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BALLOONBORNE MEASUREMENTS OF THE PINATUBO AEROSOL SIZE DISTRIBUTION AND VOLATILITY AT LARAMIE, WYOMING DURING THE SUMMER OF 1991

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Abstract. Measurements using balloonborne optical particle counters at Laramie, Wyoming during the summer of 1991 are used to study the particle size distribution and volatility of the aerosol which formed in the stratosphere following the mid-June eruptions of Mt. Pinatubo. Enhanced aerosol layers were observed below 20 km as early as 16 July, about 1 month after the eruption. During late July, a transient though substantial particle layer was observed in the 23 km region. High concentrations of large particles in this high altitude layer resulted in aerosol mass mixing ratios as large as 0.5 ppm, considerably larger than observed following the eruption of El Chichón. Aerosol volatility tests indicated that well over 90% of the particles were composed of an H_2SO_4/H_2O solution in all layers observed, indicating rapid conversion of SO_2 to H_2SO_4 and subsequent droplet growth. High concentrations of droplets suggest homogeneous or ion nucleation as the most likely aerosol production mechanism.

Introduction

The eruptions of Mt. Pinatubo (15.1°N, 120.4°E) in the Philippine Islands during 12-16 June 1991, caused a major perturbation to the stratospheric aerosol layer, which was in a near background condition prior to the eruptions [Hofmann, 1990]. Owing to the latitude and season of the eruption, the plume dispersal was generally similar to that following the eruption of El Chichón in 1982 [Hofmann, 1987]. The summer wind reversal near 20 km (easterlies above, westerlies below), with prompt meridional transport below and only sporadic transport above, again separated the plume into two layers having different characteristics during the early stages.

Seven balloon flights conducted at Laramie, Wyoming (41°N) between 16 July and 29 August, detected increases in stratospheric aerosol concentrations. The instrument complement, in addition to pressure, temperature and ozone, included in varying combinations, a condensation nuclei (CN) counter ($r \geq 0.01$ μm) , an 8 size channel, high-flow rate aerosol counter ($r \geq 0.15$, 0.25, 0.50, 1.0, 2.0, 3.0, 5.0, and 10.0 μm) and a 2 size channel, low-flow rate aerosol counter ($r \geq 0.15$ and 0.25 μm) for volatility tests. The 8-channel counter measures white light scattering at 40° from the forward direction whereas the older, 2-channel counter measures at 27° from forward scattering. Measuring aerosol scattering at 40° eliminates the double valued response inherent in the 27° counter, near the wavelength of visible light, which precluded size discrimination in the important 0.5 μm range. The detailed characteristics of these particle counters has been given previously [Hofmann and Deshler, 1991]. In addition to particle counters, a 3-stage impactor for electron microscope analysis of collected particles [Sheridan et al., 1991], was included on some of the flights. Table 1 summarizes the balloon flight information.

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TABLE 1. PINATUBO-RELATED BALLOON FLIGHTS

Date,	Instruments,	Max. Alt. (km)
16 July	A2, A2', A8, IM, O	38.8
26 July	CN, A2, A2', A8, IM, O	37.5
30 July	A2, A2', A8, IM, O	37.3
2 August	A2, A8, O	37.5
6 August	CN, A8, O	36.0
14 August	A2, A8, IM, O	36.6
29 August	CN, A8, O	35.5

CN: Condensation Nuclei, $r \geq 0.01$ μm
A2: Aerosol, 16.7 cm^3s^{-1}, $r \geq 0.15$, 0.25 μm
A2': A2 With Heated (150°C) Intake
A8: Aerosol, 167 cm^3s^{-1}, $r \geq 0.15$, 0.25, 0.50, 1.0, 2.0, 3.0, 5.0, 10.0 μm
IM: 3 Stage Impactor, $r \approx 0.12$, 0.5, 2 μm
O: Electrochemical Ozone

Observations

Figure 1 shows vertical concentration profiles of $r \geq 0.15$ μm particles for the 7 flights listed in Table 1 and compares them with a sounding on 17 June 1991 which was prior to any possible effect from the Pinatubo eruption. Where available, the data are from the low flow rate optical counter (A2 in Table 1) because on several occasions (26, 30 July and 2 August), when concentrations exceeded about 20 cm^{-3}, the high flow counter (A8) was saturated, owing to an electronic pulse counting limitation of the two coincident scattered light detectors. The enhanced concentrations of particles near the tropopause, observed in the June sounding, are believed to be the result of the Kuwaiti oil fires [Deshler and Hofmann, 1991]. The first indication of enhanced stratospheric aerosol levels, other than those which might be attributed to the oil fires, was on 16 July when increases were observed just above the local tropopause and above the higher altitude, low latitude tropopause. A second tropopause, indicated by the upper arrows in Figure 1, results from meridional transport and is a common feature at Laramie. These are often good indicators of the source of the air parcels observed at midlatitudes. By 26 July, aerosol below the upper level easterlies had increased to record concentrations of about 50 cm^{-3} at 17 km. This low altitude layer was observed on all subsequent soundings and was the result of prompt transport northward in this altitude regime.

The first evidence of a layer above 20 km, in the upper level easterlies, came on 30 July when a thin layer was observed at 23 km. Lidars, operated by the NOAA Wave Propagation Lab at Boulder, Colorado (40°N), clearly observed this layer from 29 July to 2 August [Post et al., 1991]. It was still present, although diminished in concentration, at about 22 km in the Laramie balloon sounding on 2 August but had disappeared by 6 August (see Figure 1). From the wind speeds in the vicinity of the layer, it is surmised that these observations were the result of a continental sized portion of the main plume which had broken off and was transported to midlatitudes earlier than expected. About 1 month later, probably after having circumnavigated the earth, it was

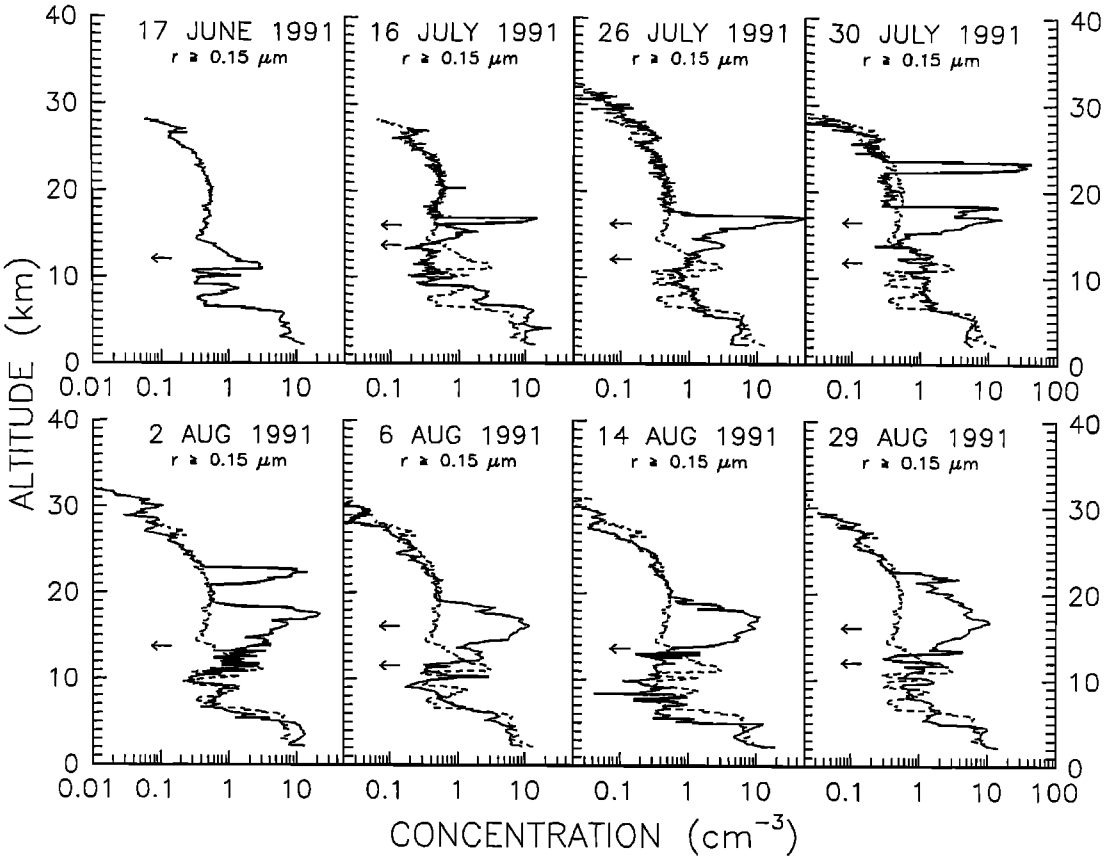

Fig. 1. Vertical profiles of $r \geq 0.15$ μm aerosol particle concentrations at Laramie, Wyoming on 17 June 1991, prior to the eruption of Pinatubo, and on 7 occasions following the eruption. The dashed curves are for the 17 June sounding. The arrows mark the observed positions of the local tropopause (lower arrows) and the coldest region, related to the low latitude tropopause (upper arrows).

again observed over Boulder by lidars, and over Laramie in the 29 August aerosol profile. The layer was at about 22 km, but aerosol concentrations had decreased. The late July - early August observation of a piece of the high-altitude plume is important because it provides measurements during the early stages of development of the main aerosol cloud, or at least a lower limit representation of it. By the end of August, a broad volcanic aerosol layer from the local tropopause to about 22 km was typical and soundings after mid-September, not discussed here, indicated excess aerosol to altitudes in the 30 km range.

Following the eruptions of Mt. St. Helens in 1980 and El Chichón in 1982, large increases in CN were observed and were interpreted as indicating direct gas to particle conversion in the plumes [Hofmann and Rosen, 1982; 1984]. Figure 2 shows several measurements of CN before and following the Pinatubo eruption. Because of large tropospheric CN concentrations, a dilution device is used below about 13 km. For the sounding on 27 June, the dilution device was not operating and the nearly constant CN concentration of about 400 cm^{-3} below about 13 km is the result of instrument saturation. This counting limit appears at a lower concentration for the high flow rate counter, discussed earlier, because of the higher sampling rate. As indicated in Figure 2, the saturation level was reached for CN in essentially all the volcanic layers in the 15-20 km region. Thus, the CN concentration in these layers was >500 cm^{-3} or about 100 times normal in the 18-20 km region. This suggests recent particle production as the coagulation lifetime at these concentrations is only a few days [Hofmann and Rosen, 1982].

To study aerosol composition, aerosol impactors and counters with sample intake heaters were flown on several occasions (see Table 1). The electron microscope analysis of the impactors are reported in a separate paper [Sheridan et al., 1991]; only the analysis of the heated intake data will be discussed here. Figure 3 compares $r \geq 0.15$ μm vertical aerosol profiles for ambient and heated air samples. The temperature used in the heated intake for the Pinatubo aerosol was 150°C, adequate to vaporize an aerosol of a 75% H_2SO_4, 25% H_2O composition, typical for midlatitude stratospheric temperatures [Hofmann and Rosen, 1983]. In the 1 May sounding prior to the Pinatubo eruption, conducted in order to determine the degree of volatility of the suspected Kuwaiti oil fire particles in the 8 to 11 km region [Deshler and Hofmann, 1991], two different instruments, one ambient (high flow counter) and one heated (low flow counter) were flown on the same balloon and both profiles in Figure 3 are for the balloon ascent. These measurements indicated that the suspected Kuwaiti particles in the 5-10 km region were about 50% non-volatile. As anticipated, the background stratospheric H_2SO_4 solution aerosol in the 15-25 km region was observed to be highly volatile at this temperature. The apparent excess concentrations of particles at 30 km on 1 May were observed in several soundings during the spring of 1991 and their source has not been identified.

The three soundings in Figure 3, after the Pinatubo eruption, used the low flow rate counter operating at ambient temperature during balloon ascent but with the intake heated during parachute descent. High altitude (>25 km) resolution in the heated mode suffers owing to the rapid initial descent; in addition, the same air

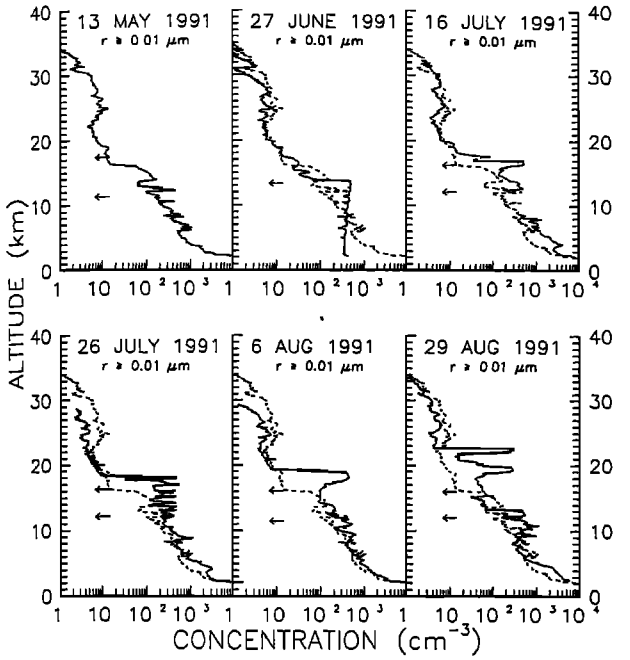

Fig. 2. Vertical profiles of condensation nuclei ($r \geq 0.01$ μm) concentrations at Laramie, Wyoming on 13 May and 27 June 1991, prior to the eruption of Pinatubo, and on 4 occasions following the eruption. The dashed curves are for the 13 May sounding. The arrows mark the observed positions of the local tropopause (lower arrows) and the coldest region, related to the low latitude tropopause (upper arrows).

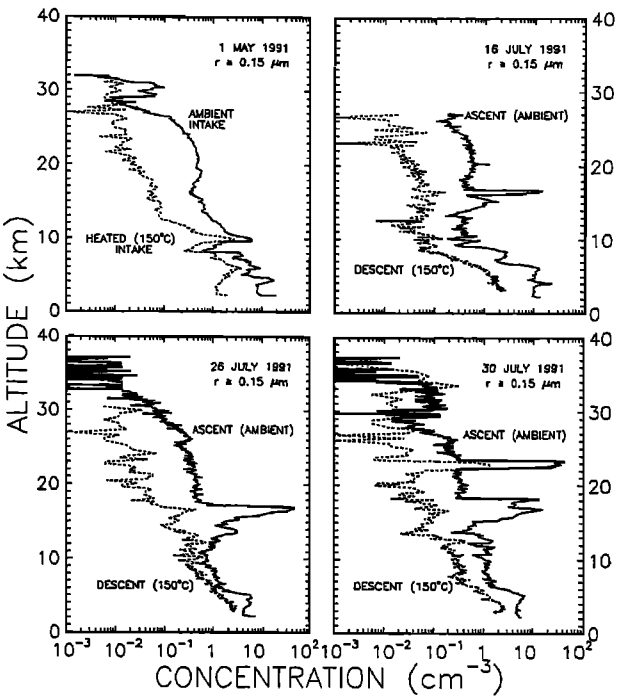

Fig. 3. Comparisons of vertical profiles of $r \geq 0.15$ μm aerosol particle concentrations at Laramie, Wyoming with and without a 150°C heater on the particle counter intake before the Pinatubo eruption (1 May) and after.

mass is not sampled during both ascent and descent. However, the high flow rate counter was operated at ambient temperature throughout each of these flights and only minimal changes in the particle profile were observed on ascent and descent. All the volcanic layers, both below and above the 20 km wind reversal, were 95-98% volatile at 150°C, indicating an H_2SO_4/H_2O composition within a month of the eruption. This verifies the expected fast chemical conversion time of SO_2 to H_2SO_4 of the order of about 38 days [McKeen et al., 1983] and the observed rapid decay of SO_2 observed with the Total Ozone Mapping Spectrometer (TOMS) instrument following the El Chichón and the Pinatubo eruptions (A. Krueger, personal communication, 1991). The H_2SO_4 vapor thus formed, results in the nucleation of new particles followed by co-condensation with water to form the observed aerosol. In the case of El Chichón, the characteristic aerosol growth time was about 45 days [Hofmann, 1987].

In addition to the amount of SO_2 injected by a volcanic eruption, the eruption latitude, injection altitude and time of year are crucial in determining the eventual stratospheric aerosol particle size [Hofmann, 1987]. Since the eruptions of Pinatubo were very similar to those of El Chichón in these respects, it is expected that the Pinatubo aerosol will display generally similar properties, i.e., large droplet sizes owing to the initial equatorial confinement of the condensing vapors. The aerosol counters used in this study measure the integral concentrations above several sizes from which differential lognormal size distributions (either single mode or bimodal) are derived. Figure 4 shows examples of these size distributions for the generally smaller aerosol below the wind reversal (16-17 km) and the larger particles above (22-23 km). The parameters of the distributions are N_i, the total concentration in the ith distribution, r_i, the median radius and σ_i, the distribution width. These parameters and the mass mixing ratio and particle surface area density derived from them are also given in Figure 4. The accuracy of these measurements in terms of size measured has been discussed by Hofmann and Deshler [1991] and is about 10% for spherical particles of known index of refraction, which is the case for the predominant volcanic sulfuric acid/water droplets. This uncertainty in size translates into an uncertainty in particle surface area density and mass density of about 20% and 30%, respectively.

Below 20 km, the aerosol can be characterized as consisting of particles with a mode radius of about 0.07 μm while above 20

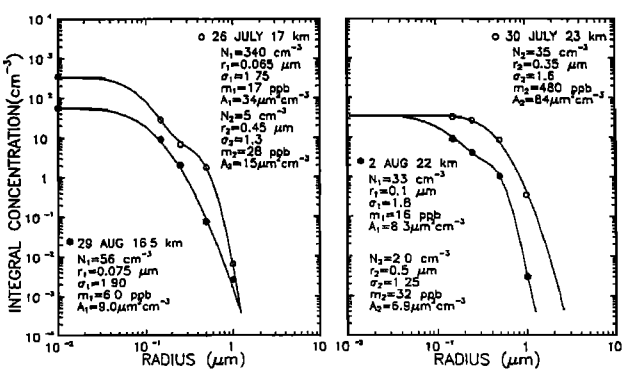

Fig. 4. Aerosol particle size distributions in two altitude regions following the Pinatubo eruption. The points are the measured integral concentrations for 0.5 km averages and the smooth curves are calculated from fitted lognormal distributions. N_i, r_i, and σ_i are parameters of the best fit single mode (i=1) or bimode (i=1 and 2) lognormal distributions (see text for definitions). A_i and m_i are the particle surface area densities and mass mixing ratios, respectively, for the ith lognormal mode.

km, in the early snapshot of the main layer, the mode radius is considerably larger, on the order of 0.35 μm for a single mode lognormal. The distributions were initially bimodal in the lower layer and also on the edges of the upper layer as observed on 2 August. The data in Figure 4 were constructed from 0.5 km averages so that the statistical uncertainties are small, reaching the size of the data points only for concentrations in the 10^{-3} cm^{-3} range. Thus, the bimodality of some of the distributions is real and not an artifact of the measurement.

Since CN were not measured on 30 July or 2 August, the possibility of an additional mode at small sizes on these days cannot be ruled out; however, numerous observations, for example in cirrus clouds, indicate that attachment of CN to large particles effectively limits their concentration when large particles are present. Thus, the presence of a considerable number of small particles in the 22 and 23 km layers on 2 August and 30 July is unlikely. If present, they would not affect the mass but could affect the surface area. The distributions were sharply limited above radii of 1 μm with very few $r \geq 2$ μm particles observed even in the dense layer at 23 km on 30 July. This observation is in agreement with the impactor results for this flight [Sheridan et al., 1991].

The aerosol mass density, mass mixing ratio and total surface area density in the 23 km layer on 30 July, of 28 $\mu g\ m^{-3}$, 0.48 ppm and 84 $\mu m^2 cm^{-3}$, respectively, considerably exceeds those observed after El Chichón [Hofmann, 1987]. However, observations in the fringes of this cloud on 2 August, indicate that particle concentrations had dropped substantially, resulting in about 10 times less mass.

The peak concentration of about 40 cm^{-3}, for $r \geq 0.15$ μm, observed at 23 km on 30 July exceeded the preeruption CN concentration at this altitude by about a factor of 10. CN concentrations exceeding 500 cm^{-3} in layers between 16 and 23 km, observed in all flights in which they were measured (see Figure 2), represent enhancements of the order of 100 over background CN concentrations in this region. These facts suggest homogeneous nucleation from the gas phase, or possibly condensation on small ions, as the particle source. The possibility of condensation on small particles ($r < 0.01$ μm), which would not be detected by the CN counter, cannot be ruled out. However, extensive work in the Arctic [Hofmann et al., 1990] and Antarctic [Hofmann and Deshler, 1991] indicates that under the high vapor supersaturation conditions that exist in the cold polar stratospheres, the concentrations of nitric acid trihydrate particles never exceed the precursor concentration of sulfuric acid CN ($r \geq 0.01$ μm), and in fact, are nearly equal to this concentration under fast cooling conditions, suggesting that small stratospheric particles ($r < 0.01$ μm), if they exist in adequate concentrations, are ineffective as condensation sites.

Summary and Conclusions

Optical particle counters, flown on balloons at Laramie following the Pinatubo eruption, detected enhanced aerosol concentrations in the 15 to 18 km region as early as 16 July, about 1 month after the eruption. A dense particle layer only a few km thick was observed at 23 km on 30 July but had disappeared by 6 August. The particles in the lower layer were as numerous as those in the upper layer, with concentrations for $r \geq 0.15$ μm reaching record levels of about 50 cm^{-3}, but were considerably smaller in size than those in the 23 km layer on 30 July where concentrations as high as 0.5 cm^{-3} were observed for $r \geq 1.0$ μm. Typical mode radii in the lower and upper layers were 0.07 and 0.35 μm, respectively. The high concentrations of large particles in the upper layer resulted in aerosol mass mixing ratios of about 0.5 ppm, and particle surface area densities of about 90 $\mu m^2 cm^{-3}$, considerably larger than observed following the eruption of El Chichón. Aerosol volatility tests indicated that 95-98% of the particles were composed of an H_2SO_4/H_2O solution in all layers observed, indicating a rapid conversion of SO_2 to H_2SO_4 and subsequent droplet growth. The high concentrations of droplets, exceeding the pre-eruption condensation nuclei (CN) concentration above 20 km by about an order of magnitude for $r \geq 0.15$ μm particles and two orders of magnitude for the CN themselves, suggest homogeneous or ion nucleation as the most likely aerosol production mechanism.

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References

Deshler, T., and D. J. Hofmann, Measurements of unusual aerosol layers in the upper troposphere over Laramie, Wyoming in the spring of 1991: Evidence for long range transport from the oil fires in Kuwait, *Geophys. Res. Lett.*, in press, 1991.

Hofmann, D. J., Perturbations to the global atmosphere associated with the El Chichón volcanic eruption of 1982, *Rev. Geophys.*, *25*, 743-759, 1987.

Hofmann, D. J., Increase in the stratospheric background sulfuric acid aerosol mass in the past 10 years, *Science*, *248*, 996-1000, 1990.

Hofmann, D. J., and J. M. Rosen, Balloon-borne observations of stratospheric aerosol and condensation nuclei during the year following the Mt. St. Helens eruption, *J. Geophys. Res.*, *87*, 11,039, 1982.

Hofmann, D. J., and J. M. Rosen, Stratospheric sulfuric acid fraction and mass estimate for the 1982 volcanic eruption of El Chichón, *Geophys. Res. Lett.*, *10*, 313-316, 1983.

Hofmann, D. J., T. Deshler, F. Arnold, and H. Schlager, Balloon observations of nitric acid aerosol formation in the Arctic stratosphere: II. Aerosol, *Geophys. Res. Lett.*, *17*, 1279-1282, 1990.

Hofmann, D. J., and T. Deshler, Stratospheric cloud observations during formation of the Antarctic ozone hole in 1989, *J. Geophys. Res.*, *96*, 2897-2912, 1991.

McKeen, S. A., S. C. Liu, and C. S. Kiang, On the chemistry of stratospheric SO_2 from volcanic eruptions, *J. Geophys. Res.*, *89*, 4873-4881, 1984.

Post, M. J., C. J. Grund, A. O. Langford, and M. H. Proffitt, Multi-wavelength lidar observations of the Pinatubo cloud over Boulder, Colorado, *Geophys. Res. Lett.*, this issue, 1991.

Sheridan, P. J., R. C. Schnell, D. J. Hofmann, and T. Deshler, Electron microscope studies of Mt. Pinatubo aerosol layers over Laramie, Wyoming during summer 1991, *Geophys. Res. Lett.*, this issue, 1991.

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