Climate/chemistry effects of the Pinatubo volcanic eruption simulated by the UIUC stratosphere/troposphere GCM with interactive photochemistry

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[1] The influence of the sulfate aerosol formed following the massive Pinatubo volcanic eruption in June 1991 on the chemical composition, temperature, and dynamics of the atmosphere has been investigated with the University of Illinois at Urbana-Champaign (UIUC) stratosphere–troposphere General Circulation Model (GCM) with interactive photochemistry (ST-GCM/PC). Ensembles of five runs have been performed for the unperturbed (control) and perturbed (experiment) conditions. The simulated repartitioning within the chlorine and nitrogen groups, as well as the ozone changes, are in reasonable quantitative agreement with observations and theoretical expectations. The simulated ozone changes in the tropics reveal the ozone mixing ratio decreases below 28 km and increases in the stratosphere above this level. However, these changes are not statistically significant in the lowermost stratosphere. The simulated total ozone loss reached 15% over the northern middle and high latitudes in winter and early spring. However, the simulated changes are statistically significant only during early winter. The magnitude of the simulated total ozone depletion is generally less than that observed, but some members of the experiment ensemble are in better agreement with the observed ozone anomalies. The model simulates a pronounced stratospheric warming in the tropics, which exceeds the warming derived from observations by 1–2 K. The model matches well the intensification of the polar-night jet (PNJ) in December 1991 and 1992, the statistically significant cooling of the lower stratosphere and warming of the surface air in boreal winter over the United States, northern Europe, and Russia, and the cooling over Greenland, Alaska, and Central Asia. *INDEX TERMS:* 0370 Atmospheric Composition and Structure: Volcanic effects (8409); 3337 Meteorology and Atmospheric Dynamics: Numerical modeling and data assimilation; 3319 Meteorology and Atmospheric Dynamics: General circulation; 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; *KEYWORDS:* Pinatubo, aerosol, chemistry, ozone, volcanic effects, climate

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1. Introduction

[2] A major explosive volcanic eruption is able to substantially alter the radiation fields in the stratosphere, and thereby affect the stratospheric heating and photolysis rates, and the stratospheric temperature and circulation patterns [*Robock*, 2000]. Additionally, the sulfate aerosol produced in situ from the oxidation of sulfate-containing gases provides the medium for relatively fast heterogeneous chemical reactions that deactivate nitrogen oxides and activate chlorine oxides [*Solomon*, 1999], which lead to changes in the ozone concentration. The ozone changes, in turn, can affect the radiation, temperature, and dynamics in the middle and lower stratosphere and alter the state of the tropospheric weather/climate system. Therefore, a theoretical and observational study of postvolcanic changes can substantially improve our understanding of the different physical and chemical processes in the atmosphere and their relationships. Studies of the consequences of volcanic eruptions can facilitate understanding of how the climate system responds to a stratospheric perturbation and enable estimation of the

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magnitude of the possible influence of stratospheric processes on the tropospheric climate/weather system. Modeling efforts and comparison of the simulated changes in postvolcanic periods with observations will also allow determination of how successfully the actual processes in the atmosphere can be simulated by a model, and what part of a model should be improved to obtain better performance.

[3] Among the major volcanic eruptions [e.g., *McCormic et al.*, 1995; *Rampino and Self*, 1984; *Robock and Mao*, 1995], Pinatubo is an exception owing to the numerous observations collected during and after its eruption in June 1991. The formation and evolution of the aerosol cloud, optical properties of the aerosol, radiative fluxes, chemical composition, temperature and the dynamical state of the atmosphere have been observed by a variety of satellite, balloon, lidar, airborne and ground-based instruments. The availability of different satellite measurements has allowed development of a data set of aerosol optical properties [*Stenchikov et al.*, 1998; *Andronova et al.*, 1999] which can be used in the simulation of the radiative and climatic effects of the Pinatubo aerosol. However, from analyses of the observations alone it is difficult to understand which processes are responsible for the observed changes of ozone and temperature following the Pinatubo eruption. Ozone, temperature, and dynamics are closely linked in the real atmosphere. Therefore, only model simulations of the consequences of the Mount Pinatubo eruption with state-of-the-art models, sensitivity studies therewith, and comparison of their results with the observations can answer the question: Which processes are mainly responsible for the observed ozone and temperature changes?

[4] During the past 8 years a number of modeling studies of the effects of the Pinatubo eruption have been performed, mainly with 2-D zonally averaged stratospheric models. These modeling efforts mainly addressed the causes of the ozone changes and the potential role of heterogeneous processes in the observed ozone depletion. The results obtained by *Brasseur and Granier* [1992], *Pitari and Rizi* [1993], *Kinnison et al.* [1994], *Tie et al.* [1994], *Jackman et al.* [1996], and *Solomon et al.* [1996] concluded that heterogeneous chemistry is the most important factor for the simulated ozone depletion in the tropical and polar areas. However, in the paper by *Rosenfield et al.* [1997] it was found that the main process responsible for about 60% of the ozone depletion in the tropics is the increased upward motion resulting from the enhanced heating rates. In this model, heterogeneous chemistry determines only about 20% of the total ozone decrease in the tropics. The other 20% has been explained by changes in the photolysis rate. This was explained as being due to the use in this simulation of a new, highly interactive 2-D model, with a parameterized description of planetary waves and a direct calculation of the eddy mixing. This example shows how important it is to include interactively in the model the chemical, hydrothermodynamical, radiative and hydrological processes that are involved in determining the influence of the Pinatubo aerosol on the atmosphere. However, some inherent limitations of 2-D models, discussed in detail by *WMO* [1999] and *Yudin et al.* [2000], hamper their ability to do this. Therefore a more appropriate tool for such studies is a General Circulation Model (GCM) with interactive photochemistry.

[5] Currently, GCMs are focusing on the simulation of the temperature and circulation changes caused by prescribed ozone and volcanic aerosol properties. *Hansen et al.* [1992] applied the GISS GCM, which has a coarse horizontal resolution of 8° latitude by 10° longitude and only 1–2 layers in the stratosphere, to simulate the post-Pinatubo atmosphere. Therefore, the dynamical interaction between the stratosphere and troposphere in that model was very simplified. *Graf et al.* [1993] investigated the relation between the northern hemisphere circulation and the surface air anomalies after the Pinatubo eruption by using the results of a perpetual January simulation with the ECHAM-2 GCM forced by an artificial decrease of solar radiation at the top of the model, and calculated the radiative heating "off-line." They concluded that the observed winter warming of the surface air over the northern hemisphere continents is associated with the enhancement of the polar-night jet (PNJ) due to anomalous radiative heating in the tropical lower stratosphere by the volcanic aerosol. A set of ensemble simulations has recently been performed by *Kirchner et al.* [1999] to study the climate response of the atmosphere to the Pinatubo eruption. They used the ECHAM-4 GCM and realistic aerosol optical properties to perform three sets of 2-yearlong simulations with and without volcanic aerosol forcing and with different sets of sea surface temperature (SST). It was concluded that the model simulates reasonably well the observed general cooling in the troposphere and winter warming near the surface over Northern Hemisphere continents. However, about a 4 K warming in the lower tropical stratosphere was simulated which exceeds the observed value by up to 2 K. It was pointed out that this overestimation of the warming can be explained by the absence of the quasi-biennial oscillation (QBO) in the model and by the cooling of the lower stratosphere due to volcanically induced ozone depletion. *Yang* [1999] and *Yang and Schlesinger* [2001, 2002] also studied the temperature and circulation changes observed following the Pinatubo eruption using the singular-value decomposition technique and the University of Illinois at Urbana-Champaign (UIUC) stratosphere–troposphere GCM, with emphasis on identification and separation of the temperature changes induced by the Pinatubo aerosol and the overlapping El Niño/Southern Oscillation (ENSO) events. Recently, a study of the atmospheric temperature response to the Pinatubo aerosol was performed using the GFDL SKYHI model [*Ramachandran et al.*, 2000]. None of these GCMs, however, included interactive photochemistry. It is difficult to predict what the response of these GCMs would be if interactive ozone chemistry was included because of the strong nonlinearity of the coupled model and the very complicated feedback system in the fully interactive model.

[6] Several GCMs with interactive chemistry [*Zhao et al.*, 1997; *Knight et al.*, 1998] have already been developed and used to estimate the changes in the stratosphere caused by the Pinatubo eruption. However, they mainly addressed the problem of ozone depletion and the partition of the reactive nitrogen and chlorine. They have not provided or discussed any information about the temperature and circulation changes in the stratosphere. Moreover, the above mentioned stratospheric GCMs are not aimed to simulate any tropospheric processes, hence they cannot be used to analyze

stratosphere–troposphere interactions. We have performed post-Pinatubo simulations with a stratosphere–troposphere GCM that has interactive photochemistry and we hope that our experiment, together with recently published results by *Al-Saadi et al.* [2001], will fill this gap.

[7] The model and experimental setup will be briefly described in section 2. The changes in the chemical composition will be addressed in section 3. Section 4 will be devoted to the temperature and zonal wind changes in the post-Pinatubo atmosphere. In section 5 we will consider the volcanic effects that occurred in December 1991. Conclusions are presented in section 6.

2. Model Description and Experimental Setup

[8] We present below the changes in the temperature, circulation and chemical composition in the post-Pinatubo atmosphere simulated by the UIUC 24-layer stratosphere–troposphere GCM with interactive photochemistry (ST-GCM/PC) which has been described in detail elsewhere [*Rozanov et al.*, 2001]. The model has a horizontal resolution of 4° latitude by 5° longitude. In the vertical direction the model extends from the Earth's surface to 1 hPa and uses sigma as its vertical coordinate.

[9] The chemical-transport part of the model simulates the time-dependent three-dimensional distributions of 42 chemical species (O_3, $O(^1D)$, $O(^3P)$, N, NO, NO_2, NO_3, N_2O_5, HNO_3, HNO_4, N_2O, H, OH, HO_2, H_2O_2, H_2O, H_2, Cl, ClO, HCl, HOCl, $ClNO_3$, Cl_2, Cl_2O_2, CF_2Cl_2, $CFCl_3$, Br, BrO, $BrNO_3$, HOBr, HBr, BrCl, $CBrF_3$, CH_3Br, CO, CH_4, CH_3, CH_3O_2, CH_3OOH, CH_3O, CH_2O, and CHO), which are determined by 199 gas-phase and photolysis reactions. The model also takes into account 6 heterogeneous reactions on/in sulfate aerosol and polar stratospheric cloud particles:

$$1.\ N_2O_5 + H_2O = 2HNO_3$$
$$2.\ ClNO_3 + H_2O = HOCl + HNO_3$$
$$3.\ BrNO_3 + H_2O = HOBr + HNO_3$$
$$4.\ ClNO_3 + HCl = Cl_2 + HNO_3$$
$$5.\ HOCl + HCl = Cl_2 + H_2O$$
$$6.\ BrNO_3 + HCl = BrCl + HNO_3$$

[10] The chemical solver is based on the pure implicit iterative Newton–Raphson scheme [*Rozanov et al.*, 1999]. The basic routine of the solver has been accelerated to improve its computational performance. A special acceleration technique for solving a sparse system of linear algebraic equations was developed and used. This technique utilizes the following main ideas: (1) the algorithm of the LU decomposition/back-substitution of the Jacobian matrix is modified to include only nonzero operations; (2) the Jacobian matrix is rearranged such that the greater the number of nonzero elements in the row of the matrix, the farther down the matrix the row is placed—this rearranging allows minimization of the number of the nonzero calculations during the LU decomposition/back-substitution process; and (3) the sequence of rows of the Jacobian matrix depends only on the photochemical reaction table used in the model and is the same for all grid cells of the model domain [*Sherman and Hindmarsh*, 1980; *Jacobson and Turco*, 1994]. To realize these ideas a nonnumerical algorithm was developed which generates a so-called loop-free FORTRAN code with only "nonzero" operations of the decomposition/back-substation process. All these operations are written explicitly as a sequence of simple FORTRAN commands. The reaction coefficients are taken from the works of *DeMore et al.* [1997] and *Sander et al.* [2000]. Photolysis rates are calculated at every step using the look-up table approach [*Rozanov et al.*, 1999].

[11] The advection of the species is performed using a Hybrid advection scheme [*Zubov et al.*, 1999]. The model time steps are 3 min for the dynamical core, and one hour for the treatment of the physical, photochemical and transport processes. PSC properties are calculated by a diagnostic thermodynamic scheme [*Hanson and Maursberger*, 1988; *Chipperfield et al.*, 1993] which describes the condensation, sedimentation and evaporation of type I and II PSC particles in a simplified manner, with coefficients of the heterogeneous reactions on stratospheric aerosol and PSC particles adopted from the works of *Hanson et al.* [1994] and *DeMore et al.* [1997], respectively.

[12] The radiation code of the model takes into account the solar and infrared radiation perturbations by the sulfate aerosol [*Yang et al.*, 2000]. Middle-level convection and penetrating convection are treated using *Arakawa and Mintz* [1974] and *Arakawa and Schubert* [1974] parameterizations, respectively. Dry-convective adjustment occurs if the temperature lapse rate between any two adjacent vertical layers is absolutely unstable, that is, exceeds the dry-adiabatic lapse rate. The links between the chemical and physical processes are maintained by the exchange of the mixing ratios of the radiatively active species between the radiation and chemistry-transport parts of the model.

[13] This model design assures that almost all physical processes invoked by the products of the volcanic eruption are taken into account. The only exception is that the formation and development of the aerosol layer is not simulated interactively. We use the time-dependent distribution of aerosol optical properties obtained from the work of *Andronova et al.* [1999] in the radiation code of the ST-GCM/PC to calculate the fluxes of solar and longwave radiation in the presence of the volcanic aerosol. The Pinatubo aerosol extinction at 1.02 micron from the same data set is used to calculate the time-dependent volcanic aerosol surface area density according to the equation proposed by *Thomason et al.* [1997]. These data have been used for the calculation of the heterogeneous reaction constants in the chemical routine of the ST-GCM/PC [*Rozanov et al.*, 1999].

[14] We have carried out two sets of 2-year-long ensemble simulations to estimate the statistical significance of the results. Each ensemble set consists of five separate 2-year-long model runs, which have been started from five different sets of initial conditions. The first ensemble set is the control run without any changes in the external forcing. In the second ensemble set we introduced the volcanic-aerosol perturbations into the radiation and chemical parts of the model. For all the simulations we used the climatological SST and sea ice distribution [*Rozanov et al.*, 2001]. Here we present the changes of several simulated quantities caused by the sulfate aerosol produced after the Pinatubo eruption. These changes, which are the difference between the

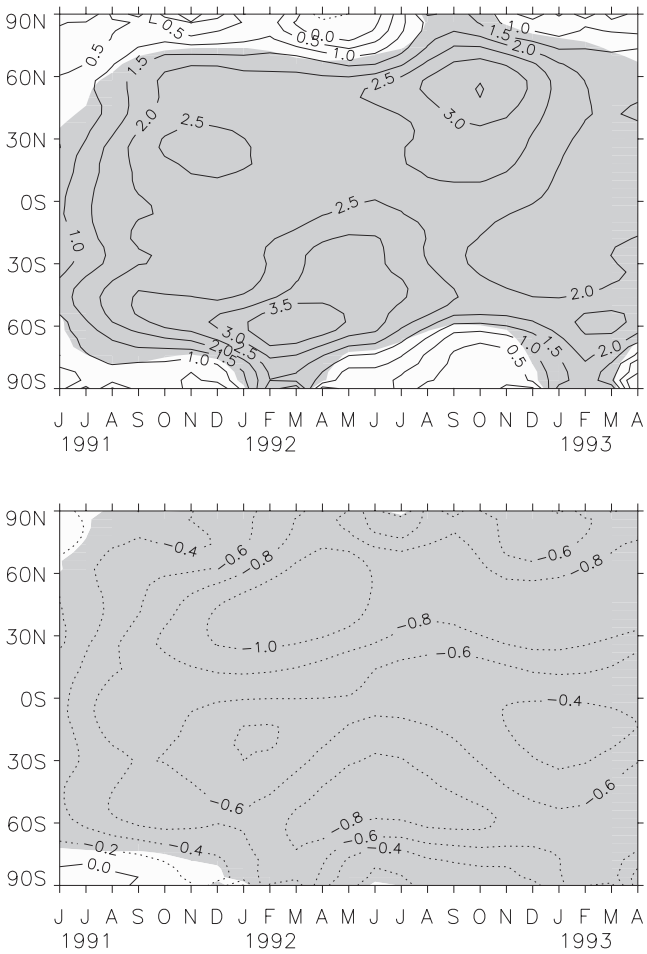

Figure 1. Simulated changes of the monthly mean total vertical column amount (10^{15} mol/cm^2) of HNO_3 (a) and NO_2 (b) after the June 1991 Pinatubo eruption. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

perturbed and control run results, can be called the Pinatubo effects. In the figures, the regions where the changes of the different quantities are statistically significant at better than the 5% level are shaded.

3. Chemical Composition of the Stratosphere

[15] Analyses of the gaseous composition of the atmosphere during the post-Pinatubo period have been presented in many publications (see the extensive reviews by *Solomon* [1999] and *Toohey* [1995]). The most important chemical processes in the post-Pinatubo atmosphere are connected with the presence of liquid sulfate aerosol, which provides a medium for heterogeneous reactions. The most important among them for the tropical lower stratosphere is the hydrolysis of N_2O_5 ($N_2O_5 + H_2O = 2HNO_3$) inside the aerosol particles. This reaction, which is rather fast in the cold tropical lower stratosphere, should lead to a substantial decrease of NO_2 and an increase of HNO_3.

[16] Figure 1 illustrates the simulated changes of the zonal monthly mean stratospheric vertical column amount of HNO_3 and NO_2. The model matches well the theoretically predicted partitioning of the total reactive nitrogen. The HNO_3 column amount started to increase just after the eruption in the tropics. Then, the HNO_3 column abundance increases into the middle and high latitudes due to both transport processes and formation from the in situ hydrolysis of N_2O_5. The HNO_3 increase reaches its maximum values ($2.5–3.5 \times 10^{15}$ mol/cm^2) in the middle latitudes during early autumn 1992 over both hemispheres. As a consequence of the N_2O_5 removal, the NO_2 burden decreases everywhere. The maximum decrease of stratospheric NO_2 ($\sim 1.0 \times 10^{15}$ mol/cm^2) occurs during winter and spring over the middle latitudes in both hemispheres. In the tropics and most of the middle latitudes, the simulated HNO_3 and NO_2 changes are statistically significant. However, in the high latitudes the HNO_3 changes are typically smaller and not statistically significant.

[17] A comparison of the simulated and observed changes of the vertical column HNO_3 and NO_2 amounts is presented in Figure 2 for three ground-based measurement points: Lauder, New Zealand (45°S, 170°E), Jungfraujoch, Switzerland (46°N, 8°E), and Sondakyla, Finland (67°N, 27°E). The observational data have been taken, respectively, from

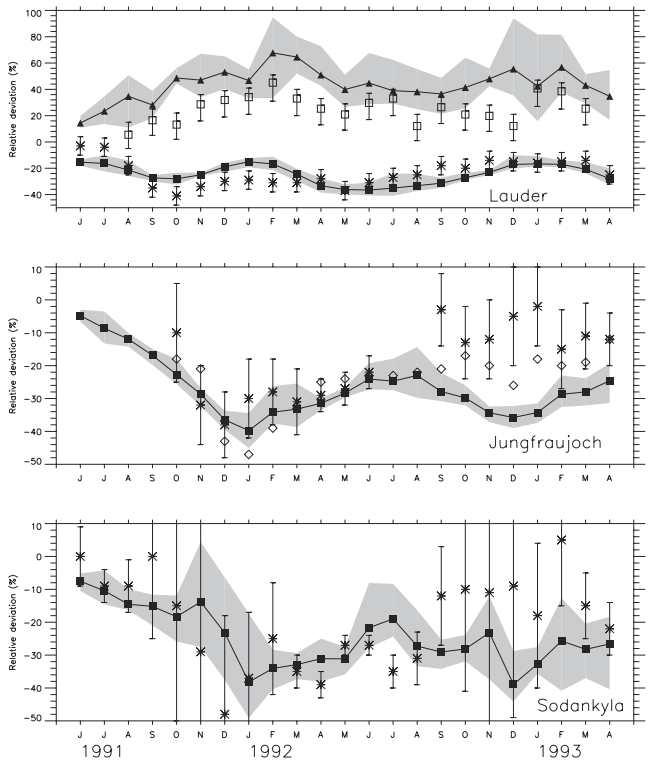

Figure 2. Simulated and observed changes of the monthly mean total vertical column (%) of HNO_3 and NO_2 after the Pinatubo eruption over Lauder (45°S, 170°E), Jungfraujoch (46°N, 8°E), and Sodankyla (67°N, 27°E) stations. The simulated changes are shown by the solid line with markers (solid triangles for HNO_3 and solid squares for NO_2). Empty squares represent the observed HNO_3 changes obtained from the work of *Koike et al.* [1994]. NO_2 changes obtained from the works of *Koike et al.* [1994] and *Van Roozendael et al.* [1997] are marked by asterisks and those from the work of *De Mazière et al.* [1998] by diamonds. Error bars denote the estimated uncertainty of the measurements. Shaded areas denote variability among ensemble members.

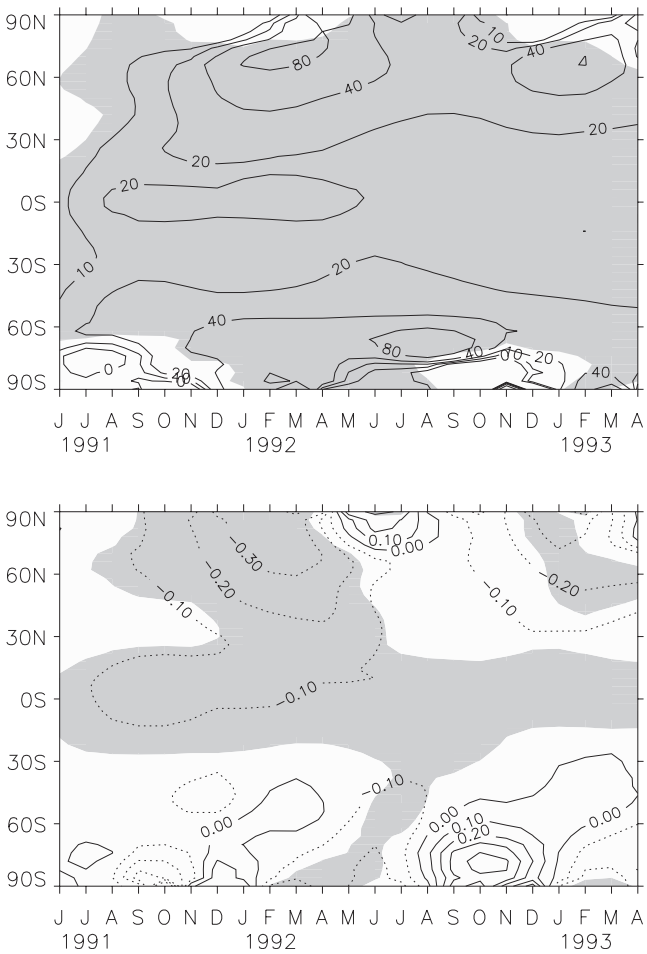

Figure 3. Simulated changes of the monthly mean total vertical column amount of ClO (a) (10^{12} mol/cm^2) and HCl (b) (10^{15} mol/cm^2) after the June 1991 Pinatubo eruption. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

the works of *Koike et al.* [1994], *Van Roozendael et al.* [1997], and *De Mazière et al.* [1998]. Taking into account the rather high level of uncertainty in the estimated HNO_3 and NO_2 changes obtained from measurements, we can conclude that the agreement of the model results with the observations is reasonable. In the Southern Hemisphere the model results reproduce rather well the measured magnitude and phase of the HNO_3 and NO_2 changes after the Pinatubo eruption, except during October 1991 and October–November 1992 when the simulated HNO_3 increase exceeded the observed by 10–20%. Over Europe the model describes well the volcanically induced NO_2 decrease during summer seasons. In the winter of 1991/1992 the model results are also in a good agreement with the observations. However, during the 1992/1993 cold seasons the model overestimates the magnitude of the NO_2 depletion. In the middle-latitude area (Jungfraujoch) the model results exceed observations by about 10%. For the high-latitude Sondakyla station the overestimation is more pronounced, but it is very hard to make a conclusion about the model's performance because the uncertainty of the measurements during winter is rather high. Later (in March–April 1993) the observed and simulated effects are again in reasonable agreement. The overestimation of the simulated NO_2 depletion implies that the heterogeneous processes in the model are too intense during the winter 1992/1993. Reasonable agreement between the simulated and observed NO_2 decrease during the winter 1991/1992, and in the southern hemisphere, allows the conclusion that the heterogeneous reaction scheme is reliable. Because the simulated warming in this area (see section 4) is slightly higher than observed, we may foresee less intensive heterogeneous NO_x to HNO_3 conversion on sulfate aerosol; however, the simulated NO_x depletion exceeds observations and this overestimation takes place for all ensemble members, i.e., it does not depend much on the simulated temperature changes. Therefore if we believe in data quality the only possibility to explain this disagreement is that the sulfate aerosol surface density in Northern Hemisphere fall/winter 1992/1993 is overestimated.

[18] Heterogeneous reactions of chlorine compounds in the stratosphere, together with low NO_2 abundance lead to the transition of passive chlorine (HCl and $ClONO_2$) into chemically active radicals. Figure 3 illustrates the changes of the stratospheric HCl and ClO zonal monthly mean total vertical column amounts simulated by the model after the eruption. The model results show that the increase of the ClO burden is most intense ($\sim 100 \times 10^{12}$ mol/cm^2) during winter in the high latitudes of both hemispheres because of the cold temperature there and the availability of the HCl and $ClONO_2$ for heterogeneous conversion. A significant ClO increase (up to $20-40 \times 10^{12}$ mol/cm^2) also takes place in the tropics and middle latitudes during almost the entire period of the model integration. The corresponding changes of HCl have a more complicated structure. In most of the global domain the vertical column tends to decrease due to heterogeneous chlorine activation. This process leads to statistically significant results mostly in the tropics, and in the Northern Hemisphere during winter 1991/1992. During the late spring and early summer, HCl has a tendency to grow. This could be explained by the fact that in the NO_2-poor postvolcanic stratosphere, fewer chlorine atoms are reacting with NO_2 to produce $ClONO_2$ and,

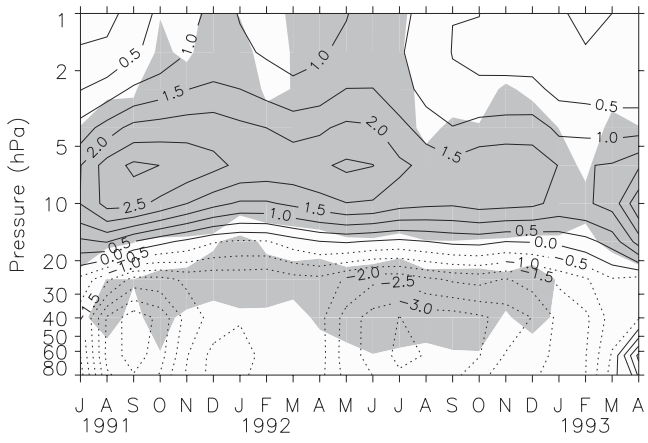

Figure 4. Simulated changes of the monthly zonal-mean O_3 mixing ratio (%) in the tropics after the June 1991 Pinatubo eruption. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

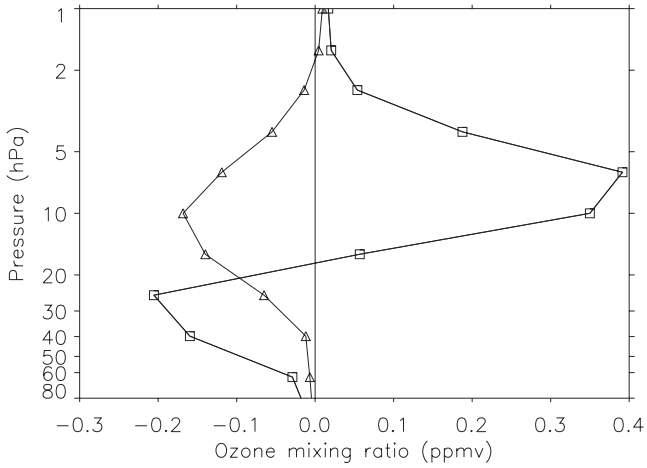

Figure 5. Changes of the monthly zonal-mean O_3 mixing ratio (ppmv) at 15°N in October 1991 simulated with (open squares) and without (open triangles) chemical perturbations due to volcanic aerosol.

therefore, the HCl concentration increases. These results, as well as the changes of HNO_3, NO_x and ClO_x mixing ratios in the lower stratosphere obtained from the same model runs and presented by *Rozanov et al.* [2000], are in reasonable agreement with the observational data and theoretical investigations presented [e.g., *Solomon*, 1999].

[19] The simulated changes of the ozone mixing ratio averaged over the tropical area (26°S–26°N) after the Pinatubo eruption in June 1991 are presented in Figure 4. The ozone mixing ratio decreases slightly in the lower stratosphere below 20 hPa mainly due to destruction by active chlorine. The elevated temperature in the post-Pinatubo tropical stratosphere (see section 4) and the enhanced intensities of the upward motion, which transport ozone-poor air from the lowermost part of the stratosphere and upper troposphere, could also play a role. However, this ozone depletion is not robust in the lowermost stratosphere, which means that the magnitude of the ozone changes is not the same for different members of the ensemble, that is, the effect strongly depends on the dynamical state of the atmosphere. Above 18 hPa the ozone mixing ratio significantly increases, albeit the magnitude is not very large. This feature can be explained by the above mentioned NO_x deactivation.

[20] To clarify the role of heterogeneous chemistry in the simulated ozone changes we performed a model ensemble run with the volcanic aerosol turned off during the photochemical calculations. Although we are preparing a separate paper comparing the results of these two ensemble runs, we present here in Figure 5 the changes of the ozone mixing ratio in October 1991 at 15°N for both simulations. The ozone mixing ratio without the aerosol-related photochemical perturbations decreases in the middle atmosphere due to the elevated temperature there and the intensified uplift of ozone-poor air from the lower stratosphere. Chlorine activation associated with heterogeneous chemistry in the basic run provides for the enhancement of the ozone destruction in the lower stratosphere. Between 20 and 30 hPa the contribution of the heterogeneous chemistry to the ozone depletion is about 50%. Above 18 hPa the influence of the heterogeneous chemistry dominates. In this area the intensive NO_x removal by the heterogeneous processes leads to the increase of the ozone mixing ratio. The simulated changes are in reasonable quantitative agreement with observational data [e.g., *Hofmann et al.*, 1993] and with other model estimations [e.g., *Rosenfield et al.*, 1997].

[21] In Figure 6 the simulated ozone mixing ratio at 50 hPa for July 1992 is compared with HALOE data measured in July 1992 and 1997. These results confirm that the model simulates the ozone mixing ratio reasonably well if the uncertainty of the measurements is taken into account. The model reproduces the tropical minimum and dynamically driven ozone increase toward midlatitudes, although the simulated near-equatorial ozone mixing ratio is slightly overestimated. However, the excessive ozone seems to have no connection to the volcanic effects because the same disagreement also takes place between the simulated ozone and HALOE data for July 1997 when the volcanic aerosol is no longer present in the stratosphere. As mentioned before, the simulated changes of the tropical ozone due to the volcanic aerosol do not exceed 0.2 ppmv and are statistically insignificant.

[22] The simulated change of total column ozone (TCO) is presented in Figure 7. The simulated tropical TCO decreases due to volcanic eruption effects, but not by more than 2%. A noticeable (more than 10%) total ozone depletion caused by the intensification of PNJ (see section 4) and chlorine activation was simulated over the high-latitude area in the Northern Hemisphere during the first winter and spring seasons after the eruption and during the second late winter–spring season in the Southern Hemisphere. The simulated magnitude of the TCO change is in reasonable agreement with the data published by *Angell* [1997b]. He found on the basis of observational data analysis that the tropical ozone depletion is only about 2%. He also detected a significant (~10%) ozone loss in the northern middle and high latitudes. The model underestimates the TCO decrease in the northern middle latitude found by *Angell* [1997b] at least in the ensemble mean.

[23] Almost the same conclusions about the underestimation of the total ozone depletion in the northern middle latitudes can be made if we compare our results with the data published by *Randel et al.* [1995] depicted in Figure 8. The total ozone depletion detected by *Randel et al.* [1995] looks much more pronounced than our results, especially between 40°N and 60°N. This can be partially explained by the fact that we compare a particular year of the observations with the ensemble mean of the simulations. Because the total ozone changes depend on the state of the atmosphere, the use of the ensemble mean inevitably smoothes and decreases the effects. This kind of analysis is a useful tool for forecasting the behavior of the atmospheric system during future volcanic eruptions because it predicts the most robust features of the post-volcanic changes, which will appear no matter what the prevailing state of the atmosphere is. However, for comparison of the simulation results with the observational data for any particular year, it is more reasonable to use that member of the simulation ensemble, which is closest to the actual meteorological situation for the chosen year. In particular, we can try to use for this purpose some a

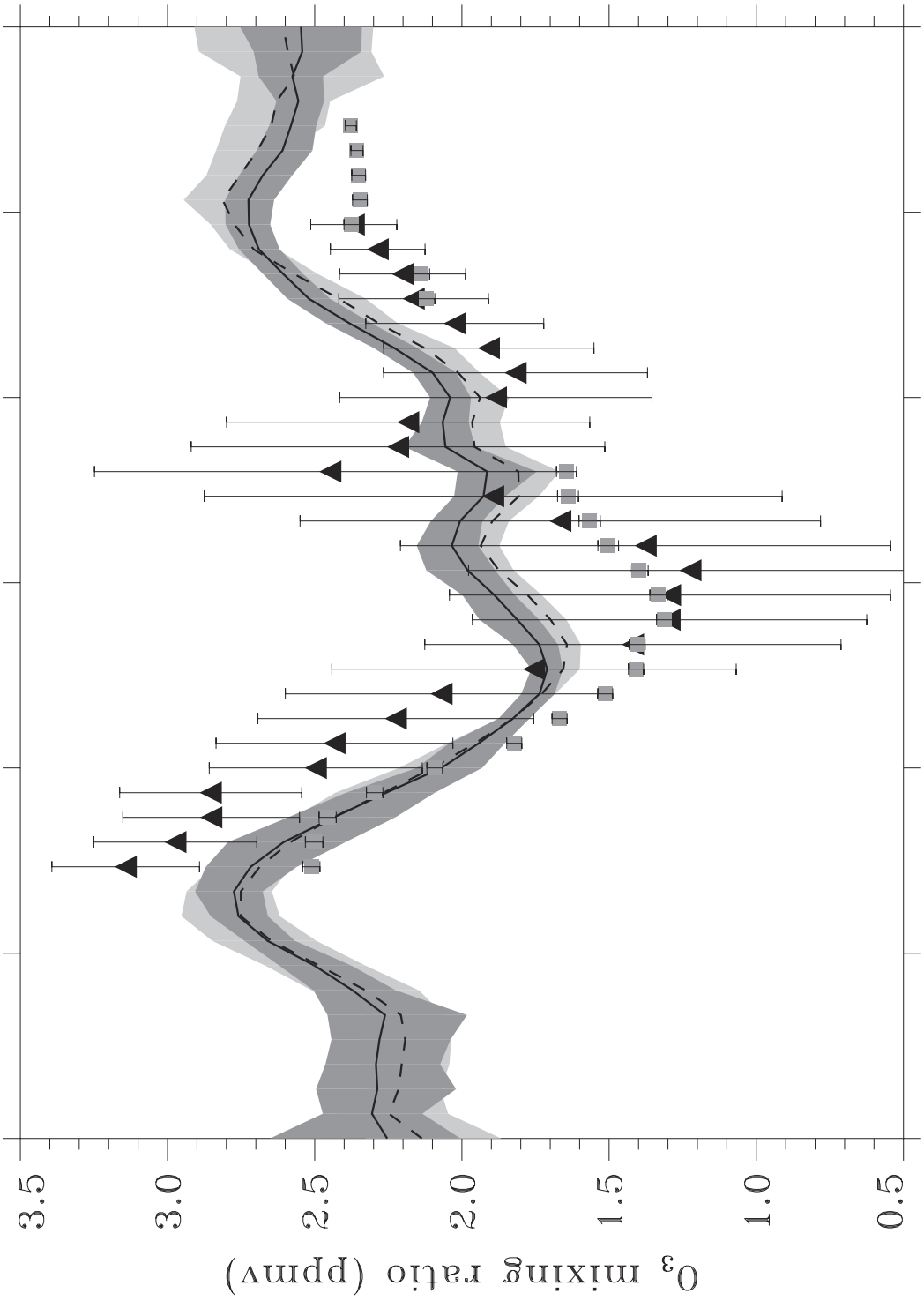

Figure 6. Monthly zonal-mean O_3 mixing ratio (ppmv) at 50 hPa. Ensemble mean simulation results are presented by solid line (control run) and dashed line (experiment) for July 1992. Shaded area represents ozone variability among ensemble members. Version 18 HALOE data are marked by filled triangles for July 1992 and by filled squares for July 1997.

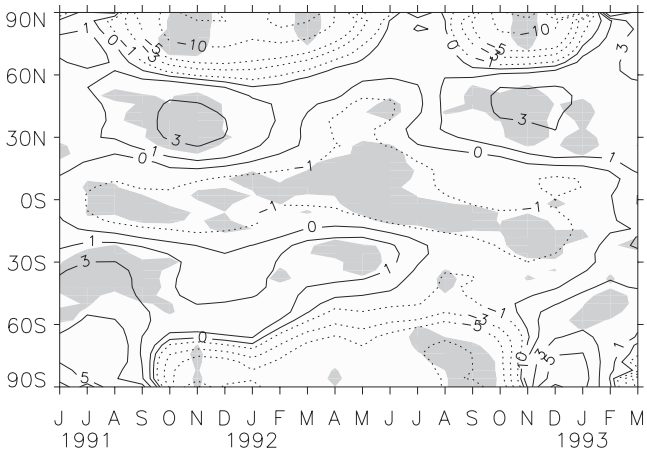

Figure 7. Simulated changes of the monthly zonal-mean total ozone (%) after the June 1991 Pinatubo eruption. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

priori information about the total ozone changes for winter–spring 1991/1992 obtained from the observational analysis and make a reasonable choice among our five ensemble members.

[24] The changes in monthly zonal-mean TCO for all five members of the ensemble are presented in Figure 8, together with the observational data obtained by *Randel et al.* [1995] from the TOMS measurements. For each ensemble member the reference is its corresponding control run. Using a priori information obtained from the observations we may conclude that at least the first and fourth members of the ensemble should be excluded from the analysis of the 1991/1992 winter–spring seasons because they produce a total ozone increase over the Northern Hemisphere. The remaining members provide a much better agreement with the total ozone changes obtained from the analysis of the observational data for this period of time. This means that the state of the atmosphere for the second, third and fifth members of the ensemble is likely closer to the actual meteorological conditions during the first winter after the Pinatubo eruption. To illustrate this conclusion we compare in Figure 9 the monthly zonal-mean zonal wind in February 1991 simulated by all members of the ensemble and obtained from ECMWF reanalysis. It is clearly seen from this figure that for the fourth member of ensemble the model completely failed to produce a strong PNJ in the middle stratosphere, while for the other members the intensity of the PNJ is comparable with the reanalysis data. For the first member of the ensemble the PNJ became rather weak in March 1992 (not shown). However, the TCO decrease during

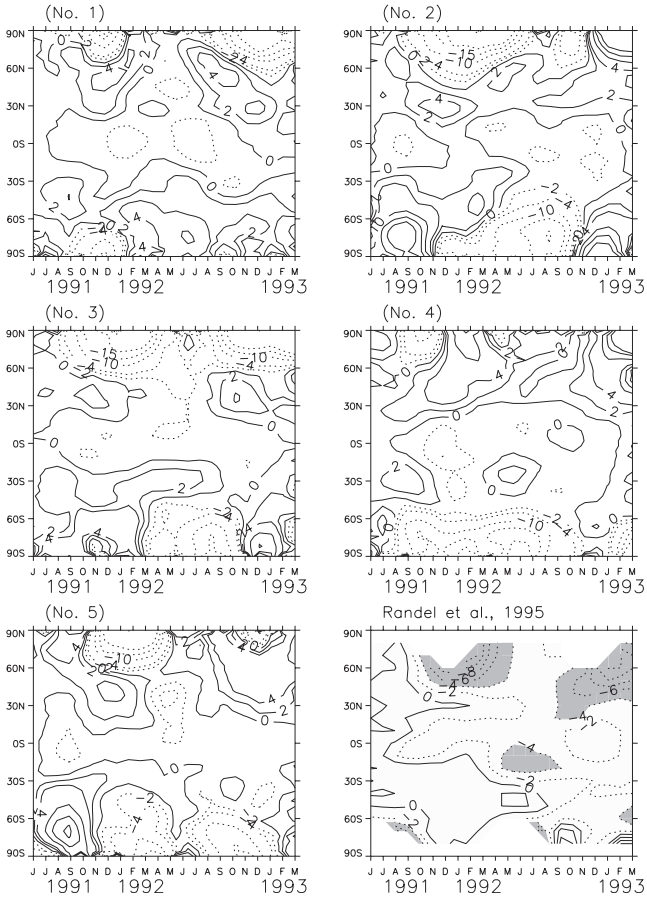

Figure 8. Monthly zonal-mean total ozone changes (%) after the eruption for all five members of ensemble and as obtained from TOMS measurements by *Randel et al.* [1995] (bottom right).

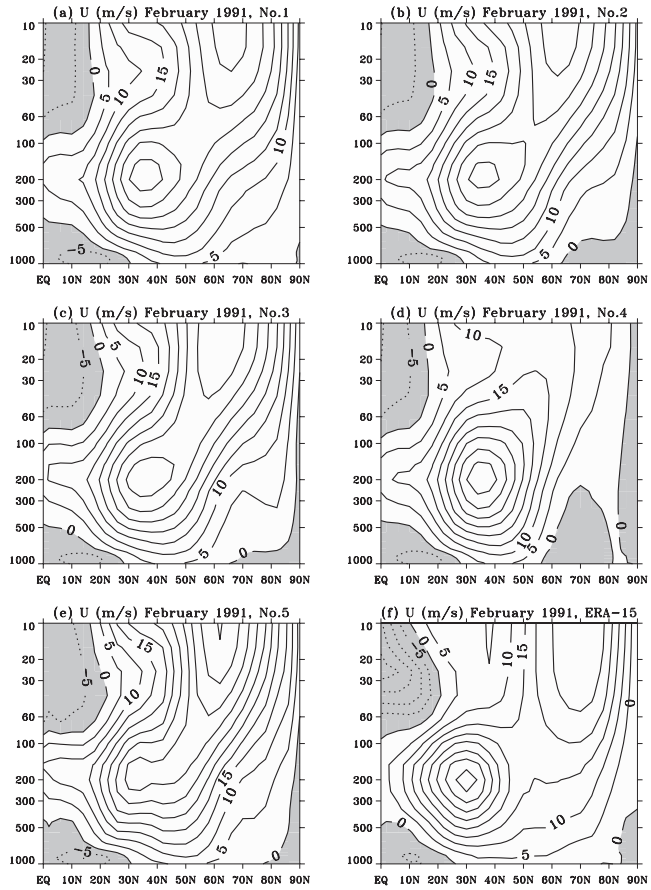

Figure 9. Monthly zonal-mean zonal wind in February 1991 simulated for all five members of the ensemble (a–e) and as obtained from ECMWF reanalysis (f).

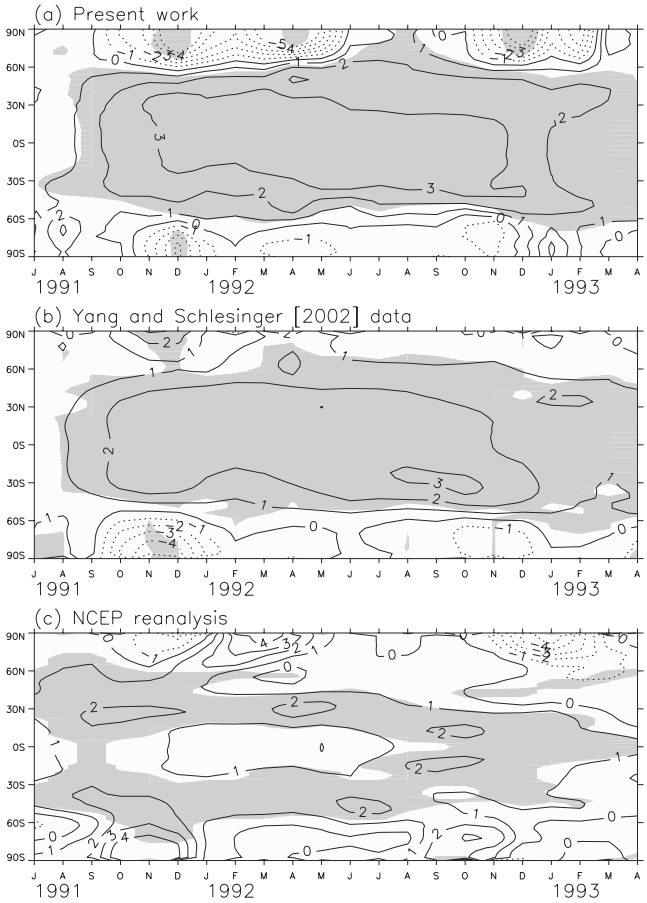

Figure 10. Anomaly of the monthly zonal-mean temperature (K) at 80 hPa after the June 1991 Pinatubo eruption: (a) present work, (b) *Yang and Schlesinger*'s [2002] results, and (c) obtained from NCEP/NCAR reanalysis. The shaded regions denote for (a) and (b) where the anomalies are judged statistically significant at or better than the 5% level and for (c) where the anomalies exceed one standard deviation.

the second winter season (1992/1993) is better reproduced by the first and third members of the ensemble.

4. Temperature and Zonal Wind in the Lower Stratosphere

[25] The time–latitude cross section of the temperature in the lower stratosphere (at 80 hPa) simulated by our model and reported by *Yang and Schlesinger* [2002], as well as temperature anomalies relative to 1968–1997 climatology obtained from NCEP/NCAR reanalysis data provided by the NOAA Climate Diagnostics Center, are presented in Figure 10. The zonal wind changes on the 10 hPa surface are presented in Figure 11.

[26] The main features of the simulated temperature changes are the warming by up to 3.5 K of the lower stratosphere in the wide area from 40°S to 40°N due to the absorption of solar near-infrared and longwave radiation by the aerosol [e.g., *Stenchikov et al.*, 1998; *Andronova et al.*, 1999] and the dynamically driven cooling over the high latitudes during the winter and spring resulting from the intensification of the PNJs.

[27] The time–latitude position of the area with elevated temperature resembles the warming obtained from the NCEP/NCAR reanalysis data. Both the observed and simulated warming starts in August 1991 and reaches 2 K in September–October 1991. The simulated temperature increase also agrees with the observed warming in the northern and southern midlatitudes throughout the period of time considered. However, our model substantially (up to 2 K) overestimates the warming in the tropical area from December 1991 to July 1992. During this particular period of time the analysis of the NCEP/NCAR data reveals only a small (less than 1 K) warming, while the simulated warming is maximal there.

[28] All the above mentioned changes also occur in our simulations in the entire lower stratosphere between 100 and 10 hPa, and they are mostly statistically significant. *Angell* [1997a] also demonstrated observationally the significant influence of the volcanic aerosol on the temperature in the stratosphere. He found that the largest warming after the Pinatubo eruption, of around 2 K after extraction of the QBO signal, occurred in the tropics between 50 and 100 hPa. He has also shown that some cooling in the high

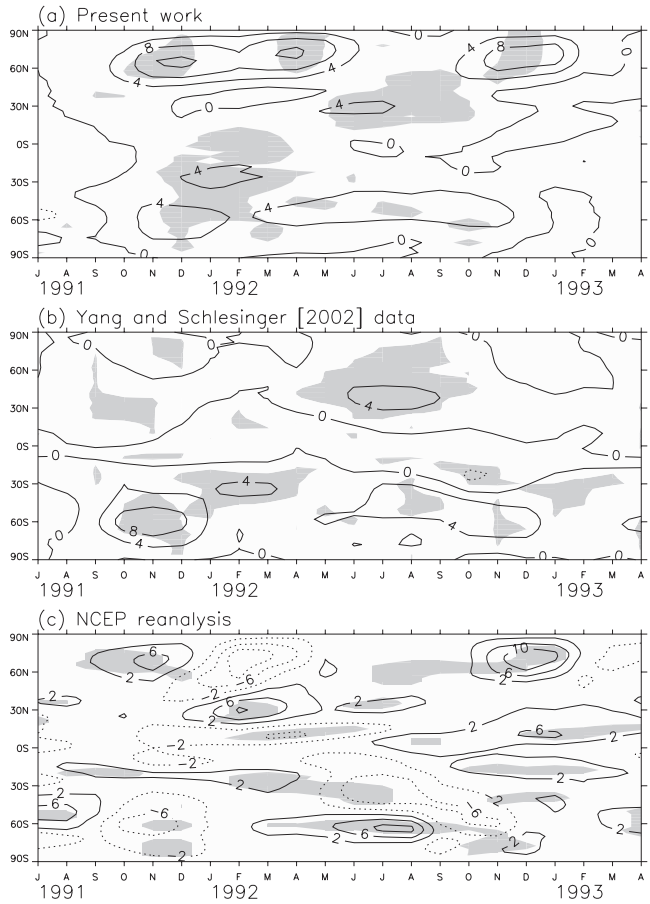

Figure 11. Anomaly of the monthly zonal-mean zonal wind (m/s) at 10 hPa after the June 1991 Pinatubo eruption: (a) present work, (b) *Yang and Schlesinger*'s [2002] results, and (c) obtained from NCEP/NCAR reanalysis. The shaded regions denote for (a) and (b) where the anomalies are judged statistically significant at or better than the 5% level and for (c) where the anomalies exceed one standard deviation.

latitudes of the lower stratosphere took place following the Pinatubo eruption.

[29] These results are in reasonable agreement with the data obtained from the NCEP/NCAR reanalysis. The post-Pinatubo temperature anomalies obtained by *Randel et al.* [1995] from the analysis of satellite data coincide well with anomalies obtained from the NCEP/NCAR data over the northern high latitudes. Both data sets reveal cooling in the lower stratosphere in November–December 1991 and winter 1992/1993. This cooling, albeit not statistically significant, reflects an intensification of the northern PNJ and also occurred in our model simulation results (see Figure 11). *Randel et al.* [1995] also found a temperature increase in this area during spring 1992, confirmed by the analysis of NCEP/NCAR data, which coincides with the deceleration of the PNJ shown in Figure 11. It should be noted that the high latitude warming obtained from the data analysis, presumably connected with the deceleration of the PNJ, should lead to the total ozone increase in spring 1992 which does not appear in the total ozone anomalies obtained by *Randel et al.* [1995]. Instead, they found pronounced ozone depletion, which is typical for the colder stratosphere and strong PNJ. Our model results reveal cooling in the high-latitude lower stratosphere and intensification of the PNJ that is opposite to the observation data. However, the model produces a total ozone decrease, which is consistent with the simulated zonal wind and temperature changes, as well as with the ozone anomalies obtained by *Randel et al.* [1995].

[30] The simulated temperature changes presented here are close to those simulated by *Kirchner et al.* [1999] with the ECHAM-4 GCM and by *Yang and Schlesinger* [2002] with the UIUC ST-GCM forced by a similar radiative aerosol perturbation. However, they are larger than the temperature changes obtained by *Ramachandran et al.* [2000] with the SKYHI GCM using the same data set of aerosol properties. *Kirchner et al.* [1999] pointed out that the ozone depletion effect is able to reduce the magnitude of the simulated warming in the tropical stratosphere by almost 1.5 K if the ozone depletion inside the aerosol cloud is about 20%. However, according to our results the ozone depletion simulated by our model (see Figure 4) in the lower tropical stratosphere does not exceed 4–5%, and this value is not large enough to substantially cool the stratosphere. The agreement of our results with the warming obtained by *Kirchner et al.* [1999] and by *Yang and Schlesinger* [2002] allows us to conclude that the direct radiative heating by the aerosol determines the magnitude of the warming of the lower tropical stratosphere.

[31] To test this conclusion we calculated the instantaneous net radiative heating around 25 km in January 1991 over the northern tropics. The net heating rate due to volcanic aerosol is about 0.3–0.4 K/day, which is rather close to the results published by *Ramachandran et al.* [2000], while the simulated ozone depletion provides radiation cooling of only about 0.05 K/day, which is 6–8 times smaller than the net heating due to the volcanic aerosol. It should be noted that the radiative cooling due to a 20% ozone loss is about 0.2 K/day, which is comparable with aerosol heating. *Kirchner et al.* [1999] showed that the corresponding temperature change caused by such intensive ozone loss could reach 1.5 K, but the observation and modeling studies do not confirm this large ozone depletion after the Pinatubo eruption [e.g., *Rosenfield et al.*, 1997]. Therefore, it is possible that the causes of the disagreement between our simulations and the analyses of the observations could be connected with the parameterization of the aerosol physical and/or radiative properties.

[32] As illustrated in Figure 11, the simulated warming of the stratosphere and the consequent increase in the pole-to-equator temperature difference leads in our model to the intensification of the PNJs. The change in the zonal-wind intensities at 10 hPa are more pronounced (more then 10 m/s) in the Northern Hemisphere than in the Southern Hemisphere and are statistically significant in December 1991 and April and December 1992. For November–December 1991 and winter 1992/1993 the simulated results agree reasonably well with the zonal wind acceleration obtained from the analysis of the NCEP/NCAR data, although the magnitude of the acceleration is slightly overestimated by the model. As discussed earlier, the disagreement in the zonal wind anomalies for spring 1992 is controversial, because the total ozone depletion obtained from the analysis of several independent data sets for spring 1992 implies an intensification of the PNJ and cooling of the lower stratosphere. In the Southern Hemisphere the acceleration of the zonal wind is significant only in December 1991. It should be noted that a statistically significant PNJ acceleration was not obtained in other GCM experiments performed up to now [e.g., *Kirchner et al.*, 1999; *Ramachandran et al.*, 2000] and in the simulations performed by *Yang and Schlesinger* [2002] with a similar model. The difference between these models consists mainly in the modification of the gravity-wave drag scheme, which allowed improvement in the representation of the northern PNJ [*Yang et al.*, 2000; *Rozanov et al.*, 2001], and the interactive treatment of the chemical processes. It seems that a successful simulation of zonal wind changes requires a model that is able to reproduce the observed distribution of the PNJ in the Northern Hemisphere.

5. Northern Hemisphere, December 1991

[33] The simulated changes in the atmosphere after the Pinatubo eruption seem to be most interesting in December 1991, when the warming in the lower tropical stratosphere produced the most pronounced changes in the atmosphere over the northern high latitudes. The latitude–altitude cross sections of the monthly zonal-mean temperature and zonal wind changes for December 1991 are presented in Figures 12 and 13. The results show substantial warming in the tropical lower stratosphere, which increases the latitudinal temperature gradient in the lower and middle stratosphere and produces a significant intensification of the northern PNJ. In comparison with the results obtained by *Kirchner et al.* [1999], the acceleration of the zonal wind in our model is much more pronounced. This can be partially explained by an additional heating in the middle stratosphere related to the ozone increase there (shown in Figure 4). The acceleration of the zonal wind is accompanied by a significant cooling of up to 10 K in the lower stratosphere over the northern high-latitude area and heating of almost the same magnitude in the upper stratosphere.

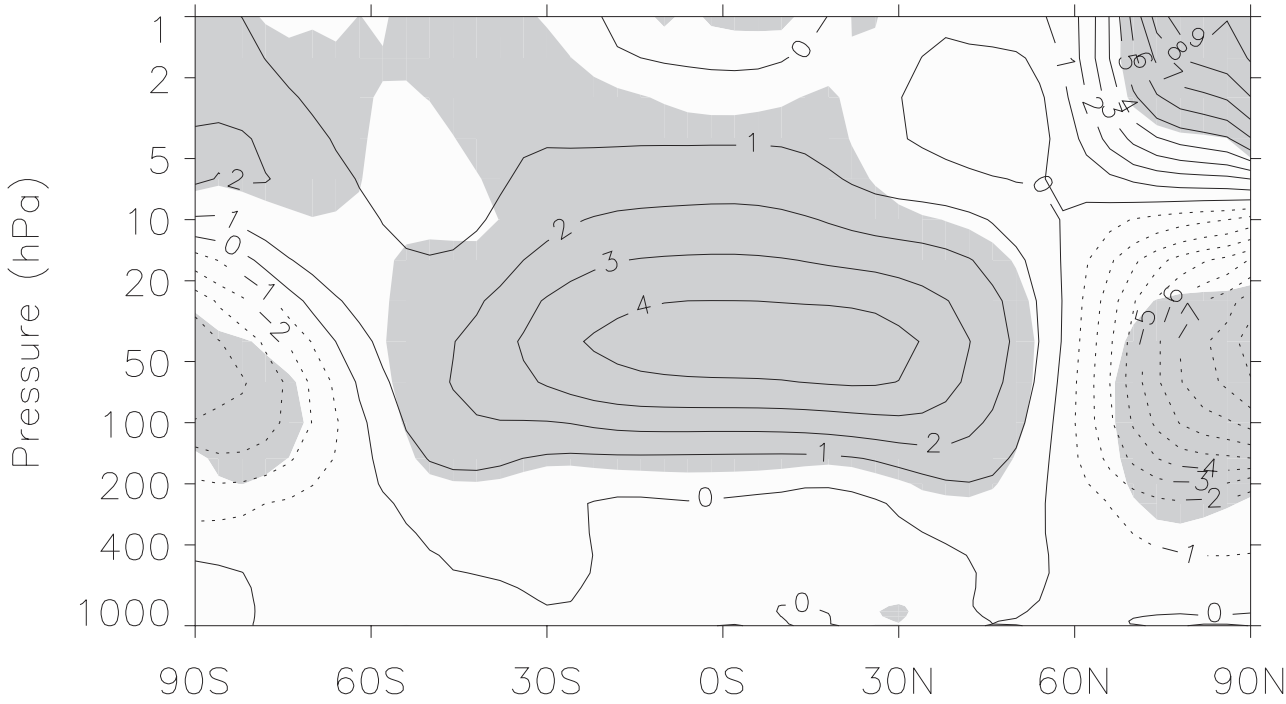

Figure 12. Simulated changes of the monthly zonal-mean temperature (K) for December 1991. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

[34] Figure 14 shows the geographical distributions of the simulated changes due to the Pinatubo eruption in geopotential height, temperature, type I polar stratospheric clouds (PSC-I) mixing ratio and total ozone for December 1991, as well as the ozone mixing ratio in March 1992. Significant geopotential height changes of up to 500 m at 63 hPa confirm the intensification of the polar vortex (see Figure 13). Subsequent cooling of the area inside the polar

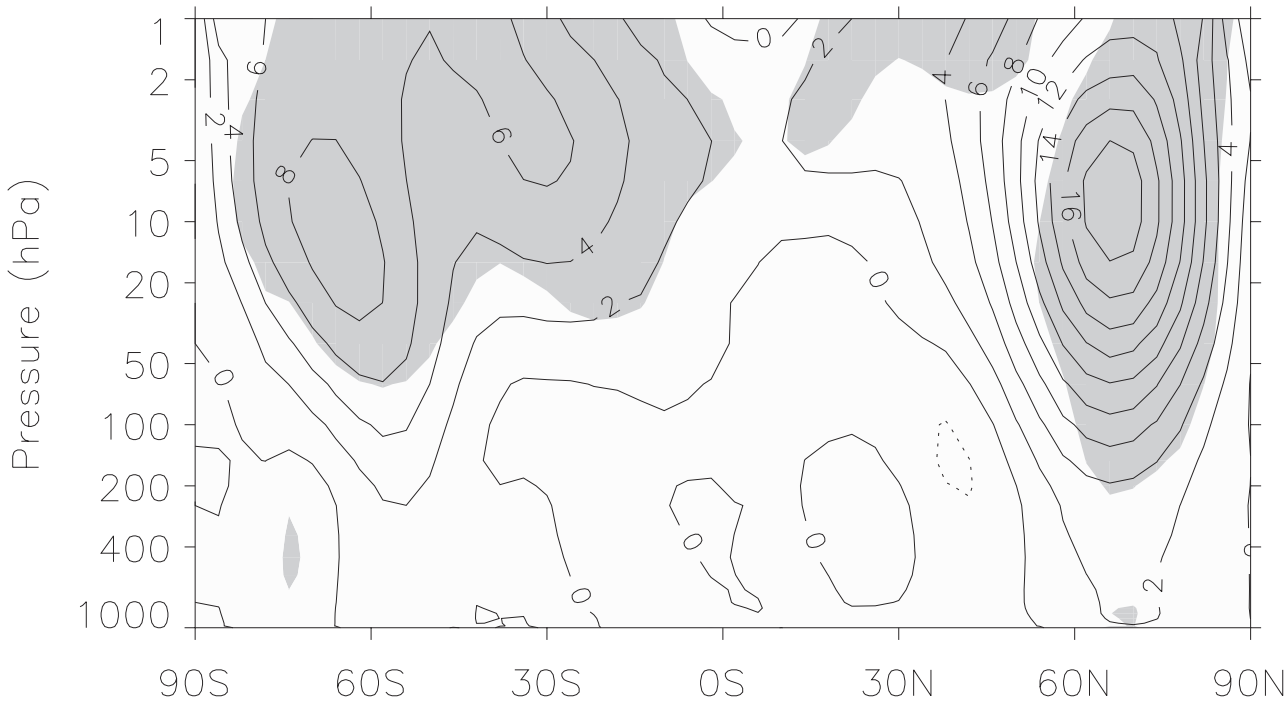

Figure 13. Simulated changes (m/s) of the monthly zonal-mean zonal wind for December 1991. The shading shows the regions where the changes are judged statistically significant at or better than the 5% level.

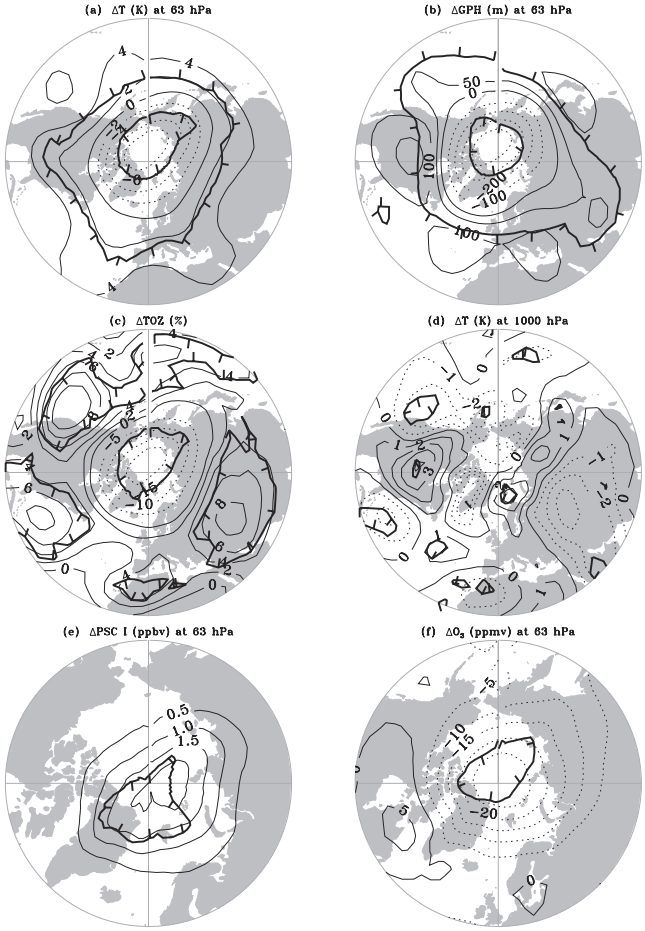

Figure 14. Simulated changes of the monthly mean (a) temperature (K) at 63 hPa, (b) geopotential height (m) at 63 hPa, (c) total ozone (%), (d) temperature (K) at 1000 hPa, and (e) PSC I mixing ratio at 63 hPa for December 1991 and ozone mixing ratio (%) at 63 hPa for March 1992. The area inside the ticked line shows the regions where the changes are judged statistically significant at or better than the 5% level.

vortex and the total ozone depletion there are also evident in Figure 14. A colder environment facilitates an intensification of the PSC formation in the high latitude area. The simulated increase of PSC-I is statistically significant over the polar area and reaches 2 ppbv. The PSC increase provides an additional medium for the chlorine activation and this effect, together with decreased intensity of the meridional ozone transport, determines the ozone mixing ratio decrease in March 1992. The data obtained from this particular model run do not allow separating the effects of different mechanisms and we are going to address this issue in follow-up papers. The ozone depletion reaches 25% and spans a wide area over the northern part of Canada, Russia, and Europe; however, this effect is statistically significant only over the polar area.

[35] The intensification of the zonal wind also could lead to changes in the tropospheric planetary-wave propagation. It was pointed out by many authors [e.g., *Robock*, 2000, and references therein] that such changes are responsible for the winter warming of the surface air in the Northern Hemisphere after a major volcanic eruption. The changes of the simulated near-surface air temperature in December 1991 (Figure 14d) confirm that our model is capable of reproducing this effect. Analysis of the temperature change reveals a very good agreement of the simulated temperature anomalies with the observed anomalies presented by *Robock* [2000]. During the winter, some statistically significant warming near the surface can be seen in the model results over North America, northern Europe, and Siberia. The model also captures well the cooling over Greenland, Alaska and Central Asia.

6. Conclusions

[36] We have presented here an analysis of the changes of the chemical composition, temperature and dynamics of the atmosphere caused by the volcanic stratospheric sulfate aerosol formed after the massive Pinatubo volcanic eruption in June 1991. The study has been performed with the UIUC ST-GCM/PC.

[37] The changes of the total vertical column amounts of HNO_3, NO_2, HCl, and ClO are in reasonable agreement with observations and theoretical expectations. The simulated ozone changes due to the Pinatubo volcanic eruption show that the ozone mixing ratio in the tropics tends to decrease below 28 km and increase in the stratosphere above this level; however, the simulated ozone changes are found not to be statistically significant in the lowermost stratosphere. The model also simulates a total ozone loss of up to 15% over the northern middle and high latitudes during winter and spring, but the magnitude of the total ozone depletion is generally less than that obtained from the observed total ozone anomalies.

[38] It should be noted that the total ozone decreases in the northern extratropics for three members of the ensemble are in better agreement with the observed total ozone anomalies, while the remaining two members generate a less pronounced total ozone loss or even some ozone increase during the spring season. This allows the conclusion that the magnitude and even sign of the simulated aerosol effects depend significantly on the background state of the model. Therefore, a numerical experiment to study the effects of any particular volcanic eruption should include more ensemble members. Those members of the ensemble which produce unrealistic changes should then be excluded on the basis of a priori information obtained from the observational data. This procedure will allow more robust estimations to be obtained of the changes in ozone and other quantities. However, this problem cannot be addressed now because of the controversies in the anomalies of different quantities obtained from the data analysis. We discussed in section 4 that for spring 1992 the NCEP/NCAR and MSU data analysis [*Randel et al.*, 1995] showed a deceleration of the PNJ and warming in the lower stratosphere. These processes should lead to the total ozone increase over the high latitudes; however, the analysis of the total ozone data provided by *Randel et al.* [1995] showed its depletion in this particular area and season. As has been shown in Figure 8, the above mentioned temperature and zonal wind anomalies are matched well by the first and fourth ensemble members, however, the ''observed'' total ozone change coincides better with the second, third and fifth ensemble members.

Therefore, the problem is what data should be used for the correct choice of the ensemble members?

[39] The model simulates a pronounced stratospheric warming in the tropics; however, the simulated warming exceeds that derived from the observations anomalies by 1–2 K. As expected, the simulated ozone changes are not homogeneous. That the ozone depletion in the lower stratosphere accompanies the even-more significant ozone increase in the middle stratosphere is one of the surprises of the interactive model. The model matches well the overall intensification of the PNJ obtained from the data analysis, which is most pronounced in December 1991 and winter 1992/1993, the boreal winter warming of the surface air over the United States, northern Europe, and Russia, and the cooling over Greenland, Alaska and Central Asia. In the work presented by *Yang and Schlesinger* [2002], the acceleration of the PNJ and the resultant winter warming are absent. In the similar experiment reported here, the acceleration of the zonal wind and the winter warming are clearly seen and are statistically significant. Therefore, a successful simulation of this feature requires a model that is able to reproduce the observed distribution of the PNJ in the Northern Hemisphere.

[40] It is also noteworthy that the simulation of winter warming and the change in total ozone are rather sensitive to the background conditions, that is, they depend on the background state of the atmosphere in the control run. In our experiment, four ensemble members did reveal winter warming in 1991, while one ensemble member did not.

[41] The model applied here is not capable of simulating the QBO, which is believed to play a significant role in the atmosphere and could alter the magnitude of the results obtained here. We plan to improve this aspect of the model and revisit this issue with the new model version as soon as it is ready.

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