Observational constraints on recent increases in the atmospheric CH_4 burden

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[1] Measurements of atmospheric CH_4 from air samples collected weekly at 46 remote surface sites show that, after a decade of near-zero growth, globally averaged atmospheric methane increased during 2007 and 2008. During 2007, CH_4 increased by 8.3 ± 0.6 ppb. CH_4 mole fractions averaged over polar northern latitudes and the Southern Hemisphere increased more than other zonally averaged regions. In 2008, globally averaged CH_4 increased by 4.4 ± 0.6 ppb; the largest increase was in the tropics, while polar northern latitudes did not increase. Satellite and in situ CO observations suggest only a minor contribution to increased CH_4 from biomass burning. The most likely drivers of the CH_4 anomalies observed during 2007 and 2008 are anomalously high temperatures in the Arctic and greater than average precipitation in the tropics. Near-zero CH_4 growth in the Arctic during 2008 suggests we have not yet activated strong climate feedbacks from permafrost and CH_4 hydrates. **Citation:** Dlugokencky, E. J., et al. (2009), Observational constraints on recent increases in the atmospheric CH_4 burden, *Geophys. Res. Lett.*, 36, L18803, doi:10.1029/2009GL039780.

1. Introduction

[2] Our ability to quantify the global methane budget is poor, particularly at the regional scales needed to assess the effectiveness of emission reduction schemes and detect climate feedbacks on CH_4 emissions. Quantifying changes in most anthropogenic CH_4 emissions is complicated because emissions from wetlands and biomass burning are dispersed over large areas, with emission rates that vary significantly inter-annually. These sources are also strongly affected by climate and land-use change.

[3] During 2007, near-record Arctic warmth [*Lawrence et al.*, 2008; *Hansen et al.*, 1999] (http://data.giss.nasa.gov/gistemp) and record low sea ice extent [*Stroeve et al.*, 2008] raise concerns that a strong climate feedback, i.e., release of organic carbon (after conversion to CH_4 or CO_2 by microbes) from melting permafrost and release of CH_4 from shallow hydrates on the continental shelf, could be activated. Methanogenesis is strongly temperature depen-

dent and occurs under anoxic conditions in saturated soils containing carbon. *Tarnocai et al.* [2009] estimated an organic carbon pool of 1024 Pg for the top 3 m of soil in the northern circumpolar permafrost region, although the uncertainty on the estimate is large. *Lawrence and Slater* [2005] used two IPCC greenhouse gas emissions scenarios in the Community Climate System Model (version 3) with explicit treatment of frozen soil processes, and they found between 50% and 90% of near-surface permafrost could melt by 2100. Although the large magnitude of these changes has been disputed [*Burn and Nelson*, 2006; *Delisle*, 2007], there is clearly potential for increased future CH_4 emissions and, thus, large positive feedbacks on climate warming from high northern latitudes.

[4] Improved understanding of the response to climate change of processes responsible for CH_4 emission can be gained by comparing our process-level understanding of CH_4 emissions to observed atmospheric inter-annual variability of CH_4. During 1997–1998, for example, the imbalance between CH_4 emissions and sinks increased by ~ 25 Tg yr^{-1} (increases of 6.3 ± 0.7 ppb in 1997 and 12.4 ± 0.7 ppb in 1998), out of average total emissions of ~ 550 Tg CH_4 yr^{-1}. Likely causes were increased tropical biomass burning, which may have also affected the CH_4 sink through changes in [OH], and warm, wet conditions in some wetland regions of the high northern latitudes and tropics [*Bousquet et al.*, 2006; *Dlugokencky et al.*, 2003]. Here we use atmospheric observations to investigate the causes of the increase in CH_4 growth rate during 2007 and 2008.

2. Experimental Methods

[5] Air sample pairs are collected approximately weekly in 2.5 L flasks from sites in NOAA's global cooperative air sampling network [*Dlugokencky et al.*, 1994]. Flasks are flushed and pressurized to ~ 1.2 atm with a portable sampler. Methane is measured by gas chromatography with flame ionization detection against the NOAA 2004 CH_4 standard scale [*Dlugokencky et al.*, 2005] and reported in dry air mole fractions (nmol mol^{-1}, abbreviated ppb). Repeatability of the measurements averages 1.5 ppb (1 s.d.). For this study, measurements from 46 globally-distributed remote boundary layer sites were fitted with curves to smooth variability with periods less than ~ 40 days [*Dlugokencky et al.*, 1994]. Synchronized points were extracted from these curves at approximately weekly intervals and smoothed as a function of latitude to define an evenly spaced matrix of surface CH_4 mole fractions as a function of time and latitude (data path: ftp://ftp.cmdl.noaa.

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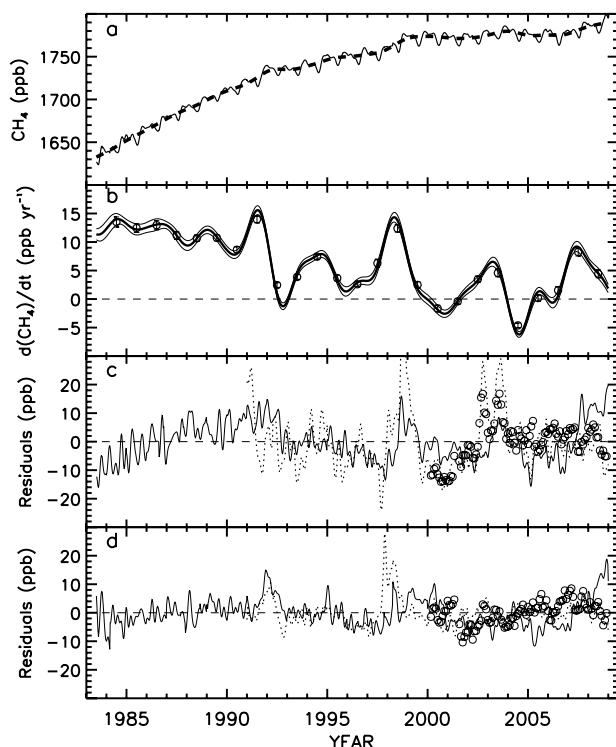
Figure 1. (a) Solid line shows globally averaged CH$_4$ dry air mole fractions; dashed line is a deseasonalized trend curve fitted to the global averages. (b) Instantaneous growth rate for globally averaged atmospheric CH$_4$ (solid line; dashed lines are $\pm 1\sigma$ [*Steele et al.*, 1992]). The growth rate is the time-derivative of the dashed line in Figure 1a. Circles are annual increases, calculated from the trend line in Figure 1a as the increase from January 1 in one year to January 1 in the next. (c) Residuals from a function fitted to zonal averages for CH$_4$ (solid line), CO (dotted line), and MOPITT CO (circles) for polar northern latitudes (53.1°N to 90°N). (d) Same as Figure 1c, but for the tropics (17.5°S to 17.5°N).

gov/ccg/ch4/flask/). This matrix was used to calculate global and zonal CH$_4$ averages. Zonal averages used are Northern Hemisphere (equator to 90°N), Southern Hemisphere (equator to 90°S), tropical (17.5°S to 17.5°N), low northern latitudes (equator to 30°N), and polar northern latitudes (53°N to 90°N).

[6] ^{13}C/^{12}C (δ^{13}C) in CH$_4$ was measured in a subset of the samples measured for CH$_4$ mole fraction. Isotopic analysis is done on an automated system using gas chromatography with isotope-ratio mass spectrometry. Using 200 mL of air, repeatability (1 s.d.) on multiple replicates of dry natural air from a cylinder is ~0.1‰ [*Miller et al.*, 2002]. CO and SF$_6$ mole fractions were measured from the same samples analyzed for CH$_4$. CO was determined using a reduction gas analyzer [*Novelli et al.*, 1994], and SF$_6$ by GC/ECD.

3. Results and Discussion

3.1. CH$_4$ Observations

[7] Globally-averaged surface CH$_4$ mole fractions (solid line) and deseasonalized trend (dashed line) at weekly

resolution are plotted in Figure 1a for 1983 to 2008. From 1999 to 2006, CH$_4$ remained nearly constant except for a small increase in 2002 to 2003. Based on measurements of CO abundance in the same samples, a potential contributor to this increase was biomass burning in Boreal regions of Asia and N. America [*van der Werf et al.*, 2006]. In Figure 1b, the derivative with respect to time of the trend is plotted as a solid line, and annual increases (from 1 January in one year to 1 January in the next) are plotted as circles. Uncertainties (1 s.d.) are calculated with a Monte Carlo method [*Steele et al.*, 1992] and only account for the sampling uncertainty resulting from the distribution of our network. Though the average annual increase from 2000 to 2006 was 0.4 ppb yr^{-1} (s.d. = 3.1 ppb yr^{-1}), the global CH$_4$ burden decreased during 3 of these years. In 2007, the global increase was 8.3 $\pm$ 0.6 ppb, in good agreement with *Rigby et al.* [2008], corresponding to an imbalance between emissions and sinks of ~23 Tg CH$_4$. Except for 1998, this is the largest observed increase since the early 1990s. The largest zonally averaged CH$_4$ increase in 2007 was observed at polar northern latitudes, 13.7 $\pm$ 1.3 ppb. Despite this large increase at Arctic latitudes, the increase during 2007 in the zonally averaged Southern Hemisphere (9.2 $\pm$ 0.3 ppb) was larger than the increase in the zonally averaged Northern Hemisphere (7.3 $\pm$ 1.3 ppb). In 2008, the global increase was 4.4 $\pm$ 0.6 ppb yr^{-1}, with the largest increase, 8.1 $\pm$ 1.6 ppb, observed at low northern latitudes. Northern polar regions resumed their low growth in 2008, with near-zero increase (0.5 $\pm$ 0.8 ppb).

[8] Methane mole fractions (Figure 2a) and δ^{13}C (Figure 2b) are plotted for Alert, Canada. Data were smoothed (lines) as discussed above. Minimum CH$_4$ values during summer occur primarily because there is a seasonal maximum in OH concentrations in the Northern Hemisphere, despite summer being the time of maximum CH$_4$ emissions from wetlands. During summer 2007, CH$_4$ at ALT was ~12 ppb greater than during the previous few summers; this anomaly continued through the winter and persisted into 2008. In late-summer 2007, we also observed that δ^{13}C in CH$_4$ was the lowest during our period of record. The changes in δ^{13}C and CH$_4$ mole fraction from 2006 to 2007 during summer suggest increased emissions from a source with δ^{13}C $\approx$ -66‰, while typical δ^{13}C from wetlands is -60‰ or lighter. Reaction of CH$_4$ with OH enriches ^{13}C, but only one third as much per mole. A decrease in [OH] would shift observed δ^{13}C in the same direction as increased emissions from ^{13}C-depleted source, but the observed change in δ^{13}C (~-0.1‰ from 2006 to 2007) is too large to be consistent with potential changes in [OH]. Decreased [OH] would also affect other species such as CO, but significant anomalies were not observed for CO at high northern latitudes in 2007. Emissions from biomass burning, with δ^{13}C values of ~-25‰, would have resulted in more positive δ^{13}C in CH$_4$.

3.2. Potential Contributions to the 2007 and 2008 Increases

[9] Emission rates of CH$_4$ from most anthropogenic sources change gradually, so the drivers of interannual variability in CH$_4$ growth rate are typically changes in emissions from biomass burning and wetlands, and changes in CH$_4$ sink rate, through changes in [OH] [*Dlugokencky et*
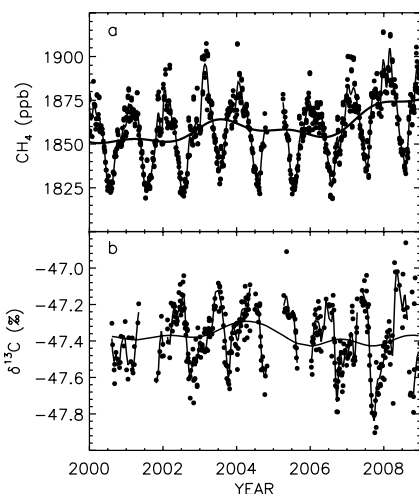
Figure 2. (a) Circles are CH$_4$ dry air mole fractions from weekly discrete samples collected at Alert, Canada. Lines are a smooth curve fitted to weekly samples and trend, as in Figure 1a. Smoothing of weekly samples eliminates variability with periods on order of one month or less. (b) Circles are δ^{13}C in CH$_4$ (‰) measured in same samples used for CH$_4$ analysis. Lines are same as in Figure 2a.

al., 1996]. The large increase in CH$_4$ at polar northern latitudes during 2007 compared to other latitude zones was coincident with anomalously high temperature (warmest year during our measurement period for northern wetland regions). Lighter than average δ^{13}C in CH$_4$ during late-summer 2007 is consistent with a wetland source. A change in [OH] at high northern latitudes is not a potentially important contributor to the CH$_4$ budget there, because [OH] is low relative to the tropics and there is a limited seasonal period when the reaction occurs. The effects of changes in [OH] at low latitudes would be transported to high latitudes. Observations of 1,1,1-trichloroethane (update of *Montzka et al.* [2000]) also suggest no significant contribution to the ~4% CH$_4$ anomaly in 2007 from decreased [OH] (inferred [OH] changes were in the range −2% to +1%, depending on assumptions made about 1,1,1-trichloroethane emissions). This result is consistent with the change in [OH] estimated by *Rigby et al.* [2008] because of their large uncertainty range (−4 ± 14%).

[10] To assess the potential for increased emissions of CH$_4$ from biomass burning at high northern latitudes, we studied surface CO measured in the same samples that were analyzed for CH$_4$. Figure 1c shows residuals from a function (2nd-order polynomial and 4 annual harmonics) fitted to zonally averaged CH$_4$ and CO for northern polar latitudes (53° to 90°N). Significant elevations from the noise are seen for CH$_4$ residuals (solid line) in 1997/98, 2002/03, and 2007/08. CO, with an emission molar ratio relative to CH$_4$ for biomass burning in the range 10 to 20 [*Christian et al.*, 2003], has large residuals (dotted line) only during the first two periods, when anomalies in Boreal biomass burning are known to have occurred. This also suggests the Arctic CH$_4$ anomaly in 2007 is related to sources other than biomass burning, or to a larger change in [OH] than is consistent with methyl chloroform observations.

[11] In the tropics, our CO measurements may be a less-reliable indicator of biomass burning emissions, because emissions are injected into the mid-troposphere, away from our surface sampling sites. Despite this, CO anomalies in the tropics were clearly seen in late-1997 and 1998 when biomass burning was wide-spread in Indonesia (Figure 1d). During 2006, 2007, and 2008, the surface CO signal was small, typical of background variability. However, in the middle troposphere, from October, 2006 through March, 2007, evidence of biomass burning comes from monthly averaged CO centered at 700 hPa detected by MOPITT (Measurements of Pollution in the Troposphere), a gas filter radiometer on board the Terra satellite from which CO vertical profiles are obtained [see, e.g., *Edwards et al.*, 2006] (http://www.acd.ucar.edu/mopitt/MOPITT/data/plots4/mapsv4_mon.html). CO anomalies up to 90 ppb were detected over the Indian Ocean. Zonally averaged monthly CO anomalies for polar northern (Figure 1c) and tropical latitudes (Figure 1d) are plotted as circles. Further evidence that some of the CH$_4$ anomaly was caused by biomass burning comes from changes in global ethane abundance observed during 2007 [*Simpson et al.*, 2006; I. Simpson, personal communication, 2008] and CH$_3$Cl (lifetime ~1.5 yr), which increased along with CH$_4$ in early-2007 at Samoa. Based on an emission ratio of 15 ppt CH$_3$Cl to 1 ppb CH$_4$ for biomass burning [*Christian et al.*, 2003] and anomalies in CH$_3$Cl of ~20 ppt at Mauna Loa and Samoa in 2007, the relative contribution of biomass burning to the CH$_4$ anomaly in the tropics during 2007 was small.

[12] A source other than biomass burning must have made significant contributions to the CH$_4$ enhancements in the tropics and extra-tropical Southern Hemisphere. Based on our analysis of gridded precipitation fields [*Schneider et al.*, 2008] (http://gcmd.nasa.gov/records/GCMD_DWD-GPCC_VASClimO.html), 2007 had the 3rd-largest and 2008 the largest positive precipitation anomalies from 1986 to 2008 for all wetland grid cells between 17.5°S and 17.5°N. In the tropics, precipitation is the dominant driver of wetland CH$_4$ emissions. Above-normal precipitation is common in some tropical regions during La Niña events; La Niña conditions started in mid-2007, waned toward the end of the year, and intensified during the first half of 2008. Vertical profiles of air samples collected near Santerém and Manaus in eastern and central Amazônia are typically enhanced in CH$_4$ relative to Atlantic background sites in the NOAA air sampling network. *Miller et al.* [2007] used these observations to infer CH$_4$ fluxes averaged over large areas (~10^5 km^2). Extension of their analysis to include 2007 and 2008 shows that, during these La Niña years (2007 and 2008), CH$_4$ emissions estimated from the profiles averaged over all seasons were ~50% greater than the average emissions calculated for 2000 to 2006. While many sources contribute to these estimated fluxes, wetlands are likely the dominant source.

[13] ENSO can affect observed CH$_4$ hemispheric averages in another way. During cold phases, interhemispheric transport may be enhanced as westerly winds at ~200 hPa in the tropics allow large scale waves to propagate through the topics into the other hemisphere. These conditions can enhance the rate of interhemispheric exchange [*Prinn et al.*, 1992], which may be in part responsible for the increased growth rate of CH$_4$ at mid- to high southern latitudes during

2007. Our measurements of the SF$_6$ latitude gradient over time are consistent with an increase of ∼15% in the rate of interhemispheric exchange. We last saw such an event for CH$_4$ during the La Niña of 1988/1989 when the CH$_4$ growth rate in the SH increased while the growth rate in the NH decreased by a comparable magnitude. Changes in growth rate were accompanied by a decrease of ∼5 ppb in the difference between NH and SH annual means in 1989. These changes are consistent with a temporary increase in interhemispheric exchange rate of ∼10% [*Steele et al.*, 1992].

4. Summary and Conclusions

[14] We measured increases in global atmospheric CH$_4$ of 8.3 ± 0.6 ppb during 2007 and 4.4 ± 0.6 ppb in 2008. These came after nearly a decade of little increase. The causes of the increases are not certain, but at least 3 factors likely contributed to the observations. First, very warm temperatures at polar northern latitudes during 2007 likely enhanced emissions from northern wetlands. Increased emission from wetlands is consistent with observations of lighter than normal δ^{13}C in CH$_4$ at our northern-most site. Since the growth rate returned to near zero in the polar Northern Hemisphere during 2008, the Arctic has not yet reached a point of sustained increased CH$_4$ emissions from melting permafrost and CH$_4$ hydrates. Second, independent observations of CH$_3$Cl (NOAA), CH$_3$CH$_3$ (by UC Irvine), and CO (by MOPITT and NOAA) are consistent with a contribution to CH$_4$ increases in the tropics by biomass burning during October and November, 2006, but comparisons with other large biomass burning events in the tropics during 1997 and 1998 suggest the fraction of enhanced emissions from biomass burning was small. Third, positive anomalies in precipitation in Indonesia and the eastern Amazon, typical during La Niña events, may have driven increased emissions from tropical wetlands, consistent with estimates of CH$_4$ fluxes derived from observations of CH$_4$ above Santarém, Brazil. We also recognize that enhanced interhemispheric transport during the ENSO cool phase may, in part, be responsible for increased CH$_4$ growth rate at mid- to high southern latitudes during 2007; this is consistent with estimates of interannual variability in the rate of interhemispheric exchange derived from SF$_6$ measurements. We emphasize that, although changing climate has the potential to dramatically increase CH$_4$ emissions from huge stores of carbon in permafrost and from Arctic hydrates, our observations are not consistent with sustained changes there yet.

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