Comparing simultaneous stratospheric aerosol and ozone lidar measurements with SAGE II data after the Mount Pinatubo eruption

G. K. Yue,[1] L. R. Poole,[1] M. P. McCormick,[1] R. E. Veiga,[2] P. -H. Wang,[3] V. Rizi,[4] F. Masci,[5] A. D'Altorio,[4] and G. Visconti[4]

Abstract. Stratospheric aerosol and ozone profiles obtained simultaneously from the lidar station at the University of L'Aquila ($42.35°N$, $13.33°E$, 683m above sea level) during the first 6 months following the eruption of Mount Pinatubo are compared with corresponding nearby Stratospheric Aerosol and Gas Experiment (SAGE) II profiles. The agreement between the two data sets is found to be reasonably good. The temporal change of aerosol profiles obtained by both techniques showed the intrusion and growth of Pinatubo aerosols. In addition, ozone concentration profiles derived from an empirical time-series model based on SAGE II ozone data obtained before the Pinatubo eruption are compared with measured profiles. Good agreement is shown in the 1991 profiles, but ozone concentrations measured in January 1992 were reduced relative to time-series model estimates. Possible reasons for the differences between measured and model-based ozone profiles are discussed.

Introduction

A major volcanic eruption such as El Chichon, and most recently Mount Pinatubo, produces considerable amounts of sulfate aerosol in the stratosphere. It has been suggested that heterogeneous chemical reactions on the surfaces of such aerosol particles in the lower stratosphere may accelerate the conversion of nitrogen pentoxide to nitric acid [*Hofmann and Solomon*, 1989; *Rodriguez et al.*, 1991]. As a result, more free chlorine radicals are available to destroy ozone. *Hofmann and Solomon* [1989] showed evidence to demonstrate the depletion of ozone due to the increase in aerosol surface area after the El Chichon eruption. Since the Pinatubo eruption was stronger than El Chichon [*McCormick and Veiga*, 1992], more ozone destruction would be expected in the aftermath of Pinatubo [*Prather*, 1992; *Rodriguez et al.*, 1994]. Total ozone levels observed in 1992 were found to be lower than in previous years [*Hofmann et al.*, 1993; *Bojkov et al.*, 1993; *Grant et al.*, 1994]. In order to critically test the relationship between aerosol properties and ozone depletion, it is essential to have simultaneous post-eruption data on both stratospheric aerosol and ozone.

One of the important sets of simultaneous aerosol and ozone measurements is that obtained by the Stratospheric Aerosol and Gas Experiment (SAGE) II [*McCormick*, 1987],

[1]Atmospheric Sciences Division, NASA Langley Research Center, Hampton, VA

[2]Science Applications International Corporation, Hampton, VA

[3]Sciences and Technology Corporation, Hampton, VA

[4]Universita' Degli Studi di L'Aquila, Italy

[5]Istituto Nazionale di Geofisica, Roma, Italy

which was launched on the Earth Radiation Budget Satellite (ERBS) on October 5, 1984. The SAGE II instrument has seven radiometric channels centered at wavelengths of 1.02, 0.94, 0.6, 0.525, 0.453, 0.448, and 0.385 μm. The measured data are inverted to obtain profiles of the aerosol volume extinction coefficient at four different wavelengths and profiles of ozone (as well as NO_2 and water vapor) concentration.

The lidar station at the University of L'Aquila ($42.35°N$, $13.33°E$, 683m above sea level) includes both aerosol and differential absorption lidar (DIAL) systems. The aerosol lidar system utilizes a Rhodamine 6G dye laser (1 Joule per pulse) and has been typically operated at the wavelength of 0.590 μm with a pulse repetition rate of about 0.15 Hz [*D'Altorio et al.*, 1981]. This system was used to detect volcanic aerosols after the Mount St. Helens eruption [*D'Altorio et al.*, 1981] and the El Chichon eruption [*D'Altorio and Visconti*, 1983]. The DIAL system for measuring stratospheric ozone profiles uses an excimer laser emitting at 0.308 μm and 0.351 μm, with typical output energies of 90 and 70 mJ per pulse, respectively [*D'Altorio et al.*, 1992]. Simultaneous aerosol and ozone lidar measurements taken by this lidar station in the post-Pinatubo period from July 1991 to December 1992 were reported by [*D'Altorio et al.*, 1993a and 1993b]. Comparison between the DIAL results and ECC ozonesonde measurements showed differences in the range of 5% to 20%. Sensitivity studies demonstrated that knowledge of the aerosol optical properties is important in retrieving the ozone concentration.

In this paper, we report the results of intercomparisons of aerosol and ozone profiles obtained nearly simultaneously (within 1 day) by the University of L'Aquila lidar systems and by SAGE II at locations within 5° in both latitude and longitude from L'Aquila. The period of comparison is between the eruption of Mount Pinatubo on June 15, 1991, and the end of January 1992, during which there were six measurement sets which meet these criteria.

An empirical time-series model for estimating ozone concentrations based on SAGE II monthly mean ozone data for the period October 1984 through June 1991 has recently been developed [*McCormick et al.*, 1992]. In this model, the ozone mixing ratio at a given altitude and latitude can be estimated by the following expression:

$$O_3 (t) = \mu + \alpha\, t + \beta \times \text{harmonics} + \text{noise}$$

where t is time, μ is a constant offset, α is the linear trend coefficient, and β is a collection of coefficients characterizing the amplitudes and phases of seasonal, semiannual, and quasi-biennial (30-month period) oscillations. The noise component represents the residual correlation. According to a paper by *McCormick et al.* [1989], the μ value is about $2.2\times10^{12}\text{cm}^{-3}$ at 30 km for latitude band $45°N$-$55°N$. The annual oscillation is about 15% of the mean value and the semiannual oscillation is about 2% of the mean value. The linear trend was estimated to be extremely small by *McCormick et al.* [1992]. The quasi-biennial amplitude is about 4% of the mean at 30 km and mid-latitudes [*Zawodny and McCormick*, 1991]. This linear regression model has been used successfully to study global trends in stratospheric column ozone and ozone profile trends [*McCormick et al.*, 1992], and as a reference for studying tropical ozone de-
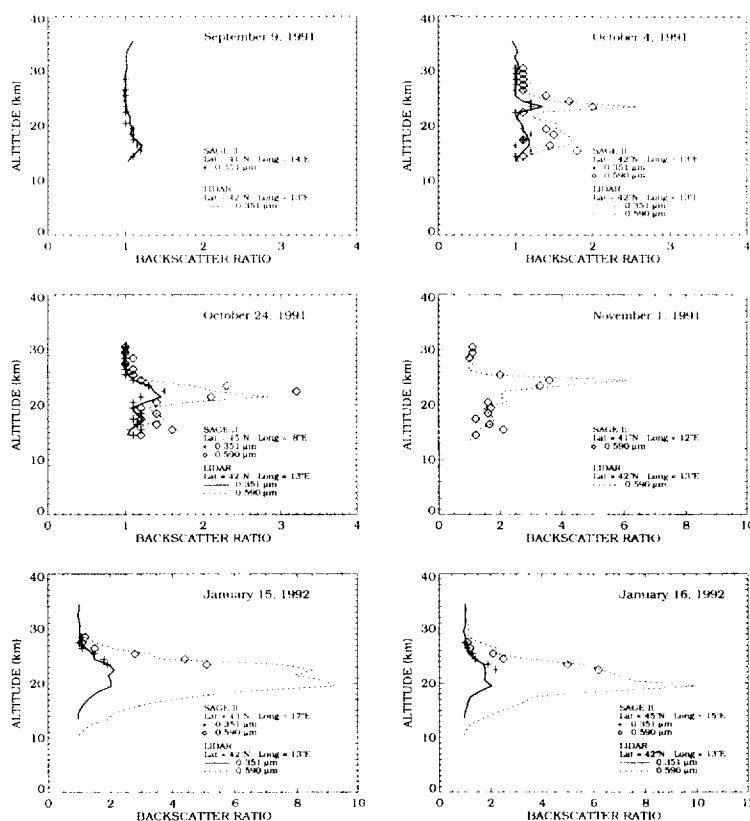
Figure 1. Profiles of backscatter ratio obtained from lidar (solid line, 0.351 μm; dotted line, 0.590 μm), and from SAGE II (cross, 0.351 μm; diamond, 0.590 μm).

creases after the Pinatubo eruption [*Grant et al.*, 1994]. The modeling results have also been used to compare with in situ measurements during the Stratospheric Photochemistry, Aerosols and Dynamics Expedition Measurements (SPADE) campaign in May 1993 [*Yue et al.*, 1994]. In this paper, we also present for comparison model-derived ozone profiles on those days for which both lidar and satellite ozone measurement profiles are available.

Lidar and SAGE II Observations

Figures 1 and 2 present the backscatter ratio (the ratio of aerosol plus molecular backscatter to molecular backscatter) and ozone concentration profiles, respectively, obtained from SAGE II and the University of L'Aquila aerosol lidar and ozone DIAL. SAGE II-measured aerosol extinction coefficients were converted to backscatter ratios at 0.351 and 0.590 μm using the method described by *Wang et al* [1989]. The time-series model-derived ozone profiles based on SAGE II measurements before the eruption of Mt. Pinatubo are also shown in Figure 2. The date and location of the measurements are given in each figure. Only backscatter data were available from the lidar on November 1, 1991. It should be noted that the original lidar measurements of ozone and backscatter ratio have a resolution of 0.3 km. However, since the vertical resolution of SAGE II profile is 1 km, the lidar profiles have been smoothed to 1-km resolution in order to make a more appropriate comparison.

The September 9, 1991, aerosol data in Figure 1 show the presence of a small aerosol layer near 16 km. The next five frames show a gradual increase in backscatter ratio and the thickness of the aerosol layer with time. This indicates the arrival of Pinatubo volcanic material and the growth of volcanic aerosols due to the conversion of SO_2 to H_2SO_4 followed by different microphysical processes including nu-

cleation, coagulation, and heterogeneous condensation on the surface of aerosol particles. No SAGE II data below 24 km are shown in January 1992 because the uncertainty of SAGE II data was so large that the inferred backscatters were unreliable at these altitudes. In general, the location of the peak and thickness of the volcanic layer shown at 0.351 μm and 0.590 μm are comparable. As pointed out by *Wang et al.* [1989], the best information on the aerosol size distribution embedded in the SAGE II multiwavelength aerosol extinction measurements is in the radius range from 0.25 to 0.80 μm. *Lenoble et al.* [1984] have noted that the retrieval method only produces an equivalent size distribution which yields the same aerosol extinctions as the true aerosol size distribution. The analysis by *Wang et al.* [1989] showed that the uncertainties of the retrieved aerosol size distribution for large particles may be as high as about 30%. It should also be noted that SAGE II measurement is a horizontal average over about 200 km. Discrepancies between the SAGE II and lidar measurements shown in Figure 1 may be partially due to the extinction-to-backscatter conversion scheme, but they may also reflect the variability of the volcanic layer in time and space since the instruments sample different volumes of air at different times.

The properties of early Pinatubo aerosols have been reported by several investigators [*Winker and Osborn*, 1992; *DeFoor et al.*, 1992; *Brock et al.*, 1993; *Deshler et al.*, 1993]. It is of interest to note that ruby lidar measurements (λ=0.6943 μm) at Hampton, Virginia, USA (37.1°N, 76.3°W) detected the arrival of Pinatubo aerosols associated with a tropospheric anticyclonic cell over Northern America in early August 1991 [*Woods et al.*, 1994]. The peak backscatter ratio and the integrated backscatter gradually increased from August 1991 through later February 1992, when maximum values were observed. On January 16, 1992, when Pinatubo aerosols were more uniformly distributed in the northern mid-latitudes, a maximum backscatter ratio of 9.6 was observed at 19.9 km in Hampton. This is com-
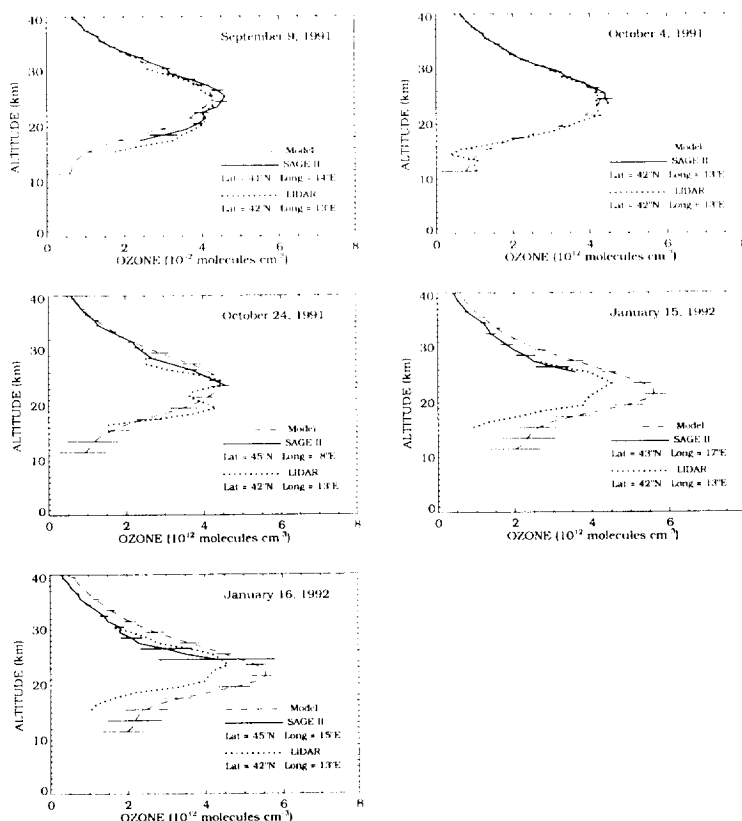
Figure 2. Profiles of ozone concentration obtained from lidar (dotted line), SAGE II (solid line), and time-series model (dashed line).

parable with the maximum backscatter ratio of about 10.0 at 19.5 km measured on the same day in Italy as shown in Figure 1.

There is a good agreement between the SAGE II and lidar ozone concentration profiles in all five frames of Figure 2. On January 15 and 16, 1992, thick volcanic layers were present over the lidar site, as demonstrated by the corresponding backscatter profiles shown in Figure 1. Both aerosol and ozone make major contributions to the extinction at 0.60 μm measured by SAGE II, and the aerosol contribution at this wavelength is estimated by extrapolating from the aerosol extinction at 1.02 μm. Thus, the presence of heavy aerosol loading reduces the reliability of the retrieved ozone concentration. For this reason, no SAGE II ozone concentration data below 24 km in January 1992 are shown in Figure 2. For data above 24 km, the small differences between SAGE II and lidar profiles are within the limits of individual uncertainties.

Comparison of time-series model-based ozone profiles with the measured profiles shows good agreement for the September 9 and October 4, 1991, profiles. Good agreement also is shown in the October 24 profiles for altitudes below 25 km. For altitudes above 25 km, the model-based ozone profile is slightly higher than the measurement profiles. However, large differences are shown in the January 15 and 16, 1992, profiles, especially at altitudes below 25 km. Lower than normal ozone measurements following the eruption of Mt. Pinatubo were reported by several investigators [Bojkov et al., 1993; Hofmann et al., 1993; Planet et al., 1994]. The ozonesonde data presented by Grant et al. [1994] have shown that tropical ozone column in 1992 was reduced relative to the historical record. The impact of the Mt. Pinatubo eruptions on the total column ozone measured from the Nimbus 7 TOMS and the NOAA - 11 SBUV/2 spectrometers has also been studied by Chandra [1993]. He concluded that about 2–4% decrease in the column ozone within a few months after the eruption can be contributed to volcanic aerosols. Schoeberl et al.

[1993] reported up to a 6% decrease in equatorial total ozone relative to climatological values about a month following the Pinatubo eruption. This is in good agreement with the modeling results of the ozone response to enhanced heterogeneous processing after the eruption [Rodriguez et al., 1994]. Although our modeling results have relatively large uncertainties and local synoptic conditions may influence ozone concentration, the relatively large departures of the lidar-derived ozone from the zonal mean climatology shown in Figure 2 may reflect that ozone has been depleted as a consequence of heterogeneous chemical reactions catalyzed by the Pinatubo aerosols. However, it should be noted that losses due to heterogeneous chemistry are much less than "catastrophic" as predicted by Prather [1992].

Conclusions

There is generally good agreement between the lidar-measured and SAGE II-deduced backscatter ratio profiles from September 1991 to January 1992. The change in the spectral characteristics of the backscatter ratio with time immediately after the arrival of Pinatubo aerosols was observed by lidar and confirmed by the SAGE II extinction measurements. The gradual increase of peak backscatter ratio with time clearly indicates the transport and growth of volcanic aerosols at mid-latitudes after the volcanic eruption. Some discrepancies between the lidar and SAGE II aerosol data are seen at lower altitudes. These may be due to the inherent spatial averaging in SAGE II measurements or the conversion of SAGE II aerosol extinction coefficient to backscatter ratio. However, they may also reflect the true variability of the volcanic layer in time and space.

There is good agreement between all SAGE II and lidar ozone concentration profiles. When time-series model estimates and lidar measurements of ozone concentrations were compared, good agreement was found in the September 9 and October 4, 1991, profiles. Model-based ozone concen-

trations at altitudes above 25 km on October 28, 1991, were slightly higher than the measured values. However, ozone concentrations measured on January 15 and 16, 1992 were reduced significantly relative to time-series model estimates. The influence of local synoptic conditions which are not included in the model may be partially responsible for the observed ozone differences. However, a major contribution may be heterogeneous chemical reactions on the surface of Pinatubo aerosols that lead to the depletion of ozone. The reduction in ozone concentration after the Pinatubo eruption reported in this study is consistent with other measurement and modeling results.

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G. K. Yue, L. R. Poole, and M. P. McCormick, Atmospheric Sciences Division, NASA Langley Research Center, Hampton, VA 23681

R. E. Veiga, Science Applications International Corporation, Hampton, VA 23666

P. -H. Wang, Sciences and Technology Corporation, Hampton, VA 23666

V. Rizi, A. D'Altorio, and G. Visconti, Dipartimento di Fisica, Universita' Degli Studi di L'Aquila, via Vetoio, 67010, Coppito L'Aquila, Italy

F. Masci, Istituto Nazionale di Geofisica, via di Vigna Murata, 605, 00143 Roma, Italy