BALLOONBORNE MEASUREMENTS OF PINATUBO AEROSOL DURING 1991 AND 1992 AT 41° N: VERTICAL PROFILES, SIZE DISTRIBUTION, AND VOLATILITY

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Abstract. Vertical profiles of aerosol have been measured approximately biweekly since June 1991 at Laramie, Wyoming (41° N). Both the total number concentration and concentration of particles > 0.15 to 10.0 μm were measured using balloonborne instruments. The aerosol size distributions were best represented when bimodal lognormal distributions were fit to the data. After an early short-lived intense aerosol layer, the stratospheric maximum surface area and mass (40 μm^2 cm^{-3}, 160 ppbm) was observed to occur approximately 180 days after the eruption. The aerosol was then observed to remain relatively homogeneous both in altitude and time during 1992, with the maximum surface area and mass remaining relatively constant between 20 to 30 μm^2 cm^{-3} and 30 to 60 ppbm.

Introduction

The eruption of Pinatubo in the Philippines in June 1991 caused a major increase in the aerosol load in the stratosphere. Early estimates of the amount of mass injected by this eruption have ranged from 20 - 30 megatons of SO_2 (McCormick and Veiga, 1992), thus making it one of the major contributors of stratospheric aerosol in the past century. Aerosol from the Pinatubo eruption was first observed at Laramie in the middle of July 1991, appearing in narrow layers primarily between 15 and 18 km, with occasional layers between 21 and 23 km (Deshler et al., 1992). The upper layers contained a large amount of material, and one on 30 July exhibited the greatest amount of mass (480 ppbm H_2SO_4) and surface area (84 μm^2 cm^{-3}) observed at Laramie. Also during this period there was evidence for homogeneous nucleation (or nucleation on ions) of the H_2SO_4 vapor. The makeup of the aerosol was found to be almost exclusively a sulfuric acid and water mixture based on *in situ* volatility measurements.

Balloonborne measurements of stratospheric aerosol have been conducted at Laramie since 1971 (Hofmann, 1990). During this period the only eruption to rival Pinatubo in its effect on the stratosphere was the eruption of El Chichón in Mexico in April 1982. Aerosol measurements following the El Chichón eruption indicated that the aerosol formed initially through homogeneous nucleation. Aircraft observations in April and May 1982 indicated condensation nuclei (CN) concentrations had increased from a background of 30 to 3000 cm^{-3} (Wilson et al., 1983). New particle formation plus condensation on pre-existing aerosol formed a bimodal size distribution (Hofmann and Rosen, 1983; Oberbeck et al., 1983), although initially the second mode of the distribution, near 1.2 μm, was likely dominated by ash (Knollenberg and Huffman, 1983). The following winter a new CN layer was observed at 30 km which

suggested an evaporation recondensation mechanism to redistribute the aerosol to a small particle phase (Hofmann et al., 1985). The aerosol surface area was observed to peak at approximately 18 μm^2 cm^{-3} at an altitude of 20 km (Hofmann, 1987) and the sulfuric acid mass mixing ratio to peak at 60 ppbm about 300 days after the eruption (Rosen and Hofmann, 1986).

The balloonborne aerosol instruments used at Laramie measure the number concentration of CN (r > 0.01 μm) and optical aerosol (r > 0.15 μm), and follow an initial design by Rosen (1964). The instrument was modified for measurements following the eruption of El Chichón (Hofmann and Rosen, 1983), and has since been modified for polar stratospheric cloud measurements (Hofmann and Deshler, 1991). In their present form the instruments measure the number concentration of aerosol with radius > 0.15 to 10.0 μm, or with radius > 0.15 to 2.0 μm, in 8 size classes. The instruments with finer size resolution have been used since April 1992. At the sensitivity of these measurements no particles > 1.5 μm were observed. The minimum detectable concentrations are approximately 0.0006 cm^{-3} for optical aerosol and 0.007 cm^{-3} for CN.

Observations and Discussion

From August 1991 until May 1992 aerosol soundings at Laramie were conducted biweekly. In June 1992 the sounding frequency was reduced to once per month and has been maintained at this level through the present time. In addition during this period there have been occasional soundings from McMurdo Station, Antarctica (78° S), in August and September of 1991 and 1992 (Deshler et al., 1992), Kiruna, Sweden (68° N), in January and February 1992 (Deshler, 1993), and in Lauder, New Zealand (45° S). Although summaries of some of these measurements are included here for comparison, the purpose of this letter is to present the results from Laramie. Figure 1 presents vertical aerosol profiles on four days compared to the pre-Pinatubo background aerosol as measured in May 1991. By November 1991, the concentration of CN had returned to background after the homogeneous nucleation observed in August (Deshler et al., 1992). The CN concentrations have remained relatively constant after this time; however, at 20 km over 50% of the CN are comprised of particles > 0.15 μm. On 18 December 1992 CN increased in concentration by a factor of 10 between 31 and 35 km. A similar increase was observed also on 16 January 1993, but not during the previous winter. The vertical profile of these CN observations is similar to those observed by Hofmann et al. (1985) the first winter following the eruption of El Chichón, although the increase of CN observed here is about a factor of 10 smaller than observed after El Chichón.

For the first two months following the Pinatubo eruption the vertical aerosol distribution was very structured and inhomogeneous (Deshler et al., 1992). After that time, the measurements at Laramie can be grouped into three general periods. The first, extending
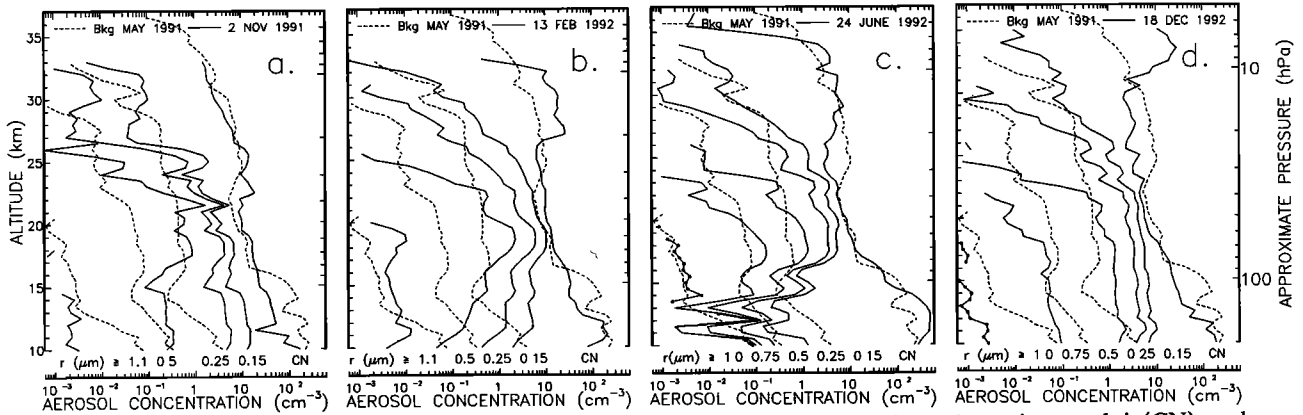
Figure 1. Vertical profiles of 0.5 kilometer averages of the concentration of condensation nuclei (CN) and particles with radius > 0.15, 0.25, 0.5, and 1.1 μm for measurements at Laramie in November 1991 and February 1992, and for particles with radius > 0.15, 0.25, 0.5, 0.75, and 1.0 μm in June and December 1992. Background measurements at Laramie in May 1991 (dashed line) of aerosol with radius > 0.15, 0.25, and 0.5 μm are also shown. In all cases no particles > 1.5 μm were observed. Since the aerosol counter is a cumulative particle counter, at any one altitude each measurement of increasing concentration corresponds to the next smaller size particle measured. The tropopause heights are: a) 9 km, b) 11 km, c) 14 km, and d) 8 km.

from September through November 1991, was characterized by aerosol filling the stratosphere between the tropopause and 27 km, but maintaining a highly layered structure, indicating that vertical mixing was not complete, Figure 1a. The layered vertical structure was replaced by a fairly homogeneous and static aerosol layer in early 1992, Figure 1b. The third period, beginning in the summer of 1992, is characterized by slowly decreasing concentrations of aerosol both at 0.15 and 1.0 μm, Figure 1c, 1d. The measurements shown in Figure 1c, 1d, were made using aerosol counters capable of the finer resolution.

In May 1992 two flights were preformed using an optical aerosol counter (r > 0.15 μm) with its intake heated to 150° C, similar to flights made soon after the eruption (Deshler et al., 1992). The results were also similar. The concentration of particles > 0.15 μm were reduced by over a factor of 50. Assuming the heated particles, which are too small to detect, are reduced to a radius of 0.1 μm, indicates that the 150° C heating causes a 70 to 99% reduction in mass for over 95% of the particles > 0.15 μm. This indicates primarily a sulfuric acid water aerosol.

To characterize the size distributions measured by these profiles we have fit the data with lognormal size distributions, usually bimodal, which may be written in differential form as,

$$n_i(r)dr = \Sigma\ N_i\ (2\pi)^{-1/2} \exp(-\alpha_i^2/2)\ d\alpha_i, \text{ where}$$

$$\alpha_i = \ln(r/r_i)\ /\ \ln\sigma_i.$$

Each lognormal distribution is then specified by three parameters, the total number concentration, N_i, the median radius, r_i, and the distribution width, σ_i. The integral size distribution, the parameter measured by the particle counters, is then given by,

$$N(>r) = \int_r^\infty (n_1(r) + n_2(r))\ dr.$$

Example size distributions are shown in Figure 2 from two of the profiles in Figure 1. In most cases bimodal distributions were necessary to fit the data below 22 km. The second mode probably arises from condensation and growth of the sulfuric acid vapor on the larger particles of

the background aerosol while the smaller mode represents the growth of the CN. Note that N_2, the total number in the second mode, roughly coincides with the concentration of background 0.15 μm particles between 15 and 25 km.
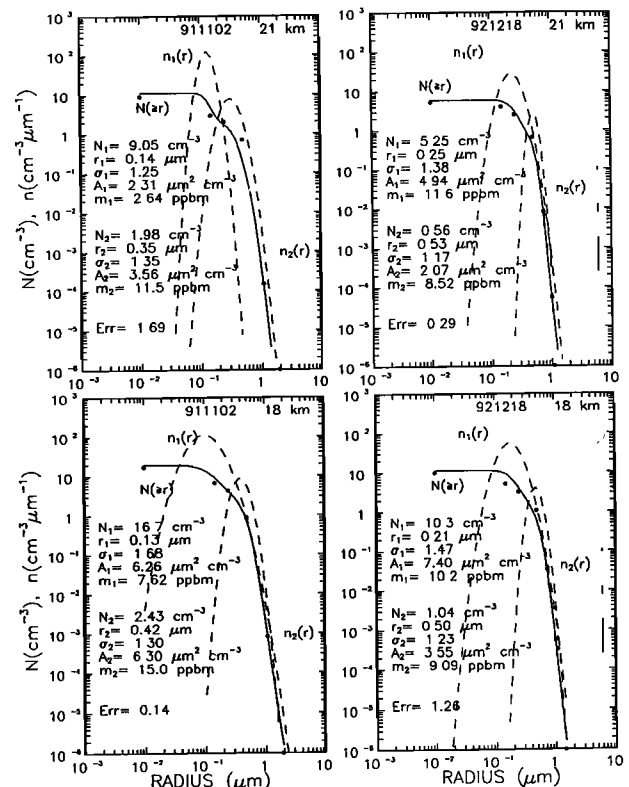

Figure 2. Bimodal lognormal size distributions fit to one kilometer averaged data (solid circles) from aerosol measurements on two days. The altitude, parameters of the size distributions, and mass and surface area calculations, are included on the plots. The error (Err) shown is the root mean square distance between a measured concentration and the calculated concentration at that point. In each case the data point at 10^{-6} cm^{-3} represents the size where no counts were observed indicating a concentration < 0.0006 cm^{-3} at that size. Uncertainties in the measurements are indicated by the vertical bars on the right hand side.

The uncertainty in the aerosol concentration measured increases as the number of particles measured decreases, see Figure 2. The impact of this uncertainty on the lognormal parameters, and the derived surface area and mass, was estimated by varying the aerosol concentrations within their uncertainty limits and refitting the size distributions. The result was an average variation in surface area and mass of 10 to 20% with a maximum variation of 60%. For the lognormal parameters the variations were less than 10% on average with a maximum near 30%.

A history of the size distribution parameters used to characterize the measurements is shown in Figure 3. Clear in this figure is the fluctuating nature of the aerosol before the end of 1991; however, beginning early in 1992

H_2SO_4 MASS MIXING RATIO (ppbm)

SURFACE AREA $(\mu m^2\ cm^{-3})$

DAYS SINCE ERUPTION

Figure 4. Time altitude contour plot of aerosol surface area $(\mu m^2\ cm^{-3})$, and mass (ppbm, assuming sulfuric acid) from the lognormal size distributions fit to the aerosol data.
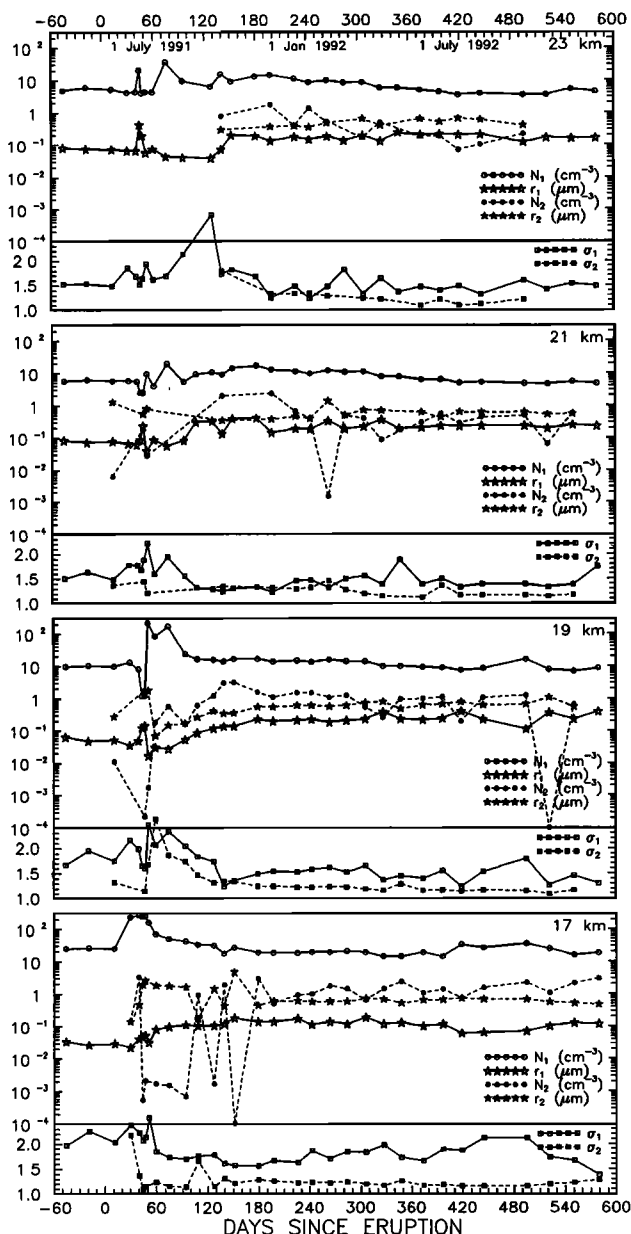

DAYS SINCE ERUPTION

Figure 3. History of size distribution parameters for unimodal or bimodal lognormal size distributions fit to the data at 4 altitudes, 17 to 23 km. When only parameters for a single mode are shown a unimodal distribution was best at that point.
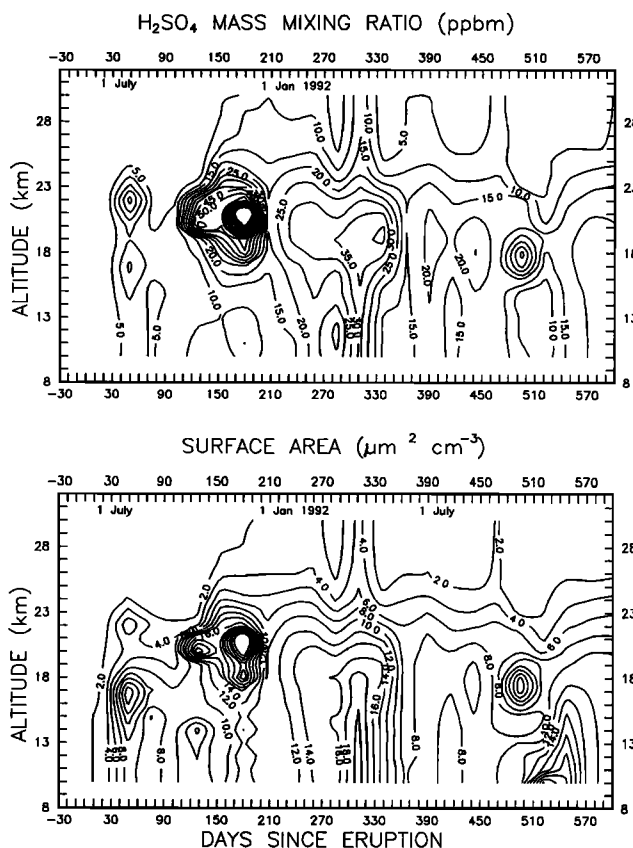
these fluctuations disappear and the size distributions do not show much variation after that point. Although there is a slight tendency for N_1 to decrease and r_1 and N_2 to increase (particularly below 20 km), parameters of the size distributions are relatively invariant after day 200.

From the size distribution parameters aerosol surface area and mass can be calculated. Contour plots displaying a time altitude history of both aerosol surface area and mass (assuming the aerosol is a 75% sulfuric acid water mixture) are shown in Figure 4. From initial values near 1 $\mu m^2\ cm^{-3}$ and 1 ppbm the aerosol surface area and mass quickly increase by a factor of 10 to 20 throughout the stratosphere below 25 km. The maximum in both quantities was observed about 150 days after the eruption between 21 and 25 km. Before this the temporal and altitude inhomogeneity of the aerosol plume is apparent. After this point both quantities become relatively more uniform with the surface area remaining between 10 and 20 $\mu m^2\ cm^{-3}$ between the tropopause and 25 km. There is more structure towards the end of this record and the peaks in the surface area have moved to lower altitudes. Aerosol mass shows a similar time altitude history; however, the mass is more concentrated between 18 and 22 km and does not show the same progression to lower altitudes.

Finally a history of the stratospheric maximum surface area and mass is shown in Figure 5. Also included in this figure for comparison are four measurements at Kiruna, Sweden, (Deshler, 1993) and three at Lauder, New Zealand. The maximum surface area and mass observed at these other locations are similar to the measurements
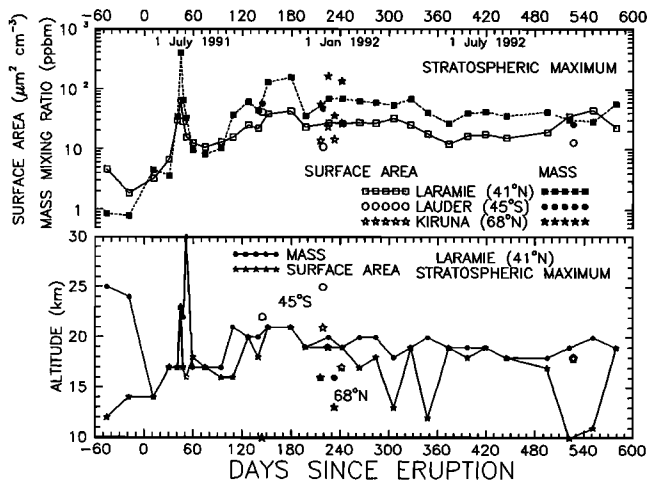
Figure 5. History and altitude of the maximum aerosol surface area and mass (assuming sulfuric acid). The results from Laramie are compared with soundings at Kiruna, Sweden, and Lauder, New Zealand.

at Laramie, particularly the measurements at 45° S. The lower altitude of the stratospheric maxima observed at Kiruna is indicative of the winter subsidence observed in the Arctic stratosphere. Although the maximum in aerosol surface area and mass was observed on 30 July at 23 km (Deshler et al., 1992), the layer was short lived and therefore weighted heavily by surrounding observations in the smoothing algorithm used to create the time altitude surfaces shown in Figure 4. In general surface area and mass peaked between 16 and 21 km and reached a relatively steady state after day 200 at 20 to 30 μm^2 cm^{-3} and 30 to 60 ppbm. These values of the mass mixing ratio are about 50% higher than a similar history for El Chichón, and the maximum in mass mixing ratio occurred sooner after the eruption in the case of Pinatubo.

Summary and Conclusions

Twenty-nine vertical aerosol profiles have been measured at Laramie, Wyoming (41° N), from June 1991 to January 1993 at approximately biweekly intervals. The aerosol size distributions were generally best characterized using bimodal lognormal distributions at altitudes below 22 km, while unimodal size distributions were adequate above 22 km. Vertical and temporal inhomogeneity in the aerosol was observed to disappear by early in 1992. The stratospheric maximum surface area and mass (40 μm^2 cm^{-3}, 160 ppbm) was reached shortly before this, 180 days after the eruption, although one aerosol layer, observed 45 days after the eruption, had a surface area and mass exceeding these maxima by a factor of two. Although the Pinatubo aerosol is decaying, and the size distributions are changing due to coagulation and gravitational settlement, the effects of these processes on changing the stratospheric maximum aerosol surface area and mass has been minimal up to January 1993.

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