were designed to avoid the plume from that eruption. For comparison, the size distribution of the stratospheric background aerosol sampled near Fairbanks, Alaska, in July 1979 is also shown in Fig. 2C. The total concentrations (C) of these two samples are similar, and they both appear to be trimodal. Because the estimated uncertainty in $\Delta C/C$ is approximately 0.06, it is not clear whether the peak between 0.1 and 1.0 μm indicates a mode or just scatter. However, judging from the relative amounts of material in the samples observed visually as well as by the electron microscope, a peak is indicated. The distinguishing feature is in the composition of the material in the size bands from 0.1 to 0.6 μm. Whereas the July 1979 background aerosol in this size range consisted mostly of carbonaceous material and relatively few sulfuric acid droplets, the material in the same size range from 17 June consisted of almost all sulfuric acid in abundant quantities. Comparison of Fig. 2C with Fig. 2B shows that the middle mode of the 17 June sample, centered at 0.22 μm, falls in the same size range as the two single modes from the 22 and 27 May samples. In addition, the abundance of sulfuric acid in the middle mode of the 17 June sample corresponds to the sulfuric acid in the 22 and 27 May samples. These similarities between the fresh plumes, sampled on 22 and 27 May, and the middle mode of the aged plume, sampled on 17 June, suggest that the middle mode contains the volcanic materials while the other modes are mainly background. Thus this middle mode might distinguish aged volcanic plumes from background.

Analyses are continuing, particularly to correlate our results with those of Sedlacek (*4*) on lower altitude particles, Gandrud and Lazrus (*7*) on total sulfate, and Inn, Vedder, and Condon (*8*) on gaseous SO_2.

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Size Distributions and Mineralogy of Ash Particles in the Stratosphere from Eruptions of Mount St. Helens

Abstract. *Samples from the stratosphere obtained by U-2 aircraft after the first three major eruptions of Mount St. Helens contained large globules of liquid acid and ash. Because of their large size, these globules had disappeared from the lower stratosphere by late June 1980, leaving behind only smaller acid droplets. Particle-size distributions and mineralogy of the stratospheric ash grains demonstrate inhomogeneity in the eruption clouds.*

Until Mount St. Helens erupted, there was little opportunity to study the nature of ash particles injected into the stratosphere by major volcanic explosions. Mossop (*1*) detected some large ash material associated with acid drops produced in the stratosphere by the eruption of Mount Agung, Bali, in 1963. Farlow *et al.* (*2*) also detected silicate particles from Mount Agung in samples from Australia (*3*). The eruption in 1974 of Volcan de Fuego, Guatemala, on the other hand, introduced few ash grains into the stratosphere (*4*). Mount St. Helens sent large intrusions of ash and liquid acid into the upper atmosphere.

We used an airborne particle sampler (*5*) carried beneath the wing of a NASA U-2 aircraft to collect samples of volcanic ash and acid droplets in the stratosphere from the first three major eruptions of Mount St. Helens. Particles struck fine wires of palladium and gold that extended from the instrument into the airstream. One of the gold wires, coated with high viscosity silicone oil, captured dry ash grains that might not stick to the bare wires.

Ash grains obtained 1 day after the eruption of 18 May 1980, were completely dry, whereas all ash collected later was covered with acid. The presence of this acid on most flights enabled the efficient capture of ash grains on the bare wires; the silicone oil trapped dry ash on the initial flight. If solid particles had bounced off the bare wires, this would have been detected by scars and dents left in the soft palladium (*5*). No such artifacts were seen except from the dry ash collections of 19 May.

These dry ash samples obtained over Montana at 14 and 17 km are composed of angular grains ranging in size from around 0.1 μm to as large as 30 μm, with median size around 0.3 μm (equivalent sphere diameters). A relative size distribution for the 14-km specimen is shown in Fig. 1a. Because of the difficulty of completely removing the grains from the viscous silicone oil in which they were imbedded, the volume of air we sampled could not be accurately determined. Therefore, the relative size distribution we obtained was normalized to a sample collected on 22 May so it could be compared to a distribution of known concentration. In this comparison the general shapes of the curves and the particle size modes are similar. Mineral compositions of these dry particles are somewhat like those of samples we collected later in that glass and plagioclase are major components; but the samples are otherwise quite different. Table 1 presents the mineralogical features of all the collections. Mineralogy is derived from elemental analyses of individual ash grains by nondispersive x-ray methods in the scanning electron microscope. Mineralogical results from the 17-km sample collected over Montana on 19 May are also in Table 1, but no size distribution was obtained.

Although the first samples of ash were very dry, flow marks in dust on the aircraft wings suggested that there were zones within the cloud containing large amounts of liquid, presumably acid, associated with the ash. The second flight to Montana on 22 May, 4 days after the initial eruption, provided an ash sample flooded with acid. Collecting wires were completely coated on the flight-facing side with coalesced acid drops encasing ash grains. Because the acid had converted to $(NH_4)_2SO_4$ in the laboratory environment (*6*), the ash grains could not be resolved in the scanning electron microscope until the sulfate crystals were removed. We did this by heating the collecting surface in an oven at 250°C for $1^1/_2$ to 2 hours. Subsequent visual examination and x-ray analyses for sulfur showed that volatile sulfates had been removed, leaving the ash grains intact and easily measured. We used this technique for all later samples where the presence of sulfates interfered with the ash grain analyses. Figure 1b presents the ash particle size distribution for this collection. Maximum sizes in this sample

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Table 1. Characteristics of Mount St. Helens ash from the stratosphere; N.F., not found; Q.P., questionably present.

Date collected (1980)	Eruption sampled	Days after eruption	Altitude (km)	Mineralogy (percent by volume)				
				Glass	Plagioclase	Hornblende	Pyroxene	Other
19 May	1	1	14	65	32	2.3	0.1	Sulfides, biotite, chromite
19 May*	1	1	17	37	16	21	13	SiO_2†
22 May	1	4	18	60	13	5.0	22	N.F.
27 May	2	2	17	80	20	N.F.	N.F.	N.F.
29 May	2	4	12	64	18	4.6	N.F.	SiO_2,† sulfides, ilmenite
3 June	1	16	12	100	Q.P.	Q.P.	N.F.	N.F.
13 June	3	1	12	83	10	2	5	N.F.

*Not included in size distribution data, Fig. 1. †Cristobalite?

did not exceed about 3 μm in equivalent sphere diameter.

We calculated log-normal functions that best fit the truncated size-frequency distributions and drew them as solid curves on the data sets in Fig. 1. The general log-normal equation is

$$\frac{dN/dr}{V} = \frac{N}{\sqrt{2\pi}\sigma_{-1}V}\,\frac{1}{r}\exp\frac{-(\ln r - \ln r_{-1})^2}{2\sigma_{-1}^2}$$

where N is total number of particles, V is sample volume in cm^3, r is particle radius, r_{-1} is median particle size, and σ_{-1} is related to the standard deviation of the size-frequency distribution. Parameter values for this equation are shown on Fig. 1. Again, nondispersive x-ray analyses provided elemental compositions from which mineralogy was derived for the 22 May sample (Table 1). In this group of particles, as in all other groups we analyzed, there were no significant variations of composition with particle size.

The stratospheric portion of the 18 May eruption cloud drifted eastward, circling the earth in about 16 days (*7*). We detected its arrival over California on 3 June, when our collector, flown at 12 km, was again flooded with acid drops and ash grains. These grains were less than 3 μm in diameter. No ash grains were found above the 12-km layer. Figure 1c shows the particle size distributions and the log-normal parameter values and Table 1 shows the mineralogy.

The second major eruption of Mount St. Helens occurred on 25 May 1980. We obtained two samples of ash flooded with acid 2 days and 4 days after the eruption (Table 1). The first collection was obtained off the British Columbia coast and the second over California. The size distributions are shown in Fig. 1, d and e, and Table 1, 27 May and 29 May. Again the ash grain sizes were less than 3 μm in diameter. Comparison of Fig. 1d with Fig. 1b suggests that the size distributions at high altitudes for the first two eruptions are similar, as are the low altitude ones (Fig. 1, c and e), but high altitude distributions do not compare favorably with low altitude ones; nor, for that matter, do any of the mineral groups compare well with each other.

We collected a sample from the third explosion on 13 June off the coast of Washington, the day after the volcano's eruption. This time our collecting surfaces were not flooded, although a dense collection of very large acid drops was obtained. Ash grains (< 3 μm in diameter) were scattered about the surfaces and stuck to the bare wires by a coating of acid around the individual grains. The size distribution (Fig. 1f) is somewhat similar (mode radius near 0.6 μm) to the other low altitude samples (Fig. 1, c and e).

An examination of the size distributions in Fig. 1 reveals that high altitude specimens generally have particle size modes around 0.3 μm in radius, and lower altitude samples have modes at about 0.6 μm. Even though the distributions coincide at the large particle end, there is a marked decrease in the small particle component at lower altitudes. However, the sizes of the ash grains do not represent the maximum sizes of the acid-ash globules. In all but the initial 19 May collection, a considerable quantity of acid encased the ash grains.

The log-normal curves fit the experimental data closely at higher altitudes (Fig. 1, b and d) and at one of the lower altitudes (Fig. 1f). They are well within the 95 percent confidence interval we determined for each size group and are confirmed by the chi-square test of fit. Such a good fit for volcanic ash is perhaps not surprising since, according to the theory of breakage (*8*), mechanical grinding, which occurs in the explosive breakup of volcanic material, produces a distribution of particles that closely fits a

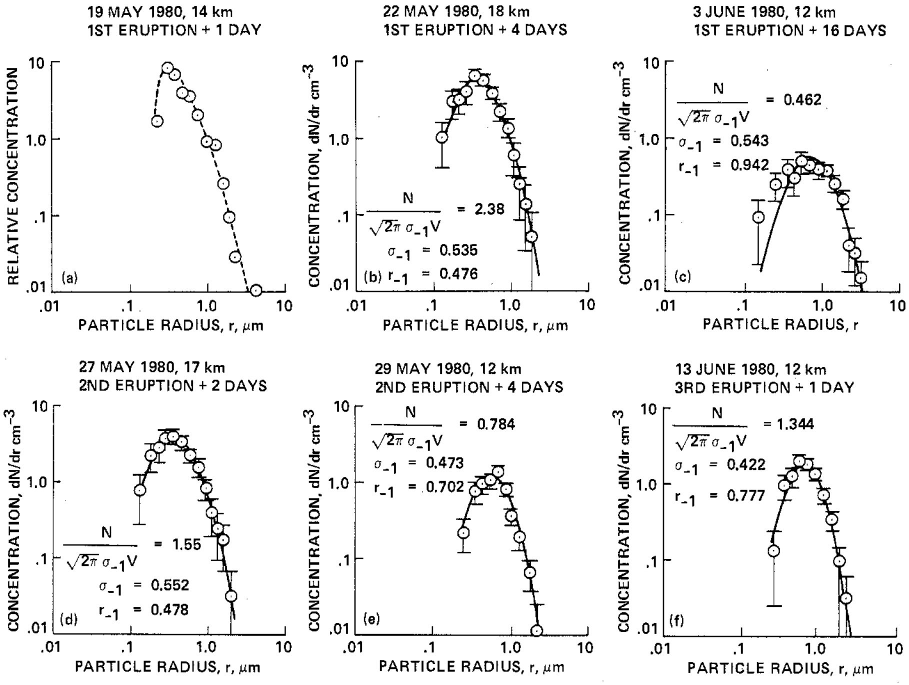

Fig. 1. Ash grain particle size distributions in the stratosphere for three Mount St. Helens eruptions. Solid curves are best fitting log-normal functions from the stated parameter values. Error bars are 95 percent confidence intervals for the population as inferred from the samples.

log-normal function. If this premise is accepted, then the two high altitude samples (22 and 27 May, Fig. 1, b and d) and one of the low altitude collections (13 June, Fig. 1f) might be considered most representative of the injected ash for their respective eruptions and altitudes. The other two low altitude samples (29 May, Fig. 1e, and 3 June, Fig. 1c) do not closely fit log-normal functions and may have been perturbed by external events such as size fractionation from fallout or meteorological effects.

The mineralogy of samples of the stratospheric ash from different parts of the cloud produced by a given eruption are not similar, nor are data from different eruptions comparable. This is probably due to the inhomogeneity of the overlying soil and rocks on Mount St. Helens. We have briefly looked at some ash samples collected at ground level from the first eruption. A coarse sample from Yakima, Washington, had about equal amounts of plagioclase and glass; less than 15 percent by volume of other phases was present. Very fine-grained ash from Pullman, Washington, was about 90 percent glass, and the rest was mostly plagioclase. Thus even the fine-grained ground sample is unlike the samples from the stratosphere after the first eruption. This further reflects the inhomogeneity of the ash produced.

Additional evidence for inhomogeneity of the ash cloud is found by comparing the two 19 May samples (Table 1). These were taken over the same location at nearly the same time but at different altitudes, and they are notably different mineralogically.

Because the sample from 3 June, collected 16 days after the first eruption, is nearly all glass, it might be assumed that mineral fragments have somehow been sorted out. But it is not likely that the later sample represents the same ash that was collected on 19 or 22 May. Conceivably a sample collected on 3 June from another altitude or locality might contain more fine-grained minerals.

Maximum stratospheric perturbations from the eruptions of Mount Agung (1963) and Volcan de Fuego (1974) were observed in mid-northern latitudes about 3 months after the eruptions (*4*), although initial effects were seen much sooner. This time lapse occurred partly because of the slow conversion of injected sulfur gases to sulfuric acid drops. Stratospheric aerosol models (*9*) were designed principally to show the long-term effects of this slow gas conversion. The injection of large globules of ash and liquid by the Mount St. Helens eruptions may cause different effects. Because the acid-coated ash particles were large, they have fallen quickly from the stratosphere, leaving behind only smaller acid droplets and whatever sulfur gases had not reacted. Consequently, it remains to be seen whether the buildup of acid aerosols a few months hence will occur as dramatically as it did after the volcanic explosions in the earlier decades.

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Absorption of Visible Radiation by Aerosols in the Volcanic Plume of Mount St. Helens

Abstract. *Samples of particles from Mount St. Helens were collected in both the stratosphere and troposphere for measurement of the light absorption coefficient. Results indicate that the stratospheric dust had a small but finite absorption coefficient ranging up to 2×10^{-7} per meter at a wavelength of 0.55 micrometer, which is estimated to yield an albedo for single scatter of 0.98 or greater. Tropospheric results showed similar high values of an albedo for single scatter.*

Increased burdens of atmospheric aerosols can lead to either a cooling or warming of the earth, depending on the optical properties of the particles (*1-3*). Several theories of climatic variability have suggested that volcanic emissions may have contributed to a cooling of the earth in the past (*4*). The eruptions of Mount St. Helens during the spring of 1980 provided an opportunity to determine if these volcanic emissions might contribute to a warming of the stratosphere as suggested by Newell (*5*), and to what extent absorption of solar radiation might reduce insolation at the ground.

Plume aerosol samples were collected in the troposphere by the University of Washington's B-23 aircraft (*6*), and in the stratosphere by the NASA U-2 aircraft (*7*). In the troposphere, the plume was too dense for the B-23 to remain in it safely for very long, so a grab-sampling procedure was used. As the aircraft flew through the fringe of the plume a 600-liter Mylar bag was filled by ram air pressure, and aerosol samples were taken from the bag. When the bag was nearly exhausted, another pass through the plume was made to refill it. For the absorption measurement, the aerosol from the bag was passed through a cyclone to remove large particles (50 percent removal efficiency for particles 0.7 μm in diameter), which made the measurement sensitive only to those particles which are not rapidly removed from the atmosphere by sedimentation. After passing through the cyclone, the particles were collected on Pallflex 2500 QAST quartz fiber filters for subsequent optical analysis.

The sampling system used on the U-2 is described in (*8*). Samples were taken with a 1-mm wide opal glass impactor that was glued into a sealed module mounted under the wing of the aircraft and was extended into the free airstream only when the aircraft was in the plume. For typical sampling conditions, the glass impactor has a theoretical collection efficiency of 50 percent for particles 0.2 μm in diameter. Despite their difference in size-dependent collection characteristics, the two samplers provide comparable measures of the absorption coefficient attributable to climatically important aerosols for two reasons: large particles removed by the cyclone have relatively short lifetimes because of their high settling velocities, while absorbing rock or mineral particles too

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