

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

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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 15th, 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, O₃, O₂, CO₂, clouds and aerosol. The
97 thermal radiation model includes absorption by water vapor, CO₂, O₃ 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°
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 (SO_4). In this study, only the
117 sulfate component is active. GOCART includes a parameterization of the chemical
118 production of SO_4 from oxidation of dimethyl sulfide (DMS) by OH during day and NO_3
119 during night, and from oxidation of sulfur dioxide (SO_2) by OH in the gas phase and by
120 H_2O_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 SO₄ to properly simulate
126 stratospheric volcanic aerosol. The settling velocity is a function of the particle's wet
127 radius. The sulfate growth factor β_{SO_4} is calculated as a function of the relative humidity
128 RH following *Petters and Kreidenweis, [2007]* as

$$129 \quad \beta_{SO_4} = \frac{r_{wet}}{r_{dry}} = \sqrt[3]{\frac{RH(1-k)-1}{RH-1}},$$

130 where the hygroscopic parameter k is equal to 1.19 and r_{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 σ 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 μm and standard deviation 1.25. This corresponds to an effective dry radius equal
139 to 0.40 μm . 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 15th, 1991. The SO₂ 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 SO₂ 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 SO₂ 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_2 is transformed into SO_4 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_4 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_2 and SO_4 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 SO₂ in the version of GEOS-5 used in this work. *Lary et*
219 *al.*, [1994] estimated that the SO₂ 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 SO₂ profiles taken with a microwave limb sounder (MLS)
223 by *Read et al.*, [1993] between 10°S and the equator on September 21st, 1991 (Figure 3).
224 The simulated profiles correspond to the September 1991 monthly mean of the SO₂
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 SO₂ profiles have a peak at
229 about 20 hPa of similar magnitude. The sensitivity tests that we performed varying the
230 injection altitude of SO₂ 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 SO₄ concentration
234 on July 15th, September 1st, November 1st and December 31st, 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 1st and
241 July 20th, 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 11th, 1991 and July 18th, 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 SO₄ 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 SO₂ 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 SO₄ 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 16th,
305 1991 at 70 hPa and on July 1st, 1991, together with the aerosol heating rates due to
306 longwave radiation. To reduce noise effects, the results for July 1st in Figure 8 and Figure
307 10 are from an eight-member ensemble of non-interactive simulations starting on the
308 midnight of July 1st, with initial conditions from July 1st 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 1st (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 SO₄ absorption of longwave radiation. The
318 winds are no longer significantly perturbed at 30 hPa by December 1991, when the
319 concentration of SO₄ becomes meridionally homogeneous. At 50 hPa, where the SO₄
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 SO₄ 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 SO₄ is responsible for the “self-
336 lofting” of the volcanic clouds and for the divergent motion from the areas with highest
337 SO₄ concentration. As already mentioned in Section 3.1, the introduction of radiative

338 interactive SO₂ 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 N₂O (not shown).

347 Climatologically, the concentration of N₂O is highest in the tropics and presents a
348 strong summer gradient between the tropics and midlatitudes. The sources of N₂O are
349 located at surface and its concentration decreases with altitude.

350 Compared to the unperturbed case, GEOS-5 simulates decreased N₂O at about 30
351 hPa and increased N₂O 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 N₂O
354 concentration can be detected after September 1992.

355 There is no sign of increased N₂O 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 SO₂ to 5 Tg.
363 In the first experiment we injected SO₂ 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 SO₄ 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 SO₂ 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 SO₄ is north
373 of the equator, while in the reference simulation it's partly in the southern hemisphere
374 already on July 15th (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 SO₂ 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 SO₂ in winter (January 15th) and in spring (April 4th, 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 SO₄ 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 SO₄ 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 SO₄ 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 N₂O, 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 SO₂ injected in the
433 atmosphere. An injected burden equal to 5 Tg SO₂ is not sufficient in our simulation to
434 take the volcanic cloud to the middle stratosphere. El Chichón injected about 7 Tg of SO₂
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 **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.
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602 Figure 3: Vertical profile of the monthly average of the SO₂ mixing ratio in
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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 SO₄ concentration from Mt. Pinatubo on July
607 15th, September 1st, November 1st and December 31st, 1991.

608 Figure 5: Horizontal distribution of SO₄ column mass between 30 hPa and the
609 top of the atmosphere on July 2nd, 1991 and on July 16th, 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 SO₂
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 SO₄
615 concentration in the reference simulation (left panel), in the non interactive
616 simulation with SO₂ injection height between 16 km and 18 km (middle panel) and in
617 the non interactive simulation with SO₂ 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 16th, 1991 (upper panel)
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624 Figure 9: Horizontal distribution of the SO₄ concentration in the reference
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626 reference simulation and the simulation without interactive aerosol at 30 hPa (upper
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628 Figure 10: Difference of the vertical velocity in mm/s between interactive and
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636 same time and location of the Mt. Pinatubo eruption.

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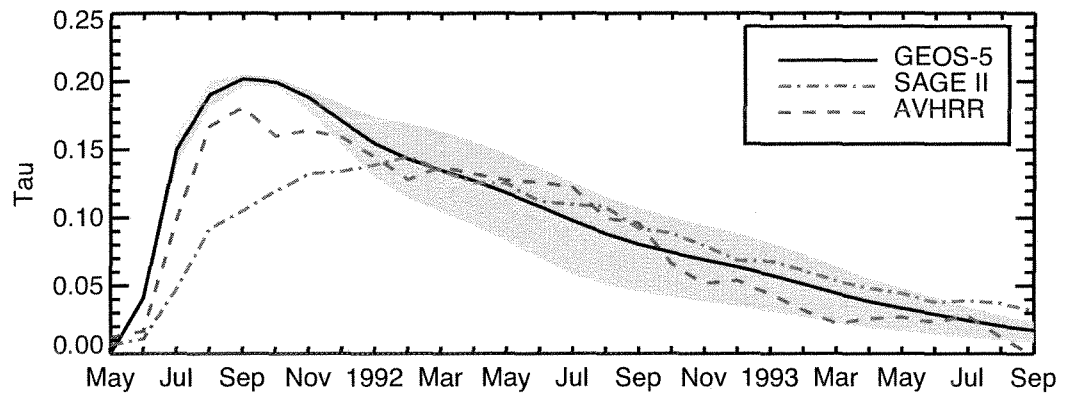
Ensemble member	<i>e</i> -folding time	
	SO ₂	SO ₄
1 st	31	373
2 nd	29	340
3 rd	26	243
4 th	34	426
5 th	29	372
6 th	29	345
7 th	26	275
8 th	34	402
Average	29.8	347
Standard deviation	2.9	57.7

638 Table 1: SO₂ and SO₄ *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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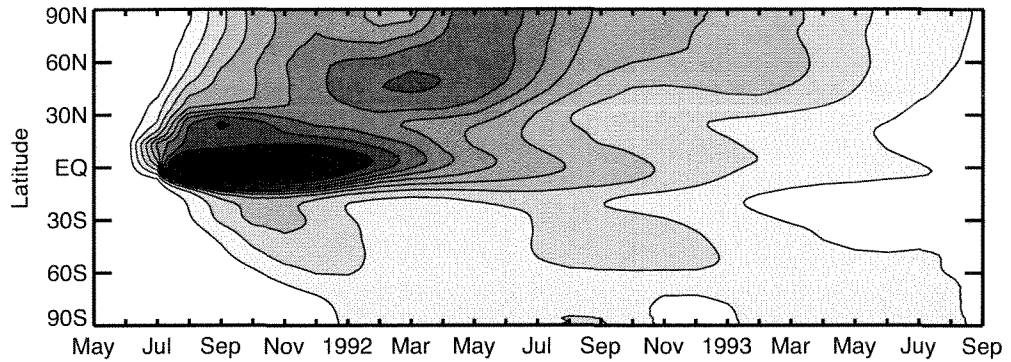


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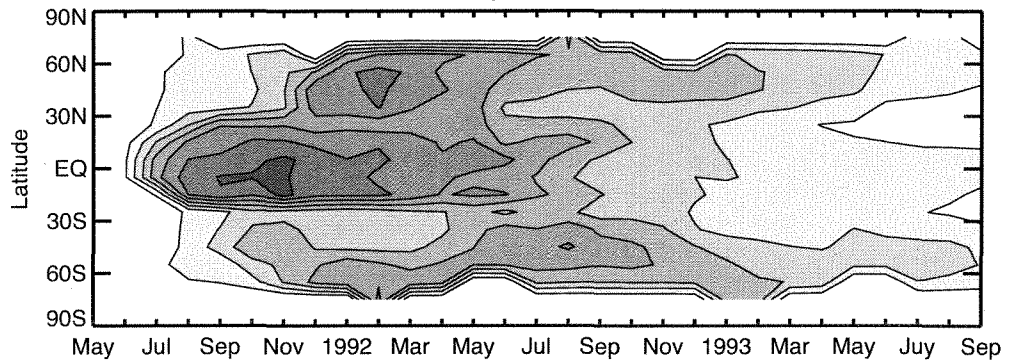
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644 been removed from the AVHRR data. The shaded area shows the variability of the
645 ensemble.

Zonal mean visible aerosol optical thickness

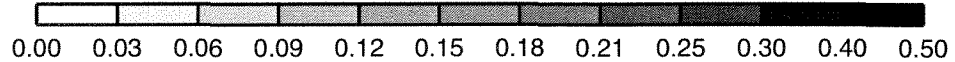
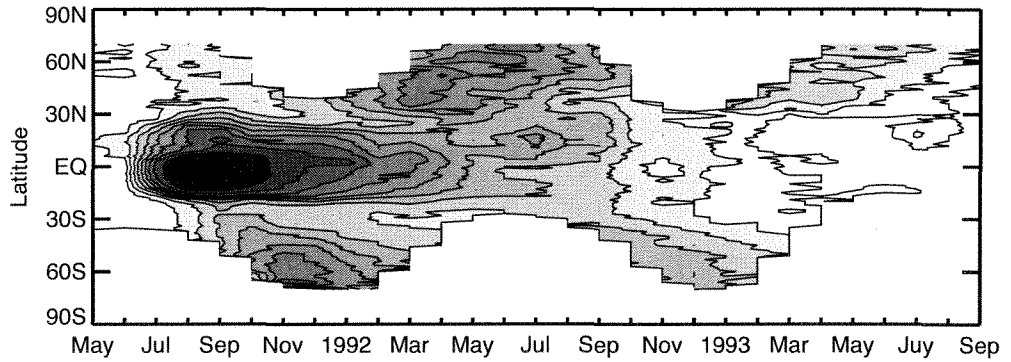
GEOS-5



SAGE-II



AVHRR



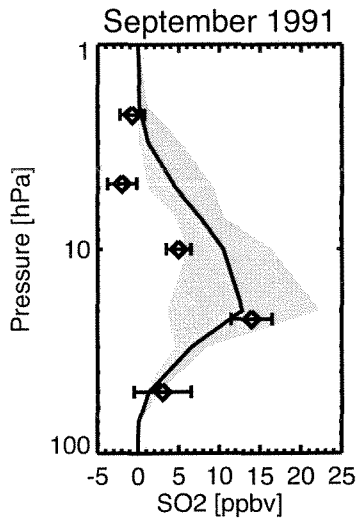
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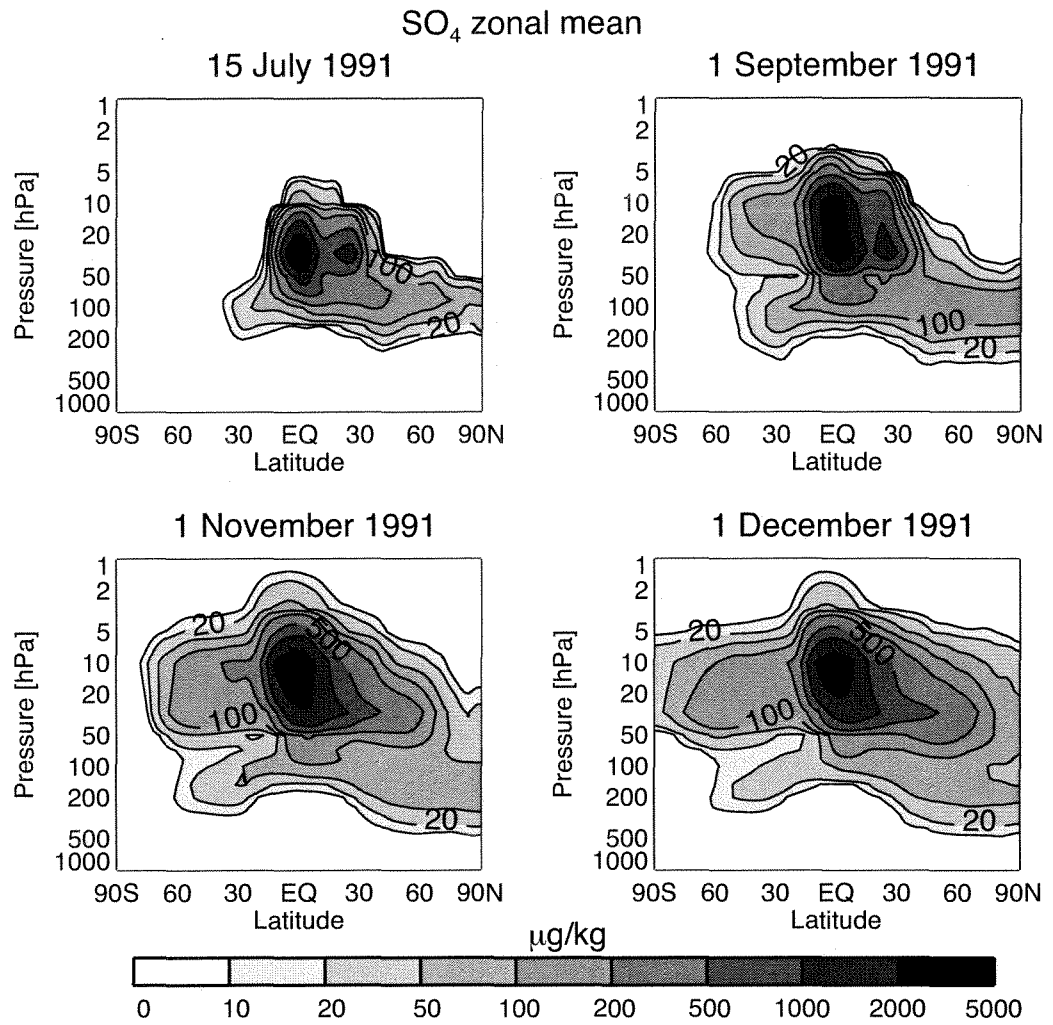
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651 Figure 3: Vertical profile of the monthly average of the SO₂ mixing ratio in
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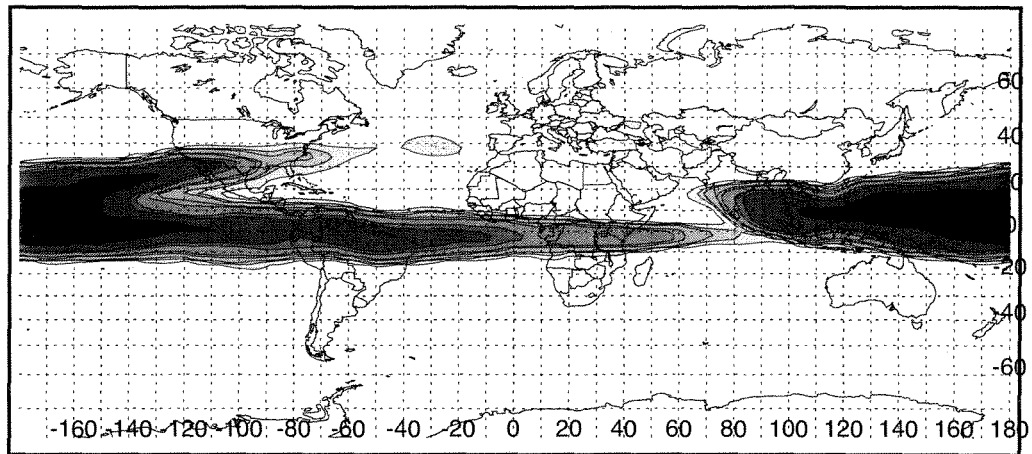
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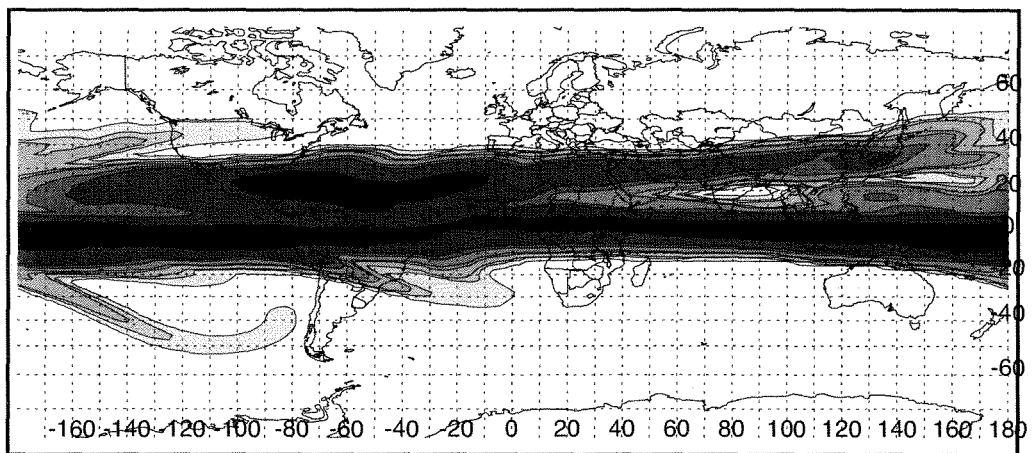
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Figure 4: Zonal mean of the SO₄ concentration from Mt. Pinatubo on July 15th, September 1st, November 1st and December 31st, 1991.

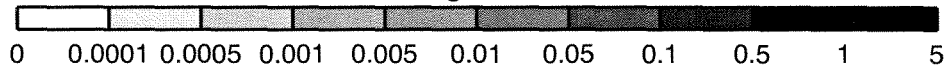
2 July 1991



16 July 1991



g/m^2

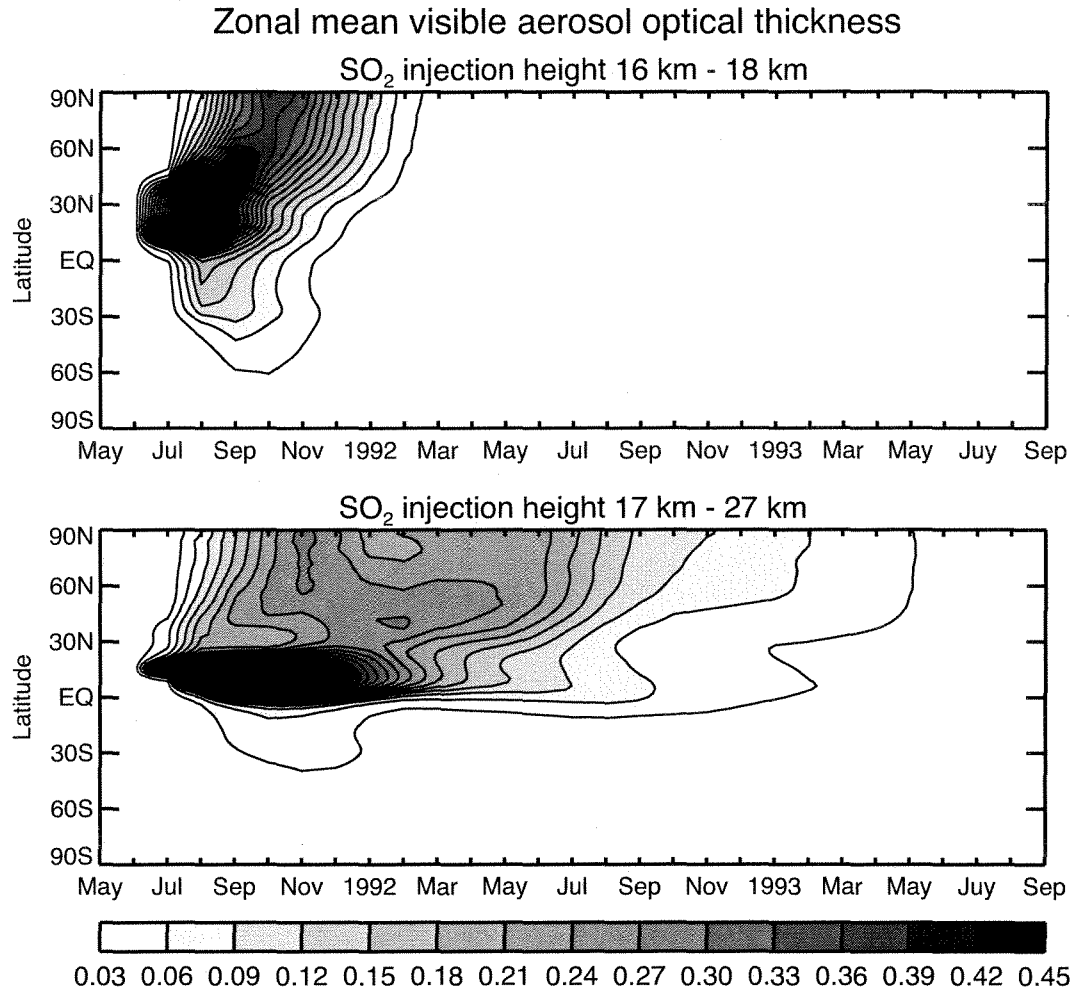


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Figure 5: Horizontal distribution of SO_2 column mass between 30 hPa and the top of the atmosphere on July 2nd, 1991 and on July 16th, 1991.



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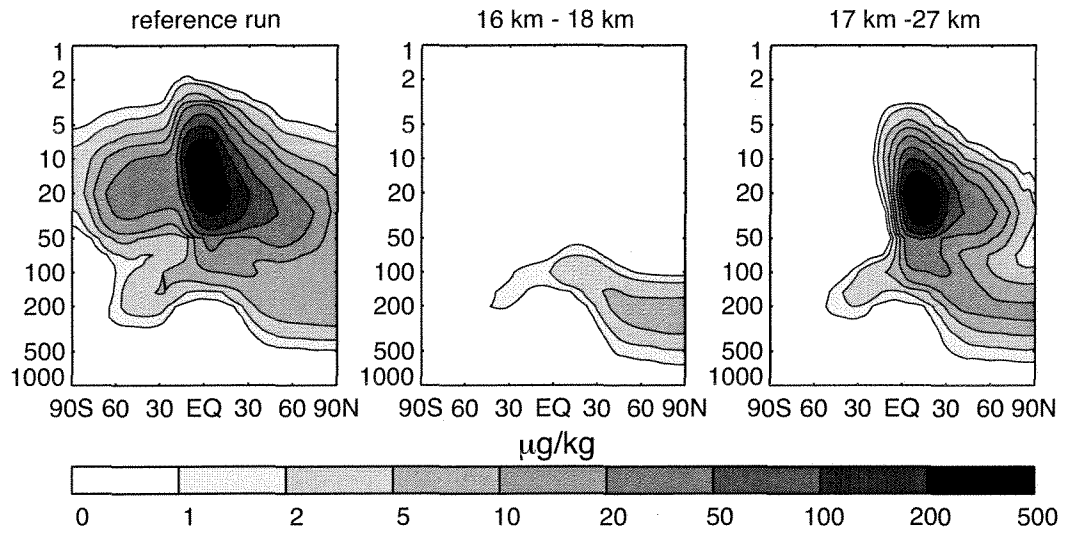
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Figure 6: Temporal evolution of the zonally averaged aerosol optical thickness at 550 nm in the ensembles with no radiatively interactive aerosol and SO₂ injection height between 16 and 18 km (upper panel) and 17 and 27 km (lower panel).

SO₄ zonal mean - December 1991



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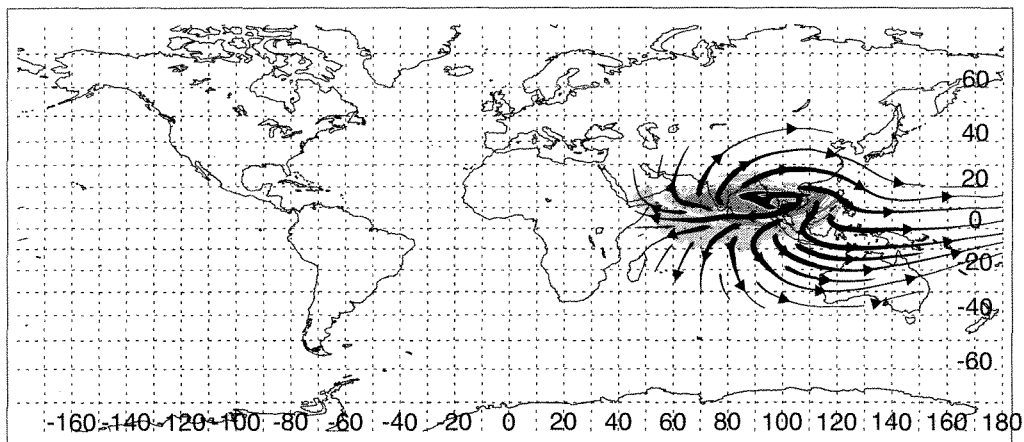
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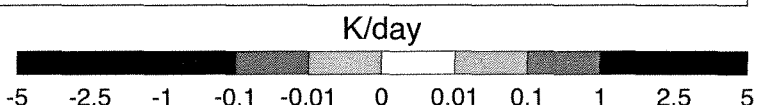
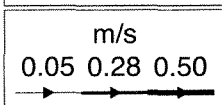
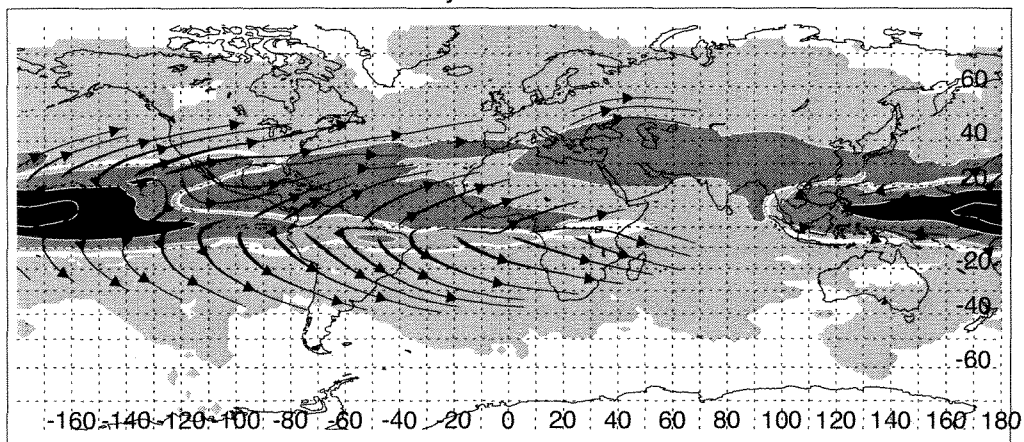
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Longwave heating rates

16 June 1991 - 70 hPa



1 July 1991 - 30 hPa



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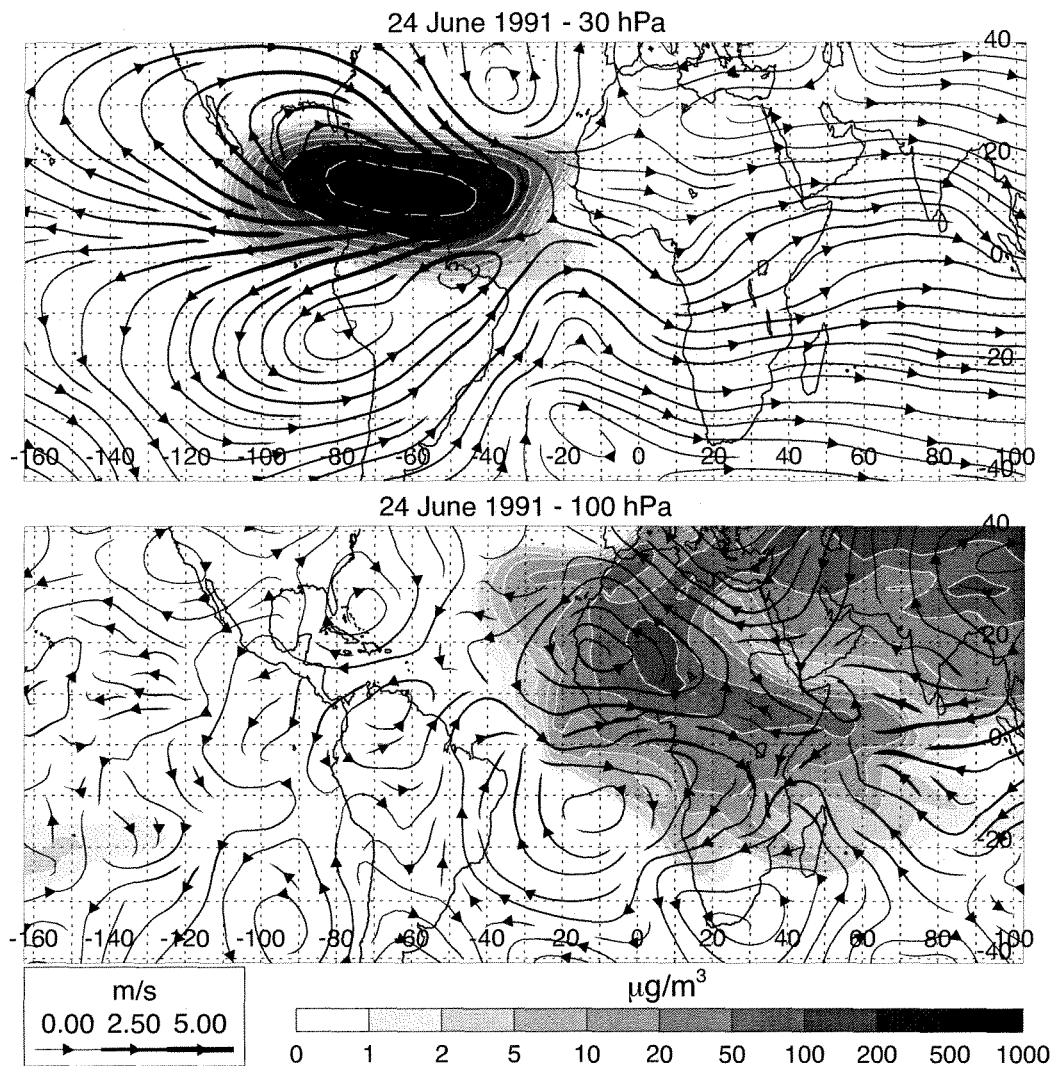
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Figure 8: Streamlines of the difference between the horizontal wind field in the interactive and in the non-interactive simulation on June 16th, 1991 (upper panel) and on July 1st, 1991 (lower panel) at 70 hPa and 30 hPa altitude, respectively. The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

SO₄ concentration and perturbation of the horizontal wind



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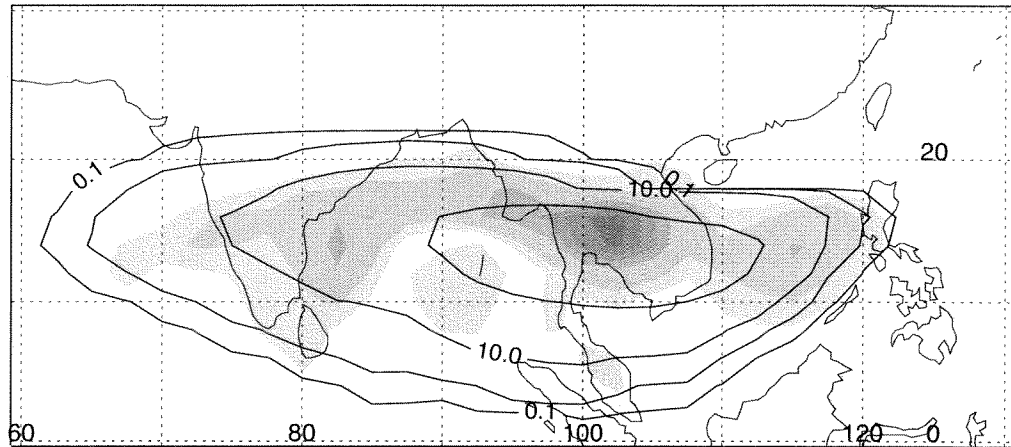
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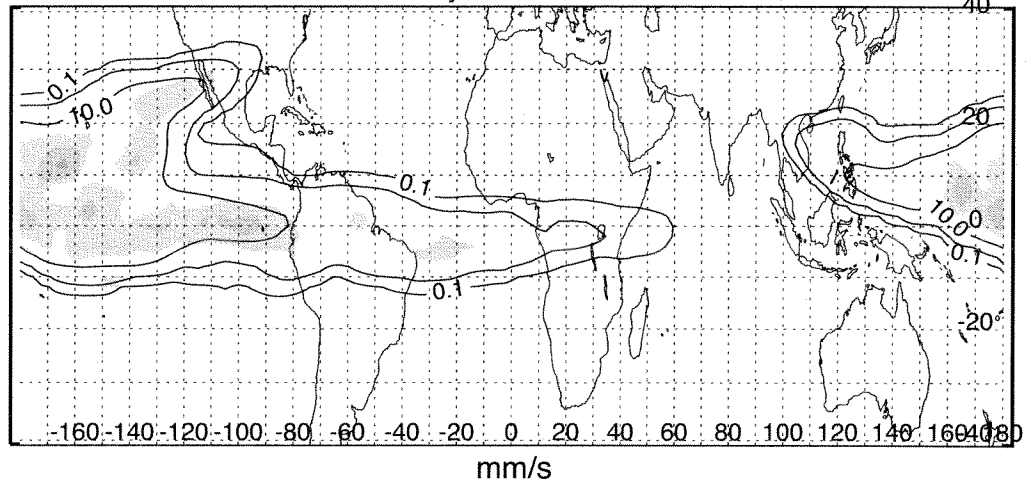
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Anomaly of the vertical velocity

16 June 1991 - 70 hPa



1 July 1991 - 30 hPa



mm/s

A horizontal color scale bar ranging from -4.0 to 4.0 mm/s. The scale is divided into 17 segments, with values: -4.0, -3.5, -3.0, -2.5, -2.0, -1.5, -1.0, -0.5, 0.0, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0. The colors transition from dark grey for negative values to light grey for positive values.

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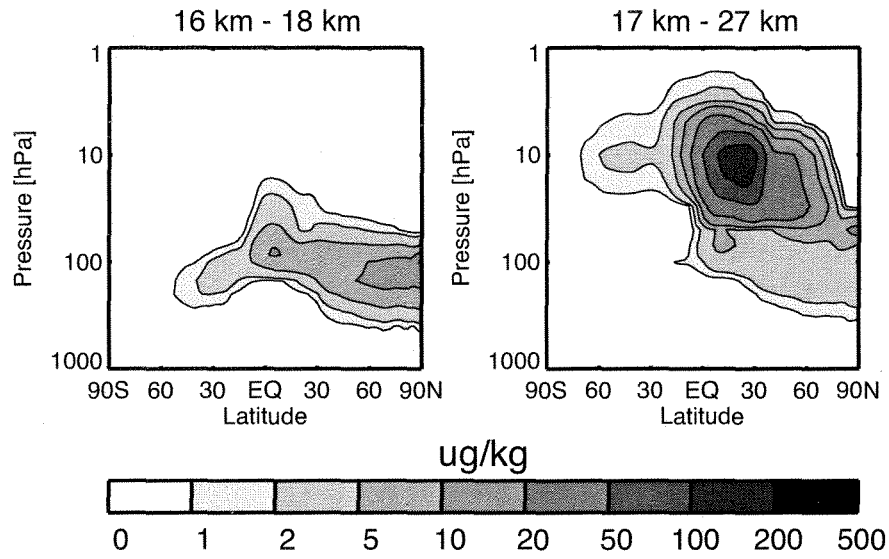
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SO₄ zonal mean



689

690 Figure 11: Zonal mean of the SO₄ concentration on October 15th, 1991 in the
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