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The response of ozone and nitrogen dioxide to the eruption of Mt. Pinatubo.

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Popular summary

On 15 June 1991 the eruption of Mt. Pinatubo, in the Philippines, injected about 20 Tg of sulfur dioxide (SO_2) in the stratosphere. The SO_2 transformed into sulfate aerosol, increasing the stratospheric aerosol loading by orders of magnitude. Such a massive aerosol perturbation is expected to change both the chemistry and dynamics of the stratosphere. On the one side, the volcanic sulfate provides additional surface for heterogeneous chemistry, lowering especially the concentrations of ozone and nitrogen dioxide (NO_2). On the other side, heating by the volcanic aerosol changes the radiative balance of the atmosphere and, therefore, the atmospheric dynamics.

In the case of the Mt. Pinatubo eruption, the aerosol heating intensified upwelling in the tropics and downwelling in the extra-tropics, strengthening the Brewer-Dobson circulation. Observations also showed a strong depletion of stratospheric NO_2 starting from three months after the eruption, indicating a strong volcanic effect on the heterogeneous chemistry. However, while a general depletion of total column ozone was observed in the northern hemisphere, observations did not detect any significant trend in the southern hemisphere, and even registered an increase in the total column ozone during the year following the eruption. The lack of ozone depletion in the southern hemisphere after the eruption of Mt. Pinatubo, contrary to the clear depletion of NO_2, has been an outstanding puzzle for many years.

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

quantifying the ozone and NO_2 anomalies due to volcanic effects on chemistry and dynamics.

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

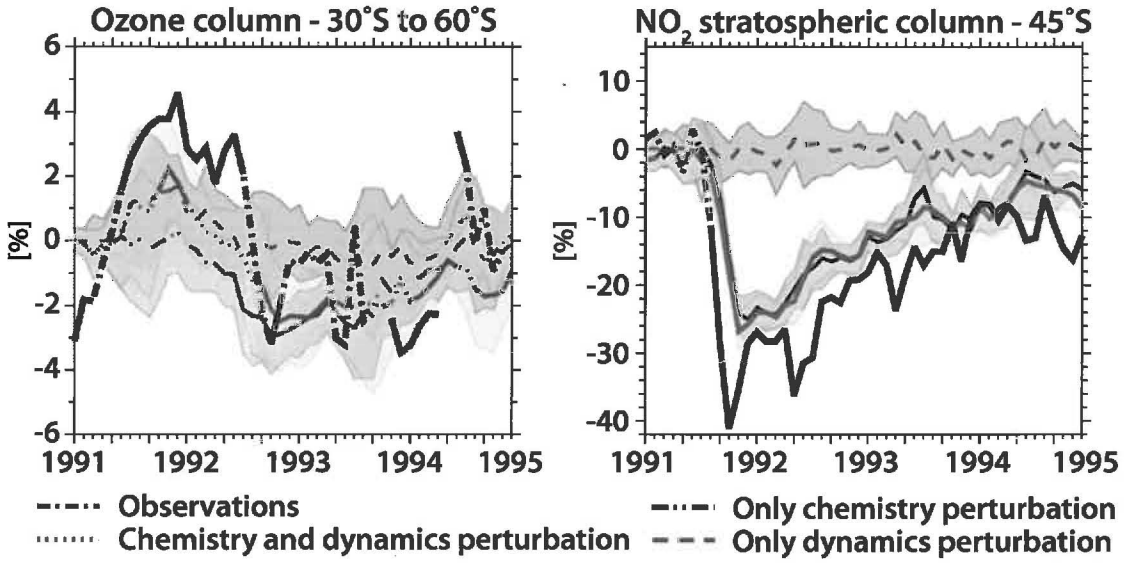

Fig. 1: Zonally averaged anomalies of the total ozone column and stratospheric NO_2 column at southern midlatitudes from observations (black) and simulations including only the chemistry perturbation (blue), only the dynamics perturbation (green), and both the chemistry and the dynamics perturbation (red). The solid lines are significant at one standard deviation level. The volcanic perturbation to the dynamics does not perturb NO_2, but induces a positive anomaly in the total ozone column.

The response of ozone and nitrogen dioxide to the eruption of Mt. Pinatubo.

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Abstract

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

1. Introduction

The volcanic eruption of Mount Pinatubo on 15 June 1991 injected about 20 Tg of sulfur dioxide (SO_2) into the stratosphere [*Bluth et al,* 1992], up to an altitude of about 30 km [*McCormick and Veiga,* 1992]. The SO_2 transformed into about 30 Tg of sulfate aerosol [*McCormick et al,* 1995], increasing the stratospheric aerosol loading by orders of magnitude over background. This perturbation persisted in the atmosphere for several years.

29 Aerosol from Mt. Pinatubo reached both the northern (NH) and southern
30 hemispheres (SH), changing the stratospheric chemistry and dynamics. The volcanic
31 sulfate provided additional surface for heterogeneous chemistry, impacting especially the
32 concentrations of ozone and nitrogen dioxide (NO_2) [*Tie and Brasseur,* 1995]. Heating by
33 this volcanic aerosol also changed the radiative balance of the atmosphere, intensifying
34 upwelling in the tropics and downwelling in the extra-tropics [e.g. *Pitari and Mancini,*
35 2002*; Aquila et al.,* 2012].

36 Observations showed depletion of stratospheric NO_2 in both hemispheres during
37 the years following the eruption [*Johnston et al.,* 1992*, Van Roozendael et al.,* 1997],
38 showing that the volcanic aerosol enhanced the heterogeneous chemistry at all latitudes.
39 Surprisingly, the observed ozone response to the volcanic perturbation was different in the
40 NH and in the SH. While column ozone generally decreased in the NH, an increase of the
41 ozone column was detected at southern mid- and high latitudes during the year following
42 the eruption [*Randel and Wu*, 1996].

43 Several model studies attribute the observed NH ozone depletion to the
44 enhancement of heterogeneous chemistry because of the volcanic aerosols [e.g. *Tie and*
45 *Brasseur,* 1995], but cannot explain why the same heterogeneous chemistry did not affect
46 the SH ozone concentration [*Stolarski et al.,* 2006]. Some studies suggested explanations
47 to the asymmetry of the ozone response other than a chemistry perturbation. *Randel and*
48 *Wu* [1996] showed that the quasi-biannual oscillation (QBO) increased ozone in the
49 extratropical SH during the 1991/1992 winter, but the effect is not large enough to explain
50 the observed increase. *Fleming et al.* [2007] and *Telford et al.* [2009] successfully
51 simulated the ozone behavior using observed meteorological fields. These studies attribute

52 the absence of an ozone depletion to interannual dynamical variability, which masked the
53 Pinatubo aerosol chemical effect in the SH. However, these studies cannot distinguish
54 between natural interannual variability and circulation changes forced by the volcanic
55 perturbation. *Poberaj et al.* [2011] performed a multiple linear regression analysis to the
56 Chemical and Dynamical Influences on Decadal Ozone Change (CANDIDOZ), stating
57 that volcanically induced chemical ozone depletion was overcompensated by the QBO and
58 by a pronounced Eliassen-Palm (EP) flux anomaly.

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

63 **2. Model and simulation**

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

74 GOCART computes the transformation of SO_2 into sulfate aerosol. The
75 simulations shown here use prescribed aerosol surface area density for heterogeneous
76 chemistry from SAM II, SAGE and SAGE-II data [*Eyring et al.,* 2008]. GEOS-5 also
77 includes an option for calculating the aerosol surface area density online using the mass of
78 sulfate aerosol and relative humidity.

79 We simulate the eruption of Mt. Pinatubo by injecting 20 Tg of SO_2 between 16
80 km and 18 km in the volcano's model grid box on 15 June 1991. No other aerosol sources
81 are included in the simulation. We performed four model experiments (Table 1), each
82 composed of 10 simulations with different sets of initial conditions typical of the year
83 2000. The results shown here are ensemble averages.

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

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

91 The third experiment (experiment CHEM) does not include radiatively interactive
92 aerosol, and uses aerosol surface area density from SAGE-II data appropriate for the
93 simulated year. In this experiment the aerosol cannot modify the simulated meteorology,
94 i.e. there is a perturbation to the stratospheric chemistry but no perturbation to the
95 dynamics.

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

99 **3. Results**

100 The comparison of the experiment FULL with the control experiment REF
101 identifies the complete perturbation of ozone and NO_2 due to Mt. Pinatubo. The
102 comparisons of experiments DYN and CHEM with experiment REF isolate the anomalies
103 of ozone and NO_2 due to the eruption effect on the atmospheric dynamics and
104 heterogeneous chemistry, respectively.

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

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

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

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

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

138 Fig. 3 shows the simulated vertical distribution of the zonal mean ozone anomaly
139 in June-July-August (JJA) 1991 (left panel) and September-October-November (SON)
140 1992 (right panel) due to the heterogeneous chemistry and dynamics (FULL-REF). In JJA
141 1991 the perturbation is completely dominated by the dynamics response, which we depict

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

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

156 **4. Conclusions**

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

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

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

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242 *Depletion: 2010,* Global Ozone Research and Monitoring Project—Report No. 50, 572
243 pp., Geneva, Switzerland, 2011.
244
245 **Table 1:** list of performed experiments.

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

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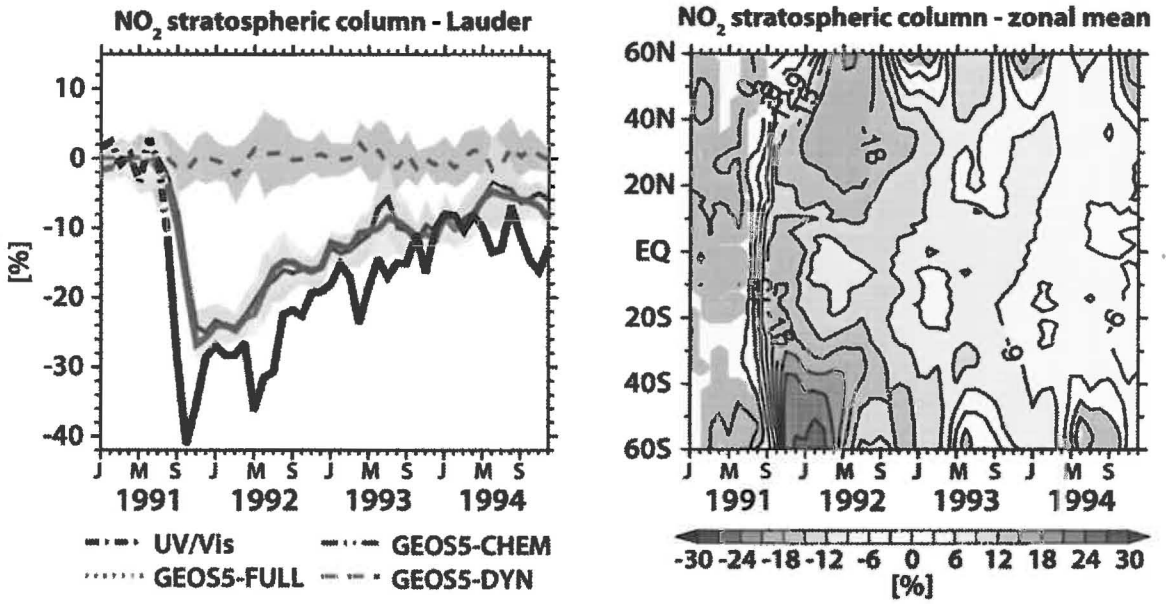
248 **Fig. 1:** NO_2 stratospheric column anomaly versus time. Left panel: The black line marks
249 the observed anomalies over Lauder, the red, blue and green lines show the simulated
250 zonal mean anomaly at 45°S. The shaded areas show the standard deviation of each
251 ensemble. Right panel: Zonal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



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255 **Fig.2:** Ozone total column anomaly versus time. Left panel: zonal mean between 30°S and
256 60°S as observed by TOMS (black line) and as simulated by GEOS-5 (blue, red and green
257 lines). The shaded areas show the standard deviation of each ensemble. Right panel: Zonal
258 mean of the simulated ozone column anomalies in experiment FULL with respect to
259 experiment REF. The solid lines (left) and bright areas (right) are significant at 1-σ level.
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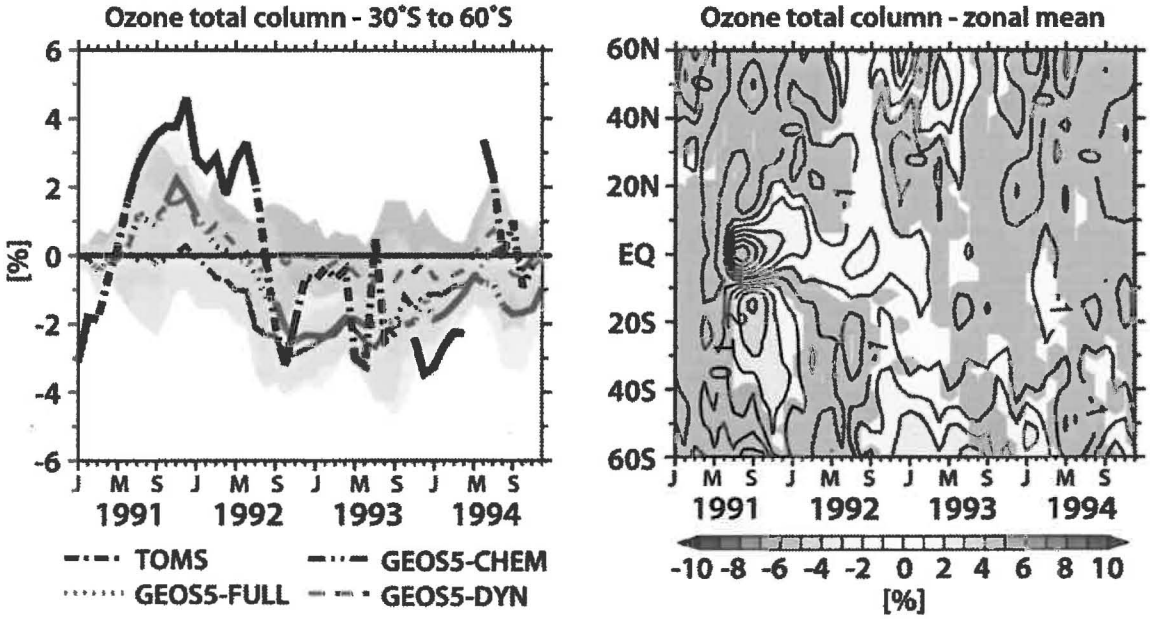

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262 **Fig. 3:** Vertical distribution of the zonal mean ozone relative anomaly [%] in June-July-
263 August 1991 (left panel) and September-October-November 1992 (right panel) of the
264 experiment FULL with respect to the experiment REF. The streamlines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

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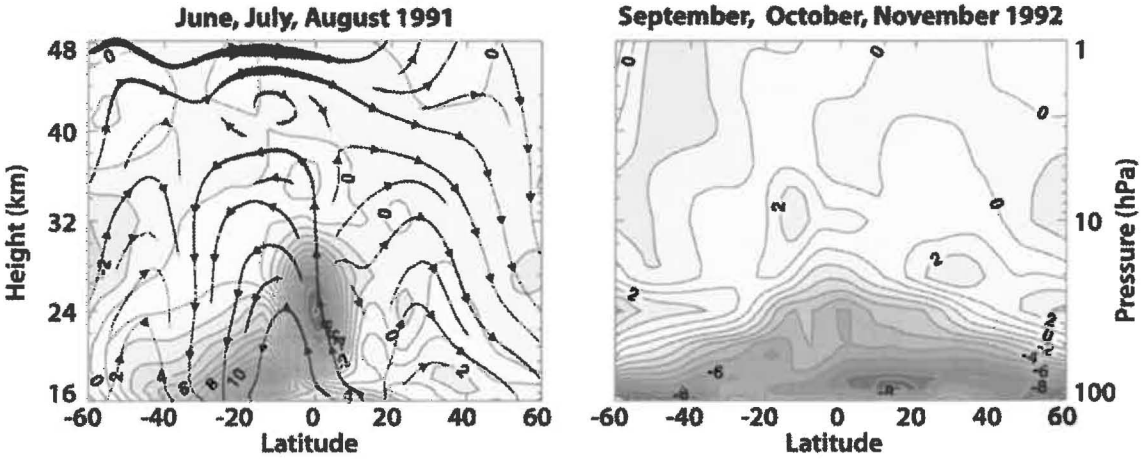

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