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

Isobel J. Simpson,[1] F. Sherwood Rowland,[1] Simone Meinardi,[1] and Donald R. Blake[1]

[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 ± 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_2H_6) and tetrachloroethene (C_2Cl_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_2Cl_4 mixing ratios were determined using GC/FID for C_2H_6 and GC with electron capture detection (HP-6890) and mass spectrometer detection (HP-5793) for C_2Cl_4. Our calibration scales for these

[1]Department of Chemistry, University of California, Irvine, California, USA.

compounds date back to the mid-to-late 1980s and are regularly updated and recalibrated with newly-prepared primary standards [*Wang et al.*, 1995; *Colman et al.*, 2001]. The measurement precision is 2% for C_2H_6, and the larger of 1% and 0.05 pptv for C_2Cl_4. The accuracy is 5% for C_2H_6, and 3% at 40 pptv for C_2Cl_4, increasing to 8% at 2 pptv.

[7] The CH$_4$ mixing ratios presented here were obtained between Jan. 1978 and Dec. 2005. Our measurement is 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 CH$_4$ 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 CH$_4$ 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 consecutive annual global CH$_4$ 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 C_2H_6 and C_2Cl_4 data. The CH$_4$, C_2H_6 and C_2Cl_4 data can be accessed at http://cdiac.ornl.gov/trends/otheratg/blake/blake.html [*Blake*, 2005].

3. Results and Discussion

3.1. General CH$_4$ Growth Rate Features

[8] In contrast to the upward growth rates through our first two decades of measurements, the global CH$_4$ 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 CH$_4$ 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 CH$_4$ 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 CH$_4$ burden than was recorded in previous seven-year increments: 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 CH$_4$ growth rate over periods of 2–3 years – varying from $+1\%$ per year to slightly negative values (-0.2%) – have been superimposed 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 pptv), 1995 (10.5 ± 0.8 pptv), 1998 (15.9 ± 0.7 pptv) and 2003 (4.9 ± 1.2 pptv), and minima during 1993 (1.1 ± 0.8 pptv), 1996 (-2.3 ± 0.8 pptv), 2000 (-2.7 ± 0.8 pptv) and 2004 (-3.8 ± 1.2 pptv).

3.2. Biomass Burning, ENSO, and CH$_4$ Growth Rate Fluctuations

[10] To understand better the causes of these fluctuations, we have compared the global CH$_4$ growth rate with concentration changes for two other OH-controlled species that we measure, C_2H_6 and C_2Cl_4, for which data are available
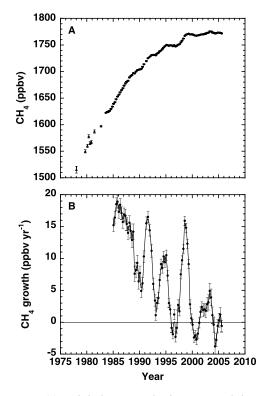

Figure 1. (a) Global tropospheric CH$_4$ mixing ratios (ppbv). Triangles: global seasonal averages. Circles: one-year running averages fit by an interpolated curve fit (a curve that passes through the data points and matches the slope at those points). Each average is plotted at the temporal mid-point 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 CH$_4$ 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 minus Jun. 2003 to Mar. 2004).

from 1996–2004. While CH$_4$ is also emitted by ethane's two major sources, natural gas use and biomass burning [*Ehhalt and Prather*, 2001], the sole major source of C_2Cl_4 is industrial solvent emissions [*Simpson et al.*, 2004, and references therein]. Because of their very different atmosphere lifetimes, CH$_4$ (8–9 years) and C_2H_6 and C_2Cl_4 (several months) have different characteristics for year-to-year data comparisons. Briefly put, the atmosphere has a strong memory for last year's CH$_4$ because most of it is still there. In contrast, it has almost no memory of last year's C_2H_6 or C_2Cl_4 because very little has survived the intervening summer. Accordingly, we plot CH$_4$ growth and C_2H_6 and C_2Cl_4 atmospheric mixing ratios in Figure 2. In principle, an observed increase in CH$_4$ could represent either an increase in emissions or a decrease in the strength of the atmospheric OH sink. Parallel mixing ratio changes by CH$_4$ and C_2H_6, 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 CH$_4$ and C_2Cl_4, as in Figure 2b, suggest that the system was not sink-driven, but rather was influenced by increased emission from source(s) that CH$_4$
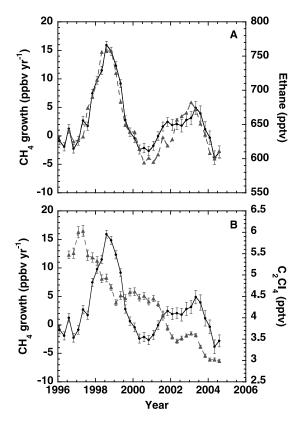
Figure 2. One-year running global growth rates of (a) CH_4 (ppbv yr^{-1}; circles) and C_2H_6 (pptv; triangles), and (b) CH_4 (pptv yr^{-1}; circles) and C_2Cl_4 (pptv; triangles) using data collected between 1996–2004. Averages and growth rates are plotted as described in Figure 1.

and C_2H_6 have in common. Moreover, when the long-term decline in the global atmospheric burden of C_2Cl_4 between 1996–2004 is taken into account [*Simpson et al.*, 2004], an anticorrelation between CH_4 and C_2Cl_4 throughout most of the measurement period becomes apparent (not shown). A detailed comparison between the trend in annual C_2Cl_4 mixing ratios and in industrial C_2Cl_4 emissions suggests that it is very unlikely that OH changes were masked by C_2Cl_4 emission variations (see *Simpson et al.* [2004] for details).

[11] Consistent with source-driven influences on recent CH_4 and C_2H_6 fluctuations, the 1998 and 2002–2003 CH_4 and C_2H_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_2H_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 ± 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 ± 3.0 pptv from 1999–2001. In addition, annual CH_4 and C_2H_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_3CCl_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_2Cl_4, which, unlike CH_4 and C_2H_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_2H_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
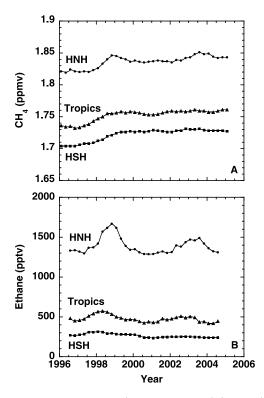

Figure 3. One-year running average mixing ratios of (a) CH_4 (ppmv) and (b) C_2H_6 (pptv) in the high northern hemisphere (HNH, 30–90°N; circles), tropics (30°N–30°S; triangles) and high southern hemisphere (HSH, 30–90°S; squares). Averages are plotted as described in Figure 1.
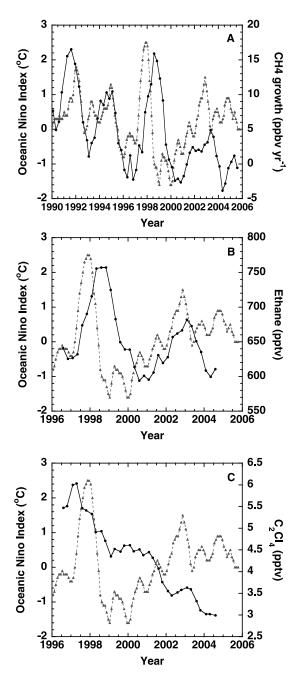
Figure 4. Time series of the Oceanic Niño Index (dashed lines) plotted with (a) one-year running global growth rates of CH_4 (ppbv yr^{-1}), and one-year running averages of (b) C_2H_6 (pptv) and (c) C_2Cl_4 (pptv) (solid lines). Averages and growth rates are plotted as described in Figure 1.

relationships between the interannual forest fire variability in Russia and climate indices including the Arctic Oscillation and the Niño 4 index ($150°E-150°W; 5°N-5°S$).

[13] To determine the extent to which biomass burning explains the most recent CH_4 and C_2H_6 growth rate anomalies in 2002–2003, we compared the measured growth rates with estimated CH_4 and C_2H_6 emissions from the fires. Based on measured CO anomalies in 2002 and 2003 [*Yurganov et al.*, 2005] and pyrogenic emission factors and their uncertainties for extratropical forests [*Andreae*

and Merlet, 2001], we estimate that the 2002 and 2003 fires respectively released 4 ± 2 and 6 ± 3 Tg CH_4, and 0.5 ± 0.2 and 0.7 ± 0.3 Tg C_2H_6. Relative to average growth measured in 1999–2001, our CH_4 growth rates for 2002 and 2003 correspond to a total emission anomaly of 14 ± 5 Tg CH_4, compared to the above estimate of 10 ± 4 Tg. Bearing in mind the uncertainties in these estimates, it appears that biomass burning explains most of the anomalous CH_4 growth in 2002–2003. Similarly, relative to growth in 1999–2001, our measured C_2H_6 growth rate anomaly for 2002–2003 is equivalent to roughly 1.0 ± 1.7 Tg C_2H_6, compared to the above estimate of 1.2 ± 0.4 Tg. The relatively large uncertainty in the estimate of 1.0 ± 1.7 Tg (as compared to CH_4) is due to ethane's shorter atmospheric lifetime and higher atmospheric variability. Again bearing in mind the uncertainties, the 2002–2003 fires appear to have made a major contribution to the measured C_2H_6 anomaly, in agreement with a strong biomass burning influence on the CH_4 growth rate in 2002–2003.

3.3. Future CH4 Levels

[14] The recent pause in the CH_4 growth rate, and its implications for future CH_4 levels, is currently under debate (Section 1). The global CH_4 mixing ratio was 1772 ± 1 ppbv in 2005, and CH_4 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 CH_4 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 CH_4 emissions is highly desirable because it would help offset radiative forcing caused by CO_2 growth and would benefit global air quality by decreasing background tropospheric O_3 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 CH_4 growth, for example as the economies of Eastern Europe and the former Soviet Union (FSU) recover and as energy consumption increases. Natural CH_4 emissions may also increase, possibly substantially, in response to increased CO_2 [*Shindell et al.*, 2004] although we have not yet seen evidence for such a large new CH_4 source. Overall we suggest that there is no reason to believe that CH_4 growth will necessarily remain at near-zero levels in the future.

4. Conclusions

[15] The global growth rate of atmospheric CH_4 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 CH_4 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 CH_4 and C_2H_6 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 CH$_4$ and C$_2$H$_6$ growth from 2000–2003 (with a peak in 2003) is consistent with boreal fire emissions that occurred during this time. The CH$_4$ and C$_2$H$_6$ growth rate anomalies also track ENSO activity, which is believed to be linked to variability in large-scale biomass burning activity. Although C$_2$Cl$_4$ 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 CH$_4$ 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 CH$_4$ 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 CH$_4$ levels may decrease if various CH$_4$ emission mitigation strategies are implemented as an offset to radiative forcing caused by CO$_2$ growth. In particular, the reduction of fossil fuel leakage has promise for mitigating greenhouse gas contributions of CH$_4$ and tropospheric O$_3$. 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 CH$_4$ 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 O$_3$, 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 CH$_4$ greenhouse gas contributions.

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D. R. Blake, S. Meinardi, F. S. Rowland, and I. J. Simpson, Department of Chemistry, University of California, Irvine, CA 92697-2025, USA. (isimpson@uci.edu)