# Atmospheric History of Carbonyl Sulfide (COS) During the 20th Century from Antarctic and Greenland Firn Air Measurements

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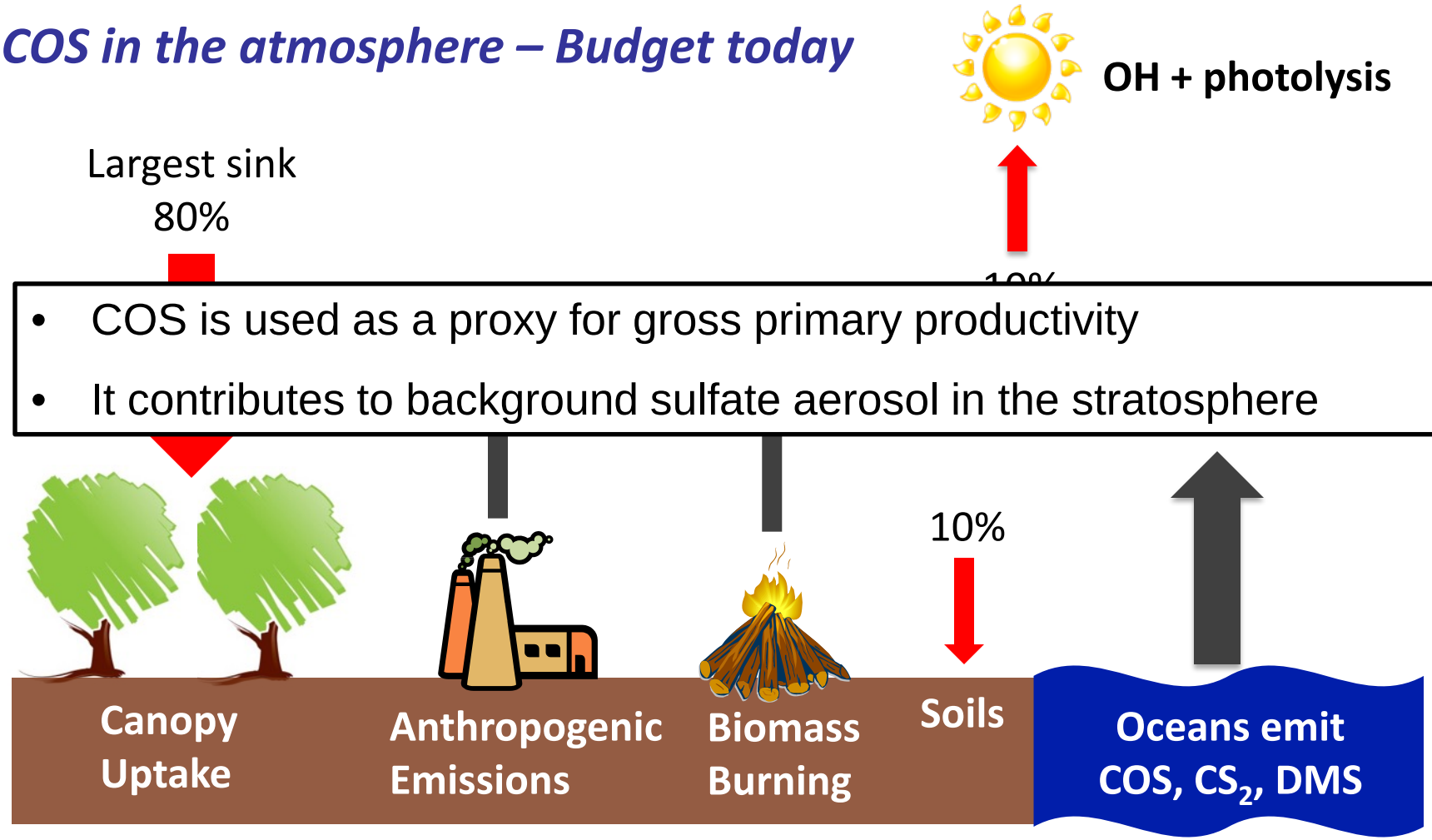
# *COS in the atmosphere – Budget today*




Current best estimates:

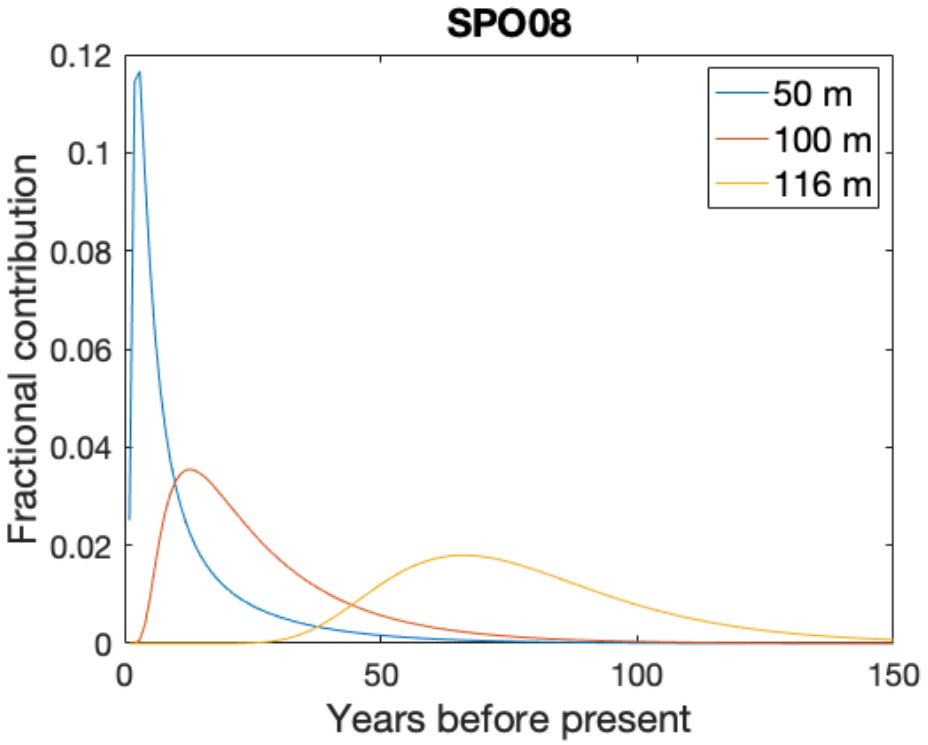
Emissions are ~1.2 TgS $y^{-1}$

Lifetime is ~2 y

- Mean trop. mixing ratio just under 500 ppt
- 1-2% more in the SH

# *Firn air COS records – background*

- Firn air ages non-linearly with depth
- Measurements from a given depth represent average levels over many years
- The deeper in the firn, the longer the time scale of averaging
- Recovery of atmospheric records requires a formal inversion using firn model outputs (figure on the left)
- Inversions benefit from knowing past atmospheric composition (ice core data helps!)



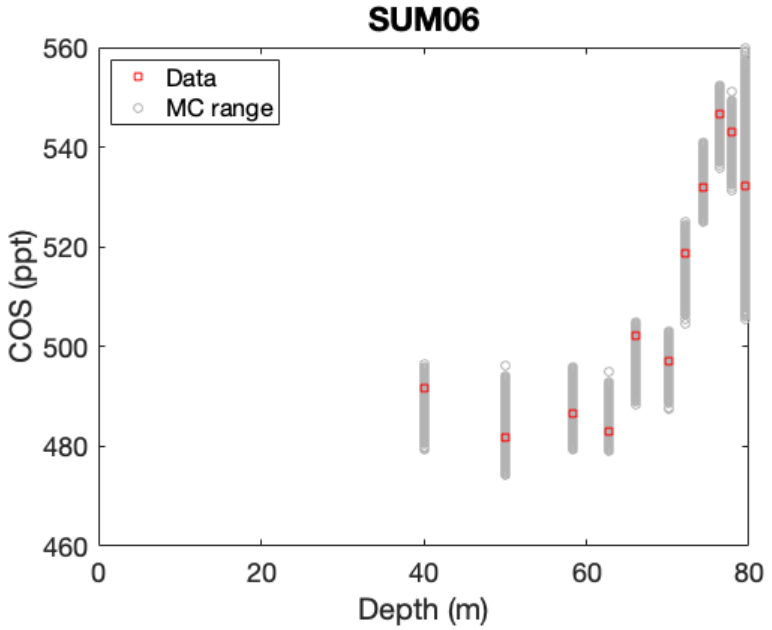

- <u>Firn data from</u>

  *3 Greenland campaigns*: Summit (SUM) in 2006 and 2013, Renland in 2015

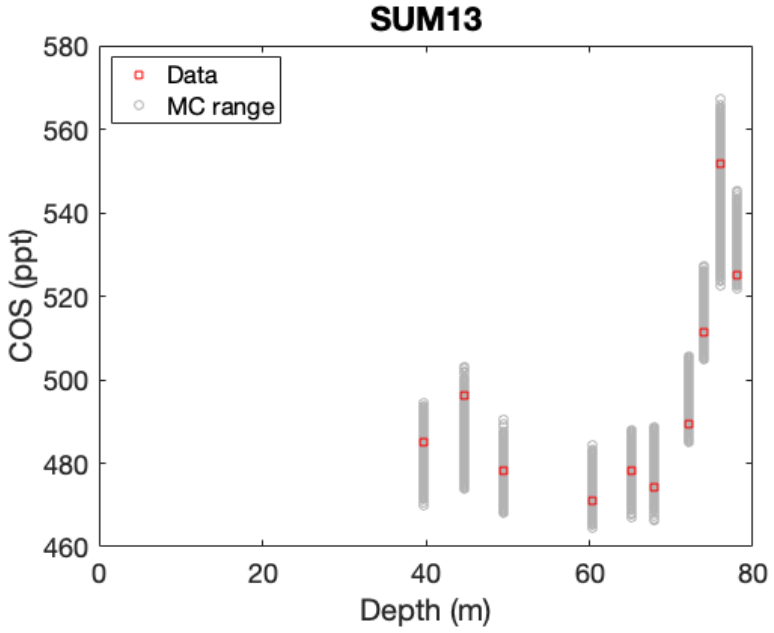
  *4 Antarctic campaigns:* S. Pole (SPO) in 2001, 2008, 2015, and Mega Dunes in 2003

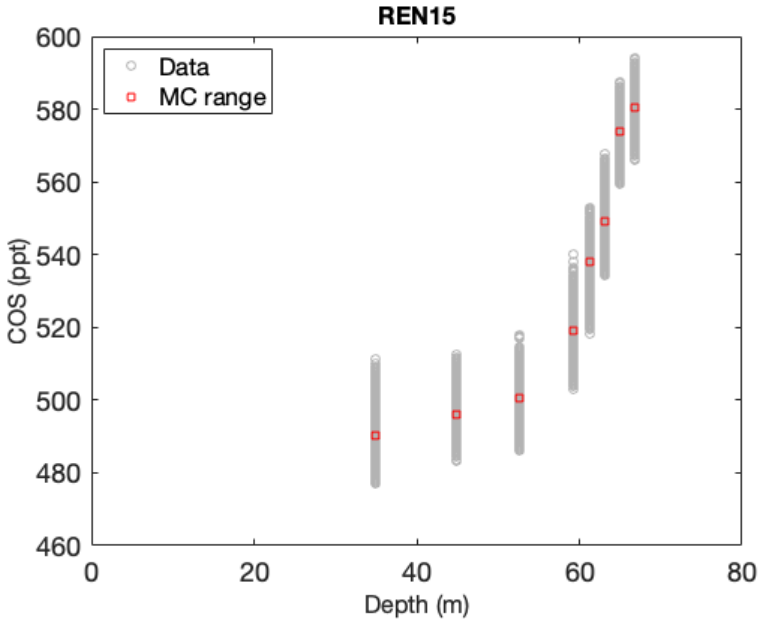
- Inversions conducted separately for NH and SH sites

# *Firn air COS measurements and MC sampling – Greenland*



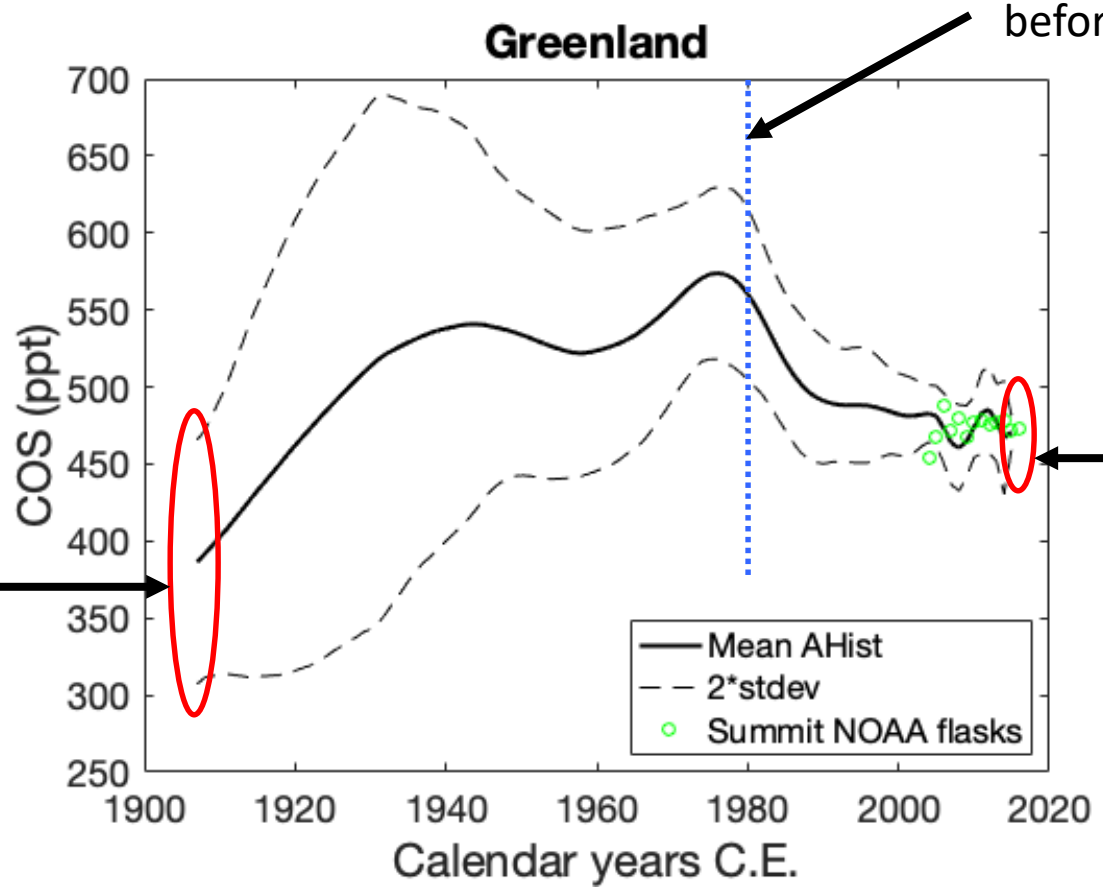

- **RED:** Measurement means show an increase at depth and turn around
- A statistical model with MCMC sampling to get full Bayesian inference (mc-stan.org)
- **GRAY:** Range covered in MC sims (20k runs) is proportional to estimated uncertainty at each sampling depth
- All sites display a COS increase at depth



# *Firn inversions – Greenland (20k MC sims)*

More than 95% of sims show peak before 1980

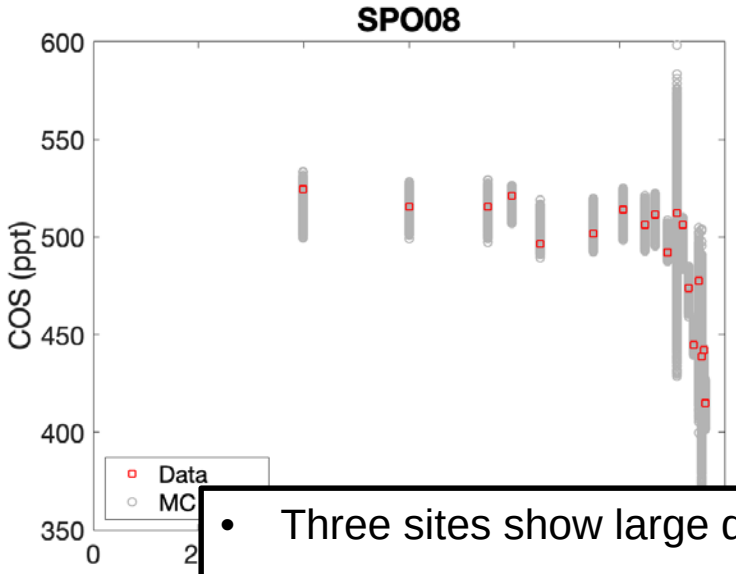


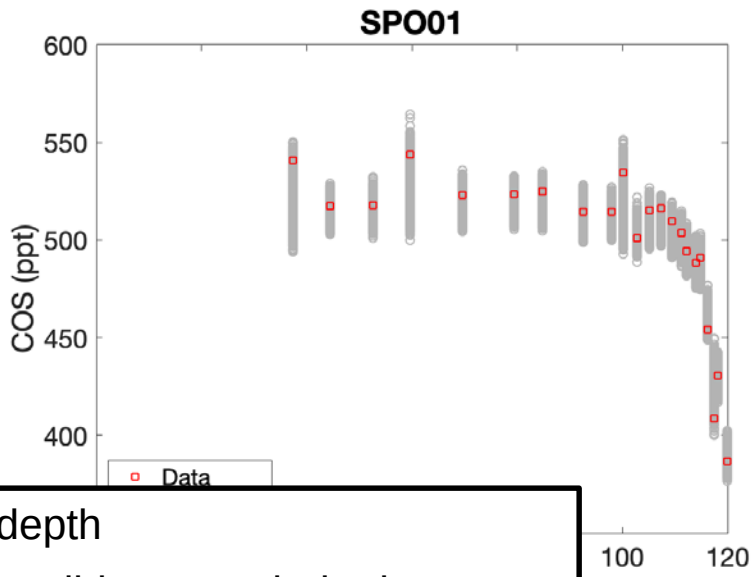

COS at $t_0$ is forced

COS at $t_{final}$ is forced

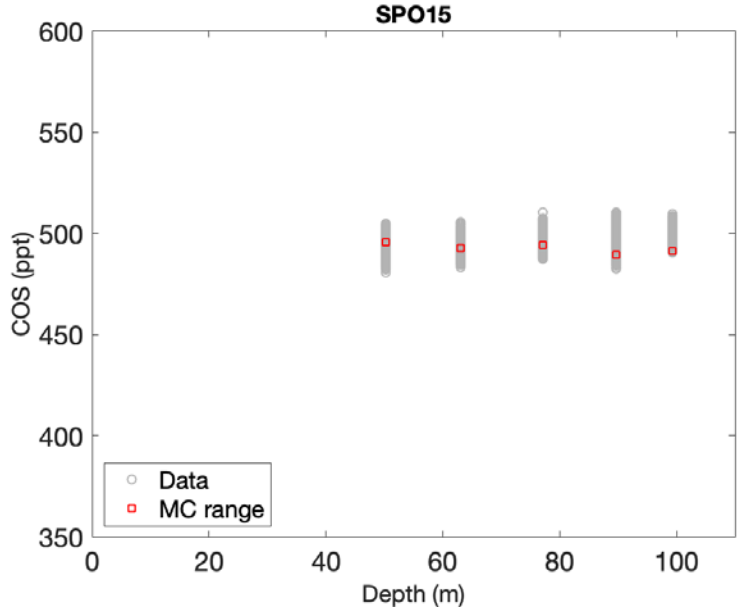
**Peak in the atmosphere in the 1970s**

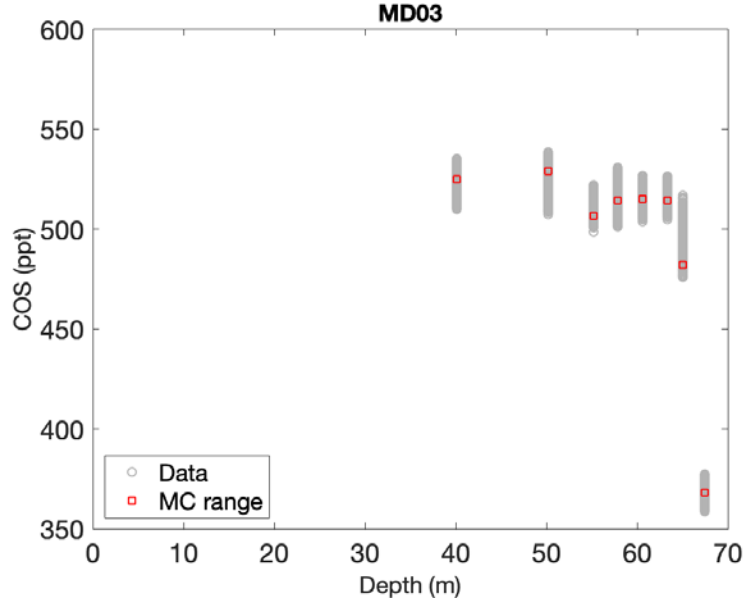
# *Firn air COS measurements – Antarctica (20k MC sims)*



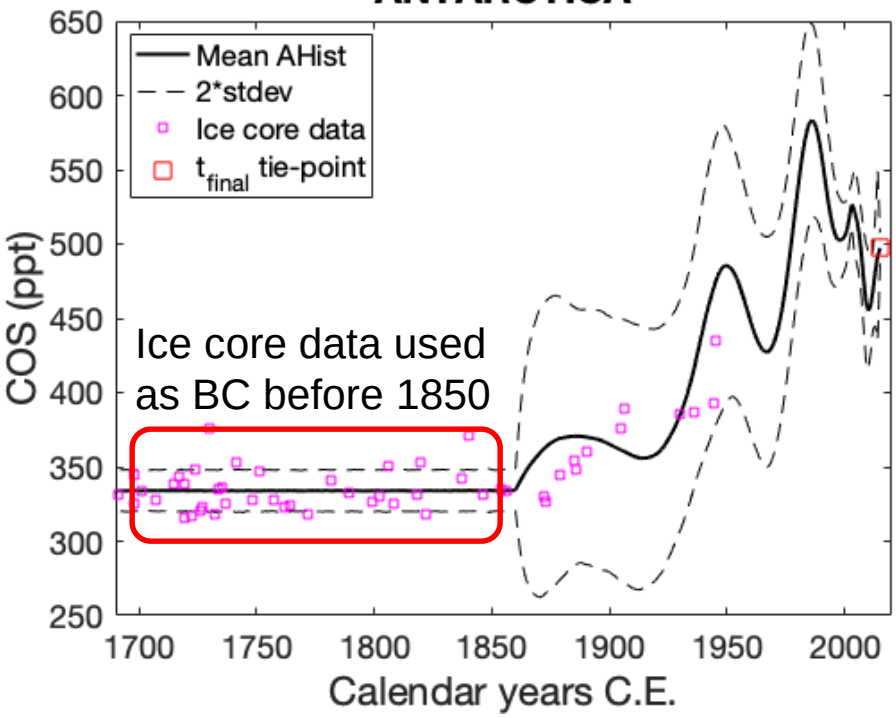




- Three sites show large decreases at depth
- SPO15 does not because the sampling did not reach the bottom

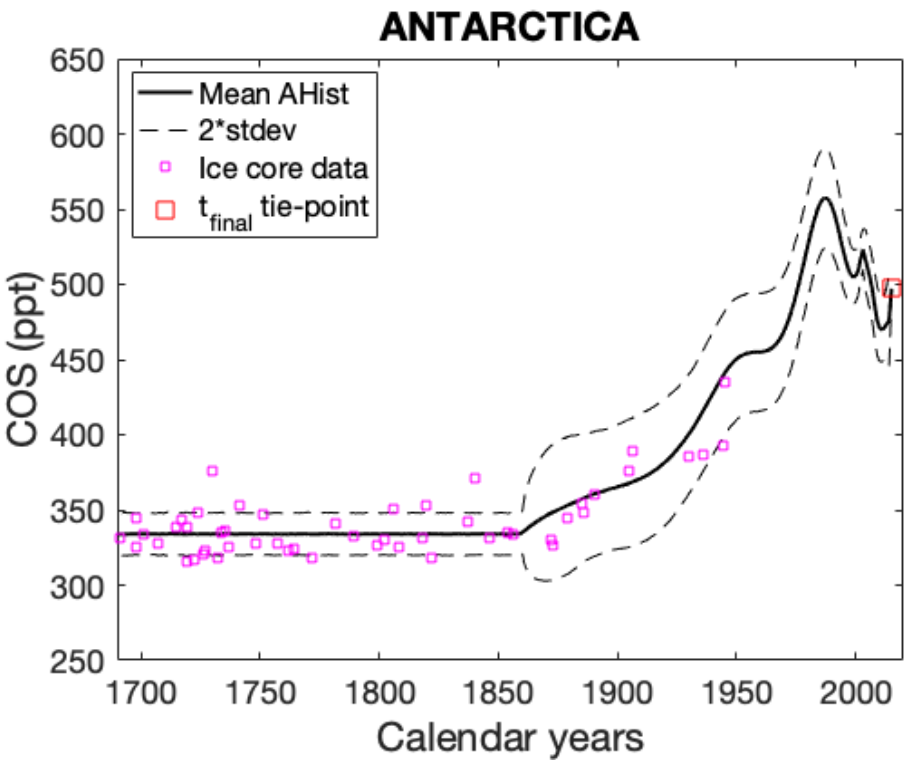


# *Firn inversion results – Antarctica (20k MC sims)*



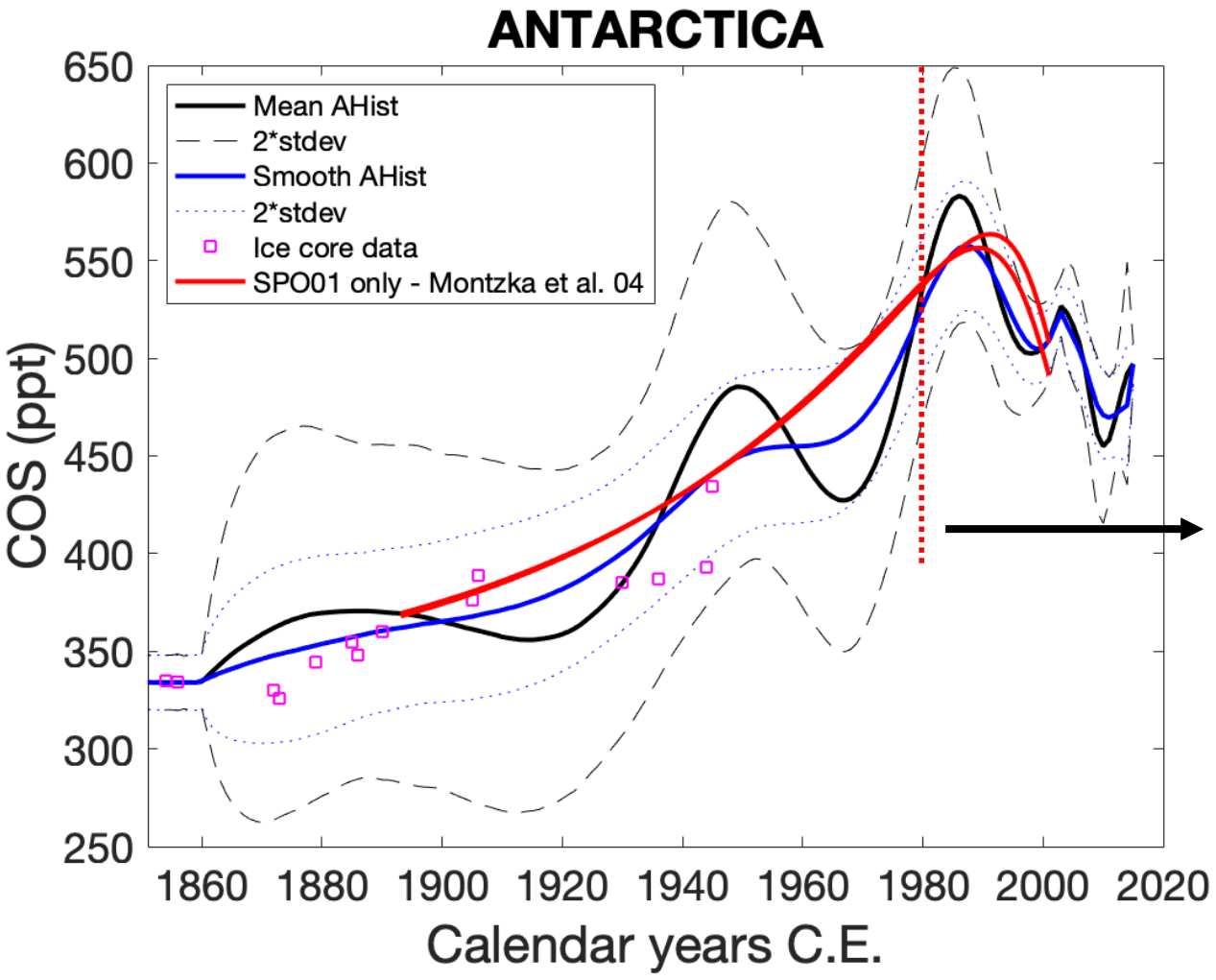

Forced smoothing




- Antarctic firn data provide a much longer atmospheric record than Greenland
- Antarctic record also displays coherent information at higher frequencies

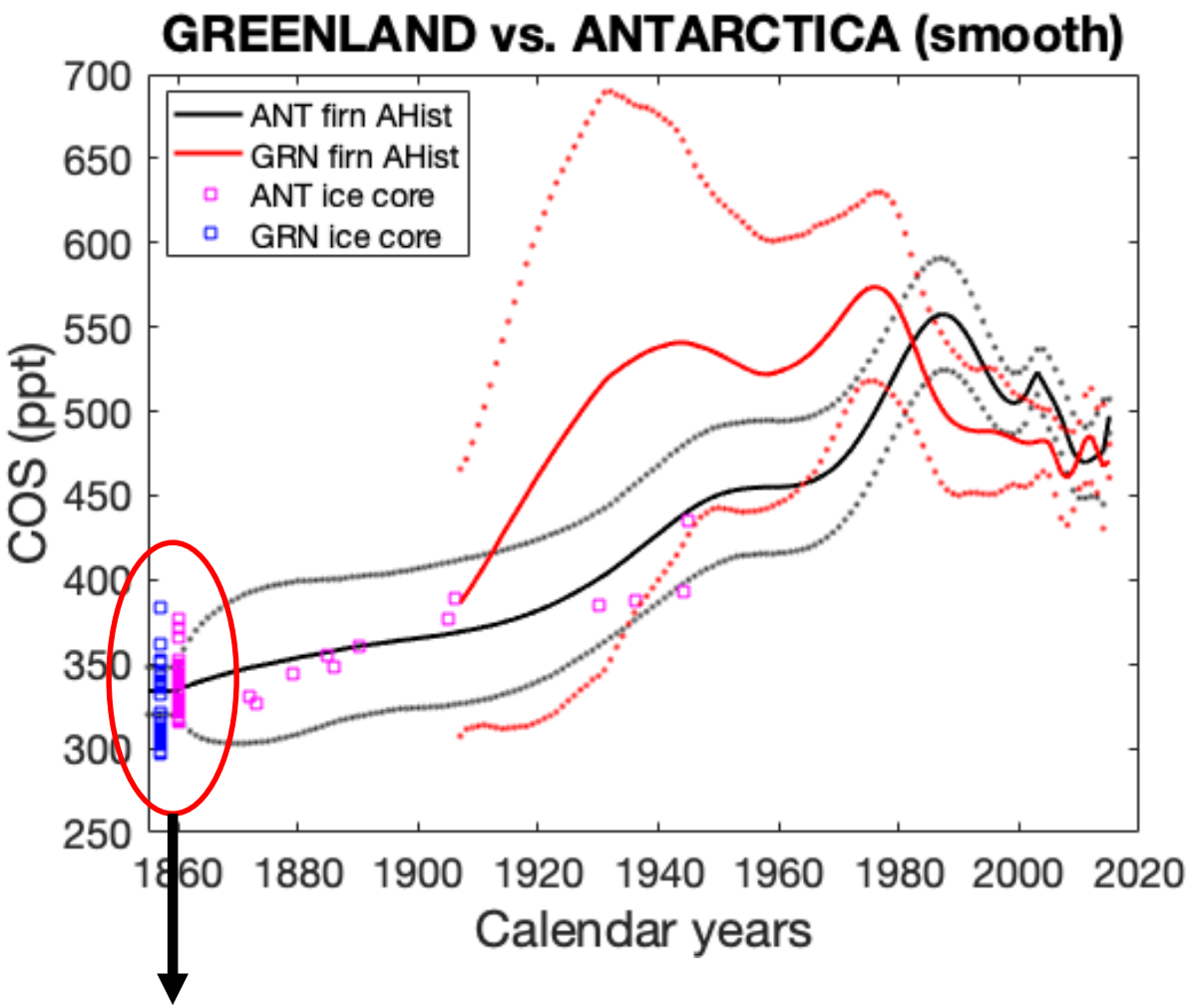
- **Record with larger variability explains the firn data better!!!**
- **Peak in the 1980s**

# *Firn inversion results – Antarctica (based on 15k MC sims)*




Peak after 1980

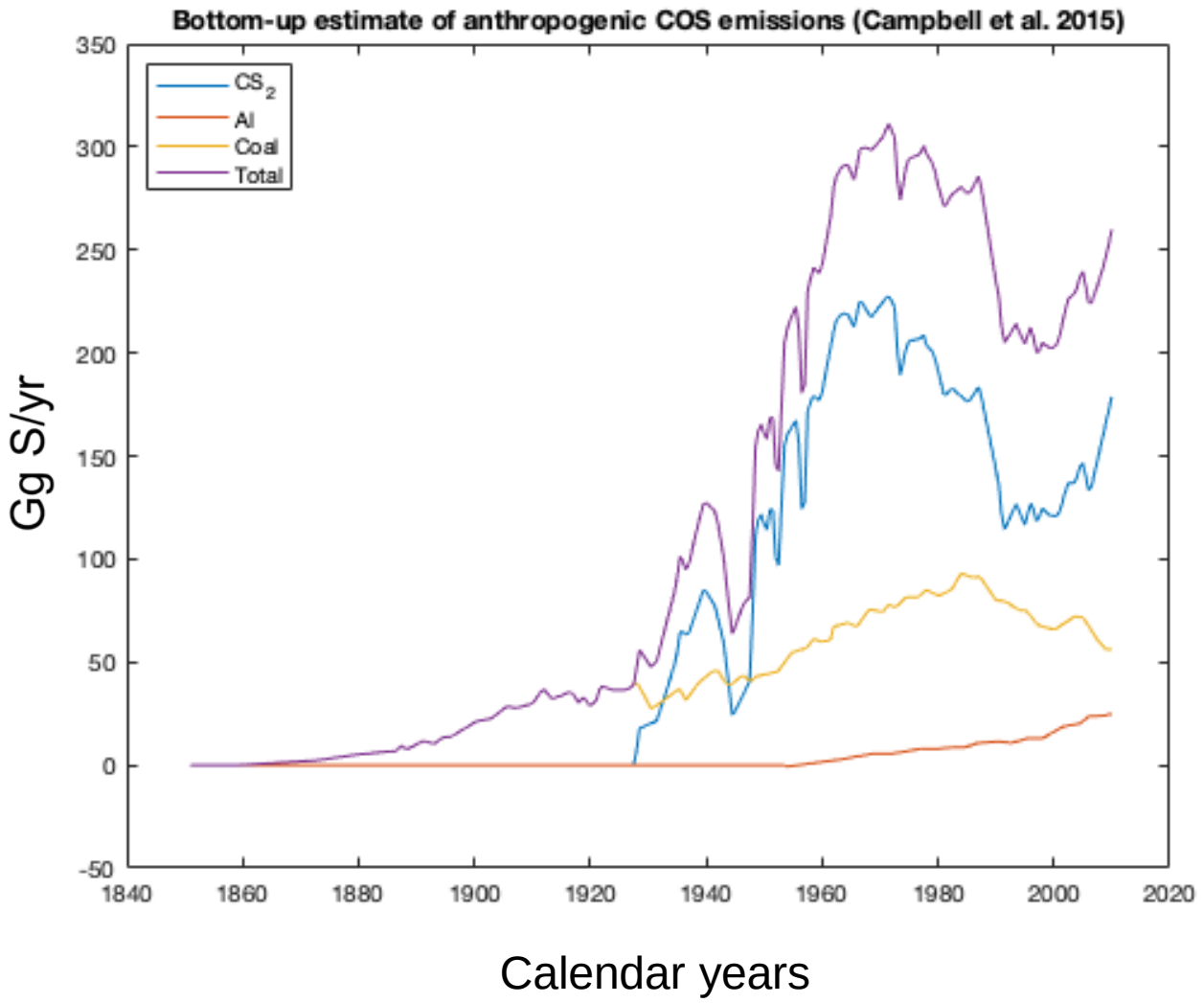
# *Firn inversions – Greenland vs. Antarctica*




**Ice core data prior to 1850 from both Greenland and Antarctica on a collapsed time scale**

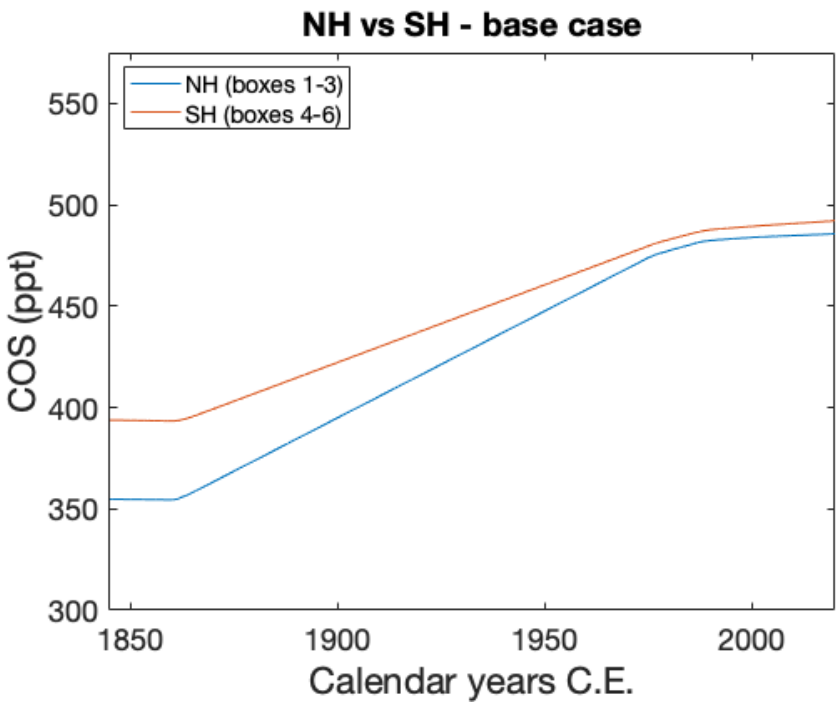
- Large increase in COS evident in both atmospheres
- Peak in both hemispheres in late 20$^{th}$ century
- NH peaks before SH
- Peak followed by decline to present day when the levels in two hemispheres are close to equal
- Is this variability driven by anthropogenic emission?

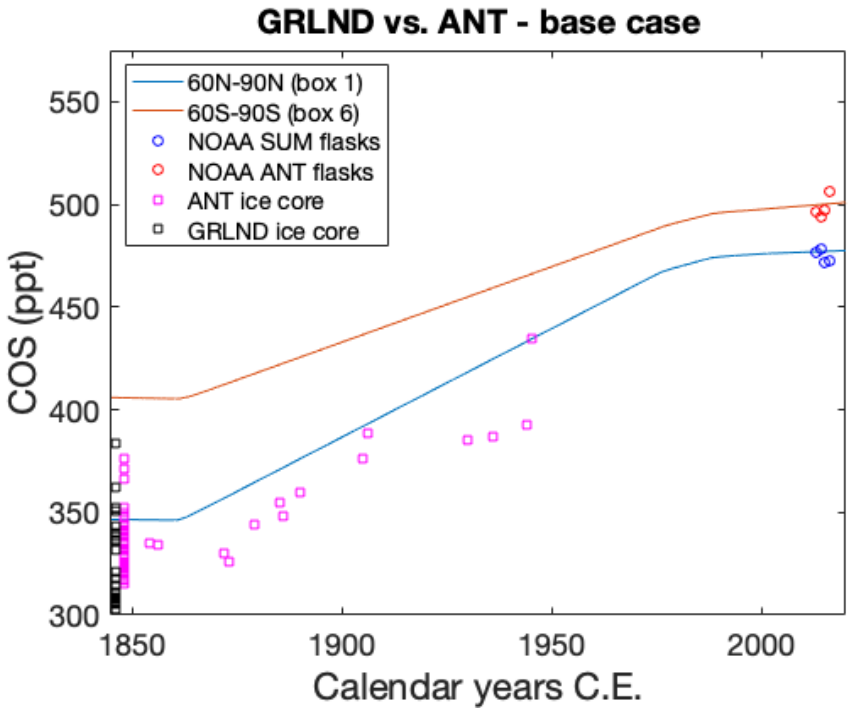
# *Bottom-up anthropogenic COS emissions estimates*




- Bottom-up estimate of anthropogenic emissions display strong ramp-up in qualitative agreement with firn records
- At the peak, anthropogenic emissions account for only 25% of the COS budget
- Present-day level is not significantly lower than the peak.

# 6-box ocean-atmosphere model – base case (current budget)



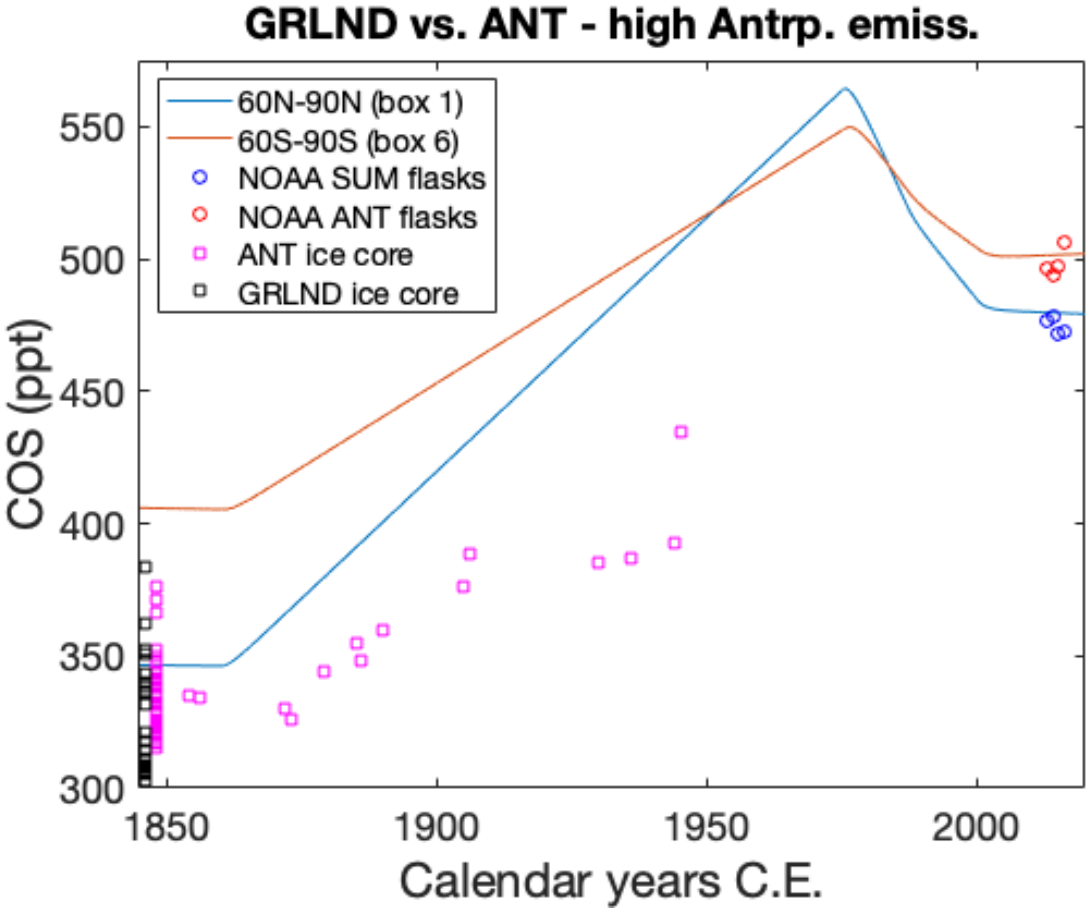

<u>From preindustrial to present</u>

- Anthropogenic emissions change similar to bottom-up inventory
- Small biomass burning increase following Campbell et al. (2017)
- Oceans set at present day productivity and respond passively
- Constant uptake rate (lifetime = 2 y)

<u>At the the Poles (60 – 90 deg)</u>

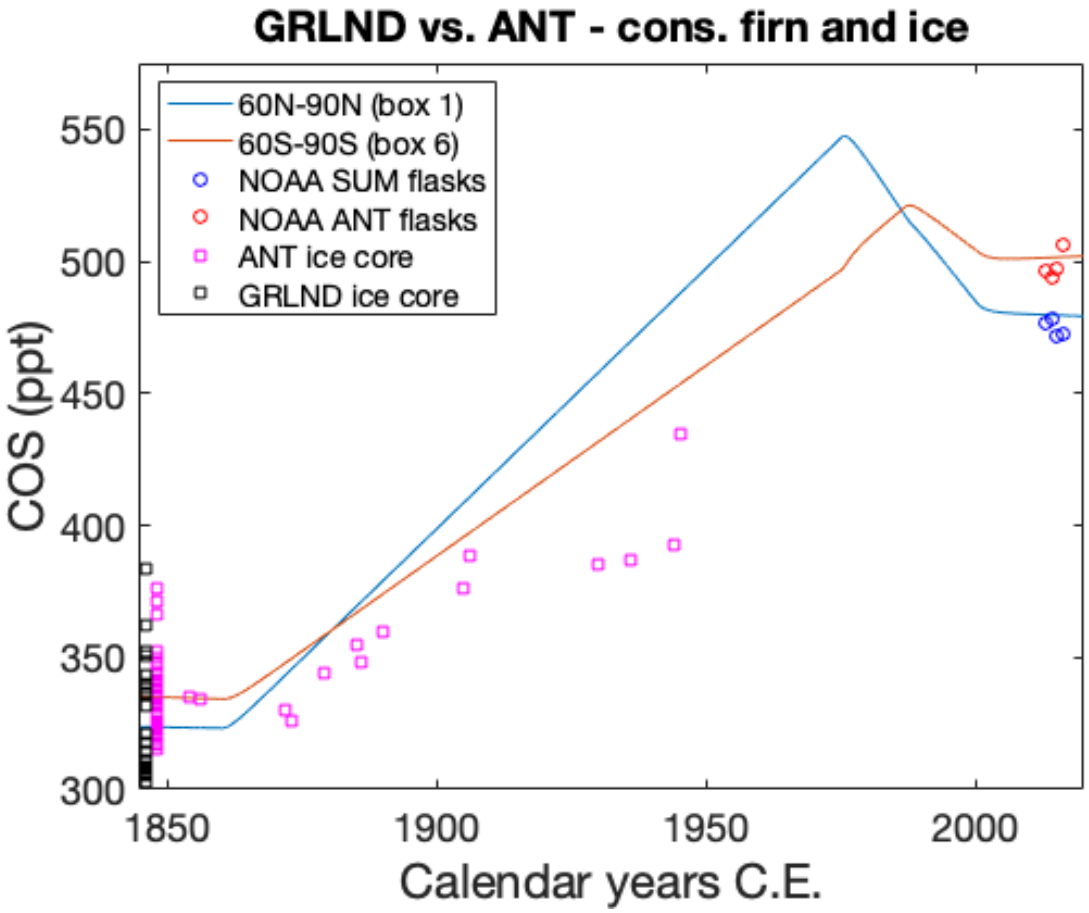
- We get present-day levels right but no peak(s) in late 20$^{th}$ century
- Antarctic levels are too high in the preindustrial atmosphere
- Antarctic always higher than Greenland

# 6-box ocean-atmosphere model – High Anthropogenic




- Doubled anthropogenic emission in late 20th century to get a peak comparable to seen in firn air records
- GRLND exceeds ANT at the peak
- GRLND and ANT peaks are simultaneous
- No impact on preindustrial SH levels

# 6-box ocean-atmosphere model – High Anthropogenic, Var. Ocean




- Lower emissions from mid-high latitude SH oceans during preindustrial era
- SH ocean emissions get back to present day levels late in the 20$^{th}$ century
- GRLND higher than ANT most of the time
- GRLND and ANT peaks separated in time

## *Conclusions*

- Inversions based on firn data from multiple sites display previously undocumented variability in NH and SH atmospheres
- Anthropogenic COS emissions were easily double the present day during the second half of the 20$^{th}$ century
- Results suggest ocean production of COS (or may be DMS) possibly changed during the 20$^{th}$ century
- Changes in removal rate cannot be ruled out, but difficult to constrain without understanding the behavior of ocean sources

## *Ongoing work*

- We are funded to measure COS in a shallow ice core from Greenland, which should provide tighter constraints on Greenland firn inversions