Dispersion of the volcanic sulfate cloud from the Mount Pinatubo eruption

Valentina Aquila¹, Luke D. Oman¹, Richard S. Stolarski¹,², Peter R. Colarco¹, Paul A. Newman¹

¹ Laboratory for Atmospheric Chemistry and Dynamics, NASA Goddard Space Flight Center, Greenbelt, Maryland, USA
² Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, MD, USA
Abstract

We simulate the transport of the volcanic cloud from the 1991 eruption of Mount Pinatubo with the GEOS-5 general circulation model. Our simulations are in good agreement with observational data. We tested the importance of initial condition corresponding to the specific meteorological situation at the time of the eruption by employing reanalysis from MERRA. We found no significant difference in the transport of the cloud. We show how the inclusion of the interaction between volcanic sulfate aerosol and radiation is essential for a reliable simulation of the transport of the volcanic cloud. The absorption of long wave radiation by the volcanic sulfate induces a rising of the volcanic cloud up to the middle stratosphere, combined with divergent motion from the latitude of the eruption to the tropics. Our simulations indicate that the cloud diffuses to the northern hemisphere through a lower stratospheric pathway, and to mid- and high latitudes of the southern hemisphere through a middle stratospheric pathway, centered at about 30 hPa. The direction of the middle stratospheric pathway depends on the season. We did not detect any significant change of the mixing between tropics and mid- and high latitudes in the southern hemisphere.

1. Introduction

Volcanic eruptions are a major source of stratospheric aerosol [Deshler, 2008]. Sulfur dioxide injected into the stratosphere by large eruptions is oxidized into sulfate aerosol and can increase the background aerosol mass by orders of magnitude. The induced perturbation of the stratospheric aerosol layer can persist for some years. During such
time the aerosol can spread over the whole globe, changing the global climate in a significant way [Robock, 2000].

Mt. Pinatubo is located in the Philippines (15.1°N, 120.4°E). Pinatubo erupted on June 15th, 1991, injecting about 20 Tg of sulfur dioxide into the atmosphere [Bluth et al., 1992]. The resulting sulfate cloud was detected at altitudes higher than 30 km [McCormick and Veiga, 1992] and, after about one year, roughly one third of the volcanic aerosol was still present in the atmosphere.

The sulfate cloud generated by the eruption of Mt. Pinatubo circled around the Earth within 3 weeks of the eruption [Guo et al., 2004; McCormick and Veiga, 1992], crossing the equator and diffusing to mid- and high latitudes in both the northern and the southern hemispheres.

Such a broad meridional spreading of the cloud is not typical of all tropical eruptions. For example, the cloud from the June 1982 El Chichón eruption, which is located 2° north of Mt. Pinatubo, was mainly confined to the northern hemisphere [McCormick and Swissler, 1983]. Young et al., [1994] first suggested that the cross-equatorial transport of the Mt. Pinatubo cloud was due to local absorption of infrared radiation from the troposphere. Timmreck et al., [1999a] confirmed this hypothesis from a theoretical point of view with a one-simulation study with the MAECHAM4 Hamburg climate model.

Niemeier et al., [2009] applied the most recent version of the MAECHAM5 Hamburg climate model, coupled to an aerosol microphysical model, to the study of the Pinatubo eruption. Other studies, such as Stenchikov et al., [1998], Kirchner et al., [1999] and Thomas et al., [2009a; 2009b] used prescribed aerosol distributions.
It is still unclear if the eruption of Mt. Pinatubo modified the circulation in the southern hemisphere. Robock *et al.*, [2007] identified no significant anomaly in the southern hemisphere circulation in their simulations with the NASA/GISS ModelE general circulation model. In contrast, Karpechko *et al.*, [2010], Marshall, [2003], Roscoe and Haigh, [2007] and Crooks and Gray, [2005] found a negative response of the Southern Annular Mode in both models and observations.

In this paper, we simulate the eruption of Mt. Pinatubo and the dispersal of the subsequent sulfate cloud with the Goddard Earth Observing System (GEOS-5) general circulation model [Rienecker *et al.*, 2008], coupled to the GOCART aerosol transport module [Colarco *et al.*, 2010] and the StratChem stratospheric chemistry module [Pawson *et al.*, 2008]. GEOS-5 is here for the first time applied to the simulation of stratospheric volcanic aerosol.

In section 2, we describe the model and the modifications introduced to simulate stratospheric volcanic aerosol. Given the large amount of observations, the eruption of Mt. Pinatubo is a good test for the ability of GEOS-5 to correctly simulate the dispersal of the volcanic cloud and the response to sudden aerosol perturbations in the stratosphere.

In section 3, we present the model results and the comparison with observations. We show that our simulations are in good agreement with observations.

Finally, in section 4 we apply GEOS-5 to the study of the interaction between Mt. Pinatubo aerosols and the stratospheric circulation, focusing on the mixing between tropics and midlatitudes.
2. The GEOS-5 general circulation model

All simulations presented in this study 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).

The GEOS-5 atmospheric general circulation model (AGCM) is able to perform weather and climate simulations used for atmospheric analyses, weather forecasts and climate simulations and predictions. GEOS-5 uses a finite-volume dynamical core [Lin, 2004] combined with a physics package that describes moist processes, radiation, turbulent mixing and surface processes.

The convective parameterization Relaxed Arakawa-Schubert (RAS) is described by Moorthi and Suarez, [1992], and is combined to a prognostic cloud scheme. The boundary-layer turbulent mixing is parameterized with the schemes by Louis et al., [1982] and Lock et al., [2000], for stable and unstable situations, respectively. The land-surface model is composed of a catchment-based hydrological model [Koster et al., 2000] and by a multi-layer snow model [Stieglitz et al., 2001]. Coupled chemistry-climate simulations can be performed using the StratChem module for stratospheric chemistry [Pawson et al., 2008].

The radiative transfer model consists of a solar radiation model [Chou and Suarez, 1999] and a thermal radiation model [Chou et al., 2001]. The solar radiation model includes absorption due to water vapor, O$_3$, O$_2$, CO$_2$, clouds and aerosol. The thermal radiation model includes absorption by water vapor, CO$_2$, O$_3$ and most of the minor trace gases, as well as clouds and aerosol.
The aerosol optical properties are read from look-up tables previously generated using the OPAC database [Hess et al., 1998]. The look-up tables contain the aerosol mass scattering and extinction coefficients as a function of relative humidity and radiation wavelength. We apply the Mie theory to calculate of the aerosol optical properties, and assume that aerosol is log-normally distributed and externally mixed.

GEOS-5 can be run both in climate or data assimilation mode. The simulations performed in this study are climate mode simulations, i.e. they provide a forecast of the climate starting from specified initial conditions. We apply GEOS-5 with resolution 2.0° x 2.5° latitude by longitude. The model has 72 vertical layers in a hybrid coordinate system from surface to 0.01 hPa.

The aerosol transport model in the GEOS-5 AGCM is based on the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model [Chin et al., 2000; 2002]. An online version of GOCART in GEOS-4, a previous version of GEOS-5, has been validated by Colarco et al., [2010]. Versions of GOCART in GEOS-5 have been already used in several recent field campaigns, as TC4 (2007), ARCTAS (2008) and GloPac (2010).

The aerosol species treated by GOCART as described in Colarco et al., [2010] are dust, sea salt, black carbon, organic carbon and sulfate (SO₄). In this study, only the sulfate component is active. GOCART includes a parameterization of the chemical production of SO₄ from oxidation of dimethyl sulfide (DMS) by OH during day and NO₃ during night, and from oxidation of sulfur dioxide (SO₂) by OH in the gas phase and by H₂O₂ in the aqueous phase.
GEOS-5 can run with radiatively interactive aerosol, which means that the aerosol concentrations simulated by GOCART can modify the meteorological fields. The simulations shown in Section 4 are performed with radiatively interactive aerosol. Some results from runs with non-interactive aerosol are presented in Section 4.

We introduced a parameterization of the settling of SO$_4$ to properly simulate stratospheric volcanic aerosol. The settling velocity is a function of the particle’s wet radius. The sulfate growth factor $\beta_{SO_4}$ is calculated as a function of the relative humidity RH following Petters and Kreidenweis, [2007] as

$$\beta_{SO_4} = \frac{r_{sett}}{r_{dry}} = \sqrt{\frac{RH(1-k)-1}{RH-1}},$$

where the hygroscopic parameter $k$ is equal to 1.19 and $r_{dry}$ the dry effective radius, which is a tuning parameter.

Assuming a lognormal distribution, the modal radius $r_m$ and the effective radius $r_e$ are related through the equation

$$r_e = r_m \exp\left[\frac{5}{2} \ln^2 \sigma\right],$$

where $\sigma$ is the standard deviation of the distribution.

We performed several sensitivity tests varying the value of the sulfate dry radius. In this work we assume that aerosol is lognormal distributed with median diameter radius to 0.35 μm and standard deviation 1.25. This corresponds to an effective dry radius equal to 0.40 μm. This modal radius is within the range of observed values for sulfate aerosol from Mt. Pinatubo (e.g. Bingen et al., [2004]; Niemeier et al., [2009]; Russell et al.,
[1996]; Stenchikov et al., [1998]), and result in good agreement with the AOT retrieved by SAGE-II and AVHRR (Section 3). The simulated e-folding time for sulfate is about one year (Table 1), as the one calculated from observations [McCormick et al., 1995].

3. Simulation of the Mt. Pinatubo eruption

We simulated the eruption of Mt. Pinatubo by injecting 20 Tg of sulfur dioxide in the grid box containing Mt. Pinatubo during the day of June 15th, 1991. The \( \text{SO}_2 \) load is initially distributed between 16 km and 18 km, and is lofted to higher altitudes within the first weeks due to the model response to radiatively interacting aerosol.

Other model studies, as e.g. [Timmreck et al., 1999b] and [Zhao et al., 1995], place the injection of \( \text{SO}_2 \) at higher altitude. They base their assumption on SAGE-II observations. At the moment of the eruption, however, SAGE-II was observing at about 70°N [Trepte et al., 1993], and observed at the latitude of Mt. Pinatubo only 15 days after the eruption. At that stage the absorption of radiation by the volcanic aerosol had already induce the lofting of the cloud itself.

We tested similar assumptions on the injection height in GEOS-5 by performing simulations with injection of \( \text{SO}_2 \) between 16 km and 25 km, 17 km and 27 km, 20 km and 27 km, 20 km and 30 km. In all these simulations, the bulk of the volcanic cloud reached altitudes much higher than observations. Our choice of a lower injection altitude results in a reasonable simulation of the SAGE-II vertical profile after a couple of weeks from the day of the eruption (Section 3.2).

We did not include any other aerosol sources in the simulations used for this work.
We performed an ensemble of eight transient simulations, each spanning from January 1991 to December 1997. The initial conditions of the ensemble members are the meteorological fields of eight different Januaries of a control simulation with no volcanic perturbation, which was initialized with climatological meteorological fields typical of the year 2000.

The injected $\text{SO}_2$ is transformed into $\text{SO}_4$ by GOCART with an average e-folding time of 29.8 days, in good agreement with observations by the Total Ozone Mapping Spectrometer (TOMS) [Bluth et al., 1992; Guo et al., 2004]. The average of the $\text{SO}_4$ e-folding times of the eight ensemble members is 347 days with a standard deviation of 57.7 days, also in good agreement with observations [Barnes and Hofmann, 1997; Nagai et al., 2010]. Table 1 shows the e-folding times of $\text{SO}_2$ and $\text{SO}_4$ for each ensemble member.

Figure 1 shows the temporal evolution of the globally averaged AOT at 550 nm. The results from our simulations are compared to SAGE II [Thomason et al., 1997] and AVHRR [Long and Stowe, 1994] observations. We removed background values from the AVHRR observations, calculated as the monthly mean AOT over the available months preceeding the eruption (June 1989 to May 1991).

The simulated peak value is in reasonable agreement with AVHRR, but is higher in magnitude and occur earlier in time than the SAGE-II data. Optical depths of about 0.15 or more, however, saturate the SAGE-II measurement [Russell et al., 1996]. Hence, the value of AOT calculated from SAGE-II observations are likely underestimated. Additionally, the sampling of SAGE-II observations is relatively sparse and can hardly
register rapid changes in the AOT. After January 1992 both SAGE-II and AVHRR observations are within the variability of the ensemble.

3.1. Horizontal dispersion of the volcanic cloud

Shortly after the eruption, the volcanic cloud is transported northward out of the tropics and southward toward the equator.

Figure 2 shows the zonal mean of the AOT as a function of time in our simulations and in the satellite observations. The model reproduces reasonably well the spreading of the cloud into the two hemispheres observed by SAGE-II and AVHRR. GEOS-5 simulates well the timing and the intensity of the tropical peak compared to AVHRR. As expected from the profiles in Figure 1, the magnitude of the simulated aerosol optical thickness is larger than the one observed by SAGE-II.

GEOS-5 simulates also the second peak in February, 1992 at 45°N, as well as the secondary peak detected by AVHRR at about 10°N in September, 1991. The high AOT values observed at 60°S in November 1991 might be due to the eruption of the Cerro Hudson volcano (72.97°W, 45.90°S) between August and October 1991, which is not included in our simulations.

GEOS-5 transports a large fraction of the cloud southward shortly after the eruption, but slightly underestimates the transport across the equator with respect to the observations. While the simulated peak is located on the equator, both SAGE-II and AVHRR detected the peak at about 5°S. The results by [Timmreck et al., 1999a] also underestimated the cross-equatorial transport. They suggested that the missing transport
might be due to the specific synoptic situation in June 1991, when a strong high over Tibet induced a southward transport of the cloud.

We tested the importance of the specific meteorological situation by performing a simulation with specified initial conditions from the Modern Era Retrospective Analyses (MERRA, Rienecker et al., [2011]). The results (not shown) are similar to the one of the reference simulations, with a peak of the AOT on the equator. This suggest that the particular meteorological situation at the moment of the eruption is not responsible for the additional southward transport.

The tests performed with different injection heights (Section 3) showed a very similar horizontal distribution of the AOT, even if the volcanic clouds reached altitudes higher than observed.

The small initial underestimation of the southward transport might be due to the lack of a radiatively interactive SO$_2$ in the version of GEOS-5 used in this work. Lary et al., [1994] estimated that the SO$_2$ heating rate can be up to 1 K/day and could therefore be significant in the early stages of the cloud’s evolution.

### 3.2. Vertical distribution of the volcanic cloud

We compare our results with SO$_2$ profiles taken with a microwave limb sounder (MLS) by Read et al., [1993] between 10°S and the equator on September 21$^{st}$, 1991 (Figure 3).

The simulated profiles correspond to the September 1991 monthly mean of the SO$_2$ vertical profiles, averaged over the latitudinal band between 10°S and 10°N. We averaged over a latitudinal band larger than the MLS observations to take into account the different transport pattern between simulations and observations.
The agreement with [Read et al., 1993] is good: both SO$_2$ profiles have a peak at about 20 hPa of similar magnitude. The sensitivity tests that we performed varying the injection altitude of SO$_2$ showed differences in the vertical profile of the volcanic cloud during the first months, but the equilibrium level where the bulk of the cloud settles was in all tests at about 20 hPa.

Figure 4 shows the vertical distribution of the zonally averaged SO$_4$ concentration on July 15$^{th}$, September 1$^{st}$, November 1$^{st}$ and December 31$^{st}$, 1991. The bulk of the cloud is between 50 hPa and 10 hPa in July 1991. The model results are in agreement with SAGE-II observational satellite data, which detected the cloud top at altitudes up to 29 km (about 10 hPa) during June, July and August 1991 [McCormick and Veiga, 1992].

Trepte et al., [1993] showed the latitude-altitude cross-section of the SAGE-II 1$\mu$m extinction ratio. Data were first collected in the tropical region between July 1$^{st}$ and July 20$^{th}$, and show values higher than the background between the tropopause and 30 km altitude, in reasonable agreement with the first panel of Figure 4.

The simulated vertical profiles for December (Figure 4, lower-right panel) also agrees with SAGE-II data, as analyzed by Vernier et al., [2011]. They detected the volcanic cloud at altitudes higher than 35 km, with its bulk between 26 and 27 km.

3.3. Cross-equatorial transport

The volcanic cloud moves to mid- and high latitudes through two main transport pathways, as shown in Figure 4. Already one week after the eruption, part of the cloud is
adveected northward through the lower stratosphere at about 100 hPa. A portion of the cloud, instead, later reaches southern higher latitudes through the middle stratosphere between 5 and 50 hPa and arrives at 90°S in the middle of November (Figure 4, lower panels).

The volcanic cloud crosses the equator during the first two weeks after the eruption, but the transport from the tropics to southern midlatitudes does not start until the middle of July and becomes significant in September (Figure 4, upper right panel).

The middle stratospheric transport regime is illustrated in Figure 5. In our simulations the volcanic cloud reaches 30 hPa about one week after the eruption (not shown) and is by then still located in the northern hemisphere. At the same time part of the cloud has already reached 40°N and 30°S latitude through the lower stratospheric pathway. At the beginning of July (Figure 5, upper panel) the volcanic cloud has dispersed longitudinally over nearly the whole globe, but is still confined in the tropical area, with a sharp gradient at 20°S. The same configuration was observed in SAGE-II data [McCormick and Veiga, 1992; Trepte et al., 1993].

About one month after the eruption we observe the first intrusion of volcanic material from the southern tropics to midlatitudes through tongue-like structures that appear in the middle stratosphere (Figure 5, lower panel). Such tongues of air have been identified by Randel et al., [1993] as the path of mixing from the tropics to midlatitudes. Trepte et al., [1993] detected in the SAGE-II observations similar intrusions detaching from the tropical cloud at 20°S between July 11th, 1991 and July 18th, 1991.

Our transport simulation of the volcanic cloud from the Mt. Pinatubo eruption is in good agreement with the observations. Both the vertical and horizontal distribution and
the timing of the mixing to mid- and high latitudes are reasonably well comparable to SAGE-II and AVHRR observations.

4. Importance of a radiative active volcanic aerosol

We investigate how the interaction between volcanic aerosol from Mt. Pinatubo and radiation changed the background mixing within the tropics and from the tropics to midlatitudes. We performed an ensemble of simulations with no interactive aerosol, and compared them to the reference simulation of the dispersal of the volcanic cloud evaluated in Section 3. We performed one additional ensemble of eight members without coupling between aerosol and radiation. Each ensemble member has exactly the same setup of the reference simulations.

Figure 6 (upper panel) shows the temporal evolution of the zonally averaged AOT at 550 nm, to be compared to the upper panel of Figure 2. In the non-interactive ensemble, most of the volcanic cloud is directed toward the northern hemisphere, faster than in the reference simulation. This is due to the different vertical distribution of the volcanic cloud: in the non-interactive case the volcanic cloud stays at much lower altitudes (Figure 7, middle panel) than in the interactive case (Figure 7, left panel).

Hence, the non-interactive cloud does not rise enough to enter the middle stratosphere, and the advection of the cloud to midlatitudes takes place only through the lower stratosphere. The e-folding time of SO$_4$ is much lower in the ensemble with non-interactive aerosol than in the reference case (74 days against 346 days).

We also performed an ensemble of three non-interactive simulations directly injecting SO$_2$ between 17 km - 27 km (Figure 6, lower panel). Also in this case the cross-
equatorial transport is not as intense as in the reference simulation, and the volcanic cloud looks even more confined to the tropics than in Figure 6a. Even if the SO$_4$ cloud reaches the middle stratosphere (Figure 7, right panel) and part of the cloud crosses the equator, it remains confined within the tropics.

The additional transport to the southern hemisphere is therefore due to the radiative interaction of volcanic aerosol, and is essential for a good simulation of the dispersal of the volcanic cloud, as observed by Timmreck et al., [1999a].

4.1. Perturbation of the background winds

In Figure 8 we show the perturbation of the horizontal wind fields induced by the interaction between radiation and volcanic aerosol. We show the difference of the horizontal winds between the interactive and the non-interactive ensembles on June 16$^{th}$, 1991 at 70 hPa and on July 1$^{st}$, 1991, together with the aerosol heating rates due to longwave radiation. To reduce noise effects, the results for July 1$^{st}$ in Figure 8 and Figure 10 are from an eight-member ensemble of non-interactive simulations starting on the midnight of July 1$^{st}$, with initial conditions from July 1$^{st}$ of the interactive run.

The sudden warming generates a divergent motion from the location of the volcanic cloud already one day after the eruption (Figure 8, upper panel). The simulated volcanic cloud is still at the same latitude as Mt. Pinatubo, and has not risen yet to altitudes higher than 50 hPa. On July 1$^{st}$ (Figure 8, lower panel) the cloud has circled nearly around the whole globe, but, in the middle stratosphere, is still confined between 20°S and 20°N.
The perturbation of the horizontal winds diffuses the sulfate northward and southward from the center of the clouds, increasing the spreading of the clouds towards the tropics, due to the heating induced by the SO$_4$ absorption of longwave radiation. The winds are no longer significantly perturbed at 30 hPa by December 1991, when the concentration of SO$_4$ becomes meridionally homogeneous. At 50 hPa, where the SO$_4$ concentration decreases (Figure 4), the winds converge towards the center of the cloud. At altitudes lower than 50 hPa no consistent perturbation is simulated.

GEOS-5 simulates the formation of two vortices at the location of the volcanic cloud during the second week after the eruption, north and south of the equator, respectively (Figure 9). This feature is similar to the response to a tropical tropospheric heating source calculated by [Gill, 1980], with a high pressure system at the top of the perturbation and a low pressure one at the bottom. A comparison with observation could identify if such a response was indeed observed.

The divergent winds are strongly related to an increased upwelling. Figure 10 shows the perturbation of the wind’s vertical velocity on the same days and levels depicted in Figure 8, and the contours of the SO$_4$ distribution. The increase of the vertical velocity is significant: in the non-interactive case the values of the vertical velocity are up to 0.5 mm/s, while in the perturbed case they reach up to 4 mm/s, in the regions with highest concentration of sulfate. In some regions the perturbation even changes the sign of the vertical wind.

The absorption of longwave radiation by volcanic SO$_4$ is responsible for the “self-lofting” of the volcanic clouds and for the divergent motion from the areas with highest SO$_4$ concentration. As already mentioned in Section 3.1, the introduction of radiative
interactive SO$_2$ could possibly increase the lofting and spreading of the cloud during the first months from the eruption.

The first stage of the volcanic cloud’s transport to the southern hemisphere is driven by the absorption of longwave radiation and comprises the transport from the latitude of the eruption across the equator and to the tropics.

Afterwards, the cloud is transported from the tropics to southern mid- and high latitudes through the structures depicted in Figure 5. We investigated if the volcanic perturbation from Mt. Pinatubo enhanced such structures, and hence the mixing between tropics and midlatitudes, by analyzing the distribution of N$_2$O (not shown).

Climatologically, the concentration of N$_2$O is highest in the tropics and presents a strong summer gradient between the tropics and midlatitudes. The sources of N$_2$O are located at surface and its concentration decreases with altitude.

Compared to the unperturbed case, GEOS-5 simulates decreased N$_2$O at about 30 hPa and increased N$_2$O at 10 hPa in the tropical region starting from September 1991, compatible with the lofting of air induced by the volcanic perturbation. The effect of the lofting weakens starting from January 1992 and no significant change in the N$_2$O concentration can be detected after September 1992.

There is no sign of increased N$_2$O transport from the tropics to midlatitudes.

The analysis of the age of air at 30 hPa leads to similar conclusion (not shown). The air in the interactive runs is younger in the tropical area between July and December 1991, implying a faster upwelling, but there is no significant difference in the following months, neither in the tropics nor at midlatitudes.
4.2. Impact of the sulfate burden on the spreading of the volcanic cloud

We performed two interactive experiments lowering the amount of injected SO₂ to 5 Tg. In the first experiment we injected SO₂ between 16 km and 18 km, in the second between 17 km and 27 km. The set up of the simulations is otherwise identical to the reference simulation. Figure 11 shows the vertical profiles of the zonal mean of SO₄ in the two experiments.

In the experiment with low injection height (Figure 11a) the volcanic cloud is mainly confined to the lower stratosphere, showing that 5 Tg of SO₂ do not produce a strong enough perturbation to raise the cloud to the middle stratosphere.

In the second experiment (Figure 11b) the cloud is injected already in the middle stratosphere and is advected to the southern hemisphere through the same middle stratospheric pathway as in the reference simulation. However, the peak of SO₄ is north of the equator, while in the reference simulation it’s partly in the southern hemisphere already on July 15th (Figure 4).

The cross-equatorial transport is slower in the experiment with low burden. The outer edges of the cloud cross the equator in August 1991 and diffuse outside the tropical area starting from October 1991, compared to the end of June 1991 and August 1991, respectively, of the reference simulation.

A lower injected sulfate burden generates a less intense perturbation, which, however, eventually produces the same transport pattern as in the reference case, when the SO₂ is injected at a higher altitude. Hence, the perturbation induced by the injection
of 5 Tg of sulfate is not strong enough to bring the cloud to the middle stratosphere but, if
directly injected at that altitude, such perturbation is sufficient to push the cloud south of
the equator and to midlatitudes.

4.3. Seasonality of the cross-equatorial transport

The cross-equatorial transport is related to the season of the eruption. We performed two
experiments injecting 20 Tg of SO$_2$ in winter (January 15$^{th}$) and in spring (April 4$^{th}$, day
of the 1982 eruption of El Chichón). The setup of the experiments was otherwise
identical to the reference simulation.

In the first experiment the volcanic cloud stays mainly in the northern
hemisphere, even if reaches the same altitude as in the reference experiment. The peak of
SO$_4$ concentration is in the northern hemisphere between 10$^\circ$N and 30$^\circ$N in February, and
during the following four months the cloud extends across the equator. The middle
stratospheric pathway is present also in this simulation, but is directed towards northern
high latitudes. It is not until June that a small amount of sulfate crosses 30$^\circ$S and reaches
the southern midlatitudes through the same middle stratospheric pathway.

In the springtime injection experiment the peak of sulfate aerosol also stays in the
northern hemisphere, between 0 and 10$^\circ$N. The edge of the cloud crosses the equator
already during the first month, and start spreading to midlatitudes in June. As in the
reference simulation, a considerable fraction of the cloud is directed to southern
midlatitudes through the middle stratospheric pathway. The seasonality of the mixing
alone, therefore, does not appear to be responsible for the different transport pattern of
the El Chichón and Mt. Pinatubo volcanic cloud.
5. Conclusions

Our GEOS-5 simulations of the transport of the volcanic cloud from the Mt. Pinatubo’s eruption are in good agreement with observations. Our simulations show that including interaction between radiation and volcanic SO$_4$ is essential to properly simulate the impact of volcanoes on the atmospheric circulation, as initially suggested by Young et al., [1994] and Fairlie, [1995].

The aerosol volcanic cloud diffuses across the globe through two main pathways: one pathway is centered in the lower stratosphere, at about 100 hPa, while the other is centered in the middle stratosphere. The volcanic cloud of Mt. Pinatubo diffuses to the northern hemisphere mainly in the lower stratosphere and to the southern hemisphere in the middle stratosphere.

We can divide the transport problem of the Pinatubo aerosol to the southern hemisphere in two stages: During the first stage, the absorption of longwave radiation from the cloud induces a lofting and a divergent motion from the center of the cloud. The self-induced transport of the cloud pushes the aerosol northward and southwards across the equator and to the tropics. The magnitude of the perturbation of the vertical velocity is closely related to the distribution of volcanic SO$_4$ and causes significant perturbations of the tropical circulation until December 1991.

The second stage, starting from about one month after the eruption, includes the transport from 30°S to southern mid- and high latitudes. Such transport takes place through tongue-like structures, which are the common way of mixing between the tropics and midlatitudes [Randel et al., 1993]. Analyzing the horizontal distribution of N$_2$O, we
could not detect any significant enhancement of the mixing between tropics and midlatitudes.

The transport across the equator is strongly dependent on the season and is much enhanced during the southern hemispheric winter. The seasonal dependence of the transport, however, does not appear to be responsible for the different transport pattern of the volcanic cloud from Mt. Pinatubo and El Chichón. Our simulations suggest that the different transport might be rather related to the amount of SO$_2$ injected in the atmosphere. An injected burden equal to 5 Tg SO$_2$ is not sufficient in our simulation to take the volcanic cloud to the middle stratosphere. El Chichón injected about 7 Tg of SO$_2$ in the atmosphere [Bluth et al., 1992], which might as well have not been enough to induce lofting to the middle stratosphere.

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Figure captions

Figure 1: Global mean of the visible aerosol optical thickness as simulated by GEOS-5 and as derived by SAGE II and AVHRR data. Background values have been removed from the AVHRR data. The shaded area shows the variability of the ensemble.

Figure 2: Zonal mean of the aerosol optical thickness at 550 nm for the Mt. Pinatubo eruption in the GEOS-5 simulations, SAGE-II and AVHRR observations. Background values have been removed from the AVHRR observations.

Figure 3: Vertical profile of the monthly average of the SO$_2$ mixing ratio in the latitudinal band between 10°S and 10°N. The black solid line represents the ensemble average and the shaded area the variability of the ensemble. The diamonds are MLS measurements by [Read et al., 1993].

Figure 4: Zonal mean of the SO$_4$ concentration from Mt. Pinatubo on July 15th, September 1st, November 1st and December 31st, 1991.

Figure 5: Horizontal distribution of SO$_4$ column mass between 30 hPa and the top of the atmosphere on July 2nd, 1991 and on July 16th, 1991.

Figure 6: Temporal evolution of the zonally averaged aerosol optical thickness at 550 nm in the ensembles with no radiatively interactive aerosol and SO$_2$ injection height between 16 and 18 km (upper panel) and 17 and 27 km (lower panel).
Figure 7: December 1991 monthly mean of the zonally averaged SO$_4$ concentration in the reference simulation (left panel), in the non interactive simulation with SO$_2$ injection height between 16 km and 18 km (middle panel) and in the non interactive simulation with SO$_2$ injection height between 17 km and 27 km (right panel).

Figure 8: Streamlines of the difference between the horizontal wind field in the interactive and in the non-interactive simulation on June 16$^{th}$, 1991 (upper panel) and on July 1$^{st}$, 1991 (lower panel) at 70 hPa and 30 hPa altitude, respectively. The shaded areas show the heating rates of sulfate from the eruption of Mt. Pinatubo due to the interaction with longwave radiation.

Figure 9: Horizontal distribution of the SO$_4$ concentration in the reference simulation and streamlines of the difference of the horizontal wind between the reference simulation and the simulation without interactive aerosol at 30 hPa (upper panel) and 100 hPa (lower panel) on June 24$^{th}$, 1991.

Figure 10: Difference of the vertical velocity in mm/s between interactive and non-interactive case at 70 hPa on June 16$^{th}$, 1991 and at 30 hPa on July 1$^{st}$, 1991 (shaded areas). The contours mark the concentration of SO$_4$, in $\mu$g/m$^3$. The average is calculated over 5 ensemble members, since the needed diagnostic was not available for all the eight ensemble members.

Figure 11: Zonal mean of the SO$_4$ concentration on October 15$^{th}$, 1991 in the simulations with low volcanic burden. In these experiments we injected 5 Tg of SO$_2$ between 16 and 18 km (left panel) and between 17 and 27 km (right panel), at the
same time and location of the Mt. Pinatubo eruption.
<table>
<thead>
<tr>
<th>Ensemble member</th>
<th>$e$-folding time</th>
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<tr>
<td></td>
<td>SO$_2$</td>
</tr>
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<td>29.8</td>
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<tr>
<td><strong>Standard deviation</strong></td>
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</table>

Table 1: SO$_2$ and SO$_4$ $e$-folding time of each ensemble member included in this study, and average values and standard deviation of the ensemble.
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SO$_4$ concentration and perturbation of the horizontal wind

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Anomaly of the vertical velocity

16 June 1991 - 70 hPa

1 July 1991 - 30 hPa

Figure 10: Difference of the vertical velocity in mm/s between interactive and non-interactive case at 70 hPa on June 16th, 1991 and at 30 hPa on July 1st, 1991 (shaded areas). The contours mark the concentration of SO$_2$, in $\mu$g/m$^3$. The average is calculated over 5 ensemble members, since the needed diagnostic was not available for all the eight ensemble members.
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