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. In contrast, the observed ozone response was largely asymmetrical with respect to the equator, with a decrease in the northern hemisphere and little change and even a small increase 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 changed both 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]. The observed ozone response to the volcanic perturbation, on the other hand, was different in the northern (NH) and in the southern hemisphere (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 depletion to the enhancement of heterogeneous chemistry because of the volcanic aerosols [e.g. *Tie and Brasseur,* 1995].

The volcanic impact on the stratospheric chemistry cannot explain the asymmetric ozone perturbation observed in the NH and SH [*Stolarski et al.,* 2006], and several studies suggested explanations 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

46 ozone depletion to interannual dynamical variability, which masked the Pinatubo aerosol
47 chemical effect in the SH. However, these studies cannot distinguish between natural
48 interannual variability and circulation changes forced by the volcanic perturbation.
49 *Poberaj et al.* [2011] performed a multiple linear regression analysis to the Chemical and
50 Dynamical Influences on Decadal Ozone Change (CANDIDOZ), stating that volcanically
51 induced chemical ozone depletion was overcompensated by the QBO and by a
52 pronounced EP flux anomaly.

53 Here, using a free-running global chemistry climate model (Section 2), we separate
54 the photochemical and dynamical contributions to the ozone and NO_2 anomalies induced
55 by the volcanic perturbation (Section 3). Section 4 presents the main conclusions of this
56 work.

57 **2. Model and simulation**

58 All simulations are performed with the Goddard Earth Observing System, Version
59 5 (GEOS-5) model [*Rienecker et al.*, 2008], a system of component models integrated
60 using the Earth System Modeling Framework (ESMF). For these simulations GEOS-5 is
61 coupled to the GOCART aerosol transport module [*Colarco et al.*, 2010] and a
62 stratospheric chemistry module [*Pawson et al.*, 2008]. The resolution is 2.0° x 2.5°
63 latitude by longitude with 72 vertical layers from surface to 0.01 hPa (~ 95 km). This
64 version of GEOS-5 does not simulate the QBO. The model is forced with observed sea
65 surface temperatures and sea ice concentrations [*Reynolds et al.*, 2002]. *Aquila et al.*
66 [2012] includes a more detailed description of the model and an evaluation of the
67 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.

90 The fourth experiment (experiment FULL) includes radiatively interactive aerosols
91 and the aerosol surface area density for the simulated year. This experiment includes the
92 volcanic perturbation to both the dynamics and the stratospheric chemistry.

93 **3. Results**

94 The comparison of the experiment FULL with the control experiment REF
95 identifies the complete perturbation of ozone and NO_2 due to Mt. Pinatubo. The
96 comparisons of experiments DYN and CHEM with experiment REF isolate the anomalies
97 of ozone and NO_2 due to the eruption effect on the atmospheric dynamics and
98 heterogeneous chemistry, respectively.

99 The left panel of Fig. 1 shows observed deviations of stratospheric NO_2 column
100 (black line) over Lauder, New Zealand with a Visible and Ultraviolet (UV/Vis)
101 spectrophotometer (*Johnston and McKenzie,* 1984) from the observed monthly means
102 over the years 1997 to 2003. In the same panel we also show the simulated anomalies of
103 the stratospheric zonal mean NO_2 column, calculated as the difference between
104 experiment REF from the experiments FULL (red line), CHEM (blue line), and DYN
105 (green line), respectively. While the experiment including only the perturbation to the
106 dynamics does not show any significant perturbation of NO_2, both experiments CHEM
107 and FULL present a decrease of stratospheric NO_2, as in the observations. This shows that
108 the perturbation of NO_2 is dominated by the volcanic effect on the stratospheric chemistry,
109 which is similar in both hemispheres (Fig. 1, right panel).

110 Experiments performed using the online calculation of the aerosol surface area
111 density (not shown) present a larger and earlier depletion of NO_2, in better agreement with
112 the observations. The online calculation of the sulfate surface area density produces a

113 higher surface area than that derived from the observations. Due to the sparse sampling,
114 SAGE-II might have underestimated the transport rate of the aerosol from the tropics to
115 midlatitudes.

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

132 Fig. 3 shows the simulated vertical distribution of the zonal mean ozone anomaly
133 in June-July-August (JJA) 1991 (left panel) and September-October-November (SON)
134 1992 (right panel) due to the heterogeneous chemistry and to the dynamics (FULL-REF).
135 In JJA 1991 the perturbation is completely dominated by the dynamics response, which

136 we depict with the streamlines of the residual circulation anomaly. The increased tropical
137 upwelling lifts air with lower ozone concentration, creating a negative equatorial anomaly
138 centered at 20 hPa (Fig. 3, right panel). At the same time, the increased upwelling drives
139 greater downwelling south of the equator, creating the positive ozone anomaly in the SH.
140 This positive anomaly is located in the SH because of the phase of the Brewer-Dobson
141 circulation at the time of the eruption, which is directed towards the winter hemisphere.
142 We performed an experiment initiating a Pinatubo-like eruption on 15 January 1991 (as
143 described in *Aquila et al.,* 2012). There the positive ozone anomaly appeared in the NH
144 (not shown), compatible with the different phase of the Brewer Dobson circulation.

145 In SON 1992 (Fig. 3, right panel) the simulated ozone anomaly is mainly due to
146 the perturbation to the chemistry. The ozone concentration is increased in the middle
147 stratosphere due to the suppression of the NO_x cycle induced by the volcanic aerosol, and
148 decreased in the lower stratosphere due to the enhancement of the HO_x and ClO_x cycles.
149 *Tie and Brasseur*, [1995], described this chemical response to the volcanic aerosol.

150 **4. Conclusions**

151 The lack of observed ozone depletion due to the eruption of Mt. Pinatubo in the
152 southern hemisphere, contrary to the clear depletion of NO_2, has been an outstanding
153 puzzle for many years [*WMO*, 2011]. We have shown that the perturbation of the
154 stratospheric dynamics by the Mt. Pinatubo eruption is responsible for the lack of an
155 observed ozone decrease in the SH. The dynamics response to the volcanic perturbation
156 dominates the changes in ozone column during the first 6 months after the eruption and
157 fades away starting in about January 1992. The chemical response, instead, produces
158 significant changes starting about one year after the eruption. The perturbations to the

159 chemistry and to the dynamics have an additive effect, resulting in the lack of ozone
160 depletion in the year following the eruption.

161 On the other hand, the NO_2 anomaly is completely driven by the chemistry
162 perturbation and is insensitive to the dynamics perturbation. The reason is the much
163 shorter timescale of the heterogeneous chemistry for depleting NO_2 compared to the
164 timescale for depleting ozone, together with the weak NO_2 vertical gradient, such that
165 changes of vertical advection do not induce large perturbations.

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Table 1: list of performed experiments.

Experiment	Radiatively interactive aerosol	Year for sulfate area density	Perturbation to the chemistry	Perturbation to the dynamics
REF	No	1979		
CHEM	No	1991-1995	X	
DYN	Yes	1979		X
FULL	Yes	1991-1995	X	X

239

240 **Fig. 1:** NO_2 stratospheric column anomaly versus time. Left panel: The black line marks
241 the observed anomalies over Lauder, the red, blue and green lines show the simulated
242 zonal mean anomaly at 45°S. The shaded areas show the standard deviation of each
243 ensemble. Right panel: Zonal mean of the simulated NO_2 column anomalies in experiment
244 FULL with respect to experiment REF. The solid lines (left) and bright areas (right) are
245 significant at 1-σ level.

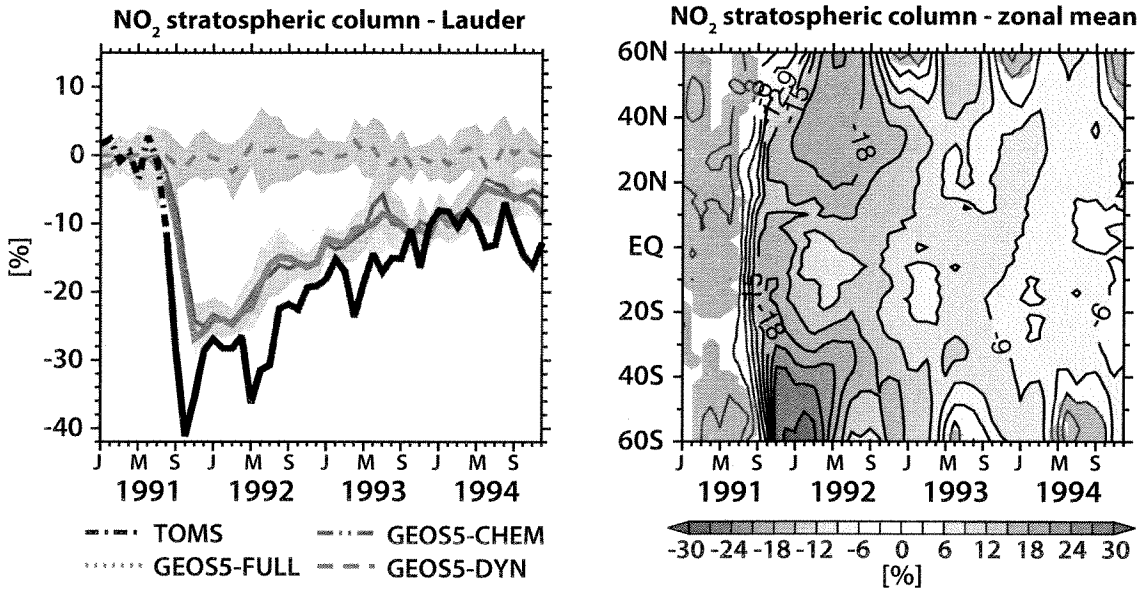

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Fig.2: : Ozone total column anomaly versus time. Left panel: zonal mean between 30°S and 60°S as observed by TOMS (black line) and as simulated by GEOS-5 (blue, red and green lines). The shaded areas show the standard deviation of each ensemble. Right 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.

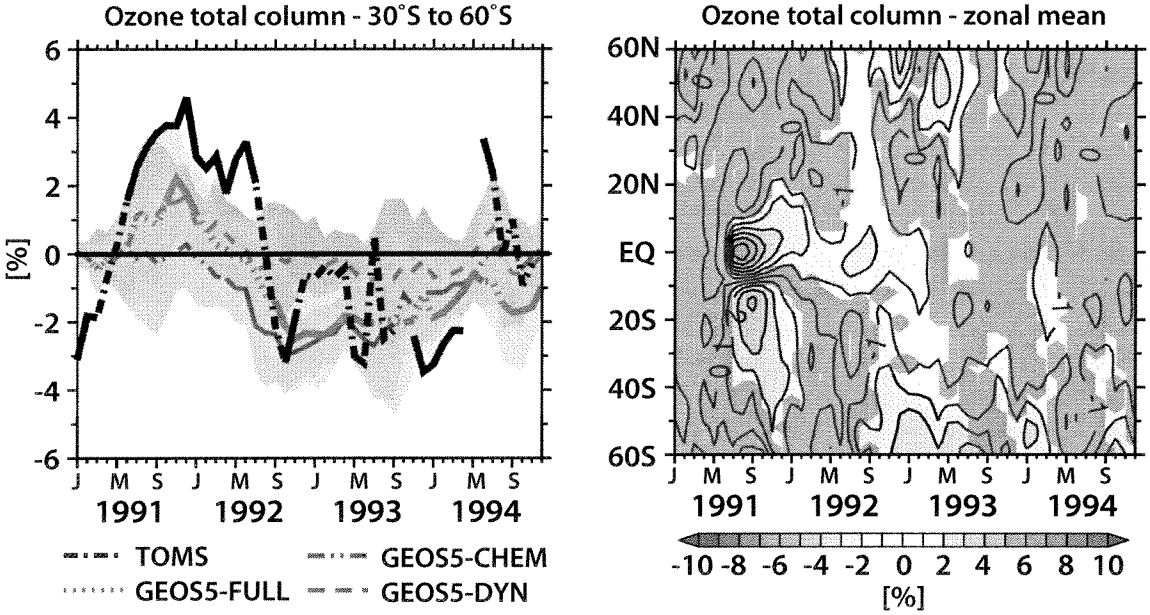

Fig. 3: Vertical distribution of the zonal mean ozone anomaly in June-July-August 1991 (left panel) and September-October-November 1992 (right panel) of the experiment FULL with respect to the experiment REF. The streamlines show the anomaly of the residual circulation.