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The role of aerosol variations in anthropogenic ozone depletion at northern midlatitudes

S. Solomon,[1] R. W. Portmann,[1,2] R. R. Garcia,[3] L. W. Thomason,[4] L. R. Poole,[4] and M. P. McCormick[4]

Abstract. Aerosol surface area distributions inferred from satelliteborne 1-μm extinction measurements are used as input to a two-dimensional model to study the effects of heterogeneous chemistry upon anthropogenic ozone depletion at northern midlatitudes. It is shown that short-term (interannual) and longer-term (decadal) changes in aerosols very likely played a substantial role along with trends in anthropogenic chlorine and bromine in both triggering the ozone losses observed at northern midlatitudes in the early 1980s and increasing the averaged long-term ozone depletions of the past decade or so. The use of observed aerosol distributions enhances the calculated ozone depletion due to halogen chemistry below about 25 km over much of the past decade, including many periods not generally thought to be affected by volcanic activity. Direct observations (especially the relationships of NO_x/NO_y and ClO/Cl_y ratios to aerosol content) confirm the key aspects of the model chemistry that is responsible for this behavior and demonstrate that aerosol changes alone are not a mechanism for ozone losses in the absence of anthropogenic halogen inputs to the stratosphere. It is also suggested that aerosol-induced ozone changes could be confused with 11-year solar cycle effects in some statistical analyses, resulting in an overestimate of the trends ascribed to solar activity. While the timing of the observed ozone changes over about the past 15 years is in remarkable agreement with the model predictions that explicitly include observed aerosol changes, their magnitude is about 50% larger than calculated. Possible chemical and dynamical causes of this discrepancy are explored. On the basis of this work, it is shown that the timing and magnitude of future ozone losses at midlatitudes in the northern hemisphere are likely to be strongly dependent upon volcanic aerosol variations as well as on future chlorine and bromine loading.

1. Introduction

Statistically significant decreases in total ozone were first observed in Antarctica, but are now well documented over much of the globe [e.g., *Farman et al.*, 1985; *Stolarski et al.*, 1991; *WMO/UNEP*, 1994]. While direct observations of chlorine monoxide and related chemical compounds have confirmed the predominant role of anthropogenic chlorinated and brominated hydrocarbons in causing the decline of Antarctic ozone [*deZafra et al.*, 1987; *Anderson et al.*, 1989; *Waters et al.*, 1993; *Solomon et al.*, 1990 and references therein], the reasons for midlatitude ozone trends have been much harder to establish in a quantitative manner. Indeed, it is well known that current numerical models of the stratosphere substantially underestimate the observed total ozone depletion in northern and southern midlatitudes [*WMO/UNEP*, 1994]. Observations of the ozone profile have shown that much of the observed column decrease has occurred in the lower stratosphere at midlatitudes (below about 25 km), but present models underpredict the observed decreases at those key altitudes [*McCormick et al.*, 1992; *WMO/UNEP*, 1994 and references therein].

Numerical model simulations suggest that chemical processes associated with sulfate aerosol chemistry should decrease midlatitude ozone abundances after major volcanic eruptions [e.g., *Hofmann and Solomon*, 1989; *Brasseur and Granier*, 1992; *Pitari and Rizi*, 1993; *Prather*, 1992; *Bekki and Pyle*, 1994; *Tie et al.*, 1994]. Support for a volcanic impact on ozone has come from observations of changes not only of ozone but also of related chemical species such as NO_2 and HNO_3 following the very major eruption of Mt. Pinatubo in 1991 [e.g., *Hofmann et al.*, 1994a,b; *Gleason et al.*, 1993; *Johnston et al.*, 1992; *Fahey et al.*, 1993; *Koike et al.*, 1994; *Rinsland et al.*, 1994; *McCormick et al.*, 1995]. For a thorough review of post-Pinatubo measurements and comparison to a typical stratospheric model, see *Tie et al.* [1994]. Measurements of the ozone profile [e.g., *Hofmann et al.*, 1994b] and the column [*Hofmann et al.*, 1994a] revealed record-low ozone amounts at midlatitudes in the northern hemisphere after Pinatubo, but there has been some debate regarding whether such observed changes in ozone can be unambiguously attributed to volcanic effects [e.g., *Chandra*, 1993; *Bojkov*, 1993; *Zerefos et al.*, 1994].

In addition to the large but transient enhancements in sulfate aerosols caused by major volcanic eruptions, there is evidence for an increase in northern hemisphere stratospheric aerosol abundances over the past several decades [*Hofmann and Rosen*, 1980; *Hofmann*, 1990]. It is not clear whether this change is due to the lingering effects of multiple volcanic events or to other sources such as carbonyl sulfide (OCS) or sulfur emissions from high-flying subsonic aircraft [see, *Hofmann*, 1991]. Nevertheless, direct observations by independent techniques including lidar [*Osborn et al.*, 1995; *Jaeger*, 1994], balloonborne [*Hofmann*, 1990], and satellite (L.W. Thomason et al., A global climatology

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[1] Aeronomy Laboratory, NOAA, Boulder, Colorado.

[2] Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder.

[3] National Center for Atmospheric Research, Boulder, Colorado.

[4] Aerosol Research Branch, NASA Langley Research Center, Hampton, Virginia.

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of stratospheric aerosol surface area density as deduced from SAGE II: 1984-1994, submitted to *Journal Geophysical Reserach*, 1995; hereinafter referred to as L.W. Thomason et al., 1995); [*Hitchman et al.*, 1994] approaches have established that northern hemisphere stratospheric sulfate aerosol abundances have increased over decadal timescales. It has been noted that consideration of the heterogeneous chemistry associated with sulfate aerosols can increase significantly the calculated trends in stratospheric ozone due to increasing abundances of chlorine and bromine compared to gas phase model calculations [*Brasseur et al.*, 1990; *Rodriguez et al.*, 1991]. However, the effect of long-term temporal changes in sulfate aerosol abundances in ozone depletion has not been evaluated prior to this study.

Here we show that both short-term (interannual) and long-term (decadal) changes in the observed abundances of stratospheric sulfate aerosols have played a major role in determining the ozone depletions observed at northern midlatitudes since about 1980. The focus of this paper is on the northern hemisphere, not only because aerosol, ozone column, and ozone vertical profile trends are relatively well documented there, but also because of the currently limited understanding of the links between polar and midlatitude ozone depletion. The much larger ozone depletion observed in the Antarctic as compared to the Arctic raises important questions regarding the impacts of dilution [see e.g., *Sze et al.*, 1989; *Grose et al.*, 1989], polar vortex "processing" [*Tuck*, 1989], and denitrification [*Fahey et al.*, 1990; *Tuck et al.*, 1994] upon midlatitude ozone trends in the southern hemisphere. Recent detailed studies show that both vortex "processing" and denitrification are limited in the Arctic [e.g., *Chipperfield et al.*, 1995; *Fahey et al.*, 1990; *Roche et al.*, 1994; *Santee et al.*, 1995].

We compare key aspects of the model chemistry to direct observations not only of ozone but also of other chemical species (especially NO_x and ClO) to show that changes in liquid aerosol can strongly influence ozone loss rates at midlatitudes. The approach used is to consider the impact of heterogeneous chemical reactions associated with sulfate aerosols upon the composition of the atmosphere, using a chemical/dynamical model driven in time dependent fashion by variations in stratospheric sulfate aerosols derived from satellite extinction measurements. We will show that the timing of calculated ozone decreases predicted by the model at northern midlatitudes since 1979 is in remarkably good agreement with satellite ozone measurements. There are important discrepancies in the amplitudes of the modeled and observed ozone responses that will be explicitly discussed.

In the next section, we summarize the methodology of this study, including the heterogeneous chemical processes represented in the model. In section 3 we present observations and model calculations of ozone columns, ozone profiles, and related chemical species that are critical to the ozone budget. Section 4 presents a discussion of results and a number of sensitivity tests that probe the dependence of the results on key variables, and section 5 summarizes the primary conclusions of this study and their implications for the future.

2. Methodology

The primary tools for this study are the observations of aerosol extinction obtained since 1979 by the Stratospheric Aerosol Measurement (SAMS) 2, Stratospheric Aerosol and Gas Experiment (SAGE) I, and SAGE II satellite instruments, and the numerical model of the stratosphere of R.R. Garcia and S. Solomon. A preliminary look at the climatology of aerosol extinction measured by these satellite experiments was presented by *Hitchman et al.* [1994], who noted the important role of both major and minor volcanic eruptions in determining the aerosol column mass extinction. The satellite observations of 1-μm aerosol extinction used here are described in detail by L.W. Thomason et al. (1995). In brief, the aerosol surface areas used here are computed from seasonal median 1-km vertical resolution satellite aerosol extinction data at a wavelength of 1-μm averaged over 10° latitude bins. This single-wavelength parameterization is based on the multispectral principal components method described by *Thomason and Poole* [1993] and yields surface areas that match multiwavelength estimates to within 20-30%. SAM 2 provides a complete record of polar aerosol extinction from late 1978 to the present, while SAGE I and SAGE II provide midlatitude data from the winter of 1978-1979 to the end of 1981 and late 1984 to the present, respectively.

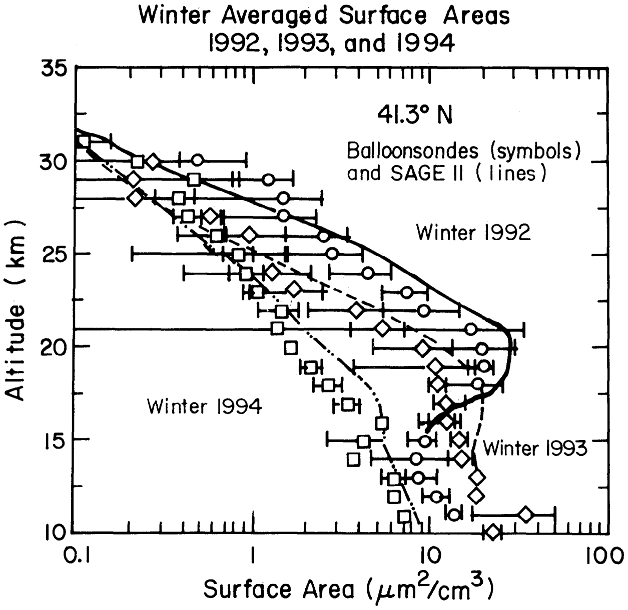

Figure 1. Observed aerosol surface areas near 40°N from satellite and balloon observations in winter 1992, 1993, and 1994. The SAGE II observations represent zonal and seasonal averages over the latitude band from 40° to 50°N, while the balloonsondes are in situ measurements from Laramie, Wyoming (41.3°N). The error bars indicate the 1-σ standard deviation of the available balloon observations in each year.

Figure 1 compares the zonally averaged aerosol surface areas derived in the winters of 1992, 1993, and 1994, with those measured by in situ dustsondes at Laramie, Wyoming. The variance of available dustsondes during each winter is shown as the error bar. Four soundings were obtained in the winter of 1992, while in 1993 and 1994, three were obtained. While there are differences in detail and the zonally averaged satellite data suggest somewhat larger surface areas (by up to 30-50%) than the local Laramie measurements at some altitudes, the comparison shows that the satellite measurements reproduce the major aspects of observed averaged seasonal changes in surface area at this location with considerable fidelity. For example, the change in altitude of the aerosol layer and relative magnitudes from one year to the next are well reproduced, providing confidence in the use of the satellite data as the basis for consideration of time dependent chemical changes associated with such sizable aerosol variations.

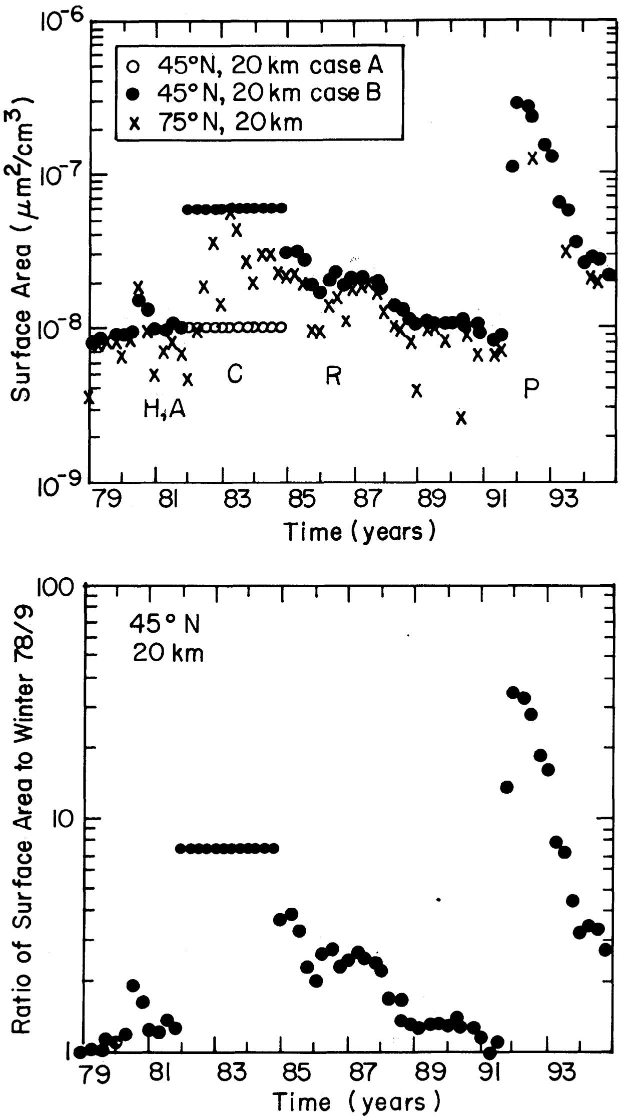

Figure 2. (top) Adopted surface areas at 20 km at 45°N and 75°N in the present study. At 75°N, satellite observations are available throughout the period from 1979 to 1995, while at 45°N, missing data from 1982 to 1984 is considered via cases A and B as shown (see text). (bottom) The ratio of the observed surface areas as a function of time to the observations in the winter of 1978-1979 is shown.

Figure 2 shows a time series of the sulfate aerosol surface areas at 20.5 km adopted in this study for 75°N and 45°N. Major volcanic eruptions are indicated by letters in the Figure. The letter H denotes St. Helens, A is Alaid, C is Chichon, R is Ruiz and P is Pinatubo. The influences of the eruptions of El Chichon (April 1982) and Pinatubo (June 1991) on aerosol surface area are profound and extend over several years. The eruption of the "mystery volcano" in late 1981 (later identified as Nyamuragira) probably also affected northern midlatitude aerosol content in the early 1980s (D. Hofmann, personal communication, 1995). The 75°N data (from SAM 2) illustrates the arrival and magnitude of El Chichon aerosol in the Arctic, but there are no midlatitude observations during this key period between SAGE I and SAGE II observations. Because of the unfortunate lack of SAGE data for the period from 1982 to 1984, two different cases have been adopted here to approximate aerosol variations at northern midlatitudes; case A (same as late 1981 throughout the period of missing data) and case B (twice that of early 1985 throughout the period of missing data) in the top figure.

Rusch et al. [1994] show that Solar Mesosphere Explorer (SME) measurements indicate a spread of El Chichon aerosol to northern midlatitudes within a year after the eruption, suggesting timing for enhanced midlatitude aerosol that lies between cases A and B. *Hofmann et al.* [1994a] observed similar timing for the arrival of El Chichon aerosol at Laramie, Wyoming, from dustsonde measurements. They report a peak surface area of 10-20 $\mu m^2/cm^3$, suggesting that the aerosol surface areas from 1982 to 1984 adopted in cases A and B are both conservative. Because global data is required for the model analysis, we do not attempt adjustments based, for example, on lidar or other observations. We will also present results for model runs using the winter 1978-1979 aerosol distributions at all times, hereafter referred to as "constant aerosols". The bottom panel shows that aerosol abundances are higher at nearly all times from 1980-1994 than the winter 1978-1979 values at 45°N, consistent with the ground-based data cited earlier. Thus the winter 1978-1979 data represents the best available information from satellite as to the likely minimum stratospheric aerosol content (which is important for the interpretation of the constant aerosol runs shown below).

The chemical/dynamical model used here has been described by *Garcia et al.* [1992] and *Garcia and Solomon* [1994]. The first paper delineates the dynamical formulation and shows that the model reproduces reasonably well the observed behavior in stratospheric tracers such as N_2O and CH_4, while the latter paper presents a detailed chemical description. The model is driven by a calculated planetary wave field whose dissipation provides both the momentum forcing that drives the stratospheric circulation and a physical basis for describing horizontal mixing [*Garcia*, 1991]. Thus the model includes a physically based representation of the "surf zone" where planetary wave breakdown mixes stratospheric species and a consistent spatial relationship between the region of mixing and the induced circulation. Reasonably realistic profiles of N_2O and other tracers are calculated both within and outside the vortex in the lower stratosphere [*Garcia et al.*, 1992]. Changes in diabatic heating (e.g., those related to the ozone changes discussed below) can also influence the circulation and temperatures calculated in the model and will be briefly discussed here in the context of ozone trends. As expected from the good comparison between observed and modeled fields of long-lived tracers such as N_2O, the modeled total ozone and ozone profiles also agree well with measurements obtained prior to the period of extensive ozone depletion in the past decade [*Garcia and Solomon*, 1994]. In addition, the calculated distributions of the key radicals species, ClO and BrO, were shown to be in general agreement with published observations (to better than about 30% in the lower stratosphere).

For the calculations presented in this paper, the solar illumination and temperature fluctuations associated with the calculated planetary wave field are included in time dependent fashion in photochemical parameters (for example, air at high latitudes is periodically exposed to sunlight due to wave displacements). Temperature changes impact the rates of heterogeneous and homogeneous chemistry, but are not fed back into the aerosol surface areas, since these are prescribed from the SAM/SAGE climatology and therefore reflect the averaged impact of temperature. *Solomon et al.* [1994] noted that the modeled good agreement with recent observations of ClO and BrO mixing ratios in the lower stratosphere at midlatitudes was difficult to reconcile with the observed large trends in ozone there using current photochemical schemes, and suggested a possible role for iodine chemistry. We will revisit the issue of photochemical schemes for ozone destruction in this paper, including the impact of the observed and highly variable aerosol distributions.

A broad range of heterogeneous chemical processes occurring on and in liquid sulfuric acid and water solutions have been studied in the laboratory and provide the basis for stratospheric model calculations. The heterogeneous chemistry included in the model is based upon the recommendations of *JPL* [1994], *Hanson et al.* [1994], *Hanson and Ravishankara* [1994], and *Hanson and Ravishankara* [1995]. A major mechanism for linking sulfate aerosol changes to ozone depletion at midlatitudes occurs via the hydrolysis of N_2O_5:

$$N_2O_5 + H_2O \rightarrow 2HNO_3 \qquad (1)$$

This process decreases the abundance of reactive nitrogen in the lower stratosphere, but increases those of reactive chlorine (ClO) and, to a lesser extent, hydrogen (HO_x) radicals. The net effect on ozone depends upon the balance between these effects, but in the lower stratosphere (below about 20 km or so), this process accelerates the net ozone loss for current levels of total chlorine as will be shown in more detail below. We adopt an efficiency (γ value) of 0.1 for this reaction.

Hanson and Ravishankara [1995] have recently shown that the hydrolysis of $BrONO_2$ on sulfate is fast under laboratory conditions:

$$BrONO_2 + H_2O \rightarrow HNO_3 + HOBr \qquad (2)$$

We adopt an efficiency for this reaction of 0.3 and also examine the sensitivity of midlatitude chemistry to higher values. This reaction is a significant source of HO_x in the lower stratosphere and thereby represents a mechanism for linking bromine trends to ozone depletion and aerosols.

We also include a set of additional reactions whose efficiency depends strongly on the water content of the aerosol and hence upon temperature [see, *Hanson et al.*, 1994; *Hanson and Ravishankara*, 1995]:

$$HCl + ClONO_2 \rightarrow Cl_2 + HNO_3 \qquad (3)$$

$$ClONO_2 + H_2O \rightarrow HOCl + HNO_3 \qquad (4)$$

$$HCl + HOCl \rightarrow Cl_2 + H_2O \qquad (5)$$

$$HCl + HOBr \rightarrow BrCl + H_2O \qquad (6)$$

The impact of these reactions upon midlatitude ozone depletion under different conditions of aerosol loading will be discussed below. Unless otherwise indicated, all of these heterogeneous processes are included in the results shown.

The chemical/dynamical model was initialized with 1970 levels of chlorine and bromine and run in time dependent fashion. Time-varying boundary conditions for chlorine and bromine source gases after 1970 are taken from *WMO/UNEP* [1994]. Changes in solar fluxes are not included in this calculation, nor are trends in stratospheric water vapor considered [see, *Oltmans and Hofmann*, 1995]. In this paper we explicitly consider only the period from 1970 to 1995, but will comment upon implications of this study for future trends in ozone. Our focus will be on the ozone trends during the period from 1978 to 1994. Since there is no SAM/SAGE data prior to 1978, we impose the winter 1978-1979 values prior to this point but recognize that volcanic eruptions such as those of Fuego in the early 1970s influenced the true aerosol content during that period [*Hofmann*, 1990; *Osborn et al.*, 1995]. We consider the impact of aerosols on heterogeneous chemistry, but do not include their direct effects on stratospheric radiation [e.g., *Michelangeli et al.*, 1989; *Pitari and Rizi*, 1993; *Tie et al.*, 1994] or dynamics [*Brasseur* and *Granier*, 1992]. These radiative and dynamical effects are expected to be important primarily in the tropics prior to dispersion of high aerosol loadings [*Tie et al.*, 1994] and are not the focus of the present study.

A companion paper by R.W. Portmann et al. (The role of aerosol variations in anthropogenic ozone depletion in the polar regions, submitted to the *Journal of Geophysical Research*, 1995; hereinafter referred to as R.W. Portmann (1995)) will present the details of polar ozone chemistry used in the model and describe the processes responsible for producing a credible Antarctic ozone "hole" in the current model. Here our focus is on northern midlatitudes, and polar processes will be considered only insofar as their influence on midlatitude ozone depletion is concerned. Temperature dependent reactions on sulfate aerosols are included in polar regions, as are reactions on water ice. We adopt the water ice chemistry described by the *JPL* [1994] whenever modeled temperatures fall below the local frost point. For temperatures below the frost point, we also assume dehydration and denitrification can occur. Irreversible removal of water and nitric acid vapor is assumed to occur on a timescale of a few days whenever temperatures fall below this threshold. We will show explicitly via sensitivity tests that polar chemistry does not signficantly affect the calculated northern midlatitude ozone changes of interest here.

In order to evaluate the effects of changes in halogen chemistry alone, the surface abundances of methane, nitrous oxide, and carbon dioxide that serve as boundary conditions to the two-dimensional model were held fixed in nearly all runs at 1.72 ppmv, 310 ppbv, and 354 ppmv, respectively. A test run using realistic time-varying boundary conditions for these radiatively and chemically important trace gases was also carried out, and the results will be briefly discussed below.

3. Results and Discussion

3.1. Ozone Column

Figure 3 shows the calculated changes in total column ozone at 45°N relative to the 1970 values for model runs using the winter 1978-1979 "constant" aerosol distributions throughout the period and for the observed aerosol variations from 1979 to 1995 as shown in Figure 2 above. The top panel shows the full annual cycle while the bottom panel has been smoothed over the annual cycle via an annual running mean. Cases A and B differ only in the adopted aerosol distribution from 1982 to 1984 as discussed above and shown in the figure.

Figure 3 shows that the model predicts significant effects upon northern midlatitude ozone abundances due to changing aerosols essentially throughout the period from 1979 to 1995. It will be shown below that this ozone depletion occurs mainly via chemical enhancements in ozone loss rates relating to chlorine/bromine chemistry. Thus like the polar stratospheric clouds that enhance Antarctic ozone depletion compared to gas phase chemistry, liquid aerosol enhancements increase the efficiency of halogen chemistry for ozone loss at midlatitudes but are not a mechanism in themselves for substantial ozone loss independent of human inputs of chlorine and bromine to the

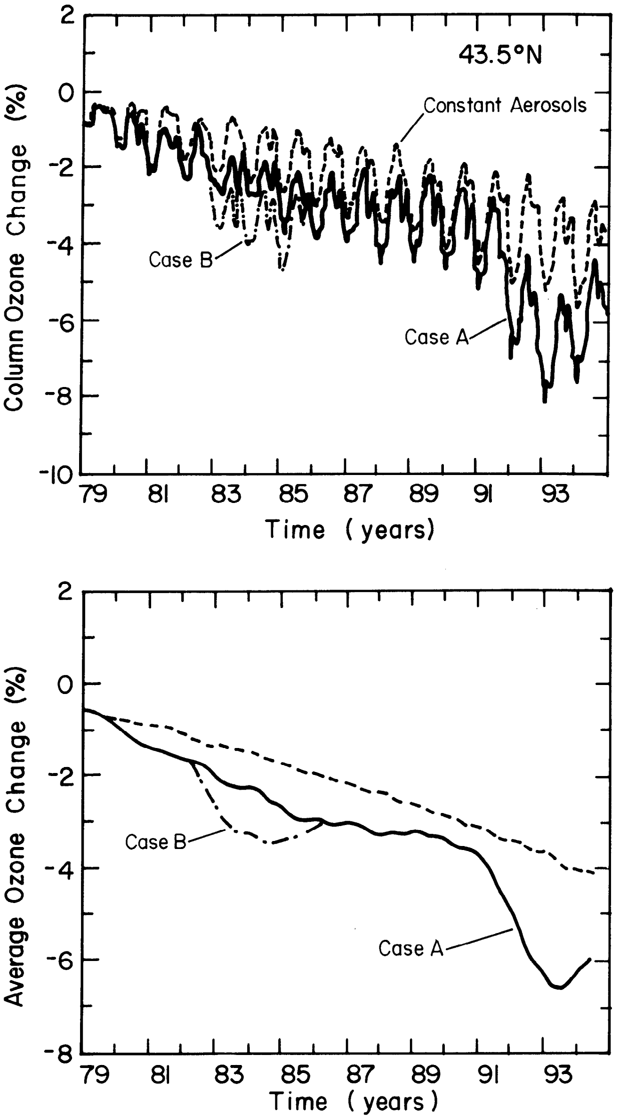

Figure 3. (top) Calculated ozone depletions at 43.5°N relative to 1970 conditions with the constant aerosol assumption for constant aerosols (at winter 1978-1979 levels) throughout the period and for cases A and B. (bottom) Same as above, but smoothed with an annual running mean filter to eliminate the seasonal cycle.

stratosphere. Indeed, *Tie and Brasseur* [1995] have shown that a major volcanic eruption in a preindustrial atmosphere (i.e., one with far less stratospheric chlorine than present conditions) would be expected to produce a net increase in total column ozone rather than a decrease based on our best current chemical understanding (see also, Figure 8 below).

The effects of El Chichon and Pinatubo upon predicted ozone levels are evident in the first year or two following those eruptions, but there are also substantial impacts in other years not generally thought to be affected by volcanic aerosols. Even in the late 1980s, the ozone content calculated using the measured aerosols differs from that expected with constant aerosols. Just after 1980, a marked decrease in ozone is predicted at 43.5°N related to enhanced aerosols then (which are largely attributed to the eruptions of St. Helens and Alaid, see *Hitchman et al.* [1994]).

One way to express the magnitude of short-term ozone fluctuations and long-term trends is to consider the departures in any given year as the anomaly relative to the long-term mean (as by *Randel et al.* [1995]). As will now be shown, these dramatically illustrate the key role likely played by aerosols in determining ozone abundances at midlatitudes.

Plate 1 compares the total ozone anomaly predicted by the model (at its grid points at 43.5°N and 53.7°N) to observations at 45°N and 55°N based upon three different analyses of observations. One set of measurements (blue squares) is the zonally averaged TOMS Version 6 data analysis of Randel (W. Randel, personal communication; as by *Randel et al.* [1995]). The QBO has been removed from the observed time series via a regression approach, and the data have been smoothed with a Gaussian filter over the annual cycle, but no solar cycle or El Nino components have been included. For further description of these data and the analysis technique see *Randel et al.* [1995]. The red diamonds show the zonally averaged TOMS Version 6 data for the same latitudes with a simple 25-month running boxcar average to smooth over both the annual cycle and the QBO. The green triangles show the zonally averaged SBUV/SBUV2 data set (the same as that used in the WMO/UNEP 1994 trend analysis, provided to us by R. Nagatani, J. Miller, and W. Planet) with the same 25-month boxcar smoothing. The good agreement between the three data sets suggests that the major anomalies observed in ozone depletion are not sensitive to the approach taken for removal of the QBO or to the details of the smoothing procedure, and are remarkably similar in the separate (but related) TOMS and SBUV date records.

The observations of these ozone trends from TOMS are within 1% of those deduced from ground-based data at northern midlatitudes during the same period, and the ground-based data display the same major features discussed above in the timing of ozone changes over the period since 1979 [*Bojkov and Fioletov*, 1995]. Hence instrumental artifacts such as calibration or errors relating to aerosol backscatter in the satellite ozone data do not substantially affect the conclusions of Figure 3 and Plate 1.

Plate 1 shows the model results smoothed with a 25-month running boxcar average for aerosol case B at both latitudes as well as those obtained when the ozone anomalies calculated for aerosol cases A and B are increased by 50%. The important 50% discrepancy in amplitude of the modeled and observed responses will be discussed further below.

While the amplitudes of the observed changes at both 45°N and 55°N are roughly 50% greater than those of the model, it is clear that nearly all features of the timing of the observed ozone changes from 1979 to 1994 are remarkably well simulated, including the marked drop in ozone around 1983-1986 (due to the enhanced aerosol associated with El Chichon and later Ruiz), the flattening of the ozone trend from 1986-1991 (due to a partial cancellation between increasing total chlorine/bromine and decreasing aerosol contents), and the dramatic depletion in 1991-1993 and signs of recovery after 1993 (due to the effect of Pinatubo's aerosol enhancement and subsequent removal). Indeed, Figure 3 and Plate 1 suggest that the combined effects of St. Helens, Alaid, Chichon and Ruiz together with trends in anthropogenic halogens represented an important "trigger" for the emergence of identifiable ozone depletion at midlatitudes during 1980-1987 [see, *Ozone Trends Report*, 1988]; the companion paper by R.W. Portmann et al. (1995) demonstrates that a similar trigger for Antarctic ozone loss occurred in the early 1980s associated with the heterogeneous chemistry of El Chichon aerosol there. Figure 3 and Plate 1 also explicitly show that the long-term trend in ozone will depend upon the time interval and approach used in the statistical analysis, due to the transient depletions associated

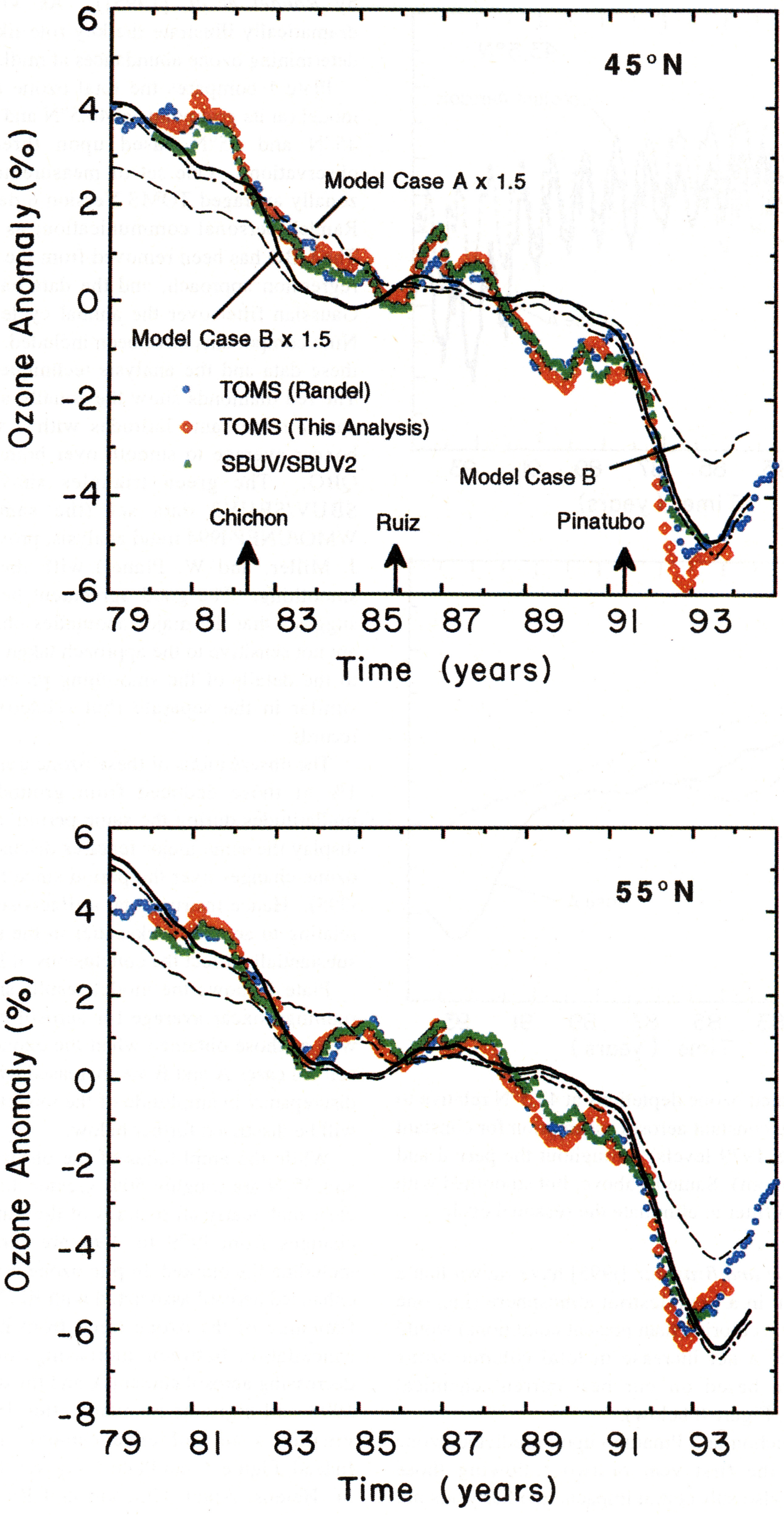

Plate 1. Observed and calculated total ozone anomalies from 1979 to 1994 at 45°N and 55°N. Blue squares show the zonally averaged TOMS Version 6 data analysis of Randel (W. Randel, personal communication; as in work by *Randel et al.* [1995], in which the QBO is removed by regression and the data are smoothed with a Gaussian smoothing function. Red diamonds show the zonally averaged TOMS Version 6 data for the same latitudes with a simple 25-month running boxcar average to smooth over both the annual cycle and the QBO. Green triangles show the zonally averaged SBUV/SBUV2 data set with the same 25-month boxcar smoothing. Model results are also smoothed with a 25-month running boxcar, and results for cases A and B are shown scaled by a factor of 1.5 along with values for case B unscaled (see text and Figure 2).

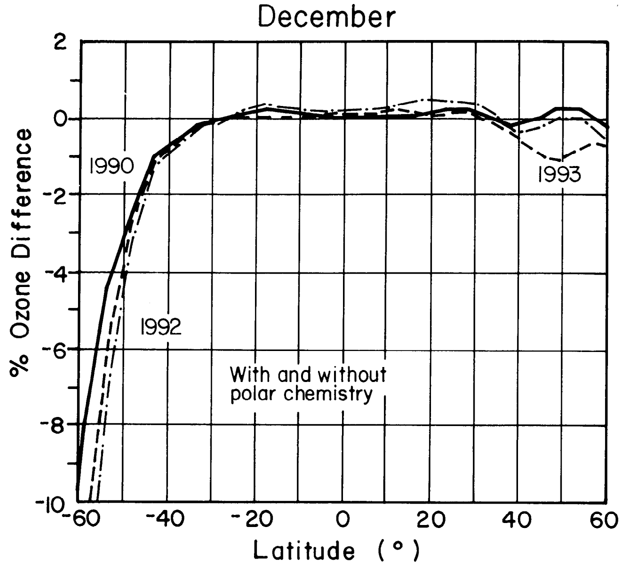

Figure 4. Calculated difference in ozone depletion as a function of latitude between model runs in which polar chemical ozone depletion is or is not considered, for December in various years.

with large aerosol changes when they take place in a stratosphere containing anthropogenically enhanced chlorine and bromine.

The enhanced ozone loss near 1989 stands out in Plate 1 as the sole major unexplained feature in the northern hemisphere ozone record and is more pronounced at 45°N than at 55°N, showing that aerosol changes alone certainly do not explain all of the ozone fluctuations observed. Factors such as stratospheric temperatures are also likely to influence year-to-year ozone variability. The Arctic stratosphere was unusually cold in the winter of 1989, which, for example, displayed the lowest January high-latitude temperatures in 26 years [*Nagatani et al.*, 1990] and substantial Arctic denitrification was directly observed [*Fahey et al.*, 1990; *Tuck et al.*, 1994]. Such processes may affect midlatitude ozone variability, but are not yet fully quantified. Nevertheless, Plate 1 suggests that heterogeneous processes associated with aerosols account for a great deal of the observed variance of total ozone depletion in northern midlatitudes since 1979. It is important to emphasize that this conclusion is based not simply upon correlations but rather upon known chemical mechanisms (discussed further below, see in particular Figure 6).

In order to separate local chemical effects from transport influences, a test run was performed for the period from 1989-1994 in which the chlorine-activating reactions (2) - (6) were turned off at latitudes poleward of 60° in both hemispheres. This mimimizes the effect of transport of ozone-depleted air out of the polar regions by eliminating the primary reactions responsible for ozone depletion there. Figure 4 illustrates the resulting changes in ozone columns at midlatitudes in December. Similar values are obtained in other months. The figure shows that the modeled ozone abundances at southern midlatitudes are strongly affected by polar processes, while northern regions are affected by less than 1%, even after Pinatubo. This does not necessarily imply that the real stratosphere behaves in the same manner, although the consistency of the 50% discrepancy between the calculated and observed ozone anomalies at both 45°N and 55°N supports the view that the shortcoming is related to the model chemistry. An unusual combination of polar processing at different locations and times giving the same discrepancy for different reasons cannot be ruled out. We discuss this issue further below.

3.2. Ozone Profile

It is well known that current photochemical models simulate reasonably well the ozone losses observed above 30 km or so at midlatitudes but fail to capture the ozone depletions observed at lower altitudes and hence underestimate the total column loss [e.g., *McCormick et al.*, 1992; *WMO/UNEP*, 1991; 1994]. The present model is no exception to this general picture, but we show here that the explicit consideration of aerosol content is highly likely to be a substantial factor in determining the shape of the profile of ozone depletion below about 30 km.

Figure 5 presents an example of the calculated vertical distributions of the ozone depletion at 45°N for October 1986 relative to October 1979 for the constant and observed (case A)

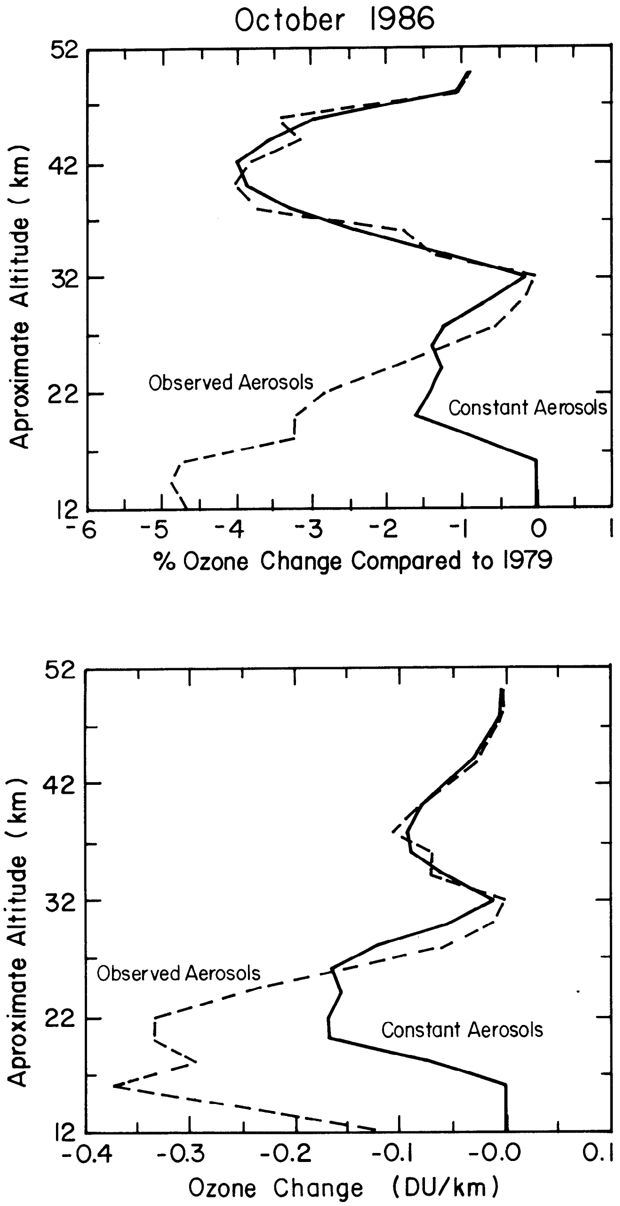

Figure 5. Calculated ozone depletion in October 1986 compared to 1979, for model runs assuming constant aerosols at the winter 1978-1979 levels, and when aerosol observations (case A) are included. (top) Figure shows percentage depletion, illustrating the very large effect of aerosols on the shape of the ozone loss profile below 25 km. (bottom) Figure shows the contribution of each altitude to the column depletion in Dobson Units (DU) per km.

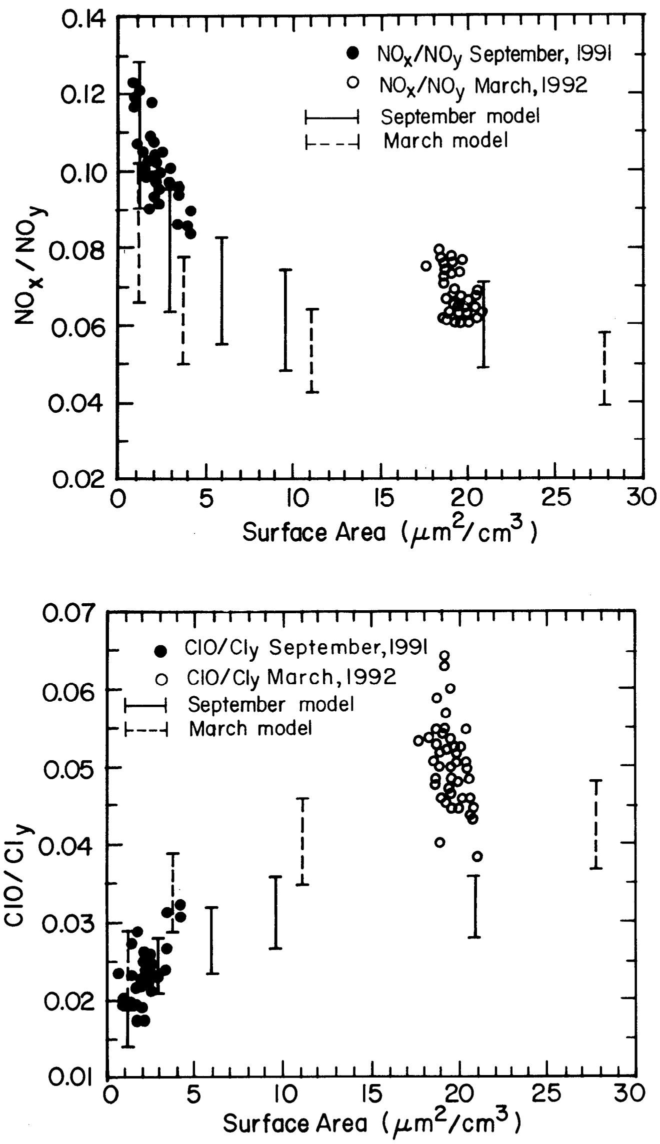

Figure 6. Calculated and observed dependence of NO_x/NO_y and ClO/Cl_y upon surface area. The data are taken from the paper by *Fahey et al.* [1993], who reported measurements made at low aerosol content in September, 1991 (left-hand side of figure) and at high aerosol content in March, 1992 (right-hand side of figure). The September data (solid circles) should be compared to the September model (solid bars) and the March data (open circles) to the March model (dashed bars).

aerosol cases. While 1986 is not generally thought of as a period perturbed by volcanic aerosols, the role of rather small aerosol enhancements (see Figure 2, bottom) in influencing ozone in the lower stratosphere during this period is quite striking, even for the conservative case A. Consideration of the observed aerosol content increases the calculated ozone depletions by more than a factor of 2 near 20 km at this time compared to predictions assuming constant aerosols (at the winter 1978-1979 levels). It is very important to note that the transience of aerosol enhancements has important effects on the details of the profile of the calculated percentage changes in ozone, especially below 20 km. The calculated ozone changes near 14 km shown in Figure 5 are mainly the result of ozone depletions that occurred higher and earlier, and have descended with time due to the mean meridional circulation of the stratosphere. In particular, the larger aerosol surface areas present near 20 km in 1985 affect the calculated ozone losses below 20 km in 1986. Thus, according to current photochemical schemes, the changing aerosol content associated with volcanic eruptions produces lingering effects that descend with time and impact the ozone depletion profile substantially, particularly below 20 km (where photochemical loss rates are believed to be relatively slow). Possible changes in photochemistry (e.g., iodine or other chemistry discussed further below) could, however, alter the timescale for chemical depletion at low altitudes and hence impact this effect.

Figure 5 shows both the calculated percentage and absolute changes in ozone, illustrating that the changes below 30 km dominate the calculated changes in the column abundance. An important new result of Figure 5 is thus the finding that the ozone changes below 30 km likely depend upon aerosol content and its past history, even for the relatively modest aerosol enhancements of a year such as 1986. For the far more perturbed winter following Pinatubo, the model suggests a 10% local ozone loss near 20 km at 40°N, considerably smaller than the 25% depletions reported at that location by *Hofmann et al.* [1994b], but much larger than predicted using constant aerosol concentrations.

Aerosols also affect the decadal trends in the lower stratosphere, as would be expected from the column trends discussed above. The present model predicts only about a 2%/decade change in ozone at 20 km in northern midlatitudes for the period since 1980 using constant aerosols. Larger changes of 3-5%/decade are obtained for the model run using observed aerosols, depending upon the time period used for trend analysis (for example, depending upon whether or not Pinatubo is included in the trend). Further analysis of SAGE and ozonesonde measurements is needed to better quantify the exact profile of observed ozone trends in the lower stratosphere, but current observations suggest larger values of the order of 10-15%/decade near 20 km, as shown by *McCormick et al.* [1992] and *WMO/UNEP* [1991, 1994].

While the total column comparisons indicate a model/measurement discrepancy of about 50%, the profile comparisons suggest larger local differences of about a factor of 2 or more near 20 km (both for highly perturbed conditions such as those following Pinatubo and over decadal timescales). These model/measurement comparisons underscore the role of time dependent aerosol variations as well as the importance of the ozone trends in the lowest part of the stratosphere, where the discrepancies are difficult to reconcile with present chemistry.

3.3. Ozone Loss Photochemistry and Budgets

We next examine explicitly the chemistry responsible for the model ozone depletions associated with aerosol changes and compare the results to observations. Figures 6 and 7 provide strong support for the accuracy of the modeled representation of the key aspects of the chemistry leading to ozone losses in response to changing aerosol abundances at northern midlatitudes. In particular, Figure 6 compares the modeled NO_x/NO_y and ClO/Cl_y values at 56 mbar at 43.5°N as a function of aerosol surface area to the observations of the same parameters by *Fahey et al.* [1993] in September 1991 (data at low aerosol on the left side of the figure) and March 1992 (data at high aerosol on the right side of the figure). As may be expected from the model's good simulations of long-lived tracers such as N_2O [*Garcia et al.*, 1992], the calculated absolute abundances of NO_y

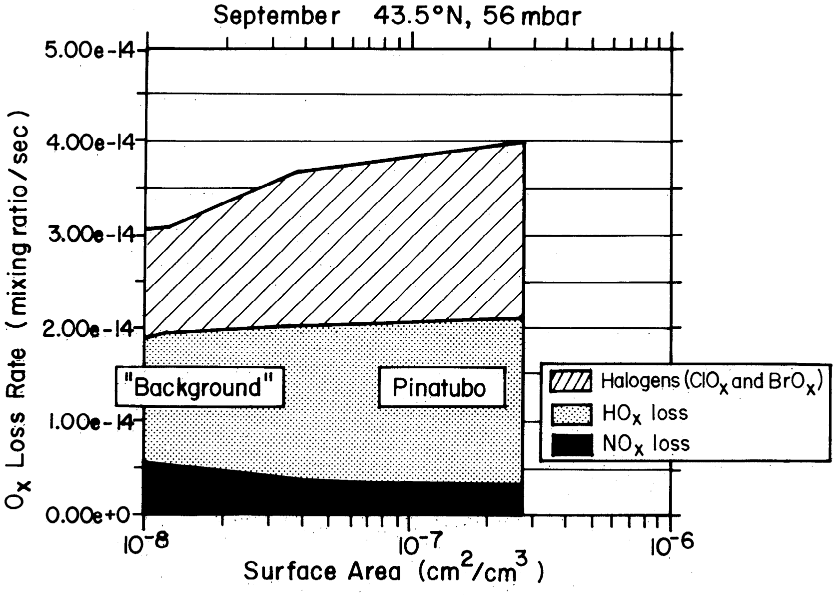

Figure 7. Calculated odd oxygen loss rates versus surface area at 43.5°N near 20 km (56 mbar) for 1990 levels of total chlorine and bromine, for NO_x, HO_x, and halogen chemistry.

and Cl_y agree to better than 20% with deductions based upon observations [*Fahey et al.*, 1990; *Woodbridge et al.*, 1995]. The trends in chlorine source gases are well defined from observations [*WMO/UNEP*, 1994] that serve as input to this calculation. Daily model values for NO_x/NO_y and ClO/Cl_y for the 2-week period centered on the measurement seasons are shown. The model ranges within each value of aerosol surface arise from seasonal changes in temperature and illumination over a 2-week period, and from the coupling of photochemistry to the modeled planetary wave amplitudes. These modeled ranges in NO_x/NO_y and ClO/Cl_y partitioning for a given aerosol content appear comparable to the observed variability. The changes in modeled aerosol abundances from low to high values represent the seasonally averaged satellite observations in 1990, 1991, 1992, 1993, and 1994.

The observations and the modeled behaviors of ClO/Cl_y and NO_x/NO_y in Figure 6 show very similar dependence on aerosol content. The increases in modeled ClO/Cl_y with increasing surface area at this altitude are mainly driven by the decreasing NO_x/NO_y abundances, which are in turn mainly a result of the hydrolysis of N_2O_5. *Stimpfle et al.* [1994] provide additional direct evidence for this key chemical interaction by showing that the observed dependence of ClO on NO_2 follows that expected from partitioning relating to $ClONO_2$. *Wennberg et al.* [1994] display data showing a similar result. They also demonstrate that the abundance of OH near 20 km at midlatitudes is nearly independent of observed NO (hence, of surface area) and the abundance of HO_2 is only weakly dependent on NO. The present model reveals a similar dependence of the HO_2/OH ratio upon NO abundance to that presented by *Wennberg et al.* [1994], as would be expected since this partitioning depends only on simple chemistry [see also *Cohen et al.*, 1994]. The calculated absolute value of OH at 18 km at 30°-50°N for 30° solar zenith angle from May to September months is 1±0.2 pptv and is not dependent on NO content, in agreement with *Wennberg et al.* [1994]. The calculated absolute values of ClO have previously been shown to be within about 30% of midlatitude observations throughout the range of altitudes from 18 to 30 km for March, 1991 (see the comparison to the data of *Avallone et al.* [1993] presented in work by *Garcia and Solomon*, [1994]).

The chemical changes shown in Figure 6 control the response of ozone loss rates to increasing surface area. Figure 7 shows the calculated September averaged ozone loss rates due to different chemical components as a function of aerosol surface area at 20 km in midlatitudes. Trends in Cl_y and Br_y have been normalized to their 1990 levels in generating this comparison, so that the halogen chemistry reflects 1990 values. Increasing surface areas decrease the abundance of NO_x and hence the NO_x-catalyzed ozone destruction cycles, while increasing somewhat those of HO_x. The figure shows that the sum of the HO_x- and NO_x-catalyzed losses under the conditions considered (midlatitudes, 20 km, September) remains nearly constant over the range of surface area considered. Except for the key reaction of ClO with BrO, bromine catalyzed losses are also nearly independent of surface area. However, the rate of chlorine-catalyzed (and chlorine-bromine catalyzed) ozone destruction increases substantially with increasing surface area due mainly to the changing ClO/Cl_y ratio shown in Figure 6, dominating the net impact on ozone at this altitude. Small enhancements in HO_2 (by about 20% over the range in surface areas shown) also contribute through their impact on the ozone-destroying reactions of ClO and BrO with HO_2.

Figure 8 (top) displays the difference in calculated O_x loss rates as a function of altitude for October 1986 when the observed aerosols are considered compared to that obtained for the constant winter 1978-1979 aerosols (as in Figure 5). This figure expands upon the conclusions drawn from Figures 6-7 by showing the changes in the various cycles with altitude for this particular latitude and season, and also indicates the competition with transport timescales (bottom). As in Figure 7, the combined HO_x+NO_x chemical loss rate remains essentially constant with changed aerosols below about 20 km. At higher altitudes, reduced NO_x levels associated with enhanced aerosols reduce the HO_x+NO_x chemical ozone loss rates. On the other hand, the halogen-induced loss is enhanced throughout the altitude range. Near 24 km and below, the net effect of HO_x, NO_x, and

ClO_x/BrO_x chemistry is a faster total chemical loss rate with increased aerosols, while at higher altitudes reduced NO_x leads to slower chemical losses for these conditions. Indeed, *Hofmann et al.* [1994b] have observed a cross over from ozone depletion below 24 km to ozone increases above this level following Pinatubo, consistent with Figure 8 and the calculations shown in Figure 5 [see also *Hofmann and Solomon*, 1995]. Above about 30 km, the O_x lifetime rapidly decreases and the indicated changes in loss rates are too small a fraction of the total rate to significantly affect the profile for the prescribed aerosol enhancements. Figure 8 shows the reasons why the ozone response to aerosols is dependent upon altitude and chlorine content, and indicates that column ozone increases can occur in response to increased aerosol for preanthropogenic conditions (or for high-altitude injections), while column decreases can be obtained under the current, largely anthropogenic burden of stratospheric chlorine [see also *Tie and Brasseur*, 1995].

Figure 8 (bottom) shows the competition between ozone transport and chemical time scales for October 1986 conditions. The figure shows that the model's timescale for transport of ozone is comparable to or faster than the chemical loss timescale below about 24 km for current chemical schemes. Thus transport is also likely to affect the ozone depletion profile in this region, and the downward propagation of chemical losses from higher altitudes and/or the horizontal transfer from other latitudes cannot be ignored. However, trends in the northern hemisphere model dynamics caused by the ozone changes are very small. Examination of the long-lived tracer and vertical velocity fields computed by the model show that the computed dynamical changes alter the modeled northern midlatitude ozone column by less than 0.2% and the profiles by less than 0.5% below 30 km.

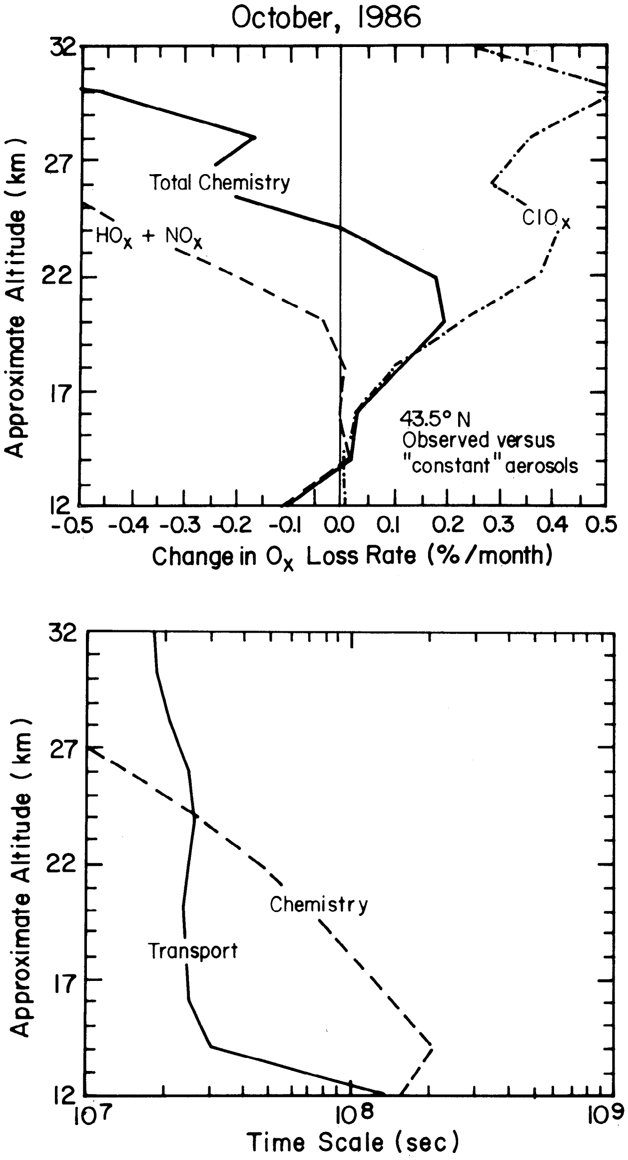

Figure 8. (top) Change in odd oxygen loss rate versus altitude at 43.5°N for October 1986 for observed (case A) versus constant winter 1978-1979 aerosols. (bottom) Timescales for photochemical destruction of odd oxygen and for transport processes under the same conditions.

4. Discussion and Sensitivity Tests

In this section we summarize the implications of the model results for the current understanding of ozone depletion, including both chemical and dynamical effects. We also indicate briefly some sensitivity tests that bear upon possible mechanisms for explaining the discrepancies between the modeled and observed ozone depletion amplitudes and indicate the possible role of aerosol-induced chemical changes in ozone for some attempts to deduce solar cycle signatures in ozone records.

Figures 6 and 7 demonstrate the relative roles of trends in chlorine versus trends in aerosol surface area for ozone losses. For example, the total chlorine content of the midlatitude lower stratosphere (Cl_y) was about 30% larger in the winter of 1984-1985 compared to that of 1978-1979, while the aerosol surface area at 20 km near 40°N changed from about 1 $\mu m^2/cm^3$ in the winter of 1978-1979 to about 4 $\mu m^2/cm^3$ in the winter of 1984-1985. Figure 6 (bottom) shows that this change in surface area increases ClO/Cl_y at this latitude and altitude by about 50%. The effect of the change in surface area upon ClO (and hence on the ozone loss rate due to halogen chemistry) is therefore greater than the effect of the Cl_y trend over this period. Figures 2, 6, and 7 taken together demonstrate that the impact of changing surface areas upon ozone depletion, through their effect on ClO/Cl_y ratios and hence on halogen-catalyzed ozone destruction near 20 km, has been comparable to or greater than the impact of slow, time dependent trends in Cl_y alone upon ozone during much of the period since 1979.

Figures 6-8 and the related discussion strongly support the midlatitude lower stratospheric chemistry used in the present model, making it unlikely that errors in modeled concentrations of ClO, NO_x, HO_x, or their response to aerosol changes could be large enough to account for the discrepancies between modeled and observed ozone trends over the past decade or two, especially when the observed discrepancies in the profile of the depletion near 20 km are considered. An approximate doubling of the calculated ozone depletion from about 15 to 25 km with unchanged depletions above is required to resolve both the discrepancies in the column and profile depletions. The model results show that the amplitude of ozone losses at midlatitudes is underestimated by this model, but that their timing is in remarkable agreement with observations, and is mainly tied to the model-calculated ClO/Cl_y response to changing aerosols. While other explanations are possible, this suggests that a solution to the discrepancy in amplitude could lie in an additional or enhanced

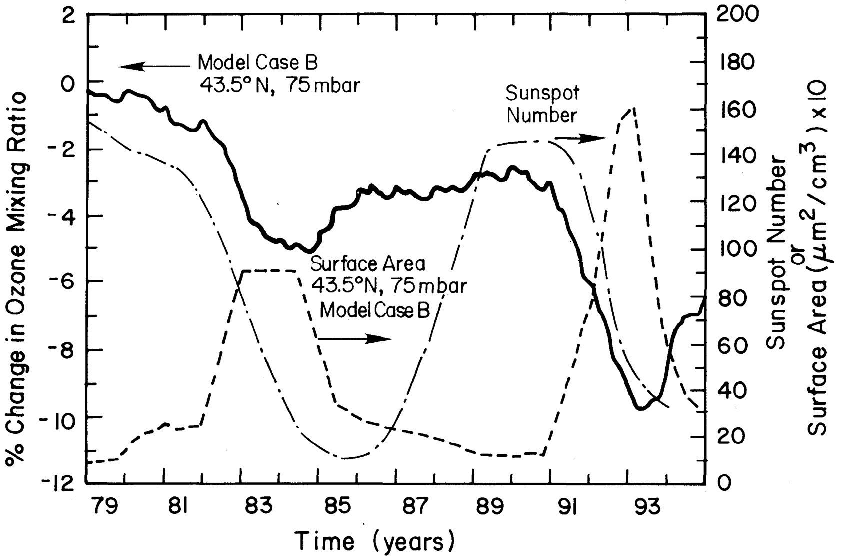

Figure 9. Calculated ozone depletion at 43.5°N, 18 km, for model case B together with the adopted SAGE/SAM aerosol surface area climatology (12-month running means) and the annually averaged sunspot number versus time.

photochemical partner for ClO. A larger amplitude ozone loss would, for example, be expected if the coupling of iodine to chlorine trends were a significant driver of ozone depletion [see, *Solomon et al.*, 1994]; such a coupling would increase the magnitude of the ozone loss associated with ClO increases but would not affect the timing of short- and long-term ozone responses relating to changing ClO concentrations. Similarly, enhanced abundances of active bromine (i.e., BrO) in the lower stratosphere (e.g., through currently unaccounted for sources such as convection of short-lived species including for example, dibromomethane, chlorobromoalkanes, iodobromoalkanes, or bromoform; see *Solomon et al.*, [1994]) could also enhance the amplitude (but not the timing) of ozone responses to changing ClO. An enhanced ozone loss due to slow changes in bromine and their coupling to, for example, iodine, may also be broadly consistent with the observations. Other chemical mechanisms (e.g., an enhanced response of HO_x to changing aerosol content through, for example, the reaction of $BrONO_2$ with H_2O or trends in water vapor) could perhaps play a role, but the former may be difficult to reconcile with the observed behavior of HO_2 and OH reported by *Wennberg et al.* [1994]. Using a sticking coefficient of 0.9 rather than 0.3 for the heterogeneous reaction of $BrONO_2$ with water, we calculate an increase in ozone depletion of about 0.5% for 45°N in 1989 and 1% in 1992, which is significant and retains the correlation with aerosol content, but is smaller than the apparent discrepancy. In a test run that included observed trends in methane, nitrous oxide, and carbon dioxide (taken from the *WMO/UNEP* [1994] assessment), calculated total ozone losses at northern midlatitudes increased by only 0.3% from 1970 to 1995, indicating that the chemical and radiative impacts of trends in these compounds are unlikely to greatly influence the midlatitude ozone column trends according to current understanding.

Figure 6 also suggests that the modeled enhancement in ClO/Cly for the highest levels of aerosol loading observed after Pinatubo (about 20 μm2/cm3) are somewhat lower than observed. Hence additional chemical mechanisms for ClO enhancements and correspondingly larger ozone depletions in highly perturbed conditions of aerosol loading are indicated. However, such large aerosol enhancements are observed only briefly in the record of the past 15 years or so, and hence are not a major factor in the longer term trends apart from the first year or so following Pinatubo and perhaps Chichon.

Temperature trends of the order of 0.5°-1°K have been observed in the lower stratosphere [*Oort and Liu*, 1993; *WMO/UNEP*, 1994]. These are too small to significantly affect photochemical rates of ozone destruction. Further, the present model simulations include the coupling between ozone and temperature and predict midlatitude temperature changes that are broadly comparable to those observed.

Dynamical processes could also influence the ozone loss amplitudes. The work of *Tie et al.* [1994] suggests that the direct dynamical affects of Pinatubo on the ozone depletion at midlatitudes are likely to be relatively small and short-lived in duration. Hence the dynamical changes relating to volcanic aerosol injections appear unlikely to explain the discrepancies. However, it is clear that rapid transport of ozone-depleted air from the vortex or latitudes just outside the vortex could enhance midlatitude ozone depletion [*Tuck*, 1989]. Since the aerosols spread relatively rapidly over most of the globe, such transport processes would remain broadly consistent with the timing of the ozone responses but, like the chemical mechanisms discussed above, would lead to increased amplitudes at midlatitudes since the magnitudes of ozone loss increase with increasing latitude. Recent three-dimensional simulations [e.g., M.P. Chipperfield et al., Simulations of Arctic winters 1991-1992 to 1994-1995 using a three-dimensional model, submitted to the *Journal of Geophysical Research*, hereinafter referred to as Chipperfield et al., 1995] do not support a large role for such processes at northern midlatitudes. It is also possible that the circulation of the stratosphere itself has changed, perhaps in response to changing planetary wave forcing [e.g., *Hood and Zaff*, 1995 and references therein]. A combination of several such effects (both chemical and dynamical) taken together is obviously also possible.

The role of the processes described here in some statistical evaluations of the magnitudes of ozone trends is also worthy of mention. Figure 9 shows the modeled local ozone change at 43.5°N, 18 km, for aerosol case B from 1979 to 1994 together with the adopted local aerosol surface area at that point and the annually averaged sunspot number. The aerosol changes indicated in the figure together with chlorine and bromine trends drive the calculated ozone changes, since solar cycle effects are not considered in the present calculation. However, the two major eruptions of El Chichon and Pinatubo both occurred a few years after solar maxima, the timescale for the removal of their aerosol inputs is of the order of several years, and the aerosol surface areas and sunspot numbers therefore are correlated to some degree. Further, both aerosol surface and sunspot number (or other solar indicators such as F10.7 cm flux) display frequencies near 10 years. Frequency analysis of the modeled ozone values shown in Figure 9 would display considerable power at a value near 10 years, but because of volcanic and not solar modulation. The eruption of Fuego in the 1970s also had a substantial effect on aerosols at midlatitudes, and happened to occur shortly after the previous solar maximum [e.g., *Hofmann et al.*, 1990; *Osborn et al.*, 1995], so that very long records may be needed to overcome this problem. Statistical methods to deduce solar effects on ozone trends often employ time lags that could further enhance the correlation shown in the figure. Depending upon the analysis technique used and the length of the records, Figure 9 suggests that some aerosol-induced ozone changes could be erroneously ascribed to solar effects in statistical analyses of ozone trends, causing an overestimate of the ozone trends ascribed to the 11-year solar cycle.

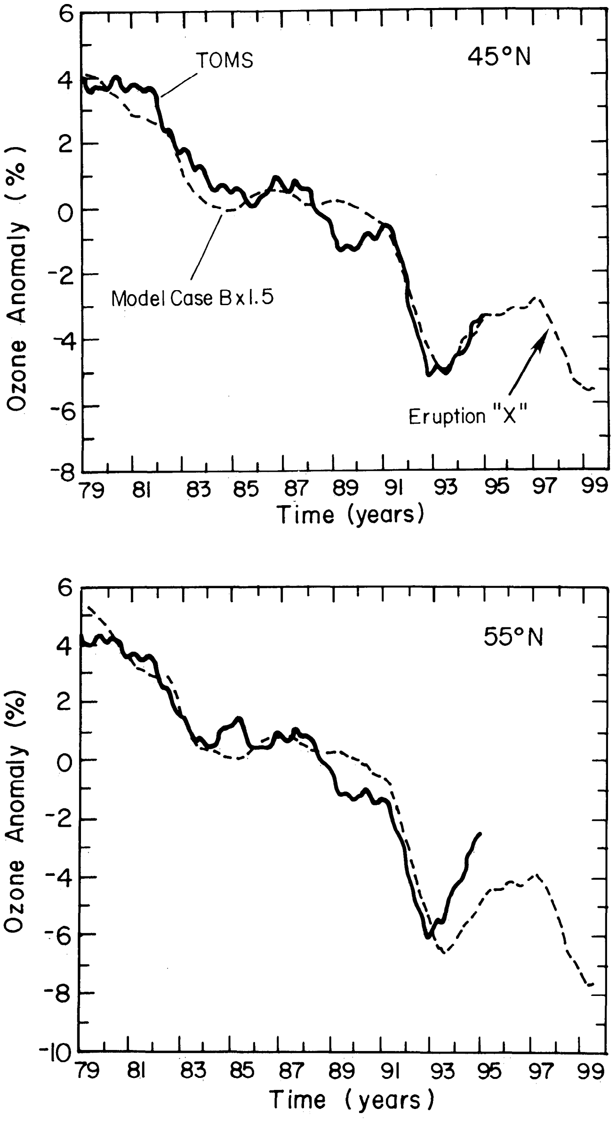

Figure 10. As in Plate 1, but extrapolated to the future. An exponential decay of Pinatubo aerosols is assumed until mid-1997, followed arbitrarily by an eruption "X" of the same characteristics as Pinatubo. TOMS data analysis is from *Randel et al.* [1995].

5. Summary and Implication for the Future

We have shown that observed variations in liquid stratospheric aerosols played an important role in determining both the trends and short-term fluctuations in ozone at northern midlatitudes since 1979. The observed timing of ozone changes at 45°N and 55°N is remarkably well simulated when aerosol observations are considered together with trends in anthropogenic chlorine and bromine, although the magnitude of the calculated changes is about 50% smaller than observations of the column and about a factor of 2 smaller than observations of the local depletion near 20 km. The modeled ozone depletions are driven mainly by changes in the NO_x/NO_y abundance of the lower stratosphere, which in turn enhances the ClO/Cl_y ratio and hence the chlorine-catalyzed ozone depletion for contemporary levels of total stratospheric chlorine. Direct observations of both NO_x/NO_y and ClO/Cl_y confirm the model chemistry at the key altitude of 20 km and support the conclusions regarding the importance of aerosols for midlatitude ozone trends and variability.

The discrepancy between the calculated and observed magnitude of the ozone depletion could be due to shortcomings in modeled photochemistry or dynamics or both. Possible explanations include new chemistry such as coupling of chlorine and/or bromine with iodine [*Solomon et al.*, 1994], an enhanced role for the reaction of $BrONO_2$ with H_2O, and/or increased bromine abundances in the lowest part of the stratosphere due, for example, to convection of short-lived gases or other sources. Trends in methane, nitrous oxide, and carbon dioxide appear unlikely to significantly affect the ozone column trends at midlatitudes according to the present model.

The observed time dependent fluctuations in aerosol content and sunspot number since 1979 display important similarities. Hence depending upon the analysis technique used, the important aerosol effects demonstrated here could be confused with solar signals in statistical studies of ozone trends.

The results presented in this paper underscore the need to carefully consider any process that could increase aerosol abundances in evaluations of future ozone trends. For example, subsonic and supersonic aircraft emissions can increase the stratospheric aerosol burden [*Hofmann*, 1991; *Fahey et al.*, 1995] and hence deplete ozone. Indeed, the present study taken together with the work of *Bekki and Pyle* [1992], *Fahey et al.* [1995], *Pitari et al.* [1993], and *Weisenstein et al.* [1991] suggests that such processes could be more important for stratospheric ozone loss than aircraft NO_x releases, at least until the stratospheric chlorine content is lowered substantially from current levels. *Bekki and Pyle* [1992] indicate that the large particle size group of global stratospheric aerosol may already have been increased by about 60% due to subsonic aircraft emissions; Figures 6-7 show that such a change could be significant in the context of ozone trends.

Finally, we briefly discuss the implications of this study for the near-term future of the ozone layer, particularly the expected chlorine peak and recovery. Figure 10 presents an extension of the results shown in Plate 1 where we assume for the purpose of illustration that the stratospheric aerosol content will decay with an exponential time scale of 3.5 years from its Pinatubo-enhanced values from 1995 to mid-1997. At that time, we arbitrarily impose an eruption of the same characteristics as Pinatubo, denoted here as eruption "X". The TOMS data shown is from the *Randel et al.* [1995] analysis.

Figure 10 shows that a reduction of stratospheric aerosol from 1993 to mid-1997 would be expected to lead to an averaged recovery of northern midlatitude stratospheric ozone depletion, although individual years (particularly unusually cold ones such as the winter of 1994-1995, see *Angell et al.*, [1995] may display variability, perhaps like that of 1989. The expectation of an averaged recovery in coming years follows from the fact that the chlorine content of the stratosphere is increasing slowly and is expected to peak around 1997 [*WMO/UNEP*, 1994], while the aerosols from Pinatubo are being removed. Unlike the period just after El Chichon, when the model results suggest that increasing stratospheric chlorine and bromine abundances approximately canceled the decreasing aerosol content to give rise to the relatively constant ozone losses observed from about 1986-1989, the period post-Pinatubo is expected to be characterized by much slower increases in halogen source gases and hence substantial recovery unless another major eruption occurs very soon. Figure 10 also shows that the future peak ozone loss at northern midlatitudes could certainly occur after the peak chlorine loading, depending upon the timing and magnitude of future volcanic eruptions such as "X". Similarly, sustained or unusually high volcanic activity (e.g., larger eruptions than Pinatubo, or several in succession) would be expected to impact strongly the future ozone layer, particularly if they occur during the coming decade of peak chlorine loading and slow recovery. While Pinatubo had a very large impact on the stratosphere, larger eruptions have been documented in the past [see *Sato et al.*, 1993; *McCormick et al.*, 1995].

In summary, the modeled response of northern midlatitude ozone over the past 15 years and comparison to observations strongly indicates that stratospheric aerosols influenced the trends and variability of anthropogenic ozone depletion due to the increasing burdens of chlorine and bromine. Stratospheric aerosols will likely also play a role in the future, as stratospheric chlorine and bromine slowly decay from their current, unprecedented levels.

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R.R. Garcia, NCAR, Boulder, CO, 80307. (e-mail: rgarcia@acd.ucar.edu)

M.P. McCormick, L.R. Poole, and L.W. Thomason, Aerosol Research Branch, NASA Langley Research Center, Hampton, VA 23665.

S. Solomon and R.W. Portmann, NOAA, R/E/AL8, 325 Broadway, Boulder, CO 80303. (e-mail: bportmann@al.noaa.gov)

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