

Effects of interannual variation of temperature on heterogeneous reactions and stratospheric ozone

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Abstract. A two-dimensional chemical/dynamical/microphysical model is used to calculate the effect of temperature interannual variability on stratospheric ozone. Two effects associated with temperature variations are considered in this paper: First, the effect on the rate coefficient of heterogeneous reactions occurring at the surface of sulfate aerosols, in particular for the enhanced sulfate aerosols following the eruption of Mount Pinatubo, and second, the effect of temperature interannual variations on the formation of polar stratospheric clouds (PSCs) and the spring Antarctic ozone depletion. The interannual variabilities of ozone concentration affected by both the processes are evaluated. The model results show that during winter 1992–1993, stratospheric temperature at high latitudes in the northern hemisphere is colder than during winter 1991–1992, leading to a larger rate of heterogeneous conversion from inactive chlorine to active chlorine in sulfate aerosols during winter 1992–1993 than during winter 1991–1992 at northern high latitudes. As a result, ozone reduction due to heterogeneous reactions following the eruption of Mount Pinatubo is enhanced in a colder winter (1992–1993) and reduced in a warmer winter (1991–1992). This result indicates that the enhanced heterogeneous conversion associated with colder temperature can explain, in part, the large ozone depletion observed by the total ozone mapping spectrometer (TOMS) experiment during winter 1992–1993. The model results also show that during winters 1986 to 1990, the formation of PSCs over Antarctica exhibits a strong interannual variation, due to temperature variability. In the colder years (1987 and 1989) the surface area of PSCs (a major factor to determine the rate of heterogeneous conversion from inactive chlorine to active chlorine) increases, and ozone concentration decreases. In the warmer years (1986 and 1988) the results are opposite. The calculated ozone interannual variation is consistent with the satellite observation (TOMS), indicating that the interannual variation of the formation of PSCs likely plays an important role for the interannual variability of the spring Antarctic ozone hole.

Introduction

Observations made after the eruption of Mount Pinatubo (Philippines) in June 1991 have revealed stratospheric ozone concentrations lower than the long-term normal value. Dobson ozone spectrophotometer data reported by Komhyr *et al.* [1994] shows that from January to April 1993, ozone concentrations were 12.6% below normal between 45°N and 65°N latitudes. Total ozone mapping spectrometer (TOMS) data reported by Herman and Larko [1994] shows that ozone amounts were about 7% to 12% below historical values at mid to high latitudes in the northern hemisphere. Hofmann *et al.* [1994] reported that balloon-borne measurements of ozone concentrations at Boulder, Colorado, and Wallops Island, Virginia, were 25% below normal during winter and spring of 1992–1993 in the 12–22 km region. Model calculations suggest that this large ozone depletion at mid to high latitudes in the northern hemisphere during winters 1991–1992 and 1992–1993 could be attributed to heterogeneous reactions occurring at the surface of volcanic sulfate aerosols after the eruption of Mount Pinatubo [Brasseur and Granier, 1992; Pitari, 1993; Rodriguez *et al.*, 1994; Bekki and Pyle, 1994; Tie *et al.*, 1994a].

Although the enhanced sulfate aerosol loading at mid to high latitudes in the northern hemisphere in winter 1992–1993 is lower

than during winter 1991–1992 [McCormick *et al.*, 1995; Osborn *et al.*, 1995], measurements show that the reduction in column ozone is approximately 3 to 6% larger in winter 1992–1993 than in winter 1991–1992 [Bojkov *et al.*, 1993; Gleason, *et al.*, 1993; Kerr *et al.*, 1993; Planet *et al.*, 1994; Randel *et al.*, 1995]. At the same time, temperature at mid to high latitudes in the lower stratospheric was substantially colder during the second winter (approximately 8 K degree cooler in early 1993 than in early 1992) [Randel *et al.*, 1995]. According to laboratory measurements [Hanson *et al.*, 1994], the rate of heterogeneous reactions, for example, the conversion from ClONO₂ to HOCl, increases significantly when temperatures decrease. In the first part of this paper we will evaluate the impacts of interannual variations of stratospheric temperature on the heterogeneous process as well as on ozone concentrations during winters of 1991 to 1993.

After the discovery of the spring Antarctic ozone depletion in the early 1980s [Farman *et al.*, 1985], the importance of polar stratospheric clouds (PSCs) on the chemistry of the lower polar stratosphere has been emphasized [Solomon *et al.*, 1986; McElroy *et al.*, 1986; Solomon, 1990]. It is now commonly accepted that PSC particles provide important sites for heterogeneous reactions to occur, causing the spring Antarctic ozone depletion. The long-term (20 years) observations of ozone concentration over Antarctica [Jiang *et al.*, 1996; Herman *et al.*, 1995] show that springtime ozone depletion increased significantly during the 1980s, especially after major volcanic eruptions [Portmann *et al.*, 1996]. Furthermore, beside the steady decreasing trend, a strong interannual variation of the Antarctic ozone hole has also been observed [Jiang *et al.*, 1996; Herman *et al.*, 1995], particu-

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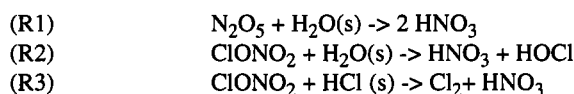
Paper number 97JD01556.

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larly during winters 1986 to 1989 (the depth and extent of the ozone hole has been relatively constant in the 1990s). In the second part of this paper, we will investigate the effect of temperature interannual variation on the formation of PSCs as well as the spring Antarctic ozone depletion during the years 1986 to 1989.

Description of Calculation

The model used in the present study is based on a two-dimensional chemical and dynamical model developed by *Brasseur et al.* [1990], coupled with detailed microphysical processes related to sulfate aerosols and PSCs formation [*Tie et al.*, 1994a, b; *De Rudder et al.*, 1996; *Tie et al.*, 1996]. In addition to the traditional gas phase chemical reactions, heterogeneous reactions at the surface of sulfate aerosol and PSC particles are considered in the chemical scheme:



Here (s) indicates that the reactants are on the surface of particles. A value of 0.1, independent of temperature, is used for the reaction probability for (R1), but the reaction probabilities for (R2) and (R3) varies strongly with the aerosol composition and hence with the ambient temperature and water abundance [*Tolbert et al.* 1988; *Hanson and Ravishankara*, 1991; *Hanson et al.*, 1994]. The reaction probabilities of (R2) and (R3) are adopted in the model according to the measurements of *Hanson et al.* [1994] and calculated using National Meteorological Center (NMC) temperature analyses. The temperature data analyzed by *Randel* [1992] is zonally averaged from 85°S to 85°N in latitude and from the surface to 45 km in altitude. The distribution and time evolution of sulfate aerosol surface area before (background aerosols) and after (enhanced aerosols) the eruption of Mount Pinatubo are calculated according to the sulfate aerosol microphysical model developed by *Tie et al.* [1994a, b]. This model has previously been used to study ozone perturbations after the eruption of Mount Pinatubo [*Tie et al.*, 1994a].

In order to identify the effect of interannual variation of temperature on heterogeneous conversions on the sulfate aerosols, a temperature field averaged over 4 years between 1991 and 1994 is used in the model to calculate reaction probabilities for a baseline calculation (case A). The seasonally varying temperatures during 1991 to 1994 have been used (case B) to compare with the baseline calculation. The background and enhanced aerosol loadings are used in both cases to calculate the effect of volcanic aerosols on stratospheric ozone associated with the presence of interannual variation of heterogeneous conversions. The formation of PSCs and its effect on ozone concentration is not included in this part calculation in order to evaluate the effect of heterogeneous reactions on the sulfate particles.

In the second part of the study we use a detailed PSCs microphysical model coupled with the two-dimensional (2-D) chemical/dynamical model to study the interannual variability of PSC formation and its effect on ozone concentration over Antarctica. The model has been previously used to study the formation of PSCs and the Antarctic ozone hole. The detailed model descriptions and results are reported by *Larsen* [1991], *De Rudder et al.* [1996], and *Tie et al.* [1996]. Some previous studies have shown that interannual variation of the Antarctic ozone hole could be attributed to several factors (i.e., dynamical transport, radiative feedback, and quasi-biennial oscillation (QBO)) [*Garcia and*

Solomon; 1987, *Schneider et al.*, 1991; *Mancini et al.*, 1991]. *Mancini et al.* [1991] also studied the issue of how the interannual variability of PSCs, which is produced by the QBO, may affect the ozone hole. In this study we will emphasize the effect of the interannual variability of PSCs, produced by temperature interannual variations, on the Antarctic ozone hole. To assess this effect, a 4-year averaged NMC temperature field (from June 1986 to July 1990) is used for a baseline calculation of the formation of PSCs (case C), and the interannual-varying temperature between June 1986 and July 1990 (case D) is used to compare with the baseline calculation. The period of 1986 to 1990 is chosen for this study because a very strong interannual variation of temperature occurs in the lower stratosphere throughout winter and spring. In order to account for longitudinal variations in the temperature (planetary waves) and the possible processing by PSC air masses flowing through localized cold environments (while the zonal mean temperature may be higher than the PSCs thresholds), we have superimposed a fluctuation temperature (temperature wave) to the zonal mean temperature. The details for the use of temperature variability is described by *DeRudder et al.* [1996].

Finally, because ozone is a radiative species [*Ramanathan and Dickinson*, 1979], the interannual variation of ozone concentration over Antarctica will produce changes in the radiative and dynamical processes which lead to additional perturbations to the ozone distribution. This feedback effect will also be evaluated in this study.

Results and Discussions

Interannual Variation of Heterogeneous Reactions on Sulfate Aerosols and Ozone

To evaluate the effect of interannual variation of heterogeneous reactions on the surface of sulfate aerosols on ozone concentrations, we first calculate the change in column ozone in the northern hemisphere from 1991 to 1994 after the eruption of Mount Pinatubo if no interannual variation of the temperature is taken into account (case A). In this case the interannual variability of heterogeneous conversions is mainly determined by the time evolution of sulfate aerosol loadings. In Figure 1 the top panel shows the ozone anomaly (percent) (compared to the ozone concentration with background aerosol) following the eruption of Mount Pinatubo observed by TOMS (the effects of QBO are excluded from these data, following the analysis of *Randel et al.* [1995]), and the lower panel shows the calculated result. The comparisons indicate that the calculated ozone changes due to the enhancement of heterogeneous conversions following the eruption of Mount Pinatubo reproduces most, but not all, of the general features deduced from satellite observations. It predicts, for example, the reduction in total ozone at high latitudes by 14% during winter 1991-1992 and winter 1992-1993, while observed ozone decreases are 10% during winter 1991-1992 and 14% during winter 1992-1993. Both calculated and observed ozone depletions are much less during winter 1993-1994 as a result of the significant decrease in sulfate aerosol loading [*McCormick et al.*, 1995; *Solomon et al.*, 1996]. The calculated ozone decrease during 1991-1992 and 1992-1993 are mainly due to chlorine activation on the surface of aerosols. Figure 2 shows the time evolution of chlorine species at 70°N and at 20 km from November 1992 to March 1992. It indicates that chlorine reservoirs (ClONO₂+HCl) decrease at high latitude during winter (top panel), while other relative chlorine species ClO_y

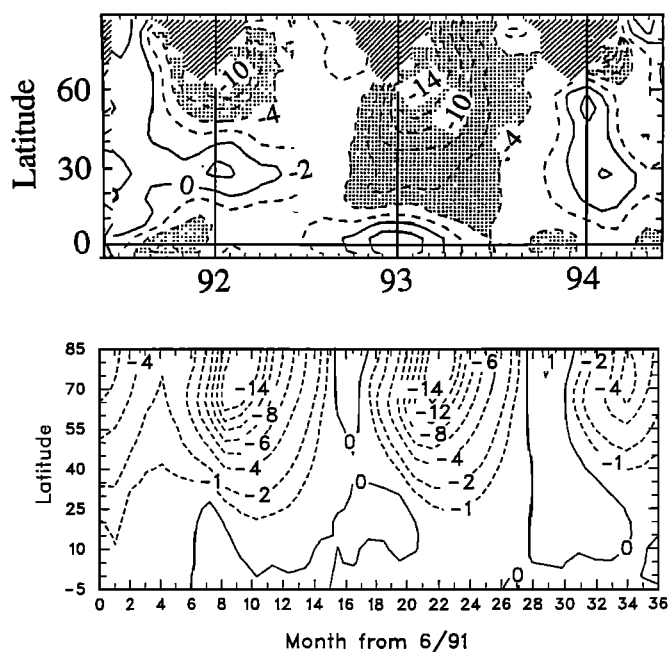


Figure 1. (top) Total ozone anomaly (percent) observed from total ozone mapping spectrometer (TOMS) [Randel *et al.*, 1995], and (bottom) calculated total ozone changes (percent) due to heterogeneous reactions occurring at the surface of enhanced sulfate aerosols following the eruption of Mount Pinatubo in the northern hemisphere. Reaction probabilities are calculated using the 1991-1994 mean temperature (case A).

(Cl+ClO+HOCl+OCIO+2Cl₂O₂+2Cl₂) increase (middle panel) as the result of heterogeneous chemical reactions. Because there is almost no sunlight between November and January, ClO concentration is very low. After sunlight returns in February, some ClO_y species are photo dissociated and are converted into Cl and ClO (bottom panel), and the increase in ClO concentration will lead to an ozone concentration decrease.

The calculated ozone decreases in this study are larger than the previous calculation of Tie *et al.* [1994a]. The reason is that the current study has included the effects of zonal temperature (wave) variations on the rate of heterogeneous reactions. Pitari [1993] shows that the rate of heterogeneous reactions may differ by more than 1 order of magnitude when it is calculated using the zonally averaged temperature field, compared to the zonal average of heterogeneous rates calculated using the actual zonally varying temperature distribution. To evaluate such an effect and to include it in the zonally averaged 2-D model, a correction factor R_c is calculated and is expressed by

$$R_c = [R(T)]/R([T])$$

where brackets denote zonal average. Thus R_c is a ratio of zonal average of heterogeneous reaction rates calculated from United Kingdom Meteorological Office temperature field to the rates calculated directly from zonal average of the temperature field. Figure 3 shows the results of R_c for reaction (R2). The upper panel shows the latitude-height distribution in January. It indicates that the maximum of R_c is about 10 at high latitude and at 25 km. The lower panel shows that R_c at 20 km is seasonally dependent. The maximum R_c occurs in winter when the planetary wave is strong, while the minimum R_c takes place in sum-

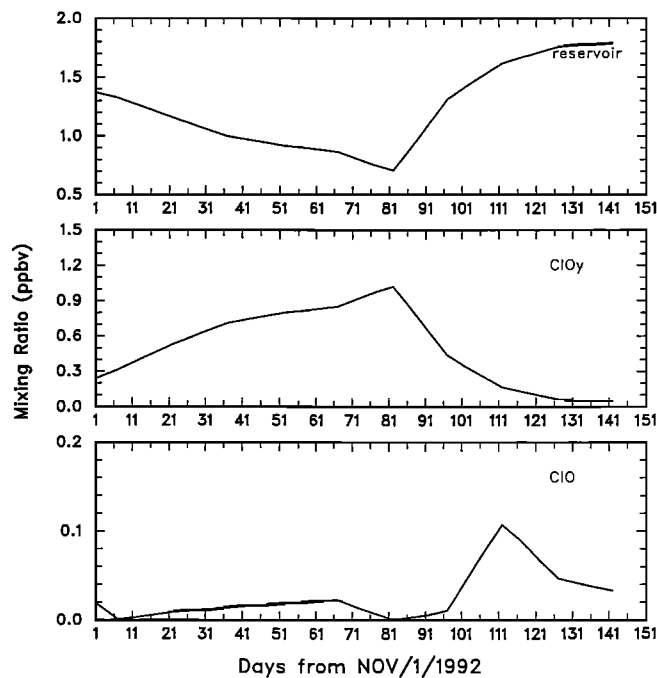


Figure 2. Calculated time evolution of chlorine species at 70°N and at 20 km during winter 1992-1993: (top) Chlorine reservoir (ClONO₂+HCl), (middle) chlorine species other than chlorine reservoir (ClO_y), and (bottom) ClO. Abbreviation ppbv, parts per billion by volume.

mer when the planetary wave is weak. The rates of heterogeneous reactions are obtained by multiplying by the correction factor in the model.

Despite the fact that the model reproduces reasonable large ozone decreases during winter 1991-1992 and winter 1992-1993,

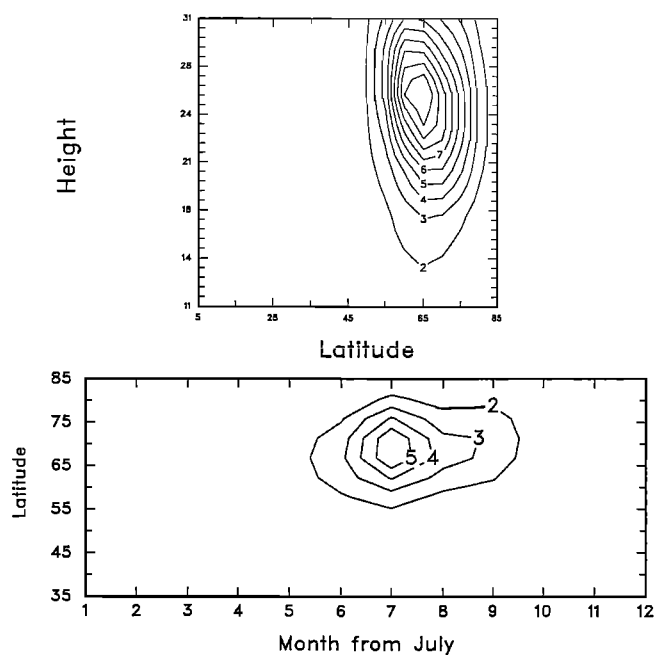


Figure 3. Calculated correction factor R_c for the heterogeneous reaction (R2). (top) Latitude-height distributions in January and (bottom) time evolution at 20 km between latitude 45°N and 85°N.

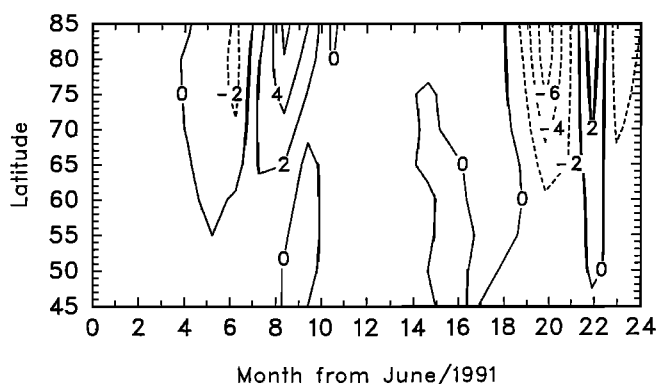


Figure 4. Temperature (NMC) anomaly (Kelvin) during winter 1991-1992 and winter 1992-1993. The temperature anomaly is calculated by the difference between NMC temperature and the averaged temperature during 1991 to 1994.

discrepancies are also noticeable. The observed ozone decrease shows substantial interannual variability, while the calculation produces a small interannual variation. The observed ozone depletion during winter 1992-1993 was larger (14%) than that during winter 1991-1992 (10%). At the same time, observed temperature in the lower stratosphere was approximately 6 to 8 K colder in winter 1992-1993 than in winter 1991-1992 (see Figure 4). This colder temperature produces a significant enhancement for the rate of heterogeneous conversions from inactive chlorine to active chlorine. Figure 5 shows that temperature at 70°N and at 20 km in 1992-1993 winter (case B) is approximately 8 K colder in maximum than the averaged temperature between 1991 and 1994 (case A). As a result, the rate of heterogeneous conversion from ClONO_2 to HOCl at 70°N is approximately 5 times higher in case B than the rate calculated in case A during winter 1992-1993. Figure 6 shows the effect of interannual variations in tem-

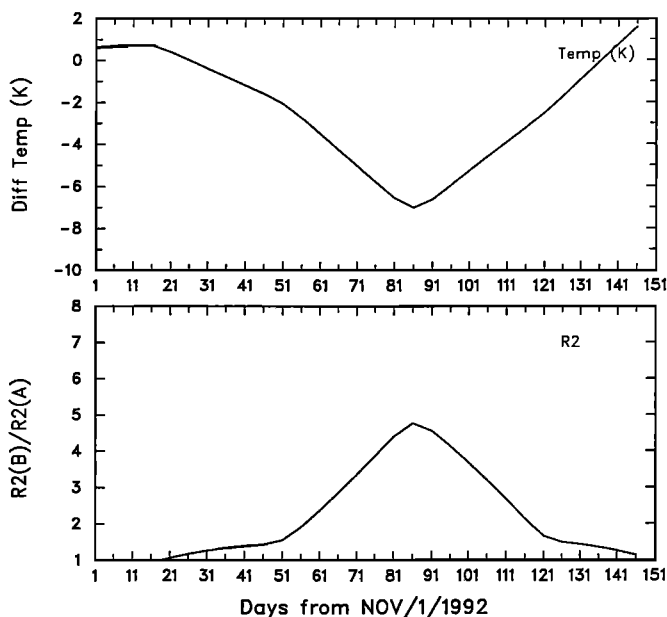


Figure 5. (top) Temperature anomaly (Kelvin) during winter 1992-1993 at 70°N and at 20 km and (bottom) the corresponding changes in the rate (ratio between case B and case A) of heterogeneous reaction (R2).

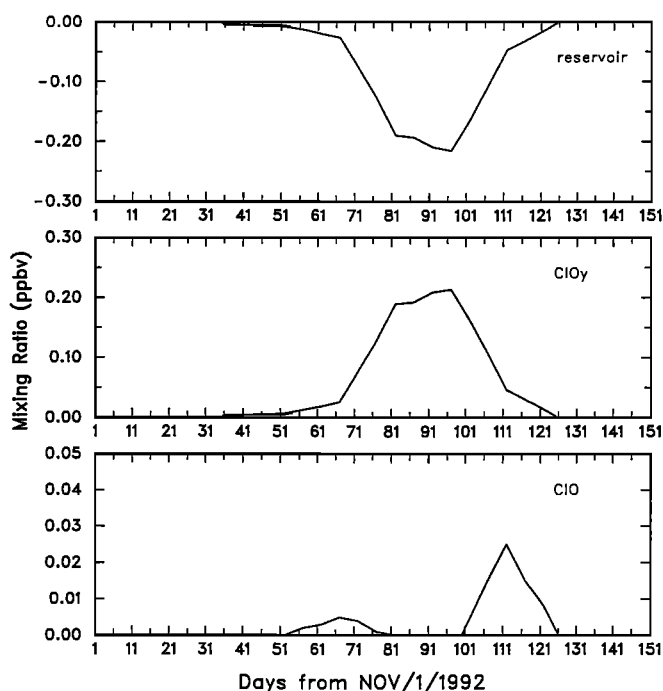


Figure 6. Calculated changes in chlorine species at 70°N and at 20 km as the result of interannual variation of heterogeneous conversion included in the model (difference between case B and case A): (top) Chlorine reservoir ($\text{ClONO}_2 + \text{HCl}$), (middle) chlorine species other than chlorine reservoir (ClO_y), and (bottom) ClO .

perature on chlorine species at 70°N and at 20 km. It indicates that the colder temperature in winter 1992-1993 produces a decrease in chlorine reservoir (top panel) and a increase in ClO_y (middle panel), indicating a enhanced heterogeneous conversion from chlorine reservoir to ClO_y . After sunlight increases in February, the increased ClO_y produces an increase in the ClO concentration (bottom panel), leading to a increase in ozone reduction.

When this enhancement in heterogeneous conversion is taken into account in the model, an additional 3% decrease in column ozone abundance occurs at high latitudes during winter 1992-1993 (see Figure 7, top). In contrast, the temperature in winter 1991-1992 is warmer in case B than in case A, leading to a 3% reduction in ozone depletion. By including the effect of temperature interannual variability on heterogeneous reactions in the model (case B), the calculated ozone reduction is now larger in winter 1992-1993 (16%) than in winter 1991-1992 (12%), which is a better agreement with the observed ozone depletion.

Interannual Variation of PSCs and Ozone

During winters 1986 to 1990, a strong temperature interannual variation over Antarctica occurred in the lower stratosphere. Figure 8 shows the temperature anomaly at 17 km from the mean values observed between 1986 and 1990. In August and September, at temperatures near the threshold for nitric acid trihydrates (PSC I) formation, the temperature is approximately 2 K warmer than average in 1986 and 1988, while it is 2 K colder in 1987 and 1989. As a consequence, a strong interannual variation in PSC I formation occurred (see Figure 9). During warmer years (1986 and 1988), PSC I surface area at 17 km over Antarctica is

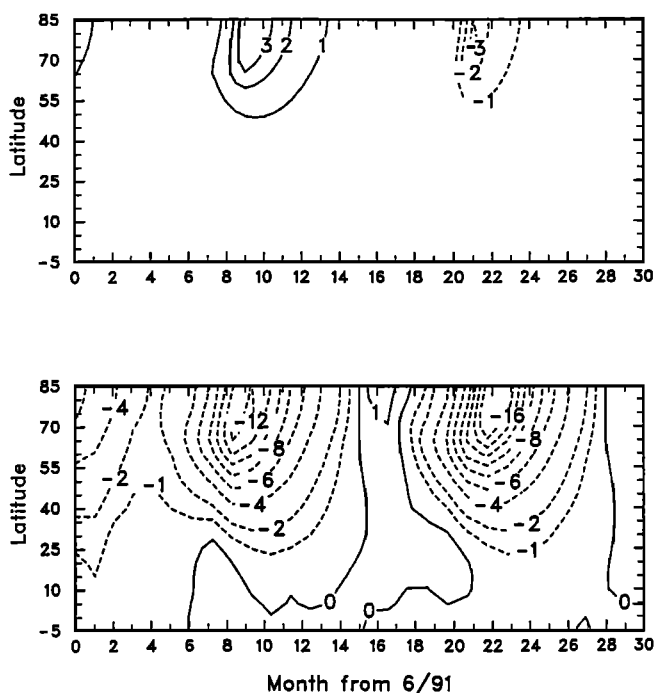


Figure 7. (top) Calculated changes in column ozone (percent) due to the interannual variation in heterogeneous reactions under volcanic aerosol condition. (bottom) Calculated total ozone changes (percent) due to the increase in sulfate aerosols following the eruption of Mount Pinatubo when interannual variation in heterogeneous reactions are taken into account in the calculations (case B).

approximately $4 \mu\text{m}^2/\text{cm}^3$ lower than the values with averaged temperature is applied for PSC calculation (case C), while it is approximately 2 to $4 \mu\text{m}^2/\text{cm}^3$ larger during colder years (1987

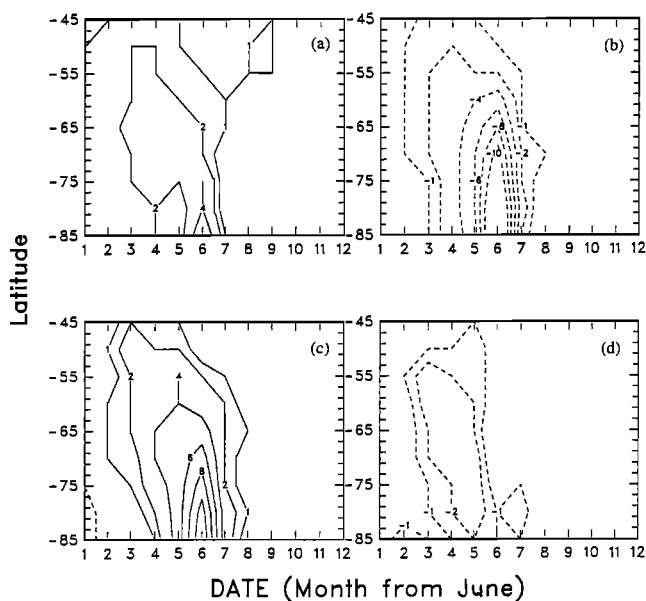


Figure 8. Time and latitude cross section of the temperature (NMC) anomaly (Kelvin) during 1986-1990 from the mean values from 1986 to 1990 at 17 km: (a) 1986-1987, (b) 1987-1988, (c) 1988-1989, and (d) 1989-1990.

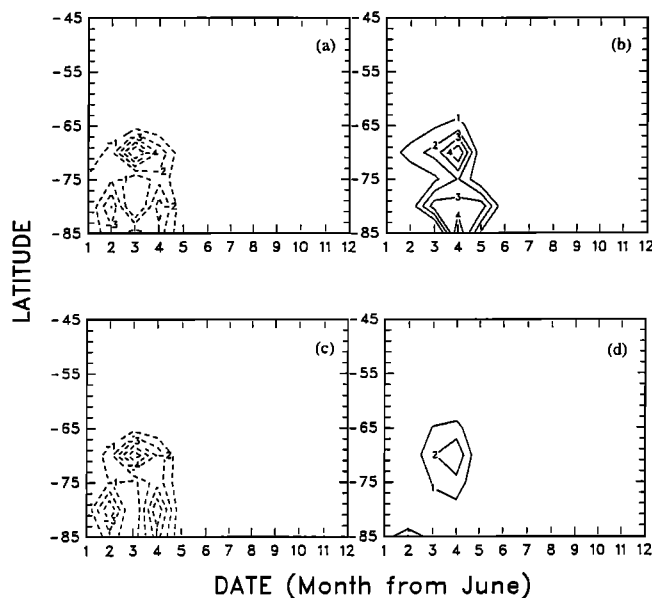


Figure 9. Calculated differences in PSC I surface area ($\mu\text{m}^2/\text{cm}^3$) between case D (actual temperature (NMC) is applied for calculating PSC formation) and case C (mean temperature is applied for calculating PSC formation) at 17 km between 1986 and 1990.

and 1989). Detailed results concerning the calculation of PSC I surface area are given by *De Rudder et al.* [1996]. As a result, during 1986 and 1988, the lower PSC I surface area reduces the heterogeneous conversion from inactive chlorine (ClONO_2 and HCl) into active chlorine (ClO and Cl), and column ozone over Antarctica is larger by up to a maximum value of 40 to 50 Dobson units (DU) (approximately 20 to 30% increase compared to case C). However, when PSC I surface area increases during 1987 and 1989, ozone decreases by a maximum of 30 to 40 DU (see Figure 10).

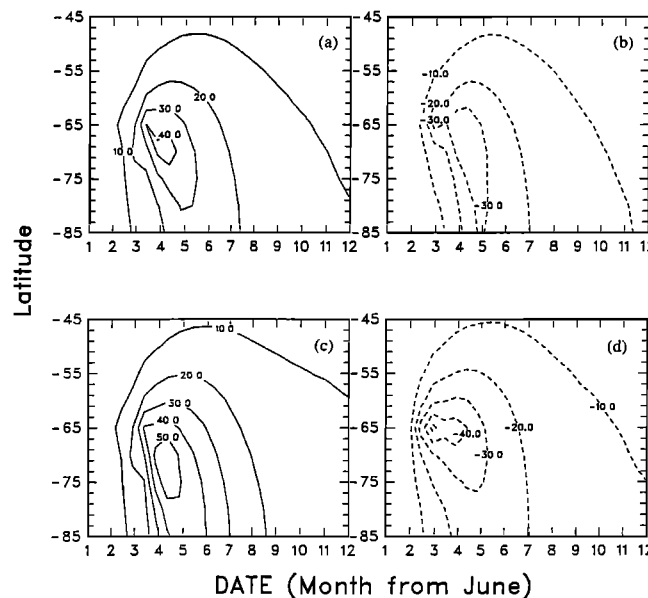


Figure 10. Same as Figure 9, but for differences in column ozone (Dobson Units).

It can be noted that the maximum increase or decrease in ozone concentration occurs at the edge of vortex (65°S to 70°S) and in very late spring (late September). It may indicate that interannual variations in temperature lead to PSC I forming for a longer time and covering a larger region over Antarctica. To exam such effect on ozone concentrations, a net change in PSC I surface area has been calculated. The net change is defined to be the following:

$$\begin{aligned} \text{net change} &= A - B \text{ (when and } B=0 \text{ during colder years)} \\ \text{net change} &= A - B \text{ (when and } A=0 \text{ during warmer years)} \end{aligned}$$

where A is calculated PSC I surface area ($\mu\text{m}^2/\text{cm}^3$) when observed temperature from 1986 to 1990 is used, and B is calculated PSC I surface area ($\mu\text{m}^2/\text{cm}^3$) when averaged temperature from 1986 to 1990 is used. The net change is expressed by an increase or a decrease in the PSC surface area when a contrast PSC surface area (A or B) is 0, so that the net change is a change if either the global extent or temporal duration of PSC occurrence changes.

The net change in PSC I surface area are shown in Figure 11. The maximum net change in the PSC I surface area is less than $1 \mu\text{m}^2/\text{cm}^3$, which is smaller than the PSC I interannual variations shown in Figure 9. However, a comparison of Figures 10 and 11 shows that the maximum changes in column ozone are correlated with the net changes in PSC I surface area, indicating that a longer duration or a larger regional covering of PSCs affects ozone concentration more effectively than the relative changes in PSC surface area density. The reason is that if PSCs are already formed, heterogeneous conversions over Antarctica are almost saturated, and further increase or decrease in PSCs does not significantly affect heterogeneous conversions and ozone concentration [Tie et al., 1996; Portmann et al., 1996].

Figure 12 shows the calculated column ozone anomaly at 70°S in Dobson units between June 1986 and May 1990 due to the interannual variation of PSC surface area and observed (TOMS)

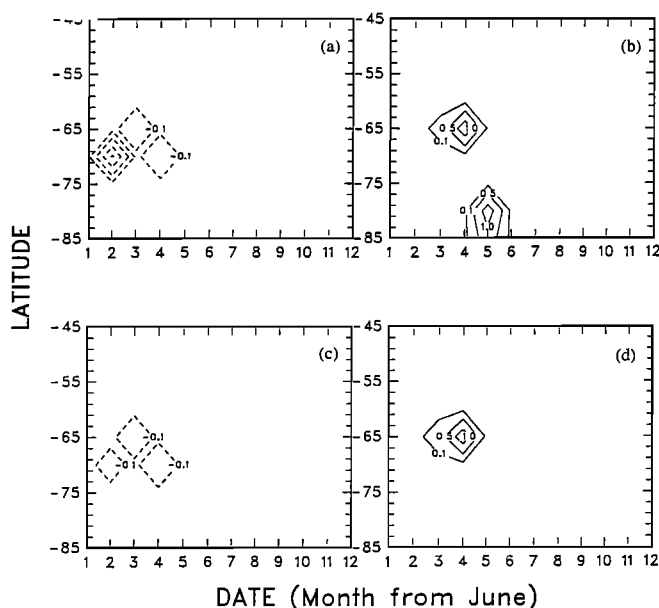


Figure 11. Same as Figure 9, except that the changes are calculated when PSC I is not formed in case C (mean temperature is applied for calculating PSC formation).

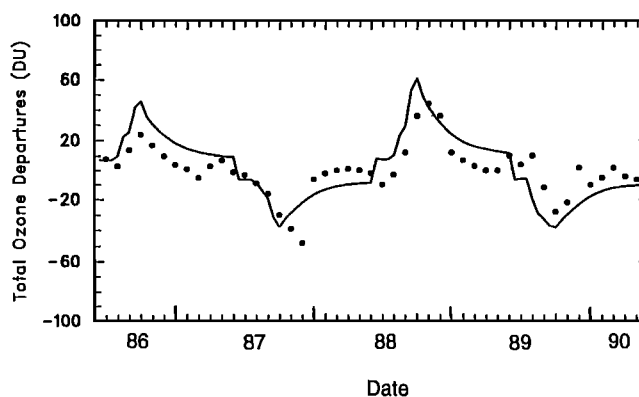


Figure 12. Calculated and observed interannual and seasonal variabilities in column ozone (Dobson Units) at 70°S between 1986 and 1990. The circles represent the observed ozone anomalies by TOMS, and the solid line is calculated by the model.

ozone interannual variations deduced from the mean value between 1986 and 1990. During this period, both calculation results and observations show a strong interannual variation in ozone column, and the calculated results agree well with the observations (TOMS). The strongest ozone anomaly occurs in October when the spring Antarctic ozone depletion is at its maximum [Solomon, 1990]. After November, the magnitude of the ozone interannual variation is reduced, as dynamical mixing starts to play a significant role. This seasonal feature suggests that the interannual variation of dynamical mixing after the vortex breaking is not a major cause for the interannual variability in total ozone concentration observed from 1986 to 1990, which would lead to a maximum interannual variation after December. The agreement between observed and calculated annual and seasonal variabilities in ozone concentration suggests that the considered processes (changes in temperature causing to changes in PSC surface area, which lead to changes in active chlorine, and

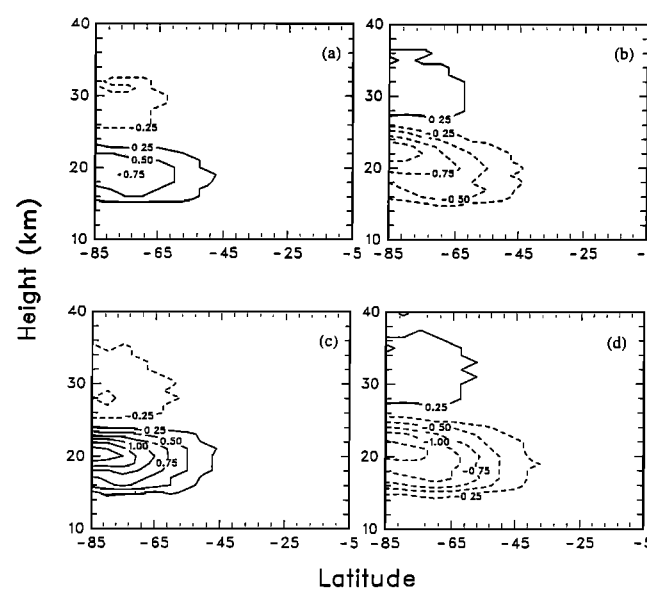


Figure 13. Calculated changes in temperature (Kelvin) resulting from ozone concentration interannual variability from 1986 to 1990 in October: (a) 1986, (b) 1987, (c) 1988, and (d) 1989.

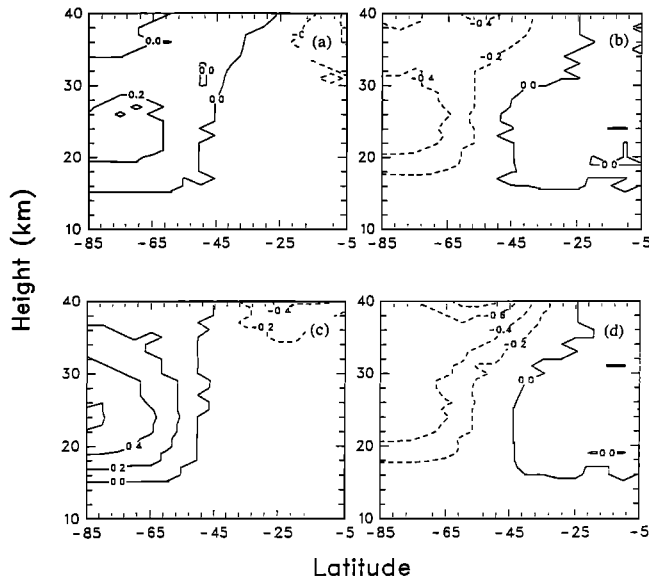


Figure 14. Same as Figure 10, but for differences in vertical motion (m/d). Positive values represent upward motion, and negative values represent downward motion.

further to changes in ozone concentration) likely play a very important role in the observed interannual variability of ozone over Antarctica from 1986 to 1990.

As ozone is an important radiative chemical species [Ramanathan and Dickinson, 1979], changes in ozone concentration in the lower stratosphere could produce perturbations in the radiative forcing. As discussed by Kiehl *et al.* [1988] and Mahlman *et al.* [1994], the Antarctic ozone hole formation could lead

to a 2 to 4 K decrease in stratospheric temperature between 10 and 200 mbar as a result of radiative cooling, as well as a 2 K increase in stratospheric temperature between 1 and 10 mbar as a result of dynamical heating in October. Such changes in temperature affect the stratospheric general circulation, which could have an effect on ozone concentration.

As the model used in this study is an interactive radiative/dynamical/chemical model, it is well adapted to investigate the effect of ozone interannual variations on radiative/dynamical processes, and the feedback effect on ozone concentrations, that is, an ozone-temperature-dynamics-ozone interactive mechanism. The calculations show that, for southern high latitudes in October during 1986 and 1988, when ozone concentration is high, radiative heating produces a 1 K increase in temperature below 25 km, and dynamical cooling produces a 0.25 to 0.5 K decrease in temperature above 25 km (see Figures 13a and 13c). For lower ozone concentration, during 1987 and 1989, the situation is just the opposite. The radiative cooling produces a 1 K decrease in temperature below 27 km and dynamical heating produces a 0.25 to 0.5 K increase in temperature above 27 km (see Figures 13b and 13d). These results are consistent with the calculation by Kiehl *et al.* [1988] and by Mahlman *et al.* [1994]. However, as the magnitude of column ozone interannual variation (approximately 40 DU) is much smaller than the magnitude of the Antarctic ozone hole (160 DU) used in the study of Kiehl *et al.* [1988], the change in temperature is smaller than the result presented by Kiehl *et al.* [1988].

Changes in heating and cooling lead to changes in the temperature but also to changes in the general circulation. The model results suggest that when the ozone concentration is high (in 1986 and 1988), the downward vertical motion is reduced (positive values indicate that the perturbation vertical motion is upward) (see Figure 14). The largest perturbations are found in

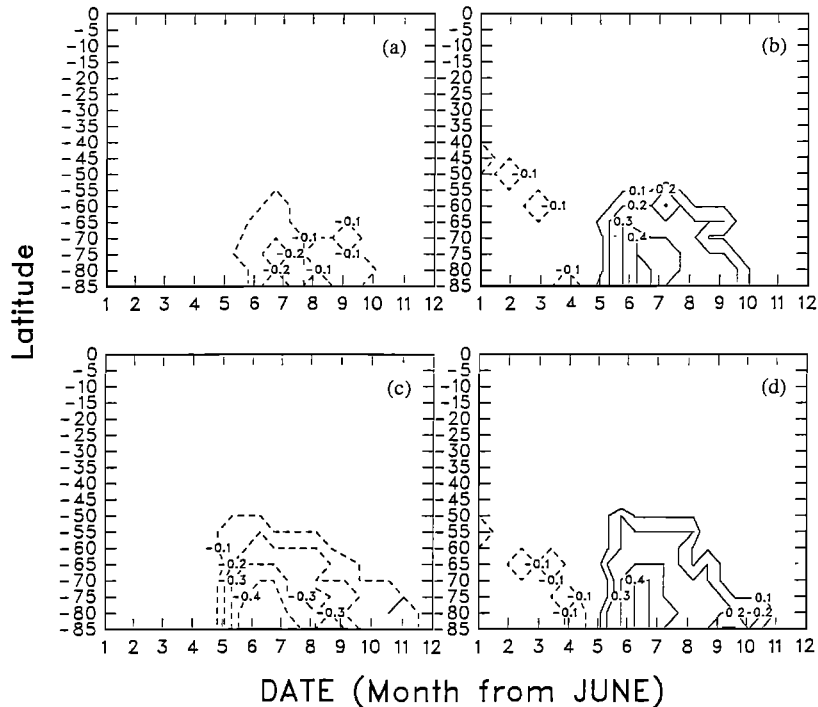


Figure 15. Calculated changes in column ozone (Dobson Units) as a result of interannual variabilities in general circulation induced by interannual variabilities in temperature formation of PSCs-ozone mechanism from 1986 to 1990: (a) 1986-1987, (b) 1987-1988, (c) 1988-1989, and (d) 1989-1990.

1988 (Figure 14c), with a maximum change of 0.5 m/d occurring at 25 km over Antarctica. Due to the reduction in descending motion, less ozone-rich air is transported into the lower stratosphere, and a decrease of approximately 0.4 DU in column ozone concentration (see Figure 15c) occurs in November. When ozone concentrations are low (in 1987 and 1989), the result is the opposite, and the column ozone increases by approximately 0.2 to 0.4 DU in November and December (see Figures 14b and 14d). This suggests that this interactive mechanism (ozone-temperature-dynamics-ozone) is not large enough to have an important effect on the interannual ozone variation over Antarctica.

Summary

Model results suggest that lower stratospheric temperature interannual variabilities affect heterogeneous reactions occurring at the surface of sulfate aerosols and PSCs, producing a significant interannual variation in the stratospheric ozone concentration. Two mechanisms were investigated in this study. First, the impact of temperature variations on heterogeneous reaction coefficients at the surface of enhanced sulfate aerosols following the eruption of Mount Pinatubo were considered. As the rate of heterogeneous conversions from inactive chlorine to active chlorine on these surfaces changes due to a decrease or an increase in temperature, the ozone destruction rate resulting from active chlorine activation was enhanced during colder winter 1992-1993, leading to a 3% ozone decrease, while it was reduced during warmer winter 1991-1992, leading to a 3% ozone increase at high latitudes in the northern hemisphere. This result improves agreement between observed and calculated ozone depletion at mid to high latitudes in the northern hemisphere during the winters 1991-1992 and 1992-1993.

Second, the effect of the interannual variation of temperature on the formation of polar stratospheric clouds (PSCs) has been considered. During winters 1987 and 1989, the lower stratospheric temperatures are 2 to 3 K colder than the averaged values between 1986 and 1990. As a result, PSC I formation persists until late September and early October (as compared to early September when averaged 1986-1990 temperature is used in the model), and the PSC I density is enhanced at the edge of the vortex (between 65°S and 70°S). During warmer winters (in 1987 and 1989), the situation is the opposite. The PSC I interannual variation as calculated by the model produces a significant interannual variation in the ozone concentration over Antarctica, which is consistent with observed column ozone variation reported by TOMS between 1986 and 1990. As ozone is a radiative chemical species, this strong interannual variation in ozone concentration affects radiative heating and cooling, as well as the general circulation, leading to a further change in ozone concentration (feedback effect). However, the magnitude of this dynamical ozone variation is very small (< 1 DU in column ozone).

Acknowledgments. The authors are grateful to Susan Solomon, Steve Massie, and Anne Smith for useful comments on the manuscript. We thank two anonymous reviewers for their good comments. One of the reviewers suggested that we should include the effect of zonal temperature wave on the calculation of heterogeneous reaction coefficients. This suggestion led to a significant improvement in our paper. We also thank Fe Wu for providing NMC temperature data. The work of XueXi Tie and Guy Brasseur is supported in part by DOE Atmospheric Chemistry Program under contract number DE-AI05-94ER619877 and by NASA under contract number S-12896-F. William Randel is partially supported under NASA grants W-16215 and W-18181. The National Center for Atmospheric Research is sponsored by the National Science Foundation.

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(Received October 18, 1996; revised May 16, 1997; accepted May 16, 1997.)