Can changes in OH explain methane growth variability?

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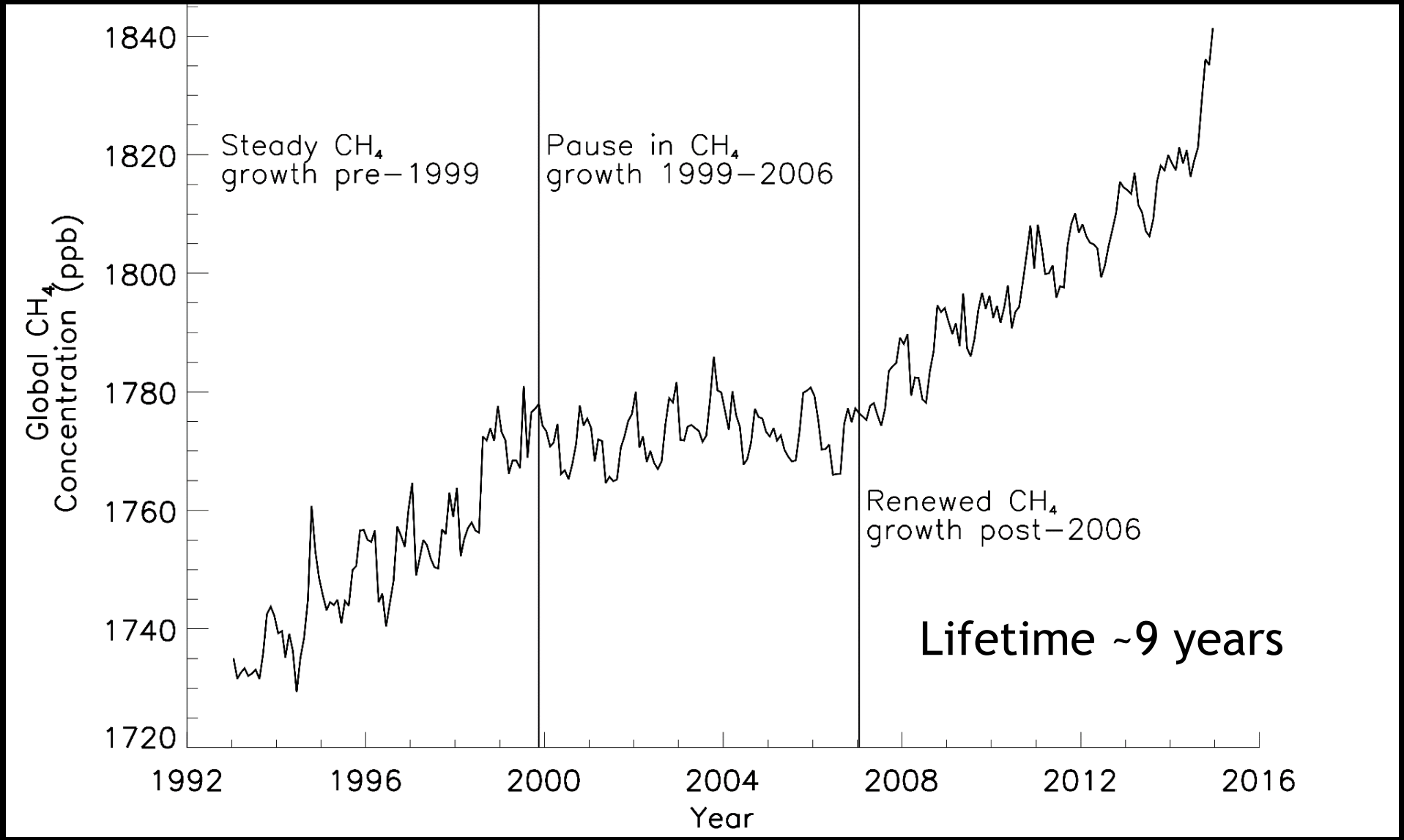
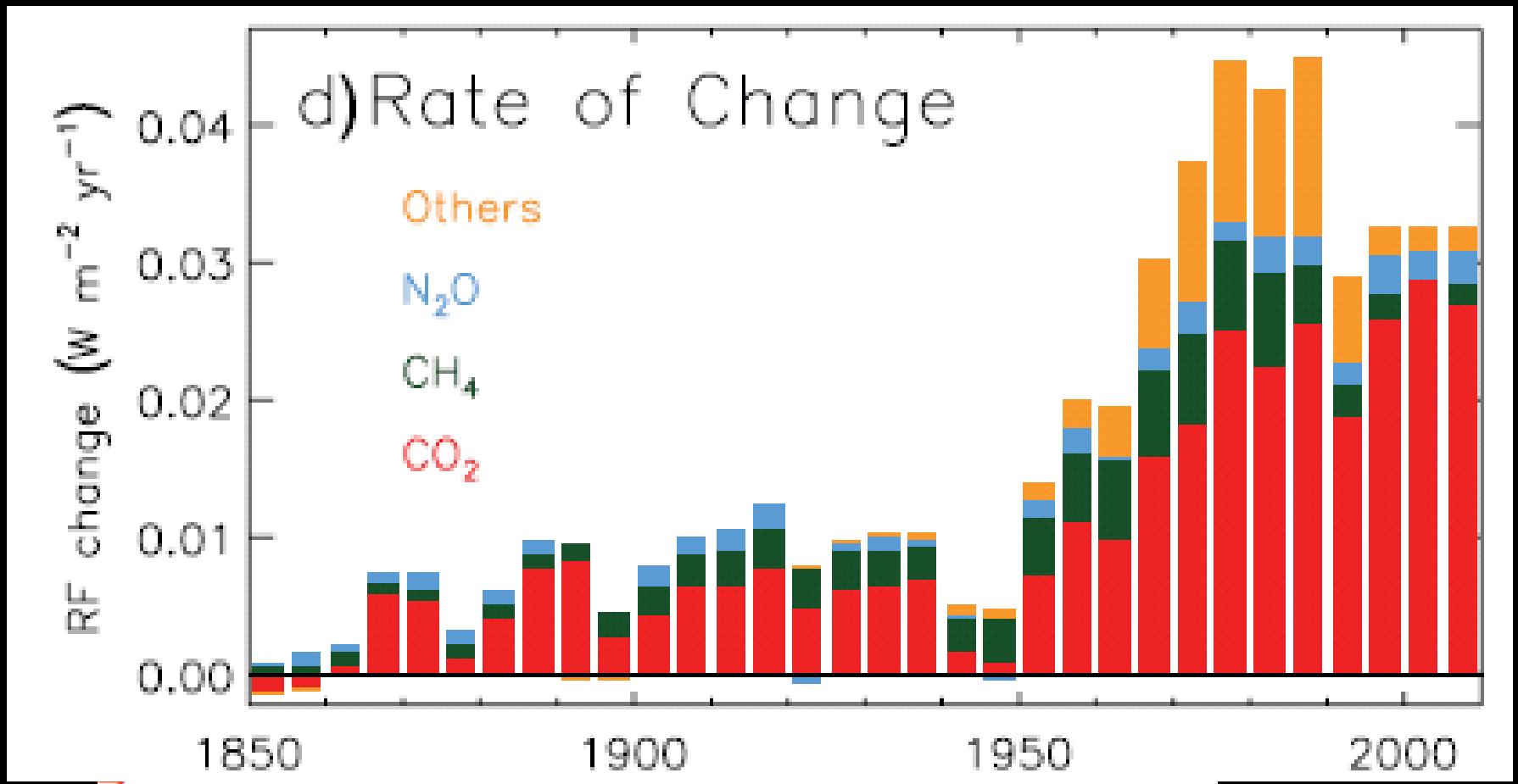
The Methane Puzzle



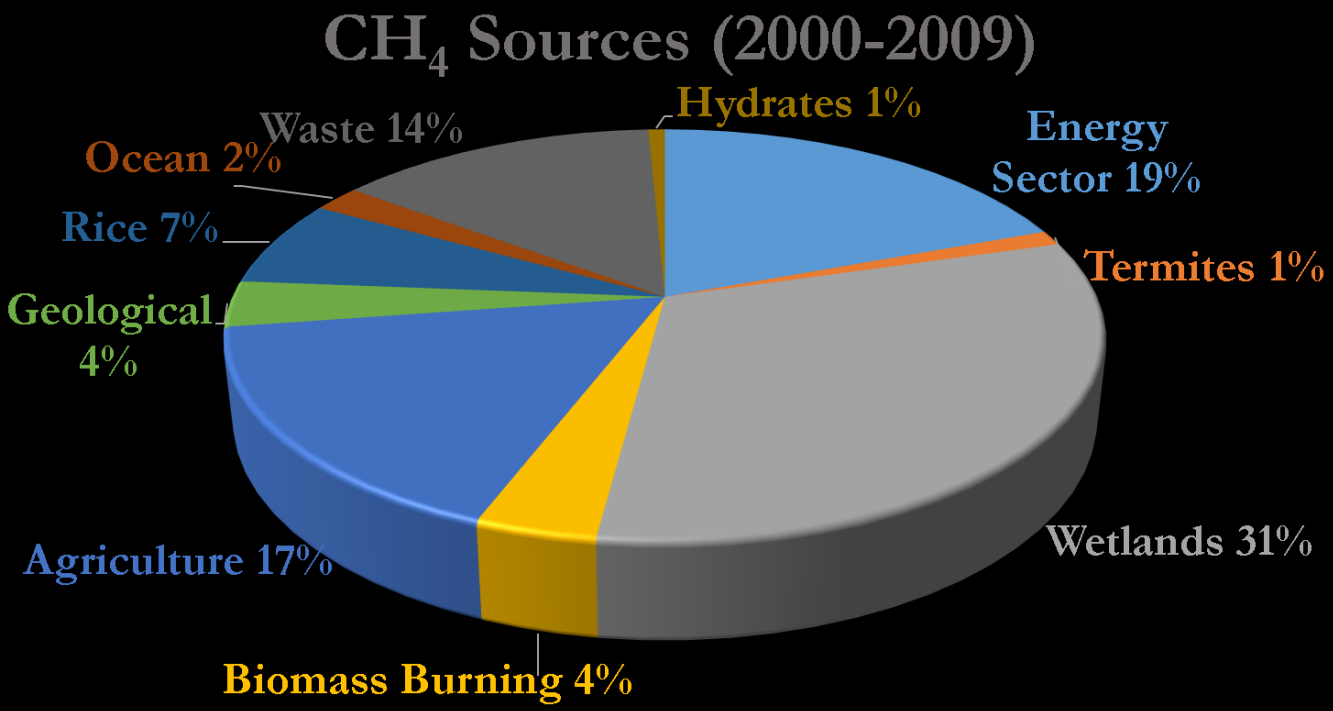
Why study methane?

- ~50% Anthropogenic/ ~50% Natural
- Lifetime ~9 years
- 0.48Wm^{-2} (~20%) of total radiative forcing by long-lived GHGs
- Pre-industrial 722 - now 1840 ppb



(IPCC, 2014)

Cause of change : Sources and Sinks



90% of loss occurs through the reaction with OH

$$CH_4 + OH \rightarrow CH_3 + H_2O$$

Cause of change : Possibilities

1) Change in Anthropogenic Emissions

Emissions continued to rise between 1999 and 2006 (Olivier *et al*. 2005)

2) Change in Natural Emissions

Wetland emissions showed only a small decrease between 1999 and 2006 (McNorton *et al*. 2016)

3) Change in Atmospheric Conditions

Increase in atmospheric loss rate between 1999-2006.

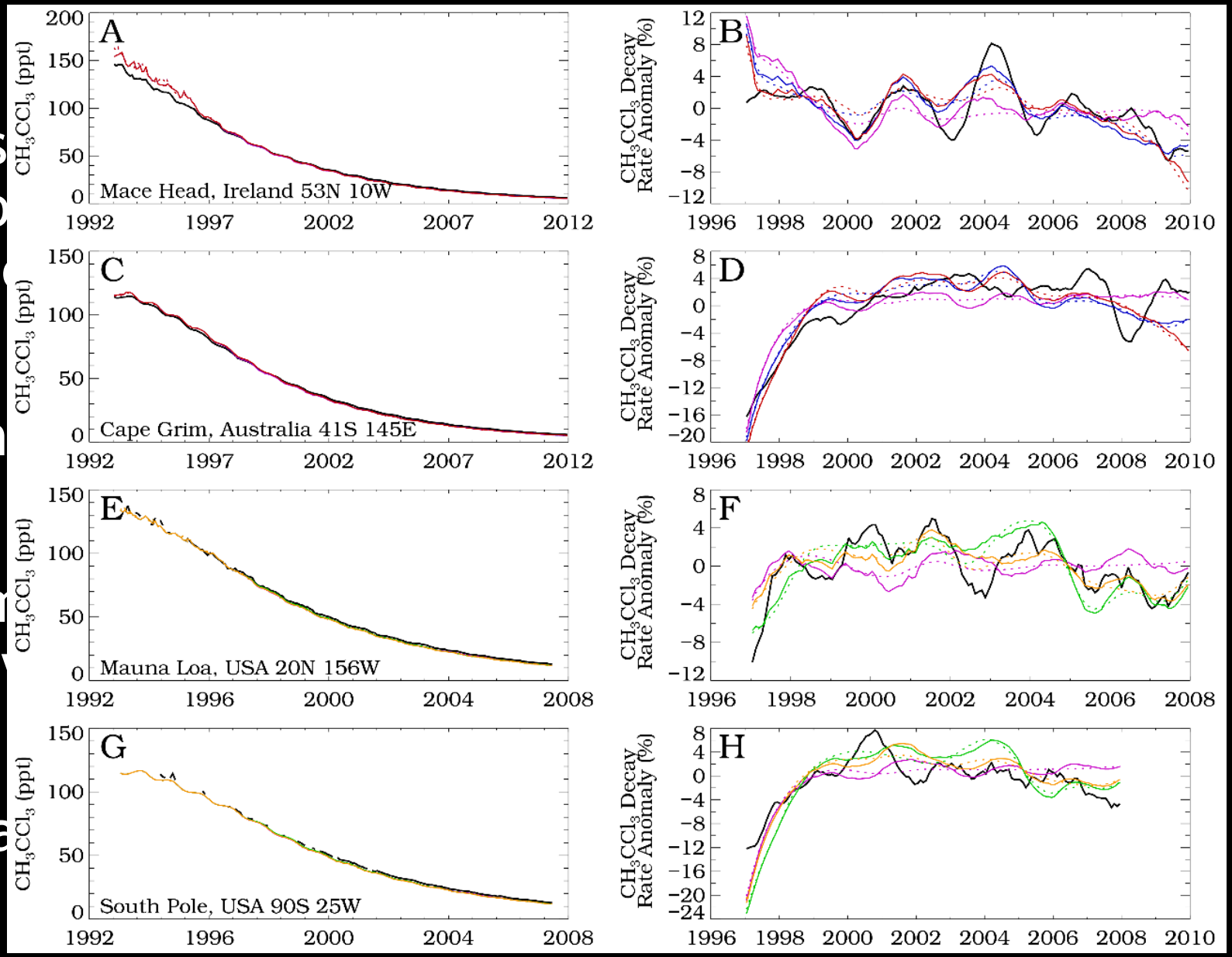
- **Transport from source to loss regions.**
- **Atmospheric temperature.**
- **Sink chemical concentrations, in particular OH (~90% of the total loss).**

Deriving OH

- Global mean OH concentrations and trends are difficult to derive using local direct measurements.
- Indirect techniques using OH-oxidised trace gases can be used for global OH concentrations.
- We used CH_3CCl_3 to derive global OH anomalies between 1997-2009.

Methyl Chloroform – CH_3CCl_3

- Us
  bo
  et
- Ph
- Em
  to
- Ma



Deriving OH

One-box-model

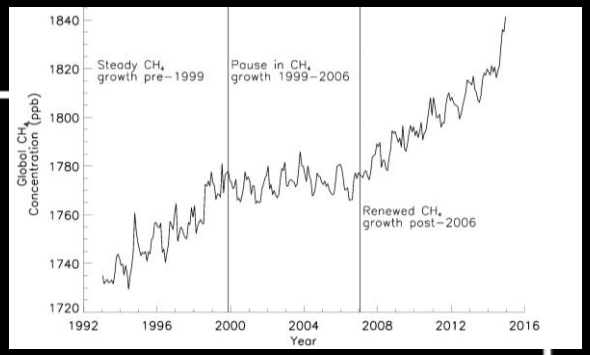
Simple atmosphere to calculate emissions/OH based on observations.

$$\frac{1}{\Delta t}(X_{t+\Delta t} - X_t) = E - L = E - k[OH][X]$$

Inversion constrained by surface CH_3CCl_3 measurements.

*Non-OH sinks are not considered (transport to stratosphere and ocean uptake).
*CH_3CCl_3 observations assumed to represent global concentrations.

Model derived OH anomalies



$$CH_4 + OH \rightarrow CH_3 + H_2O$$

A positive anomaly in OH increases CH_4 destruction.

[OH] Anomaly

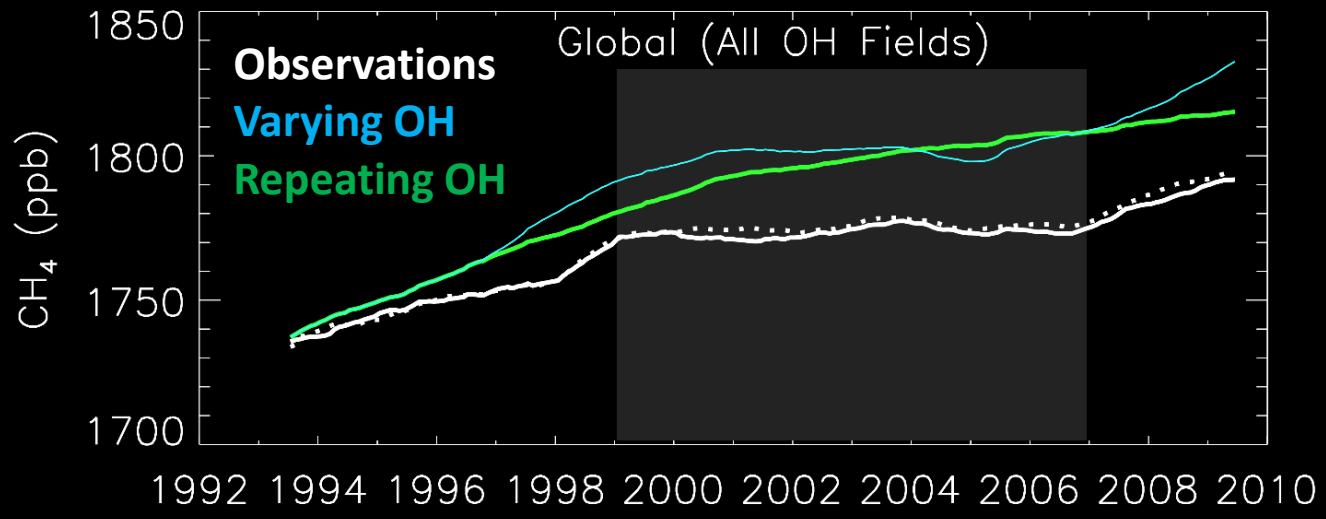
Year

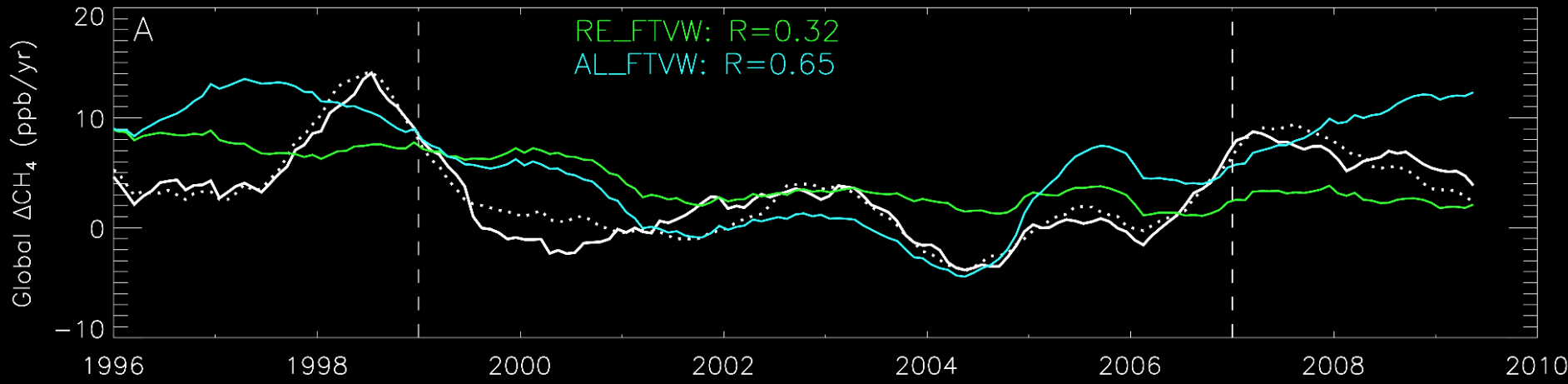
Testing derived OH on CH_4 in a global chemical transport model (CTM), TOMCAT

- Offline 6-hourly meteorology (ECMWF ERA-Interim).
- 60 σ-p levels from surface ~60km.
- Simulation from 1993-2011 with 10-day and monthly output.
- Horizontal resolution 2.8° by 2.8°.

- **Uses annually repeating OH field (Spivakovsky *et al.*2000) which has interannual anomalies applied** .
- **Constant emissions (553 Tg/yr).**

Results from TOMCAT simulation





Conclusions

- One-box model anomalies of CH3CCl3-derived [OH], negatively correlated with CH_4 growth (R = -0.32 (NOAA) and -0.64 (AGAGE)).

CH_3CCl_3

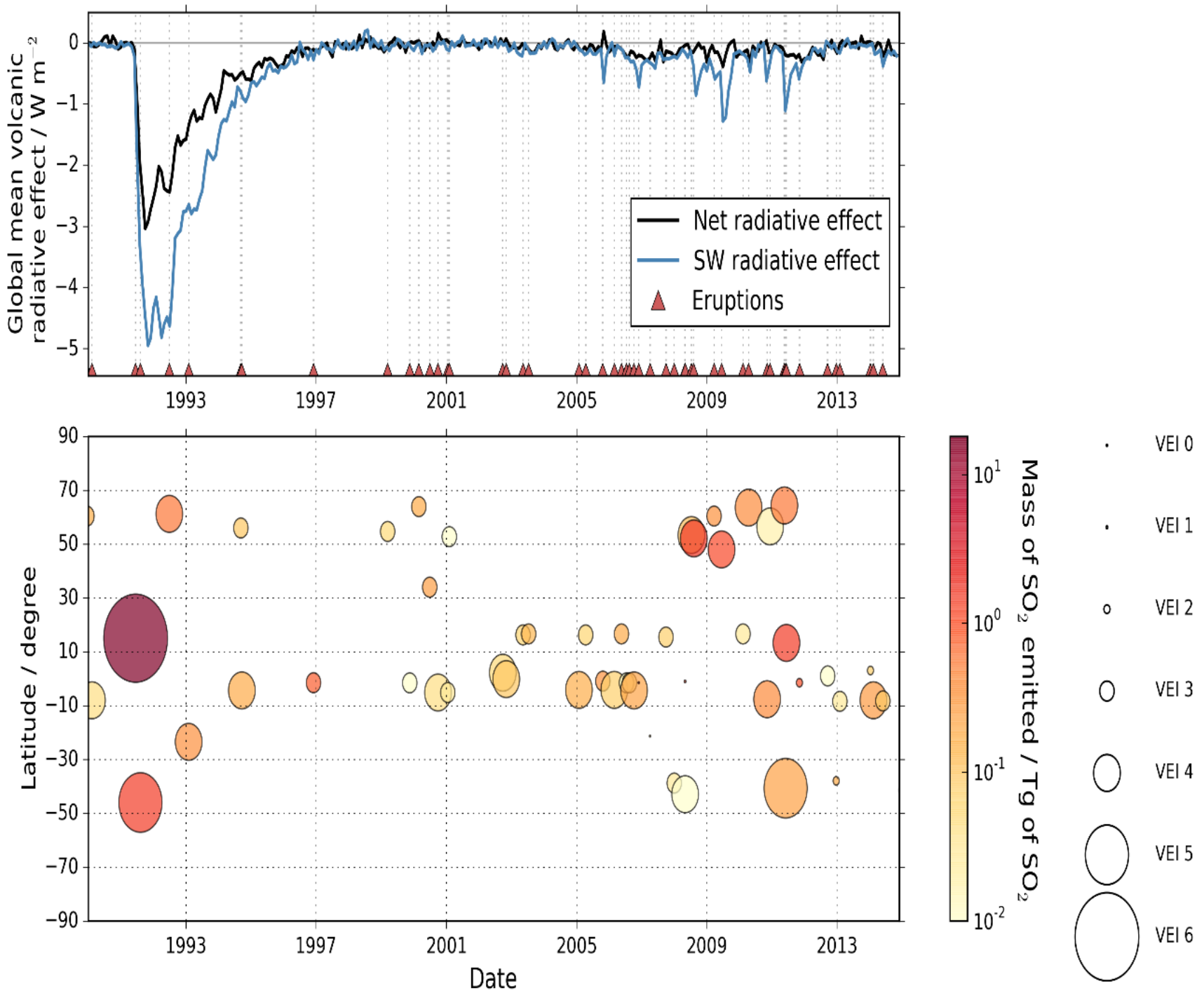
- CTM shows derived [OH] provides better CH_3CCl_3 correlation with observations (R = 0.71 – 0.90) compared with repeating [OH] (R = 0.65).

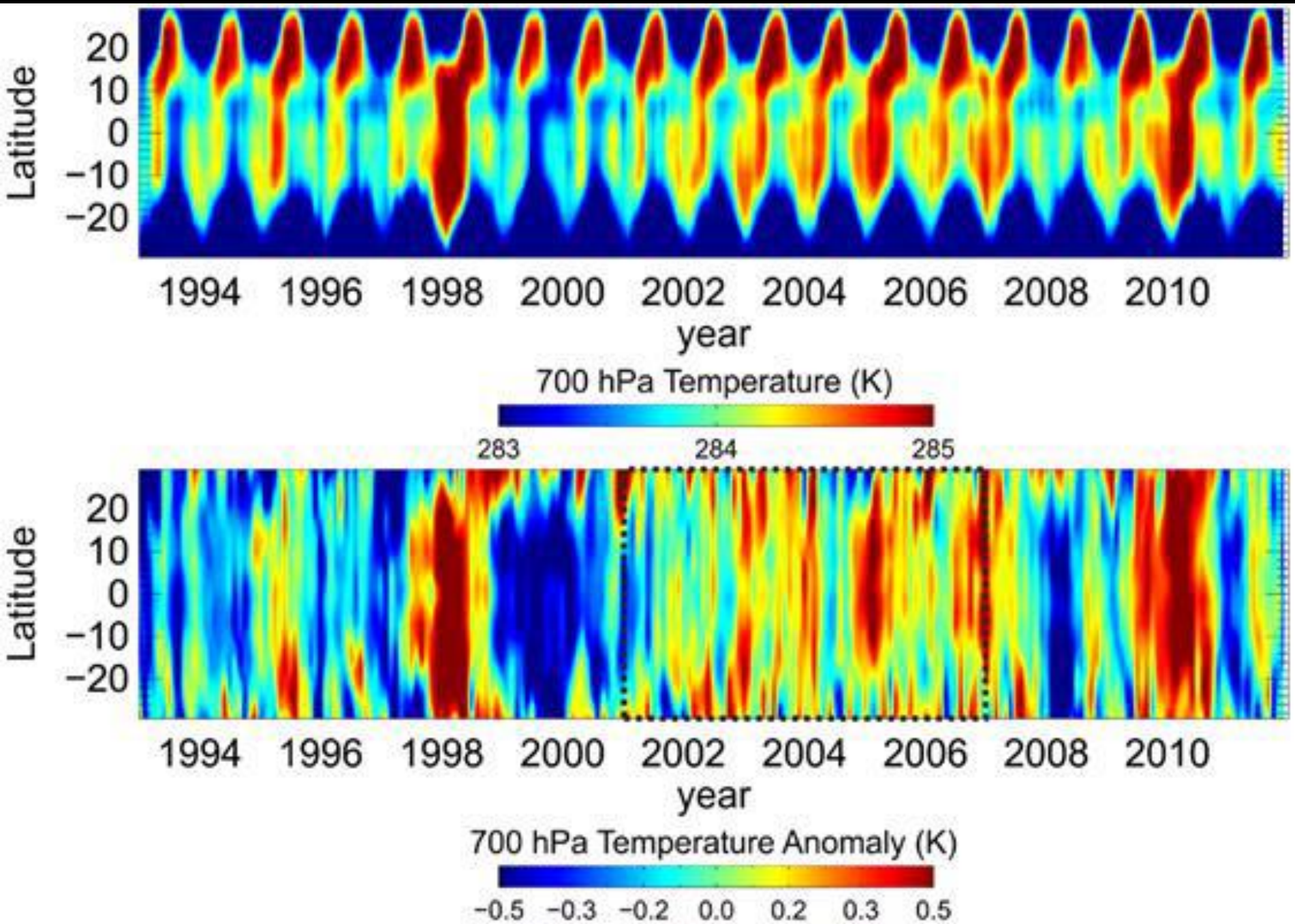
- Box-model derived [OH] used in CTM accurately predict the CH_3CCl_3 decay rate anomaly at individual stations.

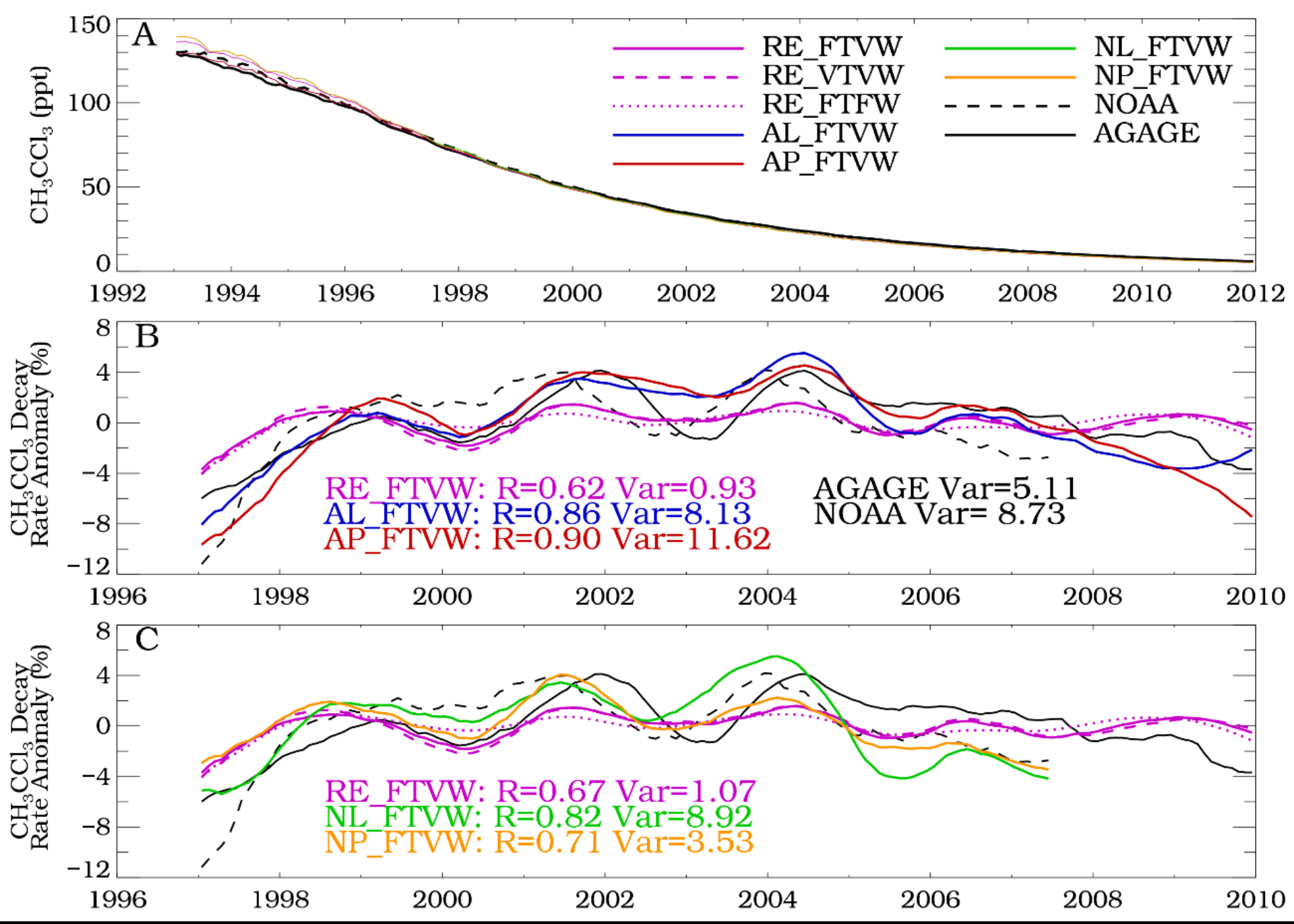
CH_4

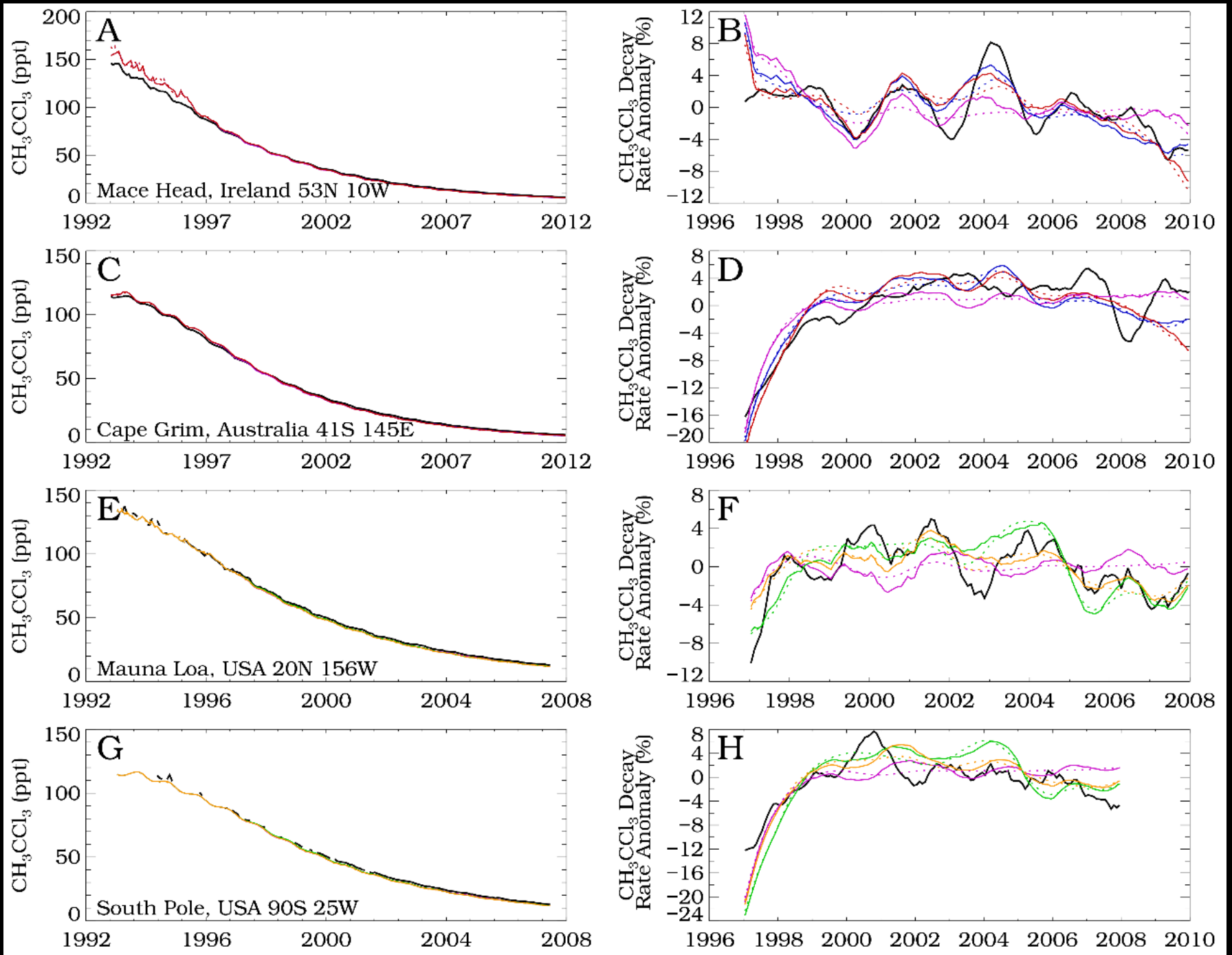
- CH_4 emissions required during the 'hiatus' (1999-2006) are higher (549.7–553.8 Tg/yr) with varying [OH] than without (546.2–548.4 Tg/yr).

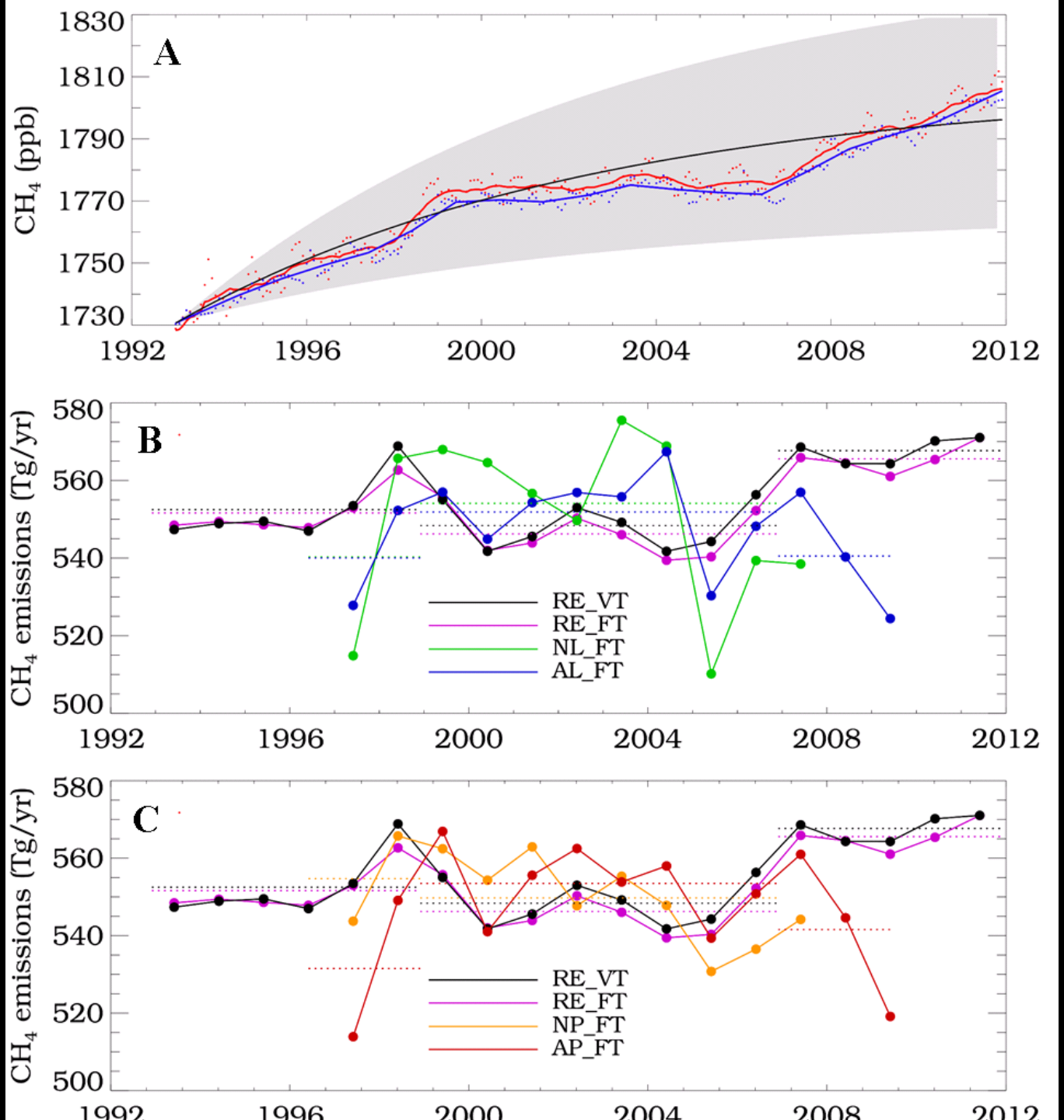
- CH_4 growth correlation between model and observations increases when box-model derived global [OH] is used (R = 0.58-0.65 v 0.32).

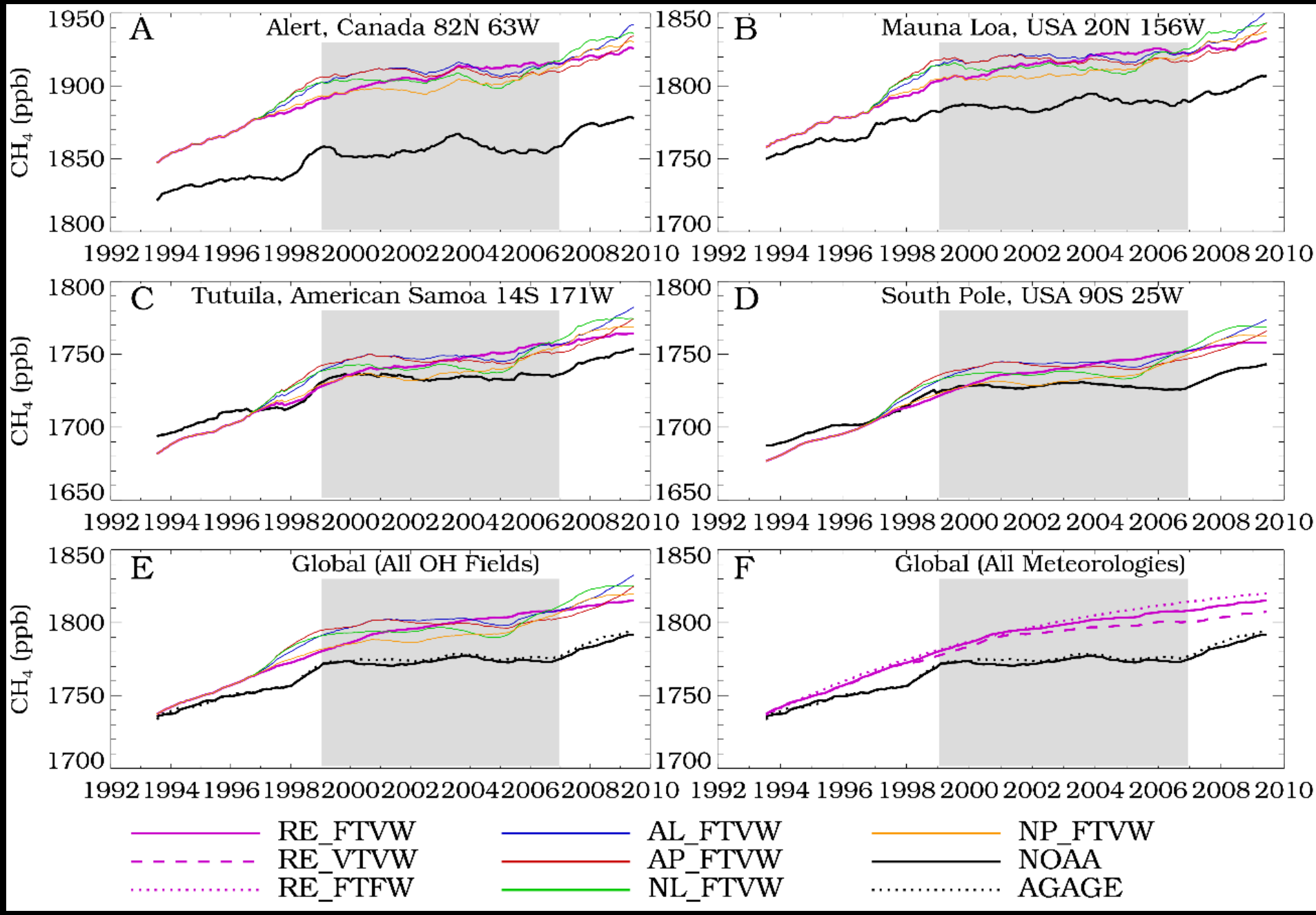

Global mean volcanic radiative effect / W m^{-2}
0
−1
−2
−3
−4
−5
1993
1997
2001
2005
2009
2013
Net radiative effect
SW radiative effect
Eruptions
Latitude / degree
90
70
50
30
10
−10
−30
−50
−70
−90
Date
Mass of SO_2 emitted / Tg of SO_2
10^{1}
10^{0}
10^{-1}
10^{-2}
VEI 0
VEI 1
VEI 2
VEI 3
VEI 4
VEI 5
VEI 6

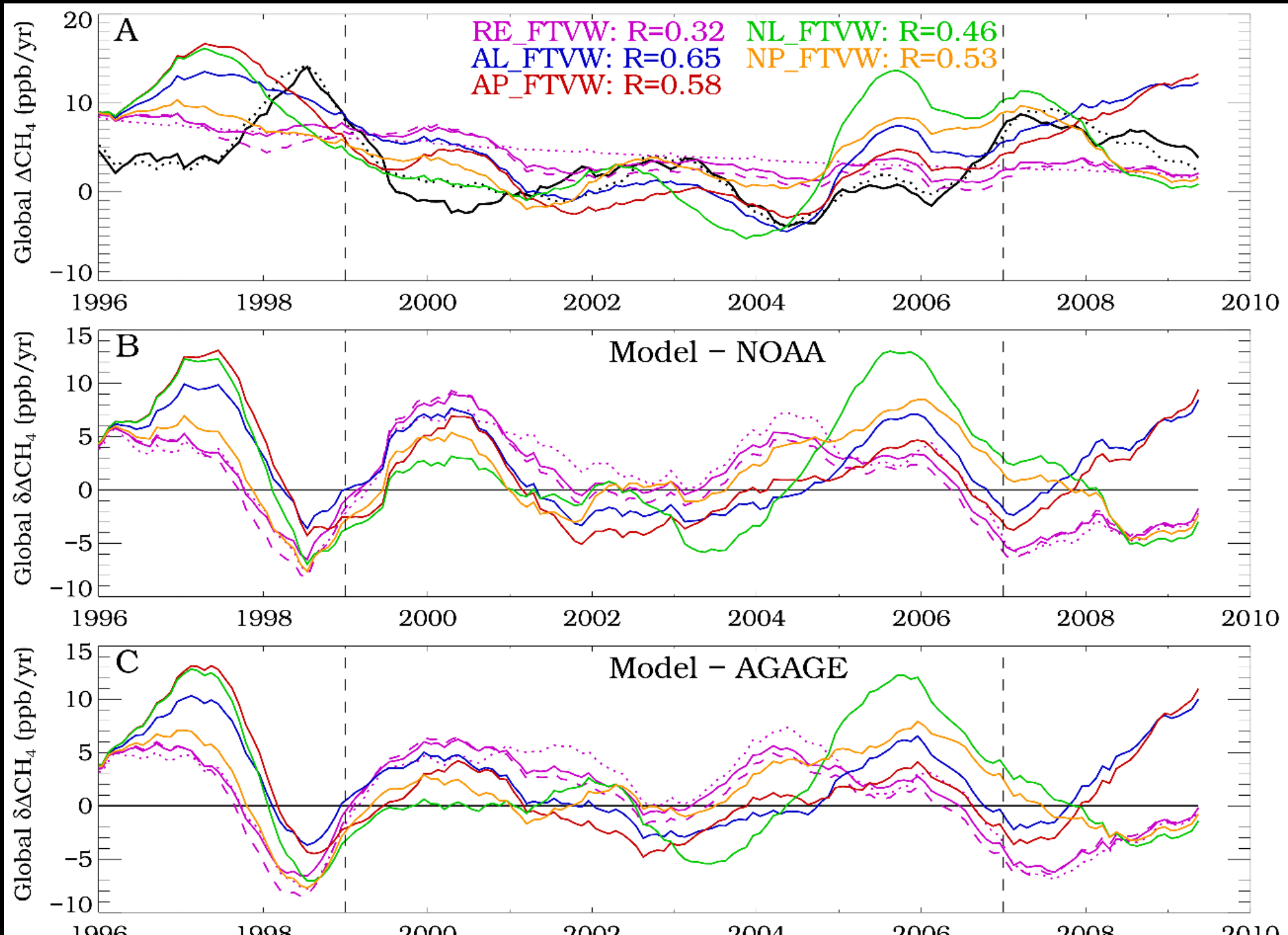

Latitude
20
10
0
−10
−20
1994
1996
1998
2000
2002
2004
2006
2008
2010
year
700 hPa Temperature (K)
283
284
285
Latitude
20
10
0
−10
−20
1994
1996
1998
2000
2002
2004
2006
2008
2010
year
700 hPa Temperature Anomaly (K)
−0.5
−0.3
−0.2
0.0
0.2
0.3
0.5


A
B
C
CH_3CCl_3 (ppt)
CH_3CCl_3 Decay Rate Anomaly (%)
RE_FTVW
RE_VTVW
RE_FTFW
AL_FTVW
AP_FTVW
NL_FTVW
NP_FTVW
NOAA
AGAGE
RE_FTVW: R=0.62 Var=0.93
AL_FTVW: R=0.86 Var=8.13
AP_FTVW: R=0.90 Var=11.62
AGAGE Var=5.11
NOAA Var= 8.73
RE_FTVW: R=0.67 Var=1.07
NL_FTVW: R=0.82 Var=8.92
NP_FTVW: R=0.71 Var=3.53


A
Mace Head, Ireland 53N 10W
B
C
Cape Grim, Australia 41S 145E
D
E
Mauna Loa, USA 20N 156W
F
G
South Pole, USA 90S 25W
H
CH3CCl3 (ppt)
CH3CCl3 Decay Rate Anomaly (%)


A
1830
1810
1790
1770
1750
1730
CH4 (ppb)
1992
1996
2000
2004
2008
2012
B
580
560
540
520
500
CH4 emissions (Tg/yr)
RE_VT
RE_FT
NL_FT
AL_FT
C
NP_FT
AP_FT


A
Alert, Canada 82N 63W
B
Mauna Loa, USA 20N 156W
C
Tutuila, American Samoa 14S 171W
D
South Pole, USA 90S 25W
E
Global (All OH Fields)
F
Global (All Meteorologies)
CH4 (ppb)
1992 1994 1996 1998 2000 2002 2004 2006 2008 2010
RE_FTVW
RE_VTVW
RE_FTFW
AL_FTVW
AP_FTVW
NL_FTVW
NP_FTVW
NOAA
AGAGE


A
RE_FTVW: R=0.32
AL_FTVW: R=0.65
AP_FTVW: R=0.58
NL_FTVW: R=0.46
NP_FTVW: R=0.53
Global ΔCH4 (ppb/yr)
20
10
0
−10
1996
1998
2000
2002
2004
2006
2008
2010
B
Model − NOAA
Global δΔCH4 (ppb/yr)
15
10
5
0
−5
−10
C
Model − AGAGE