the large eruption cloud (17 April) and in the light plume (15 May); ΔC is the mass concentration of particles in the size interval $\Delta \log D$, where D is the geometric mean diameter of particles collected in a given impaction stage of the QCM. Both plots have strong bimodal features, with maximum concentrations at diameters of 1.1 and 0.23 μm. A size-distribution plot for the ambient air obtained from measurements on 11 April just south of Barbados is shown for comparison. Bimodal and trimodal size-distributions were observed in the plumes from Fuego and Santiaguito in 1978 (*6*); however, these plumes contained particles with diameters larger than 25 μm. We did not find these large particles in the Soufriere plume on 17 April, probably because our sampling position was at the extreme edge of the cloud. On 15 May, the cloud consisted of very thin white puffs that probably did not contain large particles.

We carried out EDXRA on samples from the 17 April and 15 May collections. The elemental composition of the material by size (impaction stage) was similar for both sets of samples. Aluminum and silicon were abundant among the particles that ranged from 1.1 μm to several micrometers in size. Traces of sodium, magnesium, chlorine, potassium, calcium, and iron were also found in the large-particle mode. Sulfur, probably in the form of dilute sulfuric acid because of its liquid appearance and low volatility, was present in the submicrometer range and usually covered the particles. This liquid sulfuric acid was much more abundant in the 15 May sample, which was collected in the steamlike puffs near the crater. This sulfuric acid is shown covering the particles in the SEM photograph (Fig. 2). In addition to sulfur, iron was moderately abundant among the submicrometer-sized particles in samples from both 17 April and 15 May; only very small traces of aluminum, magnesium, and potassium were present. No measurable amount of silicon was detected in the submicrometer-sized particles. The quartz crystal on which the particles were collected has a strong silicon peak in the x-ray spectrum; therefore, very small quantities of silicon in the sample may be undetected when the background spectrum is subtracted. In comparison to samples collected in the eruption plume from Santiaguito in 1978 (*6*), the Soufriere samples were relatively simple in morphology. The submicrometer-sized particles in the Santiaguito samples were more crystalline-like. Chemically, the samples were similar except for the abundance of iron among the submicrometer-sized particles in the Soufriere samples.

Fig. 2. Scanning electron microscope photograph of particles collected on stage 8 (0.28 μm; 50 percent impaction efficiency) of the QCM impactor from the light plume of 15 May 1979. The particles are surrounded by a wet-appearing substance containing sulfur, presumed to be sulfuric acid because of its low volatility.

Small particles from volcanic eruptions, such as the submicrometer-sized particles found in the Soufriere plume, can be injected into the stratosphere. We observed submicrometer-sized sulfate particles in the stratospheric plume from the Mount St. Helens eruption (*7*). These small particles can, because of their size and optical characteristics, effectively interact with solar and terrestrial radiation. Therefore, a global distribution of such particles in the stratosphere may produce climatic effects.

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Aerosols from the Soufriere Eruption Plume of 17 April 1979

Abstract. *Aerosol samples collected from the 17 April 1979 eruption plume of Soufriere, St. Vincent, at altitudes between 1.8 and 5.5 kilometers were physically and chemically very similar to the ash that fell on the island. Higher altitude samples (7.3 and 9.5 kilometers) had a much lower ash content but comparable concentrations of sulfate, which were above the background concentration found at these altitudes.*

Within 24 hours of the largest explosion of the 1979 eruption of Soufriere, that of 1657 LCT (*1*) on 17 April, an Air Force KC-135 aircraft collected filter samples and whole-air samples from the eruption plume. Scanning electron microscopy (SEM) of representative pieces of the filters was used to determine the grain size distributions and particle morphology. Nondispersive x-ray analysis provided qualitative data on the particle compositions.

Portions of the filters were leached with ultrapure water to remove soluble material. We analyzed these extracts for soluble SO_4^{2-}, NH_4^+, and total soluble nitrogen ions (ΣN), using Autoanalyzer and colorimetric techniques. The residual filters from the leaching were ashed for 24 hours at 400°C in a covered crucible to destroy the filters. The nonsoluble, nonvolatile residual ash was weighed to obtain an estimate of the mass of ash present for comparison with the results of neutron activation analysis (NAA).

We analyzed representative pieces of the filters for trace elements by NAA. The filters, along with appropriate standards, were irradiated with neutrons in the National Bureau of Standards (NBS) reactor. The induced radioactivity was measured by Ge(Li) γ-ray detectors according to the procedures used by Germani *et al.* (*2*) for the analysis of NBS standard reference materials. The ash sample was analyzed by neutron-capture prompt γ-ray activation analysis to determine the major elemental composition and some of the minor elements (*3*). The radioactive species ^{210}Pb and ^{210}Po were analyzed by α-spectroscopy on samples that were extracted from the air filters and electroplated before the radioactivity was counted.

So that we might compare the air-fall tephra with the tephra collected from the plume, we analyzed a sample collected

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Table 1. Plume sampling and particulate analysis data. The sample locations are as follows: sample 1, from approximately 220 km north of the island to the volcano (from 14°29′N, 61°3′W to 13°13′N, 61°1′W); sample 2, a 28-km path along the north side of St. Vincent (from 13°13′N, 61°1′W to 13°35′N, 61°21′W); sample 3, from a point approximately 115 km west of the island to a point 185 km east-southeast of the island (from 12°51′N, 63°7′W to 11°18′N, 58°18′N); and sample 4, from a point 185 km east-southeast of the island to a point 30 km west-northwest of the island (from 11°18′N, 58°18′W to 13°34′N, 61°40′W).

Sample	Elevation (km)	Size range (μm)	Average particle concentration per cubic meter*	Soluble			Ash†
				ΣN	NH_4^+	SO_4^{2-}	
				ppbm = nanograms per gram of air			
1	1.8 to 5.5	0.5 to 12	0.97×10^6	0.72	0.35	1.78	22 (25 ± 8)
2	1.8	0.5 to 8	1.19×10^6	1.98	1.03	4.83	74 (118 ± 19)
3	7.3	0.5 to 2	0.07×10^6	0.17	0.07	3.62	3.7
4	9.5	0.5 to 3	0.1×10^6	0.10	0.03	2.89	~ 0

*To be used for comparative purposes only; most of the particles could have been picked up over only a fraction of each traverse. †Based on gravimetric measurements of the ashed filter residue. The numbers in parantheses represent the mass of volcanic ash that should have been present on the basis of the observed Al, Ti, Mn, and Fe concentrations in the plume and the measured composition of the ash that fell on the island, on the assumption that air has a density of 1.185 kg/m^3.

on the ground at Belmont (17 April 1979), 9 km southeast of the summit of Soufriere, by SEM and NAA. The most common pyroclast type in this air-fall sample (85 percent of the particles) is andesitic pumice. The pumice pyroclasts are equant to slightly elongate, are irregularly shaped, and range in size from 300 to 1500 μm. The average pumice pyroclast particle consists of 30 percent colorless glass, 20 percent mineral phases (plagioclase and titanomagnetite), and about 50 percent vesicles. Vesicles are ovoid to highly irregular in form and range from ovoids that are 10 μm long to coalesced irregular channels as much as 200 μm long. The pumice pyroclasts have lumpy but relatively smooth outer surfaces; holes over collapsed vesicle walls have rounded edges. When broken, the thin angular vesicle walls form a rough surface. The remaining fraction (15 percent of the particles) of air-fall tephra consisted of individual mineral grains of plagioclase (44 to 80 mole percent anorthite), orthopyroxene (0.2 mole percent wollastonite, 76 to 66 mole percent enstatite, and 24 to 34 mole percent ferrosilite), clinopyroxene (average composition 42 mole percent wollastonite, 42 mole percent enstatite, and 16 mole percent ferrosilite), and titanomagnetite.

The filter sample collected from the KC-135 aircraft at an altitude of 1.8 km contained 54 percent colorless pumiceous glass and 46 percent mineral phases (22 percent plagioclase, 11 percent clinopyroxene, 7 percent orthopyroxene, and 6 percent unidentified phases). The mineral phases are mainly angular spalls, broken from phenocrysts during the eruption. The particle sizes of the plume samples range from 0.5 to 12 μm in the sample from an altitude of 1.5 km; particle sizes ranging from 0.5 to 3 μm were observed in the higher altitude samples (Table 1). None of the glass particles in this material were large enough to show vesicularity; all appear to be fragments of vesicle walls.

Although a broad spectrum of particle sizes was produced in the eruption, only the finest grained fraction was carried higher than 10 km in the 17 April eruption column. Some of the coarser fraction, which is mineralogically similar to particles found in the 1.8-km sample, may have reached 10 km, as suggested by ground-based observers, but the flight path may have missed that part of the plume at the 9.5- and 7.3-km altitudes sampled. Gravitational settling of the larger particles, 10 to 20 μm, could not account for the difference, as the settling velocities are approximately 1 cm sec^{-1} and in the 24 hours since the eruption the particles could have settled only about 1 km (*4*).

Table 2. Chemical composition of filter samples from the Soufriere plume; SCM, standard cubic meter.

Element	Concentration (ng/SCM)		Ash
	Sample 1, altitude 1.8 to 5.5 km	Sample 2, altitude 1.8 km	
H			0.054%
B			14.1 ppm
Si			26.7%
Na		1,520	2.75%
Mg	480	2,540	2.6%
Al	3,360	15,400	9.6%
Cl		960	0.183%
K			0.52%
Ca			5.7%
Sc	0.53	2.51	
Ti	230	880	0.563%
V	3.9	18	0.027%
Mn	26	150	0.115%
Fe	1,130	6,340	5.65%
Co	0.71	4.0	
Zn		16	
As		< 60	
Se		< 0.15	
Br		~ 60	
Sb		0.61	
I		4.1	
Cs	0.084	0.38	
Ba		130	
Ce		12	
Eu	0.045	0.152	
Dy		1.1	
Yb	0.058	0.101	
Hf	0.058	0.41	
Ta		0.051	
Th/U	0.094/0.110	0.54/0.58	

Analysis of the aqueous filter extracts for ΣN and NH_4^+ revealed concentrations comparable with ambient concentrations observed at corresponding altitudes in the unperturbed atmosphere during flights of the Department of Energy (DOE) WB-57F aircraft. (The data that we are comparing the results with come from DOE Project AirStream, which utilizes the WB-57F aircraft to fly three flight profiles per year from high latitude to the equator. These data are archived at the Los Alamos Scientific Laboratory.) The SO_4^{2-} concentration increased by a factor of 4 to 5 over background samples collected at altitudes comparable to those of samples 3 and 4 during the flight to and from the vicinity of St. Vincent. Since the SO_4^{2-} concentration increased significantly although the NH_4^+ concentrations remained at ambient levels for the two higher altitude samples, we believe that these samples were collected in a fresh volcanic plume with little entrainment of air from lower altitudes during plume rise and little time for neutralization of sulfuric acid aerosols by absorption of tropospheric ammonia vapors. The presence of considerable volcanic ash together with SO_4^{2-} in the two lower altitude samples would seem to suggest that these samples were also from volcanic debris of fairly recent origin, even though the ratio of SO_4^{2-} to NH_4^+ is greater at the lower altitudes. The most interesting feature of these data is the increase in the ratio of SO_4^{2-} to ash as a function of sample altitude. This increase could be due to a decrease in the quantity of ash present because of a gravitational separation of the gas and ash plumes or to the inclusion in samples at the higher altitudes of a portion of the plume that contained mostly SO_4^{2-}.

Table 2 gives NAA results for 23 elements measured in the air filter samples. The most interesting part of the trace-element analysis is data on enrichments or depletions of particular elements relative to their abundances in the earth's crust. Because of the high chemical blanks in the IPC filters used, only a few elements could be observed at concentrations higher than those in the filters.

Only sample 2 (1.8 km) had a significant quantity of ash or detectable elements above the blank values, whereas other samples yielded data only for elements of marine origin. The relative composition of both the 1.8-km and the 1.5- to 5.5-km filter samples is the same for all of the elements measured except for some rare-earth elements, which have relatively large analytical errors. So that we might better interpret these data, we calculated enrichment factors with respect to Al, relative to Taylor's (5) crustal values, as follows:

$$EF_{\text{crust}} = \frac{(X/Al)_{\text{air}}}{(X/Al)_{\text{crust}}}$$

where X and Al refer to the concentrations of the trace element of interest and of Al, respectively. Elements with enrichment factors close to unity are not strongly enriched relative to the earth's crust. Enrichment factors up to about 5 are probably due only to variations in the mineral makeup of the source material, in this case, the volcanic ejecta. Figure 1 shows the enrichment factors for the elements determined in the plume samples. Very few elements are enriched, and a few appear to be depleted. The lack of enrichment for the heavy elements may reflect the depletion of these elements in the magma of this volcano.

In contrast, studies of other volcanoes show that eruption plumes and fumaroles have very large enrichments (6), as much as 10^5 for the volatile elements Se, As, Sb, Cl, and Br. For Soufriere, the highest enrichments are factors of only 20 to 40 for Sb and Cl, with only detection limits for Se and As. These results indicate that the aerosols sampled by the aircraft were predominantly of ash dispersed into the atmosphere, with little or no chemical fractionation taking place. This same result was observed for the plume from the eruption of Mount St. Helens on 18 May 1980 (7). Plumes formed by phreatic or magmatic-phreatic eruption may be expected to have major contributions of airborne ash and only minor amounts of volatile materials. On the other hand, plumes formed by truly magmatic eruptions, with little or no involvement of steam, might be expected to have a much larger quantity of volatile elements and therefore higher enrichments.

When one calculates the mass of ash from the trace-element measurements in the plume and the ash composition, the results are not totally in agreement with those obtained by ashing the filters and weighing the residue. This inconsistency is due either to inhomogeneities in the filter portions used or to a loss of material during the ashing procedure. In general, the composition of the plume-particulate sample is similar to that of the ash that fell on the island, but chemical differences due to some fractionation in the settling process changed the relative elemental composition slightly.

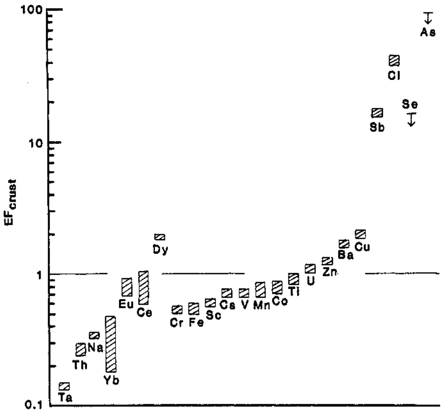

Fig. 1. Crustal enrichment factors for plume samples from the 17 April 1979 eruption of Soufriere, St. Vincent.

Analysis for the radioactive mother/daughter pair ^{210}Po and ^{210}Pb also failed to reveal an enrichment in the more volatile Po ($^{210}Po/^{210}Pb = 0.25$). Secular equilibrium would dictate a ratio of 1.0, and earlier measurements of magmatic gases in volcano plumes revealed ratios much greater than 1.0 (8). For Soufriere, we must conclude that the eruption plume is depleted in volatile elements.

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Volcanic Gases in the April 1979 Soufriere Eruption

Abstract. *Six gas samples from the 17 April 1979 Soufriere eruption plume were analyzed for carbonyl sulfide, carbon disulfide, carbon monoxide, carbon dioxide, methane, nitrous oxide, fluorocarbon-11, fluorocarbon-12, methyl chloroform, and carbon tetrachloride. Only carbon monoxide, carbon dioxide, carbonyl sulfide, and carbon disulfide were found to have increased mixing ratios as compared with those in clean tropospheric air, but the increases were not sufficient to contribute greatly to the global budgets of these four components.*

Whole-air samples were collected on a KC-135 aircraft mission near Soufriere, St. Vincent, on 18 April 1979, less than 24 hours after the 17 April eruption, which had been preceded by eruptions on 13 and 14 April. These samples have been analyzed by several gas chromatography techniques for carbonyl sulfide (COS), carbon disulfide (CS_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), fluorocarbon-12 (F-12 or CF_2Cl_2), fluorocarbon-11 (F-11 or $CFCl_3$), methyl chloroform (CH_3CCl_3), and carbon tetrachloride (CCl_4).

Two simultaneous samples were collected at each of three altitudes: 1800, 7300, and 9450 m. Methods of sample collection have been described in (1). Table 1 gives the flight times, latitudes, longitudes, and altitudes for each sample, as well as the results of the analyses of the gases. The amount of each component is reported as a dimensionless mixing ratio. Samples 1 and 2 were collected in the densest part of the plume, whereas sample pairs 3, 4 and 5, 6 were collected at higher altitudes, not necessarily entirely within the plume.

A gas chromatograph–flame photometric detector system (2) was used in analyzing for COS and CS_2. A 0.8-liter aliquot was transferred cryogenically for each sulfur-compound analysis. The samples were analyzed for CO, CO_2, and CH_4 with a gas chromatograph–flame ionization detector system and for N_2O, CF_2Cl_2, $CFCl_3$, CH_3CCl_3, and CCl_4 with a gas chromatograph–electron capture detector system (3).

To determine which of the measured species are of volcanic or partly volcanic

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