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Supplement of

Role of OH variability in the stalling of the global atmospheric CH_4 growth rate from 1999 to 2006

Joe McNorton et al.

Correspondence to: Joe McNorton (eejrm@leeds.ac.uk)

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Supplement S1: Global Box Model

To investigate the overall impact of changes in temperature, [OH] and emissions on an atmospheric gas which is removed by OH a simple global box model can be used. This model permits a first-order understanding of the factors which govern the variation in growth and the relative contribution of emissions and loss to that growth. The model integrates the global mean burden of X (Tg) based on annual mean emissions (*E*, Tg/yr) and chemical loss (*L*, Tg/yr) through the reaction X + OH → products. The modelled atmospheric burden of X ($X_{,t}$) can be integrated over a 1-year period (Δt) according to the equation:

$$\frac{1}{\Delta t}(X_{t+\Delta t} - X_t) = E - L = E - k[OH][X] \qquad (1)$$

where k (cm^3 $molecule^{-1}$ yr^{-1}) is the rate constant for the X + OH reaction (e.g. Sander et al., 2011). For CH_4 $k = 2.45\times10^{-12}\exp(-1775/T)$ while for CH_3CCl_3 $k = 1.64\times10^{-12}\exp(-1520/T)$.

When simulating CH_3CCl_3 we assume emissions from Montzka et al. (2011) or Rigby et al. (2013) and T=272.9 K. We then use equation (1) to derive the global mean [OH] which is consistent with the observed variations in CH_3CCl_3. The same procedure is used to derive global mean [OH] consistent with the observed CH_4 variations assuming E=553 Tg/yr.

Supplement S2: Spatial differences between CH_4 and CH_3CCl_3

As noted in Section 3.2.2 of the main text, anomalies in global OH derived from global CH_3CCl_3 variations might not be appropriate when considering changes in global CH_4. This is based on the fact that the same OH anomalies produce a different response when applied to CH_4 and CH_3CCl_3 in the model with and without the effect of wind and temperature variability (see Figures 2 and 5a of the main text).

To investigate the possible impact of the distribution of sites used to derive the OH variations, we compare the modelled growth rate of global CH_4, derived from model sampling at NOAA and AGAGE CH_4 sites, with the modelled decay rate anomaly of CH_3CCl_3, derived from model sampling at NOAA and AGAGE CH_3CCl_3 sites (Figure S1). The results show that, despite the spatial differences between the sampled locations, the CH_3CCl_3 decay rate anomaly correlates well (negatively) with the CH_4 growth rate, in agreement with the observations (see Figure 1a of main text).

After 1997 CH_3CCl_3 emissions diminish and the atmospheric concentration becomes spatially uniform. Site sampling of the CH_3CCl_3 decay rate anomaly after 1997 should therefore accurately represent global decay. For CH_4 the emission sources influence the spatial distribution of the atmospheric concentration and therefore the loss rate due to OH (Figure S2). In addition, for this reason variability in transport is more likely to influence the global growth rate of CH_4 than the decay rate anomaly of CH_3CCl_3 (see main text).

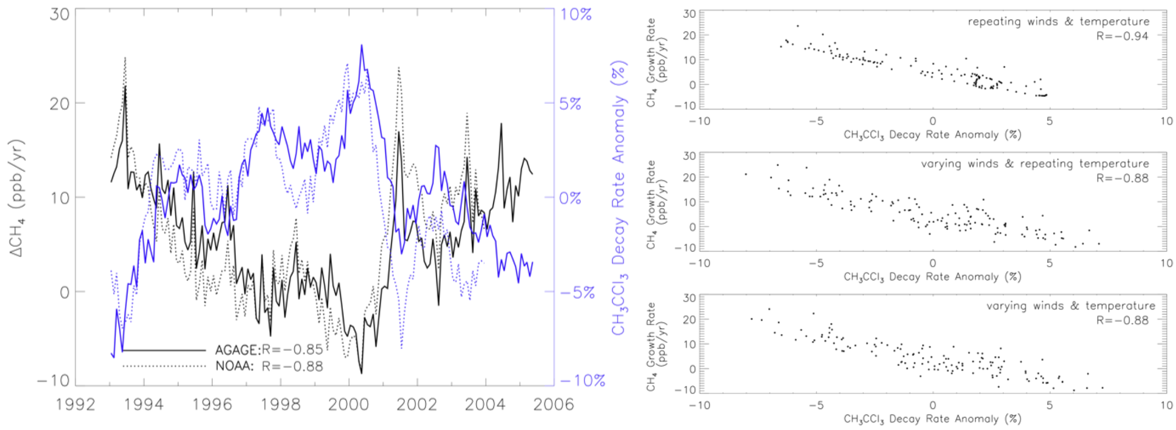

Figure S1. (Left) The smoothed variation in global annual CH_4 growth rate (ppb/yr) derived from TOMCAT 3-D CTM sampled at NOAA (black solid) and AGAGE (black dashed) CH_4 site locations (left axis). Also shown are the smoothed global CH_3CCl_3 decay rate anomalies derived from TOMCAT sampled at NOAA and AGAGE CH_3CCl_3 site locations (right axis). The legend gives represent correlation coefficients of global model CH_4 growth rate compared with the CH_3CCl_3 decay rate anomaly for AGAGE and NOAA locations. (Right) Correlation plots for global CH_4 growth rate and CH_3CCl_3 decay rate anomalies, sampled at respective locations, from the TOMCAT simulation which uses NOAA-derived OH anomalies and, (top) repeating winds and temperature, (middle) varying winds and repeating temperature and (bottom) varying winds and temperature.

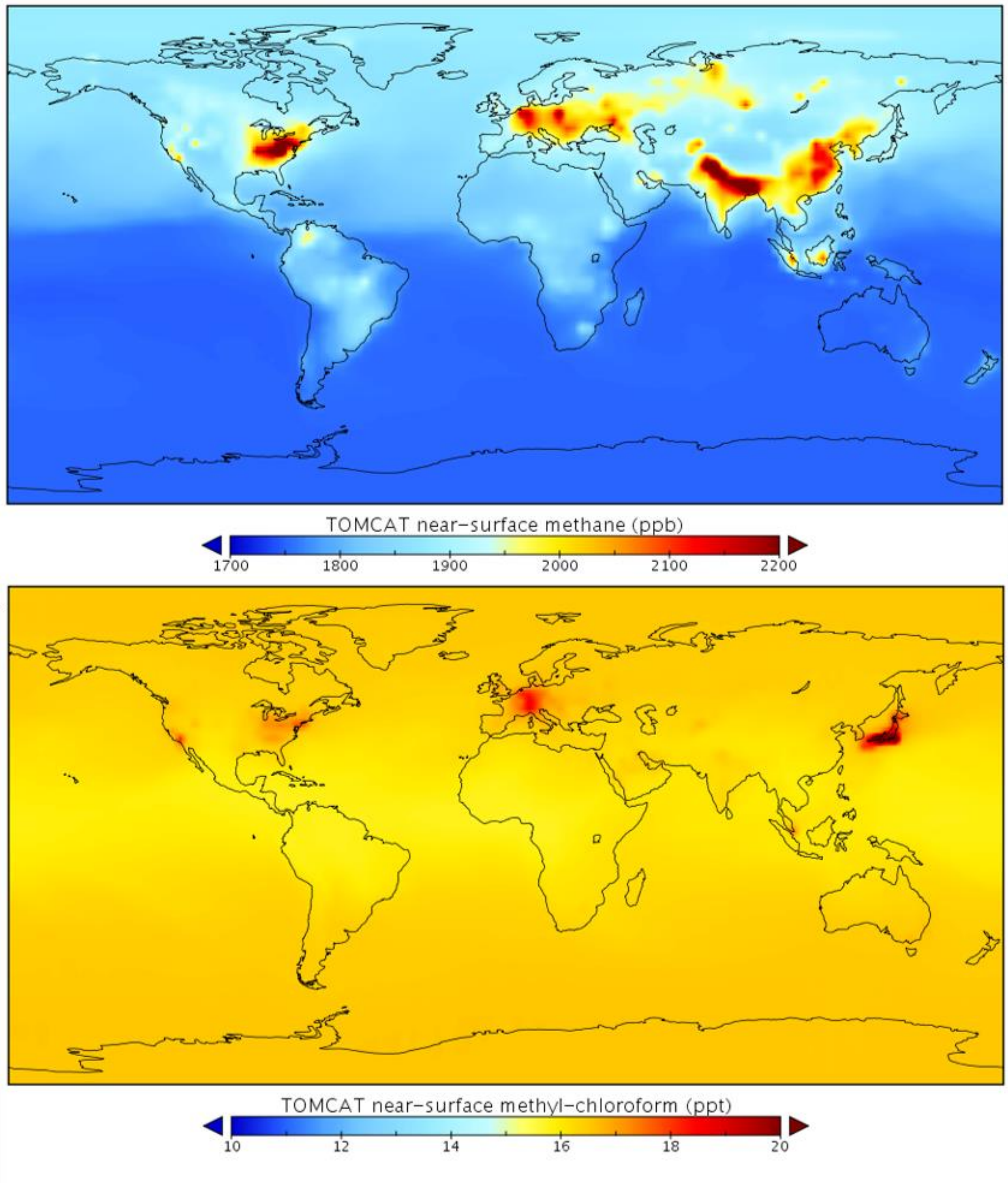

Figure S2. Monthly mean distribution of near surface (top) CH_4 (ppb) and (bottom) CH_3CCl_3 (ppt) for September 2005 from a TOMCAT simulation.