

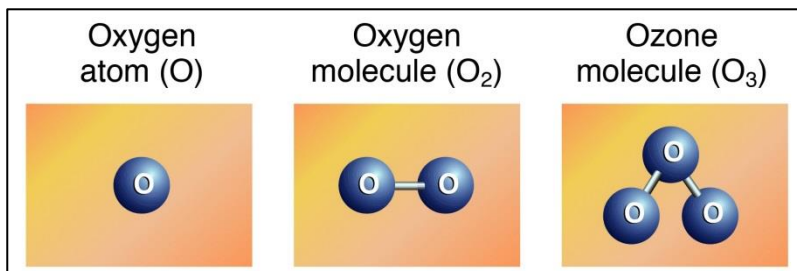
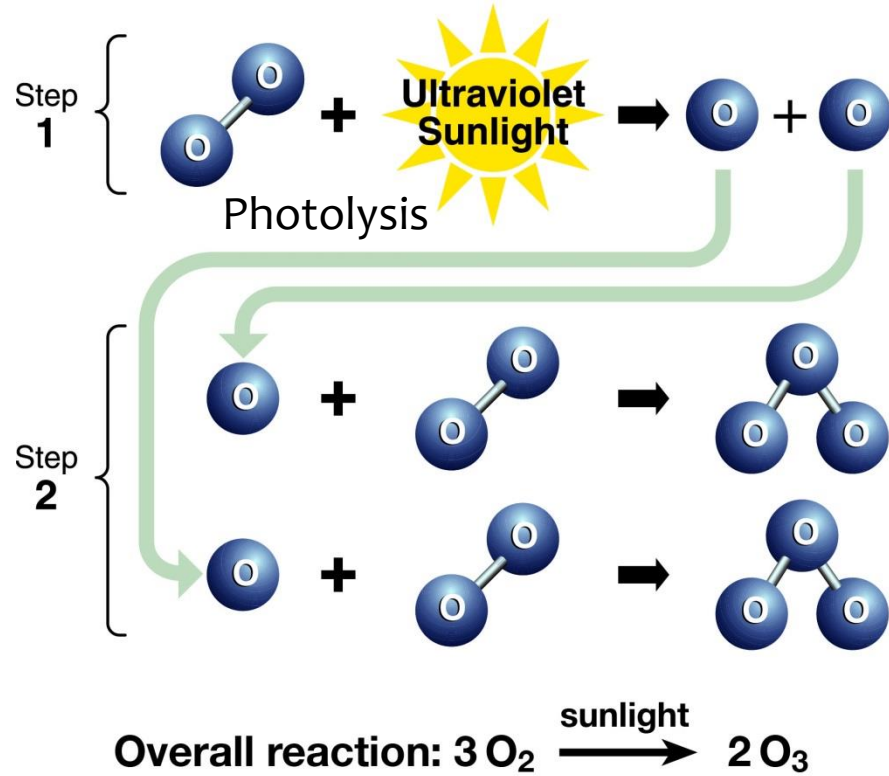
Une nouvelle menace pour la couche d'ozone ?

Bruno FRANCO

Based on Mahieu et al.: *Recent Northern Hemisphere stratospheric HCl increase due to atmospheric circulation changes*, *Nature*, 515, 104-107, doi:10.1038/nature13857

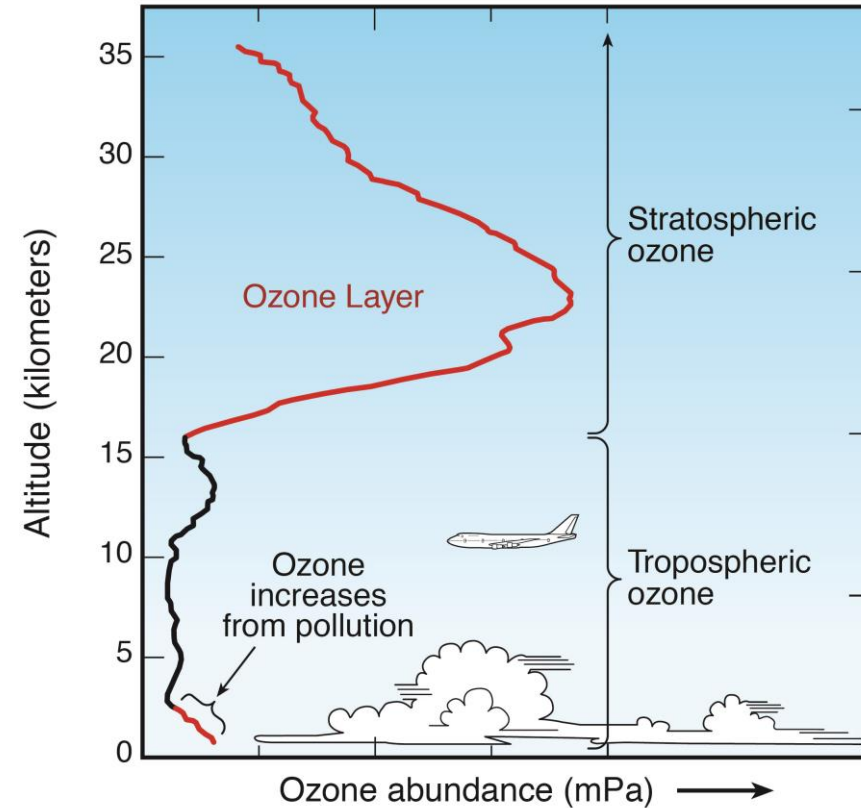
Assemblée générale AGO, ULg, 4 Décembre 2014

-> The largest O₃ production occurs in the tropical stratosphere



Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

Ozone in the Atmosphere

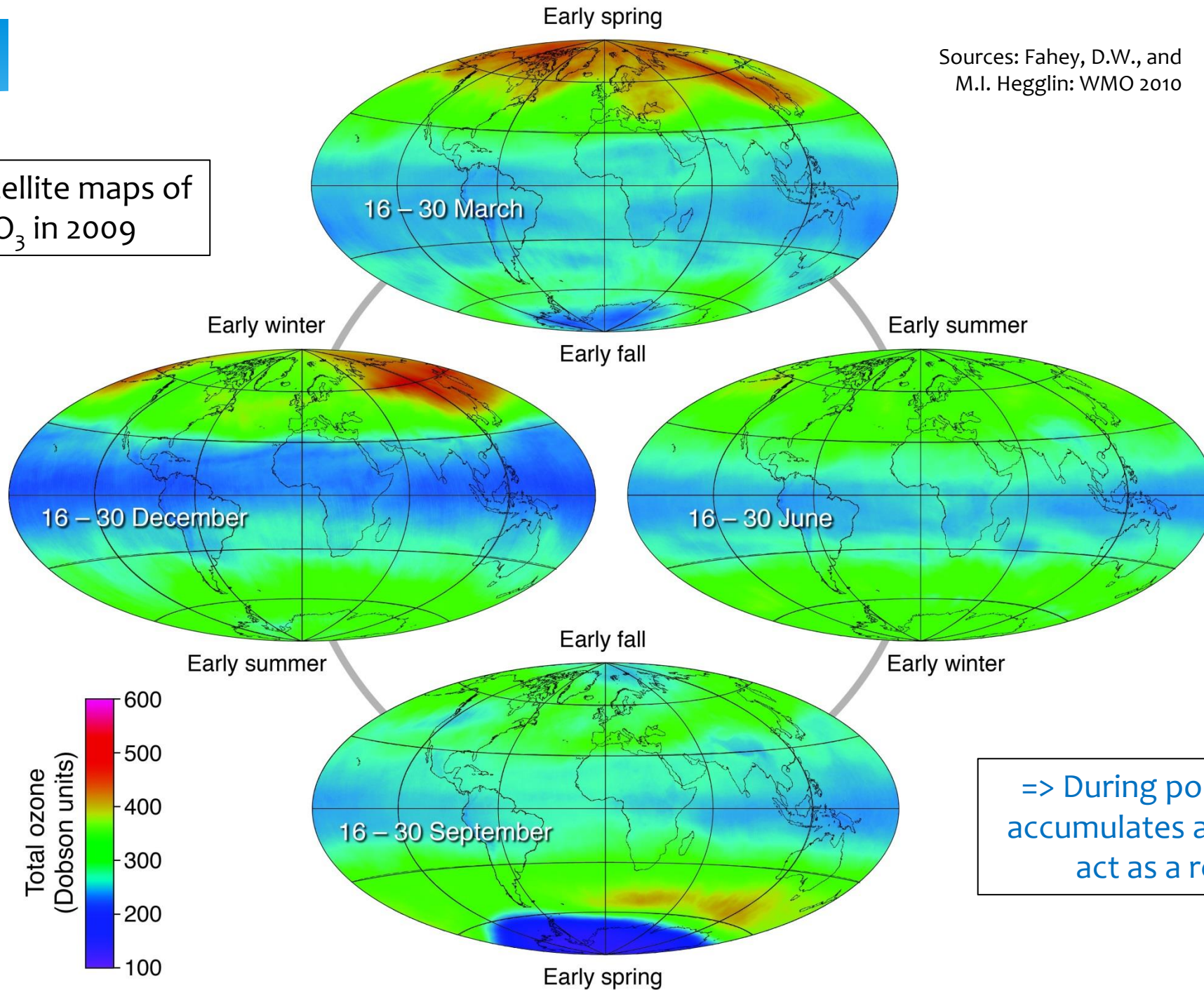


Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

=> Production/destruction of stratospheric O₃ normally **in balance**

Global satellite maps of total O₃ in 2009

Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

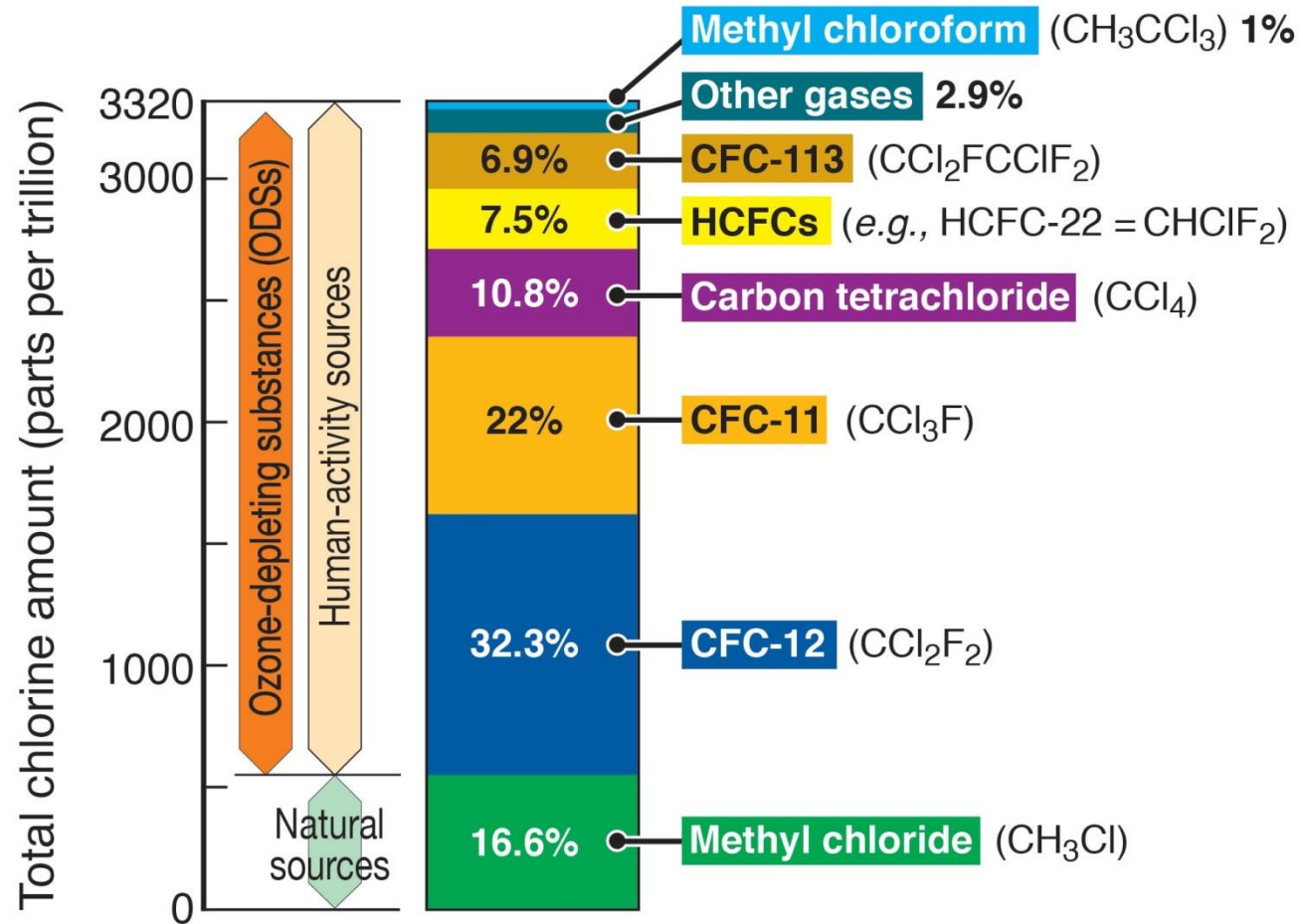


=> During polar night, O₃ accumulates and the poles act as a reservoir

=> But since the 1950s... the CFCs



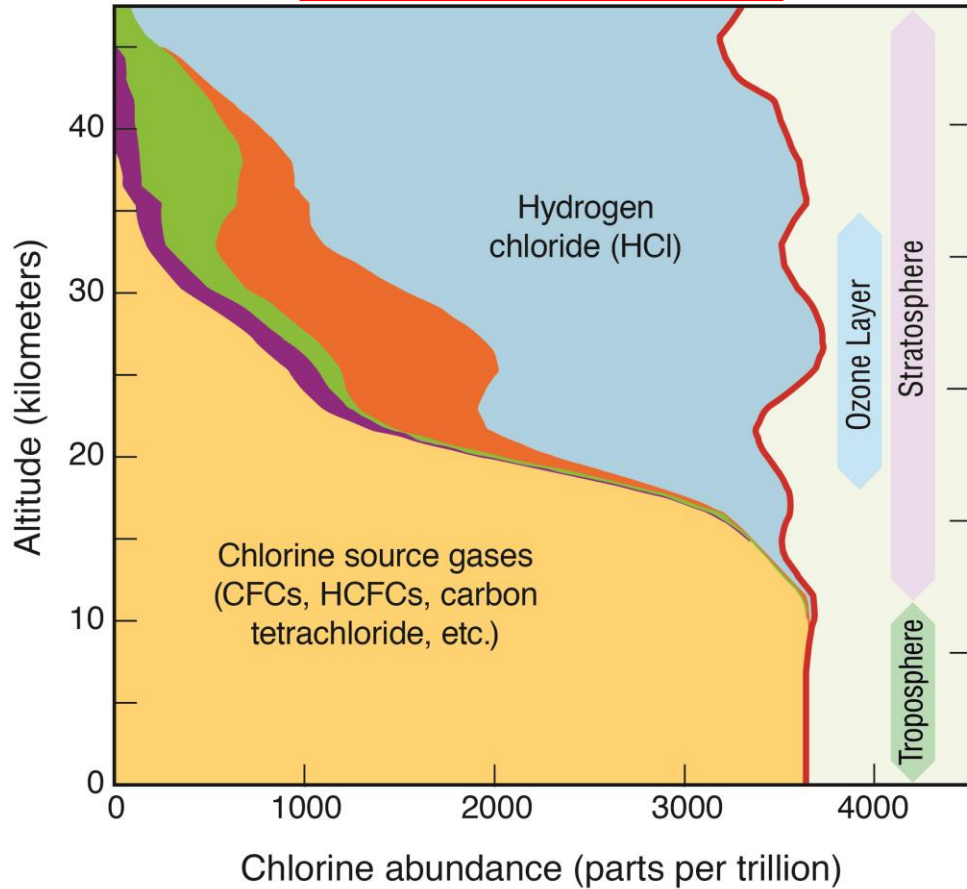
Chlorine Source Gases entering the stratosphere in 2008



Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

Measurements of Reactive Chlorine from Space

November 1994 (35° – 49°N)

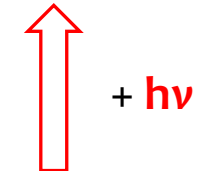
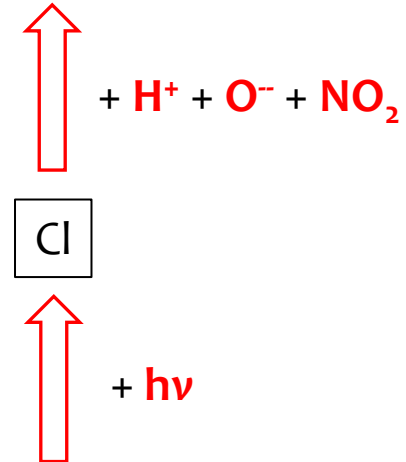


- Chlorine nitrate (CIONO₂)
- Chlorine monoxide (ClO)
- Other gases
- Available chlorine

=> at mid-latitudes

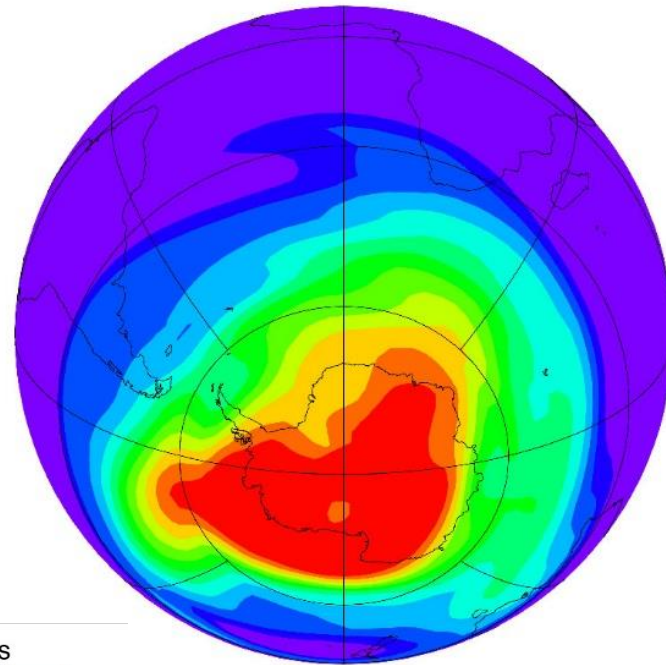


Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

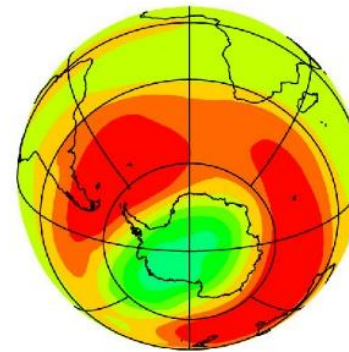


-> HCl and ClONO₂ as a reservoir of **UNREACTIVE** chlorine

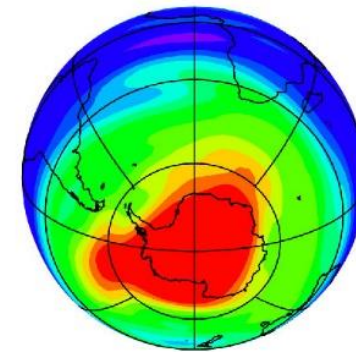
-> At the beginning of winter



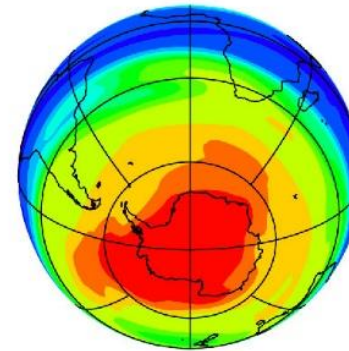
Ozone



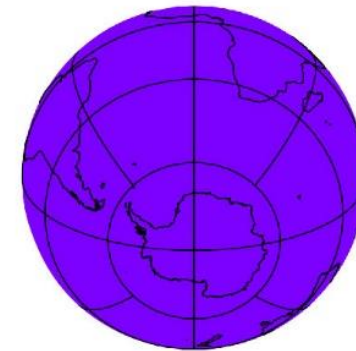
Temperature



Nitric acid (HNO₃)



Hydrogen chloride (HCl)



Chlorine monoxide (ClO)

Temperatures and chemical abundances

Low High

1800 ————— Ozone ————— 2800 ppb

-85 - - - - - Temperature - - - - - -60°C

1.0 ————— HNO₃ ————— 9.0 ppb

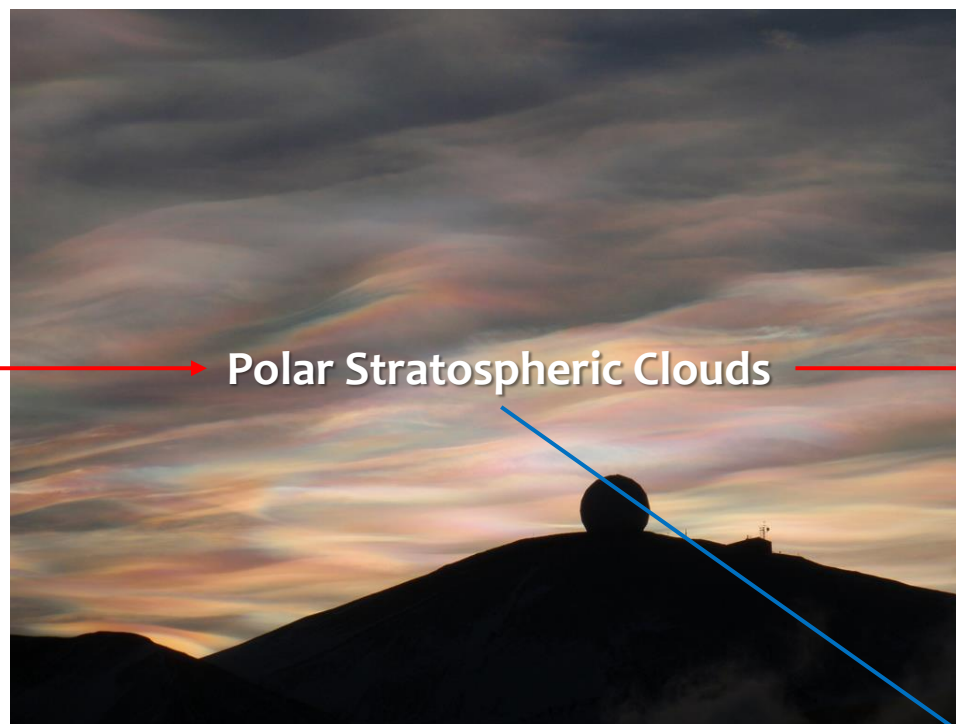
0.2 - - - - - HCl - - - - - 1.7 ppb

0.1 ————— ClO ————— 1.1 ppb

Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

-> But over high-latitudes... the PSCs

During the polar nights



HCl + ClONO₂ + ...

Polar Stratospheric Clouds

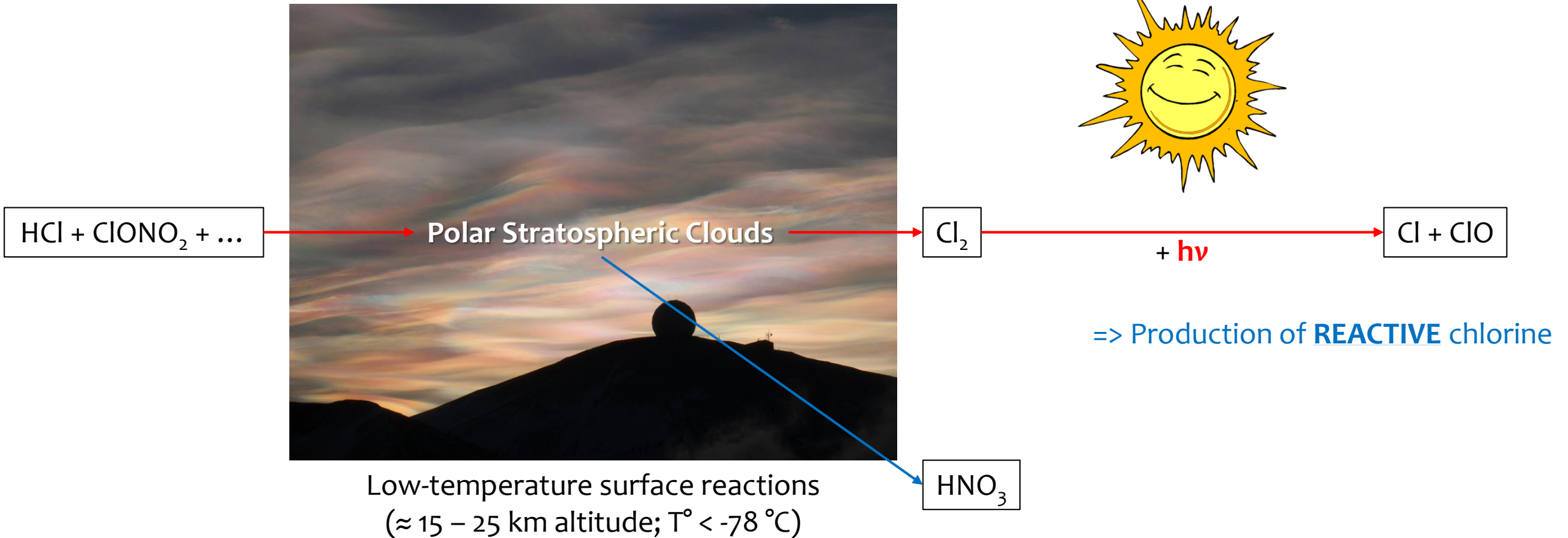
Cl₂

HNO₃

Low-temperature surface reactions
(≈ 15 – 25 km altitude; T° < -78 °C)

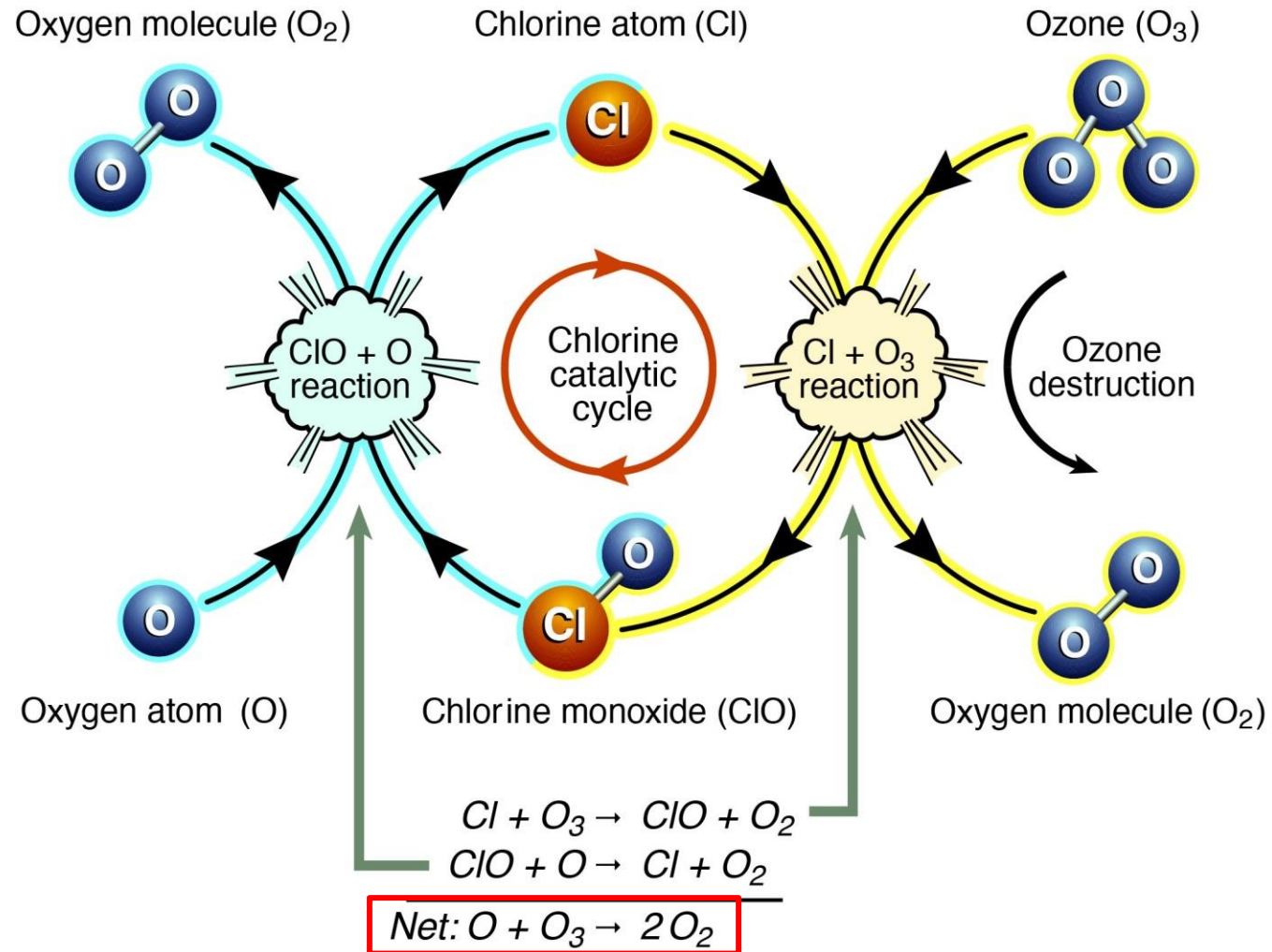
-> But over high-latitudes... the PSCs

During the polar nights

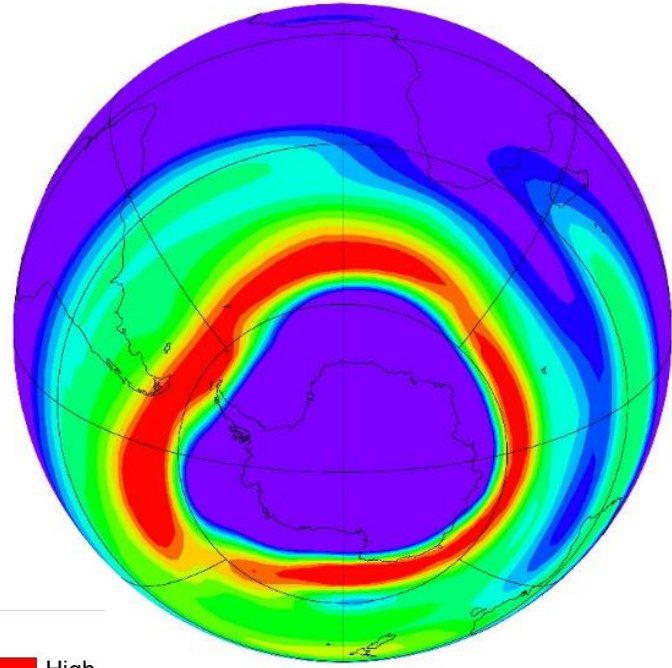


-> Chlorine catalytic cycle

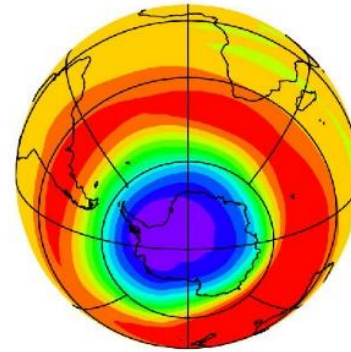
Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010



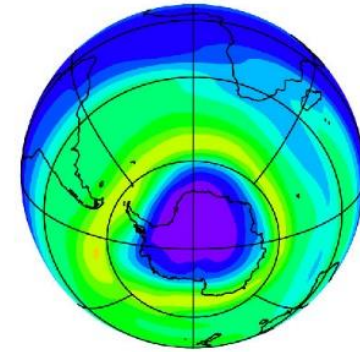
-> At the beginning of spring



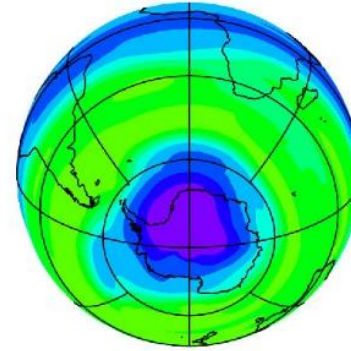
Ozone



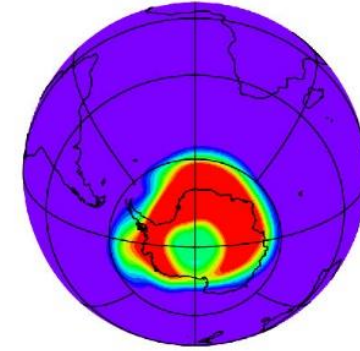
Temperature



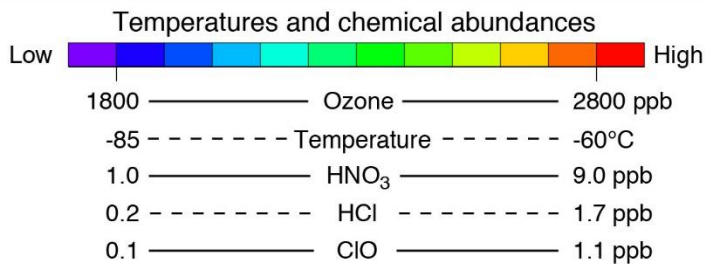
Nitric acid (HNO₃)



Hydrogen chloride (HCl)



Chlorine monoxide (ClO)

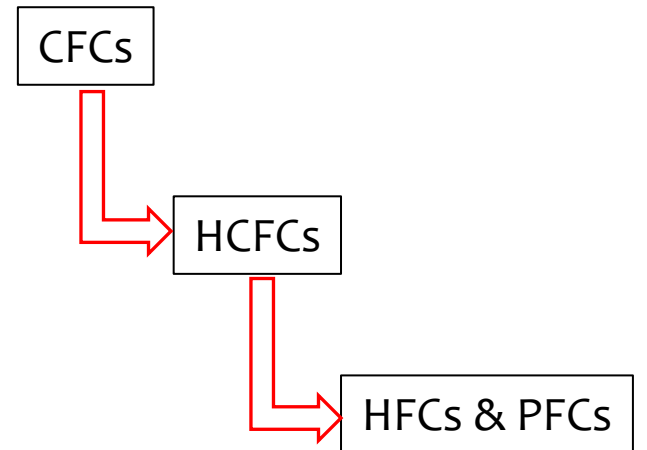
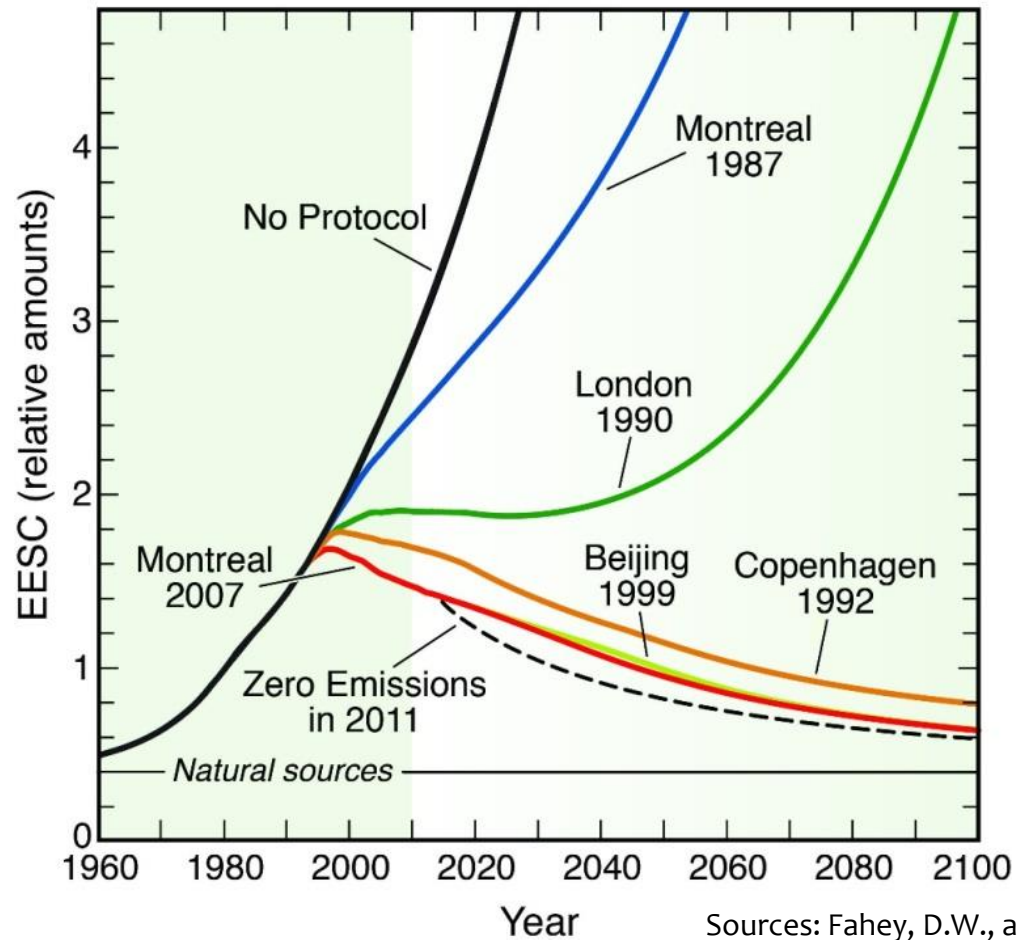


Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010



Effect of the Montreal Protocol

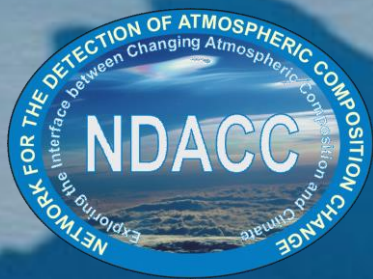
Long-term changes in equivalent effective stratospheric chlorine (EESC)

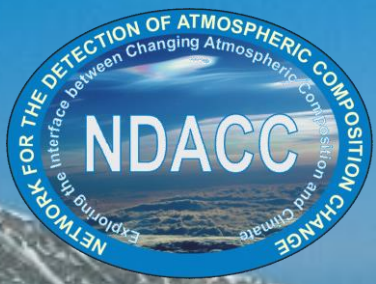


Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010

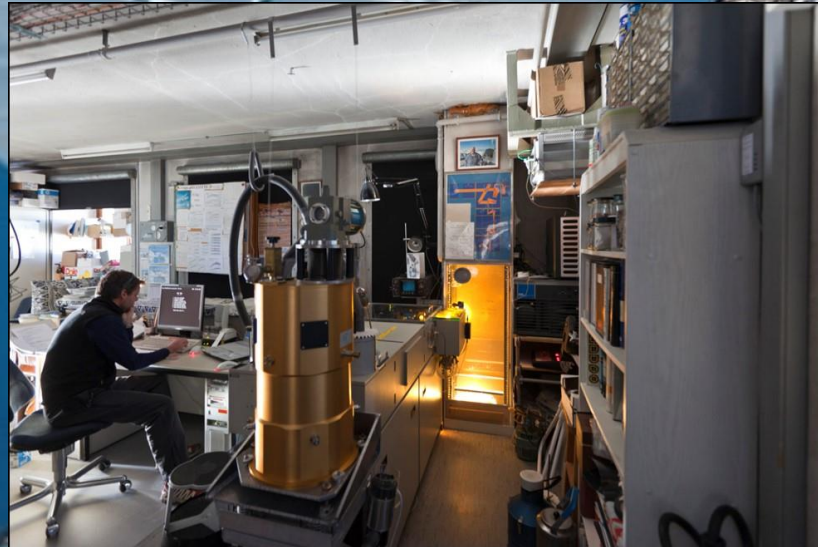
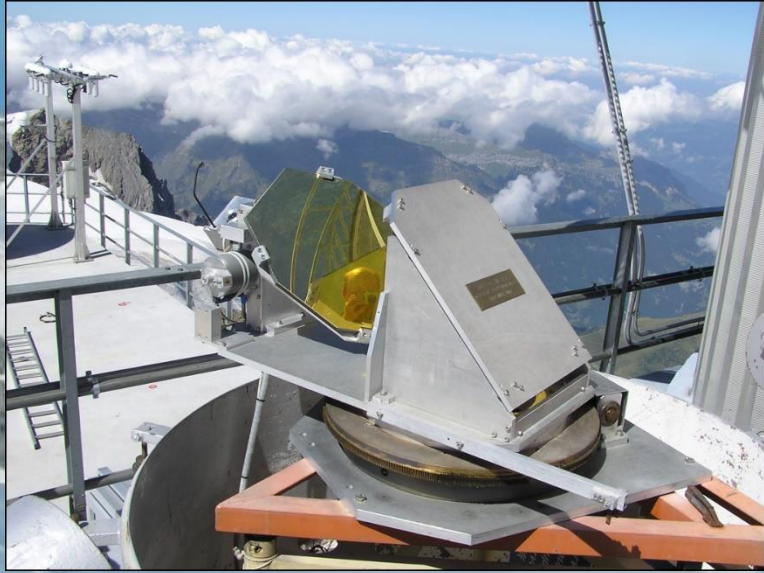
Long-term FTIR monitoring activities at Jungfrauoch (Swiss Alps; 3580 m a.s.l.)

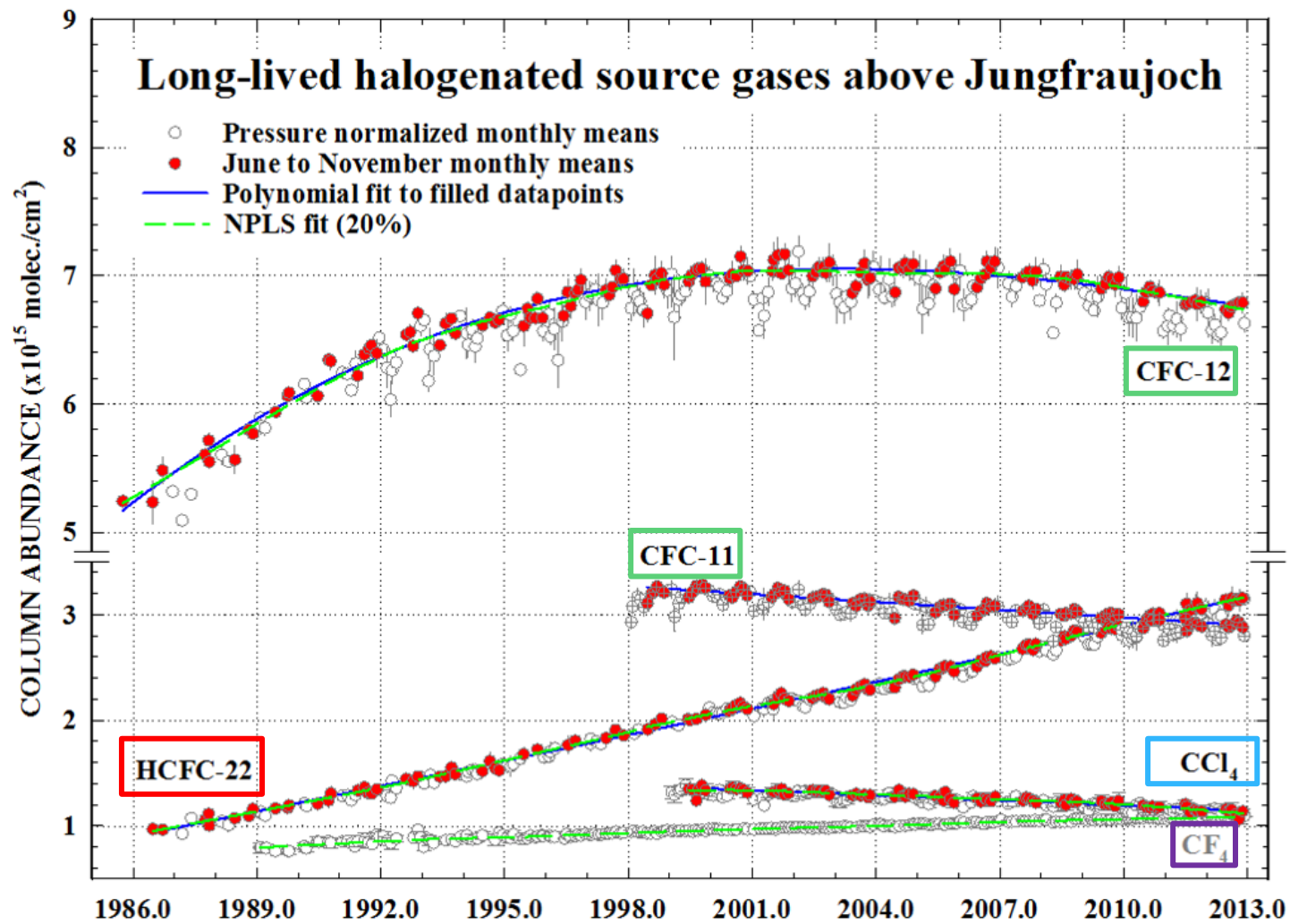
Within the framework of the Network for the Detection of Atmospheric Composition Change (NDACC; see www.ndacc.org)

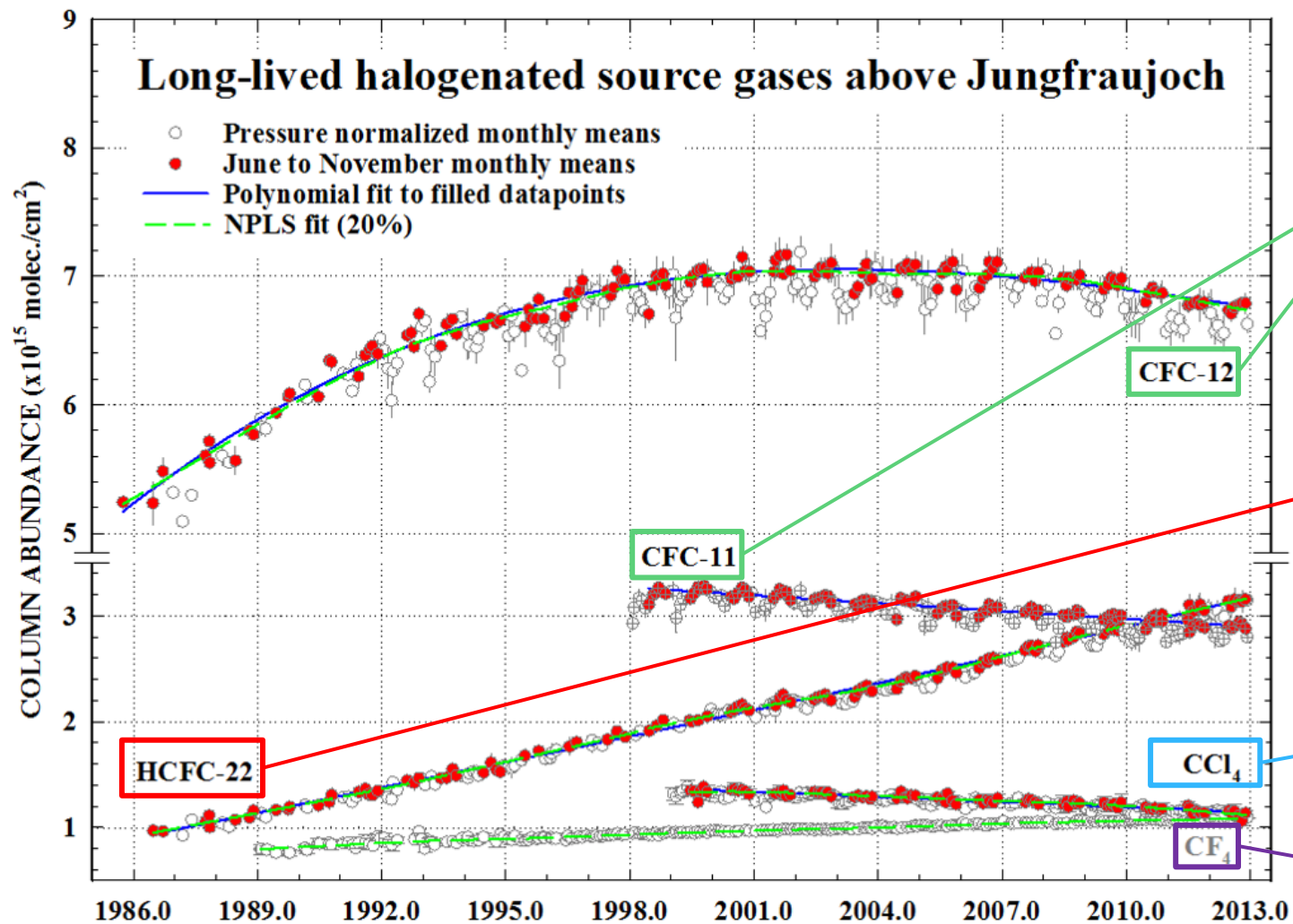




Long-term FTIR monitoring activities at Jungfrauoch (Swiss Alps; 3580 m a.s.l.)





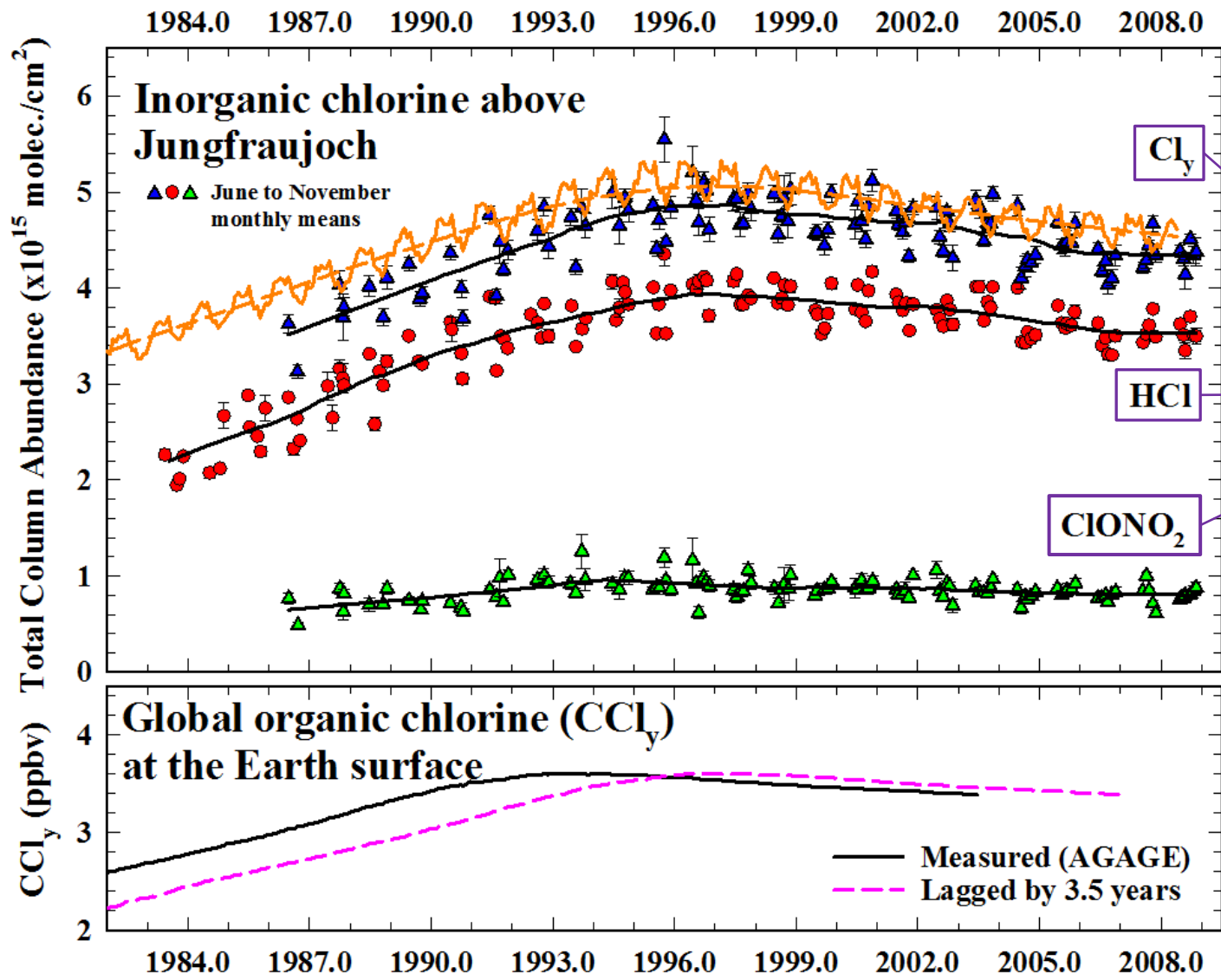


Phased out by the Montreal Protocol

Phased out by the Montreal Protocol amendments

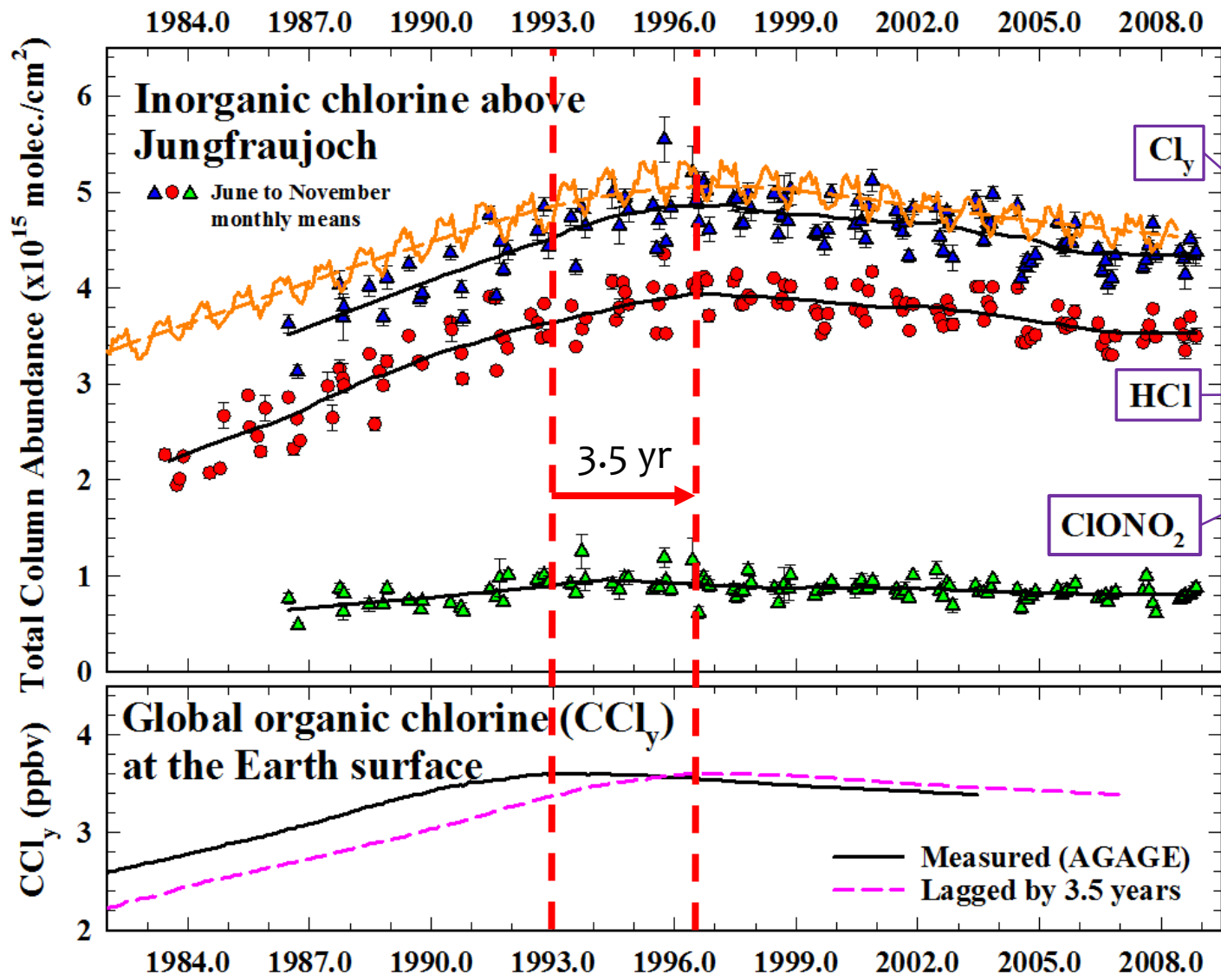
???

CF₄ (PFC) ≈ 50,000 yr lifetime !!!



$Cl_y \approx HCl + ClONO_2$

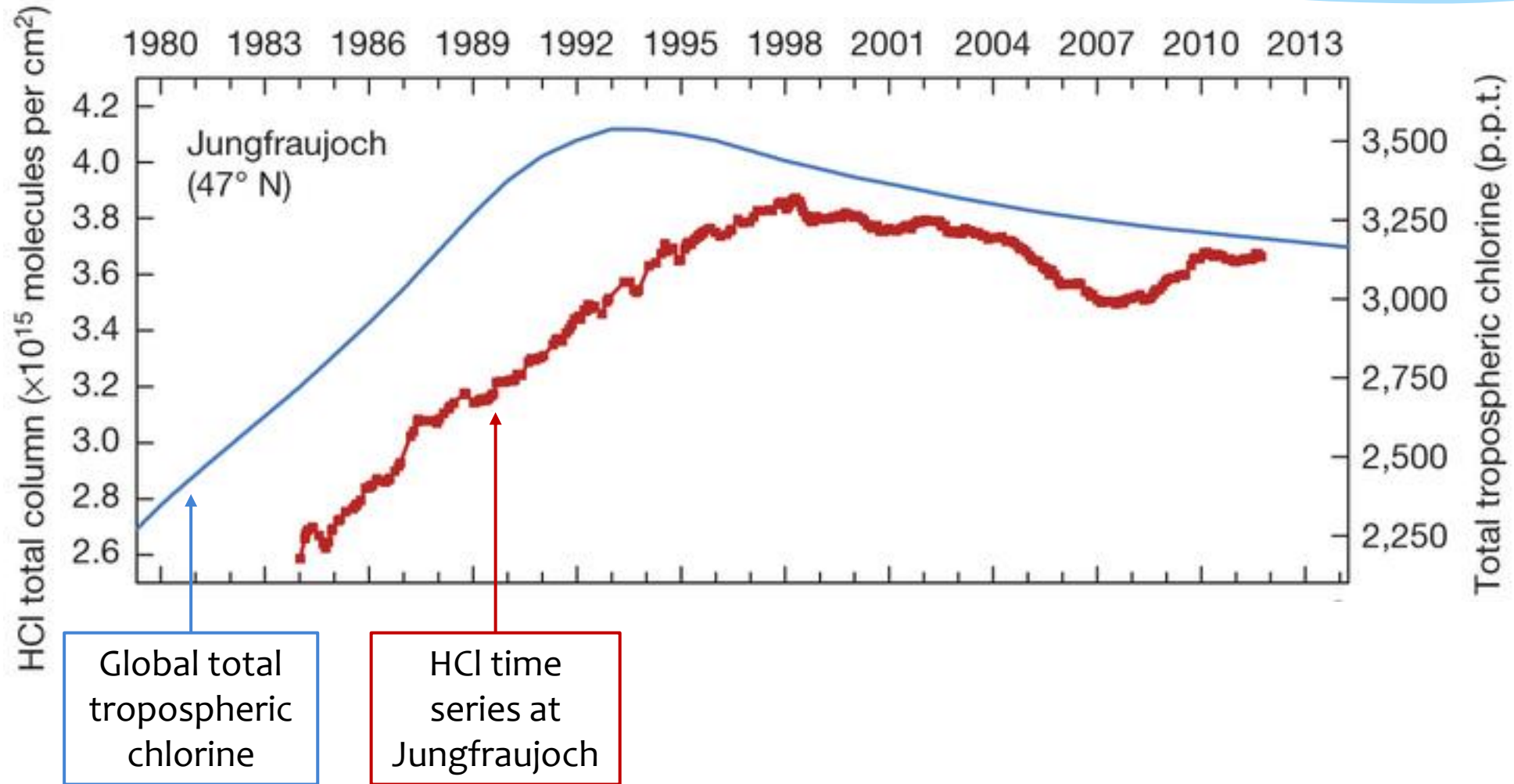
$CCl_y = CFCs + HCFCs + CCl_4 + \dots$



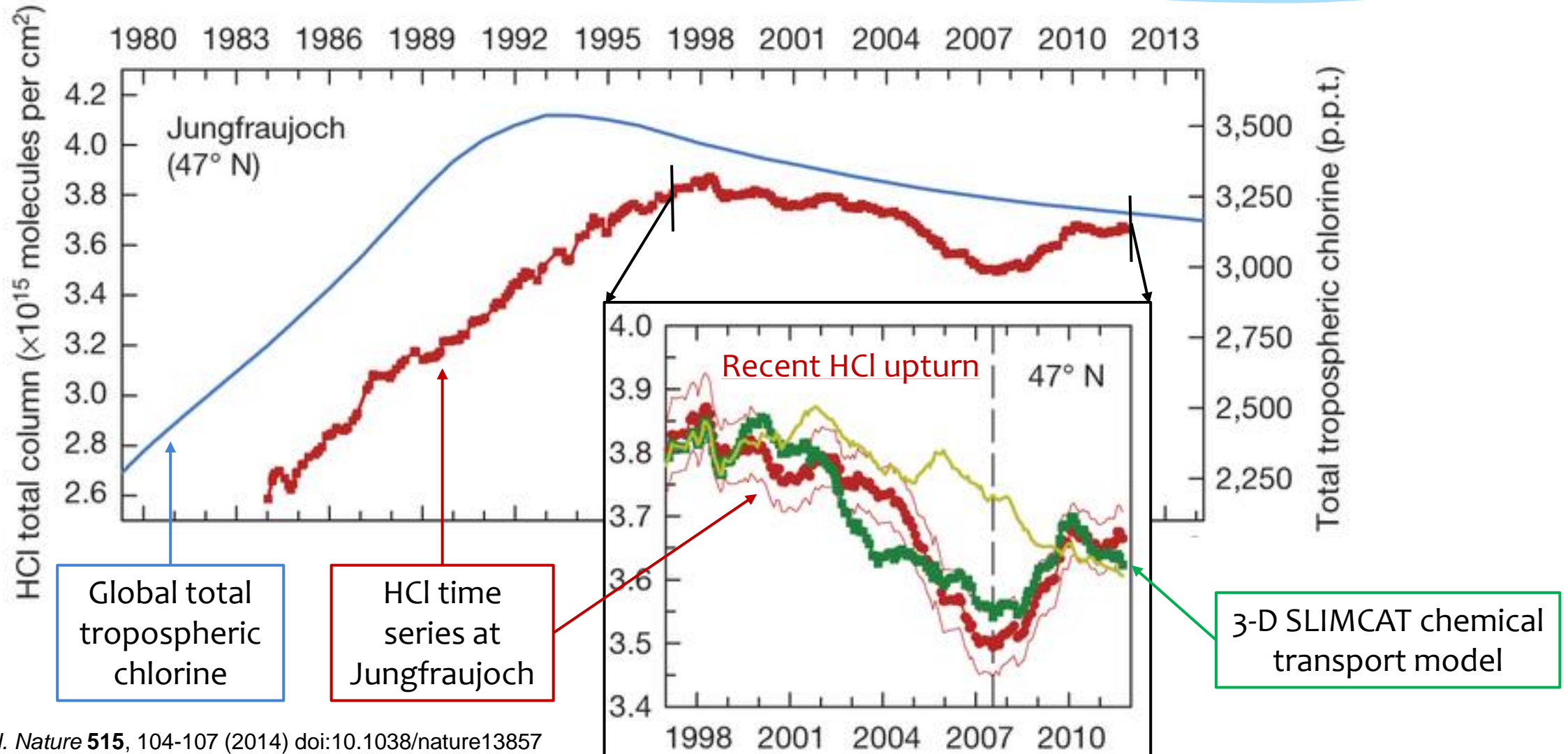
$$\text{Cl}_y \approx \text{HCl} + \text{ClONO}_2$$

$$\text{CCl}_y = \text{CFCs} + \text{HCFCs} + \text{CCl}_4 + \dots$$

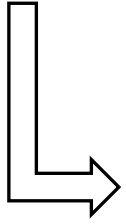
-> Monitoring of HCl at Jungfraujoch



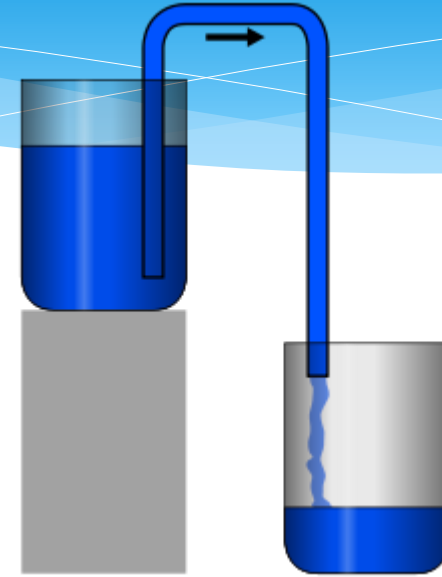
-> Monitoring of HCl at Jungfrauoch



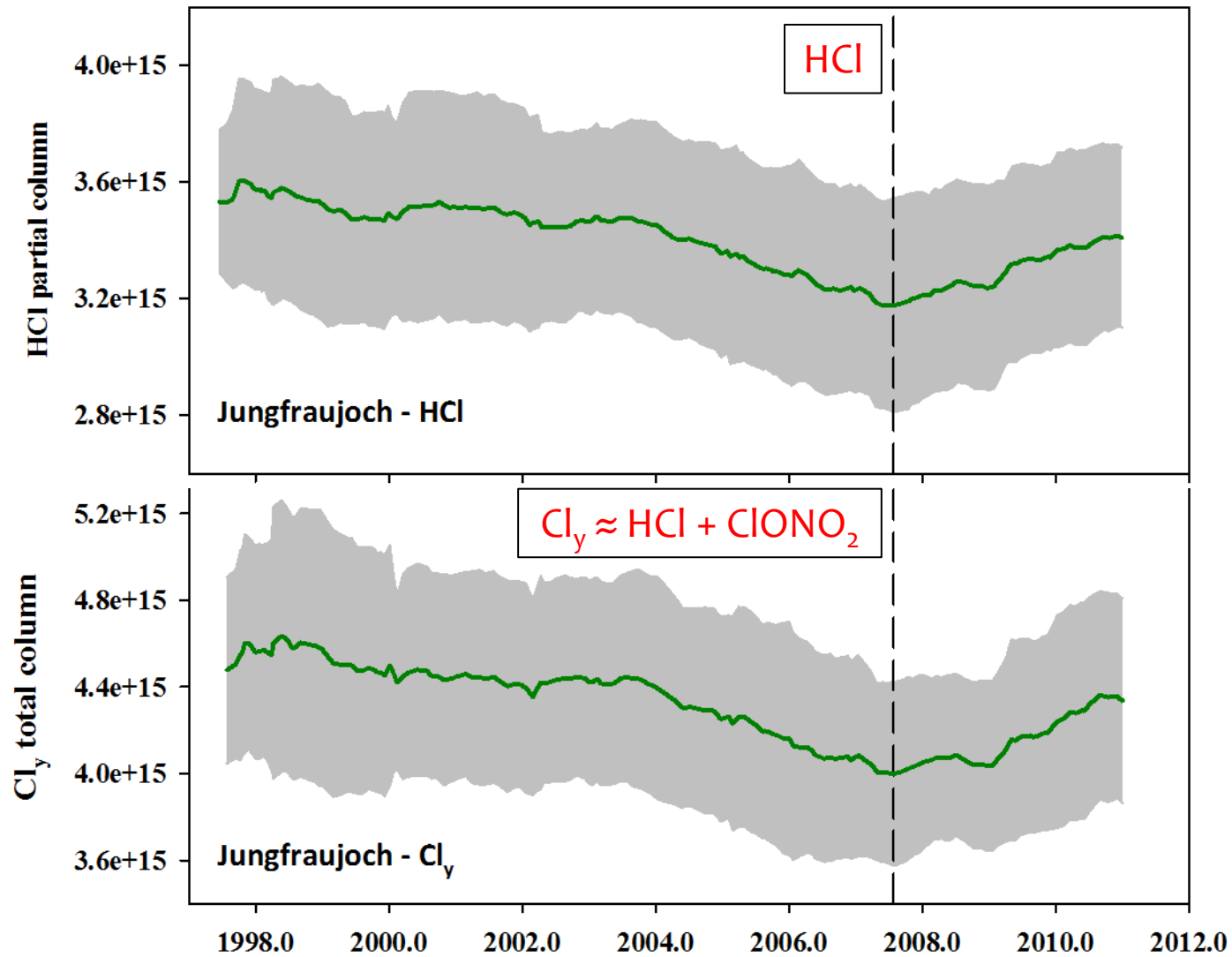
-> Recent HCl upturn: hypothesis?



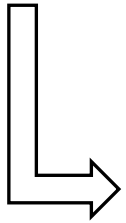
Transfer between stratospheric chlorine reservoirs?



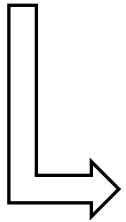
-> Transfer between stratospheric chlorine reservoirs?



-> Recent HCl upturn: hypothesis?

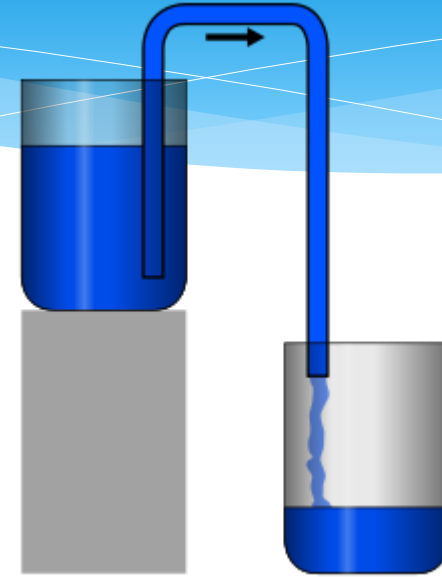


Transfer between stratospheric chlorine reservoirs?

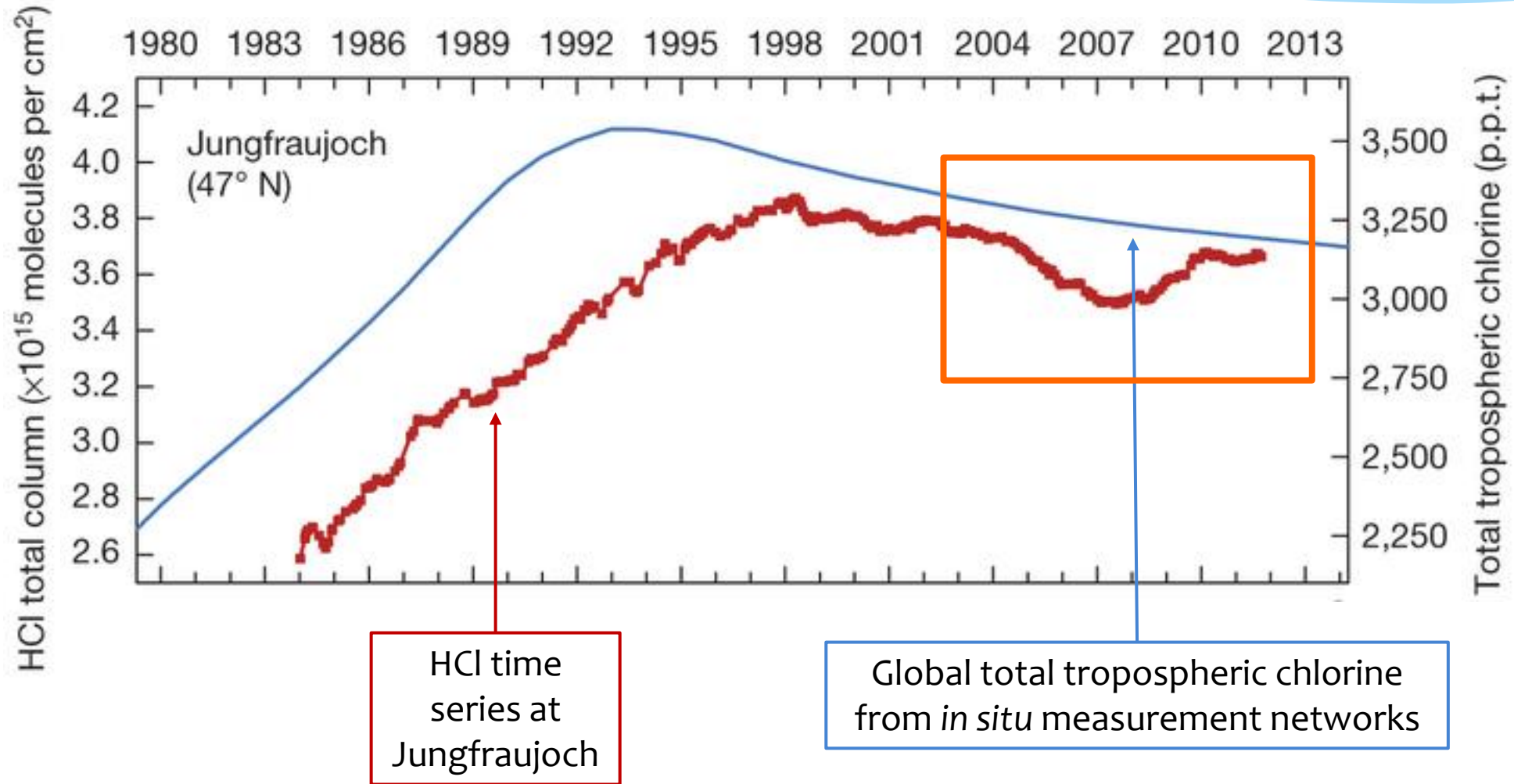


« Rogue » emissions of HCl precursors?

- Unregulated by the Montreal Protocol



-> Monitoring of HCl at Jungfraujoch



-> Recent HCl upturn: hypothesis?

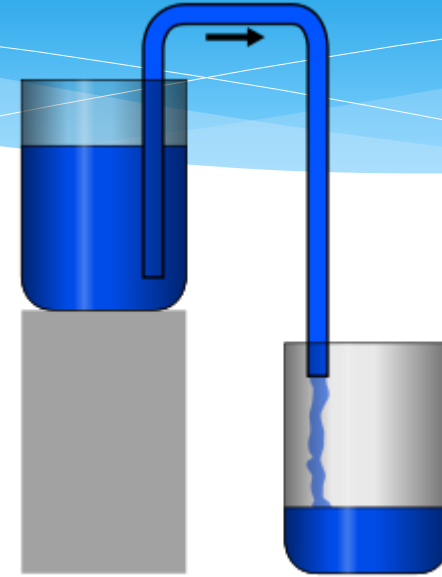
Transfer between stratospheric chlorine reservoirs?



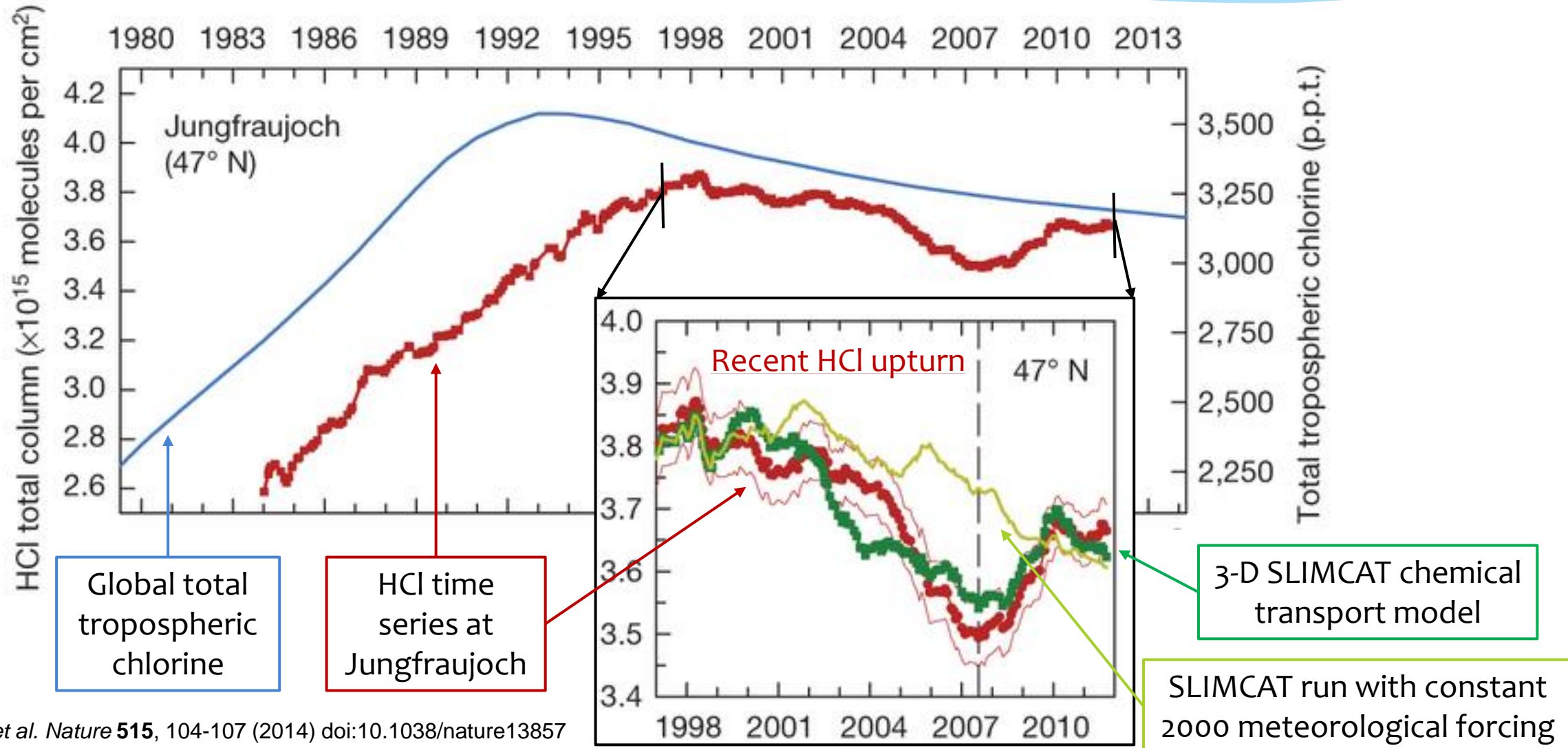
« Rogue » emissions of HCl precursors?

- Not monitored by measurement networks

(Atmospheric) circulation change?



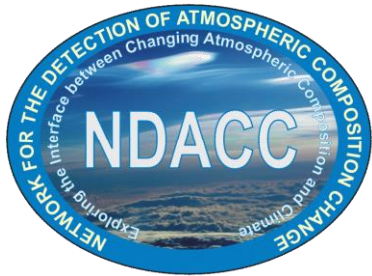
-> Monitoring of HCl at Jungfrauoch



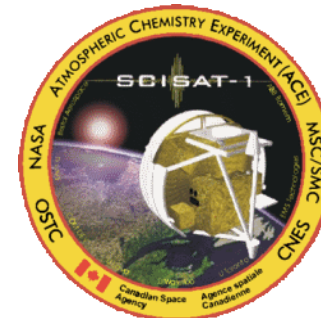
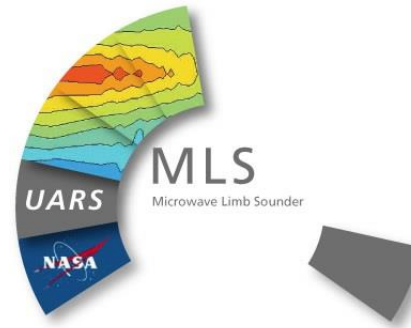
-> Investigation of the recent HCl upturn

3 satellite missions
(GOZCARDS data)

8 FTIR NDACC sites



- Ny-Ålesund (79 °N)
- Thule (77 °N)
- Kiruna (68 °N)
- Jungfraujoch (47 °N)
- Tsukuba (36 °N)
- Izana (28 °N)
- Wollongong (34 °S)
- Lauder (45 °S)

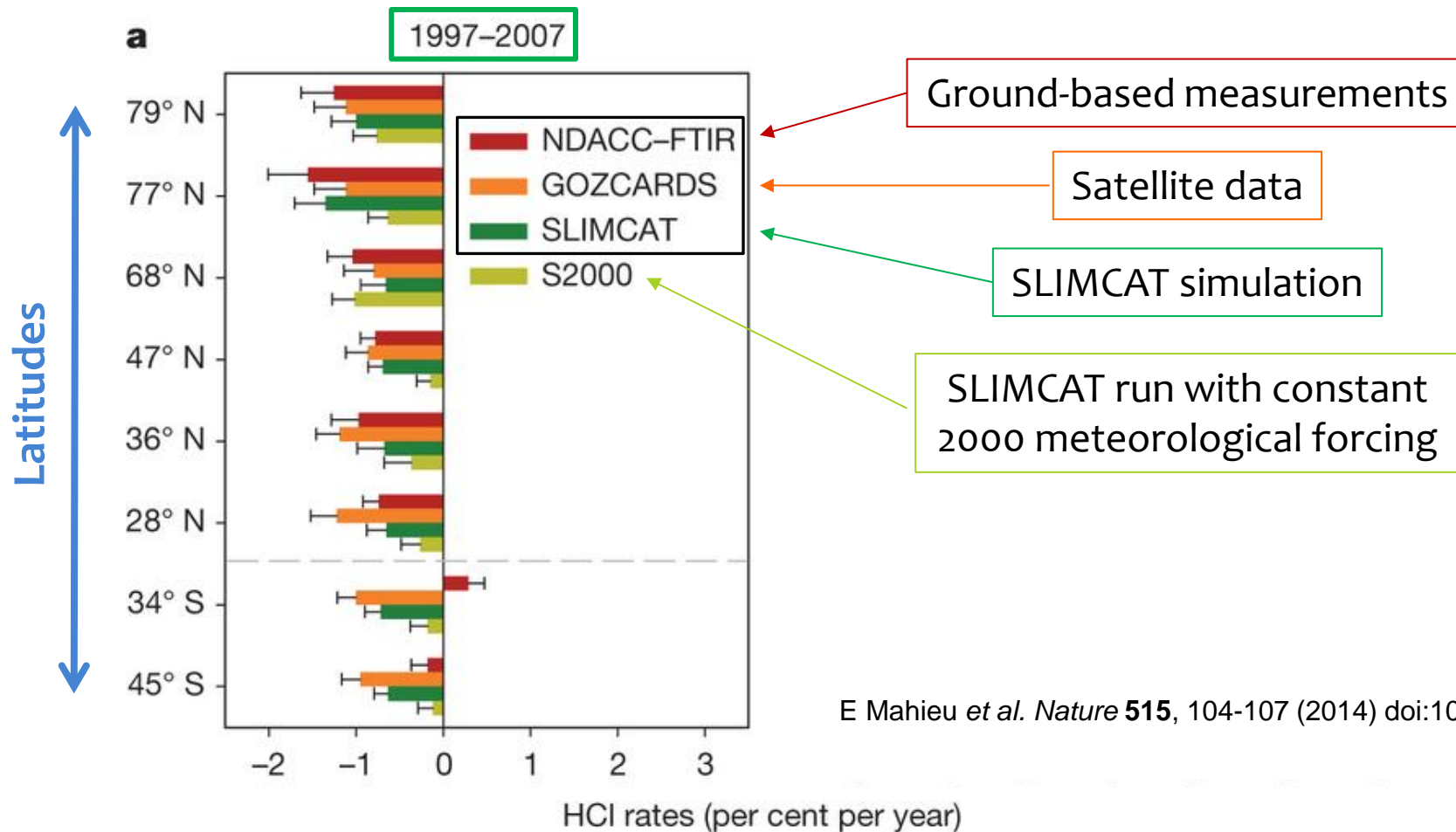


- Aura/MLS (Microwave Limb Sounder v3.3)
- HALOE (HALogen Occultation Experiment v19)
- ACE-FTS (Atmospheric Chemistry Experiment – Fourier Transform Spectrometer v2.2)

2 3-D chemical transport models

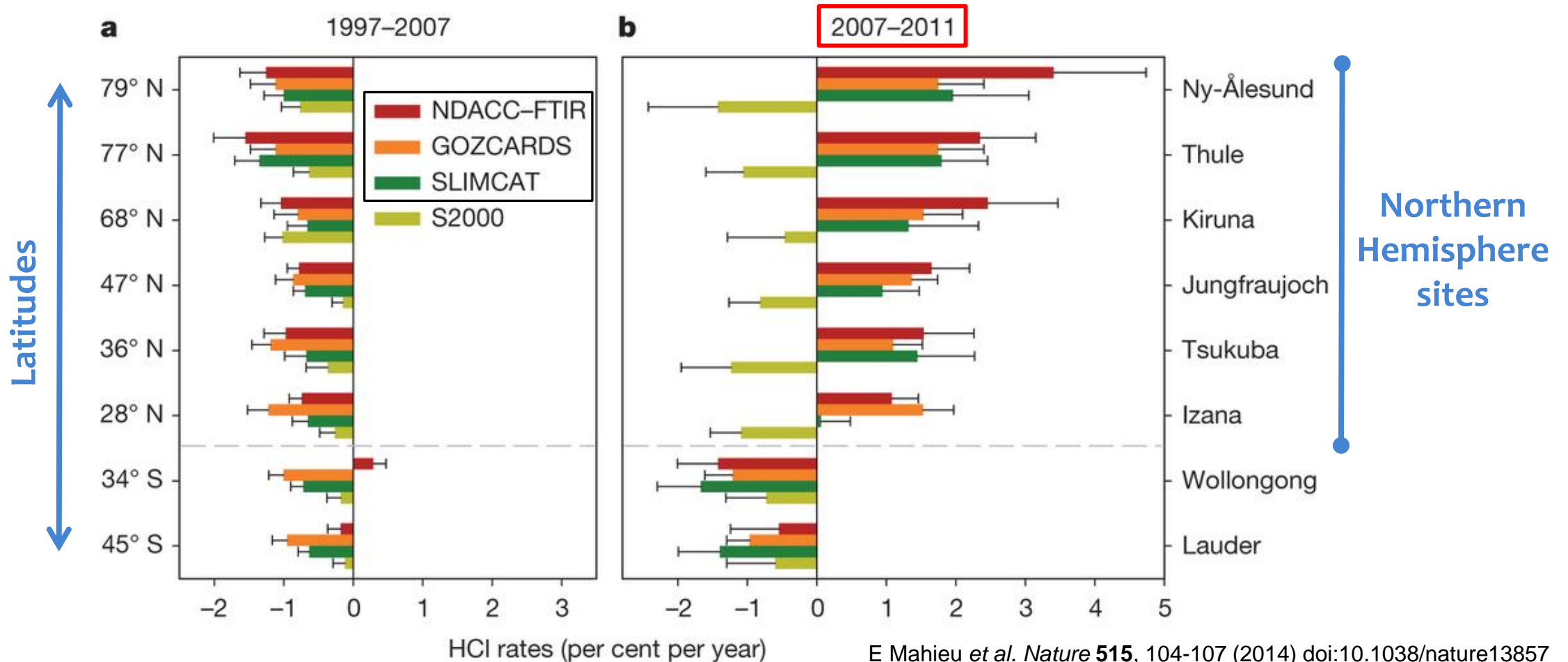
- SLIMCAT
National Centre for Atmospheric Science,
University of Leeds
- KASIMA
Karlsruhe Institute of Technology (KIT)

-> HCl relative rates of change for 8 NDACC sites



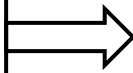
E Mahieu *et al.* *Nature* **515**, 104-107 (2014) doi:10.1038/nature13857

-> HCl relative rates of change for 8 NDACC sites



-> Evolution of stratospheric HCl from satellite observations

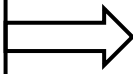
≈ 30 km altitude
(upper stratosphere)



Southern Hemisphere

Northern Hemisphere

≈ 20 km altitude
(lower stratosphere)

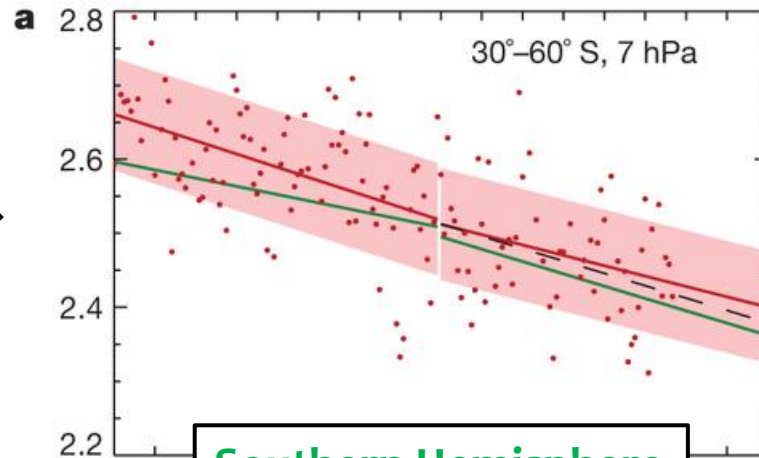


1998 2002 2006 2010

1998 2002 2006 2010

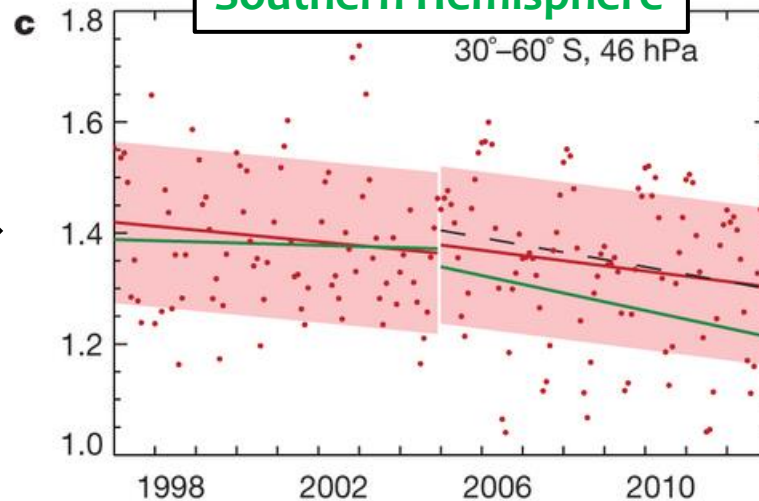
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Northern Hemisphere

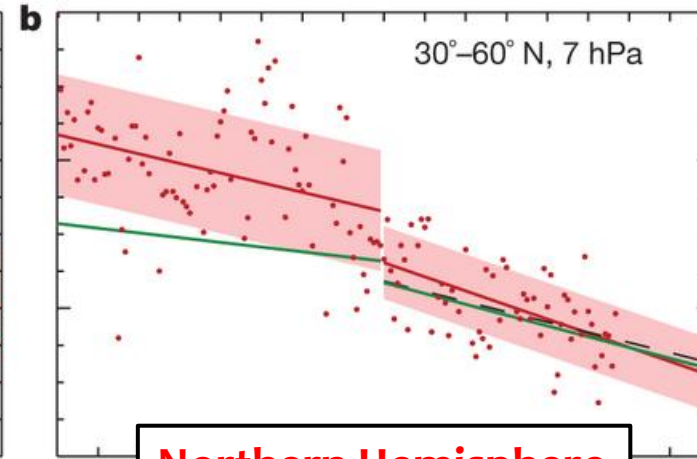
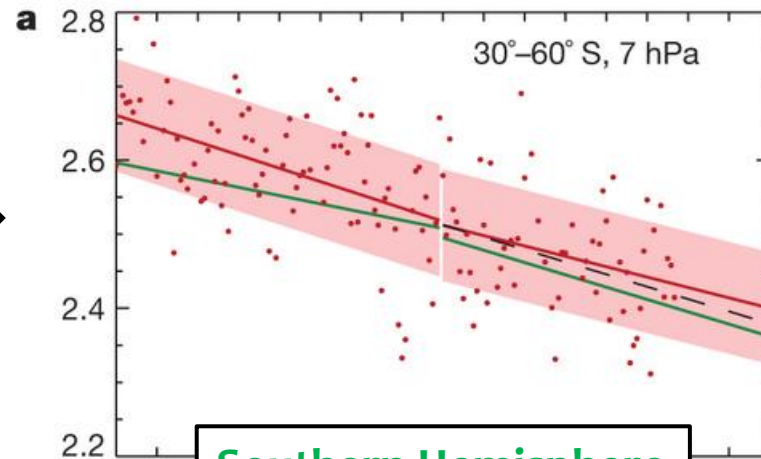
≈ 20 km altitude
(lower stratosphere)



1998 2002 2006 2010

-> Evolution of stratospheric HCl from satellite observations

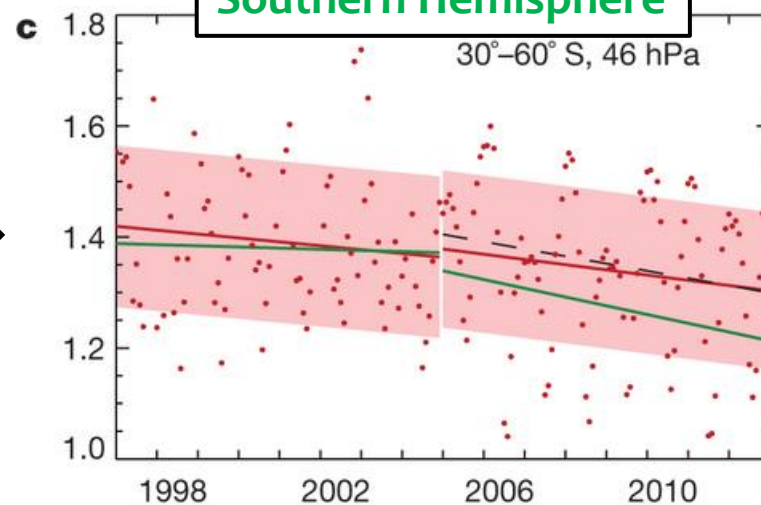
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Southern Hemisphere

Northern Hemisphere

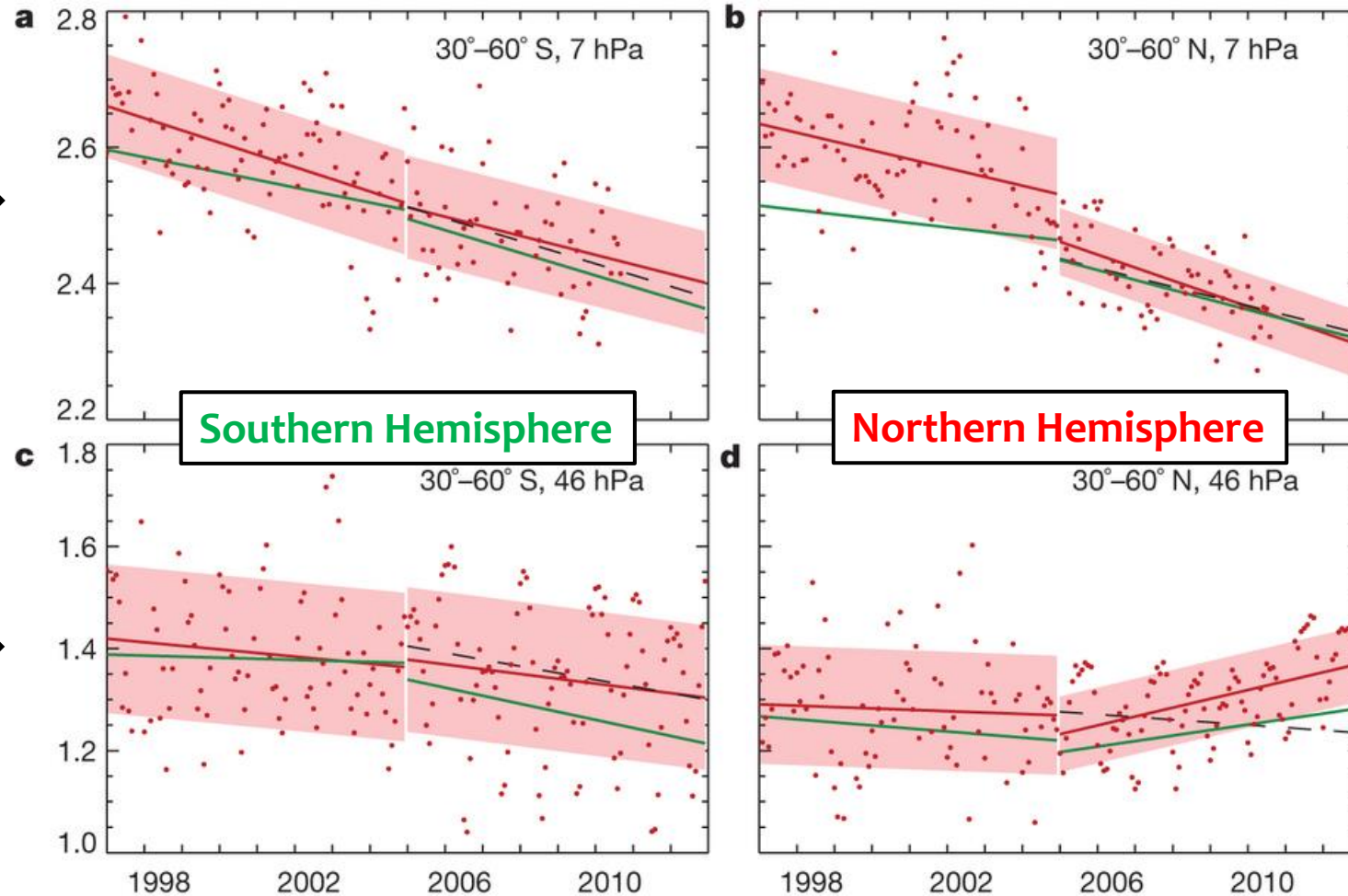
≈ 20 km altitude
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1998 2002 2006 2010

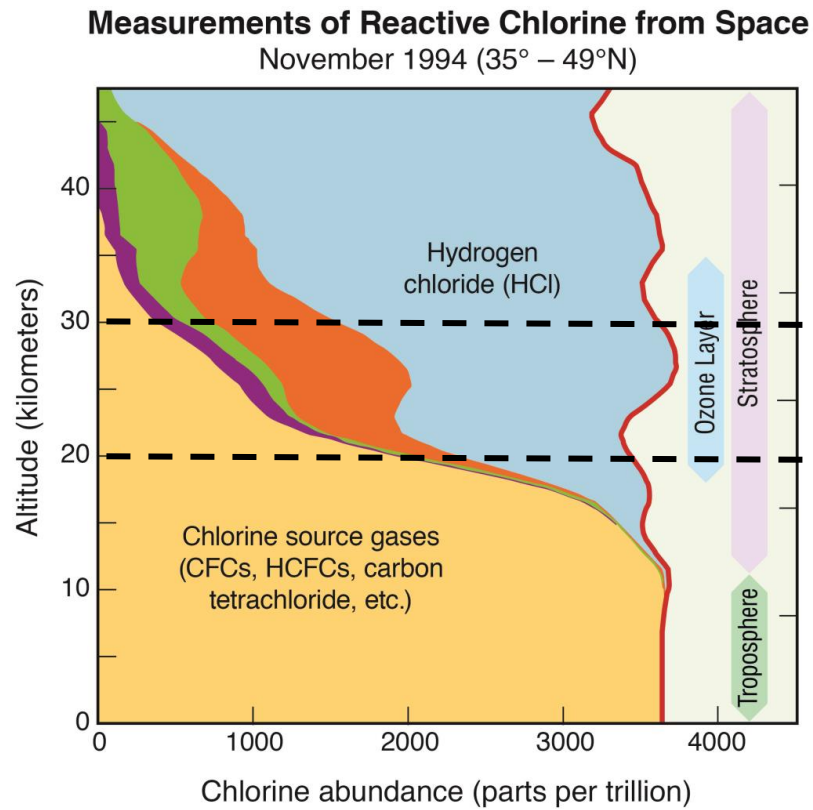
-> Evolution of stratospheric HCl from satellite observations

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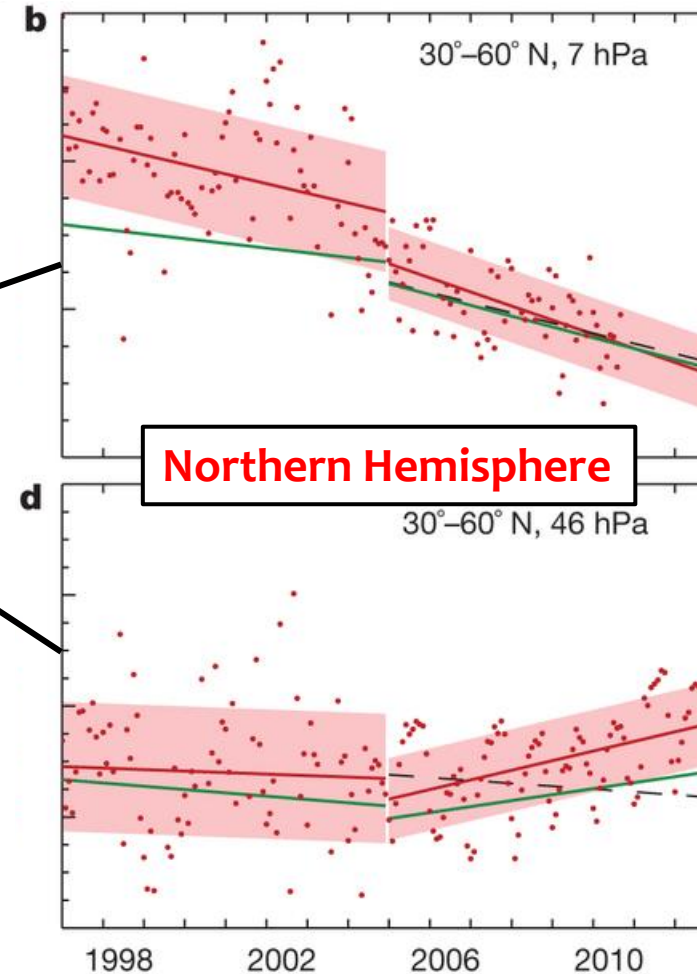
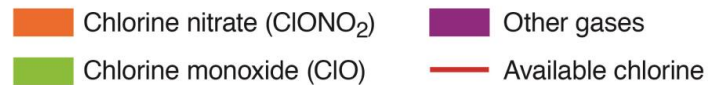


≈ 20 km altitude
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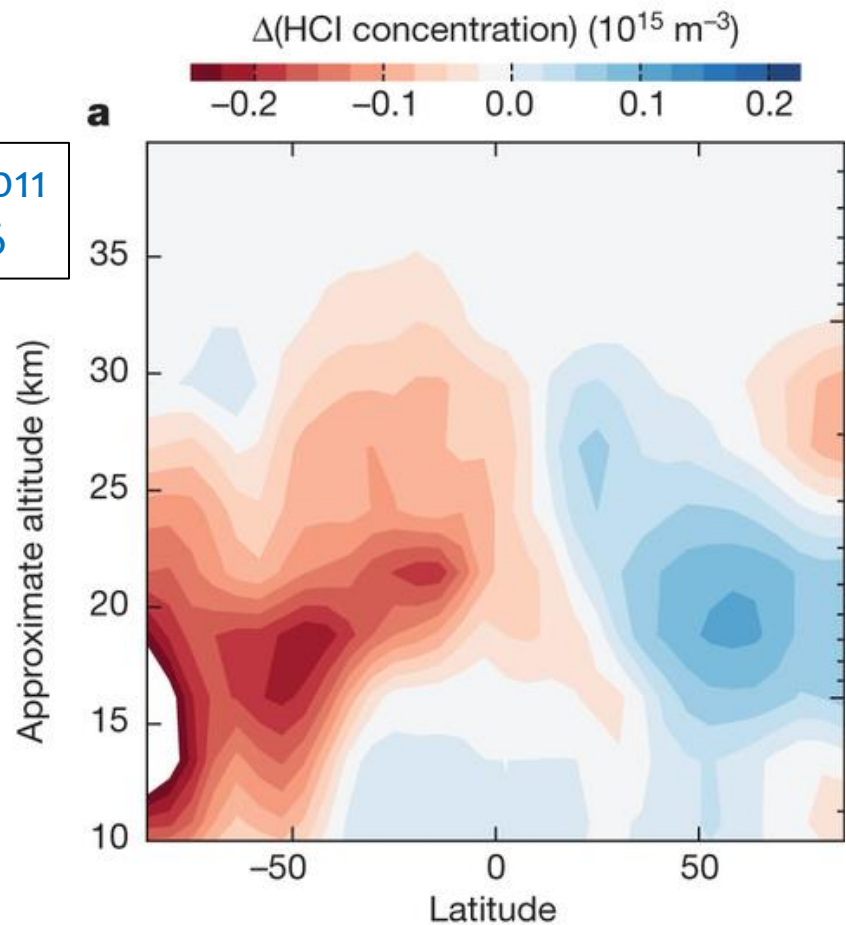


Sources: Fahey, D.W., and M.I. Hegglin: WMO 2010



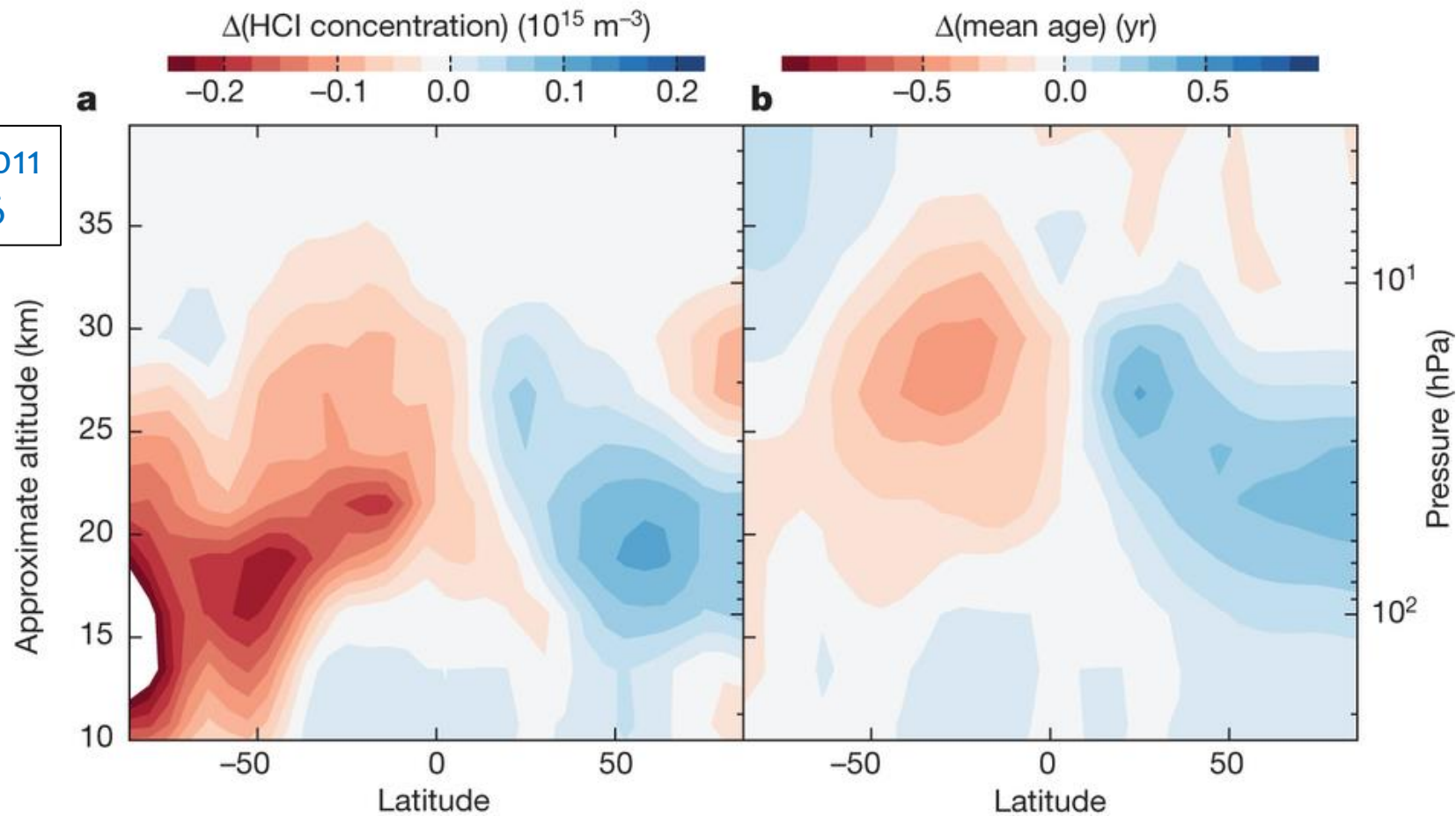
-> Spatial distribution of the HCl concentration and age-of-air changes

Between 2010/2011
and 2005/2006



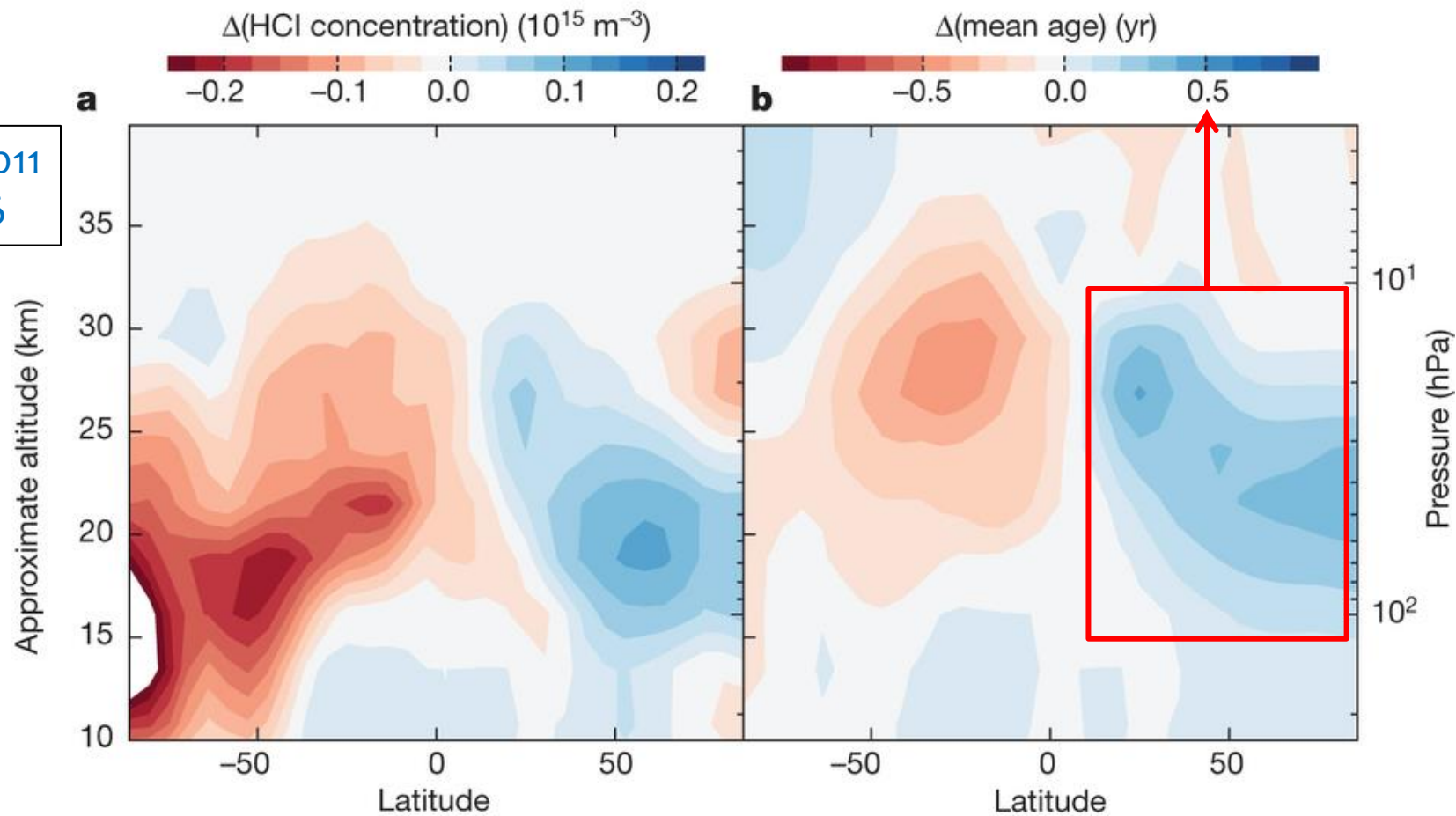
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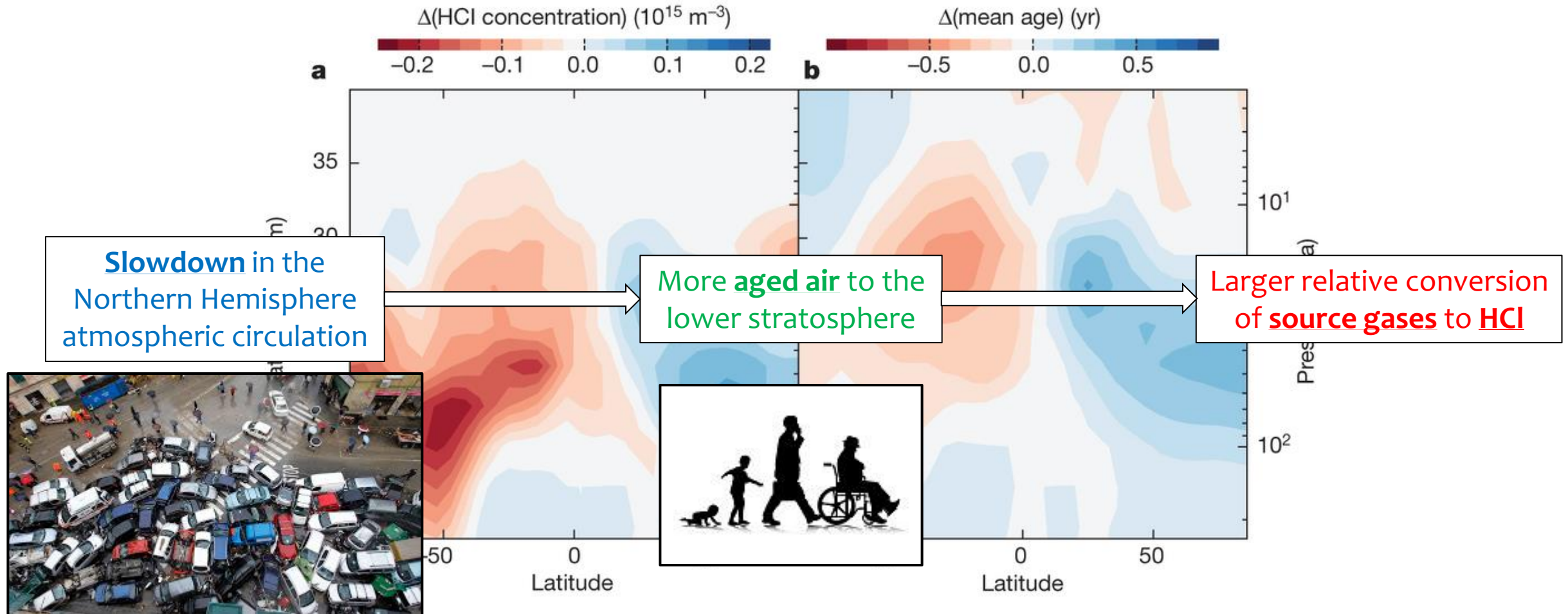


-> Spatial distribution of the HCl concentration and age-of-air changes

Between 2010/2011
and 2005/2006



-> Spatial distribution of the HCl concentration and age-of-air changes



Conclusion

- The Montreal protocol is **still on track**
 - => HCl over the 1997 – 2011 period = -0.5 %/yr
 - => overall reduction of the stratospheric chlorine loading



The Montreal Protocol **25** years of Ozone Protection

Conclusion

- The Montreal protocol is **still on track**
 - => HCl over the 1997 – 2011 period = -0.5 %/yr
 - => overall reduction of the stratospheric chlorine loading
- Short-term dynamical variability
 - => influence on other stratospheric tracers?
 - => causes still **unidentified**



The Montreal Protocol **25** years of Ozone Protection

**TO BE
CONTINUED...** →