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

J. McNorton[1,2], M. P. Chipperfield[1,2], M. Gloor[3], C. Wilson[1,2], W. Feng[1,4],
G. D. Hayman[5], M. Rigby[6], P. B. Krummel[7], S. O'Doherty[6], R. G. Prinn[8], R. F. Weiss[9],
D. Young[6], E. Dlugokencky[10], and S. A. Montzka[10]

1. School of Earth and Environment, University of Leeds, Leeds, LS2 9JT, UK.
2. National Centre for Earth Observation, University of Leeds, LS2 9JT, UK.
3. School of Geography, University of Leeds, Leeds, LS2 9JT, UK.
4. National Centre for Atmospheric Science, University of Leeds, LS2 9JT, UK.
5. Centre for Ecology and Hydrology, Wallingford, UK.
6. School of Chemistry, University of Bristol, Bristol, BS8 1TS, UK.
7. CSIRO Oceans and Atmosphere Flagship, Aspendale, Victoria, Australia.
8. Center for Global Change Science, Massachusetts Instititute of Technology, Cambridge, MA 02139, USA.
9. Scripps Institution of Oceanography, University of California, San Diego,CA 92093, USA.
10. National Oceanic and Atmospheric Administration, Boulder, USA.

Abstract

The growth in atmospheric methane (CH_4) concentrations over the past two decades has shown large variability on a timescale of ~~many~~several years. Prior to 1999 the globally averaged CH_4 concentration was increasing at a rate of 6.0 ppb/yr, but during a stagnation period from 1999 to 2006 this growth rate slowed to 0.6 ppb/yr. From~~Since~~ 2007 to 2009 the growth rate ~~has~~ again increased to 4.9 ppb/yr. These changes in growth rate are usually ascribed to variations in CH_4 emissions. We have used a 3-D global chemical transport model, driven by meteorological reanalyses and variations in global mean hydroxyl (OH) concentrations derived from CH_3CCl_3 observations from two independent networks, to investigate these CH_4 growth variations. The model shows that between 1999 and 2006, changes in the CH_4 atmospheric loss contributed significantly to the suppression in global CH_4 concentrations relative to the pre-1999 trend. The largest factor in this is relatively small variations in global mean OH on a timescale of a few years, with minor contributions of atmospheric transport of CH_4 to its sink region and of atmospheric temperature. Although changes in emissions may be important during the stagnation period, these results imply a smaller variation is required to explain the observed CH_4 trends. The contribution of OH variations to the renewed CH_4 growth after 2007 cannot be determined with data currently available.

1. Introduction

The global mean atmospheric methane (CH_4) concentration has increased by a factor of 2.5 since the pre-industrial era, from approximately 722 ppb in 1750 to 1803.2 ± 0.7 ppb in 2011 (Etheridge et al., 1998; Dlugokencky et al., 2005). Over this time period methane has accounted for approximately 20% of the total direct anthropogenic perturbation of radiative forcing by long-lived greenhouse gases (0.48 ± 0.05 W/m^2), the second largest contribution after CO_2 (Cicerone et al., 1988; Myhre et al., 2013). This long-term methane increase has been attributed to a rise in anthropogenic emissions from fossil fuel exploitation, agriculture, waste management and biomass burning (Dlugokencky et al., 2011). Predictions of future CH_4 levels require a complete understanding of processes governing emissions and atmospheric removal.

Since the mid-1980s measurements of CH_4 in discrete atmospheric air samples collected at surface sites have been used to observe changes in the interannual growth rate of CH_4 (Rigby et al., 2008; Dlugokencky et al., 2011, Kirschke et al., 2013). Nisbet et al. (2014) showed that between 1984 and 1992 atmospheric CH_4 increased at ~12 ppb/yr, after which the growth rate slowed to ~3 ppb/yr. In 1999 a period of near-zero growth began which continued until 2007. In 2007 this stagnation period ended and ~~since then~~up until 2009 average growth~~ has~~ increased again to ~4.9~~6~~ ppb/yr (Rigby et al., 2008; Dlugokencky et al., 2011).

The reasons for the pause in CH_4 growth are not well understood. Bousquet et al. (2006) performed an atmospheric transport inversion study to infer an increase in anthropogenic emissions since 1999. Similarly, the EDGAR v3.2, bottom-up anthropogenic emission inventory, an updated inventory to that used as an a priori by Bousquet et al. (2006), shows a year-on-year increase in anthropogenic CH_4 emissions between 1999 and 2006 (Olivier et al., 2005). This would suggest that a decrease in anthropogenic emissions is not the likely cause of the pause in growth during this period. A second potential explanation is a reduction in wetland emissions between 1999 and 2006, which is in part compensated by an increase in anthropogenic emissions (Bousquet et al., 2006). However, more recently, Pison et al. (2013) used two atmospheric inversions alongside a process-based model and found much more uncertainty in the role wetlands played in the pause in growth over this period. Their study found a negative trend in Amazon basin emissions between 2000 and 2006 from the process-based model and a positive trend from the inversion estimates.

Dlugokencky et al. (2003) argued that the behaviour of global mean CH_4 up to around 2002 was characteristic of the system approaching steady state, accelerated by decreasing emissions at high northern latitudes in the early 1990s and fairly constant emissions elsewhere. However, since then there have been notable perturbations to the balance of sources and sinks (Rigby et al., 2008). ~~This~~The observed growth since 2007 has been, at least partly, attributed to increases in wetland (Bousquet et al., 2011) and anthropogenic emissions (Bousquet et al., 2011~~06~~). Recent changes in emissions are not well constrained and the reasons for the renewed growth are also not fully understood (Nisbet et al., 2014).

Atmospheric chemistry has also been hypothesised to play a role in past variations in CH_4 growth rates. The major (90%) sink of atmospheric CH_4 is via reaction with the hydroxyl radical, OH. Variations in the global mean concentration of OH ([OH]), or changes to the reaction rate through changes in temperature, therefore have the potential to affect CH_4 growth. Previous studies have suggested that an increase in atmospheric OH concentration may have been at least partly responsible for a decrease in the CH_4 growth rate (Karlsdottir and Isaksen et al., 2000; Lelieveld et al., 2004; Wang et al., 2004; Fiore et al., 2006). This rise in OH has been attributed to an increase in lightning NO_x (Fiore et al., 2006), a decrease in column O_3 (Wang et al., 2004) and changes in atmospheric pollutants (Karlsdottir and Isaksen et al., 2000). The abundance of other species such as H_2O~~, O_3, CO~~ and CH_4 also determine the concentration of OH (Leliveld et al., 2004). Prinn et al. (2005) and Voulgarakis et al. (2015) suggested that major global wildfires and El Nino Southern Oscillation (ENSO) events could influence [OH] variability. ~~A reduced number of small- to moderate-magnitude volcanic eruptions during the CH_4 stagnation period (Carn et al., 2015; Mills et al., 2015) may have increased [OH], due to increased downward UV radiation. Recently, Patra et al. (2014) analysed global CH_3CCl_3 observations for 2004-2011 to derive the interhemispheric ratio of OH. In contrast to many model results which suggest higher mean [OH] in the north, they derived similar values for both hemispheres.~~

Warwick et al. (2002) investigated the impact of meteorology on atmospheric CH_4 growth rates from 1980 to 1998, i.e. well before the observed recent pause. They concluded that atmospheric conditions could be an important driver in the interannual variability (IAV) of atmospheric CH_4. In similar studies a combination of atmospheric dynamics and changes in emissions were shown to explain some of the earlier past trends in atmospheric CH_4 (Fiore et al., 2006; Patra et al., 2009). This paper builds on these studies to investigate the chemical and non-chemical atmospheric contribution to the recent variations in CH_4 growth. By ‘non-chemical’ we mean transport-related influences, although the loss of CH_4 is ultimately due to chemistry as well. We use a 3-D global chemical transport model to simulate the period from 1993 to 2011 and to quantify the impact of variations in [OH] and meteorology on atmospheric CH_4 growth.

2. Data and Models

2.1 NOAA and AGAGE CH_4 Data and Derived OH

We have used surface CH_4 observations from 19 National Oceanographic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL) cooperative global air sampling sites (Dlugokencky et al., 2014) over 1993-2009 (see Table 1). To calculate the global average concentration, measurements were interpolated across 180 latitude bins, which were then weighted by surface area. We have also used the same method to derive global mean CH_4 based on 5 sites from the Advanced Global Atmospheric Gases Experiment (AGAGE) network (Prinn et al., 2000; Cunnold et al., 2002; Prinn et al., 2015).

Montzka et al. (2011) used measurements of methyl chloroform (CH_3CCl_3) from an independent set of flasks sampled approximately weekly at a subset of NOAA air sampling sites to derive global [OH] anomalies from 1997 to 2007 and found only a small interannual variability (2.3 ± 1.5%). They argued that uncertainties in emissions are likely to limit the accuracy of the inferred inter-annual variability in global [OH], particularly before 1997. At ~~this~~that time the emissions were large but decreasing rapidly due to the phaseout of CH_3CCl_3 production and consumption, and the large atmospheric gradients were also more difficult to capture accurately with only few measurement sites. Instrument issues caused an interruption to their CH_3CCl_3 time series in 2008/9. We have averaged these (based on the red curve in Figure 3 of Montzka et al.) into yearly anomalies to produce relative interannual variations in the mean [OH]. Similarly, Rigby et al. (2013) used CH_3CCl_3 measurements from on-site instrumentation operated continuously within the 5-station AGAGE network in a 12-box model to produce yearly global [OH] anomalies from 1995 (the date from which data from all 5 stations is available) to 2010. These two timeseries, which convert anomalies in the CH_3CCl_3 decay rate into anomalies in [OH] using constant temperature, correspond to the best estimate of [OH] variability from the two measurement networks by the groups who operate them. We then applied these two series of yearly global anomalies uniformly to the global latitude-height [OH] field used in the recent TransCom CH_4 model intercomparison (see Patra et al., 2011), which itself was derived from a combination of semi-empirically calculated tropospheric OH distributions (Spivakovsky et al. 2000; Huijnen et al., 2010) and 2-D model simulated stratospheric loss rates (Velders, 1995). For consistency between the model experiments, both sets of yearly anomalies were scaled so that the mean [OH] between 1997 and 2007 (the overlap period where NOAA and AGAGE anomalies are both available) equalled the TransCom [OH] value. In the rest of this paper we refer to these two OH datasets as ‘NOAA-derived’ and ‘AGAGE-derived’.

These two calculations of yearly [OH] anomalies use slightly different assumptions for CH_3CCl_3 emissions after 2002. Before that ~~date~~year they use values from Prinn et al. (2005). The NOAA data then assumed a 20% decay in emission for each subsequent year (Montzka et al., 2011), while AGAGE used United Nations Environment Programme (UNEP) consumption values (UNEP, 2015). Holmes et al. (2013) suggested that inconsistencies in CH_3CCl_3 observations between the AGAGE and NOAA networks also limit understanding of OH anomalies for specific years due to an unexplained phasing difference of up to around 3 months. As we are interested in the impact of [OH] changes over longer time periods (e.g. 2000 – 2006), this phase difference will be less important. We have investigated the impact of the different CH_3CCl_3 observations and assumed emissions on the derived [OH] anomalies (see Section 3.1).

2.2 TOMCAT 3-D Chemical Transport Model

We have used the TOMCAT global atmospheric 3-D off-line CTM (Chipperfield, 2006) to model atmospheric CH_4 and CH_3CCl_3 concentrations. The TOMCAT simulations were forced by winds and temperatures from the 6-hourly European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalyses (Dee et al., 2011). They covered the period 1993 to 2011 with a horizontal resolution of 2.8° × 2.8° and 60 levels from the surface to ~60 km.

The TOMCAT simulations use annually repeating CH_4 emissions, which have been scaled to previous estimates of 553 Tg/yr (Ciais et al., 2013), taken from various studies (Fiore et al., 2006; Curry et al., 2007; Bergamaschi et al., 2009; Pison et al., 2009; Spahni et al., 2011; Ito et al., 2012). Annually-repeating anthropogenic emissions (except biomass burning) were calculated from averaging the EDGAR v3.2 (2009) inventory from 1993 to 2009 (Olivier and Berowski, 2001). Biomass burning emissions were calculated using the GFED v3.1 inventory and averaged from 1997 to 2009 (van der Werf et al., 2010). The Joint UK Land Environment Simulator (JULES) (Best et al., 2011; Clark et al., 2011; Hayman et al., 2014) was used to calculate a wetland emission inventory between 1993 and 2009, which was then used to produce a ~~scaled~~ mean annual cycle. Annually-repeating rice (Yan et al., 2009), hydrate, mud volcano, termite, wild animal and ocean (Matthews et al., 1987) emissions were taken from the TransCom CH_4 study (Patra et al., 2011). The methane loss fields comprised an annually repeating soil sink (Patra et al., 2011), an annually repeating stratospheric loss field (Velders, 1995) and a specified zonal mean [OH] field. This does not account for longitudinal variations in [OH], which are considered to be neglible compared to latitudinal varitions. To create a reasonable spatial distribution ~~T~~the model was spun up for 15 years prior to initialising the simulations, using emission data from 1977 to 1992 where available and annual averages otherwise. Before reinitialising the model in 1993, concentrations were scaled using the model and observed global concentrations to remove any imbalance.

Fifteen TOMCAT simulations were performed each with a CH_4 tracer and a CH_3CCl_3 tracer. The runs had differing treatments of meteorology (winds and temperature) and [OH] (see Table 2). Simulations with repeating [OH] fields (RE_xxxx) used the TransCom dataset. The other runs with varying [OH] used the NOAA-derived or AGAGE-derived [OH] fields based on the original published work or our estimates (see Section 3.1). For these runs, the mean [OH] field is used where the respective NOAA or AGAGE-derived [OH] is unavailable or uncertain (before 1997 / after 2007 for NOAA and before 1997 / after 2009 for AGAGE). The five simulations with fixed wind and temperature fields (with labels ending in FTFW) used the ERA-Interim analyses from 1996 repeated for all years. The five simulations with varying winds and fixed temperature (with labels ending in FTVW) used zonal mean temperature fields averaged from 1993-2009, any influence from the relatively small longitudinal temperature variations is unlikely to have a noticeable impact. ~~The OH anomalies are derived~~ We also derive our own [OH] anomalies from the anomaly in the CH_3CCl_3 loss rate, which combines variations in atmospheric OH concentration with variations in temperature which affect the rate constant of the CH_3CCl_3 + OH reaction. To quantify the importance of this temperature effect we also performed 5 model runs which allow both winds and temperature to vary interannually according to ERA-Interim data (labels ending VTVW). Fixed temperature simulations are used for general analysis because the derived OH anomalies already implicity contain temperature variations.

3. Results

3.1 Correlation of CH_4 variations with OH and temperature

We first investigate the extent to which variations in the observed CH_4 growth rate correlate with variations in derived [OH]. Figure 1a shows the published NOAA-derived and AGAGE-derived global [OH] anomalies along with the annual CH_4 growth rate estimated from the NOAA and AGAGE measurements. The two [OH] series show the similar behaviour of negative anomalies around 1997 and 2006/7, and an extended period of more positive anomalies in between. For the time periods covered by the NOAA (1997-2007) and AGAGE (1997-2009) CH_3CCl_3 observations, the two derived [OH] timeseries show negative correlations with the CH_4 growth from NOAA (regression coefficient, R = -0.32) and AGAGE

(R = -0.64). Only the AGAGE [OH] correlation, from the longer timeseries, is statistically significant at the 90% level. This correlation could be the result of a bidirectional effect, whereby decreased CH_4 acts to increase [OH]; however, Spivakovsky et al. (2000) showed a 25% (~450 ppb) change in model CH_4 equates to a 5-6% change in [OH]. This far exceeds the annual growth observed, therefore this effect is assumed to be small.

We can use a simple ‘global box model’ (see Supplement S1) to estimate the [OH] variations required to fit the observed CH_4 growth rate variations assuming constant CH_4 emissions and temperature (black line in Figure 1b). This provides a crude guide to the magnitude of OH variations which could be important for changes in the CH_4 budget. Our results are consistent with those of Montzka et al. (2011) who performed a similar analysis on the NOAA CH_4 data. The required [OH] rarely exceeds their CH_3CCl_3-derived interannual variability (IAV) range of [OH] (±2.3%, shown as shading in the figure). Also shown in Figure 1b are the published estimates of the global mean OH anomalies from Figure 1a, converted to concentration units (see Section 2.1). The relative interannual variations in [OH] required to fit the CH_4 observations match the CH_3CCl_3-derived [OH] variations in many years, for example from 1998-2002 (see Montzka et al., 2011). Some of the derived variations in [OH] exceed that required to match the CH_4 growth rate, with larger negative anomalies in the early and later years and some slightly larger positive values in middle of the period.

Figures 1c and 1d show our estimates of [OH] using NOAA and AGAGE observations and two assumptions of post-2000 CH_3CCl_3 emissions (see Section 2.1) in a global box model. The figures also compare our OH estimates with the NOAA-derived and AGAGE-derived [OH] anomalies based on the work of the observation groups (Figure 1a). Our results demonstrate the small impact of using different observations and post-2000 emission assumptions (compare filled and open red circles for the two panels). For these box model results there is also only a very small effect of using annually varying temperature (compare red and blue lines). In later years the choice of observations has a bigger impact than the choice of emissions on the derived [OH]. For AGAGE-derived values (Figure 1d) our estimates agree well with the published values of Rigby et al., (2013), despite the fact we use a global box model while they used a more sophisticated 12-box model. In constrast, there are larger differences between our values and the NOAA-derived OH variability published by Montzka et al. (2011) (Figure 1c), despite both studies using box models. In particular, around 2002-2003 we overestimate the positive anomaly in [OH]. We also estimate a much more negative OH anomaly in 1997 than Montzka et al., though we slightly unde~~r~~restimate the published AGAGE-derived anomaly in that year (Figure 1d). Tests show that differences between our results and the NOAA box model are due to the treatment of emissions. This suggests a larger uncertainty in the inferred low 1997 [OH] value, when emissions of CH_3CCl_3 were decreasing rapidly, although reasons why atmospheric [OH] might have been anomously low were discussed by Prinn et al. (2005). In the subsequent analysis we use the OH variability from the published NOAA and AGAGE studies as input to the 3-D model.

3.2 TOMCAT Simulations

Overall, Figure 1 shows the potential importance of small, observationally derived variations in OH concentrations to impact methane growth. We now investigate this quantitatively in the framework of a 3-D CTM.

3.2.1 Methyl Chloroform

The TOMCAT simulations include a CH_3CCl_3 tracer. This allows us to verify that our approach of using a global OH field, scaled by derived anomalies, allows the model to reproduce the observed magnitude and variability of CH_3CCl_3 decay accurately. Figure 2a shows that the model, with the imposed [OH] field, does indeed simulate the global decay of CH_3CCl_3 very well. This justifies our use of the 'offline' [OH] field, as models with interactive tropospheric chemistry can produce a large range in absolute global mean [OH] and therefore in lifetimes of gases such as CH_3CCl_3. For example, Voulgarakis et al., (2013) analysed the global mean [OH] from various 3D models and found a range of 0.~~5~~65 × 10^6 to 1.34 × 10^6 molecules cm^{-3}. Furthermore, Montzka et al., (2011) discussed how photochemical models typically show smaller interannual variability than CH_3CCl_3-derived OH, again suggesting that the models are not accurately capturing all relevant processes. Figure 2a also shows that the global mean CH_3CCl_3 from the NOAA and AGAGE networks differ by ~2.5ppt around 1993-1996, but since then this difference has become smaller.

The observed and modelled CH_3CCl_3 decay rate anomalies (calculated using the method of Holmes et al., (2013) with a 12-month smoothing) are shown in Figures 2b and 2c (different panels are used for AGAGE and NOAA comparisons for clarity). The model and observation-derived results both tend to show a faster CH_3CCl_3 decay (more positive anomaly) in the middle of the period, with slower decay at the start and end. The anomalies for the NOAA and AGAGE-derived OH show periodic variations on a timescale of 2-3 yrs but with a phase shift between the two datasets of 3 months, as noted by Holmes et al., (2013). The model runs with OH variability prescribed from the observations and varying winds also show these periodic variations with correlation coefficients ranging from 0.71 – 0.90. The correlation values for these runs using varying OH are all larger than the run using repeating OH (for RE_FTVW R=0.62 compared to AGAGE data and 0.67 compared to NOAA data). Note that for CH_3CCl_3 decay there are only small differences between the 3-D simulations which use varying temperatures and the corresponding runs which use fixed temperature (e.g. simulation RE_VTVW versus RE_FTVW). This agrees with the results of Montzka et al (2011) based on their box model. This shows that the largest contribution from the CH_3CCl_3 decay rate anomaly comes from variations in atmospheric OH concentration, rather than atmospheric temperature. The simulations with repeating winds show less variability in the CH_3CCl_3 decay rate, particularly in the period 1999-2004, but the small difference suggests that the interannual variability in the observed CH_3CCl_3 decay rate is driven primarily by the variations in the OH concentration. The remaining interannual variability in run RE_FTFW is due to variations in emissions.

Figure 3 shows the CH_3CCl_3 decay and decay rate anomalies at four selected stations, two from the NOAA network and two from the AGAGE network. The good agreement in the global CH_3CCl_3 decay in Figure 2 is also seen at these individual stations. At the AGAGE stations of Mace Head and Gape Grim, the model runs with varying OH perform better in capturing the decay rate anomalies than the runs with repeating OH. However, the impact of variability in the winds (solid lines versus dotted lines) is more apparent at these individual stations compared to the global means. At the NOAA station of Mauna Loa the model run with varying OH and varying winds also appears to perform better in capturing the observed variability in CH_3CCl_3 decay. At the South Pole the observed variability is small, except in 2000-2002. This feature is not captured by the model.

In summary, Figures 2 and 3 show that the global OH fields that we have constructed from different datasets can perform well in capturing the decay of CH_3CCl_3 and its anomalies both globally and at individual stations. Although, the interannual variability in global mean OH has been derived from these CH_3CCl_3 observations, the figures do show that the reconstructed model OH fields (which also depend on the methodology discussed in Section 2) perform well in simulating CH_3CCl_3 within the 3D model. Therefore, we would argue that these fields are suitable for testing the impact of OH variability on the methane growth rate. Even so, it is important to bear in mind that these fields may not represent the true changes in atmospheric OH, particularly if the interannual variability in CH_3CCl_3 emissions was a lot different to that assumed here. However, we would again note that we are focussing on the impact of multi-year (≥2 years) variability which appears more robustly determined by the networks under differing assumptions of temperature and emissions than year-to-year variability.

3.2.2 Methane

Figure 4 shows deseasonalised modelled surface CH_4 from the 3-D CTM simulations compared with in-situ observations from a northern high-latitude station (Alert), two tropical stations (Mauna Loa and Tutuila), a southern high-latitude station (South Pole) and the global average of the NOAA and AGAGE stations. The global comparisons are shown for simulations both with varying and repeating meteorology. Figure 5 shows the global annual CH_4 growth rates with a 12-month smoothing (panel a) and differences between the model and NOAA and AGAGE observations (panels b and c). The changes in the modelled global mean CH_4 over different time periods are given in Table 3.

Figure 4 shows that in 1993, at the end of the model spin-up, the simulations capture the global mean CH_4 level well, along with the observed values at a range of latitudes. The exception is at high northern latitudes. However, these differences are not important when investigating the change in the global growth rate. The global change in atmospheric CH_4 in all~~the~~ simulations ~~with varying winds for~~from 1993 to the end of 2009 is between 75 and 104 ppb, compared to 56 and 66 ppb in the observations.

Model run RE_FTFW does not include interannual variations in atmospheric transport or CH_4 loss. Therefore, and also given the lack of change in emissions the modelled CH_4 gradually approaches a steady state value of ~1830 ppb (Figure 4f). The rate of CH_4 growth decreases from 7.9 ppb/yr (1993-1998) to 1.4 ppb/yr (2007-2009). Compared to run RE_FTFW, the other simulations introduce variability on this CH_4 evolution.

Run RE_FTVW includes interannual variability in wind fields which may alter the transport of CH_4 from the source (emission) to the sink regions. The largest difference between runs

RE_FTFW and RE_FTVW occurs after 2000 (Figure 4f). During the stagnation period (1999-2006) run RE_FTVW has a smaller growth rate of 3.5 ppb/yr compared to 4.1 ppb/yr in run RE_FTFW, showing that variations in atmospheric transport made a small contribution to the slowdown in global mean CH_4 growth.

Compared to run RE_FTVW, runs AP_FTVW, AL_FTVW, NP_FTVW and NL_FTVW include CH_3CCl_3-derived interannual variations in [OH] which introduce large changes in modelled CH_4, which are more in line with the observations (Figure 4e and 5). These runs produce turnarounds in the CH_4 growth in 2001/2 (becomes negative) and 2005/6 (returns to being positive). For AGAGE-derived [OH] (runs AP_FTVW, AL_FTVW) the large negative anomaly in OH in 1997 produces a significant increase in CH_4 prior to the turnround in 2001.

Table 3 summarises the change in global mean CH_4 over different time periods. These periods are defined by the key dates in the observed record, i.e. 1999 and 2006 as the start and end dates of the stagnation period. Comparison of Figure 4e and Table 3 shows, however, that the timing of the largest modelled change in growth rate do not necessarily coincide with those dates. That is understandable if other factors not considered here, e.g. emission changes, are contributing to the change in global CH_4 concentration. It does mean that the summary model values in Table 3 do not capture the full impact of the changes in [OH] and winds within the stagnation period. Figure 4e shows that model runs with varying OH perform better in simulating the relative CH_4 trend from 1999 to around 2004.

Table 3 shows that runs NP_FTVW and NL_FTVW (NOAA-derived [OH]) produce a small modelled CH_4 growth of 2.5-3.1 ppb/yr during the stagnation period 1999-2006, compared to 1.0 ppb/yr for run AP_FTVW (AGAGE-derived [OH]). The AGAGE results are slightly larger than the observed growth rate of 0.6-0.7 ppb/yr. Runs AL_FTVW, AP_FTVW, NL_FTVW and NP_FTVW capture the observed strong decrease in the CH_4 growth rate. With the exception of AP_FTVW between 1999 and 2006 (p-value = 0.37) all trends, over all three time periods, are statistically significant at the 90% level. Clearly, these runs demonstrate the significant potential for relatively small variations in mean [OH] to affect CH_4 growth. Excluding the stagnation period the mean modelled CH_4 lifetime in run NP_FTVW is 9.4 years, but this decreases slightly by 0.01 years during the stagnation period. For run AP_FTVW there is a decrease of 0.18 years from 9.6 years between the same intervals. The results from all the CTM simulations during 1999-2006 indicate that the accuracy of modelled CH_4 growth is improved by accounting for interannual variability in [OH] as derived from CH_3CCl_3 observations, and interannual variability in meteorology.

The variation of [OH] after 2007 cannot be determined from the available NOAA data so run NP_FTVW used the mean [OH] field for all subsequent years. The modelled CH_4 increase of 3.5 ppb/yr underestimates the observations (4.9 ppb/yr). Should the lower [OH] of 2007 have persisted then the model would have produced a larger increase in CH_4, in better agreement with the observations. The AGAGE-derived [OH] for 2007-2009 (run AP_FTVW) produces a larger CH_4 growth relative to the previous years (8.8 ppb/yr). Runs RE_FTFW (1.4 ppb/yr) and RE_FTVW (1.8 ppb/yr) both show a decreased rate of growth during the final 5 years, consistent with a system approaching steady state.

Figure 5a shows the global CH_4 growth rate derived from the AGAGE and NOAA networks together with selected model simulations. Figures 5b and c show the differences between the model simulations and the NOAA and AGAGE observations, respectively. The runs which include variations in [OH] agree better with the observed changes, i.e. larger R values in panel (a) and the model lines are closer to the y=0 line in panels (b) and (c), especially in the first 5 years of the stagnation period. It is interesting to note that the relative impacts of wind and temperature variations are larger for CH_4 than for CH_3CCl_3 (compare simulations RE_FTFW, RE_FTVW and RE_VTVW in Figures 2 and 5a). The temperature dependences of the OH loss reactions are similar for the two species (see Supplement S1) but the impact of transport from emisson~~s~~ regions to chemical loss regions is more variable for CH_4. One possibility for this is differences in the spatial distribution of CH_4 and CH_3CCl_3 emissions. This needs to be considered when applying results derived from CH_3CCl_3 to CH_4.

4. Discussion and Conclusions

Our model results suggest that variability in atmospheric [OH]~~ and transport~~ played a key role~~s~~ in the observed recent variations in CH_4 growth, particularly during the CH_4 stagnation period between 1999 and 2006. The 3-D CTM calculations show that during the stagnation period, variations in atmospheric conditions in the tropical lower to mid-troposphere could potentially account for an important component of the observed decrease in global CH_4 growth. Within this, small increases in [OH] were the largest factor, while variations in transport from source to sink regions made a smaller contribution. Note again, however, that the ultimate loss of CH_4 is still due to chemistry. The role of atmospheric temperature variations is factored into the observationally derived OH, but model experiments show that changes in the OH concentration itself is most important. The remainder of the variation can be ascribed to other processes not considered in our runs such as emission changes. There are also measurement uncertainties to consider and the possible underrepresentation of the global mean CH_3CCl_3 which will affect the derived OH concentration. Our results are consistent with an earlier budget study which analysed 1991 to 2004 and found that variations in [OH] were the main control of variations in atmospheric CH_4 lifetime (65%), with temperature accounting for a smaller fraction (35%) (Fiore et al., 2006). However, they were not able to study the full period of the pause in CH_4 growth and did not impose observatoion-based [OH] variations. As we have noted here the CH_4 lifetime can also be affected by emissions distributions which affects transport to the main loss regions.

Prior to the stagnation period the simulation using AGAGE-derived [OH] (9.7-10.4 ppb/yr) overestimates CH_4 growth when compared to observations (6.0-7.1 ppb/yr), which degrades the agreement with the observed CH_4 variations. A likely cause of this is inaccuracies in derived [OH] in 1997 when emissions still played a large role in the observed CH_3CCl_3 and the e-fold decay had not yet stabilised (Montzka et al., 2011).

We have not accounted for expected variations in CH_4 emissions in this study. We can conclude that although global CH_4 emissions do vary year-to-year, the observed trend in CH_4 growth between 1999 and 2006 was impacted by changing atmospheric processes that affected CH_4 loss. Changes in emissions are still important over this time period and likely still dominate

CH_4 variations over other time periods. The observed changes in growth rates during ENSO events in e.g. 1998 are poorly captured by the meteorological changes considered here and can be attributed to changes in emissions through changing precipitation and enhanced biomass burning (Hodson et al., 2011). The renewed growth of CH_4 in 2007 is also poorly captured by all model simulations without varying [OH]. The observed decrease in AGAGE and NOAA-derived [OH] coincides with the increase in CH_4 growth in 2007, although the currently available data do not allow for a more detailed investigation of the possible contribution of [OH] changes in this recent increase.

Despite the differences in year-to-year variability in [OH] derived from CH_3CCl_3 observations (Holmes et al., 2013), we find that [OH] variability derived from two different networks of surface CH_3CCl_3 observations over multi-year periods provide insights into atmospheric CH_4 variations. Improved quantification of the role of OH variability will require efforts to reduce uncertainties associated with estimating [OH]. Estimates of global mean [OH] in recent years from CH_3CCl_3 observations are~~is~~ becoming increasingly difficult because CH_3CCl_3 levels are currently <5 ppt; hence this may limit the accuracy of derived [OH] and its variability in future years (Lelieveld et al., 2006). Wennberg et al. (2004) also noted that there can be time variations in the ~~small uptake~~net flux of CH_3CCl_3 by the oceans, which could potentially~~also~~ affect the derived [OH] concentrations and which were not considered in our analysis. However, the impact of interannual variability in this flux are not likely to be important. For the period considered in this study, Figure 2 of Wennberg et al., (2004) shows that the CH_3CCl_3 flux into the ocean decreased from the largest value in 1997 to almost zero in recent years, which mimics CH_3CCl_3 emissions. Including the estimated 1997 ocean flux in our box model decreased the OH anomaly for that year by 0.8%. This change would decrease in magnitude in the subsequent years. ~~and are not considered here.~~ Overall, accurate estimates of [OH] beyond 2009 will require more sophisticated analysis of CH_3CCl_3 observations, derivation from other species or improved representation of [OH] in photochemical models.

Overall our study suggests that future atmospheric trends in CH_4 are likely to be strongly influenced by not only emissions but also by changes in processes that affect atmospheric loss. ~~The accuracy of predictions would therefore be improved by including variations in [OH] and meteorology~~. Therefore, to be realistic, predictions of these future trends need to explicitly account for likely variations in [OH],the major sink, and possibly other processes related to tropospheric and stratospheric chemistry.

Acknowledgements: JRM thanks NERC National Centre for Earth Observation (NCEO) for a studentship. CW, MPC and MG acknowledge support from NERC grants GAUGE (NE/K002244/1) and AMAZONICA (NE/F005806/1). GDH acknowledges support from the European Space Agency through its Support to Science Element initiative (ALANIS Methane), NCEO and the NERC MAMM grant (NE/I028327/1). SAM acknowledges support in part from NOAA Climate Program Office's AC4 program. AGAGE is supported by NASA grants NNX11AF17G to MIT and NNX11AF15G and NNX11AF16G to SIO, by NOAA, UK Department of Food and Rural Affairs (DEFRA) and UK Department for Energy and Climate Change (DECC) grants to Bristol University, and by CSIRO and Australian Bureau of

Meteorology. MR is supported by a NERC Advanced Fellowship (NE/I021365/1). Model calculations were performed on the Arc1 and Archer supercomputers.

References

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Villani, M. G., Houweling, S., Dentener, F., Dlugokencky, E. J., Miller, J. B., Gatti, L. V., Engel, A., and Levin, I.: Inverse modeling of global and regional CH_4 emissions using SCIAMACHY satellite retrievals, J. Geophys. Res., 114, 10.1029/2009jd012287, 2009.

Best, M. J., Pryor, M., Clark, D. B., Rooney, G. G., Essery, R. L. H., Ménard, C. B., Edwards, J. M., Hendry, M. A., Porson, A., Gedney, N., Mercado, L. M., Sitch, S., Blyth, E., Boucher, O., Cox, P. M., Grimmond, C. S. B., and Harding, R. J.: The Joint UK Land Environment Simulator (JULES), model description – Part 1: Energy and water fluxes, Geosci. Model Dev., 4, 677-699, 10.5194/gmd-4-677-2011, 2011.

Bousquet, P., Ciais, P., Miller, J. B., Dlugokencky, E. J., Hauglustaine, D. A., Prigent, C., Van der Werf, G. R., Peylin, P., Brunke, E. G., Carouge, C., Langenfelds, R. L., Lathiere, J., Papa, F., Ramonet, M., Schmidt, M., Steele, L. P., Tyler, S. C., and White, J.: Contribution of anthropogenic and natural sources to atmospheric methane variability, Nature, 443, 439-443, 10.1038/nature05132, 2006.

Bousquet, P., Ringeval, B., Pison, I., Dlugokencky, E. J., Brunke, E. G., Carouge, C., Chevallier, F., Fortems-Cheiney, A., Frankenberg, C., Hauglustaine, D. A., Krummel, P. B., Langenfelds, R. L., Ramonet, M., Schmidt, M., Steele, L. P., Szopa, S., Yver, C., Viovy, N., and Ciais, P.: Source attribution of the changes in atmospheric methane for 2006–2008, Atmos. Chem. Phys., 11, 3689-3700, 10.5194/acp-11-3689-2011, 2011.

~~Carn, S. A., Yang, K., Prata, A. J., and Krotkov, N. A.: Extending the long-term record of volcanic~~ ~~SO_2~~ ~~emissions with the Ozone Mapping and Profiler Suite nadir mapper, Geophys. Res. Lett., 42, 925–932, 2015.~~

Chipperfield, M. P.: New version of the TOMCAT/SLIMCAT off-line chemical transport model: Intercomparison of stratospheric tracer experiments. Q. J. R. Meteorol Soc., 132, 1179–1203, 2006.

Ciais, P., Sabine, C., Bala, G., Bopp, L., Brovkin, V., Canadell, J., Chhabra, A., DeFries, R., Galloway, J., Heimann, M., Jones, C., Le Quere, C., Myneni, R. B., Piao, S., and Thornton, P.: Carbon and other biogeochemical cycles, in: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, 2013.

Cicerone, R. J., and Oremland, R. S.: Biogeochemical aspects of atmospheric methane, Global Biogeochem. Cycles, 2, 299-327, 1988.

Clark, D. B., Mercado, L. M., Sitch, S., Jones, C. D., Gedney, N., Best, M. J., Pryor, M., Rooney, G. G., Essery, R. L. H., Blyth, E., Boucher, O., Harding, R. J., Huntingford, C., and Cox, P. M.: The Joint UK Land Environment Simulator (JULES), Model description – Part 2: Carbon fluxes and vegetation, Geosci. Model Dev., 4, 701-722, 10.5194/gmd-4-701-2011, 2011.

Cunnold, D., Steele, L., Fraser, P., Simmonds, P., Prinn, R., Weiss, R., Porter, L., O'Doherty, S., Langenfelds, R., and Krummel, P.: In situ measurements of atmospheric methane at

GAGE/AGAGE sites during 1985–2000 and resulting source inferences, J. Geophys. Res., 107, ACH 20-21-ACH 20-18, 2002.

Curry, C. L.: Modeling the soil consumption of atmospheric methane at the global scale, Global Biogeochem. Cycles, 21, GB4012, doi:10.1029/2006GB002818, 2007.

Dee, D., Uppala, S., Simmons, A., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M., Balsamo, G., Bauer, P., et al.: The ERA‐Interim reanalysis: Configuration and performance of the data assimilation system, Q. J. R. Meteorol. Soc., 137, 553-597, 2011.

Dlugokencky, E. J., Houweling, S., Bruhwiler, L., Masarie, K., Lang, P., Miller, J., and Tans, P.: Atmospheric methane levels off: Temporary pause or a new steady‐state?, Geophys. Res. Lett., 30, doi:10.1029/2003GL018126, 2003.

Dlugokencky, E. J., Myers, R., Lang, P., Masarie, K., Crotwell, A., Thoning, K., Hall, B., Elkins, J., and Steele, L.: Conversion of NOAA atmospheric dry air CH_4 mole fractions to a gravimetrically prepared standard scale, J. Geophys. Res., 110, D18306, 2005.

Dlugokencky, E. J., Nisbet, E. G., Fisher, R., and Lowry, D.: Global atmospheric methane: budget, changes and dangers, Philos. Trans. R. Soc. A, 369, 2058-2072, 10.1098/rsta.2010.0341, 2011.

Dlugokencky, E. J., P.M. Lang, A.M. Crotwell, K.A. Masarie, M.J. Crotwell, Atmospheric Methane Dry Air Mole Fractions from the NOAA ESRL Carbon Cycle Cooperative Global Air Sampling Network, 1983-2013, Version: 2014-06-24. Available at ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/flask/surface/. Accessed July 6, 2014.

Etheridge, D. M., Steele, L. P., Francey, R. J., and Langenfelds, R. L.: Atmospheric methane between 1000 A.D. and present: Evidence of anthropogenic emissions and climatic variability, J. Geophys. Res., 103, 15,979-15,993, 10.1029/98jd00923, 1998.

Fiore, A. M., Horowitz, L. W., Dlugokencky, E. J., and West, J. J.: Impact of meteorology and emissions on methane trends, 1990–2004, Geophys. Res. Lett., 33, L12809, 10.1029/2006gl026199, 2006.

Hayman, G.D., et al., Comparison of the HadGEM2 climate-chemistry model against in-situ and SCIAMACHY atmospheric methane data, Atmos. Chem. Phys., 14, 13,257-13,280, 2014.

Hodson, E. L., Poulter, B., Zimmermann, N. E., Prigent, C., and Kaplan, J. O.: The El Niño-Southern Oscillation and wetland methane interannual variability, Geophys. Res. Lett., 38, L08810, 10.1029/2011gl046861, 2011.

Holmes, C. D., Prather, M. J., Søvde, O., and Myhre, G.: Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions, Atmos. Chem. Phys., 13, 285-302, 2013.

Huijnen, V., Williams, J., Weele, M. v., Noije, T. v., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., and de Laat, J.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, Geosci. Model Dev., 3, 445-473, 2010.

Ito, A., and Inatomi, M.: Use of a process-based model for assessing the methane budgets of global terrestrial ecosystems and evaluation of uncertainty, Biogeosciences, 9, 759-773, 10.5194/bg-9-759-2012, 2012.

Karlsdottir, S. and Isaksen, I.S.A.: Changing methane lifetime: Possible cause for reduced growth. Geophys. Res. Lett., 27(1), 93-96, 2000.

Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J., Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., et al.: Three decades of global methane sources and sinks, Nature Geosci., 6, 813-823, 2013.

Lelieveld, J., Dentener, F., Peters, W., and Krol, M.: On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere, Atmos. Chem. Phys., 4, 2337-2344, 2004.

Lelieveld, J., Brenninkmeijer, C. A. M., Joeckel, P., Isaksen, I. S. A., Krol, M. C., Mak, J. E., Dlugokencky, E., Montzka, S. A., Novelli, P. C., Peters, W. and Tans, P. P.: New Directions: Watching over tropospheric hydroxyl (OH), Atmospheric Environment, 40, 5741-5743, 2006.

Matthews, E., and Fung I.: Methane emissions from natural wetlands: Global distribution, area, and ecology of sources. Global Biogeochem. Cycles, 1, 61–86, 1987.

~~Mills, M. J., Schmidt, A., Easter, R., Solomon, S., Kinnison, D. E., Ghan, S. J., Neely III, R. R., Marsh, D. R., Conley, A., Bardeen, C. G., and Gettelman, A., Global volcanic aerosol properties derived from emissions, 1990-2014, using CESM1(WACCM), J. Geophys. Res. (under review), 2015.~~

Montzka, S. A., Krol, M., Dlugokencky, E., Hall, B., Jöckel, P., and Lelieveld, J.: Small interannual variability of global atmospheric hydroxyl, Science, 331, 67-69, 2011.

Myhre, G., Shindell, D., Bréon, F., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., and Zhang, H.: Anthropogenic and natural radiative forcing, in: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge University Press, 2013.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L., Lamarque, J.-F., Lin, M., Prather, M. J., Young, P., Bergmann, D., and Cameron-Smith, P.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 5277-5298, 2013.

Nisbet, E. G., Dlugokencky, E. J., and Bousquet, P.: Atmospheric science. Methane on the rise - again, Science, 343, 493-495, 10.1126/science.1247828, 2014.

Olivier, J.G.J., Berdowski J.J.M.: Global emissions sources and sinks, in: The Climate System, edited by: Berdowski J, Guicherit R, and Heij BJ., IISBN 9058092550, A. A. Balkema Publishers/Swets & Zeitlinger Pub., Lisse, The Netherlands, 33–78, 2001.

Olivier, J. G., Van Aardenne, J. A., Dentener, F. J., Pagliari, V., Ganzeveld, L. N., and Peters, J. A.: Recent trends in global greenhouse gas emissions: regional trends 1970–2000 and spatial distributionof key sources in 2000, Environmental Sciences, 2, 81-99, 2005.

Patra, P. K. , Takigawa, M., Ishijima, K., Choi, B.-C., Cunnold, D., J. Dlugokencky, E., Fraser, P., J. Gomez-Pelaez, A., Goo, T.-Y., Kim, J.-S., Krummel, P., Langenfelds, R., Meinhardt, F., Mukai, H., O'Doherty, S., G. Prinn, R., Simmonds, P., Steele, P., Tohjima, Y., Tsuboi, K., Uhse, K., Weiss, R., Worthy, D., and Nakazawa, T.: Growth rate, seasonal, synoptic, diurnal variations and budget of methane in the lower atmosphere, J. Meteorol. Soc. Japan, 87, 635-663, 10.2151/jmsj.87.635, 2009.

Patra, P. K., Houweling, S., Krol, M., Bousquet, P., Belikov, D., Bergmann, D., Bian, H., Cameron-Smith, P., Chipperfield, M. P., and Corbin, K.: TransCom model simulations of CH_4 and related species: linking transport, surface flux and chemical loss with CH_4 variability in the troposphere and lower stratosphere, Atmos. Chem. Phys., 11, 12,813-12,837, 2011.

Patra, P., Krol, M., Montzka, S., Arnold, T., Atlas, E., Lintner, B., Stephens, B., Xiang, B., Elkins, J., and Fraser, P.: Observational evidence for interhemispheric hydroxyl-radical parity, Nature, 513, 219-223, 2014.

Pison, I., Bousquet, P., Chevallier, F., Szopa, S., and Hauglustaine, D.: Multi-species inversion of CH_4, CO and H_2 emissions from surface measurements, Atmos. Chem. Phys., 9, 5281-5297, 2009.

Pison, I., Ringeval, B., Bousquet, P., Prigent, C., and Papa, F.: Stable atmospheric methane in the 2000s: key-role of emissions from natural wetlands, Atmos. Chem. Phys., 13, 11,609-11,623, 10.5194/acp-13-11609-2013, 2013.

Prinn, R., Weiss, R., Fraser, P., Simmonds, P., Cunnold, D., Alyea, F., O'Doherty, S., Salameh, P., Miller, B., and Huang, J.: A history of chemically and radiatively important gases in air deduced from ALE/GAGE/AGAGE, J. Geophys. Res., 105, 17,751-17,792, 2000.

Prinn, R. G.: Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, Geophys. Res. Lett., 32, L07809, 10.1029/2004gl022228, 2005.

Prinn, R.G., R.F. Weiss, P.J. Fraser, P.G. Simmonds, S. O'Doherty, P. Salameh, L. Porter, P. Krummel, R.H.J. Wang, B. Miller, C. Harth, B. Greally, F.A. Van Woy, L.P. Steele, J. Mühle, G. Sturrock, F.N. Alyea, J. Huang, and D.E. Hartley, The ALE / GAGE AGAGE Network (DB1001), Carbon Dioxide Information Analysis Center (CDIAC), U.S. Department of Energy (DOE), http://cdiac.esd.ornl.gov/ndps/alegage.html, 2015.

Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R., Huang, J., Cunnold, D. M., Steele, L. P., Krummel, P. B., and Weiss, R. F.: Renewed growth of atmospheric methane, Geophys. Res. Lett., 35, L22805, 2008.

Rigby, M., Prinn, R. G., O'Doherty, S., Montzka, S. A., McCulloch, A., Harth, C. M., Mühle, J., Salameh, P., Weiss, R. F., and Young, D.: Re-evaluation of the lifetimes of the major CFCs and CH_3CCl_3 using atmospheric trends, Atmos. Chem. Phys., 13, 2691-2702, 2013.

Sander, S.P., et al., Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies Evaluation Number 17. *JPL Publication* 10-6, Jet Propulsion Laboratory, Pasadena, USA, 2011.

Spahni, R., Wania, R., Neef, L., van Weele, M., Pison, I., Bousquet, P., Frankenberg, C., Foster, P. N., Joos, F., Prentice, I. C., and van Velthoven, P.: Constraining global methane emissions and uptake by ecosystems, Biogeosciences, 8, 1643-1665, 10.5194/bg-8-1643-2011, 2011.

Spivakovsky, C., Logan, J., Montzka, S., Balkanski, Y., Foreman-Fowler, M., Jones, D., Horowitz, L., Fusco, A., Brenninkmeijer, C., and Prather, M.: Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, J. Geophys. Res., 105, 8931-8980, 2000.

UNEP, The UNEP Environmental Data Explorer, as compiled from United Nations Envrionment Programme . United Nations Environment Programme. http://ede.grid.unep.ch, 2015.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R., Jin, Y. v., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707-11735, 2010.

Velders, G. J. M.: Description of the RIVM 2-dimensional stratosphere model, RIVM Rapport 722201002, 1995.

Voulgarakis, A., Naik, V., Lamarque, J.-F., Shindell, D. T., Young, P., Prather, M. J., Wild, O., Field, R., Bergmann, D., and Cameron-Smith, P.: Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, Atmos. Chem. Phys., 13, 2563-2587, 2013.

Voulgarakis, A., Marlier, M.E., Faluvegi, G., Shindell, D.T., Tsigaridis, K. and Mangeon, S.: Interannual variability of tropospheric trace gases and aerosols: The role of biomass burning emissions, J. Geophys. Res.: Atmos., 120(14), 7157-7173, 2015.

Wang, J.S., Logan, J.A., McElroy, M.B., Duncan, B.N., Megretskaia, I.A. and Yantosca, R.M.: A 3‐D model analysis of the slowdown and interannual variability in the methane growth rate from 1988 to 1997, Global Biogeochem. Cycles, 18(3), 2004.

Warwick, N. J., Bekki S., Law K. S., Nisbet E. G., and Pyle, J. A.: The impact of meteorology on the interannual growth rate of atmospheric methane, Geophys. Res. Lett., 29, doi:10.1029/2002GL015282, 2002.

Wennberg, P. O., Peacock, S., Randerson, J. T., and Bleck, R.: Recent changes in the air‐sea gas exchange of methyl chloroform, Geophys. Res. Lett., 31, L16112, 2004.

Yan, X., Akiyama, H., Yagi, K., and Akimoto, H.: Global estimations of the inventory and mitigation potential of methane emissions from rice cultivation conducted using the 2006 Intergovernmental Panel on Climate Change Guidelines, Global Biogeochem. Cycles, 23, GB2002, 10.1029/2008gb003299, 2009.

Tables

Table 1. List of NOAA and AGAGE stations which provided CH_4 and CH_3CCl_3 observations.

Site Code	Site Name	Lat. (°N)	Lon. (°N)	Altitude (km)	CH_4	CH_3CCl_3	Start Date^{++}	End Date
ABP	Arembepe, Brazil	-12.77	-38.17	0	NOAA		27/10/2006	12/01/2010
ALT	Alert, Canada	82.45	-62.51	0.2	NOAA	NOAA	10/06/1985	Ongoing
ASC	Ascension Island, UK	7.97	-14.4	0.09	NOAA		11/05/1983	Ongoing
BRW	Barrow, USA	71.32	-156.61	0.01	NOAA	NOAA	06/04/1983	Ongoing
CGO	Cape Grim, Australia	-40.68	144.69	0.09	NOAA/AGAGE	NOAA/AGAGE	19/04/1984	Ongoing
HBA	Halley Station, UK	-75.61	-26.21	0.03	NOAA		17/01/1983	Ongoing
ICE	Storhofdi, Iceland	63.4	-20.29	0.12	NOAA		02/10/1992	Ongoing
KUM	Cape Kumukahi, USA	19.5	-154.8	0.02	-	NOAA	-	-
LEF	Park Falls, USA	45.9	-90.3	0.47	-	NOAA	-	-
MHD	Mace Head, Ireland	53.33	-9.9	0.01	NOAA/AGAGE	AGAGE**	03/06/1991	Ongoing
MLO	Mauna Loa, USA	19.54	-155.58	3.4	NOAA	NOAA	06/05/1983	Ongoing
NWR	Niwot Ridge, USA	40.05	-105.59	3.52	NOAA	NOAA	21/06/1983	Ongoing
PAL	Pallas-Sammaltunturi, Finland	67.97	24.12	0.56	NOAA		21/12/2001	Ongoing
PSA	Palmer Station, USA	-64.92	-64	0.01	NOAA	**	01/01/1983	Ongoing
RPB	Ragged Point, Barbados	13.17	-59.43	0.02	NOAA/AGAGE	AGAGE	14/11/1987	Ongoing
SEY	Mahe Island, Seychelles	-4.68	55.53	0	NOAA		12/05/1983	Ongoing
SMO	Tutuila, American Samoa	-14.25	-170.56	0.04	NOAA	NOAA/AGAGE	23/04/1983	Ongoing
SPO	South Pole, USA	-89.98	-24.8	2.81	NOAA	NOAA	20/02/1983	Ongoing
STM	Ocean Station M, Norway	66	2	0	NOAA		29/04/1983	27/11/2009
SUM	Summit, Greenland	72.6	-38.42	3.21	NOAA	**	23/06/1997	Ongoing
THD	Trinidad Head, USA	41.1	-124.1	0.1	AGAGE	AGAGE**	09/1995	Ongoing
ZEP	Ny-Alesund, Norway/Sweden	78.91	11.89	0.47	NOAA		11/02/1994	Ongoing

++For NOAA CH_3CCl_3 data the record starts in 1992 at 7 of the 9 stations used here. It started in 1995 for KUM and 1996 for LEF.

**NOAA flask data from these sites was not used in the present study or in Montzka et al., (2011).

Table 2. Summary of the fifteen TOMCAT 3-D CTM simulations.

Run	OH time variation	Meteorology[b]	
		Winds[c]	Temperature[d]
RE_FTFW	Repeating[a]	Fixed	Fixed
RE_FTVW	Repeating[a]	Varying	Fixed
RE_VTVW	Repeating[a]	Varying	Varying
AP_FTFW	AGAGE (Rigby et al., 2013)	Fixed	Fixed
AP_FTVW	AGAGE (Rigby et al., 2013)	Varying	Fixed
AP_VTVW	AGAGE (Rigby et al., 2013)	Varying	Varying
AL_FTVT	AGAGE (this work)	Fixed	Fixed
AL_FTVW	AGAGE (this work)	Varying	Fixed
AL_VTVW	AGAGE (this work)	Varying	Varying
NP_FTFW	NOAA (Montzka et al., 2011)	Fixed	Fixed
NP_FTVW	NOAA (Monztka et al., 2011)	Varying	Fixed
NP_VTVW	NOAA (Montzka et al., 2011)	Varying	Varying
NL_FTFW	NOAA (this work)	Fixed	Fixed
NL_FTVW	NOAA (this work)	Varying	Fixed
NL_VTVW	NOAA (this work)	Varying	Varying

a. Annually repeating [OH] taken from Patra et al. (2011).
b. Varying winds and temperatures are from ERA-Interim.
c. Fixed winds using repeating ERA-Interim winds from 1996.
d. Fixed temperatures use zonal mean ERA-Interim temperatures averaged over 1993-2009.

Table 3. Calculated methane changes over different time periods from selected TOMCAT experiments and the NOAA and AGAGE observation networks.

Model run or observation network	Global mean ΔCH_4 in ~~/~~ppb (ppb/yr)			
	2009-1993	1998-1993	2006-1999	2009-2007
RE_FTFW	85.0 (5.0)	47.2 (7.9)	32.9 (4.1)	4.3 (1.4)
RE_FTVW	82.2 (4.8)	48.2 (8.0)	27.8 (3.5)	5.4 (1.8)
RE_VTVW	74.6 (4.4)	45.6 (7.6)	23.1 (2.9)	5.3 (1.8)
AP_FTVW[a]	97.7[e] (5.7)	62.3[e] (10.4)	8.2[g] (1.0)	26.4 (8.8)
AL_FTVW[b]	104.2[e] (6.1)	58.4[e] (9.7)	17.3 (2.2)	27.5 (9.2)
NP_FTVW[c]	86.2[f] (5.1)	49.7[f] (8.3)	24.8 (3.1)	10.6[f] (3.8)
NL_FTVW[d]	91.4[f] (5.4)	58.8[f] (9.8)	20.1 (2.5)	11.3[f] (3.8)
NOAA obs.	56.1 (3.3)	36.0 (6.0)	4.8 (0.6)	14.7 (4.9)
AGAGE obs.	66.3 (3.9)	42.6 (7.1)	5.6 (0.7)	17.4 (5.8)

a. Taken from Rigby et al. (2013) and Patra et al. (2011).

b. Using 1997-2009 relative annual changes in mean [OH] derived from AGAGE data (Cunnold et al., 2002).

c. Taken from Montzka et al. (2011) and Patra et al. (2011).

d. Using 1997-2007 relative annual changes in mean [OH] derived from NOAA data (Prinn et al., 2015).

e. Value using mean [OH] from 1993-1996.

f. Value using mean [OH] from 1993-1996 and 2008-2011.

g. Trend value not statistically significant at the 90% level.

Figures

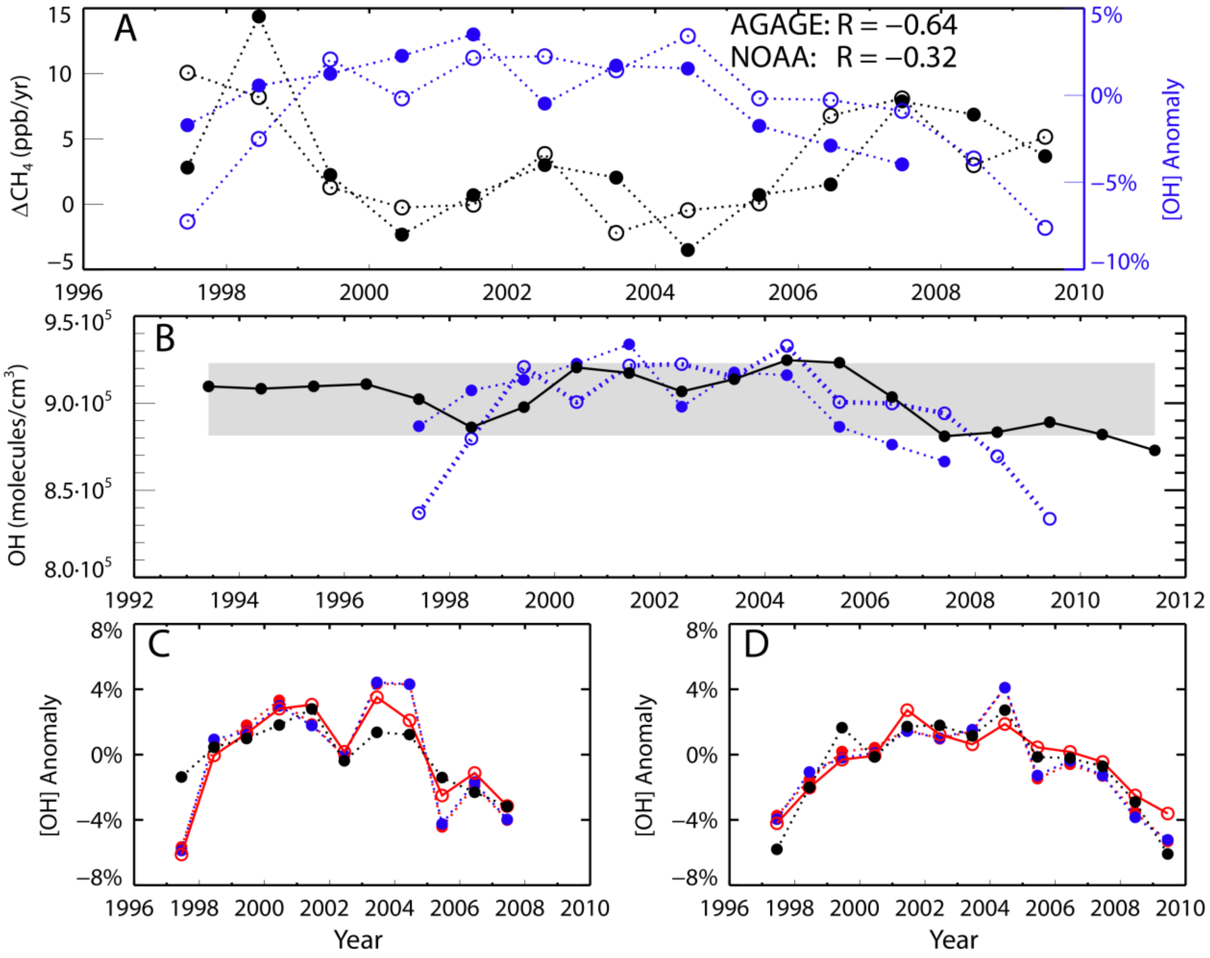

Figure 1. (a) Annual global CH_4 growth rate (ppb/yr) derived from NOAA (filled black circles) and AGAGE (open black circles) data (left hand y-axis), and published annual global [OH] anomalies derived from NOAA (filled blue circles, 1997-2007) and AGAGE (open blue circles, 1997-2009) CH_3CCl_3 measurements (right hand y-axis) (see text). (b) Annual mean [OH] (molecules/cm^3) required for global box model (see Supplement S1) to fit yearly variations in NOAA CH_4 observations assuming constant emissions and temperature (E=553 Tg/yr, T=272.9 K), based on Montzka et al. (2011) (solid black line). The shaded region denotes [OH] deviation of ±2.3% from the 1993-2011 mean. Also shown are the NOAA- and AGAGE-derived anomalies from panel (a) for an assumed mean OH (see Section 2.1). (c) Our estimates of [OH] derived from NOAA CH_3CCl_3 calculated using a global box model (Supplement S1) using repeating (blue) and varying (red) annual mean temperature and the CH_3CCl_3 emission scenario from UNEP (2015) (filled circles and dashed lines). Also shown for varying temperatures are results using the emissions of Montzka et al (2011) (red open circles and solid line) based on (Prinn et al. 2005) and the NOAA-derived values from panel (a) (black dashed line and circles). (d) As panel (c) but for OH derived from AGAGE CH_3CCl_3 observations.

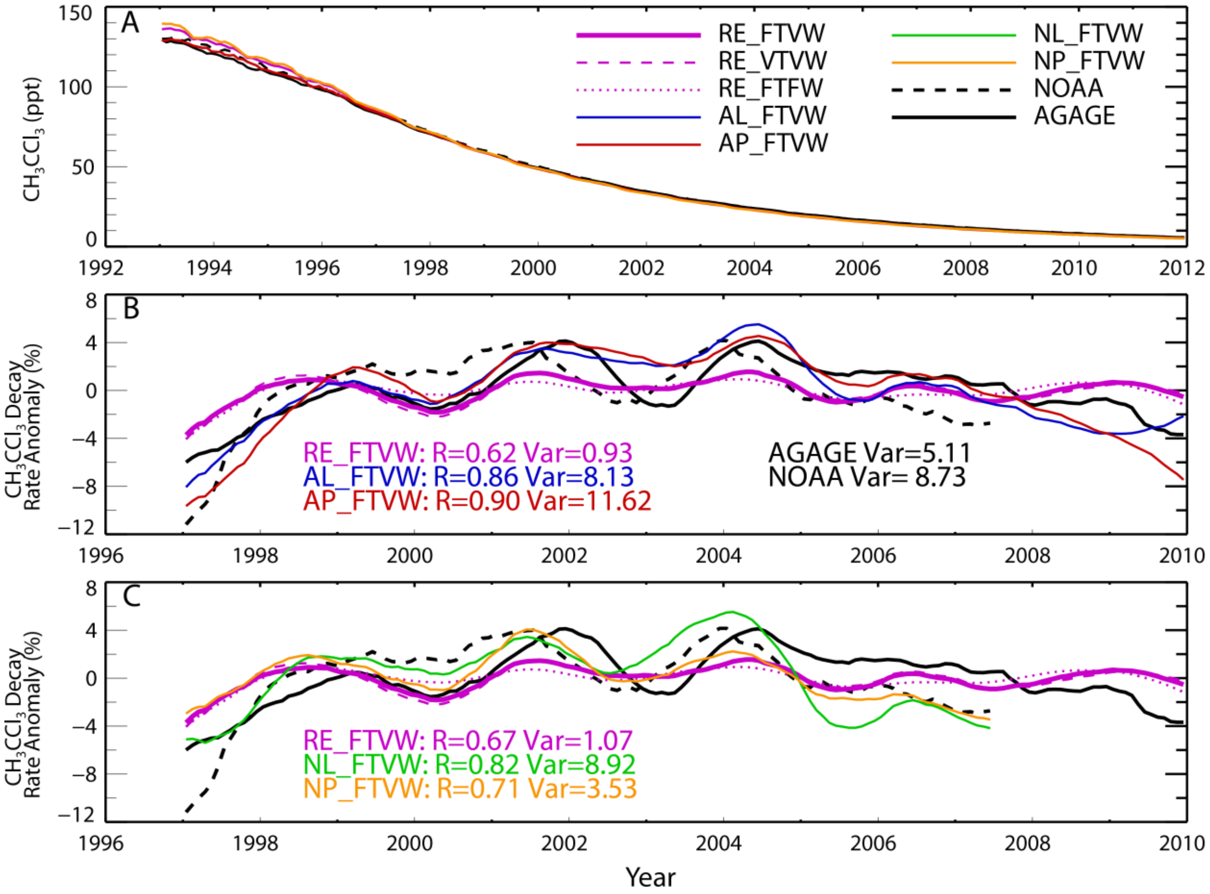

Figure 2. (a) Global mean surface CH_3CCl_3 (ppt) from NOAA (black dashed) and AGAGE (black solid) observations from 1993 to 2012. Also shown are results from five TOMCAT simulations with fixed temperatures and varying winds (see Table 1). (b) Global surface CH_3CCl_3 decay rate anomalies from NOAA and AGAGE along with model runs RE_FTVW, AL_FTVW and AP_FTVW (solid lines). Results from runs RE_FTFW and RE_VTVW are shown as a purple dotted line and dashed line, respectively. Observation and model anomalies are smoothed with a 12-month running average. Values given represent correlation coefficient when compared to AGAGE observations and variance. The decay rate anomaly is calculated from global mean CH_3CCl_3 values using equation (1) from Holmes et al., (2013), expressed as a percentage of the typical decay with a 12-month smoothing. (c) As panel (b) but for model runs NL_FTVW and NP_FTVW, along with RE_FTVW, RE_VTVW and RE_FTFW, and correlation cofficients for comparison with NOAA observations. The model results are split across panels (b) and (c) for clarity.

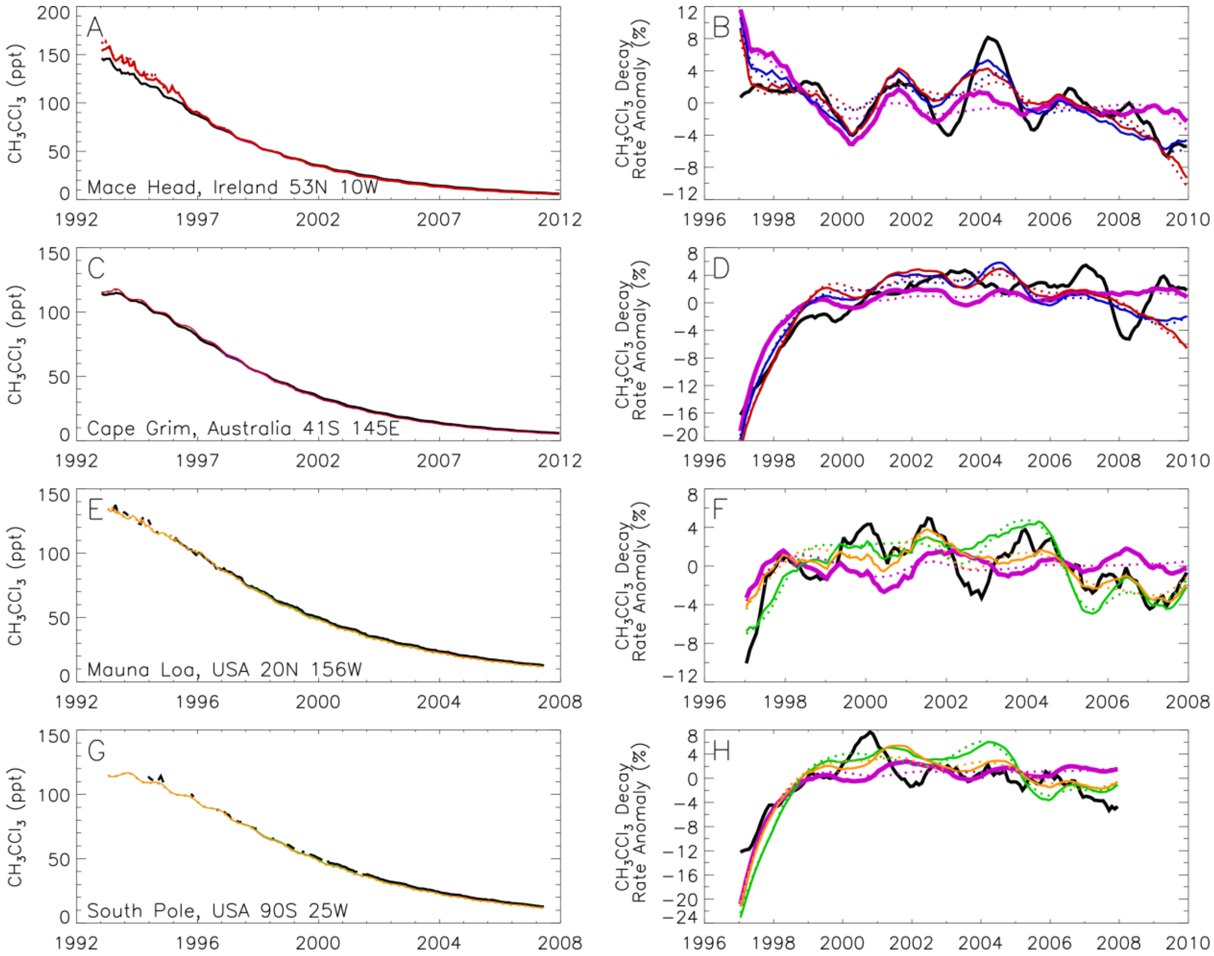

Figure 3. (Left) Observed mean surface CH_3CCl_3 (ppt) (black line) from (a) Mace Head (AGAGE), (c) Cape Grim (AGAGE), (e) Mauna Loa (NOAA) and (g) South Pole (NOAA). Also shown are results from five TOMCAT simulations with fixed temepartures and varying winds (FTVW, for legend see Figure 2a). (Right): Surface CH_3CCl_3 decay rate anomalies at the same station as the corresponding left column plot for observations (black), TOMCAT simulations with varying winds (FTVW, solid coloured lines) and TOMCAT simulations with fixed winds (FTFW, dotted lines). Comparisons at NOAA (AGAGE) stations show only comparisons with runs using NOAA (AGAGE)-derived OH, along with runs RE_FTVW and RE_FTFW in all panels.

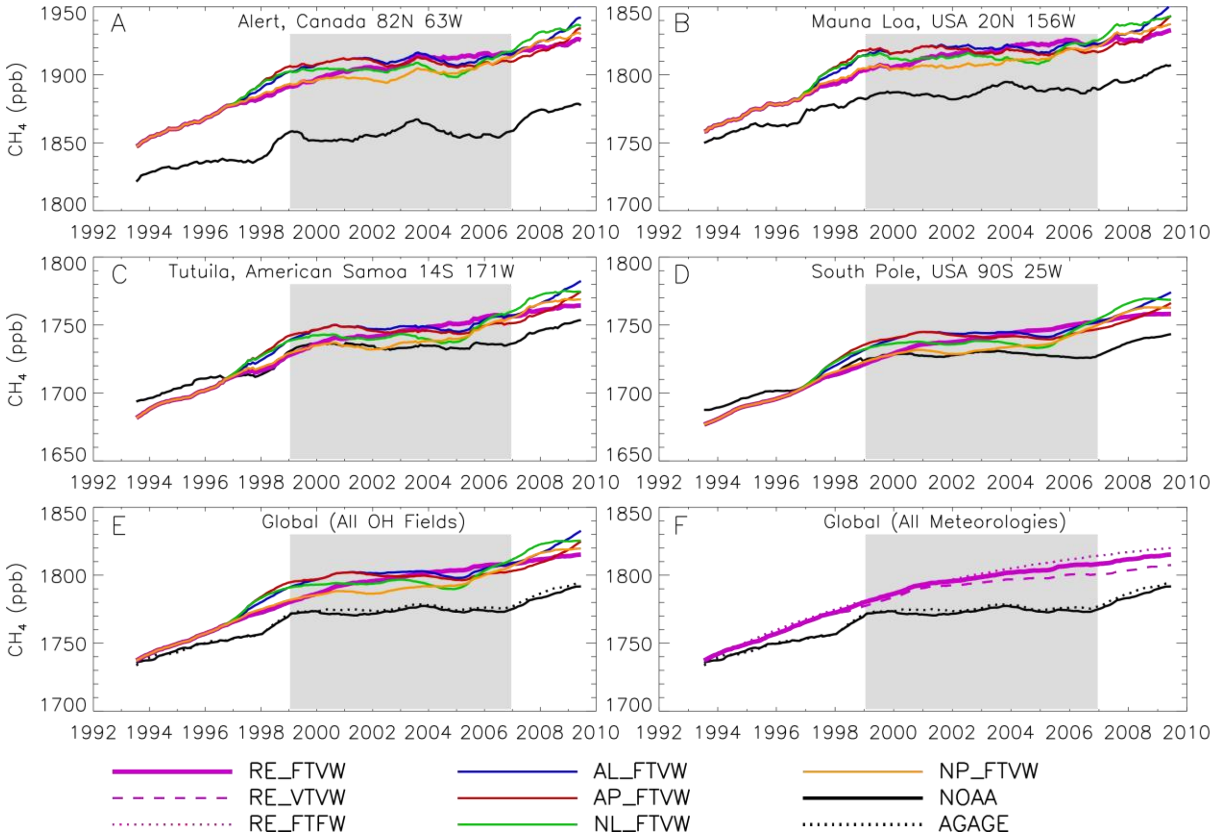

Figure 4. (a, b, c and d) Deasonalised surface CH_4 (ppb) from 4 NOAA sites (black solid line) from 1993 to 2009. Also shown are results from five TOMCAT 3-D CTM simulations with fixed temperatures and varying winds (FTVW, see **Table 2**). (e) Deasonalised global mean surface CH_4 from NOAA (black solid) and AGAGE (black dashed) observations along with five TOMCAT simulations with different treatments of OH. (f) Same as (e) but for TOMCAT simulations using repeating OH (RE) and different treatments of winds and temperature. All panels use observation and model values which are smoothed with a 12-month running average. The shaded region marks the stagnation period in the observed CH_4 growth rate.

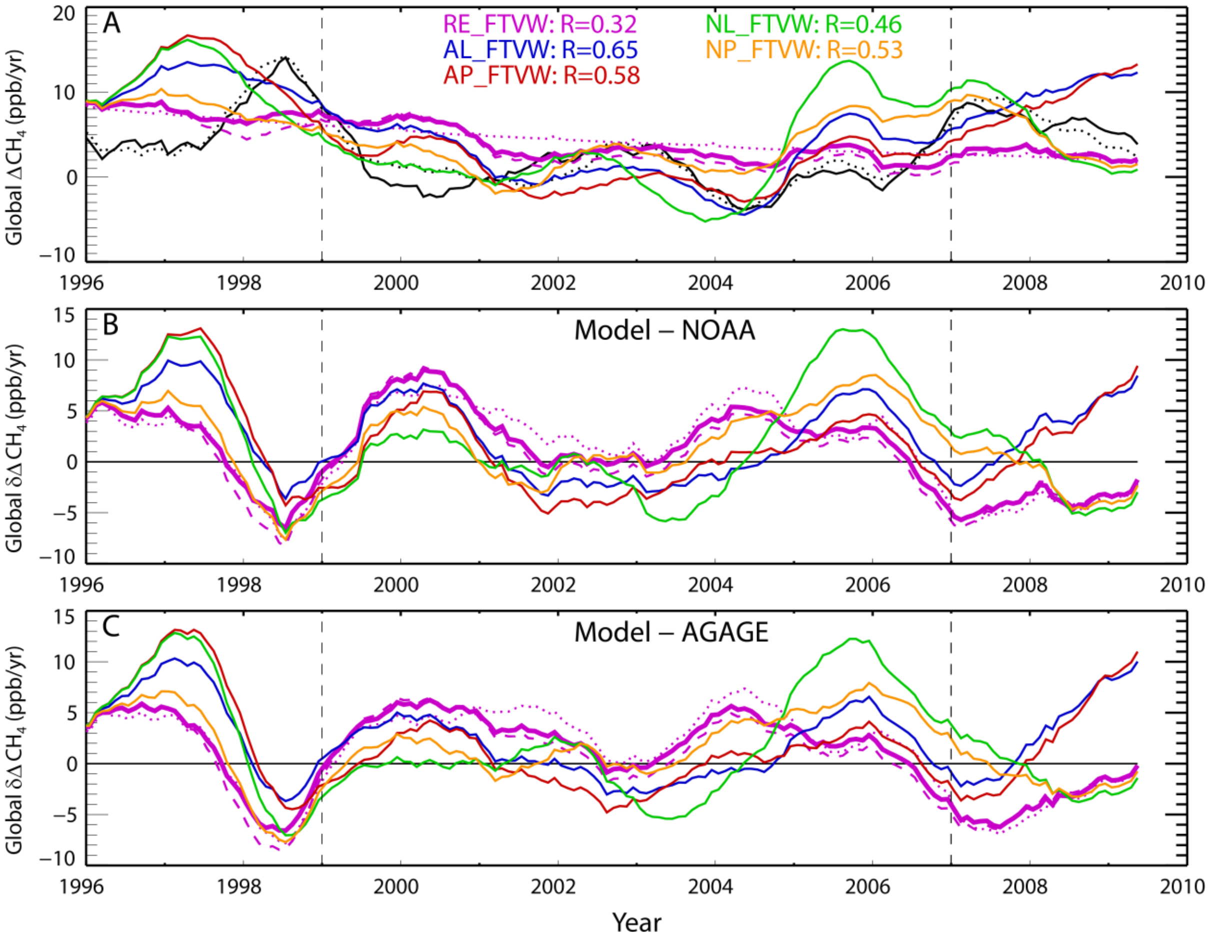

Figure 5. (a) The smoothed variation in the global annual CH_4 growth rate (ppb/yr) derived from NOAA (black solid) and AGAGE (black dashed) observations. Also shown are the smoothed growth rates from five TOMCAT 3-D CTM simulations with fixed temperatures and varying winds (FTVW, see Table 1). Values in legend give correlation coefficient between model run and NOAA observations. Also shown are results from runs RE_FTFW and RE_VTVW as a purple dotted line and dashed line, respectively (b) The difference in smoothed growth rate between TOMCAT simulations and NOAA observations shown in panel (a). (c) Same as (b) except using differences compared to AGAGE observations. The vertical dashed lines mark the start and end of the stagnation period in the observed CH_4 growth rate (1999 – 2006).

Role of OH variability in the stalling of the global atmospheric CH_4 growth rate from 1999 to 2006 by J. McNorton et al.

Response to Reviewers' Comments

We thank the reviewers for their time and constructive comments. These comments are repeated below (in normal text) followed by our responses *(in blue italics)*.

Anonymous Referee #1

GENERAL COMMENTS AND MAJOR SPECIFIC COMMENTS

This manuscript by J. McNorton et al. describes a set of chemical transport model simulations of atmospheric CH4 during the 1990s through 2000s that use specified OH fields and year-to-year OH anomalies derived from CH3CCl3 measurements by previous studies and by the authors. The authors conclude that OH variations could explain a significant portion of the observed changes in CH4 growth rate, including a drop to near zero during 1999-2006, with smaller contributions to the trends from variations in atmospheric transport and temperature.

Overall, I think this manuscript meets basic requirements for a publishable paper and has some good qualities, though it is somewhat thin on content. In its current form, it is perhaps more suited as a "letter" rather than a full-length article. Some of the work reported in the paper is mostly a confirmation that the authors can reproduce the results reported previously by others, particularly the yearly global OH anomalies derived by the authors from CH3CCl3 using a box model. And in my judgement, the paper makes a relatively small contribution to the body of scientific work, given that much of the work is not original or especially innovative. For example, the investigators used an OH distribution and yearly anomalies calculated by others. Also, the effects of transport and temperature on global CH4 loss have already been studied by others (e.g. the Warwick et al. (2002) and Fiore et al. (2006) papers cited in this paper), though perhaps not for the CH4 "stagnation period" that the current paper focusses on. Despite the shortcomings, I think the paper could become more suited for publication in *ACP* if the authors address my comments, in the process increasing the content of the paper. I do think the authors have done a good job of performing sets of CH3CCl3 and CH4 simulations that test various potential influences on CH4 trends, displaying the results thoroughly in figures and tables, and being candid about caveats and limitations of the study.

We thank the reviewer for his/her detailed review and we will make changes to the manuscript accordingly. We acknowledge that we have we have used OH anomalies calculated by others but we wanted to use the published data where available. By also using our own box model we were able to investigate differences between the two published OH anomaly datasets which were produced by different methods and based on different CH3Cl3 observations. Although the other studies noted above did look at transport and temperature effects, they did not look at the CH4 stagnation period which is of high current scientific interest.

One major specific comment is that I'm not convinced that the year-to-year variations in OH can be estimated with a high level of certainty from CH3CCl3 measurements, given various uncertainties in the modeling, including assumed emissions (especially when emissions were still significant prior to around 2000). The authors themselves acknowledge some discrepancies between their estimated OH anomalies and those of published studies (page 6, lines 208-216).

Thus, I see the findings on the contribution of OH variability to CH4 trends as somewhat speculative. The higher correlations of the varying-OH runs with the observed CH4 growth compared to the repeating-OH run in Fig. 5 could be a coincidence. A related comment is that the sub-periods delineated in Table 3 for trend calculations are rather short, so that the trends may not be robust. I think providing significance levels (p-values) for the trends would be helpful.

We agree that uncertainties exist in the OH anomalies derived from CH_3CCl_3 measurements (e.g. lines 208-216), although we would argue, as others before us have, that uncertainties on emissions play a smaller role in deriving OH anomalies after 1997, which is the main period of interest here. Furthermore, the fact that the multi-year signals derived from both global CH3CCl3 measurement programmes are reasonably consistent adds some confidence in the signals being robust. Nevertheless, even with these caveats (which we acknowledge) we think that it is still important to point out this possible role of OH variations on the observed CH_4 trend.

We agree that the sub-periods are fairly short but they are determined by the periods over which the global mean CH_4 shows variations. This length does reduce the robustness of the trends but they are the periods we need to analyse. In the revised paper we will include significance levels as suggested.

The authors make some statements in different parts of the paper that are not supported by sufficient evidence. Below, I note places where additional information or sensitivity tests could strengthen the statements.

OTHER SPECIFIC COMMENTS

This study relies entirely on the interannual OH variations inferred from CH3CCl3 observations and does not consider the OH variations suggested by other methods, including bottom-up, photochemical model calculations and top-down estimates using alternative halocarbons. The authors justify their use of specified OH with a comment near the beginning of Section 3.2.1 that "models with interactive tropospheric chemistry can produce a large range in absolute global mean [OH]", but they do not discuss the interannual variations in OH produced by such models. Montzka et al. [2011] show the OH variations derived from a photochemical model calculation as well as from various halocarbons including CH3CCl3 and note some of the differences. I think the current paper could be strengthened by considering other methods and possibly doing some sensitivity tests to assess how robust the conclusions are in the face of differing estimates of OH variations.

We agree different species could be used in principle; however previous studies, e.g. Montzka et al. (2011), which used other chemical species to derive OH, conclude that CH_3CCl_3 measurements provide the most robust and independent estimates. Other species used in their study to derive OH anomalies have much larger budget uncertainties and therefore do not provide equally reliable estimates of OH when compared with CH_3CCl_3. Both HCFC and HFC emissions are in a high state of flux because some chemicals are being phased in and out, making them much less suitable for deriving reliable changes in OH.

We also acknowledge that long-term simulations of photochemical models could be used to derive OH anomalies. However, there currently exists large uncertainty in model-derived OH, as noted by the reviewer in reference to Voulgarakis et al. (2013). As noted by Montzka et al.

(2011) photochemical models (e.g. see Leliveld et al., 2004) typically suggest a smaller-interannual variability than CH_3CCl_3-derived OH even since 1998, suggesting the models may not be accurately representing processes governing OH concentrations. Given that they calculate very different mean values it is likely that they are missing processes and will calculate different interannual variations based on the ones that they do. We believe that investigating accuracies in bottom-up photochemical models is beyond the scope of this work. We will add some brief discussion to the paper.

Section 2.1: Estimated anomalies in global OH based on CH3CCl3 measurements may not be accurate when applied to CH4 given the different spatial distributions of CH4 and CH3CCl3 and, to a lesser extent, different temperature dependences of their reaction with OH. The authors state at the end of Section 3 (lines 348-349) that this needs to be considered, but they do not actually consider it in their analysis. They should at the least emphasize this caveat more in the paper and discuss its implications for their findings.

OK. As noted by Reviewer 2 this difference between CH4 and CH3CCl3 is interesting. We will add more discussion and caveats on this point. We feel that it is a small effect.

Line 172: The runs that allow temperature to vary interannually would seem to doubly apply the temperature effect, given that the OH anomalies already implicitly contain temperature variations. Could you justify this?

We realise that there is this 'double counting' and so we use the simulations with fixed model temperature (FT) in our main analysis (see lines 168-172). By also running the model with varying temperature we can diagnose the likely contribution of temperature variations on OH + CH4 rate (see lines 358-360), even if the model run itself (VTVW) is not the most realistic. Through this we see that the temperature effect is small. We will clarify this in the revised paper.

Lines 180-182: You could discuss to what extent could the causality actually be bidirectional, i.e. high CH4 growth can sometimes result in low OH, so that OH isn't always the sole driver of the OH-CH4 correlations.

OK. We will add a statement regarding this possible bidirectional effect based on available literature. However, even with a large change in CH_4 growth rate, the total CH_4 mixing ratio in the atmosphere does not change by much. Table 6 in Spivakovsky et al (2000) shows ~5% change in model CH_4 equates to ~1% change in model OH. A 5% change in CH_4 (~100ppb), far exceeds the annual growth changes observed, therefore we believe this change to be small.

Lines 368-372: I suggest making this statement more quantitative, i.e. how large are the underestimate of OH and the overestimate of CH4 growth?

OK. The values already provided in Table 3 will be inserted into the text to quantify this statement.

Lines 376-377: Your analysis hasn't ruled out the possibility of changes in emissions being important during the 1999-2006 time period as well. Furthermore, the picture is more complex than all CH4 sources varying in the same direction; decreases in certain sources could compensate for increases in other sources.

We agree and we have tried to be careful to acknowledge that variations in emissions may still play an important role (e.g. abstract line 30, line 373-, line 395). We will further clarify this where possible in the revised version.

Lines 389-392: Is this issue relevant to your analysis? If so, could you suggest what impact it might have on your results? And if it isn't relevant, you could omit the sentence.

Yes, this is relevant and is one reason why we cannot analyse the most recent years. For the results shown in the paper measurements were only used up until 2007 (NOAA) and 2009 (AGAGE), when the methylchloroform concentration was higher. The statement is made to address future issues with the use of [OH]. We will change this sentence to offer more clarification.

Lines 392-394: Could you estimate how large of an effect this uncertainty might have on your results?

We will look at the Wennberg et al paper and add some discussion. They comment that "the loss of methylchloroform to the oceans play a small but important role". The first order effect of the ocean was as a net sink as CH3CCl3 concentrations were increasing, and potentially a very small net source as concentrations were decreasing. However, the question is how large is the interannual variability in this small term? It is difficult to imagine that any interannual variation could be large enough to affect our conclusions. We will do some estimates with the box model based on assumed extreme variations in the Wennberg sink/source (which is further reason for us to be able to run our own box model as well as use published OH values – see earlier comment).

Lines 394-397: This statement is certainly true and important, although it is not new and insightful. I suggest improving the statement so that the paper ends on a stronger note.

OK. This will statement will be modified.

Figure 5b-c: It's not clear to me from these plots that the runs with varying OH are in better agreement with observations than the run with repeating OH is. Perhaps you could also report the mean values of model minus observations over the different sub-periods.

We agree that the plot in isolation makes the difference difficult to see, which is why Table 3 provides the growth values requested. The difference over the sub-periods can be read from there.

MINOR COMMENTS

Lines 49-50: The post-2006 growth rate of ~6 ppb/yr cited here seems inconsistent with the 4.9 ppb/yr given in the abstract. Please reconcile.

OK. This will be corrected to 4.9 ppb/yr for both and the "post-2006" will be changed to 2006-2009.

Line 68: You should provide references for the statement that "the reasons for the renewed growth are also not fully understood."

OK, references will be included.

Lines 74-75: You could include additional references such as Wang et al. (2004) and Karlsdottir and Isaksen (2000). The full references are:
Wang, J. S., J. A. Logan, M. B. McElroy, B. N. Duncan, I. A. Megretskaia, and R. M. Yantosca (2004), A 3-D model analysis of the slowdown and interannual variability in the methane growth rate from 1988 to 1997, Global Biogeochem. Cycles, 18, GB3011, doi:10.1029/2003GB002180.
Karlsdottir, S., and I. S. A. Isaksen (2000), Changing methane lifetime: Possible cause for reduced growth, Geophys. Res. Lett., 27, 93– 96.

OK. These references will be added.

Lines 75-77: In addition, you could explain that Wang et al. attributed the OH trend to a decrease in column O3 amounts, and the modeled trend of Karlsdottir and Isaksen was due to changes in tropospheric pollutants.

OK, this information will be added.

Lines 79-81: I have not read those papers (Carn et al., 2015; Mills et al., 2015), but my understanding is that in the troposphere, volcanoes are a much less important aerosol source than human activities, and volcanic aerosols that reach the stratosphere actually promote ozone depletion and thus increased downward UV and OH. So I think the counter-intuitive conclusion of those papers needs some explanation here.

OK. This papers were added for completeness but this effect has not been quantified. We will remove them.

Lines 81-83: The citing of this Patra et al. paper is not really relevant to the discussion in the paragraph on OH trends, so it does not belong here.

OK, the text referring to this paper will be moved.

Line 157: This sentence is somewhat confusing, as I initially thought it meant that 1977 emissions were used for the entire 15-year spin-up. I suggest that you specify that emissions from 1977 to 1992 were used for the spin-up.

OK. This will be clarified.

Lines 315-317: I find this sentence unclear. Are you referring to the observed growth rates? Please clarify.

Theses lines refer to the modelled values. This will be clarified.

Line 351: You include "transport" in this sentence as playing a key role, but your results suggested a relatively minor role. Perhaps you could reword this sentence.

Yes. This will be corrected

Table 3: I understand your usage of “/ppb” in the heading of the table but it's not clear; maybe replace "/" with "in" or ",".

OK. This will be revised.

Anonymous Referee #2

The manuscript of McNorton et al. investigates the role of OH in driving the recent evolution of methane, especially the observed decline of its growth rates in the first half of the 2000s. The conclusion is that OH may have been a key driver of this modification of the methane growth rate.

The manuscript is well written and well within the scope of ACP. Even though there have been some key studies investigating the topic of the methane growth stagnation, this is the first paper that thoroughly investigates the role of OH. This is achieved through a series of model experiments with carefully chosen set-ups. I do not have any major concerns, but there are some (mostly minor) suggestions that I list below which I believe will improve the manuscript. Following those, I expect that it will be ready for publication.

We thank the reviewer for his/her careful reading of our manuscript and insightful comments.

GENERAL COMMENTS:

I would have expected some discussion towards the end of the paper ("Discussion and Conclusions" section) on why the previous studies that investigated this stagnation in methane growth did not come up with a similar conclusion when it comes to the role of OH. This is the new bit that this paper brings, and it needs to be understood why those conclusions were not reached before. Some brief additions to the final section commenting on this aspect would be useful.

We are not aware that other studies explicitly considered variations in OH when investigating the stagnation period. That is a motivation for this work. Lines 363-365 relate our study to a similar previous study (Fiore et al., 2006) but that study did not cover the stagnation period. We will add a few words to clarify these points.

Since the simulations start at 1993, why would the spin-up be done for 1977 conditions? That must be creating some methane imbalance in 1993, and with methane's relatively long lifetime, this will still be there in 1997, when the period of major interest begins. I may be missing something, but even in that case, it probably means that this aspect shall be clarified better.

The model is spun up from 1977 to get a reasonable spatial distribution by 1993, which does create a model-observation imbalance in 1993. This is corrected for by scaling the model 1993 global average CH4 concentration to observed data before reinitialising the model for the 1993-2009 simulation. We will clarify this in the revised version.

SPECIFIC COMMENTS:

Page 1, Lines 18-19: Sentence not very clear. How can something vary "on a timescale of many years", within two decades?

We agree the term "many years" could be confusing. In the revised version we will modify this to say that the variability is over multiple years, i.e. 2-5 years.

Page 1, Line 29: Please add "of" between "and" and "atmospheric".

OK.

Page 2, Line 49: 6ppb/yr: Number inconsistent with the abstract. Please correct the one that is wrong.

OK, the number should read 4.9 ppb/yr.

Page 2, Line 57: Please add “potential” between “second” and “explanation”.

OK.

Page 2, Line 60: “much more uncertainty” is unclear – please say a bit more.

OK. This will be modified to say that the bottom-up and top-down estimates differed.

Page 2, Line 66-67: So, the decrease in wetland emissions mentioned earlier was abandoned as a hypothesis. This paragraph needs to be connected in a clearer way with the previous one.

We agree with the reviewer that this could be confusing. The paragraph will be changed to show consistency between paragraphs L51-61 and L62-68.

Page 2, Line 71: Suggest adding “global mean” before “concentration”, as this symbol (“[OH]”) is used throughout the manuscript when referring to the global abundance.

OK.

Page 3, Lines 77-78: A recent paper by Voulgarakis et al. also included findings along these lines when it comes to the role of fires on OH variability, especially during El Nino events (see their Fig. 4c): Voulgarakis, A., M.E. Marlier, G. Faluvegi, D.T. Shindell, K. Tsigaridis, and S. Mangeon, 2015: Interannual variability of tropospheric trace gases and aerosols: The role of biomass burning emissions. J. Geophys. Res. Atmos., 120, no. 14, 7157-7173, doi:10.1002/2014JD022926.

OK, this reference will be included.

Page 3, Lines 103-110: Need to also remind the reader of the main finding of the Montzka et al. (2011) paper, i.e. the suggested small interannual variability of OH.

OK, this will be included.

Page 3, Line 107: Suggest changing “this” to “that”.

OK.

Page 4, Line 117: Suggest adding “global” between “yearly” and “anomalies”.

OK.

Page 4, Line 127: Suggest changing “date” to “year”.

OK.

Page 4, Line 152: What is meant by “scaled”? Please clarify.

More detail will be included; scaled emissions are taken from Ciais et al. (2014) top-down estimates.

Page 4, Lines 167-168: Why were zonal means of temperature used and not 3D data? That introduces one potential extra reason for differences between the runs, i.e. not just the fact that the temperature is fixed, but also that it is not 3D-varying. What is the impact of this?

We needed to create a time-averaged dataset (1993-2009) for the model run and so much of the 3-D variability would be averaged out anyway. We agree with the reviewer that using zonal mean fields might still influence the results; however, this difference is likely to be small (the major temperature variations, due to both height and latitude are considered). In any case we will add this note to the revised version.

Page 4, Lines 168-169: Suggest rephrasing to “We also derive our own OH anomalies from the anomaly in the…”.

OK.

Page 5, Line 156: Need to clarify whether the specified OH field is comprised of zonal means or whether it varies with longitude. If the former, need to discuss the implications of the lack of longitudinal variations.

The field comprises of zonal means. A sentence will be included to discuss the implications of this.

Page 6, Line 231: It should be 0.65 rather than 0.55.

OK, corrected.

Page 7, Lines 274-275: What is meant by “multi-year” here? Suggest specifying with a parenthesis.

This will be modified to now include (>1 year).

Page 7, Line 276: “year-year” -> “year-to-year”.

OK.

Page 8, Line 290-291: Why are the simulations with varying winds singled-out?

This is a miswording. The text will be changed to say that it considers all simulations not just those with varying winds.

Page 8, Line 294: Suggest adding “and also given the lack of change in emissions” after “Therefore,”.

OK.

Table 3: It is not immediately clear what is meant in the parentheses next to the numbers. I suggest writing "Global mean _CH4 in ppb" at the top row and "Global mean_CH4 per year in ppb/yr" at the bottom row of the title of those columns.

OK, this will be clarified (see also comment from Reviewer 1).

Page 8, Lines 315-317: I am not sure what is meant by this sentence. May need to be expanded or reworded.

OK, this will be reworded (see also comment from Reviewer 1).

Page 8, Line 318: In "CH4" the "4" needs to be subscripted. Also, I think a "from" is missing before "1999".

OK.

Page 9, Lines 346-349: This is interesting. But why could that be. An explanation, even a speculative one, would be nice. Is it perhaps due to somewhat different emissions regions for the two constituents, leading to different efficiencies of transport to regions of maximum loss?

OK. We do not want to add unfounded speculation to the paper but we will try to expand on this result slightly in the revised version. A different in the spatial distribution of emissions would seem to be a potentially important factor.

Page 9, Lines 357-358: I do not see why this sentence is needed.

OK. We wanted to make the point that ultimately it is always chemistry that removed CH4 and not transport. We will clarify this.

Page 10, Line 369: Please add "," before "which".

OK