

# 1 Dispersion of the volcanic sulfate cloud 2 from the Mount Pinatubo eruption

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10

11 **Abstract**

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

27 **1. Introduction**

28 Volcanic eruptions are a major source of stratospheric aerosol [*Deshler, 2008*]. Sulfur  
29 dioxide injected into the stratosphere by large eruptions is oxidized into sulfate aerosol  
30 and can increase the background aerosol mass by orders of magnitude. The induced  
31 perturbation of the stratospheric aerosol layer can persist for some years. During such

32 time the aerosol can spread over the whole globe, changing the global climate in a  
33 significant way [*Robock, 2000*].

34 Mt. Pinatubo is located in the Philippines (15.1°N, 120.4°E). Pinatubo erupted on  
35 June 15<sup>th</sup>, 1991, injecting about 20 Tg of sulfur dioxide into the atmosphere [*Bluth et al.,*  
36 1992]. The resulting sulfate cloud was detected at altitudes higher than 30 km  
37 [*McCormick and Veiga, 1992*] and, after about one year, roughly one third of the  
38 volcanic aerosol was still present in the atmosphere.

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

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

51 *Niemeier et al., [2009]* applied the most recent version of the MAECHAM5  
52 Hamburg climate model, coupled to an aerosol microphysical model, to the study of the  
53 Pinatubo eruption. Other studies, such as *Stenchikov et al., [1998]*, *Kirchner et al., [1999]*  
54 and *Thomas et al., [2009a; 2009b]* used prescribed aerosol distributions.

55           It is still unclear if the eruption of Mt. Pinatubo modified the circulation in the  
56 southern hemisphere. *Robock et al.*, [2007] identified no significant anomaly in the  
57 southern hemisphere circulation in their simulations with the NASA/GISS ModelE  
58 general circulation model. In contrast, *Karpechko et al.*, [2010], *Marshall*, [2003], *Roscoe*  
59 *and Haigh*, [2007] and *Crooks and Gray*, [2005] found a negative response of the  
60 Southern Annular Mode in both models and observations.

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

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

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

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

## 76 2. The GEOS-5 general circulation model

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78 All simulations presented in this study are performed with the Goddard Earth Observing  
79 System, Version 5 (GEOS-5) model [*Rienecker et al.*, 2008], a system of component  
80 models integrated using the Earth System Modeling Framework (ESMF).

81 The GEOS-5 atmospheric general circulation model (AGCM) is able to perform  
82 weather and climate simulations used for atmospheric analyses, weather forecasts and  
83 climate simulations and predictions. GEOS-5 uses a finite-volume dynamical core [*Lin*,  
84 2004] combined with a physics package that describes moist processes, radiation,  
85 turbulent mixing and surface processes.

86 The convective parameterization Relaxed Arakawa-Schubert (RAS) is described  
87 by *Moorthi and Suarez*, [1992], and is combined to a prognostic cloud scheme. The  
88 boundary-layer turbulent mixing is parameterized with the schemes by *Louis et al.*,  
89 [1982] and *Lock et al.*, [2000], for stable and unstable situations, respectively. The land-  
90 surface model is composed of a catchment-based hydrological model [*Koster et al.*, 2000]  
91 and by a multi-layer snow model [*Stieglitz et al.*, 2001]. Coupled chemistry-climate  
92 simulations can be performed using the StratChem module for stratospheric chemistry  
93 [*Pawson et al.*, 2008].

94 The radiative transfer model consists of a solar radiation model [*Chou and*  
95 *Suarez*, 1999] and a thermal radiation model [*Chou et al.*, 2001]. The solar radiation  
96 model includes absorption due to water vapor, O<sub>3</sub>, O<sub>2</sub>, CO<sub>2</sub>, clouds and aerosol. The  
97 thermal radiation model includes absorption by water vapor, CO<sub>2</sub>, O<sub>3</sub> and most of the  
98 minor trace gases, as well as clouds and aerosol.

99           The aerosol optical properties are read from look-up tables previously generated  
100 using the OPAC database [*Hess et al.*, 1998]. The look-up tables contain the aerosol mass  
101 scattering and extinction coefficients as a function of relative humidity and radiation  
102 wavelength. We apply the Mie theory to calculate of the aerosol optical properties, and  
103 assume that aerosol is log-normally distributed and externally mixed.

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

109           The aerosol transport model in the GEOS-5 AGCM is based on the Goddard  
110 Chemistry, Aerosol, Radiation and Transport (GOCART) model [*Chin et al.*, 2000;  
111 2002]. An online version of GOCART in GEOS-4, a previous version of GEOS-5, has  
112 been validated by *Colarco et al.*, [2010]. Versions of GOCART in GEOS-5 have been  
113 already used in several recent field campaigns, as TC4 (2007), ARCTAS (2008) and  
114 GloPac (2010).

115           The aerosol species treated by GOCART as described in *Colarco et al.*, [2010]  
116 are dust, sea salt, black carbon, organic carbon and sulfate ( $\text{SO}_4$ ). In this study, only the  
117 sulfate component is active. GOCART includes a parameterization of the chemical  
118 production of  $\text{SO}_4$  from oxidation of dimethyl sulfide (DMS) by OH during day and  $\text{NO}_3$   
119 during night, and from oxidation of sulfur dioxide ( $\text{SO}_2$ ) by OH in the gas phase and by  
120  $\text{H}_2\text{O}_2$  in the aqueous phase.

121 GEOS-5 can run with radiatively interactive aerosol, which means that the aerosol  
122 concentrations simulated by GOCART can modify the meteorological fields. The  
123 simulations shown in Section 4 are performed with radiatively interactive aerosol. Some  
124 results from runs with non-interactive aerosol are presented in Section 4.

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

$$129 \quad \beta_{\text{SO}_4} = \frac{r_{\text{wet}}}{r_{\text{dry}}} = \sqrt[3]{\frac{RH(1-k)-1}{RH-1}},$$

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

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

$$134 \quad r_e = r_m \exp\left[\frac{5}{2} \ln^2 \sigma\right],$$

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

136 We performed several sensitivity tests varying the value of the sulfate dry radius.  
137 In this work we assume that aerosol is lognormal distributed with median diameter radius  
138 to  $0.35 \mu\text{m}$  and standard deviation 1.25. This corresponds to an effective dry radius equal  
139 to  $0.40 \mu\text{m}$ . This modal radius is within the range of observed values for sulfate aerosol  
140 from Mt. Pinatubo (e.g. *Bingen et al., [2004]*; *Niemeier et al., [2009]*; *Russell et al.,*

141 [1996]; *Stenchikov et al.*, [1998]), and result in good agreement with the AOT retrieved  
142 by SAGE-II and AVHRR (Section 3). The simulated *e*-folding time for sulfate is about  
143 one year (Table 1), as the one calculated from observations [*McCormick et al.*, 1995].

### 144 3. Simulation of the Mt. Pinatubo eruption

145 We simulated the eruption of Mt. Pinatubo by injecting 20 Tg of sulfur dioxide in the  
146 grid box containing Mt. Pinatubo during the day of June 15<sup>th</sup>, 1991. The SO<sub>2</sub> load is  
147 initially distributed between 16 km and 18 km, and is lofted to higher altitudes within the  
148 first weeks due to the model response to radiatively interacting aerosol.

149 Other model studies, as e.g. [*Timmreck et al.*, 1999b] and [*Zhao et al.*, 1995],  
150 place the injection of SO<sub>2</sub> at higher altitude. They base their assumption on SAGE-II  
151 observations. At the moment of the eruption, however, SAGE-II was observing at about  
152 70°N [*Trepte et al.*, 1993], and observed at the latitude of Mt. Pinatubo only 15 days after  
153 the eruption. At that stage the absorption of radiation by the volcanic aerosol had already  
154 induce the lofting of the cloud itself.

155 We tested similar assumptions on the injection height in GEOS-5 by performing  
156 simulations with injection of SO<sub>2</sub> between 16 km and 25 km, 17 km and 27 km, 20 km  
157 and 27 km, 20 km and 30 km. In all these simulations, the bulk of the volcanic cloud  
158 reached altitudes much higher than observations. Our choice of a lower injection altitude  
159 results in a reasonable simulation of the SAGE-II vertical profile after a couple of weeks  
160 from the day of the eruption (Section 3.2).

161 We did not include any other aerosol sources in the simulations used for this  
162 work.

163           We performed an ensemble of eight transient simulations, each spanning from  
164 January 1991 to December 1997. The initial conditions of the ensemble members are the  
165 meteorological fields of eight different Januaries of a control simulation with no volcanic  
166 perturbation, which was initialized with climatological meteorological fields typical of  
167 the year 2000.

168           The injected SO<sub>2</sub> is transformed into SO<sub>4</sub> by GOCART with an average *e*-folding  
169 time of 29.8 days, in good agreement with observations by the Total Ozone Mapping  
170 Spectrometer (TOMS) [Bluth *et al.*, 1992; Guo *et al.*, 2004]. The average of the SO<sub>4</sub> *e*-  
171 folding times of the eight ensemble members is 347 days with a standard deviation of  
172 57.7 days, also in good agreement with observations [Barnes and Hofmann, 1997; Nagai  
173 *et al.*, 2010]. Table 1 shows the *e*-folding times of SO<sub>2</sub> and SO<sub>4</sub> for each ensemble  
174 member.

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

180           The simulated peak value is in reasonable agreement with AVHRR, but is higher  
181 in magnitude and occur earlier in time than the SAGE-II data. Optical depths of about  
182 0.15 or more, however, saturate the SAGE-II measurement [Russell *et al.*, 1996]. Hence,  
183 the value of AOT calculated from SAGE-II observations are likely underestimated.  
184 Additionally, the sampling of SAGE-II observations is relatively sparse and can hardly

185 register rapid changes in the AOT. After January 1992 both SAGE-II and AVHRR  
186 observations are within the variability of the ensemble.

### 187       **3.1. Horizontal dispersion of the volcanic cloud**

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

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

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

201       GEOS-5 transports a large fraction of the cloud southward shortly after the  
202 eruption, but slightly underestimates the transport across the equator with respect to the  
203 observations. While the simulated peak is located on the equator, both SAGE-II and  
204 AVHRR detected the peak at about 5°S. The results by [Timmreck *et al.*, 1999a] also  
205 underestimated the cross-equatorial transport. They suggested that the missing transport

206 might be due to the specific synoptic situation in June 1991, when a strong high over  
207 Tibet induced a southward transport of the cloud.

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

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

217 The small initial underestimation of the southward transport might be due to the  
218 lack of a radiatively interactive SO<sub>2</sub> in the version of GEOS-5 used in this work. *Lary et*  
219 *al.*, [1994] estimated that the SO<sub>2</sub> heating rate can be up to 1 K/day and could therefore  
220 be significant in the early stages of the cloud's evolution.

### 221 3.2. Vertical distribution of the volcanic cloud

222 We compare our results with SO<sub>2</sub> profiles taken with a microwave limb sounder (MLS)  
223 by *Read et al.*, [1993] between 10°S and the equator on September 21<sup>st</sup>, 1991 (Figure 3).  
224 The simulated profiles correspond to the September 1991 monthly mean of the SO<sub>2</sub>  
225 vertical profiles, averaged over the latitudinal band between 10°S and 10°N. We averaged  
226 over a latitudinal band larger than the MLS observations to take into account the different  
227 transport pattern between simulations and observations.

228           The agreement with [Read *et al.*, 1993] is good: both SO<sub>2</sub> profiles have a peak at  
229    about 20 hPa of similar magnitude. The sensitivity tests that we performed varying the  
230    injection altitude of SO<sub>2</sub> showed differences in the vertical profile of the volcanic cloud  
231    during the first months, but the equilibrium level where the bulk of the cloud settles was  
232    in all tests at about 20 hPa.

233           Figure 4 shows the vertical distribution of the zonally averaged SO<sub>4</sub> concentration  
234    on July 15<sup>th</sup>, September 1<sup>st</sup>, November 1<sup>st</sup> and December 31<sup>st</sup>, 1991. The bulk of the cloud  
235    is between 50 hPa and 10 hPa in July 1991.

236           The model results are in agreement with SAGE-II observational satellite data,  
237    which detected the cloud top at altitudes up to 29 km (about 10 hPa) during June, July  
238    and August 1991 [McCormick and Veiga, 1992].

239           Trepte *et al.*, [1993] showed the latitude-altitude cross-section of the SAGE-II 1  
240    μm extinction ratio. Data were first collected in the tropical region between July 1<sup>st</sup> and  
241    July 20<sup>th</sup>, and show values higher than the background between the tropopause and 30 km  
242    altitude, in reasonable agreement with the first panel of Figure 4.

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

### 246       **3.3. Cross-equatorial transport**

247    The volcanic cloud moves to mid- and high latitudes through two main transport  
248    pathways, as shown in Figure 4. Already one week after the eruption, part of the cloud is

249 advected northward through the lower stratosphere at about 100 hPa. A portion of the  
250 cloud, instead, later reaches southern higher latitudes through the middle stratosphere  
251 between 5 and 50 hPa and arrives at 90°S in the middle of November (Figure 4, lower  
252 panels).

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

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

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

270         Our transport simulation of the volcanic cloud from the Mt. Pinatubo eruption is  
271 in good agreement with the observations. Both the vertical and horizontal distribution and

272 the timing of the mixing to mid- and high latitudes are reasonably well comparable to  
273 SAGE-II and AVHRR observations.

#### 274 **4. Importance of a radiative active volcanic aerosol**

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

282 Figure 6 (upper panel) shows the temporal evolution of the zonally averaged AOT  
283 at 550 nm, to be compared to the upper panel of Figure 2. In the non-interactive  
284 ensemble, most of the volcanic cloud is directed toward the northern hemisphere, faster  
285 than in the reference simulation. This is due to the different vertical distribution of the  
286 volcanic cloud: in the non-interactive case the volcanic cloud stays at much lower  
287 altitudes (Figure 7, middle panel) than in the interactive case (Figure 7, left panel).  
288 Hence, the non-interactive cloud does not rise enough to enter the middle stratosphere,  
289 and the advection of the cloud to midlatitudes takes place only through the lower  
290 stratosphere. The *e*-folding time of SO<sub>4</sub> is much lower in the ensemble with non-  
291 interactive aerosol than in the reference case (74 days against 346 days).

292 We also performed an ensemble of three non-interactive simulations directly  
293 injecting SO<sub>2</sub> between 17 km - 27 km (Figure 6, lower panel). Also in this case the cross-

294 equatorial transport is not as intense as in the reference simulation, and the volcanic cloud  
295 looks even more confined to the tropics than in Figure 6a. Even if the SO<sub>4</sub> cloud reaches  
296 the middle stratosphere (Figure 7, right panel) and part of the cloud crosses the equator, it  
297 remains confined within the tropics.

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

#### 301         **4.1. Perturbation of the background winds**

302 In Figure 8 we show the perturbation of the horizontal wind fields induced by the  
303 interaction between radiation and volcanic aerosol. We show the difference of the  
304 horizontal winds between the interactive and the non-interactive ensembles on June 16<sup>th</sup>,  
305 1991 at 70 hPa and on July 1<sup>st</sup>, 1991, together with the aerosol heating rates due to  
306 longwave radiation. To reduce noise effects, the results for July 1<sup>st</sup> in Figure 8 and Figure  
307 10 are from an eight-member ensemble of non-interactive simulations starting on the  
308 midnight of July 1<sup>st</sup>, with initial conditions from July 1<sup>st</sup> of the interactive run.

309         The sudden warming generates a divergent motion from the location of the  
310 volcanic cloud already one day after the eruption (Figure 8, upper panel). The simulated  
311 volcanic cloud is still at the same latitude as Mt. Pinatubo, and has not risen yet to  
312 altitudes higher than 50 hPa. On July 1<sup>st</sup> (Figure 8, lower panel) the cloud has circled  
313 nearly around the whole globe, but, in the middle stratosphere, is still confined between  
314 20°S and 20°N.

315           The perturbation of the horizontal winds diffuses the sulfate northward and  
316 southward from the center of the clouds, increasing the spreading of the clouds towards  
317 the tropics, due to the heating induced by the SO<sub>4</sub> absorption of longwave radiation. The  
318 winds are no longer significantly perturbed at 30 hPa by December 1991, when the  
319 concentration of SO<sub>4</sub> becomes meridionally homogeneous. At 50 hPa, where the SO<sub>4</sub>  
320 concentration decreases (Figure 4), the winds converge towards the center of the cloud.  
321 At altitudes lower than 50 hPa no consistent perturbation is simulated.

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

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

335           The absorption of longwave radiation by volcanic SO<sub>4</sub> is responsible for the “self-  
336 lofting” of the volcanic clouds and for the divergent motion from the areas with highest  
337 SO<sub>4</sub> concentration. As already mentioned in Section 3.1, the introduction of radiative

338 interactive  $\text{SO}_2$  could possibly increase the lofting and spreading of the cloud during the  
339 first months from the eruption.

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

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

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

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

355 There is no sign of increased  $\text{N}_2\text{O}$  transport from the tropics to midlatitudes.

356 The analysis of the age of air at 30 hPa leads to similar conclusion (not shown).  
357 The air in the interactive runs is younger in the tropical area between July and December  
358 1991, implying a faster upwelling, but there is no significant difference in the following  
359 months, neither in the tropics nor at midlatitudes.

360       **4.2.     Impact of the sulfate burden on the spreading of the**  
361               **volcanic cloud**

362     We performed two interactive experiments lowering the amount of injected SO<sub>2</sub> to 5 Tg.  
363     In the first experiment we injected SO<sub>2</sub> between 16 km and 18 km, in the second between  
364     17 km and 27 km. The set up of the simulations is otherwise identical to the reference  
365     simulation. Figure 11 shows the vertical profiles of the zonal mean of SO<sub>4</sub> in the two  
366     experiments.

367             In the experiment with low injection height (Figure 11a) the volcanic cloud is  
368     mainly confined to the lower stratosphere, showing that 5 Tg of SO<sub>2</sub> do not produce a  
369     strong enough perturbation to raise the cloud to the middle stratosphere.

370             In the second experiment (Figure 11b) the cloud is injected already in the middle  
371     stratosphere and is advected to the southern hemisphere through the same middle  
372     stratospheric pathway as in the reference simulation. However, the peak of SO<sub>4</sub> is north  
373     of the equator, while in the reference simulation it's partly in the southern hemisphere  
374     already on July 15<sup>th</sup> (Figure 4).

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

379             A lower injected sulfate burden generates a less intense perturbation, which,  
380     however, eventually produces the same transport pattern as in the reference case, when  
381     the SO<sub>2</sub> is injected at a higher altitude. Hence, the perturbation induced by the injection

382 of 5 Tg of sulfate is not strong enough to bring the cloud to the middle stratosphere but, if  
383 directly injected at that altitude, such perturbation is sufficient to push the cloud south of  
384 the equator and to midlatitudes.

### 385 4.3. Seasonality of the cross-equatorial transport

386 The cross-equatorial transport is related to the season of the eruption. We performed two  
387 experiments injecting 20 Tg of SO<sub>2</sub> in winter (January 15<sup>th</sup>) and in spring (April 4<sup>th</sup>, day  
388 of the 1982 eruption of El Chichón). The setup of the experiments was otherwise  
389 identical to the reference simulation.

390 In the first experiment the volcanic cloud stays mainly in the northern  
391 hemisphere, even if reaches the same altitude as in the reference experiment. The peak of  
392 SO<sub>4</sub> concentration is in the northern hemisphere between 10°N and 30°N in February, and  
393 during the following four months the cloud extends across the equator. The middle  
394 stratospheric pathway is present also in this simulation, but is directed towards northern  
395 high latitudes. It is not until June that a small amount of sulfate crosses 30°S and reaches  
396 the southern midlatitudes through the same middle stratospheric pathway.

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

404 **5. Conclusions**

405 Our GEOS-5 simulations of the transport of the volcanic cloud from the Mt. Pinatubo's  
406 eruption are in good agreement with observations. Our simulations show that including  
407 interaction between radiation and volcanic SO<sub>4</sub> is essential to properly simulate the  
408 impact of volcanoes on the atmospheric circulation, as initially suggested by *Young et al.*,  
409 [1994] and *Fairlie*, [1995].

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

415 We can divide the transport problem of the Pinatubo aerosol to the southern  
416 hemisphere in two stages: During the first stage, the absorption of longwave radiation  
417 from the cloud induces a lofting and a divergent motion from the center of the cloud. The  
418 self-induced transport of the cloud pushes the aerosol northward and southwards across  
419 the equator and to the tropics. The magnitude of the perturbation of the vertical velocity  
420 is closely related to the distribution of volcanic SO<sub>4</sub> and causes significant perturbations  
421 of the tropical circulation until December 1991.

422 The second stage, starting from about one month after the eruption, includes the  
423 transport from 30°S to southern mid- and high latitudes. Such transport takes place  
424 through tongue-like structures, which are the common way of mixing between the tropics  
425 and midlatitudes [*Randel et al.*, 1993]. Analyzing the horizontal distribution of N<sub>2</sub>O, we

426 could not detect any significant enhancement of the mixing between tropics and  
427 midlatitudes.

428         The transport across the equator is strongly dependent on the season and is much  
429 enhanced during the southern hemispheric winter. The seasonal dependence of the  
430 transport, however, does not appear to be responsible for the different transport pattern of  
431 the volcanic cloud from Mt. Pinatubo and El Chichón. Our simulations suggest that the  
432 different transport might be rather related to the amount of SO<sub>2</sub> injected in the  
433 atmosphere. An injected burden equal to 5 Tg SO<sub>2</sub> is not sufficient in our simulation to  
434 take the volcanic cloud to the middle stratosphere. El Chichón injected about 7 Tg of SO<sub>2</sub>  
435 in the atmosphere [*Bluth et al.*, 1992], which might as well have not been enough to  
436 induce lofting to the middle stratosphere.

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441

441

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594

594 **Figure captions**

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

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

602 Figure 3: Vertical profile of the monthly average of the SO<sub>2</sub> mixing ratio in  
603 the latitudinal band between 10°S and 10°N. The black solid line represents the  
604 ensemble average and the shaded area the variability of the ensemble. The diamonds  
605 are MLS measurements by [Read *et al.*, 1993].

606 Figure 4: Zonal mean of the SO<sub>4</sub> concentration from Mt. Pinatubo on July  
607 15<sup>th</sup>, September 1<sup>st</sup>, November 1<sup>st</sup> and December 31<sup>st</sup>, 1991.

608 Figure 5: Horizontal distribution of SO<sub>4</sub> column mass between 30 hPa and the  
609 top of the atmosphere on July 2<sup>nd</sup>, 1991 and on July 16<sup>th</sup>, 1991.

610 Figure 6: Temporal evolution of the zonally averaged aerosol optical  
611 thickness at 550 nm in the ensembles with no radiatively interactive aerosol and SO<sub>2</sub>  
612 injection height between 16 and 18 km (upper panel) and 17 and 27 km (lower  
613 panel).

614           Figure 7: December 1991 monthly mean of the zonally averaged SO<sub>4</sub>  
615 concentration in the reference simulation (left panel), in the non interactive  
616 simulation with SO<sub>2</sub> injection height between 16 km and 18 km (middle panel) and in  
617 the non interactive simulation with SO<sub>2</sub> injection height between 17 km and 27 km  
618 (right panel).

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

624           Figure 9: Horizontal distribution of the SO<sub>4</sub> concentration in the reference  
625 simulation and streamlines of the difference of the horizontal wind between the  
626 reference simulation and the simulation without interactive aerosol at 30 hPa (upper  
627 panel) and 100 hPa (lower panel) on June 24<sup>th</sup>, 1991.

628           Figure 10: Difference of the vertical velocity in mm/s between interactive and  
629 non-interactive case at 70 hPa on June 16<sup>th</sup>, 1991 and at 30 hPa on July 1<sup>st</sup>, 1991  
630 (shaded areas). The contours mark the concentration of SO<sub>4</sub>, in µg/m<sup>3</sup>. The average is  
631 calculated over 5 ensemble members, since the needed diagnostic was not available  
632 for all the eight ensemble members.

633           Figure 11: Zonal mean of the SO<sub>4</sub> concentration on October 15<sup>th</sup>, 1991 in the  
634 simulations with low volcanic burden. In these experiments we injected 5 Tg of SO<sub>2</sub>  
635 between 16 and 18 km (left panel) and between 17 and 27 km (right panel), at the

636 same time and location of the Mt. Pinatubo eruption.

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637

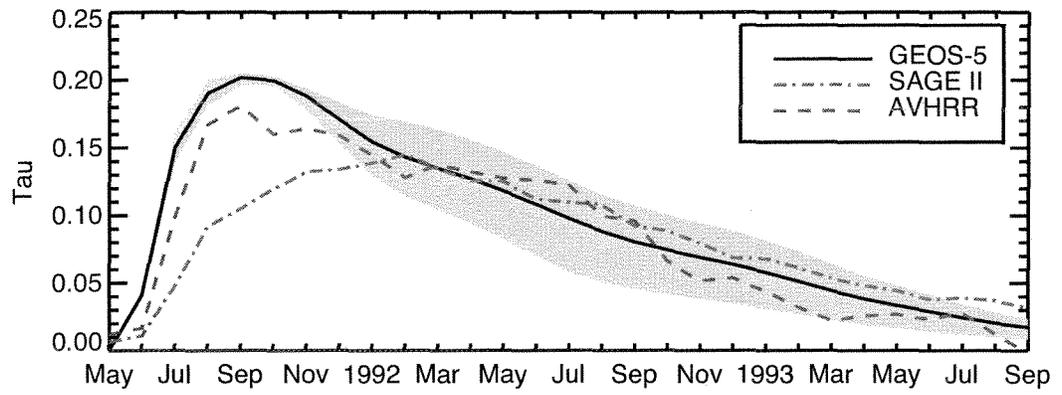
Ensemble member	<i>e</i> -folding time	
	SO <sub>2</sub>	SO <sub>4</sub>
1 <sup>st</sup>	31	373
2 <sup>nd</sup>	29	340
3 <sup>rd</sup>	26	243
4 <sup>th</sup>	34	426
5 <sup>th</sup>	29	372
6 <sup>th</sup>	29	345
7 <sup>th</sup>	26	275
8 <sup>th</sup>	34	402
Average	29.8	347
Standard deviation	2.9	57.7

638 Table 1: SO<sub>2</sub> and SO<sub>4</sub> *e*-folding time of each ensemble member included in

639 this study, and average values and standard deviation of the ensemble.

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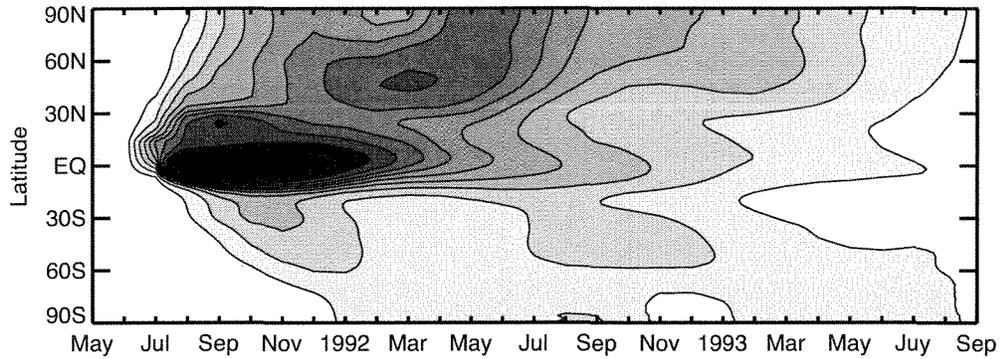


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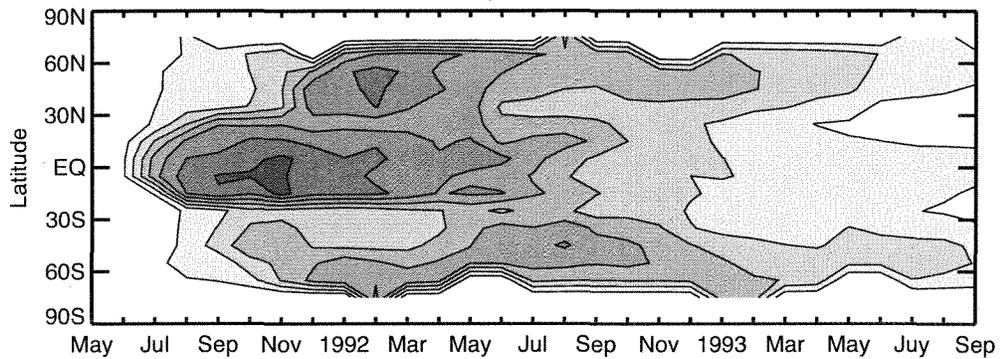
642 Figure 1: Global mean of the visible aerosol optical thickness as simulated by  
643 GEOS-5 and as derived by SAGE II and AVHRR data. Background values have  
644 been removed from the AVHRR data. The shaded area shows the variability of the  
645 ensemble.

# Zonal mean visible aerosol optical thickness

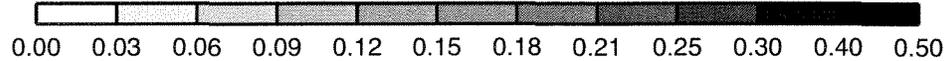
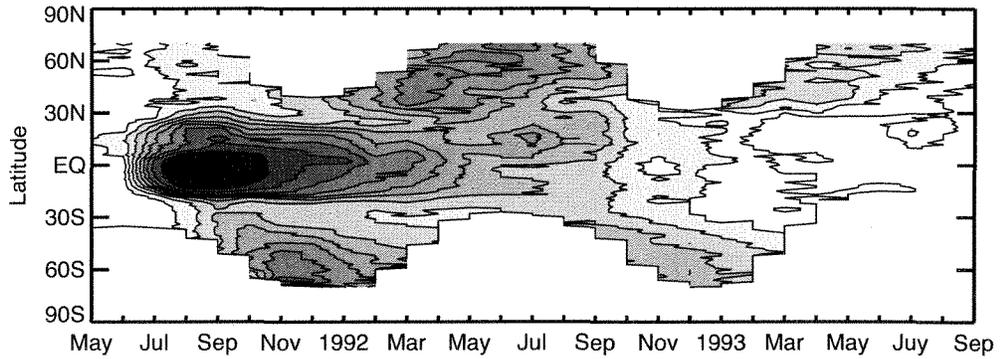
GEOS-5



SAGE-II



AVHRR



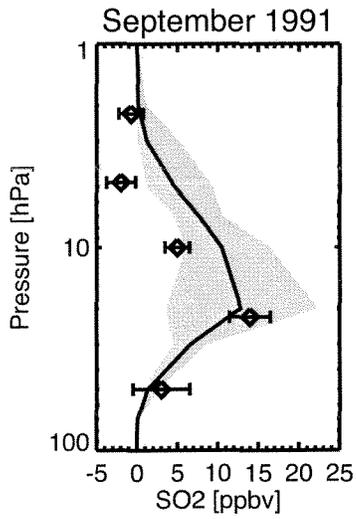
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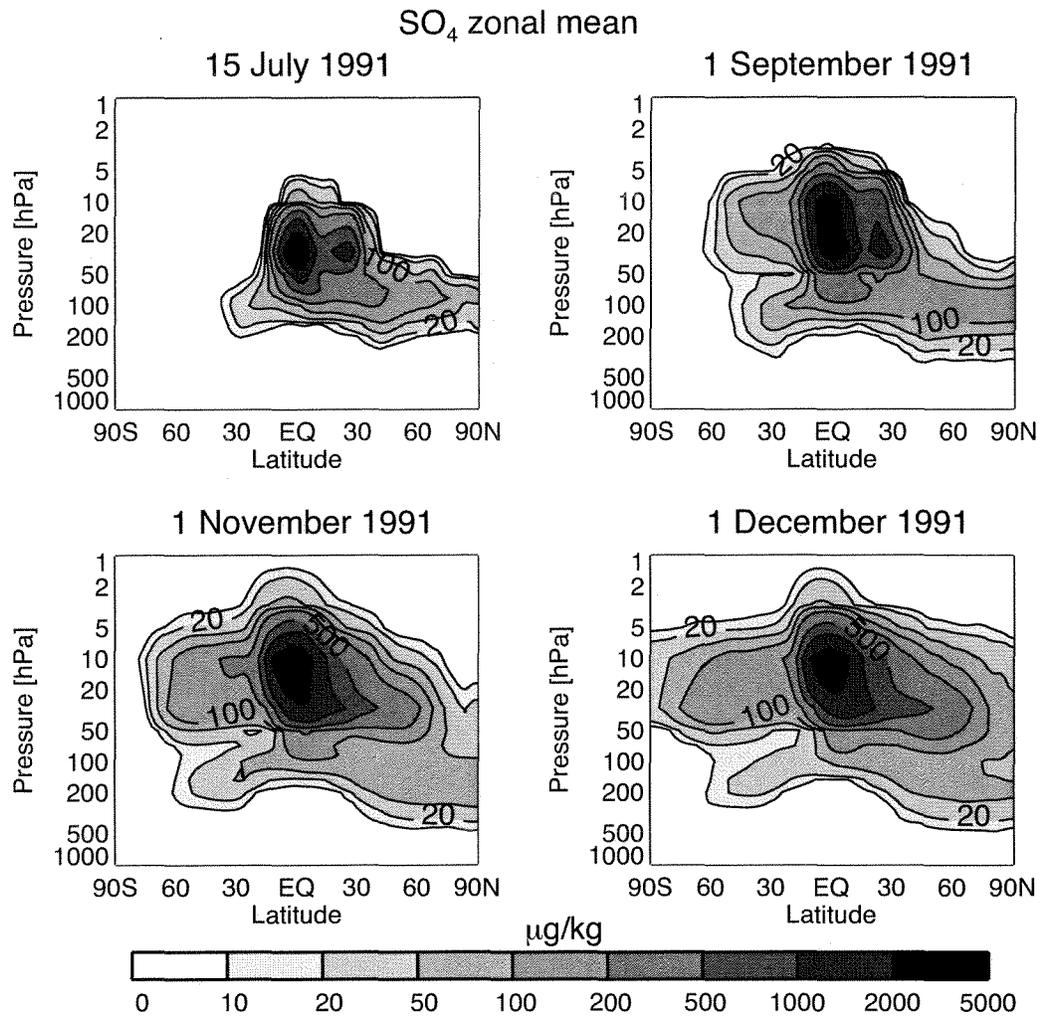
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Figure 2: Zonal mean of the aerosol optical thickness at 550 nm for the Mt. Pinatubo eruption in the GEOS-5 simulations, SAGE-II and AVHRR observations. Background values have been removed from the AVHRR observations.



650

651        Figure 3: Vertical profile of the monthly average of the SO<sub>2</sub> mixing ratio in  
 652 the latitudinal band between 10°S and 10°N. The black solid line represents the  
 653 ensemble average and the shaded area the variability of the ensemble. The  
 654 diamonds are MLS measurements by [Read *et al.*, 1993].



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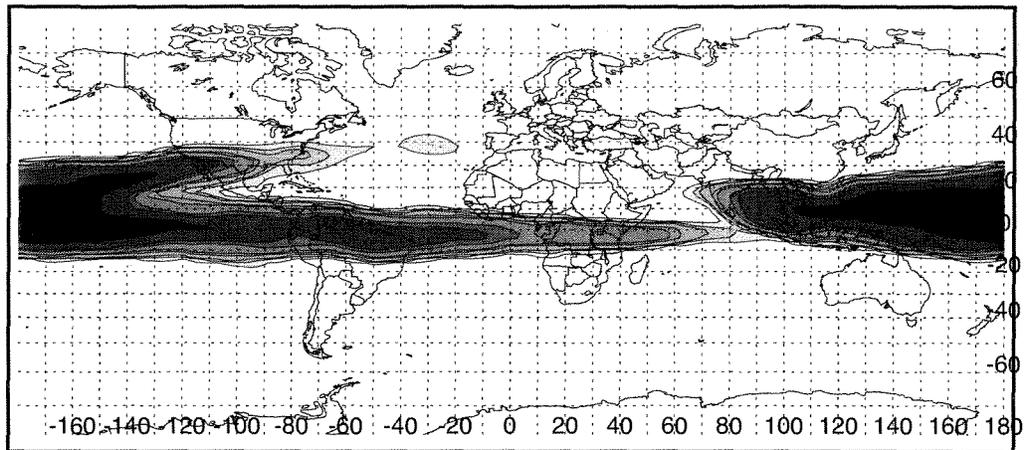
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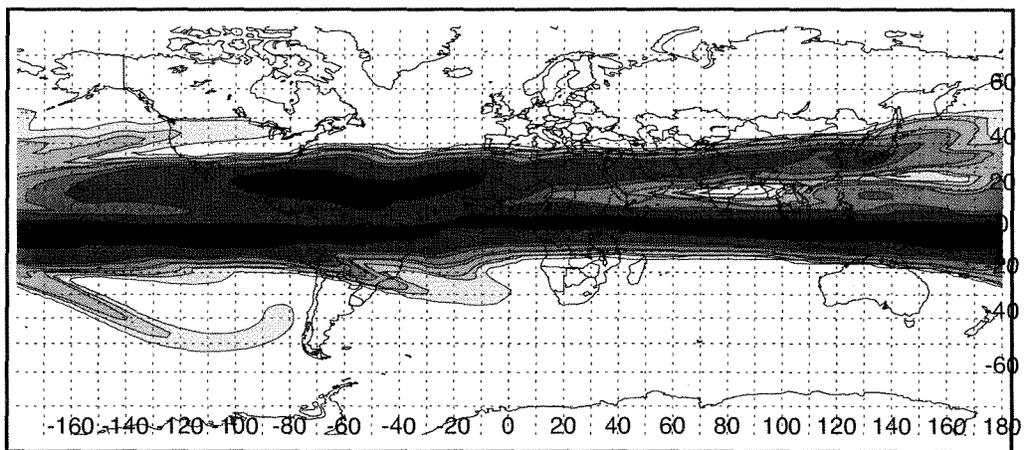
658

Figure 4: Zonal mean of the SO<sub>4</sub> concentration from Mt. Pinatubo on July 15<sup>th</sup>, September 1<sup>st</sup>, November 1<sup>st</sup> and December 31<sup>st</sup>, 1991.

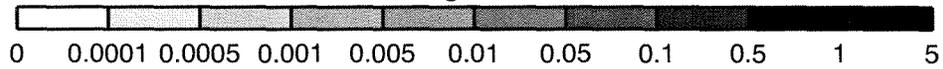
2 July 1991



16 July 1991



$\text{g/m}^2$

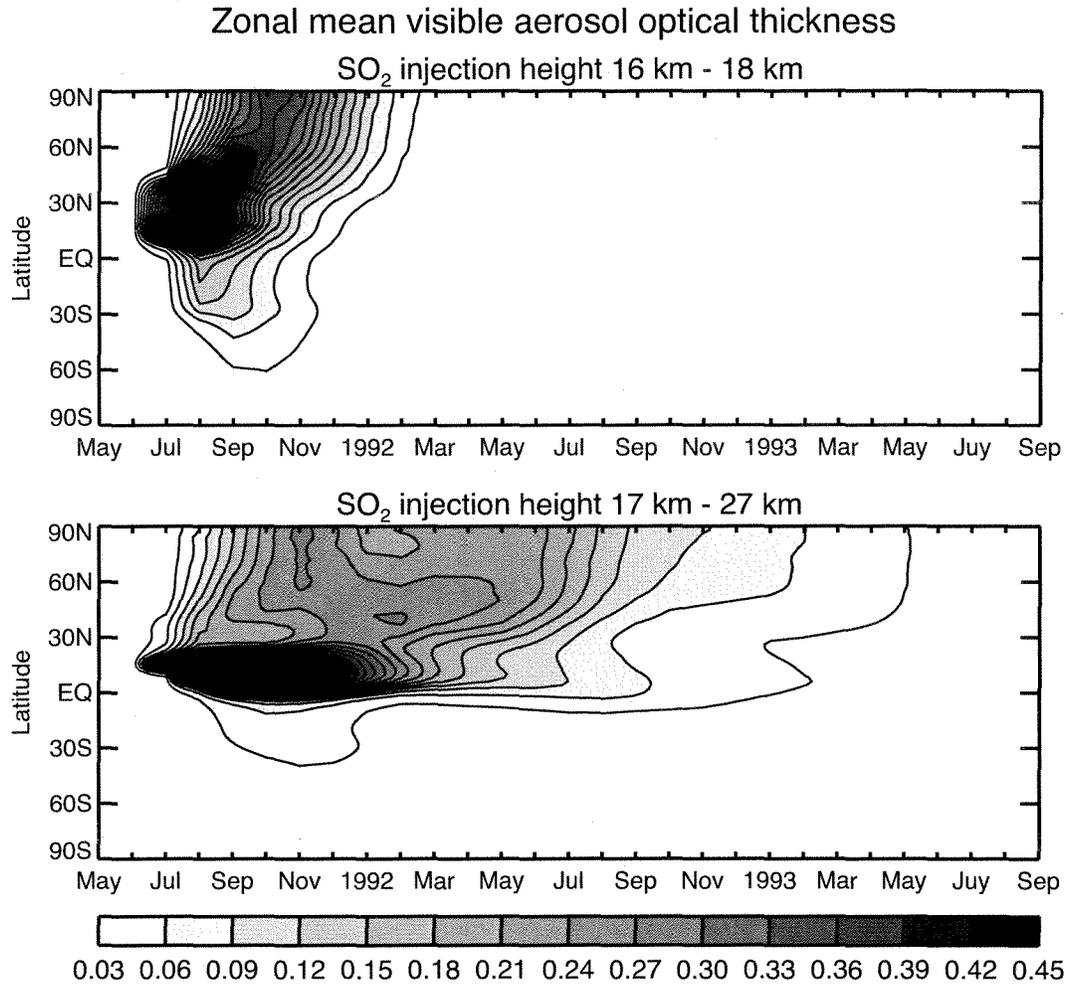


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Figure 5: Horizontal distribution of  $\text{SO}_2$  column mass between 30 hPa and the top of the atmosphere on July 2<sup>nd</sup>, 1991 and on July 16<sup>th</sup>, 1991.



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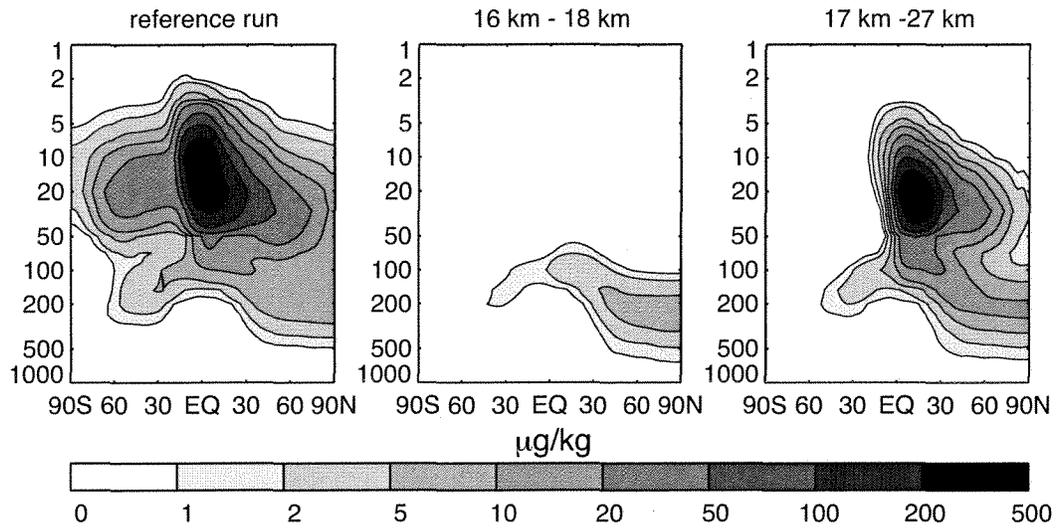
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Figure 6: Temporal evolution of the zonally averaged aerosol optical thickness at 550 nm in the ensembles with no radiatively interactive aerosol and SO<sub>2</sub> injection height between 16 and 18 km (upper panel) and 17 and 27 km (lower panel).

SO<sub>4</sub> zonal mean - December 1991



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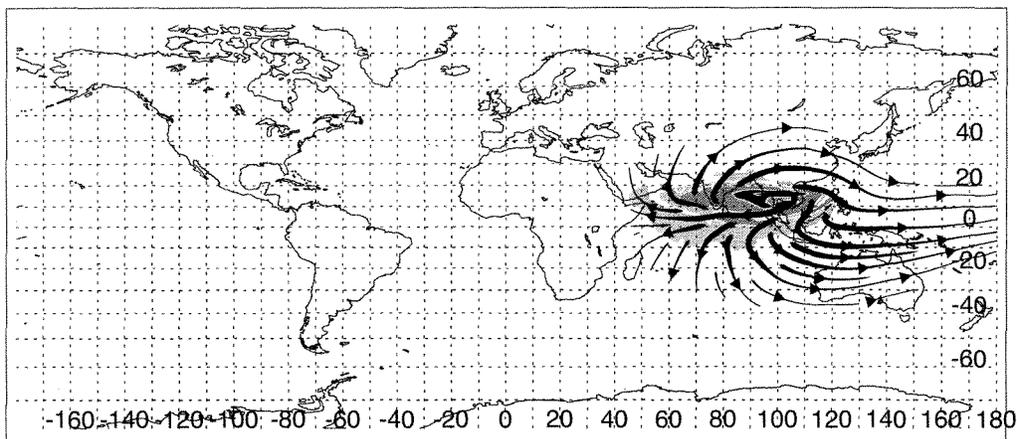
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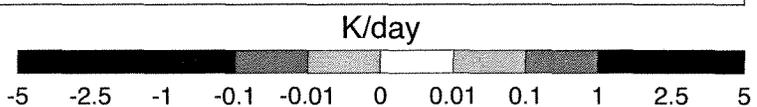
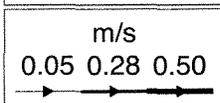
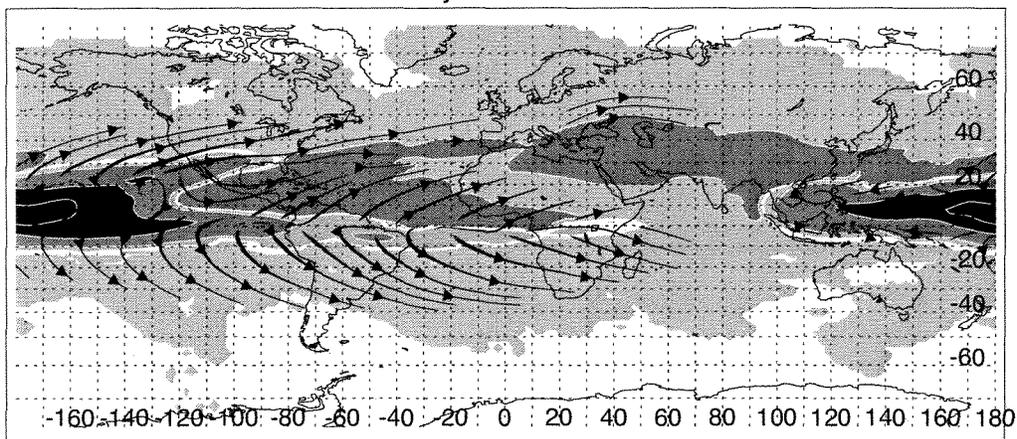
Figure 7: December 1991 monthly mean of the zonally averaged SO<sub>4</sub> concentration in the reference simulation (left panel), in the non interactive simulation with SO<sub>2</sub> injection height between 16 km and 18 km (middle panel) and in the non interactive simulation with SO<sub>2</sub> injection height between 17 km and 27 km (right panel).

## Longwave heating rates

16 June 1991 - 70 hPa



1 July 1991 - 30 hPa



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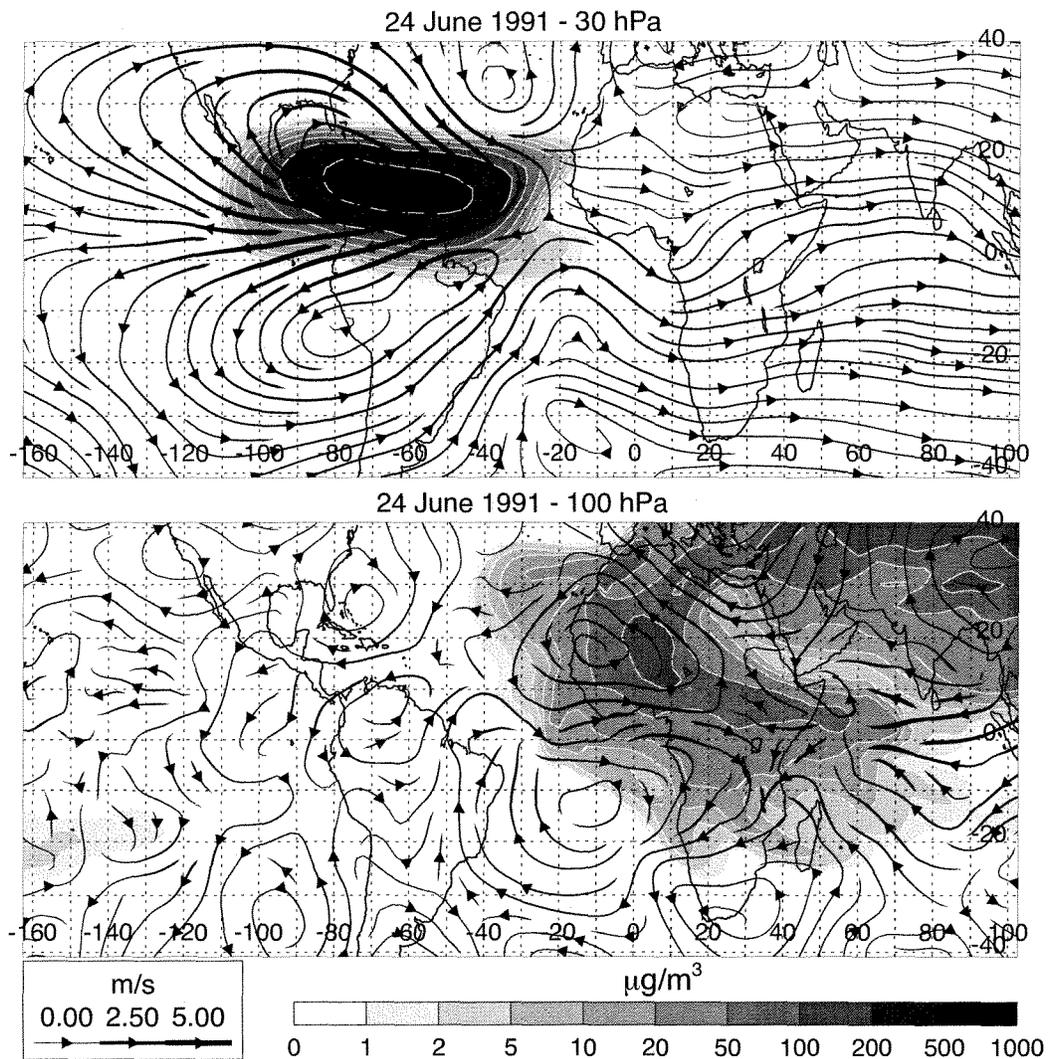
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Figure 8: Streamlines of the difference between the horizontal wind field in the interactive and in the non-interactive simulation on June 16<sup>th</sup>, 1991 (upper panel) and on July 1<sup>st</sup>, 1991 (lower panel) at 70 hPa and 30 hPa altitude,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

# SO<sub>4</sub> concentration and perturbation of the horizontal wind



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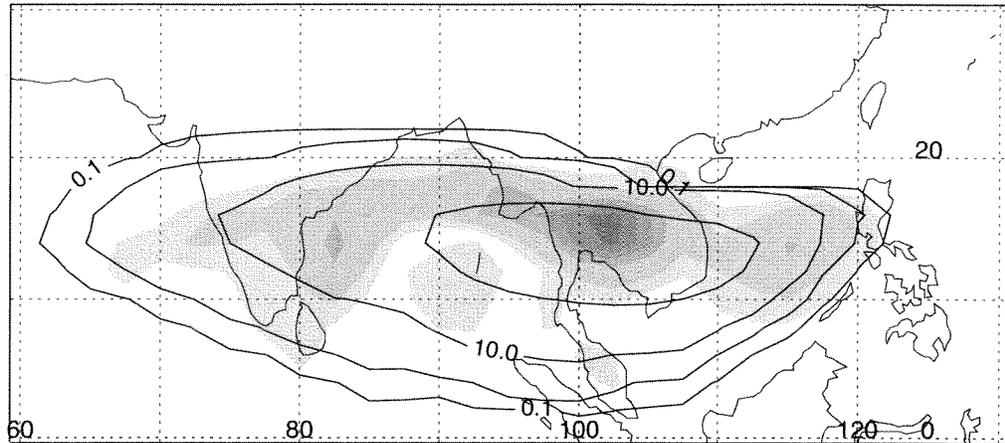
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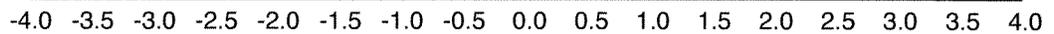
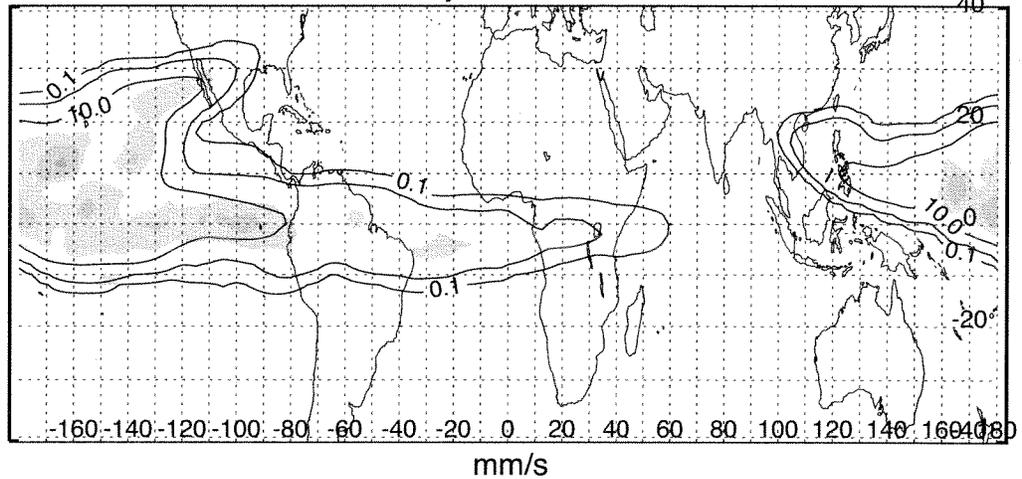
Figure 9: Horizontal distribution of the SO<sub>4</sub> concentration in the reference simulation and streamlines of the difference of the horizontal wind between the reference simulation and the simulation without interactive aerosol at 30 hPa (upper panel) and 100 hPa (lower panel) on June 24<sup>th</sup>, 1991.

# Anomaly of the vertical velocity

16 June 1991 - 70 hPa



1 July 1991 - 30 hPa



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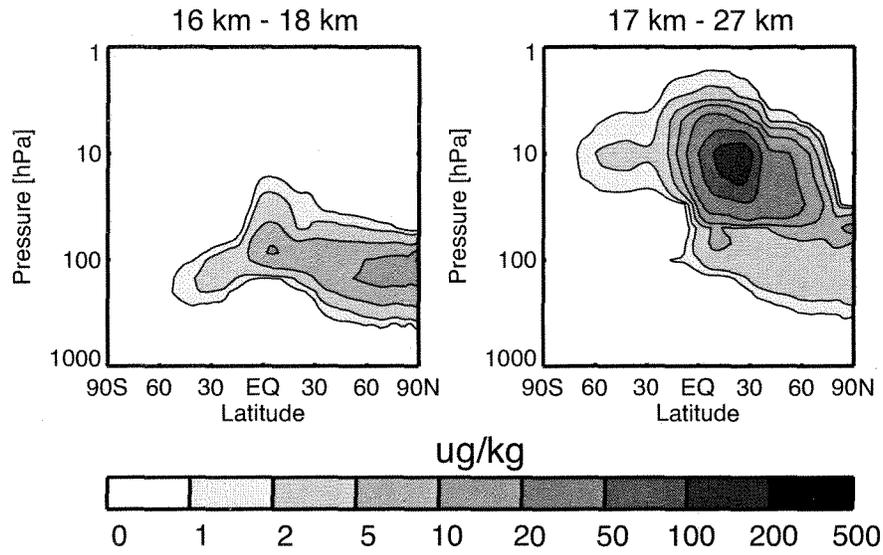
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Figure 10: Difference of the vertical velocity in mm/s between interactive and non-interactive case at 70 hPa on June 16<sup>th</sup>, 1991 and at 30 hPa on July 1<sup>st</sup>, 1991 (shaded areas). The contours mark the concentration of SO<sub>2</sub>, in μg/m<sup>3</sup>. The average is calculated over 5 ensemble members, since the needed diagnostic was not available for all the eight ensemble members.

### SO<sub>4</sub> zonal mean



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690 Figure 11: Zonal mean of the SO<sub>4</sub> concentration on October 15<sup>th</sup>, 1991 in the  
691 simulations with low volcanic burden. In these experiments we injected 5 Tg of SO<sub>2</sub>  
692 between 16 and 18 km (left panel) and between 17 and 27 km (right panel), at the  
693 same time and location of the Mt. Pinatubo eruption.