

**OH and methane  
lifetime in the  
ACCMIP simulations**

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# Analysis of present day and future OH and methane lifetime in the ACCMIP simulations

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## Abstract

Results from simulations performed for the Atmospheric Chemistry and Climate Modeling Intercomparison Project (ACCMIP) are analysed to examine how OH and methane lifetime may change from present-day to the future, under different climate and emissions scenarios. Present-day (2000) mean tropospheric chemical lifetime derived from the ACCMIP multi-model mean is  $9.8 \pm 1.6$  yr, lower than a recent observationally-based estimate, but with a similar range to previous multi-model estimates. Future model projections are based on the four Representative Concentration Pathways (RCPs), and the results also exhibit a large range. Decreases in global methane lifetime of  $4.5 \pm 9.1$  % are simulated for the scenario with lowest radiative forcing by 2100 (RCP 2.6), while increases of  $8.5 \pm 10.4$  % are simulated for the scenario with highest radiative forcing (RCP 8.5). In this scenario, the key driver of the evolution of OH and methane lifetime is methane itself, since its concentration more than doubles by 2100, and it consumes much of the OH that exists in the troposphere. Stratospheric ozone recovery, which drives tropospheric OH decreases through photolysis modifications, also plays a partial role. In the other scenarios, where methane changes are less drastic, the interplay between various competing drivers leads to smaller and more diverse OH and methane lifetime responses, which are difficult to attribute. For all scenarios, regional OH changes are even more variable, with the most robust feature being the large decreases over the remote oceans in RCP 8.5. Through a regression analysis, we suggest that differences in emissions of non-methane volatile organic compounds and in the simulation of photolysis rates may be the main factors causing the differences in simulated present-day OH and methane lifetime. Diversity in predicted changes between present-day and future was found to be associated more strongly with differences in modelled climate changes, specifically global temperature and humidity. Finally, through perturbation experiments we calculated an OH feedback factor ( $F$ ) of 1.29 from present-day conditions (1.65 from 2100 RCP 8.5 conditions) and a climate feedback on methane lifetime of  $0.33 \pm 0.13 \text{ yr K}^{-1}$ , on average.

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## 1 Introduction

Oxidation processes remove a range of environmentally important species from the atmosphere. Tropospheric oxidation heavily depends on the levels of the hydroxyl radical (OH) and its geographical distribution (Levy, 1971). Perhaps most prominently, oxidation by OH is the primary loss mechanism for methane (CH<sub>4</sub>), the second most important anthropogenic greenhouse gas in the climate system (Forster et al., 2007; Shindell et al., 2009), and an important precursor of tropospheric ozone (O<sub>3</sub>) (Logan et al., 1981). Thus, OH abundance and methane lifetime are commonly studied simultaneously. Besides its role in methane cycling, OH is also involved in removing trace gases from the atmosphere, through oxidation of atmospheric pollutants such as nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO) and non-methane volatile organic compounds (NMVOCs), and removes ozone-depleting substances such as hydrofluorocarbons (HFCs) from the atmosphere (DeMore et al., 1996). Furthermore, OH participates in the formation of atmospheric aerosols such as sulfate, nitrate and secondary organics (e.g. Koch et al., 2006).

OH production in the atmosphere is initiated by the photolysis of ozone at wavelengths smaller than 330 nm:



The product of this temperature-dependent interaction with sunlight is an excited oxygen atom (O<sup>1</sup>D), which then combines with water vapour to produce two molecules of OH:



Thus, high levels of ozone, shortwave radiation and humidity favour the production of OH, and lead to a reduction in the methane lifetime (Logan et al., 1981; Lelieveld et al., 2002). In turn, ozone depends on emissions of its precursors and on climatic conditions, while water vapour abundances are determined by temperature changes and by related underlying processes. Shortwave radiation is modified by overhead absorption

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by ozone, scattering and absorption by clouds and aerosols, and reflections from the Earth's surface (Madronich, 1987; Voulgarakis et al., 2009a). Since all these factors depend on a variety of physical and chemical processes, understanding of OH on global and regional scales is a challenge.

5 After its generation, OH has a very short lifetime, of the order of a few seconds (Lelieveld et al., 2004 and reference therein) making measurements particularly challenging. Even with in-situ measurements, its spatial variability makes it difficult to constrain OH abundances at larger spatial scales. For this reason, modelling becomes an essential tool to probe the spatial variability of OH and its drivers, as well as its effects  
10 on methane and other species, at different timescales. Globally, the main observational constraint available for the OH abundance and methane lifetime is via methyl chloroform ( $\text{CH}_3\text{CCl}_3$ , also referred to as MCF) measurements (e.g. Montzka et al., 2011; Prinn et al., 1995). Methyl chloroform has fairly well known sources (now almost zero) and very well-known concentrations, making the calculation of its loss rate, and thus of  
15 global OH concentrations feasible.

Past studies have examined the evolution of global OH and methane lifetime since preindustrial times (e.g. Wang and Jacob, 1998; Lelieveld et al., 2002), or their recent trends and interannual variability, using either observations (Bousquet et al., 2005; Manning et al., 2005; Prinn et al., 2005) or models (Dentener et al., 2003; Dalsoren  
20 et al., 2006; Fiore et al., 2006). There has also been work examining the potential future evolution of these quantities, though not exhaustive, and with contrasting results (Thompson, 1992; Lelieveld et al., 1998; Stevenson et al., 2000; Johnson et al., 2001; Prather et al., 2001; Shindell et al., 2006a, 2006b; Stevenson et al., 2006; Wild and Palmer, 2008; Zeng et al., 2010). Several of these studies have been performed  
25 with chemistry-transport models (CTMs), and often considered only ozone precursor emission effects, without accounting for simultaneous climate changes. Typically, these studies predicted OH decreases and methane lifetime increases in the future. For example, Lelieveld et al. (1998) found a 6% increase in methane lifetime from 1992 to 2050 due to increases in CO and methane emissions, both of which consume OH

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and prolong methane lifetime. However, an earlier study by Thompson (1992) suggests that future changes in OH would most likely be small, due to cancelling effects of methane/CO increases and tropospheric ozone increases. Wild and Palmer (2008), using the A2 SRES emissions scenario (IPCC, 2000), found methane lifetime increases of 13 % in 2100 compared to 2000, with a strong shift in OH abundances from oceanic to tropical continental regions, due to the differing effects of methane and ozone (methane consumes OH while ozone generates it). The model experiments performed in support of the Intergovernmental Panel for Climate Change (IPCC) Third Assessment Report (TAR) fall into this same category of experiments that did not account for climate change effects (Prather et al., 2001). All models calculated global mean OH decreases (and thus methane lifetime increases) between 2000 and 2100, ranging from 6 % to 25 %.

Global modelling studies that took both emissions and climate changes into account were first performed around the time of the publication of IPCC TAR, and, contrary to the studies that only included emission changes, found that future methane lifetime either remained unaffected or significantly decreased. This was attributed to increases in temperature, which drive a faster  $\text{CH}_4 + \text{OH}$  reaction as well as higher water vapour concentrations, increasing the rate of the  $\text{O}^1\text{D} + \text{H}_2\text{O}$  reaction (Johnson et al., 2001; Stevenson et al., 2000). More recently, Zeng and Pyle (2010) found methane lifetime decreases of 11 % by 2100, using the SRES A1B scenario as a basis. This supported the earlier findings by Shindell et al. (2006a), whose simulations with a chemistry-climate model (CCM) showed a 10 % decrease in the lifetime, though using the SRES A2 emissions scenario for 2100. Results published around the time of the IPCC Fourth Assessment Report (AR4) projected only minor changes in global OH between 2000 and 2030 (Shindell et al., 2006b; Stevenson et al., 2006). Finally, John et al. (2012) found decreasing methane lifetimes between 2000 and 2100 in three out of the four Representative Concentration Pathway (RCP) scenarios used (Meinshausen et al., 2001; van Vuuren et al., 2011), with methane lifetimes increasing only in the extreme RCP 8.5 scenario where methane abundances more than double from 2000 to 2100.

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Despite the number of studies examining the topic of OH and methane lifetime, there is still not a clear consensus on the main issues related to it, not least because there have not been systematic studies focusing on results from multiple composition-climate models, which include many of the processes affecting oxidant changes. Here, we analyse simulations performed for the Atmospheric Chemistry and Climate Modeling Intercomparison Project (ACCMIP), in support of the IPCC Fifth Assessment Report (AR5), to investigate changes in OH and methane lifetime between 2000 and 2100. This is the first study that uses the RCP scenarios in a multi-model framework to study this topic. The ACCMIP project includes a variety of CCMs, which were run for the historical period (1850 to present-day, with present-day defined as year 2000) and for the future (present-day to 2100) following the different RCPs. A wide range of chemical output from these simulations is expected to contribute to a deeper understanding of chemistry-climate interactions in long-term climate simulations (e.g. for the Coupled Model Intercomparison Project Phase 5, or CMIP5). This study complements work that is being done under ACCMIP on historical OH and methane lifetime (Naik et al., 2012a), historical and future ozone (Young et al., 2012), and ozone radiative forcing (Stevenson et al., 2012; Bowman et al., 2012; Shindell et al., 2012a). An overview of ACCMIP with an evaluation of present-day simulated climate is provided in Lamarque et al. (2012).

In Sect. 2, we will briefly describe the participating models and the simulations performed. Section 3 describes the evolution of OH and methane lifetime between present-day and future, while Sect. 4 presents the evolution of potential drivers affecting these quantities. Section 5 analyses model sensitivity experiments that were performed to isolate individual drivers of change, while Sect. 6 explores the reasons for model diversity in simulating the quantities of interest. Section 6 summarizes the conclusions of the study.



## 2 Description of models and experiments

### 2.1 Models

We have used data from 14 models, which performed the future ACCMIP simulations (see Table A1). Most of the models are CCMs, with the exception of CICERO-OsloCTM2, MOCAGE and STOC-HadAM3, which are chemistry-transport models (CTMs). The CCMs were run with an atmosphere-only configuration, with sea-surface temperature and sea-ice data coming either from coupled ocean-atmosphere model simulations or from observations. The models that are more or less linked to coupled climate models that participate in CMIP5 are: CESM-CAM-superfast (uses sea surface temperatures (SSTs)/sea-ice (SI) from CESM-CAM), CMAM (based on the preceding generation GCM, but using SSTs/SI from CanESM2), GFDL-AM3 (atmosphere-only version of GFDL-CM3), GISS-E2-R (the same runs were used both for CMIP5 and for ACCMIP), LMDzORINCA (uses SSTs/SI from IPSL-CM4 AR4 exercise), MIROC-CHEM (atmosphere-only version of MIROC-ESM-CHEM), MOCAGE (uses meteorology produced using CNRM-CM5 SSTs/SI), STOC-HadAM3 and UM-CAM (both using SSTs/SI from HadGEM2 coupled simulations). EMAC used SSTs/SI from a CMIP5 run carried out with the CMCC Climate Model, which is, like EMAC, based on ECHAM5, although differences between the atmospheric component exist in resolution and short-wave radiation code (Cagnazzo et al., 2007). GEOSSCM, HadGEM2, and NCAR-CAM3.5 used SSTs from the AR4 simulations, with the best possible correspondence between RCPs and SRES scenarios. Finally, CICERO-OsloCTM2 used ECMWF IFS model forecast data for 2006 for all simulations. Detailed model descriptions are provided in the ACCMIP overview paper of Lamarque et al. (2012a). Below we present some of the main features of the models, with an emphasis on those that are important for OH and methane lifetime.

Surface methane concentrations were prescribed in all models, except for (a) LMDzORINCA, in which specified emission fluxes were used, and (b) GISS-E2-R, in which interactive emissions for wetlands and prescribed emissions from other sectors were

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used (see Shindell et al., 2012b). For methane concentrations at the surface, most models used data from the database of Meinshausen et al. (2011), except for CICERO-OsloCTM2 and EMAC that used present-day methane values from IPCC TAR and from AGAGE (Prinn et al., 2000), respectively, scaled to match the evolution in Meinshausen et al. (2011) in the future. UM-CAM and STOC-HadAM3 are the only models having a globally constant concentration of methane. Note that methane concentrations vary between different timeslices in all models.

Anthropogenic and biomass burning emissions of  $\text{NO}_x$ , CO, NMVOCs and aerosols used in the simulations were identical in all models (Lamarque et al., 2010; 2012a). Emissions from other sources (i.e. natural) were different between the models, and varied widely (Lamarque et al., 2012a). The distribution and magnitude of lightning  $\text{NO}_x$  emissions depend on the model's convection (mostly based on Price and Rind, 1992, 1994; Price et al., 1997; Grewe et al., 2001, was used for EMAC), in all models except GEOSCCM, which used constant global lightning emissions of  $5 \text{ Tg Nyr}^{-1}$ , and CICERO-OsloCTM2 in which the distribution of lightning emissions depends on modelled convection, but with a scaling applied to produce  $5 \text{ Tg Nyr}^{-1}$  of emissions. Specifically, all models used the cloud top height in order to determine lightning flash rates and hence lightning  $\text{NO}_x$  emissions, except for CMAM, which used the convective updraft mass flux (Allen and Pickering, 2002). Note that there were some inconsistencies in the implementation of lightning  $\text{NO}_x$  emissions in HadGEM2 and MIROC-CHEM for this project, which led to significant underestimates and overestimates, respectively.

Isoprene emissions are climate-sensitive in EMAC, GEOSCCM, GISS-E2-R, and STOC-HadAM3, while the rest of the models use different kinds of estimates, except CMAM and HadGEM2 in which  $250 \text{ Tgyr}^{-1}$  and  $475 \text{ Tgyr}^{-1}$  of CO, respectively, are emitted as proxy for isoprene oxidation. Interactive emissions of other NMVOCs are also included in some models: GISS-E2-R includes climate-sensitive terpene emissions, while in GEOSCCM there are propene and CO emissions as a proxy for terpenes/methanol. EMAC and GEOSCCM include climate-sensitive soil  $\text{NO}_x$  emissions, while in the rest of the models this source is fixed. Constant fluxes are also assumed

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for oceanic CO in all models except GEOSCCM and GISS-E2-R. The spread in past, present-day and future emissions in the models is shown in Fig. 1 of Young et al. (2012).

Gas-phase chemistry schemes ranged in terms of complexity. The models with more than 100 gaseous species included are EMAC, GEOSCCM, MOCAGE, and NCAR-CAM3.5, while CESM-CAM-superfast had the lowest number of species (16). VOCs other than methane are either not included (CMAM) or included in several different ways in models, with lumping often applied to group NMVOCs in broad categories. Stratospheric ozone in HadGEM2, STOC-HadAM3, and UM-CAM was prescribed following Cionni et al. (2011; in support of CMIP5), while in LMDzORINCA the climatology of Li and Shine (1995) was used. In CICERO-OsloCTM2, monthly model climatological values of ozone and nitrogen species are used, except in the 3 lowermost layers in the stratosphere (approximately 2.5 km) where the tropospheric chemistry scheme is applied to account for photochemical ozone production (Skeie et al., 2011). A simplified scheme was used in CESM-CAM-superfast (McLinden et al., 2000). In the rest of the models, there was a full simulation of stratospheric ozone.

Photolysis treatment in some models broadly follows the approach of using pre-calculated photolysis rates and correcting for real-time atmospheric conditions (clouds, overhead ozone, and, in some cases, surface albedo). On the other hand, CICERO-OsloCTM2, GISS-E2-R, EMAC, GEOSCCM, and MIROC-CHEM used state-of-the-art, fully interactive photolysis schemes (Wild et al., 2000, for the former two; Landgraf and Crutzen, 1998, for the latter three), while HadGEM2 and UM-CAM used offline rates (Law and Pyle, 1993). These are the only two models where prognostic clouds and overhead ozone column did not affect the photolysis calculations.

For more information on model characteristics, see Lamarque et al. (2012a).

## 2.2 Experiments

The model experiments analysed here are the present-day-to-future (2000–2100) simulations performed by the ACCMIP models. The models are configured as described in Sect. 2.1. Short-lived precursor emissions (Lamarque et al., 2012a) and long-lived

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species concentrations (Meinshausen et al., 2011) follow the RCPs. There are four RCP emissions/concentrations scenarios, RCP 2.6, RCP 4.5, RCP 6.0 and RCP 8.5, with RCP 2.6 featuring the least radiative forcing, while RCP 8.5 featuring the most. The percentage change of global  $\text{NO}_x$ , CO and NMVOC emissions in these scenarios can be seen in Tables 2, S1, S2 and S3. The models were run for different “timeslices”, representative of conditions around 2000, 2030 and 2100. In a few cases, simulations were performed for 2010 and 2050 as well. The proposed simulation length for each timeslice was 4–10 yr (after spin-up) using prescribed monthly SSTs, valid for each timeslice and averaged over 10 yr (see number of simulated years for each model in Lamarque et al., 2012a). There are certain gaps in the data provided (e.g. missing variables for some models), but overall the dataset is fairly consistent.

In addition to the above-mentioned simulations, sensitivity experiments were conducted by some modelling teams: (a) simulations with present-day emissions but climatic conditions set to 2030 or 2100 levels, and (b) simulations with perturbed methane concentrations. While these experiments were only performed by a sub-set of the ACCMIP models, in combination with some further tests performed solely with GISS-E2-R, they provide further valuable insight into the processes controlling OH and methane lifetime (see Sect. 5.3).

### 3 Present-day and future OH and methane lifetime

#### 3.1 Global changes

Table 1 shows present-day (2000) global mean OH concentrations, chemical methane lifetime, and total methane lifetime, calculated from all the ACCMIP models. The chemical lifetime is calculated by dividing the global atmospheric methane burden with the global tropospheric chemical loss, while the total lifetime includes the soil and stratospheric sinks in the denominator, following Stevenson et al. (2006). The tropopause is assumed to follow the 200 hPa surface. No interpolation from the model’s native grid

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has been applied for the calculation of any global quantity. A large spread of values for these variables is evident. For example, the simulated chemical methane lifetime ranges from  $\sim 7$  yr (MOCAGE) to  $\sim 14$  yr (UM-CAM). This spread is of similar magnitude to the ACCENT (Atmospheric Composition Change: the European Network of excellence) studies (Shindell. et al., 2006b; Stevenson et al., 2006), conducted around the time of AR4. We find that these global tropospheric metrics do not depend on the definition of the tropopause (Table 1). Present-day diversity in results will be discussed further in Sect. 6.

Before presenting and analyzing the future evolution of OH and methane lifetime, we briefly discuss the historical evolution of methane lifetime in Fig. 1 (three timeslices). Multi-model mean methane lifetime increases by 2.3% from 1850 to 1980, however there is large inter-model diversity in the magnitude and sign of change across the models. Of the 14 models included here, six simulate decreases in methane lifetime with the largest reduction simulated by MOCAGE ( $-8\%$ ) while the rest simulate increases in methane lifetime with the largest increase simulated by GEOSCCM ( $14\%$ ) over the 1850 to 1980 time period. From 1980 to 2000, all models simulate decreases in methane lifetime with a mean lifetime reduction of 4%. Evolution of factors driving changes in methane lifetime and OH over the historical period are discussed in further detail in Naik et al. (2012a).

Figure 2 shows the evolution of modelled global chemical methane lifetime between present-day and 2100, for the four different RCPs. All the timeslices for which a model performed simulations have been included. The present-day lifetimes present a sizeable inter-model spread. A recent observation-based analysis (Prather et al., 2012) estimated chemical methane lifetime to be  $11.2 \pm 1.3$  yr, underestimated by most of the ACCMIP, except GISS-E2-R, HadGEM2, and LMDzORINCA.

The evolution of methane lifetime between different timeslices shows some agreement between different models, in terms of sign. More specifically, in RCP 2.6, most of the 10 models that provided data show decreases between the beginning and the middle of the 21st century, and a slow increase or stabilization later on. Notable excep-

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tions are the MOCAGE and the NCAR-CAM3.5 models, which mostly show increases throughout the period of study. We note that NCAR-CAM3.5 uses the results from a CCSM3 Commitment simulation as an equivalent for RCP 2.6, which leads to an underestimate of the climate effect (see Lamarque et al., 2012a) on methane lifetime.

The overall change between 2000 and 2100 for this scenario is minimal though, on average. For RCP 4.5, for which data from fewer models (7) are available, there is generally a tendency for methane lifetime decreases, especially after 2030. The exception is the GISS-E2-R model, where we find the opposite trend, although with smaller increases than for RCP 2.6. Overall, RCP 4.5 reveals the smallest mean global methane lifetime levels for 2100 relative to 2000. In RCP 6.0 (6 models), there is a mix of positive (2 models) and negative (4 models) trends. Positive trends occur for models that also showed positive trends throughout the RCP 2.6 simulation, though in RCP 6.0 the changes are more rapid.

The scenario with the highest level of agreement between models, in terms of sign, is RCP 8.5. Ten of the 12 models that simulated this scenario show a methane lifetime increase between 2000 and 2100. From the models that simulated both the 2030 and the 2050 timeslice (5), it is evident that the period with the sharpest increase is 2030–2050. The global mean OH evolution (Fig. 3a) also indicates that there is a sharper decline in OH in 2030–2050. This is a period of rapid NO<sub>x</sub> emission reductions (not shown; Lamarque et al., 2012a), which could be the driving factor behind the 2030–2050 feature (e.g. see Lelieveld et al., 2002). In general, since OH drives most of the methane loss in the atmosphere, OH changes correspond well to methane lifetime changes seen in the models (Tables 2, S1, S2 and S3). The models that have an opposite trend for 2000–2100 are HadGEM2 and UM-CAM. These are also the models with the largest absolute methane lifetimes for present-day. Potential reasons for these distinct features will be discussed in Sect. 5.

In Fig. 3b, we also show the ratio between northern and southern hemispheric air mass-weighted mean OH concentration (hereafter N/S ratio) for RCP 8.5. We find that whether this ratio is high or low in a particular model is not correlated with whether

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modelled global mean OH is high or low. For example, the UM-CAM model has the lowest global mean OH in 2000, and it has the highest N/S ratio. On the other hand, in NCAR-CAM3.5, both these metrics have high values relative to the average model. RCP 8.5 shows the most coherent trend (positive) in the N/S ratio, which is opposite in sign to the similarly coherent trend in global mean OH. The large increase in methane in RCP 8.5 consumes a large amount of the southern hemispheric OH in remote regions, which increases the ratio drastically. Furthermore, an equatorward redistribution of anthropogenic emissions in the future may partly shift OH production away from northern midlatitudes, reducing the fraction of global OH that exists in the Northern Hemisphere (Gupta et al., 1998; Wild and Palmer, 2008).

### 3.2 Surface OH changes

While global tropospheric OH is important in determining the lifetimes of various climate-relevant species, it is also crucial to understand the distribution and evolution of OH in the boundary layer, as it reflects the characteristics of the local photochemistry in different areas. A detailed analysis of the regional features of OH and its future changes in different models would require investigation of the distribution of emissions and of each model's regional climate response. Such a detailed analysis is not within the scope of the current study, which mainly aims to discuss the evolution of OH and methane lifetime on a global scale. However, below we present the main features of the regional behavior of OH, and we aim for further regional analysis in a future study.

Figure 4 shows the multi-model mean change in surface OH concentration between 2100 and 2000 for RCP 2.6 and RCP 8.5 (the equivalent plots for individual models are shown in Figs. S1 and S2). We focus on those two simulations because they represent the extreme cases from a climate point of view. There are almost exclusively negative changes in OH abundance in the Southern Hemisphere in RCP 8.5, which become more drastic with latitude and reach up to 50% in the Southern Ocean. In RCP 2.6, the average model shows a mixture of positive and negative changes in the Southern Hemisphere.

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Negative differences are also largely found in the Northern Hemisphere, especially (though not exclusively) in RCP 2.6, although we find strong positive changes in both scenarios over Western Europe. Otherwise, RCP 8.5 changes over heavily populated regions of the Northern Hemisphere are generally small or negative, due to decreases in  $\text{NO}_x$  emissions, similarly to RCP 2.6. The latter leads to decreased secondary OH production. The changes over Europe may be associated with a more dominant role of CO reductions (which drive OH increases), rather than  $\text{NO}_x$  effects. However, note that this effect is not seen in all models (Figs. S1 and S2). For example, in HadGEM2, positive changes over Europe are dramatic, whereas for GISS-E2-R there are solely negative changes.

Over the oceans in RCP 8.5, despite the increases of water vapour in this scenario, the dominant factor driving OH changes is methane, since it has a long-enough lifetime to travel away from its source regions and be relatively well-mixed in the troposphere. This is consistent with the ubiquitous OH reductions in RCP 8.5 (which features very large methane increases) over oceanic regions, in qualitative agreement with Wild and Palmer (2008). Negative changes are even larger at high southern latitudes (Fig. 4c), possibly due to the additional effect of stratospheric ozone depletion: ozone recovery leads to less radiation reaching the troposphere, slower photolysis and, thus, less OH being produced. Models that do not include the effects of simulated ozone in photolysis calculations (HadGEM2, UM-CAM), do not have this high-latitude feature. In RCP 2.6, the changes over oceans are mixed, despite the fact that methane decreases to a similar extent in all models. Overall, there are a variety of competing factors, the balance of which determines the complex modelled OH distribution changes. Another prominent feature of Fig. 4 is the reduction along ship tracks in RCP 2.6, due to the reduction in shipping  $\text{NO}_x$  emissions in this scenario.

The agreement with Wild and Palmer (2008) is not as evident over land as it is over oceans. Their work, which was based on the SRES A2 scenario, found OH increases over all continental areas in 2100. In our case features are more mixed, with parts of continental areas actually experiencing negative OH changes, occurring due to the



NO<sub>x</sub> emission decreases in all RCP scenarios and models (see Table 2 and S1). On the contrary, SRES A2 used in Wild and Palmer (2008) featured dramatic fossil fuel NO<sub>x</sub> emission increases of 77 Tgyr<sup>-1</sup>, globally, between 2000 and 2100. Regional climate changes will also play a role (predominately reducing OH through higher water vapour), and the way in which NMVOC chemistry is included in each model will certainly have large effects in the boundary layer. For example, in CMAM and HadGEM2, which do not include NMVOCs, there is less structure in tropical OH changes (Fig. S2), implying that differences in NMVOCs and their chemistry is a major contributor to regional oxidant trends in such regions.

### 3.3 Regional changes

To further examine regional changes in oxidizing capacity, we also show changes in OH in various tropospheric subdomains (divided in a way similar to Lawrence et al., 2001). Figure 5 shows large OH decreases in RCP 8.5 (due to methane increases) in all regions except for the tropical and northern extratropical upper troposphere. Especially in the latter, increased stratosphere-troposphere exchange (due to a strong climate impact on the Brewer-Dobson circulation in this scenario; see Kawase et al., 2011 and Young et al., 2012) is likely a driver of positive changes, through increases of ozone available to generate OH. Also, increases in upper tropospheric humidity and lightning NO<sub>x</sub> emissions in a warmer climate could partly explain this feature, which may be masked in the southern extratropics due to stratospheric ozone recovery leading to sizeable OH decreases. This may also yield larger OH decreases in the southern extratropical lower troposphere (-28%) than in the Northern Hemisphere (-22%). In RCP 2.6, there is a mixture of regions with positive and negative OH changes. The lower northern extratropics show large OH decreases, presumably due to NO<sub>x</sub> emission decreases affecting this area heavily. This effect may become smaller with altitude (middle troposphere), due to the short lifetime of NO<sub>x</sub>.

The uncertainty in these projections is large, and in some cases it exceeds the signal of changes (for individual models, see Fig. S3). The upper troposphere tends to

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yield more uncertain results. In the lower troposphere, in RCP 8.5, the strong impact of changing methane abundances results in a fairly strong and certain OH response. In RCP 2.6, where no driver changes as dramatically, the signal-to-noise ratio is smaller. For the present-day, the multi-model mean shows the largest amount of methane chemical loss is in the lower tropical troposphere (Fig. 6), in excellent quantitative agreement with Lawrence et al. (2001). It is notable that in this important region, for the RCP 2.6 scenario, there is little model agreement in predicted OH changes in the 21st century, with positive and negative changes being almost equally likely. Uncertainty in modelled future NMVOC emissions could be the driver of this feature.

## 4 Evolution of potential drivers of OH abundances

Here we provide information on important variables influencing OH and methane lifetime. We consider the evolution of these variables in the 21st century, and especially on their 2100 levels. For more details on present-day emissions and other model metrics, please see Naik et al. (2012a) and Young et al. (2012).

### 4.1 Emissions

Global emissions of  $\text{NO}_x$  decline in all the simulations in the 21st century (Tables 2, S1, S2 and S3; Lamarque et al., 2012a). In RCP 2.6, RCP 4.5 and RCP 6.0, the evolution is similar, while in RCP 8.5 the trend is less pronounced. Lightning  $\text{NO}_x$  emissions, which can be a strong driver of OH changes (Labrador et al., 2004; Fiore et al., 2006) are quite uncertain in terms of magnitude, though they consistently show stronger trends in scenarios with more rapid warming (RCP 8.5), since these also feature greater convective and lightning activity (e.g. Del Genio et al., 2007). A clear exception is CMAM, in which lightning emissions generally decrease with time. This leads to a faster decline of total  $\text{NO}_x$  emissions in this model compared to the rest. CMAM uses a lightning parameterization based on the convective updraft mass flux from a fixed pressure level

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(modified version of the method presented in Allen and Pickering, 2002), and the particular trends are due to the changing distribution of the updraft mass flux at that level. This trend could have been different had the vertical level at which the convective updraft mass flux was taken evolved with time. Note that, despite the fact that this model is an outlier in terms of lightning  $\text{NO}_x$  in the current study, decreasing lightning  $\text{NO}_x$  emissions in a warmer climate were also reported in another study (Jacobson and Streets, 2009).

CO emissions also drastically drop between 2000 and 2100, with RCP 2.6 and RCP 4.5 showing the most rapid decreases (Lamarque et al., 2012a). The good agreement between the trends in different models is due to the fact that all models used identical anthropogenic and biomass burning emissions, which dominate the totals. Large differences between models within timeslices arise from the fact that some of the models (CMAM, HadGEM2) include proxy emissions (as CO) for NMVOCs. Also, the GISS model does not include oceanic CO sources, which likely explains why it is the model with the lowest total CO emissions. NMVOC emissions have a large spread, but several models show small trends due to static isoprene emissions in all timeslices. This is the case because several of them use identical biogenic emissions for all timeslices. In models that include climate-sensitive isoprene emissions (EMAC, GISS-E2-R, STOC-HadAM3), there are detectable positive trends, especially in RCP 8.5, which features the largest warming. HadGEM2 NMVOC emissions decrease, but this is only associated with less anthropogenic and biomass burning sources of NMVOCs in 2100, as this model does not include natural hydrocarbon emissions (only proxy CO).

### 4.2 Methane concentrations

As mentioned above, methane concentrations are imposed in the models and there are no sources included, with the exception of the LMDzORINCA (past/future) and the GISS-E2-R model (future). There are clear differences between the resulting methane burdens in the different timeslices. In RCP 2.6, methane decreases steadily throughout the century, in RCP 4.5 it remains steady until 2050 and then decreases, in RCP 6.0 it

increases until 2050 and then decreases, and in RCP 8.5 it rapidly increases throughout the century and is double in 2100 compared to 2000. Most models agree fairly well in the methane burden, with the exception of the GISS-E2-R model which, especially in RCP 8.5, shows a faster methane increase and significantly higher burden by the end of the century. This is due to the fact that in future GISS-E2-R simulations, changes in methane affect OH concentration, and thus feedback to methane's own lifetime (see Fig. 2), which means that methane increases amplify themselves during the 21st century. For more discussion on GISS-E2-R methane behavior, see Shindell et al. (2012b).

### 4.3 Meteorological factors

The most important meteorological factor affecting OH and methane lifetime is tropospheric humidity (Spivakovsky et al., 2000). Higher water vapour concentrations in the troposphere, mean that more OH is produced through reaction with singlet oxygen atoms ( $O^1D$ ). Temperature can be directly and indirectly linked to OH and methane lifetime by (a) affecting the  $CH_4 + OH$  reaction rate and the absorption cross section of ozone (which is important for photolysis to produce  $O^1D$ ), and (b) through its positive effect on tropospheric humidity. There are several other direct and indirect ways in which these two factors can impact OH and methane lifetime (through effects on other chemicals, aerosols, and feedbacks into the circulation), but they are generally expected to be smaller than those described above.

Clouds, which impact photolysis, and thus affect OH levels, should have relatively small effects on a global scale (e.g. Voulgarakis et al., 2009b), and more significant effects on regional scales. However, since we do not currently have cloud data from a sufficient number of ACCMIP models, we intend to examine this driver of OH variability further in a future study.

Apart from CICERO-OsloCTM2, which used fixed meteorological fields for every simulation, global mean temperature changes are positive in all scenarios (Tables 2, S1, S2 and S3), due to the increases in greenhouse gas concentrations. The smallest temperature changes (+1.3K on average) are found in RCP 2.6 and the largest

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ones (+4.6K on average) in RCP 8.5. There is a sizeable spread in the modelled temperature changes projected for 2100 (e.g.  $\pm 90\%$  spread around the mean change for RCP 8.5). This is particularly important, as temperature can be rather effective in driving OH and methane lifetime changes (Wild, 2007), despite the fact that its relative changes across timeslices are smaller compared to other drivers (e.g. emissions). The relationship between the  $\text{CH}_4 + \text{OH}$  reaction rate constant ( $k$ ) and temperature is non-linear (see Table A1), which implies that small changes in temperature can drive relatively large changes in methane loss.

Regarding humidity, the main features of change are similar to those of temperature, but with larger relative differences between timeslices. The models with the highest global mean temperature also have the highest global mean humidity, and the inverse (see Naik et al., 2012a). For more details on ACCMIP simulated climate, see Lamarque et al. (2012a).

#### 4.4 Ozone and ozone photolysis

Tropospheric ozone can affect OH and methane lifetime directly due to the fact that its photolysis provides the  $\text{O}^1\text{D}$  atoms that react with water vapour to produce OH. Thus, increases in ozone precursors can increase the OH levels in the troposphere. Stratospheric ozone affects tropospheric OH indirectly. First of all, changes of the amounts of stratospheric ozone entering the troposphere will affect the levels of tropospheric ozone available for OH production. Perhaps more importantly, stratospheric ozone changes affect shortwave radiation reaching the troposphere to drive photolysis.

Tropospheric ozone changes in the ACCMIP simulations are shown in Tables 2, S1, S2 and S3, and documented thoroughly by Young et al. (2012). Briefly, in RCP 2.6, tropospheric ozone burden declines throughout the 21st century, due to less precursor emissions and decreasing methane concentrations. RCP 4.5 and RCP 6.0 have minor differences between 2000 and 2100, while RCP 8.5 features large increases, due to the impact of the dramatic methane increases and enhanced stratospheric influx (see also Kawase et al., 2011).

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All the scenarios show a recovery of stratospheric ozone abundances in the future, as they all take into account the measures for continued controlling emissions of ozone depleting substances. This recovery is faster in RCP 8.5, due to the effects of CO<sub>2</sub> in cooling the stratosphere leading to faster ozone recovery (e.g. Eyring et al., 2010). Of the models that simulate stratospheric ozone, those with the fastest ozone recovery are GISS-E2-R, HadGEM2 and MOCAGE. LMDzORINCA shows no stratospheric ozone changes, due to the fact that an offline ozone climatology was used (Li and Shine, 1995).

Data for photolysis rates of ozone to yield O<sup>1</sup>D (JO<sup>1</sup>D) were only provided by a fraction of the models. In most cases there is a detectable decrease in J(O<sup>1</sup>D), and it relates to the increase of overhead ozone. However, only one model (GISS-E2-R) shows global J(O<sup>1</sup>D) decreases that are sizeable (6–15 %) in all RCPs. The GISS-E2-R results on stratospheric ozone and J(O<sup>1</sup>D) may be an overestimate, as this model has been found to have an ozone hole that extends slightly too far equatorward in September-October and persists about one month too long in the polar region (Shindell et al., 2012b). Note though, that the other two models in which we found more rapid stratospheric ozone recovery did not include ozone changes in photolysis calculations (HadGEM2) or did not provide J(O<sup>1</sup>D) data (MOCAGE).

## 5 Discussion on the drivers of OH and methane lifetime changes

### 5.1 Emissions

Generally, increases in NO<sub>x</sub> emissions have been associated with more OH generation, for two reasons: (a) NO<sub>x</sub> generally leads to ozone production (except under high NO<sub>x</sub> conditions typically not represented in global models), which is the main primary source of OH, and (b) NO<sub>x</sub>-rich environments favour more efficient secondary OH production through HO<sub>x</sub> recycling processes (e.g. conversion of HO<sub>2</sub> to OH) and, thus, increase OH abundances in the troposphere (e.g. Lelieveld et al., 2002). However, despite the

fact that global NO<sub>x</sub> emissions decrease substantially in all scenarios and models between 2000 and 2100 in all scenarios, the trends in global mean OH and methane lifetime have diverse signs in different models in RCP 2.6, RCP 4.5 and RCP 6.0 (Fig. 2). Furthermore, these trends are rather small, supporting the idea that global OH may be a relatively stable quantity, despite the large fluctuations on regional scales (Lelieveld et al., 2002). RCP 8.5 is the only scenario in which NO<sub>x</sub> emissions and global OH are related in terms of sign of change (both decreasing). However, we will demonstrate in Sect. 5.2 that NO<sub>x</sub> changes are not the main driver of the global OH trends.

Emissions of CO, which consumes OH, also generally drop during the 21st century. They most likely do not play a central role in driving OH and methane lifetime changes, since (a) the latter show fairly diverse trends in RCP 2.6, RCP 4.5 and RCP 6.0, despite the large CO emissions decreases, and (b) in RCP 8.5, global mean OH decreases, which could have been explained by increasing CO emissions, while CO emissions decrease in this scenario. Emissions of NMVOCs change significantly only in GISS-E2-R, EMAC and STOC-HadAM3 (and most prominently in RCP 8.5), since these are the only models that include climate-sensitive isoprene emission. However, these relatively small increases cannot be the main driver of the sizeable OH and methane lifetime changes, which are found in most models in RCP 8.5 (see Fig. 2).

Methane burden changes (Fig. 7) do not appear to be the main driver of the evolution of OH and methane lifetime for RCP 2.6. In the latter part of the 21st century the methane burden slowly decreases, which would drive less OH consumption; however, OH and methane lifetime remain fairly unaffected. In RCP 8.5, it is likely that methane changes are a major driver of OH and methane lifetime changes, something that is examined in more detail in Sect. 5.2.

## 5.2 Sensitivity experiments

We have performed a variety of sensitivity experiments based on RCP 8.5, and a few based on RCP 6.0, in order to understand the methane lifetime trends in these simulations. RCP 8.5 has been selected as the focus, as it is a scenario with a somewhat

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better agreement between models in terms of the sign of the changes, with an increase in methane lifetime in all models except HadGEM2 and UM-CAM.

Some of the sensitivity simulations were specifically requested by ACCMIP, and were performed by more than one model. This includes (a) a simulation with ozone precursor emissions set to 2000 values, but with climate set to 2100 RCP 8.5 conditions (CI2100); (b) a simulation with present-day conditions but methane concentrations increased by 100 ppb, with this perturbation affecting the modelled chemistry only (CH42000 + 100); and (c) a simulation with 2100 RCP 8.5 conditions, but with methane concentrations perturbed by 100 ppb. In addition, we performed some extra simulations with the GISS-E2-R model, in order to examine some other potential driving factors: (d) ODS2100, in which we used present-day conditions, but set ozone depleting substance (ODS, namely chlorofluorocarbons (CFCs)) and nitrous oxide (N<sub>2</sub>O) to 2100 levels following the projections in RCP 8.5, (e) CH42100a, in which we used present-day conditions but methane concentrations corresponding to 2100 RCP 8.5 levels, and (f) CH42100b, in which we used present-day conditions but methane concentrations corresponding to 2100 RCP 6.0 levels.

By comparing results for 2000 and CI2100 (Fig. 8 and Table 3), we find that climate changes lead to methane lifetime decreases in the future, in agreement with previous studies (e.g. Stevenson et al., 2006). As explained earlier, temperature and humidity increases both drive increases in OH in the atmosphere and faster CH<sub>4</sub> + OH reaction. The faster oxidation leads to a shorter lifetime and this is augmented by the OH increase. Furthermore, drastic increases in STE in a warmer climate (Kawase et al., 2011; Young et al., 2012) lead to more ozone and, thus, more OH in the troposphere. However, the 2100 simulation shows increases in methane lifetime during the 21st century, as also discussed earlier for RCP 8.5. Thus, climate alone would have opposite effects to those found in our future simulations, in which the climate effects have probably only contributed to offsetting some of the positive changes (by ~ 1.6 yr, in 2100 for RCP 8.5). By comparing 2000, 2100 and ODS2100, it is evident that ODSs have a sizeable effect on methane lifetime. However, this is not enough to explain the changes

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between 2000 and 2100. It is methane abundance itself that actually drives the largest part of the 2000–2100 changes in RCP 8.5: in the CH42100a simulation, methane lifetime reaches and even exceeds the levels of 2100. The consumption of OH radicals by increasing methane abundances leads to a drastic decrease of methane loss rates, and thus a prolonged lifetime. The further increase above the 2100 levels would have most likely been offset had there been climate changes included in the simulation.

However, for RCP 6.0 (red bars in Fig. 8), we find that methane burden changes cannot explain the 2000–2100 increases in methane lifetime. Despite the fact that RCP 6.0 methane concentration increases are substantial in the GISS model (see Fig. 7), which is used for the sensitivity experiments, the results from CH42100b are almost identical to the 2000 simulation. Thus, when methane changes are not as dramatic as in RCP 8.5, the influence of other factors becomes more prominent. In this case, a combination of NO<sub>x</sub> emission decline and stratospheric ozone recovery leading to lower J(O<sub>1</sub>D) are the most likely drivers. Particularly J(O<sup>1</sup>D) shows a strong correlation with both the stratospheric ozone column and with tropospheric OH throughout the 21st century (Fig. 9).

### 5.3 Climate penalty and OH feedback factors

The CI2100 simulation, as well as a similar one (CI2030) with climatic conditions set to 2030 (CI2030), were performed by several ACCMIP models. Such simulations are useful in order to determine the “climate penalty factor” (defined in earlier studies as the relationship between ozone and temperature; e.g. Wu et al., 2009) for methane lifetime, i.e. the lifetime perturbation by a unit change of global temperature. From Table 3 it can be seen that we get a multi-model mean value of  $-0.31 \pm 0.14 \text{ yrK}^{-1}$  from the CI2030 simulation and  $-0.34 \pm 0.12 \text{ yrK}^{-1}$  from CI2100. The feedback factor is negative since, as discussed earlier, a warmer climate leads to more OH and an increased CH<sub>4</sub> + OH reaction rate (note that there are several other processes that are modulated by or associated with or temperature change, but those discussed here are known to be the most dominant for OH and methane lifetime). For most models, the estimate from the

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different runs is similar, though in the GISS-E2-R model the differences are more substantial. The models with the strongest methane lifetime response and the strongest response per unit temperature change are HadGEM2 and UM-CAM. This may be explained by the fact that those two models are the only ones that do not include the effect of modelled overhead ozone column on photolysis, which would have driven an offsetting of the negative climate effect on methane lifetime, since stratospheric ozone is expected to increase in a warmer climate (Eyring et al., 2010).

In addition to the above sensitivities, two models also performed runs in which methane concentrations were perturbed by a small amount (100 ppb<sub>v</sub>), in order to detect the sensitivity of oxidants and methane lifetime to changing methane abundances. 2000CH4plus100 is a simulation identical to baseline 2000, but with methane perturbed by +100 ppb<sub>v</sub>, while 2100CH4plus100 is the equivalent for 2100 RCP 8.5 conditions. Table 4 shows the “feedback factor ( $F$ )”. This is defined as the ratio of the atmospheric response time to the global atmospheric lifetime,

$$F = 1/(1 - s) \quad (1)$$

where

$$s = (\delta \ln(\tau))/(\delta \ln[\text{CH}_4]) \quad (2)$$

using values for the methane lifetime ( $\tau$ ) and concentration [ $\text{CH}_4$ ] determined from the simulations (as per Fiore et al., 2009; Prather et al., 2001). In Table 4, we also provide  $(\delta \ln(\text{OH}))/(\delta \ln[\text{CH}_4])$  which is often used in a similar context.

The  $F$  values that we get from the four simulations range from 1.23 to 1.69. The values obtained from the 2000CH4plus100 simulation are closer to the estimates of Fiore et al. (2009), which were also based on perturbed present-day conditions. The differences between the present-day and the future perturbation simulations in our study are larger than the differences between the two models’ estimates for the same perturbation. An atmosphere with very high abundances of methane and very low abundances of  $\text{NO}_x$ , such as in the RCP 8.5 scenario, would feature less OH recycling (Lelieveld et al., 2002), and so a stronger effect of methane on its own lifetime.

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## 6 Diversity in present-day and future model simulations

As mentioned earlier, one of the first obvious conclusions of this multi-model intercomparison is the diversity in present-day OH and methane lifetime. Methane chemical lifetime in our study in year 2000 is equal to  $9.8 \pm 1.6$  yr (see Table 1), with a spread of almost 7 yr, an almost identical value to that obtained from the ACCENT multi-model study, and with the same level of diversity ( $9.7 \pm 1.7$  yr; Shindell et al., 2006b). More recently, Fiore et al. (2009) reported a somewhat higher mean lifetime, but with a similar model spread ( $10.2 \pm 1.7$  yr). Note that in the IPCC TAR, the average tropospheric methane lifetime that was reported was 9.6 yr, though it was obtained from a smaller set of models (Prather et al., 2001).

The models use a variety of inputs and include many interactions that are still fairly uncertain. This includes both chemical and climate variables. Here, we examine the degree to which variation across models in present-day tropospheric OH and methane lifetime could be explained by the variation in emissions, tropospheric CO and ozone burden, atmospheric methane burden, stratospheric ozone column, global mean temperature, global mean specific humidity, and the global mean  $J(O^1D)$ . Analysis was performed over global mean values for the 2000 timeslice. Linear regression coefficients were estimated using iteratively re-weighted least squares (IRLS) regression, which is more robust than ordinary least squares (OLS) against outliers and therefore well suited to the small sample size.

Table 5 shows the regression slopes,  $p$ -values of slopes and coefficients of determination ( $R^2$ ) from the regression analysis. Present-day tropospheric OH spread in the models shows some association with NMVOC emissions and the  $J(O^1D)$  photolysis rate. The association with  $J(O^1D)$  ( $p = 0.03$ ) is more significant than with NMVOC emissions ( $p = 0.07$ ), though it is based on results from fewer models (only 8 models provided photolysis data). There were no apparent relationships for the remaining variables.

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The positive association of global OH levels with  $J(\text{O}^1\text{D})$  is shown in Fig. 10, with the UM-CAM and GISS-E2-R falling outside of the main cluster of points. UM-CAM uses offline photolysis rates calculated in the Cambridge 2-D model, and in the past it has been shown with another model (*p*-TOMCAT) that when moving from this photolysis code to a state-of-the-art one,  $J(\text{O}^1\text{D})$  and OH levels increase significantly (Voulgarakis et al., 2009a). The outlying GISS-E2-R case can in part be explained by the fact that it has the highest CO burden among all the models, which means that the photolysis effect is masked by the consumption of OH radicals by CO.

NMVOC emissions, which are more uncertain than  $\text{NO}_x$  and CO emissions, appear to have a positive association with OH (Fig. 10), meaning that on a global scale, their role in OH recycling is more important than their role in OH consumption (which can be large regionally). This contrasts the findings of e.g. Poisson et al. (2000) and Wang et al. (1998), who generally found that NMVOCs contribute to lower OH in the models. The models with the lowest NMVOC emissions are CMAM (no emissions) and HadGEM2 (no vegetation emissions). HadGEM2 has the lowest OH in ACCMIP, while CMAM is closer to the average, probably because the extra CO amount that it includes as a proxy for NMVOC oxidation is rather low ( $250 \text{ Tgyr}^{-1}$ ), and certainly lower than that in HadGEM2 ( $475 \text{ Tgyr}^{-1}$ ). UM-CAM does not have exceptionally low or high NMVOC emissions, but the fact that its photolysis is too slow makes it an outlier in terms of OH. The model with the highest abundance of OH is MOCAGE, which is likely explained by the fact that its NMVOC emissions are the highest of all models.

For methane lifetime (Table 5), there is some association with NMVOC emissions, but much weaker than for OH. The association with  $J(\text{O}^1\text{D})$  is even weaker. The reason is probably that extratropical emissions and, especially,  $J(\text{O}^1\text{D})$  (which are included in the average only with an area/volume-weighting) are less relevant for global methane chemical lifetime than for global OH, due to the fact that the bulk of methane oxidation occurs in the tropics. The association between methane lifetime and tropical emissions or  $J(\text{O}^1\text{D})$  is expected to be stronger. On the other hand, methane lifetime shows a fairly strong relationship ( $p = 0.04$ ) with CO burden, which was seen more weakly for OH.

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However, the reason why CO burden is a driver of present-day model uncertainty in ACCMIP is not that the CO sources themselves were so different between the models (there is no relationship between CO emissions and methane lifetime in Table 5), but that models have a wide range of NMVOC species emitted, and treated in a variety of ways, which leads to diversity in the amount of secondary CO produced in the atmosphere via NMVOC oxidation.

To examine what drives the inter-model differences in OH and methane lifetime in the future, we performed a similar analysis, using the difference between 2100 and 2000 values of the variables as quantities of interest. For 2100, we used data from the RCP 8.5 scenario, due to the fact that it is expected to have the strongest signals. From our analysis, the strongest relationship is with changes in CO burden, temperature and humidity, the latter two being factors that strongly depend on each other. Especially for methane lifetime and temperature/humidity the associations are very strong ( $p < 0.01$ ; see Table 5 and Fig. 11). This implies, that the differences among the models in projecting 21st century climate changes are the key driver of the differences in trends in oxidizing capacity. It is notable that the slope of the relationship between methane lifetime and temperature change ( $-0.41$ ) is not too different from the value of the climate penalty factor presented in Table 3 ( $0.34 \pm 0.12 \text{ yrK}^{-1}$ ).

CICERO-OsloCTM2, which did not take any climate changes into account, shows the largest change in methane lifetime, and CESM-CAM-superfast, which has the 2nd highest methane lifetime change, also has the second smallest temperature/humidity changes. The rest of the models also follow this relationship, but two of them (HadGEM2 and UM-CAM) are outliers, since they are the only ones with a negative methane lifetime change, though their temperature and humidity responses are not exceptional. The unique behavior of these two models was discussed in Sect. 5.3, where we found that their methane lifetime response per unit temperature change is particularly strong, when compared to the other models. The underlying reasons for this behavior may be the lack of influence of stratospheric ozone changes on photolysis in these models, as explained in Sect. 5.3.

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We also considered multiple regression analysis to identify possible interactions and multi-collinearity between predictor variables. This would possibly identify any relationships between the changes in methane lifetime between 2100 and 2000 and humidity independent of the temperature change. We found, however, that there was too little data to include any additional terms in the linear model. We note also that OLS regression estimates were weaker due to the presence of outliers among the small number of samples, but still suggestive of the same relationships. In the case of the IRLS estimates, regression coefficients that were statistically significant remained so regardless of the outlier weighting function used.

The fact that emissions, especially of  $\text{NO}_x$  and CO, and methane abundances, do not appear to be as important as NMVOC emissions and climate in driving inter-model differences in OH and methane lifetime, does not necessarily imply that emissions of such species are actually well-constrained. Rather, it means that, in terms of emissions, we performed well-constrained experiments, in order to understand what atmospheric factors can drive chemical change and diversity. This approach has been valuable, but it also has limitations, due to the fact that real uncertainty in anthropogenic and natural emissions is not accounted for. Furthermore, all the RCP scenarios that are available assume that global  $\text{NO}_x$  and CO emissions from anthropogenic sources will rapidly decrease in the 21st century, which is an assumption that restricts us from examining the evolution of tropospheric composition under a less optimistic scenario for short-lived pollutants.

## 7 Conclusions and future work

We have analysed and discussed the evolution of OH and methane lifetime between present-day and projected 2100 conditions for different RCP scenarios, as revealed by the models participating in ACCMIP. For the present-day (2000), we calculate a methane lifetime of  $9.6 \pm 1.8$  yr. We find that there is a sizeable inter-model spread in both OH and methane lifetime, which has remained almost unchanged in magnitude

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5 compared to previous multi-model studies (Shindell et al., 2006b; Fiore et al., 2009). Compared to their mean values, OH levels range by 62 % and methane lifetime ranges by 69 % across models in 2000. Based on a regression analysis, we suggest that part of this present-day variability could be explained by model differences in NMVOC emissions and the treatment of photolysis. Models with high emissions of NMVOCs and high global mean photolysis rates (which are both fairly uncertain variables) tend to have higher global mean OH levels.

10 For the future evolution of OH and methane lifetime, mixed trends are found in the different models for each of the RCPs. In particular, diagnosing coherent changes for different regions is very challenging, due to their idiosyncrasies, which are not necessarily taken into account in all models. On a global scale, the scenario with the largest changes in OH and methane lifetime is RCP 8.5. The overwhelming effect of the large methane burden increases in this scenario (doubles in 2100 compared to 2000), with a smaller contribution from the effects of stratospheric ozone recovery (which leads to slower photolysis and less OH in the troposphere), drive methane lifetime increases in most of the models. In the other RCP scenarios, where no such large perturbation is applied, the interplay between different factors leads to diverse but small changes, suggesting that OH and methane lifetime may remain fairly stable in the future. Even though RCP 8.5 shows the most coherent changes in terms of sign, the amount of change relative to 2000 is quite different among the models. We suggest that these differences mostly arise from the diversity in modelled climate changes (temperature, humidity).

25 In order to elucidate the role of individual driving factors further, future experiments should focus on sensitivity simulations, changing one factor at a time in a manner similar to Wild (2007) (but focusing on OH), and performed by a range of models. Additionally, the chemical schemes need to be assessed in more detail, since their rate coefficients and reactions remains an unknown source of uncertainty. In particular, the representation of NMVOCs and their reactions under low-NO<sub>x</sub> conditions are highly uncertain, which can lead to variations in future OH and methane lifetime projections

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(e.g. Archibald et al., 2011). Clouds, a factor on which we did not focus in this study, could be an important driver of regional changes in OH (Voulgarakis et al., 2009b), but these effects have not been examined systematically in a multi-model framework. Gas-aerosol interactions in future atmospheres could also be studied more thoroughly, using the knowledge on oxidants that is obtained through our analysis. The fact that global climate models are now being developed to include a range of processes which were not available until recently provides the possibility to understand atmospheric composition from a broader perspective, in which atmospheric chemistry is an integral part of the Earth system.

**Supplementary material related to this article is available online at:**  
**[http://www.atmos-chem-phys-discuss.net/12/22945/2012/  
acpd-12-22945-2012-supplement.pdf](http://www.atmos-chem-phys-discuss.net/12/22945/2012/acpd-12-22945-2012-supplement.pdf)**

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Models	Mean OH ( $10^5 \text{ mol cm}^{-3}$ )	$\tau_{\text{CH}_4}$ (chemical) (yr)	$\tau_{\text{CH}_4}$ (total) (yr) <sup>a</sup>
CESM-CAM-superfast	12.9	8.4	7.5
CICERO-OsloCTM2	10.4	10.0	8.7
CMAM	10.8	9.5	8.3
EMAC	11.8	9.2	8.1
GEOSCCM	11.4	9.7	8.5
GFDL-AM3	11.7	9.4	8.3
GISS-E2-R	10.6	10.6	9.2
HadGEM2	8.1	11.4	9.8
LMDzORINCA	10.3	10.4	9.1
MIROC-CHEM	12.5	8.8	7.8
MOCAGE	13.4	7.1	6.4
NCAR-CAM3.5	12.1	9.3	8.5
STOC-HadAM3	12.2	9.0	8.0
UM-CAM	6.5	13.9	11.6
Mean $\pm$ stand. dev.	$11.1 \pm 1.8$	$9.8 \pm 1.6$	$8.6 \pm 1.2$
Mean with trop1 <sup>b</sup>	$11.1 \pm 1.7$	$9.7 \pm 1.6$	–
Mean with trop2 <sup>c</sup>	$11.0 \pm 1.8$	$9.8 \pm 1.6$	–

<sup>a</sup> For the total lifetime, we add to the tropospheric chemical loss a  $30 \text{ Tgyr}^{-1}$  methane sink in soils and a  $40 \text{ Tgyr}^{-1}$  sink to the stratosphere (Stevenson et al., 2006).

<sup>b</sup> The trop1 tropopause follows the  $\text{O}_3=150 \text{ ppb}_v$  surface (e.g. Stevenson et al., 2006).

<sup>c</sup> The trop2 tropopause follows the surface defined by  $300-215 \times \cos(\text{lat})^2 \text{ hPa}$  (e.g. Shindell et al., 2006b).

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**Table 2.** Percentage (%) changes in important model metrics, between 2100 2000 (RCP 8.5). Variables examined (from left to right) are: global tropospheric air mass-weighted mean OH concentration, global chemical methane lifetime, total NO<sub>x</sub> emissions (including lightning), total lightning NO<sub>x</sub> emissions, total CO emissions, total NMVOC emissions, global atmospheric methane burden, global tropospheric ozone burden, global mean stratospheric ozone column, global volume-weighted tropospheric mean J(O<sup>1</sup>D), and global tropospheric mean temperature and humidity. Same tables for the other scenarios are shown in Tables S1, S2 and S3.

Models (RCP 8.5)	OH	$\tau_{\text{CH}_4}$	NO <sub>x</sub> Emis.	LiNO <sub>x</sub> Emis.	CO Emis.	NMVOC Emis.	CH <sub>4</sub> Burd.	O <sub>3</sub> Burd.	Strat. O <sub>3</sub>	J(O <sup>1</sup> D)	T	Q
CESM-CAM-superfast	-17.4	+21.5	-33.0	+29.7	-30.1	0.0	+112.1	+25.1	+5.3	-	0.9	16.1
CICERO-OsloCTM2	-20.6	+26.9	-29.3	0.0	-27.3	-5.8	+108.4	+10.4	+0.3	-	-	-
CMAM	-15.5	+9.1	-27.2	-45.4	-25.7	-	+114.2	+13.9	+6.4	-4.3	+2.3	+45.3
EMAC	-12.0	+5.6	-19.9	+8.9	-31.5	+21.5	+115.3	+16.2	+6.4	-4.4	+2.2	+37.5
GEOSCCM	-	-	-	-	-	-	-	-	-	-	-	-
GFDL-AM3	-6.7	-1.4	-22.4	+38.2	-30.3	-1.9	+116.1	+27.8	+8.4	-7.2	+2.5	+45.0
GISS-E2-R	-18.6	+15.9	-20.0	+26.2	-35.1	+19.8	+152.7	+27.6	+15.1	-15.0	+1.6	+28.9
HadGEM2	+1.4	-7.11	-25.8	+74.1	-24.0	-22.5	+114.7	+29.0	+10.8	-	+1.9	+35.8
LMDzORINCA	-5.8	+0.9	-31.6	+43.3	-34.7	-4.3	+105.8	+9.6	+0.1	-	+1.9	-
MIROC-CHEM	-6.4	-1.4	-6.9	+38.0	-35.4	-3.4	+116.0	+10.7	+4.2	-0.8	+2.8	+52.2
MOCAGE	-20.1	+20.1	-22.9	+19.9	-32.3	-2.8	+113.4	+28.0	+23.6	-	+1.4	+22.3
NCAR-CAM3.5	-14.1	+13.7	-26.6	+35.2	-30.3	-2.6	+113.9	+14.6	+6.2	-3.6	+1.6	+26.8
STOC-HadAM3	-13.0	+6.7	-20.8	+23.2	-32.4	+25.2	+114.2	+12.1	+5.6	-4.1	+2.3	+38.1
UM-CAM	+2.4	+0.5	-17.2	+43.6	-32.0	-4.2	+112.1	+23.2	+7.4	+0.3	+2.3	+39.0
Mean ±	-11.3	+8.5	-22.7	+24.3	-30.9	+2.3	+116.1	+19.1	+7.7	-4.9	+2.0	+35.2
stand. dev.	±7.7	±10.4	±6.7	±28.7	±3.5	±14.9	±11.4	±7.7	±6.2	±4.7	±0.5	±10.8

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**Table 3.** Methane lifetime change in the sensitivity simulations, and lifetime change per unit change of global temperature (or “climate penalty factor”), for the different ACCMIP models. The multi-model mean and standard deviation are also shown.

	$\Delta\tau_{\text{CH}_4}$ (yr)	$\Delta\tau_{\text{CH}_4}$ (yr)	$\Delta\tau_{\text{OH}}/\Delta T$ (yrK <sup>-1</sup> )	$\Delta\tau_{\text{OH}}/\Delta T$ (yrK <sup>-1</sup> )
	CI2030 – 2000	CI2100 – 2000	CI2030 – 2000	CI2100 – 2000
CESM-CAM-superfast	-0.30	-0.72	-0.29	-0.32
CICERO-OsloCTM2	–	–	–	–
CMAM	–	–	–	–
EMAC	–	–	–	–
GEOSCCM	–	–	–	–
GFDL-AM3	-0.54	-1.82	-0.32	-0.29
GISS-E2-R	-0.08	-0.88	-0.12	-0.22
HadGEM2	–	-2.40	–	-0.50
LMDzORINCA	–	–	–	–
MIROC-CHEM	-0.59	-2.08	-0.36	-0.30
MOCAGE	-0.09	-0.86	-0.21	-0.25
NCAR-CAM3.5	-0.34	-1.48	-0.40	-0.40
STOC-HadAM3	-0.31	-1.21	-0.22	-0.21
UM-CAM	-0.81	-3.08	-0.57	-0.54
Mean ± stand. dev.	-0.38 ± 0.25	-1.61 ± 0.80	-0.31 ± 0.14	-0.34 ± 0.12

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**Table 4.** Feedback factor ( $F$ ) from two of the ACCMIP models and two different simulations (one for present-day conditions (2000) and one for future (2100)).

Experiment	$\Delta \ln(\text{OH})/\Delta \ln(\text{CH}_4)$	$F$
2000CH4plus100 GISS-E2-R	-0.17	1.23
2100CH4plus100 GISS-E2-R	-0.47	1.69
2000CH4plus100 UM-CAM	-0.23	1.35
2100CH4plus100 UM-CAM	-0.34	1.60

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**Table 5.** Regression statistics for present-day and 2100 minus present-day tropospheric OH and chemical methane lifetime, and different predictor variables. We show the slope (b1), slope  $p$ -value (b1  $p$ -val) and coefficient of determination ( $R^2$ ). Variables significant at the 90 % level appear in bold and variables significant at the 95 % level appear in italics.

	b1	b1 $p$ -val	$R^2$	b1	b1 $p$ -val	$R^2$
2000	OH			$\tau_{\text{CH}_4}$		
NO <sub>x</sub> Em.	1.2x10 <sup>-1</sup>	0.38	0.02	-1.1 × 10 <sup>-1</sup>	0.28	0.02
LiNO <sub>x</sub> Em.	2.8 × 10 <sup>-1</sup>	0.27	0.09	-9.2 × 10 <sup>-2</sup>	0.63	0.00
CO Em.	-4.4 × 10 <sup>-3</sup>	0.15	0.03	2.1 × 10 <sup>-3</sup>	0.40	-0.04
NMVOc Em.	<b>2.9 × 10<sup>-3</sup></b>	<b>0.06</b>	<b>0.23</b>	-1.9 × 10 <sup>-3</sup>	0.17	0.13
CH <sub>4</sub> Burden	-2.9 × 10 <sup>-3</sup>	0.60	-0.05	1.4 × 10 <sup>-3</sup>	0.73	-0.01
CO Burden	-1.6 × 10 <sup>-2</sup>	0.11	0.26	<i>1.4 × 10<sup>-2</sup></i>	<i>0.04</i>	<i>0.33</i>
O <sub>3</sub> Burden	-4.7 × 10 <sup>-3</sup>	0.81	-0.04	7.8 × 10 <sup>-3</sup>	0.59	-0.04
Strat. O <sub>3</sub>	-1.9 × 10 <sup>-2</sup>	0.47	-0.05	1.7 × 10 <sup>-2</sup>	0.45	-0.01
J(O <sup>1</sup> D)	<i>3.2 × 10<sup>+0</sup></i>	<i>0.03</i>	<i>0.38</i>	-1.3 × 10 <sup>+0</sup>	0.24	0.33
Temp.	-3.1 × 10 <sup>-1</sup>	0.57	-0.03	2.1 × 10 <sup>-1</sup>	0.62	0.00
Hum.	2.7 × 10 <sup>+0</sup>	0.41	0.08	-1.4 × 10 <sup>+0</sup>	0.58	0.04
2100–2000						
NO <sub>x</sub> Em.	1.1 × 10 <sup>-1</sup>	0.26	0.14	-1.8 × 10 <sup>-1</sup>	0.10	0.28
LiNO <sub>x</sub> Em.	2.6 × 10 <sup>-1</sup>	0.26	0.14	-3.2 × 10 <sup>-1</sup>	0.24	0.15
CO Em.	-1.7 × 10 <sup>-2</sup>	0.54	0.04	4.3 × 10 <sup>-2</sup>	0.15	0.18
NMVOc Em.	-2.3 × 10 <sup>-3</sup>	0.53	0.05	2.0 × 10 <sup>-3</sup>	0.65	0.02
CH <sub>4</sub> Burden	-2.7 × 10 <sup>-4</sup>	0.64	0.03	4.4 × 10 <sup>-4</sup>	0.52	0.04
CO Burden	<i>-1.6 × 10<sup>-2</sup></i>	<i>0.03</i>	<i>0.41</i>	<i>1.8 × 10<sup>-2</sup></i>	<i>0.04</i>	<i>0.38</i>
O <sub>3</sub> Burden	1.3 × 10 <sup>-3</sup>	0.91	0.00	-5.7 × 10 <sup>-3</sup>	0.69	0.02
Strat. O <sub>3</sub>	-1.8 × 10 <sup>-2</sup>	0.40	0.06	6.0 × 10 <sup>-3</sup>	0.82	0.00
J(O <sup>1</sup> D)	3.9 × 10 <sup>+0</sup>	0.20	0.76	-6.2 × 10 <sup>+0</sup>	0.11	0.77
Temp.	<b>2.7 × 10<sup>-1</sup></b>	<b>0.06</b>	<b>0.33</b>	<i>-4.1 × 10<sup>-1</sup></i>	<i>0.00</i>	<i>0.55</i>
Hum.	<b>1.7 × 10<sup>+0</sup></b>	<b>0.07</b>	<b>0.34</b>	<i>-2.7 × 10<sup>+0</sup></i>	<i>0.00</i>	<i>0.52</i>

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**Table A1.** Table of participating models and their specifications. For more information, see tables in Lamarque et al. (2012a).

Model	Scenarios with simulations	Lightning NO <sub>x</sub>	Stratospheric Ozone	Photolysis scheme	Methane	$k_{\text{CH}_4+\text{OH}}$ ( $10^{-5} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1}$ ) add 2000 values
CESM-CAM-superfast (Rotman et al., 2004; Lamarque et al., 2011)	RCP 8.5	Interactive, based on model's convection (Price et al., 1997)	Linearised O <sub>3</sub> chemistry (McLinden et al., 2000)	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Madronich and Flocke, 1998)	Prescribed atmospheric concentrations with spatial variation, different for each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
CICERO-OsloCTM2 (Skeie et al., 2011)	RCP 2.6, RCP 4.5, RCP 8.5	Interactive, based on model's convection (Price et al., 1997); scaled to $5 \text{ Tg N yr}^{-1}$	Offline climatological O <sub>3</sub> , except for the bottom 3 model strat. layers (see Skeie et al., 2011)	On-line using the Fast-J2 (Wild et al., 2000; Bian and Prather, 2002); accounts for modelled O <sub>3</sub> , clouds, surf. albedo and aerosols	Prescribed surface concentrations from IPCC TAR for present-day; CMIP5 future concentrations scaled to be consistent with present-day levels	$2.45 \times 10^{-12} e^{-1775/T}$
CMAM (deGrandpré et al., 2000; Plummer et al., 2012)	RCP 4.5, RCP 8.5	Interactive, based on convective updraft mass flux (modified from Allen and Pickering, 2002)	Full stratospheric chemistry	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Chang et al., 1997)	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
EMAC (Jöckel et al., 2006; Klinger et al., 2011)	RCP 4.5, RCP 8.5	Interactive (Grewe et al., 2001)	Full stratospheric chemistry	On-line, based on modelled clouds, climatological aerosol and strat. O <sub>3</sub> (Landgraf and Crutzen, 1998)	Prescribed surface concentrations using the AGAGE data for 2000 (Prinn et al., 2000). CMIP5 concentration data are used to rescale AGAGE data to other years/ scenarios	$1.85 \times 10^{-12} \cdot T^{2.82} e^{-987/T}$
GEOSCCM (Oman, et al., 2011)	RCP 6.0	Fixed emissions with a monthly climatology, based on Price et al. (1997); scaled to $5 \text{ Tg N yr}^{-1}$	Full stratospheric chemistry	Online (FastJX); accounts for clouds, strat. O <sub>3</sub> and albedo; uses offline aerosols from GOCART	Prescribed surface (two bottom levels) concentrations, with a prescribed latitudinal gradient, but the values are normalised so that the area-weighted mean matches the CMIP5 value for the timeslice	$2.80 \times 10^{-14} \cdot T^{0.667} e^{-1575/T}$
GFDL-AM3 (Donner et al., 2011; Naik et al., 2012b)	RCP 2.6, RCP 4.5, RCP 6.0, RCP 8.5	Interactive, based on model's convection (Price et al., 1997), scaled to $\sim 3\text{--}5 \text{ Tg N yr}^{-1}$	Full stratospheric chemistry	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Madronich and Flocke, 1998)	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$

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Table A1. Continued.

Model	Scenarios with simulations	Lightning NO <sub>x</sub>	Stratospheric Ozone	Photolysis scheme	Methane	$k_{\text{CH}_4+\text{OH}}$ ( $10^{-15} \text{ cm}^3 \text{ mol}^{-1} \text{ s}^{-1}$ ) add 2000 values
GISS-E2-R (Shindell et al., 2006a, 2012b)	RCP 2.6, RCP 4.5, RCP 6.0, RCP 8.5	Interactive, based on model's convection (modified from Price et al., 1997)	Full stratospheric chemistry	Online (Fast-J2 scheme); accounts for modelled clouds, strat. O <sub>3</sub> , aerosols, surf. albedo	Emissions (interactive wetlands, non-interactive other sources)	$2.45 \times 10^{-12} e^{-1775/T}$
HadGEM2 (Collins et al., 2011)	RCP 2.6, RCP 4.5, RCP 8.5	Interactive, based on model's convection (Price and Rind, 1993)	Offline stratospheric O <sub>3</sub> from CMIP5 dataset	Look-up table (Law and Pyle, 1993); no correction for modelled fields	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
LM DzORINCA (Szopa et al., 2012)	RCP 2.6, RCP 4.5, RCP 6.0, RCP 8.5	Interactive, based on model's convection (Price et al., 1997)	Offline stratospheric O <sub>3</sub> (climatology from Li and Shine, 1995)	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Madrnich and Flocke, 1998)	Emissions	$2.45 \times 10^{-12} e^{-1775/T}$
MIROC-CHEM (Sudo et al., 2002; Watanabe et al., 2011)	RCP 2.6, RCP 6.0, RCP 8.5	Interactive, based on model's convection (Price and Rind, 1992, 1994)	Full stratospheric chemistry	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> , surf. albedo and aerosols (Landgraf and Crutzen, 1998)	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
MOCAGE (Josse et al., 2004; Teyssière et al., 2007)	RCP 2.6, RCP 6.0, RCP 8.5	Interactive, based on Price and Rind (1992) and Ridley et al. (2005)	Full stratospheric chemistry	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Madrnich and Flocke, 1998)	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
NCAR-CAM3.5 (Lamarque et al., 2011, 2012b)	RCP 2.6, RCP 4.5, RCP 6.0, RCP 8.5	Interactive, based on model's convection (Price et al., 1997; Ridley et al., 2005), scaled to $\sim 3\text{--}5 \text{ Tg Nyr}^{-1}$	Full stratospheric chemistry	Look-up table with correction for modelled clouds, strat. O <sub>3</sub> and surf. albedo, not aerosols (Madrnich and Flocke, 1998)	Prescribed surface concentrations following CMIP5, different in each timeslice	$2.45 \times 10^{-12} e^{-1775/T}$
STOC-HadAM3 (Stevenson et al., 2004)	RCP 2.6, RCP 8.5	Interactive, based on model's convection (Price and Rind, 1992; Price et al., 1997)	Offline stratospheric O <sub>3</sub> from CMIP5 dataset	1-D, two-stream model (Hough, 1988). Uses climatological O <sub>3</sub> above tropopause and modelled O <sub>3</sub> below	Prescribed globally uniform concentrations, different for each timeslice following CMIP5 dataset	$2.45 \times 10^{-12} e^{-1775/T}$
UM-CAM (Zeng et al., 2008, 2010)	RCP 2.6, RCP 4.5, RCP 8.5	Interactive, based on model's convection (Price and Rind, 1992, 1994)	Offline stratospheric O <sub>3</sub> from CMIP5 dataset	Look-up table (Law and Pyle, 1993); no correction for modelled fields	Prescribed globally uniform atmospheric concentration with no spatial variation; different for each timeslice	$1.85 \times 10^{-12} e^{-1690/T}$

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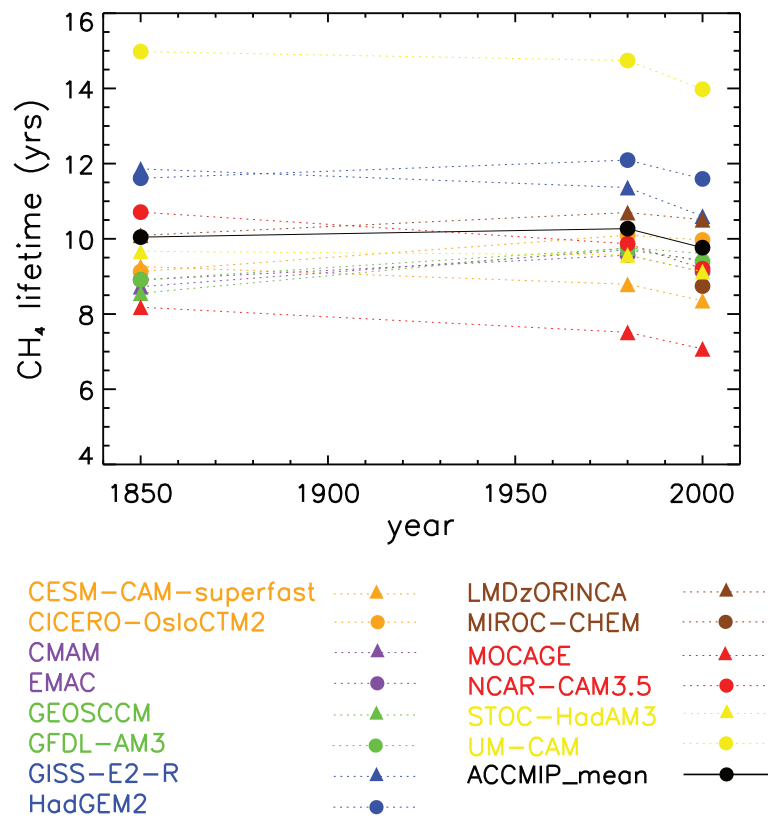
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**Fig. 1.** Evolution of global chemical methane lifetime in the ACCMIP models, for the historical period.

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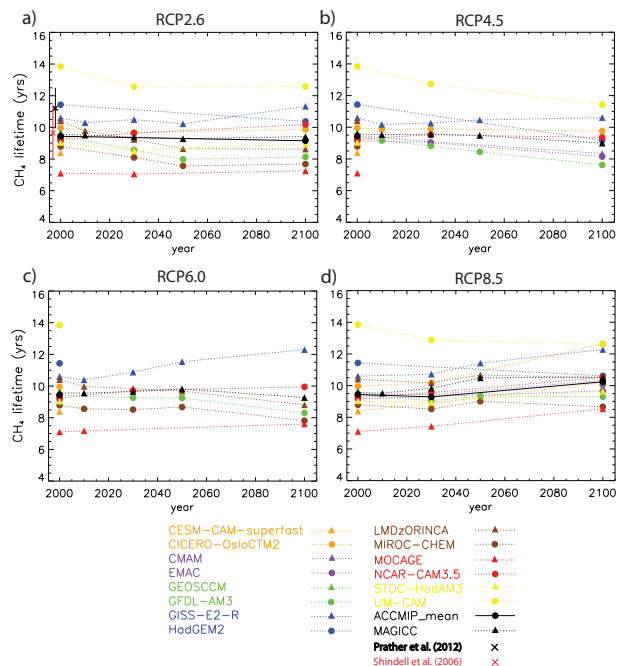
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**Fig. 2.** Evolution of global chemical methane lifetime in the ACCMIP models, for the four future RCP scenarios. Multi-model mean values (black dots connected with solid line) were only plotted for timeslices with data from at least 8 models. For comparison, the dotted black line with square points shows the lifetimes used in the MAGICC integrated assessment model. Also, in the upper left panel, the red cross for present-day shows the mean chemical lifetime from the ACCENT models (Shindell et al., 2006b), and the black cross is the observationally-based estimate made by Prather et al. (2012).

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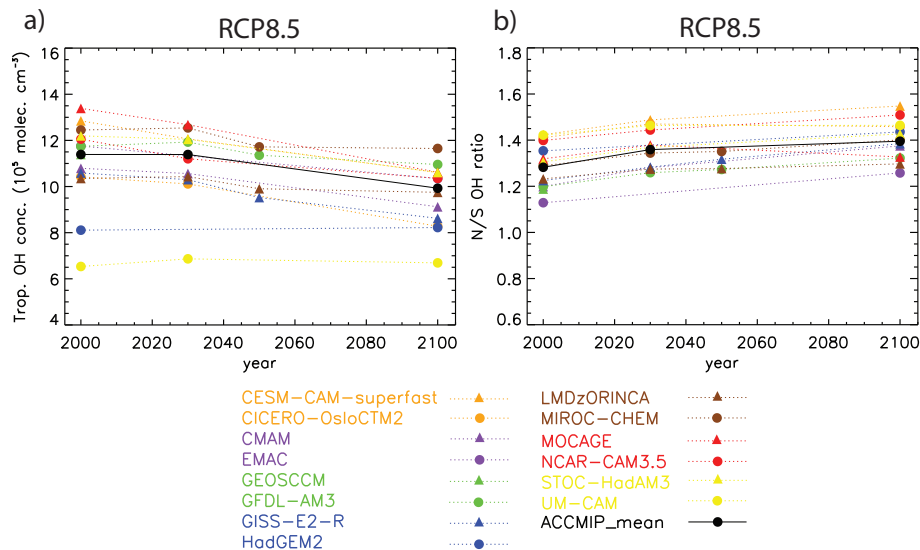
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**Fig. 3.** Evolution of air mass-weighted global tropospheric mean OH concentration **(a)** and Northern Hemisphere to Southern Hemisphere (N/S) OH ratio in the RCP 8.5 scenario.

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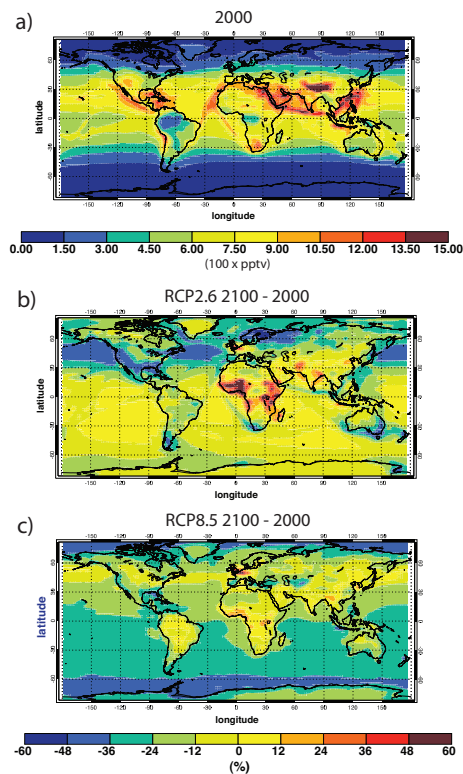
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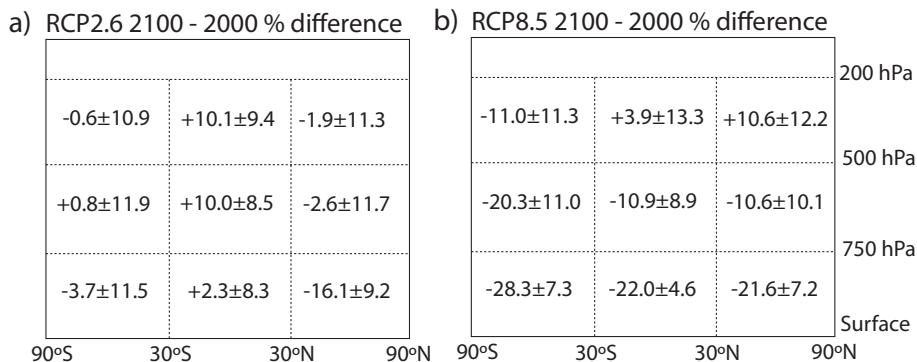


**Fig. 4.** Surface annual mean OH concentration for present-day (a), and its change between 2000 and 2100 in RCP 2.6 (b) and RCP 8.5 (c). The bottom model layer results have been used as representative for the surface.

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**Fig. 5.** Changes in regional mean OH concentration between 2100 and 2000 in various tropospheric subdomains. The range represents inter-model  $\pm 1\sigma$  spread of the change.

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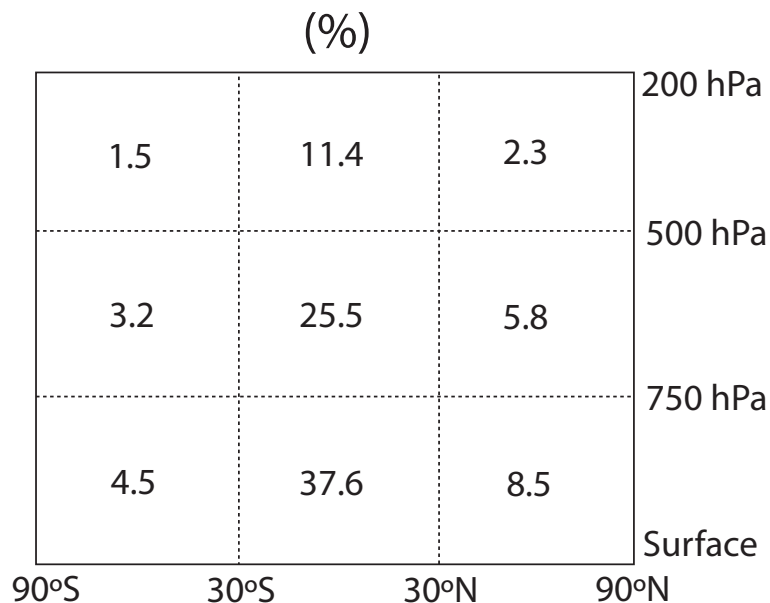
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**Fig. 6.** The percentage of global methane that is oxidised in various subdomains of the atmosphere in present-day (year 2000).

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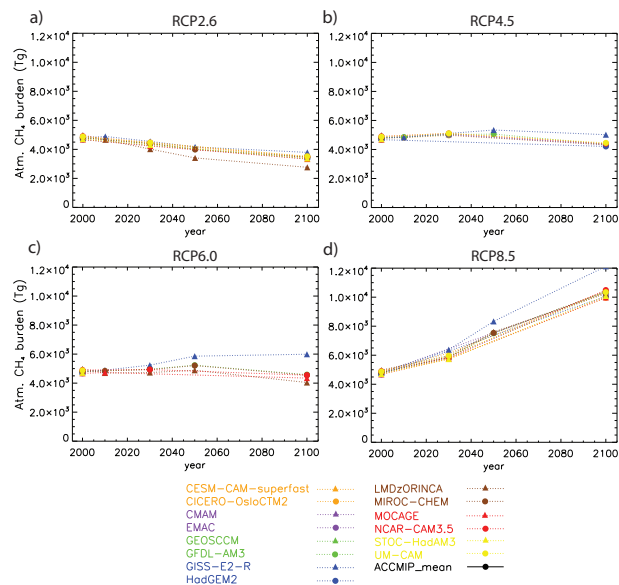
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**Fig. 7.** Evolution of global atmospheric methane burden in the ACCMIP models, for the four different future RCP scenarios. Multi-model mean values (black dots connected with solid line) were only plotted for timeslices with data from at least 8 models.

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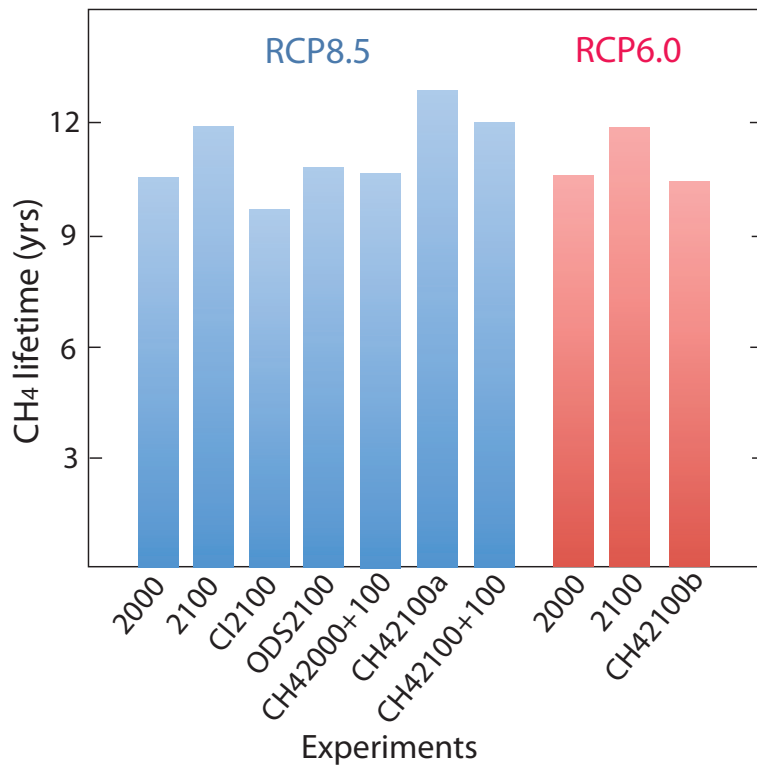
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**Fig. 8.** Tropospheric chemical methane lifetime in the sensitivity experiments performed with the GISS-E2-R model.

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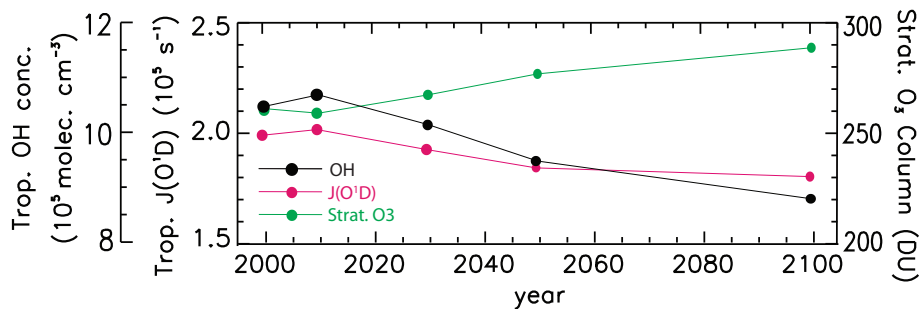
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**Fig. 9.** Global tropospheric air mass-weighted OH concentration, volume-weighted tropospheric mean J(O<sup>1</sup>D) and stratospheric mean ozone column (above 200 hPa) in the RCP 6.0 simulation from the GISS-E2-R model.

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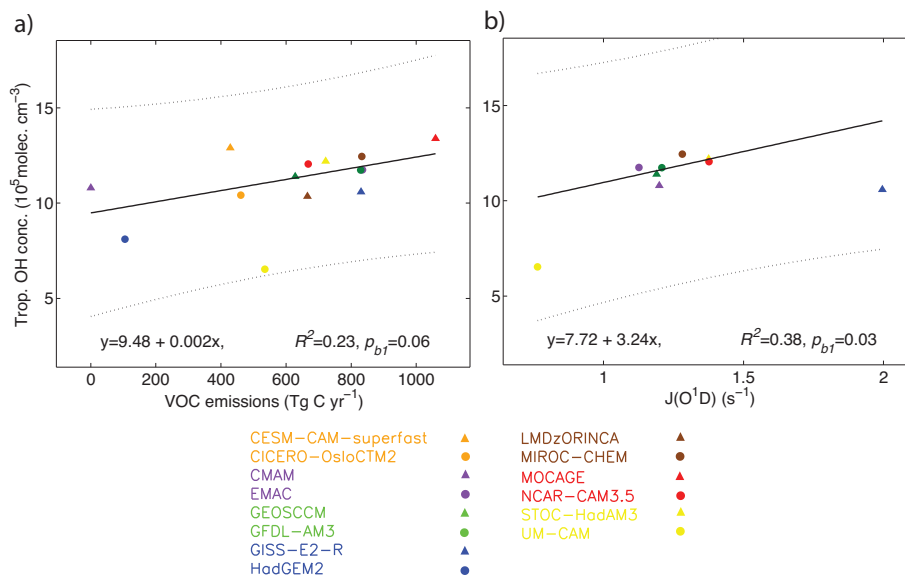
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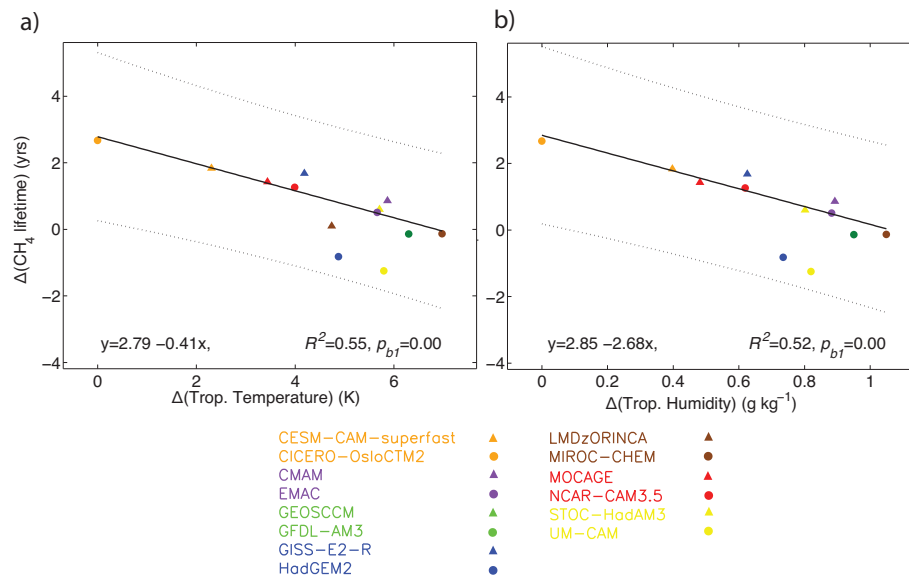


**Fig. 10.** Linear relationship between present-day (2000) global mean tropospheric OH and **(a)** global NMVOC emissions, and **(b)** global mean  $J(\text{O}^1\text{D})$ , across all models. Dashed lines show the prediction intervals at a 95% confidence level.

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**Fig. 11.** Linear relationship between 2000–2100 changes in global chemical methane lifetime and 2000–2100 changes in **(a)** global mean temperature, and **(b)** global mean humidity, across all models. Dashed lines show the prediction intervals at a 95 % confidence level.

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