Reconciling modelled and observed age of air through SF_6 sinks

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MACClim

EGU General Assembly 2020

Middle Atmosphere composition and feedbacks in a changing climate

7 May 2020

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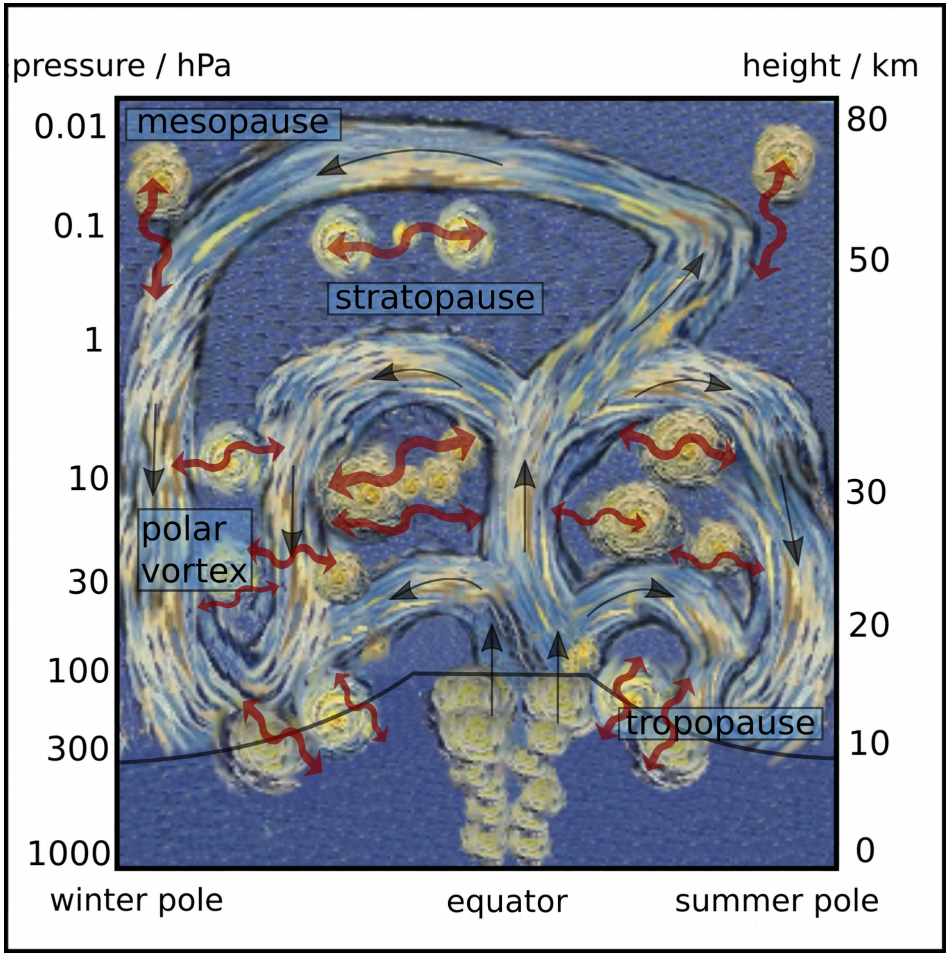
Age of Air (AoA)

- AoA ~ time elapsed since air entered stratosphere
- AoA can be derived from measurable tracers e.g. sulphur-hexafluoride: SF_6



R. Eichinger, & V. van Gogh, 2019

We use SF_6 as a tracer for AoA

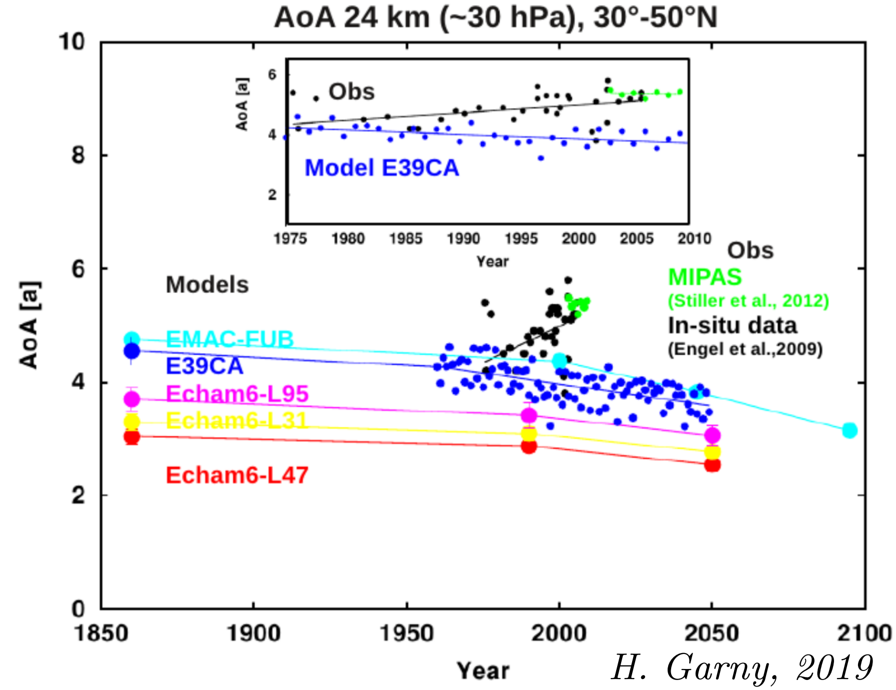
+ No sources of SF_6 in middle atmosphere
+ Relatively linear boundary conditions (near-linear increase of emissions over recent decades)
- Not fully inert: (mesospheric) sinks

- **Disagreements between observations and model simulations of AoA:** stratospheric air often older in observations (e.g. Dietmüller et al., 2018, Stiller et al., 2012, Ploeger et al., 2019) than in models

- **Discrepancies in AoA trends:** models show clear decrease of AoA over time (due to modelled acceleration of BDC), observations (e.g. Engel et al., 2009, Ray et al., 2014) show (non-significant) positive AoA trend

- **Discrepancies in tracer (SF_6) lifetime:**

Ravishankara et al., 1993:	3200 years
Reddmann et al., 2001:	400 – 10000 years
Kovács et al., 2017:	1278 years
Ray et al., 2017:	580 – 1400 years



H. Garny, 2019 personal communication

Can the inclusion of SF_6 sinks in model simulations help to reconcile simulations and observations?

Simulation Setup

- EMAC v2.54.0 — **E**CHAM **M**ESSy **A**tmospheric **C**hemistry (Jöckel et al., 2010, Jöckel et al., 2016)
- T42L90MA — T42 horizontal (2.8°x2.8°) resolution, 90 levels in the vertical, explicitly resolved middle atmosphere dynamics
- SF6 submodel — Accounts for explicit calculation of SF_6 sinks
- 4 Tracers — linear and non-linear tracer with and without sinks

Idealized Tracer
No sinks
Linear growth

	NO SINKS	WITH SINKS
LINEAR	NS, lin	WS, lin
NON-LINEAR (SF_6)	NS, SF_6	WS, SF_6

'Thought Experiments'
Artificial Tracers
For completeness

Realistic Tracer
With sinks
Non-linear growth

SF6 submodel explicitly calculates SF_6 sinks

- Based on Reddmann et al. (2001)
- SF_6 loss governed by:
  - Photodissociation
  - Electron Attachment
  - Reactions with reactant species: HCl, H, O_2, O_3, O_3P, N_2
    - Species prescribed by ESCiMo RC1-base-07 transient hindcast simulation (Jöckel et al., 2016)

$$SF_6 + h\nu \rightarrow SF_5 + F$$

$$SF_6 + e^- \rightarrow (SF_6^-)^* \longrightarrow SF_6^-$$

$$SF_6^- \xrightarrow{HCl,\ HNO_3,\ SO_2} products$$

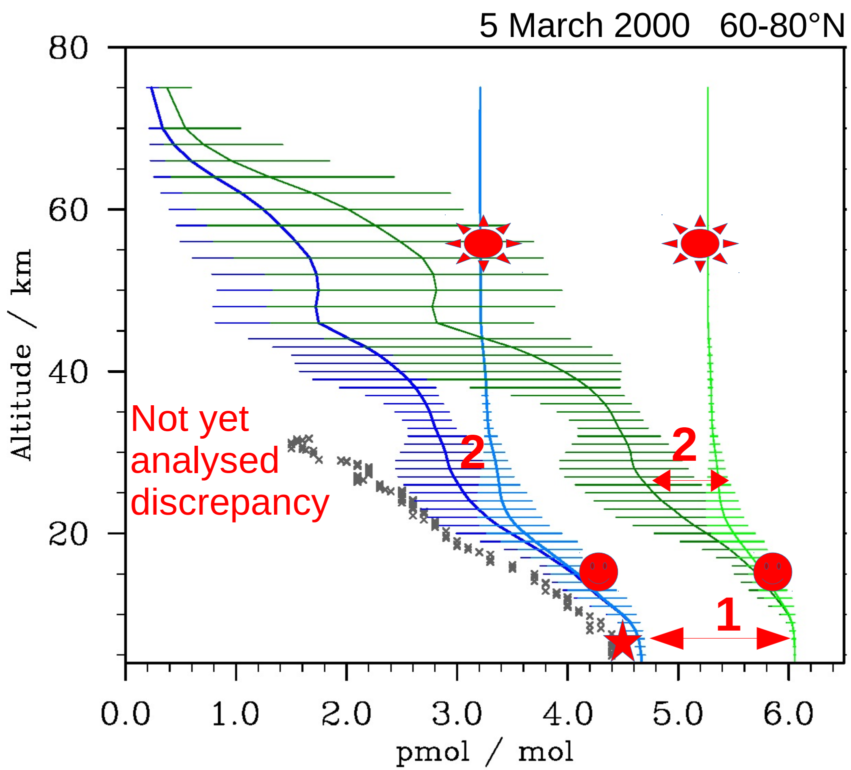
$$SF_6^- \xrightarrow{H} SF_5^+ + HF$$

$$SF_6 + O^+ \rightarrow SF_5^+ + OF$$

$$SF_6 + N_2^+ \rightarrow SF_5^+ + NF$$

$$SF_6 + O_2^- \rightarrow SF_6^- + O_2$$

SF_6 tracer: mixing ratios from SD simulation & balloon flights



1 Surface emissions (SF_6 ↔ lin)

2 Sinks ↔ Without Sinks

★ SF_6 lower boundary conditions

● No sink effect at this altitude

✹ No Sinks!

(WS, SF_6)

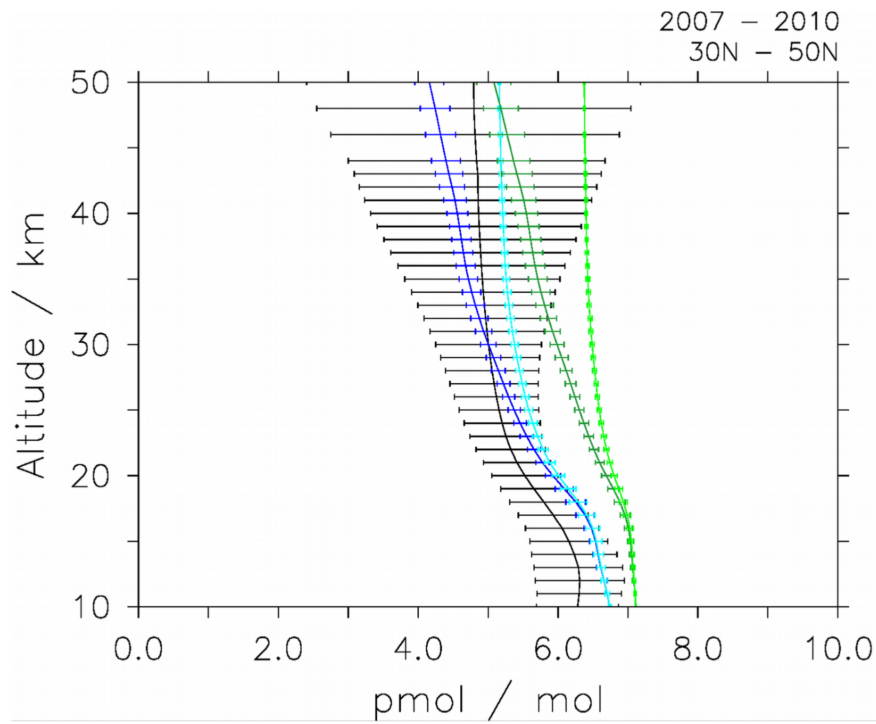
(NS, SF_6)

(WS, lin)

(NS, lin)

x Balloon Flights March 2002
Ray et al. 2017

— MIPAS 2019



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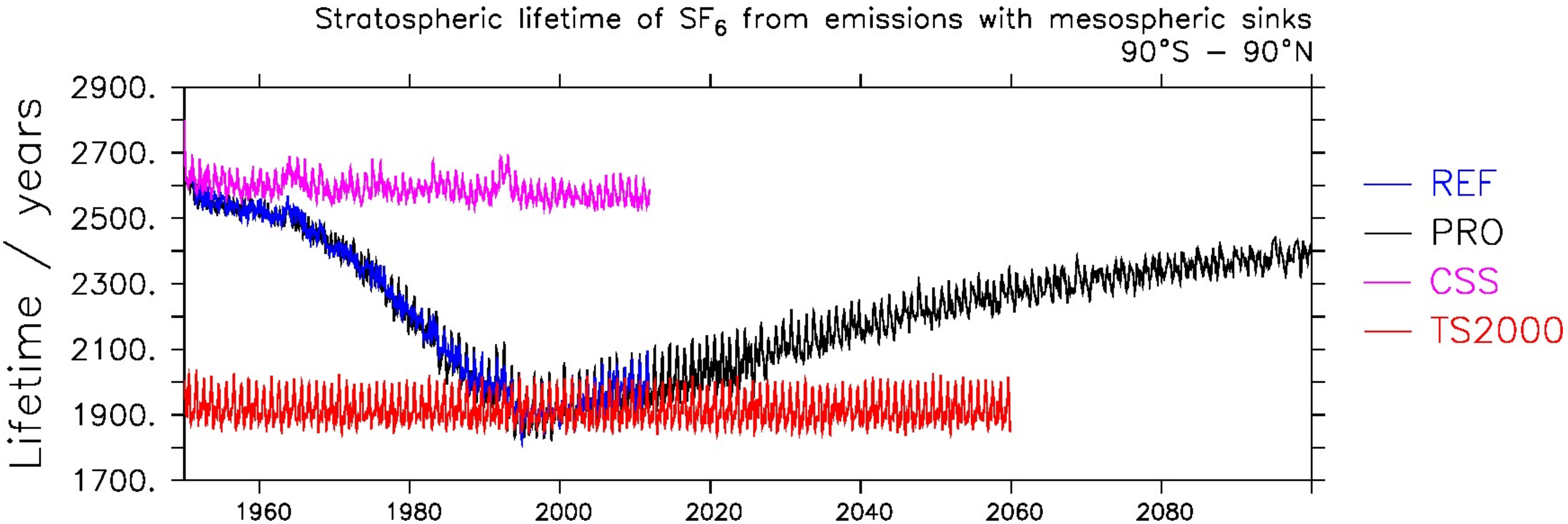
EMAC SF_6 Lifetime: 2219 years

Ravishankara et al., 1993: 3200 years
Reddmann et al., 2001: 400 – 10000 years
Kovács et al., 2017: 1278 years
Ray et al., 2017: 580 – 1400 years
Kouznetsov et al., 2019: 600 – 2900 years



Long term trend in transient simulations due to changes in reactant species. It resembles the ozone mixing ratios. However, this might be due to some simplifications.

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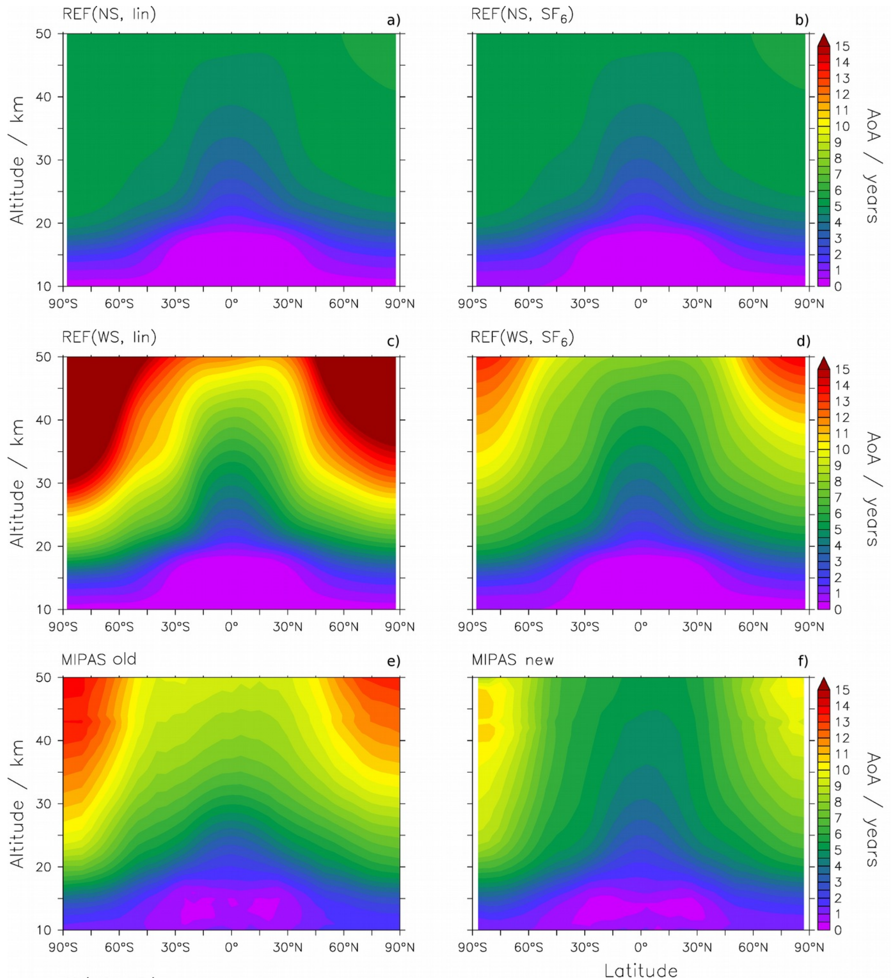
EMAC Climatologies

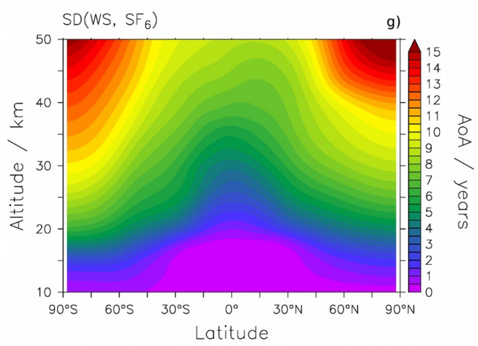
- AoA annual mean for 2002-2011 (MIPAS period)
- **AoA without sinks generally younger than with sinks:**
  - → Sinks produce smaller mixing ratios
  - → AoA seems older as reference value lies further in past
- EMAC tracer (WS, SF_6) best fit with MIPAS

MIPAS: Michelson **I**nterferometer for **P**assive **A**tmospheric **S**ounding; Atmospheric chemistry sensor on-board Envisat; Active July 2002 – April 2012

Tropics:
Good agreement between EMAC and MIPAS 'new' with regards to tropical ascent rates

High Latitudes:
Good agreement between EMAC and MIPAS 'old', especially for SD run (due to better representation of polar vortex)
→ **SON seasonal mean ?**





MIPAS 'new'
Stiller et al., 10th Limb Workshop Greifswald, 2019

MIPAS 'old'
Stiller et al., 2012
Haenel et al., 2015

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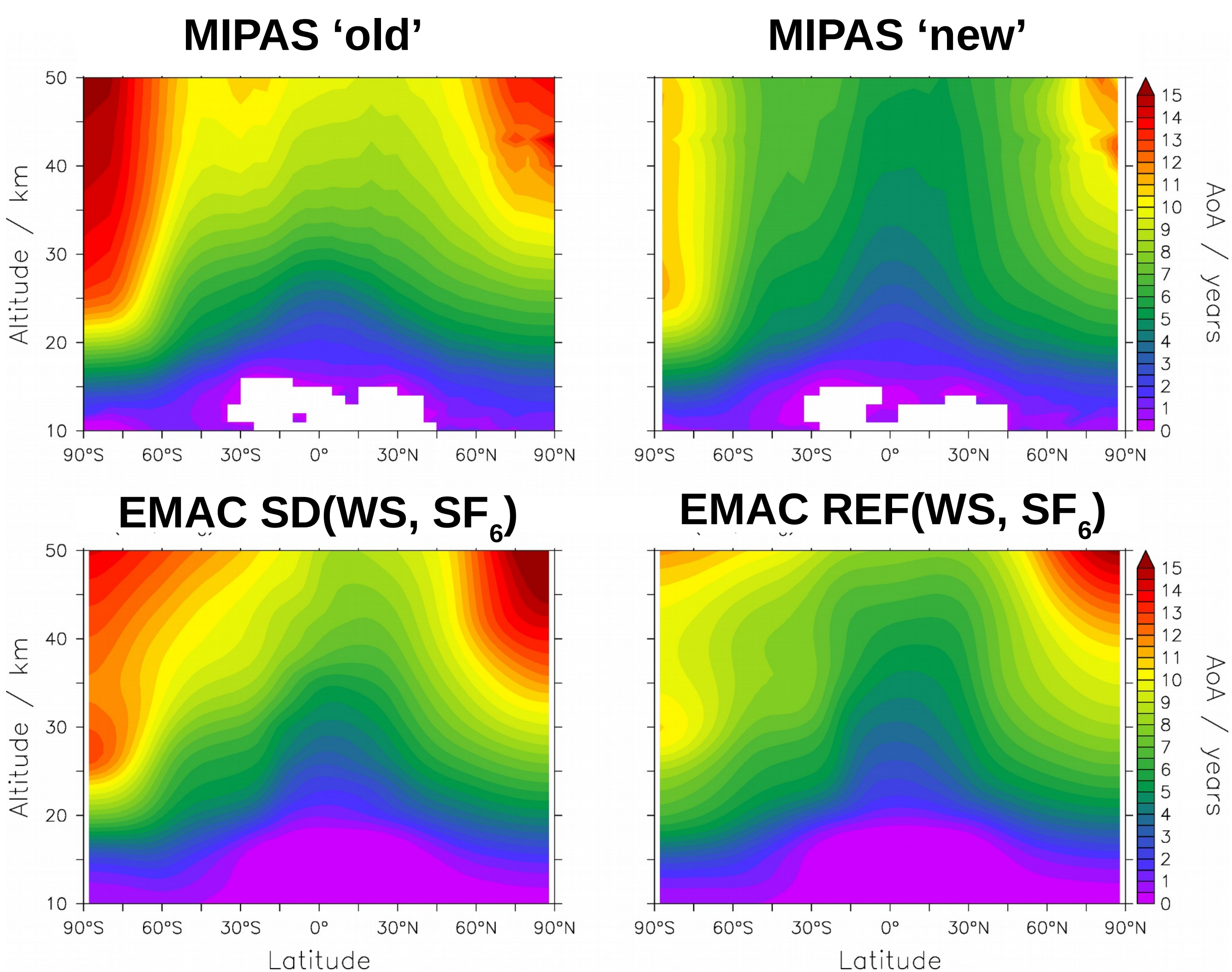
EMAC vs MIPAS on Envisat → Nudging? → SON?

- AoA 2007 – 2010 seasonal mean **SON**
- EMAC (WS, SF_6)

Antarctic vortex under-represented in EMAC
(Joeckel et al., 2016)

→Isolation and ageing of air in polar vortex better represented in SD simulation

→ however, MIPAS ‘new’ shows much lower AoA in high latitudes → further research (models and observations) required to resolve discrepancy.



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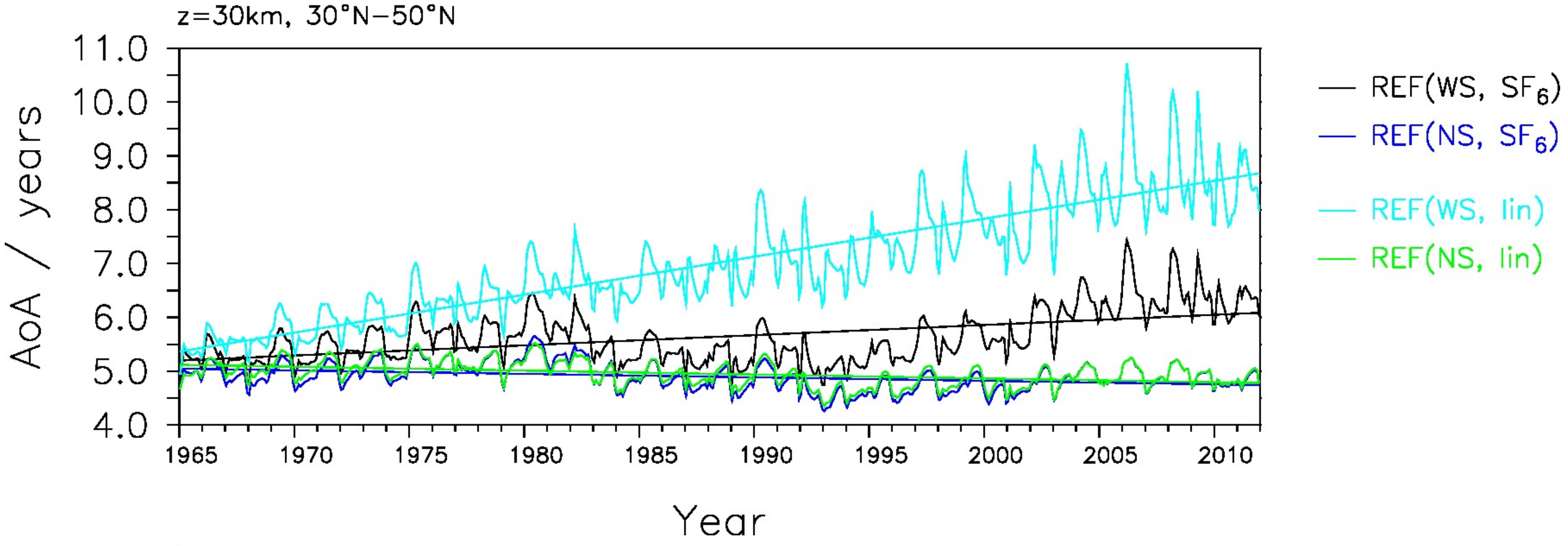
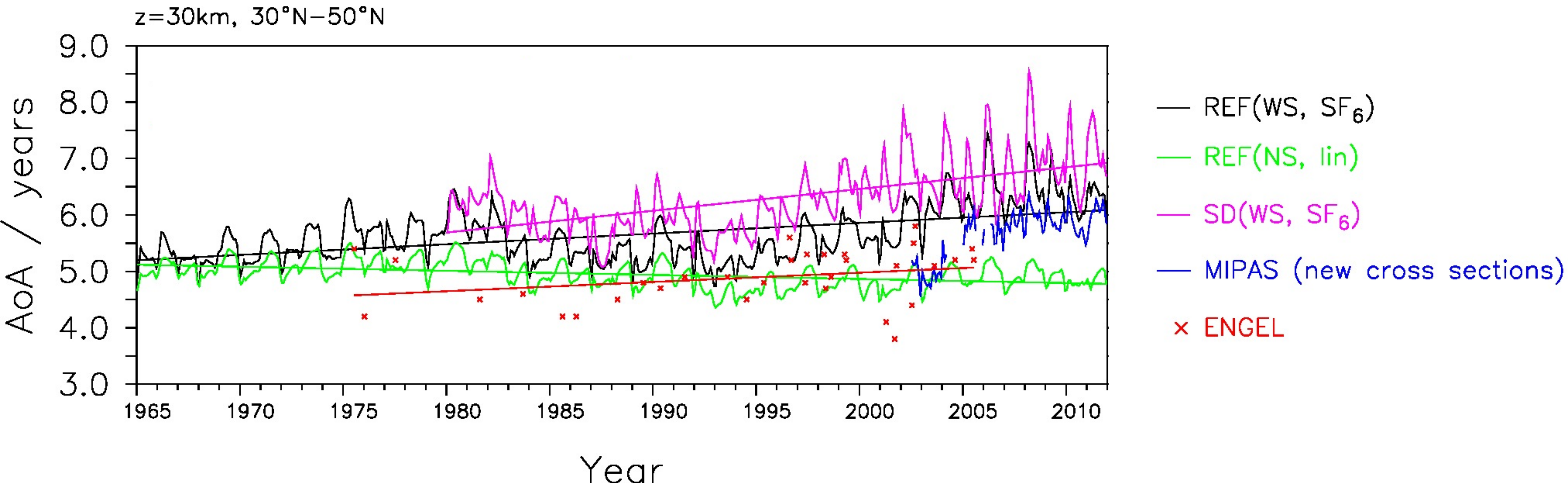
EMAC: REF timeseries

- No Sinks → Negative Trend
- (NS, lin) ~ (NS, SF_6) → Green's function in calculation of AoA (Fritsch et al., 2019)
- Sinks → Positive Trend



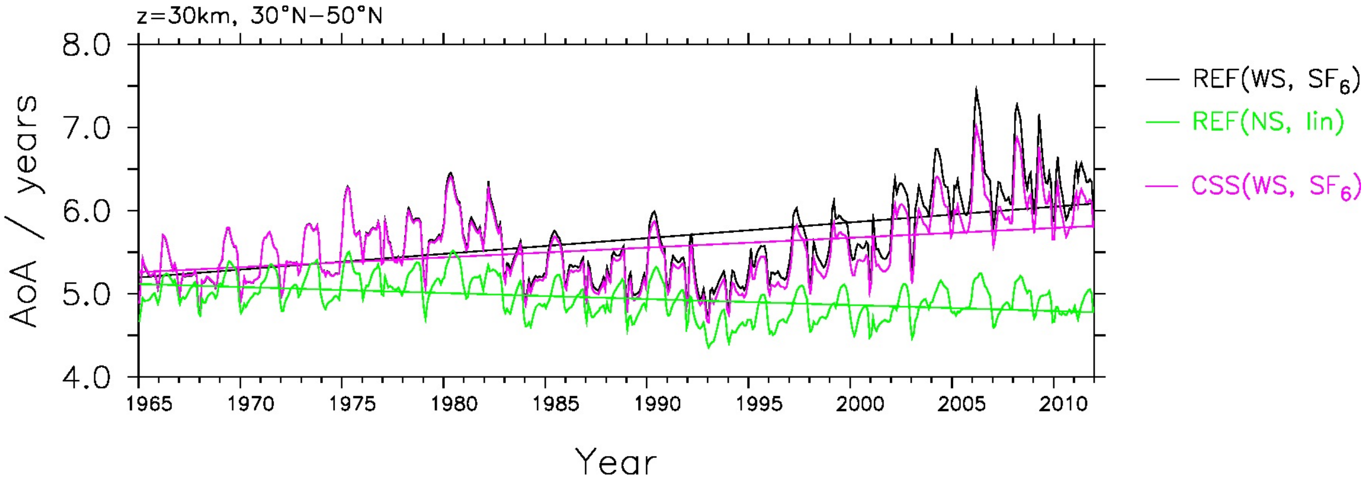
Model vs Observations

- EMAC REF(WS, SF_6) & REF(NS, lin) & SD(WS, SF_6)
- Balloon-borne measurements (Engel et al., 2009)
- MIPAS (Stiller et al., 2012; Haenel et al., 2015) with improved SF_6 retrieval scheme (Stiller et al., 2019 10th Limb Workshop, Greifswald)



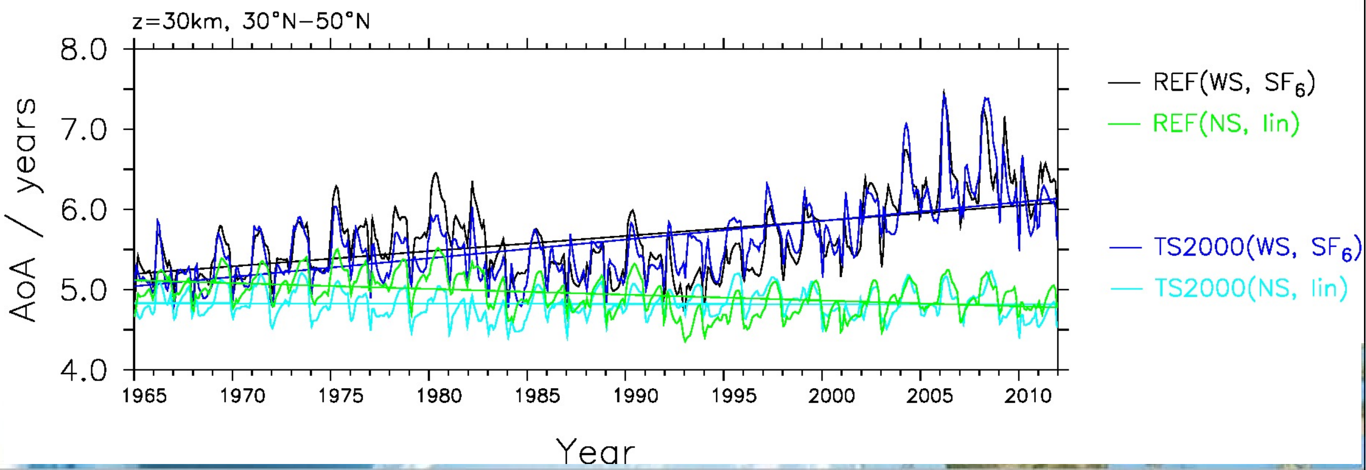
Are the reactive species in the sinks responsible for the trend?

- **CSS:** Constant mixing ratios of the reactant species
  - Also produces positive AoA trend, albeit somewhat reduced



Are changes in circulation strength responsible for the trend?

- **TS2000:** Timeslice simulation with climate conditions from 2000
  - Produces even stronger positive AoA trend than that of REF (WS, SF_6)



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Trends: REF

Following Schoeberl et al. (2000) and Hall & Plumb (1994):

Consider a tracer $\chi(t)$ with constant relative loss -kt

and with reference curve $\chi_o(t)$ with linear growth rate $\chi_o(t) = \chi_{oo}(t) \cdot t$

At any location the concentration of the tracer is:

Mixing ratio, Reference, Growth, Boundary Propagator

$$\chi(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) exp(-k\cdot\tau) G(\tau) d\tau = \chi_{oo}\left(t\cdot\widetilde{G}(k) + \frac{\partial}{\partial k}\widetilde{G}(k)\right)$$

For a passive (ie. no sinks) tracer:

Mixing ratio, Reference, Growth, Boundary Propagator

$$\chi(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) exp(\cancel{-k}\cdot\tau) G(\tau) d\tau \longrightarrow \chi_p(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) G(\tau) d\tau$$

$$= \chi_{oo}\cdot(t-\Gamma)$$

So rearranging: $\Gamma = t - \frac{\chi_p(t)}{\chi_{oo}}$ —Trend = Change over time→ $\frac{\partial\Gamma}{\partial t} = 0$, with $\chi_p(t) = \chi_{oo}\cdot t + a$

→ for a passive tracer, the trend is 0

For an active (ie. with sinks) tracer:

$$\Gamma_s = t - \frac{\chi(t)}{\chi_{oo}} = t\cdot\left(1-\widetilde{G}(k)\right) - \cancel{\frac{\partial}{\partial k}\widetilde{G}(k)}$$

Trend = Change over time →

$$\frac{\partial\Gamma_s}{\partial t} = 1 - \widetilde{G}(k) > 0$$

$\tilde{G}(k=0) = 1 \text{ whereas } \tilde{G}(k\to\infty) = 0$

Growth rate of reference mixing ratio

Mixing ratio of tracer $\chi(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) exp(-k\cdot\tau) G(\tau) d\tau$

→ "apparent AoA" rises due to the SF_6 sinks themselves

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How do SF_6 sinks affect Age of Air climatologies and trends ?

- **SF_6 lifetime: 2219 years (1900 – 2600)**
  - Within uncertainty range of previous studies
- **SF_6 sinks lead to older Age of Air**
  - Overall, the SF_6 sinks lead to good AoA agreement between the climatologies of EMAC model results and MIPAS satellite observations
- **SF_6 sinks lead to positive trends**
  - SF_6 sinks can help to reconcile the trends of models and observations (Engel et al. 2009), but the effect remains to be quantified precisely
- Positive trends are neither a result of climate change, nor of changes in reactive species involved in SF_6 depletion, **“apparent Age of Air” keeps on rising due to the SF_6 sinks themselves.** This effect overcompensates the effect of the accelerating BDC in our simulations.

THANK YOU

Look out for Loeffel et al. (2020) in ACPD coming soon

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References

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Ray et al., 2017: *Quantification of the SF_6 lifetime based on mesospheric loss measured in the stratospheric polar vortex*

Reddmann et al., 2001: *Three-dimensional model simulations of SF_6 with mesospheric chemistry*

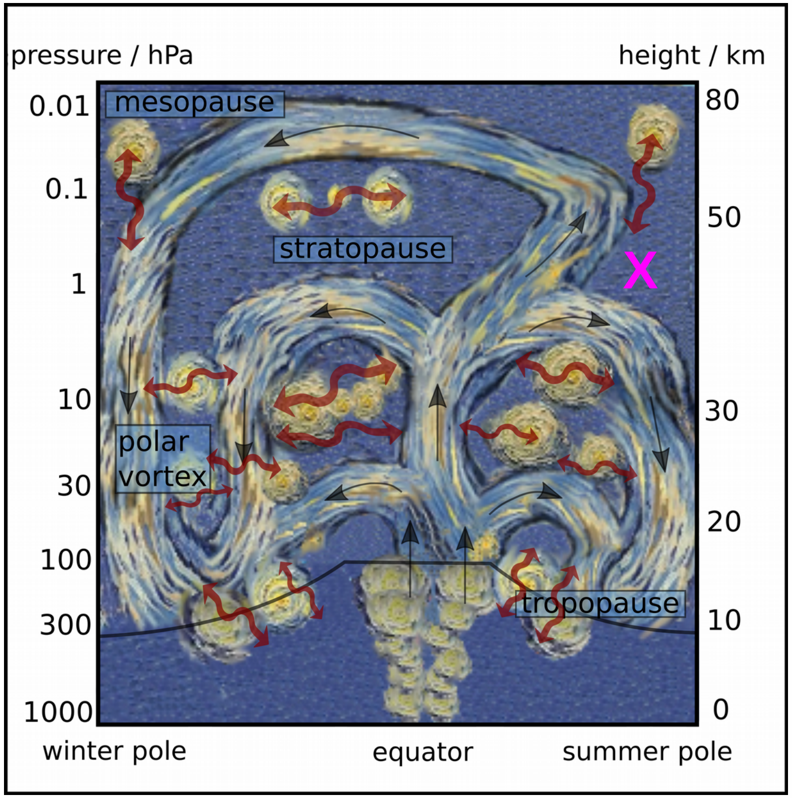
Stiller et al., 2012: *Observed temporal evolution of global mean age of stratospheric air for the 2002 to 2010 period*

Waugh, D., Hall, T. (2001): *Is upper stratospheric chlorine decreasing as expected?*

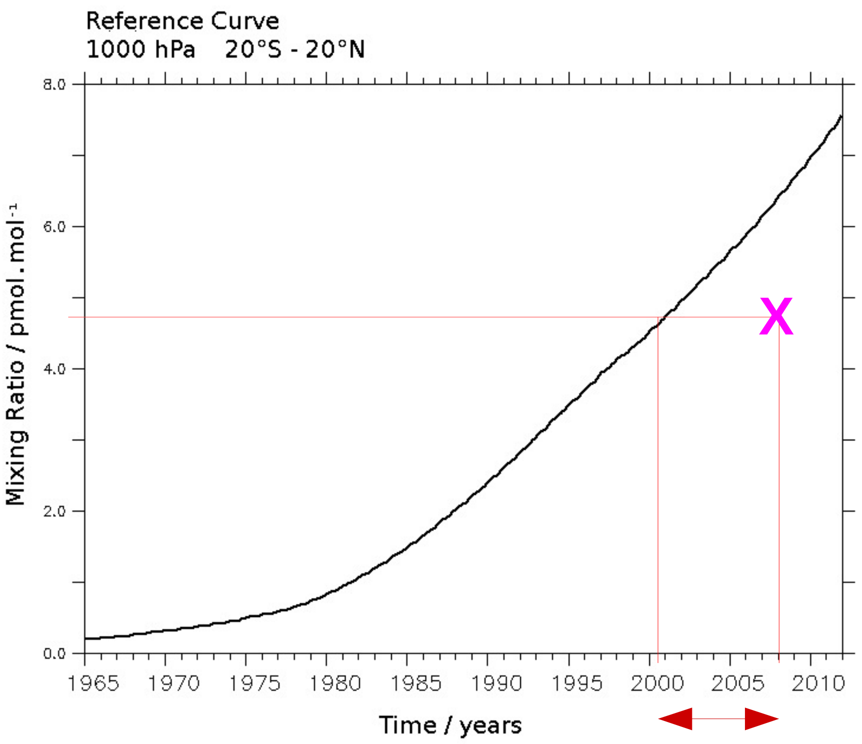
Supplementary Information

Age of Air (AoA)

- AoA ~ time elapsed since air entered stratosphere
- AoA can be derived from measurable tracers e.g. sulphur-hexafluoride: SF_6



R. Eichinger, & V. van Gogh, 2019 personal communication



Calculate AoA:

- sample the mixing ratio of SF_6 at X
- match it to the tropospheric reference
- obtain the lag time

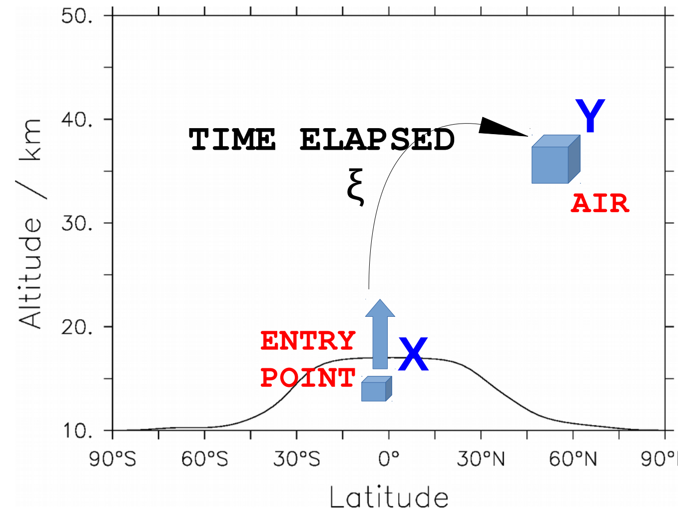
Reference Simulation		
REF Reference	Transient 1950 – 2011	• No chemistry activated other than SF6 submodel • Greenhouse gases (GHGs) (CO_2, CH_4, N_2O, O_3) and SF_6 sink reactant species transiently prescribed from ESCiMo RC1-base-07-simulation (Jöckel et al., 2016) as monthly and zonal means
Nudged Simulation		
SD Specified Dynamics	Transient 1980 – 2011	• Specified Dynamics: Newtonian relaxation of dynamics towards ERA-INTERIM (Dee et al., 2011) reanalysis data up to 1hPa
Sensitivity Experiments		
CSS Constant reaction partners for SF_6 sinks	Transient 1950 – 2011	• Same as **REF** but constant mixing ratios of the reactant species (1950 on repeat)
TS2000 Timeslice	Timeslice 1950 – 2059	• Climate conditions (GHGs, SSTs, SICs) of year 2000 Climatology taken from 1995 – 2004 • SF_6 sinks reactant species averaged over 1995 – 2004
Projection Simulation		
PRO Climate Projection	Transient 1950 - 2100	• Same as **REF** but GHGs and reactant species transiently prescribed from ESCiMo RC2-base-04-simulation (Jöckel et al., 2016) as monthly and zonal means

Age of Air: Calculation

Following the mathematical formulations and principles presented by *Hall and Plumb (1994)*:

Continuity equation for passive and conserved tracer:

$$\frac{\partial \chi}{\partial t} + \boldsymbol{L}(\chi) = 0 \qquad \chi(r,t) : \text{mixing ratio of tracer at point } r \text{ and time } t$$



Response at point r in stratosphere (**Y**):

$$\chi(r,t) = \int_{-\infty}^{t} \chi(\Omega,t')\boldsymbol{G}(r,t \mid \Omega,t')dt'$$

t' : source time

t : field time

Ω : region

$\boldsymbol{G}(r,t \mid \Omega,t')$: boundary propagator

Define:

elapsed time $\xi = t - t'$ and concentration lag time τ: elapsed time between mixing ratio at point r and its occurrence at Ω

Then:

$$\chi(r,t) = \chi(\Omega, t-\tau) \quad \Rightarrow \quad \tau(r) = \int_{0}^{\infty} \xi \boldsymbol{G}(r \mid \Omega,\xi)d\xi \;\equiv\; \Gamma(r)$$

AoA ~ time lapsed since air at Y entered stratosphere at X

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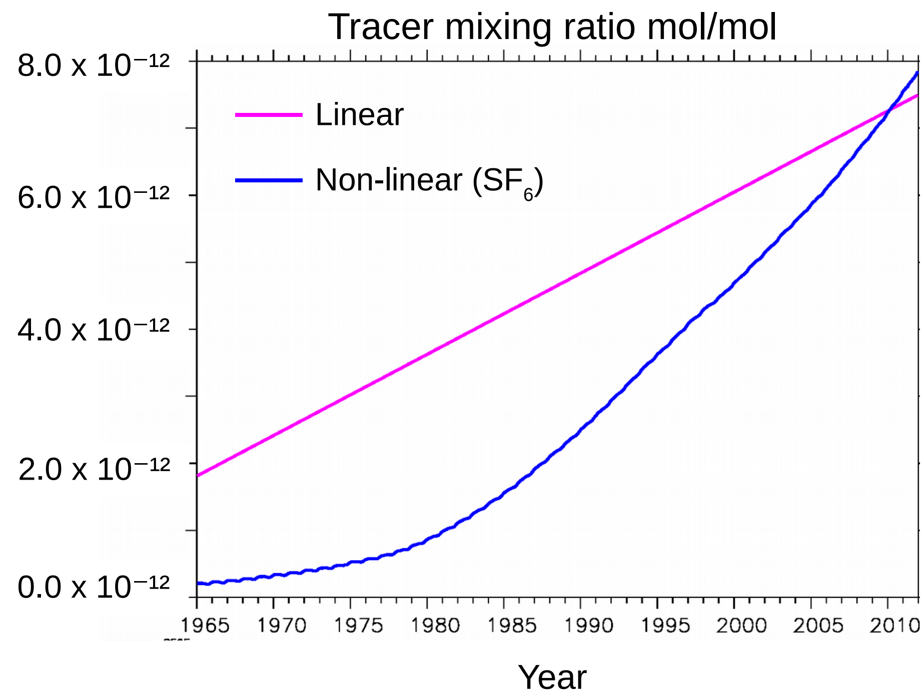
Age of Air: Calculation



We have assumed a <u>linear</u> time variation of the tracer!

SF_6 does not have a fully linear growth rate!

For a (first-order) exponentially growing tracer

with growth rate σ and spectral width Δ (measure of the spread of transit times since last tropospheric contact) the concentration time lag is:

$$\tau_{exp}(r) \approx \Gamma(r) - \sigma^{-1}\ln(1+\sigma^2\Delta^2$$

$$r) = \frac{1}{2}\int_0^\infty (\xi - \Gamma(r))^2 \boldsymbol{G}(r \mid \xi) d\xi$$

$$\xrightarrow{\sigma\Delta \ll 1} \quad \tau_{exp}(r) \approx \Gamma(r) - \sigma\Delta^2(r) \quad \Rightarrow \quad \boxed{\tau_{exp} \approx \Gamma \ \text{ if } \ \sigma^{-1} \gg \Delta^2/\Gamma}$$

Hall and Plumb (1994): $\Delta^2/\Gamma \sim 0.7 \text{ year}$

We use 1.0 (Fritsch et al., 2019)

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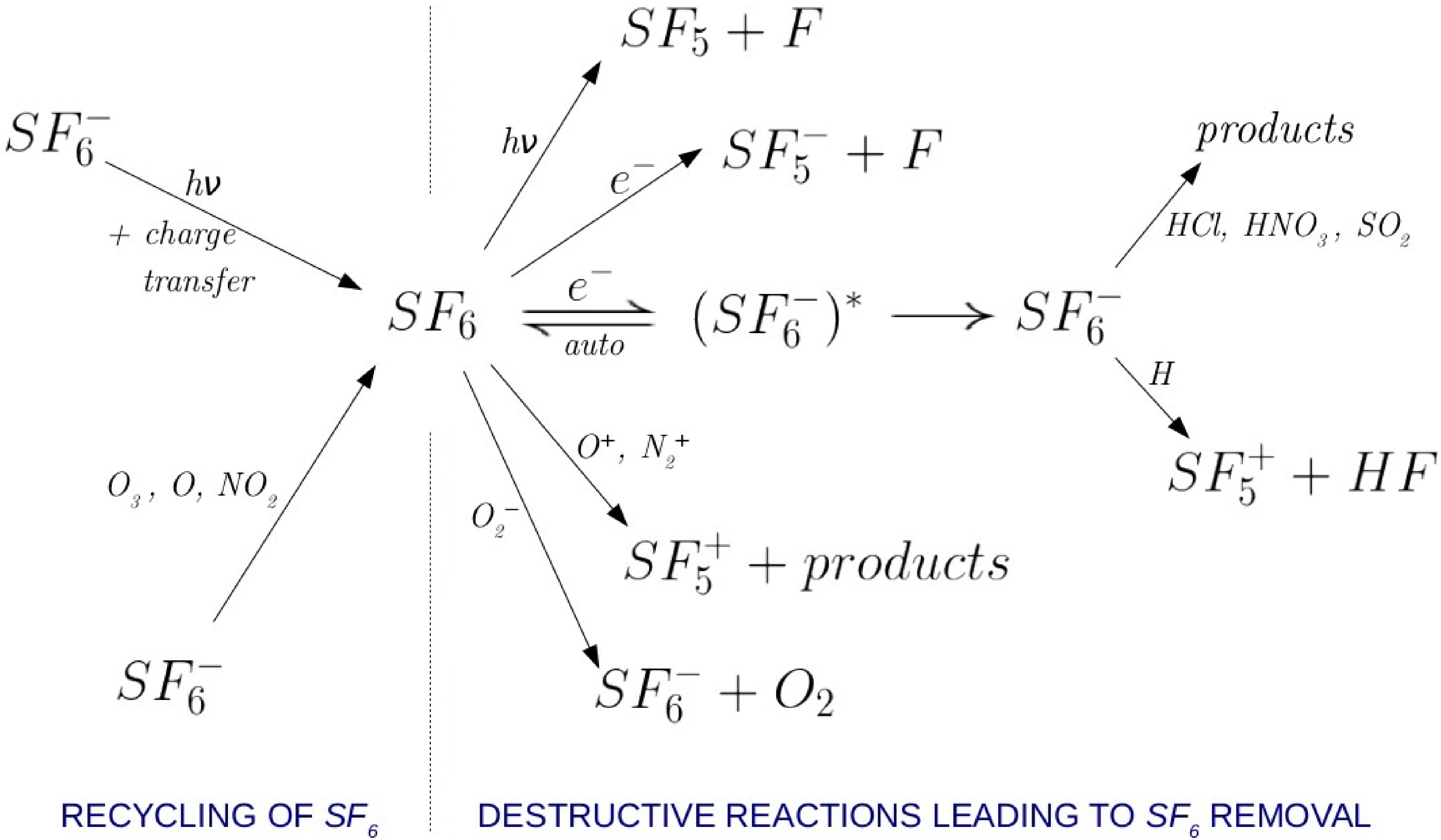
SF_6 Chemistry in the Mesosphere



AoA: EMAC vs Balloon Flights

SD(WS, SF_6) Sept. 1996
SD(WS, SF_6) Sept. 1993
SD(WS, SF_6) 1975-2005

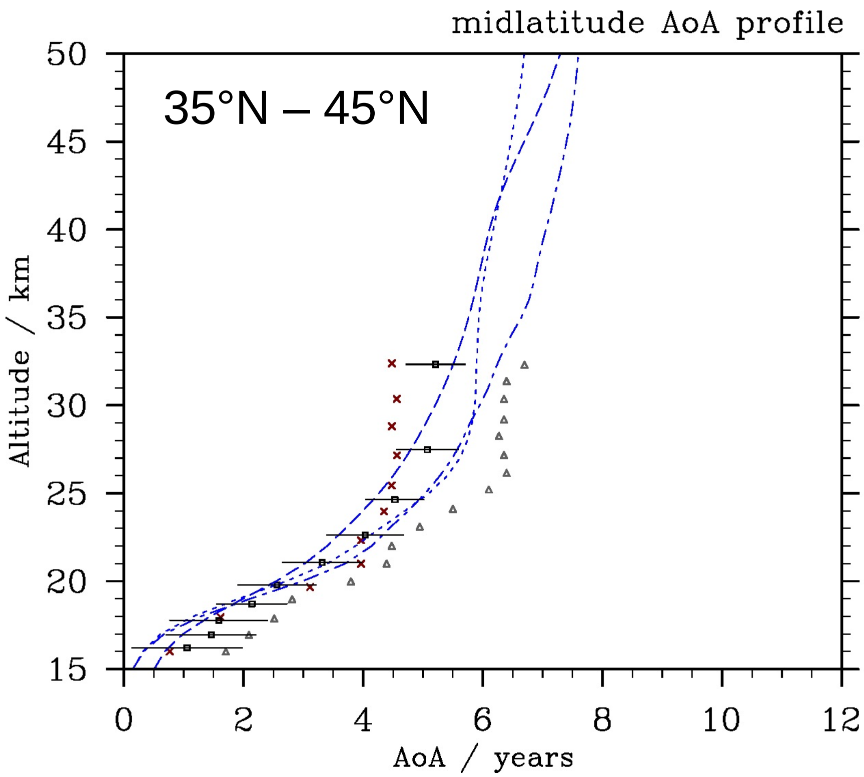
Balloon Flight Sept. 1996
Andrews et al. 2001

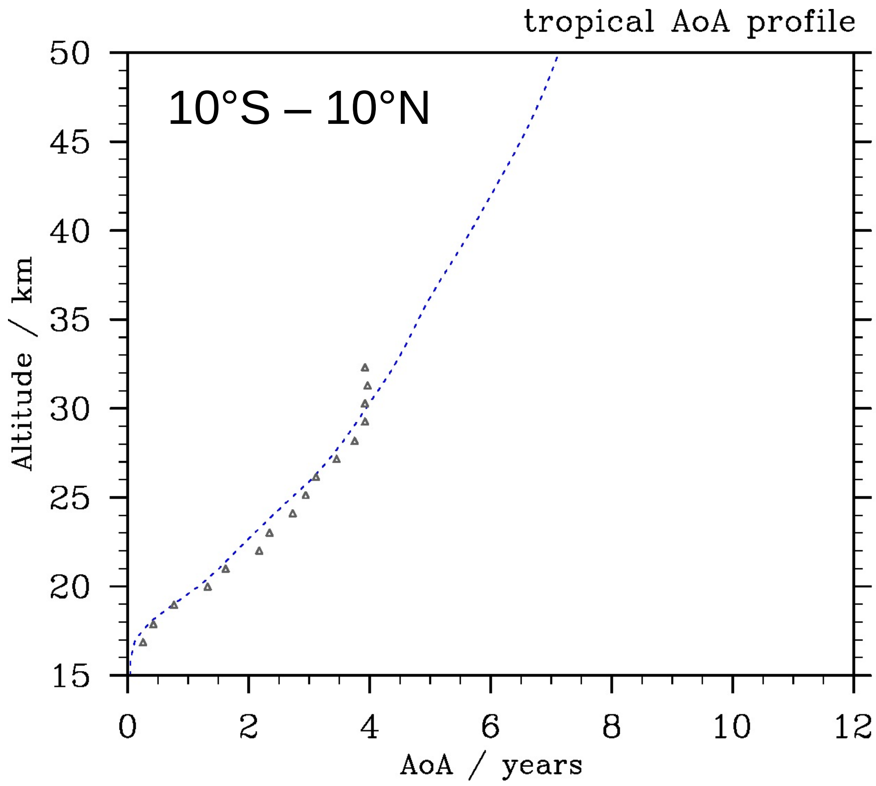
Balloon Flight Sept. 1993
Andrews et al. 2001

Balloon Flight 1975-2005
Engel et al. 2009

SD(WS, SF_6) 1992 - 1998

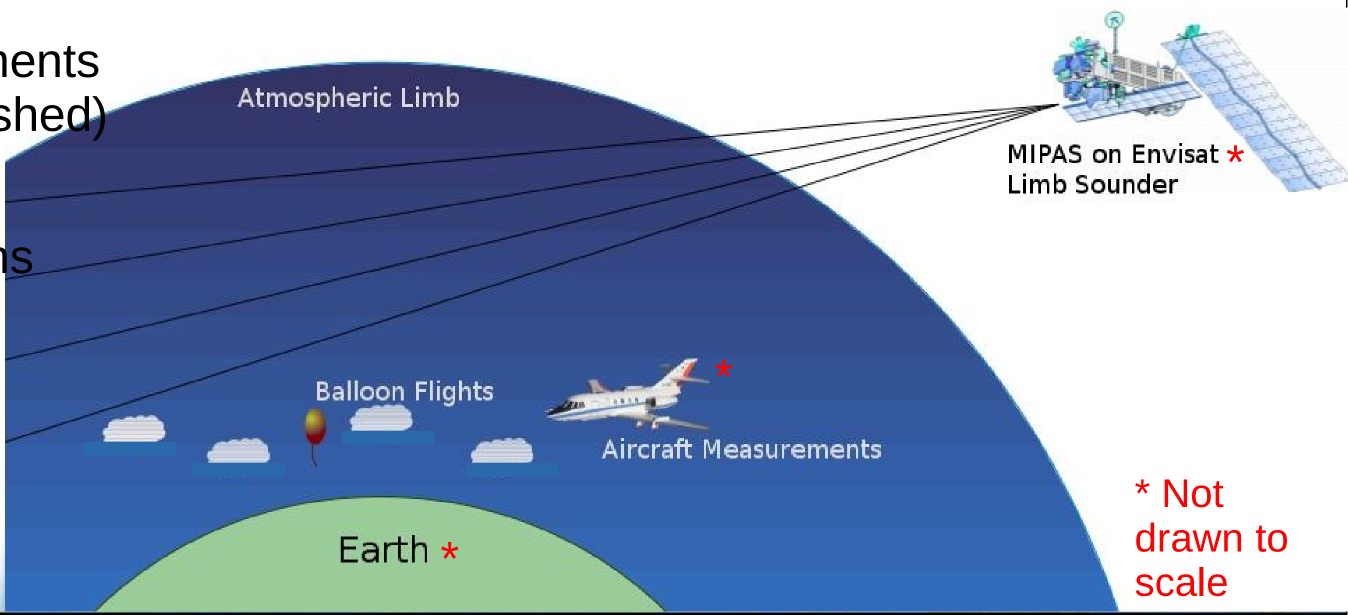
Balloon Flights 1992 – 1998
Andrews et al., 2001



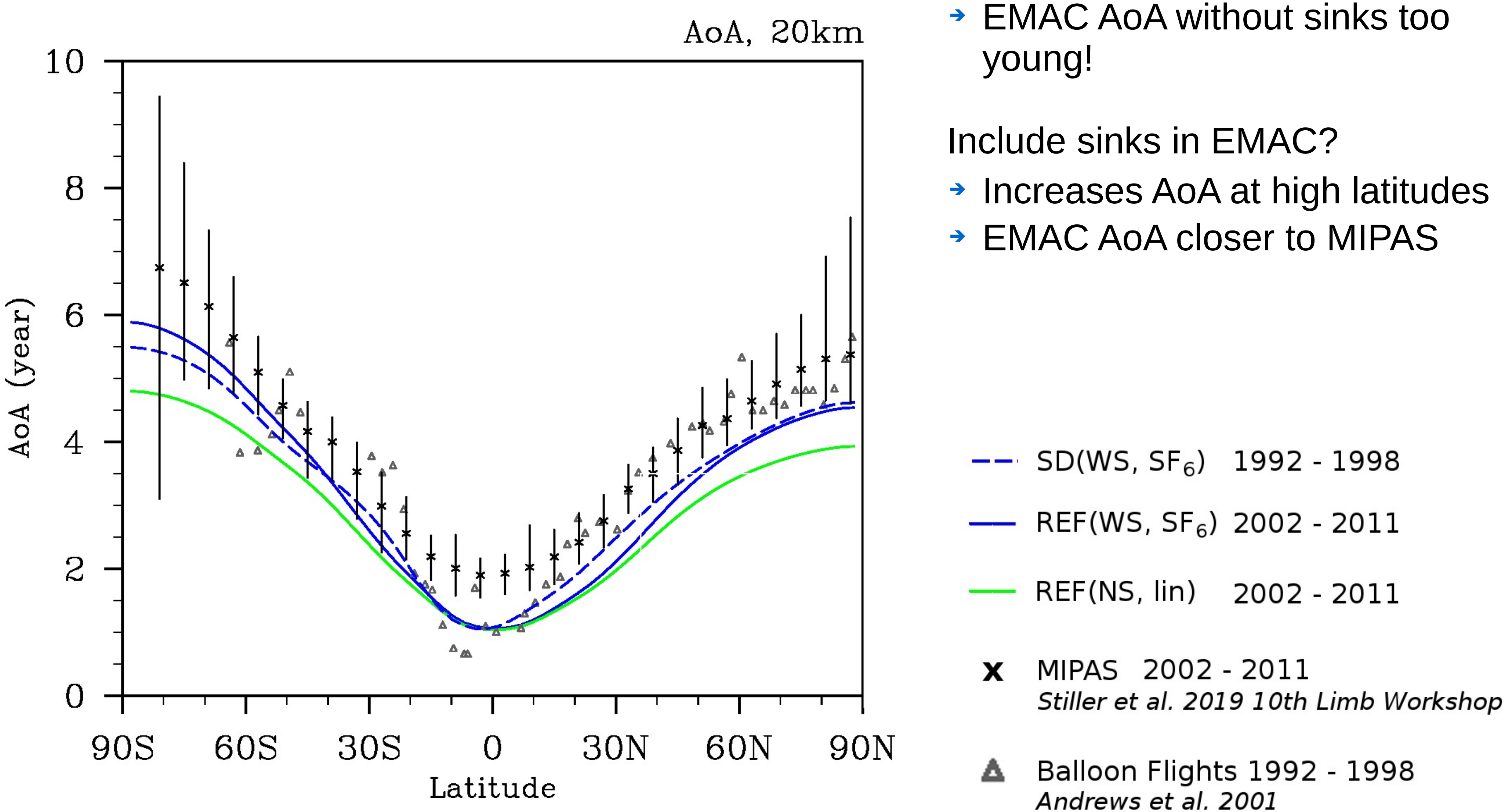


What is MIPAS ?

- **M**ichelson **I**nterferometer for **P**assive **A**tmospheric **S**ounding
- Atmospheric chemistry sensor on-board the Environmental Satellite (Envisat)
  Active July 2002 – April 2012
- Allowed for retrieval of SF_6: measured thermal emission in mid-infrared, in middle and upper atmosphere, at the atmospheric limb
- AoA from SF_6 retrieval: Stiller et al., 2012 & Haenel et al., 2015
- New version of MIPAS data exists as of 2019
  (G.Stiller, personal communication. Stiller et al., 2019, 10th Limb Workshop, Greifswald)
  - spectroscopic improvements
    (J. Harrison, to be published)
  - newly measured SF_6 absorption cross sections



AoA: EMAC vs Observations



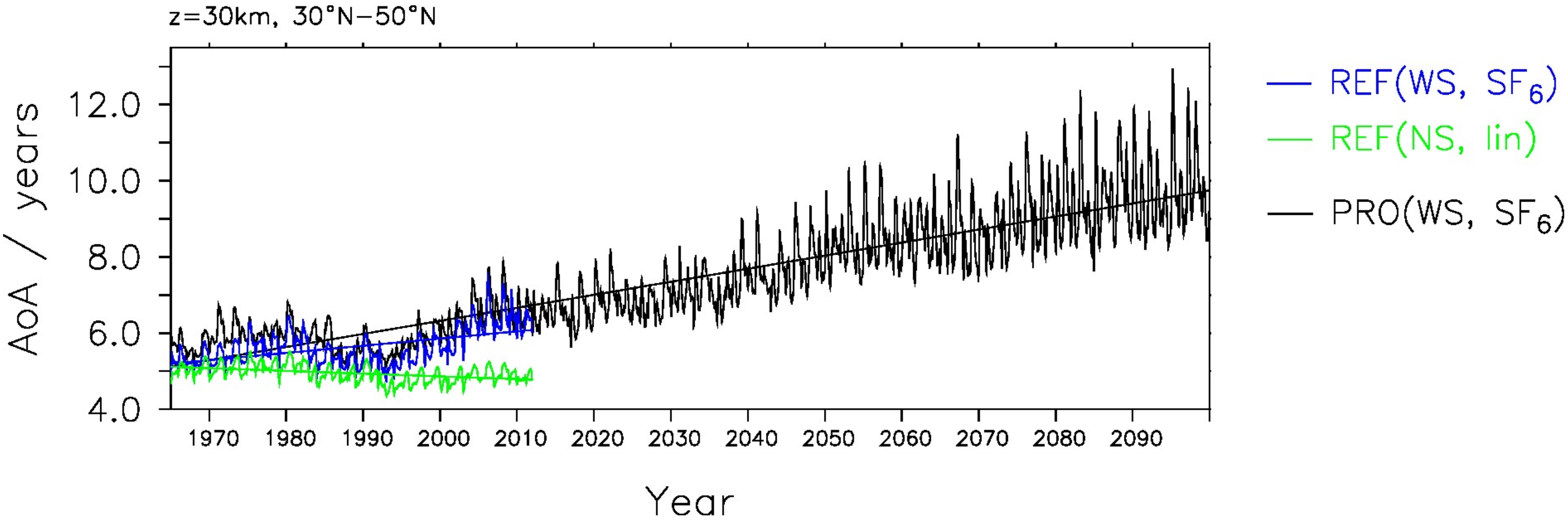
High latitudes:

- EMAC AoA without sinks too young!

Include sinks in EMAC?

- Increases AoA at high latitudes
- EMAC AoA closer to MIPAS

Climate Projection



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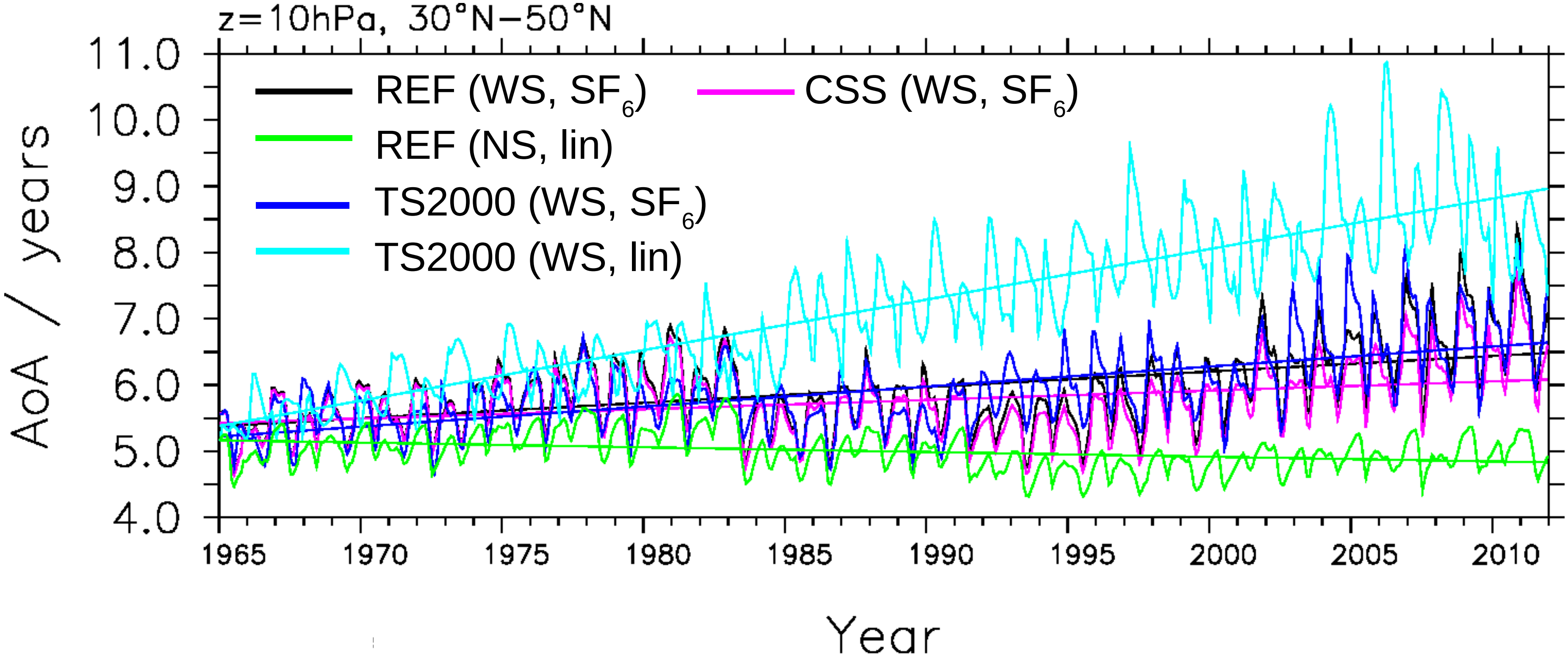
Sensitivity Experiments

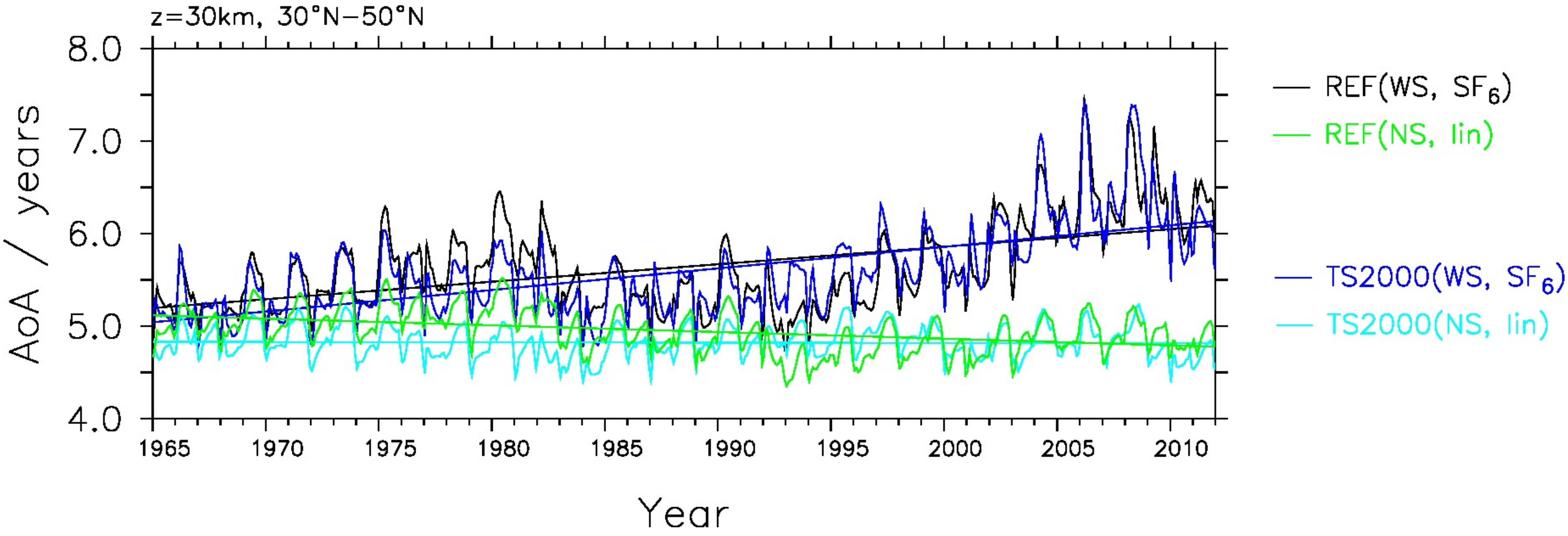
- Positive trend neither a result of climate change nor of SF_6 sinks **!**



TS2000 also answers another question:

- "80s dip" and "90s dip" not a volcanic effect, nor is it caused by the solar cycle

➔ Due to the non-linearity in SF_6 emissions: consequence of the calculation method involving Green's function (Fritsch et al., 2019)

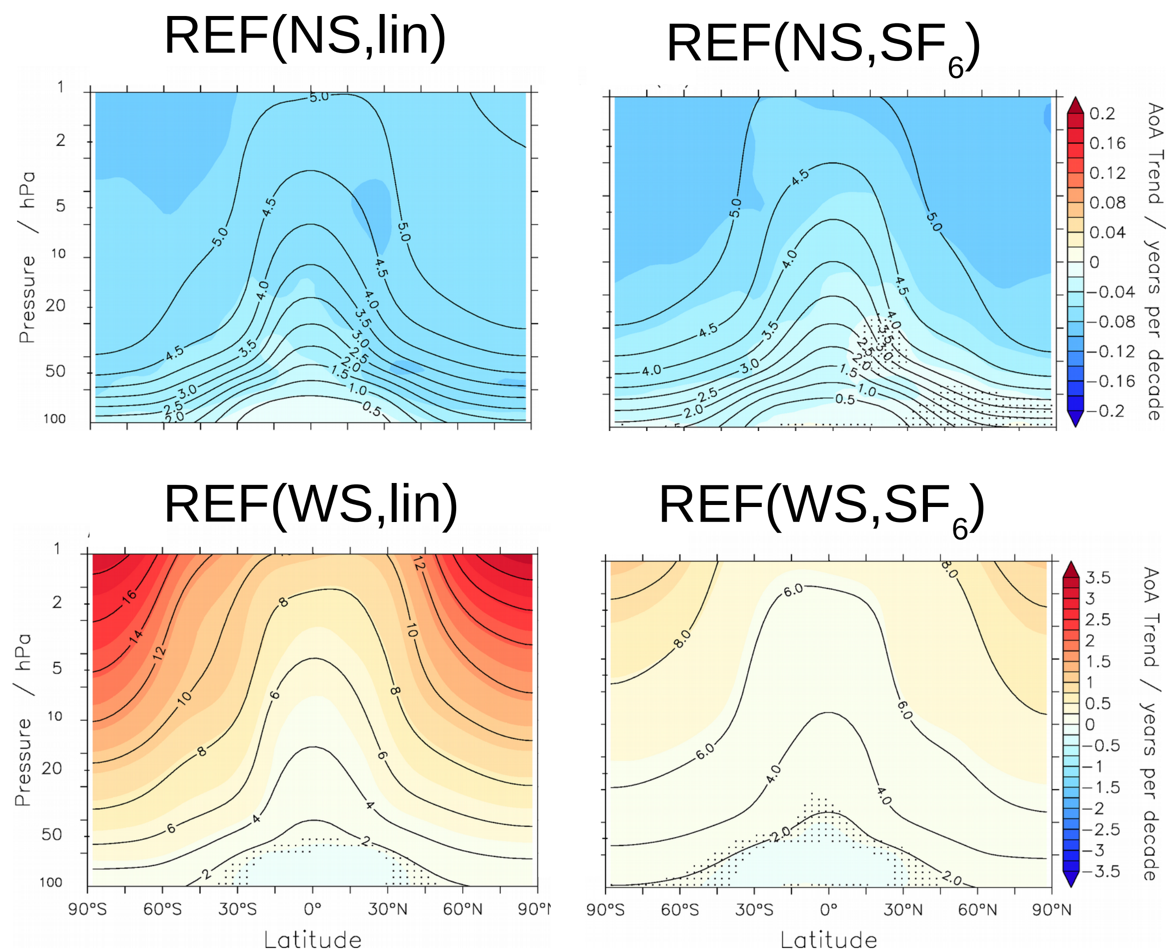


Trends throughout the stratosphere

- Linear regression at each point: trend from 1965 – 2011
- AoA contours 1995 – 2011

- Linear
  with sinks: +ive trend
  without sinks: -ive trend
- Non-linear (SF_6):
  with sinks: +ive trend
  without sinks: -ive trend

- **Sinks → positive trend**



Trends:

No Sinks:

$$\frac{\partial \Gamma}{\partial t} = 0$$

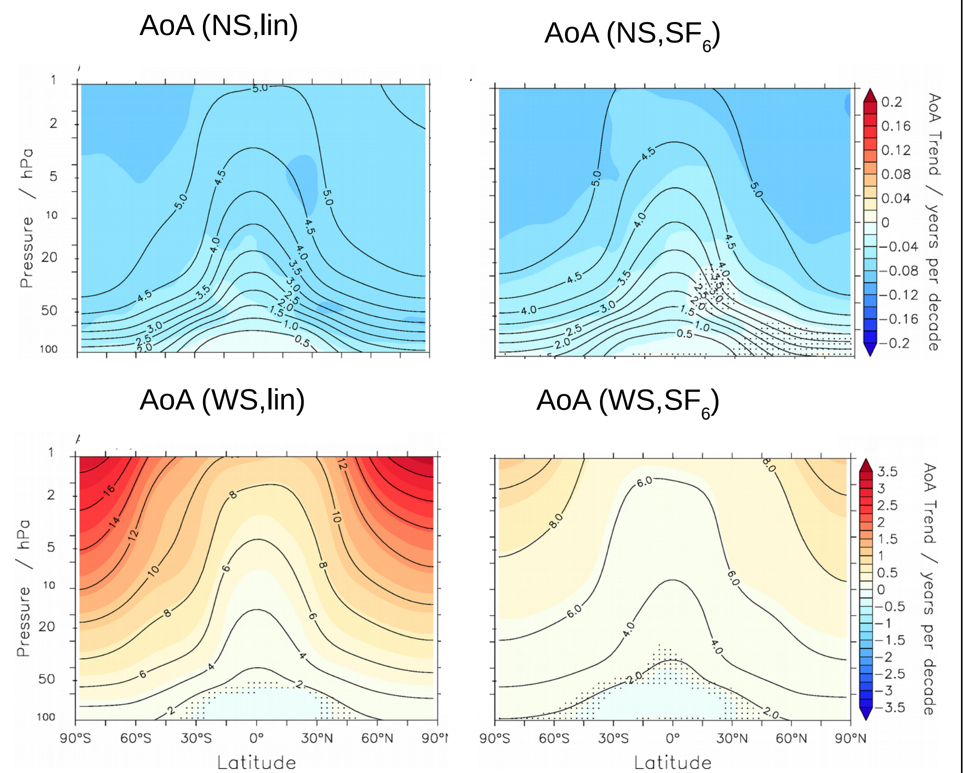
For TIMESLICE:
Without Sinks
→ No Trend
→ But negative trend due to circulation acceleration in transient simulation

With Sinks:

$$\frac{\partial \Gamma_s}{\partial t} = 1 - \widetilde{G}(k) \quad > 0$$

Gamma: AoA

k: loss rate (sinks)



With Sinks
→ Positive Trend