

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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## References and Notes

1. S. C. Mossop, *Geochim. Cosmochim. Acta* **29**, 201 (1965).
2. N. H. Farlow, G. V. Ferry, H. Y. Lem, *Space Res.* **13**, 1153 (1973).
3. The samples were provided by E. K. Bigg, CSIRO, Australia.
4. D. J. Hofmann and J. M. Rosen, *J. Geophys. Res.* **82**, 1435 (1977).
5. N. H. Farlow, G. V. Ferry, H. Y. Lem, D. M. Hayes, *ibid.* **84**, 733 (1979).
6. D. M. Hayes, K. G. Snetsinger, G. V. Ferry, V. R. Oberbeck, N. H. Farlow, *Geophys. Res. Lett.*, **7**, 974 (1980).
7. E. F. Danielsen, *Science* **211**, 819 (1981).
8. J. Aitchison and J. A. C. Brown, *The Log-Normal Distribution* (Cambridge Univ. Press, Cambridge, 1976).
9. R. P. Turco, P. Hamill, O. B. Toon, R. C. Whitten, C. S. Kiang, *J. Atmos. Sci.* **36**, 699 (1979).
10. We thank Harry Shade and Memorex Corporation for help in analyzing some of the volcano samples with their scanning electron microscope and Josef Erlichman of LFE Corporation for his analysis of some ash grain compositions. We also thank J. H. Ferry of Yakima, Wash., for the Mount St. Helens ash he sent to us and W. A. Page for the ash from Pullman, Wash.

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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  $2 \times 10^{-7}$  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  $\mu$ m 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  $\mu$ m 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

small to be collected by the impactor are not known to be produced by the mechanical processes associated with volcanic eruptions.

Absorption of visible light is measured by the integrating plate method (9-11). Particles collected on a transparent or translucent sampling substrate are placed in a beam of light in front of an opal glass slab, which acts as a Lambert scatter. This optical geometry minimizes the effect of scattering by the sampled particles, and transmission of light through the sandwich of particles, substrate, and opal glass is sensitive mainly to absorption. The absorption coefficient due to particles,  $\sigma_{ap}$ , is related to the initial and final transmission,  $I_0$  and  $I$ , respectively, by the Beer-Lambert law:  $I = I_0 \exp(-\sigma_{ap}x)$ , where  $x$  is the optical path length for the sample, that is, the air volume sampled divided by the cross-sectional area of the sampling substrate. The optical measurement is typically performed in the middle of the visible spectrum ( $\lambda = 0.55 \mu\text{m}$ ). The original integrating plate method used Nuclepore filters; however, because of a need for high face velocities, two variants on the method have been developed. The first, with collection on quartz fiber filters, uses an empirically determined multiplier of 0.35 to correct for the effects of multiple scatter and absorption within the matrix of the fiber filter (12). The second uses the opal glass surface as both particle collector and optical integrator of scattered light (8). For this variant, the sample path length is given as the product of exposure time and the true airspeed of the U-2 aircraft ( $200 \text{ m sec}^{-1}$ ).

The first tropospheric absorption measurements were obtained on 11 April 1980, during the period when minor ash and steam explosions were occurring. Sampling in the stratosphere began on 19

May, one day after the first major eruption. The stratospheric and tropospheric measurements are summarized in Table 1. The stratospheric samples (altitudes greater than 10 km above mean sea level) have much longer path lengths; these were necessary to detect the generally low absorption coefficients found in the stratosphere.

Comparison of the tropospheric values in Table 1 with previous measurements at remote locations in Washington State (12) indicates that the plume particulates less than a micrometer in diameter did not absorb visible radiation significantly in excess of background aerosol absorption. Likewise, the stratospheric measurements on 19 May and 17 June were not substantially different from values measured a year earlier (8). However, the absorption coefficients measured in the stratosphere on 22 May, 27 May, and 23 July are significantly greater than pre-eruption measurements and may be a more reliable measure of the stratospheric plume absorption coefficient. The reason for this is that the ash grains sampled on these days were flooded with acid (13) and probably stuck to the collection surface much more effectively than the dry ash encountered on 19 May. This hypothesis is supported by optical microscopic analyses of the collectors, which for the 19 May stratospheric sample showed that few ash particles adhered to the glass impactor, but many adhered to the silicone rubber adhesive used to secure the glass strip into its mounting ring. As a result, the absorption coefficient reported for 19 May should be considered as a lower bound to the true value.

Examination by optical microscopy also revealed that most of the particles appeared very slightly yellow or tan in color, and a few particles were much darker

orange or dark brown. This is consistent with mineralogical analysis indicating that most of the material is feldspar with inclusions of small amounts of colored pyroxenes as a separate phase. Estimates of  $\sigma_{ap}$  for the dark particles alone were made from the sizes and number of such particles observed in the optical microscope. This revealed that absorption by the dark particles in the 22 May sample was only a few times  $10^{-10} \text{ m}^{-1}$ , such that the bulk of the observed absorption of  $2 \times 10^{-7}$  had to be due to the weakly absorbing feldspars. Evidently an absorbing molecular species was dissolved or dispersed in the feldspar portion of the magmatic material. Since pyroxenes are ferromagnesian silicates, it is likely that the absorbing species contains iron.

An absorption coefficient of  $10^{-7} \text{ m}^{-1}$  for visible light implies a small but finite heating rate in the stratosphere of about  $2 \times 10^{-6} \text{ }^\circ\text{C}$  per second, assuming a single pass of the solar beam through an optically thin layer at 60 mbar. If this layer is irradiated for 12 hours a day, and all other factors are neglected, this yields a temperature increase of about  $0.1^\circ\text{C}$  per day. Although this heating rate is but one term in the energy balance of the stratosphere, the calculated rate of temperature increase is of the same magnitude as that suggested by Newell (5) as a result of the eruption of Mount Agung in Bali in 1963.

It is probable that absorption coefficients around  $10^{-7} \text{ m}^{-1}$ , though finite, are minimal in comparison with scattering by particles in the stratosphere. The scattering component of extinction calculated from the mass distributions obtained by the quartz crystal microbalance aboard the U-2 flights on 22 and 27 May were  $1.2 \times 10^{-5} \text{ m}^{-1}$  and  $1.1 \times 10^{-5} \text{ m}^{-1}$ , respectively (14-16). Thus, we

Table 1. Absorption coefficient ( $\sigma_{ap}$ ) measurements in plume of Mount St. Helens.

Date (1980)	Location	Altitude (km)	Optical path length (km)	$\sigma_{ap}$ ( $\text{m}^{-1}$ )	Comments
11 April	< 2 km downwind	2.4 to 2.9	2.6	$2.1 \times 10^{-6}$	
8 May	< 5 km downwind	3.5 to 3.7	2.4	$2.5 \times 10^{-5}$	Minor eruption
18 May	80 to 200 km downwind	2.7 to 5.2	2.0	$6.2 \times 10^{-6}$	
19 May	21 to 32 km downwind	2.7 to 3.2	1.1	$2.4 \times 10^{-6}$	
19 May	Washington to Wyoming	14 to 18	1900	$7.8 \times 10^{-9}$	Dry ash; particles did not adhere
22 May	Idaho and Montana	19 to 20	1330	$2.0 \times 10^{-7}$	Ash and acid mixture
23 May	< 15 km downwind	3.0 to 3.7	2.8	$9.6 \times 10^{-6}$	
27 May	British Columbia Coast	15 to 17	540	$4.7 \times 10^{-8}$	
30 May	< 15 km downwind	2.4 to 3.4	2.4	$5.5 \times 10^{-6}$	
4 June	< 25 km downwind	2.9 to 3.7	4.1	$3.3 \times 10^{-6}$	
13 June	90 km downwind	3.0	0.27	$2.8 \times 10^{-5}$	Minor eruption; insufficient sample volume
17 June	Colorado	18 to 21	1440	$3.8 \times 10^{-9}$	
23 July	Utah	17 to 18	600	$1.6 \times 10^{-8}$	

estimate single scattering albedos of 0.98 and 0.995, respectively, for the flights of these two dates. For the tropospheric samples, the integrating nephelometer aboard the B-23 was usually off-scale and above  $10^{-3} \text{ m}^{-1}$ . A typical value of  $\sigma_{\text{sp}}$  of  $5 \times 10^{-6}$  thus implies a lower limit for single scatter albedo of 0.995.

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#### References and Notes

1. R. J. Charlson and M. J. Pilat, *J. Appl. Meteorol.* **8**, 1001 (1969).
2. J. M. Mitchell, *ibid.* **10**, 703 (1971).
3. R. A. Reck, *Atmos. Environ.* **10**, 611 (1976).
4. J. B. Pollack, O. B. Toon, C. Sagan, A. Summers, B. Baldwin, W. Van Camp, *J. Geophys. Res.* **81**, 1071 (1976).
5. R. E. Newell, *Nature (London)* **227**, 697 (1970).
6. P. V. Hobbs, L. F. Radke, M. W. Eltgroth, D. A. Hegg, *Science*, **211**, 816 (1981).
7. J. B. Pollack, *ibid.*, p. 815.
8. J. A. Ogren, N. C. Ahlquist, A. D. Clarke, R. J. Charlson, *Geophys. Res. Lett.*, in press.
9. C. Lin, M. Baker, R. J. Charlson, *Appl. Optics* **12**, 1356 (1973).
10. R. E. Weiss, A. P. Waggoner, R. J. Charlson, D. L. Thorsell, J. S. Hall, L. A. Riley, *Proceedings of the Conference on Carbonaceous Particles in the Atmosphere (CONF-7803101)*, Berkeley, Calif., 1979, pp. 257-262.
11. R. E. Weiss, thesis, University of Washington (1980).
12. M. Sadler, R. J. Charlson, H. Rosen, T. Novakov, *Atmos. Environ.*, in press.
13. N. H. Farlow, personal communication.
14. R. L. Chuan, personal communication. Size distributions were averaged over the period of sampling with the opal glass impactor.
15. These Mie computations of  $\sigma_{\text{sp}}$  from Ensor and Pilat (16) assume homogeneous spheres of  $\text{H}_2\text{SO}_4$ , a specific gravity of 1.8, and refractive index of  $1.5 - 0i$ , for a wavelength of 550 nm. Additional support for these calculations is provided by satellite measurements of stratospheric extinction, which were in the range  $10^{-5}$  to  $10^{-6} \text{ m}^{-1}$  at a wavelength of  $1 \mu\text{m}$  (M. P. McCormick, personal communication).
16. D. S. Ensor and M. J. Pilat, *J. Air Pollut. Control Assoc.* **21**, 496 (1971).
17. We thank N. C. Ahlquist for designing and building the optical measurement system. Supported in part by the NASA aerosol project under grand NSG-2362 and by NSF grant DPT 79-20857.

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## Measurements of the Imaginary Part of the Refractive Index Between 300 and 700 Nanometers for Mount St. Helens Ash

**Abstract.** *The absorption properties, expressed as a wavelength-dependent imaginary index of refraction, of the Mount St. Helens ash from the 18 May 1980 eruption were measured between 300 and 700 nanometers by diffuse reflectance techniques. The measurements were made for both surface and stratospheric samples. The stratospheric samples show imaginary index values that decrease from approximately 0.01 to 0.02 at 300 nanometers to about 0.0015 at 700 nanometers. The surface samples show less wavelength variation in imaginary refractive index over this spectral range.*

The 18 May 1980 eruption of Mount St. Helens injected material into the stratosphere to an altitude of approximately 20 km, and the resulting stratospheric plume has covered major portions of the Northern Hemisphere (1). The possible effects on climate of this or of other volcanic injection of material into the stratosphere are determined by the radiative properties of the stratospheric aerosol.

In general these aerosol particles interact with solar and with thermal infrared radiation fields to produce temperature changes, both at the surface and at different levels in the atmosphere. The radiative effects of the aerosol depend on the optical constants of the aerosol and may be quite different for aerosols with differing optical constants.

These stratospheric aerosols are of two general types: a silicate ash produced directly at the time of the explosion and a sulfuric acid-water sulfate aerosol produced as a result of gas-

to-particle conversion processes that occur in the volcanic plume. The silicate aerosol has its greatest relative importance in the months immediately after an eruption, while the sulfate aerosol is of relatively greater importance in later months.

Quantitative models that describe the radiative or climatic effects of these aerosols have been discussed by several investigators (2-5). In these models, the optical effects are based on numerical calculations in which it is assumed that the particles are homogeneous spheres. Scattering coefficients for Mie's solution for scattering of electromagnetic waves (6) are related to the size and composition of the particles through a size parameter  $x = 2\pi r/\lambda$ , where  $r$  is the particle radius and  $\lambda$  is the wavelength of the incident radiation, and a complex index of refraction  $m = n_{\text{RE}} - i n_{\text{IM}}$ , where the real part  $n_{\text{RE}}$  is the ratio of the speed of light in a vacuum to its speed in the particle, and the imaginary part  $n_{\text{IM}}$  is an ab-

sorption parameter that is related to the usual Bouguer-Lambert absorption coefficient  $k$  by means of the equation

$$n_{\text{IM}} = k\lambda/4\pi \quad (1)$$

At visible wavelengths, both scattering and absorption are significant, as are details of the way in which solar energy is redistributed in the atmosphere because of particle scattering. At infrared wavelengths the optical effects are dominated by the absorption and emission characteristics of the aerosol, and the scattering behavior is of less importance.

The incident solar radiation that is scattered by the particles in the hemisphere away from the earth's surface may, as a first approximation, be considered to be lost to space. This effect acts to increase the planetary albedo, while the portion of the light scattered in the downward direction can still enter into the radiation balance of the earth-atmosphere system below the aerosol layer. The incident radiation that is absorbed in the layer by the particles also acts to decrease the planetary albedo. The relative contributions of each of the competing effects of the visible radiation lost to space by scattering and that retained by particle absorption have important consequences for the radiative effects of these particles.

Calculations of Coakley and Grams (2) suggest that particles in the size range of 0.1 to  $1 \mu\text{m}$  have a net cooling effect on the earth's surface; the cooling effects due to the interaction of the aerosol with solar radiation are of greater magnitude than the heating due to infrared effects. The net cooling calculated depends on both the visible and the infrared optical constants of the aerosol as well as the size of the particles. Data of Pollack *et al.* (5) suggest a net cooling for the sulfuric acid aerosol but, depending on the actual optical constants, either heating or cooling is possible for the silicate aerosol.

In addition, calculations of rates of in situ heating induced by stratospheric aerosol particles are extremely sensitive to  $n_{\text{IM}}$  values at visible wavelengths. Fiocco, Grams, and Mugnai (4) calculated stratospheric heating rates as a function of latitude and of the albedo of the underlying earth-atmosphere system for four different models of the spectral variation in the complex index of refraction. They found that the rates of stratospheric heating induced by the aerosol layer are most sensitive to changes in the visible values of  $n_{\text{IM}}$ .

Both the visible and the infrared optical constants of the sulfuric acid-water