



## Impact of meteorology and emissions on methane trends, 1990–2004

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[1] Over the past century, atmospheric methane (CH<sub>4</sub>) rose dramatically before leveling off in the late 1990s. The processes controlling this trend are poorly understood, limiting confidence in projections of future CH<sub>4</sub>. The MOZART-2 global tropospheric chemistry model qualitatively captures the observed CH<sub>4</sub> trend (increasing in the early 1990s and then leveling off) with constant emissions. From 1991–1995 to 2000–2004, the CH<sub>4</sub> lifetime versus tropospheric OH decreases by 1.6%, reflecting increases in OH and temperature. The rise in OH stems from an increase in lightning NO<sub>x</sub> as parameterized in the model. A simulation including annually varying anthropogenic and wetland CH<sub>4</sub> emissions, as well as the changes in meteorology, best reproduces the observed CH<sub>4</sub> distribution, trend, and seasonal cycles. Projections of future CH<sub>4</sub> abundances should consider climate-driven changes in CH<sub>4</sub> sources and sinks. **Citation:** Fiore, A. M., L. W. Horowitz, E. J. Dlugokencky, and J. J. West (2006), Impact of meteorology and emissions on methane trends, 1990–2004, *Geophys. Res. Lett.*, 33, L12809, doi:10.1029/2006GL026199.

### 1. Introduction

[2] Atmospheric methane (CH<sub>4</sub>) increased during the past century until 1998, with a general decline in the growth rate in recent decades [e.g., Steele *et al.*, 1992]. Since 1998, CH<sub>4</sub> abundances have been roughly constant, suggesting that CH<sub>4</sub> may have reached a steady state [Dlugokencky *et al.*, 2003]. The overall atmospheric CH<sub>4</sub> lifetime is 8–9 years [Prather *et al.*, 2001], reflecting a balance between diverse sources (wetlands, ruminants, energy, rice agriculture, landfills, wastewater, biomass burning, oceans, and termites) and the dominant CH<sub>4</sub> loss pathway, reaction with the hydroxyl radical (OH) in the troposphere. Recent work shows that reducing CH<sub>4</sub> emissions is a viable low-cost strategy to improve ozone air quality while slowing greenhouse warming [West and Fiore, 2005]. Our poor understanding of the processes governing the methane trend, however, limits confidence in quantitative assessments of the efficacy of methane emission controls.

[3] Prior studies with 3-D chemical transport models (CTMs) attribute much of the observed decrease in the CH<sub>4</sub> growth rate to increases in global mean OH, but differ in their explanations for the OH changes, implicating: increases in tropical tropospheric water vapor from

1979–1993 [Dentener *et al.*, 2003]; increasing NO<sub>x</sub> emissions, particularly from Southeast Asia from 1980–1996 [Karlsdóttir and Isaksen, 2000]; and trends in photolysis rates (associated with overhead ozone columns) from 1988–1997 [Wang *et al.*, 2004]. None of these studies considered the period after 1998 when observed CH<sub>4</sub> leveled off.

[4] We examine here whether existing bottom-up estimates of anthropogenic and biogenic CH<sub>4</sub> emissions are consistent with surface CH<sub>4</sub> observations from 1990 to 2004. To our knowledge, this study is the first forward modeling analysis that fully includes feedbacks between CH<sub>4</sub> and the hydroxyl radical [Prather *et al.*, 2001]. We find that recent CH<sub>4</sub> emission estimates improve the simulation of CH<sub>4</sub>, but that the large-scale trend from 1990 to 2004 is mainly controlled by small meteorologically driven changes in the CH<sub>4</sub> sink.

### 2. Methane Simulations

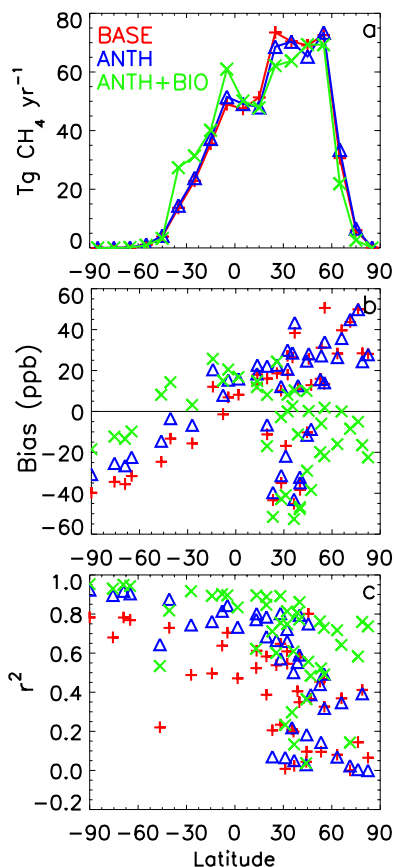
[5] We conduct transient simulations for 1990–2004 with the MOZART-2 global model of tropospheric chemistry [Horowitz *et al.*, 2003] driven by NCEP meteorological fields, at 1.9° × 1.9° horizontal resolution with 28 vertical levels. Model parameterizations based on the meteorological fields include lightning NO<sub>x</sub> (tied to convection) and stratosphere-troposphere exchange of ozone (stratospheric ozone concentrations are relaxed to observed climatologies) [Horowitz *et al.*, 2003], both of which affect tropospheric OH. We use three sets of CH<sub>4</sub> emission estimates: (i) constant 1990 emissions (BASE), (ii) BASE except for annually increasing anthropogenic emissions (ANTH), and (iii) ANTH except for annually varying biogenic emissions (ANTH+BIO). Emissions for species other than CH<sub>4</sub> are from Horowitz *et al.* [2003] and are held constant in all simulations. In the BASE simulation, CH<sub>4</sub> emissions are 261 Tg yr<sup>-1</sup> from EDGAR 2.0 anthropogenic sources (energy use, landfills, wastewater, rice, ruminants) and 72 Tg yr<sup>-1</sup> from biomass burning [Horowitz *et al.*, 2003] (see auxiliary material<sup>1</sup> Table S1 for details). We uniformly increase the global wetland emissions from Horowitz *et al.* [2003] by 40%, to 204 Tg yr<sup>-1</sup>, to reflect recent estimates [Wang *et al.*, 2004]. Figure 1a (red line) shows the latitudinal distribution of the BASE emissions.

[6] The ANTH simulation uses EDGAR 3.2 anthropogenic CH<sub>4</sub> emission estimates for 1990 and 1995 [Olivier and Berndt, 2001], and “FAST-TRACK” (FT2000) for 2000 [Olivier *et al.*, 2005]. Emissions for intermediate years are obtained by linear interpolation, with emissions for 2001–2004 held at 2000 values. Since the 1990 EDGAR 3.2 CH<sub>4</sub> emissions are lower than EDGAR 2.0, we augment

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**Figure 1.** (a) Latitudinal distribution of 1990 CH<sub>4</sub> emissions in the MOZART-2 model within 10° bands, (b) mean 1990–2004 bias (model - observed) in dry surface air (nmol mol<sup>-1</sup>, abbreviated ppb) and (c) correlation coefficient of the MOZART-2 model versus surface observations for each month in 1990–2004, for BASE (red plus signs), ANTH (blue triangles), and ANTH+BIO (green crosses). See auxiliary material (Tables S1 and S2) for details.

the wetland emissions by 20 Tg yr<sup>-1</sup> to maintain the same 1990 total as in BASE. Global anthropogenic CH<sub>4</sub> emissions are nearly constant between 1990 and 1995, but increase by 10 Tg in 2000 (Table S1), with a redistribution toward the tropics over the decade.

[7] The ANTH+BIO simulation uses inverse model estimates of wetland emissions from Wang *et al.* [2004], based on the spatial and seasonal distributions of Matthews and Fung [1987]. Wang *et al.* [2004] conducted their inversion for 1994 and then extrapolated the resulting emissions to 1988–1998 based on temperature and precipitation [Walter *et al.*, 2001]. We maintain the relative source strengths (from tundra, bogs, and swamps) from Wang *et al.* [2004], but constrain the mean 1990–1998 wetland emissions to 224 Tg CH<sub>4</sub> yr<sup>-1</sup>, as in the ANTH simulation. Wetland emissions in ANTH+BIO shift from the northern to southern hemisphere relative to BASE and ANTH (Figure 1a). The annual wetland emissions for 1990 to 1998 are 227, 229, 205, 211, 213, 218, 213, 230, and 266 Tg CH<sub>4</sub> yr<sup>-1</sup>; we apply the 1990–1998 mean in 1999–2004. The unusually large 1998 wetland emissions are associated with above-normal temperatures (globally) and precipitation

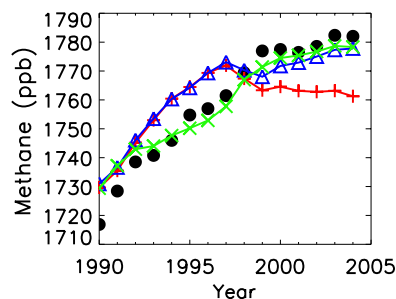
(southern tropics and north of 30°N) [Dlugokencky *et al.*, 2001; Mikaloff Fletcher *et al.*, 2004].

### 3. Atmospheric CH<sub>4</sub> Trend and Distribution

[8] We evaluate our simulations with monthly mean measurements from 42 background sampling sites in the globally distributed NOAA network (Table S2). Air is sampled at these sites in duplicate, analyzed by gas chromatography with flame ionization detection at 0.1% relative precision, and CH<sub>4</sub> abundances are reported on the NOAA04 CH<sub>4</sub> standard scale [Dlugokencky *et al.*, 1994, 2005].

[9] The BASE simulation qualitatively reproduces the observed CH<sub>4</sub> rise in the early 1990s, along with the flattening in the late 1990s (Figure 2). Since emissions are held constant in this simulation, the CH<sub>4</sub> decrease after 1998 implies an increase in the CH<sub>4</sub> sink. The lack of interannual changes in emissions of CH<sub>4</sub> or other species, or in overhead O<sub>3</sub> columns, may contribute to the systematic CH<sub>4</sub> overestimate before 1998 and the underestimate afterward. Major shortcomings of the BASE simulation include a 50% overestimate of the mean 1990–2004 gradient from the South Pole to Alert (201 ppb vs. 133 ppb observed) and a poor simulation of the seasonal cycles at northern hemispheric sites (Figures 1b and 1c, Figure S1, Table S2). The BASE model captures the observed CH<sub>4</sub> seasonality in the southern hemisphere (Figure 1c and Figure S1).

[10] The global mean abundances after 1998 increase in both ANTH and ANTH+BIO relative to the BASE simulation, with ANTH+BIO best matching the 1992–1997 observations (Figure 2). While ANTH slightly improves the negative bias in the southern hemisphere, ANTH+BIO best reproduces the observed interhemispheric gradient (Figure 1b) and captures the seasonal cycles at high northern latitude sites (Figures 1c and Figure S1), indicating that the CH<sub>4</sub> abundance and seasonal cycle at high northern latitudes is sensitive to the wetland source, as suggested by Houweling *et al.* [2000]. The large negative biases (>35 ppb) and poor correlations (r<sup>2</sup> < 0.4) at continental and coastal sites mainly between 30°N and 50°N in ANTH+BIO



**Figure 2.** Weighted global mean annual surface CH<sub>4</sub> in dry air sampled at 42 NOAA sites where a minimum of 8 years were available (see Table S2 for details). The global means are calculated consistently by averaging within latitudinal bands 30–90S, 30–0S, 0–30N, 30–60N, 60–90N and then weighting by area, for both the observations (black circles) and the MOZART-2 simulations: BASE (red plus signs), ANTH (blue triangles), and ANTH+BIO (green crosses).

suggest an inadequate resolution of regional sources in our global model [Houweling *et al.*, 2000], as these biases are greater than at sites sampling marine air in the same latitude band (Figures 1b and 1c, Table S2). In the southern tropics (15°S–2°N), the BASE CH<sub>4</sub> is typically within 10 ppb of observed, while ANTH+BIO exhibits >15 ppb positive biases, implying that the BASE southern tropical wetland source is more accurate. The overall success of the ANTH+BIO simulation suggests that the upper estimate of the recently proposed aerobic CH<sub>4</sub> source of 62–236 Tg yr<sup>-1</sup> from plants [Keppler *et al.*, 2006] is too high, although the lower range could be accommodated if the wetland source were much lower than assumed here.

#### 4. Meteorologically-Driven Changes in the CH<sub>4</sub> Sink

[11] While the global annual mean CH<sub>4</sub> abundances are best reproduced by ANTH+BIO, the BASE model simulates the observed increase for 1990–1997 (slope = 6.3 ppb yr<sup>-1</sup> in BASE vs. 6.1 ppb yr<sup>-1</sup> observed) and the flattening afterward (Figure 2). Since the BASE sources and sinks are not initially in balance, CH<sub>4</sub> abundances should increase toward a steady state. In order to examine whether meteorologically driven changes also contribute to the CH<sub>4</sub> trend, we analyze the BASE simulation where CH<sub>4</sub> emissions are held constant. The annual mean CH<sub>4</sub> lifetime against loss by tropospheric OH ( $\tau_{\text{OH}}$ ) varies with temperature and OH ( $\tau_{\text{OH}} = B/L$ ; B is the annual mean atmospheric burden;  $L = \sum k_{\text{OH}}[\text{OH}][\text{CH}_4]$ , the annual global loss by OH integrated over the troposphere). The strong temperature sensitivity of the rate constant  $k_{\text{OH}}$  ( $\sim 2\% \text{ K}^{-1}$ ) restricts the majority of the CH<sub>4</sub> sink (88%) to below 500 hPa, with 75% of this lower tropospheric loss in the tropics (30°N–30°S).

[12] In our BASE simulation, the mean annual  $\tau_{\text{OH}}$  decreases from 10.40 yr in 1991–1995 to 10.23 yr in 2000–2004 (Figure S2). By re-calculating  $\tau_{\text{OH}}$ , varying only OH or temperature while fixing the other variable at its 1990–2004 monthly mean values, we find that increases in temperature (+0.3 K in the global lower troposphere) and in OH (+1.2%, airmass-weighted below the 150 ppb O<sub>3</sub> chemical tropopause in 1990) contribute 35% (0.06 yr) and 65% (0.11 yr), respectively, to the total decrease in  $\tau_{\text{OH}}$  from 1991–1995 to 2000–2004. We estimate that the rise in CH<sub>4</sub> itself from 1991–1995 to 2000–2004 in the BASE simulation should cause OH concentrations to decrease by 0.2% [Prather *et al.*, 2001], implying a net meteorologically driven OH increase of 1.4% (1.2 + 0.2%) during this period. Trends in OH are highly uncertain, but estimates based on methyl chloroform observations suggest an increase of 0.2% yr<sup>-1</sup> over the past 25 years [Prinn *et al.*, 2005], roughly consistent with our results.

[13] We next seek to identify the cause of the OH increase in the BASE model, which includes meteorological variability but neglects interannual variability in aerosols, overhead O<sub>3</sub> columns, and surface emissions. In the BASE simulation, the annual mean 1991–2004 tropospheric OH (airmass-weighted below the chemical tropopause in 1990) correlates poorly with O<sub>3</sub> photolysis rates ( $r^2 = 0.0$ ), and more strongly with specific humidity ( $r^2 = 0.7$ ), but that correlation weakens ( $r^2 = 0.3$ ) if we neglect 1998 when

humidity was anomalously high. Strong correlations exist between annual mean OH and the tropospheric O<sub>3</sub> burden ( $r^2 = 0.6$ ), lightning NO<sub>x</sub> ( $r^2 = 0.7$ ), and the net flux of stratospheric O<sub>3</sub> to the troposphere ( $r^2 = 0.5$ ); these correlations strengthen when we neglect 1998. From 1991–1995 to 2000–2004, there are increases in the tropospheric O<sub>3</sub> burden (363 to 376 Tg), lightning NO<sub>x</sub> (2.4 to 2.7 Tg N yr<sup>-1</sup>), and the net influx of O<sub>3</sub> from the stratosphere (725 to 866 Tg O<sub>3</sub> yr<sup>-1</sup>), while specific humidity in the lower troposphere changes little (4.26 to 4.28 g H<sub>2</sub>O/kg air). We hypothesize that the coincident increases in lightning NO<sub>x</sub> and stratosphere-troposphere exchange reflect a more vigorous mixing circulation in 2000–2004.

[14] Since lightning NO<sub>x</sub> is most strongly correlated with annual mean OH, we investigate whether the rise in OH is caused by the increase in lightning NO<sub>x</sub>. In the model, lightning NO<sub>x</sub> is parameterized using empirical relationships between cloud top heights and flash frequency (separately for continental and marine) [Price *et al.*, 1997; see also Horowitz *et al.*, 2003]. This commonly used approach allows for interannual variations, but is highly sensitive to model biases in cloud top heights; flash rate parameterizations based on mass fluxes are more consistent with observations [Allen and Pickering, 2002]. To test the sensitivity of OH to lightning NO<sub>x</sub>, we decrease the lightning NO<sub>x</sub> source during model years 1997–2004 to the 1991–1995 rate (see auxiliary material). Based on this simulation, we find that the lightning NO<sub>x</sub> increase causes most of the 0.11 yr decrease in  $\tau_{\text{OH}}$  attributed to higher OH, roughly consistent with the relationship found by Labrador *et al.* [2004]. The tropospheric O<sub>3</sub> burden decreases by 2.6 Tg in this sensitivity simulation relative to the mean 2000–2004 BASE simulation, but is still  $\sim 10$  Tg higher than the BASE 1991–1995 value, reflecting a larger influx from the stratosphere in the later years. Given that the increase in lightning NO<sub>x</sub> can fully account for the decrease in  $\tau_{\text{OH}}$ , we infer that the larger stratospheric O<sub>3</sub> flux (and the resulting increase in tropospheric O<sub>3</sub> burden) contributes minimally to the enhanced OH in our simulation, consistent with the findings of Dentener *et al.* [2003]. We conclude that the 0.3 Tg N yr<sup>-1</sup> increase in lightning NO<sub>x</sub> from 1991–1995 to 2000–2004 yields the modeled increase in OH and the corresponding decrease in  $\tau_{\text{OH}}$ . Alternatively, a shift in anthropogenic NO<sub>x</sub> emissions from northern latitudes to the tropics or an increase in total emissions (both of which occur from 1990 to 2000 in the EDGAR 3.2 and FT2000 NO<sub>x</sub> inventories, not included in our simulations) could contribute to the observed CH<sub>4</sub> trend by enhancing OH [Gupta *et al.*, 1998]. Earlier work, however, suggests that the net impact of trends in all surface emissions and photolysis rates (via stratospheric O<sub>3</sub> columns) is small [Dentener *et al.*, 2003].

#### 5. Discussion

[15] Our simulations with the MOZART-2 tropospheric chemistry model show that anthropogenic and biogenic CH<sub>4</sub> source estimates are consistent with observed CH<sub>4</sub> abundances. We further use the model to identify the processes driving the CH<sub>4</sub> trend from 1990 to 2004. When emissions are held constant, the model successfully captures the observed rise in the early 1990s and flattening post-1998

but overestimates CH<sub>4</sub> abundances in the early 1990s (by 7–15 ppb) and underestimates CH<sub>4</sub> after 1998 (by 13–20 ppb). We find that the overall CH<sub>4</sub> trend is largely controlled by small (<2%) changes in the tropospheric CH<sub>4</sub> sink, decreasing the annual mean CH<sub>4</sub> lifetime by 0.17 years from 1991–1995 to 2000–2004. We attribute 65% of this lifetime change to an increase in tropospheric OH (+1.2% airmass-weighted), and the remainder to a lower tropospheric warming (+0.3 K) during this period. The OH enhancement in the model is caused by an increase in lightning NO<sub>x</sub>. The reduction in CH<sub>4</sub> lifetime in response to rising temperatures is a negative feedback on greenhouse warming, which we estimate will offset the initial warming by at most a few percent (see auxiliary material).

[16] Including annually increasing anthropogenic CH<sub>4</sub> emissions improves agreement with observations after 1998, but this simulation still fails to capture the observed interhemispheric gradient and seasonal cycles at high northern latitudes. A simulation with annually varying anthropogenic and wetland emissions best reproduces the observed global mean CH<sub>4</sub> trend, interhemispheric gradient, and seasonal cycles, indicating that CH<sub>4</sub> is sensitive to the spatial and temporal distribution of wetland emissions.

[17] Future shifts in climate can exert a strong influence on the CH<sub>4</sub> lifetime by influencing the CH<sub>4</sub>-OH reaction rate, as well as convective activity and subsequent lightning NO<sub>x</sub> production. While warmer temperatures and greater lightning activity induce negative feedbacks on greenhouse warming via CH<sub>4</sub>, increased wetland CH<sub>4</sub> emissions in a warmer, wetter climate may offset these feedbacks [Shindell et al., 2004]. Additional work is needed to determine whether enhanced lightning activity is a robust feature of a warming world or if the increase is sensitive to details of the model parameterization. More physically-based representations of lightning NO<sub>x</sub> in chemistry-climate models would enable the analysis of this feedback together with other climate-driven feedbacks, such as changes in biogenic emissions of CH<sub>4</sub> and other species.

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