tor from crustal abundances rather than the composition of ash, many elements are depleted, as the volcanic material is of dacitic composition (*12, 13*).

There are some significant changes with altitude in the plume. For example, the Si enrichment, which is constant for most samples, does show an increase in the higher reaches of the plume. A group of elements (Ca, Sc, Ti, Fe, Zn, and Th) shows a similar pattern of enrichment at higher altitudes. Although the enrichments are not large, they are significant for these elements that are not usually fractionated by volcanic processes. These enrichments may occur because the source of the higher altitude clouds is in part the hotter magmatic debris rather than simply that from the physical breakup of the material from the mountaintop. The volatile elements As, Se, and Sb show little or no enrichment, indicating either that portions of them were in the vapor phase and thus missed by particle collection, or that they were not fractionated significantly. Due to the temperatures in the stratosphere and the large quantities of ash present, it is doubtful that very much of the Se, As, and Sb would be in the vapor phase. On the other hand, there may have been some fractionation that was not observed because of the presence of the huge amounts of unfractionated ash that was released. We cannot explain the huge enrichment of Cd at high altitudes. Since the PGAA and INAA results are in good agreement, the analyses appear to be valid. There might have been Cd contamination on the filters, although no materials of high Cd concentration were used in the collection system. The source of contamination is under investigation.

The general composition of the plume material is similar to that of the ash that fell to the ground in western Washington. Only minor enrichments of volatile elements are indicated; Cd, Sb, S, and Zn were the most enriched. The plume was fractionated vertically, as judged by the slight increases of EF_{ash} for some elements at higher altitudes. The fraction of water-soluble chlorine increased greatly with altitude, but the total chlorine concentration on particles was not much greater than for ash that fell to the ground. On the basis of $^7Be/SO_4^{2-}$ ratios, most of the sulfur observed resulted from the volcanic eruption.

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Characterization of Aerosols from Eruptions of Mount St. Helens

Abstract. *Measurements of mass concentration and size distribution of aerosols from eruptions of Mount St. Helens as well as morphological and elemental analyses were obtained between 7 April and 7 August 1980. In situ measurements were made in early phreatic and later, minor phreatomagmatic eruption clouds near the vent of the volcano and in plumes injected into the stratosphere from the major eruptions of 18 and 25 May. The phreatic aerosol was characterized by an essentially monomodal size distribution dominated by silicate particles larger than 10 micrometers in diameter. The phreatomagmatic eruption cloud was multimodal; the large size mode consisted of silicate particles and the small size modes were made up of mixtures of sulfuric acid and silicate particles. The stratospheric aerosol from the main eruption exhibited a characteristic narrow single mode with particles less than 1 micrometer in diameter and nearly all of the mass made up of sulfuric acid droplets.*

The quartz crystal microbalance cascade impactor (*1*) classifies aerosol particles by size from inertial impaction. Each of the ten impactor stages contains a piezoelectric crystal microbalance which senses the mass of particles on impact. This enables the device to weigh the particles in each size interval, in real time, as sampling proceeds. Size-fractionated aerosol samples from eruptions of Mount St. Helens were collected by several aircraft, and the samples were analyzed by scanning electron microscopy and energy dispersive x-ray techniques with an electron microprobe (*2*).

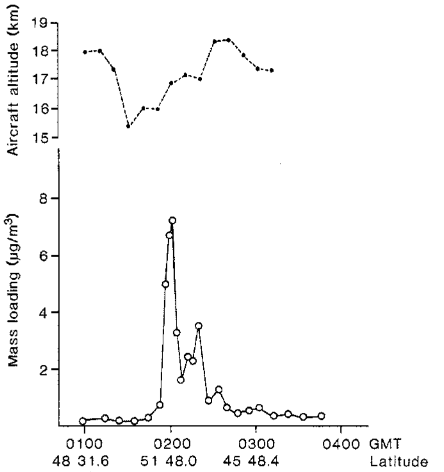

Fig. 1. Total mass loading as a function of time, position, and altitude measured by a U-2 aircraft in the eruption plume of Mount St. Helens on 27 May 1980; *GMT*, Greenwich Mean Time.

For sampling the plume in the stratosphere, the quartz crystal microbalance instrument is housed inside a NASA U-2 research aircraft. An externally mounted isokinetic sampling probe, designed for the U-2 airspeed, brings ambient air into the instrument through a control valve that is operated from the cockpit. A vacuum pump draws the sample through ten impaction stages. The geometric mean particle diameter for stage one is greater than 22 µm, and the diameters for stages two through ten are 14.8, 6.9, 3.2, 1.3, 0.54, 0.23, 0.11, 0.065, and 0.043 µm. Measurements in the plume near the volcano were made with the instrument mounted on a twin-engine QueenAire operated by the National Center for Atmospheric Research in April and on a small Navaho aircraft from the U.S. Geological Survey in August. An isokinetic sampling probe was used on the QueenAire, but a straight-through non-

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isokinetic probe was used on the Navaho. Because of the lower altitude (higher ambient pressure), the geometric mean particle diameters are higher than the values above and are as follows: greater than 30 μm for stage one and 15.3, 7.6, 3.8, 1.8, 0.92, 0.47, 0.26, 0.17, and 0.12 μm for stages two through ten, respectively.

Direct measurements in the plume just above the crater of Mount St. Helens were obtained on 7 April during the early phreatic phase of the eruptions and on 6 August during the minor phreatomagmatic phase after the major eruptions of May and June and just before the moderate eruption of 7 August. Measurements of material injected into the stratosphere by the volcano were made on 22 May, 4 days after the major eruption of 18 May, and on 27 May, 2 days after the second major eruption of 25 May. On 17 June the sampling flight was made through aged clouds, probably from the May eruptions.

A representative plot of total aerosol mass concentration against flight time is shown for the 27 May flight in Fig. 1. The location of maximum concentration, which was 30 times above the background concentration, was about 1000 km northwest of Mount St. Helens at an altitude of about 16.5 km. The plot suggests that the plume was fairly well confined in both vertical and horizontal extents, with an altitude band from about 16 to 18 km, and a horizontal spread of about 400 km. Similar data were obtained for the other stratospheric flights and close-in traverses near the mountain. From the aerosol data near the maximum concentration, details of the size distribution were extracted.

The aerosol from the phreatic eruption of 7 April was essentially monomodal; over 90 percent of the mass was larger than 10 μm in diameter, and less than 3 percent was less than 1 μm (Fig. 2A). The total mass concentration was high because the sampling aircraft flew into the eruption cloud directly over the crater. The material consisted mainly of silicates. By comparison, the size distribution of the aerosol from the premonitory (essentially continuous) emissions from the crater on 6 August, about 27 hours before a relatively strong eruption on 7 August, was trimodal, with a prominent mode centered at about 5 μm and a distinctive submicron mode at about 0.3 μm. This pattern was closely matched on 7 August about 1 hour before the eruption (Fig. 2A), although the relative magnitude of the submicron mode was submerged by the much higher absolute magnitudes of the two larger modes. Because of the low speed of the aircraft used on 6 and 7 August, and the non-isokinetic inlet probe, the large particles (> 10 μm) may have been preferentially sampled. Thus, the curves for 6 and 7 August (Fig. 2A) may overrepresent these large particles. The total mass concentration measured on 6 August (40 μg/m^3) was lower because of low level emissions and the distance from the volcano at which the sample was taken (5 km). On 7 August the intensity of emission was stronger and the sampling was made less than 1 km leeward of the crater. The material in the impactor stages corresponding to the two small modes from the 6 and 7 August samples consisted of silicates heavily mantled in sulfuric acid similar to that found in the close-in plume from Santiaguito, Guatemala, as reported by Rose *et al.* (*3*). This is significantly different from the material found in the phreatic eruption plume of 7 April, which consisted of larger, dry sulfur-free particles. Particles in the large size mode showed little or no acid mantling.

The size distributions for the 27 May sample measured between 16 and 18 km, 2 days after the 25 May eruption, and the 22 May sample measured at about 20 km, 4 days after the 18 May eruption, are shown in Fig. 2B. These are seen to be monomodal, with the sample from 27 May showing a narrower band centered at a smaller size than was exhibited by the 22 May sample. The difference may suggest growth of the particles with time. The material in the size interval corresponding to the modes in both cases were determined to be all sulfuric acid. In both samples a small amount of material, constituting less than 10 percent of the total aerosol mass and > 1 μm in diameter, was found to consist of silicates mantled in sulfuric acid. There appeared to be strong size fractionation of the aerosol in the vertical direction. We found almost all sulfuric acid droplets smaller than 1 μm at 20 km on 22 May. However, Sedlacek (*4*), sampling at a lower altitude (about 17 km) on 21 May, found mainly silicate particles larger than 2 μm.

The appearance of sulfuric acid in the stratosphere within a few days of the eruptions and the sulfuric acid mantles over silicate particles sampled near the volcano suggest that sulfuric acid is formed by short-term processes in the eruption in addition to the atmospheric processes with long-time constants postulated by Cadle (*5*) and Davis (*6*).

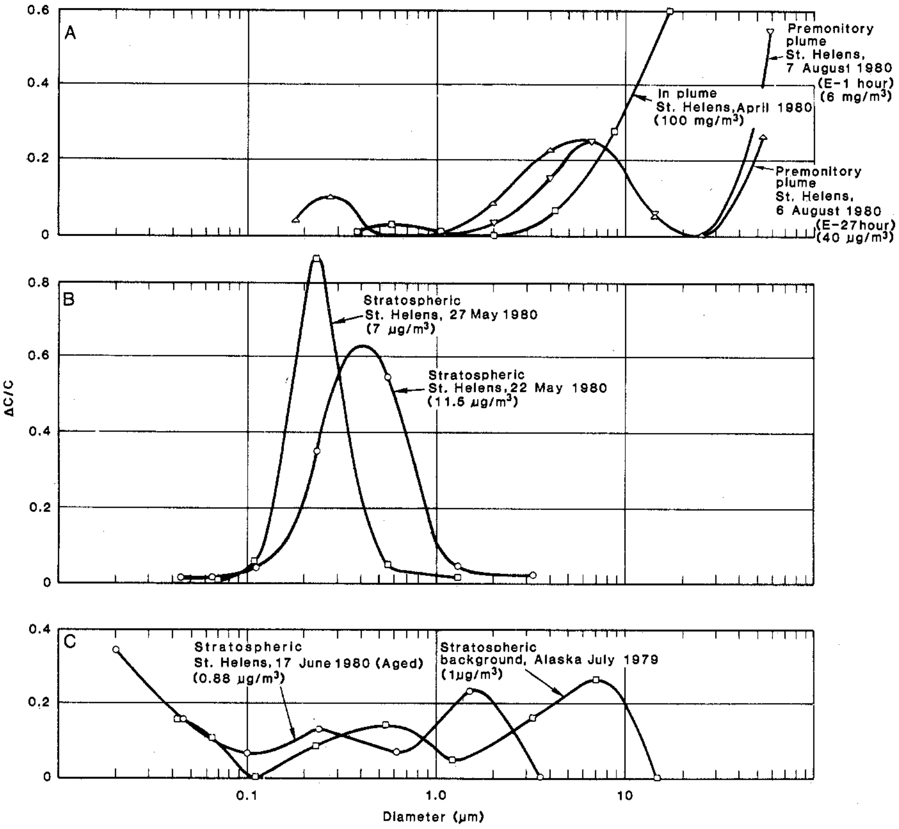

Fig. 2. Normalized plots of aerosol concentration as a function of diameter in close-in plumes and stratospheric plumes from Mount St. Helens; *E*, eruption.

The size distribution of an aged eruption plume was sampled on 17 June (Fig. 2C). While there had been an eruption on 13 June, the sampling paths on 17 June 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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