Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions

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Abstract. Accurate prediction of future methane abundances following a climate scenario requires understanding the lifetime changes driven by anthropogenic emissions, meteorological factors, and chemistry-climate feedbacks. Uncertainty in any of these influences or the underlying processes implies uncertainty in future abundance and radiative forcing. We simulate methane lifetime in three chemical transport models (CTMs) – UCI CTM, GEOS-Chem, and Oslo CTM3 – over the period 1997–2009 and compare the models’ year-to-year variability against constraints from global methyl chloroform observations. Using sensitivity tests, we find that temperature, water vapor, stratospheric ozone column, biomass burning and lightning NOx are the dominant sources of interannual changes in methane lifetime in all three models. We also evaluate each model’s response to forcings that have impacts on decadal time scales, such as methane feedback, and anthropogenic emissions. In general, these different CTMs show similar sensitivities to the driving variables. We construct a parametric model that reproduces most of the interannual variability of each CTM and use it to predict methane lifetime from 1980 through 2100 following a specified emissions and climate scenario (RCP 8.5). The parametric model propagates uncertainties through all steps and provides a foundation for predicting methane abundances in any climate scenario. Our sensitivity tests also enable a new estimate of the methane global warming potential (GWP), accounting for stratospheric ozone effects, including those mediated by water vapor. We estimate the 100-yr GWP to be 32, which is 25 % larger than past assessments.

1 Introduction

Rising atmospheric concentrations of greenhouse gases are the main cause of current and future climate change (Intergovernmental Panel on Climate Change, hereafter IPCC, 2007). Uncertainty in mapping an emission scenario onto future abundance of greenhouse gases (GHGs) thus translates almost directly into uncertainty in our ability to project climate change and its impact on nature and society. To date, IPCC has generally adopted a single trajectory for the growth of greenhouse gases in each of several different socioeconomic scenarios, thus neglecting uncertainty in those future abundances. For methane, the second most important anthropogenic GHG, these trajectories are based on simple parametric formulas for methane lifetime. In the IPCC Third Assessment Report (TAR), 4 parameters accounted for changes in the largest atmospheric methane sink, oxidation by tropospheric OH: anthropogenic emissions of CO, nitrogen oxides (NOx), and volatile organic compounds (VOCs) and the negative feedback between methane abundance and tropospheric OH (Prather et al., 2001). Other sinks, which include oxidation in the stratosphere, oxidation by tropospheric chlorine, and uptake into soil, were assessed but assumed not to change during the 21st century projections. For the upcoming IPCC 5th Assessment Report (AR5) the Representative Concentration Pathway (RCP) scenarios adopt methane trajectories calculated in the MAGICC model, which augments the TAR parametric formula with a temperature term (Meinshausen et al., 2011a).

On small spatial scales, OH concentrations and methane oxidation depend on temperature, pressure, sun elevation, clouds, surface albedo, UV attenuation by stratospheric...
ozone, and local concentrations of water vapor, ozone, CH₄, CO, NOₓ, VOCs, and aerosols (e.g. Duncan et al., 2000; Olson et al., 2006). Integrated globally and annually, some of these influences are small, but numerous studies have found that temperature, circulation, water vapor, stratospheric ozone, clouds and natural and anthropogenic emissions are important (Lei et al., 2014; Simpson et al., 2000; Dentener et al., 2003; Stevenson et al., 2005; Dalsoren et al., 2006; Fiore et al., 2006; Hess and Mahowald, 2009; Voulgarakis et al., 2010). Uncertainties in these factors and in the present-day methane budget mean that each socioeconomic emission scenario could produce a range of future methane abundances (Prather et al., 2012).

Global climate model (GCM) simulations with atmospheric chemistry provide another method for predicting future methane and other chemically reactive GHGs. An ensemble of such models can provide a range of future methane abundances for a single scenario (e.g. Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) Lamarque et al., 2012), spanning some, but likely not all, future uncertainties. This approach is computationally expensive, however, which restricts the number of socioeconomic scenarios and ensemble members that can be explored.

In this work we develop a new parametric model for global methane lifetime that accounts for climate-chemistry interactions that were neglected in previous parametric approaches. We derive the parametric factors from perturbation tests in a suite of 3 chemical transport models (CTMs), since CTMs with detailed tropospheric chemistry provide the best mechanistic representation of methane loss from tropospheric OH.

We focus on the tropospheric OH sink because other methane sinks are smaller and their intrinsic variability has a smaller impact on the total methane lifetime. The parametric model includes climate and emission factors that control the interannual variability of methane lifetime in the CTMs. These factors are likely important on decadal time scales as well. The parametric model also includes anthropogenic emissions that can change decadal trends in methane lifetime, but contribute little to interannual variability. Uncertainties in atmospheric chemistry are included in the parametric factors based on the range of perturbation responses across the CTMs. We evaluate the parametric model against 13-yr CTM simulations of methane lifetime, and against observed variability in tropospheric OH, as measured by the decay of methyl chloroform. Assuming that the same climate and emission processes will remain dominant drivers of methane lifetime throughout the 21st century, we use this parametric model to make new projections of methane abundance and its uncertainty through 2100. Finally, the perturbation tests also enable a new estimate of the ozone contribution to methane radiative forcing and global warming potential.

2 Model descriptions

We diagnose methane lifetime due to tropospheric OH, \( \tau_{\text{CH}_4 \times \text{OH}} \), from multi-year simulations in 3 different CTMs: University of Oslo CTM3, University of California, Irvine (UCI) CTM, and GEOS-Chem. All of these models are driven by assimilated meteorological data and configured to use the same emissions from anthropogenic, biogenic and biomass burning sources. We use year-specific meteorology spanning 1997–2009 for each model, except GEOS-Chem simulations with GEOS-5 meteorology, which are only 2004–2009. Sections 2.1–2.3 and Table S1 summarize features of each model and Sect. 2.4 describes the emissions.

Monthly chemistry diagnostics from each model enable us to calculate \( \tau_{\text{CH}_4 \times \text{OH}} \), defined as the total atmospheric \( \text{CH}_4 \) burden divided by its loss through reactions with tropospheric OH. All 3 models use fixed methane abundances (1760 ppb for UCI CTM and Oslo CTM3, 1775 ppb for GEOS-Chem), so variations in \( \tau_{\text{CH}_4 \times \text{OH}} \) are due solely to changes in the OH sink. Different tropopause definitions in the models have minimal effect on \( \tau_{\text{CH}_4 \times \text{OH}} \) since \( \text{CH}_4 \) oxidation between 200 hPa and the tropopause is 1.5 % of tropospheric methane loss, or less. We calculate the total methane lifetime, \( \tau_{\text{CH}_4} \), using \( \tau_{\text{CH}_4 \times \text{OH}} \) values from this work and recently estimated lifetimes for other methane sinks: tropospheric chlorine (200 yr), stratosphere (120 yr), and soil (150 yr) (Prather et al., 2012).

2.1 Oslo CTM3

Oslo CTM3 is a stratospheric and tropospheric CTM, recently described by Søvde et al. (2012). Transport is driven by pieced-forecast meteorology from the European Center for Medium-range Weather Forecasting (ECMWF) Integrated Forecast System (cycle 3r61, http://www.ecmwf.int/research/ifsdocs/CY36r1/index.html). The original T359 (∼0.55° × 0.55°) horizontal resolution and 60 layer vertical resolution of the forecast model is degraded to T42 (∼2.8° × 2.8°) resolution, while preserving the 3 h temporal resolution for all meteorological fields. Advection uses the second-order moments scheme (Prather, 1986; Prather et al., 2008) and convection follows Tiedtke (1989).

The Oslo CTM3 chemical mechanism includes a full stratospheric chemical mechanism in addition to tropospheric reactions. The tropospheric module contains 105 reactions and 51 gas-phase species, including sulfate, nitrate, and sea-salt aerosols. Nitrate aerosols influence gas-phase chemistry through HNO₃ uptake, which is a sink for reactive nitrogen through subsequent wet scavenging. Photolysis rates required in the chemistry mechanism are calculated online using the Fast-JX method (Neu et al., 2007), with cloud distributions from ECMWF meteorology and ozone concentrations calculated in the CTM. Aerosol effects on photolysis are neglected except for a small contribution from black carbon (Søvde et al., 2012), which increases OH and biases.
Table 1. Emissions.

<table>
<thead>
<tr>
<th>Source (Inventory)</th>
<th>( \text{NO}_x, \text{Tg(N) yr}^{-1} )</th>
<th>( \text{CO, Tg yr}^{-1} )</th>
<th>Isoprene, Tg yr(^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anthropogenic (RCP year 2000)</td>
<td>( 32^{b} )</td>
<td>( 609 )</td>
<td>–</td>
</tr>
<tr>
<td>Biomass burning (GFED3)</td>
<td>( 5.6^{c} )</td>
<td>( 360^{c} )</td>
<td>–</td>
</tr>
<tr>
<td>Biogenic (MEGAN)</td>
<td>–</td>
<td>( 76 )</td>
<td>523</td>
</tr>
<tr>
<td>Lightning</td>
<td>( 5^{d} )</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>Total</td>
<td>42</td>
<td>1047</td>
<td>523</td>
</tr>
</tbody>
</table>

\(^{a}\) Inventory references: RCP, Lamarque et al. (2010); van Vuuren et al. (2011), GFED3 (van der Werf et al. (2010), MEGAN Guenther et al. (2006).
\(^{b}\) Land, ship, and aviation components are 26, 5.4, and 0.85 Tg(N) yr\(^{-1}\), respectively.
\(^{c}\) Average biomass burning for 1997–2009. Emissions for individual years are 3.3–6.1 Tg(N) yr\(^{-1}\) and 263–605 Tg(CO) yr\(^{-1}\).
\(^{d}\) Average for 1997–2009 in UCI CTM and Oslo CTM3. Emissions for individual years are 4.8–5.4 Tg(N) yr\(^{-1}\).
GEOS-Chem has 5.7–6.4 Tg(N) yr\(^{-1}\) (average 6 Tg(N) yr\(^{-1}\)) over 2004–2009.

\( \tau_{\text{CH}_4 \times \text{OH}} \) low by about 10% (Bian et al., 2003; Martin et al., 2003). Oslo CTM3 shares the same chemical mechanism and some other physical process algorithms with the older Oslo CTM2, which has been extensively used for studies of present and future tropospheric composition (Isaksen et al., 2005; Hoor et al., 2009; Dalsoren et al., 2010).

2.2 UCI CTM

The UCI CTM is a tropospheric CTM, using the same meteorology, transport algorithms, and Fast-JX photolysis as Oslo CTM3. Like Oslo CTM3, the UCI CTM uses T42 horizontal resolution, but the vertical resolution in the boundary layer is reduced, so there are 57 layers total. Tropospheric chemistry of the major gas-phase species involved in \( \text{HO}_x, \text{NO}_x, \text{O}_3, \) and VOC reactions is simulated with the ASAD package (Carver et al., 1997), with updates to the mechanism and kinetics (Tang and Prather, 2010). This mechanism includes 84 reactions involving 33 species, making it simpler than the Oslo CTM3 chemical mechanism. As in Oslo CTM3, clouds from ECMWF meteorology are used for photolysis. Simplified tropospheric \( \text{O}_3 \) chemistry is simulated with Linoz (version 2, Hsu and Prather, 2009) and used for photolysis calculations and stratosphere-troposphere exchange. Aerosol effects on chemistry are neglected, which biases \( \tau_{\text{CH}_4 \times \text{OH}} \) low by up to 5% in addition to the aerosol-induced photolysis bias described above for Oslo CTM3 (Martin et al., 2003; Macintyre and Evans, 2010).

2.3 GEOS-Chem

GEOS-Chem is a tropospheric CTM, driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-5) or MERRA reanalysis (Rienecker et al., 2008, 2011). Both GEOS-5 and MERRA are produced from closely related assimilation systems, using the same spatial resolution of \( 0.5^\circ \times 0.66^\circ \) and 72 vertical layers. We use GEOS-Chem version 9-01-02 here. Most results, including all sensitivity simulations, are based on GEOS-5 meteorology, which has been degraded to \( 2^\circ \times 2.5^\circ \) and 47 layers for the CTM. GEOS-5 data are available only after 2004, however, so we also simulate 1997–2009 using MERRA meteorology at \( 4^\circ \times 5^\circ \) and 47 layers. Temporal resolution in GEOS-5 (MERRA) is 6 h (3 h) for most meteorological quantities and 3 h (1 h) for surface quantities and mixing depth.

The tropospheric chemistry mechanism in GEOS-Chem, as recently updated by Mao et al. (2010), consists of 104 species and 236 chemical reactions that simulate aerosols in addition to the \( \text{HO}_x \cdot \text{NO}_x \cdot \text{VOC} \)-ozone system. Photolysis rates are calculated with the Fast-JX method, using aerosol optical depths that are simulated internally, and ozone columns from the TOMS and SBUV satellites (until 2008) or GEOS-5 assimilation of satellite data (after 2008). For purposes of stratosphere-troposphere exchange, stratospheric ozone is simulated with Linoz.

2.4 Emissions

Emissions used in this work are representative of 1997–2010, but do not resolve trends or interannual variability in anthropogenic or biogenic emissions. To the extent possible, we use identical emissions across all models. Anthropogenic, biogenic, and biomass burning emissions of \( \text{NO}_x, \text{CO}, \) and isoprene are fully consistent in all models. Some differences in VOC emissions arise because of the different lumping schemes used in the various chemical mechanisms and because some VOC species are not simulated in all models. Lightning \( \text{NO}_x \) emissions also differ between models because they are calculated from underlying meteorology, as described below.

Table 1 summarizes emissions of key species. We use the RCP inventory for anthropogenic emissions for year 2000, repeating in each simulated year (Lamarque et al., 2010; van Vuuren et al., 2011). This inventory provides monthly gridded emissions of \( \text{NO}_x, \text{CO} \) and speciated VOCs from 11 emission activities. Aviation and shipping emissions change each month, while other anthropogenic emissions are
constant throughout the year. Biomass burning emissions are specified for each year and month by the GFED inventory (version 3, van der Werf et al., 2010). We use this instead of the climatological biomass burning emissions provided in the RCP inventory because fires are a major cause of year-to-year variability in tropospheric OH (Duncan et al., 2003; Manning et al., 2005; Dalsoren et al., 2006). Biogenic emissions of isoprene, CO, and other VOCs are from a MEGAN climatology for the 2000s decade (Guenther et al., 2006). Anthropogenic, biogenic, and biomass burning emission data are provided at 0.5° × 0.5° resolution. GEOS-Chem includes additional oceanic emissions of acetone (13 Tg yr⁻¹, Jacob et al., 2002) and acetaldehyde (57 Tg yr⁻¹, Millet et al., 2010), which are not included in other models. All emissions except aviation, biomass burning and lightning occur in the lowest model layer and are quickly mixed vertically through boundary layer convection and turbulence. Biomass burning is assumed to occur at the surface in the UCI CTM and Oslo CTM3. In Oslo CTM3, however, biomass burning emissions follow the RETRO vertical distribution (Schultz et al., 2008), which injects 35 % of equatorial emissions and 45 % of boreal emissions above the boundary layer. In a sensitivity test we inject all biomass burning into the lowest model layer in Oslo CTM3.

Lightning NOₓ emissions are calculated with similar methods in all 3 CTMs, with UCI CTM and Oslo CTM3 using identical algorithms. In all models, these emissions are derived from cloud-top heights in the underlying meteorology (Price and Rind, 1994) and scaled to match satellite-observed lightning flash rates (Christian et al., 2003). In the UCI CTM and Oslo CTM3, 2 scale factors are calculated to match observed multi-year mean flash rates over land and ocean. In GEOS-Chem scale factors are calculated for every grid column and month (Sauvage et al., 2007). Within the convective column, lightning NOₓ is distributed vertically based on NOₓ observations near thunderstorms (Ott et al., 2010). Søvde et al. (2012) provide a full description of lightning emissions in UCI CTM and Oslo CTM3, and Murray et al. (2012) do the same for GEOS-Chem. Lightning NOₓ emissions average 6 Tg(N) yr⁻¹ in GEOS-Chem and 5 Tg(N) yr⁻¹ in UCI CTM and Oslo CTM3.

3 Recent (1997–2009) variability of CH₄ lifetime

Figure 1 shows τ_CH₄×OH for 1997–2009, as simulated by the 3 CTMs. The tropospheric OH lifetimes range from 8.5 to 10.1 yr. The longest of these lifetimes (GEOS-Chem) is consistent with the constraint provided by methyl chloroform observations, 11.2 ± 1.3 yr (Prather et al., 2012), but all are within the range of contemporary tropospheric chemistry models (e.g. 9.7 ± 1.5 yr from ACCMIP, Naik et al., 2012).

These simulations show similar variability of τ_CH₄×OH in all CTMs. Common features include a sharp dip in 1998 and peak in 2000, coincident with a strong El Niño and La Niña, smaller peaks in 2004 and 2008, and general decline after 2005. These features appear robust against the various choices of chemical mechanism, meteorology, and resolution used in these CTMs. In independent work, the ECHAM model also simulates the same features, using different chemistry and emissions that include biogenic variations (Montzka et al., 2011). Oslo CTM3 has the least variability (0.65 % for σ/mean), while GEOS-Chem with MERRA meteorology has the most (1.1 %). The common features, as well as differences in their magnitude, are explained below by a small number of processes (Sect. 3.3).

3.1 Methyl chloroform comparison

Two global measurement networks have recorded the growth and decline of atmospheric methyl chloroform (MCF) since the 1970s (ALE/GAGE/AGAGE, Prinn et al., 2005), with expanded coverage since the 1990s (NOAA, Montzka et al., 2000). Like methane, atmospheric MCF is oxidized mainly by tropospheric OH, with small additional sinks in the stratosphere, oceans, and soil (Volk et al., 1997; Wennberg et al., 2004; Wang et al., 2008). Because MCF has no natural sources and the anthropogenic production is well known (McCulloch et al., 1999), MCF provides the best available constraint on global OH levels and methane lifetime. The analysis here uses observations since 1998, when anthropogenic MCF emissions became small compared to atmospheric oxidation of the residual atmospheric burden. Consequently, MCF atmospheric lifetimes can be inferred from observed decay rates without detailed accounting for emissions and transport (Montzka et al., 2011).
For each network, we calculate decay rates of MCF from monthly average concentrations provided by each network (NOAA: ftp://ftp.cmdl.noaa.gov/hats/solvents/CH3CCl3/flasks/GCMS/CH3CCL3_GCMS_flask.txt, last access: 6 August 2012; AGAGE: http://agage.eas.gatech.edu/data_archive/agage/ge-md/monthly/, last access: 4 April 2012). For site $i$ and month $t$ the observed decay rate ($\text{yr}^{-1}$) is

$$k_{i,t} = -\ln(c_{i,t+6}/c_{i,t-6}),$$

where $c_{i,t}$ is the concentration at site $i$ in month $t$. The global MCF decay rate is the average of $k_{i,t}$ across sites within a network. We calculate uncertainty in the global decay rate as the 16th–84th percentile range (i.e. $\pm 1\sigma$) of $k_{i,t}$ across sites within a network. No filling is used for months with missing data. (See Supplement for additional method details.) Averaged over 1998–2007, the global MCF decay rates from the 2 networks differ by less than 1% ($0.1811 \text{ yr}^{-1}$ for NOAA, $0.1796 \text{ yr}^{-1}$ for AGAGE), providing a strong constraint on the long-term global-mean OH and methane lifetime. This analysis, however, focuses on anomalies in the global decay rate, relative to each network’s own mean. Because the anomalies are attributed solely to tropospheric OH loss (see below) and for comparison to $\tau_{\text{CH}_4\times\text{OH}}$, the decay anomalies are divided by $r = 0.87$ to account for the tropospheric OH fraction of total MCF loss (Prather et al., 2012).

Figure 2 compares the interannual variability of simulated $\tau_{\text{CH}_4\times\text{OH}}$ in the CTMs against the MCF decay rate. In all CTMs, simulated $\tau_{\text{CH}_4\times\text{OH}}$ variations (0.7–1.1 % for $\sigma$/mean) are smaller than the upper limit imposed by the MCF constraint (2.3 %, Montzka et al., 2011). Adding variable isoprene emissions to the GEOS-Chem simulation increases the simulated variability by only 0.1 % (not shown). While the CTM $\tau_{\text{CH}_4\times\text{OH}}$ anomalies are consistently within

the observational uncertainty for both the AGAGE and NOAA networks (shaded region of Fig. 2), the year-to-year changes in the models generally do not correlate with the MCF data. Residual anthropogenic or ocean emissions could account for some MCF decay anomalies, but only if these emissions change abruptly from year to year. Emission anomalies of about 4 Gg yr$^{-1}$ would be required to cause the observed decay rate swings during 2002–2004. Meanwhile, total anthropogenic and ocean emissions for those years are estimated to be 6 and 4 Gg yr$^{-1}$, respectively, and decreasing smoothly (Wennberg et al., 2004; Prinn et al., 2005; Montzka et al., 2011). Therefore, abrupt emission changes might explain part, perhaps half, of the decay anomalies, but cannot account for the full discrepancy between simulated $\tau_{\text{CH}_4\times\text{OH}}$ and observations.

Collocated measurement sites in the NOAA and AGAGE networks provide an alternative means to evaluate possible errors in decay rates. With MCF concentrations about 30 ppt (1 ppt = 1 pmol mol$^{-1}$) in 2003 and decreasing at a rate of 6 ppt yr$^{-1}$, quantifying $\tau_{\text{CH}_4\times\text{OH}}$ anomalies of 1% requires measurement accuracy of 0.06 ppt or better for the monthly mean concentration. At all 4 collocated sites (Cape Grim, American Samoa, Trinidad Head, and Mace Head) we find differences between the networks as large as 2% (~0.1 to 0.5 pmol mol$^{-1}$) in the monthly means. (See Fig. S2) While small in absolute terms, these differences exceed the standard error in monthly means and persist for several consecutive months; thus they are likely not caused by synoptic variability and differences in sampling frequency. Because the biases change over time, they lead to differences of up to 4% in MCF decay rates at a single site. As can be seen in Fig. 2, both networks find similar magnitude of OH variability, but they differ in sign and magnitude of the anomaly at many times. These differences in observed MCF decay rates between the two networks are as large as the discrepancy between CTM $\tau_{\text{CH}_4\times\text{OH}}$ anomalies and either set of observations. Therefore, we conclude that better understanding of systematic differences between the observation networks is required before MCF can be used to constrain $\tau_{\text{CH}_4\times\text{OH}}$ and OH anomalies in specific years at the precision required (1-2 %) to test CTM interannual variability.

### 3.2 Methane lifetime sensitivity to chemistry-climate factors

Having identified consistent variations in $\tau_{\text{CH}_4\times\text{OH}}$ across multiple CTMs, we examine their causes with explicit perturbation tests. In these tests, we perturb a single climate or emission variable, simulate 3 or more years, discard the first year as spinup, and analyze the difference from the unperturbed simulation in the remaining years. Perturbations are applied to 1997–1999 for Oslo CTM3 and the UCI CTM, and, because of meteorological data availability, to 2004–2006 for GEOS-Chem with GEOS-5. The sensitivity, $\alpha$, of $\tau_{\text{CH}_4\times\text{OH}}$ to a climate or emission variable, $F$, is always
defined as

$$\alpha = \frac{d \ln(\tau_{CH_4}^{OH})}{d \ln(F)}.$$  \hspace{1cm} (2)

As such, $\alpha$ can be interpreted as the percent change in $\tau_{CH_4}^{OH}$ resulting from a 1% increase in $F$.

Table 2 reports sensitivities for the evaluated climate and emission variables. These variables include most of those identified in the literature as important influences on tropospheric OH and $\tau_{CH_4}^{OH}$ variability: temperature, water vapor, stratospheric ozone column, convective fluxes, cloud optical depth, biomass burning emissions, and anthropogenic emissions. Perturbation magnitudes are chosen to be similar to the interannual variability or decadal trend of each variable (exact magnitudes in Table S2). Perturbations are applied globally, except for stratospheric ozone, which is only perturbed over 40° S–40° N, where tropospheric OH production from UV photolysis of tropospheric O$_3$ occurs. The consequences of averaging region choice are discussed further in Sect. 3.3. Temperature and water vapor perturbations are restricted to the troposphere in Oslo CTM3, which includes full stratospheric chemistry, to avoid confounding changes in stratospheric ozone columns.

Only variables with large sensitivity, large interannual changes, or both can explain the year-to-year $\tau_{CH_4}^{OH}$ variations identified in Fig. 1. Figure 3 shows the interannual changes of 5 key variables for 1997–2009. Water vapor, having about 3% variation and $\tau_{CH_4}^{OH}$ sensitivity near 0.3, could account for about 1% interannual variability in $\tau_{CH_4}^{OH}$. Temperature, ozone column, lightning NO$_x$, and biomass burning also have sufficient sensitivity and variability to account for about 1% variation in $\tau_{CH_4}^{OH}$.

<table>
<thead>
<tr>
<th>Variable $^b$</th>
<th>UCI CTM</th>
<th>Oslo CTM3</th>
<th>GEOS-Chem</th>
<th>Literature $^c$</th>
<th>Adopted $^d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemistry-climate interactions</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Air temperature</td>
<td>−3.9</td>
<td>−2.8</td>
<td>−2.2</td>
<td>−3.0 ± 0.8</td>
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<tr>
<td>Water vapor</td>
<td>−0.32</td>
<td>−0.29</td>
<td>−0.34</td>
<td>−0.32 ± 0.03</td>
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</tr>
<tr>
<td>Ozone column, 40° S–40° N</td>
<td>+0.66</td>
<td>+0.43$^f$</td>
<td>+0.61</td>
<td>+0.28 ± 0.76$^f$ [1]</td>
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</tr>
<tr>
<td>Lightning NO$_x$ emissions</td>
<td>−0.14</td>
<td>−0.11</td>
<td>−0.24</td>
<td>−0.08 ± 0.16 [4]</td>
<td></td>
</tr>
<tr>
<td>Biomass burning emissions</td>
<td>+0.021</td>
<td>+0.013</td>
<td>+0.017</td>
<td>+0.02 ± 0.03 $^f$</td>
<td></td>
</tr>
<tr>
<td>CH$_4$ abundance</td>
<td>+0.363</td>
<td>+0.307</td>
<td>+0.274</td>
<td>+0.32 [5]</td>
<td></td>
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<tr>
<td>Optical depth, ice clouds</td>
<td>−0.036</td>
<td>N</td>
<td>N</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Optical depth, water clouds</td>
<td>+0.013</td>
<td>N</td>
<td>N</td>
<td></td>
<td></td>
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<tr>
<td>Anthropogenic emissions</td>
<td></td>
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<tr>
<td>Land NO$_x$</td>
<td>−0.15</td>
<td>−0.10</td>
<td>−0.16</td>
<td>−0.13 [5]</td>
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<tr>
<td>Ship NO$_x$</td>
<td>−0.045</td>
<td>−0.048</td>
<td>−0.017</td>
<td>−0.0412 ± 0.011 [8]</td>
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<tr>
<td>Aviation NO$_x$</td>
<td>+0.006</td>
<td>+0.050</td>
<td>+0.065</td>
<td>+0.011 [5]</td>
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<tr>
<td>CO</td>
<td>+0.006</td>
<td>+0.050</td>
<td>+0.065</td>
<td>+0.011 [5]</td>
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<tr>
<td>VOC</td>
<td>+0.045</td>
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</tbody>
</table>
| Sensitivity to climate and emissions: $^{a} \text{Sensitivity to CH}_4$ abundance calculated from perturbations applied over 1997–2009 in UCI CTM and Oslo CTM3 and 2004–2009 in GEOS-Chem. $^{b}$Italic variables are major causes of interannual $\tau_{CH_4}^{OH}$ changes (based on sensitivity and interannual changes in the variable) that are included in the 1997–2009 reconstruction of CTM $\tau_{CH_4}^{OH}$ in Sect. 3.3. Bold variables can drive decadal trends and are included in 1980–2010 prediction of $\tau_{CH_4}^{OH}$ in Sects. 4 and 5. $^{c}$1 [Karlsdottir and Isaksen 2000], [2] Krol and van Weele (1997), [3] Voulgarakis et al. (2009), [4] Labrador et al. (2004), [5] Prather et al. (2001), [6] Fiore et al. (2009), [7] Fry et al. (2012), [8] Hoer et al. (2009), [9] Myhre et al. (2011), [10] Dalson et al. (2010), [11] Holmes et al. (2011) $^{d}$Adopted values are the mean of CTMs, except for VOC, and aviation NO$_x$, which come from literature. Uncertainties are 1σ values based on CTM spread and expert assessment of literature. Terms marked N have negligible impact on interannual $\tau_{CH_4}^{OH}$ variability and are not used in the parametric model. $^{e}$Tropospheric perturbation only. $^{f}$In Oslo CTM3 stratospheric chemistry and stratosphere-troposphere exchange respond to the perturbation, whereas UCI CTM and GEOS-Chem responses are due solely to tropospheric photolysis rates. Oslo CTM3 results are rescaled to the same ozone column change as the other models. $^{g}$Response to global ozone column perturbation. $^{h}$Response to extra-tropical ozone column perturbation (poleward of 30°N). $^{i}$Sensitivities assume fire emissions are contained in the boundary layer, except where noted for Oslo CTM3. The UCI CTM value is the emission-weighted average sensitivity for 1997–2009 (Fig. 4). Values for other models are calculated from 3-yr perturbation tests and scaled to 1997–2009 means, assuming the same interannual variability as the UCI CTM. Adopted uncertainty accounts for uncertainty in emission altitude and year-to-year variability. $^{j}$Calculated with the RETRO altitude distribution, which injects 35% of equatorial (45% of boreal) biomass burning emissions above 2 km. Net sensitivity is lower because NO is longer lived at high altitudes and more effectively cancels the CO sink for OH. $^{k}$Sensitivity to CH$_4$ abundance calculated from perturbations applied over 1997–2009 in UCI CTM and Oslo CTM3 and 2004–2009 in GEOS-Chem. $^{l}$Sensitivity to CH$_4$ abundance is scaled from the 2004–2009 contribution of tropospheric OH production from CH$_4$. $^{m}$Al all anthropogenic NO$_x$ emission occurring on land, including combustion, agriculture, and waste.
These 5 climate and emission variables that we identify as important influences on $\tau_{\text{CH}_4 \times \text{OH}}$ have been recognized previously, but their sensitivities have typically not been quantified in a comparable way (e.g., Spivakovskyy et al., 2000; Dentener et al., 2003; Labrador et al., 2004; Stevenson et al., 2005; Fiore et al., 2006; Hess and Mahowald, 2009).

Convective fluxes and cloud optical depths for water and ice clouds, as diagnosed in ECMWF meteorology, vary annually by 2% and have small sensitivity ($-0.03$ to $+0.01$), so these factors have very little impact on $\tau_{\text{CH}_4 \times \text{OH}}$ ($\ll 1\%$). Due to the small impact in the UCI CTM, these perturbation tests are not repeated in the other CTMs. Our results are consistent with the known decrease in mass-weighted global OH concentrations due to clouds (Voulgarakis et al., 2010) because mass-weighted averaging emphasizes below-cloud OH concentrations and we find compensating increases in methane loss above clouds. In addition, past analyses of convective fluxes have found both positive and negative influences on $\tau_{\text{CH}_4 \times \text{OH}}$ depending on the convection scheme and perturbation used (Lawrence and Salzmann, 2008).

Methane abundance and anthropogenic NO$_x$, CO, and VOC emissions vary smoothly between years, but changed by 1–2% over the 2000–2010 (Dlugokencky et al., 2011; Granier et al., 2011). Therefore, these factors have little impact on $\tau_{\text{CH}_4 \times \text{OH}}$ variability during the 13-yr CTM simulation, but are important on multi-decadal time scales and longer.

Many of the sensitivity terms in Table 2 – specifically, water vapor, CH$_4$ abundance, and anthropogenic land NO$_x$ and CO – are consistent among the CTMs and with past estimates (Prather et al., 2001; Fiore et al., 2006; Hoor et al., 2009; Myhre et al., 2011), suggesting a good understanding of how these variables impact tropospheric methane loss. Adopted values for each sensitivity (Table 2, right column) reflect the degree of consistency among models. These adopted values are used in the parametric model described below.

For biomass burning, the CTMs show moderate agreement in the mean sensitivity when fires are assumed to emit into the boundary layer ($\alpha = 0.013$ to 0.021). The $\tau_{\text{CH}_4 \times \text{OH}}$ response to biomass burning depends strongly on the emission altitude, however, since the net sensitivity results from offsetting CO and NO$_x$ effects and the NO$_x$ lifetime depends strongly on altitude. When using the RETRO vertical distribution for wildfire emissions in Oslo CTM3, in which 35–45% of emissions enter the free troposphere, the $\tau_{\text{CH}_4 \times \text{OH}}$ sensitivity is 4 times smaller ($\alpha = 0.003$). This vertical distribution explains why Oslo CTM3 exhibits the least $\tau_{\text{CH}_4 \times \text{OH}}$ variability in the 1997–2009 simulations (Fig. 1). Sensitivity to fires also changes dramatically between years, shown in Fig. 4 for the UCI CTM. The sensitivity is greatest during years with high biomass burning emissions, and high CO/NO ratio in those emissions, both of which suppress tropospheric OH (Duncan et al., 2003; Dalsoren et al., 2006; Voulgarakis et al., 2010) and peak during El Niño years due to tropical
peat fires. Future El Niño activity cannot be predicted robustly (Collins et al., 2010) and injection altitude distributions remain uncertain so, despite some CTM consensus on present-day biomass burning sensitivity, we adopt a broad uncertainty range for this sensitivity factor.

Other sensitivities, chiefly air temperature and ship NOx, differ by 50% or more across the models. These differences are understandable, however, as consequences of modeling assumptions. For ship NOx, Oslo CTM3 and UCI CTM are nearly 3 times more sensitive than GEOS-Chem. In the UCI CTM and Oslo CTM3, ship NOx is emitted as NO, diluted into the grid volume, and the subsequent production of O3 and HNO3 are calculated by the grid-resolved chemistry. Instantaneous dilution overestimates the NOx lifetime and O3 production from ships (Chen et al., 2005), however. To compensate, GEOS-Chem instantaneously converts all ship NOx emissions to O3 and HNO3, following observed production ratios. As a result, GEOS-Chem underestimates the large-scale impact of shipping, since, in reality, 20–50% of NOx remains after 5 h following emission (Vinken et al., 2011). Although previous estimates of ship NOx are close to the high values in this work (Hoor et al., 2009; Myhre et al., 2011), the actual atmospheric sensitivity to ship NOx, likely lies somewhere between the GEOS-Chem and UCI CTM results.

Ship NOx emissions also explain the divergence of GEOS-Chem and the UCI CTM in their temperature sensitivities. Over land, both models predict similar reduction in $\tau_{CH_4 \times OH}$ in response to warming. Over the oceans, however, GEOS-Chem predicts longer $\tau_{CH_4 \times OH}$ at higher temperatures while the UCI CTM predicts the opposite. In the presence of ship NOx in the UCI CTM, higher temperatures increase both the production and loss of O3, with net excess production; OH rises in turn. In GEOS-Chem, by contrast, higher temperatures increase O3 destruction over the ocean with less opportunity for enhanced production; OH thus decreases over oceans.

The sensitivity of $\tau_{CH_4 \times OH}$ to methane abundance is closely related to the methane feedback factor, $f$, which is the ratio of methane perturbation lifetime to total budget lifetime (Prather et al., 2001). We calculate these terms from a 5% perturbation to methane abundance for 1997–2009 in the CTMs. Our multi-model mean sensitivity, 0.31 ± 0.04, is similar to past estimates (Prather et al., 2001; Fiore et al., 2009), but we derive a smaller feedback factor $f = 1.34 ± 0.06$ than has been recommended by IPCC ($f = 1.4$, Prather et al., 2001) because we use updated estimates of methane lifetime (Prather et al., 2012). Reducing the feedback factor, which was already suggested by Fiore et al. (2009), lowers the methane radiative forcing and global warming potential, as discussed in Sect. 6.

### 3.3 A parametric model for $\tau_{CH_4 \times OH}$

The sensitivity parameters in Table 2, together with the time series of corresponding climate and emission variables in Fig. 3, enable us to build a parametric model for methane lifetime representing each CTM. We combine terms linearly, so that $\tau_{CH_4 \times OH}$ is approximated by

$$\ln(\tau_{CH_4 \times OH}(t)) = \ln((\tau_{CH_4 \times OH})) + \sum \alpha_i \Delta \ln(F_i(t)),$$

(3)

where $F_i(t)$ is the time series of forcing variable $i$, $\alpha_i$ is the sensitivity of $\tau_{CH_4 \times OH}$ to that forcing variable (Eq. 2), and $\langle \tau_{CH_4 \times OH} \rangle$ is the mean lifetime in the CTM. Figure 1 shows the parametric model reconstruction of each CTM, alongside the actual calculated $\tau_{CH_4 \times OH}$. We find that 5 variables – temperature, water vapor, column ozone, biomass burning emissions, and lightning NOx emissions – explain 90% of the interannual variation in $\tau_{CH_4 \times OH}$ in all 3 CTMs over the simulated period 1997–2009. Even though the GEOS-Chem sensitivity parameters were derived from 2°C × 2.5° simulations driven by GEOS-5, the 5-parameter model performs equally well compared to the 4°C × 5° GEOS-Chem simulation driven by MERRA. The sensitivity parameters are thus robust across changes in model resolution and meteorology.

We find that 85% of methane oxidation by tropospheric OH occurs between 40°S and 40°N and that this region also controls the interannual variability of $\tau_{CH_4 \times OH}$ in the CTMs. The same latitudes also dominate the variability of global-mean temperature, water vapor, column ozone, biomass burning emissions, and lightning NOx emissions – explain 90% of the interannual variation in $\tau_{CH_4 \times OH}$ in all 3 CTMs over the simulated period 1997–2009. Even though the GEOS-Chem sensitivity parameters were derived from 2°C × 2.5° simulations driven by GEOS-5, the 5-parameter model performs equally well compared to the 4°C × 5° GEOS-Chem simulation driven by MERRA. The sensitivity parameters are thus robust across changes in model resolution and meteorology.

The atmospheric chemistry of tropospheric OH and methane involves nonlinear chemistry that could, in principle, undermine the additivity of terms in Eq. (3). We test the linearity of the system with a final perturbation test in the UCI CTM in which all 5 factors are perturbed simultaneously. The resulting change in $\tau_{CH_4 \times OH}$ differs by about 1 part in 10 from the linear addition of factors.

The CTM simulations in this work make several assumptions to simplify the perturbation analysis and enable comparisons between CTMs, but these could alter $\tau_{CH_4 \times OH}$. In particular, the simulations neglect variability in biogenic VOC emissions (Guenther et al., 2006), trends in anthropogenic emissions and their location, and trends in atmospheric methane. We compare our GEOS-Chem/MERRA simulation to an available GEOS-Chem simulation (M. Mu,
personal communication, 2012) that includes these trends and variability in emissions, uses identical meteorology and resolution, and has minor other changes (version 9-01-01). The two model configurations simulate very similar 1999–2009, meaning that year-to-year changes in biogenic and anthropogenic emissions contribute little additional τCH4×OH variability and do not alter the key parametric factors.

4 Historical (1980–2005) changes in CH4 lifetime

Having established the ability of Eq. (3) to reconstruct τCH4×OH over 1997–2009 in CTMs, we now use it to extrapolate methane lifetime over several decades for which the CTMs have not been run. We begin with the historical period 1980–2005, during which time the key atmospheric forcing variables have been relatively well observed by satellites and ground stations.

In addition to the 5 climate and emission variables identified in Sects. 3.2 and 3.3 as important influences on interannual variability, we include CH4 abundance and anthropogenic NOx, CO and VOC emissions for the historical reconstruction. NOx emissions are divided into land, ship and aviation sectors because their sensitivities differ significantly. In total, the expanded parametric model includes 11 variables. For the 11 sensitivity parameters, ai, in Eq. (3), we adopt values from the average and spread of sensitivities in the 3 CTMs (Table 2, last column).

Table 3 summarizes the data sources for historical climate and emission variables in the expanded parametric model. NASA MERRA reanalysis provides temperature and water vapor data (Bosilovich et al., 2011) and satellite observations provide ozone column data (Stolarski and Frith, 2006). As with the 5-parameter model, ozone columns are averaged over the 40°S to 40°N and other data are global averages. Historical CH4 abundance and anthropogenic and biomass burning emissions follow CMIP5 recommendations (Lamarque et al., 2010; Meinshausen et al., 2011b). Global annual lightning flash rates have varied by up to 20 % since 1998, but multi-decadal trends are not apparent (Murray et al., 2012), so we assume no change since 1980, with 10 % Gaussian uncertainty in the trend.

Figure 5 shows the reconstructed historical changes in τCH4×OH and the contribution to those changes from each climate and emission variable. We assess uncertainty in τCH4×OH by generating 105 monte carlo realizations of Eq. (3), allowing all parameters, ai, and lightning to vary independently within their uncertainties. This does not account for possible errors in emissions, ozone observations, or meteorological assimilation that are difficult to quantify. (See Fig. S6 for uncertainties in τCH4×OH due to each component.)

Our reconstructed τCH4×OH has annual variability of 1–2 % over the 1980–2005 period. Reductions in τCH4×OH occur during El Niño years (1982–1983, 1987–1988, and 1997–1998) driven mainly by water vapor and reinforced by a smaller effect from temperature. τCH4×OH is also depressed through much of the 1990s when stratospheric ozone was low, due to the solar cycle and Mt. Pinatubo, enhancing UV penetration and photolysis in the troposphere. The largest spikes in τCH4×OH occur when the solar cycle maximum and La Niña are synchronous, as in 1989 and 1999–2000. Overall, the parametric model simulates a decrease in τCH4×OH since 1980, which has also been found in numerous CTM and GCM studies (Duncan et al., 2000; Karlsdottir...
Figure 5. Recent historical $\tau_{CH_4 \times OH}$ variation (left) and its component causes (right). Lifetime reconstructed in this work from Eq. (3), with components shown at right. Shaded region shows $\pm \sigma$ uncertainty propagated from adopted parameter ranges in Table 2, but not including possible errors in reanalysis meteorology, ozone column observations, or emission inputs. All data are anomalies with respect to their 2000–2005 means. The anthropogenic (anthro) NOx component combines the separate effects of land, ship, and aircraft emissions. See Fig. S6 for uncertainties in each component.

and Isaksen, 2000; Dentener et al., 2003; Stevenson et al., 2005; Hess and Mahowald, 2009; John et al., 2012; Naik et al., 2012). This is an improvement over previous parametric approaches, which are shown in Fig. 5, that produce zero or positive trends over the same period (Prather et al., 2001; Meinhausen et al., 2011a).

Figure 6 identifies the contribution of each variable to the total change in $\tau_{CH_4 \times OH}$. Rising atmospheric methane has the largest influence on $\tau_{CH_4 \times OH}$, but the positive methane feedback effect (3.5 %) is more than compensated by negative climate and emission terms. Temperature and water vapor, which have increased due to GHGs, decrease $\tau_{CH_4 \times OH}$ by 2 %, collectively, although the water vapor effect is about 3 times larger. Halogen-driven decreases in stratospheric ozone also shortened the lifetime about 1 %. Increases in ship and land anthropogenic NOx emissions both decrease $\tau_{CH_4 \times OH}$ by 1.5 %, despite the ship source having much smaller total magnitude. Lightning NOx could also have an important impact on $\tau_{CH_4 \times OH}$, but the lightning trends are not known.

The total $\tau_{CH_4 \times OH}$ change from 1980–1985 to 2000–2005 is $-2.2 \pm 1.8$ % in our model, or $-0.12$ % yr\(^{-1}\) from a linear fit. Dentener et al. (2003), simulated a larger decrease, $-0.2$ % yr\(^{-1}\), in the 1980s that they attributed mainly to water vapor. Meteorological inputs may contribute to the difference, since water vapor trends are known to vary amongst reanalysis products (Trenberth et al., 2011). In addition, the shift of anthropogenic emissions to SE Asia, which alters the sensitivity of $\tau_{CH_4 \times OH}$ to emissions is not treated in the parametric model (e.g. Fuglestvedt et al., 1999; Karlsson and Isaksen, 2000). Methyl chloroform analyses generally suggest large decreases in $\tau_{CH_4 \times OH}$ during the 1980s followed by increases during the 1990s, which conflicts with the CTM results (Bousquet et al., 2005; Prinn et al., 2005). Assuming uncertainty of about 20 % in methyl chloroform emissions, however, reconciles the observations with the small trends found in CTMs and in our parametric model (Krol and Lelieveld, 2003; Prinn et al., 2005).
Fig. 7. Projected future methane abundance (left) and total lifetime (right) for RCP 8.5. Projected uncertainty (shaded) is the standard deviation from 10⁵ monte carlo integrations, accounting for uncertainty in the present-day budget, emissions, and climate-chemistry effects on τCH₄×OH. Our projections are compared to MAGICC model (Meinshausen et al., 2011a) and the IPCC TAR formula (Prather et al., 2001).

5 Future (2010–2100) CH₄ and CH₄ lifetime

We now apply the parametric model to predict methane and methane lifetime, with their uncertainties, for a future socioeconomic scenario. We make predictions for RCP 8.5 (Riahi et al., 2007), a scenario with rapid climate warming, but these methods apply to other scenarios as well. The prediction begins with the best estimate of present-day (2010) methane budget, including natural (202 ± 35 Tg yr⁻¹) and anthropogenic (352 ± 45 Tg yr⁻¹) emissions and lifetimes for loss by tropospheric OH (11.2 ± 1.3 yr), tropospheric Cl (200±100 yr), stratospheric reactions (120±24 yr), and soil uptake (150±50 yr) (Prather et al., 2012). The scenario specifies future anthropogenic methane emissions and we assume natural emissions could change ±20% (1σ) by 2100 due to climate feedbacks, which is about twice the change in wetland emissions since the preindustrial era (Houweling et al., 2000). We use the parametric model to predict future τCH₄×OH and assume other loss rates are constant. For future predictions we use the same expanded set of 11 parameters as were used in the historical τCH₄×OH reconstruction (Table 2, last column). Table 3 lists data sources for the future climate and emission variables. We then generate 10⁵ monte carlo realizations of future methane abundance in RCP 8.5, sampling over the uncertainties in all parametric factors and forcing variables in Eq. (3), as well as uncertainties in the present-day methane budget.

Table 4 summarizes the predicted changes in climate and emissions in RCP 8.5. In this scenario most anthropogenic emissions of ozone precursors decrease by 2100 (7–75%), although aircraft NOₓ emissions rise 123%. Biomass burning emissions, also specified by the scenario, decrease 35%, which we assume applies uniformly to all gases and aerosols from fires. The parametric model requires future global-mean air temperature and water vapor inputs consistent with the scenario. We calculate these from global-mean surface temperature in CMIP5 models that have simulated RCP 8.5 (Climate Explorer, http://climexp.knmi.nl/, accessed 18 December 2012) using the strong regression relationship between annual-mean surface temperature, air temperature and water vapor that we derive from historical reanalysis data since 1979 (See Supplement for details). We use global-mean surface temperatures to drive the parametric model because these data are more easily available and because this simplifies future applications of the parametric model to other climate scenarios. The range of future surface temperatures in the CMIP5 ensemble is propagated to uncertainty in future air temperature and water vapor using regression fitting errors from the historical reanalysis. In 2100, we calculate air temperature and water vapor to be 3.6 ± 0.9 K and 43.2 ± 9.5 % larger than 2010, respectively. For tropical stratospheric ozone, multiple models predict recovery.

![Figure 7](https://example.com/figure7.png)

![Figure 8](https://example.com/figure8.png)

**Table 4. Changes (2010–2010) in climate, emissions, and τCH₄×OH for RCP 8.5*.**

<table>
<thead>
<tr>
<th>Variable</th>
<th>Variable change</th>
<th>τCH₄×OH change, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air temperature</td>
<td>+3.6 ± 0.9 K</td>
<td>-4.2 ± 1.5</td>
</tr>
<tr>
<td>Water vapor</td>
<td>+43.2 ± 9.5 %</td>
<td>-12.9 ± 3.1</td>
</tr>
<tr>
<td>Ozone column, 40° S–40° N</td>
<td>+0.7 ± 3.0 %</td>
<td>+0.4 ± 1.7</td>
</tr>
<tr>
<td>Lightning NOₓ emissions</td>
<td>+10 ± 20 %</td>
<td>-1.2 ± 3.3</td>
</tr>
<tr>
<td>Biomass burning emissions</td>
<td>-34.8 %</td>
<td>+0.5 ± 0.3</td>
</tr>
<tr>
<td>CH₄ abundance</td>
<td>+78.2 ± 8.4 %</td>
<td>+28.9 ± 7.4</td>
</tr>
</tbody>
</table>

*Variable changes from data sources in Table 3, except CH₄ abundance, which is calculated parametric model, as described in Sect. 5. τCH₄×OH component changes derive from the variable changes and the sensitivities in Table 2, including uncertainties in both. Uncertainties are standard deviations from 10⁵ monte carlo realizations.
to 1980 levels around 2045 due to the decrease of long-lived halogenated gases (Newman et al., 2007; Austin and Scinocca, 2010; Eyring et al., 2010a), followed by GHG-driven ozone decreases through 2100 (Eyring et al., 2010a). We adopt this projection, adding uncertainty that grows to 3% of the total column in 2100. Lightning NO\textsubscript{x} emissions have been estimated in past work to grow 5–50% by the late 21st century (Wu et al., 2008), but these predictions are highly speculative due to poor mechanistic understanding of present-day global flash rates. GHG-driven climate warming tends to reduce convection globally (Held and Soden, 2006), but could intensify convection in some regions (Del Genio et al., 2007), so the total effect on lightning is unclear. In this work we assume 10% increase by 2100, but allow broad Gaussian uncertainty of 20%.

Figure 7 shows future methane and its uncertainty. Projected abundances reach 3990 ± 330 ppb in 2100, which is about 400 ppb lower than our previous work (Prather et al., 2012), which did not account for emissions and climate controls on τ\textsubscript{CH\textsubscript{4}×OH}. MAGICC predicts lower concentrations, 3750 ppb, due mainly to the strong negative sensitivity of τ\textsubscript{CH\textsubscript{4}×OH} to temperature in that model, but the MAGICC values lie within our estimated uncertainties throughout the 21st century. Statistical uncertainties in methane predictions are 8% in 2100, based on the assessed processes in the parametric model. Neglected processes – including shifting emission locations, biogenic VOC emissions, stratosphere-troposphere exchange, and cloud, dust and aerosol interactions with photolysis and chemistry – might cause additional systematic prediction errors, but we have found that including these processes in one model (GEOS-Chem/MERRA) has little impact on present-day τ\textsubscript{CH\textsubscript{4}×OH} variability (Sect. 3.3).

The parametric model predicts τ\textsubscript{CH\textsubscript{4}×OH} will increase +12.9 ± 10.8% by 2100 (Fig. 7). MAGICC gives similar results (+12.6%), but the IPCC TAR formula yields a larger result (+29.7%), consistent with their respective historical performances in Sect. 4. The ACCMIP model ensemble predicts +8.5 ± 10.4% for RCP 8.5 (Voulgarakis et al., 2012). Lightning NO\textsubscript{x} emissions likely explain the τ\textsubscript{CH\textsubscript{4}×OH} difference, since ACCMIP models calculate 24 ± 29% increase in 2100 (Voulgarakis et al., 2012), compared to our assumed increase of 10 ± 20%. Although we do not think future lightning estimates from GCMs are robust (see above), assuming an equally large lightning change in the parametric model would lower τ\textsubscript{CH\textsubscript{4}×OH} in 2100 by about 5%, after including the methane feedback. The similar central estimates and ranges of τ\textsubscript{CH\textsubscript{4}×OH} change in this work and ACCMIP, after accounting for different lightning assumptions, demonstrates that our simple parametric model represents the major chemistry-climate interactions and uncertainties affecting methane in current GCMs. This supports using the parametric model for climate scenarios where a large ensemble of GCMs with chemistry is not available and too costly to generate.

Figure 6 and Table 4 decompose the net τ\textsubscript{CH\textsubscript{4}×OH} changes in 2100 into components due to each climate and emission variable. For emission terms, all uncertainty comes from the sensitivity parameter since the scenario emissions are taken as given, while uncertainty in other terms is a combination of possible errors in the forcing variable and sensitivity. Methane feedback has the largest influence, with an individual contribution of +28.9 ± 7.4% on NO\textsubscript{x} emission reductions over land also force τ\textsubscript{CH\textsubscript{4}×OH} upwards (+11.2 ± 2.5%), which is opposite to NO\textsubscript{x} influence in recent decades. Other climate and emission components are zero or negative, with water vapor having the largest effect (−12.9 ± 3.1%). Stratospheric ozone and lightning NO\textsubscript{x} contribute little to τ\textsubscript{CH\textsubscript{4}×OH} changes, but they make a significant contribution to the uncertainty. MAGICC predicts similar τ\textsubscript{CH\textsubscript{4}×OH} changes to the parametric model because its temperature response is similar to the combined effects of temperature and water vapor in our work, and sensitivities to the other dominant terms – land NO\textsubscript{x} and methane feedback – have changed little since the IPCC TAR, on which MAGICC is based. The parametric model and MAGICC will differ in socioeconomic and climate scenarios where other emissions, ozone, or lightning drive τ\textsubscript{CH\textsubscript{4}×OH} changes.

6 Methane global warming potential

Global Warming Potentials (GWP) are useful for comparing the radiative forcing (RF) caused by emissions of various GHGs having different absorbances and atmospheric lifetimes. The methane GWP customarily accounts for the direct RF from the emitted gas, as well as indirect RF caused by methane-induced increases in ozone, stratospheric water vapor, and feedback on the methane lifetime (Forster et al., 2007). Here we evaluate the methane GWP implied by the perturbation experiments. Radiative forcing of methane and ozone are calculated for the control simulation and a simulation with 5% more methane, using the University of Oslo radiative transfer model (Myhre et al., 2011). A small perturbation is used to satisfy the linearity assumption in the methane feedback factor derivation. In addition, we test the effect of methane-induced water vapor on stratospheric ozone, with an additional Oslo CTM3 simulation in which stratospheric water vapor was increased to maintain equilibrium with the CH\textsubscript{4} perturbation. To our knowledge, this indirect, H\textsubscript{2}O-mediated effect on ozone has not been included in prior assessments of methane GWP.

Table 5 summarizes ozone changes and RF results for all simulations, normalized to 1 ppb CH\textsubscript{4} perturbations. Tropospheric ozone changes in GEOS-Chem and the UCI CTM (2.9 and 4.0 DU ppm(CH\textsubscript{4})\textsuperscript{-1}, respectively) are within the range of previous multi-model studies (Holmes et al., 2011; Boucher, 2012; Fry et al., 2012). Oslo CTM3, however, exhibits larger tropospheric changes (5.1 DU ppm(CH\textsubscript{4})\textsuperscript{-1}), likely due to the effects of stratospheric chemistry on the
Upper troposphere. Stratospheric ozone increases twice as much as tropospheric ozone (10.4 DU ppm(CH₄)^{-1}), but the change is small compared to the total stratospheric column. We find that stratospheric water vapor produced by oxidation of methane causes small decreases in stratospheric ozone (−4.3 DU ppm(CH₄)^{-1}). Although these stratospheric effects have greater uncertainty because they are assessed from a single model, the stratospheric chemistry mechanism in Oslo CTM3 is able to reproduce recent stratospheric ozone variability (Isaksen et al., 2012).

Ozone generally has greater radiative forcing efficiency in the troposphere than in the stratosphere (Forster and Shine, 1997), so tropospheric ozone changes tend to dominate the ozone RF components. In our 3 models, the methane RF through tropospheric ozone is 30–50% of the direct methane RF. After including stratospheric changes, ozone contributes up to 65% of the direct RF. Previous IPCC assessments have assumed 25% for purposes of calculating GWP (Shine et al., 1995; Forster et al., 2007), similar to a recent estimate of 21% based on tropospheric changes alone (Fry et al., 2012). Methane perturbation data from the TAR (3.67 DU(O₃)ppm(CH₄)^{-1}) (Prather et al., 2001), however, suggest that tropospheric ozone RF is about 40% of the methane RF (154 mW m^{-2} ppm(CH₄)^{-1}, assuming efficiency of 42 mW m^{-2} DU(O₃)^{-1}) (Ramaswamy et al., 2001).

Accounting for both direct and indirect effects, the methane RF efficiency, F_e, is 620 mW m^{-2} ppm(CH₄)^{-1} in steady-state. A 1 Tg pulse emission of methane raises the atmospheric abundance by δ = 0.364 ppb, which decays at a rate f τ_{CH₄}, where f = 1.34 is the methane feedback on its lifetime. We use τ_{CH₄} = 9.14 yr (Prather et al., 2012). Neglecting delays between emission time and stratospheric impacts, the 100-yr absolute GWP is δ f τ_{CH₄} F_e = 2.76 mW yr m^{-2}, compared to 0.087 mW yr m^{-2} for CO₂. Thus, the methane GWP_{100} is 31.8. Our result is higher than several previous reports, generally near 25 (Forster et al., 2007; Fry et al., 2012), mainly because we include stratospheric ozone effects, but also because the updated and longer methane lifetime used here (Prather et al., 2012). IPCC TAR recommended f = 1.4 (Prather et al., 2001), which would imply an even larger GWP, but since f depends on τ_{CH₄}, the two must be chosen consistently. Uncertainty in the GWP is difficult to assess without further modeling and analysis of stratospheric impacts, but it is likely ±20% or larger.

### 7 Conclusions

Over 1997–2009, the three CTMs in this work exhibit common variability in methane lifetime, which is also shared by other published model studies. The simulated τ_{CH₄×OH} anomalies generally lie within the constraints provided by
global methyl chloroform observations, but correlate poorly with year-to-year changes inferred from the NOAA and AGAGE networks. However, while both networks provide strong and mutually consistent estimates of the long-term mean $\tau_{CH_2O}$, their year-to-year anomalies differ from each other by as much as they differ from the CTMs. These differences in global $\tau_{CH_2O}$ anomalies between networks can partially be explained by small discrepancies ($\sim 0.1$ ppt) in the monthly mean methyl chloroform concentrations at collocated measurement sites.

We quantitatively explain $\tau_{CH_2O}$ variability in the CTMs with 5 climate and emission variables: air temperature, water vapor, ozone column, biomass burning emissions, and lightning NOx emissions. A parametric model built on these 5 factors reproduces 90% of the variability in methane lifetime during 1997–2009. For projections over many decades, the parametric model also includes 5 factors for anthropogenic emissions and one for methane feedback. The ensemble of 3 models provides a measure of uncertainty in each parametric factor, which we use to project past and future methane and its lifetime, with uncertainties. While this approach lacks the full complexity of atmospheric chemistry that can be included through multi-decadal simulations of a CTM or GCM, the advantage is that it can be rapidly applied to many climate data sets or socioeconomic scenarios.

Using the parametric model to reconstruct methane lifetime for 1980–2005, we estimate $2.2 \pm 1.8$ % decrease in $\tau_{CH_2O}$, in contrast to previous parametric approaches that suggested increases or no change. Our decrease is the same direction of change as previous CTM studies and consistent with methyl chloroform observations, but smaller in magnitude than many past estimates. The parametric model may underestimate the trend because it does not consider the location of anthropogenic emissions. We also use the parametric model to project future methane abundance, which implicitly assumes that the emission and climate processes that controlled methane lifetime in the recent past will remain dominant throughout the 21st century. For the RCP 8.5 scenario, our projected methane abundances are larger than the CMIP5 recommendations, which are based on the MAGICC model, but the uncertainty encompasses the difference. Our projected change in methane lifetime is consistent with an ensemble of GCMs containing atmospheric chemistry, providing further evidence that the parametric model includes the key processes. Uncertainty in 2100 methane abundance is 10% based on the processes we have assessed here, which include a simple treatment of natural methane emissions. Water vapor, anthropogenic NOx emissions, and methane feedback on its OH sink are the major drivers of $\tau_{CH_2O}$ in both the historical reconstruction and future RCP 8.5 scenario.

We also provide a new estimate of the indirect components of methane RF. Based on three CTMs, tropospheric ozone contributes 30–50% of the direct methane RF, compared to 25% that has been used in previous IPCC assessments (Forster et al., 2007). Using one model with stratospheric chemistry (Oslo CTM3), we calculate the stratospheric chemical effects of methane, including those mediated by water vapor. Combining the troposphere and stratosphere results, the total methane-induced ozone RF is 50% of the direct methane RF. Based on these data, the 100-yr methane GWP is 32, which is higher than several previous estimates around 25.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys.net/13/285/2013/acp-13-285-2013-supplement.pdf.

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