



Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J., Cunnold, D. M., Steele, L. P., Krummel, P. B., Weiss, R. F., O'Doherty, S. J., Salameh, P. K., Wang, H. J., Harth, C. M., Muhle, J., & Porter, L. W. (2008). Renewed growth of atmospheric methane. *Geophysical Research Letters*, 35(22).  
<https://doi.org/10.1029/2008GL036037>

Publisher's PDF, also known as Version of record

Link to published version (if available):  
[10.1029/2008GL036037](https://doi.org/10.1029/2008GL036037)

[Link to publication record in Explore Bristol Research](#)  
PDF-document

This article was first published by the American Geophysical Union in *Geophysical Research Letters* [Volume 35, Issue 22, 20/11/2008]. DOI: 10.1029/2008GL036037. Copyright 2008 by the American Geophysical Union.

## University of Bristol - Explore Bristol Research

### General rights

This document is made available in accordance with publisher policies. Please cite only the published version using the reference above. Full terms of use are available:  
<http://www.bristol.ac.uk/red/research-policy/pure/user-guides/ebr-terms/>



## Renewed growth of atmospheric methane

M. Rigby,<sup>1</sup> R. G. Prinn,<sup>1</sup> P. J. Fraser,<sup>2</sup> P. G. Simmonds,<sup>3</sup> R. L. Langenfelds,<sup>2</sup> J. Huang,<sup>1</sup> D. M. Cunnold,<sup>4</sup> L. P. Steele,<sup>2</sup> P. B. Krummel,<sup>2</sup> R. F. Weiss,<sup>5</sup> S. O'Doherty,<sup>3</sup> P. K. Salameh,<sup>5</sup> H. J. Wang,<sup>4</sup> C. M. Harth,<sup>5</sup> J. Mühle,<sup>5</sup> and L. W. Porter<sup>6,7</sup>

Received 17 September 2008; accepted 16 October 2008; published 20 November 2008.

[1] Following almost a decade with little change in global atmospheric methane mole fraction, we present measurements from the Advanced Global Atmospheric Gases Experiment (AGAGE) and the Australian Commonwealth Scientific and Industrial Research Organisation (CSIRO) networks that show renewed growth starting near the beginning of 2007. Remarkably, a similar growth rate is found at all monitoring locations from this time until the latest measurements. We use these data, along with an inverse method applied to a simple model of atmospheric chemistry and transport, to investigate the possible drivers of the rise. Specifically, the relative roles of an increase in emission rate or a decrease in concentration of the hydroxyl radical, the largest methane sink, are examined. We conclude that: 1) if the annual mean hydroxyl radical concentration did not change, a substantial increase in emissions was required simultaneously in both hemispheres between 2006 and 2007; 2) if a small drop in the hydroxyl radical concentration occurred, consistent with AGAGE methyl chloroform measurements, the emission increase is more strongly biased to the Northern Hemisphere. **Citation:** Rigby, M., et al. (2008), Renewed growth of atmospheric methane, *Geophys. Res. Lett.*, 35, L22805, doi:10.1029/2008GL036037.

### 1. Introduction

[2] Methane (CH<sub>4</sub>) is an important greenhouse gas and plays a significant role in ozone layer chemistry [Forster et al., 2007, Denman et al., 2007]. Its average concentration in the atmosphere is largely determined by a balance between emission from the surface and destruction by hydroxyl free radicals (OH) in the troposphere. Dominant emission sources include boreal and tropical wetlands (including rice paddies), anthropogenic activity (for example, from fossil fuels and ruminant animals), and biomass burning.

[3] Previous measurements of atmospheric CH<sub>4</sub> showed a persistent increase in the global burden throughout the

latter part of the twentieth century [Dlugokencky et al., 1994; Cunnold et al., 2002], followed by a period of little change since 1999 [Dlugokencky et al., 2003]. Before 2007, the last significant global growth event occurred during the *El Niño* of 1997 and 1998. This period was associated with increased emission from wetland and rice growing regions and biomass burning [Chen and Prinn, 2006; Langenfelds et al., 2002]. A decrease in the OH concentration may also have occurred, possibly due to the increased levels of carbon monoxide (CO), the main OH sink, released in widespread wildfires, and through changes in water vapor, cloud cover and temperature [Prinn et al., 2005; Bousquet et al., 2006].

[4] Here we report measurements from the AGAGE and CSIRO networks that show renewed growth from the end of 2006 or beginning of 2007 until the most recent measurements. We investigate this observation further using a simple model of atmospheric transport and chemistry to attempt to quantify the required increase in hemispheric-scale emissions assuming either inter-annually repeating OH, or variable annual mean OH levels consistent with methyl chloroform observations.

### 2. Recent AGAGE/CSIRO Measurements

[5] The atmospheric CH<sub>4</sub> data presented in this paper were collected by two global monitoring networks. AGAGE makes high-precision, high-frequency (approximately every 40 minutes) measurements of the most radiatively important non-carbon dioxide (CO<sub>2</sub>) “greenhouse” gases and many ozone-depleting species [Prinn et al., 2000]. The network consists of five stations located in coastal regions at latitudes from 53°N to 41°S (Table 1). The gas chromatograph/flame ionization CH<sub>4</sub> measurements are described in detail by Cunnold et al. [2002]. Monthly average AGAGE mole fractions are used here in which pollution events have been removed using a statistical technique in order to better represent “baseline” air masses [see, e.g., O'Doherty et al., 2001]. Whilst the CSIRO flask network collects data at a much lower frequency than AGAGE, it provides an independent and complementary data set, and a wider latitudinal site distribution [Langenfelds et al., 2002]. Flasks are collected from eight locations around the globe (Table 1) and pollution events are avoided by conditional sampling of air from unpolluted wind sectors. Measurements from both networks are reported here on the Tohoku University scale [Cunnold et al., 2002].

[6] The errors in the measured mole fractions include the monthly variability in the baseline and an instrumental precision error, generously taken to be 4 nmol mol<sup>-1</sup> [Chen and Prinn, 2006]. Since it is not possible to determine the variability at the CSIRO flask sites, this is assumed to be

<sup>1</sup>Center for Global Change Science, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA.

<sup>2</sup>Centre for Australian Weather and Climate Research, CSIRO Marine and Atmospheric Research, Aspendale, Victoria, Australia.

<sup>3</sup>School of Chemistry, University of Bristol, Bristol, UK.

<sup>4</sup>School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, Georgia, USA.

<sup>5</sup>Scripps Institution of Oceanography, UCSD, La Jolla, California, USA.

<sup>6</sup>Australian Government Bureau of Meteorology, Melbourne, Victoria, Australia.

<sup>7</sup>Deceased 7 December 2007.

**Table 1.** AGAGE and CSIRO Measurement Sites

| ID  | Name                          | Network     | Latitude | Longitude |
|-----|-------------------------------|-------------|----------|-----------|
| alt | Alert, Canada                 | CSIRO       | 82       | -62       |
| mhd | Mace Head, Ireland            | AGAGE       | 53       | -9        |
| thd | Trinidad Head, California     | AGAGE       | 41       | -124      |
| mlo | Mauna Loa, Hawaii             | CSIRO       | 19       | -155      |
| rpb | Ragged Point, Barbados        | AGAGE       | 13       | 42        |
| smo | Cape Matatula, American Samoa | AGAGE       | -14      | -170      |
| cfa | Cape Ferguson, Queensland     | CSIRO       | -19      | 147       |
| cgo | Cape Grim, Tasmania           | AGAGE/CSIRO | -41      | 145       |
| mqa | Macquarie Island, Australia   | CSIRO       | -55      | 159       |
| cya | Casey, Antarctica             | CSIRO       | -67      | 17        |
| maa | Mawson, Antarctica            | CSIRO       | -68      | 63        |
| spo | South Pole, Antarctica        | CSIRO       | -89      | -24       |

equal to that of the nearest AGAGE station in each month. The CSIRO mole fraction errors also include an estimate of the uncertainty in the flask baseline, using the deviation of the monthly average from a curve fit to a longer timescale ( $\sim 80$  days) record [Langenfelds *et al.*, 2002].

### 3. Renewed Methane Growth

[7] Monthly mean and annual running mean mole fractions at each AGAGE monitoring site and five CSIRO sites from January 1997 to April 2008 are presented in Figure 1 (a subset of CSIRO sites are shown for clarity; the three additional high-latitude Southern Hemisphere (SH) sites show very similar behaviour to the measurements at the South Pole and Cape Grim). Also shown is the annual running mean growth rate at each site. The growth rate is calculated simply as the annual average year-on-year calendar month mole fraction change (i.e., the 2007 average growth rate is the mean of the differences  $y_{01/2007} - y_{01/2006}$ ,  $y_{02/2007} - y_{02/2006} \dots y_{12/2007} - y_{12/2006}$ , where  $y$  is the monthly mean methane mole fraction). As discussed above, a relatively rapid growth of between 5 and 15 nmol mol<sup>-1</sup> year<sup>-1</sup> was observed at all sites during the *El Niño* of 1998. In the following years, although some growth was seen in the Northern Hemisphere (NH) in 2002–2003 (possibly linked to biomass burning [Simmonds *et al.*, 2005]), the global average mole fraction remained relatively stable between 1999 and 2006. However, from around early-2007 we see renewed global growth, the rate of which had reached almost 10 nmol mol<sup>-1</sup> yr<sup>-1</sup> at all locations by late-2007.

[8] The pattern of this recent anomaly provides some information about the behaviour of the global sources and sinks that may be responsible. Positive annual average growth rate is found to first appear almost simultaneously at all latitudes around early-2007, and rises at a similar rate across the globe until the most recent measurements. Given that the inter-hemispheric mixing time is of the order of one year, we can infer that if an increase in CH<sub>4</sub> emission is solely responsible, output must have risen almost simultaneously from both hemispheres. Alternatively, a decrease in the OH sink may have occurred, potentially reducing the required emissions rise in one or both hemispheres.

[9] To further investigate the relative roles of these processes and to quantify the emissions change required to produce the measured rise in atmospheric mole fraction, we present an inverse analysis using a simplified model of atmospheric transport and chemistry. In the first part of our

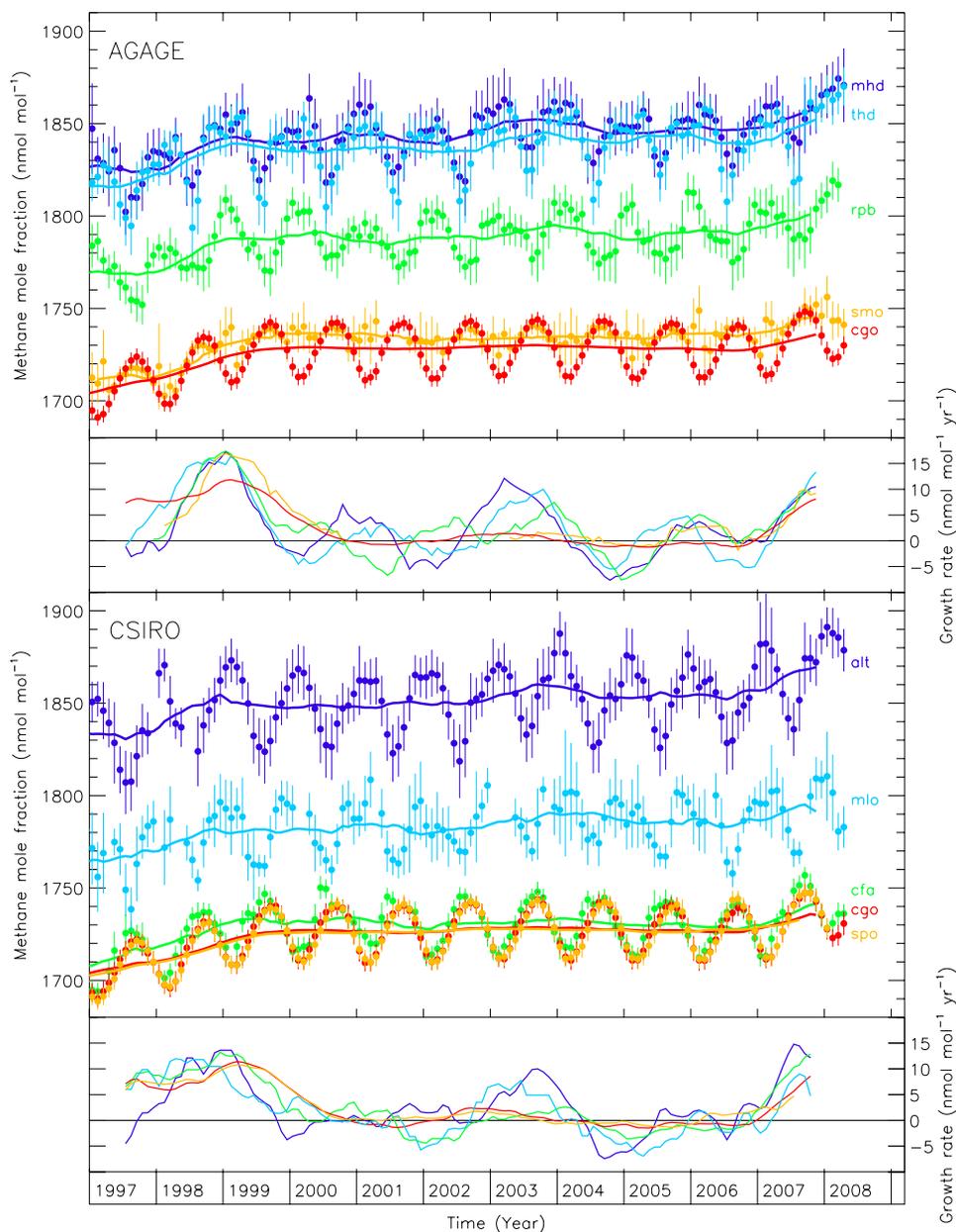
analysis we assume no change in the annual-average OH sink and optimally estimate the contribution of each hemisphere to the required change in global emissions. Our approach enables us to identify the approximate location of any emission increases, but not to attribute these increases to particular emission processes.

[10] Previous studies have identified inter-annual variations in OH concentrations inferred from measurements of methyl chloroform (CH<sub>3</sub>CCl<sub>3</sub>), whose timing, but not necessarily amplitude, appears to be well defined [Prinn *et al.*, 2005; Bousquet *et al.*, 2005]. In the final part of this paper, we extend our previous CH<sub>3</sub>CCl<sub>3</sub> analysis [Prinn *et al.*, 2005] to include more recent years and use the derived OH levels to determine the subsequent CH<sub>4</sub> emission change required to match the observations.

### 4. Modeling and Inversion Methodology

[11] Rather than using a three-dimensional model of atmospheric chemistry and transport [Chen and Prinn, 2006; Bousquet *et al.*, 2006], we utilized the flexibility and computational efficiency afforded by a parameterized two dimensional, twelve-box model which allows inclusion of model parametric errors in the inversions, and which has been previously demonstrated to be reliable for inversion of a number of species including CH<sub>4</sub> and CH<sub>3</sub>CCl<sub>3</sub> [Cunnold *et al.*, 2002; Prinn *et al.*, 2005]. The model consists of eight “tropospheric” and four “stratospheric” boxes that resolve the latitudinal bands 90°S–30°S, 30°S–0°N, 0°N–30°N, and 30°N–90°N, and the vertical layers 0–200, 200–500, and 500–1000mb. OH concentrations in each box follow prescribed seasonal cycles. However, annual mean levels can be specified and we present model output for both inter-annually repeating and inter-annually varying OH. The model includes a parameterization of inter-annual variations in inter-hemispheric transport rate related to the Southern Oscillation Index (SOI), and imposes inter-annually varying temperatures (see auxiliary material).<sup>1</sup> Following Cunnold *et al.* [2002], the lifetime of CH<sub>4</sub> in the stratospheric boxes was set to 20 years (the inversion is relatively insensitive to this value since the removal of CH<sub>4</sub> is dominated by the reaction with tropospheric OH). No soil sink strength was specified, so that the emission rates presented here are net (source-soil sink) surface exchange rates.

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2008GL036037.



**Figure 1.** Monthly average  $\text{CH}_4$  mole fraction (with 12-month running mean, solid line) and annual mean growth rate at AGAGE and CSIRO sites after filtering of pollution events, January 1997–April 2008. Error bars are estimated using the method outlined in the text. Measurement site names and locations are shown in Table 1.

[12] To compare our thirteen  $\text{CH}_4$  point measurement records to the box model mole fractions, semi-hemispheric averages were required. This was formulated by weighting the relative contribution of each station to the semi-hemispheric mean by the inverse square of the estimated station measurement error. The weighted standard error in this box mean was then calculated and used in the inversion.

[13] Following the inversion approach of *Chen and Prinn* [2006], the sensitivity of the  $\text{CH}_4$  mole fractions to monthly pulses of emission from each semi-hemisphere was calculated and used in the inversion to estimate emissions from each month in each semi-hemisphere. *A priori* emission rates derived by *Cunnold et al.* [2002] were assumed, with a 30% error to allow for seasonal and inter-annual variations. Upon comparison with the observations using the Kalman

filter [*Prinn, 2000*], a new set of monthly emissions were derived. The model output with this new estimate was then compared to the observations again and a second estimate obtained to ensure convergence. The  $\text{CH}_4$  mole fractions in the box model are found not to be very sensitive to the differences between emissions from tropical and high latitude semi-hemispheres [see *Cunnold et al., 2002*] (see also auxiliary material). However, the annual mean hemispheric growth rate was well modelled and therefore the derived semi-hemispheric emission rates were aggregated and are presented here as hemispheric annual running mean anomalies. Emission errors were calculated as the sum of the inversion error, an estimate of model parametric error made by running the inversion with perturbed transport parameters [*Prinn et al., 2005*], and an estimate of the influence of

inter-annual OH concentration changes made by running the inversion with annual mean OH levels perturbed by plus and minus one standard deviation.

[14] In the second part of this analysis, the box model was used to estimate the sensitivity of  $\text{CH}_3\text{CCl}_3$  mole fraction to changes in annual-average OH concentrations [Prinn *et al.*, 2005]. The Kalman filter was then employed to find the optimal OH concentration using AGAGE  $\text{CH}_3\text{CCl}_3$  measurements and emission estimates based on production and consumption data from the *United Nations Environment Programme* [2005, 2008; McCulloch and Midgley, 2001]. Since emission estimates are not yet available from 2006 onwards, we linearly extrapolated those of previous years and assumed the same error. The error in our derived OH levels includes inversion and modeling error in addition to the significant emission rate error [Prinn *et al.*, 2005]. Whilst these OH concentrations are sensitive to the projected emissions estimates, the mean values obtained for a range of reasonable emissions fall within this estimated uncertainty. These uncertainties were carried through to our derived monthly  $\text{CH}_4$  emissions estimates.

## 5. Results and Discussion

### 5.1. Inter-annually Repeating OH

[15] Figure 2a shows the annual running mean global, NH and SH emission rate anomalies required to best fit the observed mole fractions, assuming annually repeating OH concentrations. An analysis of the optimized model mole fractions and a posteriori error covariance matrix is presented in the auxiliary material. Between 2006 and 2007 a rise in annual mean emissions of  $40 \pm 43$  Tg/yr was derived globally, made up of a  $22 \pm 35$  Tg/yr increase in the NH and  $18 \pm 24$  Tg/yr in the SH. Figure 2a shows that the emissions are substantially elevated compared to the average, and almost as large as during the growth anomaly of 1998. Although poorly constrained, this analysis indicates that the observations are consistent with a substantial increase in emissions occurring in both hemispheres almost simultaneously in 2007. This finding stems from the near synchronous increase in observed mole fraction across the globe, despite an approximately one year inter-hemispheric exchange time. It therefore seems unlikely that an increase in emission from one hemisphere alone could be solely responsible for the recent increase under inter-annually repeating OH concentrations.

[16] Our analysis cannot attribute this required emissions increase to specific processes. However, we can briefly speculate about the sources most likely to be responsible. Unlike the 1998 anomaly, 2007 was not an *El Niño* year and therefore the processes involved may not be the same. Boreal and tropical wetland emissions are thought to dominate inter-annual  $\text{CH}_4$  emission variability [Chen and Prinn, 2006; Bousquet *et al.*, 2006], and may therefore play a significant role in the recent  $\text{CH}_4$  rise. These emissions are found to be sensitive to an interaction of a number of factors including temperature, water table level and rainfall [e.g., Walter *et al.*, 2001]. Perhaps the most striking climatic anomaly in 2007 was the high ( $\sim +4\text{K}$  compared to 1961–1990) annual mean temperature over Siberia, where extensive wetlands are found [National Climatic Data Center, 2008]. Provided that precipitation and water-table

levels were not anomalously low during 2007, these temperatures may have led to an increase in emissions. However, a more detailed study is required to confirm whether this anomaly could be the source of the NH emissions increase required by our analysis. SH wetland sources are largely confined to tropical regions. Therefore, if increased wetland emissions are the source of the SH  $\text{CH}_4$  anomaly, we would expect them to be limited to this region.

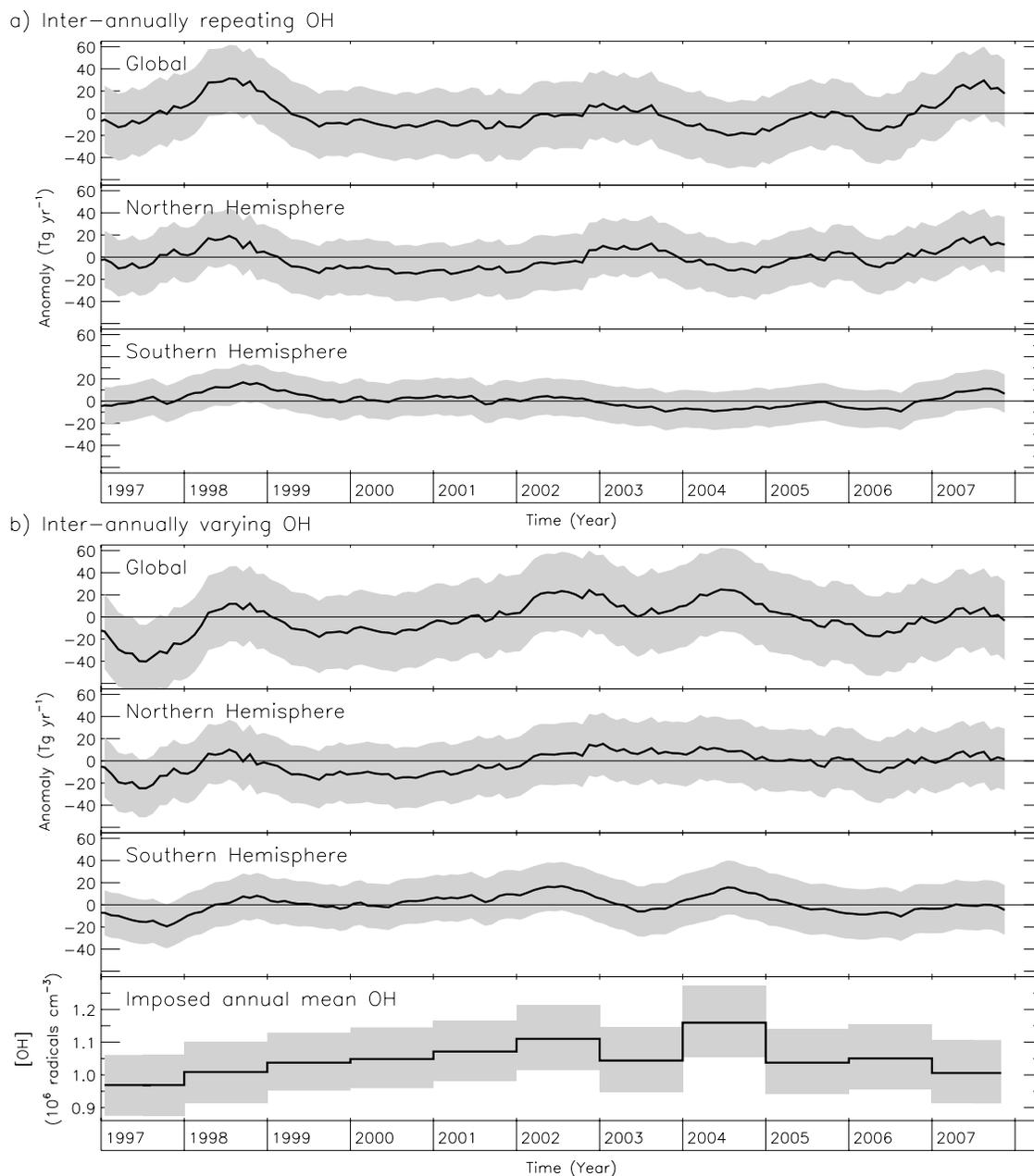
[17] Of the other major sources, we expect biomass burning to have a potentially larger influence on the recent increase than emissions linked to human activity, since these emissions should change less abruptly than the one-year required to explain the rise. If  $\text{CH}_4$  emissions from wildfires have a strong role, we would expect an accompanying increase in atmospheric CO concentration. However, AGAGE and CSIRO surface measurements only find a small CO growth anomaly, limited to the SH during 2006/2007, with no significant growth in the NH. Therefore, whilst a biomass burning contribution to the recent  $\text{CH}_4$  increase may be possible, a corresponding CO rise is not clear.

### 5.2. Inter-annually Varying OH

[18] We find a generally declining OH concentration from 2004 onwards (Figure 2b (bottom)), and a decrease from 2006 to 2007 of  $4 \pm 14\%$ . The reason for this drop is not entirely clear and it is not, in any case, statistically significant. However, because this change falls within typical inter-annual OH variability based on previous studies (e.g., Prinn *et al.* [2005] find a standard deviation in the annual mean of  $\sim 6.5\%$  and Bousquet *et al.* [2005] find values of 8.5% or 3.8% when optimization of  $\text{CH}_3\text{CCl}_3$  emissions are omitted from or included in the inversion respectively), it is useful to examine the effects of such a drop on our emission estimates.

[19] Figure 2b shows the global, NH and SH  $\text{CH}_4$  emission rate anomalies derived using variable annual average OH levels with repeating annual cycles. Figure 2b indicates an increase in emissions between 2006 and 2007 of  $20 \pm 50$  Tg/yr globally, with  $13 \pm 39$  Tg/yr in the NH and  $7 \pm 32$  Tg/yr in the SH. However, compared to the average across the whole period, the 2007 emissions were not unusually high. This is a result of the generally declining OH levels from 2004 inferred by our  $\text{CH}_3\text{CCl}_3$  analysis. Whilst the 2006–2007 emissions increases are, again, poorly constrained, they indicate that if our derived OH levels are correct, the OH decline between 2006 and 2007 substantially reduces the required recent emissions growth and appears to account for much of the SH mole fraction increase. It seems likely, however, that a relatively large NH emission rate rise is still required between these two years.

[20] It should be noted that whilst we have imposed a change in the global OH level, more localized changes may have occurred. For example, it is possible that low OH concentration occurred in only one hemisphere during 2007 compared to 2006, thereby reducing the required  $\text{CH}_4$  emissions increase in that region. However, with our analysis it is not possible to draw statistically significant conclusions about specific year-to-year hemisphere-scale OH changes. Even smaller scale regional effects are also known to take place and will require three-dimensional modeling to resolve. For example, OH radicals can be



**Figure 2.** Annual running mean global, NH and SH emission rate anomaly under (a) inter-annually repeating OH and (b) inter-annually varying OH derived from  $\text{CH}_3\text{CCl}_3$  inversion. Figure 2b (bottom) shows the imposed mass-weighted tropospheric annual mean OH concentration. The x-axis values refer to the center of the 12-month averaging period for all except the lowest panel, for which calendar year average OH levels are shown. The same normalized seasonal variations in OH are used in the inter-annually varying and inter-annually repeating cases. Shaded areas indicate the 1- $\sigma$  error in the derived quantities.

recycled in the presence of nitrogen oxides resulting in generally higher OH concentrations over “polluted” land regions than over the ocean [Lelieveld *et al.*, 2004]. Any changes in the land/ocean OH distribution in recent years cannot be resolved by our model.

## 6. Conclusions

[21] The  $\text{CH}_4$  mole fraction growth rate during 2007 was significantly elevated at all AGAGE and CSIRO sites simultaneously for the first time in almost a decade. If

inter-annually repeating OH levels are assumed, we find that a substantial increase in emissions from both hemispheres was necessary between 2006 and 2007 to fit the observations. OH levels derived from  $\text{CH}_3\text{CCl}_3$  inversion indicate a  $4 \pm 14\%$  decrease in concentration between 2006 and 2007. Whilst this inferred OH drop is not statistically significant, a change of this nature was found to shift the required emissions increase more strongly to the NH and decrease the overall magnitude of the required global total rise in emissions. Given the well known preponderance of

the NH in contributing to global total CH<sub>4</sub> emissions, NH dominance in the required 2006–2007 emissions increase may be reasonable.

[22] **Acknowledgments.** The AGAGE network is supported primarily by NASA grants to MIT and UCSD/SIO with contributions also by NOAA, CSIRO, the Australian BoM, UK DEFRA and the NSF Arctic Research Program. The CSIRO flask network is supported by CSIRO Australia, Australian Bureau of Meteorology, Australian Institute of Marine Science, Australian Antarctic Division, NOAA USA, and the Meteorological Service of Canada.

## References

- Bousquet, P., D. A. Hauglustaine, P. Peylin, C. Carouge, and P. Ciais (2005), Two decades of OH variability as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform, *Atmos. Chem. Phys.*, *5*, 2635–2656.
- Bousquet, P., et al. (2006), Contribution of anthropogenic and natural sources to atmospheric methane variability, *Nature*, *443*, 439–443, doi:10.1038/nature05132.
- Chen, Y., and R. G. Prinn (2006), Estimation of atmospheric methane emissions between 1996 and 2001 using a three-dimensional global chemical transport model, *J. Geophys. Res.*, *111*, D10307, doi:10.1029/2005JD006058.
- Cunnold, D. M., et al. (2002), In situ measurements of atmospheric methane at GAGE/AGAGE sites during 1985–2000 and resulting source inferences, *J. Geophys. Res.*, *107*(D14), 4225, doi:10.1029/2001JD001226.
- Denman, K., et al. (2007), Couplings between changes in the climate system and biogeochemistry, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., chap. 7, pp. 501–587, Cambridge Univ. Press, Cambridge, U. K.
- Dlugokencky, E. J., L. P. Steele, P. M. Lang, and K. A. Masarie (1994), The growth rate and distribution of atmospheric methane, *J. Geophys. Res.*, *99*, 17,021–17,043.
- Dlugokencky, E. J., S. Houweling, L. Bruhwiler, K. A. Masarie, P. M. Lang, J. B. Miller, and P. P. Tans (2003), Atmospheric methane levels off: Temporary pause or a new steady-state?, *Geophys. Res. Lett.*, *30*(19), 1992, doi:10.1029/2003GL018126.
- Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., chap. 2, pp. 131–234, Cambridge Univ. Press, Cambridge, U. K.
- Langenfelds, R. L., R. J. Francey, B. C. Pak, L. P. Steele, J. Lloyd, C. M. Trudinger, and C. E. Allison (2002), Interannual growth rate variations of atmospheric CO<sub>2</sub> and its δ<sup>13</sup>C, H<sub>2</sub>, CH<sub>4</sub>, and CO between 1992 and 1999 linked to biomass burning, *Global Biogeochem. Cycles*, *16*(3), 1048, doi:10.1029/2001GB001466.
- Lelieveld, J., F. J. Dentener, W. Peters, and M. C. Krol (2004), On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere, *Atmos. Chem. Phys.*, *4*, 2337–2344.
- McCulloch, A., and P. M. Midgley (2001), The history of methyl chloroform emissions: 1951–2000, *Atmos. Environ.*, *35*, 5311–5319, doi:10.1016/S1352-2310(01)00306-5.
- National Climatic Data Center (2008), Climate of 2007: Annual report, <http://www.ncdc.noaa.gov/oa/climate/research/2007/ann/global.html>, NOAA, Asheville, N. C.
- O'Doherty, S., et al. (2001), In situ chloroform measurements at Advanced Global Atmospheric Gases Experiment atmospheric research stations from 1994 to 1998, *J. Geophys. Res.*, *106*, 20,429–20,444.
- Prinn, R. G. (2000), Measurement equation for trace chemicals in fluids and solution of its inverse, in *Inverse Methods in Global Biogeochemical Cycles*, *Geophys. Monogr. Ser.*, vol. 114, edited by P. Kasibhatla et al., pp. 3–18, AGU, Washington, D. C.
- Prinn, R. G., et al. (2000), A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, *J. Geophys. Res.*, *105*, 17,751–17,792.
- Prinn, R. G., et al. (2005), Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, *Geophys. Res. Lett.*, *32*, L07809, doi:10.1029/2004GL022228.
- Simmonds, P., A. Manning, R. Derwent, P. Ciais, M. Ramonet, V. Kazan, and D. Ryall (2005), A burning question: Can recent growth rate anomalies in the greenhouse gases be attributed to large-scale biomass burning events?, *Atmos. Environ.*, *39*, 2513–2517, doi:10.1016/j.atmosenv.2005.02.018.
- United Nations Environment Programme (2005), *Production and Consumption of Ozone Depleting Substances Under the Montreal Protocol 1986–2004*, 75 pp., Nairobi. (Available from <http://www.unep.org/ozone/>)
- United Nations Environment Programme (2008), United Nations Environment Programme Ozone Secretariat Data Access Centre, [http://ozone.unep.org/Data\\_Reporting/Data\\_Access/](http://ozone.unep.org/Data_Reporting/Data_Access/), Nairobi.
- Walter, B. P., M. Heimann, and E. Matthews (2001), Modeling modern methane emissions from natural wetlands: 2. Interannual variations 1982–1993, *J. Geophys. Res.*, *106*, 34,207–34,219.

D. Cunnold and H. Wang, School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332–0340, USA.

P. Fraser, P. Krummel, R. Langenfelds, and P. Steele, Centre for Australian Weather and Climate Research, CSIRO Marine and Atmospheric Research, Aspendale, Vic 3195, Australia.

C. Harth, J. Mühle, P. Salameh, and R. Weiss, Scripps Institution of Oceanography, UCSD, La Jolla, CA 92093–0244, USA.

J. Huang, R. Prinn, and M. Rigby, Center for Global Change Science, Massachusetts Institute of Technology, Cambridge, MA 02138, USA. (mrigby@mit.edu)

S. O'Doherty and P. G. Simmonds, School of Chemistry, University of Bristol, Bristol BS8 1TS, UK.