Temporal variation of HCl and HF at Tsukuba related to the change of the meridional circulation in the northern lower stratosphere

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Why HCl, HF ?

To check the effect of Montreal Protocol

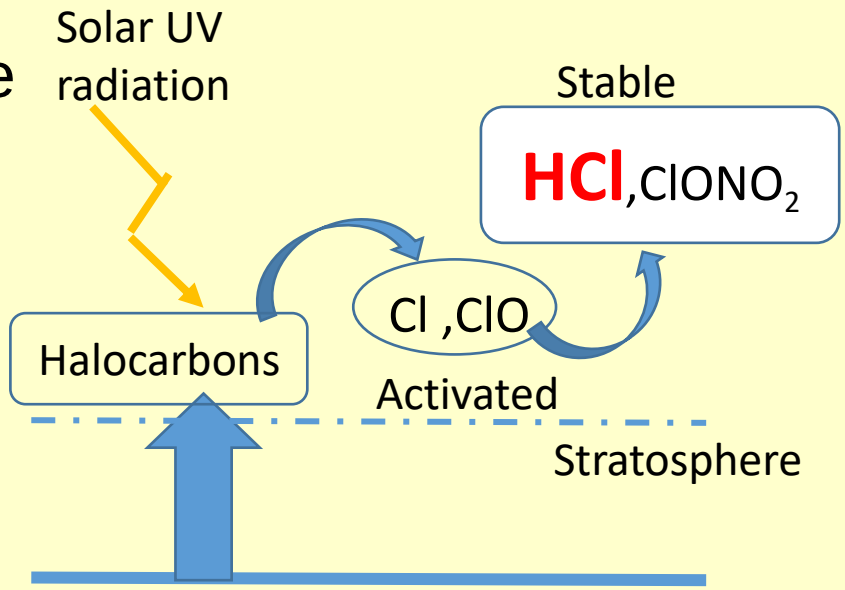
HCl:

Mainly distributed in the stratosphere

Source: CFCs (+ Natural)

Reservoir of chlorine species

(chlorine destroys ozone)



$$CF_2Cl_2 + h\nu \rightarrow CF_2Cl + Cl \qquad (\lambda < 320nm)$$

$$CF_2Cl + O_2 + M \rightarrow CF_2ClO_2 + M$$

$$CF_2ClO_2 + NO \rightarrow CF_2ClO + NO_2$$

$$CF_2ClO + M \rightarrow COF_2 + Cl + M$$

$$Cl + CH_4 \rightarrow HCl + CH_3$$

HF:

Mainly distributed in the stratosphere

Source: CFCs, HFCs etc.

Stable (tracer of transport)

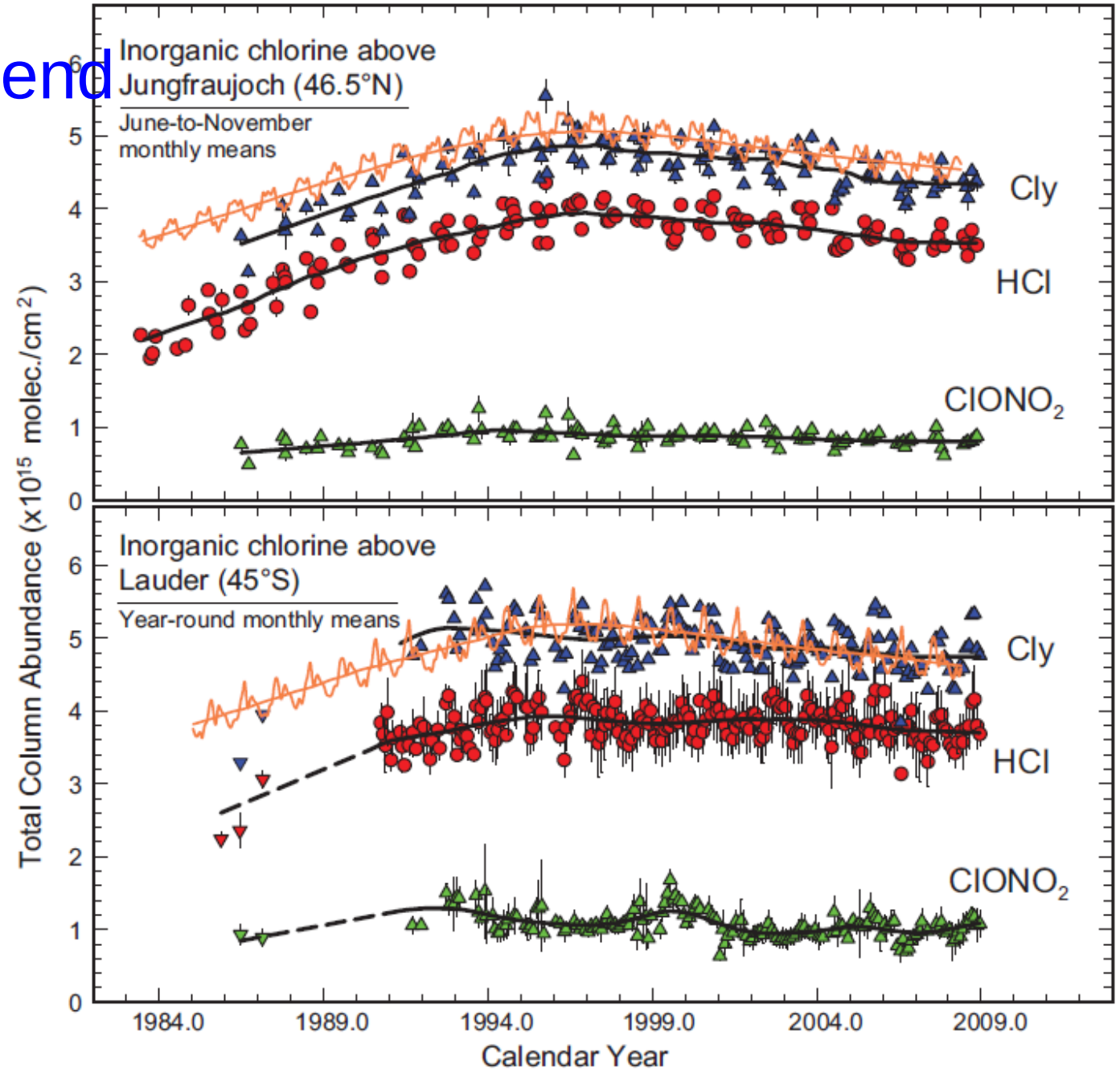
$$COF_2 + h\nu \rightarrow FCO + F$$

$$F + CH_4 \rightarrow HF + CH_3$$

$$F + H_2O \rightarrow HF + OH$$

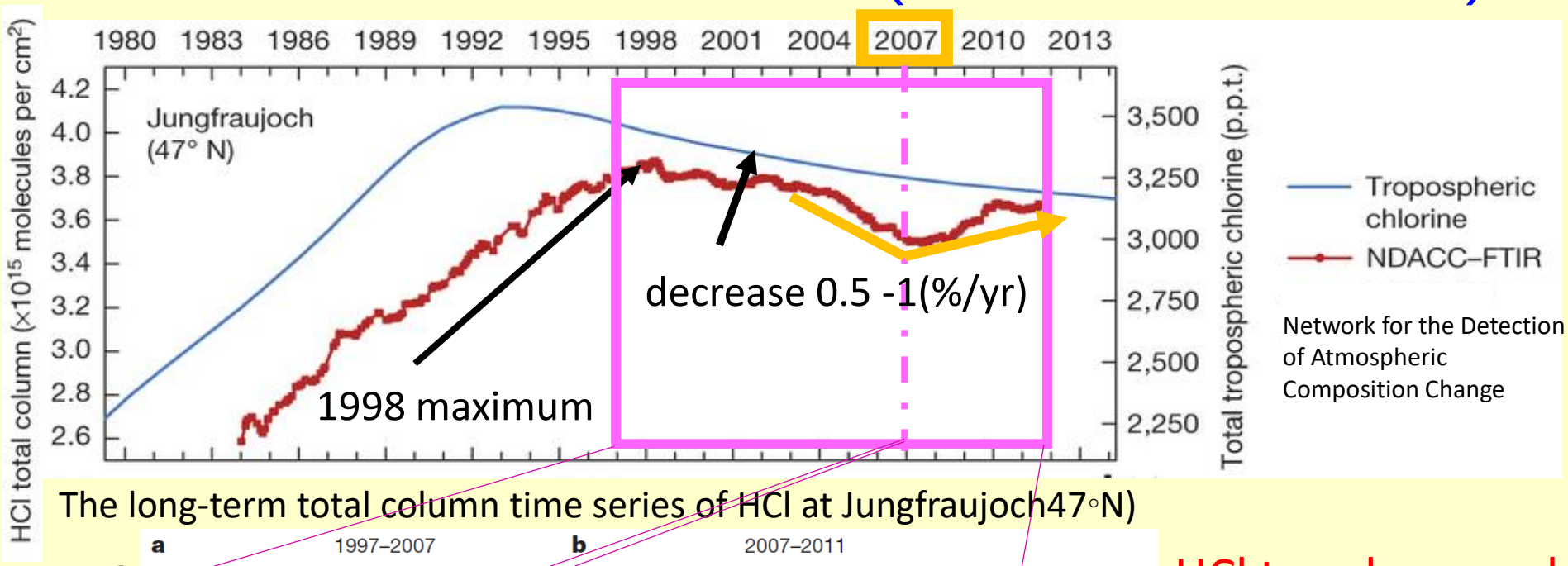
Observed trend of total Cl

Decrease from late ‘90

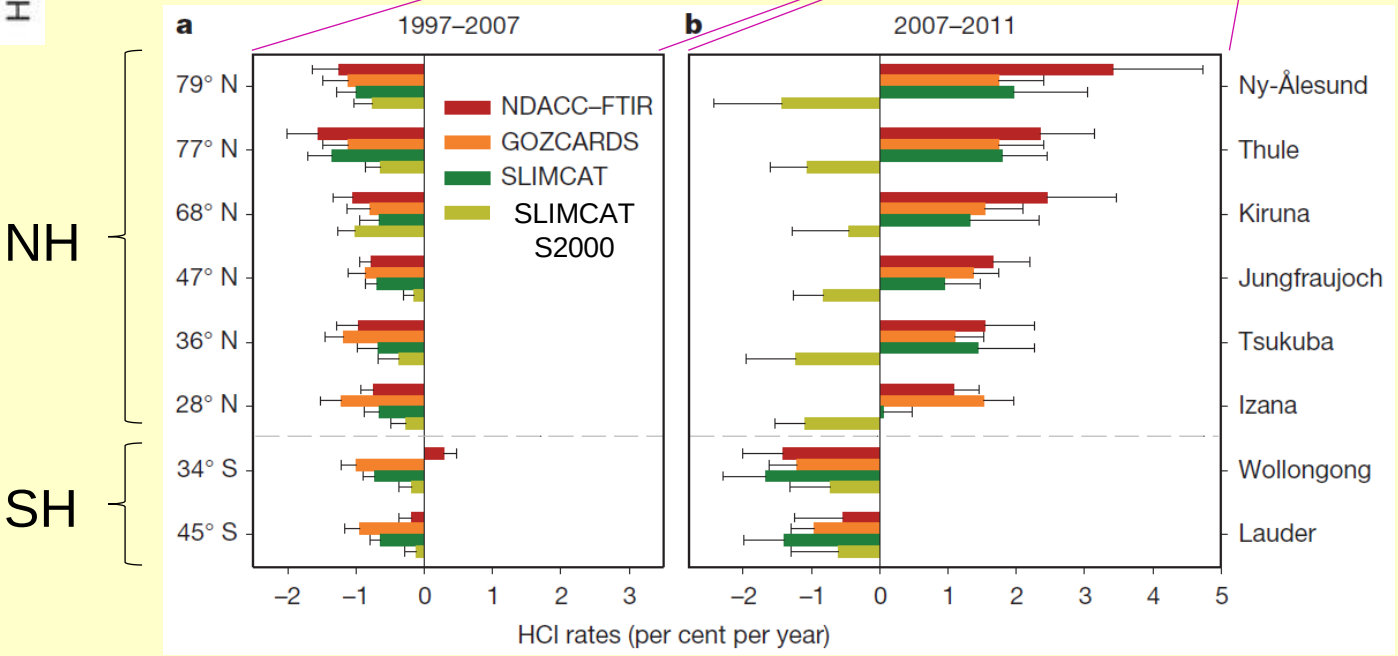


[WMO, Scientific Assessment of Ozone Depletion: 2010, 2011]

HCl trend reversal after 2007 (Mahieu et al., 2014)



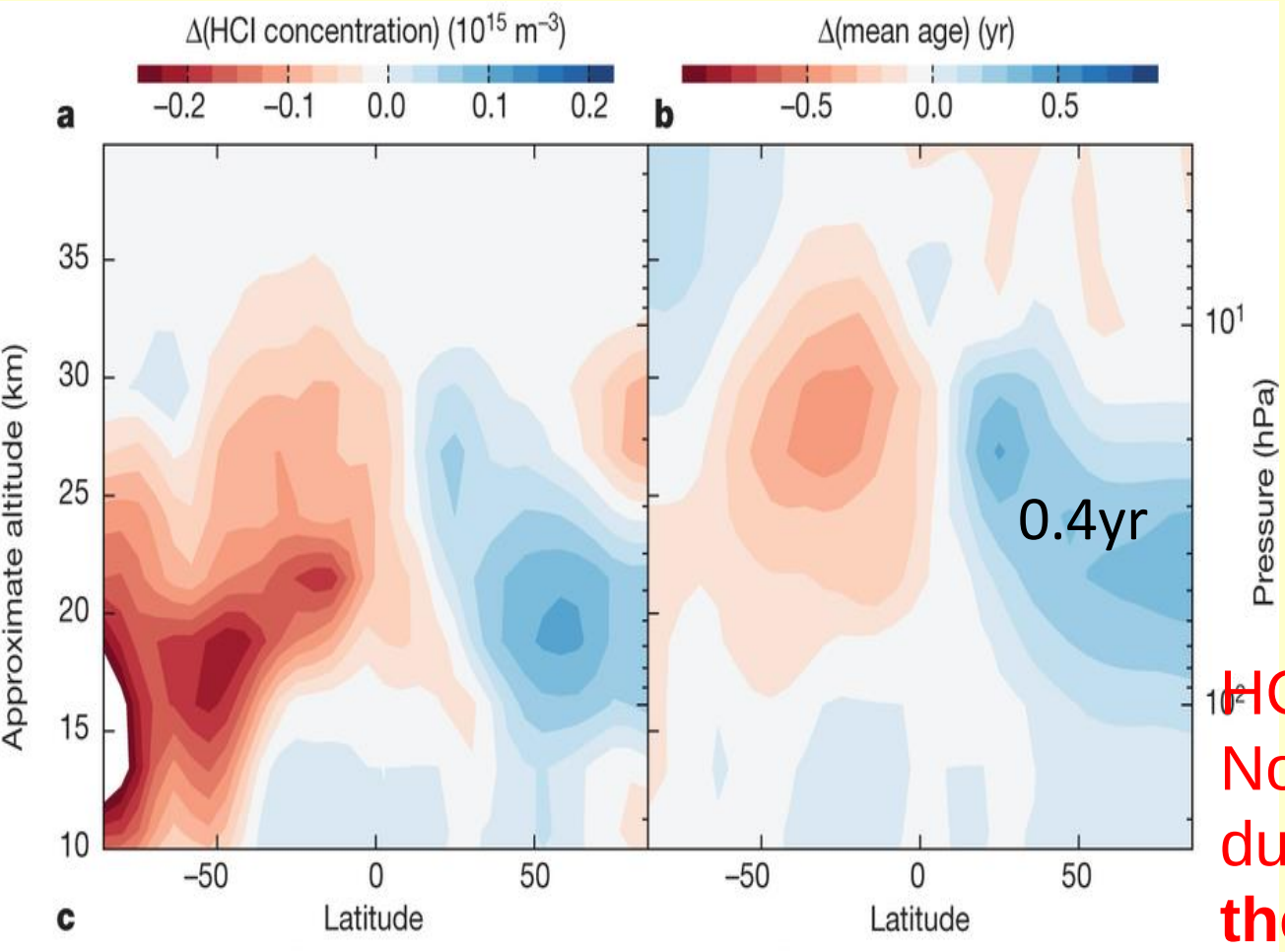
The long-term total column time series of HCl at Jungfraujoch47◦N)



HCl relative rates of change for 8 NDACC sites

HCl trend reversal (decrease to increase) occurred at 2007 only in the northern hemisphere.

Trend reversal due to circulation change



Mean differences between 2010/2011 and 2005/2006

The age-of-air increased in the Northern stratosphere. (circulation become slower)

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Conversion from source gases into HCl progressed.

⇩

HCl increased.

HCl increase in the Northern hemisphere is due to a **slowdown of the atmospheric circulation**, occurring over several consecutive years.

Results of previous study

Dynamical variability which occurred on a timescale of a few years, characterized by a persistent slowing of stratospheric circulation after 2005 brought HCl-enriched air into the Northern hemisphere lower stratosphere.

E.Mahieu et al.,[2014]

Make sure that HCl total column decreases again after the previous study. ⟵ Last year's presentation

Purpose of this study

Investigate the recent trend (till 2018) of HCl and HF total column at Tsukuba observed with FTIR and compare it with MIROC3.2 Chemistry-Climate Model (CCM) results.

Instrument

Bruker 120HR(125HR from 2010)

at Tsukuba（36.1°N, 140.1°E, 31 m A.S.L.）

Total column retrieval

SFIT4 v0.9.4.4

HCl: MW1: 2727.73 - 2727.83 cm^{-1} HCl, O_3, HDO

MW2: 2775.70 - 2775.80 cm^{-1} HCl, N_2O, O_3

MW3: 2925.80 - 2926.00 cm^{-1} HCl, NO_2, CH_4, O_3

HF:

4038.81 - 4039.07 cm^{-1}

HF, H_2O, CH_4, HDO

Bruker 120HR (left) and 125HR (right) in NIES building

CCM

Version 3.2 of the Model for Interdisciplinary Research on Climate (MIROC3.2)

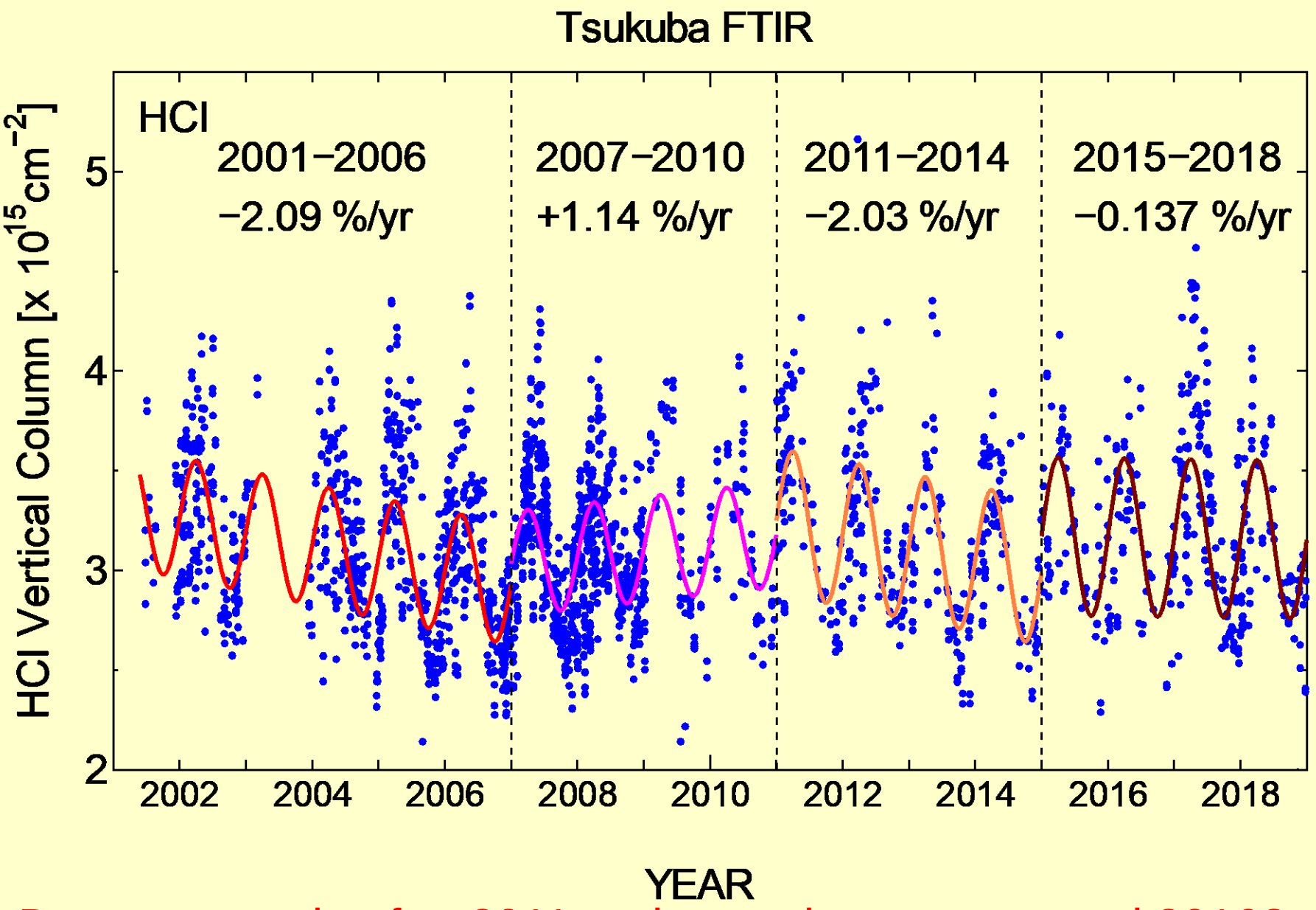
spectral model with a T42 horizontal resolution (2.8° × 2.8°) and 34 vertical atmospheric layers above the surface. The top layer is located at approximately 80 km (0.01 hPa).

The horizontal wind velocity in the CCM were nudged toward the ERA–Interim data.
The transport is calculated by a semi–Lagrangian scheme.

The reaction-rate and absorption coefficients are based on JPL–2010.

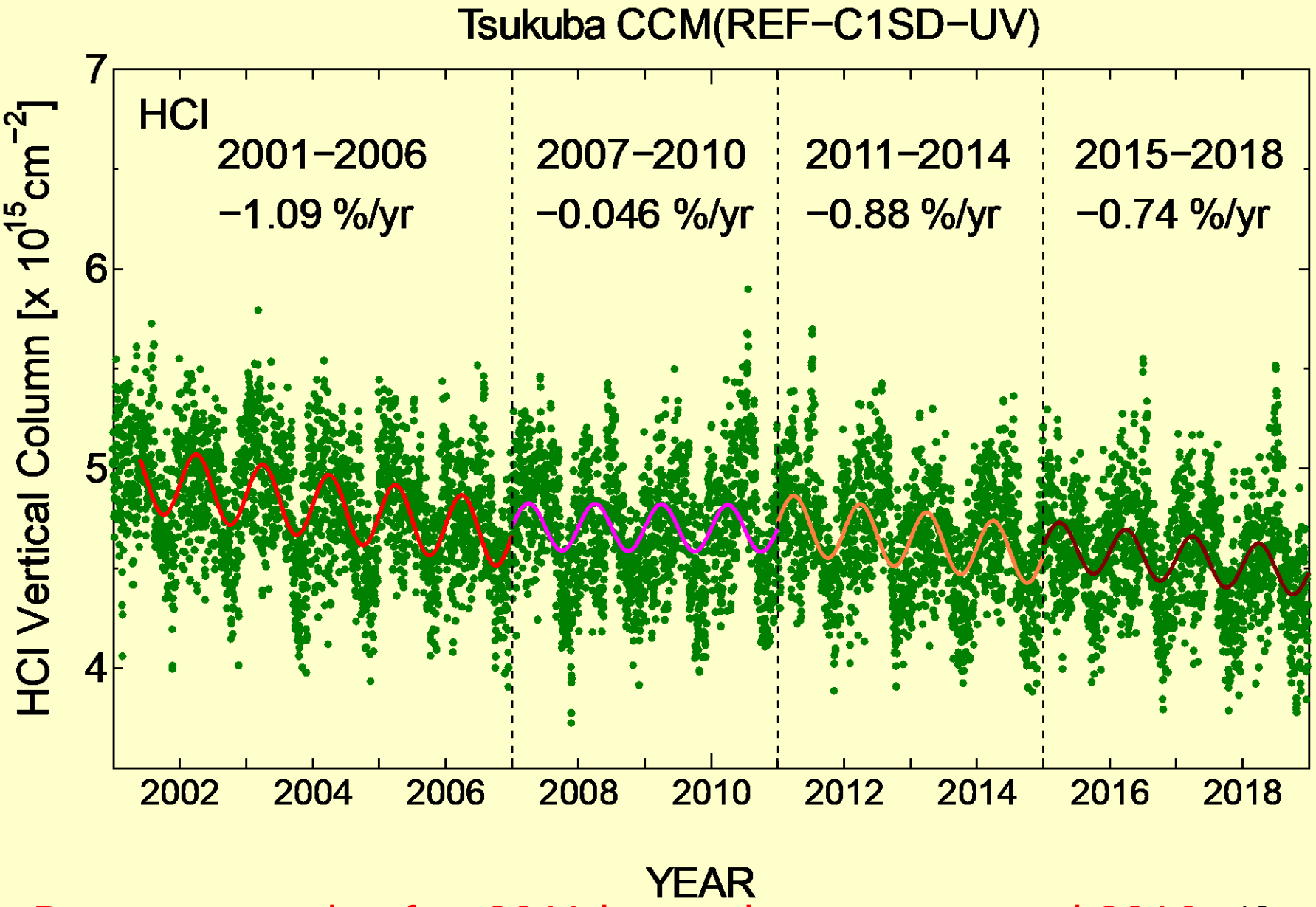
Emission Scenario:
The evolution of surface concentration of ODSs is from the World Meteorological Organization (WMO) baseline (A1) scenario (WMO, 2011). That of GHGs is based on the observation until 2005 then the RCP6.0 scenario is used after 2005.

Results: Temporal variation of HCl total column



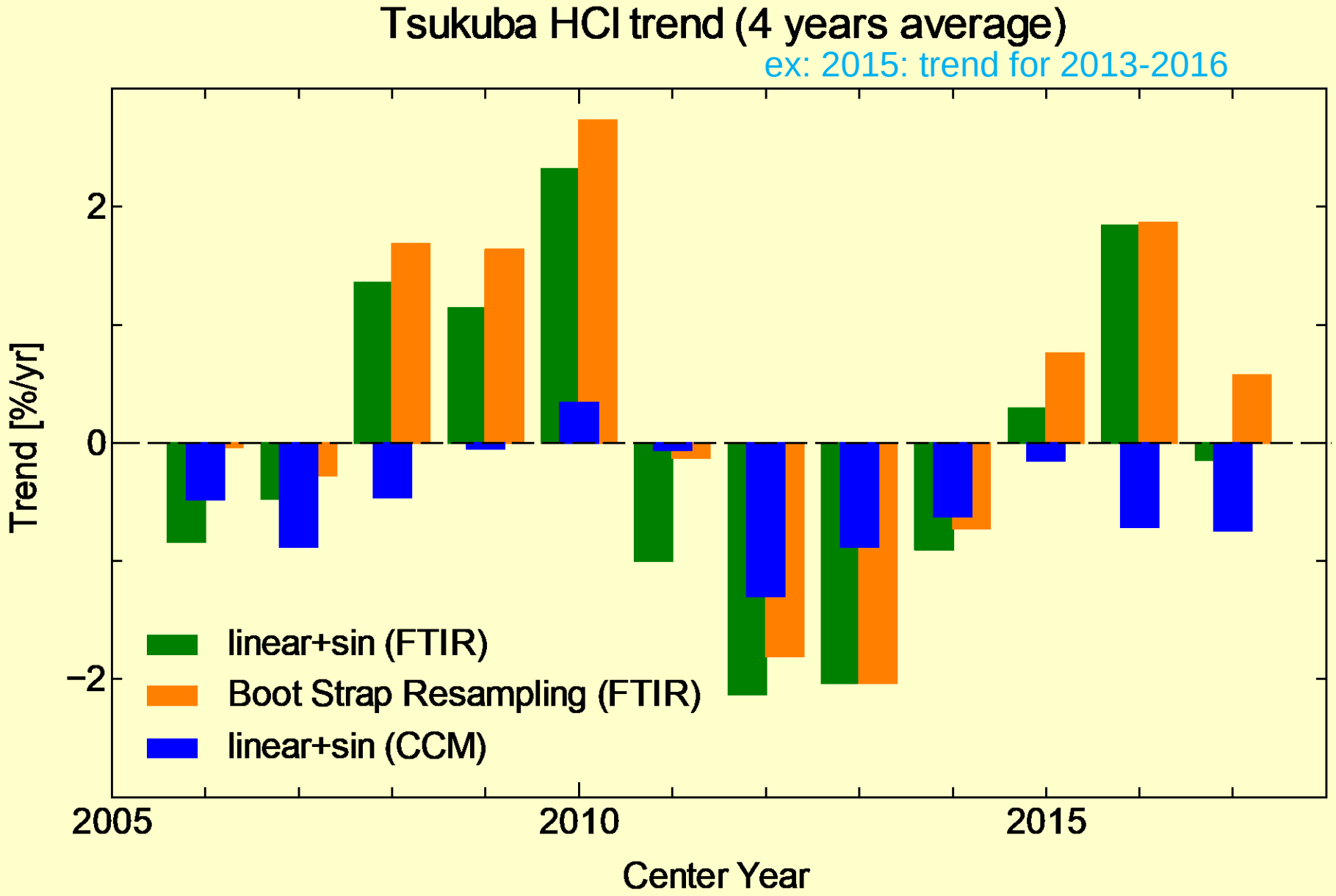
Decrease again after 2011 and some increase around 2016?

Temporal variation of HCl by CCM



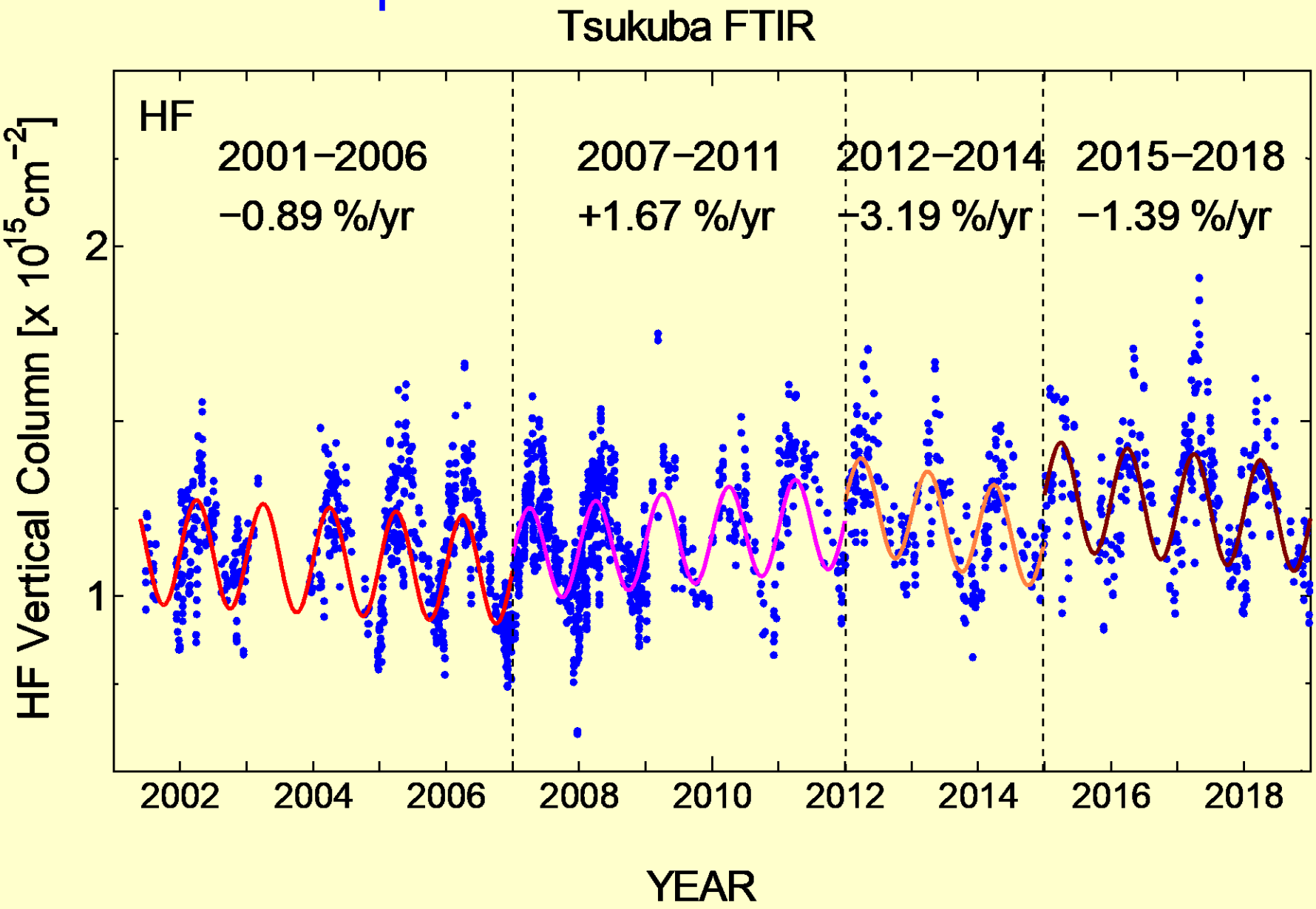
Decrease again after 2011 but no increase around 2016

Results: trend difference between Obs. and CCM



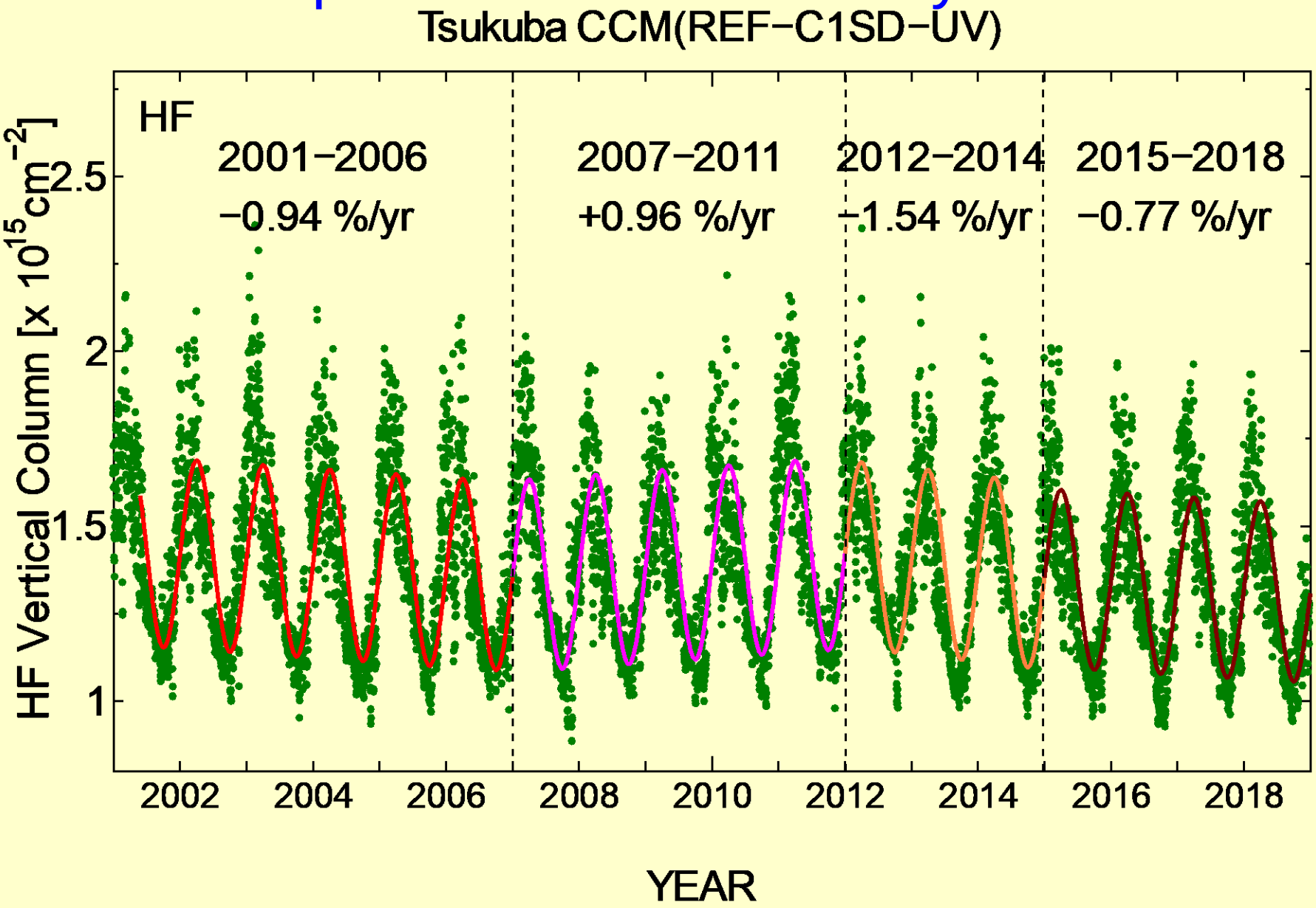
Trends of observation and CCM are different after 2015

Results: Temporal variation of HF total column



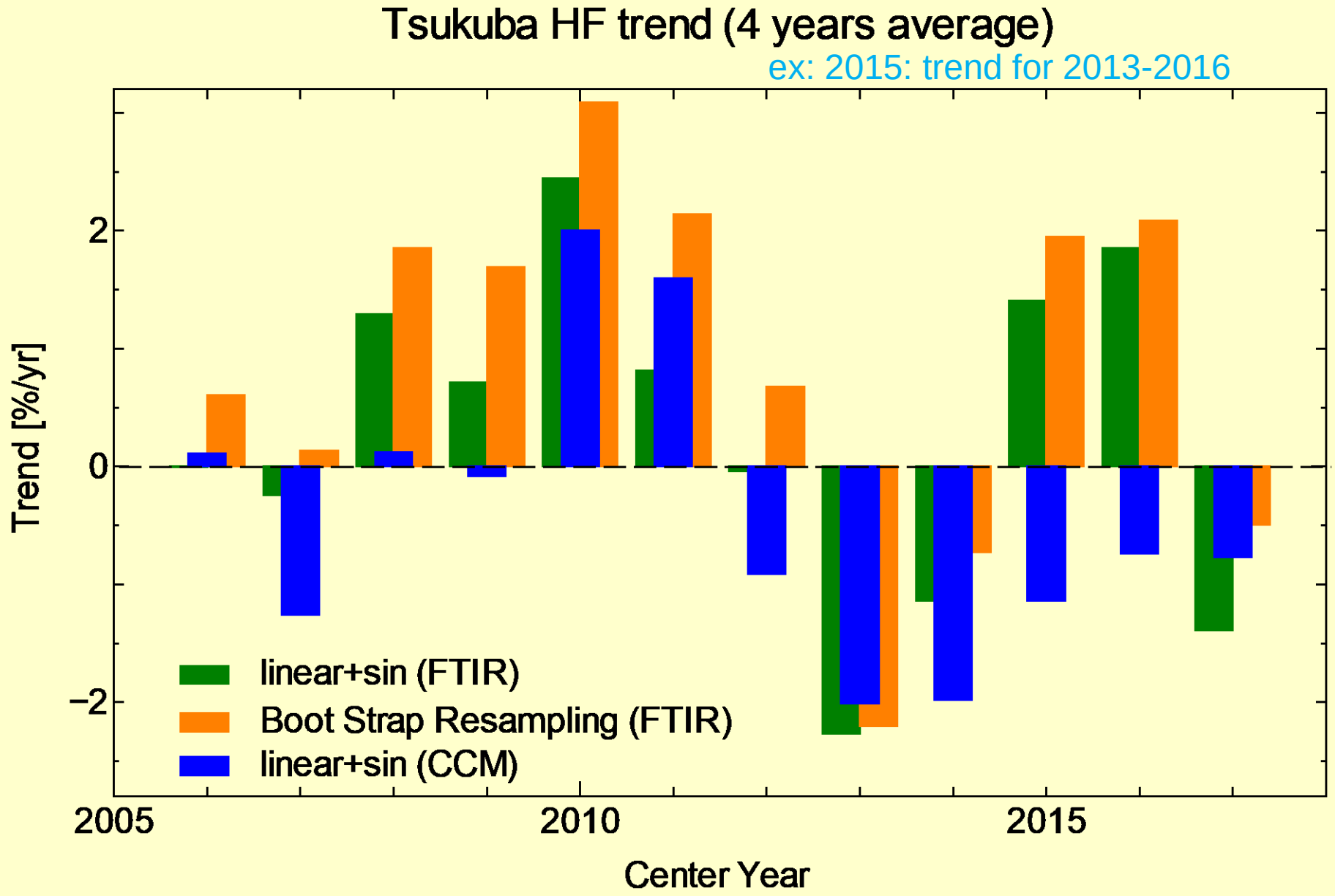
Decrease after 2012 and some increase around 2016?

Temporal variation of HF by CCM



Decrease after 2012 and slowdown after 2015

Results: trend difference between Obs. and CCM



Trends of observation and CCM are different after 2015

Why the trends of observation and CCM are different after 2015?

Dynamical variation in the northern hemisphere from the mass stream function change calculated using ERA-Interim (Last year's presentation)

2003-2006 to 2007-2010 deceleration ⇨ HCl, HF: decrease to increase

2007-2010 to 2011-2014 acceleration ⇨ HCl, HF: increase to decrease

2011-2014 to 2015-2018 deceleration (but weak)

⇨ HCl, HF: decrease to decrease but smaller rate

CCM results are consistent with dynamic variation (CCM also use ERA-Interim)

Observed trends are consistent with dynamic variation except for the last period (2011-2014 to 2015-2018)

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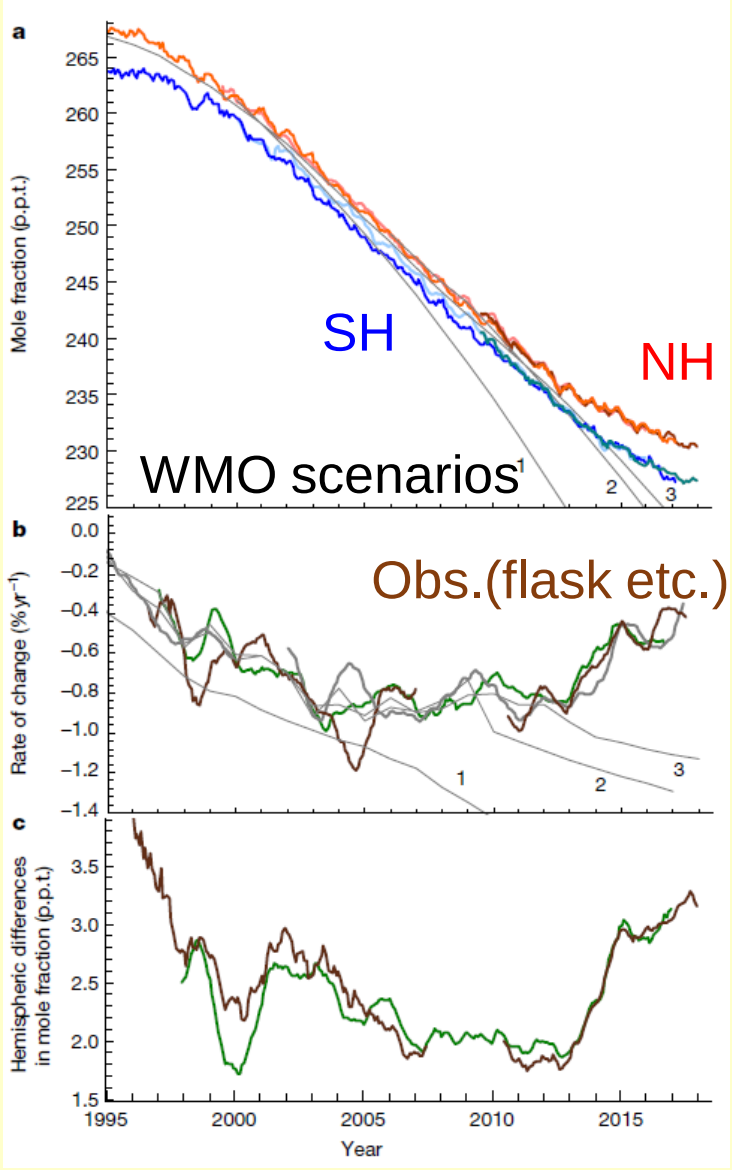
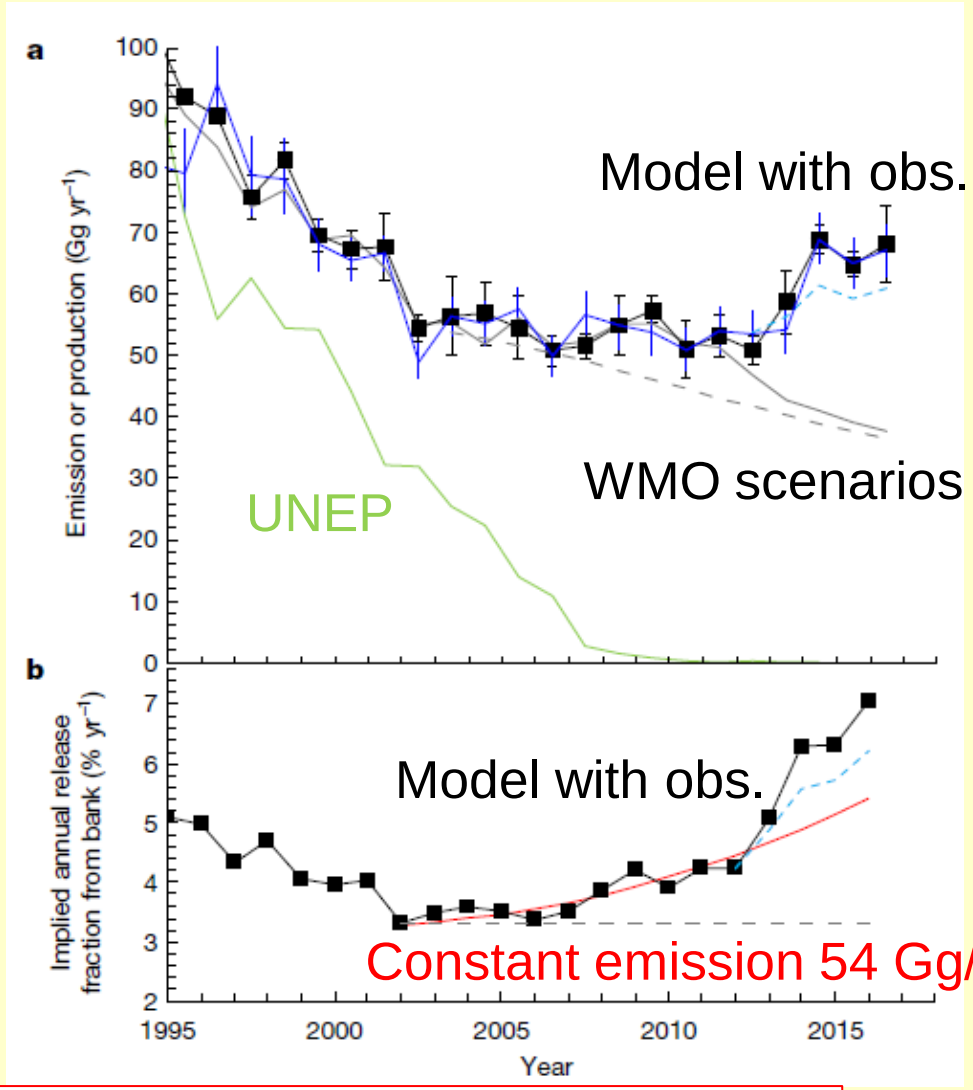
Maybe due to the extra emission of chlorine and fluorine

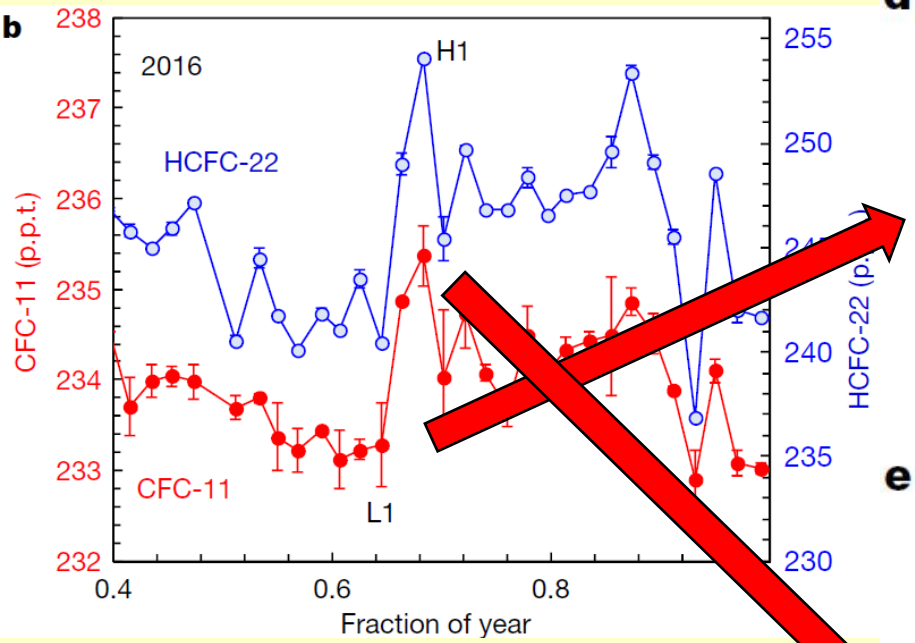
Increase after 2015 is due to emission change?

[Montzka et al., 2018]

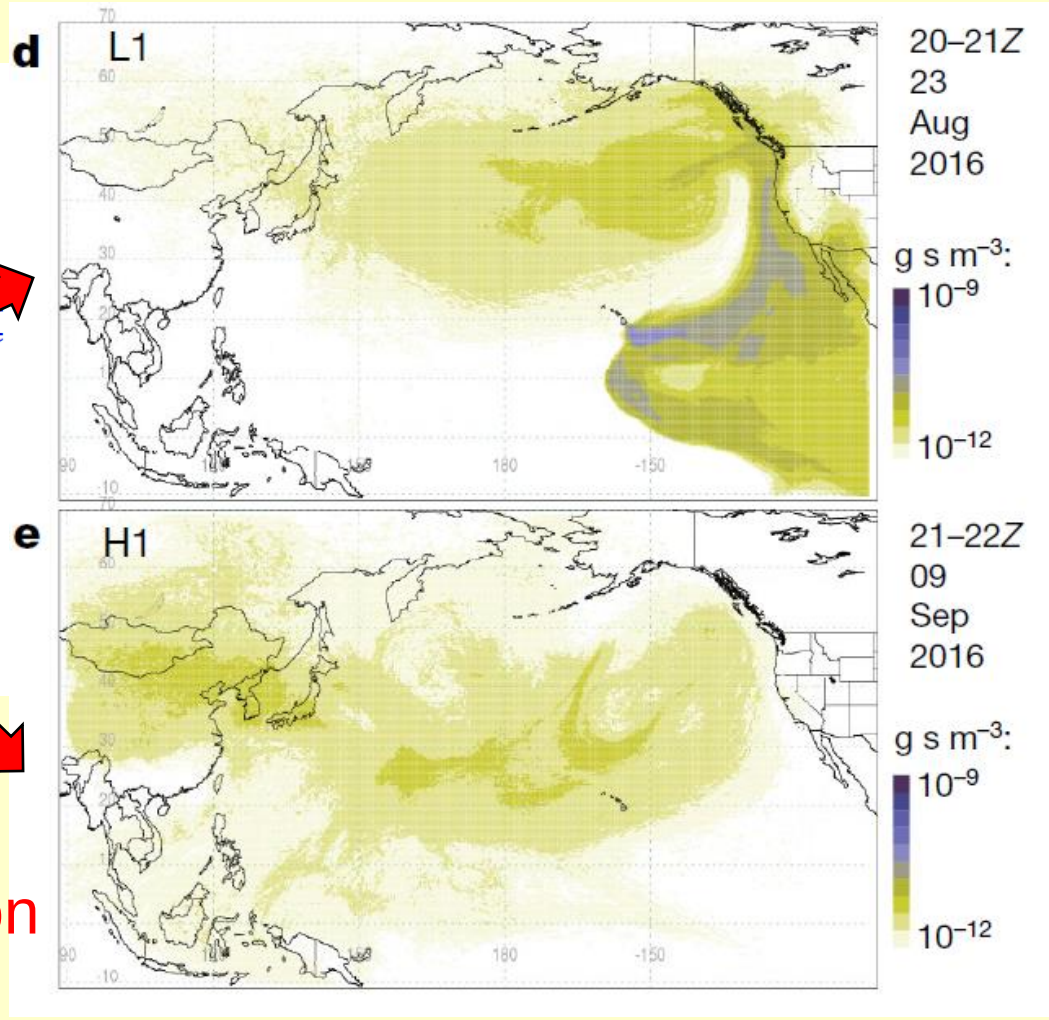




CFC-11 decline rate slowed after 2012



Observed concentration at MLO (Hawa



Backward trajectory from MLO (Hawaii)

Emission from China
(Some of the illegal production of CFC-11 have already stopped in 2018.)

There are some possibility that the emission change in CFC-11 affects the increase of HCl and HF after 2015.
(It needs several years to transport the air from the surface to stratosphere)

Conclusion

- HCl and HF total column were retrieved from the spectra observed with FTIR at Tsukuba between 2001 and 2018 and compared with CCM resuls.

- The trends of HCl and HF total column are different between the observation and CCM after 2015.

- There are some possibility that the emission change in CFC-11 affects the increase of HCl and HF after 2015.