

the stratosphere are of greater relevance to climatic studies than the properties of tropospheric material since some material can remain in the stratosphere for months to years, whereas material in the troposphere can be removed by rain in days (11). The reports that follow provide a reasonably complete definition of the radiative properties of volcanic aerosols in the stratosphere and will therefore permit an assessment of their impact on regional and global radiation budgets and hence on weather and climate. For example, both the absorption and scattering coefficients of the volcanic aerosols can be determined from the ACE measurements (2, 14). Whether the aerosols cause an increase or decrease in the fraction of sunlight reflected back to space depends on their single scattering albedo (ratio of scattering to the sum of the scattering and absorption coefficients), the fraction of light scattered into the backward hemisphere, and the albedo of the underlying atmosphere and surface (15). The very high single scattering albedo of the Mount St. Helens stratospheric aerosols (2), in conjunction with the backscattering ratio inferred from their measured size distribution (3, 9, 16), implies that they caused the earth's albedo to increase and hence the fraction of absorbed sunlight to decrease (17).

Volcanic aerosols warm the layers of the atmosphere in which they reside by absorbing both incident sunlight and thermal radiation from the lower, warmer regions of the atmosphere, and they warm or cool the troposphere and surface depending on the competition between the decrease in the amount of sunlight reaching the tropopause and the increase in the amount of downward directed thermal radiation emitted by the lower stratosphere (15). The size of the stratospheric volcanic particles ( $> 0.2 \mu\text{m}$ ), in conjunction with their high single scattering albedo and sulfuric acid and silicate composition, probably result in a dominance of the first of these factors; thus a cooling is to be expected (17). Much more complete analyses of these data will permit an assessment of the short- and long-term climatic effects of the eruptions of Mount St. Helens as well as an extension of these results to other volcanic episodes, such as the period from 1880 to 1915, when major volcanic events were frequent and the climate was colder than during subsequent volcanically quiescent times (18).

JAMES B. POLLACK  
NASA Ames Research Center,  
Moffett Field, California 94035

#### References and Notes

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2. J. A. Ogren, R. J. Charlson, L. F. Radke, and S. K. Domonkas (*ibid.*, p. 834) obtained measurements of the aerosols' absorption coefficient at a wavelength of  $0.55 \mu\text{m}$  in both the stratosphere and troposphere. Since the absorption was probably due primarily to the ash component, the shape of the spectral absorption curve for ash found by Patterson (19) can be used to extend these results to other visible wavelengths.
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13. B. Gandrud and A. Lazrus, *ibid.*, p. 826. Also see (10) on the temporal evolution of the mass density and composition of the volcanic aerosols in the stratosphere.
14. The absorption coefficient is found from laboratory transmission measurements made by Ogren *et al.* (2) on collected samples, while the scattering coefficient is obtained by applying Mie scattering theory to the measured size distributions.
15. J. B. Pollack, O. B. Toon, C. Sagan, A. Summers, B. Baldwin, W. Van Camp, *J. Geophys. Res.* **81**, 1071 (1976); J. A. Coakley, Jr., and G. W. Grams, *J. Appl. Meteorol.* **15**, 679 (1976). See also (19).
16. Mie theory can be used for this purpose since the particles were typically separate sulfuric acid droplets or ash particles with a sulfuric acid coating (3, 9).
17. J. B. Pollack, O. B. Toon, D. Wiedman, *Geophys. Res. Lett.*, in press; J. B. Pollack, paper presented at the International Radiation Symposium, Fort Collins, Colo., August 1980. See also (15).
18. J. B. Pollack, O. B. Toon, C. Sagan, A. Summers, B. Baldwin, W. Van Camp, *Nature (London)* **263**, 551 (1976).
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20. The success of the U-2 flights through the volcanic plumes of Mount St. Helens was due in large measure to the efforts of W. Page, E. Danielsen, and the U-2 pilots and ground support personnel at NASA Ames Research Center. These ACE activities were sponsored by the Climate Program Office of NASA.

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## Airborne Studies of the Emissions from the Volcanic Eruptions of Mount St. Helens

**Abstract.** *The concentrations of particles less than 10 micrometers in diameter in the ash emissions from Mount St. Helens have been more than 1000 times greater than those in the ambient air. Mass loadings of particles less than 2 micrometers in diameter were generally several hundred micrograms per cubic meter. In the ash clouds, produced by the large eruption on 18 May 1980, the concentrations of several trace gases generally were low. In other emissions, significant, but variable, concentrations of sulfur gases were measured. The 18 May eruption produced nuées ardentes, lightning flashes, and volcanic hail.*

Mount St. Helens, which is situated about 140 km south of Seattle in the Cascade Mountains of Washington State, ended its 123 years of dormancy on 20 March 1980 with a single small earthquake registering 4.1 on the Richter scale (1). Seismic activity increased steadily, becoming nearly continuous by 25 March.

The first eruption, which occurred at 2036 Universal Time (2) on 27 March 1980, ejected volcanic material to an altitude of 4.9 km (3). Airborne measurements and observations made from the University of Washington's research aircraft (4) began on 28 March when periodic (about every 5 to 10 minutes) eruptions of ash, and more frequent but shorter duration emissions of steam, reached up to 6.1 km (Fig. 1A). The size spectra of particles were measured in a cloud of ash 13 km downwind of the mountain (Fig. 2A). The number concentrations of particles with diameters be-

tween 1 and  $10 \mu\text{m}$  in this cloud were up to 1000 times greater than in the ambient air, but particles  $< 1 \mu\text{m}$  in diameter were found in concentrations similar to those in the ambient air. The mass concentrations of particles  $< 2 \mu\text{m}$  in diameter in the ash cloud and in the ambient air were about 600 and  $30 \mu\text{g m}^{-3}$ , respectively. Gaseous sulfur, almost entirely reduced sulfur, which we presume by its smell to be  $\text{H}_2\text{S}$ , was detected in concentrations of up to 240 ppb (5).

Between 28 March and 17 May, when the volcanic activity was quite variable, seven flights were made. The measurements on 2, 4, 8, and 11 April showed concentrations of total gaseous sulfur ranging from 15 to 38 ppb with quite variable contributions from  $\text{H}_2\text{S}$  and  $\text{SO}_2$ . However, on 8 May the total gaseous sulfur concentrations had returned to values similar to those measured on 28 March.

Early on 18 May, seismic activity

and visible emissions had virtually ceased when suddenly a major eruption occurred at 1532. A large portion of the north face of the mountain collapsed in avalanches and pyroclastic flows, devastating a wide area north and east of the mountain; this eruption caused the first recorded human deaths due to volcanic activity in the continental United States. The explosive eruption ejected material to an altitude of about 20 km, and ash was carried eastward over Washington, Idaho, and Montana within a few hours.

We started our airborne measurements in the vicinity of Mount St. Helens at  $\sim 1900$  on 18 May. Violent emissions of ash were still continuing at this time (Fig. 1B), numerous *nuées ardentes* (glowing avalanches) were cascading down the sides of the mountain, and lightning flashes, presumably caused by charge separation during particle collisions in the very dense clouds of ash, were common. We attempted to obtain our first measurements at  $\sim 2000$  at the edge of the dark ash plume  $\sim 37$  km downwind and east of the volcano but were driven back as the aircraft was bombarded with clumps of loosely aggregated material (up to fist size) that disintegrated on hitting the plane; similar bombs, both dry and wet, were observed on the ground 12 km from the summit of Mount St. Helens within a few minutes of the eruption (6). We believe these bombs were formed by the rapid aggregation of ash particles in the plume; we propose they be termed volcanic hail (7).

We obtained measurements of particle size distribution at the periphery of the ash plume,  $\sim 9.3$  km downwind at  $\sim 2010$  (Fig. 2B). For particles 0.01 to 10  $\mu\text{m}$  in diameter, the concentrations in the plume varied from a factor of 4 to over 1000 times those in the ambient air. For particles  $< 2 \mu\text{m}$  in diameter, the mass loading in the plume was  $\sim 95 \mu\text{g m}^{-3}$  compared to  $< 0.1 \mu\text{g m}^{-3}$  in the ambient air. Optical polarization and x-ray diffraction measurements showed that the particles consisted primarily of lithic rock fragments, largely plagioclases, with some apparently juvenile glasses.

The concentrations of sulfur gases measured at the periphery of the main ash cloud on 18 May were generally only slightly greater than those found in the ambient air; nitrogen oxides and ozone remained at or below ambient concentrations. It is possible that the small quantity of sulfur gas reflects absorption of reactive trace gases on the ash plume's very large total particle surface area. However, the ash samples we have ana-

lyzed showed generally small amounts of soluble materials such as sulfate, while samples from the stratosphere collected on U-2 flights showed the main cloud to steadily increase in acidity over a period of days, evidently indicating the presence of significant trace gas concentrations (8). Thus these observations may reflect temporal or even altitude variations.

In addition to the dark ash plume, smaller whitish-looking clouds of effluents were observed on 18 and 19 May along the edges of the main plume (Fig. 1C) (9). These clouds were small enough to be penetrated completely by our aircraft. Particle size distribution measured near the center of such a cloud (Fig. 2C) showed that for particles  $< 0.1 \mu\text{m}$  in diameter, the concentrations were similar to those in the ambient air, but the concentrations of particles from  $\sim 0.1$  to 30  $\mu\text{m}$  in diameter were up to 100 times greater than in the ambient air. The mass concentrations of particles  $< 2 \mu\text{m}$  in di-

ameter in the whitish clouds and in the ambient air were  $\sim 30$  and  $5 \mu\text{g m}^{-3}$ , respectively. The concentration of total sulfur gases in these clouds, primarily  $\text{SO}_2$ , was at least 13 ppb, compared to  $\sim 6$  ppb in the ambient air. Nitrogen oxides and ozone concentrations in the whitish clouds were similar to those in the ambient air.

Our final measurements on this flight were taken near the top of the main ash plume ( $\sim 5.2$  km downwind) late in the evening, when the volcanic activity was much less than earlier in the day. These measurements showed a high concentration of total sulfur gases ( $\sim 1$  ppm) that consisted primarily of  $\text{H}_2\text{S}$ .

The emissions of particles and gases from the volcano measured on our next flight, much later on 19 May, were considerably lower than on the previous flight, although they were still considerable. Particle size spectra were measured on the periphery of the main ash emissions (Fig. 2D),  $\text{H}_2\text{S}$  concentrations

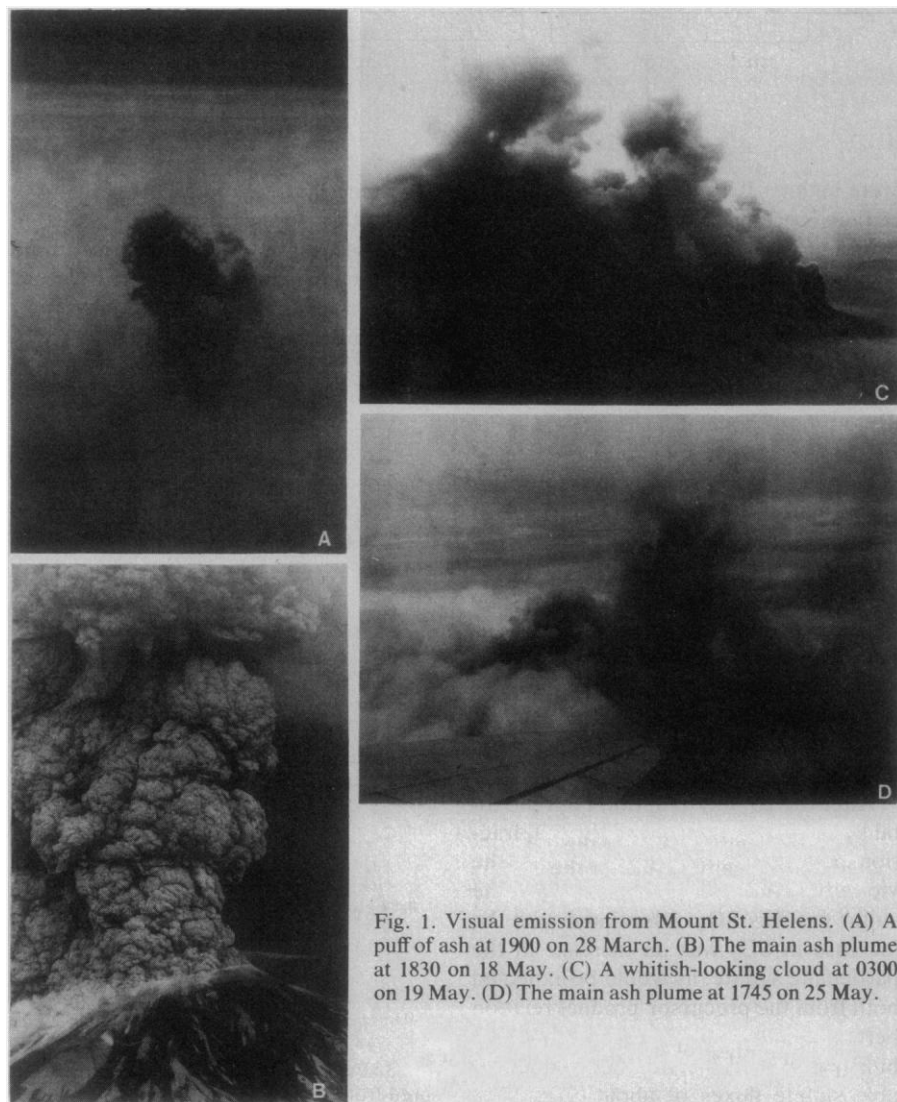


Fig. 1. Visual emission from Mount St. Helens. (A) A puff of ash at 1900 on 28 March. (B) The main ash plume at 1830 on 18 May. (C) A whitish-looking cloud at 0300 on 19 May. (D) The main ash plume at 1745 on 25 May.

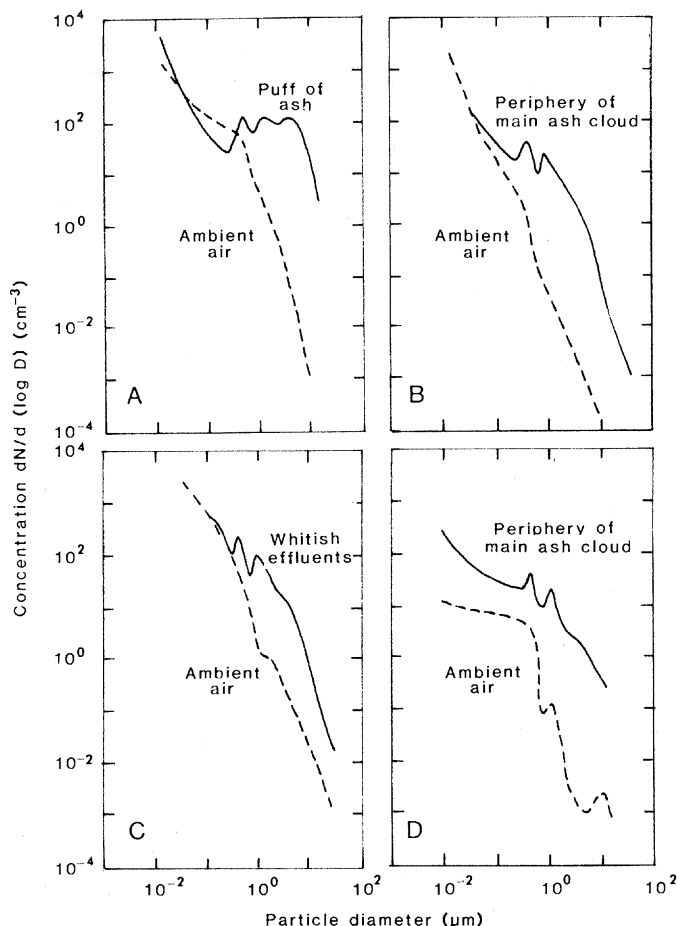


Fig. 2. Size spectra of small particles: (A) in a puff of ash at an altitude of 3 km and 13 km downwind of the volcano at 2239 on 28 March; (B) at the periphery of the dark ash cloud at an altitude of 3 km and 9.3 km downwind of the volcano at 2010 on 18 May; (C) near the middle of the whitish-looking clouds at an altitude of 3.2 km and 130 km downwind of the volcano at 0230 on 19 May; (D) at the periphery of the main ash cloud at an altitude of 2.7 km and 22 km downwind of the volcano at 2140 on 19 May.

were high ( $\sim 0.5$  ppm), but  $\text{SO}_2$  concentrations were near ambient values.

Mount St. Helens exploded violently for the second time at 0932 on 25 May when ash was ejected to an altitude of 13.7 km (Fig. 1D). Particle size measurements obtained in the periphery of the main ash plume just after 1800 were similar to those obtained on 18 May. Unlike the 18 May measurements, we detected consistently high amounts of total sulfur gases, up to 110 ppb, with 13 to 95 ppb of  $\text{H}_2\text{S}$  and 3 to 6 ppb of inert sulfur-containing gases.

Measurements on 30 May and 4 June showed continuing gaseous emissions but much reduced ash emissions. A significant fraction of the smaller particles on these 2 days consisted of soluble material, with quantities of  $\text{SO}_4^{2-}$ ,  $\text{Br}^-$ ,  $\text{F}^-$ , and  $\text{Cl}^-$ .

After the 25 May eruption both the total sulfur gas emissions and the  $\text{SO}_2$  fraction increased dramatically, and after the violent eruption of 13 June most of the sulfur gas was  $\text{SO}_2$ . Associated with the increase in  $\text{SO}_2$  was an increase in the particulate sulfate, as would be expected both from the precursor-product relation between  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  and the possibilities of direct injection of  $\text{H}_2\text{SO}_4$ . Soluble sulfate fluxes of about  $25 \text{ g sec}^{-1}$

were measured on both 4 and 18 June. During this period a significant fraction of the aerosol contained spherical fume particles and haloed particles that appear to have been impure  $\text{H}_2\text{SO}_4$  droplets similar to those found at Augustine Volcano, Alaska, in January 1976 and April 1977

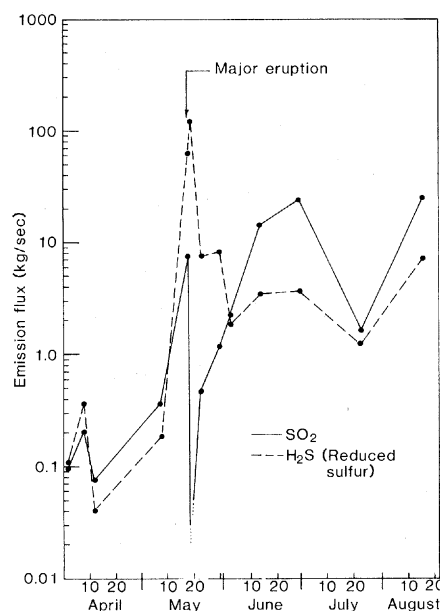


Fig. 3. Emission fluxes of  $\text{SO}_2$  and  $\text{H}_2\text{S}$  measured from Mount St. Helens.

(10). On 18 June the hot outgassing plume generated a small cumulus cloud that capped the caldera. The cloud droplets were found to have a  $\text{pH} \leq 1$ .

Elevated  $\text{NO}_2$  concentrations were observed only on one occasion. On 13 June, after the third violent eruption sequence, concentrations approaching 400 ppb were observed at a range of 8 km, resulting in an estimated emission flux of  $7 \text{ kg sec}^{-1}$ .

The gaseous sulfur emissions summarized in Fig. 3 as total fluxes were deduced from multiple traverses of the plumes and simultaneous measurements of horizontal winds. The emission fluxes for 18 May, however, are based on more fragmentary data of plume volume and are, therefore, subject to large potential error. These fluxes are quite similar to those we measured from the Augustine Volcano during its eruptive phase in February 1976 (9).

PETER V. HOBBS  
LAWRENCE F. RADKE  
MARK W. ELTGROTH  
DEAN A. HEGG

Department of Atmospheric Sciences,  
University of Washington,  
Seattle 98195

#### References and Notes

1. All seismic data in this report are taken from the logs of S. Malone at the Department of Geophysics, University of Washington, Seattle.
2. All times are Universal Time.
3. All heights are given above mean sea level.
4. P. V. Hobbs, D. A. Hegg, M. W. Eltgroth, and L. F. Radke have described the research instrumentation aboard this aircraft [*Atmos. Environ.* **13**, 935 (1979)].
5. Measurements of gaseous sulfur were made with a flame photometric detector with scrubbers which remove sequentially from the total sulfur stream the reduced sulfur species and then both reduced and oxidized species, allowing separation of oxidized sulfur (presumed  $\text{SO}_2$ ), reduced sulfur (presumed on the basis of smell to be  $\text{H}_2\text{S}$ ), and the comparatively inert species such as carbonyl sulfide and  $\text{CS}_2$ .
6. K. Rohnholm, personal communication.
7. Vulcanologists have referred to similar bombs as accretionary lapilli. These are most commonly observed to be less than 1 cm in diameter but have been observed as large as 15 cm during the 1902 eruptions of Mont Pelée. However, given the apparent similarities to hail formation, we consider volcanic hail to be a far more descriptive term.
8. J. A. Ogren, R. T. Charlson, L. F. Radke, S. K. Domonkos, *Science* **211**, 834 (1981); NASA/FAA Bull. Upper Atmos. Programs Issue 80-3 (1980).
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11. We thank all members of our airborne crew for their help in obtaining these measurements. We are grateful to T. Irving for optical and x-ray analyses of ash samples. Our measurements were made possible by prompt actions on funding by B. Fogle, G. Bierly, R. Dirks, and D. Senich of the National Science Foundation (grant DPP 79-20857), and by W. Wilson of the Environmental Protection Agency (grant R005263-02). Contribution 562, Atmospheric Sciences Department, University of Washington.

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