Influence of biomass burning during recent fluctuations in the slow growth of global tropospheric methane

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[1] During the past 15 years the annual growth rate of tropospheric methane (CH$_4$) has shown striking changes over 2–3 year periods, varying from $+1\%$ yr$^{-1}$ to slightly negative values ($-0.2\%$ yr$^{-1}$). These fluctuations are superimposed on an overall slowdown of the CH$_4$ growth rate since the 1980s. Here we use our complementary measurement of other compounds (ethane, tetrachloroethene) to confirm the influence of biomass burning on large global CH$_4$ pulses in 1998 and 2002–2003. Methane growth rate fluctuations also track ENSO indices, most likely via the influence of ENSO activity on large-scale biomass burning. We also report the seventh year of near-zero growth of global CH$_4$ levels (Dec. 1998 – Dec. 2005). The global CH$_4$ mixing ratio was $1772 \pm 1$ ppbv in 2005, and CH$_4$ increases of 118–376 ppbv between 2000–2020 (ten scenarios in the 2001 IPCC report, to levels around 1900+ ppbv by 2020, now appear quite unlikely. Citation: Simpson, I. J., F. S. Rowland, S. Meinardi, and D. R. Blake (2006), Influence of biomass burning during recent fluctuations in the slow growth of global tropospheric methane, Geophys. Res. Lett., 33, L22808, doi:10.1029/2006GL027330.

1. Introduction

[2] Methane (CH$_4$) has been second only to carbon dioxide (CO$_2$) in enhanced climatic forcing from 1750 to the present. Methane also plays critical roles in atmospheric chemistry, for example as an important source of tropospheric ozone (O$_3$), which is also a key greenhouse gas. The global CH$_4$ mixing ratio has more than doubled since the Industrial Revolution to a current level in excess of 1750 ppbv [Simpson et al., 2002; Dlugokencky et al., 2003]. By comparison, although ice core records show that CH$_4$ levels vary with climate, the CH$_4$ maxima during interglacial periods have not exceeded 780 ppbv in a record that now extends to 420,000 BP [Petit et al., 1999]. The main anthropogenic CH$_4$ sources include ruminants, energy use, rice agriculture, biomass burning, and landfills [Ehhalt and Prather, 2001]. Measurements of the amount of $^{14}$CH$_4$ in atmospheric methane indicate a contribution of 65–75% “modern” carbon, implying that CH$_4$ associated with fossil fuels is less significant than sources with modern biological involvement.

[3] Our extensive direct measurements of the global CH$_4$ distribution began in 1978 and showed that the global tropospheric CH$_4$ burden increased by 11% in just ten years (1978–1987), at an average increase of 16 ± 1 ppbv yr$^{-1}$ [Blake and Rowland, 1988]. The CH$_4$ growth rate slowed to 5–10 ppbv yr$^{-1}$ (0.3–0.6% yr$^{-1}$) by the late 1980s and continued to decline into the 1990s, though with striking fluctuations in its annual growth rate [Dlugokencky et al., 1994, 1998; Simpson et al., 2002]. These unpredicted growth rate fluctuations have been linked primarily to non-cyclical events such as the eruption of Mt. Pinatubo in 1991, the Indonesian and boreal wildfires of 1997 and 1998, respectively, and anomalous wetland emissions in 1998 [e.g., Dlugokencky et al., 1996, 2001; Duncan et al., 2003; Wang et al., 2004; Prinn et al., 2005]. [4] In contrast to the variability of the 1990s, the early 2000s have been characterized by a temporary pause in methane’s growth rate [Dlugokencky et al., 2003]. This pause and its implications for future CH$_4$ levels are currently under debate in the literature, in particular whether the near-zero CH$_4$ growth may represent the arrival at a new steady-state [Dlugokencky et al., 1998; Prinn et al., 2005] or whether it is a temporary plateau with no implications for future changes in CH$_4$ levels [Simpson et al., 2002; Dlugokencky et al., 2003]. Here we present the most recent global atmospheric CH$_4$ mixing ratios, discuss future CH$_4$ levels, and, in order to better understand the causes of recent fluctuations in methane’s growth rate, compare its concentration changes to those of two molecules, ethane (C$_2$H$_6$) and tetrachloroethene (C$_2$Cl$_4$), whose atmospheric removal also depends upon reaction with hydroxyl radicals (OH).

2. Experimental

[5] Our global CH$_4$ monitoring program began in 1978 and has been described by Simpson et al. [2002]. Briefly, we collect 60–80 whole air samples at more than 40 remote sites in the Pacific basin (71°N to 47°S) four times a year. Each air sample is collected into a conditioned, evacuated 2-L stainless steel canister equipped with a bellows valve, and is returned to our UC-Irvine laboratory for CH$_4$ analysis using gas chromatography (GC, HP-5890A) with flame ionization detection (FID). The use of primary CH$_4$ calibration standards dating back to late 1977 ensures that our measurements are internally consistent. The measurement accuracy is ±1% and our analytical precision at atmospheric mixing ratios is currently about 1 ppbv. Occasional CH$_4$ samples are discarded because of evidence for nearby source effects.

[6] Although CH$_4$ is measured separately, extensive analysis is also made for more than 100 compounds from the same air canisters. Ethane and C$_2$Cl$_4$ mixing ratios were determined using GC/FID for C$_2$H$_6$ and GC with electron capture detection (HP-6890) and mass spectrometer detection (HP-5793) for C$_2$Cl$_4$. Our calibration scales for these
mixing ratios presented here were obtained
mixing ratio is the average of 4 consecutive
growth rates

[7] The CH₄ mixing ratios presented here were obtained
essentially a Pacific basin surface average, and we infer it to
be globally representative because of the rapid east-west
mixing in the troposphere. One-year running growth rates
use data collected since we began regular seasonal sampling
in Apr. 1983. Our procedures for constructing a global
average from 16 equal-area latitudinal bands have been
described by Simpson et al. [2002, 2004]. Briefly, the global
seasonal CH₄ mixing ratio is the mean of the 16 band
averages, and its uncertainty is the sum of standard errors
(SEs) for each band, added in quadrature, divided by 16.
The annual CH₄ mixing ratio is the average of 4 consecutive
seasonal means, and its uncertainty is the sum of the SEs of
the seasonal means, added in quadrature, divided by 4. The
annual growth rate is the difference between two consecu-
tive annual global CH₄ mixing ratios. Its uncertainty is the
sum of the SEs of the two annual means from which it was
calculated, added in quadrature. The same procedures were
applied to the C₂H₆ and C₂Cl₄ data. The CH₄, C₂H₆ and
C₂Cl₄ data can be accessed at http://cdiac.ornl.gov/trends/
otheratg/Blake/Blake.html [Blake, 2005].

3. Results and Discussion

3.1. General CH₄ Growth Rate Features

[8] In contrast to the upward growth rates through our
first two decades of measurements, the global CH₄ burden
is now in its seventh year of very slow net atmospheric
growth (Dec. 1998 to Dec. 2005, Figure 1). During this time
the rolling annual CH₄ growth rate ranged between −3.8 ±
1.2 and 4.9 ± 1.2 ppbv yr⁻¹ (−0.2 to +0.3% yr⁻¹), and the
global CH₄ mixing ratio in 2005 (1772 ± 1 ppbv) was
larger than that measured in 1998 (1767 ± 1 ppbv) by about
5 ppbv. This is a much slower increase in the global CH₄
burden than was recorded in previous seven-year incre-
ments: 1998 levels were 42 ppbv larger than those in 1991
(1725 ± 1 ppbv), and 1991 levels were 96 ppbv larger than
those in 1984 (1629 ± 1 ppbv). Our global record begins in
1978 at 1515 ppbv.

[9] Striking changes in the annual CH₄ growth rate over
periods of 2–3 years – varying from +1% per year to
slightly negative values (−0.2%) – have been superim-
posed on its general slowdown (Figure 1b). Based on
rolling averages our group has recorded four such growth
rate peaks and troughs, with maxima occurring during
1991 (16.5 ± 0.9 ppbv), 1995 (10.5 ± 0.8 ppbv), 1998 (15.9 ±
0.7 ppbv) and 2003 (4.9 ± 1.2 ppbv), and minima during 1993
(1.1 ± 0.8 ppbv), 1996 (−2.3 ± 0.8 ppbv), 2000 (−2.7 ± 0.8
ppbv) and 2004 (−3.8 ± 1.2 ppbv).

3.2. Biomass Burning, ENSO, and CH₄ Growth Rate
Fluctuations

[10] To understand better the causes of these fluctuations,
we have compared the global CH₄ growth rate with con-
centration changes for two other OH-controlled species that
we measure, C₂H₆ and C₂Cl₄, for which data are available
from 1996–2004. While CH₄ is also emitted by ethane’s
two major sources, natural gas use and biomass burning
[Ehhalt and Prather, 2001], the sole major source of C₂Cl₄
is industrial solvent emissions [Simpson et al., 2004, and
references therein]. Because of their very different atmo-
sphere lifetimes, CH₄ (8–9 years) and C₂H₆ and C₂Cl₄
(several months) have different characteristics for year-to-
year data comparisons. Briefly put, the atmosphere has a
strong memory for last year’s CH₄ because most of it is still
there. In contrast, it has almost no memory of last year’s
C₂H₆ or C₂Cl₄ because very little has survived the inter-
vening summer. Accordingly, we plot CH₄ growth and
C₂H₆ and C₂Cl₄ atmospheric mixing ratios in Figure 2. In
principle, an observed increase in CH₄ could represent
either an increase in emissions or a decrease in the strength
of the atmospheric OH sink. Parallel mixing ratio changes
by CH₄ and C₂H₆, as in Figure 2a, can be expected for a
change in the OH sink, or by parallel emission patterns such
as those observed during biomass burning. Uncorrelated
mixing ratio changes for CH₄ and C₂Cl₄, as in Figure 2b,
suggest that the system was not sink-driven, but rather was
influenced by increased emission from source(s) that CH₄

Figure 1. (a) Global tropospheric CH₄ mixing ratios (ppbv). Triangles: global seasonal averages. Circles: one-
year running averages fit by an interpolated curve fit (a curve that passes through the data points and matches
the slope at those points). Each average is plotted at the
temporal mid-point of the four seasons from which the
average was calculated (e.g. Nov. 1, 2004 for Jun. 2004
to Mar. 2005). (b) One-year running global CH₄ growth rates
(ppbv yr⁻¹). Each growth rate is plotted at the temporal
mid-point of the second year from which the growth rates
were calculated (e.g. Nov. 1, 2004 for Jun. 2004 to Mar. 2005
and C$_2$H$_6$ have in common. Moreover, when the long-term decline in the global atmospheric burden of C$_2$Cl$_4$ between 1996–2004 is taken into account [Simpson et al., 2004], an anticorrelation between CH$_4$ and C$_2$Cl$_4$ throughout most of the measurement period becomes apparent (not shown). A detailed comparison between the trend in annual C$_2$Cl$_4$ mixing ratios and in industrial C$_2$Cl$_4$ emissions suggests that it is very unlikely that OH changes were masked by C$_2$Cl$_4$ emission variations (see Simpson et al. [2004] for details).

[11] Consistent with source-driven influences on recent CH$_4$ and C$_2$H$_6$ fluctuations, the 1998 and 2002–2003 CH$_4$ and C$_2$H$_6$ peaks coincide with major biomass burning in Indonesia in autumn 1997, in Russia in 1998 (with an October maximum), and in Russia during summer/autumn of 2002 and 2003 [Duncan et al., 2003; Yurganov et al., 2004, 2005; Kasischke et al., 2005]. For example, the three largest seasonal C$_2$H$_6$ differences in our record occur in Dec. 1997 (165 pptv higher than the Dec. 1996 average), Sep. 1998 (160 pptv) and Sep. 2002 (115 pptv), compared to an average seasonal difference ($\pm 1\sigma$) of $-9 \pm 66$ pptv from 1999–2001. The highest seasonal CH$_4$ differences occur in Dec. 1998, Sep. 1998 and Mar. 1998 (21, 16 and 14 ppbv, respectively), compared to an average seasonal difference ($\pm 1\sigma$) of $-0.3 \pm 3.0$ pptv from 1999–2001. In addition, annual CH$_4$ and C$_2$H$_6$ mixing ratios show an earlier peak in the tropics (30°N–30°S) than in the high northern hemisphere (HNH, 30–90°N) that is consistent with influence from the 1997 tropical Indonesian fires followed by the 1998 boreal fires, with an additional perturbation in the HNH that reached a maximum with the boreal fires of 2003 (Figure 3). Simmonds et al. [2005] have also linked CH$_4$ growth rate anomalies in 1998–1999 and 2002–2003 at Mace Head, Ireland (53°N) to large-scale biomass burning.

[12] The hydrocarbon growth rate fluctuations also show a similar pattern to the Oceanic Niño Index (ONI) – a measure of the oceanic temperature anomaly during El Niño and La Niña events – in the Niño 3.4 region (120°–170°W; 5°N–5°S) (Figures 4a and 4b). Similarly, Jones and Cox [2005] found a statistical link between El Niño activity and CO$_2$ growth rate anomalies, though they state that the 2002 and 2003 CO$_2$ growth rates cannot be fully explained on this basis. Prinn et al. [2001] have likewise noted the sensitivity of annual CH$_3$CCl$_3$ cycles to the El Niño-Southern Oscillation (ENSO), based on measurements in Samoa. Although they state that global OH is lower during El Niño years, we do not see this reflected in C$_2$Cl$_4$, which, unlike CH$_4$ and C$_2$H$_6$, does not clearly follow the ONI trend (Figure 4c) even when it has been detrended for its long-term decline (not shown). Instead, we suggest that drought conditions during El Niño years lead to increased fire activity and therefore increased release of hydrocarbons such as CH$_4$ and C$_2$H$_6$ from biomass burning. In support of this, Carmona-Morena et al. [2005] have recently shown the high likelihood that ENSO events and fire activity are connected, and Baltzer et al. [2005] found significant

![Figure 2](image_url) Figure 2. One-year running global growth rates of (a) CH$_4$ (ppbv yr$^{-1}$; circles) and C$_2$H$_6$ (pptv; triangles), and (b) CH$_4$ (pptv yr$^{-1}$; circles) and C$_2$Cl$_4$ (pptv; triangles) using data collected between 1996–2004. Averages and growth rates are plotted as described in Figure 1.

![Figure 3](image_url) Figure 3. One-year running average mixing ratios of (a) CH$_4$ (ppmv) and (b) C$_2$H$_6$ (pptv) in the high northern hemisphere (HNH, 30–90°N; circles), tropics (30°N–30°S; triangles) and high southern hemisphere (HSH, 30–90°S; squares). Averages are plotted as described in Figure 1.
The recent pause in the CH₄ growth rate, and its implications for future CH₄ levels, is currently under debate (Section 1). The global CH₄ mixing ratio was 1772 ± 1 ppbv in 2005, and CH₄ increases (predicted by the 2001 IPCC report [Ehhalt and Prather, 2001]) of 67–111 ppbv between 2000–2010, to levels around 1850 ppbv by 2010 [Ehhalt and Prather, 2001], now appear extremely unlikely. Likewise, predicted CH₄ increases of 118–376 ppbv between 2000–2020, to levels of 1900 ppbv or more by 2020, also appear unlikely, and opportunities exist for still further reductions. With its easily perturbed budget, reducing CH₄ emissions is highly desirable because it would help offset radiative forcing caused by CO₂ growth and would benefit global air quality by decreasing background tropospheric O₃ levels [Hansen et al., 2000; Fiore et al., 2002; Dentener et al., 2004]. However, Wang et al. [2004] suggest that there is significant potential for CH₄ growth, for example as the economies of Eastern Europe and the former Soviet Union (FSU) recover and as energy consumption increases. Natural CH₄ emissions may also increase, possibly substantially, in response to increased CO₂ [Shindell et al., 2004] although we have not yet seen evidence for such a large new CH₄ source. Overall we suggest that there is no reason to believe that CH₄ growth will necessarily remain at near-zero levels in the future.

**4. Conclusions**

[15] The global growth rate of atmospheric CH₄ has been near-zero for the past seven years, averaging 0.7 ± 2.6 ppbv yr⁻¹. This temporary pause follows an overall slowdown of methane’s growth rate in the 1980s, and a series of striking and unpredicted growth rate fluctuations in the 1990s. Barring an exceptional increase in emissions, the predicted CH₄ concentrations for 2100 now fall well below the midpoint of the 2001 IPCC report estimates [Ehhalt and Prather, 2001], and opportunities exist for still further reductions.

[16] The contribution of biomass burning emissions to the most recent growth rate fluctuations of CH₄ and C₂H₆ is...
clear in our data set. A measurable increase in hydrocarbons is coincident with heavy biomass burning in Indonesia in 1997 and in Russia in 1998. Prolonged, positive CH4 and C2H6 growth from 2000–2003 (with a peak in 2003) is consistent with boreal fire emissions that occurred during this time. The CH4 and C2H6 growth rate anomalies also track ENSO activity, which is believed to be linked to variability in large-scale biomass burning activity. Although C2H4 is also an OH-controlled species it did not track the hydrocarbon growth rate fluctuations. This suggests that ENSO activity and hydrocarbon fluctuations are connected via perturbations to hydrocarbon sources, rather than perturbations to OH or to meteorological fields.

[17] Although the CH4 budget is currently in approximate balance, there is no reason to believe that it will necessarily remain so in the future, given the possibilities that CH4 levels may increase as a result of increased natural gas and energy use, climate change feedbacks, and/or a decrease in global OH, or conversely that CH4 levels may decrease if various CH4 emission mitigation strategies are implemented as an offset to radiative forcing caused by CO2 growth. In particular, the reduction of fossil fuel leakage has promise for mitigating greenhouse gas contributions of CH4 and tropospheric O3. Because the leaking fossil fuels have high value in the market, these mitigation steps can in some cases even be economically favorable. Our regional survey of the southwestern United States [Katzenstein et al., 2003] showed non-urban CH4 concentrations elevated as much as 20% above background levels, and other light alkanes as much as 100 times background. The study also showed that reactive nitrogen compounds were present at levels sufficient to produce tropospheric O3, another greenhouse gas, in these non-urban locations. Such regional surveys are comparatively rare, and extensions to other areas are likely to disclose other favorable areas for mitigation of CH4 greenhouse gas contributions.

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References

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