



RESEARCH ARTICLE

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How Useful Is a Linear Ozone Parameterization for Global Climate Modeling?

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- Linear scheme can be a useful tool to interactively represent ozone at low computational cost
- Responses of ozone to 4xCO₂ are very similar for an explicit and a linear ozone scheme
- Linear scheme underestimates amplitudes of circulation-induced variability patterns

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Abstract The explicit calculation of stratospheric ozone in global climate models still comes at a high computational cost. Here, the usefulness of a linear ozone parameterization in a global climate model is assessed by comparing it to an explicit chemistry scheme and to observations. It is shown that the annual mean total ozone column and the tropical ozone profile agree well for the linear and the explicit chemistry schemes and the observations. Ozone variability caused by the quasi-biennial oscillation and by extratropical quasi-stationary planetary waves is reproduced qualitatively, but its magnitude is underestimated in particular in the simulations using the linear parameterization. The response of ozone to a quadrupling of CO₂ simulated with both schemes is in the range of earlier simulations with explicit schemes. This concerns in particular ozone decreases in the tropical lower stratosphere and increases above predicted as a consequence of a strengthening of tropical upwelling and potentially affecting climate sensitivity. This study demonstrates that despite existing weaknesses a linear ozone parameterization can be a useful tool to represent stratospheric ozone in climate models at negligible computational cost.

Plain Language Summary Ozone is the main absorber of solar radiation in the stratosphere. Feedbacks between ozone, temperature, and circulation are therefore crucial for stratospheric variability and responses to external forcings like anthropogenic greenhouse gas emissions. However, the comprehensive representation of stratospheric ozone in global climate models comes at a high computational cost. Here, we analyze to what extent a computationally cheap linear ozone parameterization can overcome this problem. It is shown that a linear ozone scheme reproduces well key features of the mean ozone distribution but underestimates variability induced by circulation patterns like the quasi-biennial oscillation of winds in the tropical stratosphere. The response of ozone to an idealized global warming scenario (instantaneous quadrupling of the atmospheric CO₂ concentration) simulated with the linear ozone scheme is in the range of earlier model studies with more complex chemistry schemes. We demonstrate that despite existing weaknesses, a linear ozone parameterization can be a useful tool to represent stratospheric ozone in climate models at negligible computational cost.

1. Introduction

Ozone is the most important absorber of solar radiation in the stratosphere. The ozone distribution is, therefore, crucial for stratospheric temperature and circulation and has been shown to have impacts down to the Earth's surface. Consequently, an increasing number of climate models include comprehensive representations of stratospheric chemistry, which comes at a high computational cost. In this paper we try to answer the question how useful a traditional linear ozone parameterization is for global climate modeling taking into account recent advances in the knowledge of ozone-circulation and ozone-climate feedbacks.

Several recent studies with comprehensive chemistry-climate models show large changes in ozone in the tropical lower stratosphere as a response to a quadrupling of CO₂ (Dietmüller et al., 2014; Marsh et al., 2016; Muthers et al., 2014; P. J. Nowack et al., 2015). The studies report impacts of these ozone changes on effective climate sensitivity that range between about 0% and −20%. Chiodo et al. (2018) found that the spread in the reduction of lower tropical stratospheric ozone between these different models emerges from the spread in the strength of the change in tropical upwelling. In an idealized model study with a one-dimensional radiative convective equilibrium model representing the tropical atmosphere, Dacie et al. (2019) showed that differences in the tropical ozone changes could contribute to the large spread in the chemistry-climate feedback. In contrast to this, Chiodo and Polvani (2019) found that global mean radiative effects from three different ozone anomalies prescribed in a three-dimensional coupled climate model are very similar and

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close to 0 because effects in the longwave and shortwave parts of the spectrum and in tropics and extratropics almost cancel. However, the potential chemistry-climate feedback provides a strong motivation for the use of interactive stratospheric ozone in global climate models.

More motivation comes from effects of stratospheric ozone on circulation and feedbacks between the two. For example, the quasi-biennial oscillation (QBO) of zonal winds in the tropical stratosphere is known to produce also an oscillation in ozone, and it has been shown that the simulated QBO differs for model experiments with interactive and prescribed ozone (Tian et al., 2006). Furthermore, planetary waves in the middle- and high-latitude stratosphere create corresponding wave signatures in ozone, which provide a feedback on the waves and also influence the zonal mean circulation (e.g., Albers & Nathan, 2012; Gabriel et al., 2007; Kirchner & Peters, 2003; Peters et al., 2015; Silverman et al., 2018). While it is known since about two decades that in particular the Antarctic ozone hole affects atmospheric circulation down to the surface (e.g., Thompson & Solomon, 2002), more recent model studies have indicated that also the aforementioned stratospheric ozone-dynamics feedbacks influence stratosphere-troposphere coupling (Haase & Matthes, 2019; Lin et al., 2017; Romanowsky et al., 2019).

However, in climate model intercomparison experiments like the fifth Coupled Model Intercomparison Project (CMIP5) most models still do not operate with interactive atmospheric chemistry due to high computational cost but prescribe, in general, monthly and zonal mean ozone fields. Ozone-circulation feedbacks are obviously ignored when ozone fields are prescribed.

Beside explicit calculation of ozone, another option to represent ozone are parameterizations. Nowack et al. (2018) proposed a parameterization based on machine learning that has, however, not yet been included in a general circulation model. Other ozone parameterizations calculate the ozone as a linearization of the net chemical production term in the ozone continuity equation. The advantage of such an approach is its computational efficiency. In this study, we assess the usefulness of the linear ozone parameterization from Cariolle and Teyss  re (2007), commonly called “Cariolle scheme,” for global climate modeling by comparing it to an explicit chemistry scheme and to observations. We focus on two major aspects of which the importance is increasingly realized as described above, first, dynamically induced variability in ozone and second, the response of ozone to global warming.

Cariolle and D  qu   (1986) were the first to develop a linear ozone parameterization based on sensitivity calculations with a chemistry-transport model containing a more comprehensive explicit chemistry scheme. Their parameterization depends only on temperature and ozone concentration; hence, no other chemical species need to be modeled. An updated version of the Cariolle scheme includes also a term for heterogeneous chemistry (Cariolle & Teyss  re, 2007). It is included in the operational forecast model of the European Centre for Medium-Range Weather Forecasts and in the ERA5 reanalysis model (Hersbach et al., 2018). The Cariolle scheme has been widely used to analyze past ozone trends and ozone variability (Hadjinicolaou et al., 2005; J  rar et al., 2006; Pyle et al., 2005).

Similar linear parameterizations have been developed by McLinden et al. (2000) (LINOZ) and McCormack et al. (2006) (CHEM2D-OPP). Geer et al. (2007) compared three ozone parameterizations (Cariolle, LINOZ and CHEM2D-OPP) for data assimilation and stated that all three schemes work well in the stratosphere and mesosphere.

Already a quarter century ago Mahfouf et al. (1994) made use of an early version of the Cariolle scheme to account for ozone changes in a general circulation model study of the climate response to a doubling of CO₂. They found a similar dipole response of tropical stratospheric ozone to global warming as reported by Chiodo et al. (2018) but with a much smaller magnitude. One could argue that today the relevance of computationally cheap ozone schemes is much smaller than at that time because running explicit chemistry schemes over climate time scales at typical climate model resolutions has become much more affordable. However, there are also good arguments to invest additional computing power rather in very high, convection-permitting model resolutions (e.g., Satoh et al., 2019) or the computation of very large ensembles (e.g., Maher et al., 2019), where again the cost of comprehensive explicit chemistry schemes would be prohibitive. Given the more recent knowledge on the importance of the role of ozone for climate studies as discussed above, we think it is timely to ask again the question how well current linear ozone schemes can perform in global climate modeling.

To answer this question, we use simulations with the general circulation model Hamburg Model of the Neutral and Ionized Atmosphere (HAMMONIA) coupled to both the explicit chemistry model MOZART (Model of Ozone and Related Tracers Kinnison et al., 2007) and the Cariolle scheme, enabling a direct comparison of the two schemes. We concentrate on two major points: how well the Cariolle scheme reproduces (a) the climatological ozone distribution and variability induced by the QBO and extratropical planetary waves and (b) the ozone response to a quadrupling of CO_2 . Regarding (a), we compare the linear scheme to observations and reanalysis data. A strong forcing like quadrupled CO_2 allows us to study the limitations of the linearization of the parameterization.

This paper is organized as follows: Section 2 describes the model setup and the linear Cariolle scheme along with a short summary of the used observational and reanalysis data. Section 3.1 compares the Cariolle scheme to the observations, and section 3.2 shows the response of the ozone parameterization to quadrupling of CO_2 . Finally, section 4 summarizes and discusses the main outcomes and limitations of this study.

2. Model and Observational Data

2.1. HAMMONIA

HAMMONIA is the chemistry and general circulation model used in this study. It is an upward extension of the general circulation model ECHAM5 (Roeckner et al., 2006) and treats atmospheric dynamics, radiation, and chemistry interactively from the surface to the thermosphere (2×10^{-7} hPa, ~ 250 km altitude). The atmospheric chemistry is calculated by MOZART (Kinnison et al., 2007). A more detailed description is given by Schmidt et al. (2006) and Meraner et al. (2016).

For this study, HAMMONIA is run with a triangular truncation at Wave Number 63 (T63), which corresponds to a horizontal resolution of about 1.9° in latitude and longitude. In the vertical, 119 layers are used of which the thickness in the middle atmosphere ranges from ~ 800 m near the tropopause to ~ 3 km near the mesopause.

We implemented the Cariolle scheme (see section 2.2) in HAMMONIA and, hence, have now two ways to predict ozone: either by the Cariolle parameterization or by the MOZART explicit chemistry scheme. In contrast to the model used in Meraner et al. (2016) ion chemistry is not considered. We carried out two timeslice experiment sets, in which either the Cariolle ozone or the MOZART ozone interacts with the radiation. All experiments used in this study calculate both ozones; however, only one ozone tracer per experiment interacts with the radiation. Hence, Cariolle ozone is an additional output in the MOZART simulations. This has the advantage that both ozone schemes can be compared excluding any difference from circulation. The Cariolle scheme provides parameters only up to 0.01 hPa. Above this height the MOZART explicit chemistry is used in all experiments to stabilize the upper atmospheric temperature in HAMMONIA preventing the model from crashing.

All boundary conditions are set constant to minimize transient impacts. Stratospheric aerosols are set to constant low values of the Year 1999; likewise, we simulate solar minimum (e.g., the $F_{10.7}$ solar flux is set to $68.7 \times 10^{-22} \text{ W} \cdot \text{m}^{-2} \cdot \text{Hz}^{-1}$). Sea surface temperature (SST) and sea ice cover as well as the upper and lower boundaries of chemistry species are set to an average annual cycle of the period 1979–2008. Carbon dioxide is fixed at 359.09 ppmv (and to 1436.37 ppmv for the $4\times\text{CO}_2$ case).

We simulate two different climatic conditions: the late twentieth century (average over 1979–2008, named Control hereafter) and “global warming” (quadrupled atmospheric CO_2 concentration and $+6.7$ K globally uniform SST increase, named $4\times\text{CO}_2$ hereafter). The increase in SST is taken from a multimodel mean from CMIP5 coupled models (Andrews et al., 2012). We ran 15 years for each experiment but analyzed only the final 8 years, discarding the first 7 as spin-up.

As both Control experiments show no or only a weak indication of a QBO (not shown), we performed an additional set of experiments with retuned gravity wave source parameters (see section 2.1). We tuned the QBO by increasing the source term of the nonorographic gravity wave parameterization (Hines, 1997), the uniform, isotropic root-mean-square wave wind speed at a fixed launching level of 830 hPa, from 0.7 to 1.0 m/s. These experiments were started from the spun-up states and run for only 5 years. The, in total, six experiments of this study are listed in Table 1.

Table 1

List of experiments with either MOZART or Cariolle ozone tracer interacting with radiation for two different climatic conditions (late twentieth century or global warming), and with two RMS wave wind speeds (σ) of the gravity wave source function

Experiment	Ozone tracer	SST (K)	CO ₂ (ppmv)	σ (m/s)
MOZART Control	MOZART	290.7	359.09	0.7
Cariolle Control	Cariolle	290.7	359.09	0.7
MOZART 4xCO ₂	MOZART	297.4	1436.37	0.7
Cariolle 4xCO ₂	Cariolle	297.4	1436.37	0.7
QBO MOZART Control	MOZART	290.7	359.09	1.0
QBO Cariolle Control	Cariolle	290.7	359.09	1.0

Note. The sea surface temperature is a global and annual mean.

The implementation of the Cariolle scheme in addition to the MOZART scheme in HAMMONIA has increased the total run time of the model by about 1%. In the ICON general circulation model (not used in this study; Giorgetta et al., 2018) the Cariolle scheme can be used optionally instead of a prescribed ozone climatology and increases the run time by about 5%. While exact numbers will depend on the specific computer, implementation, and model configuration, the computational cost of such a linearized ozone scheme can be considered negligible in comparison to complex schemes like MOZART, which usually increase the computing time of general circulation models by more than a factor of 2.

2.2. Cariolle Scheme: Linear Ozone Parameterization

We implemented the Cariolle scheme as described by Cariolle and Déqué (1986) and Cariolle and Teyssède (2007) in HAMMONIA. The scheme describes the net chemical ozone production in terms of a first-order Taylor series depending on ozone volume mixing ratio x_{O_3} , temperature T , the overhead ozone column $\overline{\mathcal{N}}_{O_3} = \int_z^\infty x_{O_3}(h)dh$ at any point of the atmosphere, and the zenith angle α . The ozone tendency is calculated using eight coefficients $A_1 \dots, A_8$, which are prescribed as zonal means and are interpolated from monthly means for every model time step:

$$\frac{dx_{O_3}}{dt} = A_1 + A_2(x_{O_3} - A_3) + A_4(T - A_5) + A_6(\overline{\mathcal{N}}_{O_3} - A_7) + A_8x_{O_3} \times \chi_{\{T \leq 195K\}} \times \chi_{\{\cos \alpha > 0\}}, \quad (1)$$

where χ_B is the characteristic function of set B ; that is, $\chi_B(x) = 1$ if $x \in B$ and $\chi_B(x) = 0$ otherwise. The coefficients A_3, A_5, A_7 represent the point in the multidimensional space at which the Taylor series was constructed and mean ozone mixing ratio, temperature, and the overhead ozone column at this point. The coefficient A_1 represents a net chemical ozone tendency, and A_2, A_4, A_6 the partial derivatives of this tendency with respect to ozone mixing ratio, temperature and the overhead ozone column, respectively. The coefficient A_8 accounts for heterogeneous chemistry in the Antarctic region.

The first two terms in equation (1) correspond to the traditional linearization of net photochemical loss and production rates of ozone, where $\tau = -1/A_2$ can be interpreted as the photochemical relaxation time. A_1 is most important in the low-latitude middle stratosphere. At altitudes where photochemistry is in a steady state ($A_1 = 0$), the first two terms reduce to a relaxation toward the zonal mean profile A_3 . The temperature dependency is described by the third term. Higher stratospheric temperatures slow down the production cycle of ozone causing lower ozone concentrations. The fourth term in equation (1) models the influence of the local ultraviolet (UV) flux on ozone concentrations. Less (more) ozone in the overhead atmosphere leads to an increase (decrease) of UV flux reaching lower altitudes and in turn to increasing (decreasing) ozone production due to molecular oxygen dissociation. This mechanism has its largest influence in the stratosphere and upper mesosphere. UV radiation also plays an important role in the activation of catalytic ozone destruction. Finally, the fifth term accounts for heterogeneous chemistry and is introduced by Cariolle and Teyssède (2007). Ozone is lost when the polar stratospheric temperature is lower than 195 K, and the Sun is over the horizon due to heterogeneous ozone destruction from chlorine activation on the surface of polar stratospheric cloud particles in high-latitude winter and spring.

We use Version 2.9 (from March 2017) of the A_i coefficients of the Cariolle scheme. They were generated by perturbing the 2-D photochemical model MOBIDIC (Cariolle & Brard, 1985) by $\pm 20\%$ for ozone mixing ratio and overhead ozone column and by ± 10 K for temperature (Cariolle & Teyssède, 2007).

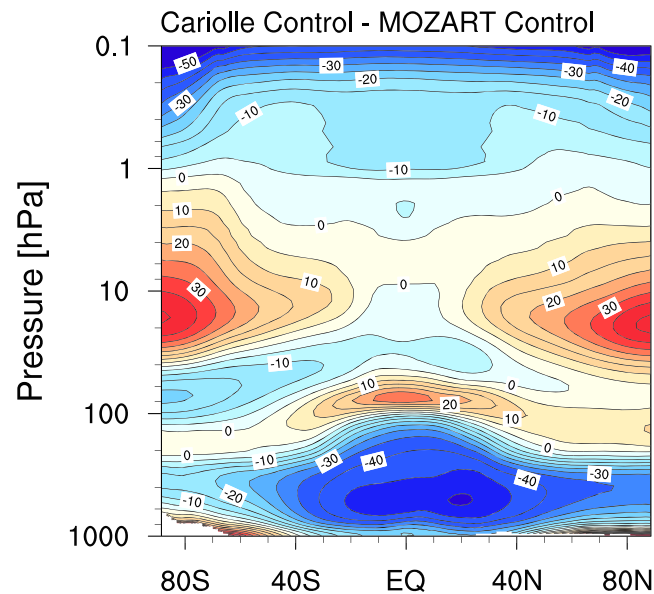


Figure 1. Relative difference in annual mean ozone volume mixing ratio (%) between Cariolle Control and MOZART Control.

Cariolle and Déqué (1986) found that the largest deviation in ozone distribution between their parameterization and their explicit chemistry is less than 12% (see their Figure 4). The largest errors are located in the lower stratosphere. However, we compare the Cariolle scheme to a different model with a different explicit chemistry. Hence, we expect larger differences between the Cariolle scheme and the MOZART chemistry than those found by Cariolle and Déqué (1986). Indeed, Figure 1 shows deviations up to 50%, mainly in the troposphere and polar lower stratosphere. MOZART simulates more ozone in the troposphere and less in the polar lower stratosphere than the Cariolle scheme and in turn the ARPEGE model used as original reference. Hence, the MOZART and ARPEGE chemistries differ quite substantially. Despite the larger deviations, our comparison method sheds light on the usefulness of the Cariolle scheme without prior tuning in climate models.

2.3. Observational Data

We compare results obtained with the linear scheme not only to an explicit chemistry scheme but also to observational and reanalysis data. In this section, we briefly describe the used data in order of appearance.

First, we evaluate the total column ozone using the National Institute of Water and Atmospheric Research-Bodeker Scientific v3.3 (NIWA-BS) total column ozone database (Bodeker et al., 2005; Struthers et al., 2009). These data combine total ozone measurements from various sources: the Global Ozone Monitoring Experiment 2 (GOME-2) instruments, four Solar Backscatter Ultraviolet (SBUV) instruments, the Ozone Monitoring Instrument (OMI), and the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument.

Second, the tropical ozone profile is compared to two climatologies: the Climatology 2011 from McPeters and Labow (2012) and the CMIP5 ozone data set from Cionni et al. (2011). The first climatology is based on observations of the Microwave Limb Sounder (MLS) instrument on the Aura satellite from 2004 to 2010 and data from balloon sondes from 1988 to 2010. The latter climatology has been created in support of CMIP5 using a multiple linear regression analysis of SAGE I+II satellite observations and polar ozonesonde measurements for the period from 1979 to 2009. We averaged each climatology between 10°S and 10°N to obtain a tropical ozone profile.

Third, we evaluate the ozone anomalies induced by the QBO using winds from the Free University of Berlin data, which is based on soundings from Singapore, and the ozone profiles from the Southern Hemisphere Additional Ozonesonde (SHADOZ) network (Witte et al., 2017), which provides ozone sonde records from seven tropical stations. We average over data from three stations around the equator (San Cristobal, Nairobi, and Kuala Lumpur) to obtain a monthly mean tropical ozone profile of high vertical resolution

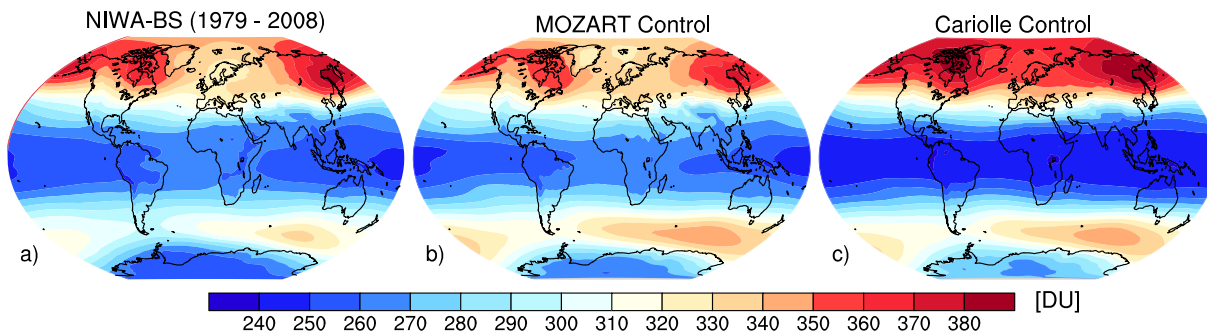


Figure 2. Total ozone column (DU) from (a) National Institute of Water and Atmospheric Research-Bodeker Scientific (NIWA-BS) observational data and two HAMMONIA simulations: (b) MOZART Control and (c) Cariolle Control.

(1,013–3 hPa). Measurements from 1998 to 2017 are used for Nairobi and Kuala Lumpur, while for San Cristobal only data until 2016 are available. More details on SHADOZ and its comparison to satellite data can be found in Witte et al. (2017) and Thompson et al. (2017).

Finally, Modern-Era Retrospective Analysis for Research and Applications 2 (MERRA-2; Rienecker et al., 2011) reanalysis data is used to evaluate the zonal asymmetry of ozone in the Northern Hemisphere. An average over 28 years (1980–2007) is used.

3. Results

3.1. Ozone Distribution and Variability

First, we concentrate on the simulations of the late twentieth century, that is, on the experiments MOZART Control and Cariolle Control, analyzing whether the Cariolle scheme reproduces a reasonable ozone distribution and variability. In the next section, our focus lies on the response to global warming.

Important features of the total ozone column are well reproduced by the explicit and the linear chemistry (see Figure 2). Compared to the National Institute of Water and Atmospheric Research-Bodeker Scientific data (see section 2.3), the model simulations exhibit too much ozone in the southern high latitudes. But the global means of both schemes (MOZART Control: 292 DU; Cariolle Control: 287 DU) agree well with the observations (287 ± 3 DU). The differences in total ozone column between the Cariolle and MOZART chemistries are within 10%. However, the Cariolle scheme models a stronger latitudinal gradient in total ozone column than the MOZART scheme. In the tropics, the Cariolle ozone column is substantially smaller than the MOZART column. This is almost entirely caused by much less ozone in the tropical troposphere in Cariolle compared to MOZART (see Figure 1). The difference in the ozone columns calculated between 1,000 and 100 hPa is 14.8 DU and almost identical to the difference in the total tropical ozone column of 14.3 DU. Although the Cariolle parameters are prescribed zonally, the zonal structure is similar to the two other data sets. More specifically, a wave-two structure is reproduced in the northern high latitudes and a wave-one pattern in the southern midlatitudes. The wave structure in the northern high latitudes is further analyzed in section 3.1.2.

Of course, different meteorology simulated in the relatively short experiments could cause part of the differences in ozone. However, the column ozone distribution calculated by the Cariolle scheme in the MOZART Control experiment (not shown), which experiences exactly the same meteorology as the MOZART ozone presented in Figure 2, is very similar to the distribution from Cariolle Control. The described differences can, hence, be attributed to the different chemistry schemes.

As the suggested ozone feedback on climate sensitivity has been attributed to changes in the tropics, next, we compare the tropical (averaged over 10°S to 10°N) ozone profiles from the Cariolle and MOZART schemes (Figure 3). While the profiles from both Control simulations are very similar, compared to the observation-based climatologies, both HAMMONIA simulations show too little ozone in the upper stratosphere (10–1 hPa). Below 10 hPa the ozone profile is well reproduced and within one standard deviation of the annual mean from the observed climatology (see Figure 3b). In general, we conclude that both the comprehensive and the linear scheme simulate a reasonable tropical ozone profile.

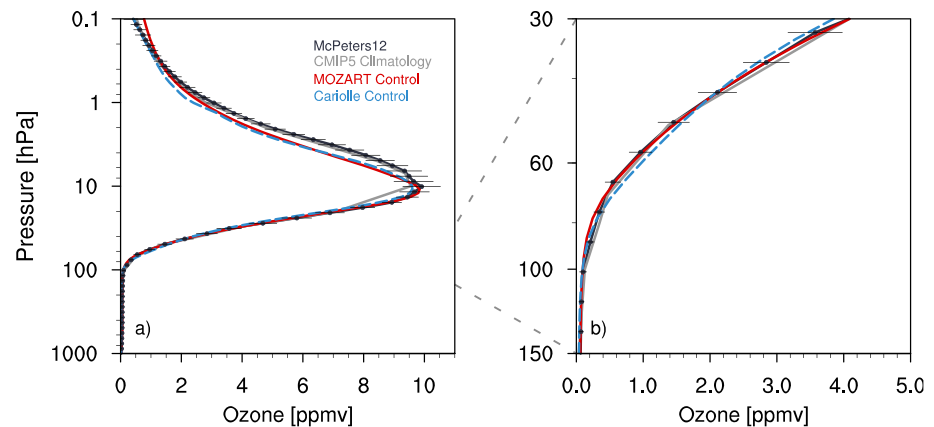


Figure 3. (a) Tropical (10°S to 10°N) ozone profiles (ppmv) for two climatologies (McPeters & Labow, 2012 and Cionni et al., 2011) and two HAMMONIA simulations: MOZART Control (red solid line) and Cariolle Control (blue, dashed line). The error bars represent 1σ standard deviation from the McPeters and Labow (2012) climatology. (b) The same as (a) but zoomed in on the lowermost stratosphere (200–20 hPa).

In the following we analyze two aspects of ozone variability: tropical vertical anomalies caused by the QBO and zonal inhomogeneities attributed to extratropical planetary waves.

3.1.1. Sensitivity to the QBO

The QBO is an oscillation of zonal winds between easterlies and westerlies in the tropical stratosphere (see Baldwin et al., 2001, and references therein). The QBO dominates the interannual variability of tropical stratospheric ozone.

The tuned-QBO simulations show a QBO-like oscillation, but with a period shorter than in observations. The FU-Berlin data for 1987–2018 (see section 2.3) show a mean QBO period of 27.8 ± 3.5 months, whereas the MOZART QBO simulation has a mean QBO period of 20.5 months and the Cariolle QBO simulation of 23.5 months. However, the QBO amplitude at 20 hPa is well reproduced in the model simulations. In the observations, the QBO oscillates between 16.0 ± 4.1 m/s (west phase) and -32.0 ± 5.5 m/s (east phase). In our model simulations, the equatorial zonal wind varies between 16.3 (17.1) m/s for the west phase and -27.0 (-27.3) m/s for the east phase in the MOZART (Cariolle) QBO experiment.

We calculate QBO ozone anomalies by removing the mean from QBO-east and QBO-west composites. We use the zonal wind speed at 20 hPa as criterion: If its monthly mean is below -5 m/s, this month is classified as QBO-east phase, a month with a wind speed above $+5$ m/s is classified as QBO-west phase. All months for each phase are then averaged. We compare the ozone anomalies from the two QBO experiments to a tropical mean of the SHADOZ database (see section 2.3). As SHADOZ does not provide zonal wind speeds, we use the FU-Berlin QBO data to distinguish between QBO-east and QBO-west phase for the SHADOZ data.

Figure 4 shows the ozone anomalies for the QBO-east and QBO-west phases. In general, ozone is reduced during a QBO-east phase and increased during a QBO-west phase in the lower stratosphere. During a QBO-east phase the strength of the lower stratospheric tropical upwelling is increased as part of the secondary circulation caused by the QBO, and vice versa for the QBO-west phase. This leads to a negative (positive) ozone anomaly during a QBO-east (QBO-west) phase (Gray & Pyle, 1988). The Cariolle and MOZART chemistries reproduce the structure of the observed ozone anomalies qualitatively, but the amplitudes of the ozone anomalies around 30 hPa are smaller for both model simulations. Especially, the Cariolle ozone generates only about half of the amplitude at 30 hPa compared to the observations. At least a part of the general underestimation of the QBO signal may be related to deficiencies in the simulated QBO winds, but a perfect match cannot be expected in a free running simulation.

More important for the evaluation of the usefulness of linear ozone schemes is again the question whether the differences between MOZART and Cariolle ozone anomalies are due to the different chemistry schemes or merely due to different dynamics. Therefore, we use the Cariolle ozone calculated in the MOZART Control experiment, which has the advantage that the dynamics are the same as for the MOZART ozone. Below 10 hPa the ozone anomalies calculated by the Cariolle scheme in both experiments (cf. gray and blue lines in Figure 4) are very similar. Thus, the difference between the MOZART and Cariolle ozone anomalies are

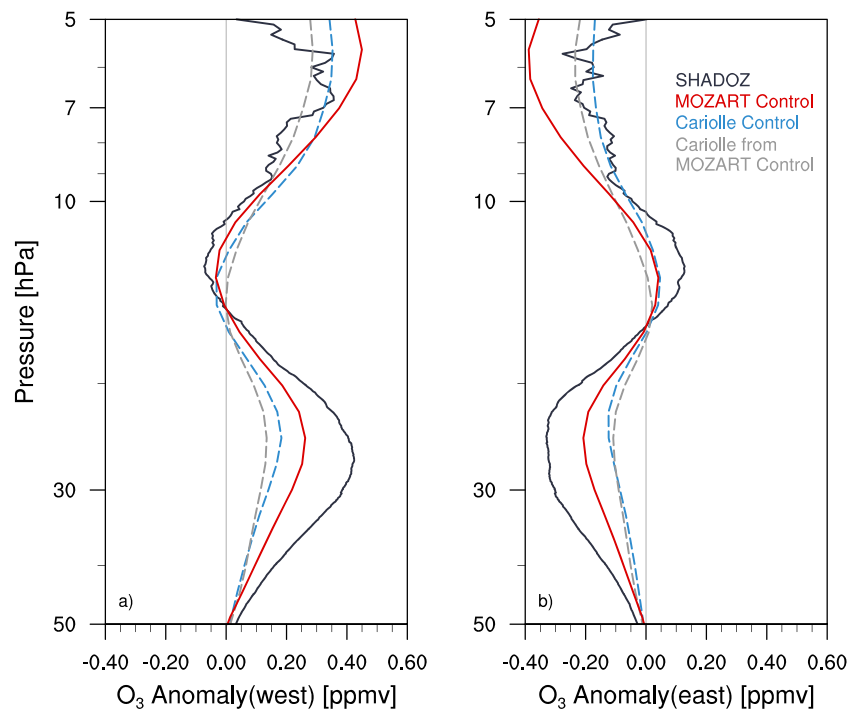


Figure 4. Ozone anomalies (ppmv) averaged between 5°S to 5°N for (a) QBO-west and (b) QBO-east phase for SHADOZ ozonesondes and two HAMMONIA experiments: MOZART QBO (red, solid line) and Cariolle QBO (blue, dashed line). The gray, dashed line represents the Cariolle ozone from the MOZART QBO experiment.

mainly due to the different chemistry schemes. Part of the difference between observed and simulated ozone anomalies may be due to differences in the simulated and real secondary stratospheric circulation patterns caused by the QBO, but the weaker amplitude of the Cariolle ozone in comparison to MOZART indicates a general limitation of the linear scheme.

3.1.2. Zonal Asymmetry

The second variability feature we analyze are the ozone deviations from the zonal mean in middle and high latitudes. The zonal asymmetry in ozone can provide a radiative feedback on the wave structure in temperature and circulation and affect the zonal mean state and stratosphere-troposphere coupling, as mentioned in the introduction.

Figure 5 shows the quasi-stationary zonal ozone asymmetry at 10 hPa in the Northern Hemisphere for January. A predominant wave-one structure is evident in the reanalysis data and in all simulations. However,

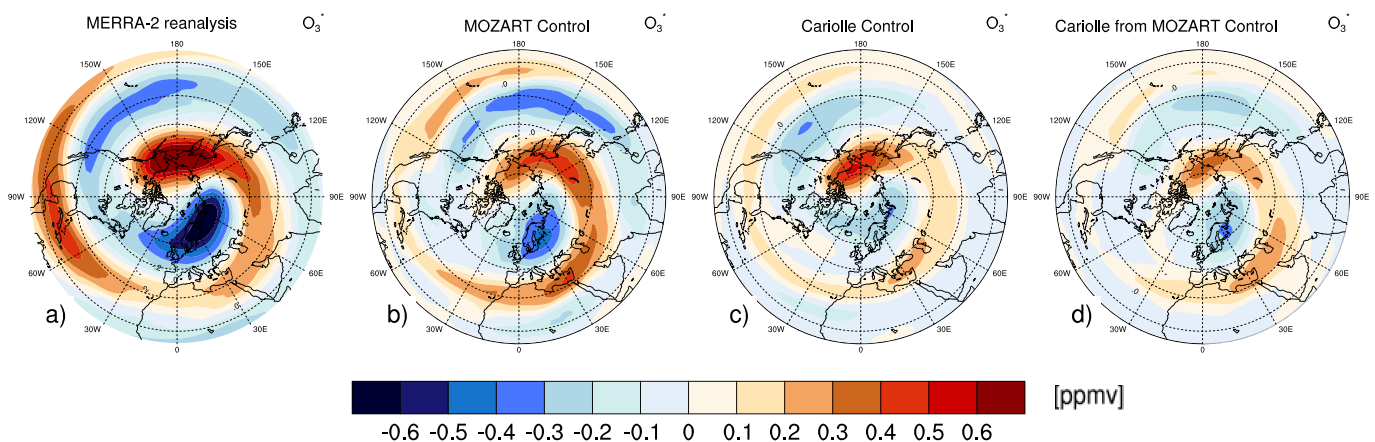


Figure 5. Deviations from zonal mean ozone (O_3^*) (ppmv) at 10 hPa for January from (a) MERRA-2 reanalysis data (1979–2007), (b) MOZART Control, (c) Cariolle Control, and (d) Cariolle ozone from the MOZART Control experiment.

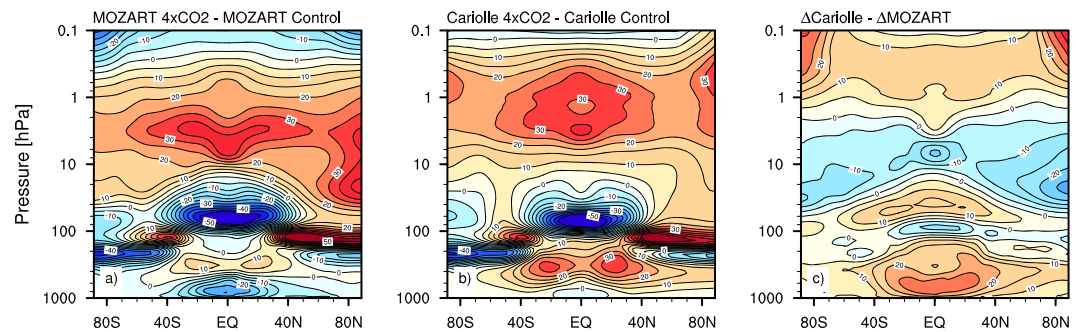


Figure 6. Relative response in annual mean zonal mean ozone (%) for the $4\times\text{CO}_2$ experiments for (a) MOZART chemistry, (b) Cariolle chemistry, and (c) its difference.

compared to the reanalysis data the amplitudes in the model simulations are too weak, particularly for the Cariolle ozone. Different dynamics between the reanalysis and the model simulations may contribute to this. Indeed, the zonal temperature deviation is much weaker in the model than in the reanalysis. But similarly as in the case of QBO variability, the amplitude of the wave structure in Cariolle ozone is also weaker than in MOZART ozone when both are taken from MOZART Control (Figures 5b and 5d) and have, hence, experienced the same dynamics. One reason for the weaker Cariolle ozone amplitude is likely that the Cariolle parameters are prescribed as zonal averages, which may dampen the wave structure. Furthermore, at this altitude, the zonal mean meridional ozone gradient is smaller in the Cariolle experiment than in the MOZART experiment resulting in a weaker contribution of meridional advection to ozone anomalies. Both HAMMONIA simulations show a much smaller meridional ozone gradient than the reanalysis data explaining at least part of the differences between model and reanalysis data. Analysis of simulated ozone patterns in high-latitude austral spring (not shown) indicates a similar relation between wave amplitude (weaker in Cariolle than in MOZART) and zonal mean meridional gradient.

3.2. Ozone Changes Under Quadrupling of CO_2

To assess the response of the linear ozone scheme to global warming, we carried out experiments with quadrupled atmospheric CO_2 and a 6.7 K SST increase (see section 2.1). This SST increase is estimated as mean equilibrium response for CMIP5 models. Thus, the imposed temperature increase is higher than what coupled atmosphere-ocean models in general simulate only 100 or 150 years after a quadrupling of CO_2 as equilibrium is not reached at that time.

Figure 6 shows the response of ozone in the $4\times\text{CO}_2$ experiments for the two different chemistry schemes. In both schemes ozone decreases in the tropical lower stratosphere by $\sim 50\%$ and increases above by $\sim 35\%$. However, the maximum increase is for the Cariolle scheme at a higher altitude than in MOZART. In the lower stratosphere (60–20 hPa) the explicit chemistry simulates a stronger decrease in ozone for $4\times\text{CO}_2$ than the linear chemistry. Furthermore, the Cariolle scheme simulates a larger ozone increase in the tropical troposphere (about 30%) than the MOZART scheme (10%). Note that neither scheme is specifically designed for a realistic representation of tropospheric chemistry. Again, differences in the responses of the Cariolle ozone between the Cariolle and MOZART simulations are small (not shown), which indicates that all above mentioned differences between MOZART and Cariolle are caused by the different chemistries and not by different dynamics.

Ozone changes simulated in our $4\times\text{CO}_2$ experiments are well in the range of results from other chemistry-climate models. Chiodo et al. (2018) compared the ozone response to instantaneous quadrupling of CO_2 in four different chemistry-climate models and reported decreases in the tropical lower stratosphere between about 30% and 50% and increases in the upper stratosphere by 40% to 50%. Figure 7a compares the absolute changes in tropical mean ozone for $4\times\text{CO}_2$ from the MOZART and Cariolle scheme to these four models. The difference between the chemistry schemes in HAMMONIA is as large as differences between different chemistry-climate models are in general. Chiodo et al. (2018) have identified a strong intermodel correlation between changes in upwelling in the tropical tropopause region and changes in tropical lower stratospheric ozone. Figure 7c is a reprint of Figure 6b from (Chiodo et al., 2018) showing this correlation but with both HAMMONIA experiments added. Concerning this quantity, the responses of the HAMMONIA experiments

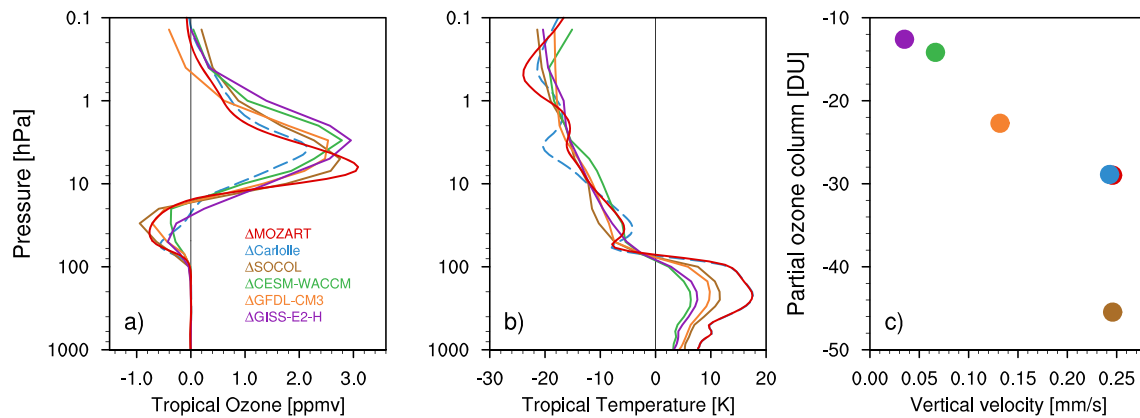


Figure 7. Tropical (10°S to 10°N) response in annual mean (a) ozone (ppmv) and (b) temperature (K) for the HAMMONIA experiments and four other chemistry-climate models (GISS-EH-2, GFDL-CM3, CESM-WACCM, and SOCOL). (c) Scatterplot of vertical velocity (mm/s) at 100 hPa versus lower stratospheric (100–20 hPa) ozone column (DU) averaged between 22°S and 22°N in response to quadrupling of CO₂. The data of the other models is from Chiodo et al. (2018).

are almost identical. The correlation between changes in vertical wind and ozone drops from -0.98 to -0.86 after including both HAMMONIA experiments, but their responses are well in the range of the other models.

The described ozone changes contribute to the temperature responses in the $4\times\text{CO}_2$ experiments (Figure 8), which are very similar for the linear and explicit chemistry schemes. The zonal mean differences between the chemistry schemes are less than 3 K everywhere. The largest discrepancies emerge in the tropical middle atmosphere and resemble changes due to different QBO phases. The temperature changes from both schemes are in good agreement with the results of the four chemistry-climate models presented by Chiodo et al. (2018) (Figure 7b). The larger temperature response of HAMMONIA in the troposphere is caused by the larger SST increase compared to the chemistry-climate models which are coupled to an ocean and have not fully equilibrated at the time for which results are shown. However, all models exhibit a similar lower stratospheric cooling to which the ozone reduction contributes up to slightly more than 2 and 3 K in the simulations by Marsh et al. (2016) and Nowack et al. (2015), respectively. Part of the ozone feedback on climate sensitivity reported by several studies would be caused by less water vapor entering the stratosphere due to the cooling effect of the ozone reduction on the tropical tropopause (e.g., Dacie et al., 2019).

We conclude that the responses of ozone in the tropical lower stratosphere in the $4\times\text{CO}_2$ experiments are very similar for the two chemistry schemes and also in the range of what is predicted by other chemistry-climate models. This is likely related to ozone in the tropical lower stratosphere being rather controlled by transport than chemistry. Figure 9 compares the photochemical lifetime of ozone and vertical transport constants. The photochemical lifetime is estimated as $-1/A_2$ from equation (1) (see also Figure 3 from Cariolle & Teysse re,

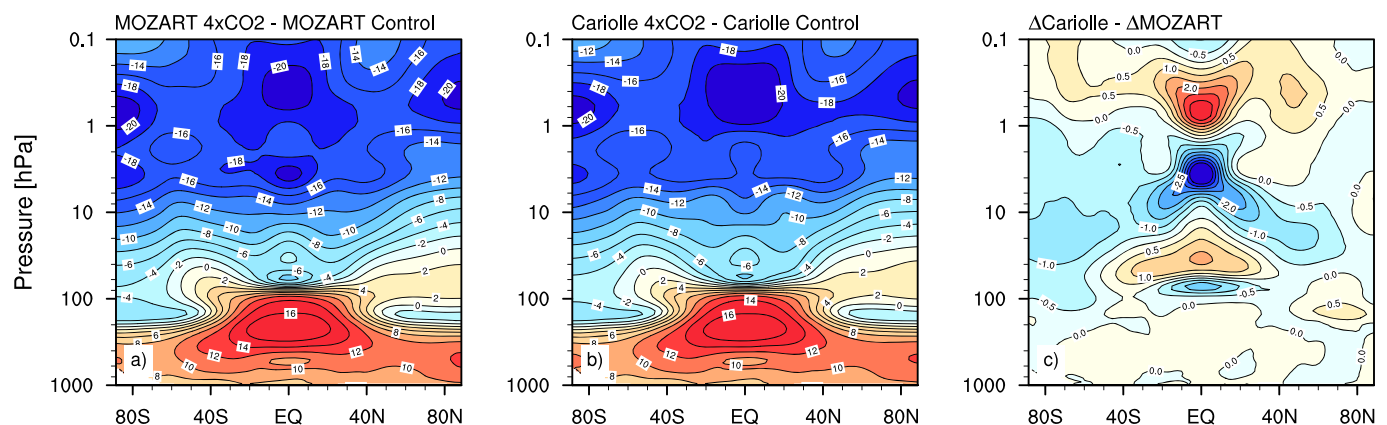


Figure 8. Response in annual mean zonal mean temperature (K) for the $4\times\text{CO}_2$ experiments for (a) MOZART chemistry, (b) Cariolle chemistry, and (c) its difference.

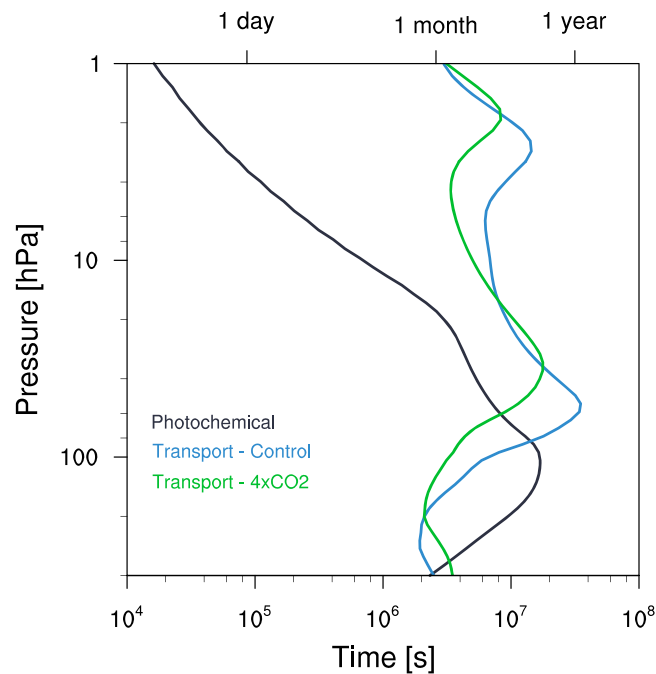


Figure 9. Profiles of tropical (10°S to 10°N) and annual mean photochemical lifetime and time constant of vertical transport (s) for Cariolle Control and Cariolle $4\times\text{CO}_2$.

2007). The vertical transport constant is approximated by $-w/H_i$, where we assume for the scale height of ozone $H_i = 5$ km (Brasseur & Solomon, 2005). For w , we use the tropical mean of residual vertical wind. In the lower stratosphere (between 300 and 70 hPa) the photochemical lifetime is larger than the dynamical time constant. The vertical transport constants for MOZART Control and Cariolle Control are comparable inducing similar changes in ozone in the $4\times\text{CO}_2$ experiments. This result is in line with the findings of Chiodo et al. (2018), who showed that the changes in ozone are highly negatively correlated to changes in the lower stratospheric upwelling (see Figure 7c).

4. Summary and Conclusion

In this study, we assessed how useful a linear ozone parameterization is for global climate modeling. Simulations with the general circulation and chemistry model HAMMONIA were carried out including the linear Cariolle ozone scheme (Cariolle & Teyss  dre, 2007). We compared simulations with this ozone parameterization to simulations with the comprehensive MOZART chemistry scheme. With respect to the climatological distribution and variability of ozone, we compared the simulations further to observations, with respect to their responses to global warming to earlier simulations with comprehensive chemistry-climate models. Running the MOZART and Cariolle schemes in parallel in HAMMONIA, while letting only one of the ozone fields interact with the model's radiation code, enables to identify which differences between simulated fields can be uniquely traced back to the chemistry schemes.

Differences in total column ozone between the Cariolle scheme and the explicit chemistry are within 10%. Similarly, the tropical ozone profile agrees very well for both chemistry schemes and with observations. However, this can be assumed to be a specific feature of this particular scheme and not general to linear approaches.

In terms of ozone variability we concentrated on signals caused by the QBO and by extratropical quasi-stationary planetary waves. Qualitatively, for both variability features the Cariolle scheme agrees well with the MOZART scheme and with observations. However, the amplitudes of the variability patterns simulated by the linear approach are up to about 50% smaller than by the explicit scheme. The relaxation toward a climatological mean state, implicit in linear approaches, obviously reduces amplitudes of circulation-induced variability patterns on time scales of the QBO as of quasi-stationary planetary waves.

The responses of ozone in the 4xCO₂ experiments are almost identical for the comprehensive and the linear ozone scheme, and both responses are well in the range of simulations with other chemistry-climate models (Chiodo et al., 2018). The reduction of ozone in the lower tropical stratosphere, which has been identified as particularly relevant for an ozone feedback on climate sensitivity, differs only slightly between the two schemes. As ozone in this region is sensitive mostly to changes in tropical upwelling and the chemical lifetime is relatively long, intrinsic deficiencies of linear approaches should a priori not matter much, here. A possible issue could be an upward shift of the tropopause, because some of the coefficients used in the Cariolle and other linear schemes have relatively strong vertical gradients in this region. Anyhow, it is impossible to say which of the responses is more realistic because also in this region they are in the range of responses reported by Chiodo et al. (2018).

It is clear that even with today's computational capacities comprehensive chemistry schemes are too expensive for many modeling approaches in climate research like large ensembles or convective-permitting resolutions. From our analyses we conclude that computationally cheap linear ozone schemes like the Cariolle scheme can be useful tools in such cases. For the zonal mean response of stratospheric ozone to global warming, possible deficiencies of the linear approach seem not to matter much even for a relatively strong forcing as the quadrupling of the atmospheric CO₂ concentration. With respect to ozone-circulation feedbacks, our analysis shows that comprehensive chemistry schemes are superior to the linear scheme in reproducing more realistic amplitudes of variability patterns. However, the linear scheme has certainly advantages over the prescription of climatological ozone fields also in this respect.

In this study, we only used the Cariolle scheme, but there is no reason to expect that other linear ozone parameterizations should perform in principle very differently. Their application in global climate change simulations, for example, of CMIP type, should be advantageous in comparison to the still widespread prescription of ozone climatologies. We expect even smaller differences between explicit and linear chemistry schemes when the coefficients for the linear scheme were derived from the chemistry model to which the linear scheme is compared, which was not the case in this study. More detailed parameterization approaches have been developed (e.g., SWIFT—Kreyling et al., 2018; Wohltmann et al., 2017), which may work better in particular in situations where ozone chemistry behaves very nonlinear. Machine learning approaches as proposed by Nowack et al. (2018) may provide another option in the future. However, also the further development of linear parameterizations to include, for example, effects of solar variability, volcanic eruptions, or even orbit variability could be envisioned.

Data Availability Statement

Primary data of the model simulations and scripts used in the analysis and other supporting information that may be useful in reproducing this work are archived by the Max Planck Institute for Meteorology and can be accessed online (<http://hdl.handle.net/21.11116/0000-0005-61B8-E>). The NIWA-BS data set can be obtained from the www.bodekerscientific.com/data/total-column-ozone website. The Climatology 2011 can be obtained from the Goddard anonymous ftp account (<ftp://toms.gsfc.nasa.gov/pub/ML&urlscore;climatology>). The CMIP5 ozone climatology is available at the website (cmip.llnl.gov/cmip5/forcing.htm#ozone&urlscore;forcing). The zonal mean zonal wind data from the FU Berlin can be obtained from the website (www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/index.html). The SHADOZ data are available at the tropo.gsfc.nasa.gov/shadoz/ website. The MERRA-2 data can be obtained from the NASA Goddard Earth Sciences Data and Information Services Center. Last access to all data was on 25 April 2019 for this study.

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