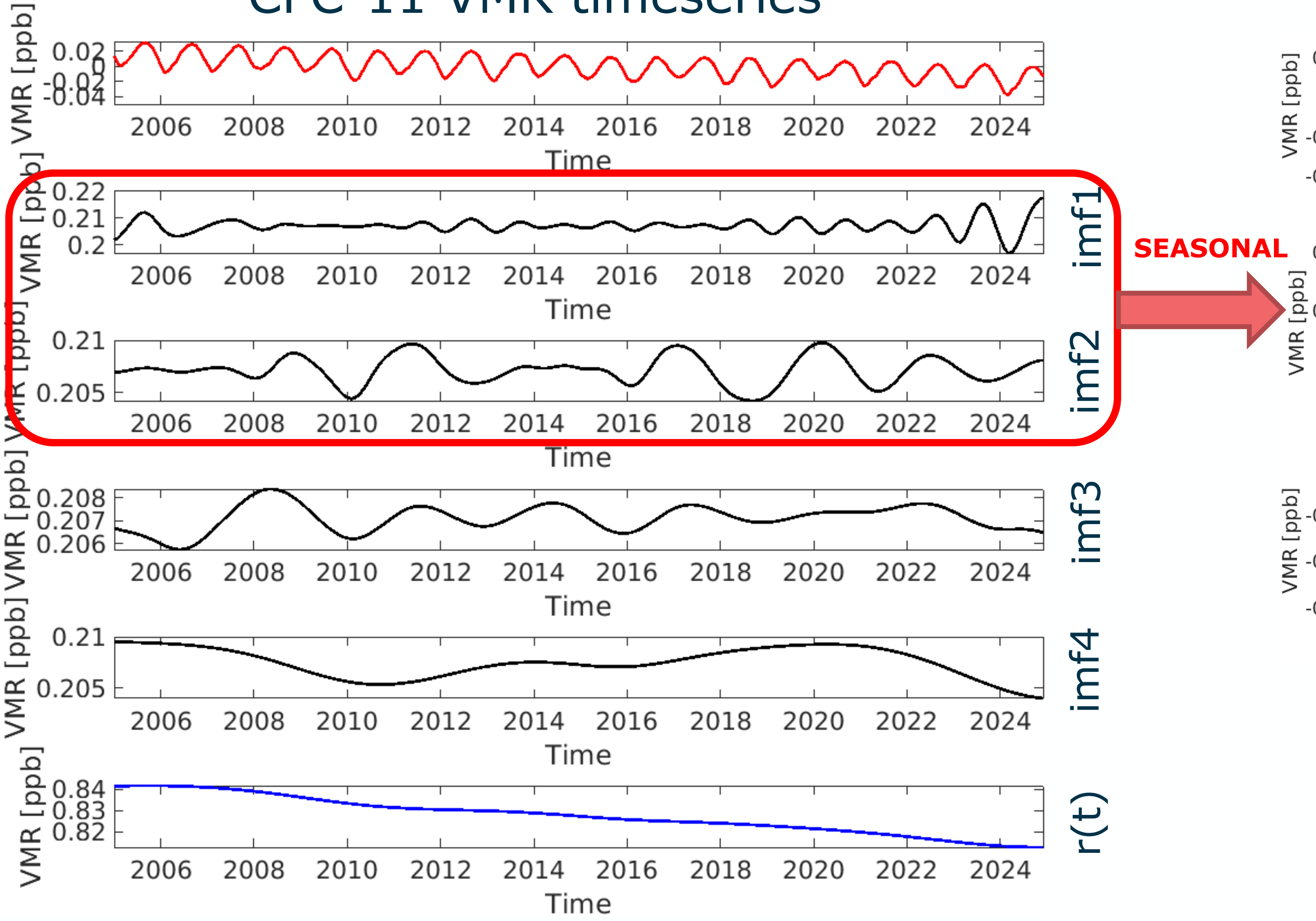
Use of Complete Ensemble Empirical Mode Decomposition with **Adaptive Noise** (CEEMDAN).

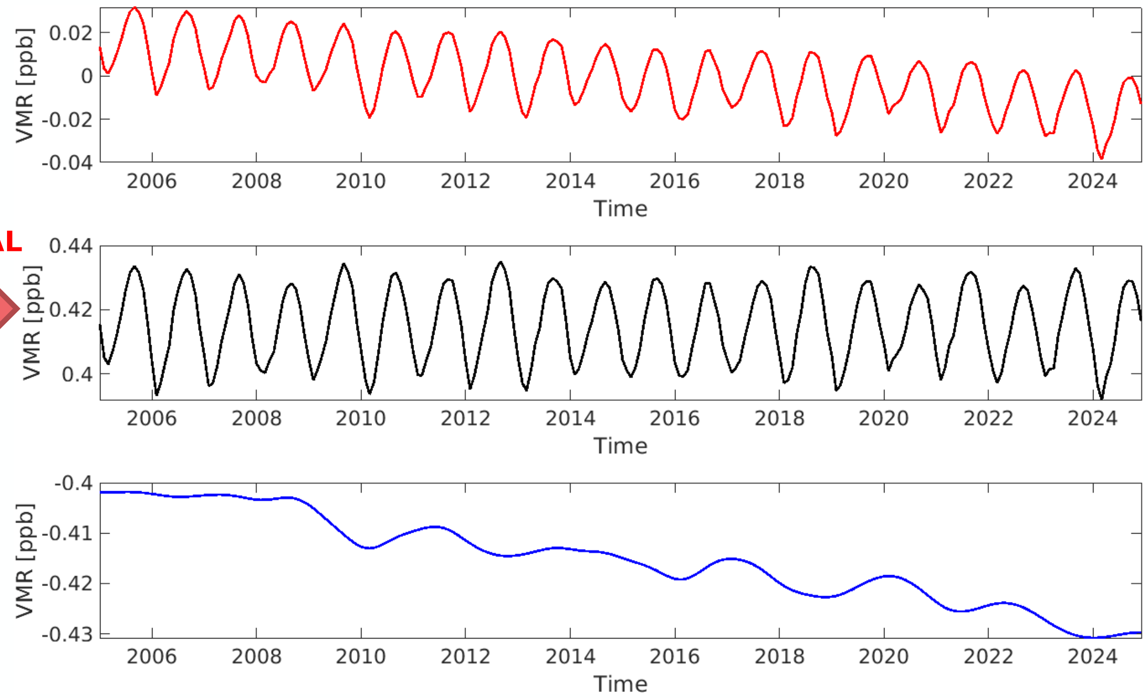
- Incorporates noise in an ensemble framework, improving stability.
- Utilizes adaptive noise to refine decomposition precision.



INTRINSIC MODE FUNCTIONS

CFC-11 VMR timeseries



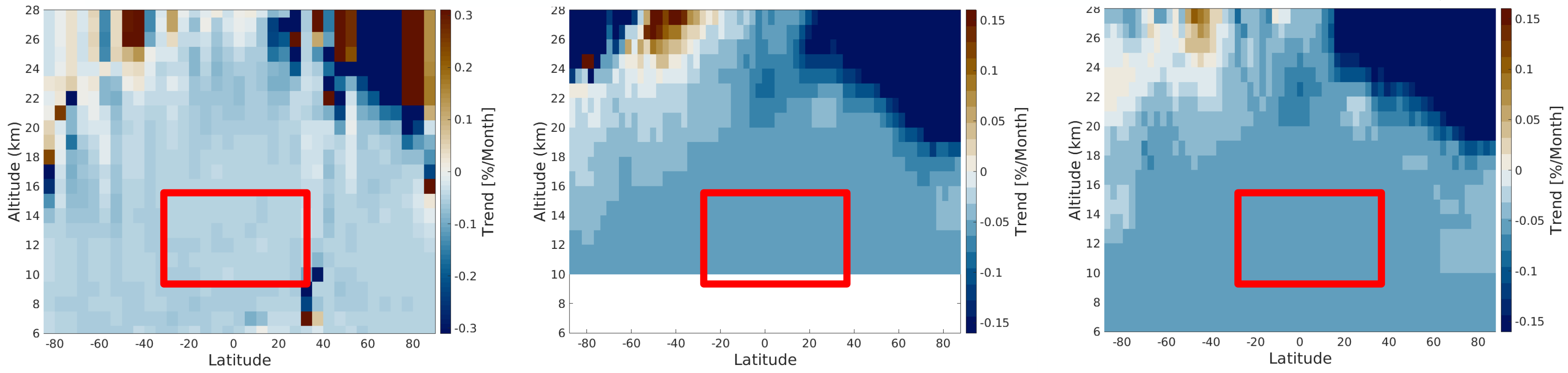


DE-SEASONALISED

GLOBAL AND REGIONAL TRENDS

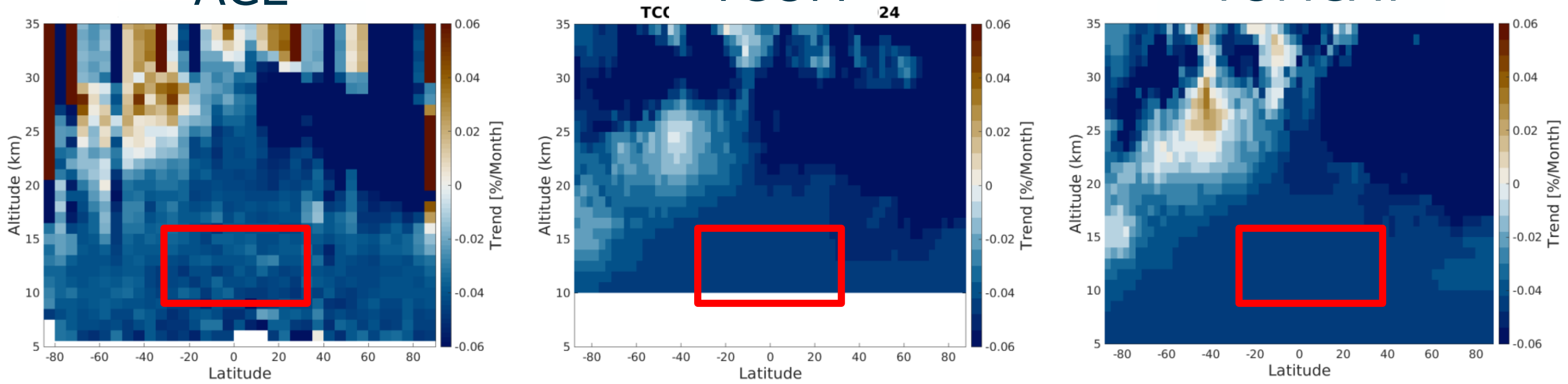
CFC-11 2005-2024 trends

ACE | TCOM | TOMCAT



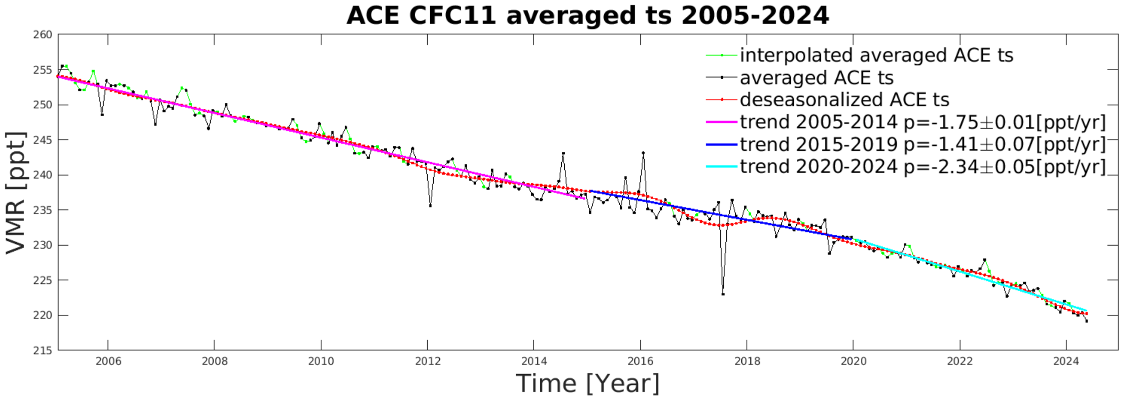
CFC-12 2005-2024 trends

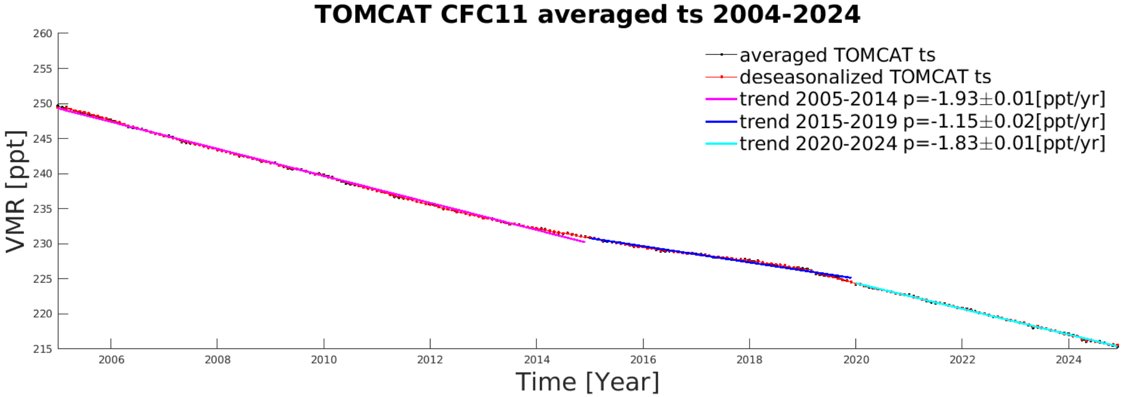
ACE | TCOM | TOMCAT

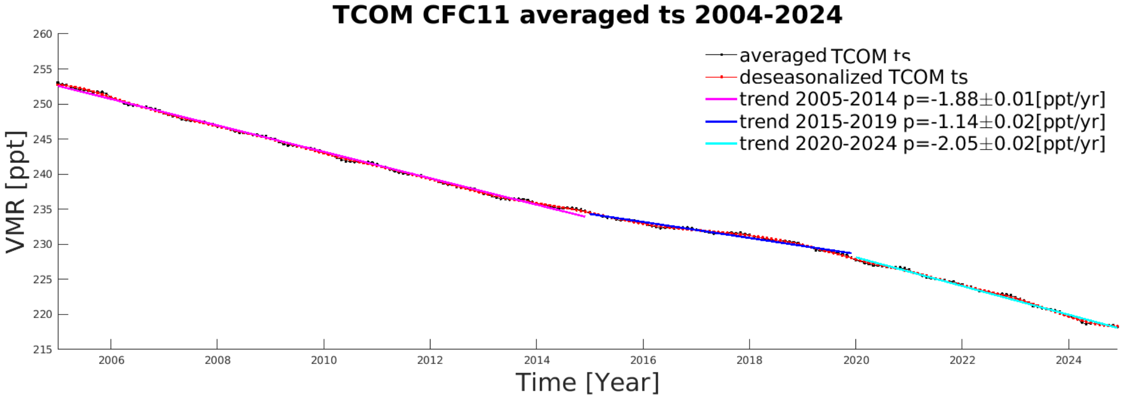


CFC-11 and CFC-12 VMR averaged between 9.5–15.5 km and 30ºS-30ºN (Schmidt et al., 2024).

GLOBAL AND REGIONAL TRENDS: CFC-11







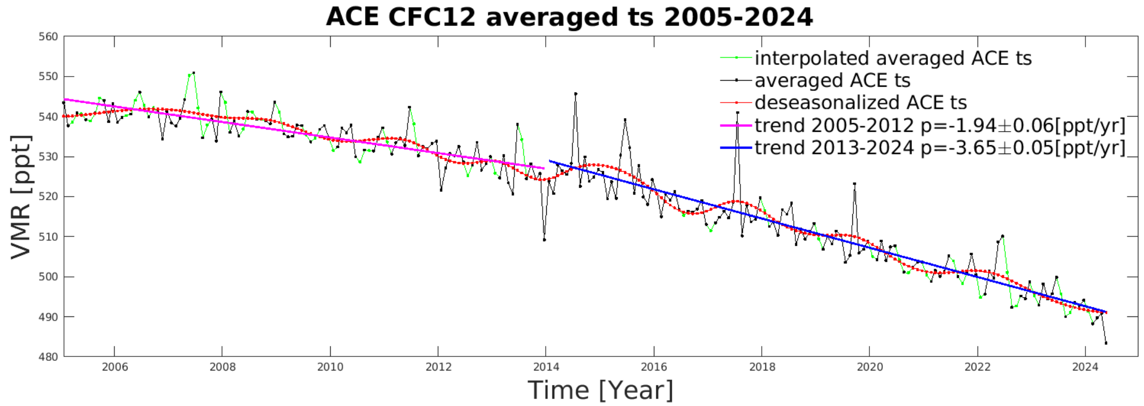
	Slope 2005-2014 [ppt/yr]	**Slope 2015-2019 [ppt/yr]**	**Slope 2019-2024 [ppt/yr]**
ACE v5.2 (Schmidt 2024)	-1.89 ± 0.02	-1.00 ± 0.05	-2.12 ± 0.06
NOAA NH (Schmidt 2024)	-2.07 ± 0.01	-1.03 ± 0.01	-2.74 ± 0.04
ACE v5.2	-1.75 ± 0.01	-1.41 ± 0.07	-2.34 ± 0.05
TOMCAT	-1.93 ± 0.01	-1.15 ± 0.02	-1.83 ± 0.01
TCOM	-1.88 ± 0.01	-1.14 ± 0.02	-2.05 ± 0.02

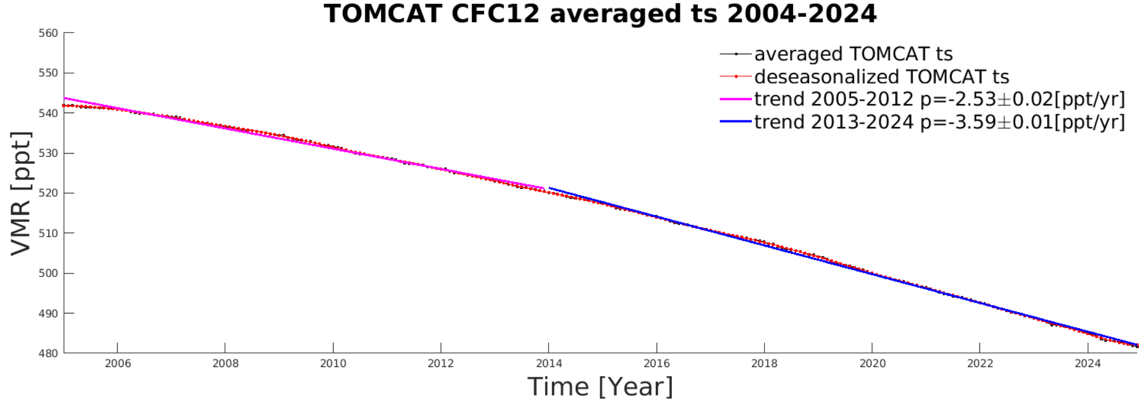
Decrease is expected under MP regulations

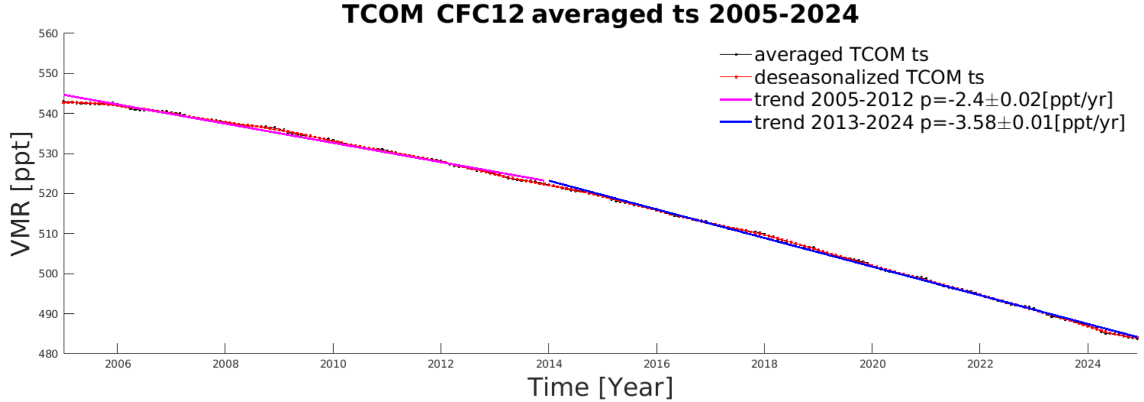
GLOBAL AND REGIONAL TRENDS: CFC-12

	Slope 2005-2012 [ppt/yr]	Slope 2013-2024 [ppt/yr]
ACE v5.2 (Schmidt 2024)	-1.44 ± 0.08	-3.16 ± 0.04
NOAA NH (Schmidt 2024)	-1.47 ± 0.05	-3.20 ± 0.02
ACE v5.2	-1.94 ± 0.06	-3.65 ± 0.05
TOMCAT	-2.53 ± 0.02	-3.59 ± 0.01
TCOM	-2.40 ± 0.02	-3.58 ± 0.01

Decrease is expected under MP regulations

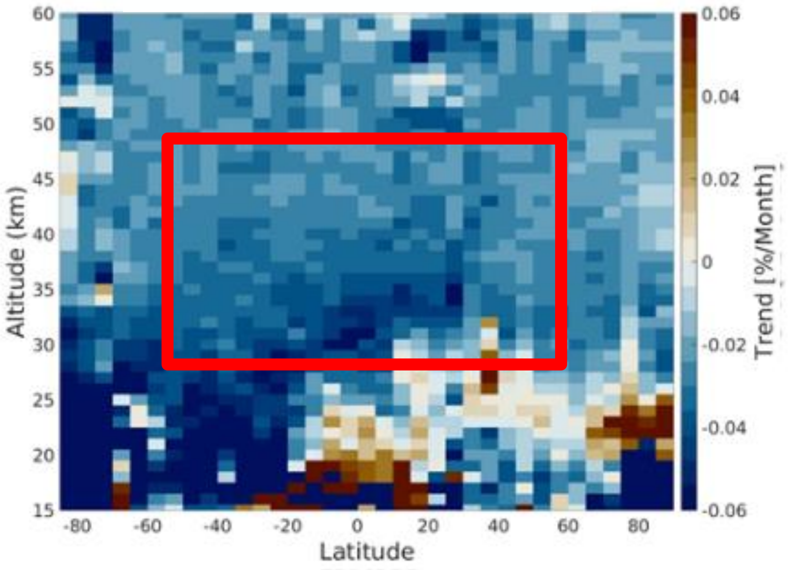




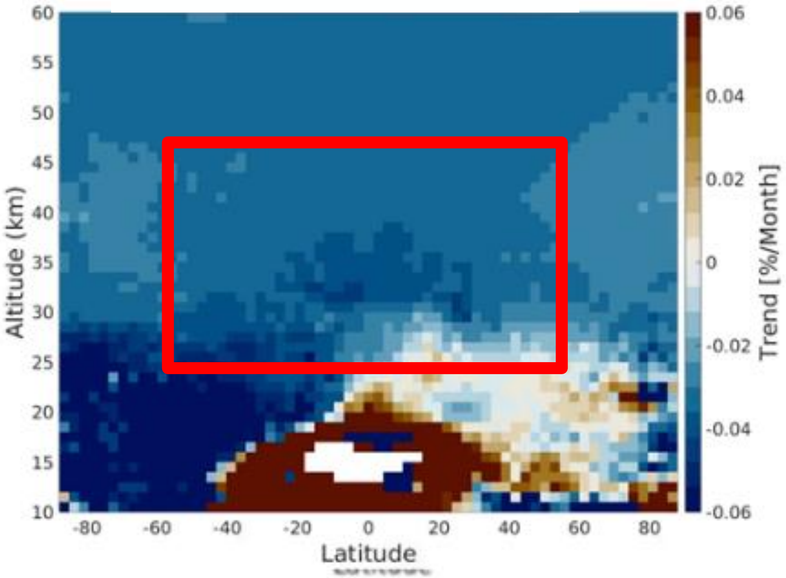


HCl 2005-2024 trends

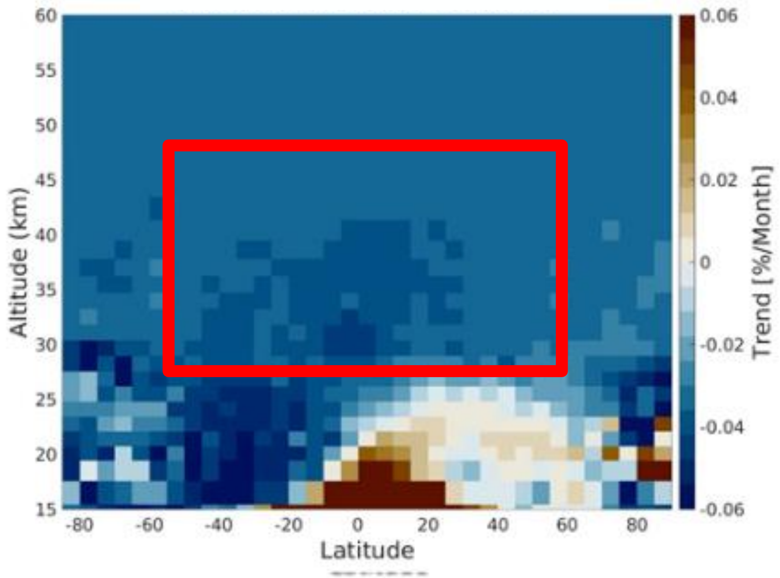
ACE



TCOM

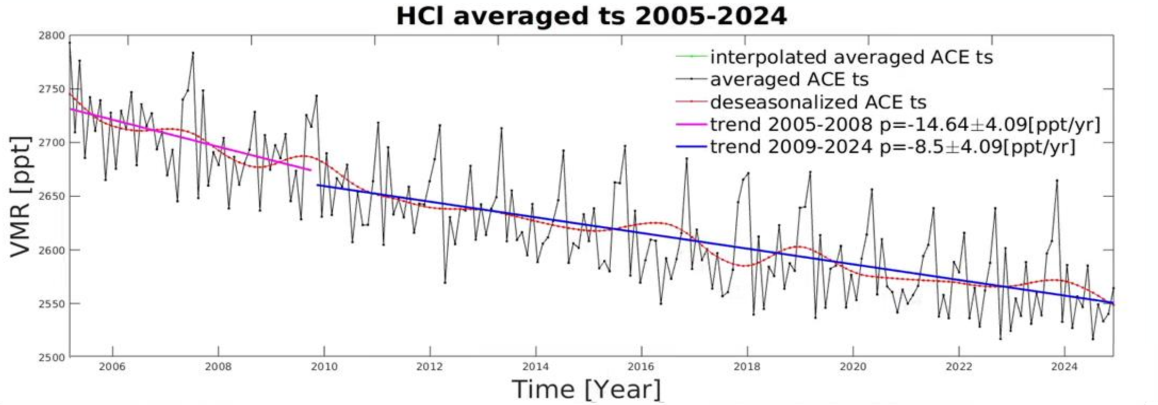


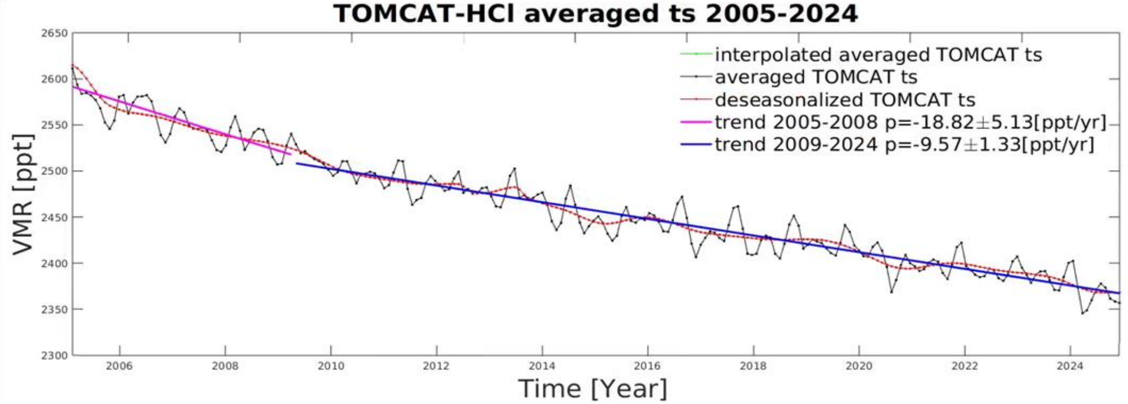
TOMCAT

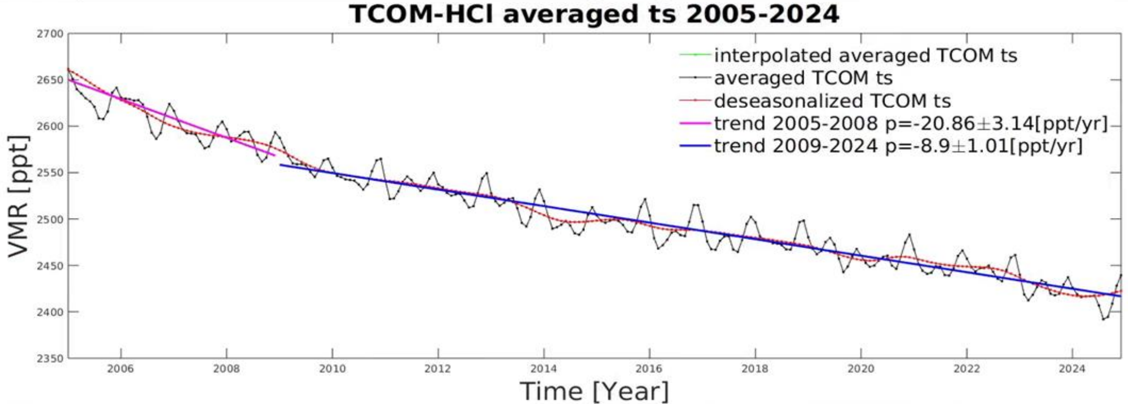


HCl VMR averaged between 28.5-48.5 km and 60°S-60°N (Schmidt et al., 2024).

GLOBAL AND REGIONAL TRENDS: HCl







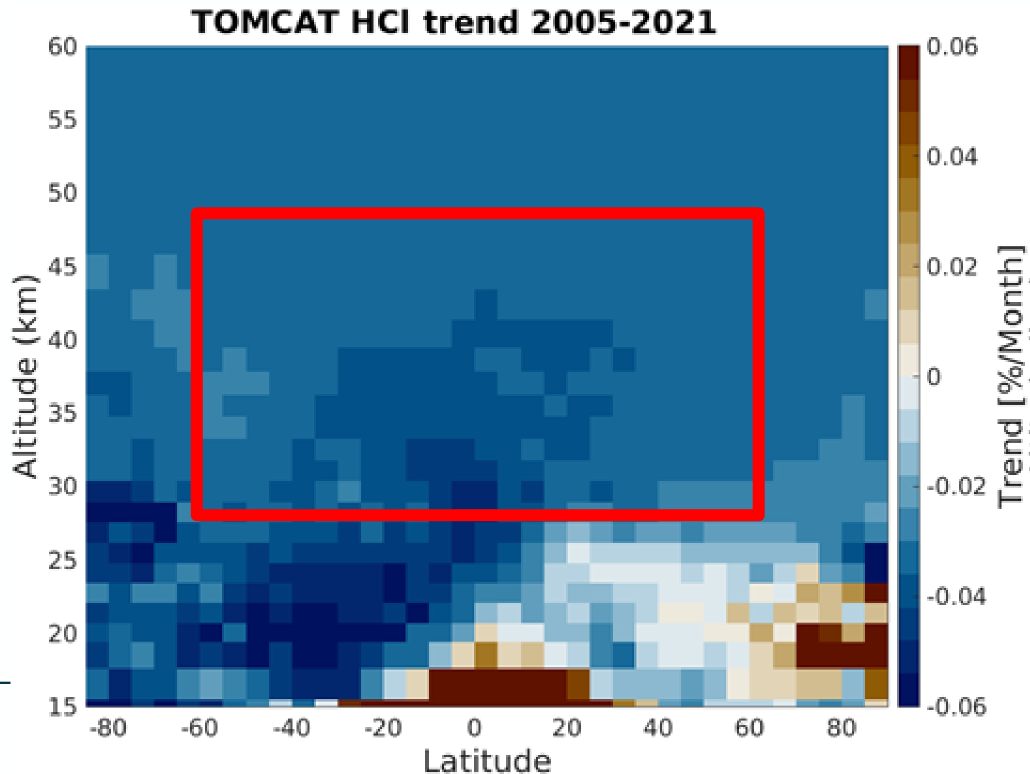
	Slope 2005-2008 [ppt/yr]	Slope 2009-2024 [ppt/yr]
ACE v5.2 (Schmidt)	-20.86 ± 2.66	-8.64 ± 0.32
ACE v5.2	-14.64 ± 4.09	-8.50 ± 4.09
TOMCAT	-18.82 ± 5.13	-9.57 ± 1.33
TCOM	-20.86 ± 3.14	-8.90 ± 1.01

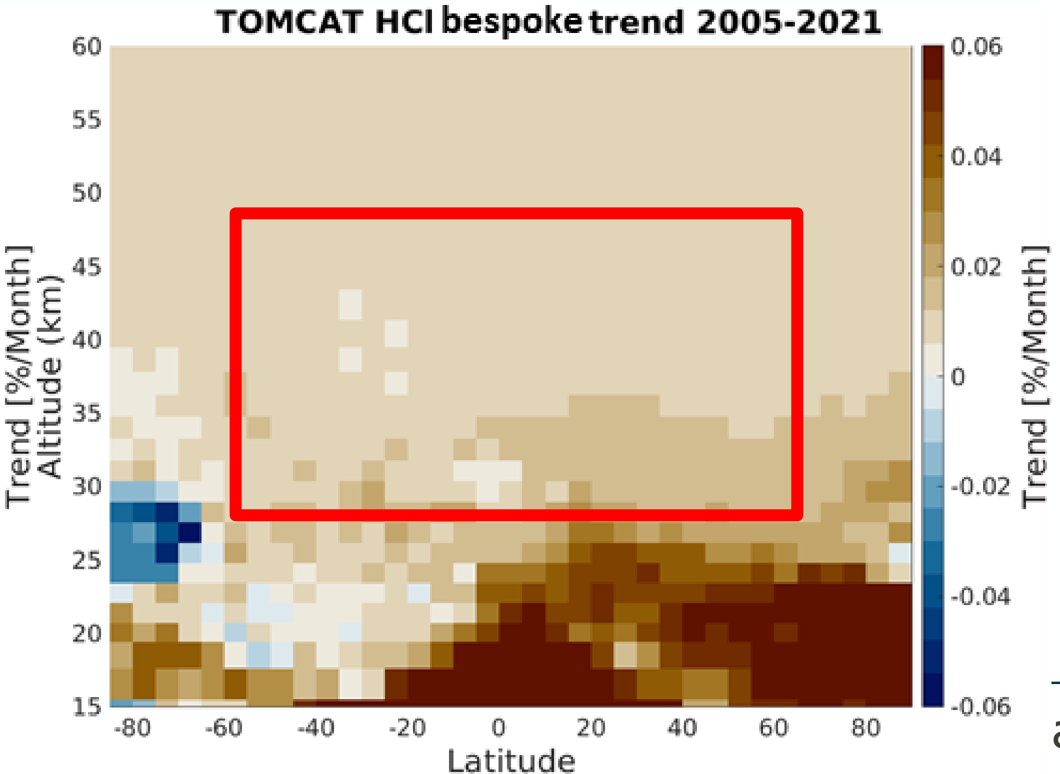
Decrease is expected under MP regulations

TOMCAT BESPOKE MODEL RUNS

Two configurations were compared:

- a control run reproducing the observed decline in chlorine
- a bespoke run with chlorine source gas concentrations fixed to their peak 1995 values.





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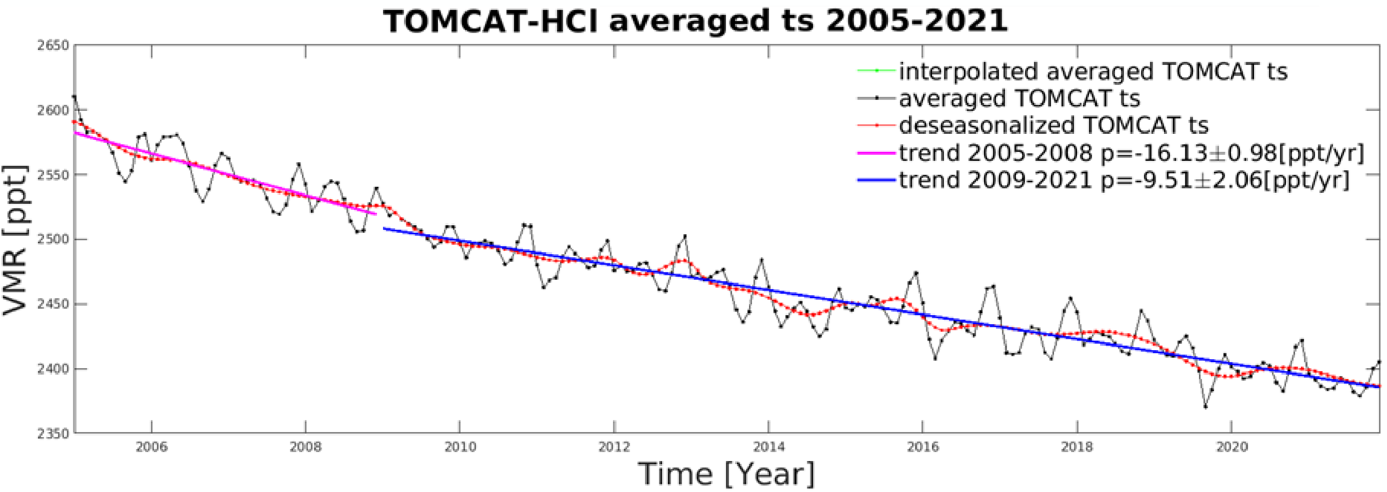
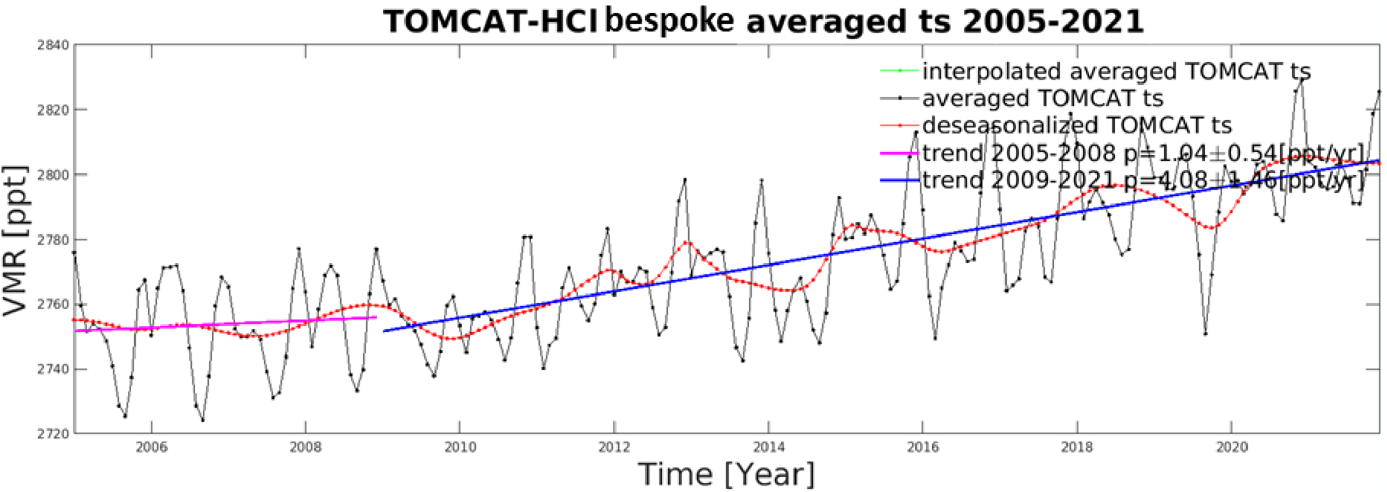
TOMCAT BESPOKE MODEL RUNS





	Slope 2005-2008 [ppt/yr]	Slope 2009-2024 [ppt/yr]
ACE v5.2 (Schmidt)	-20.86 ± 2.66	-8.64 ± 0.32
ACE v5.2	-14.64 ± 4.09	-8.50 ± 4.09
TOMCAT	-18.82 ± 5.13	-9.57 ± 1.33
TCOM	-20.86 ± 3.14	-8.90 ± 1.01
BESPOKE	1.04 ± 0.54	4.08 ± 1.46

The increasing HCl concentrations of the bespoke model run are linked to increasing stratospheric ozone loss.

SUMMARY

- Defined a **new methodology** based on the EMD, a technique particularly effective in analysing **non-stationary** signals, where the frequency content of the signal varies over time.

- Used the EMD-based approach to **de-seasonalise** CFC-11, CFC-12 and HCl timeseries and calculate significant trends.

- Evaluated **trends** of CFCs (CFC-11 and CFC-12) HCl over 2005-2024 from three different data sources against published ACE-FTS trends, and new TCOM datasets.

- Demonstrated TOMCAT model's **sensitivity** of stratospheric chlorine loading to source gas emissions.

- The comparison between the control and constant-Cl runs confirms that changes in chlorine source gas concentrations directly influence the modelled HCl trends with the increasing trend related to ozone loss.

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