JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 100, NO. D9, PAGES 18,855–18,862, SEPTEMBER 20, 1995

Pinatubo aerosols and the behavior of ozone inferred from backscatter ultraviolet measurements on the space shuttle

Guoyong Wen and John E. Frederick
Department of the Geophysical Sciences, University of Chicago, Illinois

Ernest Hilsenrath
Laboratory for Atmospheres, NASA Goddard Space Flight Center, Greenbelt, Maryland

Abstract. Backscattered radiation measurements from the shuttle solar backscatter ultraviolet spectral radiometer were used to examine aerosol optical properties and a possible ozone decrease in the tropical region after the eruption of Mt. Pinatubo. Observations made on two flights separated in time by 10 months show that backscattered ultraviolet radiation was greatly enhanced after the eruption as compared to the earlier measurements. The enhancements in backscattered radiation exhibit a special wavelength dependent signature. The peak percentage enhancement was found near 302 nm for solar zenith angles greater than 20°, corresponding to the latitude band 14°S–14°N. Radiative transfer calculations indicate that the wavelength dependence of the enhancement is consistent with an ozone decrease in the altitude region of the Pinatubo aerosol clouds. The Pinatubo aerosol optical depth was found to vary in the range 0.07–0.53 in the ultraviolet region at tropical latitudes. Furthermore, a 16–22 Dobson unit decrease of ozone in the altitude region occupied by the aerosol cloud is consistent with the spectral dependence of the enhanced backscattered radiance.

Introduction

A good deal of attention has focused on the potential effects of a volcanic eruption on stratospheric ozone. Unusually low ozone amounts were reported worldwide from late 1991 to 1993 [*Bojkov et al.*, 1993; *Chandra*, 1993; *Gleason et al.*, 1993; *Kerr et al.*, 1993; *Komhyr et al.*, 1994; *Hofmann et al.*, 1994], possibly related to the enhancement of sulfur aerosol loading in the stratosphere after the eruption of Mt. Pinatubo (15.14°N, 120.35°E) on June 15–16, 1991. Heterogeneous reactions on aerosols in dense volcanic clouds could lead to localized loss of ozone in the midlatitude stratosphere [*Hofmann and Solomon*, 1989; *Prather*, 1992]. At tropical latitudes the effects of radiative heating, changes in circulation patterns, and changes in photolysis rates associated with aerosol become major effects [*Brasseur and Granier*, 1992; *Kinne et al.*, 1992; *Tie et al.*, 1994].

In this study we use measurements from the shuttle solar backscatter ultraviolet (SSBUV) spectral radiometer to investigate the optical properties of Pinatubo aerosols and possible changes in ozone amount. The backscattered ultraviolet (UV) radiation in the tropical region observed during August 3–6, 1991, is compared with that from an unperturbed atmosphere 10 months before. In order to reduce effects from tropospheric clouds and make the analysis more straightforward we used a modified spatial-spectral coherence technique to eliminate cloudy scenes. Then a discrete-ordinate radiative transfer model was employed to interpret anomalies in backscattered UV radiation after the Pinatubo eruption.

Paper number 95JD01839.
0148-0227/95/95JD-01839$05.00

Data Description

The SSBUV experiment was designed to provide an in-orbit calibration check of the SBUV/2 instruments flying on National Oceanic and Atmospheric Administration (NOAA) operational satellites [*Hilsenrath et al.*, 1988; *Frederick et al.*, 1990; *Hilsenrath et al.*, 1995]. The SBUV/2 sensors are flying on the NOAA 9, NOAA 11, and NOAA 14 spacecraft to provide continuous global ozone observations. The SSBUV instrument is identical to the SBUV/2, except for modifications which allow for flights on the shuttle. SSBUV has flown seven times since 1989, and one more flight is planned for 1995.

The SSBUV instrument consists of a grating spectrometer which operates from 160 to 405 nm. For ozone measurements the spectrometer steps consecutively through 12 wavelengths (252.2, 273.6, 283.2, 287.7, 292.4, 297.7, 302.0, 306.0, 312.7, 317.7, 331.4, and 340.0 nm) over 32 s, including an 8-s retrace. In the nadir the instrument views with an 11.3 × 11.3 degree field of view, corresponding to an approximate 50-km-square footprint which moves about 220 km during the entire spectral scan. Coincident and coaligned with the spectrometer data are photometer observations fixed in wavelength at 379 nm. The purpose of the photometer is to provide a continuous measure of relative scene brightness to pair with each step of the spectrometer.

Since SSBUV data are used as a calibration transfer standard, the calibration of the instrument is tracked with great precision [*Cebula et al.*, 1989; *Hilsenrath et al.*, 1993]. The radiometric stability has changed by only a few percent during its flight record, and this is tracked to better than 1% over the entire spectral range. Wavelength stability is also very high and has been tracked to better than 0.02 nm. The changes in radiances between pre- and post-Pinatubo conditions reported here are significantly larger than the uncertainties in the SSBUV calibrations.

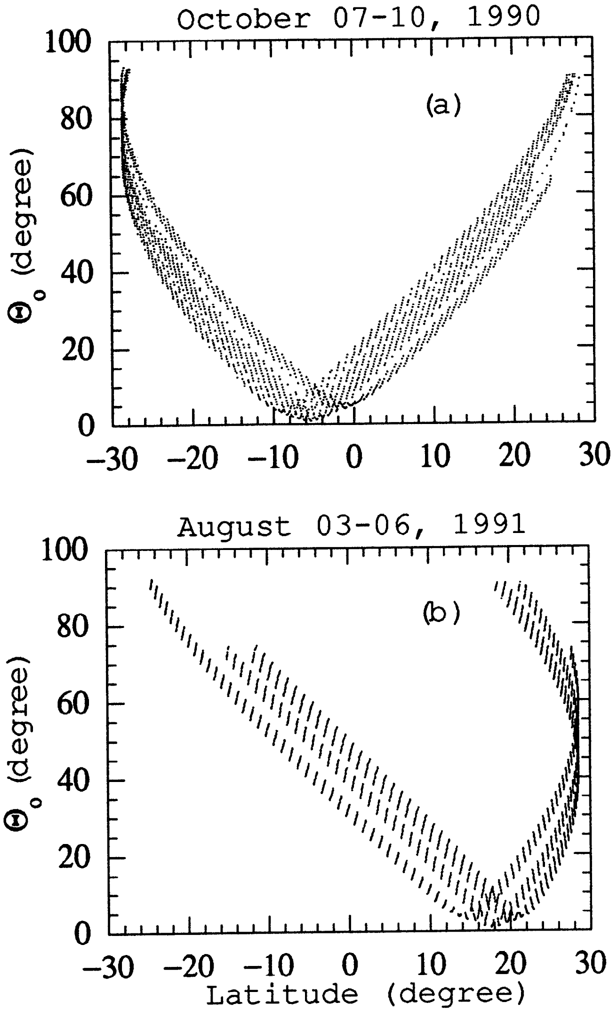

Figure 1. Latitude coverage and solar zenith angle variation of shuttle solar backscatter ultraviolet measurements for flights of (a) October 7–10, 1990, and (b) August 3–6, 1991.

The two flights of SSBUV studied here, October 7–10, 1990, and August 3–6, 1991, were confined to the latitude band 30°S–30°N, as shown in Figure 1. As the shuttle flies from west to east, the solar zenith angle (SZA) changes. A wide range of SZA is a unique feature of the flights, and this is useful in examining the optical properties of Pinatubo aerosols and related changes in ozone amounts. Even though both flights were over the tropical region, the orbits were not identical, and the SZA at a given latitude differs considerably between the flights. The measurements in the latitude band 20°N–30°N during the 1991 flight correspond to a range of SZA from 0° to 90°, while the SZA encompasses only 45°–90° for the 1990 flight over the same latitude band. In order to reduce effects related to latitudinal variations in ozone and differing SZAs we compared the observations posteruption and preeruption in the latitude band 20°S–20°N.

A directional albedo may be defined as the ratio of backscattered radiance to the incident solar irradiance normalized by the cosine of the SZA [*Wen and Frederick*, 1994]. This is

$$A(w, \theta_0) = \pi I(w, \theta_0)/[F(w) \cos(\theta_0)] \qquad (1)$$

where I and F are the backscattered UV radiance in the vertical direction and the incident solar irradiance at SZA equal to θ_0 and wavelength w, respectively. A similar definition applies to the photometer albedo at 379 nm, where one measurement is available to pair with each of the 12 values of w in (1).

The enhancement in backscattered albedo at wavelength w after the Pinatubo eruption may be defined as the relative difference between the albedos of posteruption and those 10 months before:

$$\text{Enhancement} = [A_2(w, \theta_0) - A_1(w, \theta_0)]/A_1(w, \theta_0) \qquad (2)$$

where $A_2(w, \theta_0)$ and $A_1(w, \theta_0)$ are the albedo at posteruption, measured on the 1991 flight of SSBUV, and that from an unperturbed atmosphere observed during the 1990 flight, respectively.

Observations

Identification of Cloud-Free Conditions

Backscattering by tropospheric clouds, especially those associated with horizontally inhomogeneous or broken scenes, is extremely difficult to model properly [*Wen and Frederick*, 1995]. In order to avoid these difficulties we examine only measurements from cloud-free scenes. The technique for identifying clear atmospheres proceeds as follows. Solar radiation at 379 nm can penetrate to the Earth's surface, and therefore the SSBUV photometer measurements may be used to identify clouds and define the surface reflectivity. It is expected that consecutive photometer measurements from a clear atmosphere will show less variability than those from a horizontally inhomogeneous cloudy scene. A cloudy atmosphere, on the other hand, is more effective in reflecting solar radiation than a clear sky. Hence the criteria used to identify radiances from a clear atmosphere include both small absolute photometer readings and small spatial variability.

Figure 2 shows the spatial variation of the photometer data recorded during complete step scans of SSBUV for an overhead sun and for larger SZAs, where each point is based on 12 albedos. The horizontal axis is the difference between the first and the last of the 12 photometer albedos observed during one step scan. The vertical axis represents the standard deviation of the 12 photometer albedos in the scan corresponding to the point on the horizontal axis. A "V structure" is apparent in Figure 2. These V structures arise from spatial variations in tropospheric optical properties along the path of SSBUV. The cluster at the bottom of the V structure represents measurements from horizontally homogeneous atmospheric conditions. The left branch of the V structure represents measurements when the shuttle flies from a clear atmosphere to a cloudy atmosphere or from optically thin clouds to optically thick clouds. The points in the right branch of the V structure are observed when the shuttle flies from a cloudy to a clear atmosphere or from optically thick clouds to optically thin clouds. A standard deviation of 0.01 in the photometer is chosen as a cutoff value to define homogeneous scenes in the following study.

Among the measurements for horizontally homogeneous scenes those from a clear atmosphere may be separated from those for homogeneous cloudy scenes by examining the frequency distribution of the measurements. Histograms of measurements from homogeneous scenes are constructed as shown in Figure 3. The frequency distributions suggest that the albedos clustered at small values are from clear atmospheres. A few measurements outside the major cluster are probably from horizontally homogeneous cloudy atmospheres and are excluded from further analysis.

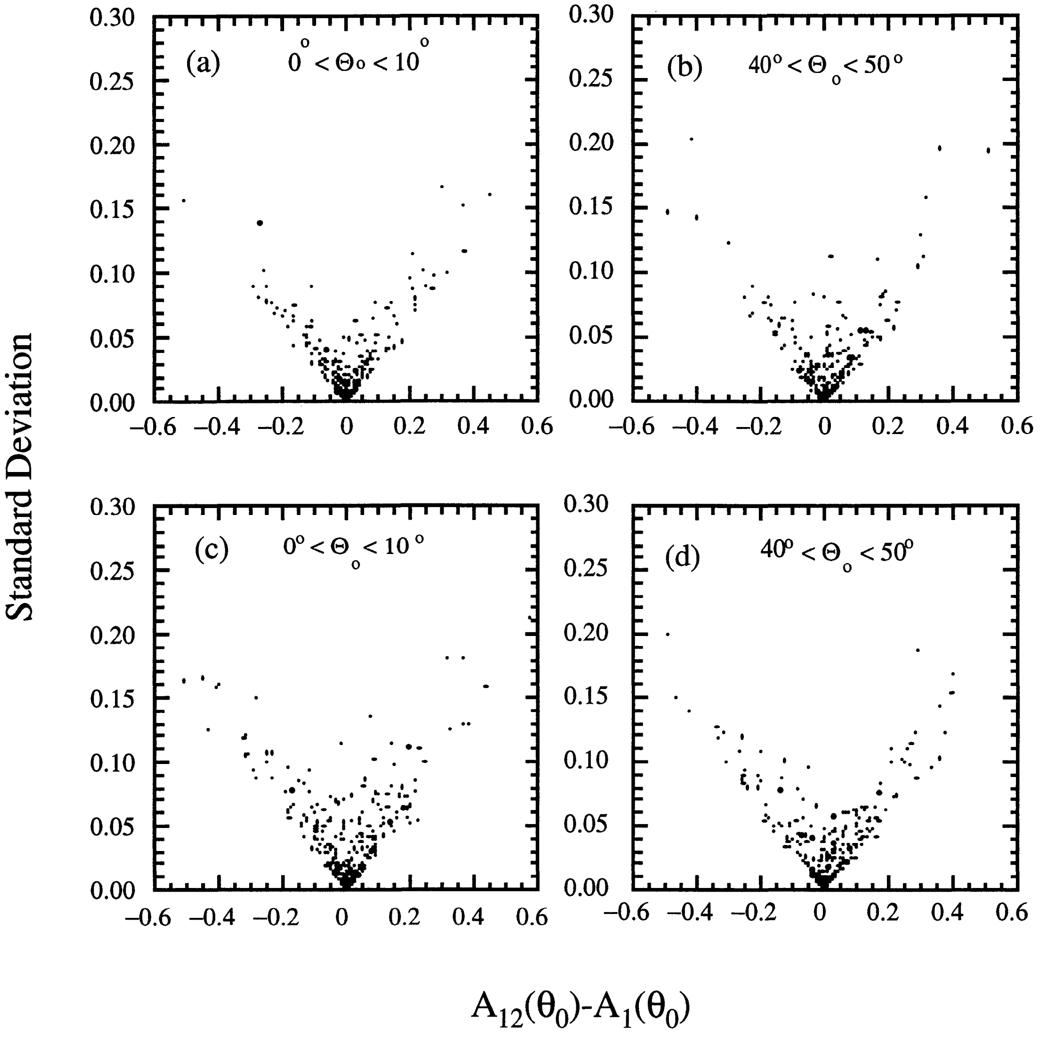

Figure 2. Spatial variation of the photometer albedos at 379 nm for (a) 1990 flight with solar zenith angle 0° $< \theta_0 <$ 10°; (b) 1990 flight and 40° $< \theta_0 <$ 50°; (c) 1991 flight and 0° $< \theta_0 <$ 10°; (d) 1991 flight and 40° $< \theta_0 <$ 50°. The abscissa, A_{12}-A_1, is the difference between the last (twelfth) and the first photometer albedo in one complete scan, and the ordinate represents the standard deviation of the corresponding 12 photometer albedos.

It is interesting to note that the peaks of the clear-sky portion of the albedo histograms shifted to larger values after the Pinatubo eruption. The peaks shifted from 0.33 to 0.35 for an overhead sun and from 0.31 to 0.33 for SZAs of 40°–50°. Because the Pinatubo aerosol clouds may be horizontally homogeneous over the distance covered by one SSBUV step scan, about 220 km, and the optical depths of the aerosol clouds are small compared to those of tropospheric clouds, the shifts in the histograms may reflect the enhancement of backscattered radiation from Pinatubo aerosols.

Enhancements in the Spectral Albedo

As the shuttle flies from west to east in the tropical region, the SZA changes rapidly. For example, the local SZA varies from 0° to 60° in the latitude band 20°S–20°N. To minimize the effects of these changes, we compared the backscattered spectral albedos from the SSBUV flight in 1991 with those measured 10 months earlier, before Pinatubo, in the same bins of SZA. Since most of the measured UV radiation at wavelengths less than 290 nm is backscattered above 30 km, where the volcanic disturbance was small, we examine only the measurements at wavelengths greater than 290 nm.

The enhancements in spectral albedo shown in Figure 4 define the relative difference between the averaged albedos from posteruption and those from preeruption for each of six SZA bins. Both posteruption and preeruption measurements in Figure 4 are from cloud-free scenes over the ocean. This reduces ambiguities related to changes in reflectivity of clouds and underlying surfaces. The enhancements are small for SZAs less than 20°. The changes in the backscattered albedo are only about 2–3% at 340 nm where ozone absorption is weak. The enhancements become more pronounced as SZA increases. The enhancements for SZAs greater than 20° generally increase with wavelength from 290 nm, have a peak around 302 nm, and then decrease toward longer wavelengths. The backscattered albedos were increased by 10–25% at the peak and 7–8% at the long wavelength end of the spectra.

Radiative Transfer Studies

A discrete ordinates radiative transfer model, originally developed by *Stamnes et al.* [1988], is used to examine optical properties of Pinatubo aerosol clouds and possible changes in ozone amounts after the eruption. The model solves the one-

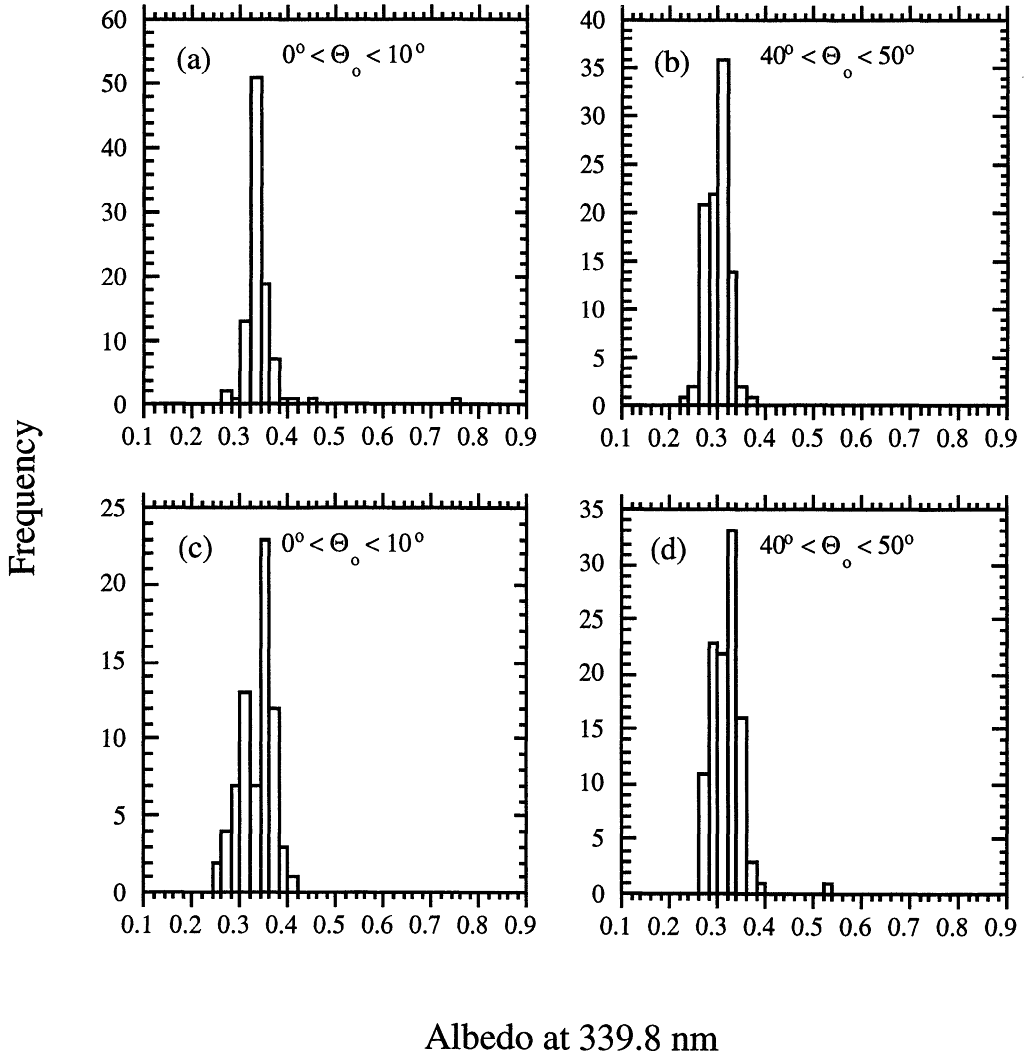

Figure 3. Frequency distributions of the photometer albedos from homogeneous scenes with standard deviation less than 0.01 for (a) 1990 flight with solar zenith angle 0° < θ_0 < 10°; (b) 1990 flight and 40° < θ_0 < 50°; (c) 1991 flight and 0° < θ_0 < 10°; (d) 1991 flight and 40° < θ_0 < 50°.

dimensional radiative transfer equation numerically for a vertically inhomogeneous atmosphere. The model has been used previously to evaluate changes in ozone abundance within the El Chichon aerosol clouds [*Wen and Frederick*, 1994]. The accuracy of the model depends on the number of streams used. As in the earlier work by *Wen and Frederick* [1994], we use 32 streams for an accuracy better than 1%. The U.S. Standard Atmosphere (1976) is used for the vertical profile of total number density. The Rayleigh scattering cross sections are taken from *World Meteorological Organization* [1985], and the absorption cross sections of *Molina and Molina* [1986] are used to calculate ozone extinction. For a given total ozone amount, statistical relations [*Bojkov*, 1969] are applied to estimate the vertical profile.

The Pinatubo eruption injected SO_2 into the stratosphere, and this gas eventually converted to sulfuric acid aerosol. A large increase of stratospheric aerosol optical depth has been found in a number of observations after the eruption [*McCormick and Veiga*, 1992; *Stowe et al.*, 1992; *Valero and Pilewskie*, 1992], and this increase acts to enhance the backscattered UV radiation. In this study an aerosol size distribution observed on August 2, 1991, [*Deshler et al.*, 1992] was used to calculate the scattering phase function via Mie theory [*Bohren and Huffman*, 1983]. Similar to the El Chichon aerosol phase function [*Wen and Frederick*, 1994], based on the size distribution of *Hofmann and Rosen* [1984], the Pinatubo aerosol phase function in Figure 5 has a strong forward peak and a secondary increase in the backward direction. The forward peak of the calculated phase function of Pinatubo aerosols is not as pronounced as that for El Chichon. This is primarily a result of the presence of more large droplets in the distribution used for the El Chichon aerosols than in the distribution assumed for Pinatubo.

As the size distribution of Pinatubo aerosol varies in space and time, so does the phase function. It is unrealistic and impossible to adopt a vertical profile of aerosol size distribution for each scene viewed by SSBUV. Several observed Pinatubo aerosol size distributions from different locations and times were reported to have similar means and standard deviations but different total number densities in the lognormal distribution [*Russell et al.*, 1993]. Because the total number density will not affect the shape of the aerosol phase function, we expect the single phase function used here to be a reasonable model for use in all of the radiative transfer calculations.

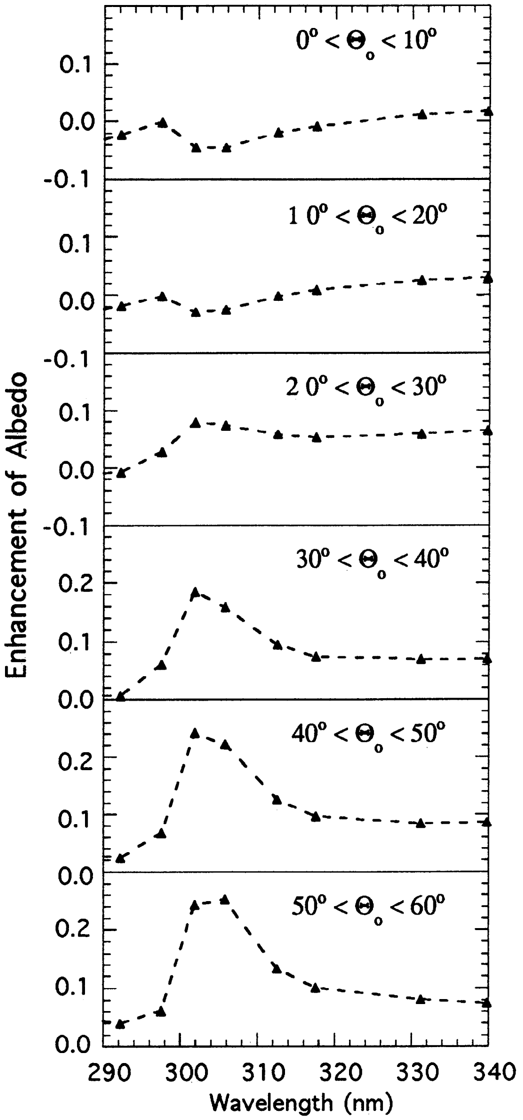

Figure 4. Averaged enhancements of spectral albedos for cloud-free atmospheres over the ocean. These were obtained by comparing the albedos after the Pinatubo eruption and those one year before in the same solar zenith angle bin. The latitude range is 20°S–20°N.

Since absorption by ozone is very small at 340.0 nm, the observed enhancement of spectral albedo at this wavelength was used to infer the aerosol optical depth. For simplicity the calculation is restricted to measurements over the ocean. The retrieved optical depths are presented in Table 1 for each SZA bin. From Figure 1 it is evident that each individual SZA bin is confined to a certain latitude band. The aerosol optical depth in Table 1 is therefore associated with these different latitude bands.

The backscattered UV radiation at the top of the atmosphere is the integrated result of scatterings from different levels. The backscattered UV radiation at shorter wavelengths comes primarily from higher altitudes, while that at longer wavelengths is contributed from lower levels. Hence the spectral dependence of the enhancement in backscattered UV radiation will depend on the vertical distribution of aerosols. Observations from the Stratospheric Aerosol and Gas Experiment and lidars indicate that most Pinatubo aerosol particles were concentrated between 16 and 26 km and appeared to have a one- or two-layer vertical structure [*McCormick and Veiga*, 1992; *Winker and Osborn*, 1992 a, b; *DeFoor et al.*, 1992; *Jager*, 1992; *Labitzke and McCormick*, 1992]. In order to have a realistic vertical distribution of aerosol loading, three different vertical distributions of aerosol optical depth similar to the observed backscatter ratios of *DeFoor et al.* [1992] are used in the calculations. These appear in Figure 6.

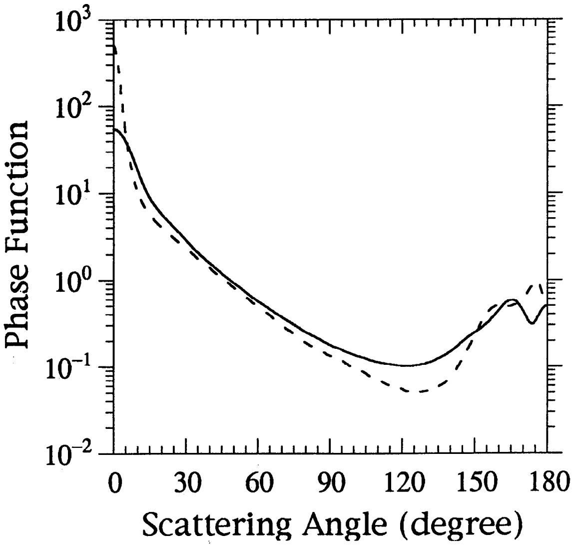

Figure 5. Phase functions computed for Pinatubo (solid curve) and El Chichon (dashed curve) aerosols.

The modeled enhancements in backscattered radiances from aerosol clouds with different optical depths are compared with observations in Figure 7. It is apparent that the aerosol clouds alone cannot produce the wavelength dependent enhancements over the entire range from 290 to 340 nm, regardless of the vertical distribution of aerosol optical depth. With different aerosol optical depths the modeled enhancements could resemble the observations either near the peak or at the longer end of the spectrum. A similar situation was noted for the El Chichon aerosol clouds [*Wen and Frederick*, 1994].

Since backscattering from aerosol clouds alone cannot produce the observed enhancements, the likely alternative is a decrease in stratospheric ozone abundance between the SSBUV flights of 1990 and 1991. It is expected that a decrease in the ozone amount within the aerosol layer will have little effect on backscattered UV radiation at both the short and long ends of the spectral region studied here. As stated previously, this is because the observed radiation at short wavelengths is backscattered from altitudes higher than affected by the eruption, while that at longer wavelengths is not altered by ozone.

Table 1. Derived Optical Depths of Pinatubo Aerosol Clouds for Given Solar Zenith Angles and Latitude Ranges

SZA, deg	Latitude, deg	Optical Depth
0–10	10–20N	0.07
10–20	6–18N	0.10
20–30	0–14N	0.27
30–40	6S–10N	0.35
40–50	10S–5N	0.47
50–60	12S–0	0.53

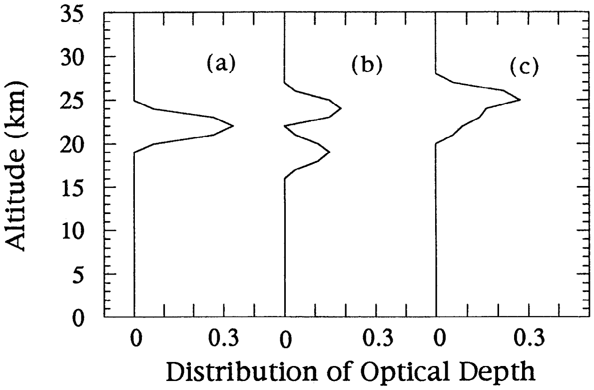

Figure 6. Three vertical profiles of aerosol optical depths after the Pinatubo eruption.

In the radiative transfer study a series of calculations was performed for various ozone decreases within the aerosol clouds. The vertical distribution of the percentage decrease in the ozone amount was assumed to be the same as the aerosol optical depth profiles in Figure 6. The modeled enhancements for three dense aerosol clouds are compared with observations in Figure 8. It is evident that the modeled enhancements closely resemble the observations when certain ozone decreases are assumed. A decline in column ozone of about 16–22 Dobson units (DU), being 6–8.5% out of the total of 260 DU, within the aerosol clouds is needed to produce enhancements close to the observations over the whole spectral region from 290 to 340 nm. This reduction in ozone corresponds to 20–27% of the unperturbed amount contained in the altitude region of the aerosol layer.

Summary and Discussion

Measurements of backscattered UV irradiance from SSBUV show large enhancements after the Pinatubo eruption as compared to data obtained 10 months earlier. The enhancements at SZAs greater than 30° increase with wavelength, reach a peak near 20–25% at about 302 nm, and decrease toward longer wavelengths. A smaller value and a weaker wavelength dependence were found for SZAs less than 30°. The apparent SZA dependence of the enhancements may reflect spatial variations owing to the coupling between SZA and latitude in the SSBUV measurements.

Aerosol optical depths were estimated based on an observed aerosol size distribution [*Deshler et al.*, 1992] and a discrete ordinates radiative transfer model [*Stamnes et al.*, 1988]. The aerosol optical depths associated with Pinatubo were found to range from 0.07 to 0.53 within the latitude band 12°S–20°N. An apparent systematic increase of the optical depth with SZA was found. This relationship may arise in part from the dependence of local SZA on latitude. This seems to be the case based on the aerosol optical depths inferred from advanced very high resolution radiometer measurements [*Stowe et al.*, 1992], where the optical depth over southern latitudes was evidently larger than on the northern side of the equator. Another factor which may contribute to the SZA dependence is the change in scattering phase function associated with changes in aerosol size distribution. A change of size distribution will certainly lead to a change in the phase function and hence a change in the retrieved aerosol optical depth. The systematic increase of the enhancement with SZA may indicate a different aerosol size distribution in the equatorial region as compared to the one observed by *Deshler et al.* [1992] over Wyoming.

Radiative transfer calculations indicate that aerosol clouds alone cannot produce the observed enhancement in backscattered UV radiation. In addition to scattering by aerosols, a decline in ozone amount between October 1990 and August 1991 in the altitude region of the aerosol layer is necessary to match the observed enhancements in irradiance between 290 and 340 nm. The data imply a decrease in column ozone of

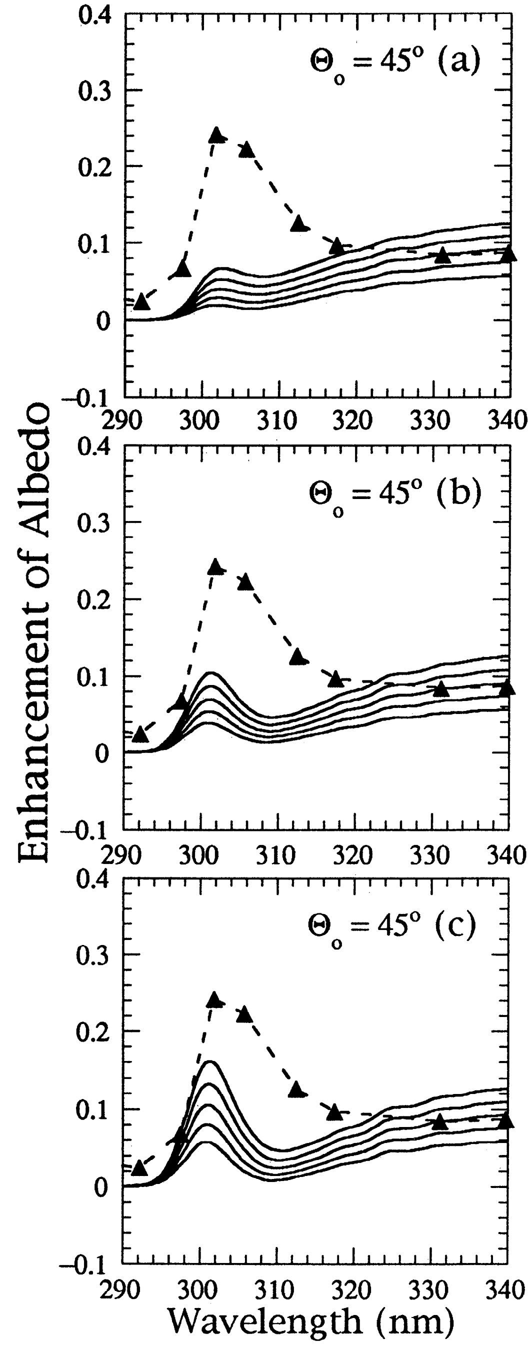

Figure 7. Comparison of observations (triangles with dashed lines) and model results for an aerosol layer with corresponding vertical profiles (a, b, c) in Figure 6 and optical depths (solid curves) of 0.3, 0.4, 0.5, 0.6, and 0.7 from lower to upper curves.

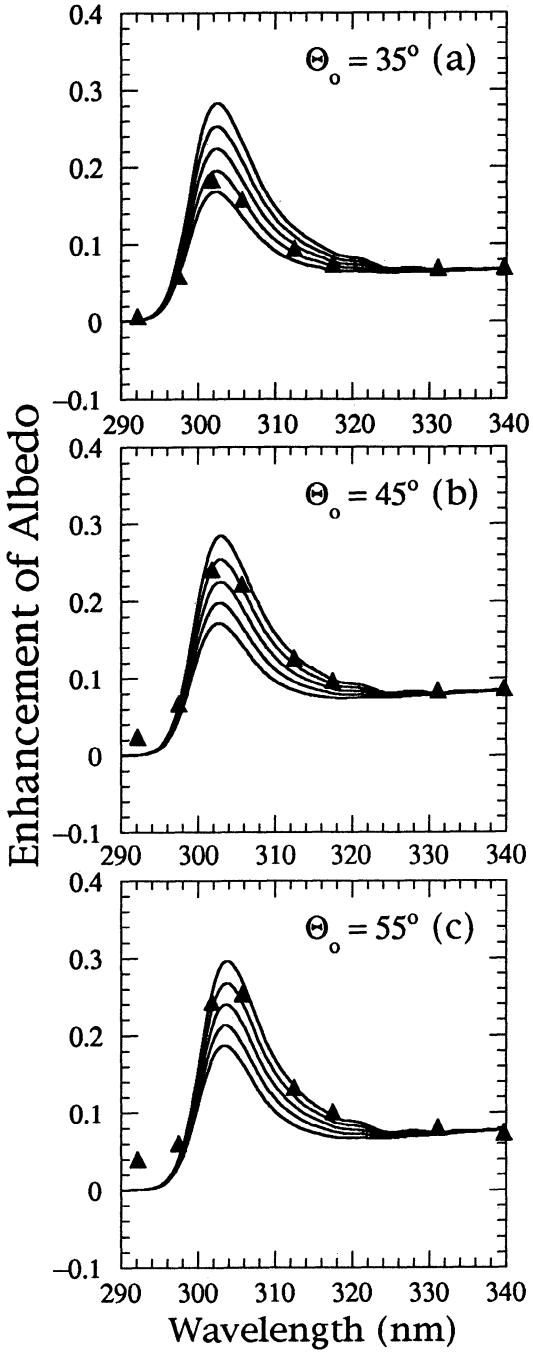

Figure 8. The observed (triangles) and modeled (curves) enhancements in spectral albedos, including both aerosol scattering and an ozone decrease by 5, 6, 7, 8, and 9% of the unperturbed column amount of 260 Dobson units from lower to upper curves: solar zenith angle (a) $30° < \theta_0 < 40°$, aerosol optical depth of 0.35; (b) $40° < \theta_0 < 50°$, aerosol optical depth of 0.47; (c) $50° < \theta_0 < 60°$, aerosol optical depth of 0.53.

about 16–22 DU. Changes in column ozone associated with Pinatubo have also been inferred from balloon-borne measurements made over Natal, Brazil [*Schoeberl et al.*, 1993] before and after the eruption. The posteruption data were taken, coincidentally, with the August 1991 flight of SSBUV. The balloon data indicate a local ozone depletion of about 20% in the altitude range 25–28 km, which was near the upper part of the aerosol layer. Changes in total column ozone observed in the balloon soundings were of the order of a few percent relative to the tropical ozone climatology of *Kirchoff et al.* [1991]. These changes are consistent with inferences based on SSBUV.

As stated above, the retrieved aerosol optical depth is dependent on the phase function adopted in the radiative transfer calculations. However, a change in the phase function would not alter the wavelength dependence of the enhancement due to aerosol scattering. Sensitivity studies show that the inferred ozone decrease within the aerosol clouds does not depend on details of the phase function. A decline in column ozone of several percent is required to produce the observed spectral dependence in the enhanced albedos, but it is not necessarily true that all of this change is attributable to the effects of Pinatubo. Natural variability related to the quasi-biennial oscillation (QBO) could also contribute to the changes in observed ozone amounts between 1990 and 1991. The stratospheric ozone abundance in the equatorial region was experiencing negative anomalies related to the phase of the QBO after the Pinatubo eruption [*Chandra*, 1993]. This could account for a decrease in column ozone of about 5% from 1990 to 1991. This leaves an estimated decline of 1–3.5% resulting from the perturbation provided by Pinatubo.

Acknowledgments. We are grateful to K. Stamnes for providing a copy of the radiative transfer code. This research was supported by the National Aeronautics and Space Administration under grant NAG5-873.

References

Bohren, C. F., and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, John Wiley, New York, 1983.

Bojkov, R. D., Computing the vertical ozone distribution from its relationship with total ozone amount, *J. Appl. Meteor.*, *8*, 284–292, 1969.

Bojkov, R. D., C. S. Zerefos, D. S. Balis, I. C. Ziomas, and A. F. Bais, Record low total ozone during northern winters of 1992 and 1993, *Geophys. Res. Lett.*, *20*, 1351–1354, 1993.

Brasseur, G. P., and C. Granier, Mount Pinatubo aerosols, chlorofluorocarbons and ozone depletion, *Science*, *257*, 1239–1242, 1992.

Cebula, R. P., E. Hilsenrath, and B. Guenther, Calibration of the shuttle borne Solar Backscatter Ultraviolet Spectrometer, *Proc. SPIE Int. Soc. Opt. Eng.*, *1109*, 205–218, 1989.

Chandra, S., Changes in stratospheric ozone and temperature due to the eruptions of Mt. Pinatubo, *Geophys. Res. Lett.*, *20*, 33–36, 1993.

DeFoor, T. E., E. Robinson, and S. Ryan, Early lidar observations of the June 1991 Pinatubo eruption plume at Mauna Loa Observatory, Hawaii, *Geophys. Res. Lett.*, *19*, 187–190, 1992.

Deshler, T., D. J. Hofmann, B. J. Johnson, and W. R. Rozier, Balloonborne measurements of the Pinatubo aerosol size distribution and variability at Laramie, Wyoming during Summer 1991, *Geophys. Res. Lett.*, *19*, 199–202, 1992.

Frederick, J. E., X. Niu, and E. Hilsenrath, An approach to the detection of long term trends in upper atmospheric ozone from space, *J. Atmos. Oceanic Technol.*, *7*, 734–740, 1990.

Gleason, J. F., et al., Record low global ozone in 1992, *Science*, *260*, 523–526, 1993.

Hilsenrath, E., D. Williams, and J. Frederick, Calibration of long term data sets from operational satellites using the space shuttle, *Proc. SPIE Int. Soc. Opt. Eng.*, *924*, 215–222, 1988.

Hilsenrath, E., D. E. Williams, R. T. Caffrey, R. P. Cebula, and S. J. Hynes, Calibration and radiometric stability of the Shuttle Solar Backscatter Ultraviolet (SSBUV) experiment, *Metrologia*, *30*, 243–248, 1993.

Hilsenrath, E., R. P. Cebula, M. T. Deland, K. Laamann, S. Taylor, C. Wellemeyer, and P. K. Bhartia, Calibration of the NOAA 11 SBUV/2 ozone data set from 1989 to 1993 using inflight calibration data and SSBUV, *J. Geophys. Res.*, *100*, 1351–1366, 1995.

Hofmann, D. J., and J. M. Rosen, Balloonborne particle counter observation of the El Chichon aerosol layers in the 0.01–1.8 μm radius range, *Geofis. Int.*, *23*(2), 155–185, 1984.

Hofmann, D. J., and S. Solomon, Ozone destruction through heterogeneous chemistry following the eruption of El Chichon, *J. Geophys. Res.*, *94*, 5029–5041, 1989.

Hofmann, D. J., S. J. Oltmans, W. D. Komhyr, J. M. Harris, J. A. Lathrop, A. O. Langford, T. Deshler, B. J. Johnson, A. Torres, and W. A. Mathews, Ozone loss in the lower stratosphere over the United States in 1992–1993: Evidence for heterogeneous chemistry on the Pinatubo aerosol, *Geophys. Res. Lett.*, *21*, 65–68, 1994.

Jager, H., The Pinatubo eruption cloud observed by lidar at Garmisch-Partenkirchen, *Geophys. Res. Lett.*, *19*, 191–194, 1992.

Kerr, J. B., D. J. Wardle, and D. W. Tarasick, Record low ozone values over Canada in early 1993, *Geophys. Res. Lett.*, *20*, 1979–1982, 1993.

Kinne, S., O. B. Toon, and M. J. Prather, Buffering of stratospheric circulation by changing amounts of tropical ozone: A Pinatubo case study, *Geophys. Res. Lett.*, *19*, 1927–1930, 1992.

Kirchoff, V. W., R. A. Barnes, and A. L. Torres, Ozone climatology at Natal, Brazil from in-situ ozonesonde data, *J. Geophys. Res.*, *96*, 10,899–10,909, 1991.

Komhyr, W. D., R. D. Grass, R. D. Evans, R. K. Leonard, D. M. Quincy, D. J. Hofmann, and G. L. Koenig, Unprecedented 1993 ozone decrease over the United States from Dobson spectrometer observations, *Geophys. Res. Lett.*, *21*, 201–203, 1994.

Labitzke, K., and M. P. McCormick, Stratospheric temperature increase due to Pinatubo aerosols, *Geophys. Res. Lett.*, *19*, 207–210, 1992.

McCormick, M. P., and R. E. Veiga, SAGE II measurements of early Pinatubo aerosols, *Geophys. Res. Lett.*, *19*, 155–158, 1992.

Molina, L. T., and M. J. Molina, Absolute absorption cross section of ozone in the 185- to 350-nm wavelength range, *J. Geophys. Res.*, *91*, 14,501–14,508, 1986.

Prather, M., Catastrophic loss of stratospheric ozone in dense volcanic clouds, *J. Geophys. Res.*, *97*, 10,187–10,191, 1992.

Russell, P. B., et al., Pinatubo and pre-Pinatubo optical-depth spectra: Mauna Loa measurements, comparisons, inferred particle size distributions, radiative effects, and relationship to lidar data, *J. Geophys. Res.*, *98*, 22,969–22,985, 1993.

Schoeberl, M. R., P. K. Bhartia, E. Hilsenrath, and O. Torres, Tropical ozone loss following the eruption of Mt. Pinatubo, *Geophys. Res. Lett.*, *20*, 29–32, 1993.

Stamnes, K., S. Tsay, W. Wiscombe, and K. Jayaweera, Numerically stable algorithm for discrete-ordinate-method radiative transfer in multiple scattering and emitting layered media, *Appl. Opt.*, *27*, 2502–2509, 1988.

Stower, L. L., R. M. Carey, and P. P. Pellegrino, Monitoring the Mt. Pinatubo aerosol layer with NOAA 11 AVHRR data, *Geophys. Res. Lett.*, *19*, 159–162, 1992.

Tie, X., G. P. Brasseur, B. Briegleb, and C. Granier, Two-dimensional simulation of Pinatubo aerosol and its effect on stratospheric ozone, *J. Geophys. Res.*, *99*, 20,545–20,562, 1994.

Valero, F. P., and P. Pilewskie, Latitudinal survey of spectral optical depths of the Pinatubo volcanic cloud-derived particle sizes, columnar mass loadings, and effects on planetary albedo, *Geophys. Res. Lett.*, *19*, 163–166, 1992.

Wen, G., and J. E. Frederick, Ozone within the El Chichon aerosol cloud inferred from solar backscatter ultraviolet continuous-scan measurements, *J. Geophys. Res.*, *99*, 1263–1271, 1994.

Wen, G., and J. E. Frederick, The effects of horizontally extended clouds on backscattered ultraviolet sunlight, *J. Geophys. Res.*, *100*, 16,387–16,393, 1995.

Winker, D. M., and M. T. Osborn, Airborne lidar observations of the Pinatubo volcanic plume, *Geophys. Res. Lett.*, *19*, 167–170, 1992a.

Winker, D. M., and M. T. Osborn, Preliminary analysis of observations of the Pinatubo volcanic plume with a polarization-sensitive lidar, *Geophys. Res. Lett.*, *19*, 171–174, 1992b.

World Meteorological Organization, *Atmospheric Ozone 1985*, *Rep. 16*, p. 908, Global Ozone Res. and Monit. Proj., Geneva, Switzerland, 1985.

J. E. Frederick and G. Wen, Dept. of the Geophysical Sciences, University of Chicago, 5734 South Ellis Ave., Chicago, IL 60637. (e-mail: frederic@rainbow.uchicago.edu; wen@rainbow.uchicago.edu)

E. Hilsenrath, Laboratory for Atmospheres, Code 916, NASA Goddard Space Flight Center, Greenbelt, MD 20771. (e-mail: hilsen@ssbuv.gsfc.nasa.gov)

(Received November 15, 1994; revised June 3, 1995;
accepted June 7, 1995.)