Inter-model comparison of global hydroxyl radical (OH) distributions and their impact on atmospheric methane over the 2000-2016 period

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Abstract

The modeling study presented here aims to estimate how uncertainties in global hydroxyl radical (OH) distributions, variability, and trends may contribute to resolve discrepancies between simulated and

50   observed methane (CH_4) changes since 2000. A multi-model ensemble of 14 OH fields were analysed and were aggregated into 64 scenarios to force the offline atmospheric chemistry transport model LMDz with a standard CH_4 emission scenario over the period 2000-2016. The multi-model simulated global volume-weighted tropospheric mean OH concentration([OH]) averaged over 2000-2010 ranges between 8.7×10^5 and 12.8×10^5 molec cm^{-3}. The inter-model differences in tropospheric OH burden and vertical

55   distributions are mainly determined by the differences in the nitrogen oxide (NO) distributions, while the spatial discrepancies between OH fields are mostly due to differences in natural emissions and VOC chemistry. From 2000 to 2010, most simulated OH fields show an increase of 0.1-0.3×10^5 molec cm^{-3} in the tropospheric mean [OH], with year-to-year variations much smaller than during the historical period 1960-2000. Once ingested into the LMDz model, these OH changes translated into a 5 to 15 ppbv

60   reduction in CH_4 mixing ratio in 2010, which represent 7%-20% of the model simulated CH_4 increase due to surface emissions. Between 2010 and 2016, the ensemble of simulations showed that OH changes could lead to a CH_4 mixing ratio uncertainty of $> \pm 30$ ppbv. Over the full 2000-2016 time period, using a common state-of-the-art but non-optimized emission scenario, the impact of [OH] changes tested here can explain up to 54% of the gap between model simulations and observations. This result emphasizes

65   the importance of better representing OH abundance and variations in CH_4 forward simulations and emission optimizations performed by atmospheric inversions.

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1 Introduction

70 The hydroxyl radical (OH) is the main oxidizing agent in the troposphere (Levy, 1972). OH is produced

by the reaction of water vapor with excited oxygen atoms (O^{1D}), produced by ozone (O_3) photolysis (λ

<340nm). In the troposphere, OH is rapidly removed by reactions with carbon monoxide (CO), methane

(CH_4) and non-methane volatile organic compounds (NMVOCs) to generate hydroperoxyl radical (HO_2)

or organic peroxy radicals (RO_2), resulting in a short lifetime of a few seconds (Logan et al., 1981;

75 Lelieveld et al., 2004). HO_2 and RO_2 can further react with nitrogen oxide (NO) to regenerate OH (Crutzen,

1973; Zimmerman et al., 1987). At high latitudes, such a secondary production plays an important role,

because the OH primary production is limited by the supply of O^{1D} and water vapor (Spivakovsky et al.,

2000). The abundance of OH reflects the combined effects of atmospheric composition (tropospheric O_3,

and NO, CO, CH_4, and NMVOCs) and of meteorological factors such as humidity, UV radiation, and

80 temperature.

Due to its short lifetime, global [OH] is difficult to estimate from direct measurements. Current

understanding on global [OH] has been obtained either from inversion of 1-1-1trichloroethane (methyl

chloroform, MCF) (Prinn et al., 2005; Bousquet et al., 2005; Montzka et al., 2011; Rigby et al., 2017;

85 Turner et al., 2017), or using atmospheric chemistry models (Naik et al., 2013; Voulgarakis et al., 2013,

Lelieveld et al., 2016). The former approach relies on the fact that OH is the main sink of MCF and on

the hypotheses that emissions and concentrations of MCF are well known and well measured, respectively.

The latter approach relies on chemistry-transport modeling with chemistry schemes of varying complexity.

The global mass-weighted tropospheric mean [OH] in the 2000s calculated by atmospheric chemistry

90 models was found to be about 11.5×10^5 molec cm^{-3}, with an inter-model dispersion of ±15% (Naik et

al., 2013; Voulgarakis et al., 2013). Atmospheric chemistry models usually calculate higher [OH] over

the Northern hemisphere than the Southern hemisphere (N/S ratio>1) (Naik et al., 2013) whereas MCF

and [14]CO observations indicate a N/S ratio slightly smaller than 1 (Brenninkmeijer et al., 1992; Bousquet

et al., 2005; Patra et al., 2014).

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OH determines the lifetime of most pollutants and non-CO_2 greenhouse gases including CH_4, the second most important anthropogenic greenhouse gas after carbon dioxide (CO_2) (Ciais et al., 2013). About 90% of tropospheric CH_4 is removed by reacting with OH (Ehhalt et al., 1974; Kirschke et al., 2013; Saunois et al., 2016). The tropospheric CH_4 chemical lifetime against OH oxidation (global tropospheric CH_4

100   burden divided by annual CH_4 tropospheric loss by OH) calculated by the models that participated in the Atmospheric Chemistry and Climate Model Inter-comparison Project (ACCMIP) is 9.3 ± 1.6 years, and the CH_4 total lifetime including all sink processes is 8.3 ± 0.8 years (Naik et al., 2013; Voulgarakis et al., 2013), smaller than that of 9.1 ± 0.9 years lifetime constrained by observations (Prather et al., 2012).

105   The tropospheric CH_4 burden has more than doubled compared to the pre-industrial era due to anthropogenic activities and climates change, resulting in about 0.62 W m^{-2} additional radiative forcing (Etminan et al., 2016). The global mean CH_4 growth rate decreased to near zero in the early 2000s but resumed increasing at $\sim$5ppbv yr^{-1} since 2006 and reached more than 10 ppbv yr^{-1} in 2014 and in 2018 (Ed Dlugokencky, NOAA/ESRL, 2019). The growth rate of CH_4 is determined by the imbalance of its

110   sources, primarily from anthropogenic activities (agriculture, waste, fossil fuel production and usage, and biomass burning) but also from natural emissions (mainly wetland and other inland waters), and sinks (OH oxidation, other chemical reactions with chlorine and oxygen radicals, and soil uptake). The precise scenario of the stagnation and renewed CH_4 growths still remains unclear (e.g. Rigby et al 2017; Saunois et al., 2017; Nisbet et al., 2019).

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Several studies have linked such CH_4 variations to inter-annual variations and trend of OH. Based on MCF inversions, McNorton et al. (2016) concluded that an increase in [OH] significantly contributed to the stable atmospheric CH_4 before 2007; Rigby et al. (2008) found that a decrease of $4\pm14\%$ in [OH]

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could partly explain the CH$_4$ growth between 2006 and 2007; Bousquet et al. (2011) found a smaller

120   decrease in [OH] (<1% over the two years) and attributed the increase in CH$_4$ mostly to enhanced

emissions over tropical regions; Montzka et al. (2011) also calculated a small inter-annual variation of

2.3±1.5% in [OH] during 1998 to 2007. More recently, based on multi-species box model inversions,

Rigby et al. (2017) and Turner et al. (2017) inferred a decrease of 8±11% and 7% in [OH] during 2004-

2014 and 2003-2016 respectively. Both of these studies suggested that such a decrease in [OH] is

125   equivalent to an increase of more than 20 Tg yr^{-1} in CH$_4$ emissions, and therefore could significantly

contribute to explain the post-2007 CH$_4$ atmospheric growth, although a solution with constant OH cannot

be discarded. Meanwhile, atmospheric chemistry models have calculated only a small increase of [OH]

(decrease in CH$_4$ lifetime) during the early 2000s, and usually show much smaller year-to-year variations

and long-term trends than most of the MCF-based inversions (e.g. Voulgarakis et al., 2013; Holmes et al.,

130   2013; Dalsøren et al., 2016). The discrepancy between individual process-based models and MCF-proxy

approaches, and the uncertainties, limit our ability to be conclusive on the role of [OH] changes to explain

the CH$_4$ changes over the past decades.

To better understand OH distributions, trends, and influences on CH$_4$ since 2000, we have performed an

135   inter-model comparison of 14 OH fields, including 11 derived from chemistry transport and chemistry-

climate models that took part in the phase 1 of the Chemistry-Climate Model Initiative (CCMI) (Hegglin

and Lamarque, 2015; Morgenstern et al., 2017), 2 from different configurations of the LSCE atmospheric

chemistry transport model LMDz-INCA (Hauglustaine et al., 2004;Szopa et al., 2013), and 1 from the

TRANSCOM 2011 inter-comparison exercise (Patra et al., 2011). Using this ensemble of OH fields, our

140   aim is to estimate a range for the contribution of changes in [OH] to the atmospheric CH$_4$ variations since

2000, and to relate this contribution to characteristics of the different OH fields. Compared with previous

inter-model studies of OH based ACCMIP model which simulated time-slices (constant emissions) (Naik

et al., 2013; Voulgarakis et al., 2013), OH fields from CCMI and INCA model simulations are time-

dependent, which allow us to address the question of year to year variability. In the following, our analysis

145   first provides a brief description of the OH fields used in this study and the LMDz offline model (section

2). Section 3 compares the OH fields, analyses the factors contributing to inter-model differences and

presents their inter-annual variability. Section 4 presents and discusses the impact of the different OH

fields on the global CH_4 burden and growth rates simulated by LMDz. Section 5 summarizes the results

and conclusion.

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2 Method

2.1 OH fields

The CCMI project aims to conduct a detailed evaluation of atmospheric chemistry models in order to

155   assess uncertainties in the models' projections of various climate-related topics such as tropospheric

composition (Hegglin and Lamarque, 2015; Morgenstern et al., 2017). The CCMI OH fields used in our

study are obtained from 10 different models and 2 CCMI reference experiments: REF-C1 (covering the

time period 1960-2010) and REF-C2 (covering 1960-2100). The REF-C1SD experiment is not analyzed

since it is conducted by only a part of the models and covers only a shorter time period (1980-2010). The

160   REF-C1 experiment is driven by state-of-the-art historical forcings and sea surface temperatures (sst) and

sea ice concentrations (sic) based on observations, while the REF-C2 experiment is using either coupled

ocean and sea ice modules or prescribes sst and sic obtained from another climate model. Since the REF-

C1 experiment is supposed to be more realistic regarding sea surface conditions, our analysis focused on

OH fields from the REF-C1 experiment before 2010 and only tested the influences of OH on CH_4

165   simulations after 2010 by applying the inter-annual variability from the REF-C2 experiment. Detailed

descriptions of CCMI simulations can be found in Morgenstern et al. (2017).

In this study, we used only the CCMI models that include coupled tropospheric ozone chemistry as listed

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in Table 1. Note that EMAC offers fields at two different model resolutions. The level of detail in chemical

170   mechanism, in particular with respect to included NMVOCs, varies among the models. For example, CMAM does not include any NMVOC species, but added 250 Tg CO emissions to account for CO production from isoprene oxidation. UMUKCA-UCAM only include HCHO and SOCOL3 only include HCHO and C_5H_8. Other models include multiple primary NMVOC species and more complex VOC chemistry.

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The anthropogenic emissions recommended for the two CCMI reference simulations are the MACCity inventory (Granier et al., 2011) for 1960-2000. After 2000, the REF-C1 experiment continued to use the MACCity inventory, while the REF-C2 used the RCP6.0 inventory (Masui et al., 2011). The CMAM model did not follow this procedure and used the ACCMIP historical database of emissions (Lamarque

180   et al., 2010) until 2000 followed by RCP8.5 emissions (Riahi et al., 2011) instead. CCMI model simulations also include natural emissions from lightning, soil and biogenic sources. Lightning NO_x emission are calculated based on meteorological   data such as cloud top height (Price and Rind, 1994; Grewe et al., 2001) and updraft mass flux (Allen and Pickering, 2002). Soil NO_x emissions are calculated interactively in EMAC and GEOSCCM using the scheme described by Yienger and Levy (1995), but are

185   prescribed in other models. Biogenic NMVOC emissions in CESM and GEOSCCM are calculated based on the distribution of plant functional types and meteorology conditions with MEGAN, whereas the other models apply prescribed biogenic NMVOC emissions.

The CCMI models do not represent CH_4 emissions explicitly but prescribe CH_4 surface mixing ratios

190   according to the RCP6.0 scenario (global mean of ~1750ppbv averaged from over 2000-2010) with different spatial distributions: GEOSCCM, CESM and EMAC models consider the full latitudinal gradient and prescribe CH_4 surface mixing ratios about 50 ppbv higher over the Northern hemisphere than over the Southern hemisphere; while CMAM, MRI-ESM1r1, and SOCOL3 use global uniform

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195   values. Photolysis rates are calculated either following online schemes such as FAST-JX (Neu et al., 2007;
Telford et al., 2013) by GEOSCCM, HadGEM3-EA, UMUKA-UCAM, JVAL (Sander et al., 2014) by
EMAC, or based on look-up tables with online cloud corrections by the rest of the models used in this
study. Kinetics and photolysis data are mainly from Sander et al. (2011) with a few exceptions. More
information on model characteristics can be found in Morgenstern et al. (2017) and references listed in
Table 1.

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Additionally to CCMI OH fields, we also included 2 OH fields simulated by the Interaction with
Chemistry and Aerosols (INCA) coupled to the general circulation model of the Laboratoire de
Meteorologie Dynamique (LMD), LMDz (Sadourny and Laval, 1984; Hourdin and Armengaud, 1999;
Hourdin et al., 2006; Hauglustaine et al., 2004). The two INCA simulations are driven by different
205   versions of the LMDz GCM (INCA NMHC-AER-S covering time period 2000-2010 (Terrenoire et al.,
2019), and INCA NMHC covering time period 2000-2009 (Szopa et al., 2013)), which provide different
water vapor fields, and include different chemistry and emissions. The INCA NMHC-AER-S used the
latest version of INCA model including both gas phase (NMHC) and aerosol (AER) chemistry in the
troposphere and the stratosphere (S) (Terrenoire et al., 2019), while INCA NMHC used a former version
210   that only includes tropospheric gas-phase chemistry (Szopa et al., 2013). Anthropogenic emissions for
INCA NMHC-AER-S and INCA NMHC are from the Evaluating the Climate and AirQuality Impacts of
Short-Lived Pollutants (ECLIPSE) inventory (Stohl et al.,2015) for 2005 and from the RCP85 emission
inventory (Riahi et al., 2011)) for 2010, respectively.

215   Finally, we included in this study the OH field used in TransCom simulations, which results from a
combination of the semi-empirical tropospheric 3-dimensional OH field from Spivakovsky et al. (2000)
and a 2-dimensional simulated stratospheric OH for year 2000. The tropospheric OH was calculated using
prescribed chemical species (O_3, nitrogen oxides, and CO) as well as meteorological fields (temperature,

humidity, and cloud optical depth) to fit the observations. The original tropospheric [OH] has been

220   reduced by 8% to match CH_3CCl_3 observations (Patra et al., 2011). The TransCom OH field is only

climatological (one year of monthly fields).

In total, we compared 14 OH fields: 11 from CCMI, 2 from the online LMDz-INCA model and 1 from

TransCom. We analyzed spatial distributions and annual variations of OH fields by calculating volume-

225   weighted tropospheric mean [OH]. Since employing different weightings can results in large differences

in mean [OH] (Lawrence et al., 2001), we also calculated dry air mass-weighted tropospheric mean [OH]

to better compare with previous studies.

2.2 LMDz model simulations

230   ### 2.2.1 Model description and setup

We have run the offline version LMDz5B of the LMDz model (Locatelli et al., 2015) at a horizontal

resolution of 3.75 °×1.85 ° with 39 vertical layers up to 3 hPa to access the impact of OH on tropospheric

CH_4. All monthly mean OH fields have been interpolated to the LMDz model grid. The transport of

atmospheric tracers is driven by prescribed air mass fluxes provided by the general circulation model

235   LMDz with horizontal wind fields nudged to ERA-Interim re-analysis meteorology data produced by the

European Center for Medium-Range Weather Forecasts (Dee et al, 2011). The vertical transport is

parameterized according to updates of the Emanuel (1991) scheme for convection and of the Louis (1979)

scheme for boundary layer mixing (Hourdin et al., 2016; Locatelli et al., 2015). Chemical sinks of CH_4

are calculated using prescribed three-dimensional OH and O^{1D} fields. No chlorine-related sink is

240   simulated in this version of the model. To assess the influences of OH only, all LMDz simulations used

the same O^{1D} fields generated by INCA model simulations. The reaction rate co-efficient (k) for CH_4

destruction by OH in the model is computed depending on temperature following Sander et al. (2011):

$$k = 2.45 \times 10^{-12} e^{-1775 \times (\frac{1}{T})} \qquad (1)$$

The LMDz model has been applied in various studies focusing on long-lived gases such as CH_4, CO_2 and

245   MCF (Bousquet et al., 2005; Pison et al., 2009; Lin et al., 2018). It has also been used in model inter-

comparison projects such as the TransCom experiment (Patra et al., 2011) with the simplified chemistry

module SACS (Pison et al., 2009) and CCMI (Morgenstern et al., 2017) but only with the stratospheric

chemistry model PEPROBUS (Jourdain et al., 2008).

250   The CH_4 emissions input to LMDz simulations are provided by the Global Carbon Project (GCP) methane

and include anthropogenic and biofuel emissions from EDGARv4.3.2 (Janssens-Maenhout et al., 2017),

the mean wetland emissions from Poulter et al., (2017), fire emissions from the Global Fire Emissions

Database Version 4.1 (GFED4) (Randerson et al., 2018), termite emissions as described by Saunois et al.

(2016), geological emissions based on the spatial distribution of Etiope (2015), ocean emissions from

255   Lambert and Schmidt (1993) and soil uptake from Ridgwell et al. (1999). EDGARv4.3.2 data, available

until 2012, were extrapolated from 2013 to 2016 using economical statistics according to the methodology

described by Saunois et al., (2016). Anthropogenic and fire emissions vary from 2000 to 2016 while

natural emissions are applied as a climatology.

260   The spatial distributions and annual variations of the CH_4 emissions during the study period are shown in

Fig.1. CH_4 emissions range from 10 to 40 kg ha^{-1} yr^{1} over most natural ecosystems and can exceed 100

kg ha^{-1} yr^{-1} over wetlands in Canada, South America, and Central Africa, as well as over densely

populated regions such as South and East Asia. Global net CH_4 emissions (soil uptake included) increased

by 15% from 482 Tg yr^{-1} in 2000 to 552 Tg yr^{-1} in 2016. Of this 70 Tg yr^{-1} increase, 60 Tg yr^{-1} (85%) are

265   emissions from the Northern hemisphere, mainly contributed by livestock (18 Tg yr^{-1}, 25%), oil and gas

(16 Tg yr^{-1}, 23%), coal burning (17 Tg yr^{-1}, 24%) and waste (13Tg yr^{-1}, 18%). The three emission peaks

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in 2002, 2006 and 2015 are driven by biomass burning. This CH_4 emission scenario is state-of-the-art but has not been optimized for the simulated CH_4 mixing ratios to fit the observations.

270   2.2.2 Model simulations

Two sets of experiments (steady-state and transient simulations) have been performed to examine the impacts of the input OH fields on the global CH_4 burden as well as the CH_4 spatial distribution and annual variation. These tests excluded the OH fields from CESM1-CAM4chem and EMAC-L47MA, since they are similar to those of CESM1-WACCM and EMAC-L90MA, respectively. We also discarded the OH

275   fields from HadGEM3-ES and UMUKCA-UCAM because output from these two models has been supplied on too coarse vertical pressure levels. Finally, 10 different OH fields (seven from CCMI, two from LMDz-INCA and one from Transcom) were used in the two sets of simulations.

Initially, for each OH field described in Sect. 3, we ran 30 consecutive years of LMDz simulations (with

280   recycled same emissions, sinks, and meteorology of the year 2000) to allow the simulation to reach a steady-state (CH_4 has an approximate lifetime of 9 years in the atmosphere). This step aims to examine the impact of the magnitude and distribution of OH on the global CH_4 burden.

Secondly, we performed transient simulations starting from the year 2000, which are forced by time-

285   varying OH fields as well as time-varying emissions and meteorology fields. In order to compare the impacts of the different OH fields on realistic CH_4 mixing ratios, for each simulation (except the one using the OH fields from INCA NMHC), the OH field has been scaled to get the same LMDz simulated CH_4 loss as the one calculated by INCA NMHC in 2000, as INCA is the OH field consistently obtained using the LMDz transport. Then a series of LMDz model simulations is conducted to investigate the

290   impact of the various OH fields on CH_4 growth rates between 2000 and 2016 as summarized in Table 2.

The standard simulations (Run_standard in Table 2) using the 10 different OH fields (7 are from CCMI

REF-C1), included annual variations and were performed from 2000 to 2010. Since REF-C1 experiments

are only available up to 2010, the influence of OH on CH_4 mixing ratios after 2010 have been tested based

on alternative scenarios. First, for CCMI simulations, we tested a scenario that takes into account the

annual variability from the REF-C2 experiments (Run_REF-C2 in Table 2). Previous ACCMIP model

experiments showed slightly decreasing or increasing [OH] from 2000 to 2030 according to the largest or

lowest radiative forcing pathways (RCP8.5 or RCP2.6), respectively (Voulgarakis et al., 2013). Top-down

approaches suggested that global OH decreased by 0.5-1% annually from 2003 to 2016 (Rigby et al.,

2017; Turner et al., 2017). In order to assess the recent change in OH, we tested two additional scenarios

between 2010 and 2016: one with [OH] increase of $+1‰$ yr^{-1} (Run_OH_inc) and one with [OH] decrease

of -1% yr^{-1} (Run_OH_dec) . To assess influences from OH alone, we also conducted additional

simulations of the period 2000 to 2016 with annually repeated prescribed [OH] equal to the year 2000

(Run_fix_OH) for each OH field. The differences between these constant OH simulations and the

corresponding time-varying OH simulations indicate the impact of OH inter-annual variations and trends

on atmospheric CH_4 changes.

3 Analysis of OH fields

3.1 Spatial distributions of tropospheric OH

Fig. 2 shows the spatial distributions of volume-weighted tropospheric mean [OH] averaged from 2000

to 2010. Based on the 14 OH fields we have assembled, the global mean volume-weighted tropospheric

[OH] vary from 8.7×10^5 to 12.8×10^5 molec cm^{-3}. SOCOL simulated the highest [OH], which

overestimation of [OH] have been reported by Staehelin et al. (2017). To better compare with previous

studies, we also calculated dry air mass-weighted tropospheric mean [OH] in table 4, which vary from

9.4×10^5 to 14.4×10^5 molec cm^{-3}. This (large) range is consistent with previous multi-model results given

by IPCC (2011) and the ACCMIP project (Naik et al., 2013; Voulgarakis et al), as well as with inversions

based on MCF observations (Bousquet et al., 2005; Rigby et al., 2017). The model spread remains large

as ~50% of the minimum value, as noted in previous studies (e.g. Naik et al., 2013).

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Table 3 summarizes their inter-hemispheric ratios of tropospheric OH and mean values over four

latitudinal bands. The inter-hemispheric ratios (N/S ratios) of CCMI and INCA OH fields are within the

range of 1.2-1.5, similar to those from the ACCMIP project (Naik et al., 2013). In contrast, the TransCom

OH field has a ratio of 1.0, which is more consistent with that of MCF and ^{14}C constrained OH

325   (Brenninkmeijer et al., 1992; Krol and Lelieveld, 2003; Bousquet et al., 2005). However, as discussed by

Spivakovsky et al. (2000), the TransCom OH field may overestimate Southern Extra-tropics OH by ~25%.

The lower N/S ratios inferred from MCF observations are mainly due to high [OH] over the Southern

Tropics (35% higher than Northern Tropics) (Bousquet et al., 2005). In contrast, process-based simulated

OH is 10-26% more abundant over the Northern Tropics than over the Southern Tropics, and 35% to >

330   90% higher over 30°N-90°N than 30°S-90°S in the CCMI models. Previous studies have attributed the

inconsistency between the simulated and the observed OH N/S ratios to a model overestimation of O_3 and

underestimation of CO over the Northern Hemisphere (Naik et al., 2013; Young et al., 2013; Strode et al.,

2015), as well as to a lack of OH recycling due to the presence of VOCs over rainforest (mainly located

in the Southern Tropics) (Lelieveld et al., 2008; Archibald et al., 2011).

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We further assessed the simulated OH spread by comparing the detailed spatial distributions of OH fields

in Fig. 2. Nearly all CCMI models and two versions of the INCA model simulated high [OH] over eastern

North American and South and East Asia, which is related to higher tropospheric O_3 concentrations

(Cooper et al., 2014; Lu et al., 2018) and NO_x emissions from human activities (Lamarque et al., 2010;

340   Miyazaki et al., 2012). High [OH] over these emission hotspots dominate the aforementioned simulated

large N/S ratio. Some models also simulated high OH values over the African savanna plains (MOCAGE

and INCA excluded), regions with intense biomass burning (van der Werf et al., 2006) and soil NO_x

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emissions (Yienger and Levy 1995; Vinken et al., 2014). The O_3 concentrations used to generate the

TransCom OH field were larger in the Southern Tropics than in the Northern latitudes (Spivakovsky et

345   al., 2000), in contrast to recent observations (Cooper et al., 2014). Therefore, TransCom shows the highest

[OH] over the Southern Tropics during biomass burning seasons (Spivakovsky et al., 2000) and thus a

lower N/S ratio.

Despite consistency on high OH values over regions influenced by human activities and biomass burning,

350   models show the largest discrepancies over some natural ecosystem such as tropical rainforests. For

example, INCA, CESM, HadGEM3-Es, MRI-ESM1r1, MOCAGE and GEOSCCM simulated overall low

[OH] (4×10^5-14×10^5 molec cm^{-3}) over tropical rainforests, despite differences in details, while EMAC,

CMAM, SOCOL3 and UMUKCA-UCAM simulated overall high [OH] (16×10^5- more than 25×10^5

molec cm^{-3}). Besides these, inter-model differences also exist in remote areas such as over the open ocean.

355   Most simulated OH fields show higher concentrations over continents or coastal areas due to higher

precursor emissions, while MRI-ESM1r1, EMAC, and GEOSGCM also simulated high values ($>15\times10^5$

molec cm^{-3}) over the open ocean. Factors contributing to these inter-model differences are further

discussed in Sect. 3.3

3.2 Vertical distributions

Figure 3 shows the vertical distribution of OH fields and Table 4 provides the volume-weighted mean

[OH] averaged over the troposphere and over three pressure latitudinal intervals representing the planetary

boundary layer, the mid-troposphere, and the upper troposphere (surface-750, 750-500, and 500-250 hPa,

respectively). At the global scale, the mean tropospheric concentration of TransCom OH increases by a

365   factor of nearly two from the surface (7×10^5 molec cm^{-3}) to 600hPa (13×10^5 molec cm^{-3}) and then

decreases rapidly with altitude (7×10^5 molec cm^{-3} at 250hPa). UMUKA-UCAM, HadGEM3-ES, CMAM,

MOCAGE, and SOCLO3 on the other hand all show a continuous decrease of [OH] with altitude from

the surface to the upper troposphere (e.g. the global mean concentrations of MOCAGE OH decreases

from 23.6×10^5 molec cm^{-3} at the surface to 6.4×10^5 molec cm^{-3} at 250hPa). Other OH profiles show much

370 smaller vertical variations in the troposphere (standard deviations of mean value below 200hPa $< 2 \times 10^5$

molec cm^{-3}).

Model simulated OH vertical distributions can also be different over land versus ocean (Fig. 3) and

between the different latitudinal bands (Fig. S1). For example, SOCOL3 [OH] continuously decreases

375 with altitude over both, land and ocean; MOCAGE OH increases from the surface (14.9×10^5 molec cm^{-3})

to 800hPa (18.2×10^5 molec cm^{-3}) and then decreases over land but almost continuously decreases over

the ocean; CMAM and UMUKCA-UCAM only show significant vertical variations in [OH] over land.

Vertical variations of most OH fields can be attributed to mid and low latitude regions, except for those

of SOCOL3 and MOCAGE, that also decrease with altitude over mid and high northern latitudes (45 °N

380 -90 °N, see Fig. S1).

3.3 Factors contributing to inter-model differences

Tropospheric OH is produced primarily through the reaction of O^{1D} with H$_2$O and secondarily through

the reaction of NO with HO$_2$ and RO$_2$, and is removed primarily by reacting with CO and CH$_4$ (Logan et

385 al., 1981). Hence, factors controlling inter-model OH discrepancies can be complex as differences in

model emissions, chemistry, and dynamics can together impact [OH]. Here we propose a qualitative

analysis focusing on both, emissions and chemical mechanisms. A more quantitative analysis would

require a detailed model output of production and loss pathways and is beyond the scope of this work.

390 To analyse inter-model differences in OH vertical distributions, we compared the O^{1D} photolysis rates,

specific humidity, and CO and NO mixing ratios in table 5. The inter-model variations (calculated as

standard deviation/multi-model mean) in tropospheric O^{1D} photolysis rates, specific humidity, and CO

16

mixing ratios are usually <10%-20%, while NO mixing ratios show a larger variation of 38% (12-32pptv). MRI-ESM1r1 simulated the highest NO tropospheric mixing ratio, mainly attributable to high values
395   above 200hPa, where OH formation is limited by H_2O. In addition, MRI-ESM1r1 has ~20% more CO emissions than MOCAGE and GEOSCCM (Fig. S2), leading to about 10 ppbv higher CO mixing ratios, offsetting (for [OH]) its higher NO_x emissions and NO mixing ratios. The high NO mixing ratios near the surface and mid-troposphere simulated by SOCOL3 (48 pptv below 750hPa and 10 pptv from 750 to 500hPa), MOCAGE (26 pptv below 750hPa and 14 pptv from 750 to 500hPa) and CMAM (17 ppbv
400   below 750hPa) are consistent with their high tropospheric and near-surface [OH].

Besides emissions, previous studies have reported additional factors leading to high surface NO and NO_2. The overestimation of NO by MOCAGE could be due to the lack of N_2O_5 heterogeneous hydrolysis on tropospheric aerosol, which is an efficient sink for NO_x (Teyssèdre et al., 2007). SOCOL3 does not include
405   N_2O_5 heterogeneous hydrolysis and also overestimates tropospheric NO production by NO_2 photolysis compared to other models, due to issues with the look-up tables used in the calculation of photolysis rates (Revell et al. 2018). We conclude here that physical and chemical processes related to NO production and loss can have a large impact on OH burden and its vertical distribution. In this context, an improved representation of the partitioning between NO and other nitrogen species in the models seems of great
410   importance to correctly simulate tropospheric [OH].

Concerning the spatial distributions, as aforementioned in Sect. 3.1, the largest model discrepancies are found over tropical rain forests. The [OH] over tropical rainforest regions are mostly sensitive to natural emissions including NO_x and NMVOCs, which vary among the models. Previous studies showed that
415   [OH] is more sensitive to soil and lightning emissions than to wildfires, because the former sources only emit NO_x (OH source), whereas the latter emits NO_x, CO and VOCs together (OH sources and OH sinks, see Murray et al., 2014). Soil NO_x emissions in CCMI models range from around 4 Tg N yr^{-1} in MOCAGE

to more than 7 Tg N yr^{-1} in GEOSCCM and 9Tg N yr^{-1} in CMAM (Naik et al., 2013; Yienger and Levy,

1995). In particular, lower NO$_x$ emissions over South America and Africa in MOCAGE might be linked

420   to lower [OH] over this region (Fig. S2). Isoprene and other NMVOCs remove about 3% and 7% of

troposheric OH on a global scale, respectively (Spivakovsky et al., 2000; Murray et al., 2014) and can

be more important over tropical regions with higher emission rates (Sindelarova et al., 2014). The higher

[OH] over tropical rainforests simulated by CMAM and UMUKCA-UCAM may be due to a lack of or

the lower OH destruction by VOCs in these models. Therefore, the inter-model differences in OH spatial

425   distributions over tropical rainforests may result from differences in natural emissions of VOC species

and different related chemical reactions.

3.4 Inter-annual variations of OH

Figure 4 shows the time series of volume-weighted troposheric mean [OH] from 1960 to 2010 (from

430   REF-C1 CCMI comparison). During this period, all OH fields show small year-to-year variations,

remaining within $\pm 0.5 \times 10^5$ molec cm^{-3}. CCMI models simulated significantly different OH long-term

evolutions from 1960 to 1980. For example, [OH] continuously decrease in the CMAM and HadGEM3-

ES simulations ($\sim -0.3 \times 10^5$ molec cm^{-3}); and increase in SOCOL3 ($\sim +0.6 \times 10^5$ molec cm^{-3}), UMUKCA-

UCAM ($\sim +0.5 \times 10^5$ molec cm^{-3}), and MOCAGE ($\sim +0.5 \times 10^5$ molec cm^{-3}) during this period, while other

435   models show no obvious long-term trend. After 1980 (1990 for CMAM), all models show stabilized or

slightly increasing [OH]. For our period of interest (after 2000) and focusing on the anomaly in [OH]

compared to the 2000-2010 mean (Fig. 4b), OH year-to-year variations are found to be smaller than in

previous decades and [OH] only increases by about 0.1-0.3$\times 10^5$ molec cm^{-3} from 2000 to 2010.

440   Previous atmospheric chemistry model studies have concluded that anthropogenic activities lead to only

a small perturbation of the OH burden, as the increased OH production tend to be compensated by an

increased loss through reactions with CO and CH$_4$ (Lelieveld et al., 2000; Naik et al., 2013). The ensemble

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of ACCMIP models simulated large divergent OH changes (even in their signs) from 1850 to 2000, but

revealed a consistent increase of 3.5±2.2% from 1980 to 2000 (Naik et al., 2013). Here, for the same

445 period the increase of CCMI [OH] is 4.6±2.4%, consistent with the ACCMIP project (Naik et al., 2013)

and with other atmospheric chemistry model studies (Dentener et al., 2003; John et al., 2012; Holmes et

al., 2013; Dalsøren et al., 2016). The year-to-year variations of most CCMI and INCA OH fields are much

smaller than the OH inter-annual variability based on MCF observations (e.g. Bousquet et al., 2005;

Montzka et al., 2011), which can reach 8.5±1.0% from 1980 to 2000, and 2.3±1.5% from 1998 to 2007,

450 as compared to 2.1±0.8% and 1.0±0.5% here for these two periods.

We further analyzed regional [OH] trends from 2000 to 2010 in Fig. 5. Instead of dividing subdomains as

Naik et al. (2013) did, we calculated the trend for each model grid-cell to identify and distinguish regions

with different trends. Most models show significant positive [OH] trends over tropical regions (0.05-

455 0.1×10^5 molec cm^{-3} yr^{-1}) and over East and Southeast Asia (>0.1×10^5 molec cm^{-3} yr^{-1}). From 2000 to 2010,

NO_x emissions in the MACCity (RCP85) inventory increased by 83% over East Asia, which is much

faster than the CO increase (8%) (Riahi et al., 2011) and can explain the strong positive trend of CCMI

simulations. Over the rest of the extra-tropical regions such as North America and Western Europe, the

models disagree on the sign of OH change. In the Southern hemisphere, where biogenic and fire emissions

460 dominate, most OH fields do not show clear trends and the inter-model differences are even larger. For

example, MOCAGE simulated and OH decrease of >0.1×10^5 molec cm^{-3} yr^{-1} over the Amazon, South

Africa, and Indonesia, whereas MRI-ESM and EMAC-L90MA simulated positive OH trends over these

regions; HadGEM3-ES simulated a significant decrease of the OH trend over most of the extra-tropical

regions in the Southern hemisphere, while CMAM simulated slightly positive trends in this hemisphere.

465

In the following, we investigate how the differences in mean [OH] and variations presented in this section

affect CH_4 burden and its variations for the period 2000-2016.

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4 Influences of OH fields on CH₄ simulations

470

4.1 Global total CH₄ burden

We now present the results based on the first set of LMDz experiments, where the LMDz model was run

for 30 years recycling the year 2000 until the steady-state is reached. The simulations using the OH fields

as given by CCMI and INCA models provide a wide range of values for the tropospheric global mean

475   CH₄ mixing ratios (Table 6), from 1204 ppbv (SOCOL3, with a global volume-weighted tropospheric

mean [OH] of 12.8×10^5 molec cm^{-3}) to 1822 ppbv (INCA NMHC-AER-S, with a global volume-

weighted tropospheric mean [OH] of 8.7×10^5 molec cm^{-3}). It appears that the global CH₄ burden is not

only sensitive to the global mean [OH], but also to its vertical distribution. Indeed, the OH radicals in the

lower troposphere are more efficient to oxidize CH₄ molecules, because the CH₄+OH reaction rate

480   increases with temperature (Eq. 1). When considering the standard atmosphere, the reaction rate

corresponding to the surface temperature of 288K (5.2×10^{-15} s^{-1}) is more than twice that for the 500hPa

temperature of 253K (2.2×10^{-15} s^{-1}). Despite similar volume-weighted tropospheric mean [OH] of ~10.4

$\times 10^5$ molec cm^{-3}, MOCAGE simulated much lower CH₄ mixing ratios (1275 ppbv) than CMAM (1540

ppbv) and MRI-ESM1r1 (1639 ppbv) because of its higher near surface [OH] (19×10^5 molec m^{-3}) (Table

485   4). The spatial distribution of the OH radicals also slightly influences CH₄ oxidation. Indeed, the [OH] of

EMAC-L90MA are higher than those of CESM-WACCM for both, tropospheric (11.1×10^5 versus 10.7

$\times 10^5$ molec cm^{-3}) and near-surface (12.5×10^5 versus 12.4×10^5 molec cm^{-3}) means, but a slightly higher

CH₄ burden is found for the former (1579 versus 1575ppbv, Table 6). This is because EMAC-L90MA

simulated higher [OH] over the ocean, while CESM-WACCM OH is more concentrated over land closer

490   to CH₄ source regions. The model experiments also emphasize that volume-weighted tropospheric

concentrations cannot fully indicate the atmospheric oxidizing efficiency for CH₄, as has been discussed

by Lawrence et al. (2001). Tropospheric mean [OH] weighted by reaction rates with CH₄, which consider

both temperature and CH_4 distributions, can be a better indicator for CH_4 oxidation (Lawrence et al., 2001).

495

4.2 Impacts on CH_4 spatial distribution and growth rate

In order to address the question of inter-annual variability of atmospheric CH_4, we scaled each OH field globally to get the same CH_4 loss (for the year 2000) as the one obtained with INCA NMHC OH field (see Sect. 2.2.2). The scaling used a single global scaling factor (per OH field). As listed in Table 4, after

500   scaling most OH fields have volume-weighted tropospheric mean concentrations closer to INCA NMHC (9.7×10^5 molec cm^{-3}), within the range of 9.0-10.4×10^5 molec cm^{-3}. One exception is MOCAGE, with tropospheric mean [OH] scaled to 7.7×10^5 molec cm^{-3}, due to its distinct vertical distribution (Sect.3.2). This scaling of OH makes it possible to start model experiments at the same initial CH_4 burden. Although slightly modifying the magnitude of the global mean [OH], this scaling maintained the spatial and

505   temporal differences and trend over the 2000-2010 period.

4.2.1 Spatial distributions of tropospheric CH_4 mixing ratios

We used the scaled OH fields to perform simulations between 2000 and 2010. Figure 6 shows the spatial distribution of tropospheric CH_4 mixing ratios for the simulation Run_standard (Table. 2, driven by OH

510   with inter-annual variations) averaged over 2000-2010. Although all simulations started from the same initial conditions and OH fields were scaled to give the same global CH_4 loss as INCA NMHC in 2000, using the differently scaled OH fields still generated a spread of 8 ppbv in the 11-year average tropospheric mean CH_4 mixing ratios. The LMDz model using the TransCom OH fields (without inter-annual variability) simulated the highest CH_4 mixing ratios (1735 ppbv), while the one using the CMAM

515   OH (with slightly increasing trends during the decade) simulated the lowest (1727 ppbv). Differences between the global tropospheric mean [OH] cannot explain these differences (see Table 4). Clearly the different spatial (horizontal and vertical) and temporal variations of the OH fields (as described in Sect.

21

3), which were kept in this experiment, significantly modify the simulated CH_4 mixing ratios (Table 7 and Fig. 6).

520

Looking at latitudinal CH_4 mixing ratios, the inter-model differences appear larger than in the global mean (Fig. 6 and Table 7). The model spreads of the mean CH_4 mixing ratios over 60 °S-90 °S and 60 °N-90 °N range from 1771 to 1794ppbv and 1784 to 1812ppbv, respectively. Here, we define the N/S gradient of CH_4 as the difference in mean CH_4 mixing ratio between the latitudinal bands 60 °N - 90 °N and from 60 °S

525 - 90 °S. With the TransCom OH field (N/S ratio =1.0), the model simulated 12-43% larger N/S gradients of CH_4 (129 ppbv) than other simulations (90-115 ppbv) driven by OH fields with higher N/S OH ratios of 1.2-1.5. Previous model studies have attributed the overestimation of the CH_4 N/S ratio to an underestimation of model inter-hemispheric exchange time (e.g. Zimmermann et al., 2018). Our results show that uncertainties in OH distributions can also contribute to such model biases.

530

4.2.2 Changes in CH_4 mixing ratios

To assess the influence of OH inter-annual variations on CH_4 mixing ratios, we calculated the difference in the simulated CH_4 between the standard run (Run_standard) and the simulations with fixed [OH] (Run_fix_OH, Table 2). The Run_fix_OH simulations show that global tropospheric mean CH_4 mixing

535 ratios increased by 75 ppbv from 2000 to 2010 (Fig. 7, black dashed lines), due to the enhanced emissions (Fig.1). The increase in [OH] can obviously reduce CH_4 growth. An increase in [OH] by 0.1-0.3×10^5 molec cm^{-3} (1%-3%) (Fig. 7, orange lines) during this period leads to a reduction of the CH_4 mixing ratios by 5-15 ppbv by 2010 (Fig. 7, blue lines).The largest reductions are found when using CESM1-WACCM and CMAM OH fields, given the continuous OH growth in these models. Compared to Run_fix_OH, we

540 estimated that such reductions in CH_4 mixing ratios offset 7- 20% of the CH_4 increase driven by the rising CH_4 emissions of our scenario over the period 2000 to 2010.

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After 2010, CCMI REF-C2 experiments simulated increasing, relatively stable, or decreasing OH variations, thus having a variable influence on CH_4 variations. Over the period 2011-2016, [OH]

545   simulated by EMEC-L90MA, CESM-WACCM and CMAM stabilizes at a level $0.2\text{-}0.4 \times 10^5$ molec cm^{-3} higher than the concentrations in 2000, further reducing CH_4 mixing ratios by up to 20-30 ppbv in 2016 (Fig. 7, blue lines). Other OH fields have similar concentrations over 2010-2016 as in the early 2000s (Fig. 7, orange lines), thus simulating CH_4 mixing ratios that remain close to Run_fix_OH with differences less than a few +/-ppbv.

550

As large uncertainties remain regarding the inter-annual variations and trend of OH after 2010, we have tested two additional OH scenarios: Run_OH_inc (with an annual increase of 1‰ yr^{-1}) and Run_OH_dec (with an annual decrease of 1% yr^{-1}), to assess the uncertainty range of the impact of OH changes (the orange areas in Fig. 7) on CH_4 mixing ratios (the blue areas in Fig. 7). In these two scenarios, the mean

555   [OH] of run_OH_dec is $\sim 7 \times 10^5$ molec cm^{-3} (7%) lower than run_OH_inc in 2016. If OH decreases at 1% yr^{-1} after 2010, by 2016, the differences in CH_4 mixing ratios between Run_OH_dec and Run_fix_OH range -7–30ppbv, with the lower end (-7ppbv) simulated by OH from CESM1-WACCM given its highest [OH] in 2010. On the contrary, Run_OH_inc simulated 3-39 ppbv lower CH_4 mixing ratios compared to Run_fix_OH (the blue areas in Fig. 7). As such, uncertainties in the OH trend can clearly lead to $> \pm 30$

560   ppbv changes in CH_4 mixing ratios (the gray areas in Fig. 7) after only 6 years of simulations, as compared to the fix-OH case.

It is now interesting to compare the range of simulated [CH_4] changes induced by OH scenarios to changes in surface CH_4 observations, in order to quantify how much of the model-observation mismatch could

565   potentially be attributed to uncertainties in [OH] and its variability (Fig. 8). To do so, we used surface CH_4 observations from the National Oceanic and Atmospheric Administration (NOAA) networks and selected stations with 17 years continuous records over 2000-2016. Since the simulated absolute CH_4

mixing ratios largely depend on the initial conditions and OH fields, we compared changes in the

simulated and observed global CH_4 mixing ratios starting at the same point in 2000. The observed CH_4

570   shows zero growth between 2000 and 2006 and then increases by 5.6 ppbv yr^{-1} between 2006 and 2012

(6.4 ppb yr^{-1} for 2006-2010) and by 9.4 ppbv yr^{-1} after 2012. In this study we do not expect to fit these

CH_4 trends as this inter-comparison was not conducted with a set of optimized emissions. It has already

been noticed that standard CH_4 emission inventories lead to overestimated CH_4 mixing ratios (e.g.

Saunois et al., 2016). Indeed, neither Run_standard nor Run_fix_OH simulations do capture the

575   stagnation before 2006, and overestimated surface CH_4 increments by 2.5-5.2 ppbv yr^{-1} during the period

2006-2010. Based on Run_fix_OH, on average over 2000-2016 and depending on the OH scenario, we

found that [OH] changes can emphasize the model-observation mismatch by up to 19%, or fill the gap

between model simulations and observations by up to 54% (figure 8). Such comparisons strongly suggest

that a better understanding of OH inter-annual variations and trends is required in order to simulate more

580   reliable CH_4 trends in atmospheric chemistry models. Atmospheric chemistry transport model (Dalsøren

et al., 2016) and box model studies (Rigby et al., 2017; Turner et al., 2017) have pointed out that variations

in OH can partly explain the recent CH_4 trends. However, current top-down estimates of CH_4 emissions

usually assume constant [OH] (Saunois et al., 2017) and attribute the model-observation discrepancies

only to surface emissions rather than changes in [OH]. Our results confirm the potentially significant role

585   played by the still uncertain OH changes in the actual changes of methane emissions since 2000.

5 Conclusions

We have analyzed 14 OH fields (11 from CCMI experiments, 2 from INCA model simulations, 1 from

590   TransCom) to investigate the inter-model differences in the spatial distributions and trends of tropospheric

OH, and estimated the influences of OH spatio-temporal distributions on tropospheric CH_4 by feeding

them in different simulations with LMDz offline chemistry transport model.

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595 Simulated global volume-weighted tropospheric mean [OH] are within the range of 8.7×10^5-12.8×10^5 molec cm^{-3}, which is consistent with the (large) multi-model range of previous estimates. CCMI and INCA models simulated larger [OH] in the Northern hemisphere than in the Southern hemisphere (N/S ratio of 1.2-1.5), with consistently high OH values over anthropogenic emission hotspots in North America, East and South East Asia, while TransCom OH shows a N/S ratio close to 1.0. In the vertical, TransCom OH reaches its maximum value at about 600 hPa, while CCMI and INCA OH fields either

600 continuously decrease with altitude or show very small vertical variations in the troposphere. The factors most likely responsible for these inter-model differences include i) large NO mixing ratios leading to high surface and mid-tropospheric [OH] (Teyssèdre et al., 2007; Pevell et al. 2018), and ii) different natural emissions as well as VOC species and chemical mechanisms driving spatial model discrepancies over natural ecosystems.

605

Simulated OH fields show small year-to-year variations, within $\pm 0.5 \times 10^5$ molec cm^{-3} during 1960-2010. From 2000 to 2010, year-to-year variations in OH are smaller than in previous decades and all OH fields increase by about 0.01-0.03×10^5 molec cm^{-3} yr^{-1}. Such an increase in OH is mainly attributed to the significant positive OH trend over East and Southeast Asia ($>0.1 \times 10^5$ molec cm^{-3} yr^{-1}), in response to more

610 OH production by NO$_x$ than OH loss by CO (Riahi et al., 2011).

The inter-model differences in tropospheric OH burden generate a wide range of CH$_4$ burdens (1204-1882ppbv) when used to simulate steady-state CH$_4$ mixing ratios in the atmospheric chemistry model LMDz. Our findings suggest that not only different global mean [OH], but also differences in the

615 horizontal and vertical distributions between OH fields are responsible for this range (CH$_4$ destruction rates by OH increase with temperature).

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The CH$_4$ simulations for 2000-2016 using OH with inter-annual variation show that inter-model differences of the OH N/S ratio lead to 12-43% differences in the CH$_4$ N/S gradient. For the time period 620   2000-2010, we found that a 1%-3% increase in [OH] leads to a 5-15 ppb reduction of the CH$_4$ mixing ratio until 2010, accounting for 7-20% of the simulated emission driven CH$_4$ increase over this period. After 2010, the ensemble of OH scenarios tested here leads to differences in CH$_4$ mixing ratio of up to 30 ppb by 2016. Comparing with surface observations, we found that [OH] changes can emphasize the model-observation mismatch by up to 19%, or fill the gap between model simulations and observations 625   by up to 54% (Figure 8). Therefore, addressing the OH variability in CH$_4$ source inversions seems critical to avoid a wrong attribution of CH$_4$ changes to emission changes only. Future work is needed to quantify the impact of this ensemble of OH fields on CH$_4$ emissions obtained by inversion and to generate improved OH fields to be used in CH$_4$ inversion studies.

630   **Author contributions**

YZ, MS, and PB designed the study, analyzed data and wrote the manuscript. BZ and XL helped with data analysis and model simulations. JC, RJ, and AS discussed the results. ED provided the atmospheric in situ data. Other co-authors provided numerical model outputs. All co-authors commented on the manuscript.

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References

660 Allen, D. J., and Pickering, K. E.: Evaluation of lightning flash rate parameterizations for use in a global
chemical transport model, Journal of Geophysical Research: Atmospheres, 107, ACH 15-11-ACH 15-21,
doi:10.1029/2002JD002066, 2002.

Bednarz, E. M., Maycock, A. C., Abraham, N. L., Braesicke, P., Dessens, O., and Pyle, J. A.: Future
Arctic ozone recovery: the importance of chemistry and dynamics, Atmos. Chem. Phys., 16, 12159–
665 12176, doi:10.5194/acp-16-12159-2016, 2016.

Bousquet, P., Hauglustaine, D. A., Peylin, P., Carouge, C., and Ciais, P.: Two decades of OH variability
as inferred by an inversion of atmospheric transport and chemistry of methyl chloroform, Atmos. Chem.
Phys., 5, 2635-2656, doi:10.5194/acp-5-2635-2005, 2005.

Archibald, A. T., Levine, J. G., Abraham, N. L., Cooke, M. C., Edwards, P. M., Heard, D. E., Jenkin, M.
670 E., Karunaharan, A., Pike, R. C., Monks, P. S., Shallcross, D. E., Telford, P. J., Whalley, L. K., and Pyle,

J. A.: Impacts of HOx regeneration and recycling in the oxidation of isoprene: Consequences for the composition of past, present and future atmospheres, Geophysical Research Letters, 38, L05804, doi:10.1029/2010GL046520, 2011.Bousquet, P., Ringeval, B., Pison, I., Dlugokencky, E. J., Brunke, E. G., Carouge, C., Chevallier, F., Fortems-Cheiney, A., Frankenberg, C., Hauglustaine, D. A., Krummel, P.

675   B., Langenfelds, R. L., Ramonet, M., Schmidt, M., Steele, L. P., Szopa, S., Yver, C., Viovy, N., and Ciais, P.: Source attribution of the changes in atmospheric methane for 2006–2008, Atmospheric Chemistry and Physics, 11, 3689-3700, doi:10.5194/acp-11-3689-2011, 2011.

Brenninkmeijer, C. A. M., Manning, M. R., Lowe, D. C., Wallace, G., Sparks, R. J., and Volz-Thomas, A.: Interhemispheric asymmetry in OH abundance inferred from measurements of atmospheric 14CO,

680   Nature, 356, doi:50, 10.1038/356050a0, 1992.

Ciais, P., Sabine, C., Bala, G., Bopp, L., Brovkin, V., al, e., and House, J. I.: Carbon and Other Biogeochemical Cycles, in: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change Change, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 2013.

685   Colin, P., and David, R.: Possible implications of global climate change on global lightning distributions and frequencies, Journal of Geophysical Research: Atmospheres, 99, 10823-10831, doi:10.1029/94JD00019, 1994.

Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N.,

690   Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Quarterly Journal of the Royal Meteorological Society, 137, 553-597, doi:10.1002/qj.828, 2011.

695   Ed Dlugokencky, NOAA/ESRL ,www.esrl.noaa.gov/gmd/ccgg/trends_ch4/, 2018

Emanuel, K. A.: A Scheme for Representing Cumulus Convection in Large-Scale Models, Journal of the Atmospheric Sciences, 48, 2313-2329, doi:10.1175/1520-0469(1991)048<2313:ASFRCC>2.0.CO;2, 1991.

Cooper, O. R., Parrish, D. D., Ziemke, J., Balashov, N. V., Cupeiro, M., Galbally, I. E., Gilge, S., Horowitz,

700   L., Jensen, N. R., Lamarque, J. F., Naik, V., Oltmans, S. J., Schwab, J., Shindell, D. T., Thompson, A. M., Thouret, V., Wang, Y., and Zbinden, R. M.: Global distribution and trends of tropospheric ozone: An observation-based review, Elementa: Science of the Anthropocene, 2, 000029, doi: 10.12952/journal.elementa.000029, 2014.

Crutzen, P.: A discussion of the chemistry of some minor constituents in the stratosphere and troposphere,

705   Pure and Applied Geophysics, 106, 1385-1399, doi: 10.1007/bf00881092, 1973.

Dalsøren, S. B., Myhre, C. L., Myhre, G., Gomez-Pelaez, A. J., Søvde, O. A., Isaksen, I. S. A., Weiss, R. F., and Harth, C. M.: Atmospheric methane evolution the last 40 years, Atmospheric Chemistry and

Atmospheric
Chemistry
and Physics
Discussions

Physics, 16, 3099-3126, doi:10.5194/acp-16-3099-2016, 2016.

710 Dentener, F., Peters, W., Krol, M., van Weele, M., Bergamaschi, P., and Lelieveld, J.: Interannual variability and trend of CH4 lifetime as a measure for OH changes in the 1979–1993 time period, Journal of Geophysical Research: Atmospheres, 108, 4442, 10.1029/2002jd002916, 2003.

Deushi, M. and Shibata, K.: Development of a Meteorological Research Institute chemistry-climate model version 2 for the study of tropospheric and stratospheric chemistry, Pap. Meteorol. Geophys., 62, 1–46, 2011.

715 EHHALT, D. H.: The atmospheric cycle of methane, Tellus, 26, 58-70, doi:10.1111/j.2153-3490.1974.tb01952.x, 1974.

Etminan, M., Myhre, G., Highwood, E. J., and Shine, K. P.: Radiative forcing of carbon dioxide, methane, and nitrous oxide: A significant revision of the methane radiative forcing, Geophysical Research Letters, 43, 12,614-612,623, doi:10.1002/2016GL071930, 2016.

720 Etiope, G.: Natural Gas Seepage: The Earth's Hydrocarbon Degassing, in, Springer International Publishing, Cham, 195 pp.,, 2015.

Garcia, R. R. and Boville, B. A.: Downward control of the meanmeridional circulation and temperature distribution of the polar winter stratosphere, J. Atmos. Sci., 51, 2238–2245, 1994.

Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G. J., Heil, A., Kaiser, 725 J. W., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F., Liousse, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M. G., Smith, S. J., Thompson, A., van Aardenne, J., van der Werf, G. R., and van Vuuren, D. P.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period, Climatic Change, 109, 163, doi:10.1007/s10584-011-0154-1, 2011.

730 Grewe, V., Brunner, D., Dameris, M., Grenfell, J. L., Hein, R., Shindell, D., and Staehelin, J.: Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes, Atmospheric Environment, 35, 3421-3433, https://doi.org/10.1016/S1352-2310(01)00134-0, 2001.

Guth, J., Josse, B., Marécal, V., Joly, M., and Hamer, P.: First implementation of secondary inorganic aerosols in the MOCAGE version R2.15.0 chemistry transport model, Geosci. Model Dev., 9, 137–160, 735 doi:10.5194/gmd-9-137-2016, 2016.

Hardiman, S. C., Butchart, N., O'Connor, F. M., and Rumbold, S. T.: The Met Office HadGEM3-ES chemistry–climate model: evaluation of stratospheric dynamics and its impact on ozone, Geosci. Model Dev., 10, 1209-1232, doi: 10.5194/gmd-10-1209-2017, 2017.

Hauglustaine, D. A., Hourdin, F., Jourdain, L., Filiberti, M.-A., Walters, S., Lamarque, J.-F., and Holland, 740 E. A.: Interactive chemistry in the Laboratoire de Météorologie Dynamique general circulation model: Description and background tropospheric chemistry evaluation, Journal of Geophysical Research: Atmospheres, 109, doi:10.1029/2003JD003957, 2004.

Holmes, C. D., Prather, M. J., Søvde, O. A., and Myhre, G.: Future methane, hydroxyl, and their uncertainties: key climate and emission parameters for future predictions, Atmospheric Chemistry and

Atmospheric
Chemistry
and Physics
Discussions
Open Access
EGU

745 Physics, 13, 285-302, 10.5194/acp-13-285-2013, 2013.

Hourdin, F., and Armengaud, A.: The Use of Finite-Volume Methods for Atmospheric Advection of Trace Species. Part I: Test of Various Formulations in a General Circulation Model, Monthly Weather Review, 127, 822-837, doi:10.1175/1520-0493(1999)127<0822:tuofvm>2.0.co;2, 1999.

Hourdin, F., Mauritsen, T., Gettelman, A., Golaz, J.-C., Balaji, V., Duan, Q., Folini, D., Ji, D., Klocke, D.,
750 Qian, Y., Rauser, F., Rio, C., Tomassini, L., Watanabe, M., and Williamson, D.: The Art and Science of Climate Model Tuning, Bulletin of the American Meteorological Society, 98, doi:589-602, 10.1175/bams-d-15-00135.1, 2017.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., Bergamaschi, P., Pagliari, V., Olivier, J. G. J., Peters, J. A. H. W., van Aardenne, J. A., Monni, S., Doering, U., and
755 Petrescu, A. M. R.: EDGAR v4.3.2 Global Atlas of the three major Greenhouse Gas Emissions for the period 1970-2012, Earth Syst. Sci. Data Discuss., 2017, 1-55, doi:10.5194/essd-2017-79, 2017.

Jourdain, L., Bekki, S., Lott, F., and Lefèvre, F.: The coupled chemistry-climate model LMDz-REPROBUS: description and evaluation of a transient simulation of the period 1980–1999, Ann. Geophys., 26, 1391-1413, doi: 10.5194/angeo-26-1391-2008, 2008.

760 Hegglin, M. I. and Lamarque, J.-F.: The IGAC/SPARC Chemistry-Climate Model Initiative Phase-1 (CCMI-1) model data output, NCAS British Atmospheric Data Centre, [ADD ACCESS DATE], available at:http://catalogue.ceda.ac.uk/uuid/ 9cc6b94df0f4469d8066d69b5df879d5, 2015.

Hunke, E. C. and Lipscombe, W. H.: CICE: the Los Alamos sea ice model documentation and software user's manual, Version 4.0, LA-CC-06-012, Los Alamos National Laboratory, New Mexico, 2008.

765 John, J. G., Fiore, A. M., Naik, V., Horowitz, L. W., and Dunne, J. P.: Climate versus emission drivers of methane lifetime against loss by tropospheric OH from 1860–2100, Atmospheric Chemistry and Physics, 12, 12021-12036, doi:10.5194/acp-12-12021-2012, 2012.

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), Geosci. Model Dev.,
770 3, 717–752, doi:10.5194/gmd-3-717-2010, 2010.

Josse, B., Simon, P., and Peuch, V.-H.: Radon global simulations with the multiscale chemistry and transport model MOCAGE, Tellus B, 56, 339–356, 2004.

Jöckel, P., Tost, H., Pozzer, A., Kunze, M., Kirner, O., Brenninkmeijer, C. A. M., Brinkop, S., Cai, D. S., Dyroff, C., Eckstein, J., Frank, F., Garny, H., Gottschaldt, K.-D., Graf, P., Grewe, V., Kerkweg, A., Kern,
775 B., Matthes, S., Mertens, M., Meul, S., Neumaier, M., Nützel, M., Oberländer-Hayn, S., Ruhnke, R., Runde, T., Sander, R., Scharffe, D., and Zahn, A.: Earth System Chemistry integrated Modelling (ESCiMo) with the Modular Earth Submodel System (MESSy) version 2.51, Geosci. Model Dev., 9, 1153–1200, doi:10.5194/gmd-9-1153-2016, 2016.

Jonsson, A. I., de Grandpré, J., Fomichev, V. I., McConnell, J. C., and Beagley, S. R.: Doubled CO2-
780 induced coolingin the middle atmosphere: Photochemical analysis of the ozone radiative feedback, J. Geophys. Res, 109, D24103, doi:10.1029/2004JD005093, 2004.

Krol, M., and Lelieveld, J.: Can the variability in tropospheric OH be deduced from measurements of 1,1,1-trichloroethane (methyl chloroform)?, Journal of Geophysical Research: Atmospheres, 108, doi:10.1029/2002JD002423, 2003.

785 Lamarque, J. F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017-7039, doi:10.5194/acp-10-7017-2010, 2010.

790 Lambert, G., and Schmidt, S.: Reevaluation of the oceanic flux of methane: Uncertainties and long term variations, Chemosphere, 26, 579-589, doi:https://doi.org/10.1016/0045-6535(93)90443-9, 1993.

Lawrence, M. G., Jöckel, P., and von Kuhlmann, R.: What does the global mean OH concentration tell us?, Atmos. Chem. Phys., 1, 37-49, 10.5194/acp-1-37-2001, 2001.

Lelieveld, J., and Dentener, F. J.: What controls tropospheric ozone?, Journal of Geophysical Research: 795 Atmospheres, 105, 3531-3551, doi:10.1029/1999JD901011, 2000.

Lelieveld, J., Dentener, F. J., Peters, W., and Krol, M. C.: On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere, Atmos. Chem. Phys., 4, 2337-2344, doi:10.5194/acp-4-2337-2004, 2004.

Lelieveld, J., Butler, T. M., Crowley, J. N., Dillon, T. J., Fischer, H., Ganzeveld, L., Harder, H., Lawrence, 800 M. G., Martinez, M., Taraborrelli, D., and Williams, J.: Atmospheric oxidation capacity sustained by a tropical forest, Nature, 452, 737-740, doi:10.1038/nature06870, 2008.

Lelieveld, J., Gromov, S., Pozzer, A., and Taraborrelli, D.: Global tropospheric hydroxyl distribution, budget and reactivity, Atmospheric Chemistry and Physics, 16, 12477-12493, doi: 10.5194/acp-16-12477-2016, 2016.

805 Levy, M. B., and Wadycki, W. J.: LIFETIME VERSUS ONE-YEAR MIGRATION IN VENEZUELA*, Journal of Regional Science, 12, 407-415, doi:10.1111/j.1467-9787.1972.tb00362.x, 1972.

Lin, X., Ciais, P., Bousquet, P., Ramonet, M., Yin, Y., Balkanski, Y., Cozic, A., Delmotte, M., Evangeliou, N., Indira, N. K., Locatelli, R., Peng, S., Piao, S., Saunois, M., Swathi, P. S., Wang, R., Yver-Kwok, C., Tiwari, Y. K., and Zhou, L.: Simulating CH_4 and CO_2 810 over South and East Asia using the zoomed chemistry transport model LMDz-INCA, Atmospheric Chemistry and Physics, 18, 9475-9497, doi:10.5194/acp-18-9475-2018, 2018.

Locatelli, R., Bousquet, P., Hourdin, F., Saunois, M., Cozic, A., Couvreux, F., Grandpeix, J. Y., Lefebvre, M. P., Rio, C., Bergamaschi, P., Chambers, S. D., Karstens, U., Kazan, V., van der Laan, S., Meijer, H. A. J., Moncrieff, J., Ramonet, M., Scheeren, H. A., Schlosser, C., Schmidt, M., Vermeulen, A., and Williams, 815 A. G.: Atmospheric transport and chemistry of trace gases in LMDz5B: evaluation and implications for inverse modelling, Geosci. Model Dev., 8, 129-150, 10.5194/gmd-8-129-2015, 2015.

Logan, J. A., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Tropospheric chemistry: A global perspective, Journal of Geophysical Research, 86, 7210, doi:10.1029/JC086iC08p07210, 1981.

Atmospheric
Chemistry
and Physics
Discussions

Lu, X., Hong, J., Zhang, L., Cooper, O. R., Schultz, M. G., Xu, X., Wang, T., Gao, M., Zhao, Y., and
820   Zhang, Y.: Severe Surface Ozone Pollution in China: A Global Perspective, Environmental Science &
      Technology Letters, 5, 487-494, doi:10.1021/acs.estlett.8b00366, 2018.

Madec, G.: NEMO ocean engine, Institut Piere-Simon Laplace (IPSL), France, No. 27, ISSN No. 1288-
      1619, 2008.

Marsh, D., Mills, M. J., Kinnison, D. E., Garcia, R. R., Lamarque, J.-F., and Calvo, N.: Climate change
825   from 1850–2005 simulated in CESM1 (WACCM), J. Climate, 26, 7372–7391, doi:10.1175/JCLI-D-12-
      00558.1, 2013.

Masui, T., Matsumoto, K., Hijioka, Y., Kinoshita, T., Nozawa, T., Ishiwatari, S., Kato, E., Shukla, P.,
      Yamagata, Y., and Kainuma, M.: An emission pathway for stabilization at 6 Wm^{-2} radiative forcing,
      Climatic change, 109, 59, 2011.

830   McNorton, J., Chipperfield, M. P., Gloor, M., Wilson, C., Feng, W., Hayman, G. D., Rigby, M., Krummel,
      P. B., amp, apos, Doherty, S., Prinn, R. G., Weiss, R. F., Young, D., Dlugokencky, E., and Montzka, S. A.:
      Role of OH variability in the stalling of the global atmospheric CH_4 growth rate
      from 1999 to 2006, Atmospheric Chemistry and Physics, 16, 7943-7956, 10.5194/acp-16-7943-2016,
      2016.

835   Miyazaki, K., Eskes, H. J., and Sudo, K.: Global NO_x emission estimates derived from an assimilation of
      OMI tropospheric NO_2 columns, Atmos. Chem. Phys., 12, 2263-2288, doi:10.5194/acp-12-2263-2012,
      2012.

Montzka, S. A., Krol, M., Dlugokencky, E., Hall, B., Jöckel, P., and Lelieveld, J.: Small Interannual
      Variability of Global Atmospheric Hydroxyl, Science, 331, 67-69, doi:10.1126/science.1197640, 2011.

840   Molod, A., Takacs, L., Suarez, M., Bacmeister, J., Song, I.-S., and Eichmann, A.: The GEOS-5
      Atmospheric General Circulation Model: Mean Climate and Development from MERRA to Fortuna,
      NASA Technical Report Series on Global Modeling and Data Assimilation, NASA TM-2012-104606, 28,
      117 pp., 2012.

Molod, A., Takacs, L., Suarez, M., and Bacmeister, J.: Development of the GEOS-5 atmospheric general
845   circulation model: evolution from MERRA to MERRA2, Geosci. Model Dev., 8, 1339–1356,
      doi:10.5194/gmd-8-1339-2015, 2015.

Morgenstern, O., Braesicke, P., O'Connor, F. M., Bushell, A. C., Johnson, C. E., Osprey, S. M., and Pyle,
      J. A.: Evaluation of the new UKCA climate-composition model – Part 1: The stratosphere, Geosci. Model
      Dev., 2, 43–57, doi:10.5194/gmd-2-43- 2009, 2009.

850   Morgenstern, O., Braesicke, P., O'Connor, F. M., Bushell, A. C., Johnson, C. E., Osprey, S. M., and Pyle,
      J. A.: Evaluation of the new UKCA climate-composition model – Part 1: The stratosphere, Geosci. Model
      Dev., 2, 43–57, doi:10.5194/gmd-2-43-2009, 2009.

Morgenstern, O., Hegglin, M. I., Rozanov, E., amp, apos, Connor, F. M., Abraham, N. L., Akiyoshi, H.,
      Archibald, A. T., Bekki, S., Butchart, N., Chipperfield, M. P., Deushi, M., Dhomse, S. S., Garcia, R. R.,
855   Hardiman, S. C., Horowitz, L. W., Jöckel, P., Josse, B., Kinnison, D., Lin, M., Mancini, E., Manyin, M.

Atmospheric
Chemistry
and Physics
Discussions

Open Access

E., Marchand, M., Marécal, V., Michou, M., Oman, L. D., Pitari, G., Plummer, D. A., Revell, L. E., Saint-Martin, D., Schofield, R., Stenke, A., Stone, K., Sudo, K., Tanaka, T. Y., Tilmes, S., Yamashita, Y., Yoshida, K., and Zeng, G.: Review of the global models used within phase 1 of the Chemistry–Climate Model Initiative (CCMI), Geoscientific Model Development, 10, 639-671, doi:10.5194/gmd-10-639-2017, 2017.

Murray, L. T., Mickley, L. J., Kaplan, J. O., Sofen, E. D., Pfeiffer, M., and Alexander, B.: Factors controlling variability in the oxidative capacity of the troposphere since the Last Glacial Maximum, Atmos. Chem. Phys., 14, 3589-3622, doi:10.5194/acp-14-3589-2014, 2014.

Naik, V., Voulgarakis, A., Fiore, A. M., Horowitz, L. W., Lamarque, J. F., Lin, M., Prather, M. J., Young, P. J., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R., Eyring, V., Faluvegi, G., Folberth, G. A., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., van Noije, T. P. C., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R., Shindell, D. T., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., and Zeng, G.: Preindustrial to present-day changes in tropospheric hydroxyl radical and methane lifetime from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmospheric Chemistry and Physics, 13, 5277-5298, doi:10.5194/acp-13-5277-2013, 2013.

Neu, J. L., Prather, M. J., and Penner, J. E.: Global atmospheric chemistry: Integrating over fractional cloud cover, Journal of Geophysical Research: Atmospheres, 112, D11306, doi:10.1029/2006JD008007, 2007.

Nielsen, J. E., Pawson, S., Molod, A., Auer, B., da Silva, A. M., Douglass, A. R., Duncan, B., Liang, Q., Manyin, M., and Oman, L. D.: Chemical mechanisms and their applications in the Goddard Earth Observing System (GEOS) earth system model, Journal of advances in modeling earth systems, 9, 3019-3044, 2017.

Nisbet, E. G., Manning, M. R., Dlugokencky, E. J., Fisher, R. E., Lowry, D., Michel, S. E., Myhre, C. L., Platt, S. M., Allen, G., Bousquet, P., Brownlow, R., Cain, M., France, J. L., Hermansen, O., Hossaini, R., Jones, A. E., Levin, I., Manning, A. C., Myhre, G., Pyle, J. A., Vaughn, B., Warwick, N. J., and White, J. W. C.: Very strong atmospheric methane growth in the four years 2014-2017: Implications for the Paris Agreement, Global Biogeochemical Cycles, 33, doi:10.1029/2018GB006009, 2019.

O'Connor, F. M., Johnson, C. E., Morgenstern, O., Abraham, N. L., Braesicke, P., Dalvi, M., Folberth, G. A., Sanderson, M. G., Telford, P. J., Voulgarakis, A., Young, P. J., Zeng, G., Collins, W. J., and Pyle, J. A.: Evaluation of the new UKCA climatecomposition model – Part 2: The Troposphere, Geosci. Model Dev., 7, 41–91, doi:10.5194/gmd-7-41-2014, 2014.

Oman, L. D., Ziemke, J. R., Douglass, A. R., Waugh, D. W., Lang, C., Rodriguez, J. M., and Nielsen, J. E.: The response of tropical tropospheric ozone to ENSO, Geophys. Res. Lett., 38, L13706, doi:10.1029/2011GL047865, 2011.

Oman, L. D., Douglass, A. R., Ziemke, J. R., Rodriguez, J. M., Waugh, D. W., and Nielsen, J. E.: The ozone response to ENSO in Aura satellite measurements and a chemistry-climate simulation, J. Geophys. Res., 118, 965–976, doi:10.1029/2012JD018546, 2013.

33

Patra, P. K., Houweling, S., Krol, M., Bousquet, P., Belikov, D., Bergmann, D., Bian, H., Cameron-Smith, P., Chipperfield, M. P., Corbin, K., Fortems-Cheiney, A., Fraser, A., Gloor, E., Hess, P., Ito, A., Kawa, S. R., Law, R. M., Loh, Z., Maksyutov, S., Meng, L., Palmer, P. I., Prinn, R. G., Rigby, M., Saito, R., and Wilson, C.: TransCom model simulations of CH4 and related species: linking transport, surface flux and chemical loss with CH4 variability in the troposphere and lower stratosphere, Atmospheric Chemistry and Physics, 11, 12813-12837, doi:10.5194/acp-11-12813-2011, 2011.

Patra, P. K., Krol, M. C., Montzka, S. A., Arnold, T., Atlas, E. L., Lintner, B. R., Stephens, B. B., Xiang, B., Elkins, J. W., Fraser, P. J., Ghosh, A., Hintsa, E. J., Hurst, D. F., Ishijima, K., Krummel, P. B., Miller, B. R., Miyazaki, K., Moore, F. L., Mühle, J., O'Doherty, S., Prinn, R. G., Steele, L. P., Takigawa, M., Wang, H. J., Weiss, R. F., Wofsy, S. C., and Young, D.: Observational evidence for interhemispheric hydroxyl-radical parity, Nature, 513, doi:219, 10.1038/nature13721, 2014.

Pison, I., Bousquet, P., Chevallier, F., Szopa, S., and Hauglustaine, D.: Multi-species inversion of CH4, CO and H2 emissions from surface measurements, Atmos. Chem. Phys., 9, 5281-5297, doi:10.5194/acp-9-5281-2009, 2009.

Poulter, B., Bousquet, P., Canadell, J. G., Ciais, P., Peregon, A., Saunois, M., Arora, V. K., Beerling, D. J., Brovkin, V., and Jones, C. D.: Global wetland contribution to 2000–2012 atmospheric methane growth rate dynamics, Environmental Research Letters, 12, 094013, 2017.

Prather, M. J., Holmes, C. D., and Hsu, J.: Reactive greenhouse gas scenarios: Systematic exploration of uncertainties and the role of atmospheric chemistry, Geophysical Research Letters, 39, L09803, doi:10.1029/2012GL051440, 2012.

Price, C., and Rind, D.: Modeling Global Lightning Distributions in a General Circulation Model, Monthly Weather Review, 122, 1930-1939, doi:10.1175/1520-0493(1994)122<1930:mgldia>2.0.co;2, 1994.

Prinn, R. G., Huang, J., Weiss, R. F., Cunnold, D. M., Fraser, P. J., Simmonds, P. G., McCulloch, A., Harth, C., Reimann, S., Salameh, P., O'Doherty, S., Wang, R. H. J., Porter, L. W., Miller, B. R., and Krummel, P. B.: Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, Geophysical Research Letters, 32, L07809, doi:10.1029/2004GL022228, 2005.

Randerson, J.T., G.R. van der Werf, L. Giglio, G.J. Collatz, and P.S. Kasibhatla. 2018. Global Fire Emissions Database, Version 4.1 (GFEDv4). ORNL DAAC, Oak Ridge, Tennessee, USA.doi: https://doi.org/10.3334/ORNLDAAC/1293

Revell, L. E., Tummon, F., Stenke, A., Sukhodolov, T., Coulon, A., Rozanov, E., Garny, H., Grewe, V., and Peter, T.: Drivers of the tropospheric ozone budget throughout the 21st century under the medium-high climate scenario RCP 6.0, Atmos. Chem. Phys., 15, 5887–5902, doi:10.5194/acp-15-5887-2015, 2015.

Revell, L. E., Stenke, A., Tummon, F., Feinberg, A., Rozanov, E., Peter, T., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Butchart, N., Deushi, M., Jöckel, P., Kinnison, D., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L. D., Pitari, G., Plummer, D. A., Schofield, R., Stone, K., Tilmes, S., Visioni,

930   D., Yamashita, Y., and Zeng, G.: Tropospheric ozone in CCMI models and Gaussian process emulation
       to understand biases in the SOCOLv3 chemistry–climate model, Atmos. Chem. Phys., 18, 16155-16172,
       doi:10.5194/acp-18-16155-2018, 2018.
       Riahi, K., Rao, S., Krey, V., Cho, C., Chirkov, V., Fischer, G., Kindermann, G., Nakicenovic, N., and
       Rafaj, P.: RCP 8.5—A scenario of comparatively high greenhouse gas emissions, Climatic Change, 109,
935   33-57, 10.1007/s10584-011-0149-y, 2011.
       Ridgwell, A. J., Marshall, S. J., and Gregson, K.: Consumption of atmospheric methane by soils: A
       process-based model, Global Biogeochemical Cycles, 13, 59-70, doi:10.1029/1998GB900004, 1999.
       Rigby, M., Prinn, R. G., Fraser, P. J., Simmonds, P. G., Langenfelds, R. L., Huang, J., Cunnold, D. M.,
       Steele, L. P., Krummel, P. B., Weiss, R. F., O'Doherty, S., Salameh, P. K., Wang, H. J., Harth, C. M.,
940   Mühle, J., and Porter, L. W.: Renewed growth of atmospheric methane, Geophysical Research Letters,
       35, 10.1029/2008gl036037, 2008.
       Rigby, M., Montzka, S. A., Prinn, R. G., White, J. W. C., Young, D., O'Doherty, S., Lunt, M. F., Ganesan,
       A. L., Manning, A. J., Simmonds, P. G., Salameh, P. K., Harth, C. M., Muhle, J., Weiss, R. F., Fraser, P.
       J., Steele, L. P., Krummel, P. B., McCulloch, A., and Park, S.: Role of atmospheric oxidation in recent
945   methane growth, Proc Natl Acad Sci U S A, 114, 5373-5377, doi:10.1073/pnas.1616426114, 2017.
       Sadourny, R., and Laval, K.: January and July performances of the LMD general circulation model, New
       Perspectives in Climate Modeling, In New Perspectives in Climate Modeling, A.L. Berger and C. Nicolis
       (eds.), Elsevier Press, Amsterdam, 173-197, 1984.
       Sander, R., Jöckel, P., Kirner, O., Kunert, A. T., Landgraf, J., and Pozzer, A.: The photolysis module
950   JVAL-14, compatible with the MESSy standard, and the JVal PreProcessor (JVPP), Geosci. Model Dev.,
       7, 2653-2662, doi:10.5194/gmd-7-2653-2014, 2014.
       Sander, S. P., Abbatt, J., Barker, J. R., Burkholder, J. B., Friedl, R. R., Golden, D. M., Huie, R., Kurylo,
       M. J., Moortgat, G. K., Orkin, V. L., and Wine, P. H.: Chemical kinetics and photochemical data for use
       in atmospheric studies evaluation number 17, Pasadena, CA: Jet Propulsion Laboratory, National
955   Aeronautics and Space Administration, 2011.
       Saunois, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J. G., Dlugokencky, E. J., Etiope,
       G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F. N., Castaldi, S., Jackson, R. B.,
       Alexe, M., Arora, V. K., Beerling, D. J., Bergamaschi, P., Blake, D. R., Brailsford, G., Brovkin, V.,
       Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Höglund-
960   Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H. S., Kleinen, T., Krummel, P., Lamarque, J. F.,
       Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K. C., Marshall, J., Melton, J. R.,
       Morino, I., Naik, V., O'Doherty, S., Parmentier, F. J. W., Patra, P. K., Peng, C., Peng, S., Peters, G. P.,
       Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W. J., Saito, M., Santini, M., Schroeder, R., Simpson,
       I. J., Spahni, R., Steele, P., Takizawa, A., Thornton, B. F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis,
965   A., van Weele, M., van der Werf, G. R., Weiss, R., Wiedinmyer, C., Wilton, D. J., Wiltshire, A., Worthy,
       D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z., and Zhu, Q.: The global methane budget 2000–

Atmospheric
Chemistry
and Physics
Discussions

2012, Earth Syst. Sci. Data, 8, 697-751, doi:10.5194/essd-8-697-2016, 2016.

Scinocca, J. F., McFarlane, N. A., Lazare, M., Li, J., and Plummer, D.: Technical Note: The CCCma third generation AGCM and its extension into the middle atmosphere, Atmos. Chem. Phys., 8, 7055–7074,
970   doi:10.5194/acp-8-7055-2008, 2008.

Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J. F., Kuhn, U., Stefani, P., and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmospheric Chemistry and Physics, 14, 9317-9341, doi:10.5194/acp-14-9317-2014, 2014.

975   Solomon, S., Kinnison, D. E., Bandoro, J., and Garcia, R.: Simulations of polar ozone depletion: an update, J. Geophys. Res., 120, 7958–7974, doi:doi:10.1002/2015JD023365, 2015.

Spivakovsky, C. M., Logan, J. A., Montzka, S. A., Balkanski, Y. J., Foreman-Fowler, M., Jones, D. B. A., Horowitz, L. W., Fusco, A. C., Brenninkmeijer, C. A. M., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, Journal of
980   Geophysical Research: Atmospheres, 105, 8931-8980, doi:10.1029/1999jd901006, 2000.

Stenke, A., Schraner, M., Rozanov, E., Egorova, T., Luo, B., and Peter, T.: The SOCOL version 3.0 chemistry-climate model: description, evaluation, and implications from an advanced transport algorithm, Geosci. Model Dev., 6, 1407–1427, doi:10.5194/gmd-6-1407-2013, 2013.

Staehelin, J., Tummon, F., Revell, L., Stenke, A., and Peter, T.: Tropospheric Ozone at Northern Mid-
985   Latitudes: Modeled and Measured Long-Term Changes, Atmosphere, 8, 163, 2017.

Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian, R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes, C., Hodnebrog, Ø., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund, M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J., Quennehen, B., Raut,
990   J. C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, Ø., Shine, K. P., Skeie, R. B., Wang, S., Yttri, K. E., and Zhu, T.: Evaluating the climate and air quality impacts of short-lived pollutants, Atmos. Chem. Phys., 15, 10529-10566, doi: 10.5194/acp-15-10529-2015, 2015.

Strode, S. A., Duncan, B. N., Yegorova, E. A., Kouatchou, J., Ziemke, J. R., and Douglass, A. R.: Implications of carbon monoxide bias for methane lifetime and atmospheric composition in chemistry
995   climate models, Atmos. Chem. Phys., 15, 11789-11805, doi: 10.5194/acp-15-11789-2015, 2015.

Szopa, S., Balkanski, Y., Schulz, M., Bekki, S., Cugnet, D., Fortems-Cheiney, A., Turquety, S., Cozic, A., Déandreis, C., Hauglustaine, D., Idelkadi, A., Lathière, J., Lefevre, F., Marchand, M., Vuolo, R., Yan, N., and Dufresne, J.-L.: Aerosol and ozone changes as forcing for climate evolution between 1850 and 2100, Climate Dynamics, 40, 2223-2250, doi:10.1007/s00382-012-1408-y, 2013.

1000   Telford, P. J., Abraham, N. L., Archibald, A. T., Braesicke, P., Dalvi, M., Morgenstern, O., O'Connor, F. M., Richards, N. A. D., and Pyle, J. A.: Implementation of the Fast-JX Photolysis scheme (v6.4) into the UKCA component of the MetUM chemistry-climate model (v7.3), Geosci. Model Dev., 6, 161-177, doi:10.5194/gmd-6-161-2013, 2013.

Atmospheric
Chemistry
and Physics
Discussions

Terrenoire, E., D. A. Hauglustaine, R. Valorso, and A. Cozic, Impact of present and future aircraft NOx
1005   and aerosol emissions on atmospheric composition and radiative forcing of climate, Atmos. Chem. Phys.
in preparation, 2019.

Teyssèdre, H., Michou, M., Clark, H. L., Josse, B., Karcher, F., Olivié, D., Peuch, V. H., Saint-Martin, D.,
Cariolle, D., Attié, J. L., Nédélec, P., Ricaud, P., Thouret, V., van der A, R. J., Volz-Thomas, A., and
Chéroux, F.: A new tropospheric and stratospheric Chemistry and Transport Model MOCAGE-Climat for
1010   multi-year studies: evaluation of the present-day climatology and sensitivity to surface processes, Atmos.
Chem. Phys., 7, 5815-5860, doi:10.5194/acp-7-5815-2007, 2007.

Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Ma, P.-L., Liu, X., Ghan, S., Bardeen, C.,
Arnold, S., Deeter, M., Vitt, F., Ryerson, T., Elkins, J.W., Moore, F., Spackman, J. R., and Val Martin, M.:
Description and evaluation of tropospheric chemistry and aerosols in the Community Earth System Model
1015   (CESM1.2), Geosci. Model Dev., 8, 1395–1426, doi:10.5194/gmd-8-1395-2015, 2015.

Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith, A. K.,
Neely, R. R., Conley, A., Vitt, F., Val Martin, M., Tanimoto, H., Simpson, I., Blake, D. R., and Blake,
N.: Representation of the Community Earth System Model (CESM1) CAM4-chem within the
Chemistry-Climate Model Initiative (CCMI), Geosci. Model Dev., 9, 1853-1890, doi:
1020   https://doi.org/10.5194/gmd-9-1853-2016, 2016.

Turner, A. J., Frankenberg, C., Wennberg, P. O., and Jacob, D. J.: Ambiguity in the causes for decadal
trends in atmospheric methane and hydroxyl, Proc Natl Acad Sci U S A, 114, 5367-5372,
doi:10.1073/pnas.1616020114, 2017.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S., and Arellano Jr, A. F.:
1025   Interannual variability in global biomass burning emissions from 1997 to 2004, Atmos. Chem. Phys., 6,
3423-3441, 10.5194/acp-6-3423-2006, 2006.

Vinken, G. C. M., Boersma, K. F., Maasakkers, J. D., Adon, M., and Martin, R. V.: Worldwide biogenic
soil NOx emissions inferred from OMI NO2 observations, Atmos. Chem. Phys., 14, 10363-10381, doi:
10.5194/acp-14-10363-2014, 2014.

1030   Voulgarakis, A., Naik, V., Lamarque, J. F., Shindell, D. T., Young, P. J., Prather, M. J., Wild, O., Field, R.
D., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring,
V., Faluvegi, G., Folberth, G. A., Horowitz, L. W., Josse, B., MacKenzie, I. A., Nagashima, T., Plummer,
D. A., Righi, M., Rumbold, S. T., Stevenson, D. S., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.:
Analysis of present day and future OH and methane lifetime in the ACCMIP simulations, Atmospheric
1035   Chemistry and Physics, 13, 2563-2587, 10.5194/acp-13-2563-2013, 2013.

Wang, Y., and Jacob, D. J.: Anthropogenic forcing on tropospheric ozone and OH since preindustrial
times, Journal of Geophysical Research: Atmospheres, 103, 31123-31135, doi:10.1029/1998JD100004,
1998.

Yienger, J. J., and Levy, H.: Empirical model of global soil-biogenic NOχ emissions, Journal of
1040   Geophysical Research: Atmospheres, 100, 11447-11464, doi:10.1029/95JD00370, 1995.

Atmospheric
Chemistry
and Physics
Discussions

Open Access

EGU

Yukimoto, S., Yoshimura, H., Hosaka, M., Sakami, T., Tsujino, H., Hirabara, M., Tanaka, T. Y., Deushi, M., Obata, A., Nakano, H., Adachi, Y., Shindo, E., Yabu, S., Ose, T., and Kitoh, A.: Meteorological Research Institute Earth System Model Version 1 (MRIESM1)

Walters, D. N., Williams, K. D., Boutle, I. A., Bushell, A. C., Edwards, J. M., Field, P. R., Lock, A. P.,
1045   Morcrette, C. J., Stratton, R. A., Wilkinson, J. M., Willett, M. R., Bellouin, N., Bodas- Salcedo, A., Brooks, M. E., Copsey, D., Earnshaw, P. D., Hardiman, S. C., Harris, C. M., Levine, R. C., MacLachlan, C., Manners, J. C., Martin, G. M., Milton, S. F., Palmer, M. D., Roberts, M. J., Rodrǵuez, J. M., Tennant, W. J., and Vidale, P. L.: The Met Office Unified Model Global Atmosphere 4.0 and JULES Global Land 4.0 configurations, Geosci. Model Dev., 7, 361–386, doi:10.5194/gmd-7-361-2014, 2014.
1050   Zimmerman, P. R., Chatfield, R. B., Fishman, J., Crutzen, P. J., and Hanst, P. L.: Estimates on the production of CO and H2 from the oxidation of hydrocarbon emissions from vegetation, Geophysical Research Letters, 5, 679-682, doi:10.1029/GL005i008p00679, 1978.

Zimmermann, P. H., Brenninkmeijer, C. A. M., Pozzer, A., Jȍckel, P., Zahn, A., Houweling, S., and Lelieveld, J.: Model simulations of atmospheric methane and their evaluation using AGAGE/NOAA
1055   surface- and IAGOS-CARIBIC aircraft observations, 1997–2014, Atmospheric Chemistry and Physics Discussions, doi: 1-45, 10.5194/acp-2017-1212, 2018.

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Atmospheric
Chemistry
and Physics
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Tables

1080 **Table 1.** List of CCMI models included in this study with model versions and references.[1]

Model	Version	References
CESM1-CAM-chem	CCMI_23	Tilmes et al.(2015, 2016)
CESM1-WACCM	CCMI_30	Solomon et al. (2015); Garcia et al. (2016); Marsh et al. (2013)
CMAM	v2.1	Jonsson et al. (2004); Scinocca et al. (2008)
EMAC(offers two resolutions: EMAC-L47MA and EMAC-L90MA)	v2.51	Jöckel et al. (2010, 2016)
GEOSCCM	v3	Molod et al. (2012, 2015); Oman et al. (2011, 2013); Nielsen et al. (2017)
HadGEM3-ES	HadGEM3 GA4.0, NEMO 3.4, CICE, UKCA, MetUM8.2	Walters et al.(2014); Madec(2008); Hunke and Lipscombe(2008); Morgenstern et al.(2009); O'Connor et al.(2004); Hardiman et al.(2017)
MOCAGE	v2.15.1	Josse et al. (2004); Guth et al. (2016)
MRI-ESM1r1	v1.1	Yukimoto et al. (2012, 2011); Deushi and Shibata (2011)
SOCOL3	v3	Revell et al. (2015); Stenke et al. (2013)
UMUKCA-UCAM	MetUM 7.3	Morgenstern et al. (2009); Bednarz et al. (2016)

[1] The table refers to Table 2 in Morgenstern et al. (2017)

Table 2. List of LMDz experiments and model setups.

	Simulation period	Inter annual variability in [OH]
Run_standard	2000-2010	2000-2010
Run_REF-C2	2011-2016	2010 apply inter-annual variability from REF-C2
Run_OH_inc	2011-2016	2010 apply annual growth rate of 1‰
Run_OH_dec	2011-2016	2010 apply annual decrease rate of 1%
Run_fix_OH	2000-2016	Constant OH (year 2000)

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Table 3. Inter-hemispheric ratios (N/S) of hemispheric mean OH and volume-weighted tropospheric mean [OH] for four latitude bands (in 10^5 molec.cm^{-3}) averaged over the years 2000 to 2010.

OH fields	N/S ratio	90 °S-30 °S (10^5 molec.cm^{-3})	30 °S-0 ° (10^5 molec.cm^{-3})	0 °-30 °N (10^5 molec.cm^{-3})	30 °N-90 °N (10^5 molec.cm^{-3})
TransCom	1.0	5.8	12.7	11.8	6.2
INCA NMHC-AER-S	1.3	4.7	10.6	12	7.5
INCA NMHC	1.2	5.7	11.9	13.4	7.8
CESM1-CAM4Chem	1.4	5.7	12.4	15.3	9.2
CESM1-WACCM	1.3	5.9	12.3	15.1	9.3
CMAM	1.2	5.6	13.1	14.3	8.3
EMAC-L47MA	1.2	6	13.5	15.6	8.4
EMAC-L90MA	1.2	6.3	13.8	15.7	8.6
GEOSCCM	1.2	6.5	14.8	16.8	9.1
HadGEM3-ES	1.4	4.1	10.4	12.5	8.1
MOCAGE	1.5	5.5	11.4	14.3	10.2
MRI-ESM1r1	1.2	4.7	13.7	15.3	7.3
SOCOL3	1.5	6.8	13.5	17.0	14.0
UMUKCA-UCAM	1.3	5.6	13.7	14.9	9.9

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Table 4. Global mean [OH] averaged over the troposphere and three vertical pressure levels (in 10^5 molec cm^{-3}) over the years 2000 to 2010.

	Tp-v[1]	Tp-m[2]	750[3]	500	250	Tp scaled[2]
TransCom	9.1	10.0	9.9	12.8	9.2	9.5
INCA NMHC-AER-S	8.7	9.4	11.3	10.4	7.8	9.3
INCA NMHC	9.7	10.4	11.8	11.4	8.9	9.7
CESM1-CAM4Chem	10.7	11.3	12.2	12.3	10.7	/
CESM1-WACCM	10.7	11.4	12.4	12.5	10.7	9.9
CMAM	10.4	11.3	14.3	11	10.5	9.3
EMAC-L47MA	10.9	11.3	12.1	12	10.3	/
EMAC-L90MA	11.1	11.5	12.5	12.2	10.2	10.3
GEOSCCM	11.8	12.3	12.3	13.7	12	10.4
HadGEM3-ES	8.8	9.9	12.7	10.8	7.7	/
MOCAGE	10.4	12.5	19	13.5	7.7	7.7
MRI-ESM1r1	10.3	10.6	12.2	10.4	9.4	10.2

SOCOL3	12.8	14.4	19.4	15.1	10.9	9.0
UMUKCA-UCAM	11.0	11.9	14.9	11.7	10.5	/

[1] Tp-v refers to the volume-weighted tropospheric mean [OH].

[2] Tp-m refers to the mass-weighted tropospheric mean [OH]

[3] 750 refers to the volume-weighted average from the surface to 750hPa, 500 refers to the volume-weighted average from 750hPa to 500 hPa, and 250 refers to the volume-weighted average from 500 to 250hPa.

[4] Tp scaled refer to the volume-weighted global tropospheric mean [OH] after scaling to the same CH_4 loss as with INCA NMHC in 2000.

Table 5. Global volume-weighted mean O^{1D} photolysis rate, specific humidity, CO and NO mixing ratios averaged over the whole troposphere and three pressure altitude levels for CCMI models over 2000 to 2010.[1]

	O^{1D} photolysis rate 10^5 s^{-1}				Specific humidity (10^{-3} g/kg)			
	750	500	250	Tp[2]	750	500	250	Tp
CESM1-CAM4Chem	0.9	1.3	1.5	1.3	11.7	4.2	0.9	4.7
CESM1-WACCM	1.0	1.3	1.6	1.3	11.6	4.1	0.9	4.7
CMAM	0.9	1.2	1.3	1.1	10.1	3.5	0.8	4
EMAC-L47MA	0.7	1	1.3	1.1	11.8	4.1	0.8	4.7
EMAC-L90MA	0.7	1	1.3	1.1	11.6	4.2	0.8	4.7
GEOSCCM	0.6	0.8	0.9	0.8	11.4	4.5	1.1	4.9
MOCAGE	/	/	/	/	11.0	3.7	0.8	4.3
MRI-ESM1r1	0.7	0.9	1.1	0.9	11.8	4.2	0.9	4.8
SOCOL3	0.7	0.8	0.9	0.8	11.6	4.6	1.0	4.8
	CO ppbv				**NO pptv**			
	750	500	250	Tp	750	500	250	Tp
CESM1-CAM4Chem	76	71	70	71	9	4	12	13
CESM1-WACCM	75	70	69	70	9	5	12	12
CMAM	77	68	64	69	17	4	17	26
EMAC-L47MA	85	77	70	75	8	4	11	14
EMAC-L90MA	84	76	69	74	8	5	11	17
GEOSCCM	78	74	73	74	9	5	13	13
MOCAGE	67	68	67	67	26	14	17	20
MRI-ESM1r1	93	86	83	86	10	5	20	32
SOCOL3	79	73	74	74	48	10	14	25

[1] HadGEM3-ES and UMUKCA-UCAM are not analyzed since model output has been regridded to too

coarse vertical pressure levels.

[2] Tp refers to the total troposheric average, 750 refers to the average from the surface to 750hPa, 500 refers to the average from 750hPa to 500hPa, and 250 refers to the average from 500hPa to 250hPa.

1110 **Table 6.** Global mean tropospheric CH_4 mixing ratios as simulated by LMDz using different OH fields and repeating year 2000 over 30 times.

CH_4 mixing ratio (ppbv)			
INCA NMHC-AER-S	1822	CESM1-WACCM	1575
TransCom	1776	**CMAM**	1540
INCA NMHC	1709	**GEOSCCM**	1503
MRI-ESM1r1	1693	**MOCAGE**	1275
EMAC-L90MA	1579	**SOCOL3**	1204

Table 7. LMDz simulated CH_4 mixing ratios (in ppbv) averaged over each latitudinal band and the years
1115 2000 to 2010 simulated from the standard experiment (Run_standard) using different OH fields.

	90 °S-60 °S	60 °S-0 °	0 °-60 °N	60 °N-90 °N	N/S gradient[1]
TransCom	1683	1697	1769	1812	129
INCA NMHC-AER-S	1687	1698	1757	1795	108
INCA NMHC	1687	1700	1762	1802	115
CESM1-WACCM	1688	1701	1757	1794	106
CMAM	1682	1694	1756	1796	114
EMAC-L90MA	1685	1698	1759	1798	113
GEOSCCM	1688	1701	1764	1803	115
MOCAGE	1686	1699	1753	1788	102
MRI-ESM1r1	1691	1702	1762	1803	112
SOCOL3	1694	1707	1754	1784	90

[1] N/S gradient is defined as the difference between 60 °N to 90 °N and 60 °S to 90 °S.
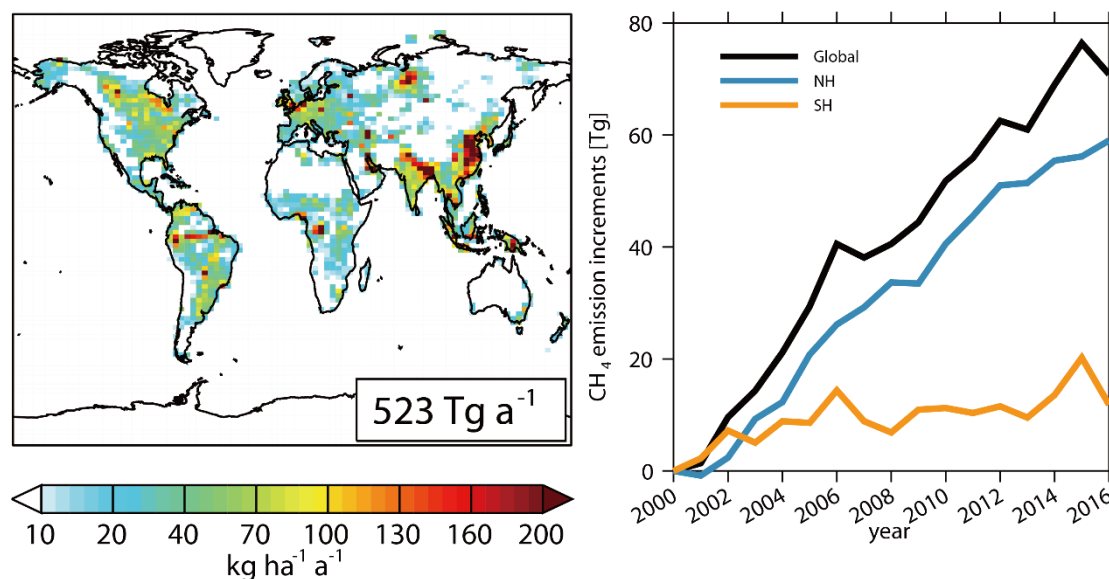
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Figure 1. Spatial distribution of global CH$_4$ emissions averaged between 2000 and 2016 (left) and a time series of CH$_4$ emissions relative to year 2000 emissions (482 Tg CH$_4$ a^{-1}) (right) for the globe (black line), Northern hemisphere (NH, blue line) and Southern hemisphere (SH, yellow line), respectively.
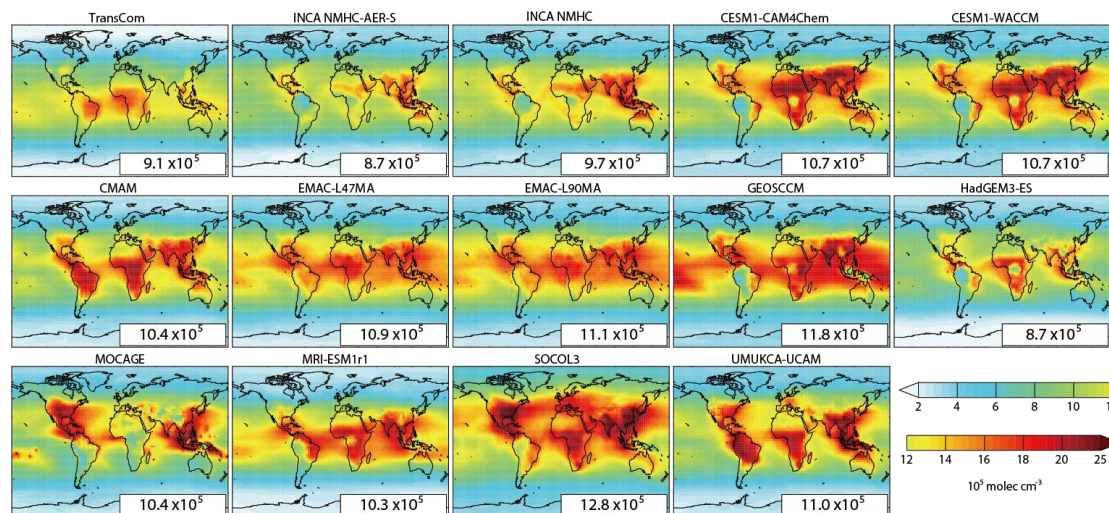

Figure 2. The spatial distributions of volume-weighted tropospheric mean OH fields of TransCom, INCA, and CCMI models averaged for 2000-2010. Global mean values (10^5 molec cm^{-3}) are shown as insets.
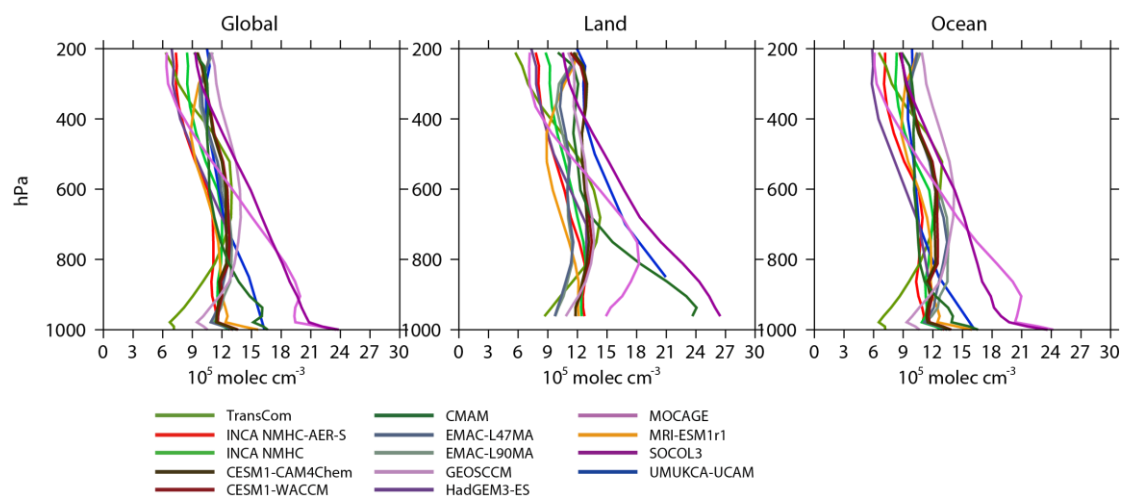
Figure 3. Vertical distributions of [OH] averaged over the globe (left), land (middle) and ocean (right) for 2000-2010.
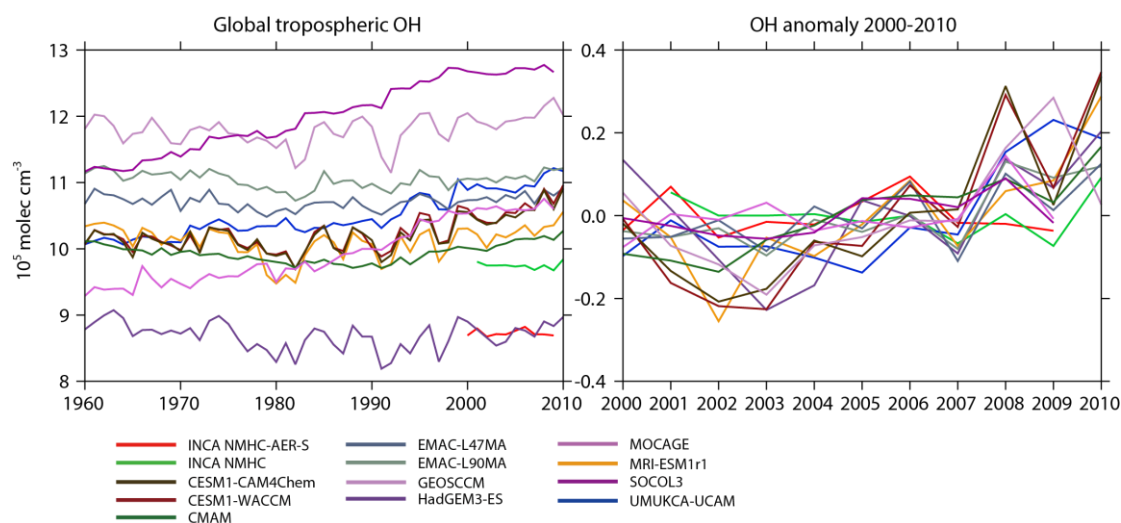
Figure 4. Left: Inter-annual variations of global volume-weighted tropospheric mean [OH] from CCMI and INCA model simulations from 1960 to 2010. Right: OH anomaly during 2000-2010, in reference to the mean concentration over the period 2000-2010 for each model.
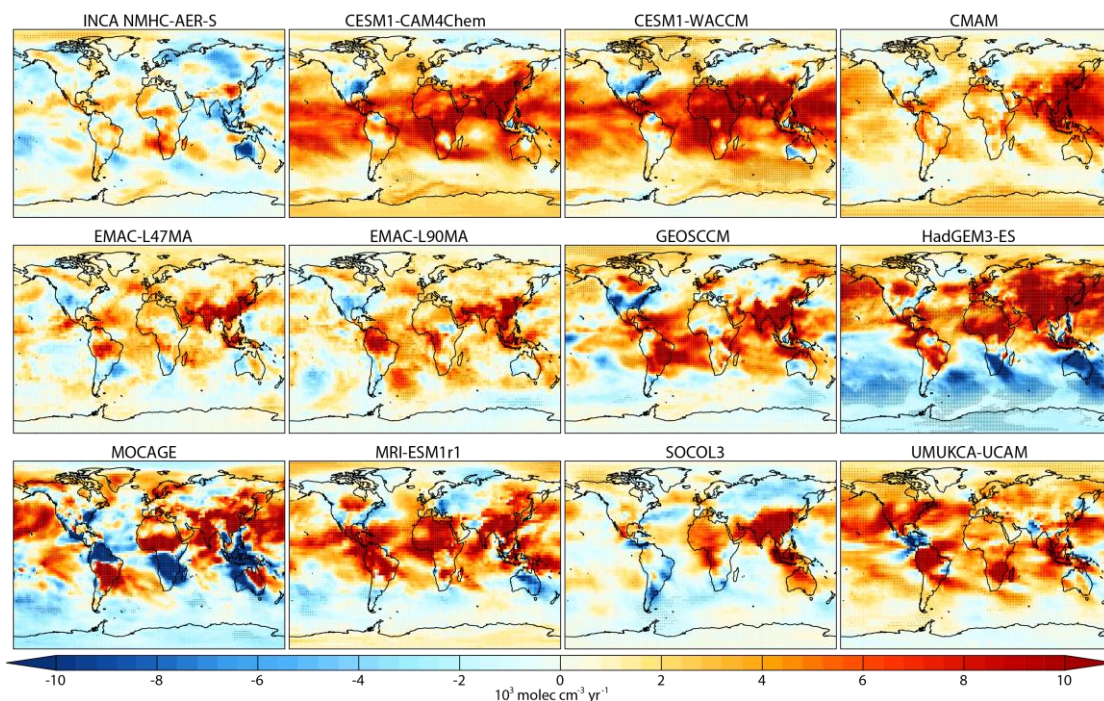
Figure 5. Spatial distribution of tropospheric OH trends from 2000 to 2010 (in 10^3 molec cm^{-3} yr^{-1}). Black dots denote model grid-cells with statistically significant trends (p-value < 0.05)
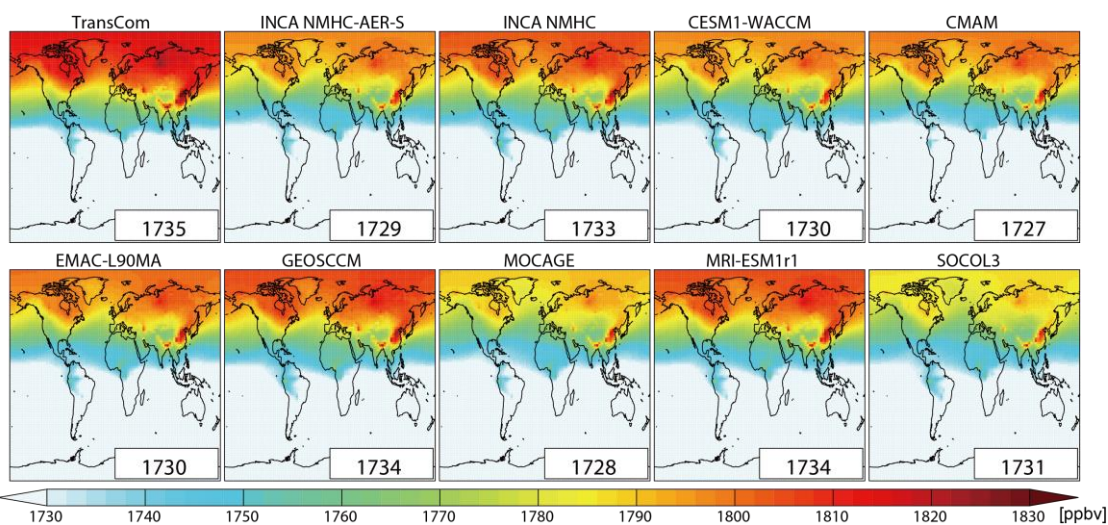

Figure 6. Spatial distribution of volume-weighted tropospheric mean CH_4 mixing ratios averaged from 2000 to 2010 as simulated by LMDz with different OH fields in the LMDz model. The global mean values in units of ppbv are shown as insets.
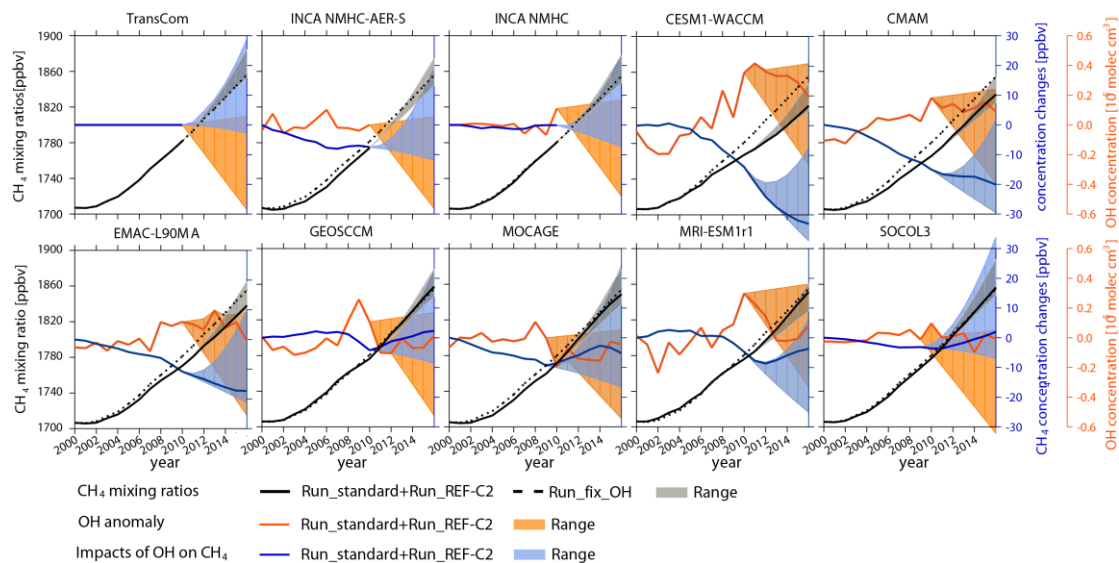
Figure 7. Time-series of global tropospheric CH_4 mixing ratios and [OH] associated with the model experiments listed in Table 2. The black lines represent the evolution of CH_4 mixing ratios with varying (solid lines) or with constant (dashed lines) OH. The varying OH case is obtained using OH inputs from Run_standard from 2000 to 2010 followed by Run_REF-C2 from 2011 to 2016 (see Table 2). The blue solid lines represent the corresponding differences between the simulations with varying OH and with constant OH. The orange solid line represents the corresponding anomalies in tropospheric [OH] (with the average over 2000-2010 as reference). The shaded areas correspond to the range obtained from all simulations over 2010-2016 (Table 2) for tropospheric CH_4 mixing ratios (grey), for changes in tropospheric CH_4 mixing ratios (blue) and for changes in tropospheric [OH] (orange).

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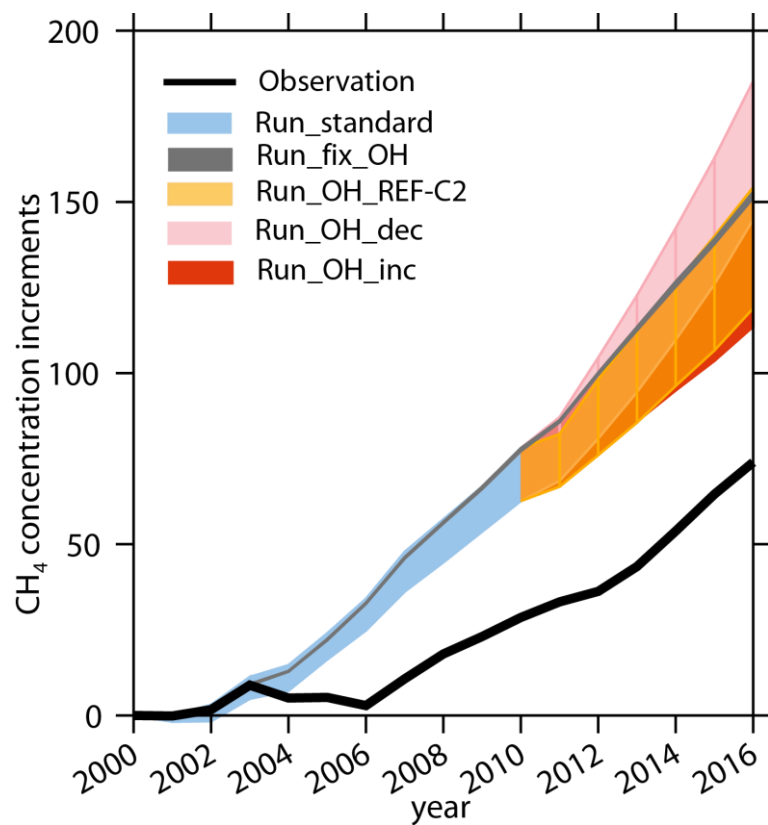
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Figure 8. Time series of surface CH_4 mixing ratio increments compared to 2000 for NOAA observations (black line) and model ranges from all the LMDz experiments collected at observation sites (shades) and described in the text and in Table 2.

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