

The Response of Ozone and Nitrogen Dioxide to the Eruption of Mt. Pinatubo at Southern and Northern Midlatitudes

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ABSTRACT

Observations have shown that the mass of nitrogen dioxide decreased at both southern and northern midlatitudes in the year following the eruption of Mt. Pinatubo, indicating that the volcanic aerosol had enhanced nitrogen dioxide depletion via heterogeneous chemistry. In contrast, the observed ozone response showed a northern midlatitude decrease and a small southern midlatitude increase. Previous simulations that included an enhancement of heterogeneous chemistry by the volcanic aerosol but no other effect of this aerosol produce ozone decreases in both hemispheres, contrary to observations. The authors' simulations show that the heating due to the volcanic aerosol enhanced both the tropical upwelling and Southern Hemisphere extratropical downwelling. This enhanced extratropical downwelling, combined with the time of the eruption relative to the phase of the Brewer-Dobson circulation, increased Southern Hemisphere ozone via advection, counteracting the ozone depletion due to heterogeneous chemistry on the Pinatubo aerosol.

1. Introduction

The 15 June 1991 volcanic eruption of Mount Pinatubo injected about 20 Tg of sulfur dioxide (SO₂) into the stratosphere (Bluth et al. 1992), up to an altitude of about 30 km (McCormick and Veiga 1992). The SO₂ transformed into about 30 Tg of sulfate aerosol (McCormick et al. 1995), increasing the stratospheric aerosol loading by orders of magnitude over background. This perturbation decreased with time but a signature of the eruption was detectable in the atmosphere for about 5 yr (Robock et al. 2000).

Aerosol from Mt. Pinatubo reached both the Northern Hemisphere (NH) and Southern Hemisphere (SH), changing the stratospheric chemistry and dynamics. The volcanic sulfate provided additional surfaces for the into HNO₃, resulting in the depletion of nitrogen dioxide (NO₂) (e.g., Fahey et al. 1993; Tie and Brasseur 1995; Portmann et al. 1996; Solomon et al. 1996; Tabazadeh et al. 2002). This heterogeneous reaction generally leads to an increase in ClO and OH, and subsequently to an increase of ozone depletion (e.g., Tie and Brasseur 1995). Additionally, heating by the volcanic aerosol changed

heterogeneous chemical reaction that transform N₂O₅

the radiative balance and dynamics of the atmosphere (e.g., Robock and Mao 1992; Graf et al. 1993; Stenchikov et al. 2002; Robock et al. 2007; Karpechko et al. 2010), intensifying the upwelling in the tropics and downwelling in the extratropics (Kinne et al. 1992; Pitari and Mancini 2002; Aquila et al. 2012).

The observed depletion of stratospheric NO₂ in both hemispheres during the years following the eruption (Johnston et al. 1992; Van Roozendael et al. 1997; Danilin et al. 1999) provided evidence that the volcanic aerosol had enhanced the heterogeneous chemistry at all latitudes. Surprisingly, the observed ozone response

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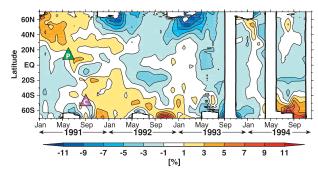


FIG. 1. Zonal mean of the ozone column anomaly (%) from TOMS observations. The baseline is the 1979–90 column ozone mean from TOMS. Effects due to the seasonal cycle, QBO, ENSO, solar cycle, and changing chlorine and bromine have been removed by using a seasonally varying regression analysis (Stolarski et al. 1991). The green and purple triangles mark the locations and times of the eruption of Mt. Pinatubo and Cerro Hudson, respectively.

to the volcanic perturbation was different between the NH and SH. While column ozone generally decreased in the NH, an increase of the ozone column was detected at southern mid- and high latitudes during the year following the eruption (Zerefos et al. 1994; Randel et al. 1995; Randel and Wu 1996). Figure 1 shows the anomaly of the total ozone column due to the eruption of Mt. Pinatubo as derived from Total Ozone Mapping Spectrometer (TOMS) observations.

Several model studies attributed the observed NH ozone depletion to the enhancement of heterogeneous chemistry because of the volcanic aerosols (e.g., Tie and Brasseur 1995) but could not explain why the same heterogeneous chemistry did not affect the SH ozone concentration (Stolarski et al. 2006). Some studies suggested explanations other than a chemistry perturbation for the asymmetry of the ozone response. Randel and Wu (1996) showed that the quasi-biennial oscillation (QBO) increased ozone in the extratropical SH during the 1991/92 winter, but the effect is not large enough to explain the observed increase. Fleming et al. (2007) and Telford et al. (2009) successfully simulated the ozone behavior using observed meteorological fields. These studies attribute the absence of ozone depletion to interannual dynamic variability, which masked the Pinatubo aerosol chemical effect in the SH. However, these studies cannot distinguish between natural interannual variability and circulation changes forced by the volcanic perturbation. Poberaj et al. (2011) performed a multiple linear regression analysis to the Chemical and Dynamical Influences on Decadal Ozone Change (CANDIDOZ), stating that volcanically induced chemical ozone depletion was overcompensated by the QBO and by a pronounced Eliassen–Palm (EP) flux anomaly.

Here, we use a free-running global chemistry-climate model (section 2) to separate the photochemical and dynamical contributions to the ozone and NO₂ anomalies induced by the volcanic perturbation alone (section 3).

2. Model and simulation

All simulations are performed with the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM). The general circulation model is the Goddard Earth Observing System, version 5 (GEOS-5; Rienecker et al. 2008), a system of component models integrated using the Earth System Modeling Framework (ESMF). For these simulations, GEOS-5 is coupled to the Georgia Institute of Technology-Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) module (Colarco et al. 2010) and a stratospheric chemistry module (Pawson et al. 2008). The resolution is 2.0° latitude $\times 2.5^{\circ}$ longitude with 72 vertical layers from surface to 0.01 hPa (~80 km). This version of GEOSCCM does not simulate the QBO. The model is forced with observed sea surface temperatures and sea ice concentrations (Reynolds et al. 2002). Aquila et al. (2012) includes a more detailed description of the model and an evaluation of the simulation of the Mt. Pinatubo cloud's transport within GEOSCCM.

GOCART computes the transformation of SO_2 into sulfate aerosol. The simulations shown here use a prescribed aerosol surface area density (SAD) from the Stratospheric Aerosol Measurement II (SAM II), Stratospheric Aerosol and Gas Experiment (SAGE), and SAGE-II data (Eyring et al. 2008) for heterogeneous chemistry. GEOSCCM also includes an option for calculating the aerosol SAD online using the simulated mass of sulfate aerosol and the relative humidity.

We simulate the Mt. Pinatubo eruption by injecting 20 Tg of SO_2 between 16 and 18 km in the volcano's model grid box on 15 June 1991. No other aerosol sources are included in the simulation. We performed four model experiments, each composed of 10 simulations with different sets of initial conditions typical of the year 2000, when the values of effective equivalent chlorine were similar to the early 1990s (Clerbaux and Cunnold 2007). The results shown here are ensemble averages.

The first experiment (experiment REF) is a control ensemble that does not include any volcanic perturbation. This experiment uses prescribed aerosol SAD from SAM II and SAGE observations from 1979 for the heterogeneous chemistry—a period when the stratospheric aerosol layer is relatively unperturbed (Thomason et al. 1997). The heterogeneous chemistry in experiment REF is representative of the chemistry that takes place on the stratospheric background aerosol in absence of major volcanic eruptions.

Experiment	Radiatively interactive aerosol	Year(s) for sulfate area density	Perturbation to the chemistry	Perturbation to the dynamics
REF	No	1979	No	No
CHEM	No	1991–95	Yes	No
DYN	Yes	1979	No	Yes
FULL	Yes	1991–95	Yes	Yes

The second experiment (experiment DYN) includes radiatively interactive aerosol; that is, the aerosol can modify the simulated meteorology. As in experiment REF, experiment DYN uses aerosol SAD from 1979. Hence, this experiment perturbs the stratospheric dynamics but does not perturb the stratospheric chemistry.

The third experiment (experiment CHEM) does not include radiatively interactive aerosol and uses aerosol SAD from SAGE-II data appropriate for the simulated year; that is, the heterogeneous chemistry takes place on an aerosol layer perturbed by the Mt. Pinatubo eruption. This experiment includes a Pinatubo perturbation of the stratospheric chemistry but no perturbation of the stratospheric dynamics.

The fourth experiment (experiment FULL) includes radiatively interactive aerosols and aerosol SAD for the Pinatubo period. This experiment includes the volcanic perturbation to both the stratospheric dynamics and chemistry.

The comparison of the experiment FULL with the control experiment REF identifies the complete perturbation of ozone and NO_2 due to Mt. Pinatubo. Comparisons of experiments DYN and CHEM with REF isolate the anomalies of ozone and NO_2 due to the effects of the eruption on the atmospheric dynamics and heterogeneous chemistry, respectively. Table 1 summarizes the performed model experiments.

3. Results

Figure 2 shows the observed deviations of the stratospheric NO₂ column (black line) over Lauder, New Zealand, from a visible and ultraviolet (UV/Vis) spectrophotometer (Johnston and McKenzie 1984). The baseline for the calculation of the anomalies is the observed monthly means over the years 1997–2003, which is a period free of major volcanic eruptions. The plot shows the simulated zonal mean anomalies at 45°S of the stratospheric NO₂ column, calculated as the difference between the experiments FULL (red), CHEM (blue), and DYN (green) and the experiment REF. The DYN experiment including only the perturbation to the dynamics does not show any significant perturbation of NO₂. Both the CHEM (blue) and FULL (red) experiments

produce a decrease of stratospheric NO_2 in good agreement with the observations. The perturbation of NO_2 is dominated by the volcanic effect on the stratospheric chemistry, which is relatively symmetric between the hemispheres (Fig. 3). Heterogeneous reactions on sulfate aerosols explain the repartitioning between NO_x ($NO + NO_2$) and the reactive nitrogen reservoir NO_y ($NO + NO_2 + NO_3 + 2N_2O_5 + HNO_3 + HO_2NO_2 + CIONO_2 + BrONO_2$) that is observed after the eruption of Mt. Pinatubo at northern and southern midlatitudes (Fahey et al. 1993 and Koike et al. 1994).

The simulated NO₂ depletion is smaller in magnitude and peaks 1 month later than the observations. Koike et al. (1994) presented results from the Lawrence Livermore National Laboratory (LLNL) (Wuebbles et al. 1991) and Atmospheric and Environmental Research (AER) (Rodriguez et al. 1991, 1994) models, which also used aerosol fields from SAGE-II and also underestimate the NO₂ depletion. We performed experiments using the online calculation of the aerosol SAD (not shown), and found a larger and earlier depletion of NO₂ in better agreement with the observations.

GEOSCCM produces a reasonably good simulation of the vertical distribution of the Pinatubo aerosol in the posteruption period (Aquila et al. 2012). Figure 4 (left panel) displays the SAGE-II 525-nm zonal mean

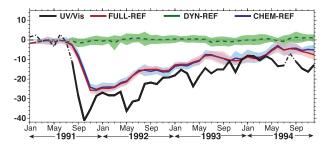


FIG. 2. Stratospheric NO_2 column anomalies (%) at 45°S. The black line shows the anomalies at Lauder, New Zealand (45.5°S), calculated from UV/Vis observations. The baseline is a 1997–2003 monthly climatology calculated from these Lauder observations. The solid black line is significantly different from zero at 1σ level. The simulated 45°S anomalies for FULL – REF (red), DYN – REF (green), and CHEM – REF (blue) are also shown. Shading shows the standard deviation of each ensemble. The solid red, green, and blue lines are significantly different from zero at the 95% confidence level, while the broken lines are not significant.

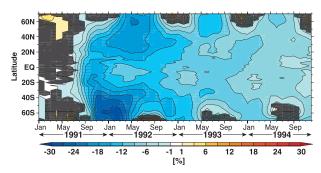


FIG. 3. Zonal mean stratospheric NO_2 column anomalies in experiment FULL with respect to experiment REF. The gray shaded areas are not significantly different from zero at 95% confidence level.

extinction coefficients for the 1991 September-November (SON) period. Features to note include the large extinctions in the tropics and the SH midlatitude profile with double peaks at about 40 and 200 hPa. The online model simulation produces a somewhat different SAD vertical profile in the SH midlatitudes, with peaks at 20 and 200 hPa (Fig. 4, right panel, blue line). The upper peak is slightly higher than is observed in SAGE-II. The analysis of SAGE-II data by Kinnison et al. (1994) also revealed this two-peak SH vertical structure. The prescribed monthly mean profile from SAGE-II and used herein (red line; Eyring et al. 2008) only exhibits a single peak in aerosol SAD at about 70 hPa. The reasons for the discrepancy between this SAGE-II single peak profile shape (Fig. 4, right panel, red line) and the SAGE-II extinction observations (left panel) possibly results from the coarse averaging resolution used in deriving the prescribed values indicated by the red line.

The volcanic perturbation to the dynamics modifies the ozone concentrations. Figure 5 compares the 30°-60°N and 30°-60°S zonal total ozone column anomalies from TOMS data (Herman et al. 1996; McPeters et al. 1996) with the GEOSCCM simulations. The observed anomalies (black) are calculated as the deviation from the ozone column mean over 1979-90 after removing the effects of the seasonal cycle, QBO, El Niño-Southern Oscillation (ENSO), solar cycle, and changing stratospheric chlorine and bromine. The data show a negative NH anomaly (Fig. 5, top panel) from December 1991 that is significantly different from zero (all solid lines in the figure are significantly different from zero) for about 2 yr after the eruption. The experiment CHEM also shows a negative NH anomaly, while the experiment DYN has no significant ozone anomaly. The negative anomaly present in the experiment FULL is not significantly different from the anomaly of CHEM at the 95% confidence level. Hence, the simulated NH anomaly is

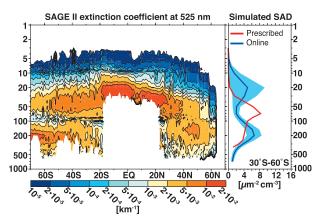


FIG. 4. (left) Zonal mean vertical profile of the SAGE-II aerosol extinction coefficient at 525 nm. (right) Zonal mean vertical profiles at 30°-60°S of the prescribed aerosol surface area density adopted for our simulations (red line, Eyring et al. 2008) and the online simulation from GEOSCCM (blue line). The blue shaded areas show the standard deviation of the 10-member ensemble used for the online calculation of the SAD. Both panels are September, October, and November 1991 averages.

primarily caused by the Pinatubo-forced heterogeneous chemical effect.

In the SH, TOMS recorded a positive anomaly for about 1 yr after the eruption (Fig. 5, bottom panel). This SH positive anomaly is simulated by both the DYN and FULL experiments. By June 1992 the DYN positive ozone anomaly is gone, while CHEM produces a significant negative perturbation starting about March 1992. This CHEM ozone response is similar in both hemispheres. The ozone anomaly in FULL is essentially the sum of the DYN and CHEM anomalies. The ozone anomaly in FULL is significantly different from CHEM at 95% level during two periods: October 1991–January 1992 and March–July 1992 (gray shaded areas of Fig. 5). These differences demonstrate that the ozone anomaly from the Pinatubo dynamical response is larger or comparable to the chemical perturbation for about 1 yr posteruption.

The positive ozone anomaly at SH midlatitudes in experiments DYN and FULL is induced by the dynamical perturbation. The absorption of largely longwave radiation by the volcanic aerosol leads to enhanced upwelling in the tropical lower stratosphere. By continuity, this enhanced tropical upwelling enhances the downwelling in the extratropics (Aquila et al. 2012). The ozone anomaly due to advection by this enhanced circulation is negative in the tropics and positive at midlatitudes (Fig. 6). This volcanic perturbation of the dynamics lasts as long as there is a strong gradient of the aerosol concentration between the tropics and midlatitudes. This mean circulation perturbation by Pinatubo was created by the volcanic aerosol radiative forcing (Aquila et al. 2012).

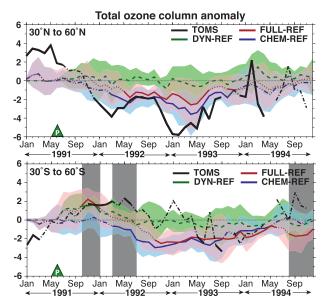


FIG. 5. Zonal mean total ozone column anomalies for (top) 30° – 60° N and (bottom) 30° – 60° S. The black line shows TOMS anomalies (%) after removing the effects of the seasonal cycle, QBO, ENSO, solar cycle, and changing chlorine and bromine (see Fig. 1 for details). The solid black lines are significantly different from zero at 1σ level. The simulated anomalies for FULL – REF (red), CHEM – REF (blue), and DYN – REF (green) are also shown. The color shaded areas show the standard deviation of each ensemble. Solid red, blue, and green lines are significantly different from zero at the 95% confidence level. The gray shaded times show where the FULL and CHEM simulations are significantly different from one another at the 95% confidence level. The green triangle marks the month of the Pinatubo eruption.

The anomalous ozone distributions (Fig. 7) are shown in the immediate posteruption period [June–August (JJA) 1991, left panel] and the late posteruption period (SON 1992, right panel) by differencing the FULL and REF simulations. In JJA 1991 the perturbation is

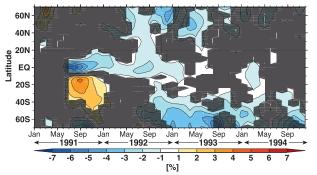


FIG. 6. Zonal mean anomalies of total ozone column between the FULL and REF simulations. The gray shaded areas are not significantly different from zero at 95% confidence level.

completely dominated by the dynamics response, which we depict with the streamlines of the residual circulation anomaly. The increased tropical upwelling lifts air with lower ozone concentration, creating a negative equatorial anomaly centered at 20 hPa or 24 km. At the same time, the downwelling south of the equator creates an SH positive ozone anomaly. This positive anomaly is located in the SH because of the phase of the Brewer–Dobson circulation at the time of the eruption, which is directed toward the winter (SH) hemisphere. We performed an experiment initiating a Pinatubo-like eruption on 15 January 1991, as described in Aquila et al. (2012). There, the positive ozone anomaly appeared in the NH (not shown)—compatible with the different phase of the Brewer–Dobson circulation.

In SON 1992 (Fig. 7, right panel) the simulated ozone anomaly is mainly due to the perturbation to the chemistry. In the middle stratosphere the ozone concentration is increased, because the presence of the volcanic aerosol suppresses the NO_x cycle that destroys ozone. In the lower stratosphere the presence of the

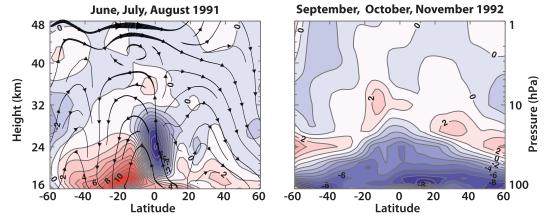


FIG. 7. Altitude vs latitude distributions of the zonal mean ozone anomalies (%) in (left) immediate post-Pinatubo period of JJA 1991 and (right) late post-Pinatubo period of SON 1992. The anomalies are calculated by differencing the FULL and REF simulations. The streamlines in the left panel show the residual circulation anomalies.

volcanic aerosol enhances the HO_x and ClO_x cycles and causes ozone depletion, as was also described in Tie and Brasseur (1995).

4. Conclusions

The lack of observed ozone depletion due to the eruption of Mt. Pinatubo in the Southern Hemisphere, in spite of the clear depletion of NO2, has been an outstanding puzzle for many years (Douglass and Fioletov 2011). We have shown that the perturbation of the stratospheric dynamics by the eruption of Mt. Pinatubo is responsible for the lack of an observed ozone decrease in the SH during the first year after the eruption. In the SH the dynamical response to the volcanic perturbation dominates the changes in ozone column during the first 6 months after the eruption and fades away starting about January 1992. The chemical response, instead, produces significant changes in the SH starting about 1 yr after the eruption. The perturbations to the chemistry and to the dynamics have an additive effect, resulting in the lack of ozone depletion at southern midlatitudes in the year following the eruption.

The NO_2 anomaly is completely driven by the volcanic-induced chemical perturbation and is insensitive to the dynamic perturbation. The reason is the much shorter time scale of the heterogeneous chemistry for depleting NO_2 compared to the time scale for depleting ozone, together with the weak NO_2 vertical gradient, such that changes of vertical advection do not induce large perturbations.

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REFERENCES

- Aquila, V., L. D. Oman, R. S. Stolarski, P. R. Colarco, and P. A. Newman, 2012: Dispersion of the volcanic sulfate cloud from a Mount Pinatubo-like eruption. *J. Geophys. Res.*, 117, D06216, doi:10.1029/2011JD016968.
- Bluth, G. J. S., S. D. Doiron, C. C. Schnetzler, A. J. Krueger, and L. S. Walter, 1992: Global tracking of the SO₂ cloud from the

- June, 1991 Mount Pinatubo eruptions. *Geophys. Res. Lett.*, **19**, 151–154, doi:10.1029/91GL02792.
- Clerbaux, C., and Coauthors, 2007: Long-lived compounds. Scientific assessment of ozone depletion: 2006, World Meteorological Organization Global Ozone Research and Monitoring Project Rep. 50, 1–64.
- Colarco, P., A. Da Silva, M. Chin, and T. Diehl, 2010: Online simulations of global aerosol distributions in the NASA GEOS-4 model and comparisons to satellite and groundbased aerosol optical depth. *J. Geophys. Res.*, 115, D14207, doi:10.1029/2009JD012820.
- Danilin, M. Y., and Coauthors, 1999: Nitrogen species in the post-Pinatubo stratosphere: Model analysis utilizing UARS measurements. J. Geophys. Res., 104 (D7), 8247–8262.
- Douglass, A., and Coauthors, 2011: Stratospheric ozone and surface ultraviolet radiation. Scientific assessment of ozone depletion: 2010, World Meteorological Organization Global Ozone Research and Monitoring Project Rep. 52, 1–76.
- Eyring, V., and Coauthors, 2008: Overview of the new CCMVal reference and sensitivity simulations in support of upcoming ozone and climate assessments and the planned SPARC CCMVal Report. SPARC Newsletter, No. 30, World Climate Research Program, Geneva, Switzerland, 20–26.
- Fahey, D. W., and Coauthors, 1993: In situ measurements constraining the role of sulphate aerosols in mid-latitude ozone depletion. *Nature*, 363, 509–514, doi:10.1038/363509a0.
- Fleming, E. L., C. H. Jackman, D. K. Weisenstein, and M. K. W. Ko, 2007: The impact of interannual variability on multi-decadal total ozone simulations. *J. Geophys. Res.*, 112, D10310, doi:10.1029/2006JD007953.
- Graf, H.-F., I. Kirchner, A. Robock, and I. Schult, 1993: Pinatubo eruption winter climate effects: Model versus observations. *Climate Dyn.*, **9**, 81–93, doi:10.1007/BF00210011.
- Herman, J. R., and Coauthors, 1996: Meteor-3 Total Ozone Mapping Spectrometer (TOMS) data products user's guide. NASA Reference Publ. 1393, 55 pp.
- Johnston, P. V., and R. L. McKenzie, 1984: Long-path absorptionmeasurements of tropospheric NO₂ in rural New Zealand. *Geophys. Res. Lett.*, 11, 69–72.
- —, —, J. G. Keys, and W. A. Matthews, 1992: Observations of deplete stratospheric NO₂ following the Pinatubo volcanic eruption. *Geophys. Res. Lett.*, 19, 211–213.
- Karpechko, A. Y., N. P. Gillett, M. Dall'Amico, and L. J. Gray, 2010: Southern Hemisphere atmospheric circulation response to the El Chichón and Pinatubo eruptions in coupled climate models. *Quart. J. Roy. Meteor. Soc.*, 136, 1813–1822, doi:10.1002/qj.683.
- Kinne, S., O. B. Toon, and M. J. Prather, 1992: Buffering of stratospheric circulation by changing amounts of tropical ozone—A Pinatubo study. *Geophys. Res. Lett.*, 19, 1927–1930.
- Kinnison, D. E., K. E. Grant, P. S. Connell, D. A. Rotman, and J. D. Wuebbles, 1994: The chemical and radiative effects of the Mount Pinatubo eruption. J. Geophys. Res., 99 (D12), 25 705–25 731.
- Koike, M., N. B. Jones, W. A. Matthews, P. V. Johnston, R. L. McKenzie, D. Kinnison, and J. Rodriguez, 1994: Impact of Pinatubo aerosols on the partitioning between NO₂ and HNO₃. *Geophys. Res. Lett.*, **21**, 597–600, doi:10.1029/94GL00303.
- McCormick, M. P., and R. E. Veiga, 1992: SAGE-II measurements of early Pinatubo aerosols. *Geophys. Res. Lett.*, **19**, 155–158, doi:10.1029/91GL02790.
- —, L. W. Thomason, and C. R. Trepte, 1995: Atmospheric effects of the Mt Pinatubo eruption. *Nature*, **373**, 399–404, doi:10.1038/373399a0.

- McPeters, R. D., and Coauthors, 1996: Nimbus-7 Total Ozone Mapping Spectrometer (TOMS) data products user's guide. NASA Reference Publ. 1384, 67 pp.
- Pawson, S., R. S. Stolarski, A. R. Douglass, P. A. Newman, J. E. Nielsen, S. M. Frith, and M. L. Gupta, 2008: Goddard Earth Observing System chemistry-climate model simulations of stratospheric ozone-temperature coupling between 1950 and 2005. J. Geophys. Res., 113, D12103, doi:10.1029/2007JD009511.
- Pitari, G., and E. Mancini, 2002: Short-term climatic impact of the 1991 volcanic eruption of Mt. Pinatubo and effects on atmospheric tracers. *Nat. Hazards Earth Syst. Sci.*, 2, 91–108, doi:10.5194/nhess-2-91-2002.
- Poberaj, C. S., J. Staehelin, and D. Brunner, 2011: Missing stratospheric ozone decrease at Southern Hemisphere middle latitudes after Mt. Pinatubo: A dynamical perspective. *J. Atmos. Sci.*, 68, 1922–1945.
- Portmann, R. W., S. Solomon, R. R. Garcia, L. W. Thomason, L. R. Poole, and M. P. McCormick, 1996: Role of aerosol variations in anthropogenic ozone depletion in the polar regions. *J. Geophys. Res.*, **101** (D17), 22 991–23 006.
- Randel, W. J., and F. Wu, 1996: Isolation of the ozone QBO in SAGE II data by singular-value decomposition. *J. Atmos. Sci.*, **53**, 2546–2559.
- —, —, J. M. Russell III, J. W. Waters, and L. Froidevaux, 1995: Ozone and temperature changes in the stratosphere following the eruption of Mount Pinatubo. *J. Geophys. Res.*, **100** (D8), 16 753–16 764.
- Reynolds, R. W., N. A. Rayner, T. M. Smith, D. S. Stokes, and W. Wang, 2002: An improved in situ and satellite SST analysis for climate. J. Climate, 15, 1609–1625.
- Rienecker, M. M., and Coauthors, 2008: The GEOS-5 Data Assimilation System—Documentation of versions 5.0.1, 5.1.0, and 5.2.0. NASA Tech. Rep. Series on Global Modeling and Data Assimilation NASA/TM-2008-104606, Vol. 27, 101 pp.
- Robock, A., 2000: Volcanic eruptions and climate. *Rev. Geophys.*, **38**, 191–219.
- —, and J. Mao, 1992: Winter warming from large volcanic eruptions. Geophys. Res. Lett., 19, 2405–2408, doi:10.1029/92GL02627.
- —, T. Adams, M. Moore, L. Oman, and G. Stenchikov, 2007: Southern Hemisphere atmospheric circulation effects of the 1991 Mount Pinatubo eruption. *Geophys. Res. Lett.*, 34, L23710, doi:10.1029/2007GL031403.
- Rodriguez, J. M., M. K. W. Ko, and N. D. Sze, 1991: Role of heterogeneous conversion of N₂O₅ on sulphate aerosols in global ozone losses. *Nature*, 352, 134–137, doi:10.1038/352134a0.

- —, —, C. W. Heisey, G. K. Yue, and M. P. McCormich, 1994: Ozone response to enhanced heterogeneous processing after the eruption of Mt. Pinatubo. *Geophys. Res. Lett.*, **21**, 209–212
- Solomon, S., R. W. Portmann, R. R. Garcia, L. W. Thomason, L. R. Poole, and M. P. McCormick, 1996: The role of aerosol variations in anthropogenic ozone depletion at northern midlatitudes. *J. Geophys. Res.*, 101 (D3), 6713–6727.
- Stenchikov, G., A. Robock, V. Ramaswamy, M. D. Schwarzkopf, K. Hamilton, and S. Ramachandran, 2002: Arctic Oscillation response to the 1991 Mount Pinatubo eruption: Effects of volcanic aerosols and ozone depletion. J. Geophys. Res., 107, 4803, doi:10.1029/2002JD002090.
- Stolarski, R. S., P. Bloomfield, R. D. McPeters, and J. R. Herman, 1991: Total ozone trends deduced from Nimbus 7 TOMS data. Geophys. Res. Lett., 18, 1015–1018.
- —, A. R. Douglass, S. Steenrod, and S. Pawson, 2006: Trends in stratospheric ozone: Lessons learned from a 3D chemical transport model. J. Atmos. Sci., 63, 1028–1041.
- Tabazadeh, A., K. Drdla, M. Schoeberl, P. Hamill, and O. B. Toon, 2002: Arctic "ozone hole" in a cold volcanic stratosphere. *Proc. Natl. Acad. Sci. USA*, 99, 2609–2612, doi:10.1073/ pnas.052518199.
- Telford, P., P. Braesicke, O. Morgenstern, and J. Pyle, 2009: Reassessment of causes of ozone column variability following the eruption of Mount Pinatubo using a nudged CCM. Atmos. Chem. Phys., 9, 4251–4260, doi:10.5194/acp-9-4251-2009.
- Thomason, L. W., G. S. Kent, C. R. Trepte, and L. R. Poole, 1997: A comparison of the stratospheric aerosol background periods of 1979 and 1989–1991. J. Geophys. Res., 102 (D3), 3611–3616.
- Tie, X., and G. Brasseur, 1995: The response of stratospheric ozone to volcanic eruptions: Sensitivity to atmospheric chlorine loading. *Geophys. Res. Lett.*, **22**, 3035–3038.
- Van Roozendael, M., and Coauthors, 1997: Ground-based observations of stratospheric NO₂ at high and midlatitudes in Europe after the Mount Pinatubo eruption. *J. Geophys. Res.*, 102, 19 171–19 176.
- Wuebbles, D. J., D. E. Kinnison, K. E. Grant, and J. Lean, 1991: The effect of solar flux variations and trace gas emissions on recent trends in stratospheric ozone and temperature. *J. Geomagn. Geoelectr.*, 43, 709–718.
- Zerefos, C. S., K. Tourpali, and A. F. Bais, 1994: Further studies on possible volcanic signal to the ozone layer. *J. Geophys. Res.*, **99** (D12), 25 741–25 746.