



Tropospheric-stratospheric chemistry in the CNRM climate model ARPEGE-climat 7.0.1

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Abstract.

This study presents the evaluation of chemistry schemes implemented in ARPEGE-Climat 7.0.1, the latest version of the atmospheric model component of the CNRM climate model (CNRM-ESM) which will be used for future research. Three chemistry schemes of increasing complexity are considered: the stratospheric-upper-tropospheric REPROBUS scheme, and the stratospheric-tropospheric RELACS and RACMOBUS schemes. Atmosphere-only (AMIP) simulations (1979–2014) have been evaluated in terms of key chemical species against the previous generation of AMIP CNRM simulations (CNRM-ESM2-1), which ran with REPROBUS only, and against the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis of atmospheric composition in the troposphere. In the stratosphere, ARPEGE-Climat 7.0.1 provides improved total column ozone representation, reducing biases previously identified in CNRM-ESM2-1. Stratospheric water vapour is also better simulated due to increased moistening, though differences in methane oxidation between RELACS and RACMOBUS require further investigation. Additionally, issues remain with nitric acid in both RELACS and RACMOBUS, and for the RACMOBUS hydrogen chloride and nitrous oxide distributions. In the troposphere, RELACS and RACMOBUS show significant differences in carbon monoxide, with RACMOBUS simulating too low carbon monoxide throughout the entire troposphere, and in peroxy acetyl nitrate (PAN) with RELACS simulating among the highest PAN levels typically modelled. Nitrogen species concentrations are excessive at high northern latitudes in winter, partially due to missing nitrogen pentoxide heterogeneous loss processes. Tropospheric ozone is generally well represented, closely following reanalysis data. Overall, ARPEGE-Climat 7.0.1 marks significant progress in CNRM atmospheric chemistry modelling, supporting future climate-chemistry interaction studies.



1 Introduction

Atmospheric chemistry is highly interactive with other components of the Earth's system and plays a crucial role in climate change studies. The atmosphere's oxidizing capacity regulates the chemical loss rate, influencing the abundance and atmospheric lifetime of key greenhouse gases such as methane (CH_4 , Saunio et al., 2020) and nitrous oxide (N_2O , Tian et al., 2024). Understanding tropospheric chemical composition is particularly important, as secondary aerosols, both organic and inorganic, form from gaseous precursors and remain the largest source of uncertainty in the Earth's radiative budget. Additionally, gaseous species such as O_3 and nitrogen oxides (NO_x , $\text{NO} + \text{NO}_2$), along with aerosols, contribute to poor air quality and adverse health effects (Manisalidis et al., 2020). Atmospheric chemistry, air quality, and climate change are also closely interconnected (von Schneidemesser et al., 2015). Finally, atmospheric chemistry interacts with vegetation, as, for example, O_3 deposition can reduce photosynthetic carbon assimilation (Sicard et al., 2017).

For all the reasons mentioned above, the importance of representing atmospheric chemistry in climate models has long been recognized (Hameed et al., 1980). This recognition has led to numerous initiatives aimed at addressing chemistry-climate interactions. One of the first coordinated efforts was the CCMVal-1 activity, which evaluated the ability of Chemistry-Climate Models (CCMs) to simulate key processes influencing the distribution of stratospheric ozone (Eyring et al., 2006). This effort contributed to the 2006 WMO/UNEP Scientific Assessment of Ozone Depletion. A second phase, CCMVal-2, established a more structured and coordinated approach, incorporating common external forcings, recommendations for chemical kinetics, and detailed diagnostic requirements (Morgenstern et al., 2010a). This phase resulted in a series of publications among which a very detailed report (Eyring V. and Waugh, 2010), and significantly improved the representation of stratospheric chemistry. However, tropospheric chemistry remained largely absent or highly simplified in most of the participating models.

A follow-up to the CCMVal-2 activity is the Chemistry-Climate Model Initiative (CCMI), aimed at improving our understanding of the links between atmospheric composition and climate. The first phase, CCMI-1, involved 20 models, most of which explicitly represented tropospheric chemistry with varying levels of complexity (Morgenstern et al., 2017a). CCMI continues to support ozone assessments and serves as a complement to the Coupled Model Intercomparison Project (CMIP, Eyring et al., 2016). Within CMIP, the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013) was the first coordinated effort to support Assessment Reports, complementing CMIP5 simulations with time slice experiments. For CMIP6, the AerChemMIP project (Collins et al., 2017) was endorsed to quantify the climate and air quality impacts of aerosols and chemically reactive gases. AerChemMIP involved various global atmosphere-ocean coupled climate models, with different modelling groups contributing different approaches. For example, Griffiths et al. (2021a) analysed tropospheric ozone using five models, while Keeble et al. (2021) evaluated stratospheric ozone and water vapour across 22 models, six with full stratospheric chemistry, three with simplified stratospheric schemes, and 13 using prescribed CMIP6 ozone fields. Regarding tropospheric chemistry, Young et al. (2018) provide a comprehensive review of the development of global models, analysing ozone simulations from their participation in the most recent multi-model assessments, the eighth since 2000.

Turning to the future, Sanderson et al. (2023) argue for the need of more interactive Earth System Models (ESMs) to allow emissions driven simulations instead of concentration driven ones, thus better addressing the overall uncertainties in



climate scenarios. The UKESM is an example of such models that took the interactivity a step further than interactive full atmospheric chemistry with climate. After merging of previously developed stratospheric (Morgenstern et al., 2009) and tropospheric (O'Connor et al., 2014b) schemes (see Archibald et al., 2020b), the model has been further developed coupling the chemistry scheme with interactive emissions from wetland and dry deposition of CH₄ to study its global CH₄ cycle by performing CH₄ emission driven simulations (Folberth et al., 2022).

Chemistry and climate modeling at CNRM has contributed to several coordinated model evaluations, including CCMVal-2 (e.g., Strahan et al. (2011); Austin et al. (2010); Morgenstern et al. (2010b)), CCMI (e.g., Morgenstern et al. (2017a); Maycock et al. (2018a); Dhomse et al. (2018)), ACCMIP (e.g., Lamarque et al. (2013); Naik et al. (2013); Young et al. (2013)), and CMIP6 (e.g., Smith et al. (2020); Morgenstern et al. (2022); Skeie et al. (2020); Keeble et al. (2021)). These contributions have involved two types of models: a Chemistry Transport Model, incorporating either stratospheric or full-atmosphere chemistry, and an atmospheric climate model with "online" stratospheric chemistry.

The logical next step at CNRM was to implement a comprehensive representation of atmospheric chemistry throughout the entire atmosphere in the latest version of the atmospheric climate model, ensuring its applicability for future studies. Beyond documenting and evaluating these developments, this paper investigates the benefits of increasing complexity in chemical schemes for the representation of atmospheric composition in climate models. The paper is structured as follows: in addition to the Introduction and Conclusion sections, Section 2 presents the new ARPEGE-Climat model along with its chemistry schemes and the simulation setup used for evaluation. Section 3 then details and discusses the results obtained.

2 Methodology

2.1 The ARPEGE-Climat global atmospheric model

The present study has been conducted with a new version of the ARPEGE-Climat global atmosphere model which is the atmospheric component of the Centre National de Recherches Météorologiques (CNRM) climate model. This new version, referred to as 7.0.1, is an update of version 6.3 (Roehrig et al., 2020) which was used for CMIP6. Version 7.0.1 is based on cycle 48t1_op1 of the ARPEGE/IFS (Integrated Forecast System), developed jointly by Météo-France and the European Center for Medium-range Weather Forecasts (ECMWF) for both Numerical Weather Prediction (NWP) and climate applications. This version is aimed, among other applications, to serve as a basis for CMIP7 simulations, and has first been used in Drugé et al. (2024).

As other atmospheric models, ARPEGE-Climat consists in a dry dynamical core and a suite of physical parameterisations that represent diabatic processes. In short, ARPEGE-Climat 7.0.1 very largely shares the choices made for the ARPEGE NWP version which means that these choices differ from those of the ARPEGE-Climat CMIP6 version. This is the case in particular for the parameterisations of the deep convection and of the radiation. Specifically, the convection scheme is based on the work of Tiedtke (1989), with many subsequent modifications (as reported in Bechtold et al., 2008, 2014; Becker et al., 2021). The radiation scheme used is "ecRad" (Hogan and Bozzo, 2018), which has been operational in the IFS NWP model since 2017.



In our climate simulations, the atmospheric dynamics and physics are computed on a T127 triangular grid truncation or
85 associated reduced Gaussian grid, which corresponds to a spatial resolution of approximately 150 km in both longitude and
latitude. ARPEGE-Climat is a 'high-top' model with 91 vertical levels, extending from the surface to 0.01 hPa in the meso-
sphere. These levels use hybrid σ -pressure coordinates, with 15 levels below 1500 m. To simulate surface state variables
and fluxes, ARPEGE-Climat uses the SURFace EXternalisée (SURFEX) modelling platform, in its version 8.1 here which is
present in cycle 48t1 of the ARPEGE/IFS system (Le Moigne et al., 2020). This platform operates over the same grid and
90 with the same time step as the rest of the model. Physical processes at the land surface are represented using the Interaction
Soil-Biosphere-Atmosphere (ISBA) land surface model (Noilhan and Mahfouf, 1996). The chemistry schemes described in
the next section are one of the "physical" processes of the model, they are in operations at each physical time step (i.e., 900 s),
which is also the dynamics time step.

2.2 The chemistry schemes

95 A chemical scheme describing the chemistry of the upper troposphere and stratosphere, known as REPROBUS-C_v2 (or
simply REPROBUS), was first implemented as an "on-line" scheme in ARPEGE-Climat and evaluated in Michou et al. (2011).
This model version contributed to the CCMI initiative (e.g., Maycock et al., 2018b; Morgenstern et al., 2017a; Wales et al.,
2018; Ayarzagüena et al., 2018) before being integrated into the CNRM-ESM2-1 Earth System Model (Michou et al., 2020b),
where it contributed to the AerChemMIP simulations of CMIP6 (see references in the introduction). More recently, we have
100 ported REPROBUS into ARPEGE-Climat 7.0.1. However, REPROBUS does not account for non-methane ozone chemistry
in the lower troposphere, and it is important to note that chemical evolutions are computed down to 560 hPa only (for details
see Morgenstern et al., 2017a; Michou et al., 2020a). To address this limitation, two additional chemical schemes have been
introduced in ARPEGE-Climat 7.0.1.

The first implemented tropospheric chemical scheme is RACM (Regional Atmospheric Chemistry Mechanism, Stockwell
105 et al., 1997). It is designed to represent atmospheric composition across a wide range of conditions, from remote to polluted
environments, and from the surface to the upper troposphere. This mechanism includes both inorganic and organic chemistry,
with organic species grouped based on their reactivity with the hydroxyl radical (OH). To enable comprehensive atmospheric
composition modelling, RACM and REPROBUS have been combined into a single scheme called RACMOBUS. This scheme
represents a total of 115 gaseous species, evolving through 381 chemical reactions, including 53 photolysis reactions. This
110 chemical scheme has been implemented in various atmospheric chemistry models. For example, RACMOBUS is the chemical
mechanism used in the Chemistry-Transport Model MOCAGE (*MOdèle de Chimie Atmosphérique à Grande Échelle*, Cussac
et al., 2020), which contributed to the Chemistry and Climate Model Initiative (CCMI, Morgenstern et al., 2017b). Additionally,
RACMOBUS is integrated into ECMWF's on-line atmospheric composition model. Specifically, in this configuration named
IFS (MOCAGE), it has been developed and evaluated at both the global scale in the troposphere (Huijnen et al., 2010) and at
115 a regional scale, focusing on the lower troposphere and air quality (Williams et al., 2022).

The second implemented tropospheric chemical scheme is ReLACS (Regional Lumped Atmospheric Chemical Scheme,
Crassier et al., 2000). Derived from RACM, ReLACS aims to reduce computational cost while maintaining strong scientific



performance. This reduction is achieved through a reactivity-weighted approach, aggregating organic compounds into a smaller number of lumped classes than RACM. Similar to RACM, ReLACS is combined with REPROBUS in a merged scheme referred to as RELACS (note the difference in the capitalization of the first letter "e"). It includes 87 chemical species that evolve through 247 chemical reactions, of which 45 are photolysis reactions. The RELACS chemical scheme had been integrated into the MOCAGE CTM. Its implementation was evaluated in the work of Teyss  dre et al. (2007) where the scheme was found to perform satisfactorily overall, with detailed analysis of biases against observations.

Although the three chemistry schemes share many similarities, they also differ in several aspects, including their complexity, their approach to mass conservation, such as the specific treatment in RACMOBUS that distinguishes the stratosphere from the troposphere, and the set of constants involved in chemical reactions and the physical evolution of chemical species. These specificities have been adjusted in previous studies. Here, we present an analysis of the current state of these schemes and their implementation.

2.3 Physical evolutions of chemical species

Large-scale transport of chemical species, treated as prognostic variables, is managed by the ARPEGE-Climat semi-Lagrangian scheme, which is also used to simulate the advection of humidity and temperature (see Roehrig et al., 2020). Since semi-Lagrangian schemes are not fully mass-conservative, a local mass fixer algorithm based on Bermejo and Conde (2002) is applied to all transported species. At subgrid scales, the model's turbulence and convection schemes (see Section 2.1) handle the transport of chemical species.

Wet deposition, which removes atmospheric compounds through precipitation, is calculated by the scheme described in Flemming et al. (2015b). This approach is an update of the earlier Harvard scheme (Liu et al., 2001) in which scavenging flux calculations in convective updrafts have been decoupled from the convection scheme. The wet deposition process involves in-cloud scavenging, where species are removed during the formation of rain or snow droplets, and below-cloud scavenging, where falling droplets collect gaseous species. Additionally, precipitation re-evaporation can release previously scavenged compounds back into the atmosphere when conditions allow. The efficiency of wet deposition depends on the solubility of each species, determined using Henry's law equilibrium. In ARPEGE-Climat, effective Henry's law coefficients are calculated for a droplet pH of 5, following the values provided in Sander (2023). The total wet deposition flux, accounting for rain-out, washout, and evaporation, is computed separately for convective and large-scale precipitation to properly consider differences in precipitation within a grid cell and variations in droplet fall speed.

Dry deposition refers to the removal of gaseous compounds from the atmosphere in the absence of precipitation, through interactions with the surface. In ARPEGE-Climat, dry deposition fluxes are calculated by the SURFEX surface model (Le Moigne et al., 2020) before being applied to atmospheric concentrations. For a given species, the dry deposition flux is determined by the following equation: $F = -v_d c$, where F represents the dry deposition flux, v_d is the deposition velocity, and c is the concentration of the chemical species near the surface. v_d is estimated, as commonly done in models (e.g., Michou et al. (2005); Flemming et al. (2015a)), as the sum of three resistance terms: $v_d = \frac{1}{R_a + R_b + R_c}$. The aerodynamic resistance (R_a) accounts for the vertical transfer of species in the air layer near the surface and depends only on dynamic quantities. It is estimated following



Slinn and Slinn (1980). The resistance of the quasi-laminar layer (R_b) is calculated classically as detailed for instance in Michou and Peuch (2002). The surface resistance (R_c) represents the absorption resistance of vegetation or soil and is calculated following Wesely (1989). It accounts for multiple components, including vegetation resistance, lower canopy resistance, and ground resistance. We modified the surface resistance over water from Seinfeld and Pandis (2006), which produced unsatisfactory deposition velocities, not only for O_3 , where the surface resistance was an order of magnitude too high, but also for many Volatile Organic Compounds (VOCs), where it was conversely too low. These adjustments were made to better align with the limited available observations of deposition velocities, as detailed in the thesis report of Aguirre Martine (2004).

2.4 Lightning NO_x emissions

In ARPEGE-Climat, NO_x emissions from lightning are calculated following Meijer et al. (2001), using a linear function of the flash rate. The flash rate itself is estimated as a linear function of convective precipitation, with a conversion rate of 13,700 flashes per meter. Over oceans, emissions are set to one-tenth of their continental counterparts, following Huijnen et al. (2010) and Levy II et al. (1996). The conversion factor from flash energy to NO_x emissions is taken as 10^{17} molecules per Joule, a widely accepted coefficient in the previously mentioned literature. The energy per flash (E) is assumed to be the same for both cloud-to-ground (CG) and intra-cloud (IC) flashes, with $E_{CG} = E_{IC} = 6.7 \times 10^8 J$. The fraction of CG flashes relative to total flashes is determined using the fourth-order polynomial function of cold cloud thickness from Price and Rind (1993). Lightning NO_x emissions are then distributed vertically following the approach of Pickering et al. (1998), where all IC lightning NO_x is released between the $T = -15^\circ C$ level and the cloud top, while all CG lightning NO_x is distributed between the surface and $T = -15^\circ C$ level. Finally, a calibration coefficient is applied to the emission flux to ensure that the total annual amount of nitrogen emitted remain consistent with an expected yearly amount. According to Verma et al. (2021), despite advancements in observational and modelling techniques, estimates of global lightning NO_x emissions remain within a wide range of $5 \pm 3 \text{ Tg(N).yr}^{-1}$. In our simulations these emissions have been set to $2.5 \text{ Tg(N).yr}^{-1}$ after various sensitivity tests. The 5 Tg(N).yr^{-1} case is being presented and discussed in section 3.3.1. The horizontal and vertical distributions of the resulting annual average of lightning NO_x emissions in our simulations is shown in Figure A1. Since emissions are scaled to match a predefined total nitrogen release into the atmosphere, the spatial distribution is expected to remain largely unchanged from one year to the next.

2.5 Simulation description

We conducted with ARPEGE-Climat 7.0.1 three atmosphere-only (AMIP) simulations covering the period 1979–2014, using respectively the three chemistry schemes described in Section 2.2, with initial state from the CNRM-ESM2-1 AMIP simulation (see Section 2.6). Additionally, we performed simulations to assess the sensitivity of results to the magnitude of lightning NO_x emissions (see Section 3.3.1 and to the level of relaxation applied in the troposphere for long-lived gases (see Section 3.3.2) and summary in Table 1). In all simulations, the 3D concentrations of various trace gases interact with the atmospheric radiative code at each call of the radiative scheme (every hour) for both the longwave and shortwave parts of the spectrum see (for details, see Michou et al., 2020a). These simulations employed CMIP6 forcings, including greenhouse gases (GHG) and



Table 1. Characteristics of the simulations analysed, including the simulation name, model name, chemistry scheme, simulation type (AMIP for this analysis, or "sensi." for a sensitivity test), and the period covered (see text for details).

Name	Model	Chem. scheme	Type	Period
ESM2-REPRO	CNRM-ESM2-1	REPROBUS-C_v2	CMIP6 AMIP	1979-2014
ARP7-REPRO	ARPEGE-Climat 7.0.1	REPROBUS-C_v2	CMIP6 AMIP	1979-2014
ARP7-RELACS	ARPEGE-Climat 7.0.1	RELACS	CMIP6 AMIP	1979-2014
ARP7-RACM	ARPEGE-Climat 7.0.1	RACMOBUS	CMIP6 AMIP	1979-2014
ARP7-RACM5Tg	ARPEGE-Climat 7.0.1	RACMOBUS, 5Tg lightning	sensi.	1979-2014
ARP7-RELACS5Tg	ARPEGE-Climat 7.0.1	RELACS, 5Tg lightning	sensi.	1979-2014
ARP7-RELACS_relaniv	ARPEGE-Climat 7.0.1	RELACS	sensi.	1979-1988 (or less years)

185 ozone-depleting substance (ODS) concentrations following Meinshausen et al. (2017), volcanic aerosols based on Thomason et al. (2018), solar forcing from Matthes et al. (2017), and sea surface temperatures and sea ice as indicated in Eyring et al. (2016).

All emissions, except for lightning-generated NO_x , were accounted for using emission inventories, CMIP6 datasets for anthropogenic and biomass burning sources, along with other datasets for additional sources as specified below. Anthropogenic
190 emissions were taken from the Community Emissions Data System (CEDS, Hoesly et al., 2018), including emissions of CO , CH_4 , NH_3 , NO_x , SO_2 , and Non Methane VOCs (NMVOCs). This dataset, covering 1750–2014, was developed by integrating country- and regional-scale inventories with energy consumption data. For biomass burning emissions, we used the BB4CMIP CMIP6 inventory (van Marle et al., 2017). It spans the same period as CEDS (1750–2014) and reconstructs emissions for earlier years using a combination of satellite observations (1997–2014), fire proxies, and fire models. Biogenic NMVOC emissions
195 from plants were included using the CAMS-GLOB-BIO v3.1 dataset (Sindelarova et al., 2022). This dataset, produced using the Model of Emission of Gases and Aerosols from Nature (MEGANv2.1, Guenther et al., 2012), is driven by a meteorological reanalysis. Since it only covers 2000–2019, we created a monthly climatology by averaging the monthly emission fluxes, which we applied throughout our simulation period. Other natural sources of reactive gases were also considered. Soil NO_x emissions were based on the CAMS-GLOB-SOIL v2.3 inventory (Simpson and Darras, 2021), while oceanic CO emissions followed the
200 RETRO inventory (Lana et al., 2011). Similarly, oceanic emissions of dimethyl sulfide (DMS) were taken from the CAMS-GLOB-OCE v3.1 dataset (Schultz et al., 2008). For details on total emissions, we refer to the original publications cited above. Additionally, to provide context on the magnitude of emissions in our simulations, Table 2 presents the 2005 annual totals for NO_x , CO and NMVOC emissions considered in the RELACS and RACMOBUS chemical schemes.

2.6 Reference data sets

205 We base our analysis on the following reasoning. For the troposphere, we rely on an atmospheric composition reanalysis, which is described in detail in Section 2.6.1. In the stratosphere, we assume that the validity of the REPROBUS scheme has



Table 2. Emissions accounted for in the RELACS and RACM chemical schemes, with annual totals for the year 2005. For NO + NO₂: Tg(N) yr⁻¹, for CO: Tg(CO) yr⁻¹, and for all NMVOCs: Tg(C) yr⁻¹. See text details on the chemical schemes.

RELACS spec.	N. elt (N or C)	Total	RACM spec.	N. elt (N or C)	Total
NO + NO ₂	1	47	NO + NO ₂	1	47
CO	1	1042	CO	1	1042
ISOTOT	5	433	C ₅ H ₈	5	392
			API	10	84
ALKANEbis	4	133.9	HC ₃	2.9	94.9
			HC ₅	4.8	29.1
			HC ₈	7.9	4.21
ALKENEbis	3.7	47.5	OLT	3.8	23.0
			DIEN	4	10.8
			C ₂ H ₄	2	28.54
ALD	2.4	15.9	ALD	2.4	15.9
AROMATIC	7.58	20.9	TOL	7.1	11.0
			XYL	8.9	12.2
ETH	2	7.8	C ₂ H ₆	2	7.8
HCHO	1	4.5	HCHO	1	4.5
KET	3.5	11.1	KET	3.5	11.1
-			CH ₃ COCHO	3	1.7

already been established, along with its strengths and weaknesses, particularly in recent multi-model climate comparisons (e.g., CMIP6). Therefore, our "reference" dataset is the CNRM-ESM2-1 CMIP6 AMIP simulation (see 2.6.2).

2.6.1 The CAMS EAC4 reanalysis

210 Several chemical reanalysis products of varying interest have recently become available (Sekiya et al., 2025). For this study, we selected the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis, namely the EAC4 (ECMWF Atmospheric Composition Reanalysis 4) product. Indeed it covers a period relevant to climate evaluation, regularly updated for near-real time periods (2003-2024), and has shown satisfactory agreement with in situ observations (see Sekiya et al. (2025)). Further evaluation details can be found in Wang et al. (2020), including vertical validation against aircraft campaigns, as well as in Wagner et al. (2021) for O₃, CO, and NO₂, and in Williams et al. (2022) for NO_x, HNO₃, and PAN (Peroxy Acetyl Nitrate). The most comprehensive description of EAC4 is provided in Inness et al. (2019b), but we summarize its key characteristics here. The assimilation system is based on the operational 4D-Var system from IFS cy42r1, running on a T255 grid (80 km resolution). The dynamical system uses the same input observations as the ERA5 reanalysis. The relevant assimilated trace gas retrievals include seven different products for O₃ (total columns, profiles, and partial columns from SCIAMACHY, MIPAS, MLS, OMI,

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220 GOME-2, and SBUV/2), as well as the CO total column retrieval from MOPITT and three different tropospheric column re-
trievals (from SCIAMACHY, OMI, and GOME-2) for NO₂. Tropospheric chemistry is modelled using the IFS(CB05) chemical
scheme (Flemming et al., 2015b), while stratospheric chemistry follows the linear scheme of Cariolle and Teyss  re (2007),
a simpler approach compared to REPROBUS. This makes EAC4 particularly relevant for evaluating tropospheric chemical
composition and justifies the use of the CNRM-ESM2-1 AMIP simulation for the stratosphere. Anthropogenic emissions are
225 prescribed from the MACCity inventory (Granier et al., 2011), biomass burning emissions follow the GFASv1.2 inventory
(Kaiser et al., 2012), and biogenic emissions are based on the MEGAN-MACC dataset (Sindelarova et al., 2022). It is impor-
tant to note that these inventories differ from those used in our simulations, which should be considered when interpreting the
evaluation results. Throughout this paper, we refer to the EAC4 reanalysis.

2.6.2 The CNRM-ESM2-1 CMIP6 AMIP simulation

230 The CNRM-ESM2-1 CMIP6 AMIP simulation employed the REPROBUS scheme with all necessary forcings, identical to
those used in our simulations with the ARPEGE-Climat 7.0.1 scheme. This simulation did not, however, include emissions, as
they are not relevant in this context. These characteristics make it particularly useful for comparison, as differences in chemical
concentrations with the concentrations obtained running ARPEGE-Climat-7.0.1 can then be attributed to variations in model
dynamics, such as transport and temperature, or to the influence of tropospheric chemistry. Analyses of its outputs have been
235 presented in several publications (see Section 2.2). Here, we use outputs from the 1979–2014 period, which we refer to as
ESM2-REPRO outputs (see Table 1).

3 Results and discussion

In the following section and in Section 3.2.2 we conduct a qualitative rather than quantitative evaluation of several key chemical
species, first in the stratosphere then in the troposphere.

240 3.1 Evaluation of stratospheric air composition

We analyse here several key stratospheric species, (i.e., O₃, H₂O, CH₄, HNO₃, NO₂, N₂O and HCl), as discussed in Teyss  re
et al. (2007), highlighting major differences and similarities between the ARP7-REPRO, ARP7-RELACS, and ARP7-RACM
simulations in comparison to the ESM2-REPRO simulation. Additionally, we contextualize our findings with CMIP6 analyses.
For components whose behaviour has not yet been extensively examined in CMIP6 publications, to our knowledge, we also
245 refer to the key conclusions of Michou et al. (2011), which documented the performance of the REPROBUS scheme within a
climate model closely related to the one used in CNRM-ESM2-1.

O₃ is the most abundant chemically active trace gas in the stratosphere. It is primarily produced through the photolysis of O₂,
a process most efficient in the tropical middle stratosphere. O₃ is also subject to destruction via catalytic reactions involving
nitrogen oxides, chlorine, and bromine species, though these reactive compounds can be temporarily sequestered in reservoir
250 species. Despite O₃ concentrations reaching over 10 ppm in parts of the stratosphere, it exists in a chemical equilibrium with



trace species present at much lower concentrations. Additionally, O_3 serves as a major radiative agent, with its distribution strongly influenced by stratospheric temperature and transport processes.

Water vapour and CH_4 play an important role in stratospheric chemistry. For example, the oxidation of CH_4 generates HO_x radicals which catalytically destroy O_3 , and are involved in many stratospheric reactions. Furthermore, water vapour is a key constituent of polar stratospheric clouds (PSC), which play a unique role in the chemistry of the polar vortex (Noël et al., 2018). Noël et al. (2018) provide a clear description of the various processes influencing the distributions of these two constituents in the stratosphere, distributions that are closely related. As a first approximation, it can be considered that the destruction of one molecule of CH_4 leads to the production of two molecules of H_2O (Teyssède et al., 2007; Noël et al., 2018).

HNO_3 is likely the main reservoir of nitrogen species in the lower stratosphere and acts as a sink for nitrogen compounds in the stratosphere. A significant removal involves uptake in polar vortices by PSCs that sediment to lower altitudes. This uptake strongly depends on temperature. NO_2 in the stratosphere is another key species that plays a major role in the O_3 budget, particularly in the lower stratosphere. N_2O is significant due to its warming potential and its impact on stratospheric O_3 . This GHG is well mixed in the troposphere due to its long lifetime there (about 100 years), and its concentration decreases with altitude in the stratosphere due to photodissociation and production of NO , which catalyses the O_3 destruction (Strong et al., 2008). In the stratosphere, its distribution is mainly governed by the Brewer-Dobson circulation. HCl is considered a reservoir species. Under specific conditions, typically upon return of sunlight in spring, it can be converted into reactive chlorine species that contribute to the O_3 destruction. Chlorofluorocarbons and other long-lived species that contain chlorine are the primary sources of HCl when broken by photolysis.

3.1.1 Ozone

Typical total column ozone (TCO) diagnostics are presented in Figs. 1 (as time series) and 2 (as zonal climatologies). In Fig. 1, the TCO time series for the three ARP7 simulations (ARP7-REPRO, ARP7-RELACS, and ARP7-RACM) closely follow each other, particularly after the first few years of simulations. This holds across various latitude bands and periods analysed (note that at high latitudes, these periods represent only one month, March for northern latitudes and October for southern latitudes). Using the same chemistry scheme (i.e., REPROBUS), the ARP7-REPRO simulation exhibits higher TCO values than the ESM2-REPRO simulation over northern latitudes (both mid and high), as well as globally, of about 10 %. This represents an improvement (see Michou et al., 2020a), where TCO in the same regions were underestimated by the same amount, and may be linked to a better representation of stratospheric circulation in the ARPEGE-Climat 7.0.1 climate model compared to the CNRM-ESM2-1 model. Overall, ARP7-REPRO TCO is in good agreement with the EAC4 reanalysis (red line), which falls well within observed variability, as documented in Michou et al. (2020a). Compared to the results of Keeble et al. (2021), who analysed historical CMIP6 simulations from 22 models, all ARP7 simulations fall within the spread of these CMIP6 models (see their Fig. 6). Keeble et al. (2021) evaluated model performance for annual and seasonal TCO against observations from 2000 to 2014, concluding that CNRM-ESM2-1 was the closest to the reference, with relatively small biases compared to other CMIP6 models. It was also one of the few models to reproduce the observed negative trends in TCO over northern mid- and high latitudes. Here, we confirm that all time series correctly capture the trend shifts across all subregions except

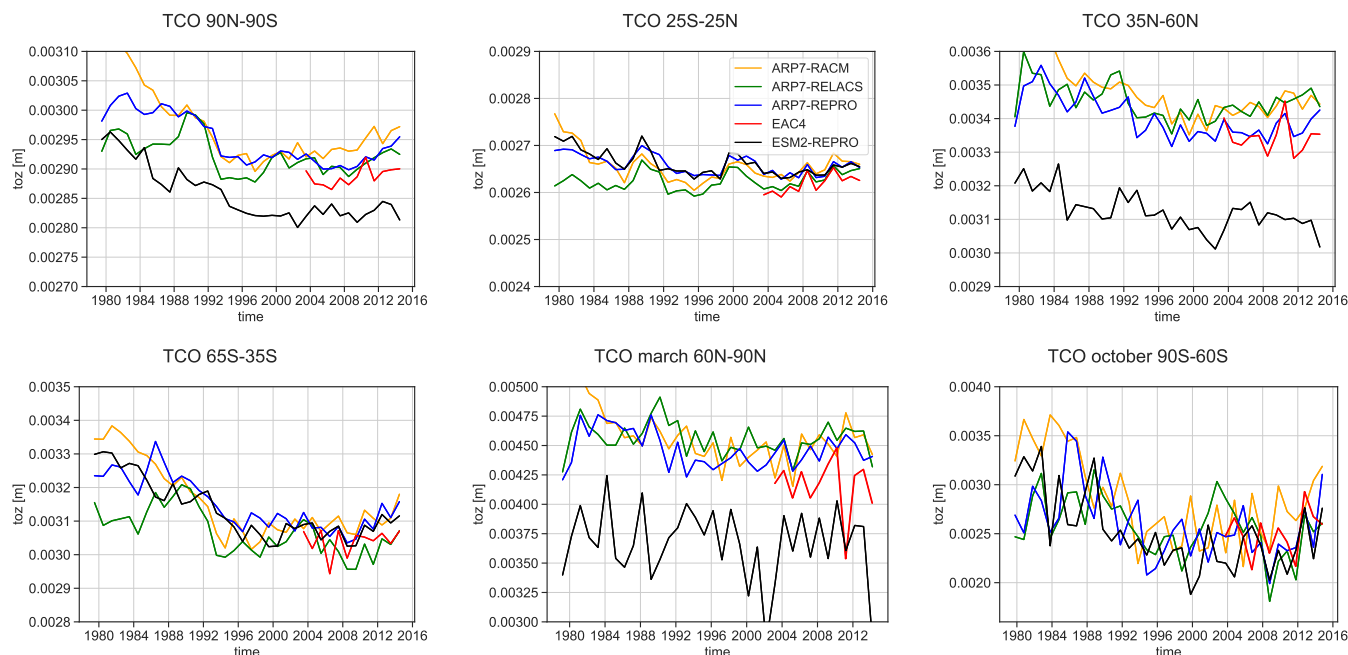


Figure 1. Time series (1979–2014) of yearly mean area-weighted TCO (m) over different latitude bands and periods, following Michou et al. (2020a) Fig. 10. Note that y-axes may vary across sub-images. ARP7-RACM (orange), ARP7-RELACS (green), ARP7-REPRO (blue), ESM2-REPRO (black), and EAC4 reanalysis (red).

285 in the tropics (25S–25N), between the pre-2000 period (negative trend) and the post-2000 period (neutral to positive trends). Zeng et al. (2022) quantified the contribution of individual forcings to changes in total, stratospheric, and tropospheric column ozone using AerChemMIP CMIP6 atmosphere-only simulations from six models, including CNRM-ESM2-1. They identified that the ODS forcing, the dominant driver of TCO changes, is too strong in CNRM-ESM2-1 in the Arctic, and to a lesser extent in the Antarctic. In the ARP7 simulations, the larger Arctic TCO columns compared to the ESM2-REPRO simulations, particularly linked to higher winter (DJF) temperatures at these latitudes (not shown), help mitigate this deficiency. Zeng et al. (2022) also showed that increasing tropospheric O₃ since the late 1970s has played an increasingly important role in TCO changes, particularly in the tropics and northern mid-latitudes. This effect appears to be confirmed in Fig. 1 for the 35N–60N latitude band, where higher TCO values in the ARP7-RELACS and ARP7-RACM simulations, compared to ARP7-REPRO, emerge from 2000 onwards.

295 The monthly climatologies (1990–1999 period, chosen for consistency with Michou et al. (2020a) which analysed this same period) of zonal TCO further support the observations discussed above. Differences from the ESM2-REPRO climatology are smallest for the ARP7-REPRO simulation. It is worth noting that the EAC4 climatology spans 2003–2014, which may partially explain the positive bias over southern high latitudes in September and October. However, the even less pronounced O₃ hole in the ARP7-RACM simulation suggests a potential deficiency in this case. Overall, the 1990–1999 climatologies for the ARP7

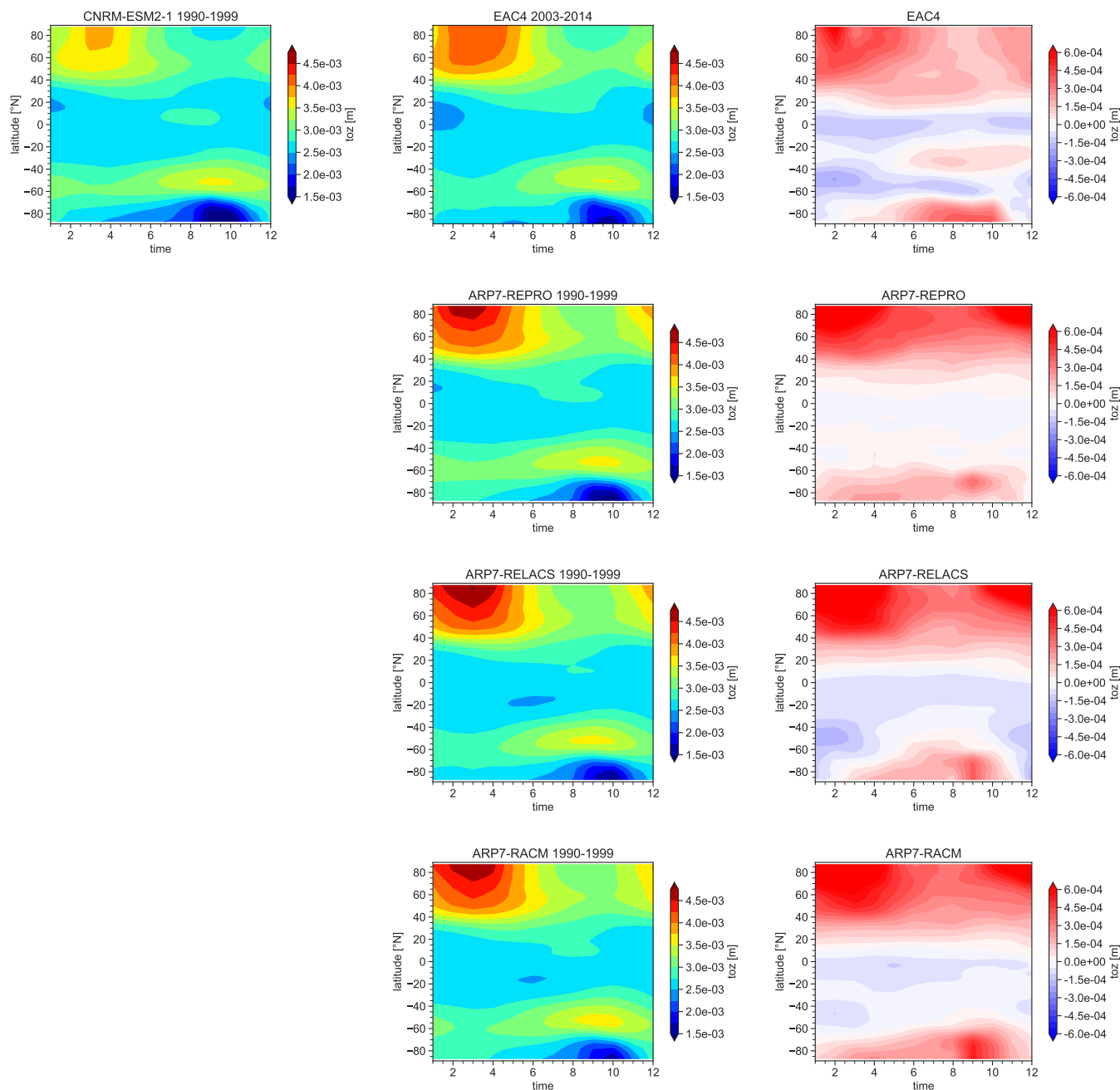


Figure 2. Monthly zonal TCO (m) climatologies over 1990–1999, following Michou et al. (2020a) Fig. 5. Absolute values are shown for the ESM2-REPRO simulation, while differences (simulation minus ESM2-REPRO) are presented in the following order: EAC4 reanalysis, and the ARP7-REPRO, ARP7-RELACS, and ARP7-RACMOBUS simulations. Note that the EAC4 climatology covers a different period (2003–2014).



simulations with lower TCO in the tropics and higher O₃ elsewhere compared to the ESM2-1 simulation lead to a reduction in the biases identified in Michou et al. (2020a) across all latitudes and seasons.

Zonal mean climatologies (1979–2014) of O₃ in the stratosphere (300 to 1 hPa) are shown in Fig. 3 for March and in Appendix Fig. A2 for September. Differences from the ESM2-REPRO simulations are generally similar across all simulations, with the largest deviations occurring at high northern latitudes in spring and around the O₃ concentration peak near 10 hPa, where the difference represents between 5 and 10 % of ozone mixing ratios. The same pattern is observed in September, likely due to variations in the strength of the polar vortices between the CNRM-ESM2-1 and ARPEGE-Climat 7.0.1 models, as well as greater model variability at these latitudes. A significant negative bias at 1 hPa compared to observations, falling outside the range of all CCMVal-2 models, was previously identified in Michou et al. (2011) and attributed to excessive upper-stratospheric warming. In the simulations presented here, this negative bias at 1 hPa has been eliminated in the ARP7-REPRO and ARP7-RELACS simulations (though the exact diagnostics from Michou et al. (2011) are not shown), while it remains, albeit to a lesser extent, in the ARP7-RACM simulation. Since temperatures are very similar across all ARP7 simulations, they cannot be solely responsible for this negative bias of ARP7-RACM.

3.1.2 Water vapor and methane

The spatial and temporal distributions of CH₄ in the ARP7-REPRO and ARP7-RELACS simulations are very similar from 1 hPa down to 100 hPa (see Fig. 3), although their entry levels in the tropical lower stratosphere differ due to the level of GHG relaxation, with higher entry levels for ARP7-REPRO (see Section 3.3.2). The CH₄ oxidation process that results in stratospheric moistening leads to a very similar CH₄/H₂O equilibrium throughout most of the stratosphere in both ARP7-REPRO and ARP7-RELACS. In ARP7-RACM, however, the moistening is lower: with the same H₂O entry level in the equatorial lower stratosphere constrained by temperature (see Fig. 4), H₂O and CH₄ concentrations are lower in ARP7-RACM compared to the other ARP7 simulations. The chemical production of H₂O appears less efficient in ARP7-RACM, with more CH₄ entering from the troposphere, similar H₂O entry at the cold point tropopause (see Fig. 4) and, ultimately, less water in the entire stratosphere. Keeble et al. (2021) explore the expected and observed relationship $H_2O = 7.0 - 2.0 \times CH_4$ (Eyring V. and Waugh, 2010; Archibald et al., 2020a) for six CMIP6 models and find a wide range of results. In the CNRM-ESM2-1 model, H₂O increases by more than two molecules for every molecule of CH₄ oxidized, although there is a negative bias in H₂O to observations, as is the case in most CMIP6 climate models (18 models provided H₂O outputs). The production of H₂O through CH₄ oxidation remains an open question (Keeble et al., 2021). However, we note in Fig. 4 a clear improvement in the equatorial temperature at 100 hPa, transitioning from the CNRM-ESM2-1 model to the ARPEGE-Climat 7.0.1 model, with higher temperatures, more in line with those of the other CMIP6 models.

3.1.3 Other species

The similarities in the zonal distributions of ARP7-REPRO and ARP7-RELACS, previously observed for CH₄ and H₂O, also extend to N₂O and HCl; this resemblance is however less pronounced for NO₂ and HNO₃ concentrations (see Figs. 5 and 6).

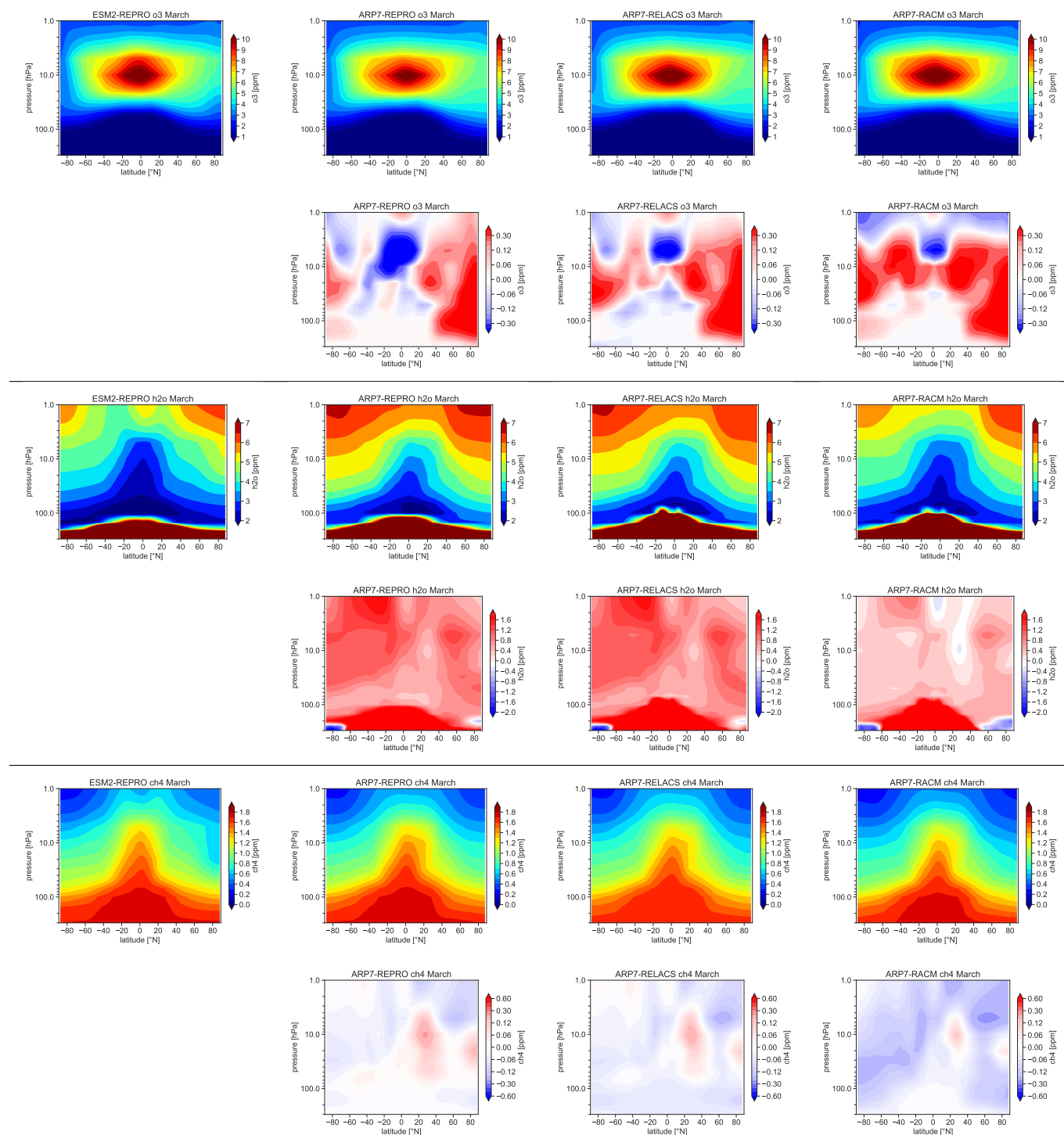


Figure 3. Mean zonal climatology (1979-2014, 300 to 1 hPa) of various species, in March (see September in the Annex). Each set consists of two lines: O₃ (ppm), H₂O (ppm), and CH₄ (ppm). The first line presents absolute values for, in order, ESM2-REPRO, ARP7-REPRO, ARP7-RELACS, and ARP7-RACM simulations. The second line shows the differences (simulation minus ESM2-REPRO) for the ARP7-REPRO, ARP7-RELACS, and ARP7-RACM simulations.

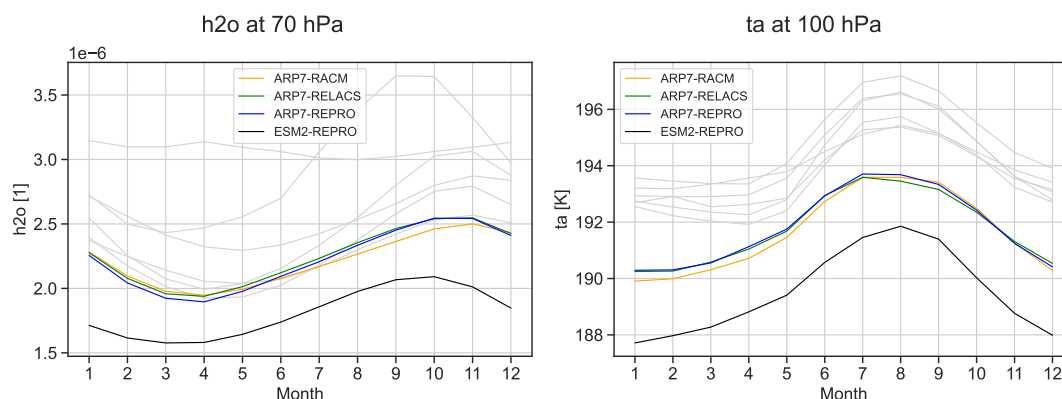


Figure 4. Annual cycles of H_2O mixing ratio (mol mol^{-1}) at 70 hPa and temperature (K) at 100 hPa, within the 15S-15N latitude band (as in Keeble et al. (2021)), for the ARP7-RACM (in orange), ARP7-RELACS (in green), ARP7-REPRO (in blue), and ESM2-REPRO (in black) simulations, along with other CMIP6 model AMIP simulations (in grey).

For HNO_3 , the concentrations in all three ARP7 simulations are lower than those of ESM2-REPRO at high latitudes, with larger negative biases for ARP7-RELACS and even greater negative biases for ARP7-RACM. These negative biases are even more pronounced in the southern latitudes in September (see Fig. A3). Differences between ESM2-REPRO and ARP7-REPRO, with the same chemistry scheme, could be linked to polar temperatures. Indeed, lower polar temperatures may lead to the formation of more PSCs, resulting in enhanced uptake and sedimentation of HNO_3 . However, this is not what we observe in model temperature (not shown) with ARPEGE-Climat 7.0.1 having higher polar stratospheric temperatures than CNRM-ESM2-1 in all seasons. Since there are only small temperature differences between ARP7-RELACS and ARP7-RACM, the discrepancies in HNO_3 concentrations likely point to differences in the denitrification scheme. We further compared our results with those of Michou et al. (2011), who analyzed annual cycles from models and observations at various altitudes and latitudinal bands. We find that the EMS2-REPRO and ARP7-REPRO simulations consistently fall within the observed range, even at 200 hPa between 40S and 60S, an area previously highlighted by Michou et al. (2011) as problematic. However, ARP7-RELACS and ARP7-RACM exhibit notably lower concentrations than ESM2-REPRO, with annual cycles significantly weaker than the observations presented in Michou et al. (2011). More recently, Archibald et al. (2020a), in their evaluation of the Strat-Trop chemical mechanism in the UK Earth System Model, noted that satellite observations from 2004–2013 revealed considerably higher stratospheric HNO_3 concentrations compared to the older satellite dataset used in Michou et al. (2011). This suggests that the evolution of HNO_3 in the stratosphere within the ARP7-RELACS and ARP7-RACM simulations warrants further investigation.

The distributions of NO_2 in March and September capture some of the characteristics cited in Teyss  dre et al. (2007) for the NO_x species, including a vertical gradient with maxima around 5 hPa. Negative biases compared to ESM2-REPRO are observed in most of the stratosphere (except for the tropical upper stratosphere) for all three ARP7 simulations, with larger biases for ARP7-RACM. A problematic NO_2 concentration at 1 hPa was highlighted in the conclusions of Michou et al.

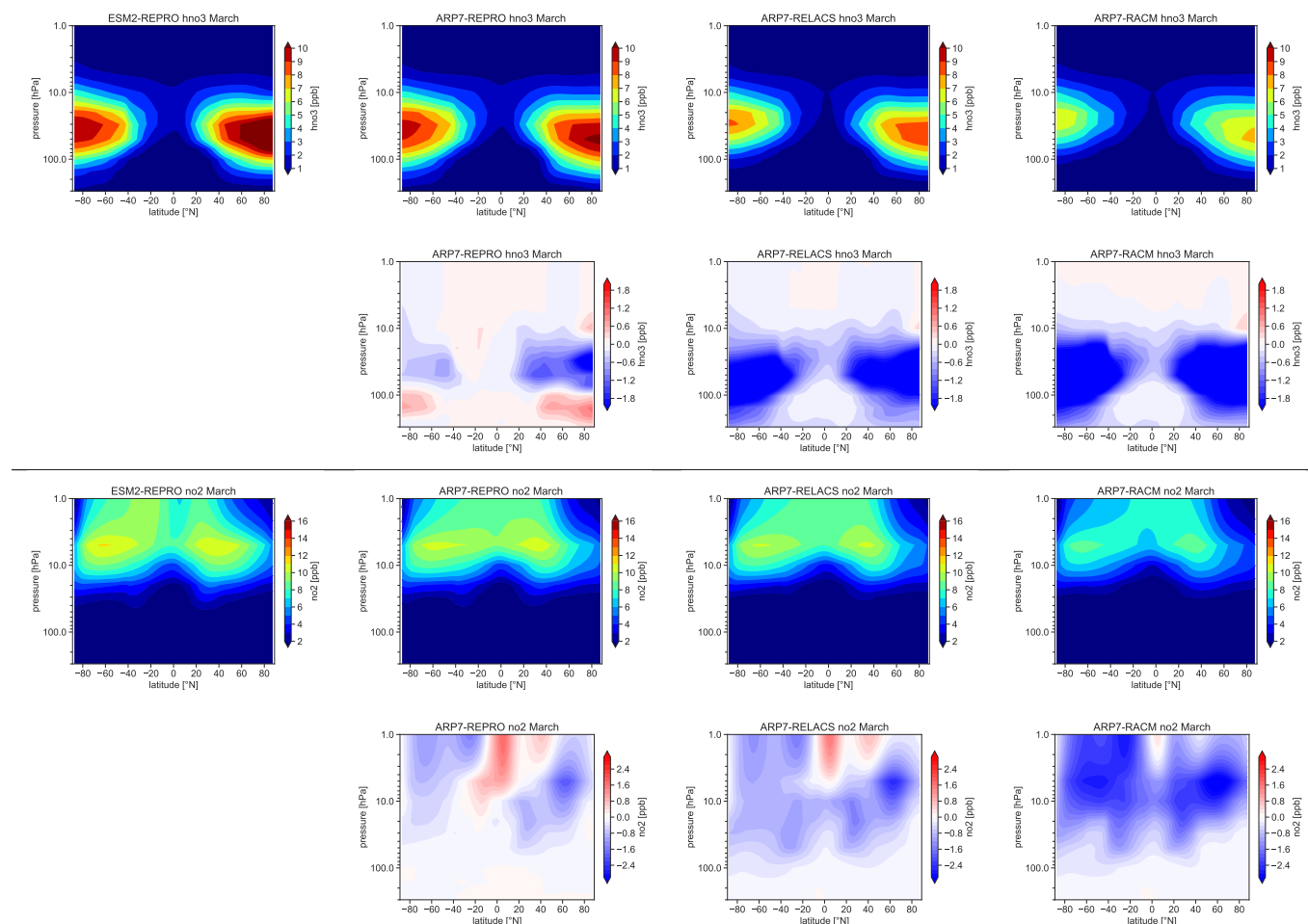


Figure 5. As in Fig. 3 for HNO_3 (ppb) and NO_2 (ppb).

(2011), where the hypothesis was that too warm temperatures in this part of the stratosphere resulted in much too high NO_2 compared to observations. In ARPEGE-Climat 7.0.1, with a colder upper stratosphere than that of Michou et al. (2011), NO_2 at 1 hPa remains too high (not shown), although ARP7-RACM NO_2 is now within the spread of the CCMVal-2 models shown in Michou et al. (2011).

N_2O distributions in the ESM2-REPRO, ARP7-REPRO, and ARP7-RELACS simulations are largely similar, except in the tropical latitudes of the middle stratosphere. This tropical bias, also observed in the CH_4 distribution, may stem from differences in stratospheric transport between the CNRM-ESM2-1 and ARPEGE-Climat 7.0.1 climate models, particularly regarding mixing across the subtropical barriers. The negative bias in ARP7-RACM compared to ESM2-REPRO, already noted for CH_4 , extends throughout the stratosphere, indicating a lower stratospheric entry level. This is further confirmed in Fig. 17, which shows the N_2O trend across the entire simulation period (1979–2014). The entry-level concentration appears

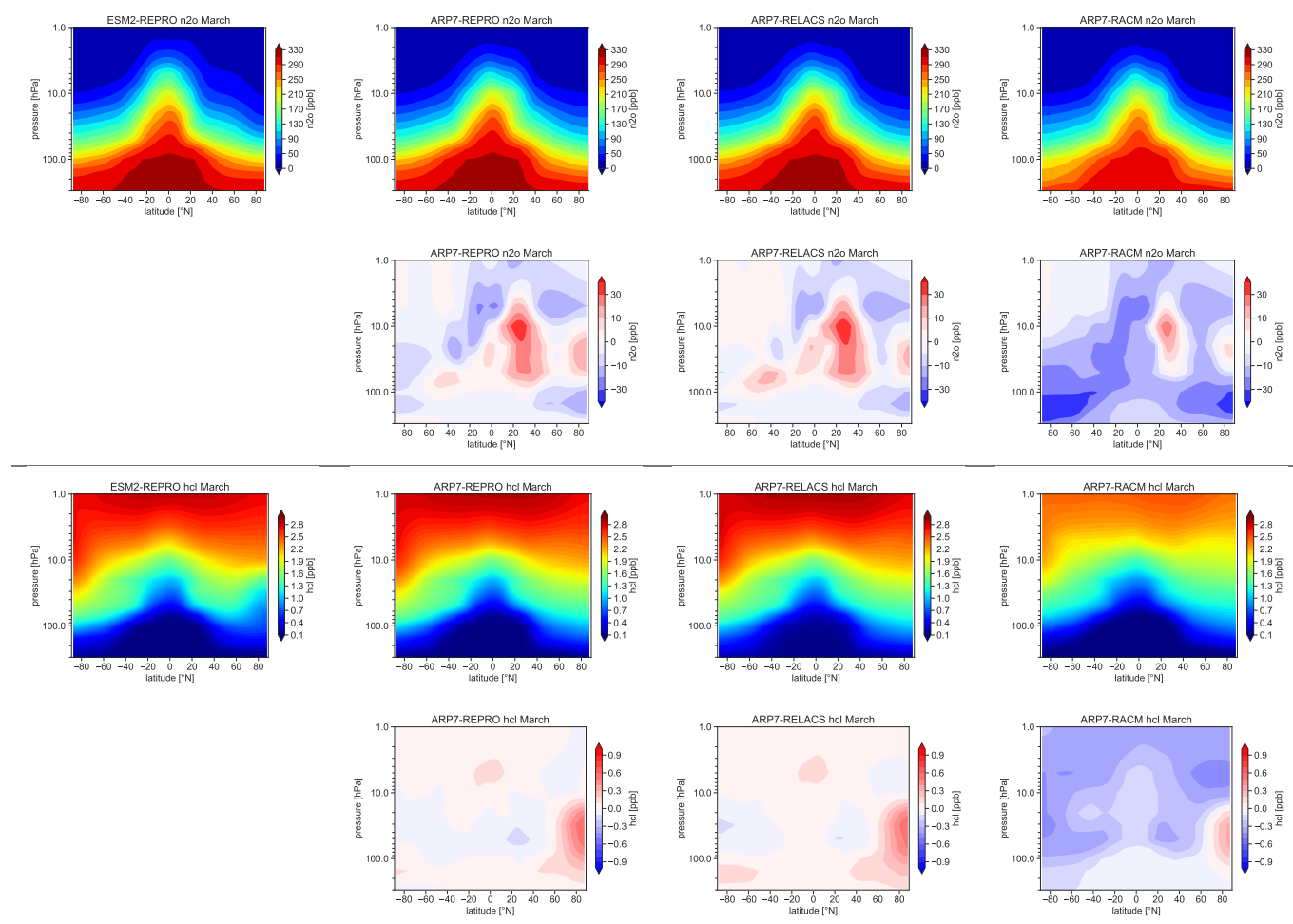


Figure 6. As in Fig. 3 for N₂O (ppb) and HCl (ppb).



too low compared to observations (e.g., 319 ppb in 2005 observations (Strong et al., 2008) vs. 313 ppb in ARP7-RACM), highlighting the need for further investigation.

365 For HCl, the distributions of the ARP7-REPRO and ARP7-RELACS outputs are similar, with the largest differences observed in the spring high latitudes, around 40 hPa, where they show higher concentrations compared to ESM2-REPRO. This suggests either lower sequestration of HCl on PSCs in winter, possibly due to smaller PSC areas (this can be confirmed by higher polar temperatures in the ARPEGE-Climat 7.0.1 model compared to CNRM-ESM2-1), and/or a reduced transformation of HCl sequestered in PSCs into more reactive chlorine species upon the return of sunlight. The negative bias of ARP7-RACM
370 throughout the stratosphere results in values in the upper stratosphere being too low given the maximum total chlorine imposed by CMIP6. We confirm these too low values for ARP7-RACM with plots at 1 hPa (not shown), as presented in Michou et al. (2011). This deficiency is undoubtedly linked to an insufficient amount of HCl entering the stratosphere, likely due to excessive scavenging of this species in the troposphere (not shown).

3.2 Evaluation of tropospheric air composition

375 The following analysis focuses on key atmospheric components, O₃, CO, NO₂, OH, HNO₃, and PAN, which are commonly evaluated in climate models with tropospheric chemistry (e.g., O'Connor et al. (2014a); Archibald et al. (2020a); Williams et al. (2022)). We outline below key aspects that illustrate the close interconnections between these chemical species.

Tropospheric O₃ is a critical atmospheric component, acting both as a greenhouse gas and an air pollutant (Young et al., 2018). It primarily forms through photochemical reactions between precursor gases, such as NO_x and NMVOCs, in the presence
380 of sunlight. Its concentrations, which can reach up to 100 ppb, are highest in the lower to middle troposphere and vary by region, season, and altitude. O₃ is also closely linked to OH, which plays a crucial role in its production and destruction. Other key influences on its tropospheric budget include stratospheric transport, and dry deposition. NO_x species originate mainly from anthropogenic sources, such as fossil fuel combustion, but also from natural sources like lightning. Their chemistry is complex (Seinfeld and Pandis, 2006): the photolysis of NO₂ leads to O₃ formation, while NO rapidly destroys O₃. However, in the
385 presence of other radicals NO₂ is quickly regenerated. Major sinks for NO_x include the production of HNO₃ via reactions with NO₂ and the formation of PAN. OH is a highly reactive species at the core of atmospheric chemistry, often referred to as “the atmosphere’s cleansing agent” (Naik et al., 2013). It serves as the primary oxidant for trace gases such as CH₄, CO, and NMVOCs, regulating their removal. NO_x in general influences OH levels in opposing ways, generating OH, but in highly polluted environments, NO₂ becomes a dominant sink for OH through HNO₃ formation (Stevenson et al., 2020). HNO₃ is one
390 of the most significant acidic air pollutants, reaching several ppb in polluted air and a few tens of ppt in clean air. CO is another key atmospheric component, emitted primarily from incomplete combustion, making it a useful tracer of anthropogenic activity (Faïn et al., 2025). It also acts as a precursor to tropospheric O₃ and, due to its longer lifetime (~two months compared to a few days for NMVOCs), is more abundant. Additionally, CO is a major sink for OH, directly affecting the oxidizing capacity of the troposphere. Among primary tropospheric pollutants, CO is one of the most extensively monitored on a global scale via satellite
395 observations (Froidevaux et al. (2025) and references therein). PAN primarily forms through the reaction of NMVOC-derived radicals with NO₂ and acts as a temporary reservoir for NO_x, facilitating its long-range transport, particularly in the cold upper

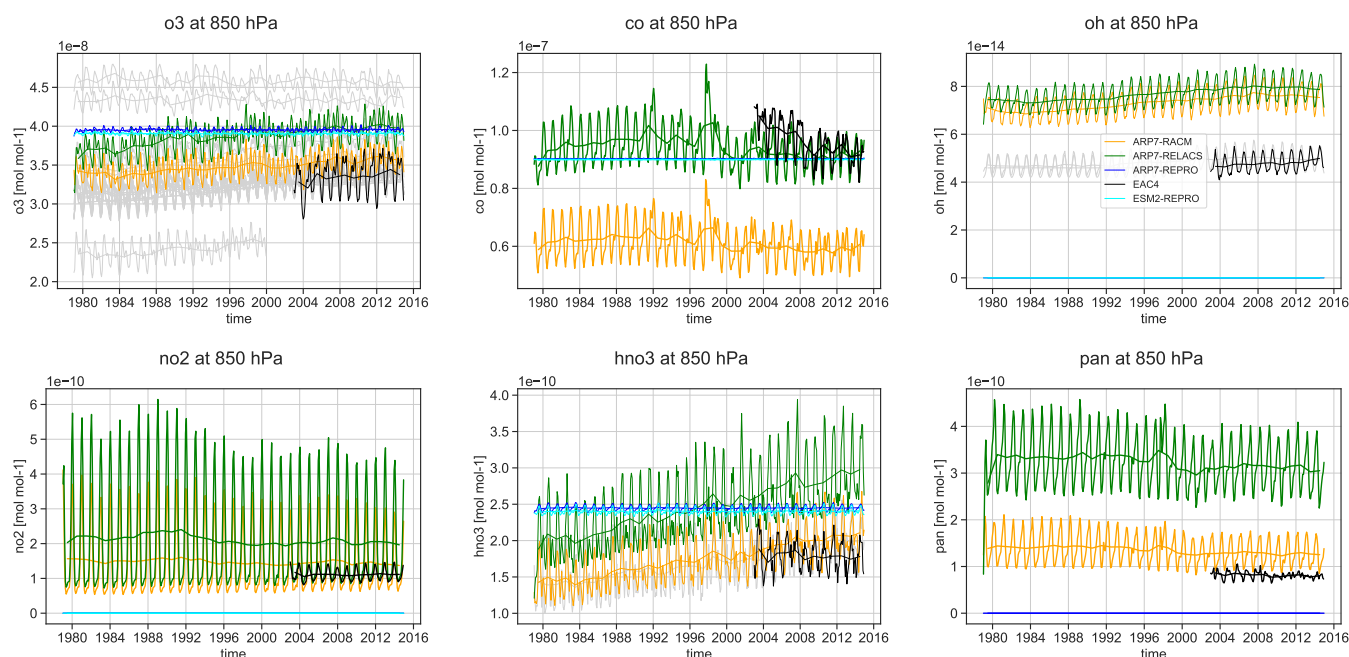


Figure 7. Time series (1979-2014) of monthly and annual global mean mixing ratios (mol mol^{-1}) at 850 hPa. In the following order: O_3 , CO , OH , NO_2 , HNO_3 , and PAN. EAC4 reanalysis in black (2003-2014), ARP7-RACM simulation in orange, ARP7-RELACS in green, ARP7-REPRO in dark blue, ESM2-REPRO in cyan. Note that PAN is not a species of the REPROBUS scheme (ESM2-REPRO simulation). CMIP6 model AMIP simulations (outputs depending on the species) other than ESM2-REPRO in grey.

troposphere (Fiore et al., 2018). Since its lifetime greatly increases at lower temperatures, it is found in larger amounts in the free troposphere. Under warmer conditions, PAN undergoes thermal decomposition, releasing NO_2 .

In addition to insights from the published literature, we enhance our study by incorporating outputs, primarily O_3 ones, from additional CMIP6 models processed using the ESMValTool package (Righi et al., 2020).

3.2.1 Trends and variability of various species

Fig. 7 is aimed at identifying large trends over the 1979-2014 period, as well as annual and interannual variability. It shows time series of global mean concentrations at 850 hPa for the six chemical species listed above. In all the line plots, our reference dataset, the EAC4 reanalysis, is in black, while data from CMIP6 models appear in light grey, and the four ARPEGE-Climat 7.0.1 simulations are presented in other colours.

Some characteristics are common to all species: in particular, the REPROBUS scheme produces similar concentrations in both the ARPEGE-Climat 7.0.1 model and the CNRM-ESM2-1 model. Time series in this case show no trend, and much smaller variability compared to the time series from the reanalysis or the other two chemistry schemes. As by essence the REPROBUS scheme computes chemical concentrations of transported species below 560 hPa using relaxation rather than a



410 chemistry solver (see Michou et al. (2020a)), some REPROBUS time series at 850 hPa appear coherent with the reanalysis time series (e.g., for HNO_3). Other species diverge significantly (e.g., NO_2) because REPROBUS attributes a quasi-null mixing ratio value to non-transported species below 560 hPa. Another feature of these time series is that the RELACS and the RACM time series differ in their mean for O_3 , CO , HNO_3 and PAN.

For O_3 , RACM is closer to EAC4 than RELACS, noting that the dispersion of the CMIP6 model time series is very large
415 and encompasses all ARPEGE-Climat 7.0.1 time series. For CO , no CMIP6 dataset could be processed with ESMValTool. However, the ARP7-RELACS time series is found to closely align with the reanalysis, whereas the ARP7-RACM values appear too low. Additionally, the ARP7-RELACS and ARP7-RACM CO time series closely have the same interannual variations. The case of nitrogen-containing compounds (i.e., NO_2 , HNO_3 and PAN) is particularly interesting. While the monthly means of NO_2 for ARP7-RACM and ARP7-RELACS are close to those of the reanalysis during half of the year, around the northern
420 hemisphere summer, these monthly means differ significantly during the remainder of the year, see also the mean annual cycles of NO_2 in Figure 13). However, none of the NO_2 time series exhibit a marked trend. A similar winter concentration bias for NO_2 was identified in Teyss  dre et al. (2007), which also found a positive bias in HNO_3 concentrations in the lower part of the troposphere, a pattern also observed in the ARP7-RELACS concentrations here. Teyss  dre et al. (2007) had already identified this winter-time overestimation, and two of the hypotheses made at that time, namely how the various species within the NO_x
425 family are handled and the absence of heterogeneous loss of N_2O_5 on tropospheric aerosols, remain valid. The case of the PAN time series will be discussed further in section 3.2.4.

Regarding trends, a clear positive trend in ozone (O_3) is observed in all of our simulations, the reanalysis, and most CMIP6 models, over the whole 1979-2014 period. On the other hand, the negative trend for carbon monoxide (CO) is more pronounced in the reanalysis, and much less so in the ARP7-RELACS and ARP7-RACM simulations. This trend warrants further
430 investigation, as the assimilation of column data (specifically from the MOPITT instrument) tends to be less effective near the surface (Wagner et al., 2021). In the case of HNO_3 , its concentration increases significantly in the ARP7-RACM and ARP7-RELACS simulations, while it remains constant in the reanalysis. We attribute this clear positive trend in the global time series to significant trends in the India-China region (not shown), which are linked to positive NO_2 trends in that area (Hoesly et al., 2018). Differences in physical removal processes between our simulations and the reanalysis may be influencing this behavior,
435 warranting further investigation.

3.2.2 Ozone

Fig. 8 presents the zonal mean (2003-2014) distribution in the troposphere (from the surface to 100 hPa) from the EAC4 reanalysis as well as from the ESM2 and ARPEGE-Climat 7.0.1 simulations, along with the absolute differences of the four AMIP simulations, listed in Table 1, with respect to the reanalysis. The two simulations using the REPROBUS scheme show
440 comparable differences with EAC4 in the southern hemisphere but larger negative differences for the ESM2-REPRO simulation in the northern hemisphere upper troposphere. This clearly reflects differences in stratosphere to troposphere exchanges, likely linked to higher stratospheric O_3 levels in the ARP7-REPRO simulation. Biases in the lower troposphere, below 500 hPa, are smaller in the simulations with the tropospheric schemes than in those with the REPROBUS scheme, with the best performance



seen in the ARP7-RACM simulation. Higher O₃ levels in the northern hemisphere in both ARP7-RELACS and ARP7-RACM
445 could be due to larger stratospheric downwelling, or to higher O₃ precursor concentrations. The first hypothesis should be
viewed with caution, given uncertainties regarding this phenomenon (Young et al., 2018), and since it does not extend further
up in the troposphere. As for the second hypothesis, while higher NO₂ concentrations for ARP7-RACM and ARP7-RELACS
simulations compared to EAC4 are evident in Fig. 12, the same is not true for CO concentrations (see Fig. 10). We note that
this positive bias of O₃ over major polluted regions is a common feature in climate models (see Sekiya et al. (2025) and
450 references therein, Young et al. (2018)). Additionally, the EAC4 surface O₃ is biased low compared to four other reanalyses
across most continental regions (Jones et al., 2025), which helps to contextualize the positive biases of ARP7-RACM and
ARP7-RELACS. A final striking difference between the ARP7-RELACS and ARP7-RACM simulations concerns the southern
high latitudes of the UTLS region. In this region the two schemes yield very different O₃ concentrations, possibly reflecting
higher transfer of O₃ from the stratosphere to the troposphere in the ARP7-RACM simulation, in line to its slightly higher
455 stratospheric concentrations. This is however consistent with Griffiths et al. (2021a) Fig. 2, which indicates that inter-model
variability peaks in the Southern Hemisphere between 100 and 250 hPa. Finally, not surprisingly, the overall zonal distributions
of the simulations with full tropospheric chemistry, with for instance higher levels of O₃ in the northern hemisphere, align more
closely with the multi-model mean of Griffiths et al. (2021b), compared to the simulations with the REPROBUS scheme.

Annual cycles of O₃ at 250, 500 and 700 hPa, and over two latitudinal bands in the northern hemisphere and two in the
460 southern one, as presented in Griffiths et al. (2021a), are shown in Fig. 9. General features of these annual cycles are as
follows: in all cases, the annual cycles of the ARP7-RACM and ARP7-RELACS simulations follow closely each other, while
the annual cycles of the two REPROBUS simulations (ESM-2-REPRO and ARP7-REPRO) exhibit distinct values and at
700 hPa show flat annual cycles because of the chemical scheme itself. Focusing on ARP7-RACM and ARP7-RELACS, the
annual cycles replicate those of the reanalysis more accurately in the southern hemisphere than in the northern hemisphere.
465 In the southern hemisphere, they show an adequate phase in all cases although the peak in October at 250 and 500 hPa is not
marked enough. Notably, in the southern extratropics, a few of CMIP6 models (grey lines) have seasonal cycles anti-correlated
with the EAC4 reanalysis, similarly to both of our REPROBUS simulations, this would suggest models without tropospheric
chemistry. In the northern hemisphere, ARP7 outputs at 700 hPa are outside the range of the CMIP6 models in the winter
months, and specifically in the extratropics both ARP7-RACM and ARP7-RELACS show a second peak in October which is
470 not present in the reanalysis. This issue may be related to the annual cycle of NO₂ which also shows problems in the northern
extratropics (see section 3.2.4). Additionally, the spring peak in the northern extratropics occurs one or two months earlier
in the ARP7 simulations, which could be associated to a prematurely occurring peak in the annual cycle of CO (see section
3.2.3). This peak is better reproduced by the ARP7-REPRO simulation at 500 hPa. ARP7-REPRO also shows better agreement
than ARP7-RACM and ARP7-RELACS with the reanalysis at 250 hPa northern tropics. In contrast, both ARP7-RACM and
475 ARP7-RELACS fail to reproduce the seasonality of the reanalysis at both at 500 and at 250 hPa in the northern tropics.

To contextualize our results, some deficiencies identified here were also observed in Teyssèdre et al. (2007) Fig. 19 and 20,
such as the too early spring peaks at 500 hPa (e.g., in stations like Sofia, Wallops Island or Churchill) or the second seasonal
peak in autumn (e.g., at Wallops Island). Furthermore, Jones et al. (2025) report that, at 500 hPa, the largest discrepancies

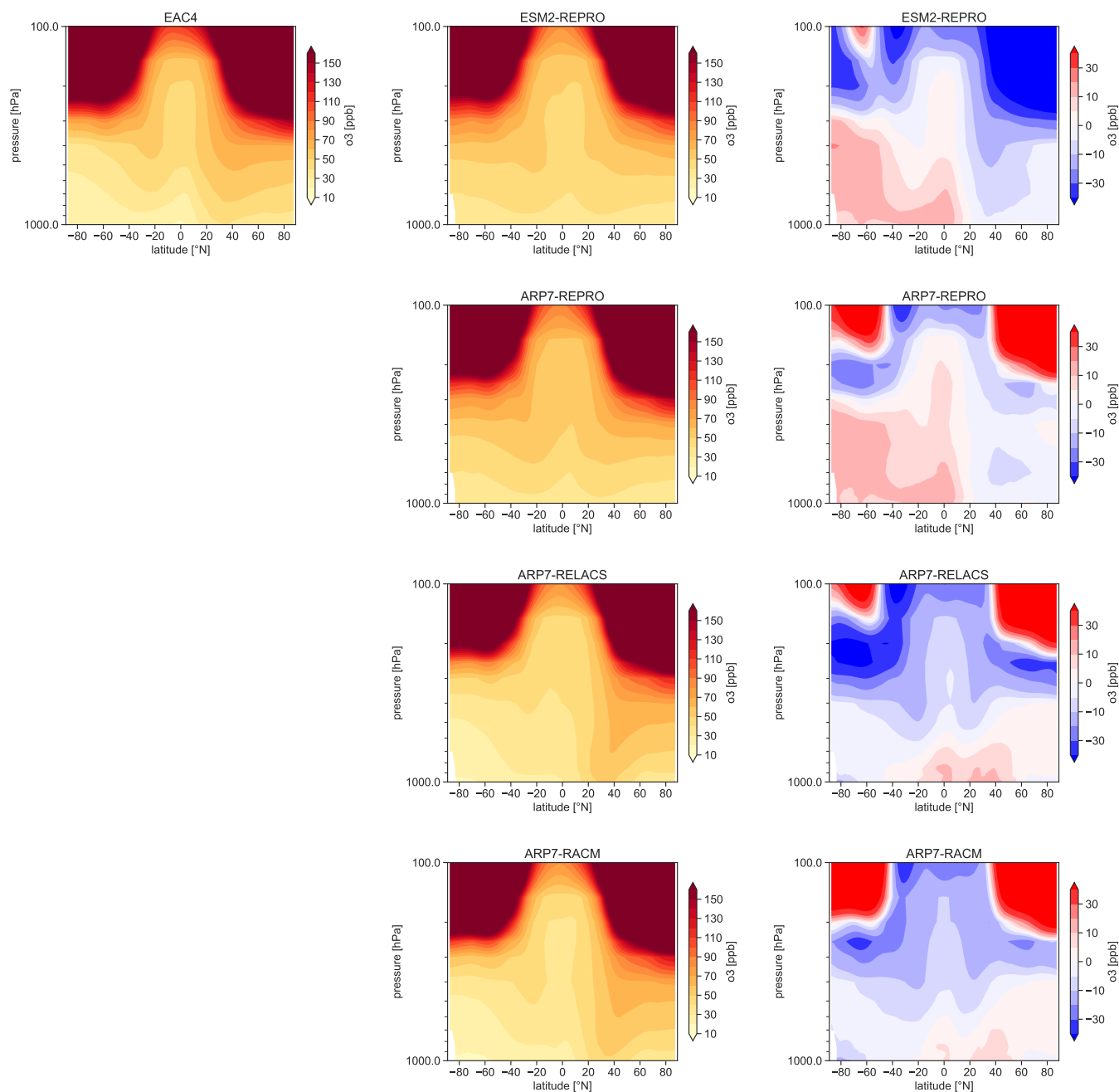


Figure 8. Mean (2003-2014) zonal climatology of tropospheric (surface to 100 hPa) O₃ (ppb). First column: EAC4 reanalysis (absolute values, scale as in Griffiths et al. (2021a)), second column ESM2-REPRO, ARP7-REPRO, ARP7-RELACS and ARP7-RACM simulations, third column differences (simulation minus EAC4).

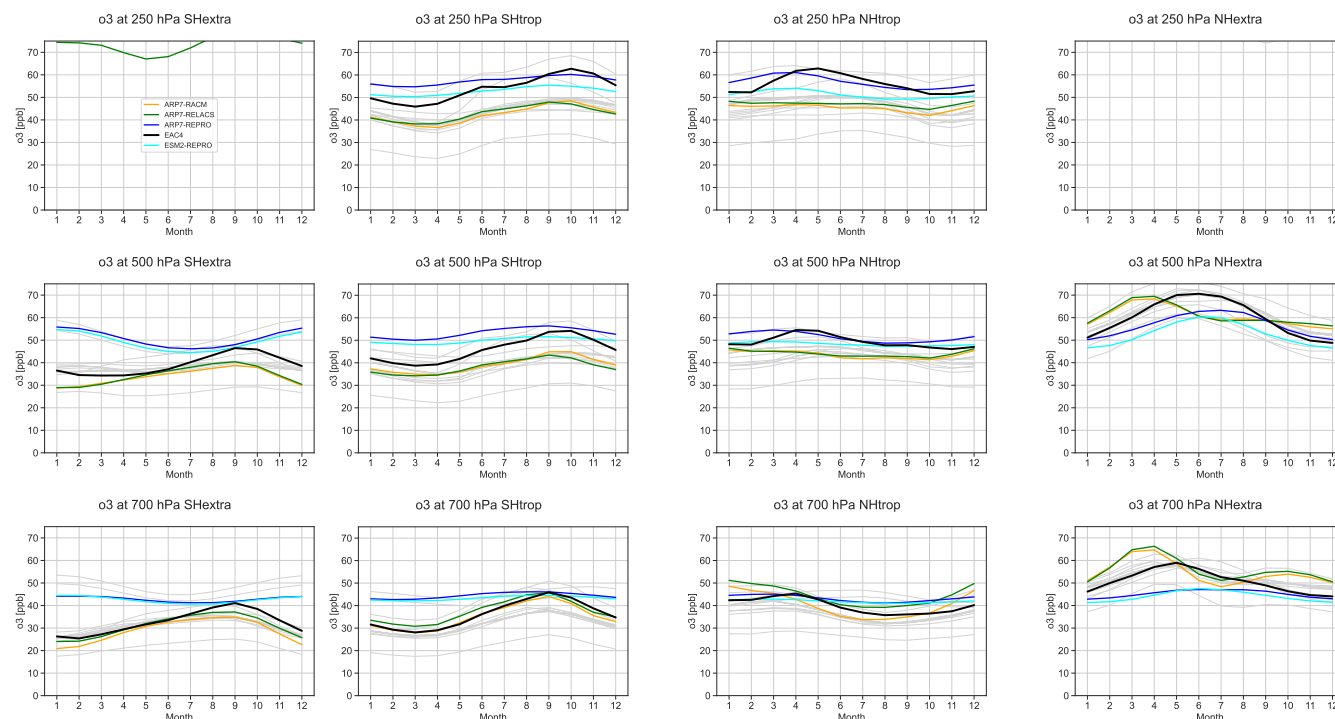


Figure 9. Mean annual cycles (2003-2014) of O_3 (ppb) at 250 hPa (first row), 500 hPa (second row) and 700 hPa (third row), over four latitudinal bands (from left to right: 90 to 30S, 30S to 0, 0 to 30N and 30N to 90N), as in Griffiths et al. (2021a). EAC4 reanalysis in black, ESM2-REPRO simulation in cyan, ARP7-RACM in orange, ARP7-RELACS in green, ARP7-REPRO in dark blue. Additional CMIP6 models in grey. No plotting at 250 hPa in the extratropics, as in Griffiths et al. (2021a).

between five chemical reanalyses, including EAC4, are observed in regions of the northern tropics. Finally, over the northern hemisphere extratropical continents (40N–70N), EAC4 typically exhibits seasonal biases in monthly mean O_3 , with a negative bias in winter and a positive bias in summer (Wagner et al., 2021). In the free troposphere (750–350 hPa), however, EAC4 does not show any discernible bias. In the tropics, Wagner et al. (2021) reports a systematic overestimation of O_3 in EAC4, both in the free troposphere (750–200 hPa) compared to ozonesondes and aircraft observations, and at the surface compared to in situ measurements.

3.2.3 Carbon monoxide

Fig. 10 presents the mean zonal distributions of CO, analogous to Fig. 8. Similar to O_3 , the CO distributions of the REPROBUS simulations are very similar, with hemispheric differences compared to the reanalysis: negative in the northern hemisphere and positive in the southern hemisphere. This reflects the fact that CO remains relatively constant throughout the troposphere in both the ESM2-1-REPRO and ARP7-REPRO simulations. In contrast, the CO distributions of the ARP7-RACM and ARP7-RELACS simulations show more significant differences, with particularly pronounced negative biases for ARP7-

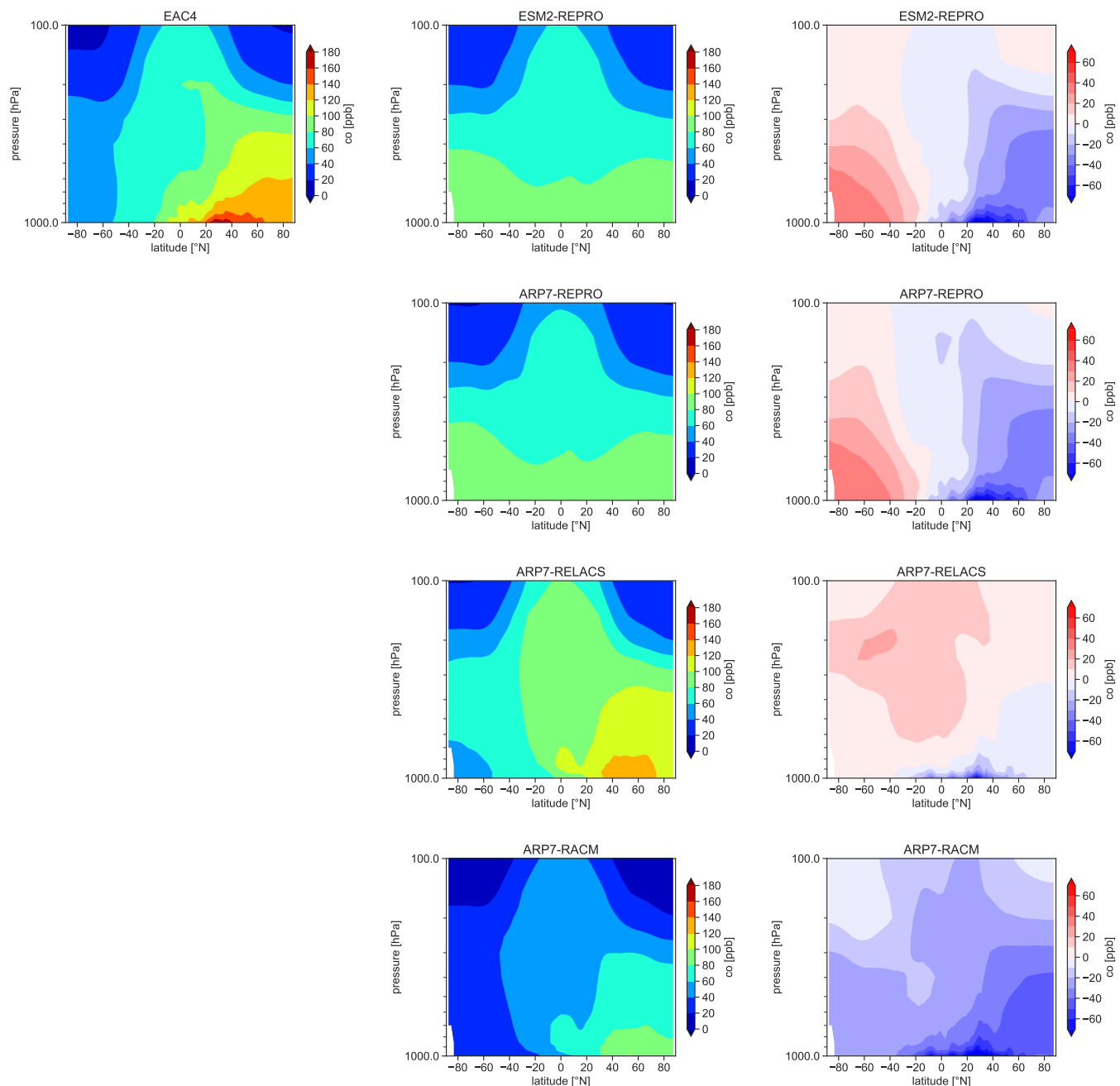


Figure 10. Mean (2003-2014) zonal climatology of tropospheric (surface to 100 hPa) CO (ppb). Same presentation as in Fig. 8.

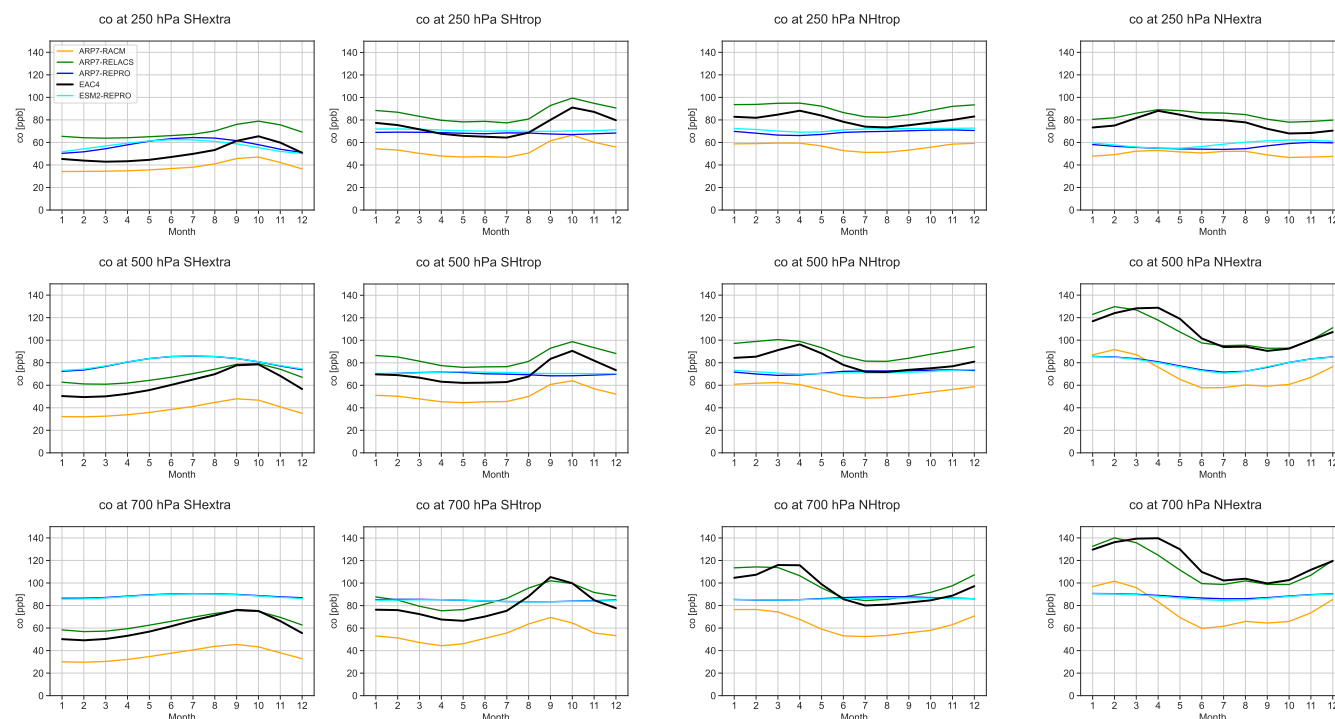


Figure 11. Mean annual cycles (2003-2014) of CO (ppb) at 250 hPa (first line), 500 hPa (second line) and 700 hPa (third line), over four latitudinal bands (from left to right: 90 to 30S, 30S to 0, 0 to 30N and 30N to 90N). EAC4 reanalysis in black, ESM2-REPRO simulation in cyan, ARP7-RACM in orange, ARP7-RELACS in green, ARP7-REPRO in dark blue.

RACM throughout the troposphere. The biases for ARP7-RELACS are generally smaller than those for ARP7-RACM, and their signs (negative in the northern hemisphere and positive in the southern hemisphere) are consistent with those reported in the literature (Shindell et al., 2006).

The differences in CO between ARP7-RACM and ARP7-RELACS may be due to differences in sources or sinks. Faïn et al. (2025) provides a synthesis of the carbon monoxide modern budget: primary emissions (mostly from fossil fuel combustion, with the remainder from biomass burning) and secondary production (primarily from CH₄ oxidation, with the remainder from NMVOCs oxidation) each accounts to 50% of CO sources, whereas oxidation by OH accounts for 93% of the CO sink, with the rest being removed through dry deposition. Since primary emissions are the same in all our ARPEGE-Climat 7.0.1 simulations, we can hypothesize that secondary production might differ, possibly due to slightly different CH₄ concentrations throughout the troposphere (see, for instance, Fig. 17) or due to differences in the COVs reactivity as the COVs lumping differs between RELACS and RACMOBUS (Crassier et al., 2000). Indeed, Tilmes et al. (2016) observed in the CCM1 simulations of the CESM1 CAM4-chem model an underestimation of the CO tropospheric column over most latitudes and time of the year, as we observe in the ARP7-RACM simulations. They indicated that, in the absence of any other straightforward evidence, a large underestimation of ethane concentrations likely contributed to this bias. The underestimation of CO in the northern hemisphere

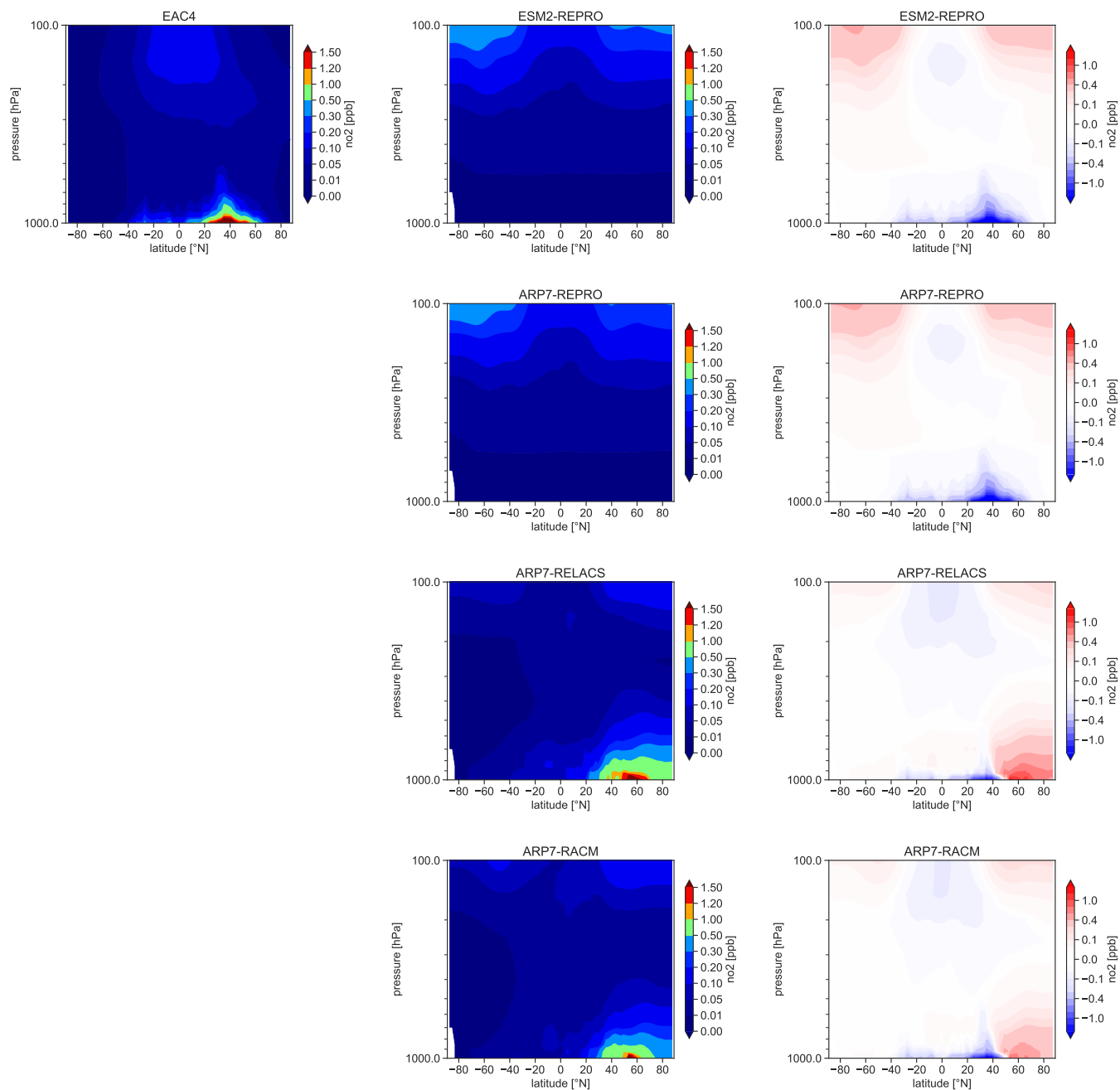


Figure 12. Mean (2003-2014) zonal climatology of tropospheric (surface to 100 hPa) NO_2 (ppb). Same presentation as in Fig. 8.

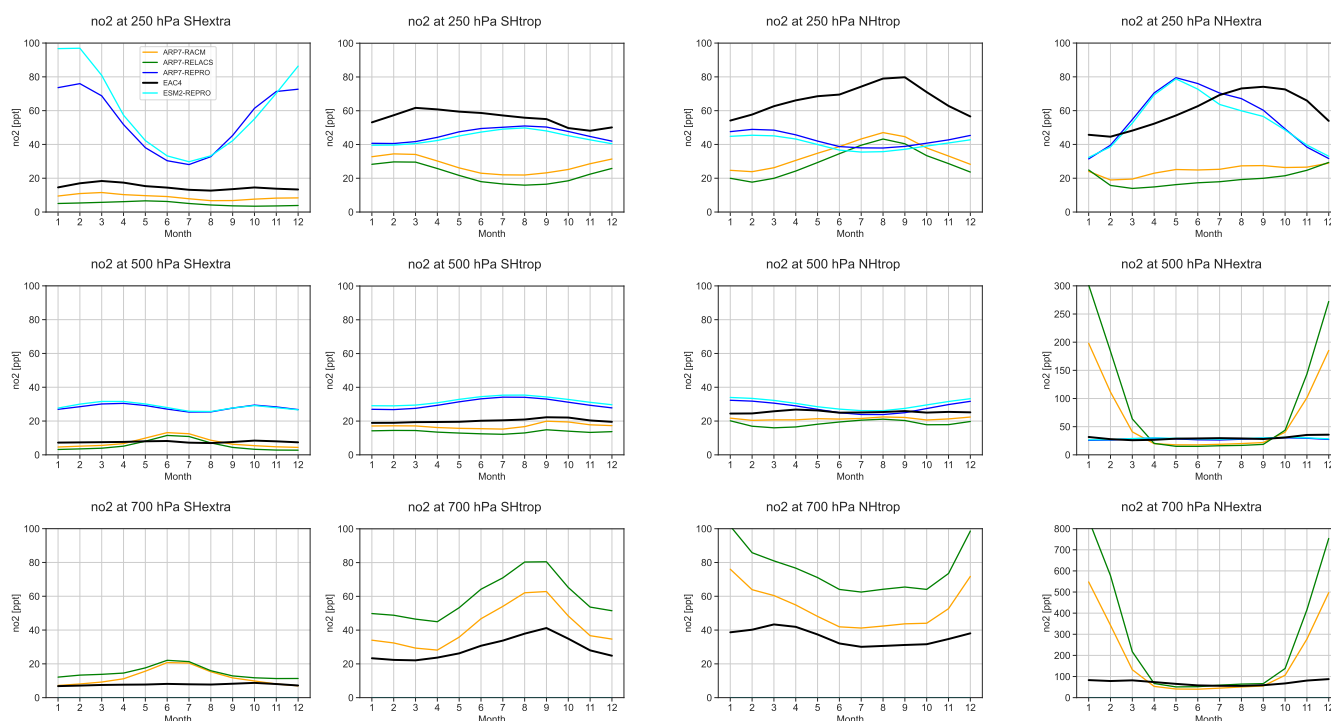


Figure 13. Mean annual cycles (2003–2014) of NO_2 (ppt) at 250 hPa (first row), 500 hPa (second row) and 700 hPa (third row), over four latitudinal bands (from left to right: 90° to 30°S, 30°S to 0°, 0° to 30°N and 30°N to 90°N). EAC4 reanalysis in black, ESM2-REPRO simulation in cyan, ARP7-RACM in orange, ARP7-RELACS in green, ARP7-REPRO in dark blue. Note that y-axis scale differs for each 30°N–90°N image, and at 700 hPa the ESM2-REPRO concentrations are not shown because of their very low values ($\sim 1\text{e}-20$).

is common in models and has been treated either as a cause or an effect of a general overestimation of OH (Strode et al. (2015); Zhao et al. (2023) and references herein). However, as OH and CO are major losses for each other, this complicates the determination of how much CO bias drives OH bias versus OH bias driving CO bias. Strode et al. (2015) performed several sensibility analyses to investigate the low bias in CO that could put some light on the results we get in the ARP7-RACM simulations. They show the importance of the meteorology and of the CO emissions, but in our case this cannot explain the difference we get between ARP7-RACM and ARP7-RELACS. They also highlight the OH latitudinal distribution but we have similar ones in ARP7-RACM and ARP7-RELACS (see Fig. 15). So further work is required to understand and correct the too low CO concentrations we get in the ARP7-RACM simulations.

The annual cycles of CO are presented in Fig. 11, similarly as O_3 appears in Fig. 9. Besides the previously discussed biases in ARP7-RACM, the cycles in both simulations with tropospheric chemistry align well with the EAC4 reanalysis. The ESM2-REPRO and the ARP7-REPRO lines, generally overlapping, have very flat annual cycles as no emissions are considered by the REPROBUS chemistry scheme. ARP7-RELACS has not only the smallest but also non-systematic positive or negative biases. This runs counter to results in which models systematically underestimated concentrations in the northern



hemisphere (Shindell et al., 2006). Shindell et al. (2006) attributed this underestimation to too low anthropogenic emissions in the northern hemisphere. Our ARP7-RELACS results appear in line with the conclusions of Faïn et al. (2025) who analysed surface concentrations of CO of 16 CMIP5 and 7 CMIP6 models in high latitudes against ice archive reconstructions. Faïn et al. (2025) conclude in improved CO emissions in the CMIP6 exercise, both of anthropogenic and of biomass burning sources. In the northern extratropics, the annual peak occurs two months early at 700 and 500 hPa, while in the southern latitudes the timing and intensity of the peak is correct, although during the rest of the year ARP7-RELACS overestimates the reanalysis. The annual cycles in the tropics at 250 hPa match rather well those presented in Froidevaux et al. (2025) Fig. 11, with peaks in October (respectively April) for the southern (respectively northern) hemisphere, in link with biomass burning injection and its subsequent transport towards the upper troposphere.

The evaluation of EAC4 against aircraft measurements and surface observations in Wagner et al. (2021) indicates that EAC4 generally underestimates CO mixing ratios in the free troposphere by 10 to 20%, so this provides overall good confidence in the RELACS tropospheric values. At the surface over both Europe and East Asia EAC4 also shows a negative bias while in contrast surface validation over North America reveals partially positive biases. These surface biases appear to be primarily driven by emissions, as inferred from comparisons with a previous ECMWF reanalysis (MACC), which operated with different or unscaled CO emissions (Wagner et al., 2021). While the hypothesis of emissions being the primary driver of biases is often suggested, it does not seem to apply in the specific case of ARP7-RACM. Despite using the same emissions, ARP7-RELACS and ARP7-RACM produce CO fields that exhibit a nearly constant difference throughout the year.

3.2.4 Nitrogen species

Fig. 12 presents the zonal distributions of NO₂ from our simulations and their biases relative to the EAC4 reanalysis. NO₂ is a relatively short-lived species, and its emissions are tightly linked to anthropogenic sources. Consequently, its zonal distribution is maximal at mid-latitudes in the northern hemisphere. This is well captured with the ARP7-RELACS and ARP7-RACM simulations while in parallel the REPROBUS-based simulations show a clear negative bias over these latitudes. However, we observe lower concentrations in the model between 40S and 40N close to the surface which could be attributed to lower biomass burning emissions in this region. In contrast, the RELACS and RACMOBUS schemes produce too much NO₂ over the northern high latitudes, up to 600 hPa. The positive bias is more pronounced in ARP7-RELACS than in ARP7-RACM. We analyse further differences between RELACS and RACM below when discussing the NO₂ annual cycles.

In the free troposphere, the biases with the reanalysis have opposite signs in the 40S-40N band, with negative biases inside and positive biases outside of it. This could result from differences in NO_x emissions from lightning, which are a major primary source over the tropics and southern mid-latitudes or from aircraft emissions which can dominate in northern mid-latitudes depending on the season (Shah et al., 2023). Note that lightning emissions have been here adjusted (see section 3.3.1) and that aircraft emissions are not considered. We could not find any specific details on how the reanalysis deals with these types of emissions in Inness et al. (2019a). The differences observed between our simulations and the reanalysis may also be the signature of diverse NO_x sources from the stratosphere.



Here again, as for CO, the diversity of ARPEGE-Climat NO₂ zonal distributions can only be attributed to the chemistry scheme or to quantity transported from the stratosphere, as all other contributions are the same. The higher ARP7-RELACS NO₂ concentrations at 700 hPa appear consistent with larger ARP7-RELACS O₃ concentrations at these levels, where the chemical regime is expected to be NO_x limited.

555 Annual cycles of NO₂ are shown in Fig. 13. At 700 hPa, in all four latitude bands, the annual cycles from the reanalysis are flatter than those from our simulations, with winter maxima in the extratropics much larger in the ARP7-RELACS and ARP7-RACM simulations. Undoubtedly, the higher NO_x emissions in our simulations from the CEDS inventory contribute to this. Furthermore, as we noted in section 3.2.1, Teyssèdre et al. (2007) had related such a bias to the lack of heterogeneous loss of N₂O₅ on tropospheric aerosols. Interestingly, Inness et al. (2019a) also show the positive impact of this reaction which
560 requires coupling between a gaseous chemistry scheme and an aerosol scheme. Additionally, Young et al. (2018) conclude that gas-aerosol interactions and heterogeneous chemistry are recent advances in understanding that have yet to be incorporated into most models. Meanwhile, measurements from the WINTER campaign showed that N₂O₅ hydrolysis accounted for 58% of the chemical loss of NO_x (see Williams et al. (2022) and references therein). At 700 hPa in the southern hemisphere, the annual peaks coincide with biomass burning activity, with the highest concentrations observed in the ARP7-RELACS simulation.

565 Concentrations of the reanalysis are lower at 500 hPa than at 700 hPa, but higher at 250 than at 500 hPa. This pattern is not present in the ARP7-REPRO and ARP7-RELACS simulations. The shape of their annual cycles at 500 hPa in the extratropics clearly shows the influence of the concentrations at 700 hPa, particularly during the November to February period in the northern extratropics, and their values are quite close to those of the reanalysis for most months. In contrast, at 250 hPa almost nearly each line of annual cycles appears to have its own distinct pattern. For example, in the extratropics at 250
570 hPa the reanalysis and the REPROBUS simulations exhibit clear annual cycles, though peaking 5 months apart. Meanwhile, the ARP7-RELACS and ARP7-RACM simulations show almost no annual cycle. The annual cycles from the REPROBUS simulations are also noteworthy in this context.

Regarding two additional aspects of the EAC4 reanalysis, Inness et al. (2019a) note that the NO₂ columns are generally underestimated, particularly in winter. However, the assimilation of this quantity has led to improved columns, especially during
575 winter when the lifetime of NO₂ is longer, compared to reanalyses that did not incorporate such data. They also emphasize the need for further validation of the NO₂ fields, highlighting the lack of representative observations at the EAC4 grid scale, to determine whether the wintertime underestimation stems from persistent model deficiencies or inaccuracies in emissions. Very recently, Horner et al. (2024) confirmed the critical lack of routine observations of the vertical distribution of tropospheric nitrogen oxides. They developed global tropospheric vertical profiles using four years of satellite data from the TROPOMI
580 instrument, covering five discrete vertical layers. These profiles were used to evaluate the representation of the global vertical distribution of tropospheric NO_x in the GEOS-Chem chemical transport model. Such a dataset would be invaluable for assessing both the strengths and weaknesses of reanalysis products and ARPEGE-Climat NO₂ concentrations.

Neither PAN nor HNO₃ are assimilated species in the EAC4 reanalysis. However, since they serve as good tracers of total nitrogen oxides, we analyse them in comparison to the reanalysis. Figure 14 presents their zonal distributions in the ARP7-
585 RELACS and ARP7-RACM simulations.

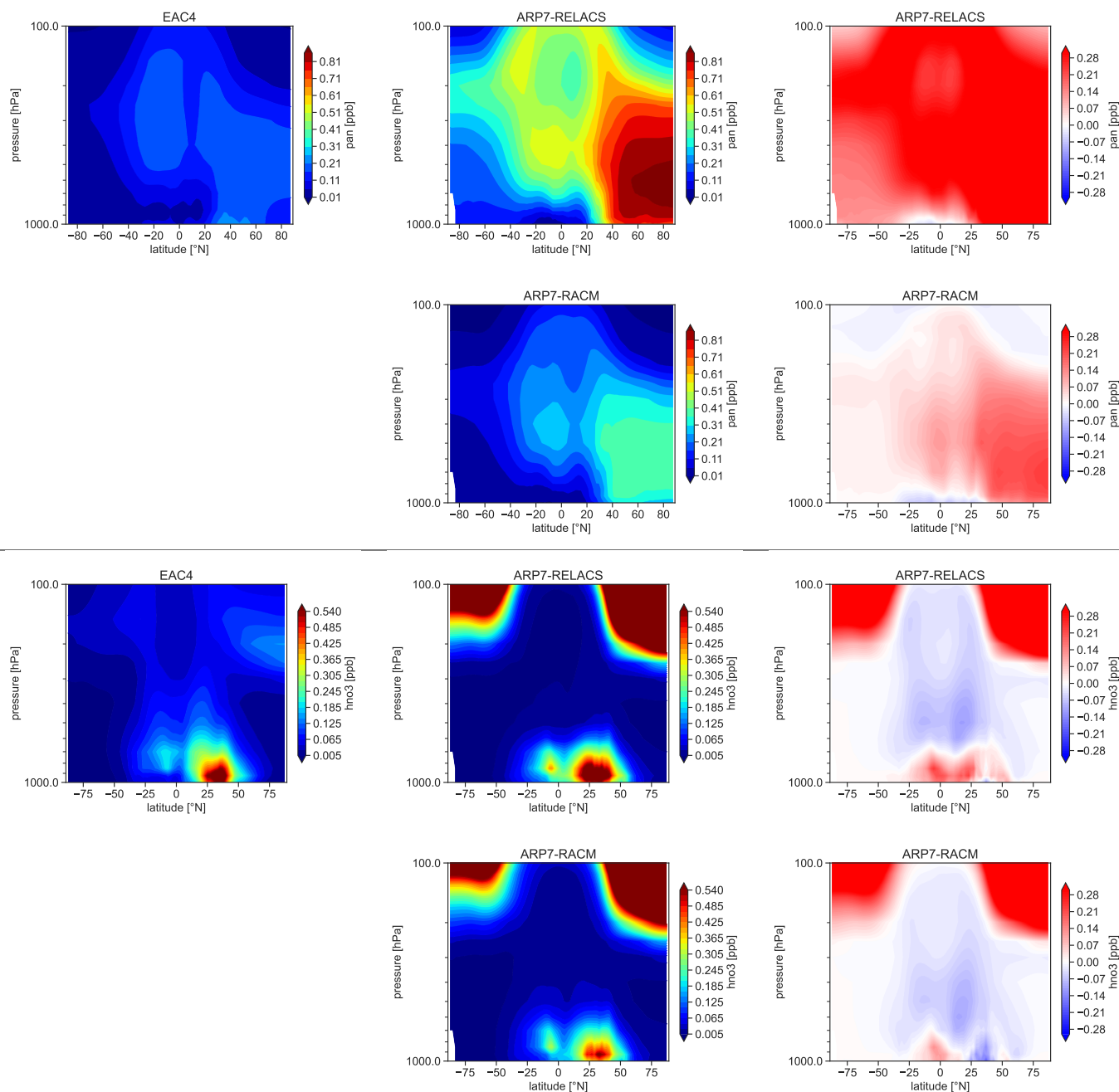


Figure 14. Mean (2003-2014) zonal climatology of tropospheric (surface to 100 hPa) PAN (ppb), first two rows, and HNO₃, last two rows. First column: EAC4 reanalysis (absolute values), second column ARP7-RELACS and ARP7-RACM simulations, third column differences (simulation minus EAC4).



The spatial distributions in both the reanalysis and simulations exhibit similar patterns for PAN. As expected, PAN mixing ratios are higher in the Northern Hemisphere, where pollution levels are greater, and in the tropical mid-troposphere, where it is transported to upper levels and lower temperature lengthens its lifetime. However, the maximal values appears close to the surface in the reanalysis and not in our simulations. Moreover, mixing ratios vary significantly across the three zonal plots.

590 In the northern hemisphere, up to 500 hPa, ARP7-RACM mixing ratios are approximately twice those of the reanalysis and ARP7-RELACS values are four to five times higher. While these elevated mixing ratios may seem problematic, several studies provide context. For instance, in-situ mountain sites have measured climatological PAN mixing ratios exceeding 200 ppt year-round, and even surpassing 1000 ppt in winter when NMVOCs accumulate under low-photochemistry conditions (Fiore et al. (2018)). Model simulations struggle to reproduce these observed values, with some underestimating them significantly and
595 others overestimating them (see Fiore et al. (2018), Fig. S1). Comparatively, the climatological annual cycles of ARP7-RACM are within the range of the 14 models presented in Fiore et al. (2018) at five mountain sites, whereas ARP7-RELACS exhibits too large PAN concentrations of about 1200-1400 ppt (not shown) at the sites with altitudes around 3000 m. Regarding its zonal distribution, HNO₃ in ARP7-RELACS shows higher concentrations in the boundary layer compared to ARP7-RACM (as also noted in the time series at 850 hPa; see Fig. 7), while mixing ratios are similar throughout the rest of the atmosphere.

600 To our knowledge, PAN and HNO₃ from the RELACS scheme have not been evaluated in any published studies. However, the RACMOBUS scheme has been extensively analysed in Williams et al. (2022) over the United States. Williams et al. (2022) evaluated outputs, including PAN and HNO₃, from three chemical schemes embedded in the IFS model, among which the RACMOBUS scheme. We note that in this study RACMOBUS had undergone several updates compared to our version, particularly in terms of heterogeneous NO_x chemistry. Their findings highlight that differences among the three schemes are
605 most pronounced during winter for both PAN and HNO₃. They report an overall underestimation of PAN formation due to insufficient organic precursors in summer, whereas in winter, RACMOBUS overestimates PAN concentrations relative to other chemistry modules due to excessive PAN stability. They also emphasize the need for further measurements to better constrain wintertime variations and acknowledge the significant uncertainty in wintertime N₂O₅ conversion into HNO₃, which leads to a general overestimation of HNO₃. Furthermore, Williams et al. (2022) note that HNO₃ modelling performs better in the
610 reanalysis than in the three updated chemistry schemes.

3.2.5 Hydroxyl radical

The zonal mean climatology of OH in the troposphere is shown in Fig. 15. ARP7-RACM and ARP7-RELACS overestimate the reanalysis from the surface up to 600 hPa, and underestimate it aloft, with negative biases larger than positive one. Biases are more pronounced in the tropical troposphere where OH concentrations are the highest. Similar biases had been identified
615 for the RELACS scheme in Teyssède et al. (2007). Negative biases in the lower troposphere in the REPROBUS simulations reveal the absence of chemistry there.

Apart from the uncertainties in OH diagnostics inherent to its very short lifetime, the following studies shed some light on OH modelling. Using a novel approach based on OH precursor observations and a box model, Zhao et al. (2023) concluded that the overestimation of OH in models is primarily due to their underestimation of CO and total ozone column (TOC), as well

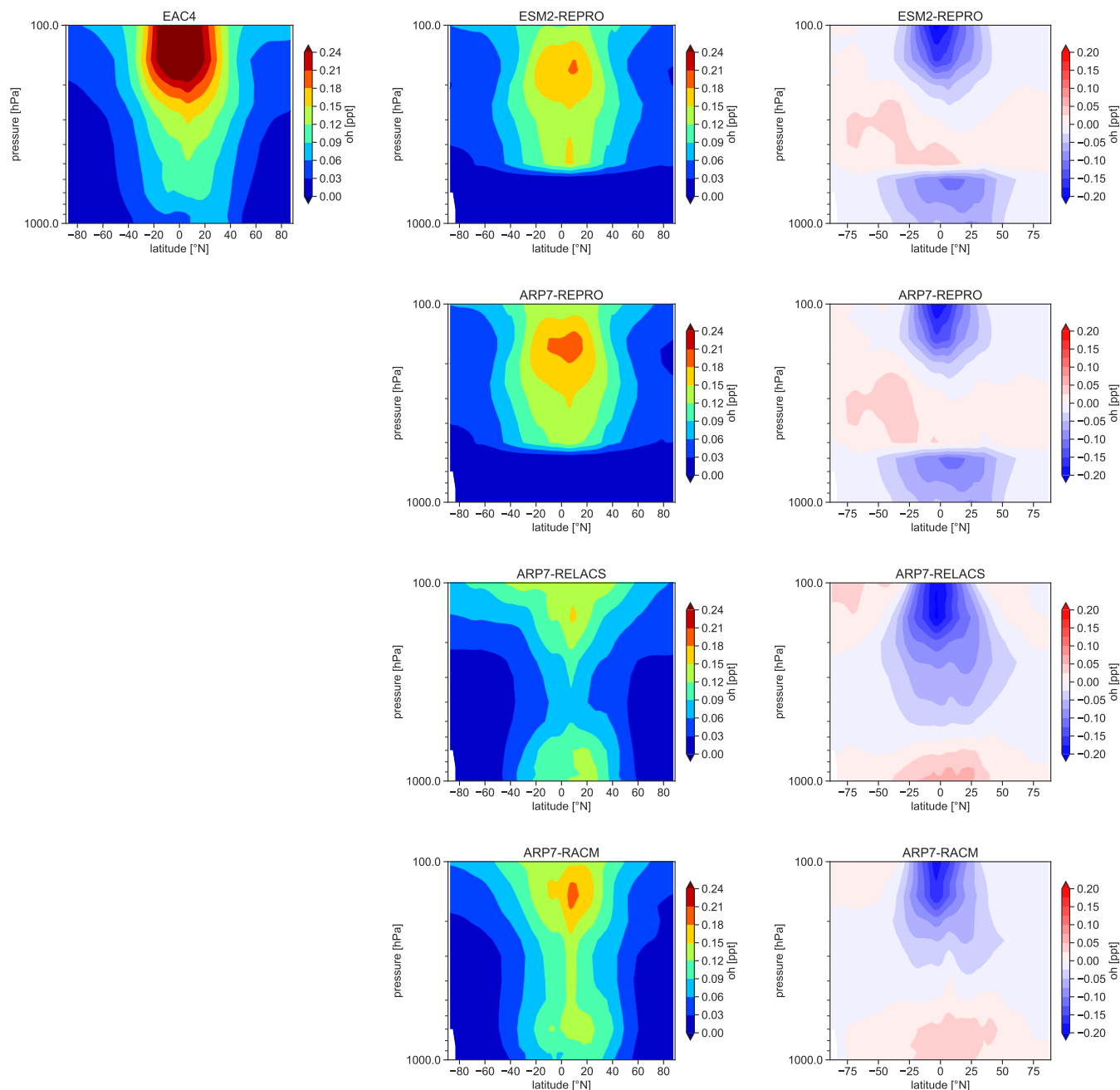


Figure 15. Mean (2003-2014) zonal climatology of tropospheric (surface to 100 hPa) OH (ppt). Same presentation as in Fig. 8.



as their overestimation of NO_2 . However, at the regional scale, they noted that biases in H_2O can lead to even larger OH biases than those induced by CO. Despite these insights, their study does not provide a clear path forward for improving our OH fields. In fact, despite similar TOC but significantly different CO levels, ARP7-RELACS and ARP7-RACM produce relatively similar OH fields.

Many other studies have investigated OH modelling, particularly its sensitivity to different atmospheric species, though a comprehensive understanding remains elusive (Stevenson et al. (2020) and references therein). For instance, when analysing ACCMIP simulations, Voulgarakis et al. (2013) found that MOCAGE, using a version of the RACMOBUS scheme similar to that in ARPEGE-Climat 7.0.1, had the highest OH concentrations among all ACCMIP models. They attributed this to the model's exceptionally high NMVOC emissions, whereas Naik et al. (2013) suggested that such high NMVOC emissions should lead to a greater OH sink. Wild et al. (2020) reported that model intercomparison studies reveal substantial variability in OH estimates, reflecting underlying uncertainties. However, they noted that little progress has been made in identifying the root causes of these discrepancies or the weaknesses in process representation that could lead to improved models. In their sensitivity study, they examined key variables to quantify the sources of model diversity in tropospheric O_3 and OH. While they identified clear sensitivities for O_3 , they concluded that the large inter-model differences in OH reactivity remain a major challenge, requiring further investigation.

3.3 Sensitivity studies

3.3.1 Tropospheric ozone and lightning NO_x emissions

The tropospheric ozone burden remains a key metric for model evaluation, as it provides a reliable indicator of the oxidizing capacity of the troposphere. For both the RELACS and RACMOBUS mechanisms, we carried out paired simulations in which lightning NO_x emissions were varied from $2.5 \text{ Tg(N) yr}^{-1}$, the value used throughout the rest of this study, to the more commonly adopted value of 5 Tg(N) yr^{-1} . This sensitivity test was designed to evaluate the model's response to lightning NO_x emissions, which are known to significantly influence the free troposphere, where the ozone production regime is typically NO_x -limited (Verma et al., 2021). We calculated the tropospheric ozone burden using the inclusive pressure definition, in accordance with the methodology described by Griffiths et al. (2021b). This approach involves summing the ozone mass in all grid cells located entirely below the tropopause, as well as those intersected by it. The tropopause pressure was diagnosed online using the temperature lapse-rate method, consistent with the WMO definition. Figure 16 illustrates the evolution of the tropospheric ozone burden throughout the simulation period (1979–2014). It also includes burden estimates from several multi-model studies, namely ACCMIP (Young et al., 2013) and CMIP6 (Griffiths et al., 2021b), as well as the estimate from the Tropospheric Ozone Assessment Report (TOAR, Gaudel et al., 2018).

The time series of tropospheric ozone burden in the $2.5 \text{ Tg(N) yr}^{-1}$ simulations, ARP7-RELACS and ARP7-RACM, show good agreement with other modeling studies and literature estimates, and successfully reproduce the observed positive trend. The ARP7-RELACS burden is slightly higher than that of ARP7-RACM, due to enhanced ozone concentrations in the lower troposphere, as shown in Fig. 8. Both ARP7-RELACS and ARP7-RACM burdens lie at the lower end of the CMIP6 model

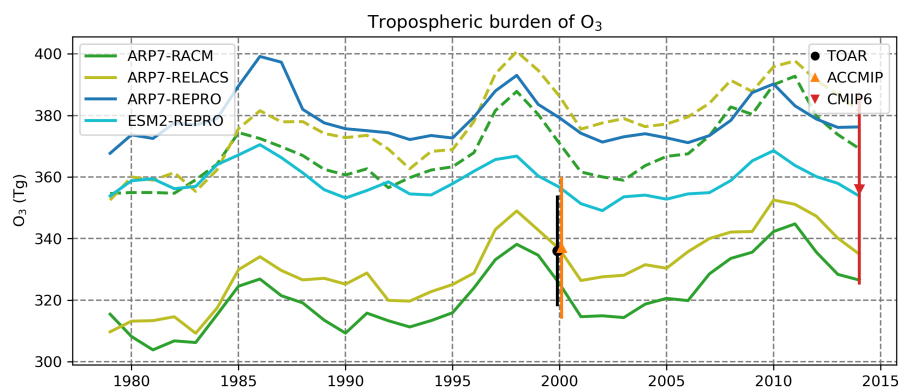


Figure 16. Time-series of the O_3 tropospheric burden (in Tg) for different ARPEGE-Climat 7.0.1 simulations, using the RELACS, RACMOBUS and REPROBUS chemical schemes, for two different totals of lightning NO_x emissions (2.5 and 5 Tg(N) yr^{-1} , respectively in solid lines and dotted lines). The estimates, presented as points, are from TOAR, along with ACCMIP and CMIP6 multi-model means (see text for more details).

range for 2014; however, it is worth noting that Griffiths et al. (2021b) reported a general positive bias in CMIP6 models relative to the most recent observationally constrained estimates of the tropospheric ozone burden.

655 In the sensitivity simulations using 5 Tg(N)·yr^{-1} lightning NO_x emissions, tropospheric ozone burdens increase by approximately 40 Tg compared to the $2.5 \text{ Tg(N)·yr}^{-1}$ simulations. Although 5 Tg(N)·yr^{-1} is the most commonly used estimate for annual lightning NO_x emissions, in our case, it results in an overestimation of the tropospheric ozone burden. Since the $2.5 \text{ Tg(N)·yr}^{-1}$ value remains within the accepted uncertainty range ($5 \pm 3 \text{ Tg(N)·yr}^{-1}$), it appears appropriate for continued use in ARPEGE-Climat, though this calibration may need to be revised when future versions include aircraft emissions (estimated
660 at $0.7 \text{ Tg(N)·yr}^{-1}$ around the year 2000, with 60% emitted above 700 hPa (Terrenoire et al., 2022)).

Our findings highlight the strong sensitivity of the tropospheric ozone burden to lightning NO_x emissions. This is consistent with results from Archibald et al. (2020a), who reported substantial sensitivity in their model, where a pronounced positive ozone bias aligned with regions of intense lightning NO_x emissions, based on a total of 6 Tg(N)·yr^{-1} . Given the inherent uncertainties in estimating lightning NO_x emissions, calibrating total annual emissions within the accepted range offers a
665 promising approach to better constrain the troposphere's oxidizing capacity.

Finally, we evaluated the tropospheric ozone burden in the simulations including only upper-tropospheric and stratospheric chemistry (ARP7-REPRO and ESM2-REPRO). As expected, in the absence of emissions, these simulations fail to capture the observed positive trend. Their tropospheric burdens exhibit a positive bias, although they remain within the range of the 5 Tg(N)·yr^{-1} simulations discussed previously. This level of bias is acceptable for a model configuration that does not include
670 tropospheric processes, despite the lack of realistic spatial distributions or seasonal variability. The ARP7-REPRO simulation shows the largest bias, which is linked to higher ozone mixing ratios in the lower stratosphere—particularly in the northern



hemisphere (see Fig. 3). In this setup, ozone concentrations in the lower troposphere are nudged towards values representative of the upper troposphere, which are themselves predominantly influenced by stratosphere–troposphere exchange processes.

3.3.2 Sensitivity to the model level relaxation for CH₄

Figure 17 presents sensitivity tests on the relaxation level of well-mixed gases in the troposphere, including GHGs and ODSs. The global mean concentrations of CH₄ and N₂O at 1000 hPa and 850 hPa are shown for the ESM2-REPRO and ARP7-REPRO simulations, where the relaxation level is set at 560 hPa (model level 66), and for the ARP7-RELACS and ARP7-RACM simulations, where it is set at the model level 80 (i.e., 900 hPa in a standard atmosphere). Additionally, two extra RELACS simulations are included: one where the relaxation level is set at the lowest model level (91, or ~1013 hPa), a setting typically used in ESMs (F. O'Connor, personal communication), and another simulation where the relaxation level is also level 91 but includes CH₄ emissions.

Several conclusions emerge: First, ARPEGE-Climat 7.0.1 exhibits clear malfunctions when the relaxation level is set at 1013 hPa, as evidenced by the rapid decline in both CH₄ and N₂O concentrations within a few years. Second, CH₄ concentrations in ARP7-RELACS are lower than in ESM2-REPRO, with the difference partially reduced when CH₄ emissions are included (red line). However, since ARP7-RELACS is considered a concentration-driven simulation, and the methane levels obtained in ARP7-RELACS appear satisfactory without signs of drift, we chose not to include CH₄ emissions in this simulation. In the ARP7-RACM simulations the choice was made to consider CH₄ emissions in order to get a more realistic CH₄ zonal gradient and possibly improve the CO distribution, but this had only a minor impact in the end. This explains why ARP7-RACM CH₄ values are higher than those of ARP7-RELACS. Finally, Figure 17 shows that although N₂O concentrations at 1000 and 850 hPa in ARP7-RACM quickly reach equilibrium within a few months, this equilibrium level appears too low, warranting further investigation. These too low concentrations had already been underlined in the analysis of stratospheric N₂O (see Section 3.1.3).

4 Conclusions

This paper aims at evaluating and comparing three chemistry schemes of various complexity implemented in the latest version of the CNRM atmospheric climate model, ARPEGE-Climat 7.0.1, which is based on the operational NWP model at Météo-France and will be used for future scientific applications. These schemes include the upper tropospheric-stratospheric REPROBUS scheme and two tropospheric-stratospheric schemes of increasing complexity, RELACS and RACMOBUS.

We conducted AMIP-type simulations (1979–2014) using these three schemes, called ARP7-REPRO, ARP7-RELACS, and ARP7-RACM, with all CMIP6 forcings, and we evaluated several key chemical species. The model setup was selected based on sensitivity tests, particularly regarding lightning emissions (2.5 Tg(N) yr⁻¹) and the relaxation level of GHGs and ODSs (900 hPa), which we present in this paper.

In the stratosphere, we evaluated our simulations against the AMIP CMIP6 simulation of the CNRM-ESM2-1 model (Michou et al., 2020a; Séférian et al., 2019; Voldoire et al., 2019; Roehrig et al., 2020; Decharme et al., 2019), which used the

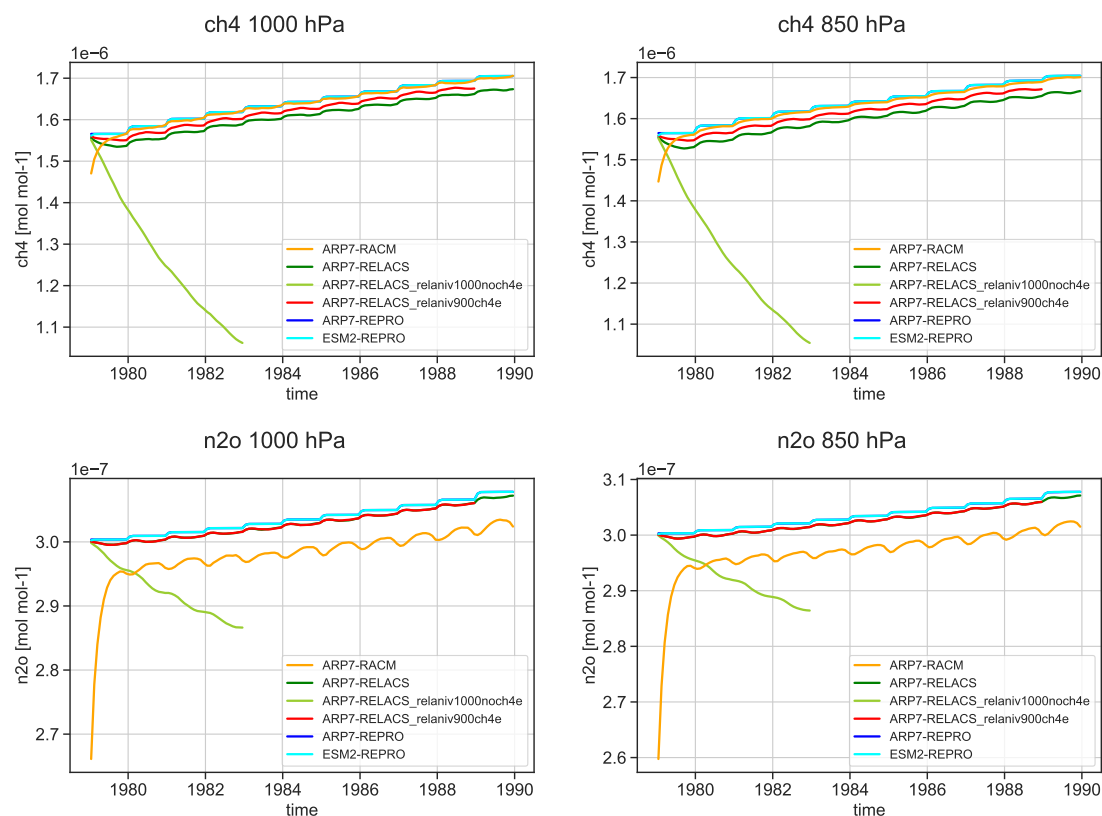


Figure 17. Global mean time series of CH₄ (first row) and N₂O (second row), at 1000 hPa (left column) and at 850 hPa (right column) for the ESM2-REPRO (in cyan), ARP7-REPRO (in dark blue), ARP7-RELACS (in green), ARP7-RACM (in orange) simulations, along with two sensitivity tests, one relaxing GHGs and ODSs at 1000 hPa (in yellow green), and another at 900 hPa considering CH₄ emissions (in red). See text for details.

REPROBUS scheme and has been extensively documented. This simulation, referred to here as ESM2-REPRO, provides a useful benchmark.

Total column ozone (TCO) in ARPEGE-Climat 7.0.1 is well represented, both in terms of trends across different latitudinal bands and in its zonal climatology (1990–1999). Notably, two biases identified in the ESM2-REPRO simulations, excessive TCO in the tropics and underestimated TCO at northern latitudes, are reduced in all three ARP7 simulations, likely due to improved stratospheric circulation in the climate model. Additionally, the increasing influence of the tropospheric ozone column on TCO is satisfactorily captured. The zonal climatology of O₃ is also improved in the top of the stratosphere in ARP7-REPRO and ARP7-RELACS.



Regarding H₂O, the new simulations exhibit greater stratospheric moistening, with more water vapor entering the stratosphere due to higher cold point tropopause temperatures, an improvement. However, differences in the CH₄ oxidation process between the ARP7-RELACS and ARP7-RACM simulations require further investigation.

715 Other stratospheric species in ARPEGE-Climat 7.0.1 also require further investigation. In both ARP7-RELACS and ARP7-RACM, the zonal climatology of HNO₃ at high latitudes is problematic. Moreover, the REPROBUS scheme produces unexpectedly diverse HNO₃ evolutions in the three ARP7 simulations, despite the underlying climatology being very similar. Additionally, N₂O and HCl in ARP7-RACM are consistently underestimated throughout the stratosphere, possibly due to conservation treatments and excessive tropospheric scavenging of HCl.

720 For the troposphere, we used the EAC4 CAMS reanalysis as a reference dataset, analyzing time series, climatological zonal means, and annual cycles at different altitudes and latitudinal bands. The RELACS and RACMOBUS schemes result in significantly different CO and PAN concentrations across the troposphere.

In particular, CO is systematically underestimated in the ARP7-RACM simulation, displaying a persistent negative bias throughout the evaluation period. We ruled out low emissions as the cause of this bias, since ARP7-RACM and ARP7-RELACS
725 use identical emissions, and CO concentrations in ARP7-RELACS are satisfactory. Other potential factors, such as OH distributions, were also examined, but they appear similar in both simulations and do not exhibit major biases. However, OH remains a key source of model variability and an area requiring further understanding. Additional work is needed to resolve the CO distribution issue in ARP7-RACM.

Regarding nitrogen species, the mean annual climatologies of ARP7-RACM and ARP7-RELACS show excessive concentrations in the lower troposphere at high northern latitudes, reaching up to 600 hPa. Analysing time series and annual cycles
730 reveals that while model concentrations align well with reanalysis data from April to October, they become significantly overestimated during the rest of the year, with even larger biases in the ARP7-RELACS simulation. This issue had already been identified specifically for the RELACS chemical scheme and was attributed, at least in part, to the absence of heterogeneous loss of N₂O₅ on tropospheric aerosols. Addressing this limitation provides a clear avenue for model improvement. Higher up
735 in the troposphere, we observed the influence of localized emissions, such as those from lightning, and emphasized the need for better observational constraints on the vertical distribution of NO₂, beyond just column measurements. PAN concentrations in the ARP7-RELACS simulation also exhibit problematic behaviour, requiring further investigation.

Moreover, regarding tropospheric O₃, both tropospheric schemes produce similarly satisfactory concentrations throughout the troposphere, though some differences arise, notably due to stratosphere-to-troposphere transport. Time series at 850 hPa
740 closely follow those of the reanalysis, particularly in the ARP7-RACM simulation. This is also true for climatological annual cycles in the southern hemisphere, while in the northern hemisphere, our simulations appear reliable, considering the deficiencies identified in the EAC4 reanalysis.

Finally, an evaluation was conducted in terms of tropospheric burden of ozone, in addition to a sensitivity test to the amount of NO_x emitted from lightning. It was found that both the RACMOBUS and RELACS tropospheric schemes manage to reproduce the performances of other models and latest literature on tropospheric ozone, and even more so when scaling down the
745 lightning NO_x emissions from 5 to 2.5 Tg(N).yr⁻¹.



In summary, this work builds on a community-wide interest in exploring atmospheric composition resulting from chemistry schemes of varying complexity embedded within a single climate model framework. In our case, using “cousin” schemes, we obtain broadly similar climatological ozone characteristics in both the stratosphere and troposphere. However, the climatological features of several species involved in ozone chemistry differ significantly. These discrepancies may be partially attributed to the varying levels of chemical complexity, but also to the intrinsic non-linearity of atmospheric chemistry, as well as potential compensating biases or errors within the model components themselves. Brown-Steiner et al. (2018), who analysed three chemical mechanisms of differing complexity within the CESM climate model, reached conclusions closely aligned with ours: while their schemes produced comparable large-scale patterns of surface ozone, this consistency did not extend to nitrogen oxides or CO, particularly during winter and in regions with strong biogenic emissions. They also emphasized that model development choices can have lasting impacts on simulation results, something that may also apply to our case. Importantly, Brown-Steiner et al. (2018) highlight the scientific value of evaluating and using chemistry schemes of differing complexity within a shared modelling framework.

In line with this perspective, our study represents a substantial advancement in the representation of atmospheric chemistry in the CNRM climate model compared to previous versions. We have shown that this chemistry is well suited to the challenges of representing chemistry-climate interactions in an Earth system model, and we have identified key areas for future development that will further improve the model. Additionally, coupling the outputs of gaseous chemistry to the aerosol scheme TACTIC (Michou et al., 2020b), which is also embedded in ARPEGE-Climat 7.0.1, would enhance interactivity. Future developments could further incorporate interactive biogenic emissions, for example through the use of the MEGAN model (Guenther et al., 2012). Furthermore, with the explicit representation of CH₄ sinks via tropospheric oxidation, ARPEGE-Climat 7.0.1 now supports CH₄ emission-driven simulations, in which emissions are prescribed instead of surface concentrations. However, this increased complexity comes with higher computational costs. Moreover, as Young et al. (2018) point out, “it is unclear whether the implementation of complex chemistry schemes would improve the representation of global-scale ozone distributions and the chemistry relevant to climate.”. We will carefully evaluate these factors in our future development efforts.

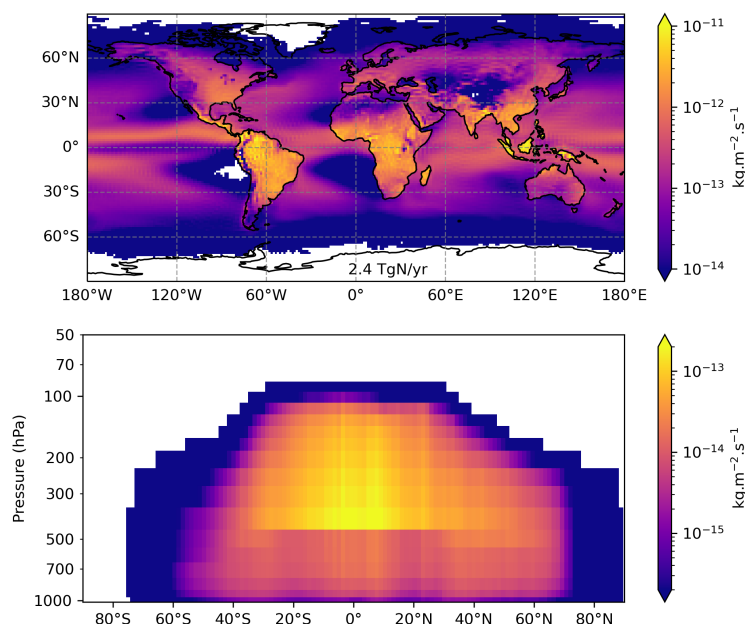


Figure A1. Emissions fluxes of NO_x from lightning ($\text{kg m}^{-2} \text{s}^{-1}$). Top row: vertically integrated. Bottom row: zonally averaged. Note that the two plots use different logarithmic scales.

770 *Code availability.* Météo-France belongs to the ACCORD consortium (<http://www.accord-nwp.org/>, last access 22 April 2025) for the development of forecasting systems for numerical weather prediction (NWP), within which it cooperates on the development of a shared system of model codes. ACCORD was established in 2021 and initially brought together members of the consortia ALADIN, LACE, and HIRLAM. The newly developed ARPEGE-Climat code forms part of the shared system of model codes. According to the ACCORD Memorandum of Understanding and in particular its annexes IX and X, for non-commercial research purposes, code can be distributed upon signature of

775 a licence agreement (see <http://www.accord-nwp.org/?ACCORD-MoU-2021-2025>, for conditions, last access: 22 April 2025). The newly surface developed code will be included in the next official version of SURFEX v9.0. The official release of SURFEX v8.1 offline is available at <https://www.umr-cnrm.fr/surfex/spip.php?rubrique141> (last access: 22 April 2025).

Data availability. The CAMS reanalysis can be downloaded on the Atmosphere Data Store (<https://ads.atmosphere.copernicus.eu/>). Forcing datasets as well as CNRM-ESM2-1 and other CMIP6 simulations are available through the ESGF platform (<https://esgf-node.ipsl.upmc.fr/search/cmip6-ipsl/>). The simulation output (netcdf files) used in this paper are available in open access on the IPSL thredds platform (<https://thredds-su.ipsl.fr/thredds/catalog/ESM2025/Simulations/Cussac2025/catalog.html>).

Appendix A: Additional figures

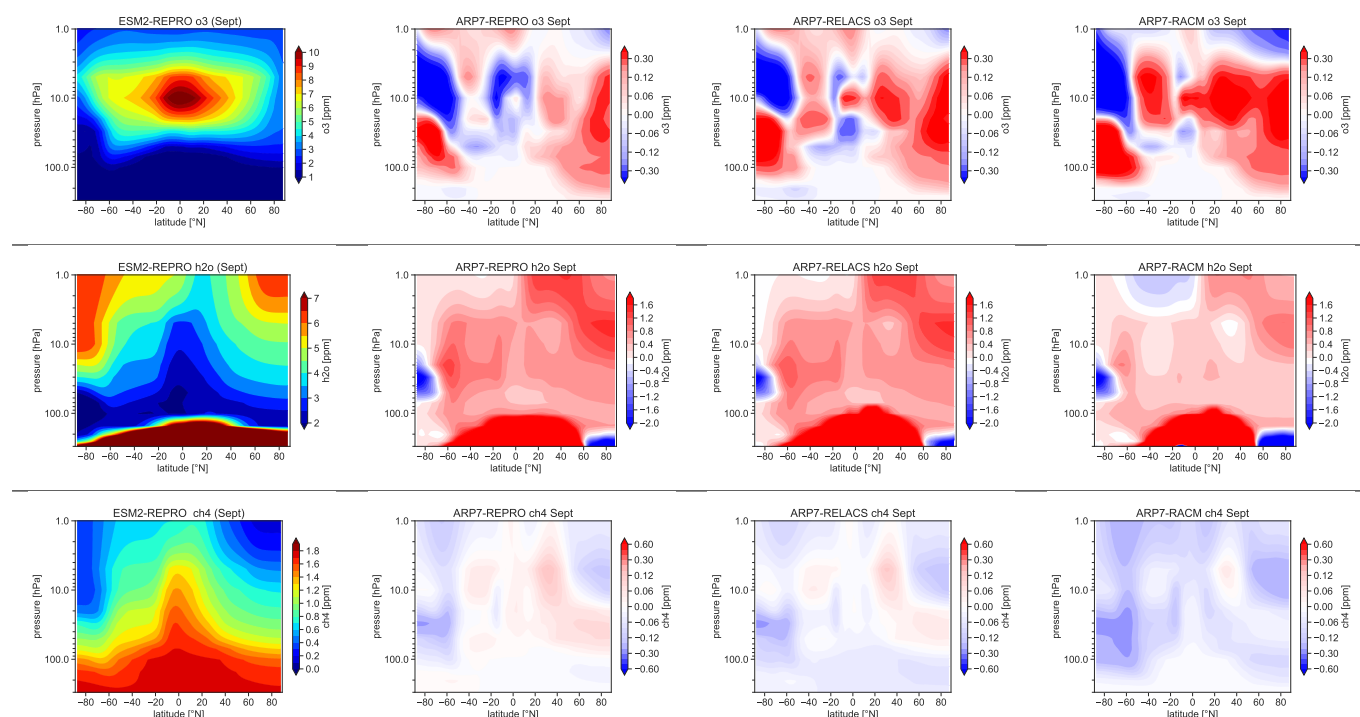


Figure A2. Mean zonal climatology (1979-2014, 300 to 1 hPa) of various species, in September (see March in main text). First line: O_3 (ppm), second line: H_2O (ppm), third line: CH_4 (ppm). First column ESM2-REPRO simulation (absolute values) and subsequent columns differences (simulation minus ESM2-REPRO), for the ARP7-REPRO, ARP7-RELACS, and ARP7-RACMOBUS simulations.

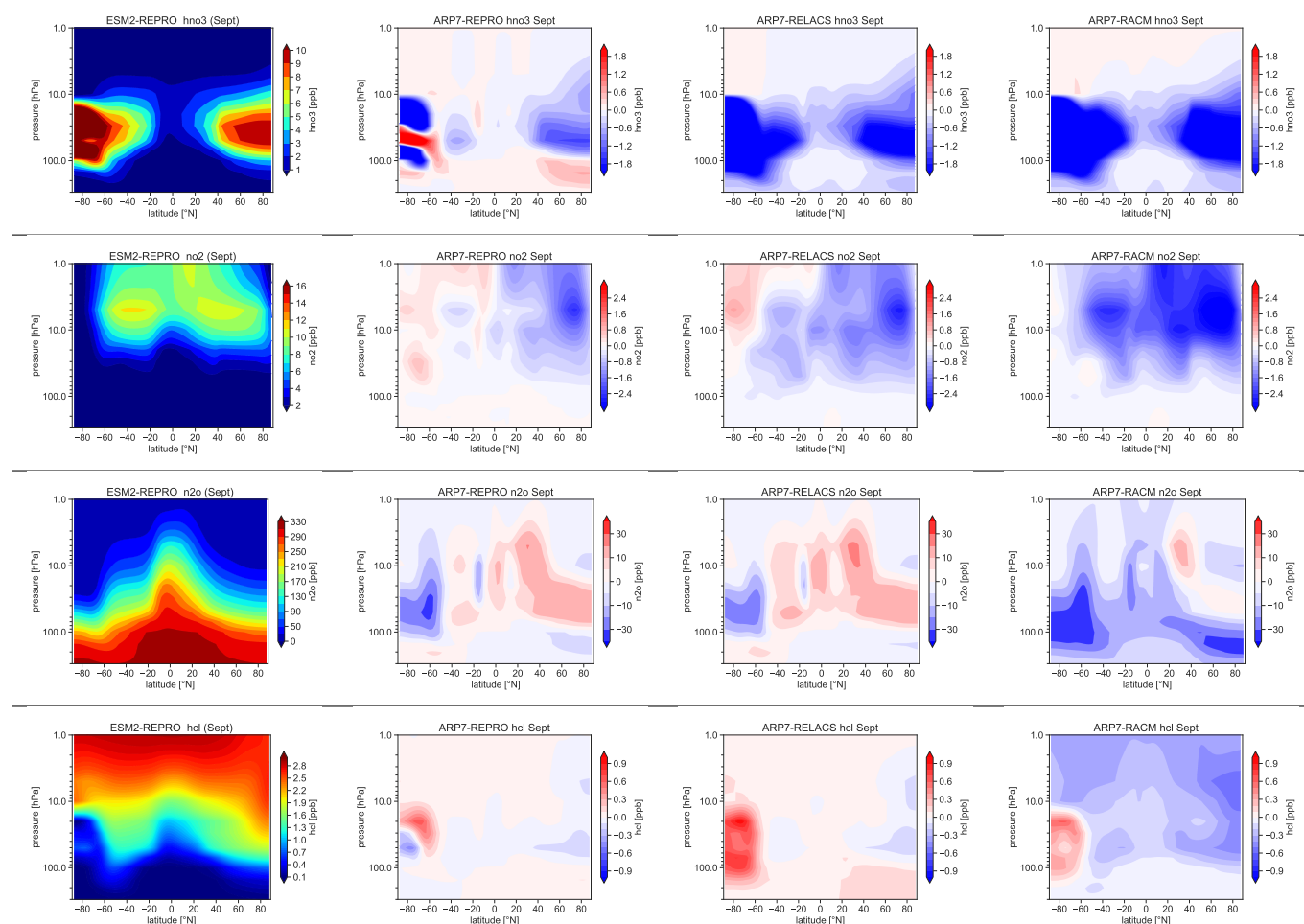


Figure A3. Mean zonal climatology (1979-2014, 300 to 1 hPa) of various species, in September (see March in main text). First line: HNO_3 (ppb), second line: NO_2 (ppb), third line: N_2O (ppb), fourth line: HCl (ppb). First column ESM2-REPRO simulation (absolute values) and subsequent columns differences (simulation minus ESM2-REPRO), for the ARP7-REPRO, ARP7-RELACS, and ARP7-RACM simulations.



Author contributions. MC and MM implemented the chemistry schemes into the atmospheric component of the latest version of the CNRM climate model, with valuable input from all co-authors. They also designed the evaluation presented in the paper and wrote the paper, with
785 contributions from all co-authors.

Competing interests. Some authors are members of the editorial board of journal Geoscientific Model Development.

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790 (Andela et al., 2024b, doi:10.5281/zenodo.3387139) and the ESMValTool code (Andela et al., 2024a, doi:10.5281/zenodo.3401363). The authors also acknowledge the support of the entire team in charge of the CNRM climate models. Supercomputing time was provided by the Météo-France/DSI supercomputing centre.



References

- Andela, B., Broetz, B., de Mora, L., Drost, N., Eyring, V., Koldunov, N., Lauer, A., Mueller, B., Predoi, V., Righi, M., Schlund, M., Vegas-Regidor, J., Zimmermann, K., Adeniyi, K., Arnone, E., Bellprat, O., Berg, P., Bock, L., Bodas-Salcedo, A., Caron, L.-P., Carvalhais, N., Cionni, I., Cortesi, N., Corti, S., Crezee, B., Davin, E. L., Davini, P., Deser, C., Diblen, F., Docquier, D., Dreyer, L., Ehbrecht, C., Earnshaw, P., Gier, B., Gonzalez-Reviriego, N., Goodman, P., Hagemann, S., Hardacre, C., von Hardenberg, J., Hassler, B., Heuer, H., Hunter, A., Kadow, C., Kindermann, S., Koirala, S., Kuehbachner, B., Lledó, L., Lejeune, Q., Lembo, V., Little, B., Loosveldt-Tomas, S., Lorenz, R., Lovato, T., Lucarini, V., Massonnet, F., Mohr, C. W., Amarjiit, P., Pérez-Zanón, N., Phillips, A., Russell, J., Sandstad, M., Sellar, A., Senftleben, D., Serva, F., Sillmann, J., Stacke, T., Swaminathan, R., Torralba, V., Weigel, K., Sarauer, E., Roberts, C., Kalverla, P., Alidoost, S., Verhoeven, S., Vreede, B., Smeets, S., Soares Siqueira, A., Kazeroni, R., Potter, J., Winterstein, F., Beucher, R., Kraft, J., Ruhe, L., Bonnet, P., and Munday, G.: ESMValTool, <https://doi.org/10.5281/zenodo.12654299>, 2024a.
- Andela, B., Broetz, B., de Mora, L., Drost, N., Eyring, V., Koldunov, N., Lauer, A., Predoi, V., Righi, M., Schlund, M., Vegas-Regidor, J., Zimmermann, K., Bock, L., Diblen, F., Dreyer, L., Earnshaw, P., Hassler, B., Little, B., Loosveldt-Tomas, S., Smeets, S., Camphuijsen, J., Gier, B. K., Weigel, K., Hauser, M., Kalverla, P., Galytska, E., Cos-España, P., Pelulessy, I., Koirala, S., Stacke, T., Alidoost, S., Jury, M., Sényi, S., Crocker, T., Vreede, B., Soares Siqueira, A., Kazeroni, R., Hohn, D., Bauer, J., Beucher, R., Benke, J., Martin-Martinez, E., and Cammarano, D.: ESMValCore, <https://doi.org/10.5281/zenodo.12638409>, 2024b.
- Archibald, A. T., O'Connor, F. M., Abraham, N. L., Archer-Nicholls, S., Chipperfield, M. P., Dalvi, M., Folberth, G. A., Dennison, F., Dhomse, S. S., Griffiths, P. T., Hardacre, C., Hewitt, A. J., Hill, R. S., Johnson, C. E., Keeble, J., Köhler, M. O., Morgenstern, O., Mulcahy, J. P., Ordóñez, C., Pope, R. J., Rumbold, S. T., Russo, M. R., Savage, N. H., Sellar, A., Stringer, M., Turnock, S. T., Wild, O., and Zeng, G.: Description and evaluation of the UKCA stratosphere–troposphere chemistry scheme (StratTrop vn 1.0) implemented in UKESM1, *Geoscientific Model Development*, 13, 1223–1266, <https://doi.org/10.5194/gmd-13-1223-2020>, 2020a.
- Archibald, A. T., O'Connor, F. M., Abraham, N. L., Archer-Nicholls, S., Chipperfield, M. P., Dalvi, M., Folberth, G. A., Dennison, F., Dhomse, S. S., Griffiths, P. T., Hardacre, C., Hewitt, A. J., Hill, R. S., Johnson, C. E., Keeble, J., Köhler, M. O., Morgenstern, O., Mulcahy, J. P., Ordóñez, C., Pope, R. J., Rumbold, S. T., Russo, M. R., Savage, N. H., Sellar, A., Stringer, M., Turnock, S. T., Wild, O., and Zeng, G.: Description and evaluation of the UKCA stratosphere–troposphere chemistry scheme (StratTrop vn 1.0) implemented in UKESM1, *Geoscientific Model Development*, 13, 1223–1266, <https://doi.org/10.5194/gmd-13-1223-2020>, 2020b.
- Austin, J., Scinocca, J., Plummer, D., Oman, L., Waugh, D., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M., Cugnet, D., Dameris, M., Dhomse, S., Eyring, V., Frith, S., Garcia, R. R., Garny, H., Gettelman, A., Hardiman, S. C., Kinnison, D., Lamarque, J. F., Mancini, E., Marchand, M., Michou, M., Morgenstern, O., Nakamura, T., Pawson, S., Pitari, G., Pyle, J., Rozanov, E., Shepherd, T. G., Shibata, K., Teyssède, H., Wilson, R. J., and Yamashita, Y.: Decline and recovery of total column ozone using a multimodel time series analysis, *Journal of Geophysical Research: Atmospheres*, 115, <https://doi.org/10.1029/2010JD013857>, 2010.
- Ayarzagüena, B., Polvani, L. M., Langematz, U., Akiyoshi, H., Bekki, S., Butchart, N., Dameris, M., Deushi, M., Hardiman, S. C., Jöckel, P., Klekociuk, A., Marchand, M., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L. D., Plummer, D. A., Revell, L., Rozanov, E., Saint-Martin, D., Scinocca, J., Stenke, A., Stone, K., Yamashita, Y., Yoshida, K., and Zeng, G.: No robust evidence of future changes in major stratospheric sudden warmings: a multi-model assessment from CCM1, *Atmospheric Chemistry and Physics*, 18, 11 277–11 287, <https://doi.org/10.5194/acp-18-11277-2018>, 2018.
- Bechtold, P., Köhler, M., Jung, T., Doblas-Reyes, F., Leutbecher, M., Rodwell, M. J., Vitart, F., and Balsamo, G.: Advances in simulating atmospheric variability with the ECMWF model: From synoptic to decadal time-scales, *Quarterly Journal of the Royal*



- 830 Meteorological Society: A journal of the atmospheric sciences, applied meteorology and physical oceanography, 134, 1337–1351, <https://doi.org/10.1002/qj.289>, 2008.
- Bechtold, P., Semane, N., Lopez, P., Chaboureau, J.-P., Beljaars, A., and Bormann, N.: Representing equilibrium and nonequilibrium convection in large-scale models, *Journal of the Atmospheric Sciences*, 71, 734–753, <https://doi.org/10.1175/JAS-D-13-0163.1>, 2014.
- Becker, T., Bechtold, P., and Sandu, I.: Characteristics of convective precipitation over tropical Africa in storm-resolving global simulations, *Quarterly Journal of the Royal Meteorological Society*, 147, 4388–4407, <https://doi.org/10.1002/qj.4185>, 2021.
- 835 Bermejo, R. and Conde, J.: A Conservative Quasi-Monotone Semi-Lagrangian Scheme, *Monthly Weather Review*, 130, 423 – 430, [https://doi.org/10.1175/1520-0493\(2002\)130<0423:ACQMSL>2.0.CO;2](https://doi.org/10.1175/1520-0493(2002)130<0423:ACQMSL>2.0.CO;2), 2002.
- Brown-Steiner, B., Selin, N. E., Prinn, R., Tilmes, S., Emmons, L., Lamarque, J.-F., and Cameron-Smith, P.: Evaluating simplified chemical mechanisms within present-day simulations of the Community Earth System Model version 1.2 with CAM4 (CESM1.2
- 840 CAM-chem): MOZART-4 vs. Reduced Hydrocarbon vs. Super-Fast chemistry, *Geoscientific Model Development*, 11, 4155–4174, <https://doi.org/10.5194/gmd-11-4155-2018>, 2018.
- Cariolle, D. and Teyssède, H.: A revised linear ozone photochemistry parameterization for use in transport and general circulation models: multi-annual simulations, *Atmospheric Chemistry and Physics*, 7, 2183–2196, <https://doi.org/10.5194/acp-7-2183-2007>, 2007.
- Collins, W. J., Lamarque, J.-F., Schulz, M., Boucher, O., Eyring, V., Hegglin, M. I., Maycock, A., Myhre, G., Prather, M., Shindell, D., and
- 845 Smith, S. J.: AerChemMIP: quantifying the effects of chemistry and aerosols in CMIP6, *Geoscientific Model Development*, 10, 585–607, <https://doi.org/10.5194/gmd-10-585-2017>, 2017.
- Crassier, V., Suhre, K., Tulet, P., and Rosset, R.: Development of a reduced chemical scheme for use in mesoscale meteorological models, *Atmospheric Environment*, 34, 2633–2644, [https://doi.org/10.1016/S1352-2310\(99\)00480-X](https://doi.org/10.1016/S1352-2310(99)00480-X), 2000.
- Cussac, M., Marécal, V., Thouret, V., Josse, B., and Sauvage, B.: The impact of biomass burning on upper tropospheric carbon
- 850 monoxide: a study using MOCAGE global model and IAGOS airborne data, *Atmospheric Chemistry and Physics*, 20, 9393–9417, <https://doi.org/10.5194/acp-20-9393-2020>, 2020.
- Decharme, B., Delire, C., Minvielle, M., Colin, J., Vergnes, J.-P., Alias, A., Saint-Martin, D., Séférian, R., Sénési, S., and Voldoire, A.: Recent Changes in the ISBA-CTRIP Land Surface System for Use in the CNRM-CM6 Climate Model and in Global Off-Line Hydrological Applications, *Journal of Advances in Modeling Earth Systems*, 11, 1207–1252, <https://doi.org/10.1029/2018MS001545>,
- 855 2019.
- Dhomse, S. S., Kinnison, D., Chipperfield, M. P., Salawitch, R. J., Cionni, I., Hegglin, M. I., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Bednarz, E. M., Bekki, S., Braesicke, P., Butchart, N., Dameris, M., Deushi, M., Frith, S., Hardiman, S. C., Hassler, B., Horowitz, L. W., Hu, R.-M., Jöckel, P., Josse, B., Kirner, O., Kremser, S., Langematz, U., Lewis, J., Marchand, M., Lin, M., Mancini, E., Marécal, V., Michou, M., Morgenstern, O., O'Connor, F. M., Oman, L., Pitari, G., Plummer, D. A., Pyle, J. A., Revell, L. E., Rozanov, E., Schofield,
- 860 R., Stenke, A., Stone, K., Sudo, K., Tilmes, S., Visionsi, D., Yamashita, Y., and Zeng, G.: Estimates of ozone return dates from Chemistry-Climate Model Initiative simulations, *Atmospheric Chemistry and Physics*, 18, 8409–8438, <https://doi.org/10.5194/acp-18-8409-2018>, 2018.
- Drugé, T., Nabat, P., Michou, M., and Mallet, M.: Radiative and climate effects of aerosol scattering in long-wave radiation based on global climate modeling, *EGUsphere*, 2024, 1–28, <https://doi.org/10.5194/egusphere-2024-3659>, 2024.
- 865 Eyring, V., Butchart, N., Waugh, D. W., Akiyoshi, H., Austin, J., Bekki, S., Bodeker, G. E., Boville, B. A., Brühl, C., Chipperfield, M. P., Cordero, E., Dameris, M., Deushi, M., Fioletov, V. E., Frith, S. M., Garcia, R. R., Gettelman, A., Giorgetta, M. A., Grewe, V., Jourdain, L., Kinnison, D. E., Mancini, E., Manzini, E., Marchand, M., Marsh, D. R., Nagashima, T., Newman, P. A., Nielsen, J. E., Pawson, S., Pitari,



- G., Plummer, D. A., Rozanov, E., Schraner, M., Shepherd, T. G., Shibata, K., Stolarski, R. S., Struthers, H., Tian, W., and Yoshiki, M.: Assessment of temperature, trace species, and ozone in chemistry-climate model simulations of the recent past, *Journal of Geophysical Research: Atmospheres*, 111, <https://doi.org/10.1029/2006JD007327>, 2006.
- Eyring, V., Bony, S., Meehl, G. A., Senior, C. A., Stevens, B., Stouffer, R. J., and Taylor, K. E.: Overview of the Coupled Model Intercomparison Project Phase 6 (CMIP6) experimental design and organization, *Geoscientific Model Development*, 9, 1937–1958, <https://doi.org/10.5194/gmd-9-1937-2016>, 2016.
- Eyring V., S. T. and Waugh, D.: SPARC, 2010: SPARC CCMVal Report on the Evaluation of Chemistry-Climate Models. V. Eyring, T. Shepherd and D. Waugh (Eds.), SPARC Report No. 5, WCRP-30/2010, WMO/TD – No. 40, <https://doi.org/www.sparc-climate.org/publications/sparc-reports/>, 2010.
- Faïn, X., Szopa, S., Naïk, V., Martinerie, P., Etheridge, D. M., Rhodes, R. H., Trudinger, C. M., Petrenko, V. V., Fourteau, K., and Place, P.: Preindustrial-to-present-day changes in atmospheric carbon monoxide: agreement and gaps between ice archives and global model reconstructions, *Atmospheric Chemistry and Physics*, 25, 1105–1119, <https://doi.org/10.5194/acp-25-1105-2025>, 2025.
- Fiore, A. M., Fischer, E. V., Milly, G. P., Pandey Deolal, S., Wild, O., Jaffe, D. A., Staehelin, J., Clifton, O. E., Bergmann, D., Collins, W., Dentener, F., Doherty, R. M., Duncan, B. N., Fischer, B., Gilge, S., Hess, P. G., Horowitz, L. W., Lupu, A., MacKenzie, I. A., Park, R., Ries, L., Sanderson, M. G., Schultz, M. G., Shindell, D. T., Steinbacher, M., Stevenson, D. S., Szopa, S., Zellweger, C., and Zeng, G.: Peroxy acetyl nitrate (PAN) measurements at northern midlatitude mountain sites in April: a constraint on continental source–receptor relationships, *Atmospheric Chemistry and Physics*, 18, 15 345–15 361, <https://doi.org/10.5194/acp-18-15345-2018>, 2018.
- Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geoscientific Model Development*, 8, 975–1003, <https://doi.org/10.5194/gmd-8-975-2015>, 2015a.
- Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geoscientific Model Development*, 8, 975–1003, <https://doi.org/10.5194/gmd-8-975-2015>, 2015b.
- Folberth, G. A., Staniasek, Z., Archibald, A. T., Gedney, N., Griffiths, P. T., Jones, C. D., O'Connor, F. M., Parker, R. J., Sellar, A. A., and Wiltshire, A.: Description and Evaluation of an Emission-Driven and Fully Coupled Methane Cycle in UKESM1, *Journal of Advances in Modeling Earth Systems*, 14, e2021MS002 982, <https://doi.org/10.1029/2021MS002982>, 2022.
- Froidevaux, L., Kinnison, D. E., Gaubert, B., Schwartz, M. J., Livesey, N. J., Read, W. G., Bardeen, C. G., Ziemke, J. R., and Fuller, R. A.: Tropical upper-tropospheric trends in ozone and carbon monoxide (2005–2020): observational and model results, *Atmospheric Chemistry and Physics*, 25, 597–624, <https://doi.org/10.5194/acp-25-597-2025>, 2025.
- Gaudel, A., Cooper, O. R., Ancellet, G., Barret, B., Boynard, A., Burrows, J. P., Clerbaux, C., Coheur, P.-F., Cuesta, J., Cuevas, E., Doniki, S., Dufour, G., Ebojie, F., Foret, G., Garcia, O., Granados-Muñoz, M. J., Hannigan, J. W., Hase, F., Hassler, B., Huang, G., Hurtmans, D., Jaffe, D., Jones, N., Kalabokas, P., Kerridge, B., Kulawik, S., Latter, B., Leblanc, T., Le Flochmoën, E., Lin, W., Liu, J., Liu, X., Mahieu, E., McClure-Begley, A., Neu, J. L., Osman, M., Palm, M., Petetin, H., Petropavlovskikh, I., Querel, R., Rahpoe, N., Rozanov, A., Schultz, M. G., Schwab, J., Siddans, R., Smale, D., Steinbacher, M., Tanimoto, H., Tarasick, D. W., Thouret, V., Thompson, A. M., Trickl, T., Weatherhead, E., Wespes, C., Worden, H. M., Vigouroux, C., Xu, X., Zeng, G., and Ziemke, J.: Tropospheric Ozone Assessment



- 905 Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, *Elementa: Science of the Anthropocene*, 6, 39, <https://doi.org/10.1525/elementa.291>, 2018.
- Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G. J., Heil, A., Kaiser, 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
910 of air pollutants at global and regional scales during the 1980–2010 period, *Climatic Change*, 109, 163, <https://doi.org/10.1007/s10584-011-0154-1>, 2011.
- Griffiths, P. T., Murray, L. T., Zeng, G., Shin, Y. M., Abraham, N. L., Archibald, A. T., Deushi, M., Emmons, L. K., Galbally, I. E., Hassler, B., Horowitz, L. W., Keeble, J., Liu, J., Moeini, O., Naik, V., O'Connor, F. M., Oshima, N., Tarasick, D., Tilmes, S., Turnock, S. T., Wild, O., Young, P. J., and Zanis, P.: Tropospheric ozone in CMIP6 simulations, *Atmospheric Chemistry and Physics*, 21, 4187–4218,
915 <https://doi.org/10.5194/acp-21-4187-2021>, 2021a.
- Griffiths, P. T., Murray, L. T., Zeng, G., Shin, Y. M., Abraham, N. L., Archibald, A. T., Deushi, M., Emmons, L. K., Galbally, I. E., Hassler, B., Horowitz, L. W., Keeble, J., Liu, J., Moeini, O., Naik, V., O'Connor, F. M., Oshima, N., Tarasick, D., Tilmes, S., Turnock, S. T., Wild, O., Young, P. J., and Zanis, P.: Tropospheric ozone in CMIP6 simulations, *Atmospheric Chemistry and Physics*, 21, 4187–4218, <https://doi.org/10.5194/acp-21-4187-2021>, 2021b.
- 920 Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geoscientific Model Development*, 5, 1471–1492, <https://doi.org/10.5194/gmd-5-1471-2012>, 2012.
- Hameed, S., Cess, R. D., and Hogan, J. S.: Response of the global climate to changes in atmospheric chemical composition due to fossil fuel burning, *Journal of Geophysical Research: Oceans*, 85, 7537–7545, <https://doi.org/10.1029/JC085iC12p07537>, 1980.
- 925 Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), *Geoscientific Model Development*, 11, 369–408, <https://doi.org/10.5194/gmd-11-369-2018>, 2018.
- Hogan, R. J. and Bozzo, A.: A flexible and efficient radiation scheme for the ECMWF model, *Journal of Advances in Modeling Earth*
930 *Systems*, 10, 1990–2008, <https://doi.org/10.1029/2018MS001364>, 2018.
- Horner, R. P., Marais, E. A., Wei, N., Ryan, R. G., and Shah, V.: Vertical profiles of global tropospheric nitrogen dioxide (NO₂) obtained by cloud slicing the TROPOspheric Monitoring Instrument (TROPOMI), *Atmospheric Chemistry and Physics*, 24, 13 047–13 064, <https://doi.org/10.5194/acp-24-13047-2024>, 2024.
- Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, *Geoscientific Model Development*, 3, 445–473, <https://doi.org/10.5194/gmd-3-445-2010>, 2010.
- Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.:
940 The CAMS reanalysis of atmospheric composition, *Atmospheric Chemistry and Physics*, 19, 3515–3556, <https://doi.org/10.5194/acp-19-3515-2019>, 2019a.



- Inness, A., Ades, M., Agustí-Panareda, A., Barré, J., Benedictow, A., Blechschmidt, A.-M., Dominguez, J. J., Engelen, R., Eskes, H., Flemming, J., Huijnen, V., Jones, L., Kipling, Z., Massart, S., Parrington, M., Peuch, V.-H., Razinger, M., Remy, S., Schulz, M., and Suttie, M.: The CAMS reanalysis of atmospheric composition, *Atmospheric Chemistry and Physics*, 19, 3515–3556, <https://doi.org/10.5194/acp-19-3515-2019>, 2019b.
- Jones, D., Prates, L., Qu, Z., Cheng, W., Miyazaki, K., Sekiya, T., Inness, A., Kumar, R., Tang, X., Worden, H., Koren, G., and Huijnen, V.: Assessment of regional and interannual variations in tropospheric ozone in chemical reanalyses, *EGUsphere*, 2025, 1–41, <https://doi.org/10.5194/egusphere-2024-3759>, 2025.
- Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M., Schultz, M. G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system based on observed fire radiative power, *Biogeosciences*, 9, 527–554, <https://doi.org/10.5194/bg-9-527-2012>, 2012.
- Keeble, J., Hassler, B., Banerjee, A., Checa-Garcia, R., Chiodo, G., Davis, S., Eyring, V., Griffiths, P. T., Morgenstern, O., Nowack, P., Zeng, G., Zhang, J., Bodeker, G., Burrows, S., Cameron-Smith, P., Cugnet, D., Danek, C., Deushi, M., Horowitz, L. W., Kubin, A., Li, L., Lohmann, G., Michou, M., Mills, M. J., Nabat, P., Olivie, D., Park, S., Seland, Ø., Stoll, J., Wieners, K.-H., and Wu, T.: Evaluating stratospheric ozone and water vapour changes in CMIP6 models from 1850 to 2100, *Atmospheric Chemistry and Physics*, 21, 5015–5061, <https://doi.org/10.5194/acp-21-5015-2021>, 2021.
- Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, *Geoscientific Model Development*, 6, 179–206, <https://doi.org/10.5194/gmd-6-179-2013>, 2013.
- Lana, A., Bell, T. G., Simó, R., Vallina, S. M., Ballabrera-Poy, J., Kettle, A. J., Dachs, J., Bopp, L., Saltzman, E. S., Stefels, J., Johnson, J. E., and Liss, P. S.: An updated climatology of surface dimethylsulfide concentrations and emission fluxes in the global ocean, *Global Biogeochemical Cycles*, 25, <https://doi.org/10.1029/2010GB003850>, 2011.
- Le Moigne, P., Besson, F., Martin, E., Boé, J., Decharme, B., Etchevers, P., Faroux, S., Habets, F., Lafaysse, M., Leroux, D., et al.: The latest improvements in SURFEX v8. 0 of the Safran-Isba-Modcou hydrometeorological model over France, *Geoscientific Model Development Discussions*, 2020, 1–32, <https://doi.org/10.5194/gmd-13-3925-2020>, 2020.
- Levy II, H., Moxim, W. J., and Kasibhatla, P. S.: A global three-dimensional time-dependent lightning source of tropospheric NO_x, *Journal of Geophysical Research: Atmospheres*, 101, 22 911–22 922, <https://doi.org/10.1029/96JD02341>, 1996.
- Liu, H., Jacob, D. J., Bey, I., and Yantosca, R. M.: Constraints from ²¹⁰Pb and ⁷Be on wet deposition and transport in a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *Journal of Geophysical Research: Atmospheres*, 106, 12 109–12 128, <https://doi.org/10.1029/2000JD900839>, 2001.
- Manisalidis, I., Stavropoulou, E., Stavropoulos, A., and Bezirtzoglou, E.: Environmental and Health Impacts of Air Pollution: A Review, *Frontiers in Public Health*, 8, <https://doi.org/10.3389/fpubh.2020.00014>, 2020.
- Matthes, K., Funke, B., Andersson, M. E., Barnard, L., Beer, J., Charbonneau, P., Clilverd, M. A., Dudok de Wit, T., Haberreiter, M., Hendry, A., Jackman, C. H., Kretzschmar, M., Kruschke, T., Kunze, M., Langematz, U., Marsh, D. R., Maycock, A. C., Misios, S., Rodger, C. J., Scaife, A. A., Seppälä, A., Shangguan, M., Sinnhuber, M., Tourpali, K., Usoskin, I., van de Kamp, M., Verronen, P. T., and Versick, S.: Solar forcing for CMIP6 (v3.2), *Geoscientific Model Development*, 10, 2247–2302, <https://doi.org/10.5194/gmd-10-2247-2017>, 2017.



- Maycock, A. C., Matthes, K., Tegtmeier, S., Schmidt, H., Thiéblemont, R., Hood, L., Akiyoshi, H., Bekki, S., Deushi, M., Jöckel, P., Kirner,
980 O., Kunze, M., Marchand, M., Marsh, D. R., Michou, M., Plummer, D., Revell, L. E., Rozanov, E., Stenke, A., Yamashita, Y., and
Yoshida, K.: The representation of solar cycle signals in stratospheric ozone – Part 2: Analysis of global models, *Atmospheric Chemistry
and Physics*, 18, 11 323–11 343, <https://doi.org/10.5194/acp-18-11323-2018>, 2018a.
- Maycock, A. C., Randel, W. J., Steiner, A. K., Karpechko, A. Y., Christy, J., Saunders, R., Thompson, D. W. J., Zou, C.-Z., Chrysanthou,
A., Luke Abraham, N., Akiyoshi, H., Archibald, A. T., Butchart, N., Chipperfield, M., Dameris, M., Deushi, M., Dhomse, S., Di Genova,
985 G., Jockel, P., Kinnison, D. E., Kirner, O., Ladstadter, F., Michou, M., Morgenstern, O., O'Connor, F., Oman, L., Pitari, G., Plummer,
D. A., Revell, L. E., Rozanov, E., Stenke, A., Visionsi, D., Yamashita, Y., and Zeng, G.: Revisiting the Mystery of Recent Stratospheric
Temperature Trends, *Geophysical Research Letters*, 45, 9919–9933, <https://doi.org/10.1029/2018GL078035>, 2018b.
- Meijer, E. W., van Velthoven, P. F. J., Brunner, D. W., Huntrieser, H., and Kelder, H.: Improvement and evaluation of the parameterisation of
nitrogen oxide production by lightning, *Physics and Chemistry of the Earth, Part C: Solar, Terrestrial & Planetary Science*, 26, 577–583,
990 [https://doi.org/10.1016/S1464-1917\(01\)00050-2](https://doi.org/10.1016/S1464-1917(01)00050-2), 2001.
- Meinshausen, M., Vogel, E., Nauels, A., Lorbacher, K., Meinshausen, N., Etheridge, D. M., Fraser, P. J., Montzka, S. A., Rayner, P. J.,
Trudinger, C. M., Krummel, P. B., Beyerle, U., Canadell, J. G., Daniel, J. S., Enting, I. G., Law, R. M., Lunder, C. R., O'Doherty, S.,
Prinn, R. G., Reimann, S., Rubino, M., Velders, G. J. M., Vollmer, M. K., Wang, R. H. J., and Weiss, R.: Historical greenhouse gas
concentrations for climate modelling (CMIP6), *Geoscientific Model Development*, 10, 2057–2116, <https://doi.org/10.5194/gmd-10-2057->
995 2017, 2017.
- Michou, M. and Peuch, V.: Échanges en surface dans le modèle de chimie transport multi-échelles MOCAGE, *Revue des sciences de l'eau /
Journal of Water Science*, 15, 173–203, <https://doi.org/https://doi.org/10.7202/705492ar>, 2002.
- Michou, M., Laville, P., Serça, D., Fotiadis, A., Bouchou, P., and Peuch, V.-H.: Measured and modeled dry deposition velocities over the
ESCOMPTE area, *Atmospheric Research*, 74, 89–116, <https://doi.org/https://doi.org/10.1016/j.atmosres.2004.04.011>, 2005.
- 1000 Michou, M., Saint-Martin, D., Teyssède, H., Alias, A., Karcher, F., Olivie, D., Voldoire, A., Josse, B., Peuch, V.-H., Clark, H., Lee, J. N., and
Cheroux, F.: A new version of the CNRM Chemistry-Climate Model, CNRM-CCM: description and improvements from the CCMVal-2
simulations, *Geoscientific Model Development*, 4, 873–900, <https://doi.org/10.5194/gmd-4-873-2011>, 2011.
- Michou, M., Nabat, P., Saint-Martin, D., Bock, J., Decharme, B., Mallet, M., Roehrig, R., Séférian, R., Sènesi, S., and Voldoire, A.: Present-
Day and Historical Aerosol and Ozone Characteristics in CNRM CMIP6 Simulations, *Journal of Advances in Modeling Earth Systems*,
1005 12, e2019MS001 816, <https://doi.org/10.1029/2019MS001816>, e2019MS001816 10.1029/2019MS001816, 2020a.
- Michou, M., Nabat, P., Saint-Martin, D., Bock, J., Decharme, B., Mallet, M., Roehrig, R., Séférian, R., Sènesi, S., and Voldoire, A.: Present-
Day and Historical Aerosol and Ozone Characteristics in CNRM CMIP6 Simulations, *Journal of Advances in Modeling Earth Systems*,
12, e2019MS001 816, <https://doi.org/10.1029/2019MS001816>, 2020b.
- 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
1010 climate-composition model – Part 1: The stratosphere, *Geoscientific Model Development*, 2, 43–57, <https://doi.org/10.5194/gmd-2-43->
2009, 2009.
- Morgenstern, O., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M. P., Cugnet, D., Deushi, M., Dhomse, S. S., Garcia,
R. R., Gettelman, A., Gillett, N. P., Hardiman, S. C., Jumelet, J., Kinnison, D. E., Lamarque, J.-F., Lott, F., Marchand, M., Michou, M.,
Nakamura, T., Olivie, D., Peter, T., Plummer, D., Pyle, J. A., Rozanov, E., Saint-Martin, D., Scinocca, J. F., Shibata, K., Sigmond, M.,
1015 Smale, D., Teyssède, H., Tian, W., Voldoire, A., and Yamashita, Y.: Anthropogenic forcing of the Northern Annular Mode in CCMVal-2
models, *Journal of Geophysical Research: Atmospheres*, 115, <https://doi.org/https://doi.org/10.1029/2009JD013347>, 2010a.



- Morgenstern, O., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M. P., Cugnet, D., Deushi, M., Dhomse, S. S., Garcia, R. R., Gettelman, A., Gillett, N. P., Hardiman, S. C., Jumelet, J., Kinnison, D. E., Lamarque, J.-F., Lott, F., Marchand, M., Michou, M., Nakamura, T., Olivie, D., Peter, T., Plummer, D., Pyle, J. A., Rozanov, E., Saint-Martin, D., Scinocca, J. F., Shibata, K., Sigmond, M., Smale, D., Teyss  re, H., Tian, W., Voldoire, A., and Yamashita, Y.: Anthropogenic forcing of the Northern Annular Mode in CCMVal-2 models, *Journal of Geophysical Research: Atmospheres*, 115, <https://doi.org/https://doi.org/10.1029/2009JD013347>, 2010b.
- 1020 Morgenstern, O., Hegglin, M. I., Rozanov, E., O'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., Hardiman, S. C., Horowitz, L. W., J  ckel, P., Josse, B., Kinnison, D., Lin, M., Mancini, E., Manyin, M. E., Marchand, M., Mar  cal, 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, <https://doi.org/10.5194/gmd-10-639-2017>, 2017a.
- 1025 Morgenstern, O., Hegglin, M. I., Rozanov, E., O'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., Hardiman, S. C., Horowitz, L. W., J  ckel, P., Josse, B., Kinnison, D., Lin, M., Mancini, E., Manyin, M. E., Marchand, M., Mar  cal, 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, <https://doi.org/10.5194/gmd-10-639-2017>, 2017b.
- 1030 Morgenstern, O., Kinnison, D. E., Mills, M., Michou, M., Horowitz, L. W., Lin, P., Deushi, M., Yoshida, K., O'Connor, F. M., Tang, Y., Abraham, N. L., Keeble, J., Dennison, F., Rozanov, E., Egorova, T., Sukhodolov, T., and Zeng, G.: Comparison of Arctic and Antarctic Stratospheric Climates in Chemistry Versus No-Chemistry Climate Models, *Journal of Geophysical Research: Atmospheres*, 127, e2022JD037123, <https://doi.org/https://doi.org/10.1029/2022JD037123>, e2022JD037123 2022JD037123, 2022.
- 1035 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., Dals  ren, 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, <https://doi.org/10.5194/acp-13-5277-2013>, 2013.
- 1040 No  l, S., Weigel, K., Bramstedt, K., Rozanov, A., Weber, M., Bovensmann, H., and Burrows, J. P.: Water vapour and methane coupling in the stratosphere observed using SCIAMACHY solar occultation measurements, *Atmospheric Chemistry and Physics*, 18, 4463–4476, <https://doi.org/10.5194/acp-18-4463-2018>, 2018.
- 1045 Noilhan, J. and Mahfouf, J.-F.: The ISBA land surface parameterisation scheme, *Global and planetary Change*, 13, 145–159, [https://doi.org/10.1016/0921-8181\(95\)00043-7](https://doi.org/10.1016/0921-8181(95)00043-7), 1996.
- 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 climate-composition model – Part 2: The Troposphere, *Geoscientific Model Development*, 7, 41–91, <https://doi.org/10.5194/gmd-7-41-2014>, 2014a.
- 1050 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 climate-composition model – Part 2: The Troposphere, *Geoscientific Model Development*, 7, 41–91, <https://doi.org/10.5194/gmd-7-41-2014>, 2014b.



- 1055 Pickering, K. E., Wang, Y., Tao, W.-K., Price, C., and Müller, J.-F.: Vertical distributions of lightning NO_x for use in regional and global chemical transport models, *Journal of Geophysical Research: Atmospheres*, 103, 31 203–31 216, <https://doi.org/10.1029/98JD02651>, 1998.
- Price, C. and Rind, D.: What determines the cloud-to-ground lightning fraction in thunderstorms?, *Geophysical Research Letters*, 20, 463–466, <https://doi.org/10.1029/93GL00226>, 1993.
- Righi, M., Andela, B., Eyring, V., Lauer, A., Predoi, V., Schlund, M., Vegas-Regidor, J., Bock, L., Brötz, B., de Mora, L., Diblen, F., Dreyer, L., Drost, N., Earnshaw, P., Hassler, B., Koldunov, N., Little, B., Loosveldt Tomas, S., and Zimmermann, K.: Earth System Model Evaluation Tool (ESMValTool) v2.0 – technical overview, *Geoscientific Model Development*, 13, 1179–1199, <https://doi.org/10.5194/gmd-13-1179-2020>, 2020.
- 1060 Roehrig, R., Beau, I., Saint-Martin, D., Alias, A., Decharme, B., Guérémy, J.-F., Voldoire, A., Abdel-Lathif, A. Y., Bazile, E., Belamari, S., Blein, S., Bouniol, D., Bouteloup, Y., Cattiaux, J., Chauvin, F., Chevallier, M., Colin, J., Douville, H., Marquet, P., Michou, M., Nabat, P., Oudar, T., Peyrillé, P., Piriou, J.-M., Salas y Mélia, D., Séférian, R., and Sénési, S.: The CNRM Global Atmosphere Model ARPEGE-Climat 6.3: Description and Evaluation, *Journal of Advances in Modeling Earth Systems*, 12, e2020MS002075, <https://doi.org/10.1029/2020MS002075>, e2020MS002075 10.1029/2020MS002075, 2020.
- 1065 Sander, R.: Compilation of Henry’s law constants (version 5.0.0) for water as solvent, *Atmospheric Chemistry and Physics*, 23, 10901–12 440, <https://doi.org/10.5194/acp-23-10901-2023>, 2023.
- 1070 Sanderson, B. M., Booth, B. B. B., Dunne, J., Eyring, V., Fisher, R. A., Friedlingstein, P., Gidden, M. J., Hajima, T., Jones, C. D., Jones, C., King, A., Koven, C. D., Lawrence, D. M., Lowe, J., Mengis, N., Peters, G. P., Rogelj, J., Smith, C., Snyder, A. C., Simpson, I. R., Swann, A. L. S., Tebaldi, C., Ilyina, T., Schleussner, C.-F., Seferian, R., Samset, B. H., van Vuuren, D., and Zaehle, S.: The need for carbon emissions-driven climate projections in CMIP7, *EGUsphere*, pp. 1–51, <https://doi.org/10.5194/egusphere-2023-2127>, 2023.
- Saunois, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P. M., Covey, K., Curry, C. L., Etiope, G., Frankenberg, C., Gedney, N., Hegglin, M. I., Höglund-Isaksson, L., Hugelius, G., Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen, K. M., Joos, F., Kleinen, T., Krummel, P. B., Langenfelds, R. L., Laruelle, G. G., Liu, L., Machida, T., Maksyutov, S., McDonald, K. C., McNorton, J., Miller, P. A., Melton, J. R., Morino, I., Müller, J., Murguía-Flores, F., Naik, V., Niwa, Y., Noce, S., O’Doherty, S., Parker, R. J., Peng, C., Peng, S., Peters, G. P., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W. J., Rosentretter, J. A., Segers, A., Simpson, I. J., Shi, H., Smith, S. J., Steele, L. P., Thornton, B. F., Tian, H., Tohjima, Y., Tubiello, F. N., Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T. S., van Weele, M., van der Werf, G. R., Weiss, R. F., Worthy, D., Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: The Global Methane Budget 2000–2017, *Earth System Science Data*, 12, 1561–1623, <https://doi.org/10.5194/essd-12-1561-2020>, 2020.
- 1080 Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J. G., Held, A. C., Pereira, J. M. C., and van het Bolscher, M.: Global wildland fire emissions from 1960 to 2000, *Global Biogeochemical Cycles*, 22, <https://doi.org/10.1029/2007GB003031>, 2008.
- Séférian, R., Nabat, P., Michou, M., Saint-Martin, D., Voldoire, A., Colin, J., Decharme, B., Delire, C., Berthet, S., Chevallier, M., Sénési, S., Franchisteguy, L., Vial, J., Mallet, M., Joetzjer, E., Geoffroy, O., Guérémy, J.-F., Moine, M.-P., Msadek, R., Ribes, A., Rocher, M., Roehrig, R., Salas-y Mélia, D., Sanchez, E., Terray, L., Valcke, S., Waldman, R., Aumont, O., Bopp, L., Deshayes, J., Etche, C., and Madec, G.: Evaluation of CNRM Earth-System model, CNRM-ESM 2-1: role of Earth system processes in present-day and future climate, *Journal of Advances in Modeling Earth Systems*, <https://doi.org/10.1029/2019MS001791>, 2019.
- 1090 Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*. 2nd Edition, 2006.



- Sekiya, T., Emili, E., Miyazaki, K., Inness, A., Qu, Z., Pierce, R. B., Jones, D., Worden, H., Cheng, W. Y. Y., Huijnen, V., and Koren, G.: Assessing the relative impacts of satellite ozone and its precursor observations to improve global tropospheric ozone analysis using multiple chemical reanalysis systems, *Atmospheric Chemistry and Physics*, 25, 2243–2268, <https://doi.org/10.5194/acp-25-2243-2025>, 2025.
- Shah, V., Jacob, D. J., Dang, R., Lamsal, L. N., Strode, S. A., Steenrod, S. D., Boersma, K. F., Eastham, S. D., Fritz, T. M., Thompson, C., Peischl, J., Bourgeois, I., Pollack, I. B., Nault, B. A., Cohen, R. C., Campuzano-Jost, P., Jimenez, J. L., Andersen, S. T., Carpenter, L. J., Sherwen, T., and Evans, M. J.: Nitrogen oxides in the free troposphere: implications for tropospheric oxidants and the interpretation of satellite NO₂ measurements, *Atmospheric Chemistry and Physics*, 23, 1227–1257, <https://doi.org/10.5194/acp-23-1227-2023>, 2023.
- Shindell, D. T., Faluvegi, G., Stevenson, D. S., Krol, M. C., Emmons, L. K., Lamarque, J.-F., Pétron, G., Dentener, F. J., Ellingsen, K., Schultz, M. G., Wild, O., Amann, M., Atherton, C. S., Bergmann, D. J., Bey, I., Butler, T., Cofala, J., Collins, W. J., Derwent, R. G., Doherty, R. M., Drevet, J., Eskes, H. J., Fiore, A. M., Gauss, M., Hauglustaine, D. A., Horowitz, L. W., Isaksen, I. S. A., Lawrence, M. G., Montanaro, V., Müller, J.-F., Pitari, G., Prather, M. J., Pyle, J. A., Rast, S., Rodriguez, J. M., Sanderson, M. G., Savage, N. H., Strahan, S. E., Sudo, K., Szopa, S., Unger, N., van Noije, T. P. C., and Zeng, G.: Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes, *Journal of Geophysical Research: Atmospheres*, 111, <https://doi.org/10.1029/2006JD007100>, 2006.
- Sicard, P., Anav, A., De Marco, A., and Paoletti, E.: Projected global ground-level ozone impacts on vegetation under different emission and climate scenarios, *Atmospheric Chemistry and Physics*, 17, 12 177–12 196, <https://doi.org/10.5194/acp-17-12177-2017>, 2017.
- Simpson, D. and Darras, S.: Global soil NO emissions for Atmospheric Chemical Transport Modelling: CAMS-GLOB-SOIL v2.2, *Earth System Science Data Discussions*, pp. 1–35, <https://doi.org/10.5194/essd-2021-221>, 2021.
- Sindelarova, K., Markova, J., Simpson, D., Huszar, P., Karlicky, J., Darras, S., and Granier, C.: High-resolution biogenic global emission inventory for the time period 2000–2019 for air quality modelling, *Earth System Science Data*, 14, 251–270, <https://doi.org/10.5194/essd-14-251-2022>, 2022.
- Skeie, R. B., Myhre, G., Hodnebrog, Ø., Cameron-Smith, P. J., Deushi, M., Hegglin, M. I., Horowitz, L. W., Kramer, R. J., Michou, M., Mills, M. J., Olivié, D. J. L., Connor, F. M. O., Paynter, D., Samset, B. H., Sellar, A., Shindell, D., Takemura, T., Tilmes, S., and Wu, T.: Historical total ozone radiative forcing derived from CMIP6 simulations, *npj Climate and Atmospheric Science*, 3, 32, <https://doi.org/10.1038/s41612-020-00131-0>, 2020.
- Slinn, S. A. and Slinn, W. G. N.: Predictions for particle deposition on natural waters, *Atmospheric Environment* (1967), 14, 1013–1016, [https://doi.org/10.1016/0004-6981\(80\)90032-3](https://doi.org/10.1016/0004-6981(80)90032-3), 1980.
- Smith, C. J., Kramer, R. J., Myhre, G., Alterskjær, K., Collins, W., Sima, A., Boucher, O., Dufresne, J.-L., Nabat, P., Michou, M., Yukimoto, S., Cole, J., Paynter, D., Shiogama, H., O'Connor, F. M., Robertson, E., Wiltshire, A., Andrews, T., Hannay, C., Miller, R., Nazarenko, L., Kirkevåg, A., Olivié, D., Fiedler, S., Pincus, R., and Forster, P. M.: Effective radiative forcing and adjustments in CMIP6 models, *Atmospheric Chemistry and Physics*, 20, 9591–9618, <https://doi.org/10.5194/acp-20-9591-2020>, 2020.
- Stevenson, D. S., Zhao, A., Naik, V., O'Connor, F. M., Tilmes, S., Zeng, G., Murray, L. T., Collins, W. J., Griffiths, P. T., Shim, S., Horowitz, L. W., Sentman, L. T., and Emmons, L.: Trends in global tropospheric hydroxyl radical and methane lifetime since 1850 from AerChem-MIP, *Atmospheric Chemistry and Physics*, 20, 12 905–12 920, <https://doi.org/10.5194/acp-20-12905-2020>, 2020.
- Stockwell, W. R., Kirchner, F., Kuhn, M., and Seefeld, S.: A new mechanism for regional atmospheric chemistry modeling, *Journal of Geophysical Research: Atmospheres*, 102, 25 847–25 879, <https://doi.org/10.1029/97JD00849>, 1997.
- Strahan, S. E., Douglass, A. R., Stolarski, R. S., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M. P., Cugnet, D., Dhomse, S., Frith, S. M., Gettelman, A., Hardiman, S. C., Kinnison, D. E., Lamarque, J.-F., Mancini, E., Marchand, M., Michou, M., Morgenstern,



- O., Nakamura, T., Olivié, D., Pawson, S., Pitari, G., Plummer, D. A., Pyle, J. A., Scinocca, J. F., Shepherd, T. G., Shibata, K., Smale, D., Teyssèdre, H., Tian, W., and Yamashita, Y.: Using transport diagnostics to understand chemistry climate model ozone simulations, *Journal of Geophysical Research: Atmospheres*, 116, <https://doi.org/https://doi.org/10.1029/2010JD015360>, 2011.
- 1135 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 climate models, *Atmospheric Chemistry and Physics*, 15, 11 789–11 805, <https://doi.org/10.5194/acp-15-11789-2015>, 2015.
- Strong, K., Wolff, M. A., Kerzenmacher, T. E., Walker, K. A., Bernath, P. F., Blumenstock, T., Boone, C., Catoire, V., Coffey, M., De Mazière, M., Demoulin, P., Duchatelet, P., Dupuy, E., Hannigan, J., Höpfner, M., Glatthor, N., Griffith, D. W. T., Jin, J. J., Jones, N., Jucks, K., Kuellmann, H., Kuttippurath, J., Lambert, A., Mahieu, E., McConnell, J. C., Mellqvist, J., Mikuteit, S., Murtagh, D. P., Notholt, J., Piccolo, 1140 C., Raspollini, P., Ridolfi, M., Robert, C., Schneider, M., Schrems, O., Semeniuk, K., Senten, C., Stiller, G. P., Strandberg, A., Taylor, J., Tétard, C., Toohey, M., Urban, J., Warneke, T., and Wood, S.: Validation of ACE-FTS N₂O measurements, *Atmospheric Chemistry and Physics*, 8, 4759–4786, <https://doi.org/10.5194/acp-8-4759-2008>, 2008.
- Terrenoire, E., Hauglustaine, D. A., Cohen, Y., Cozic, A., Valorso, R., Lefèvre, F., and Matthes, S.: Impact of present and future aircraft NO_x and aerosol emissions on atmospheric composition and associated direct radiative forcing of climate, *Atmospheric Chemistry and Physics*, 22, 11 987–12 023, <https://doi.org/10.5194/acp-22-11987-2022>, 2022.
- 1145 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 multi-year studies: evaluation of the present-day climatology and sensitivity to surface processes, *Atmospheric Chemistry and Physics*, 7, 5815–5860, <https://doi.org/10.5194/acp-7-5815-2007>, 2007.
- 1150 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 multi-year studies: evaluation of the present-day climatology and sensitivity to surface processes, *Atmospheric Chemistry and Physics*, 7, 5815–5860, <https://doi.org/10.5194/acp-7-5815-2007>, 2007.
- Thomason, L. W., Ernest, N., Millán, L., Rieger, L., Bourassa, A., Vernier, J.-P., Manney, G., Luo, B., Arfeuille, F., and Peter, T.: A global 1155 space-based stratospheric aerosol climatology: 1979–2016, *Earth System Science Data*, 10, 469–492, <https://doi.org/10.5194/essd-10-469-2018>, 2018.
- Tian, H., Pan, N., Thompson, R. L., Canadell, J. G., Suntharalingam, P., Regnier, P., Davidson, E. A., Prather, M., Ciais, P., Muntean, M., Pan, S., Winiwarter, W., Zaehle, S., Zhou, F., Jackson, R. B., Bange, H. W., Berthet, S., Bian, Z., Bianchi, D., Bouwman, A. F., Buitenhuis, E. T., Dutton, G., Hu, M., Ito, A., Jain, A. K., Jeltsch-Thömmes, A., Joos, F., Kou-Giesbrecht, S., Krummel, P. B., Lan, X., Landolfi, 1160 A., Lauerwald, R., Li, Y., Lu, C., Maavara, T., Manizza, M., Millet, D. B., Mühle, J., Patra, P. K., Peters, G. P., Qin, X., Raymond, P., Resplandy, L., Rosentreter, J. A., Shi, H., Sun, Q., Tonina, D., Tubiello, F. N., van der Werf, G. R., Vuichard, N., Wang, J., Wells, K. C., Western, L. M., Wilson, C., Yang, J., Yao, Y., You, Y., and Zhu, Q.: Global nitrous oxide budget (1980–2020), *Earth System Science Data*, 16, 2543–2604, <https://doi.org/10.5194/essd-16-2543-2024>, 2024.
- Tiedtke, M.: A comprehensive mass flux scheme for cumulus parameterization in large-scale models, *Monthly weather review*, 117, 1779–1800, 1165
- 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), *Geoscientific Model Development*, 9, 1853–1890, <https://doi.org/10.5194/gmd-9-1853-2016>, 2016.
- 1170 van Marle, M. J. E., Kloster, S., Magi, B. I., Marlon, J. R., Daniau, A.-L., Field, R. D., Arneth, A., Forrest, M., Hantson, S., Kehrwald, N. M., Knorr, W., Lasslop, G., Li, F., Mangeon, S., Yue, C., Kaiser, J. W., and van der Werf, G. R.: Historic global biomass burning emissions for CMIP6 (BB4CMIP) based on merging satellite observations with proxies and fire models (1750–2015), *Geoscientific Model Development*, 10, 3329–3357, <https://doi.org/10.5194/gmd-10-3329-2017>, 2017.
- Verma, S., Yadava, P. K., Lal, D. M., Mall, R. K., Kumar, H., and Payra, S.: Role of Lightning NO_x in Ozone Formation: A Review, *Pure and Applied Geophysics*, 178, 1425–1443, <https://doi.org/10.1007/s00024-021-02710-5>, 2021.
- 1175 Voldoire, A., Saint-Martin, D., Sénéci, S., Decharme, B., Alias, A., Chevallier, M., Colin, J., Guérémy, J.-F., Michou, M., Moine, M.-P., Nabat, P., Roehrig, R., Salas y Mélia, D., Séférian, R., Valcke, S., Beau, I., Belamari, S., Berthet, S., Cassou, C., Cattiaux, J., Deshayes, J., Douville, H., Ethé, C., Franchistéguy, L., Geoffroy, O., Lévy, C., Madec, G., Meurdesoif, Y., Msadek, R., Ribes, A., Sanchez-Gomez, E., Terray, L., and Waldman, R.: Evaluation of CMIP6 DECK Experiments With CNRM-CM6-1, *Journal of Advances in Modeling Earth Systems*, 11, 2177–2213, <https://doi.org/10.1029/2019MS001683>, 2019.
- 1180 von Schneidmesser, E., Monks, P. S., Allan, J. D., Bruhwiler, L., Forster, P., Fowler, D., Lauer, A., Morgan, W. T., Paasonen, P., Righi, M., Sindelarova, K., and Sutton, M. A.: Chemistry and the Linkages between Air Quality and Climate Change, *Chemical Reviews*, 115, 3856–3897, <https://doi.org/10.1021/acs.chemrev.5b00089>, PMID: 25926133, 2015.
- 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 Chemistry and Physics*, 13, 2563–2587, <https://doi.org/10.5194/acp-13-2563-2013>, 2013.
- 1185 Wagner, A., Bennouna, Y., Blechschmidt, A.-M., Brasseur, G., Chabrilat, S., Christophe, Y., Errera, Q., Eskes, H., Flemming, J., Hansen, K. M., Inness, A., Kapsomenakis, J., Langerock, B., Richter, A., Sudarchikova, N., Thouret, V., and Zerefos, C.: Comprehensive evaluation of the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis against independent observations: Reactive gases, *Elementa: Science of the Anthropocene*, 9, 00 171, <https://doi.org/10.1525/elementa.2020.00171>, 2021.
- 1190 Wales, P., Salawitch, R., Nicely, J., Anderson, D., Canty, T., Baidar, S., Dix, B., Koenig, T., Volkamer, R., Chen, D., Huey, L., Tanner, D., Cuevas, C., Fernandez, R., Kinnison, D., Lamarque, J., Saiz-Lopez, A., Atlas, E., Hall, S., Navarro, M., Pan, L., Schauffler, S., Stell, M., Tilmes, S., Ullmann, K., Weinheimer, A., Akiyoshi, H., Chipperfield, M., Deushi, M., Dhomse, S., Feng, W., Graf, P., Hossaini, R., Jöckel, P., Mancini, E., Michou, M., Morgenstern, O., Oman, L., Pitari, G., Plummer, D., Revell, L., Rozanov, E., Saint-Martin, D., Schofield, R., Stenke, A., Stone, K., Visionsi, D., Yamashita, Y., and Zeng, G.: Stratospheric Injection of Brominated Very Short-Lived Substances: Aircraft Observations in the Western Pacific and Representation in Global Models, *Journal of Geophysical Research: Oceans*, 123, 5690–5719, <https://doi.org/10.1029/2017JD027978>, 2018.
- 1195 Wang, Y., Ma, Y.-F., Eskes, H., Inness, A., Flemming, J., and Brasseur, G. P.: Evaluation of the CAMS global atmospheric trace gas reanalysis 2003–2016 using aircraft campaign observations, *Atmospheric Chemistry and Physics*, 20, 4493–4521, <https://doi.org/10.5194/acp-20-4493-2020>, 2020.
- 1200 Wesely, M.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, *Atmospheric Environment* (1967), 23, 1293–1304, [https://doi.org/10.1016/0004-6981\(89\)90153-4](https://doi.org/10.1016/0004-6981(89)90153-4), 1989.



- 1205 Wild, O., Voulgarakis, A., O'Connor, F., Lamarque, J.-F., Ryan, E. M., and Lee, L.: Global sensitivity analysis of chemistry–climate model budgets of tropospheric ozone and OH: exploring model diversity, *Atmospheric Chemistry and Physics*, 20, 4047–4058, <https://doi.org/10.5194/acp-20-4047-2020>, 2020.
- Williams, J. E., Huijnen, V., Bouarar, I., Meziane, M., Schreurs, T., Pelletier, S., Marécal, V., Josse, B., and Flemming, J.: Regional evaluation of the performance of the global CAMS chemical modeling system over the United States (IFS cycle 47r1), *Geoscientific Model Development*, 15, 4657–4687, <https://doi.org/10.5194/gmd-15-4657-2022>, 2022.
- 1210 Young, P. J., Archibald, A. T., Bowman, K. W., Lamarque, J.-F., Naik, V., Stevenson, D. S., Tilmes, S., Voulgarakis, A., Wild, O., Bergmann, D., Cameron-Smith, P., Cionni, I., Collins, W. J., Dalsøren, S. B., Doherty, R. M., Eyring, V., Faluvegi, G., Horowitz, L. W., Josse, B., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Plummer, D. A., Righi, M., Rumbold, S. T., Skeie, R. B., Shindell, D. T., Strode, S. A., Sudo, K., Szopa, S., and Zeng, G.: Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmospheric Chemistry and Physics*, 13, 2063–2090, <https://doi.org/10.5194/acp-13-2063-2013>, 2013.
- 1215 Young, P. J., Naik, V., Fiore, A. M., Gaudel, A., Guo, J., Lin, M. Y., Neu, J. L., Parrish, D. D., Rieder, H. E., Schnell, J. L., Tilmes, S., Wild, O., Zhang, L., Ziemke, J., Brandt, J., Delcloo, A., Doherty, R. M., Geels, C., Hegglin, M. I., Hu, L., Im, U., Kumar, R., Luhar, A., Murray, L., Plummer, D., Rodriguez, J., Saiz-Lopez, A., Schultz, M. G., Woodhouse, M. T., and Zeng, G.: Tropospheric Ozone Assessment Report: Assessment of global-scale model performance for global and regional ozone distributions, variability, and trends, *Elementa: Science of the Anthropocene*, 6, 10, <https://doi.org/10.1525/elementa.265>, 2018.
- 1220 Zeng, G., Morgenstern, O., Williams, J. H. T., O'Connor, F. M., Griffiths, P. T., Keeble, J., Deushi, M., Horowitz, L. W., Naik, V., Emmons, L. K., Abraham, N. L., Archibald, A. T., Bauer, S. E., Hassler, B., Michou, M., Mills, M. J., Murray, L. T., Oshima, N., Sentman, L. T., Tilmes, S., Tsigaridis, K., and Young, P. J.: Attribution of Stratospheric and Tropospheric Ozone Changes Between 1850 and 2014 in CMIP6 Models, *Journal of Geophysical Research: Atmospheres*, 127, e2022JD036452, <https://doi.org/https://doi.org/10.1029/2022JD036452>, e2022JD036452 2022JD036452, 2022.
- 1225 Zhao, Y., Saunois, M., Bousquet, P., Lin, X., Hegglin, M. I., Canadell, J. G., Jackson, R. B., and Zheng, B.: Reconciling the bottom-up and top-down estimates of the methane chemical sink using multiple observations, *Atmospheric Chemistry and Physics*, 23, 789–807, <https://doi.org/10.5194/acp-23-789-2023>, 2023.