How reliably can we estimate inter-annual changes in global emissions of long-lived trace gases from atmospheric measurements?

Steve Montzka[1], Geoff Dutton[1,2], Eric Ray[2,3], Robert Portmann[3], Martyn Chipperfield[4]

[1]NOAA Global Monitoring Laboratory, USA
[2]CIRES, Univ. of Colorado, USA
[3]NOAA Chemical Sciences Laboratory, USA,
[4]Univ. of Leeds, UK

Thanks to:

C. Siso, D. Nance, B.D. Hall, F. Moore, J.W. Elkins, D. Mondeel, M. Gentry

S. Davis[3], W. Feng[4] (3-D modeling)

Station personnel at NOAA sites and cooperative institutes, and

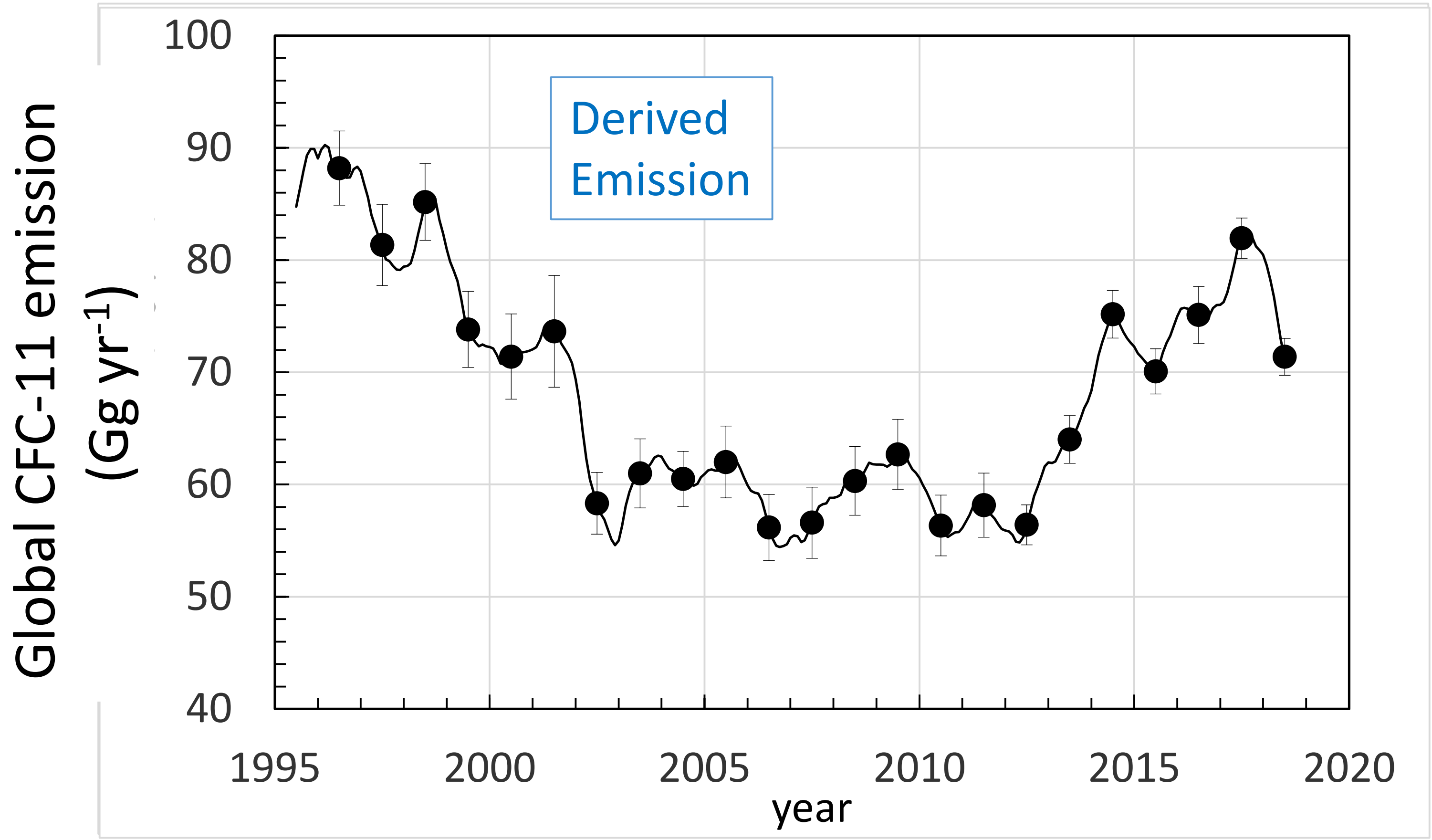
& the AGAGE community of scientists

NOAA-GML's virtual Global Monitoring Annual Conference
22 June 2020

CFC-11 global emission derived from remote atmospheric measurements

Hourly measurements at 5 sites

Weekly measurement at 12 sites



→**Emissions derived** with simple mass balance considerations (3-box-model analysis):

$$\underline{dG_{F11}/dt} = \textbf{Emission} - k^*G$$

Measured

Are these inter-annual changes real?

Is the 2017-2018 difference robust?

Uncertainties (2 to 4 Gg yr^{-1}) include measurement precision & consistency, atmospheric variability, & an estimate of network representation of the true global surface mean

Uncertainties (2 to 4 Gg yr^{-1}) don't explicitly include:

* calibration consistency:

0.1% error in annual mole fraction

→ 5 Gg yr^{-1} emission error

- NOAA inter-annual calibration consistency is ~0.03%

- Annual global mean variability (NOAA vs AGAGE) is also ~0.03%

or

* Variability in atmospheric transport and dynamics

particularly between loss region and measurement locations at Earth's surface

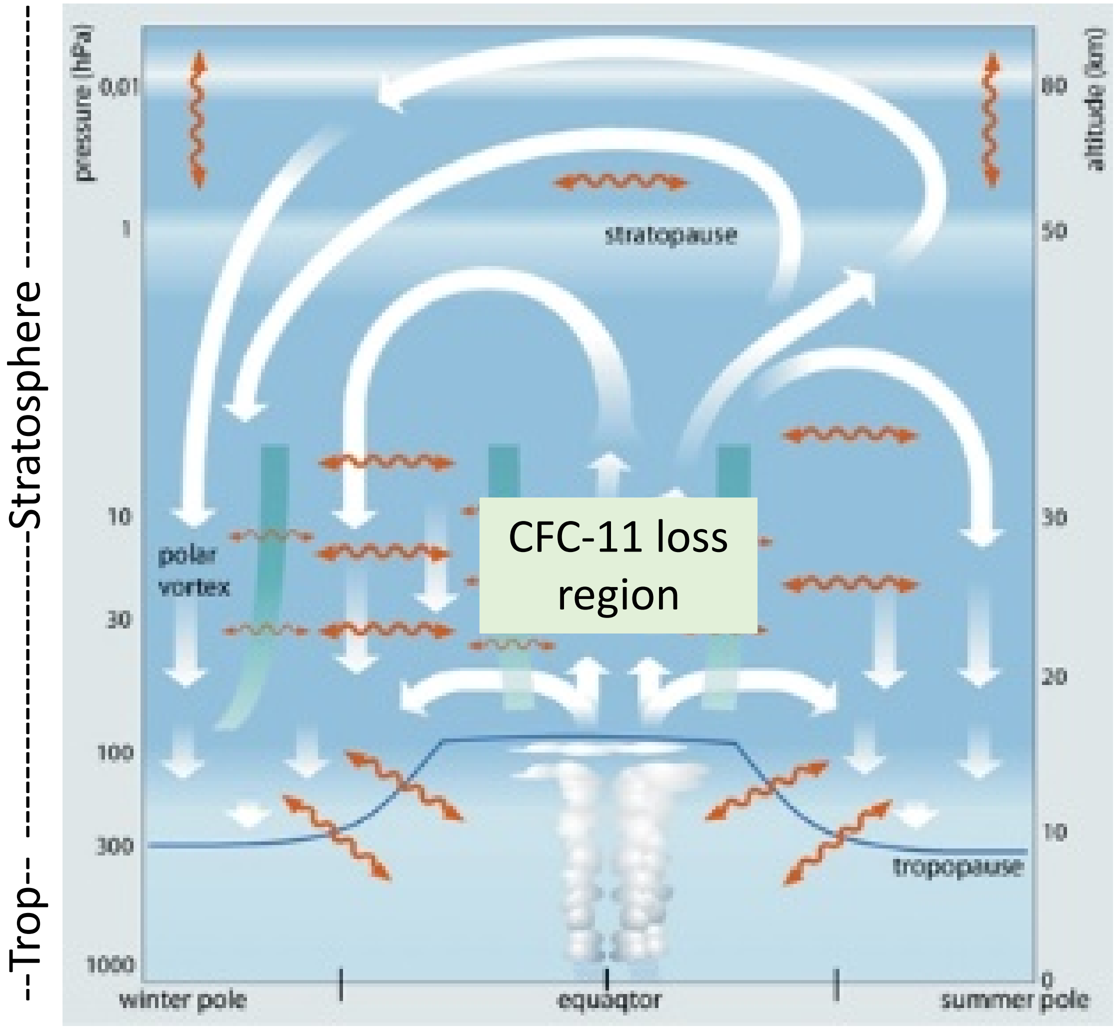
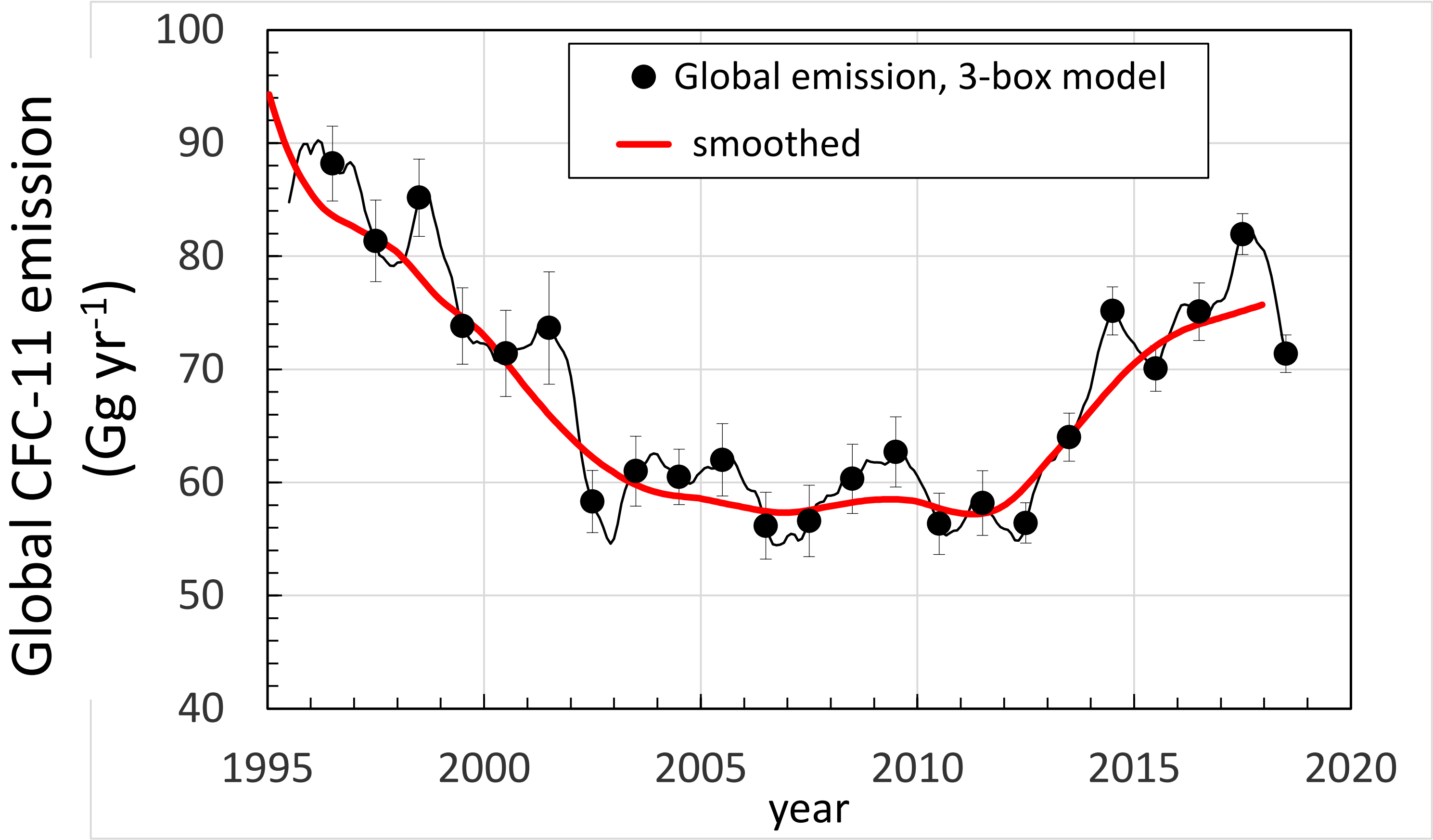


Image from web: https://www.goethe-university-frankfurt.de/47669287/Atmospheric_Tracers

Investigating the influence of variability in dynamics and air transport on derived emissions (e.g., see Ray *et al*., 2020*)



From the observations,

- **derive a smoothed emission history**
- **use the smoothed emission history as input to**:

1) a simple 3-box model

2) two 3-D global models using different meteorology

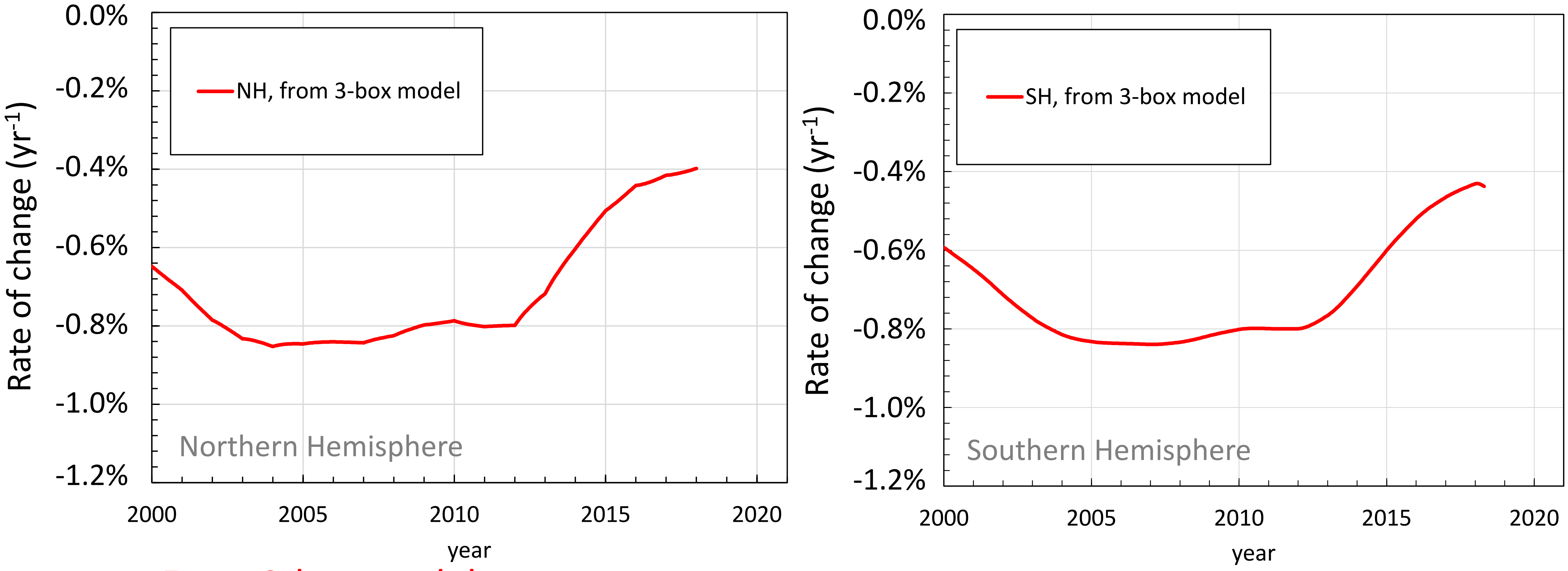
Then:

Assess *measured* vs. *simulated* mole fraction rates of change

Ray *et al*. The influence of the ***stratospheric Quasi-Biennial Oscillation*** on trace gas levels at the Earth's surface. *Nat. Geo*. **13**, 22-27 (2020). https://doi.org/10.1038/s41561-019-0507-3

Using the smoothed emission history as input:

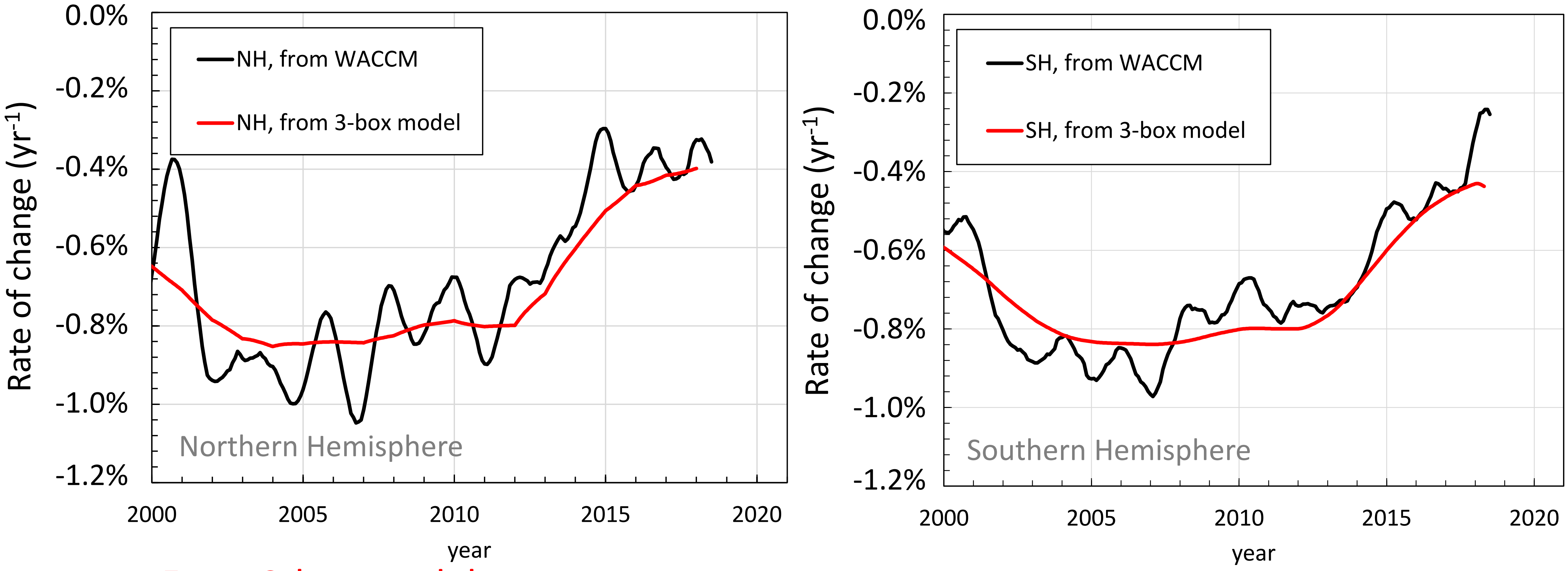
Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)



From 3-box model

Using the smoothed emission history as input:

Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)



From 3-box model

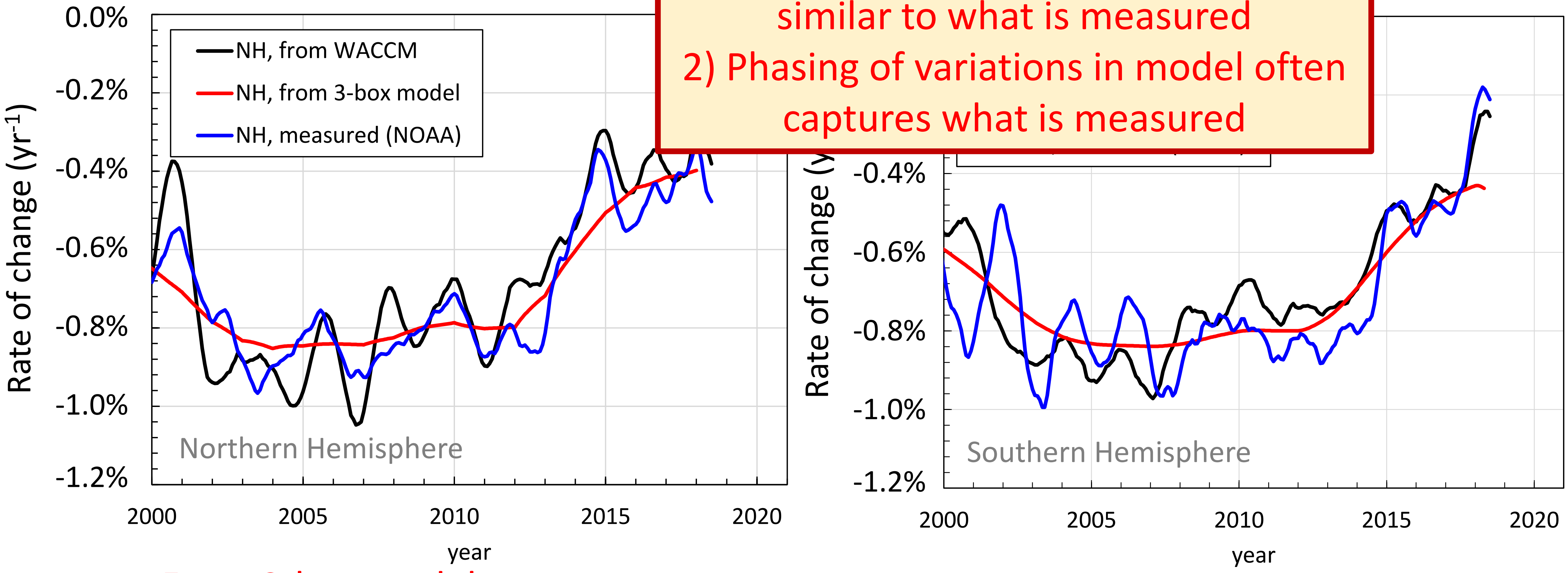
From 3-D model WACCM using specified dynamics from MERRA2

Using the smoothed emission history as input:

Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)

1) Inter-annual variability in model is similar to what is measured

2) Phasing of variations in model often captures what is measured



From 3-box model

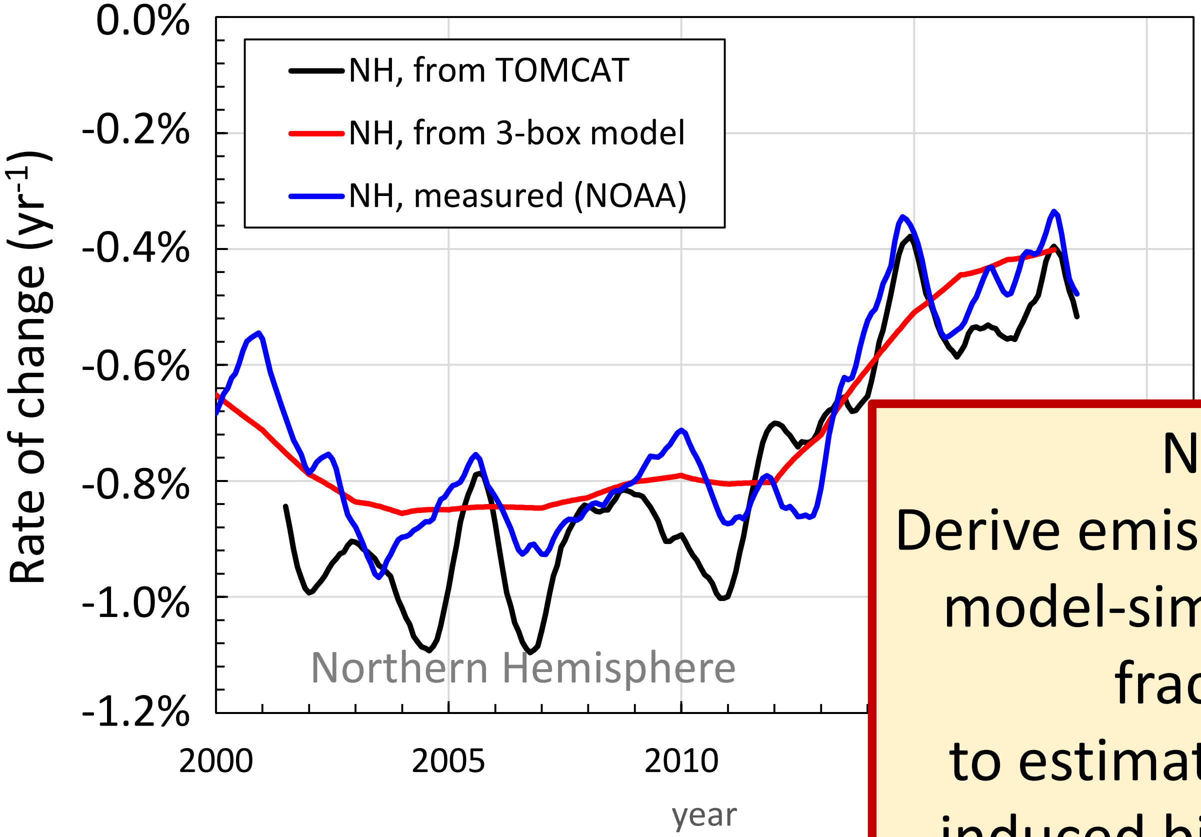
From 3-D model WACCM using specified dynamics from MERRA2

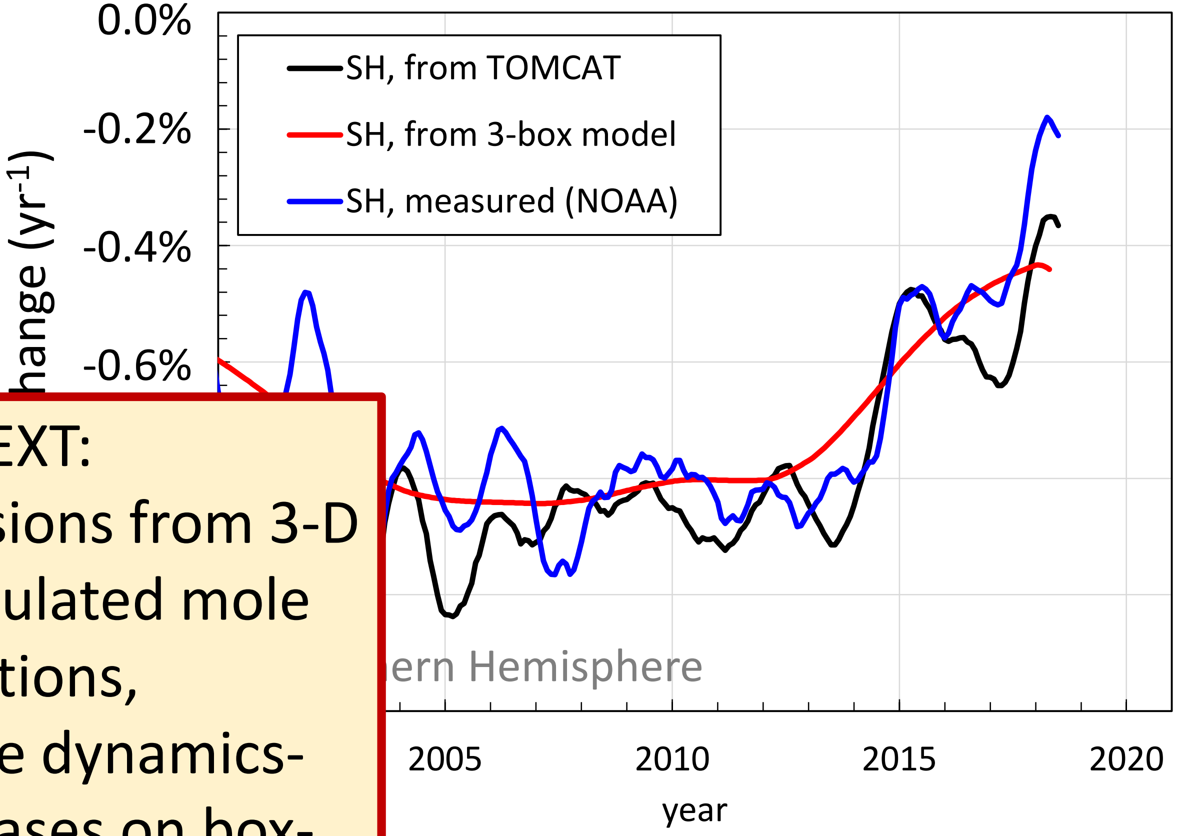
From measurements, 8 sites in NH, 4 sites in SH

Using the smoothed emission history as input:

Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)

And with a different 3-D model:





NEXT:
Derive emissions from 3-D model-simulated mole fractions,
to estimate dynamics-induced biases on box-model emissions

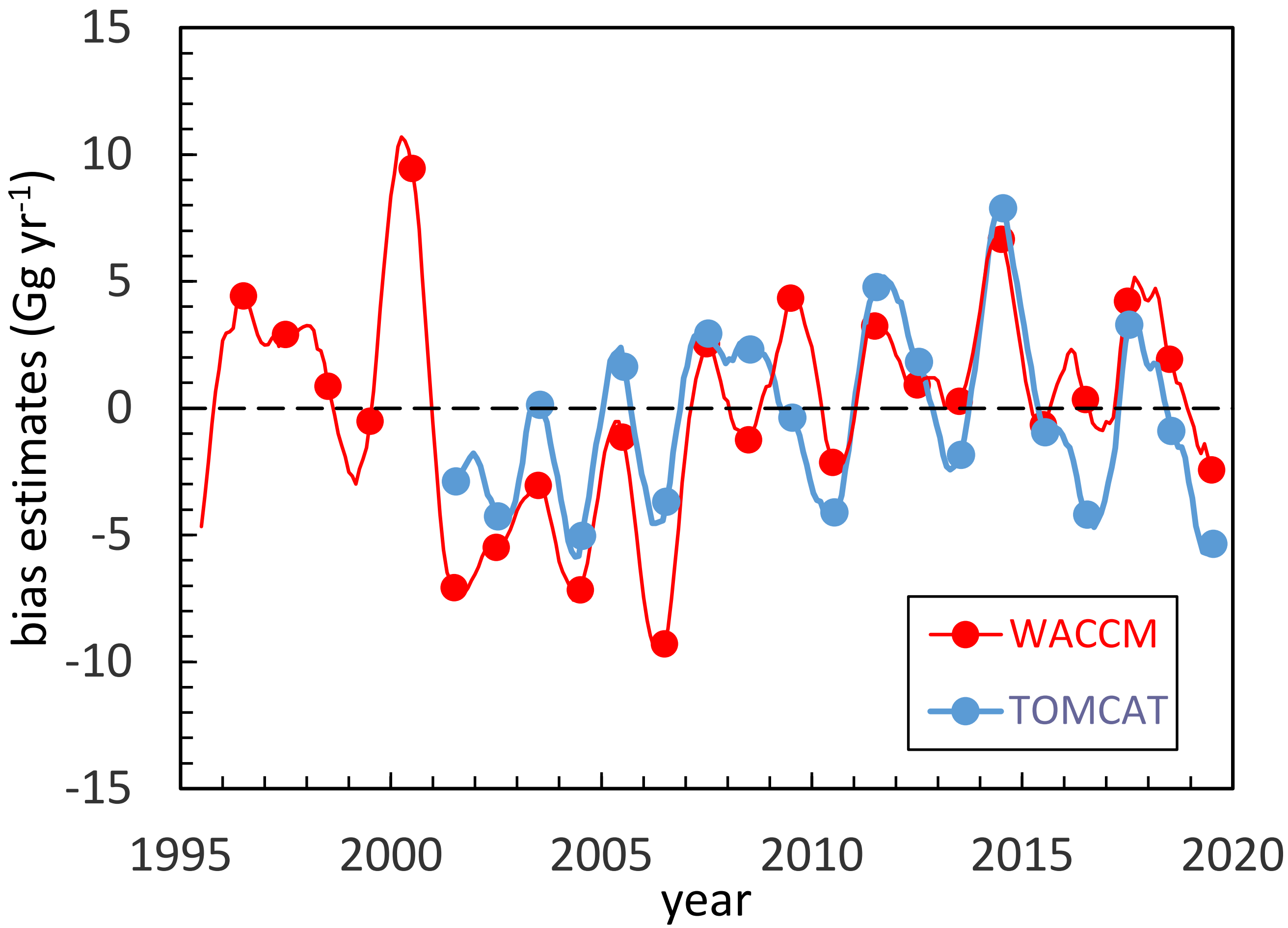
From 3-box model

From 3-D **chemical tra**[...] **ERA5**

From measurements, 8 sites in NH, 4 sites in SH

Dynamics-related biases on inferred CFC-11 emissions
→ obtained from the difference between:

* Smoothed input emissions &
* Emissions derived from 3-D model-simulated mole fractions



From both models:

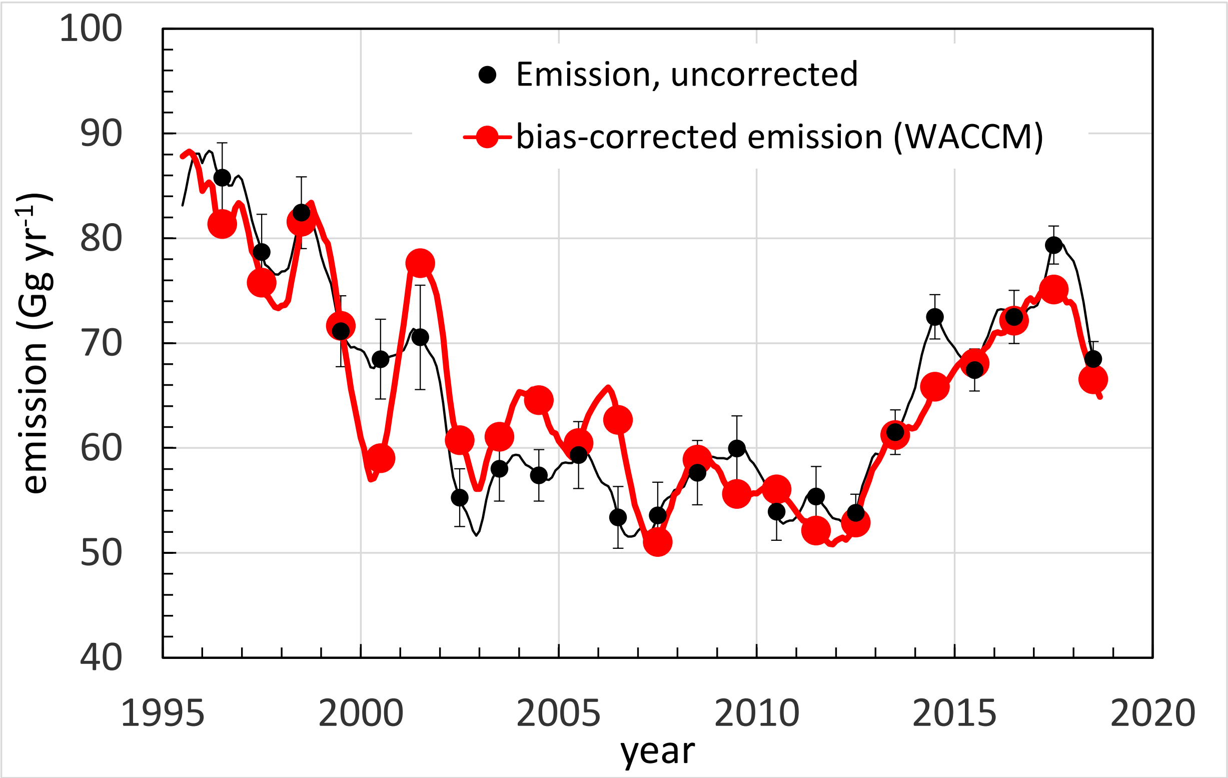
Inter-annual changes

- ***typically have the same sign,***
- ***often a similar magnitude:***
  - mean inter-annual bias: 5 Gg yr^{-1},
  - as high as 15 Gg yr^{-1}
  (compared to 2 - 4 Gg yr^{-1} uncertainty)

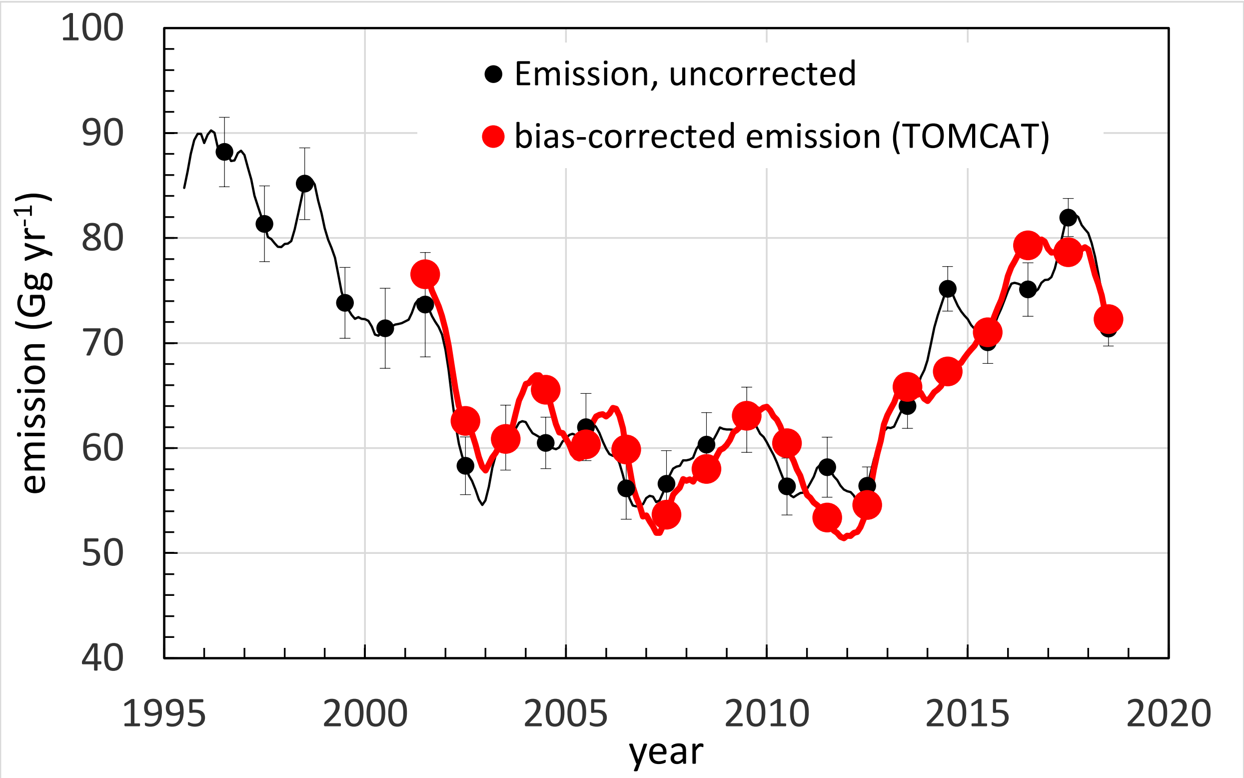
WACCM suggests ***a significant shift in 2000***, reflecting a known perturbation in the stratospheric circulation (Randel et al., 2006)

Inferred global CFC-11 emissions including dynamics-related biases derived from 3-D models

From WACCM



From TOMCAT



→ Smoother emission changes implied after 2010, perhaps to be expected

Pre-2010 variability is sometimes enhanced

→ real?

Enhanced errors in observations or models?

Summary:

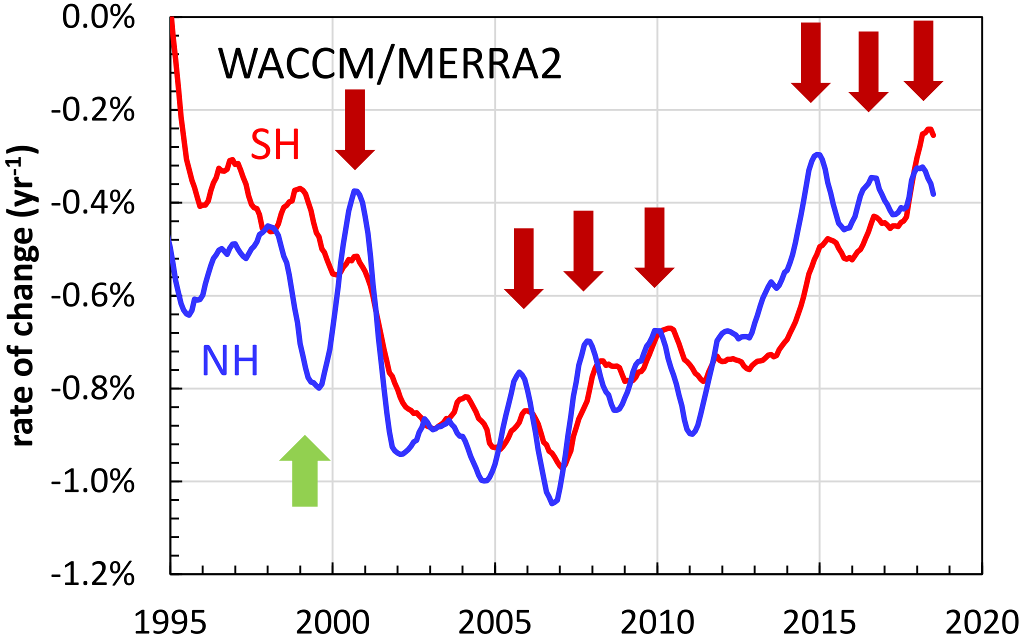
Improvements in measurement capabilities (precision, consistency, global coverage) yield uncertainties in derived annual emissions of 2 to 4 Gg yr^{-1} are implied.

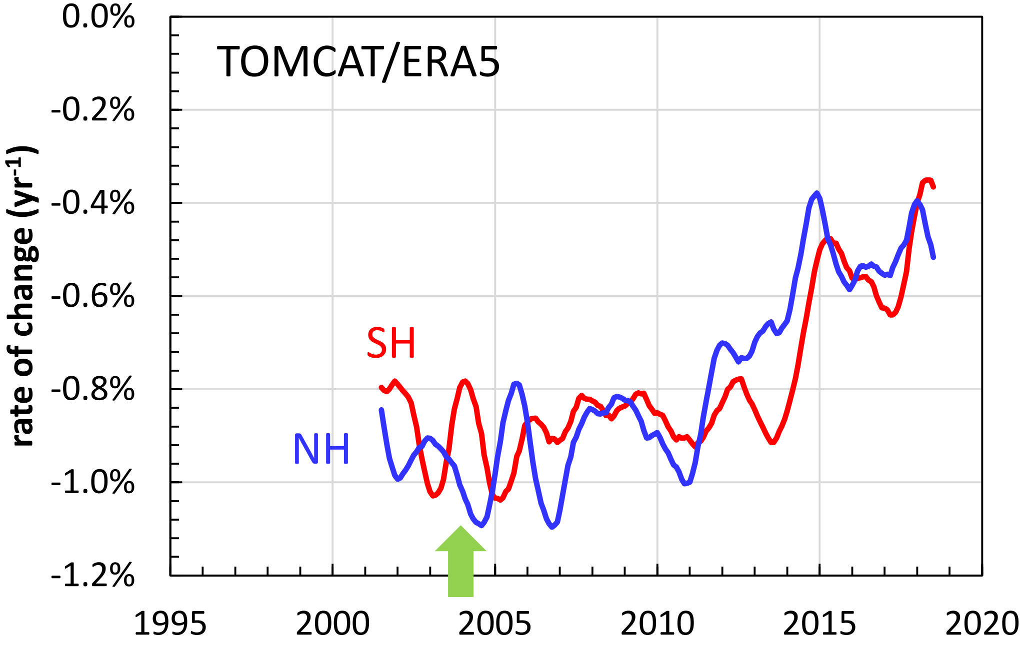
3-D models with reanalysis meteorology suggest that larger biases in year-to-year emission changes can stem from variability in dynamics.

→ some dynamics-related biases can persist for multiple years (post 2000)

Models do a good job of simulating measured interannual variability in mole fraction trends in some years, not all.

Assessing emission changes on a year-to-year basis, (e.g., for rapid feedback to policymakers) requires an accurate estimate of these non-emissive influences on derived global emissions.





NH vs SH rates from 3-D models:

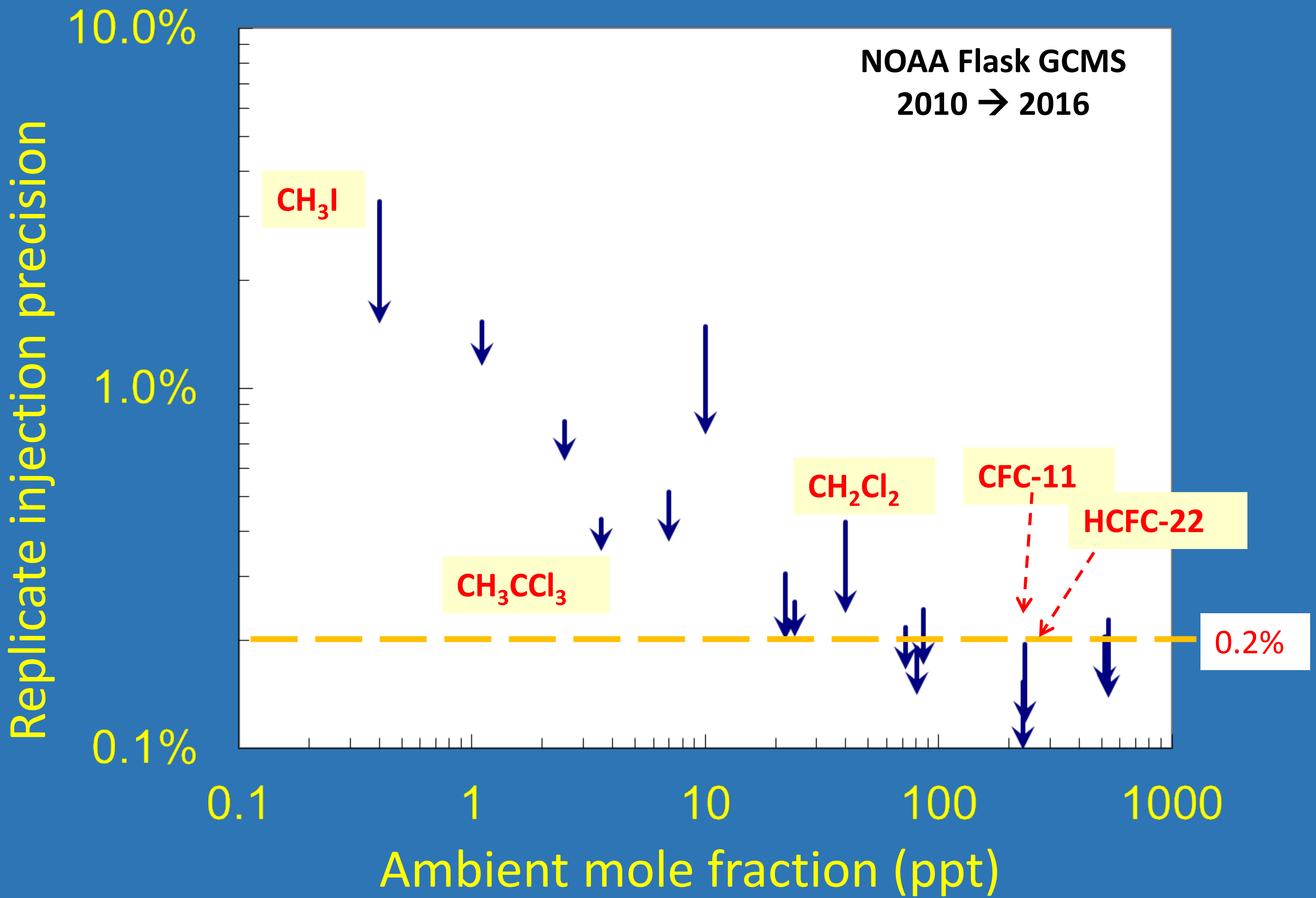
Much of the variability has similar phasing in the two hemispheres

→ variability out of phase less often (N –S exchange?)

→ implying source of variability as begin the BDC or strat-trop exchange (e.g., QBO as in Ray *et al.*, 2020).

Looking at uncertainties: measurement precision at ppt-levels.

→ mean replicate injection precision *vs.* mole fraction:



Estimating uncertainty in global mean mole fraction from 12 measurement sites:

Network 1	Network 2	Network 3	Network 480
$X_{10} \pm \sigma$	$X_9 \pm \sigma$	$X_{10} \pm \sigma$	$X_9 \pm \sigma$
$X_9 \pm \sigma$	$X_9 \pm \sigma$	$X_{10} \pm \sigma$	$X_8 \pm \sigma$
$X_8 \pm \sigma$	$X_8 \pm \sigma$	$X_8 \pm \sigma$	$X_8 \pm \sigma$
$X_7 \pm \sigma$	$X_8 \pm \sigma$	$X_7 \pm \sigma$	$X_6 \pm \sigma$
$X_6 \pm \sigma$	$X_6 \pm \sigma$	$X_7 \pm \sigma$	$X_6 \pm \sigma$
$X_5 \pm \sigma$	$X_5 \pm \sigma$	$X_4 \pm \sigma$	$X_3 \pm \sigma$
$X_4 \pm \sigma$	$X_5 \pm \sigma$	$X_4 \pm \sigma$	$X_3 \pm \sigma$
$X_3 \pm \sigma$	$X_1 \pm \sigma$	$X_2 \pm \sigma$	$X_3 \pm \sigma$
$X_2 \pm \sigma$	$X_1 \pm \sigma$	$X_2 \pm \sigma$	$X_2 \pm \sigma$
$X_1 \pm \sigma$	$X_1 \pm \sigma$	$X_1 \pm \sigma$	$X_2 \pm \sigma$

G_1 G_2 G_3 . . . . G_{480} → $G \pm \sigma$ <<< Global mean

a) Annual site means (X_j) are derived from a random draw of monthly mole fractions given the measured s.d. (σ).

b) Sites used in estimating a global mean (G) and randomly chosen.

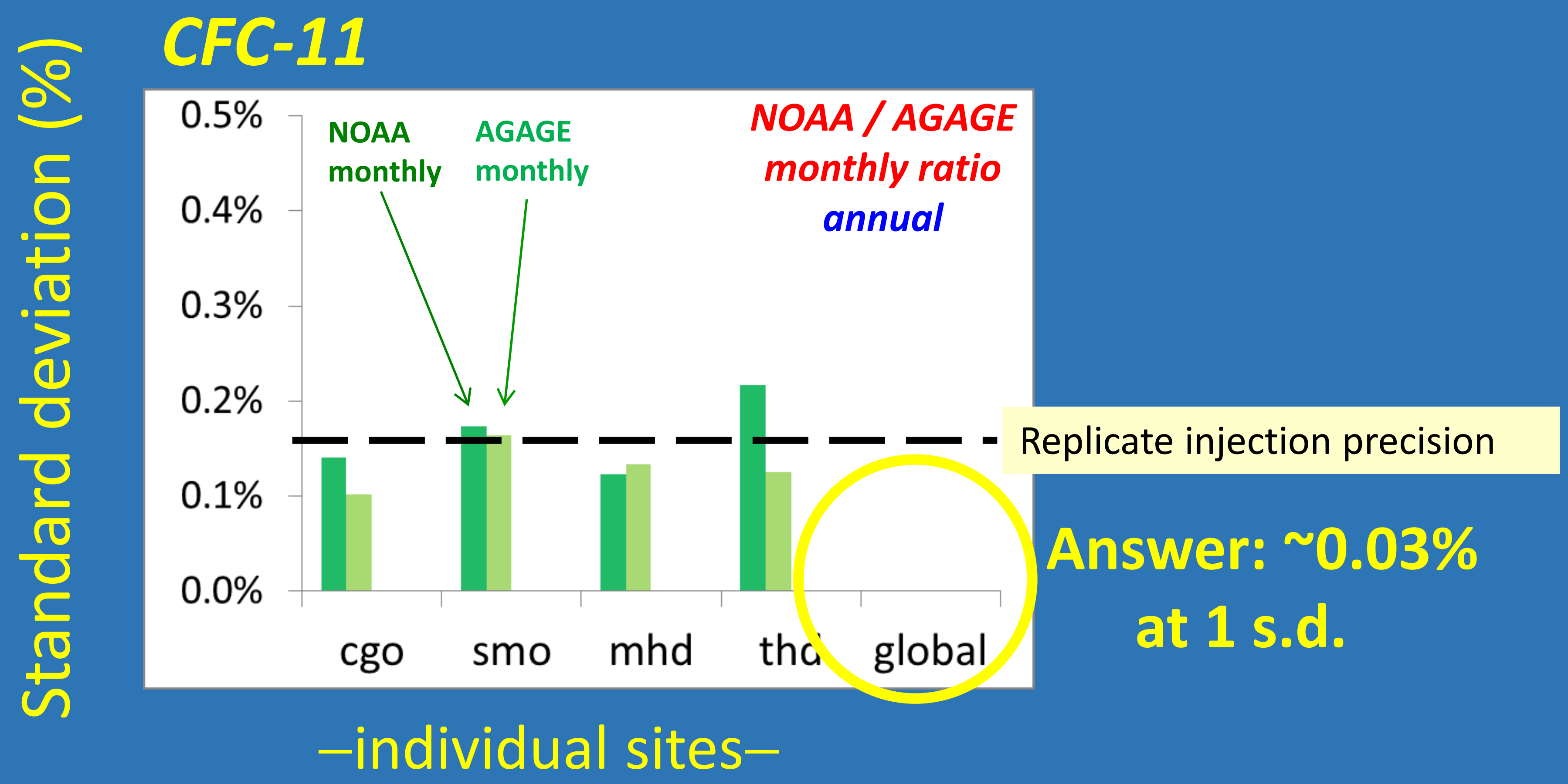
c) multiple network representations give an estimate of G and σ

Use G ± σ in simple box model to estimate emission uncertainty

Bootstrap analysis with replacement; Dlugokencky *et al.*, 1994

Looking at uncertainties: atmospheric variability.

NOAA: 4 – 5 samples/month
8-12 sites
AGAGE: 300 samples/month
5 sites
2010-2015



Answer: ~0.03% at 1 s.d.

Which is similar to our (NOAA) estimate of inter-annual calibration consistency.

Errors of ± 0.03% → ± 1.5 Gg on annual emission