Top-Down Estimates of tropospheric OH: Past and Future

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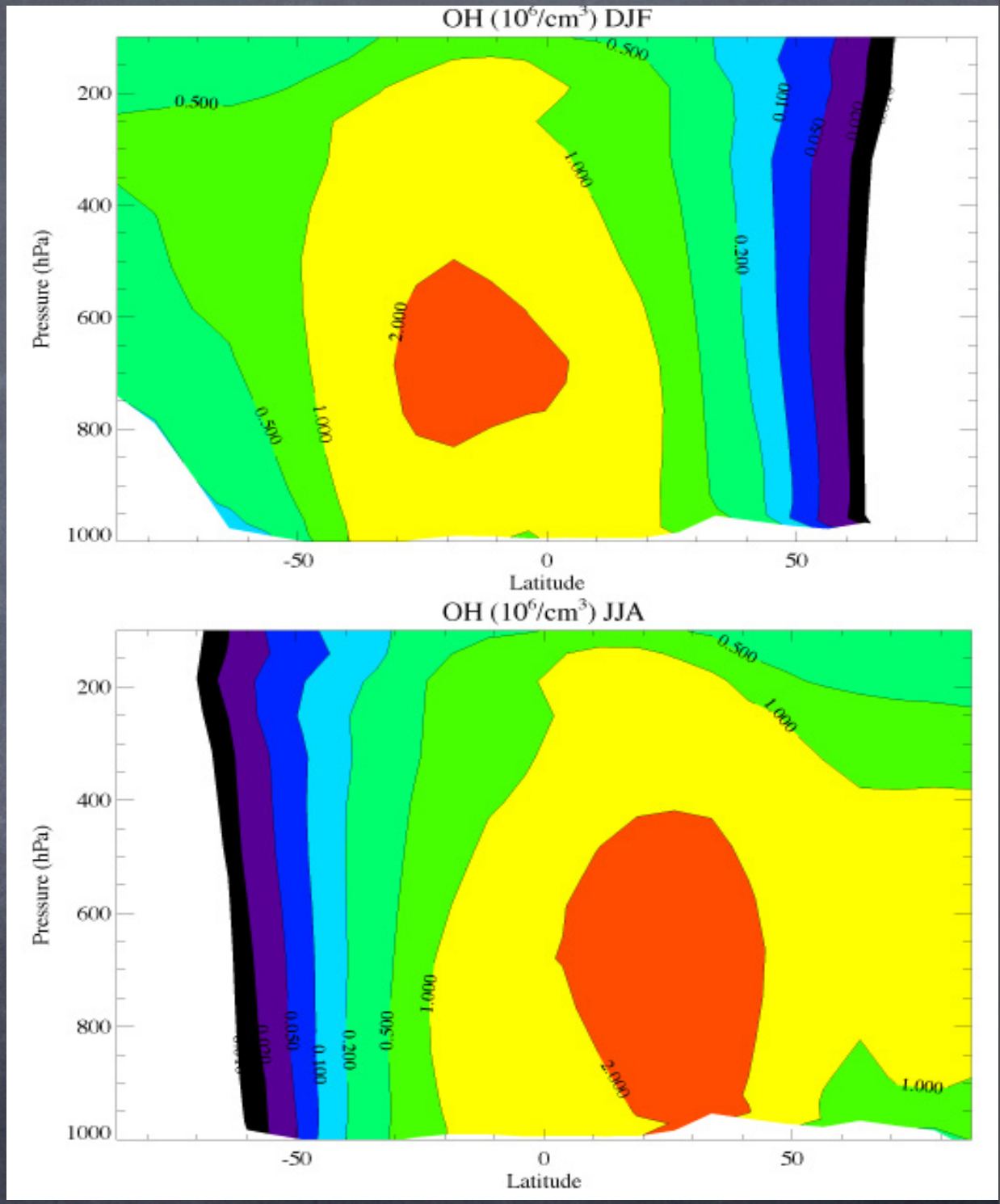
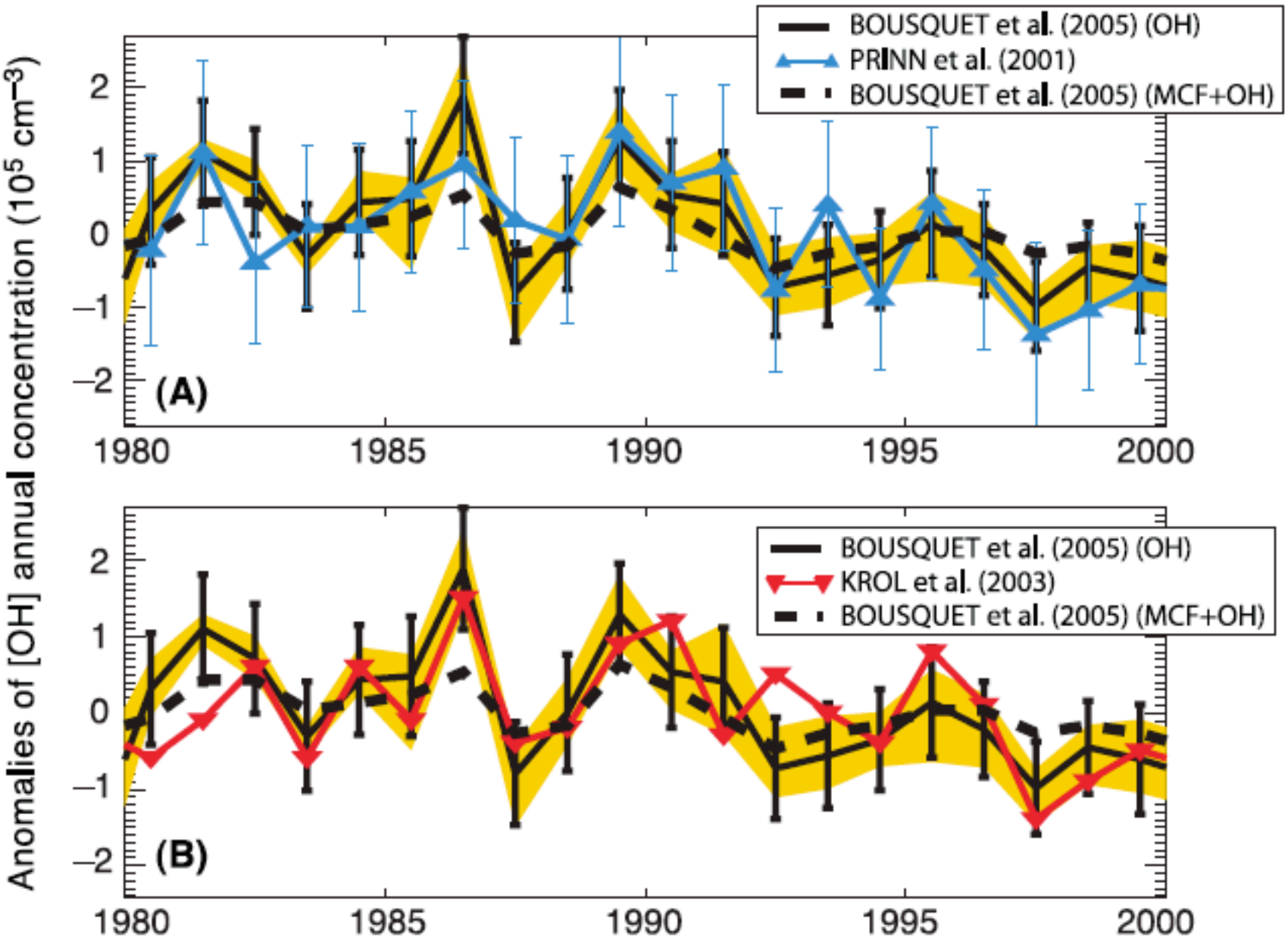
TM-meeting June 2008

OH in the troposphere

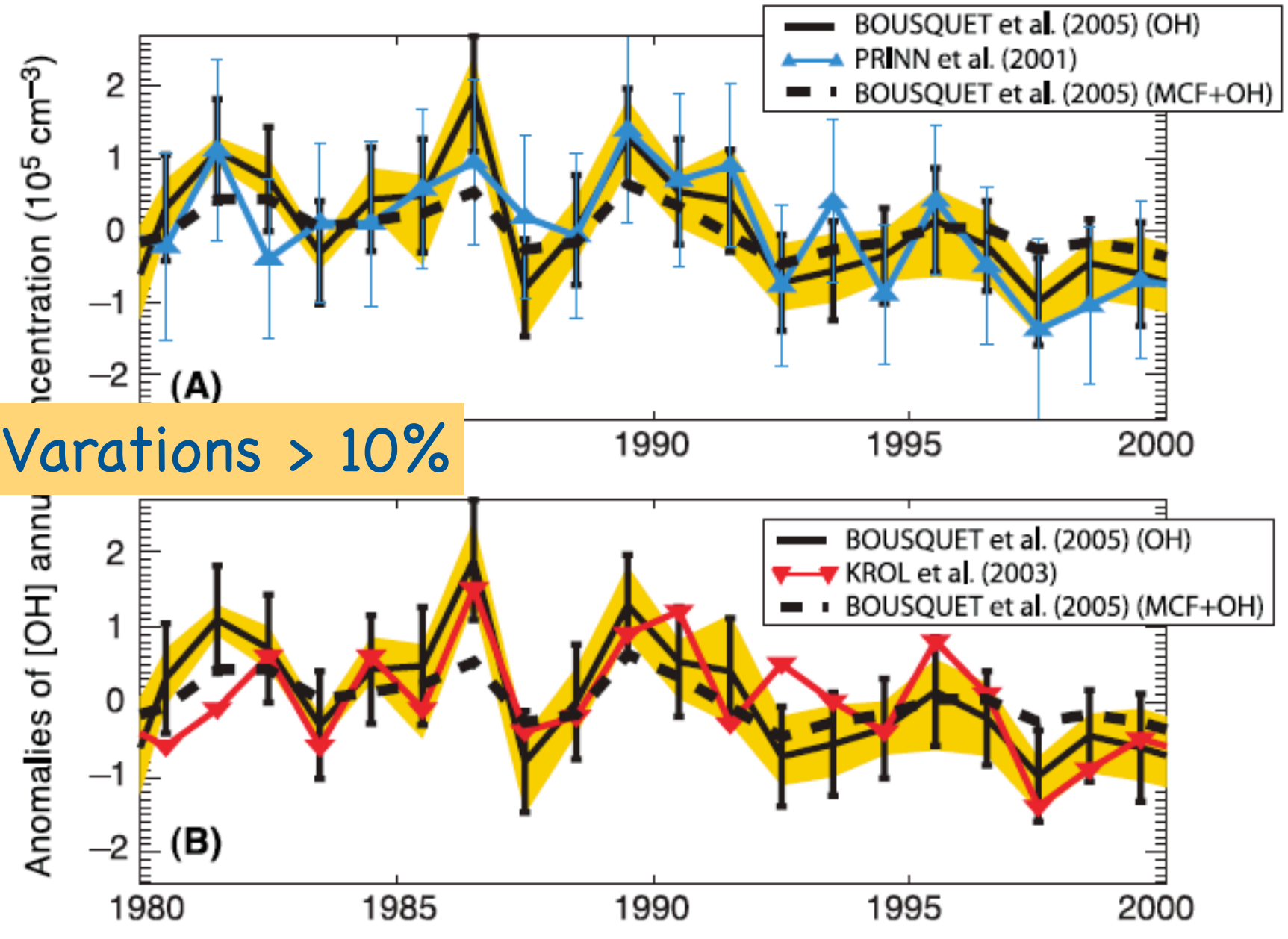
Mean: ≈1x10^6 molecules/cm^3



IPCC (AR4), Chapter 2



IPCC (AR4), Chapter 2



OH Varations > 10%

Interpretation …

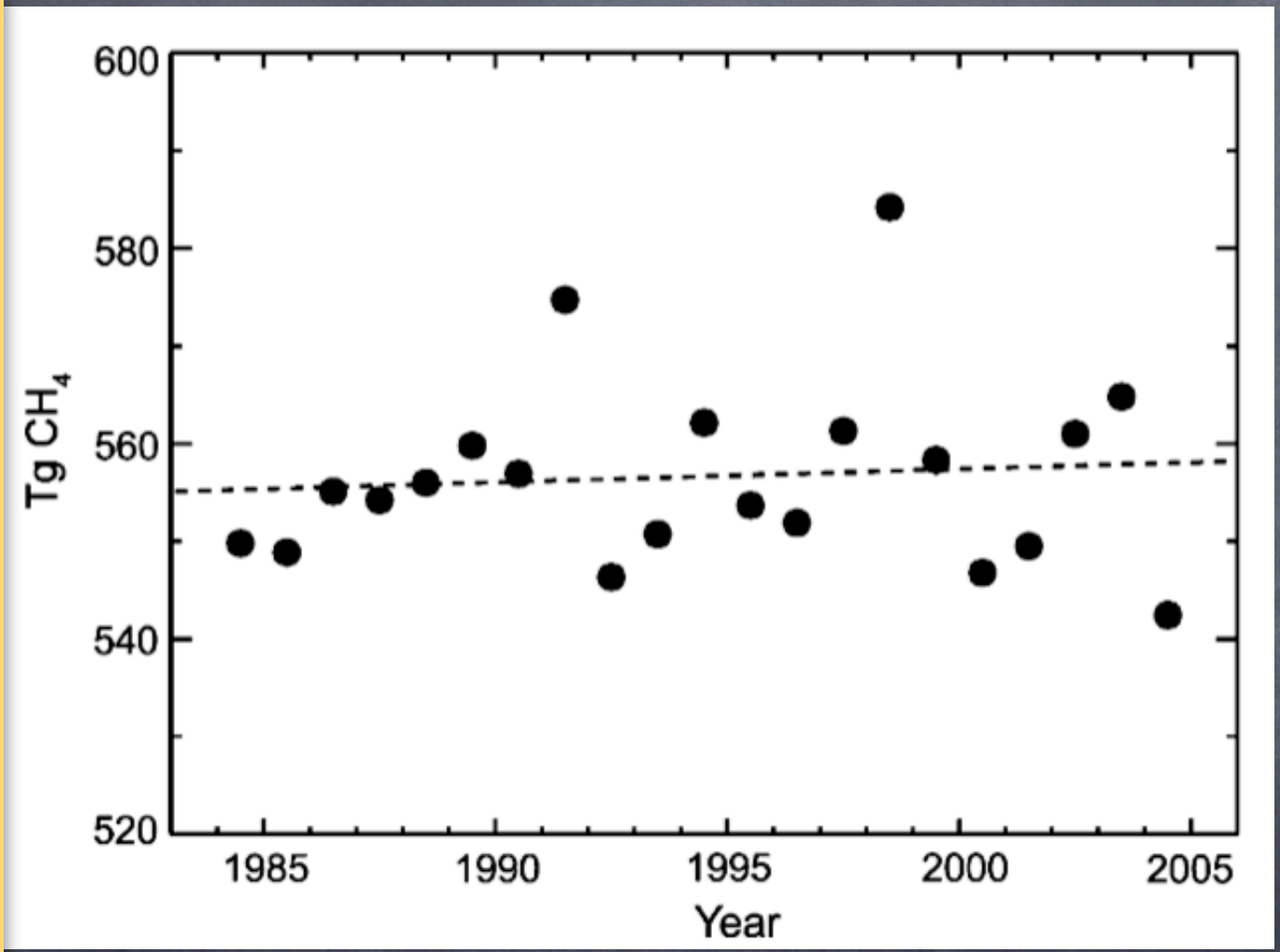
2.8). This implies that these interannual OH variations are real, but only their phasing and not their amplitude, is well defined. Bousquet et al. (2005) also deduced that OH in the SH shows a zero to small negative trend, in qualitative agreement with Prinn et al. (2001). Short-term variations in OH were also recently deduced by Manning et al. (2005) using 13 years of ^{14}CO measurements in New Zealand and Antarctica. They found no

Conclusion mostly based on methyl chloroform (MCF)

MCF is phased out, emissions and hence the atmospheric burden are declining since 1992

Are these OH variations real?

Yearly CH_4 Source Required to Explain the Observed Global Burden (OH = C)

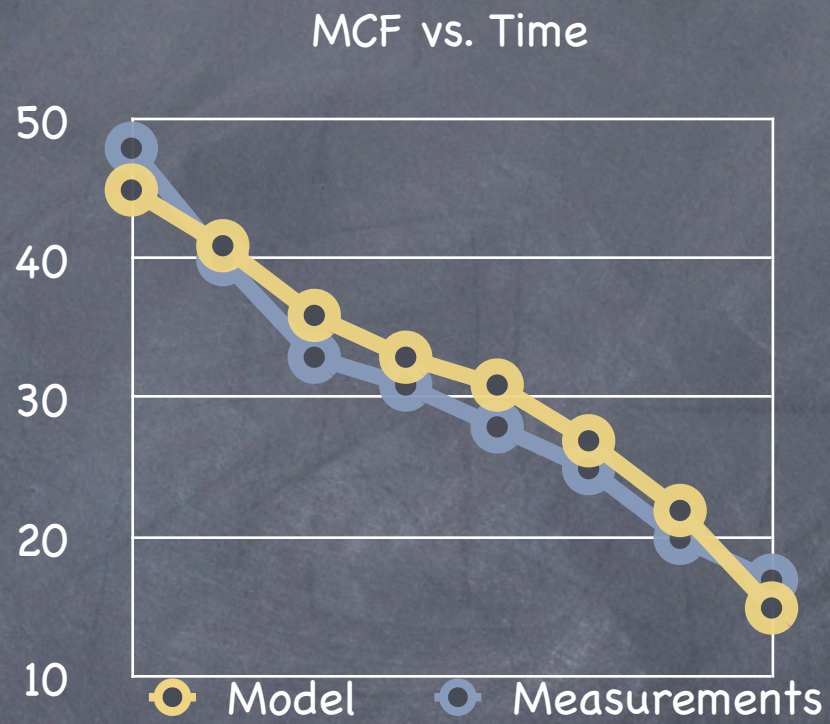


Lifetime CH_4 (OH oxidation) 10 years

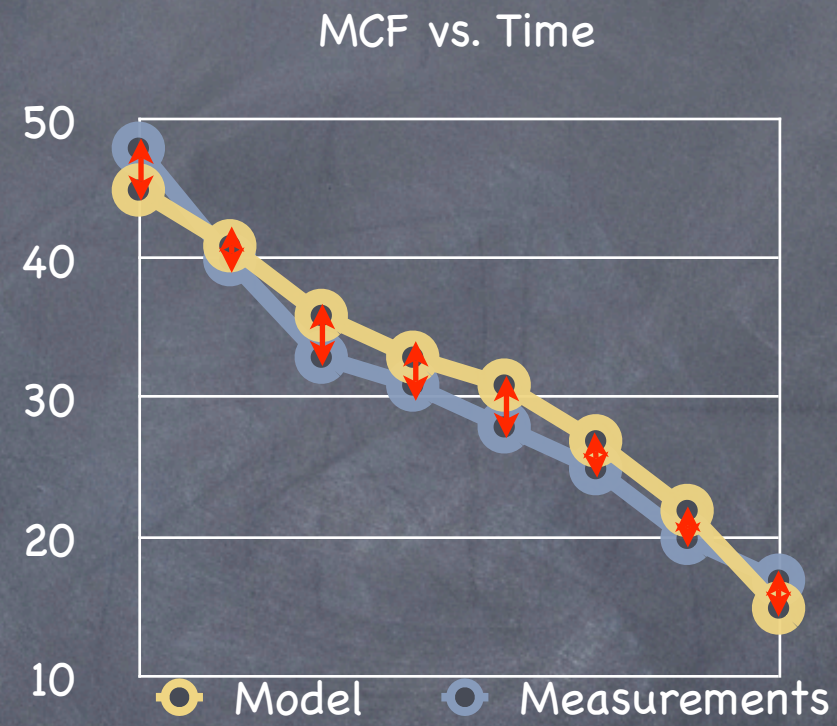
10% OH variation: 50 Tg CH_4!

Lelieveld et al., 2006

Inverse Modelling: General Approach

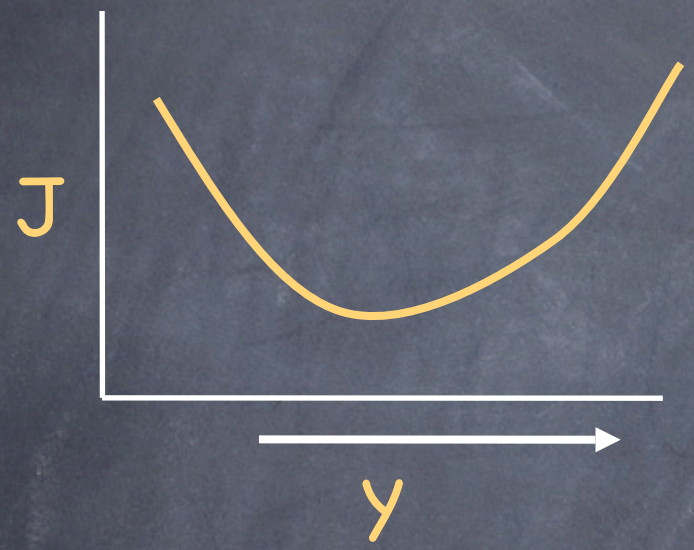


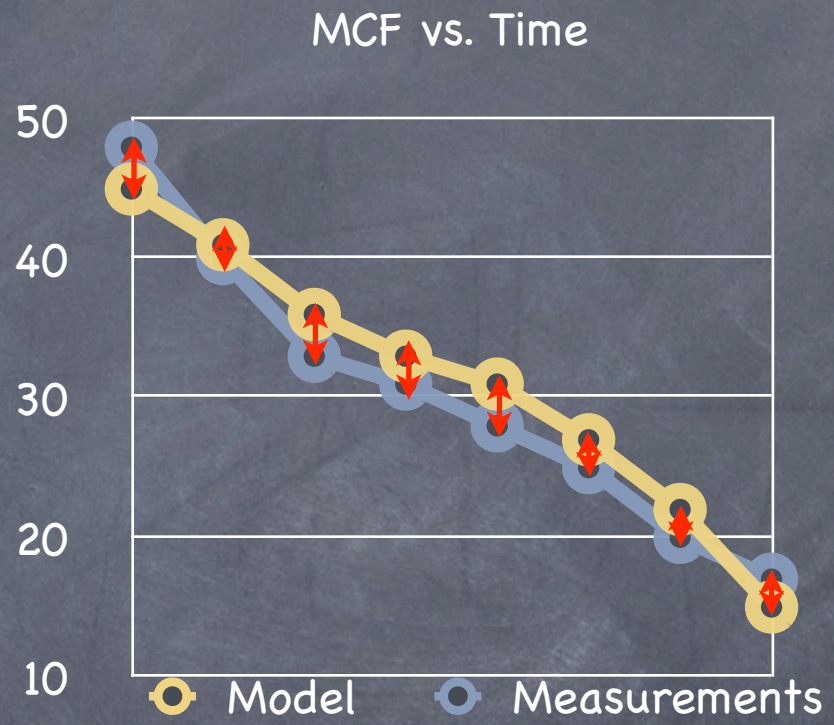
Inverse Modelling: General Approach



$$J(y) = \Sigma_n \, (\text{model}(y) \leftrightarrow \text{measurement})^2/\sigma^2$$

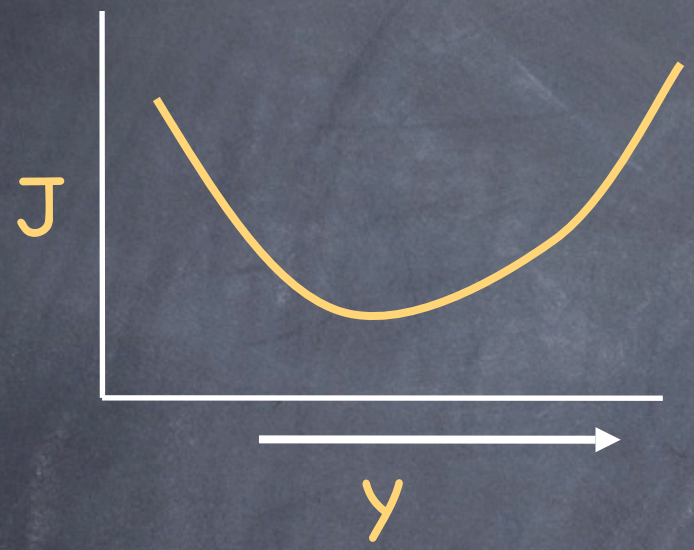
Inverse Modelling: General Approach

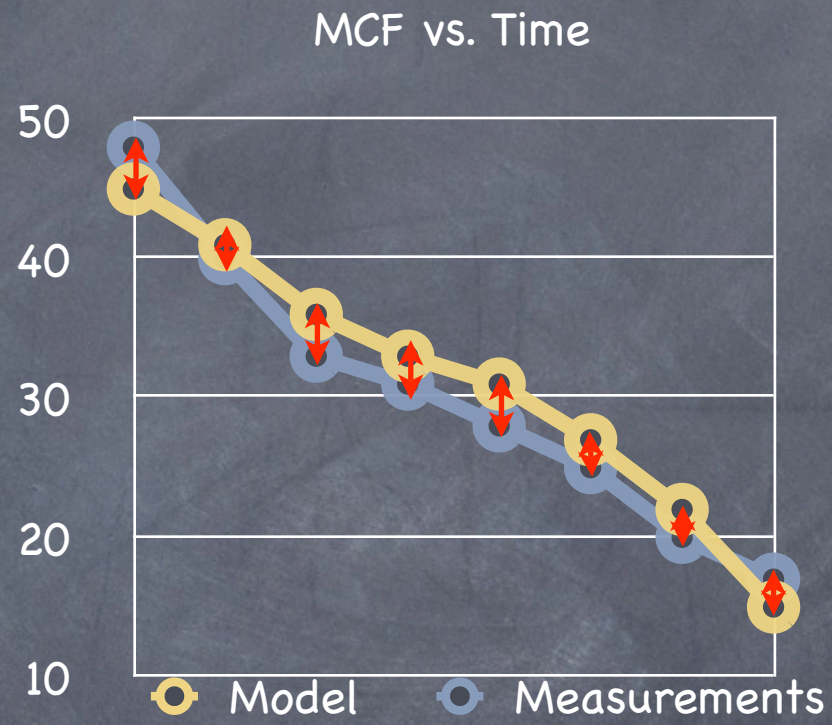




$$J(y) = \Sigma_n \text{ (model}(y) \leftrightarrow \text{measurement)}^2/\sigma^2$$

Inverse Modelling: General Approach





$J(y) = \Sigma_n (\text{model}(y) \leftrightarrow \text{measurement})^2/\sigma^2$

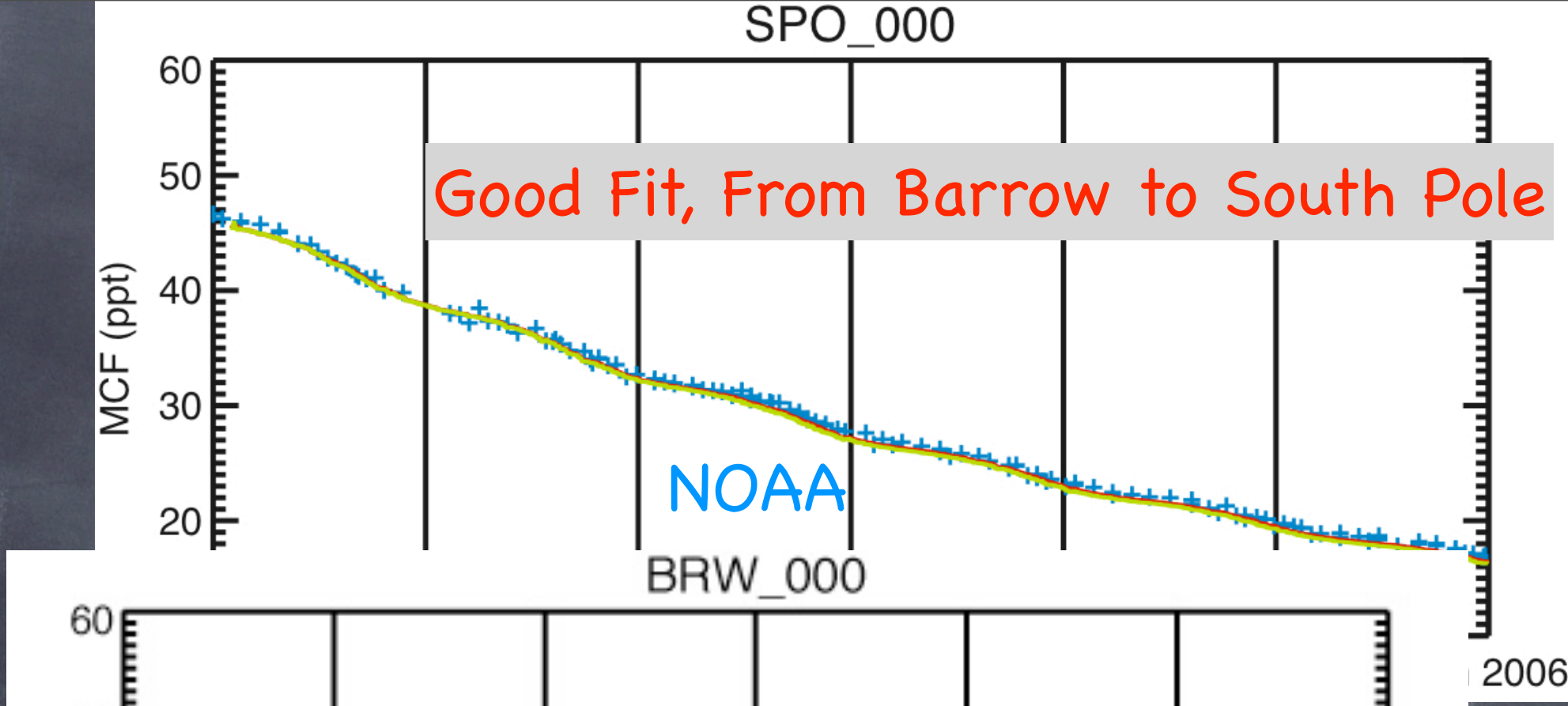
y = OH, MCF emissions, ..

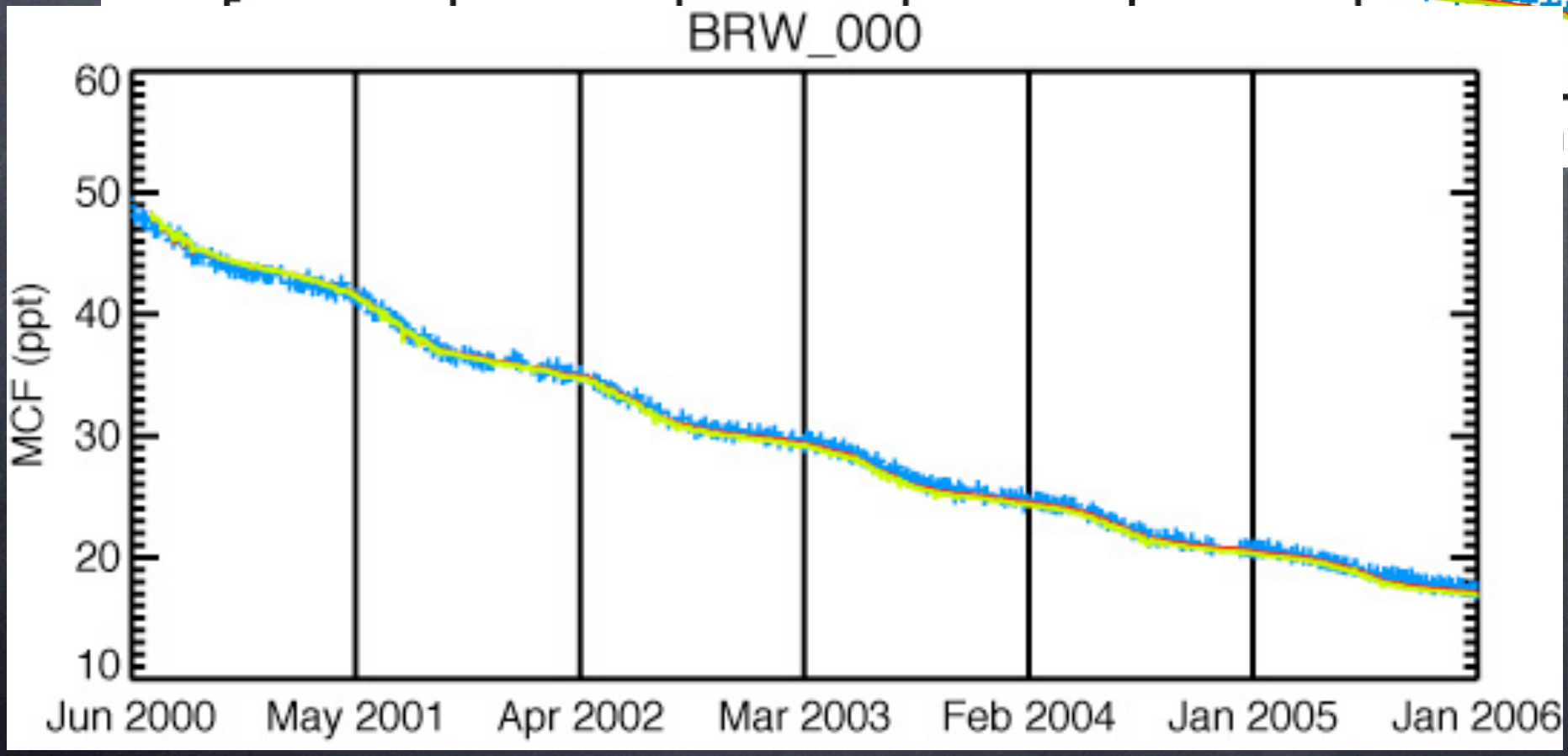
Inversion Details

- Inversion often data-limited (not enough measurements to fully constrain the unknowns y)
- Prior assumptions about OH and emissions are required
- Inversion results MCF depend strongly on prior assumptions about emission uncertainty and OH uncertainty

What about the post 2000 period MCF?

- With Vanishing MCF Emissions good estimates for OH are expected (e.g. Spivakovsky et al. 2000)
- Step 1: Forward Model Simulations


SPO_000
60
50
40
30
20
MCF (ppt)
Good Fit, From Barrow to South Pole
NOAA
2006


BRW_000
60
50
40
30
20
10
MCF (ppt)
Jun 2000
May 2001
Apr 2002
Mar 2003
Feb 2004
Jan 2005
Jan 2006

Step 2: develop suitable inverse modelling framework to derive the 'optimal' OH (and Emissions) (e.g. Bousquet et al., 2005)

Here: 4D-VAR data-assimilation

TM5-4DVAR

EUROPEAN COMMISSION
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Joint Research Centre

Institute for Environment and Sustainability

Joint Research Centre

model parameters

observations

$$J(\vec{x}) = \frac{1}{2}(\vec{x} - \vec{x}_b)^T \mathbf{B}^{-1}(\vec{x} - \vec{x}_b) + \frac{1}{2}\sum_{i=1}^{n}\left(\vec{y}_{OBS,i} - \vec{H}_i(\vec{x})\right)^T \mathbf{R}_i^{-1}\left(\vec{y}_{OBS,i} - \vec{H}_i(\vec{x})\right)$$

covariance matrix

covariance matrix

a priori estimate

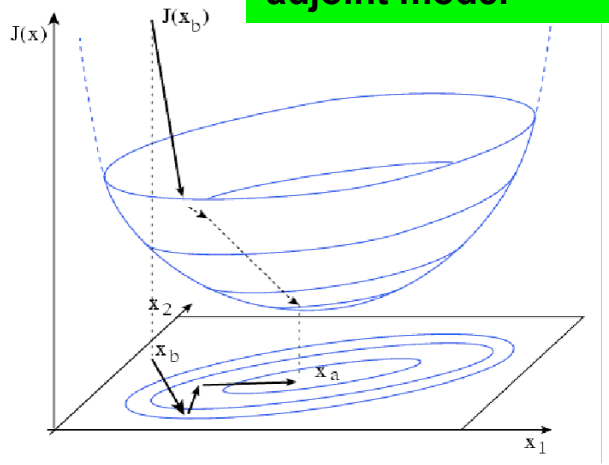
parameters (dimension: n_{PARA})

observations (dimension: n_{OBS})

model simulations corresponding to observations

$$\vec{\nabla} J(\vec{x}) = \mathbf{B}^{-1}(\vec{x} - \vec{x}_B) - \sum_{i=1}^{n} \mathbf{M}_1^T \cdots \mathbf{M}_{i-1}^T \mathbf{M}_i^T \mathbf{H}_i^T \mathbf{R}_i^{-1}\left(\vec{y}_{OBS,i} - \vec{H}_i(\vec{x})\right)$$

adjoint model



atmospheric transport model: TM5

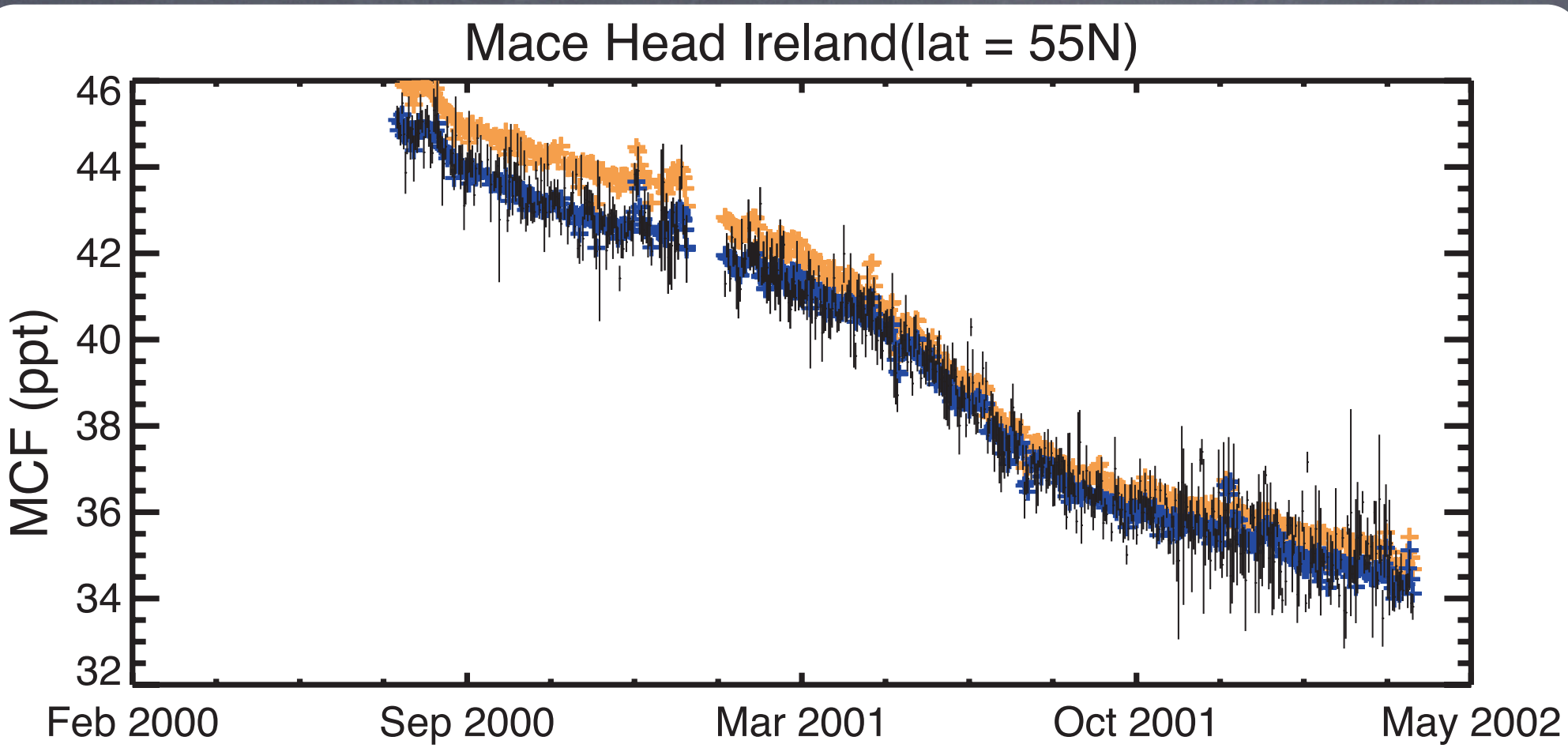
[Krol et al., 2005]

ECMWF meteorology

minimization ECMWF conjugate gradient

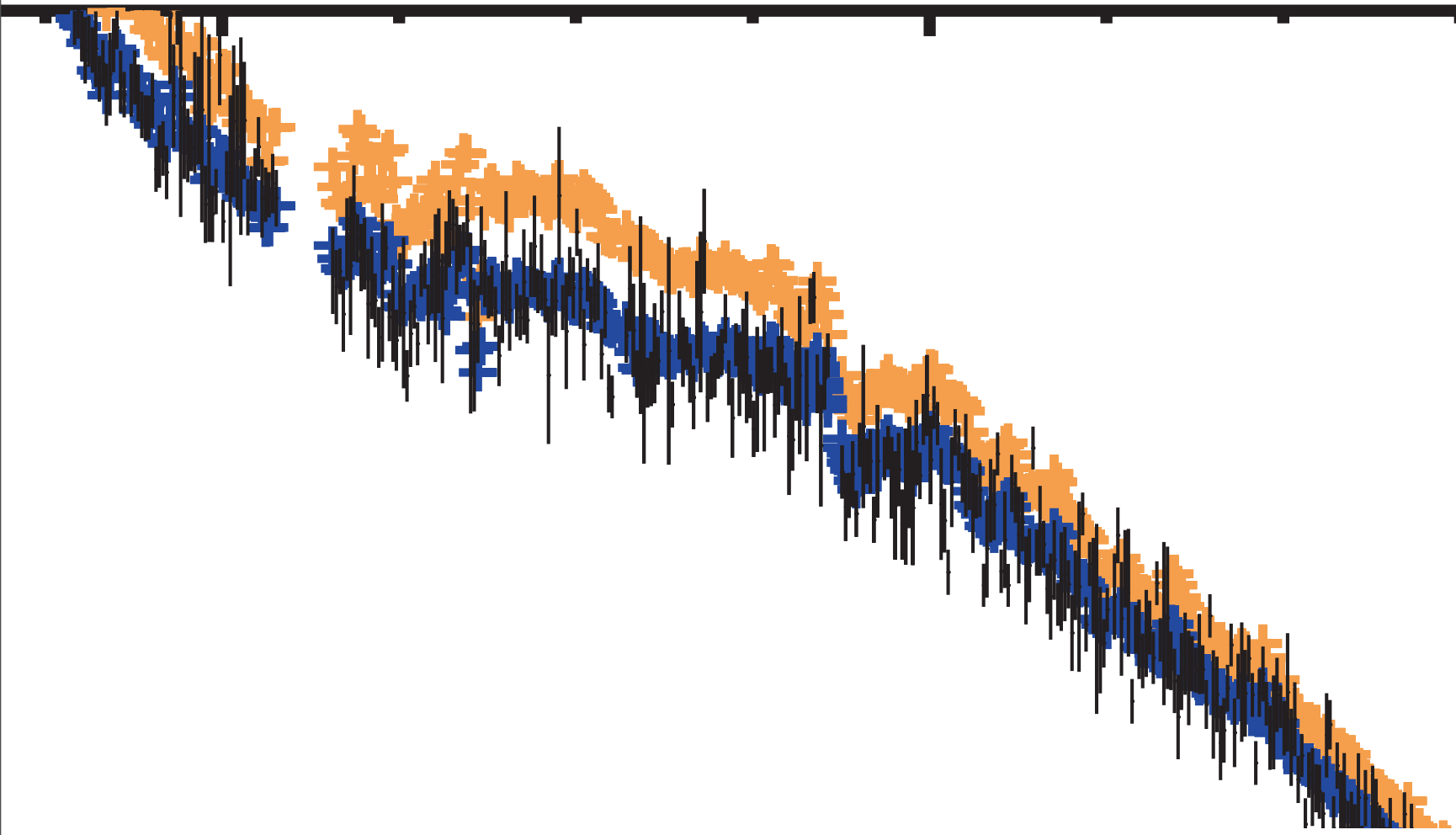
n_{PARA} = ~ 10^{5}-10^{6}

n_{OBS} = ~ 10^{4}-10^{6}



Optimised MCF emissions
and Initial MCF field

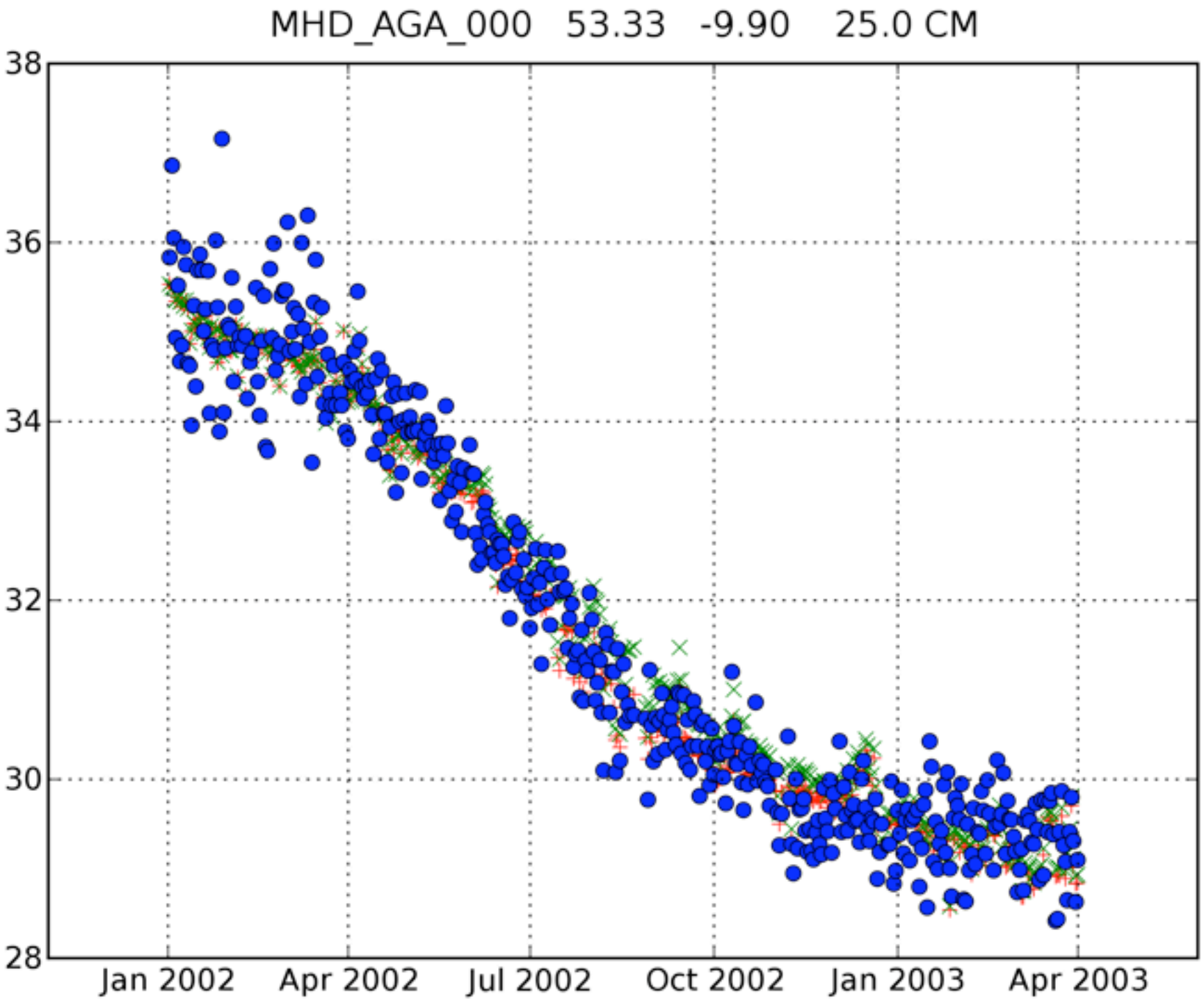
Ragged Point, Barbados (lat = 13N)

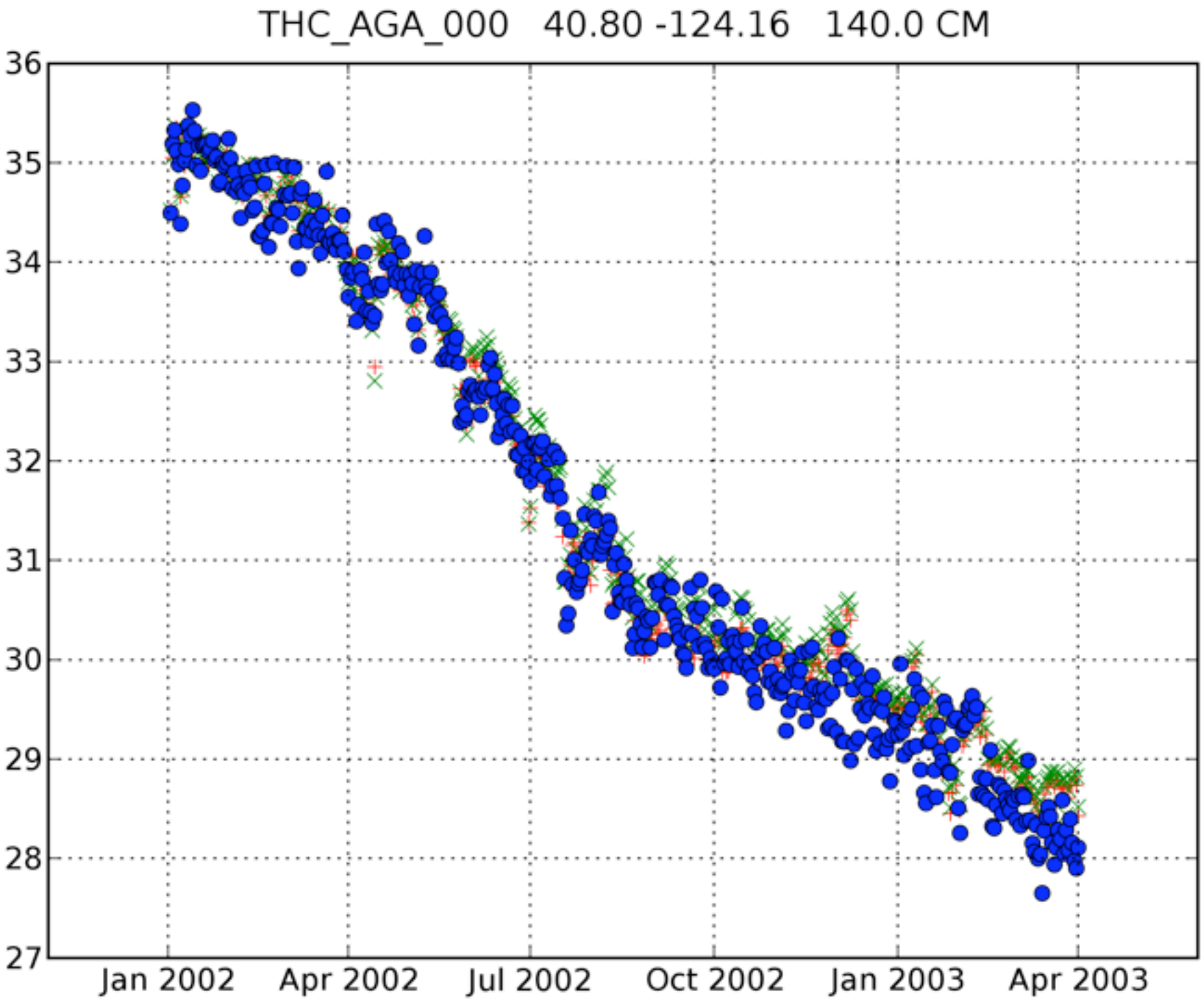


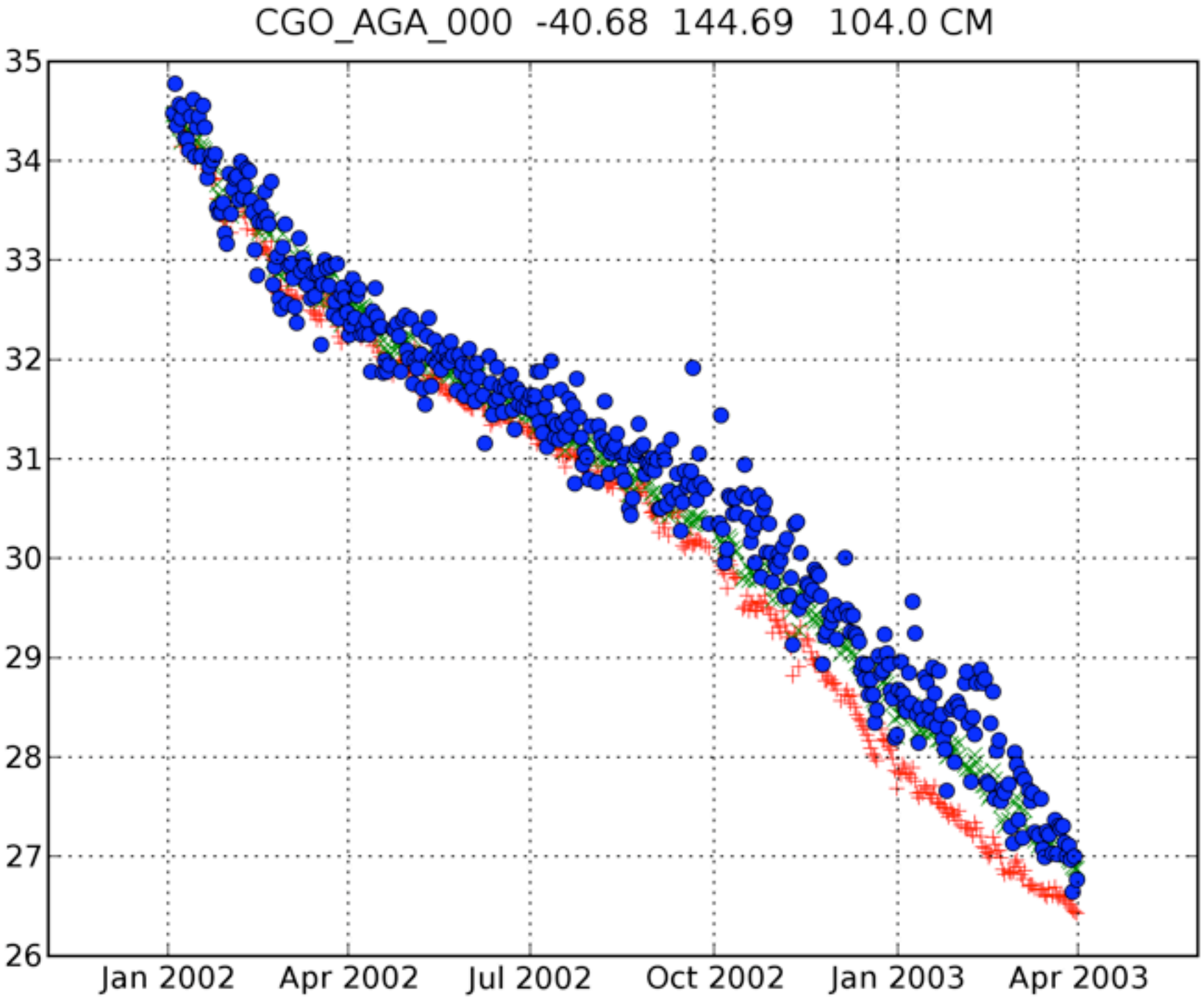
Model Reproduces Variability

Next Step

- Optimize OH concentration in the 2000-2006 period with the 4DVAR-TM5 approach


MHD_AGA_000 53.33 -9.90 25.0 CM
38
36
34
32
30
28
Jan 2002
Apr 2002
Jul 2002
Oct 2002
Jan 2003
Apr 2003


THC_AGA_000 40.80 -124.16 140.0 CM
36
35
34
33
32
31
30
29
28
27
Jan 2002
Apr 2002
Jul 2002
Oct 2002
Jan 2003
Apr 2003


CGO_AGA_000 -40.68 144.69 104.0 CM
35
34
33
32
31
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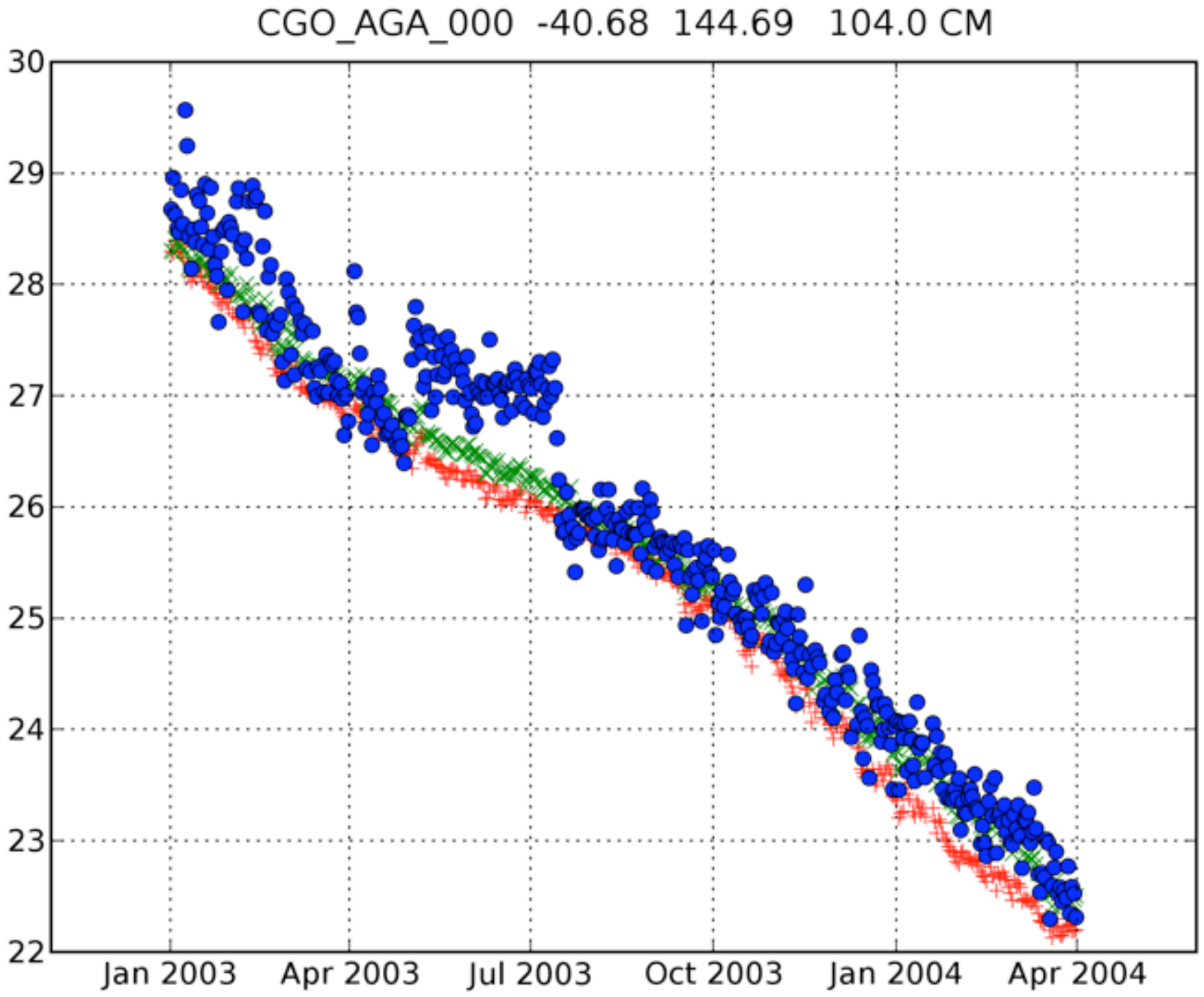
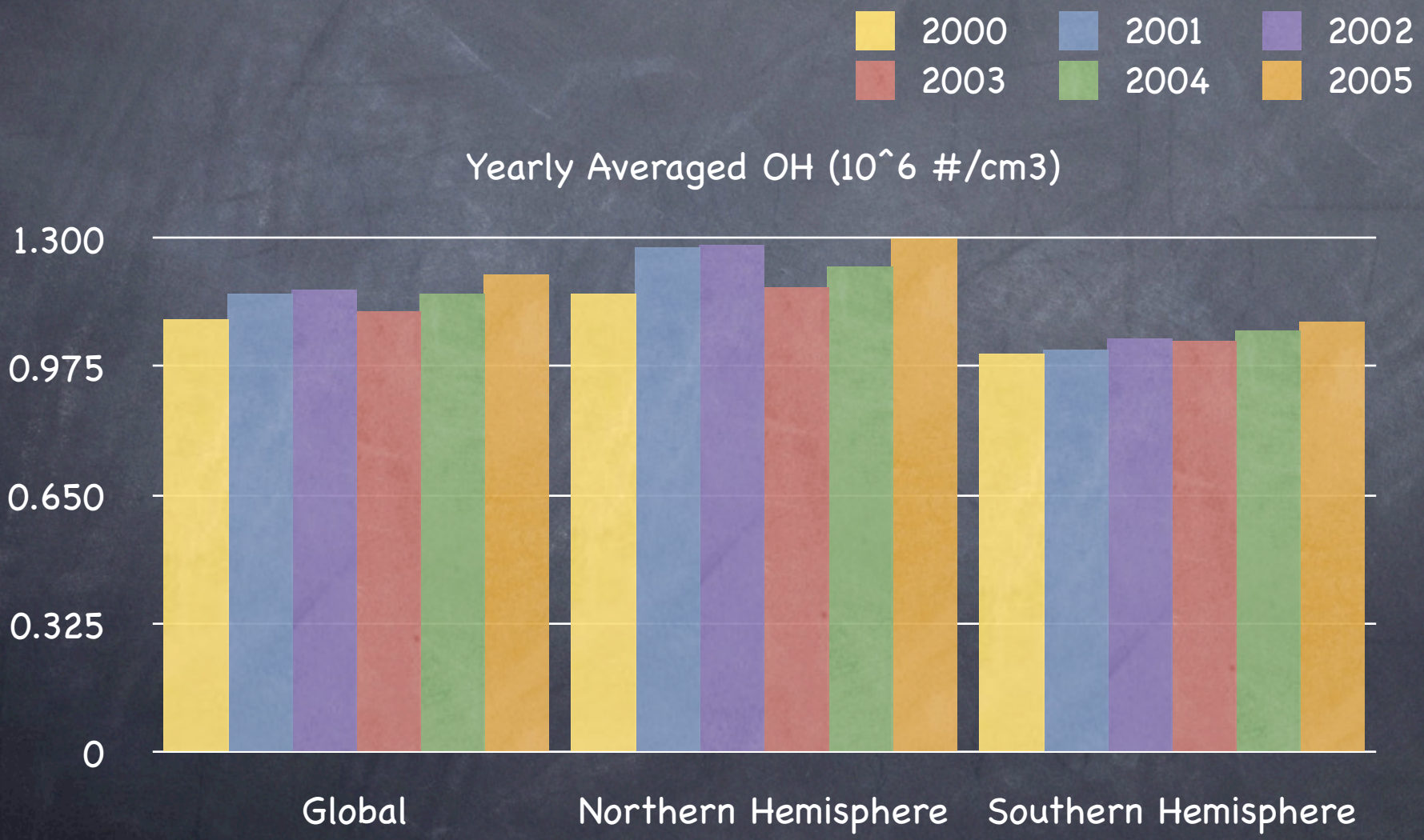
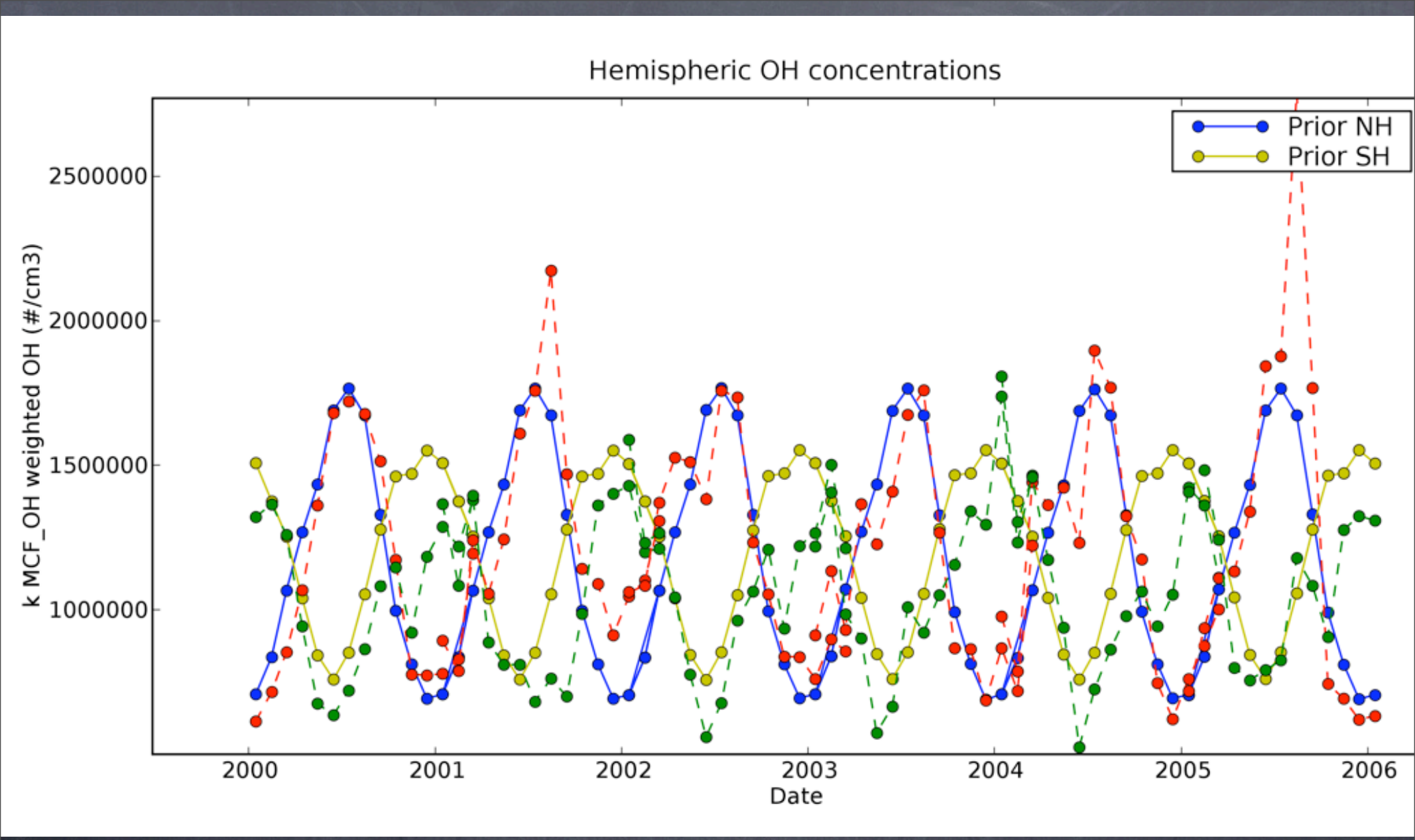
29
28
27
26
Jan 2002
Apr 2002
Jul 2002
Oct 2002
Jan 2003
Apr 2003

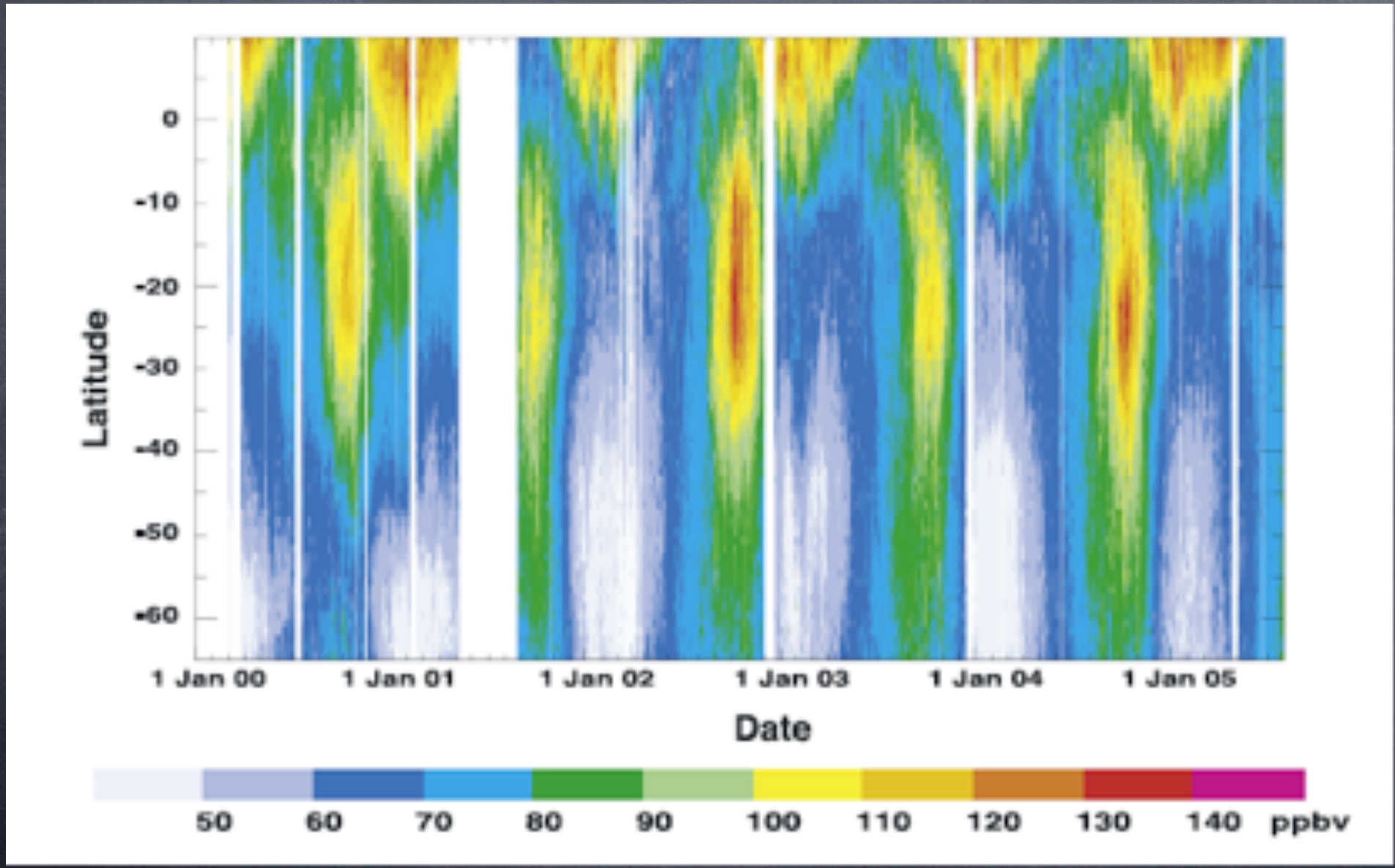

CGO_AGA_000 -40.68 144.69 104.0 CM
30
29
28
27
26
25
24
23
22
Jan 2003
Apr 2003
Jul 2003
Oct 2003
Jan 2004
Apr 2004

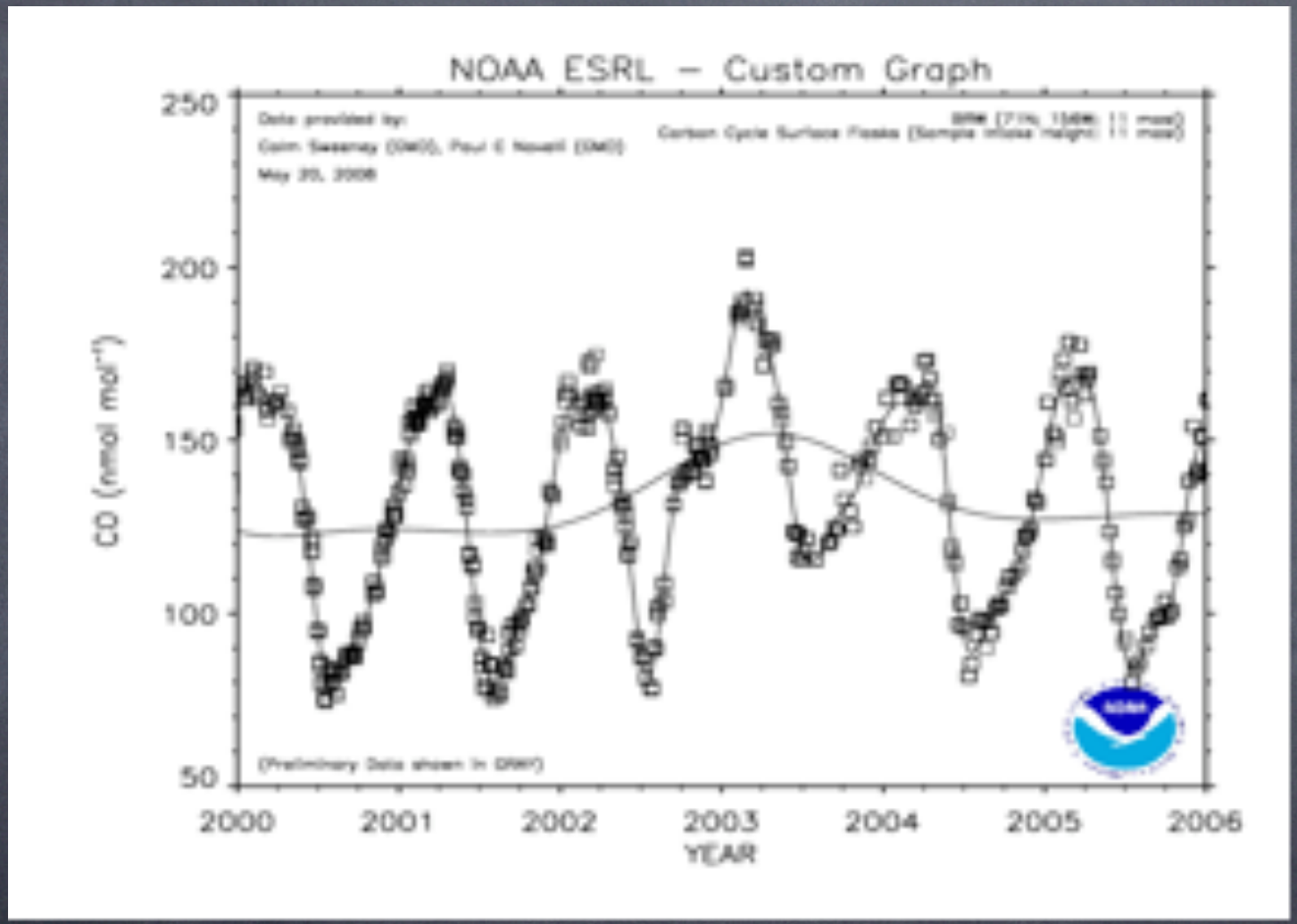
Optimised OH-concentrations




Hemispheric OH concentrations
Prior NH
Prior SH
k MCF_OH weighted OH (#/cm3)
2500000
2000000
1500000
1000000
2000
2001
2002
2003
2004
2005
2006
Date

MOPITT 700 hPa CO zonal mixing ratio (ppbv), from Edwards et al. 2006.





NH anomaly caused by boreal fires 2002-2003

Conclusions

- OH fluctuations: not supported by the methane budget
- Derived OH fluctuations since 2000 generally small
- Existing fluctuations align with 'known' perturbations in atmospheric CO (main OH sink)
- Derived OH fluctuations in 1980s and 1990s are probably the result of an overstated accuracy of the MCF emissions