Inter-annual variability in atmospheric hydroxyl as inferred from measurements of CH_3CCl_3, CH_4, and other trace gases

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Contributions from
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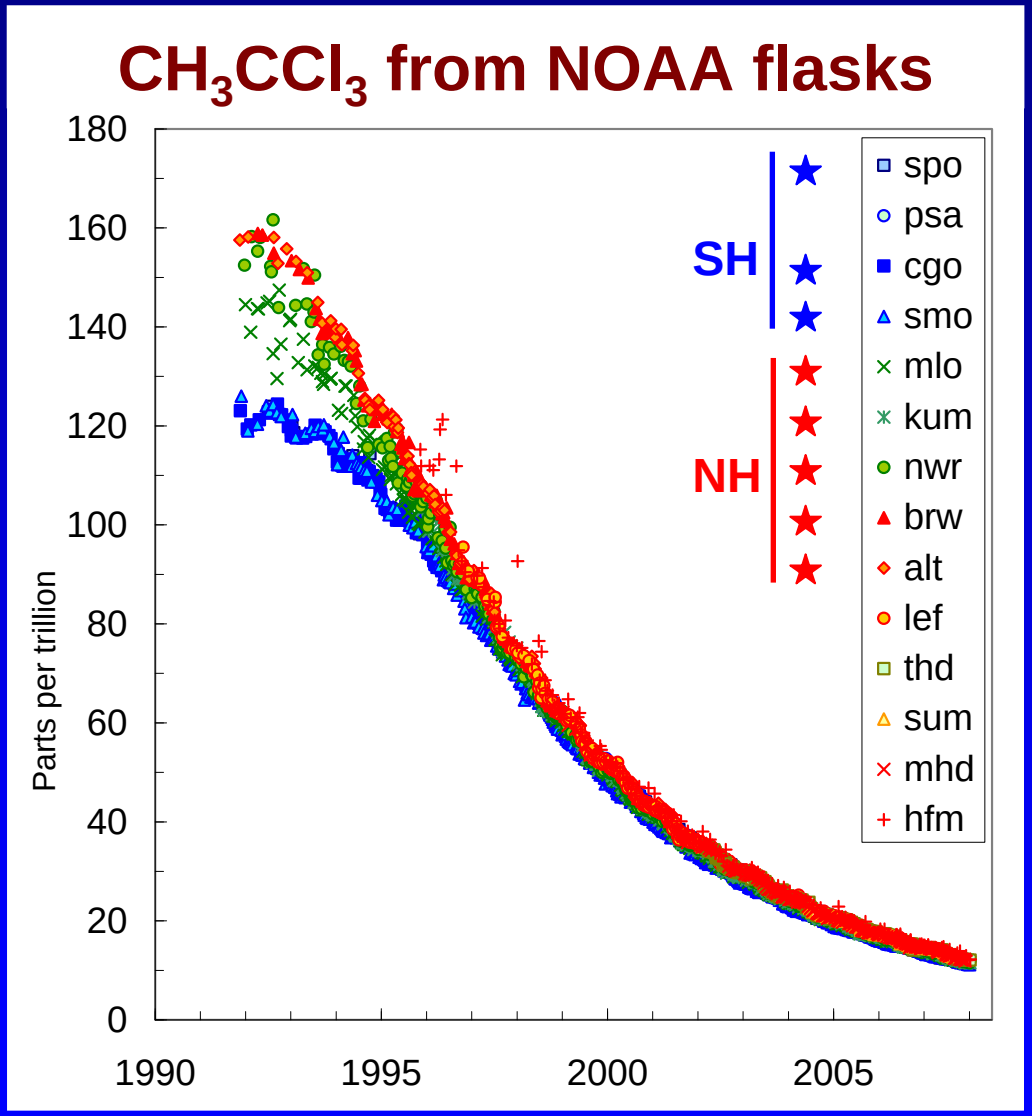
- NOAA HATS group
- NOAA CCGG group
- Station Personnel

Collaborations with:
- J. Lelieveld
- R. Prinn & AGAGE



Why the concern over OH?

*** It is central to atmospheric chemistry on global and local scales**

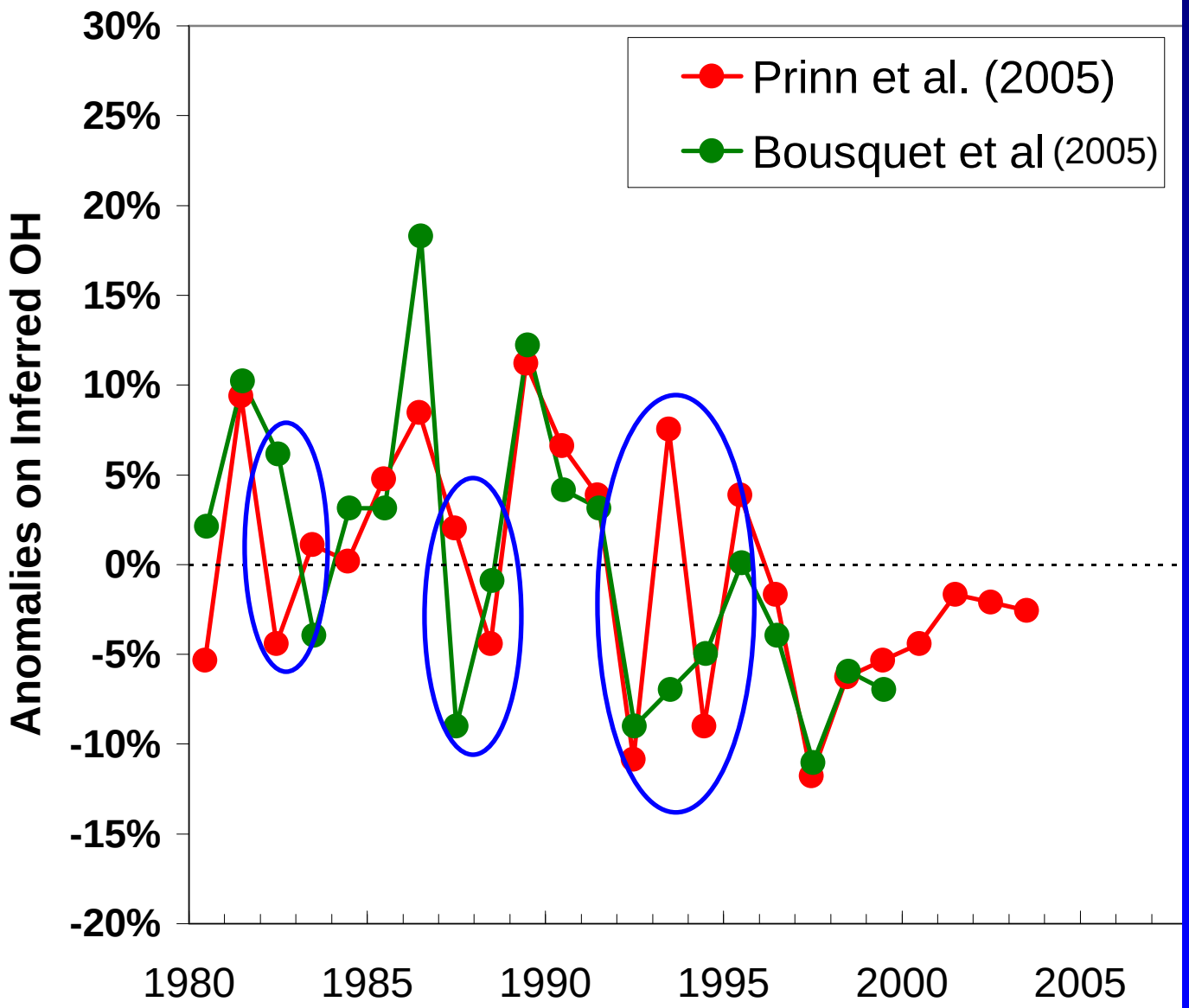
- **→ Transforms & removes trace gases**
  - ** *determines lifetime of many non-CO_2 GHGs*
- **→ Aerosol formation (S → SO_4^{2-})**
- **→ Central to tropospheric ozone chemistry**

*** Both natural and anthropogenic processes influence OH production and loss**

- **→ H_2O, light (clouds), O_3, NO_x, CO, CH_4, HCs**

*** Mean OH on large scales integrates the atmospheric response to large-scale forcings…**

a) 'Global' OH inferred from AGAGE CH_3CCl_3 and industrial emission history…



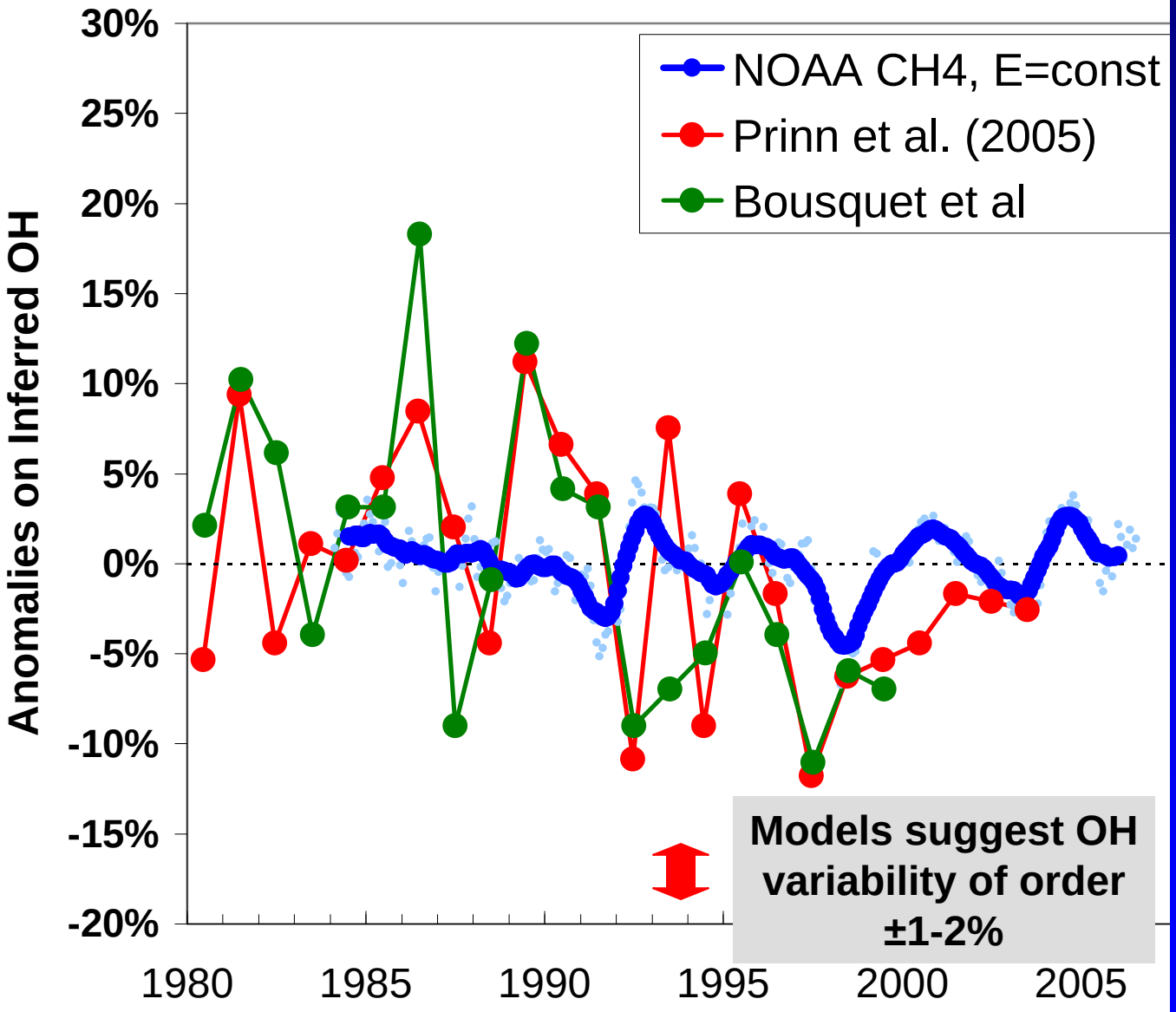
AGAGE 12-box or 3-D transport model with inter-annually varying meteorology…

OH Inter-annual variability = 7 (±6)%

…up to 23%

AGAGE data

b) Global' OH inferred from NOAA CH_4 data and *constant* emissions…



Inter-annual OH variability:

from CH_3CCl_3: Mean = 7(± 6)%

from CH_4 (constant emiss): Mean = 1.6(±1.1)% Max = 4.4%

Dlugokencky CH_4 data

Why the concern over OH *variability*?

Is 'global' mean OH highly sensitive to:

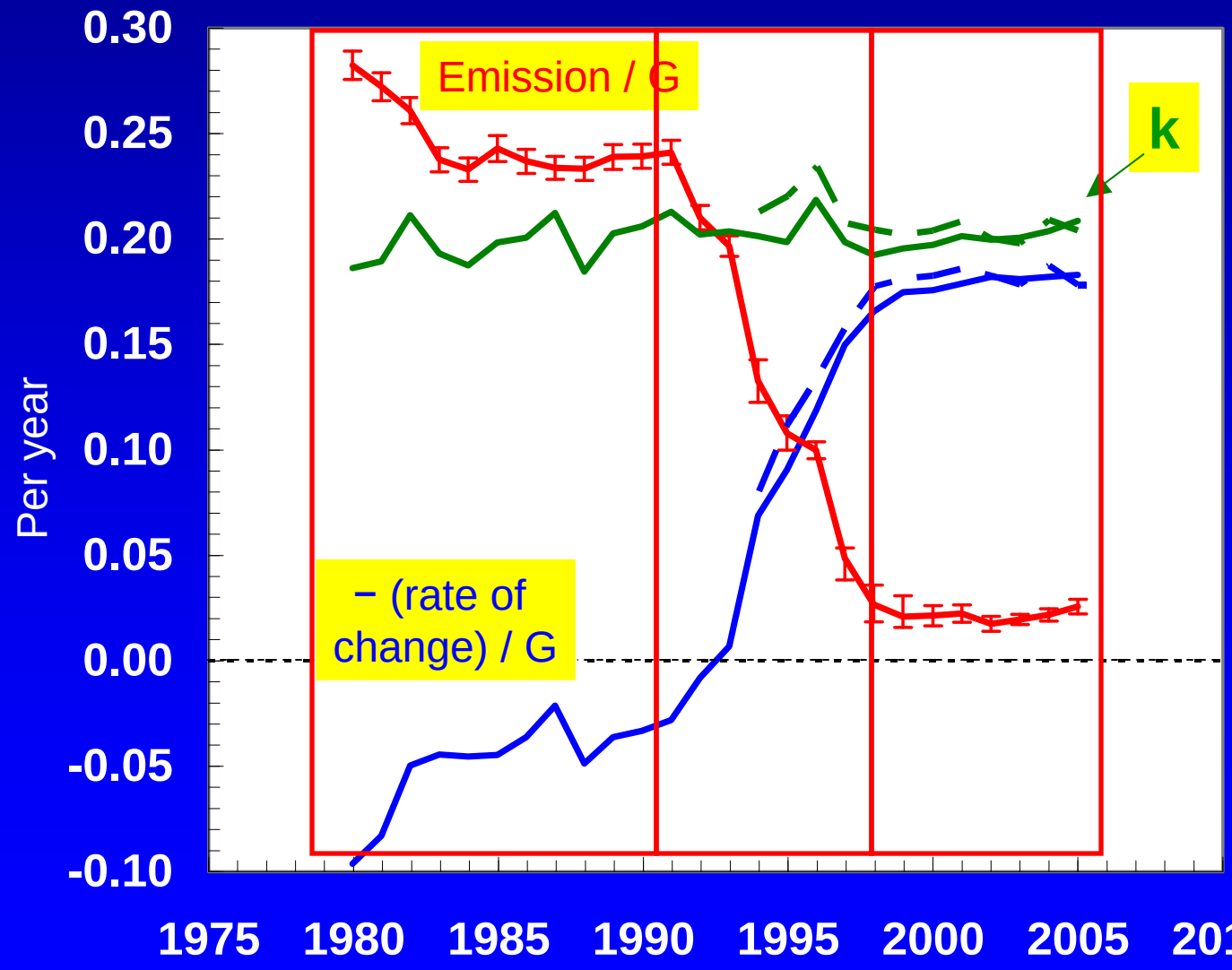
* Inter-annual or decadal changes in the atmospheric environment? Global pollution magnitudes?

How constant is the atmospheric cleansing capacity? Or trace gas lifetimes?

Is OH production dominated by primary or secondary formation pathways?

How have factors influencing estimates of k (and OH) from CH_3CCl_3 changed over time?

k(OH +…) = Emissions / G − rate of change / G

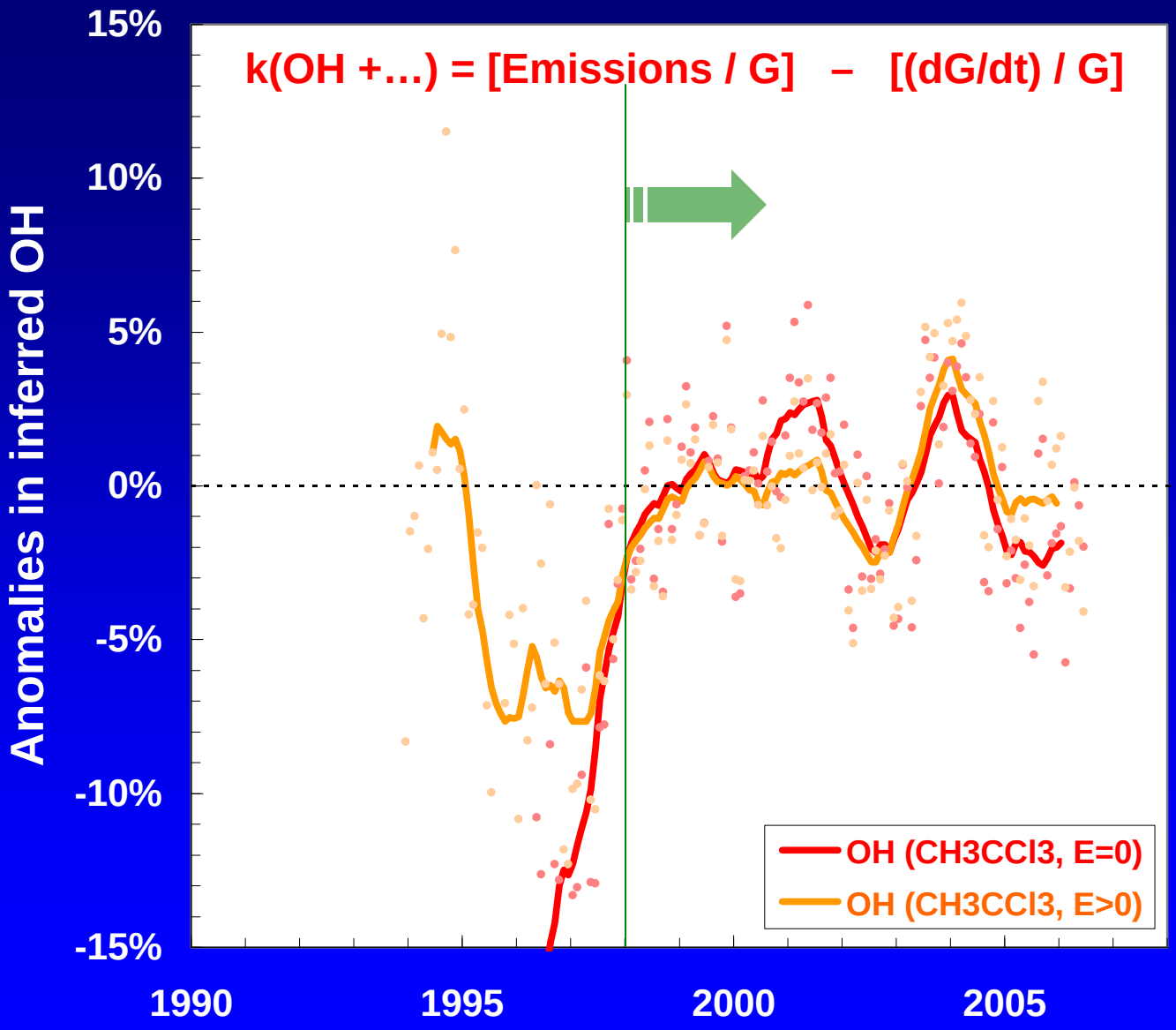


Solid lines:
AGAGE obs.

Dashed lines:
NOAA obs.

Emissions:
Prinn *et al*. (2005) via industry

Inferring OH from CH_3CCl_3 with and without Emissions:



During 1998-2006 (8-yr period):

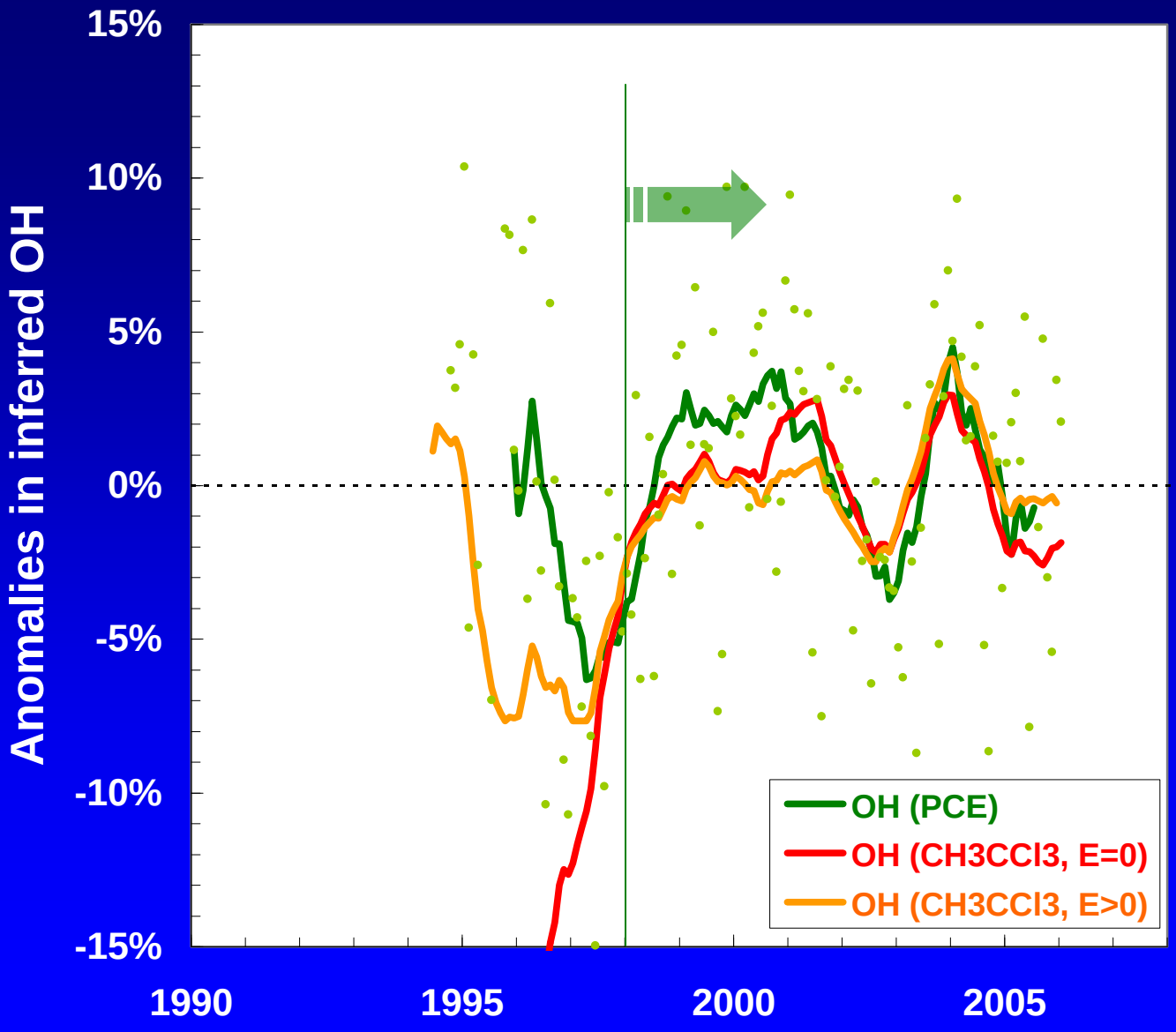
Mean inter-annual variability implied for OH = 1.8 - 2.3%

Conclusion insensitive to emission magnitude and data source

NOAA flask data

R. Prinn 'base' emissions through 2004 used in E>0 results

Inferring OH from C_2Cl_4 (PCE) changes (smoothly varying E)…

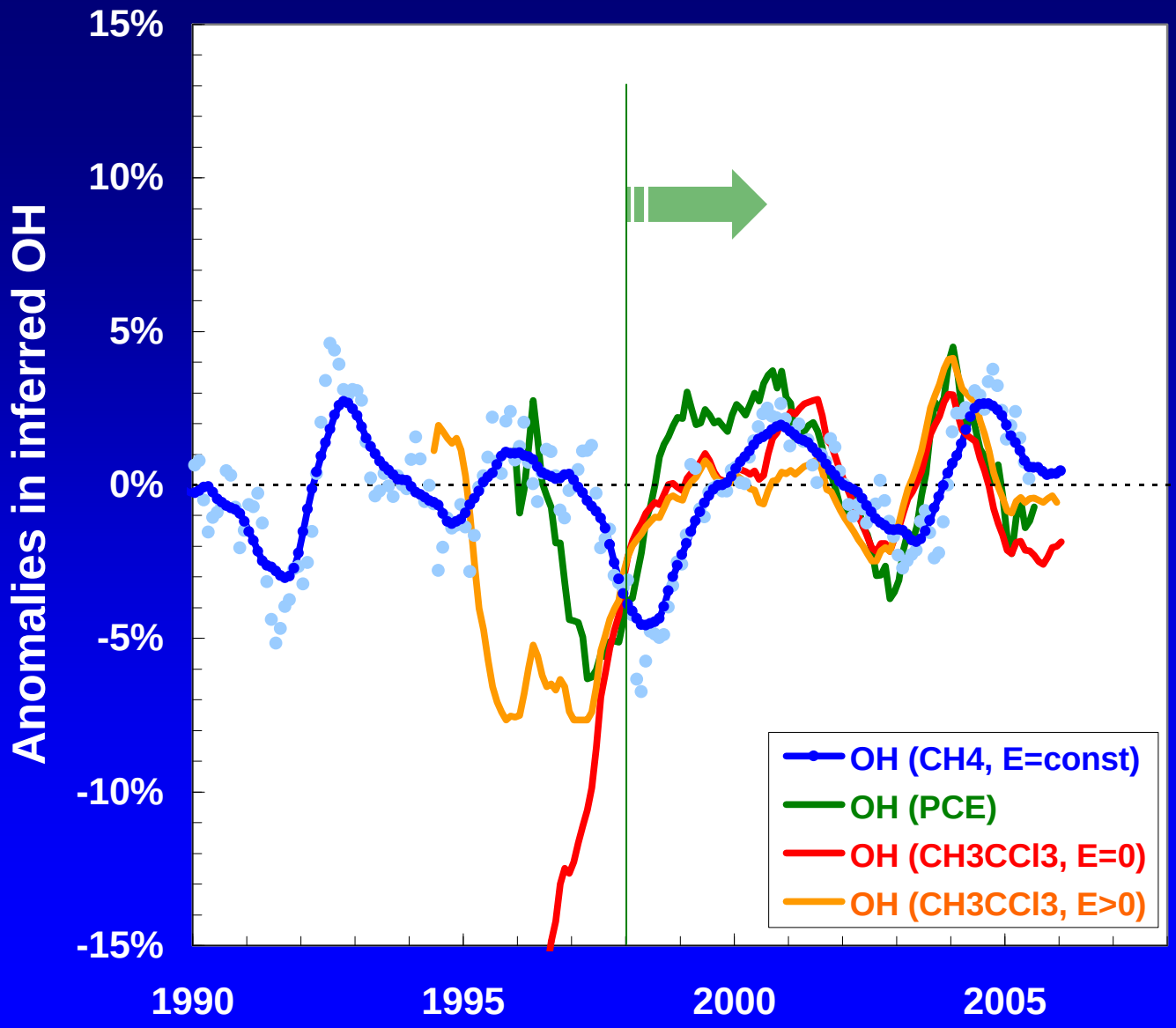


C_2Cl_4:
Much shorter lifetime than CH_3CCl_3,

Emissions variability not well characterized…

Similar inter-annual variability implied for OH

OH inferred from CH_3CCl_3, C_2Cl_4, and CH_4



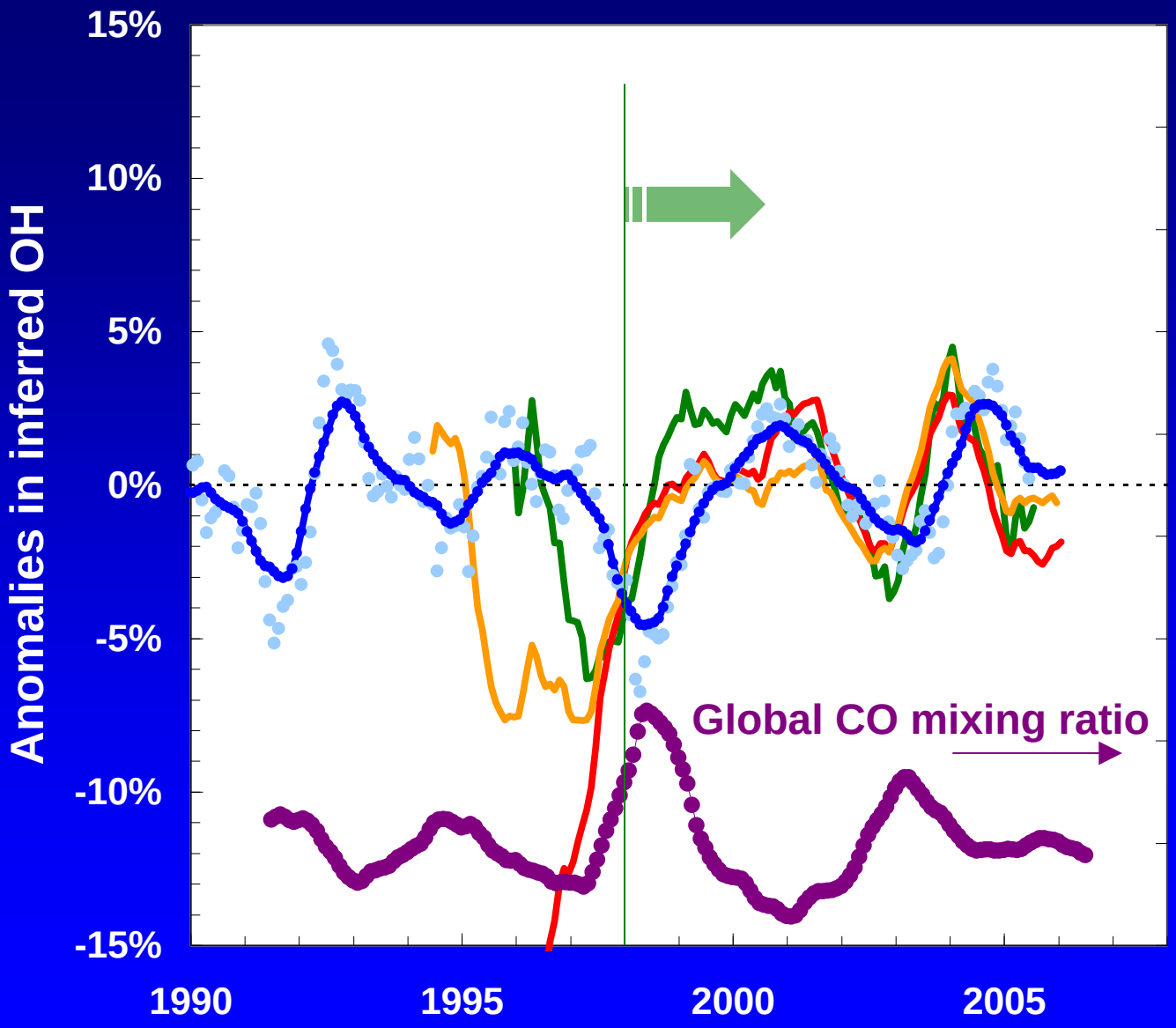
Since 1998:

Much more consistent picture of OH variability from CH_4, CH_3CCl_3, and C_2Cl_4…

OH inter-annual variability of ≤ 2%

NOAA flask data

What could have affected OH during 1998-2006?

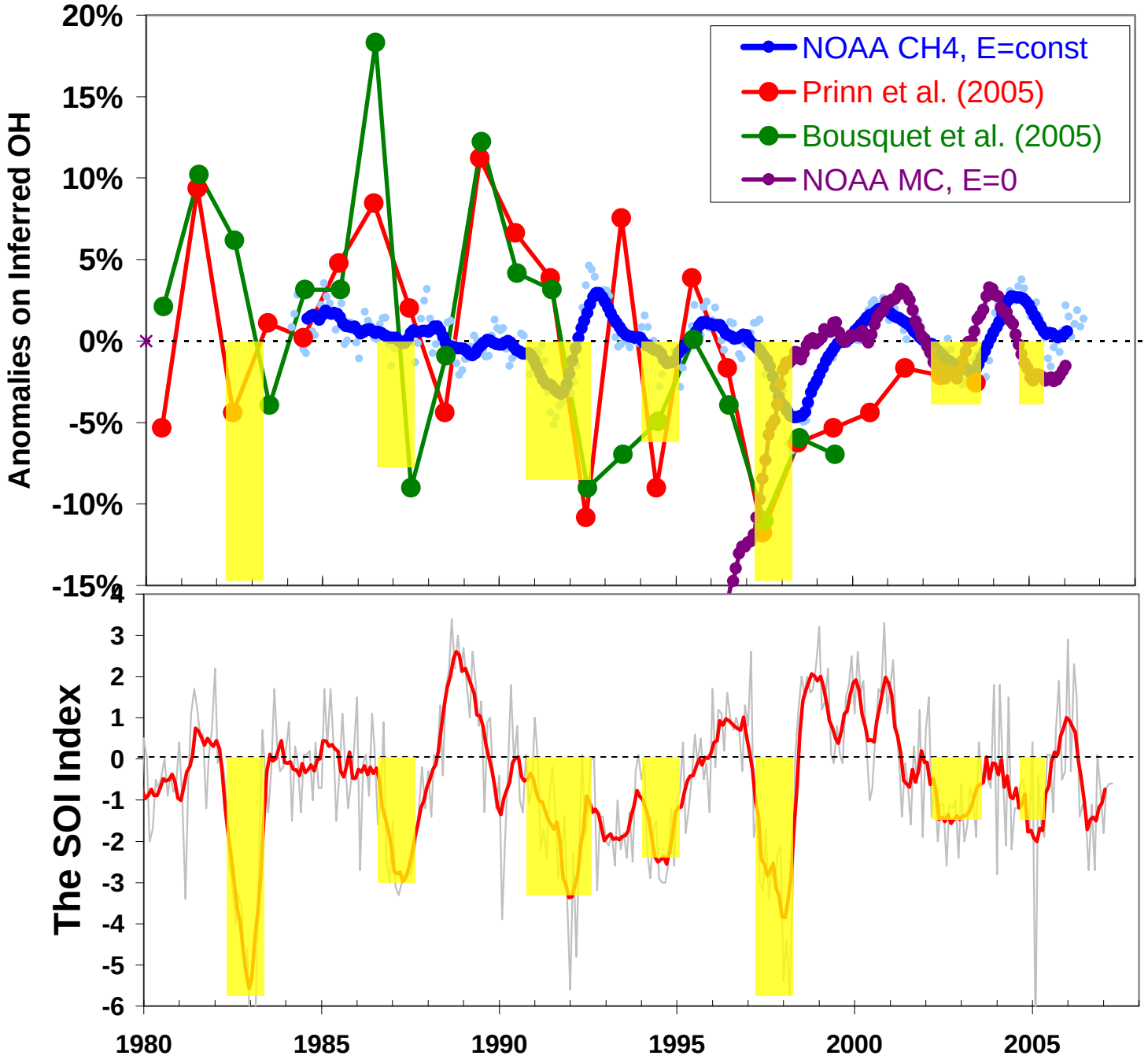


CO is a main sink for OH

Scale is 1:3, OH vs CO—based upon model expectations

Carbon Monoxide mixing ratio anomalies

CO data from P. Novelli



On the influence of El Niños

Is there a measurable climate influence on global atmospheric chemistry?

Is it realized in OH via its influence on trace gases, clouds, or… ?

Conclusions:

Research-quality 'monitoring' data can provide substantial added value: Insights into pressing questions concerning atmospheric chemistry and climate are possible.

Regarding 'global' OH variability:

In contrast to previous years, since 1998 CH_3CCl_3, C_2Cl_4 and CH_4 imply that ***variability in 'global' OH is ≤ 2% inter-annually***.

We argue that ***since 1998 CH_3CCl_3 has been a more precise tool*** for sensing OH inter-annual variations.

These results imply that ***OH is highly buffered*** against large inter-annual changes.

Interesting correlations were observed between inferred OH variations, CO mixing ratios, and the SOI (El Niño).

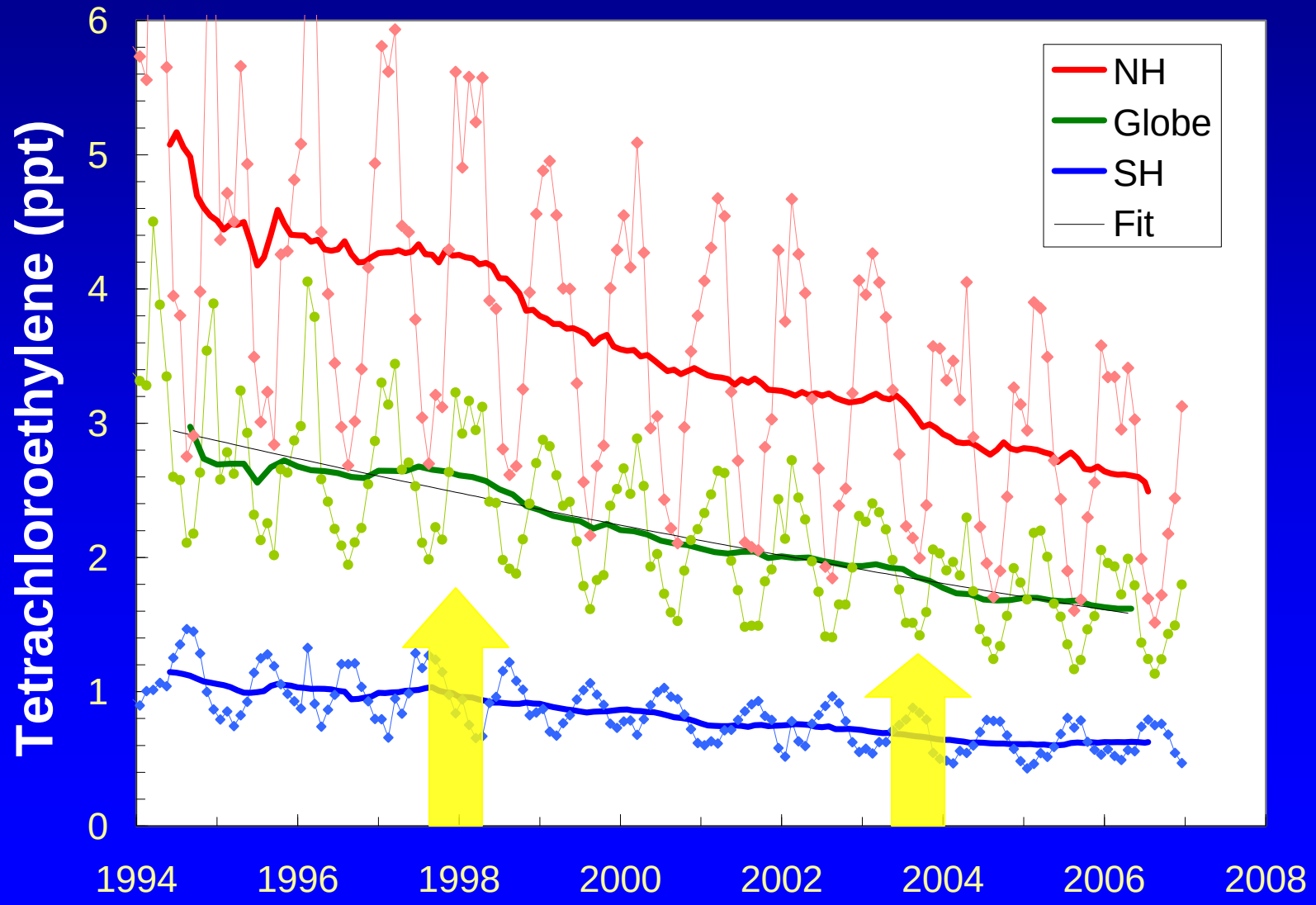
Can one infer OH variability from C_2Cl_4?

Predominantly OH loss

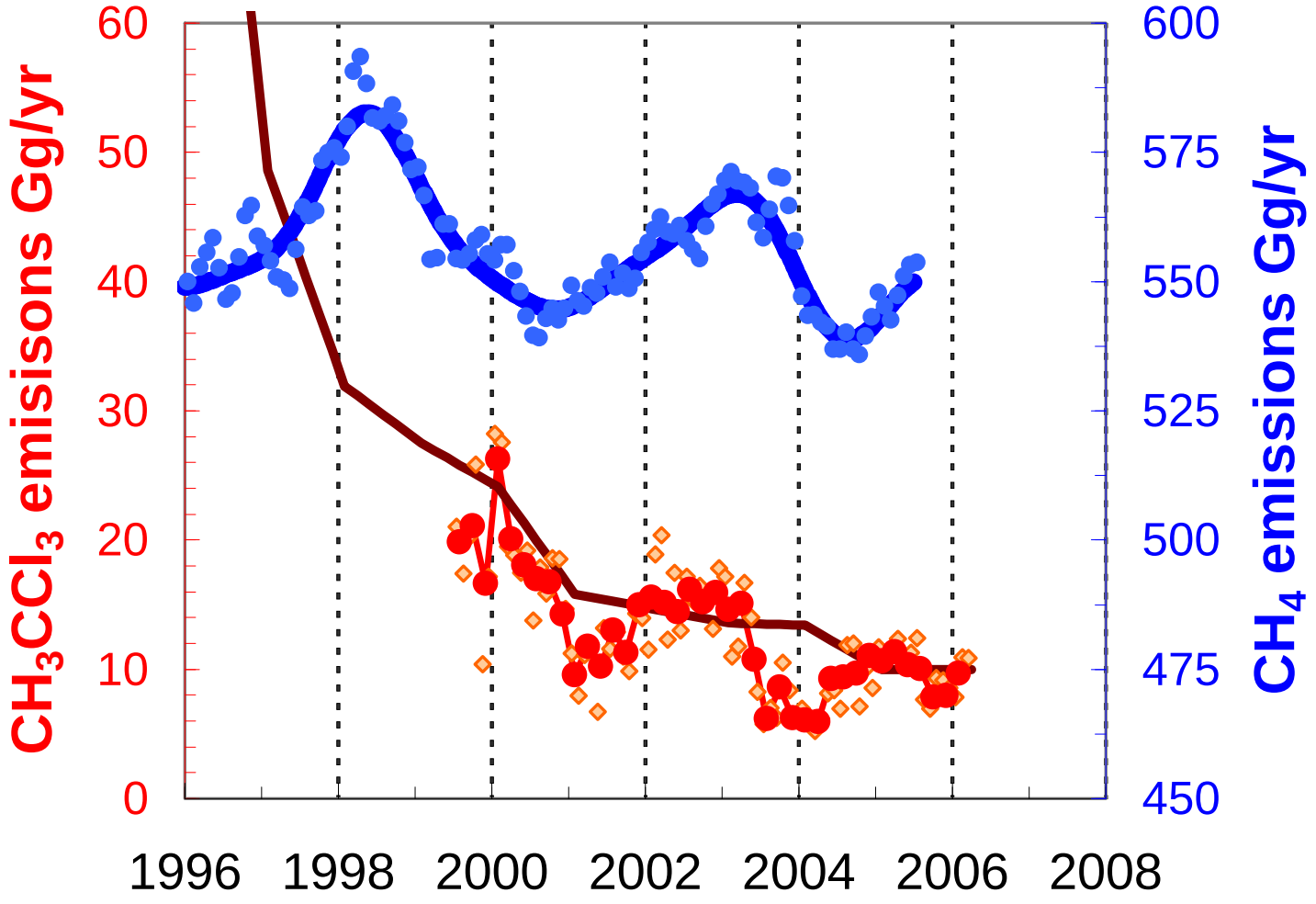
Anthropogenic source is not well characterized

No biomass burning source

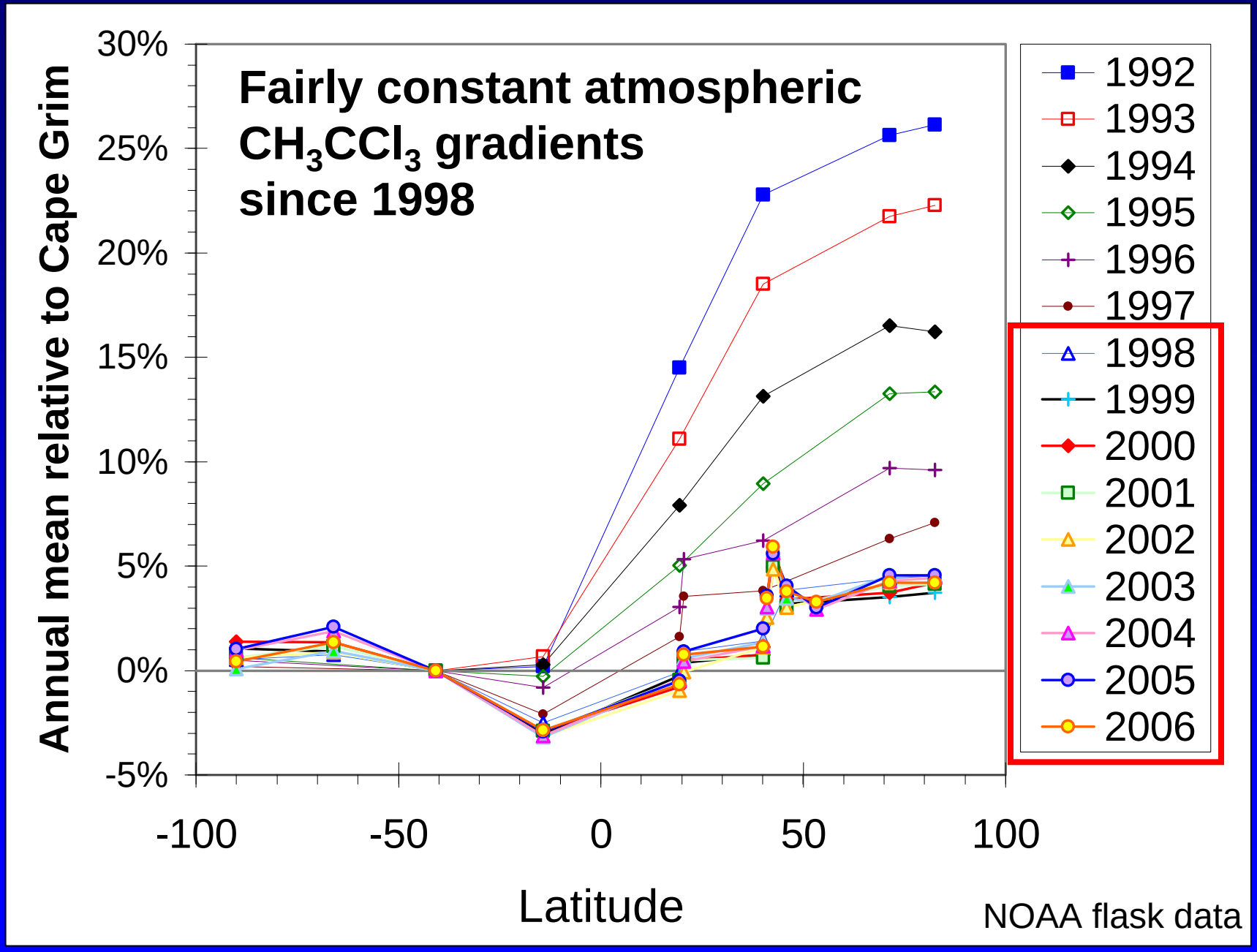
(OH calculated with emissions derived from poly fit to global mean…)



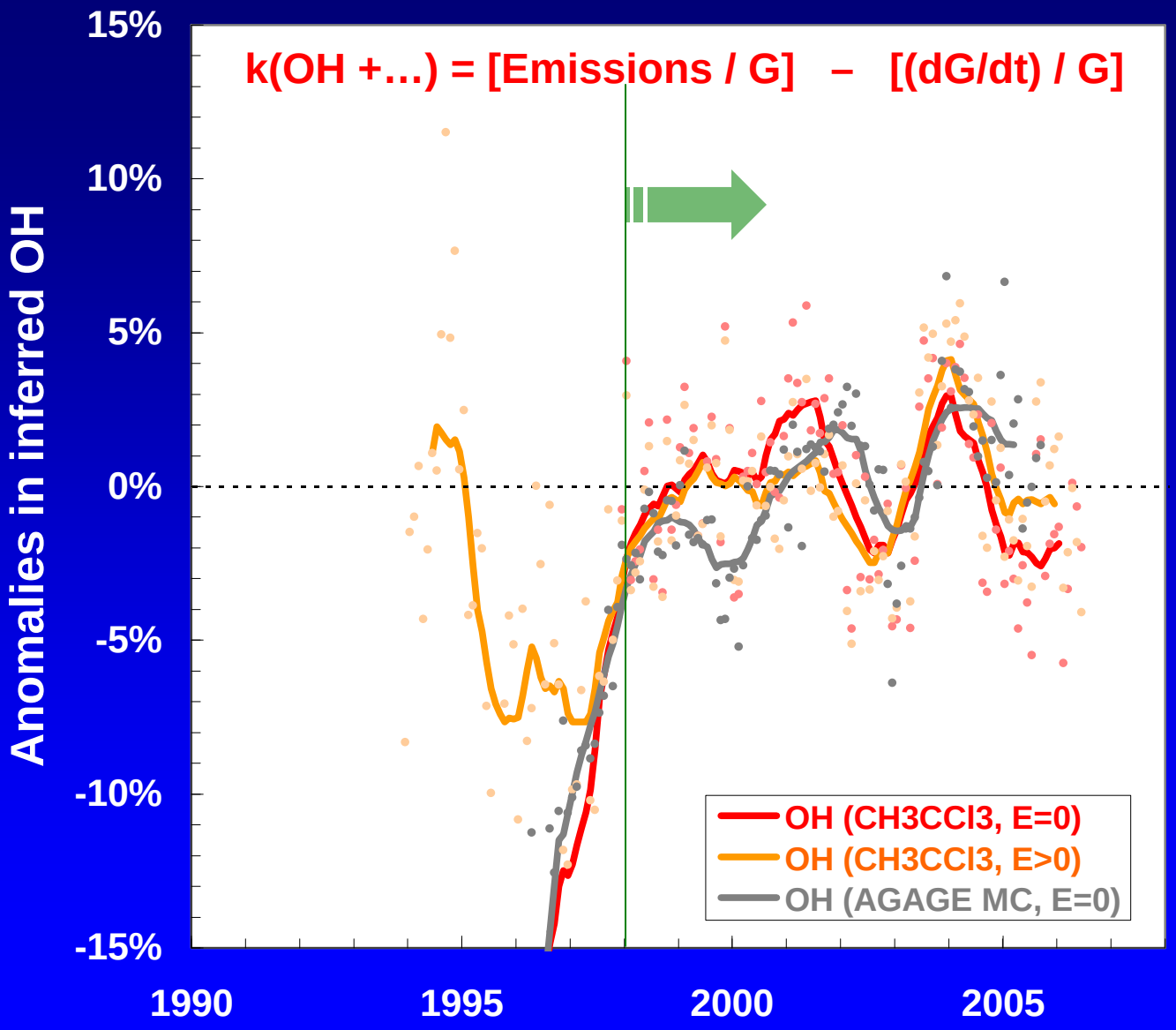
Variability implied for emissions if zero variability in losses (OH):



Industrial
MC emissions in brown


Fairly constant atmospheric
CH_3CCl_3 gradients
since 1998
Annual mean relative to Cape Grim
30%
25%
20%
15%
10%
5%
0%
-5%
-100
-50
0
50
100
Latitude
1992
1993
1994
1995
1996
1997
1998
1999
2000
2001
2002
2003
2004
2005
2006
NOAA flask data

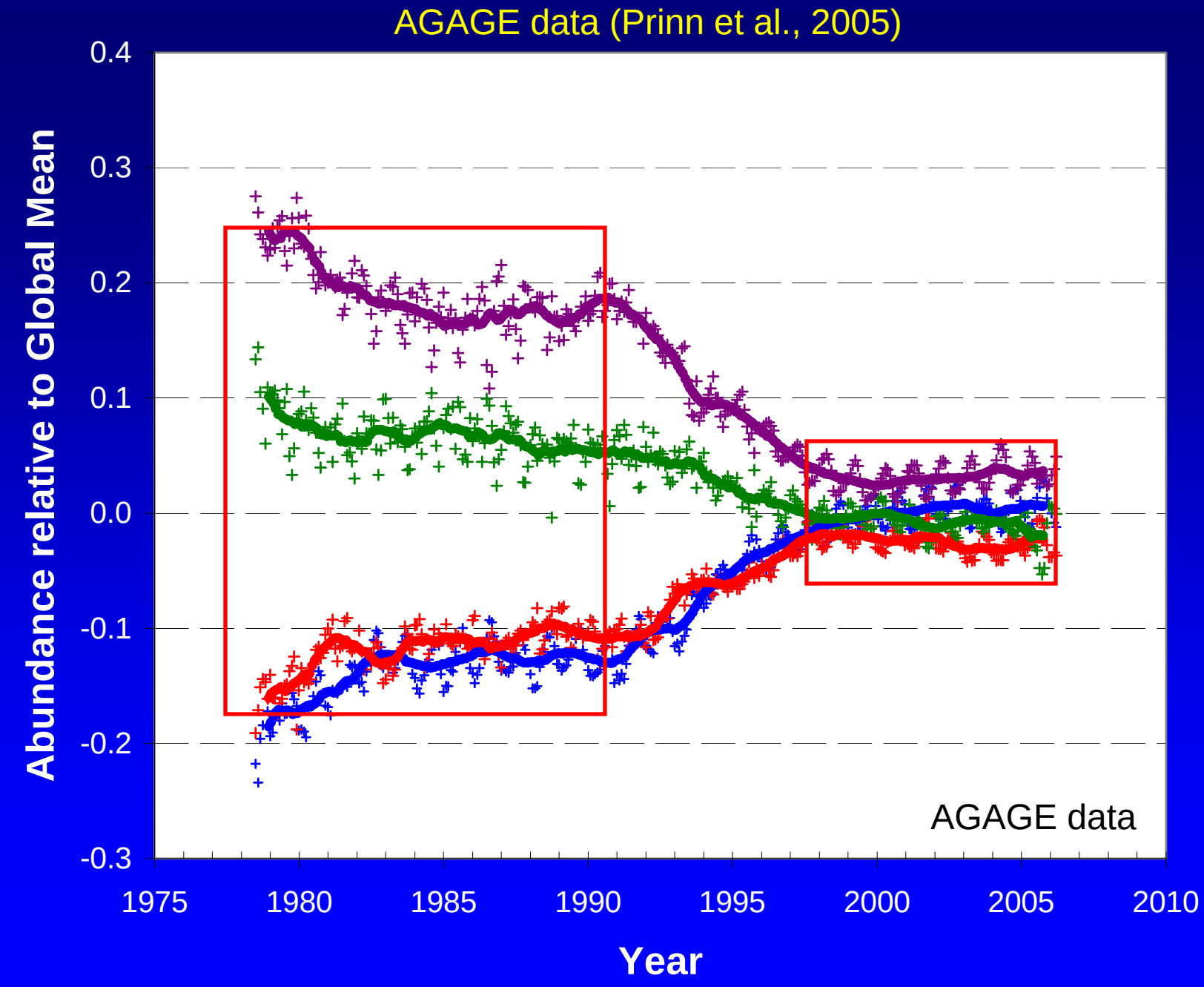
Inferring OH from CH_3CCl_3 with and without Emissions:



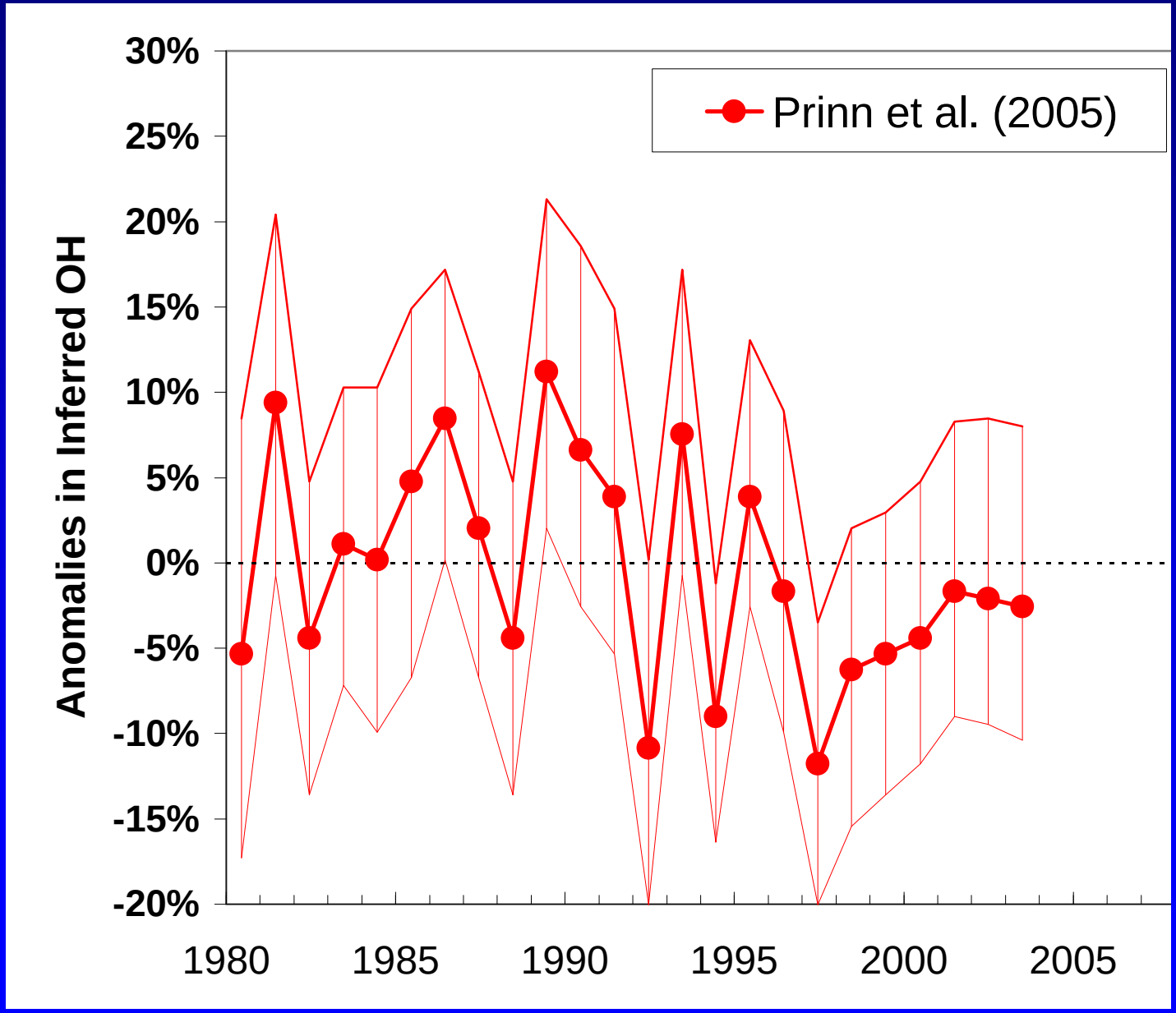
Similar conclusion can be drawn from AGAGE CH_3CCl_3 data since 1998

R. Prinn 'base' emissions through 2004 used in E>0 results

Pre-1990 atmospheric gradients are much larger than post 1997…



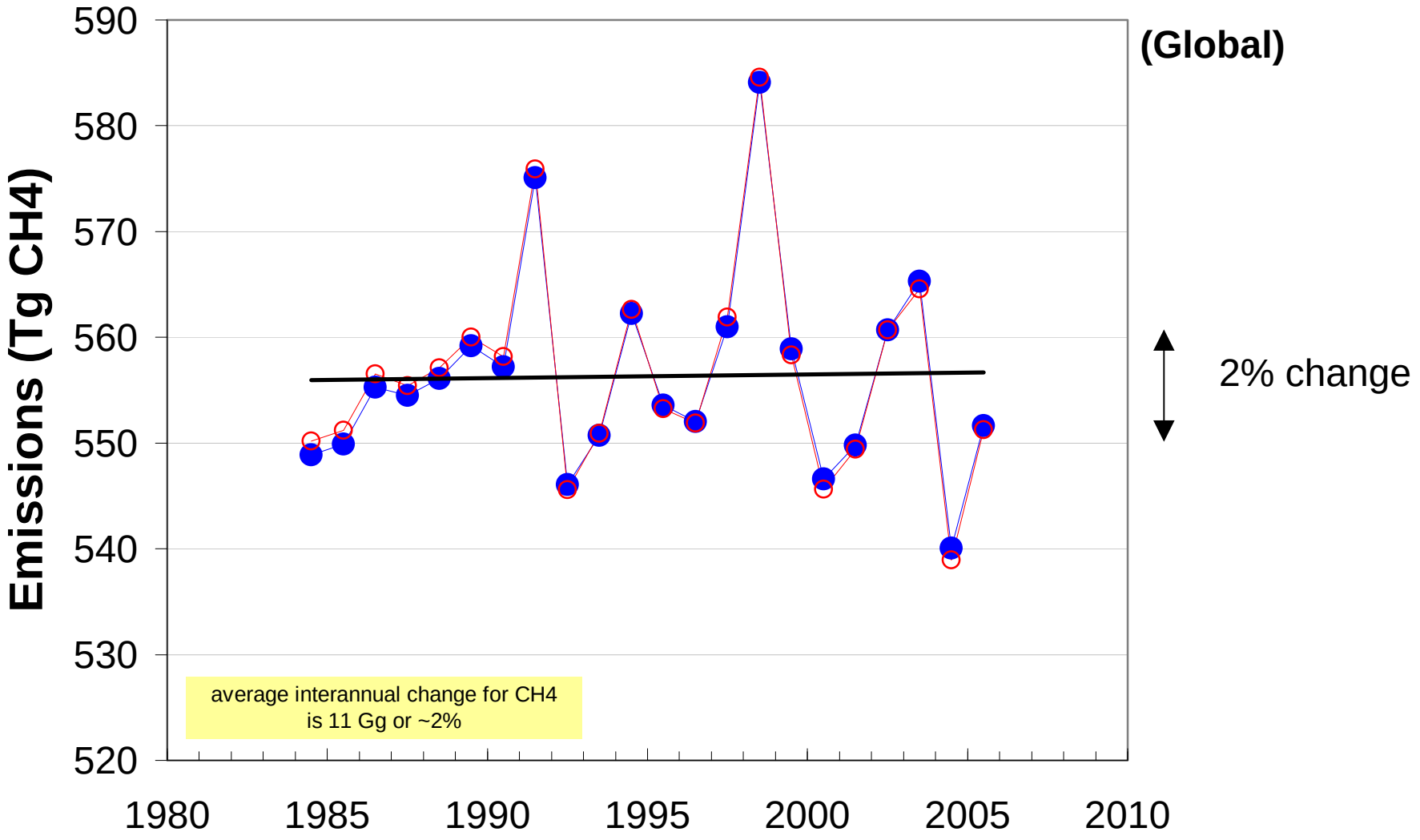
a) OH inferred from AGAGE CH_3CCl_3 and industrial emissions…



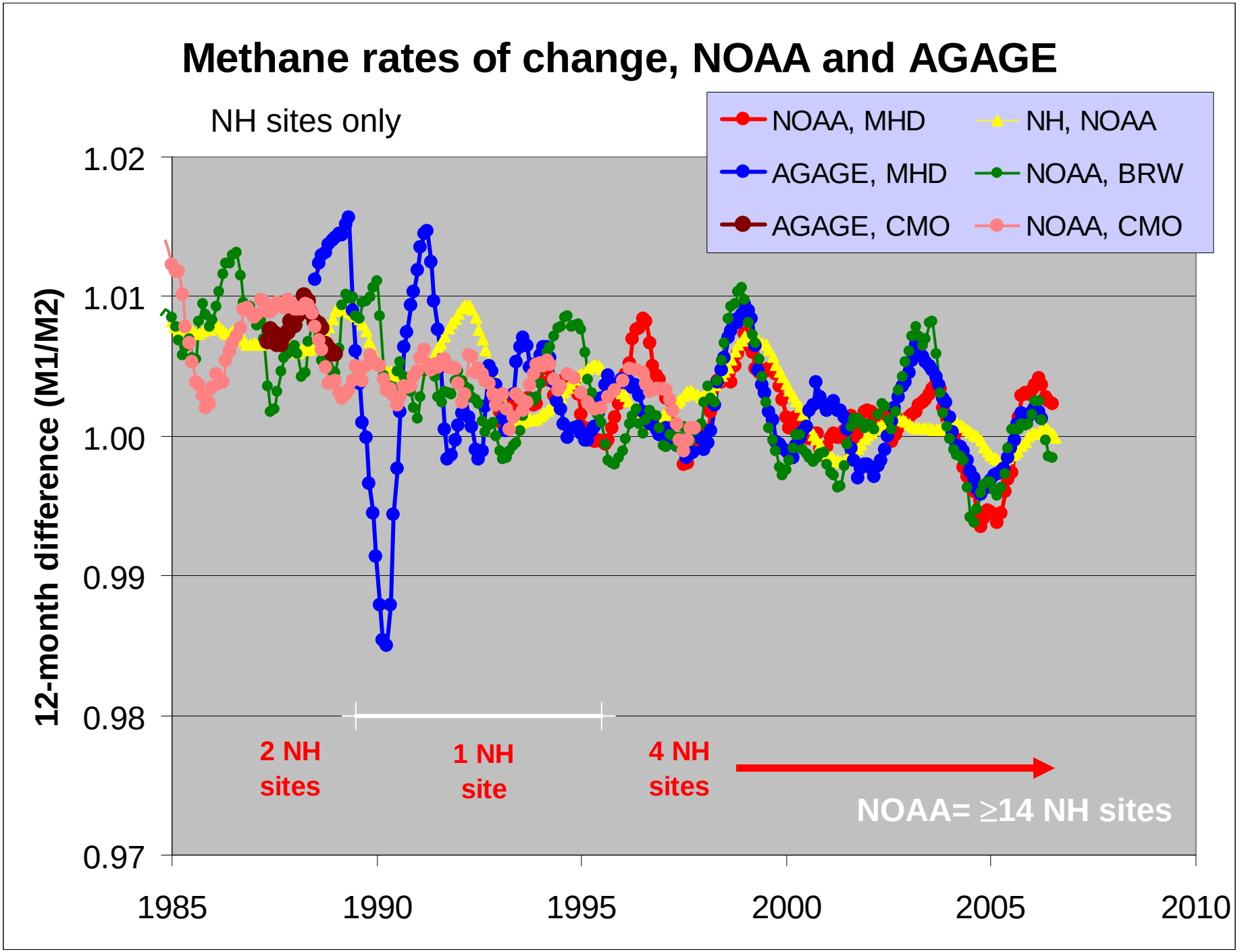
AGAGE multi-box model calculation

OH Inter-annual variability = 7 (±6)%

Methane emissions assuming constant loss



From E. Dlugokencky


Methane rates of change, NOAA and AGAGE
NH sites only
NOAA, MHD
NH, NOAA
AGAGE, MHD
NOAA, BRW
AGAGE, CMO
NOAA, CMO
12-month difference (M1/M2)
1.02
1.01
1.00
0.99
0.98
0.97
1985
1990
1995
2000
2005
2010
2 NH sites
1 NH site
4 NH sites
NOAA= ≥14 NH sites