The response of ozone and nitrogen dioxide to the eruption of Mt. Pinatubo.
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Popular summary
On 15 June 1991 the eruption of Mt. Pinatubo, in the Philippines, injected about 20 Tg of sulfur dioxide (SO$_2$) in the stratosphere. The SO$_2$ transformed into sulfate aerosol, increasing the stratospheric aerosol loading by orders of magnitude. Such a massive aerosol perturbation is expected to change both the chemistry and dynamics of the stratosphere. On the one side, the volcanic sulfate provides additional surface for heterogeneous chemistry, lowering especially the concentrations of ozone and nitrogen dioxide (NO$_2$). On the other side, heating by the volcanic aerosol changes the radiative balance of the atmosphere and, therefore, the atmospheric dynamics.

In the case of the Mt. Pinatubo eruption, the aerosol heating intensified upwelling in the tropics and downwelling in the extra-tropics, strengthening the Brewer-Dobson circulation. Observations also showed a strong depletion of stratospheric NO$_2$ starting from three months after the eruption, indicating a strong volcanic effect on the heterogeneous chemistry. However, while a general depletion of total column ozone was observed in the northern hemisphere, observations did not detect any significant trend in the southern hemisphere, and even registered an increase in the total column ozone during the year following the eruption. The lack of ozone depletion in the southern hemisphere after the eruption of Mt. Pinatubo, contrary to the clear depletion of NO$_2$, has been an outstanding puzzle for many years.

The authors identify the reason of the missing ozone depletion in the volcanic perturbation to the stratospheric chemistry. Using the Goddard Earth Observing System GEOS-5, the authors performed simulations with no volcanic perturbation, including only the volcanic perturbation to the stratospheric chemistry and to the stratospheric dynamics, respectively, and including both these effects. The comparison of the four experiments allows to separately
quantifying the ozone and NO₂ anomalies due to volcanic effects on chemistry and dynamics.

The results of this work show that the perturbation of the stratospheric dynamics by the Mt. Pinatubo eruption is responsible for the lack of observed ozone depletion in the southern hemisphere. By increasing the upwelling in the equatorial region, the volcanic aerosol brings air with lower ozone concentration in the stratosphere and creates a negative anomaly of the total ozone column in the equatorial region. At the same time, the increased upwelling drives greater downwelling south of the equator, inducing a positive ozone anomaly in the southern hemisphere that counteracts the chemical depletion of ozone.

![Ozone column - 30°S to 60°S](image1)

![NO₂ stratospheric column - 45°S](image2)

**Fig. 1:** Zonally averaged anomalies of the total ozone column and stratospheric NO₂ column at southern midlatitudes from observations (black) and simulations including only the chemistry perturbation (blue), only the dynamics perturbation (green), and both the chemistry and the dynamics perturbation (red). The solid lines are significant at one standard deviation level. The volcanic perturbation to the dynamics does not perturb NO₂, but induces a positive anomaly in the total ozone column.
The response of ozone and nitrogen dioxide to the eruption of Mt. Pinatubo.

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Abstract

Observations have shown that the global mass of nitrogen dioxide decreased in both hemispheres in the year following the eruption of Mt. Pinatubo, indicating an enhanced heterogeneous chemistry. In contrast, the observed ozone response was largely asymmetrical with respect to the equator, with a decrease in the northern hemisphere and little change in the southern hemisphere. Simulations including enhanced heterogeneous chemistry due to the presence of the volcanic aerosol reproduce a decrease of ozone in the northern hemisphere, but also produce a comparable ozone decrease in the southern hemisphere, contrary to observations. Our simulations show that the heating due to the volcanic aerosol enhanced both the tropical upwelling and the extratropical downwelling. The enhanced extratropical downwelling, combined with the time of the eruption relative to the phase of the Brewer-Dobson circulation, increased the ozone in the southern hemisphere and counteracted the ozone depletion due to heterogeneous chemistry on volcanic aerosol.

1. Introduction

The volcanic eruption of Mount Pinatubo on 15 June 1991 injected about 20 Tg of sulfur dioxide (SO$_2$) into the stratosphere [Bluth et al, 1992], up to an altitude of about 30 km [McCormick and Veiga, 1992]. The SO$_2$ 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 persisted in the atmosphere for several years.
Aerosol from Mt. Pinatubo reached both the northern (NH) and southern hemispheres (SH), changing the stratospheric chemistry and dynamics. The volcanic sulfate provided additional surface for heterogeneous chemistry, impacting especially the concentrations of ozone and nitrogen dioxide (NO$_2$) [Tie and Brasseur, 1995]. Heating by this volcanic aerosol also changed the radiative balance of the atmosphere, intensifying upwelling in the tropics and downwelling in the extra-tropics [e.g. Pitari and Mancini, 2002; Aquila et al., 2012].

Observations showed depletion of stratospheric NO$_2$ in both hemispheres during the years following the eruption [Johnston et al., 1992, Van Roozendael et al., 1997], showing that the volcanic aerosol enhanced the heterogeneous chemistry at all latitudes. Surprisingly, the observed ozone response to the volcanic perturbation was different in the NH and in the 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 [Randel and Wu, 1996].

Several model studies attribute the observed NH ozone depletion to the enhancement of heterogeneous chemistry because of the volcanic aerosols [e.g. Tie and Brasseur, 1995], but cannot explain why the same heterogeneous chemistry did not affect the SH ozone concentration [Stolarski et al., 2006]. Some studies suggested explanations to the asymmetry of the ozone response other than a chemistry perturbation. Randel and Wu [1996] showed that the quasi-biannual oscillation (QBO) increased ozone in the extratropical SH during the 1991/1992 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 an ozone depletion to interannual dynamical 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, using a free-running global chemistry climate model (Section 2), we separate
the photochemical and dynamical contributions to the ozone and NO₂ anomalies induced
by the volcanic perturbation alone (Section 3). Section 4 presents the main conclusions of
this work.

2. Model and simulation

All simulations are performed with the Goddard Earth Observing System, Version
5 (GEOS-5) model [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 GOCART aerosol transport module [Colarco et al., 2010] and a
stratospheric chemistry module [Pawson et al., 2008]. The resolution is 2.0° x 2.5°
latitude by longitude with 72 vertical layers from surface to 0.01 hPa (~ 95 km). This
version of GEOS-5 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 GEOS-5.
GOCART computes the transformation of SO$_2$ into sulfate aerosol. The simulations shown here use prescribed aerosol surface area density for heterogeneous chemistry from SAM II, SAGE and SAGE-II data [Eyring et al., 2008]. GEOS-5 also includes an option for calculating the aerosol surface area density online using the mass of sulfate aerosol and relative humidity.

We simulate the eruption of Mt. Pinatubo by injecting 20 Tg of SO$_2$ between 16 km 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 (Table 1), each composed of 10 simulations with different sets of initial conditions typical of the year 2000. The results shown here are ensemble averages.

The first experiment (experiment REF) is a control ensemble that does not include any volcanic perturbation.

The second experiment (experiment DYN) includes radiatively interactive aerosol. This experiment uses for heterogeneous chemistry prescribed aerosol surface area density from SAM II and SAGE observations relative to 1979, when the stratospheric aerosol layer is considered to be in an unperturbed condition [Thomason et al., 1997]. Hence, this experiment includes only the volcanic perturbation to the stratospheric dynamics.

The third experiment (experiment CHEM) does not include radiatively interactive aerosol, and uses aerosol surface area density from SAGE-II data appropriate for the simulated year. In this experiment the aerosol cannot modify the simulated meteorology, i.e. there is a perturbation to the stratospheric chemistry but no perturbation to the dynamics.
The fourth experiment (experiment FULL) includes radiatively interactive aerosols and the aerosol surface area density for the simulated year. This experiment includes the volcanic perturbation to both the dynamics and the stratospheric chemistry.

3. Results

The comparison of the experiment FULL with the control experiment REF identifies the complete perturbation of ozone and NO$_2$ due to Mt. Pinatubo. The comparisons of experiments DYN and CHEM with experiment REF isolate the anomalies of ozone and NO$_2$ due to the eruption effect on the atmospheric dynamics and heterogeneous chemistry, respectively.

The left panel of Fig. 1 shows observed deviations of stratospheric NO$_2$ column (black line) over Lauder, New Zealand with a Visible and Ultraviolet (UV/Vis) spectrophotometer (Johnston and McKenzie, 1984) from the observed monthly means over the years 1997 to 2003. In the same panel we also show the simulated anomalies of the stratospheric zonal mean NO$_2$ column, calculated as the difference between experiment REF and the experiments FULL (red line), CHEM (blue line), and DYN (green line), respectively. While the experiment including only the perturbation to the dynamics does not show any significant perturbation of NO$_2$, both experiments CHEM and FULL present a decrease of stratospheric NO$_2$, as in the observations. This shows that the perturbation of NO$_2$ is dominated by the volcanic effect on the stratospheric chemistry, which is similar in both hemispheres (Fig. 1, right panel).

Experiments performed using the online calculation of the aerosol surface area density (not shown) present a larger and earlier depletion of NO$_2$, in better agreement with the observations. The online calculation of the sulfate surface area density produces a
higher surface area than that derived from the observations. Due to the sparse sampling, SAGE-II might have underestimated the transport rate of the aerosol from the tropics to midlatitudes.

The ozone concentration responds differently to the inclusion of the volcanic perturbation to the dynamics. The left panel of Fig. 2 compares the anomalies of the total ozone column, zonally averaged between 30°S and 60°S, as calculated from Total Ozone Mapping Spectrometer (TOMS) data (Herman et al., 1996, McPeters et al., 1996) and as simulated by GEOS-5. The observed anomalies (black line) are calculated as the deviation from the 1987-1990 monthly means after eliminating the depletion due to increasing chlorine. The data show a positive anomaly for one year after the eruption, simulated also in the experiments DYN and FULL. This positive anomaly is induced by the absorption of largely longwave radiation by the volcanic aerosol, which leads to an increase in the Brewer-Dobson circulation (Aquila et al., 2012) and to a subsequent positive anomaly of ozone total column in the southern hemisphere (Fig. 2, right panel). The experiment DYN does not produce any significant perturbation after the initial positive anomaly, while the experiment CHEM shows a significant negative perturbation starting from April 1992.

The ozone anomaly in the experiment FULL is essentially the sum of the DYN and CHEM anomalies, but the dynamical positive anomaly has delayed the negative anomaly from April to August 1992.

Fig. 3 shows the simulated vertical distribution of the zonal mean ozone anomaly in June-July-August (JJA) 1991 (left panel) and September-October-November (SON) 1992 (right panel) due to the heterogeneous chemistry and dynamics (FULL-REF). In JJA 1991 the perturbation is 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 (Fig. 3, right panel). At the same time, the increased upwelling drives greater downwelling south of the equator, creating the positive ozone anomaly in the SH. 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 towards the winter 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. 3, right panel) the simulated ozone anomaly is mainly due to the perturbation to the chemistry. The ozone concentration is increased in the middle stratosphere due to the suppression of the NO\(_x\) cycle induced by the volcanic aerosol, and decreased in the lower stratosphere due to the enhancement of the HO\(_x\) and ClO\(_x\) cycles. Tie and Brasseur, [1995], described this chemical response to the volcanic aerosol.

4. Conclusions

The lack of observed ozone depletion due to the eruption of Mt. Pinatubo in the southern hemisphere, contrary to the clear depletion of NO\(_2\), has been an outstanding puzzle for many years [WMO, 2011]. We have shown that the perturbation of the stratospheric dynamics by the Mt. Pinatubo eruption is responsible for the lack of an observed ozone decrease in the SH. The dynamics response to the volcanic perturbation dominates the changes in ozone column during the first 6 months after the eruption and fades away starting in about January 1992. The chemical response, instead, produces significant changes starting about one year after the eruption. The perturbations to the
chemistry and to the dynamics have an additive effect, resulting in the lack of ozone
depletion in the year following the eruption.

On the other hand, the NO$_2$ anomaly is completely driven by the chemistry
perturbation and is insensitive to the dynamics perturbation. The reason is the much
shorter timescale of the heterogeneous chemistry for depleting NO$_2$ compared to the
timescale 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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**Table 1:** list of performed experiments.

<table>
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<th>Experiment</th>
<th>Radiatively interactive aerosol</th>
<th>Year for sulfate area density</th>
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<td>No</td>
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<tr>
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<td>1991-1995</td>
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Fig. 1: NO$_2$ stratospheric column anomaly versus time. Left panel: The black line marks the observed anomalies over Lauder, the red, blue and green lines show the simulated zonal mean anomaly at 45°S. The shaded areas show the standard deviation of each ensemble. Right panel: Zonal mean of the simulated NO$_2$ column anomalies in experiment FULL with respect to experiment REF. The solid lines (left) and bright areas (right) are significant at 1-σ level.
**Fig. 2:** Ozone total column anomaly versus time. Left panel: zonal mean between 30°S and 60°S as observed by TOMS (black line) and as simulated by GEOS-5 (blue, red and green lines). The shaded areas show the standard deviation of each ensemble. Right panel: Zonal mean of the simulated ozone column anomalies in experiment FULL with respect to experiment REF. The solid lines (left) and bright areas (right) are significant at 1-σ level.
Fig. 3: Vertical distribution of the zonal mean ozone relative anomaly [%] in June-July-August 1991 (left panel) and September-October-November 1992 (right panel) of the experiment FULL with respect to the experiment REF. The streamlines show the anomaly of the residual circulation.