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PROPERTIES AND DECAY OF STRATOSPHERIC AEROSOLS IN THE ARCTIC FOLLOWING THE 1991 ERUPTIONS OF MOUNT PINATUBO

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Abstract. Sunphotometer observations made from an aircraft several months after the June 1991 eruptions of Mount Pinatubo are used to quantify the spectral opacity of the Arctic stratosphere. Ancillary surface-based measurements are presented in support of the aircraft data that show large increases in stratospheric optical depth attributed to the presence of volcanic aerosols. Visible optical depths greater than 0.2 were observed during flight segments flown above the tropopause. An inversion algorithm and the optical depth data are used to infer effective aerosol size distributions. The distributions tend to be bimodal, having a large-particle mode radius of about 0.50 μm and a small-particle mode of higher concentration with radii less than 0.18 μm. Surface measurements made during spring 1992 and 1993 are also used to estimate a time constant (e-folding time) of about 13.5 months assuming that the Arctic stratosphere's opacity decays exponentially; this estimate is larger than decay times observed following other major volcanic eruptions. Our results suggest that any climate perturbations in the Arctic caused by the eruptions of Pinatubo may be significant and will very likely persist longer than any volcanically-induced changes observed there during the past century.

1. Introduction

The June 1991 eruptions of Mount Pinatubo ejected massive amounts of debris and sulfur dioxide gas into the stratosphere. It is estimated that some 20 million tons of SO_2 [*Bluth et al.*, 1992] was dispersed widely by upper-level winds. Within 2 months fragments of the volcanic plume consisting of sulfuric acid particles were observed as far north as 40° latitude [*Deshler et al.*, 1992] and by the following spring anomalously large aerosol optical depths were observed at high northern latitudes by polar-orbiting satellites [*C. Long and L. Stowe*, Using the NOAA AVHRR to study stratospheric aerosol optical thickness following the Mount Pinatubo eruption, submitted to *Geophys. Res. Lett.*, 1993].

Increased concentrations of aerosols in the stratosphere can perturb the radiative balance of the entire earth/atmosphere system [e.g., *Minnis et al.*, 1993; *Hansen et al.*, 1992; *Dutton and Christy*, 1992]. This "volcanic forcing" is characterized by tropospheric cooling caused by an increase in the planetary albedo and by stratospheric warming primarily due to infrared absorption by aerosol particles. After reaching a peak, the stratosphere's opacity normally decays exponentially in time at a rate dependent on the magnitude, time of year and location of the eruption [e.g., *Gerber and Deepak*, 1984; *Hofmann and Rosen*, 1987]. Hofmann and Rosen [1987], for example, attributed the prolonged lifetime of large sulfuric acid particles resulting from the 1982 emissions of El Chichon to vaporization of the particles at unusually high altitudes caused by stratospheric heating, followed by growth of new aerosols as the vapor diffused to lower levels. Although the net radiative effect of atmospheric aerosols depends on many factors, the optical thickness and effective size distribution of the aerosols are most important [*Lacis et al.*, 1992]. Aerosol optical thickness is a nondimensional parameter used to quantify the spectral extinction of direct solar irradiance by aerosols integrated along a path between an observer and the sun. If measurements of extinction can be obtained over a suitable range of wavelengths, then an effective size distribution of an aerosol layer can be inferred [*King et al.*, 1978].

During the spring of 1992 an extensive series of *in situ* measurements were made using airborne techniques as part of the Fourth Arctic Gas and Aerosol Sampling Program (AGASP-IV) in conjunction with the Arctic Leads Experiment (LEADEX). Nearly 1300 spectral measurements of solar irradiance were made from near the surface into the stratosphere using handheld sunphotometers during seven flights of the NOAA WP-3D aircraft. We focus here on an analysis of the stratospheric data to quantify the spectral opacity and infer effective size distributions for the Pinatubo aerosols present in the Arctic. Ancillary surface measurements are presented in support of the aircraft data analysis and are further used to estimate a decay rate of stratospheric optical depth following the period of peak aerosol concentration.

2. Measurements

The data are derived from two types of radiometric observations: (1) airborne and surface-based measurements made using sunphotometers, and (2) direct beam, wideband solar irradiance measurements made at a ground station. The primary data are derived from spectral measurements made during flight segments flown above the tropopause. Additional sunphotometer measurements were made at the surface near Resolute, N.W.T., as part of the 1992 and 1993 Seasonal Sea Ice Monitoring and Modeling Site (SIMMS) field programs [*Reddan et al.*, 1992], and in the vicinity of Anchorage, Alaska. The sunphotometer observations were made using two handheld, dual-channel instruments that sense directly transmitted solar irradiance at 380 and 500 nm, and at 778 and 862 nm, respectively; each channel having a nominal half-bandwidth of 5 nm and a field of view of 2.4°. The wideband (350-695 nm) pyrheliometer data were collected at the NOAA Climate Monitoring and Diagnostics Laboratory's Barrow Observatory (CMDL/BRW). The "wideband method" used to estimate optical depth was described by *Dutton and Christy* [1992]. Only data collected during cloud-free periods are analyzed. The locations and dates corresponding to the various measurement periods are shown in Figure 1. The curved vectors are back-trajectories representing stratospheric winds prior to each flight.

Airborne observations were made through optical glass windows whenever viewing was possible within ±30° of the solar azimuth relative to a plane perpendicular to the aircraft heading [e.g., *Dutton et al.*, 1989]. The sunphotometers were carefully calibrated at high-altitude sites before and after

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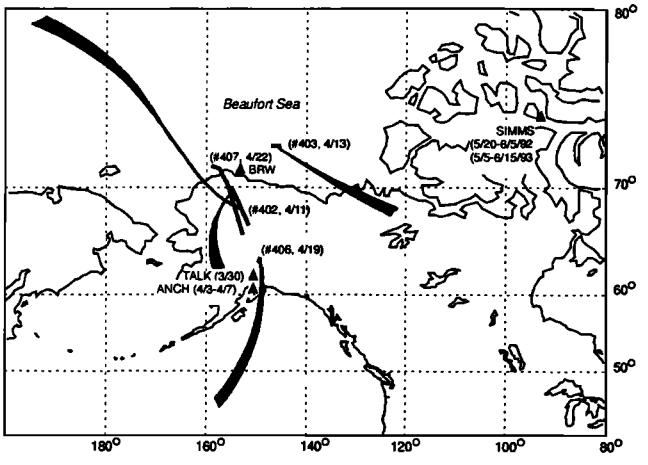

Fig. 1. Distribution of sunphotometer measurements made during AGASP-IV/LEADEX stratospheric flight segments and at surface locations (ANCH, TALK, and SIMMS). Wideband pyrheliometric measurements were made at the Barrow Observatory (BRW). Curved vectors represent stratospheric winds for 36 hours prior to each flight (back-trajectories).

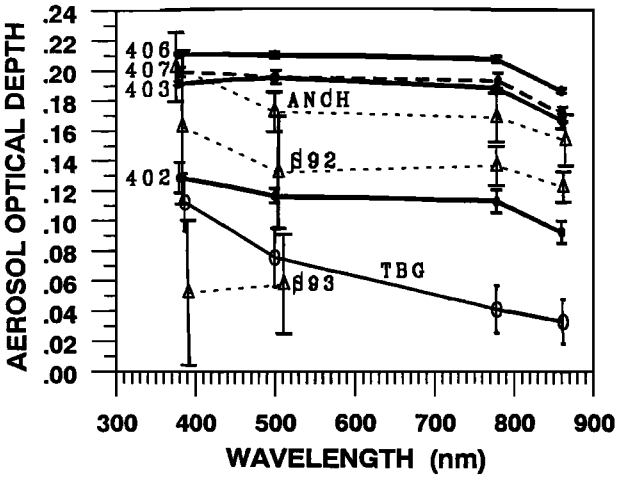

Fig. 2. Mean stratospheric spectral aerosol optical depths derived for the flight segments and surface locations shown in Figure 1. Slight spectral offsets are used to clearly indicate values at plus and minus one standard deviation (vertical bars). S92 and S93 are averages of the SIMM'92 and SIMMS'93 data, respectively (only 380 and 500 nm measurements were made in 1993); ANCH is the average of the Anchorage (ANCH) and Talkeetna (TALK) data. The TBG curve (adapted from Dutton et al. [1984]) gives the values and estimated ranges of the tropospheric background corrections used to derive stratospheric optical depths from the surface-based measurements.

AGASP-IV and again prior to SIMMS'93 in accordance with recommended procedures using the Langley plot method [*Shaw*, 1983]. Each instrument was determined to be stable with precision of about 0.002 optical depth units (ODUs). All optical depth values reported in this paper account for Rayleigh scattering, ozone absorption at 500 nm, and changes in relative airmass as a function of time and location. Attenuation by the optical glass was also accounted for when reducing the aircraft data. Thus, from total slant-path irradiance measurements, columnar aerosol optical depths were computed. Estimated uncertainties due to all sources of error are no more than 0.02 ODUs [*Reddy et al.*, 1990], or less than 10% of typical values reported here. The wideband measurements are accurate to within ±0.04 ODUs [*Dutton and Christy*, 1992].

3. Results

3.1. Stratospheric Aerosol Optical Depths

Only direct measurements made above the tropopause and the clear-sky, ground-based measurements were selected for analysis. Tropopause heights for each flight were determined on the basis of the analyses of *Herbert et al.* [1993]; these range from about 7 km to 10 km, depending on geographical location and synoptic conditions. Stratospheric components were estimated from the surface measurements of total-column optical depth by subtracting predetermined "background" values for the troposphere, a method also used by *Asano et al.* [1993]. For our purposes the March 1979-1982 monthly mean background values of spectral aerosol optical depth estimated for BRW [*Dutton et al.*, 1984] were systematically subtracted from the respective measured values. Note, however, that March background values are typically smaller than those for April and larger than those for May or June based on recent wideband analyses [Dutton, unpublished data]. Figure 2 shows the mean stratospheric aerosol optical depths, plus and minus one standard deviation (±1σ), derived from observations made at the corresponding times and locations shown in Figure 1. The tropospheric background values used to correct the surface measurements are also shown with ranges of uncertainty indicated.

Several features of Figure 2 are notable. First and most important, the values derived for all times and locations are 1 to 2 orders of magnitude greater than stratospheric background levels [*Toon and Pollack*, 1976]; these values also exceed similar measurements made in the Arctic approximately a year after El Chichon erupted [*Spinhirne and King*, 1985; *Dutton et al.*, 1984]. Second, optical depths derived from the aircraft observations tend to be rather flat spectrally in the visible range compared with the 1992 surface results. Third, three of the flights, 403, 406 and 407, indicate a high degree of homogeneity in time and space, as evidenced by their small standard deviations, similar magnitudes, and spectral dependencies. Fourth, the values for flight 402 are about 60% lower than those measured during flight 407 despite their having similar flight tracks and altitudes relative to the tropopause. Fifth, the curve for SIMMS'92 also falls below all of the AGASP curves except for that of flight 402, suggesting that either temporal and/or spatial variations occurred over the period and geographical region represented by these data, or that the SIMMS data are negatively biased due to incorrect assumptions made regarding the tropospheric background corrections. And last, the values for SIMMS'93 are significantly lower and have a different spectral signature than those for SIMMS'92, indicating a decay in opacity from one year to the next as well as a change in microphysical characteristics. These similarities and differences are discussed in Section 4.

3.2. Inferred Aerosol Size Distributions

Based on the data presented in Figure 2, effective aerosol size distributions were inferred using the constrained linear inversion algorithm of *King et al.* [1978]. The radius sensitivity ($r_{min} \leq r \leq r_{max}$) [*Spinhirne and King*, 1985] determined for our particular set of measurements was within the range r_{min}=0.10 ±0.02 μm and r_{max}=1.10 ±0.10 μm. We assumed an index of refraction of 1.45-0.0*i* based on earlier

in situ observations of the Pinatubo aerosol layers [*Deshler et al.*, 1992]. The inversion results are presented in Figure 3. Each curve shows the total number concentration dN (cm^{-2}) for seven radius increments [$dlog(r)$]. The vertical bars indicate the range of number density determined by inverting the mean spectral optical depth data $\pm 1\sigma$. We find that, for the period and geographical region of interest, the inferred size spectra tend to fall into two groups. Both are bimodal, having a large-particle mode centered at about 0.50 μm radius and a small-particle mode of higher concentration peaking below about 0.18 μm. These results suggest that the volcanic aerosols present in the Arctic about 10 months after Pinatubo erupted were slightly larger than the newly formed particles observed over Laramie, Wyoming several weeks after the eruption [*Deshler et al.*, 1992] but were smaller than those estimated by *Asano et al.* [1993] in Japan approximately 2 months before the Arctic observations were made. Their independent determinations of size spectra also show the bimodal feature noted here attributed to the superposition of a monodisperse large-particle (volcanic) mode onto a small-particle (background) mode. Similar bimodal size spectra were inferred using photometric measurements made at high northern latitudes about a year after El Chichon erupted [*Spinhirne and King*, 1985].

3.3. Time Decay of Stratospheric Opacity

To evaluate the decay of the Pinatubo aerosol layer(s), we analyzed the BRW optical depth data and the spectrally-weighted average of the 380 and 500 nm SIMMS observations for the successive 1992 and 1993 spring periods. At BRW the optical depth reached an average peak value of about 0.19 during early May 1992 and declined thereafter (updated from *Dutton and Christy* [1992]). Assuming that optical depth τ decays exponentially in time t after reaching its peak value τ_{peak}, i.e., $\tau(t) = \tau_{peak}\exp(-t/T)$, a fit to the BRW data yields a time constant T of 13.3 months. A corresponding fit to the SIMMS data yields a time constant of 13.7 months, assuming that the peak opacity also occurred during May 1992 and that the BRW seasonal background tropospheric corrections were valid for the SIMMS site. We give credence to these estimates because independent measurements from geographically distinct sites were used to obtain very similar results noting, however, that the actual overall decay of volcanic aerosols may not be well represented by a smooth exponential function [*Hofmann and Deshler*, 1987]. At least another year of monitoring is needed before our results can be corroborated. Our preliminary analysis suggests that the Pinatubo aerosols are being systematically removed from the Arctic stratosphere, but at a slower rate than estimated following earlier volcanic eruptions. On the basis of *in situ* midlatitude measurements of aerosol size distributions, *Hofmann and Deshler* [1987] estimated the decay rate (e-folding time) of total stratospheric mass after the El Chichon eruption to be about 10.3 months and between 8 and 10 months following the 1974 eruption of Fuego in Guatemala. High-latitude satellite observations of 1.0 μm stratospheric aerosol optical depth [*McCormick and Trepte*, 1987] also exhibited faster decay rates following the eruptions of Mt. St. Helens in 1980 and El Chichon in 1982 than we report here.

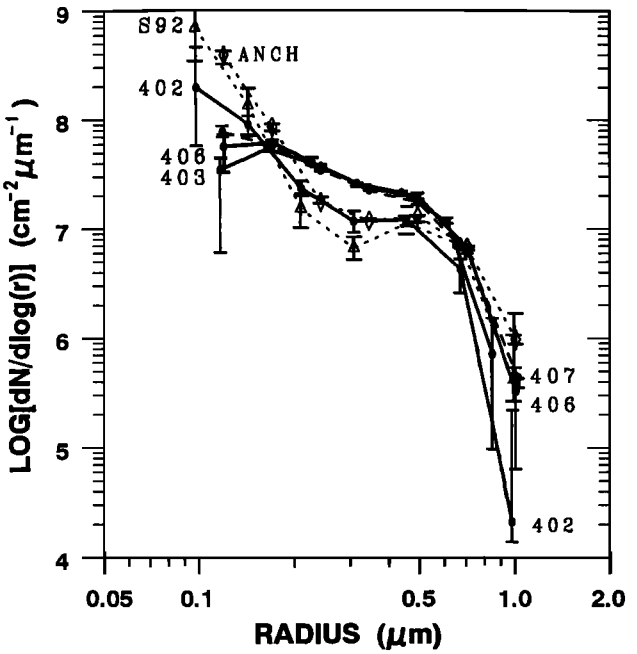

Fig. 3. Effective aerosol size distributions showing the range of number concentrations dN at each radius interval [$dlog(r)$] inferred from the optical depth data shown in Figure 2.

4. Discussion

It appears from Figure 2 that temporal and/or spatial variations in stratospheric aerosols occurred in the Arctic during spring 1992. The lower relative opacity noted for flight 402 can be explained by differences in synoptic conditions during the respective flight periods [*Herbert et al.*, 1993]. During flight 402 strong northerly winds transported polar air into the region whereas for the later three flights moderately weak southerly flow was generally observed. An analysis of isentropic back-trajectories [e.g., *Harris and Bodhaine*, 1983] based on the ECMWF 2.5° gridded data supports the synoptic analyses. Figure 1 shows 36 hour back-trajectories representing the flow at altitudes ranging from about 14.5 to 15.8 km referenced to the respective midflight segments. This analysis suggests that relatively clean Arctic air displaced or mixed with lower latitude air, effectively reducing the stratosphere's opacity prior to flight 402. The formation of the polar vortex during the previous autumn/winter probably prevented high concentrations of Pinatubo aerosols from penetrating the central Arctic. Similar gradients related to the position, size, and shape of the polar vortex were observed several months after El Chichon erupted [*McCormick et al.*, 1983].

The relatively large 380 nm optical depths measured during flight 402 and at all the surface locations during spring 1992 (Figure 2) indicate fundamental differences in the aerosols' microphysical properties compared with the results of other flights or the SIMMS'93 data which show less variation over the visible wavelengths. The apparent increase in attenuation at 380 nm is most likely due to the presence of higher concentrations of small particles (Figure 3) that have greater extinction efficiencies as the ratio of size-to-wavelength (r/λ) increases [*van de Hulst*, 1981]. Because such enhancements are most pronounced in the results derived from highly variable surface measurements we suspect that tropospheric haze, not visible against the "milky" appearing stratosphere, may have contaminated these particular results. The sharp decreases evident at the large-particle end of the inferred size spectra are attributed to the observed decreases in opacity for $\lambda > 778$ nm.

Finally, we speculate that the apparent longevity of the relatively large Pinatubo aerosols in the Arctic may be due to the vaporization and regrowth processes discussed by *Hofmann and Rosen* [1987], possibly augmented by general circulation patterns that favor the heating of the Arctic

stratosphere during winter [*Robock and Mao*, 1992] and the accumulation of aerosols at high latitudes.

5. Summary and Conclusions

A representative set of sunphotometer data collected during spring 1992 and 1993 was used to quantify the opacity of the Alaskan/Canadian Arctic stratosphere and to characterize the microphysical properties of volcanic aerosols transported to that region following the eruptions of Mount Pinatubo. The observed visible optical depths (≥0.2) exceeded any previous high-latitude measurements made in the aftermath of earlier volcanic eruptions. Airborne and surface measurements made prior to the AGASP-IV flights and later in the Canadian Arctic suggest that spatial and/or temporal variations occurred in the aerosol layer(s), probably in response to dynamical forcings associated with the position and extent of the polar vortex. The microphysical properties of the Pinatubo aerosols can be described in terms of an effective size distribution that is bimodal, having a large-particle (volcanic) mode at 0.50 μm and a more concentrated small-particle (background) mode peaking below 0.18 μm. On the basis of our limited data, the decay rate (e-folding time) of optical depth is estimated to be greater than 13 months, which suggests a slower decay than has been observed for earlier post-volcanic periods. Because of the increased opacity of the stratosphere and the apparent longevity of the volcanic aerosols present in the Arctic, direct radiative forcing in that region and indirect climate perturbations (via feedbacks) on a much broader geographical scale may result. Careful monitoring and analysis of surface and satellite data in conjunction with meteorological studies are essential before the results presented in this paper can be confirmed or a complete assessment of the extent of Mount Pinatubo's impact on Arctic climate can be made.

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