Stratospheric Ozone and Temperature Simulated from the Preindustrial Era to the Present Day

JOHN AUSTIN
Princeton, New Jersey

LARRY W. HOROWITZ, M. DANIEL SCHWARZKOPF, R. JOHN WILSON, AND HIRAM LEVY II
NOAA/Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey

(Manuscript received 19 March 2012, in final form 10 September 2012)

ABSTRACT

Results from the simulation of a coupled chemistry–climate model are presented for the period 1860 to 2005 using the observed greenhouse gas (GHG) and halocarbon concentrations. The model is coupled to a simulated ocean and uniquely includes both detailed tropospheric chemistry and detailed middle atmosphere chemistry, seamlessly from the surface to the model top layer centered at 0.02 hPa. It is found that there are only minor changes in simulated stratospheric temperature and ozone prior to the year 1960. As the halocarbon amounts increase after 1970, the model stratospheric ozone decreases approximately continuously until about 2000. The steadily increasing GHG concentrations cool the stratosphere from the beginning of the twentieth century at a rate that increases with height. During the early period the cooling leads to increased stratospheric ozone. The model results show a strong, albeit temporary, response to volcanic eruptions. While chlorofluorocarbon (CFC) concentrations remain low, the effect of eruptions is shown to increase the amount of HNO₃, reducing ozone destruction by the NOₓ catalytic cycle. In the presence of anthropogenic chlorine, after the eruption of El Chichón and Mt. Pinatubo, chlorine radicals increased and the chlorine reservoirs decreased. The net volcanic effect on nitrogen and chlorine chemistry depends on altitude and, for these two volcanoes, leads to an ozone increase in the middle stratosphere and a decrease in the lower stratosphere. Model lower-stratospheric temperatures are also shown to increase during the last three major volcanic eruptions, by about 0.6 K in the global and annual average, consistent with observations.

1. Introduction

Climate model simulations participating in the Coupled Model Intercomparison Project phase 3 (CMIP3) in support of the Intergovernmental Panel on Climate Change Fifth Assessment Report (IPCC) Fourth Assessment Report (Solomon et al. 2007) were restricted in two important ways by the computer power then available. First, the models specified zonally symmetric stratospheric ozone. As a result, the model dynamics (in particular planetary waves) are unrelated to the ozone field, therefore causing the two fields not to be coupled in the way that they are in the atmosphere. Detailed atmospheric behavior in the vicinity of the tropopause is also poorly simulated as the ozone climatology used for model forcing tends to be described as a function of pressure, rather than relative to the tropopause. As a result, cooling rates in the vicinity of the tropopause are poorly simulated in comparison with measurements (Forster et al. 2007). Second, about half of the CMIP3 models prescribed constant ozone, rather than using a time-varying climatology (Son et al. 2008).

In addition, the models had poor stratospheric resolution, with many models having an upper boundary as low as 5 hPa. Since the Brewer–Dobson circulation in the atmosphere rises to at least 1 hPa before turning and affecting the lower stratosphere middle- and high-latitude region, the whole of the stratospheric circulation of the models does not correspond closely with the atmospheric behavior. As a result such models are unable to represent properly the impact of stratospheric...
changes on the troposphere. These changes have been better understood in the last decade using observational and theoretical studies (e.g., Baldwin and Dunkerton 2001; Thompson and Solomon 2002; Perlwitz et al. 2008; Son et al. 2009). In particular, three of the aforementioned studies have pointed out the significance of the Antarctic ozone hole for tropospheric climate. Thus, a proper treatment of stratospheric influences requires a detailed stratospheric chemistry scheme and a well-resolved stratosphere, such as typically used for stratospheric purposes (e.g., Morgenstern et al. 2010).

Simulations of a climate model with tropospheric chemistry have previously been completed for the preindustrial period by Shindell et al. (2006), but their model excluded stratospheric processes. Simulations of the Canadian Middle Atmosphere Model have also recently been completed (Scinocca et al. 2008) with the coupled stratospheric chemistry of de Grandpré et al. (2000). This model includes a coupled ocean and stratospheric chemistry, but not tropospheric chemistry. Thus previous climate models have lacked either tropospheric chemistry or stratospheric chemistry, or both. This gap has now been filled, in preparation for CMIP5, by the Geophysical Fluid Dynamics Laboratory (GFDL) Coupled Model version 3 (CM3) (Donner et al. 2011). The atmospheric model incorporates a detailed aerosol scheme and coupled tropospheric chemistry (Naik et al. 2012, manuscript submitted to J. Geophys. Res.), as well as the usual climate model processes. In particular, the model has a comprehensive stratosphere with a resolution high enough for stratospheric studies (Austin and Wilson 2010). Also, the chemistry scheme includes all of the important stratospheric reactions. The development of a fully coupled atmosphere–ocean climate model such as CM3, including detailed, seamless stratospheric and tropospheric chemistry, was a key overall recommendation of the 2010 Stratosphere–Troposphere Processes and Their Role in Climate (SPARC) Chemistry–Climate Model Validation (CCMVal) report (SPARC CCMVal 2010). In this paper, the stratospheric temperature results and ozone distributions from the model are investigated to determine the likely effects of natural and anthropogenic processes on the historical stratosphere (1860–2005), including the effects of volcanoes and human produced halocarbons.

2. Description of model and simulations

We use the fully coupled atmosphere–ocean climate model described in detail by Donner et al. (2011). The model includes an online atmospheric chemistry scheme that incorporates both tropospheric and stratospheric processes. The tropospheric chemistry is based on Horowitz et al. (2003) and Horowitz (2006) as described by Naik et al. (2012, manuscript submitted to J. Geophys. Res.) The stratospheric chemistry is based on Austin and Wilson (2010) and includes the full range of gas phase reactions covering the HOx, NOx, ClOx, and BrOx catalytic cycles. Heterogeneous reactions are included in ternary solutions, and on the surfaces of nitric acid trihydrate (NAT) and ice polar stratospheric clouds (PSCs). All chemical reactions are included throughout the model atmosphere. Thus, for example, nonmethane hydrocarbon reactions are simulated in the stratosphere as well as the troposphere, even though in the former case the molecular concentrations may be very low and scarcely affect the main chemistry. This ensures that there is no artificial transition level in the model where the reaction set changes, thus increasing the overall validity of the results.

For the radiative forcing, a limited number of chlorofluorocarbon species were considered (CFC-11, CFC-12, CFC-113, HCFC-22), but for the halogen chemistry all known contributors were included in parameterized form [as described in detail by Austin and Wilson (2010)]. These contributors include additional bromine from very short-lived organic source gases. The CFCs, HCFCs, halons, and short-lived species were included by specifying volume mixing ratios at the model lower boundary condition as a function of time (SPARC CCMVal 2010). The model includes an 11-yr solar cycle affecting radiation and photolysis, but the impact of this solar variability on the model chemistry has not been assessed. The model does not simulate a quasi-biennial oscillation (QBO).

The model has been run for a range of experiments for the IPCC Fifth Assessment Report (AR5). In this paper, we present results from the ensemble runs for the historical atmosphere, in which all forcings (greenhouse gas and halocarbon concentrations, solar fluxes, and volcanic aerosols) were specified from observations. Five ensemble members are presented, where the initial conditions were taken 50 years apart from a long 1860 control run of the same model. There were no statistically significant stratospheric trends in the 1860 control for either ozone or temperature.

Similar simulations, but for a limited range of experiments (essentially 1960 to 2100 with all forcings), were completed with stratospheric models for the CCMVal report (SPARC CCMVal 2010). The GFDL model used there is compared in general terms with the results obtained in this work. However, we have not performed a thorough comparison of the current model with the CCMVal results.
3. Model results: Ozone

a. Ozone evolution

Figure 1 shows the ensemble mean of the annually averaged total column ozone simulated for the period 1860 to 2005 as a function of latitude and time. In the tropics there is no clear trend. In middle latitudes there is a tendency for the ozone column to increase during the simulation in both hemispheres until about 1980. Thereafter the ozone columns start to decrease, especially in the Southern Hemisphere. Although it is only a seasonal phenomenon, the Antarctic ozone hole is particularly noticeable as a decrease in the annual-mean column ozone from about the late 1980s. Ozone loss in the Arctic is somewhat smaller and barely perceptible in this framework. Overall, the model results tend to be biased low in the Arctic by up to 40 Dobson units (DU) and high in the southern midlatitudes by up to 20 DU compared with measurements of the contemporary atmosphere. This is consistent with the known performance of the previous stratospheric version of the climate model (Austin and Wilson 2006), which does not include tropospheric chemistry. A comparison between model and Total Ozone Mapping Spectrometer (TOMS) data is shown in Fig. 2. This comparison is similar to that presented in Donner et al. (2011) and indicates that Arctic and Antarctic spring ozone are too low compared with observations, although in the annual mean (excluding the period of darkness) the Arctic is in agreement with observations. Improvements of photolysis rate calculations and explicit simulation of CFCs (rather than the parameterized method used here) have significantly reduced the ozone biases over both poles. However, no century-length simulations have been performed with this improved model because of computer time limitations.

The increased column ozone in the midlatitudes is due to the excessive strength of the Brewer–Dobson circulation, which transports more ozone from the tropics (e.g., Shepherd 2008). After about 1990 the reduction of midlatitude column ozone is due to chlorine. This is due to ozone loss occurring locally, as well as in the ozone hole, followed by transport to midlatitudes (Austin and Wilson 2006; Shepherd 2008; see also section 6 herein).

A more detailed indicator of ozone change is provided by the vertically varying ozone mixing ratio, as shown in Fig. 3. Overall, global mean ozone (top) changes very little until the middle 1970s when upper-stratospheric halogen loss began. The upper-stratospheric change had little impact on the total column in Fig. 1. Below 10 hPa a number of significant ozone changes occurred, including ozone increases around 1883 and 1902 and also in the 1960–80 period. These results are investigated in more detail in section 5. The occurrence of the ozone hole, which started to become significant after 1980, is noticeable in the global average in the lower stratosphere in Fig. 3 (top).

During the spring periods, interannual variability is larger, particularly in the polar regions, but ozone depletion is also larger there. The middle and bottom panels in Fig. 3 show the ozone mixing ratio results obtained for March and September averaged poleward of 60° latitude for (boreal) north and (austral) south, respectively. Prior to 1980 there is a considerable amount of interannual variability between 7 and 100 hPa. After 1980, in the Arctic (Fig. 3, middle) ozone loss is significant in two regimes: in the upper stratosphere, where gas phase chlorine has a significant role; and in the lower stratosphere, where heterogeneous chlorine chemistry (with substantial interannual variability) is important. In the middle stratosphere the ozone concentration is frequently higher than the historical (model 1860) value due to stratospheric cooling, which slows ozone catalytic destruction. In the Antarctic (Fig. 3, bottom) similar behavior occurs, but the midstratospheric interannual variability tends to be larger in the 1980s when the Antarctic ozone hole was developing. Also, there is an indication in Fig. 3 (bottom) that the ozone hole extends too far in the vertical, by about 5 km at both its top and base. This weakness occurred also in previous simulations of the model with simplified tropospheric chemistry (Austin et al. 2010). The reason for this has now been identified as mainly due to the parameterized
CFC concentrations (Austin and Wilson 2010) used in the model, as mentioned above, as well as temperature biases in the polar regions. While absolute values of ozone differ from observations by more than 10% at times, it is anticipated that overall trends are reliable for both ozone and temperature.

b. Ozone trends

In this subsection we investigate the absolute and relative ozone mixing ratio trends. The annual mean ozone trends for the first 100 years of the model simulation are indicated in Fig. 4. The trends were calculated...
from annual mean values and autocorrelation is excluded. The trend is significantly different from zero (shading) in the upper stratosphere and in the troposphere. In both cases ozone amounts have increased. In the middle stratosphere simulated ozone has decreased, with a slightly larger trend in the Southern Hemisphere where it is statistically significant. Although statistically significant, the stratospheric ozone change is at most about 3% (0.3% decade$^{-1}$) from 1860 to 1959, while the stratospheric ozone trend in the control run (not shown) is negligible throughout the domain, as expected.

The contemporary ozone trends are shown in Fig. 5. The stratosphere results are in good agreement with previous simulations by the Atmospheric Model with Transport and Chemistry (AMTRAC) and AMTRAC version 3 (AMTRAC3) (Austin and Wilson 2006, 2010). After 1980, model ozone changes are statistically significant except for small regions in the middle and lower stratosphere. The results show the observed upper-stratospheric ozone loss (Randel and Wu 2007)
(due to chlorine increase), the loss of ozone in the Antarctic ozone hole, and the loss of Arctic ozone. Since the control run ozone trends are negligible, the model’s ozone changes will be taken to result from the external forcings applied during the simulations, which include not only changes in halogen-containing species, but also changes in CH$_4$, H$_2$O, and N$_2$O.

4. Model results: Temperature

a. Temperature evolution

The globally averaged temperature anomaly in the model as a function of pressure and time is shown in Fig. 6 for the ensemble mean of the experiments. Since in the global average the effects of dynamical variations are removed, those remaining are due to chemistry, radiation, and aerosols. From about 1920, the model stratosphere begins to cool, similar to the cooling found by Schwarzkopf and Ramaswamy (2008) in the GFDL CM2 model. After about 1980, the cooling in the current simulation is reinforced by additional cooling associated with ozone loss. The eruptions of Krakatau (1883), Santa Maria (1902), Agung (1963), El Chichón (1982), and Mt. Pinatubo (1991) induce a notable warming in the lower stratosphere and a cooling in the middle troposphere. The influence of these eruptions on stratospheric ozone is explored in detail in section 5.

b. Temperature trends

The zonal mean temperature trends as a function of latitude and pressure for the period 1860–1959 are shown in Fig. 7 (left). As noted previously, there is significant
stratospheric cooling. Since 1960, the simulated temperature trends are much larger (Fig. 7, middle and right) and are in agreement with previous studies. Between 1960 and 1980 the stratospheric cooling is primarily due to CO$_2$ increase (e.g., Shine et al. 2003). After 1980, the increased cooling is due to ozone depletion (e.g., Shine et al. 2003). The simulated Antarctic ozone hole has contributed to a substantial cooling in the lower stratosphere. Since the control run shows negligible trends in temperature, as in the case of ozone, the results are taken to arise from the combination of external model forcings.

Obtaining stratospheric temperature measurements suitable for trend determination is complicated by the sparseness of the data network and the change in instruments during the observational record (e.g., Randel et al. 2009). The global average observations after adjustment (Haimberger et al. 2008; Shine et al. 2008; Randel et al. 2009) are presented compared with model results in Fig. 8. The model trends are consistent across the five ensemble members. As in Austin et al. (2009), the model agrees well with observations throughout the altitude range of the measurements, except possibly for a small altitude range near 5 hPa. This difference from models has been previously noted (Shine et al. 2003), although recent adjustments to the satellite data (Shine et al. 2008), included here, have reduced the discrepancy. The remaining differences between models and observations may have been caused in part by the large vertical extents of the Stratospheric Sounding Unit (SSU) weighting functions.

5. The influence of volcanic eruptions on atmospheric chemistry and temperature

a. Volcanic aerosol amounts

The major volcanic eruptions are clearly visible in Fig. 9, which shows the globally averaged stratospheric aerosol surface area density (SAD) at 17 and 63 hPa in the model. The SAD was derived from 1-micron constructed aerosol optical depths based on observations (Stenchikov et al. 2006). From the optical depths the SAD is calculated using the relationship of Thomason and Poole (1997). In this framework, Mt. Pinatubo (1991) has a peak aerosol loading almost as large as Krakatau (1883), while Santa Maria (1902) has slightly less peak SAD than Agung (1963) and El Chichón (1982). There were also additional eruptions in the twentieth century, which had peak aerosol loadings of less than half that of Agung and will be neglected in the rest of this study. The

![Fig. 6. Annually averaged simulated global temperature anomaly (K) relative to 1860 as a function of pressure, for the period 1860 to 2005 for the ensemble mean.](image)

![Fig. 7. Mean temperature trend (K decade$^{-1}$) for the periods (left) 1860–1959, (middle) 1960–79, and (right) 1980–99 for the ensemble mean. The shaded region indicates where the temperature trends are significantly different from zero at the 95% confidence level.](image)
periods of high aerosol loadings approximately coincide in both the middle and upper stratosphere.

b. The ozone volcanic response

The response of global mean ozone to volcanic aerosol loadings is demonstrated in Fig. 10. In contrast to the volcanic aerosols, the short time scale variations in ozone computed by the model are different in the middle and lower stratosphere. At 17 hPa the model ozone amount increased following each of the major eruptions as well as the minor eruptions, although the magnitude of the ozone change was not strongly dependent on the volcanic aerosol amount. In the lower stratosphere the simulated ozone amount remained constant to within about 0.02 ppmv from 1860 to 1980 (Fig. 10, lower panel). Following the eruptions of El Chichón and Mt. Pinatubo during a period of increased chlorine loading, there were significant ozone reductions.

The volcanic effects on ozone are shown in Fig. 11 for the two years before and after each of the five major eruptions since 1860. The mean values for the two years prior to the eruptions have been subtracted from the ozone values and the annual cycle averaged over the 2-yr

Fig. 10. Globally averaged ozone mixing ratio on the 17-hPa and 63-hPa surfaces. The results plotted are relative to the monthly varying mean over the first 100 years of the simulation and are an average of the five ensemble runs. The thin vertical lines correspond to the times of major volcanic eruptions.
period has also been removed. In the middle stratosphere (Fig. 11, top), the ozone increased following each of the five eruptions. The maximum volcanic response occurred near the peak in aerosol loading, typically about one year after the eruption start, and varies between $0.82 \pm 0.03$ ppmv increase for Krakatau and $0.25 \pm 0.05$ ppmv increase for Mt. Pinatubo (95% confidence intervals based on the prevolcanic ozone variance). In the lower stratosphere (Fig. 11, middle), the ozone response was negligible except for El Chichón ($-0.06 \pm 0.01$) and Mt. Pinatubo ($-0.10 \pm 0.01$). In the latter case the peak loss occurred about two years after the eruption start. The net effect on the total ozone column (Fig. 11, bottom) agrees with previous work (e.g., Tie and Brasseur 1995): the early volcanoes led to an ozone column increase whereas, after stratospheric chlorine levels increased, volcanoes decreased the ozone column.

c. Chemical radical concentrations

The perturbation to stratospheric chemistry due to volcanic aerosols is further diagnosed by examining the ClOx and NOx amounts (Fig. 12). Results relative to the eruption start are shown in Fig. 13. To remove the large seasonal variation in both NOx and ClOx, the annual cycle for the 2-yr period prior to each eruption has been removed from all values. The results are similar at both levels, although the magnitude of the changes differs. Prior to 1965, when virtually all the atmospheric chlorine was natural, the chlorine radical concentrations increased significantly at the time of each of the eruptions, although the largest change was only about 2 ppt at 63 hPa (10 ppt at 17 hPa). However, by the time of the El Chichón eruption, background chlorine levels were twice as large, and the volcanic effect was substantially larger. By the time of the eruption of Mt. Pinatubo, chlorine levels had risen still higher, and the volcanic impact was over 7 ppt at 63 hPa (30 ppt at 17 hPa). Thus, the two later eruptions have much larger effects on chlorine radical amounts. By contrast, the impact of the eruptions on NOx amounts tended to be largest for the earliest two eruptions. The eruption of Krakatau was accompanied by a NOx reduction of about 2 ppbv at 17 hPa (0.5 ppbv at 63 hPa), similar to the change during the eruption of Santa Maria. For the later volcanoes the NOx impact was similar to that for Santa Maria, but NOx recovered faster.

The results obtained are consistent with those expected due to heterogeneous reactions on sulfate aerosols (Hofmann and Solomon 1989; Granier and Brasseur 1992; Koike et al. 1994), the most important of which are as follows:

$$N_2O_5 + H_2O \rightarrow 2HNO_3$$ \hspace{1cm} (R1)
$$ClONO_2 + HCl \rightarrow Cl_2 + HNO_3.$$ \hspace{1cm} (R2)
Reaction R1 takes nitrogen from active forms (NOx) and converts it to the reservoir species HNO3. Reaction R2 takes chlorine from the reservoirs HCl and ClONO2 and converts it to Cl2, which generates Cl by photolysis. Thus, the general effect of the volcanic aerosols is to reduce nitrogen radicals and increase chlorine radicals. These two effects have opposite impacts on ozone. The net effect depends on altitude and the overall amount of chlorine loading, as noted previously by Tie and Brasseur (1995) using a somewhat simpler model. In the results shown here, ozone increased significantly in the middle stratosphere for the three early volcanoes because the NOx effect was larger than the chlorine effect. For the last two volcanoes, the ozone increase in the middle stratosphere was smaller because of the increasing impact of chlorine. In the lower stratosphere, where the NOx effect on ozone is small, the chlorine and NOx effects were approximately in balance and ozone was unaffected by the first three major volcanoes. However, for the later volcanoes, El Chichón and Pinatubo, the chlorine effect was larger than the NOx effect and ozone decreased. Overall, the first three volcanoes increased column ozone in the model (dominated by middle stratospheric processes), and the last two decreased column ozone (dominated by lower-stratospheric effects).

The response of radical concentrations to aerosol amount is shown in Fig. 14 as a sequence of scatterplots for the two pressure levels considered. The results illustrate the general decrease in NOx and increase in ClOx with increasing aerosol amounts but with the sensitivity to aerosols dependent on each particular eruption. The chlorine response to aerosols is particularly variable because of the substantial change in background concentration (a factor of 4 at the upper level and 6 at the lower level) for the five volcanoes. The results illustrate, for example, that for Santa Maria and Krakatau the chlorine perturbation is small (due to low background chlorine levels) and that in the model this allows reaction R1 to dominate in the heterogeneous scheme. This has resulted in a larger NOx response during those eruptions for a given aerosol amount (Fig. 14). However, although these variations can be generally understood, there is often not a direct relationship between aerosol and ClO because of the different time scales that operate and control these processes in the model, including the background ozone and chemical seasonal cycles.

**Fig. 12.** Globally averaged ClO and NOx (∑NO + NO2) mixing ratios simulated on the 17-hPa and 63-hPa surfaces. The results plotted are 12-month running means and are averaged over the five ensemble runs. The thin vertical lines correspond to the times of major volcanic eruptions.
d. Chemical reservoir concentrations

Figure 15 shows the key reservoir species that are affected by the volcanic aerosol. The results have been plotted after removing the annual cycle and in the case of the chlorine species after removing, from the whole period, the linear trend calculated for the 24 months prior to each eruption. A rapid increase in HNO$_3$ following the eruptions is apparent and reflects the decrease in NO$_x$ seen in Fig. 13. The eruptions also influence chlorine partitioning with a decrease in reservoir species corresponding to the increase in ClO in Fig. 13. It should be noted that the results at the lower level (63 hPa) are sensitive to the details of the procedure for removal of the annual cycle and, even over a 2-yr period, the Cl$_y$ trend was significant, particularly for the pre-Pinatubo period. During the eruption itself, the simulated change in reservoir chlorine is only about 5%, with an increase in HOCl to balance the chlorine budget. These caveats aside, the reductions in reservoir chlorine for the high chlorine loading eruptions (Mt. Pinatubo and El Chichón) are notable.

e. Temperature

The volcanic impact on stratospheric temperature since 1960 is clearly seen in Fig. 16, which shows the global average values weighted in the vertical by the Microwave Sounding Unit (MSU) channel 4 weighting function, which peaks in the lower stratosphere. Expressed as an anomaly from the 1980 mean values, the model is in good agreement with MSU and Radiosonde Innovation Composite Homogenization (RICH) observations throughout the period shown. The results are similar to those found in coupled chemistry–climate models (Austin and Wilson 2006; Eyring et al. 2006; Austin et al. 2009) as well as Santer et al. (2006) and follow the general behavior of a rapid warming during the eruptions followed by a rapid cooling afterward with stabilization of temperature for a period, as discussed also by Ramaswamy et al. (2006). The overall cooling trend is a well-established effect due to increases in CO$_2$ (e.g., Shine et al. 2003). The size of the volcanic temperature signal is in good agreement with observations during the eruptions of Agung, El Chichón, and Mt. Pinatubo, of about 0.6 K in the annual average.
The temperature changes for the largest five eruptions since 1860 are shown in Fig. 17, concentrating on the 4-yr period centered on the start of each eruption. In the middle stratosphere (upper panel), the correlation with ozone (Fig. 11) is high, indicating that ozone change (induced by chemistry) is likely to be the largest contributor. In the lower stratosphere, the temperature is seen to increase consistently for each eruption despite the absence of an ozone increase. Indeed, for the latest two eruptions, the ozone amount decreased (Fig. 11), but this was not sufficient to overcome the direct heating by aerosols. An example of the different processes controlling temperature at the two levels is illustrated by the Krakatau results. The timing indicated is relative to the first significant increase in aerosols (January 1883), which was nevertheless small compared with the major eruption in August 1883. Whereas nitrogen chemistry can be affected by small amounts of aerosols (e.g., Krakatoa results in Figs. 13 and 15), temperature effects require ozone changes (Fig. 11, top) or substantial aerosol changes in the lower stratosphere (Fig. 17, lower).

6. Meridional transport and model age

A measure of atmospheric transport is given by the stratospheric age, which is essentially the mean time taken for air parcels to reach a given stratospheric location (e.g., Hall and Plumb 1994). Stratospheric age has acquired particular significance because of its connection with the Brewer–Dobson circulation (Neu and Plumb 1999; Austin and Li 2006), which in turn has been shown to accelerate in future simulations (Butchart and Scaife 2001; Butchart et al. 2006; SPARC CCMVal 2010, ch. 4; Butchart et al. 2010). The increased circulation is believed to result primarily from the increase in greenhouse gas (GHG) concentrations (Butchart et al. 2006; Garcia et al. 2007), although ozone impacts are also considered to play a role (Li et al. 2008). In Fig. 18 (left and middle) the age diagnostic from the model has relatively minor fluctuations from 1860 until about 1975. Thereafter the stratospheric age decreased until the end of the simulation. A consistent picture occurred at both levels considered. Since the GHG increases began to...
impact stratospheric temperature from about 1940 or perhaps earlier (Fig. 6), the implication is that ozone trends are playing an important role in the current simulations, consistent with the Li et al. (2008) study. The lack of a significant trend in the age of air prior to 1960 is also consistent with the lack of a trend in tropical lower-stratospheric ozone (Fig. 4) where photochemistry is too slow to induce changes. However, as the age of air starts to decline, more steeply, ozone in the lower stratosphere also decreases (Fig. 5, lower right). Thus, the chemical ozone change in the middle and upper stratosphere enhances the circulation, which decreases ozone in the tropical lower stratosphere. Overall, the model age is biased low compared with other models and with observations, which is probably due to a meridional circulation that is too strong (e.g., SPARC CCMVal 2010, ch. 5). In the CCMVal report, model age decreased in all models (SPARC CCMVal 2010, ch. 4; Butchart et al. 2010) but, since those simulations did not begin until 1960, the current work extends considerably the period of understanding for this quantity.

There is no clear volcanic signal in the age of air. Although ozone amounts have a small influence on the age, the effective multiyear average of the age diagnostic reduces the sensitivity of this quantity to a relatively temporary phenomenon such as a volcanic eruption.

The tropical transporting velocity, as measured by the transformed Eulerian mean $w^*$ at 70 hPa (Fig. 18, right), show the same changes as the age, but of opposite sign (see also, e.g., Austin and Li 2006). Thus, from about 1975 onward, $w^*$ increased significantly, leading to increased upward transport of ozone. Similarly, the meridional circulation is enhanced in midlatitudes after 1975, leading in particular to the increase in midlatitude ozone shown in Fig. 1. Comparison with other models shows that $w^*$ in AMTRAC3 was higher than the multimodel mean of Butchart et al. (2010), which likely explains the overestimate of column ozone in middle latitudes compared with observations (Fig. 2).

7. Conclusions

We have presented results from a set of experiments using the GFDL CM3 model in which the climate is simulated from the preindustrial to the present day using the observed external forcings and coupled
stratospheric and tropospheric chemistry. Among climate models, CM3 would be considered a “high top” model, while at the same time including a coupled ocean to allow realistic simulation of the future climate. Regarding models with a coupled ocean, it is unique in representing atmospheric chemistry seamlessly (i.e., using the same set of reactions with no artificial boundaries) from the surface to the model top layer centered at 0.02 hPa. A simpler version of the model—without the coupled ocean and with the tropospheric organic chemistry reduced to methane chemistry and its derivatives—was used in the intermodel comparison described in the CCMVal report (SPARC CCMVal 2010) and in the 2010 WMO–United Nations Environment Programme (UNEP) ozone report (WMO 2011, chapter 3). Both dynamics and chemistry have been extensively validated in the former report and overall the model was found to perform as well as or better than most models.

FIG. 18. Globally averaged stratospheric age on the (left) 17-hPa and (middle) 63-hPa surfaces and (right) residual vertical velocity $\vec{w}$ at 70 hPa (averaged from $10^\circ$S to $10^\circ$N). Annual means averaged over the five ensemble members.
In this work, we have presented the results from five ensemble members integrated for the period 1860 to 2005. The results indicate a fairly stable atmosphere from 1860 to 1960 with only small trends, primarily a slight cooling in the stratosphere due to GHG increases. In response to the stratospheric cooling, ozone concentrations increased in the upper stratosphere. In the lower stratosphere, ozone decreased primarily due to upward motion associated with the changing Brewer–Dobson circulation, diagnosed by decreasing stratospheric age and increasing transformed Eulerian mean vertical velocity \( \overrightarrow{w} \). The lack of large changes in \( O_3 \) temperature, and age prior to 1960 may well have been expected on the basis of previous simulations. However, null trends simulated here might be considered more reliable because of the incorporation of a wider range of processes. Unfortunately, few stratospheric measurements are available prior to 1960 to support the results of our model simulations. After 1960, the stratospheric trends accelerated due to well-established processes: stratospheric cooling from GHG increases and stratospheric ozone loss from CFC increases. Volcanic eruptions were found to have a significant effect on the results, especially for the five major eruptions: Krakatau (1883), Santa María (1902), Agung (1963), El Chichón (1982), and Mt. Pinatubo (1991). These led to a decrease in NO\(_x\) amounts in the middle and lower stratosphere and a corresponding increase in the reservoir HNO\(_3\). Simultaneously, ClO\(_x\) amounts increased. Thus the net effect on ozone changed substantially, consistent with previous work (Tie and Brasseur 1995) and, in addition, varied with altitude. In the middle stratosphere, ozone increased for all five eruptions but, in the lower stratosphere, the ozone amount was relatively unaffected except for the last two eruptions, by which time chlorine concentrations had risen substantially above background levels. The net effect on the ozone column therefore depends on the balance of the different catalytic cycles operating at different altitudes. For natural levels of chlorine, volcanoes tend to increase the ozone column while, for high anthropogenic levels of chlorine, volcanic eruptions decrease ozone columns. By contrast, a consistent signal was seen in the simulated temperature response. Although the quantitative details varied depending on the eruption, a significant temperature increase was seen during each one, consistent with observations when these existed. It follows that, while halogen amounts remain high, both temperature and ozone in the lower stratosphere will remain sensitive to volcanic eruptions.

Overall, simulated temperature and ozone trends are in good agreement with observed trends providing confidence in future projections of the model. With that aim, the simulations described in this work have now been continued until the end of the twenty-first century, following the four Representative Concentration Pathways (RCPs) used in CMIP5 simulations and may be used to investigate the timing of ozone recovery in the context of previous calculations (WMO 2011, ch. 3).

Acknowledgments. We thank Arlene Fiore and Vaishali Naik for their helpful comments on an earlier version of this paper. Three anonymous reviewers are also thanked for providing additional useful comments.

REFERENCES


Garcia, R. R., D. Marsh, D. Kinnison, B. A. Boville, and F. Sassi, 2007: Simulations of secular trends in the middle atmosphere...