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SMALL PARTICLES IN PLUMES OF MOUNT ST. HELENS

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**Abstract.** Particles in the size range of 0.1-25 micrometers were sampled by aircraft carrying a quartz crystal microbalance in the Mount St. Helens plume on three dates in August and September 1980. Two of the sampling dates represented 'typical' emissions of the volcano between plinian eruptions. One sampling flight was made 1-4 hours before the small plinian eruption of August 7, 1980, when the plume had become discontinuous and visibly darker. Size distributions were determined, and individual particles were studied by using scanning electron microscopy. The plume sampled on August 7, before the eruption, contained mainly approximately 2 micrometer diameter silicic glass particles, fragments of the Mount St. Helens magma. The 'typical' plumes sampled on September 22 and August 6 had much smaller concentrations of particles, trimodal size distributions with peaks at 10, 0.4, and 0.1 micrometers. The particles were largely nonsilicate and apparently represented Cu-Zn oxide (10 micrometer peak), Al sulfate, chloride and oxide(?), and sulfuric acid (smallest size peak). The characterization of small particles in the plume may give information about approaching activity and possibly about changing conditions in the subsurface magma body.

Introduction

Sampling of the particles in the plumes of Mount St. Helens on 3 days during the month of August and September 1980 was carried out by using the Quartz Crystal Cascade Microbalance (QCM) [Chuan, 1975]. This sampling device is capable of determining the size distribution of aerosol particles in the size range of less than 0.1-25  $\mu\text{m}$ , by employing a cascade. It also allows direct SEM examination of each of 10 size fractions, because particles accumulate on extractable stages. The stages are piezoelectric quartz crystals, monitored electronically to give real-time size distributions. The QCM has been widely applied in studies of the atmosphere and has also been used before at active volcanoes [Rose et al., 1980; Chuan et al., 1981].

Results of study of the aerosol particles sampled by U-2 aircraft with the QCM from the May 1980 activity of Mount St. Helens documented the predominance of silicate ash and acid droplets within the 0.1-25  $\mu\text{m}$  diameter range [Chuan et al.,

1981]. Acid droplets less than 1  $\mu\text{m}$  made up the bulk of the stratospheric aerosol in the days following the May 18 eruption.

Sampling and Volcanic Activity

The plume of Mount St. Helens was sampled on August 6 and 7, using the regular U. S. Geological Survey gas sampling aircraft, and on September 22, 1980, as part of the RAVE project, using the NASA P-3 aircraft [Hart et al., 1981]. This was a period when the volcano was alternating between periods of heavy gas fuming and short-lived plinian eruptions. Samples taken on August 6 and September 22 were taken in typical heavy gas fuming periods according to all available data. Estimates of the  $\text{SO}_2$  flux of the plume on these dates are typical for the August - September 1980 periods (Table 1).

Visual observations also support our assessment of these plumes as typical of the August - September period. The plume sampled on August 7 between 1 and 3 PM was unusual in several respects: (1) It was darker in color, appearing to contain fine particles. (2) It was discontinuous in intensity, puffing at intervals of about 3 min.

Continuous harmonic tremor had begun at about 12:30 PM and the eruption of August 7 began at 4:27 PM. Thus the plume sampled on August 7 was immediately precursive to a small plinian eruption. Although dust from high winds and avalanche dispersal of surface material is sometimes abundant in the crater region, this was not the case on any of the three dates discussed here. We believe the contribution of surface dust to the plumes we sampled is minor.

Description of the Particles

The size distributions of particles sampled on August 6 and 7 and September 22 are shown in Figure 1. The precursive August 7 sampling had much higher total concentration, from 68 to 388 micrograms/ $\text{m}^3$  and a unimodal size distribution with a peak in the 2 micrometer size fraction. We examined the particles using the scanning electron microscope (Figure 2). The particles were largely silicate and have the morphology and spectral signature of silicic glass, the dominant component of the St. Helens dacite magma. They are similar to other volcanic eruption cloud particles, from Mount St. Helens [Chuan et al., 1981] and other explosive volcanoes [Rose et al., 1980].

Size distributions of the September 22 and August 6 plume particles differ sharply from the

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TABLE 1. Measurements of the Flux of the Gaseous Plume of Mount St. Helens on Selected Dates

Date	SO <sub>2</sub> t/d*	CO <sub>2</sub> t/d*	CO <sub>2</sub> /SO <sub>2</sub>
Aug. 6, 1980†	780	3700	4.7
Aug. 7, 1980†	870	3000	3.4
Sept. 22, 1980‡	1300	nd	-
Sept. 22, 1980‡	940	nd	-
Sept. 22, 1980†	1450	7500	5.2
Average Aug.-Sept.	1150	6000	5.2

nd = not determined

† United States Geological Survey flights: Casadevall et al. [1981]; Harris et al. [1981].

‡ Hart et al. [1981] data from R.E. Stoiber.

\*t/d = metric tonnes per day

August 7 sample (Figure 1). The August 6 sampling is trimodal with peaks at 10, 3, and 0.1 micrometers. The size distribution measured on September 22 is affected by problems related to moisture condensing and evaporating on the stages, so we could not directly measure the masses of particles larger than about 5 micrometers. A peak in the mass loading occurred near 0.1 micrometer diameter with the suggestion of a second mode be-

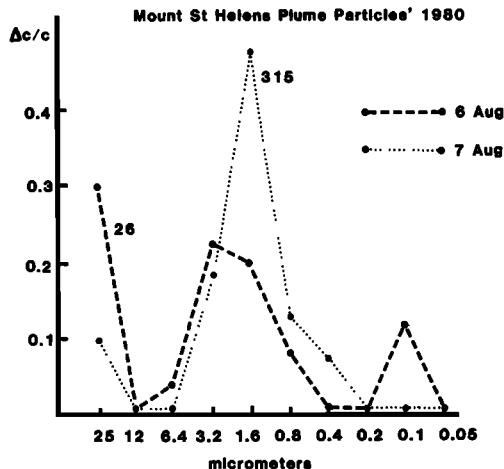


Fig. 1. Typical size distributions of particles in the size range of 25-0.1 micrometers for Mount St. Helens volcanic plume of August 6 and 7. The total concentrations of particles are listed in the figure as well. Size distributions are obtained in flight by the QCM instrument [Chuan, 1975] and assume an average density of about 2.0 g/cc. The September 22, 1980, size distribution could not be obtained electronically in the >5 micrometer stages, but we know that the <5 micrometer size distribution was similar to August 6, and post-flight examination shows that there was a particle size peak in the 10 micrometer range. Total concentrations of particles on September 22 were 24-53 micrograms/m<sup>3</sup>, also similar to August 6 measurements.

ginning at about 3 micrometers. Data from the QCM stages corresponding to larger particles were ambiguous, but based on post-flight examination of the stages, a third peak in the size distribution occurred in the 10 micrometer size range. Thus the size distributions for September 22 were similar to those of August 6. The total particle concentrations are much less, ranging from 24 to 53 micrograms/m<sup>3</sup>. SEM examination (Figure 3) shows that the chemistry and morphology of these particles is strikingly different from the August 7 sample. This conclusion is based on the September 22 sampling, from which stages 3 (6 micrometers), 5 (1.6 micrometers), 7 (0.4 micrometers), and 9 (0.1 micrometers) were examined in detail. Silicate particles were only of minor significance. The smallest particles (Figure 3A) contain minor Mg and Al but consist mostly of sulfuric acid. We have previously documented [Rose et al., 1980] the abundance of sulfuric acid droplets in the submicron size fractions of particles on volcanic clouds. Larger particles in the September 22 plume sampling are different from volcanic eruption clouds, because they are largely non-silicate and contain much Cu, Zn, Al, S, and sometimes Cl (Figure 3). Based on energy dispersive analysis with the SEM, we believe the particles represent Cu-Zn oxides, Al-sulfates, and chlorides along with a few silicate particles, all coated with variable amounts of sulfuric acid. The Cu-Zn particles are particularly abundant in the larger size fraction. These always contain Cu and Zn in a 2-1 atomic abundance (Table 2,b). An oxide with this Cu:Zn proportion is unknown in nature and in the laboratory. Also in Table 2 are analytical data on zincite (ZnO) and tenorite (CuO), which have been identified before at volcanoes.

TABLE 2. Chemistry of Cu and Zn Oxides in Volcanic Aerosol

	a*	b†	c††
ZnO	80.4	33.9	-
Fe <sub>2</sub> O <sub>3</sub>	1.9	-	tr
CuO	1.4	59.5	100.0
CdO	6.0	-	-
SiO <sub>2</sub>	10.2	-	tr
Al <sub>2</sub> O <sub>3</sub>	-	1.3	tr
SO <sub>3</sub>	-	5.3	-

All analyses recalculated to 100%; energy dispersive X-ray analysis; tr refers to less than 0.2%.

\*Zincite, phreatic ash of Mount St. Helens [Thomas et al., 1981].

†Cu-Zn oxide, September 22, 1981, plume of Mount St. Helens.

††Tenorite, from Harter fumarole, Cerro Negro Volcano, Nicaragua, 1969. (M. A. Malinconico, personal communication, 1979).

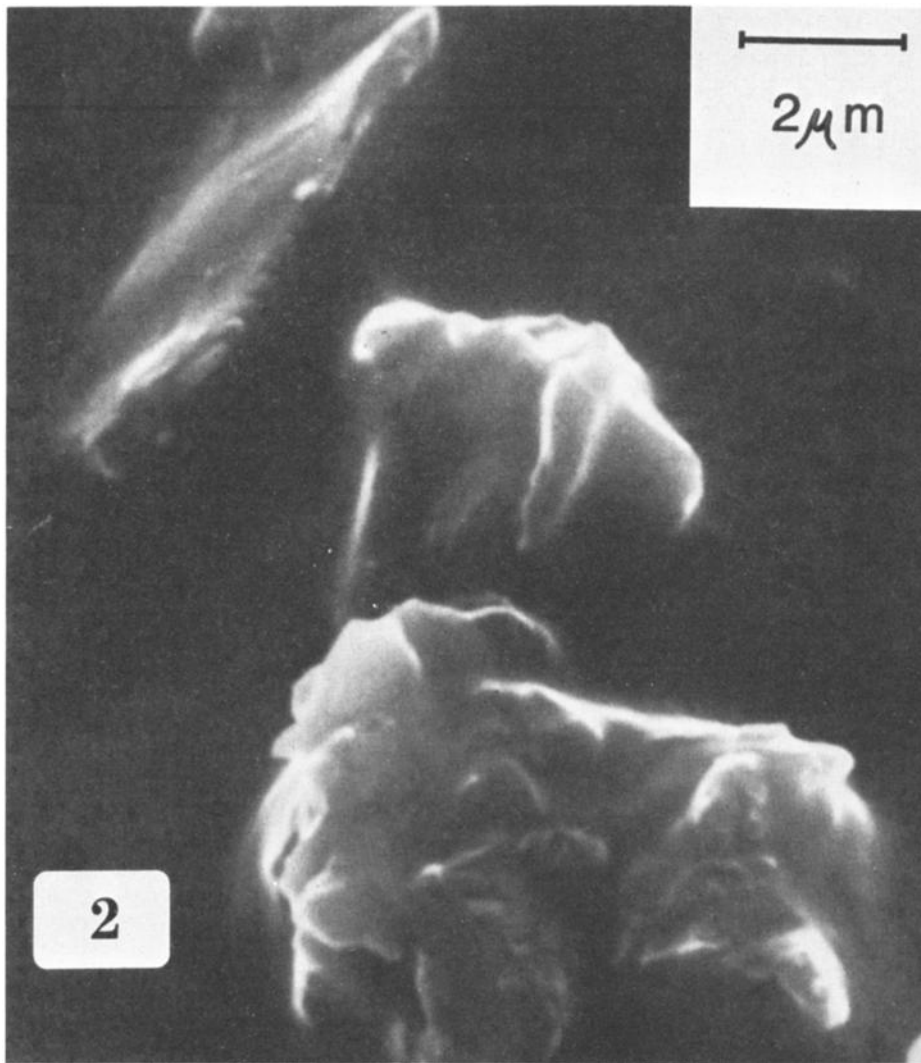


Fig. 2. Scanning electron micrograph of ash particles in the August 7, 1980, sample, from stage 5 (1.6 micrometers) of the QCM instrument. This is the stage which sampled the largest mass of particles in the unimodal size distribution. Energy dispersive X-ray analysis of these particles and comparison with standard patterns demonstrates that they have the spectral pattern of silica-rich glass, with Si, Al, K, Na, and Ca peaks in the correct proportions. The particles also show the angular 'shard' morphology of silicic tephra [Rose et al., 1980]. A vesicular glass with 72% SiO<sub>2</sub> is the dominant component of the St. Helens tephra of 1980.

#### Discussion and Interpretations

Our results emphasize the differences between the particles found in two types of volcanic plumes. One type, termed 'typical' which occurs during periods of relative inactivity, has far less particulate matter. The other 'atypical' type, which occurs during active eruption periods, had abundant particles, which represent pieces of finely divided magma and lithic material from the vent. The typical volcanic plume is more analogous to volcanic fume, while the atypical one is an ash eruption cloud. Oskarsson [1980] has summarized some other relevant geochemical processes that occur in volcanic plumes. At temperatures near that of the magma, gaseous compounds react

and some tiny salt particles are formed that are carried along with the plume [see also Naughton et al., 1974]. At lower temperatures, absorption of some gaseous compounds on silicate and salt particles occur. At temperatures of 300°C and below sulfuric acid droplets begin to form, and these, being hygroscopic, dissolve H<sub>2</sub>O and other components from the gas [see also Rose, 1977]. In precursive and active plumes, silicate fragments may dominate in the particle populations [Rose et al., 1980]. In typical plumes the salt and acid particles dominate although the total concentration of particles is less.

The August 6 and September 22 plumes of Mount St. Helens represent typical volcanic plumes. The August 7 plume had the characteristics of an active

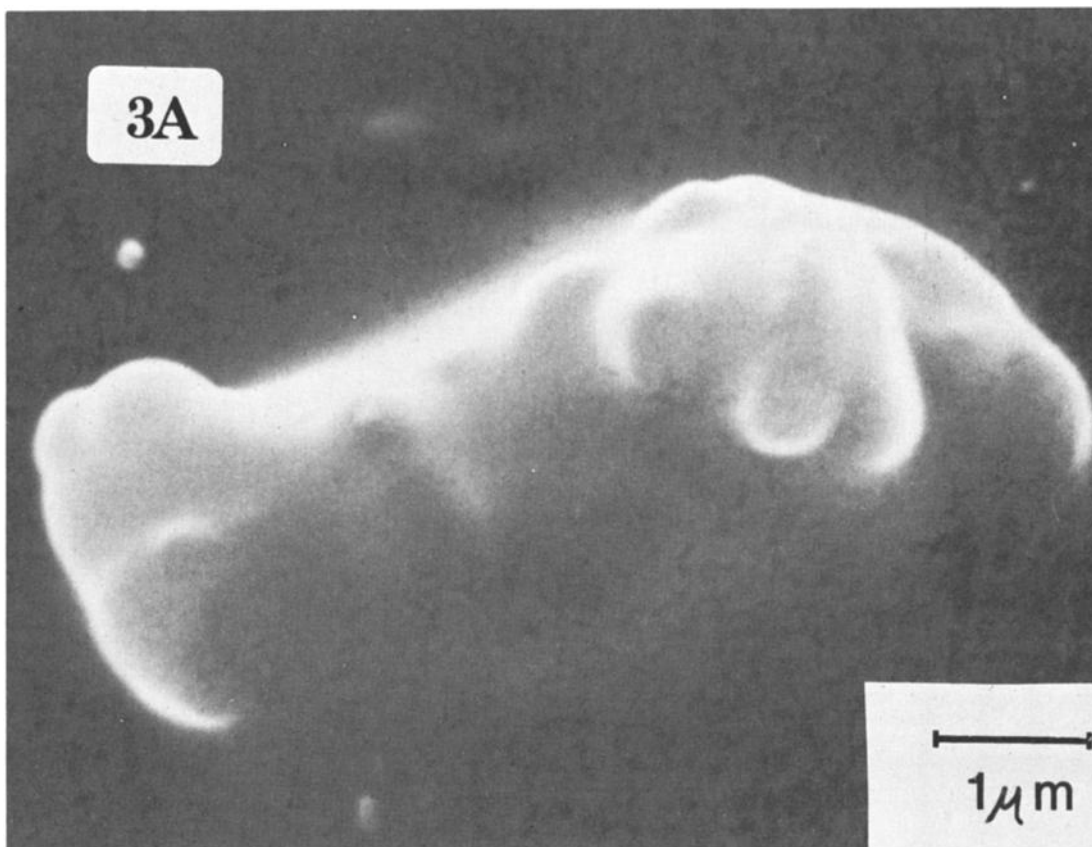


Fig. 3a

Fig. 3. Scanning electron micrographs of selected particles representative of the September 22, 1980, sampling of the Mount St. Helens volcanic plume. (A) One of the largest particles on stage 9 (0.1 micrometers) of the QCM instrument. The large size of the particle on this stage may be explained by its low density and/or a composite nature. The energy dispersive X-ray pattern for this particle is dominated by the sulfur elemental peak. The only other elements detected are minor amounts of Mg and Al. The morphology of the particle is globular, suggesting that it has a thick coating of liquid. We interpret it to be largely sulfuric acid, perhaps with a Mg-Al sulfate core. The spectral pattern for all of stage 9 is similar to this particle and this suggests that sulfuric acid is the dominant type of material in this size range. (B) A particle from stage 7 (0.4 micrometer) of the QCM instrument. This irregularly shaped particle has droplets on its surface and on the stage surrounding it. The energy dispersive X-ray pattern of this grain shows major amounts of Al and S and minor Zn, Cu, and Cl. We interpret the particle to be aluminum sulfate (hydrated?). It is representative of many Al-rich particles found especially in the intermediate stages of the QCM instrument. (C) Typical particles from stage 3 (6 micrometers) of the QCM instrument. These have a flaky morphology and obvious surface particles (both droplets and tiny crystals). The energy dispersive analysis of these shows major Cu and Zn, in 2-1 elemental proportion, and minor S and Al. We interpret them to be a Cu-Zn oxide with coating of sulfuric acid and Al-salts. Such particles dominate the large size fractions of the September 22, 1980, QCM samples. A quantitative estimate of the composition of the particle at top is given in Table 2, column b.

cloud. This is significant since sampling occurred 3 hours before a major eruption. It shows that particle sampling can identify a precursive eruption cloud.

Interpretation of the typical plume particles is aided by studies of volcanic fume [Naughton et al., 1974] and fumarolic incrustations [Stoiber and Rose, 1974; Oskarsson, 1981]. Table 3 gives a list of nonsilicate solid phases that have been suggested to be in volcanic fume or

volcanic clouds by various authors. Many species are listed.

We are sure this list is not complete. The formation of these compounds is not well understood. Even so, we are beginning to understand some of the environmental characteristics that may dictate which of these many phases may be found in a particular plume. During September 1980 the passive plumes at Mount St. Helens Cu-Zn oxide and Al-salts (mostly sulfate) as well as sulfuric

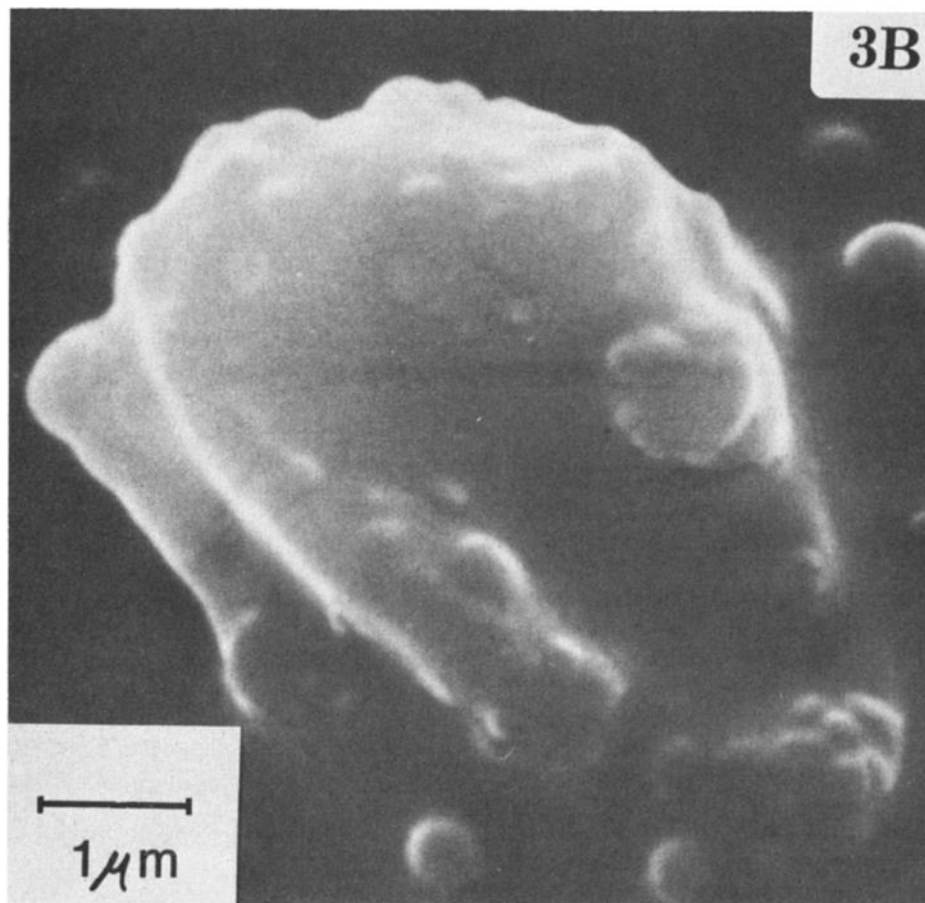


Fig. 3b

acid seem to be the dominant nonsilicate particles. For other volcanoes different solid phases have been dominant. In the eruption clouds of Fuego and Cerro Negro,  $\text{CaSO}_4$  and  $\text{NaCl}$  were important phases [Rose et al., 1973]. At Erebus, Al-salts are very important to the fume [Keys, 1980]. In the early phreatic eruptions of Mount St. Helens, Thomas et al. [1981] reported  $\text{ZnO}$  particles (Table 2,a). Stoiber and Rose [1974] reported that certain volcanoes have characteristic minor element enrichments in their fumarolic deposits, which is a reflection of the volcano fume content.

Mizutani [1970] was one of the first to report the relation of trace elements in fumarolic gases to the degree of oxidation. Oskarsson [1981] further emphasized the effect of degree of oxidation of the gases and showed how the dominance of either HF or  $\text{CHl}$  in the halide fraction of the gas might affect the salts formed.

What conditions explain the Mount St. Helens particles? There is a suggestion that the early gaseous emissions of Mount St. Helens contained more  $\text{H}_2\text{S}$  than  $\text{SO}_2$  and that  $\text{SO}_2$  became the dominant sulfur species in the plume after May 1980 [Hobbs et al., 1981]. This is plausible, because thermodynamically the T- $\text{PO}_2$  conditions of the Mount St. Helens magma are near the cross-over from  $\text{H}_2\text{S}$  to  $\text{SO}_2$  dominance [Gerlach and Nordlie,

1975]. A slight change in temperature or  $f\text{O}_2$  could effect a change in the dominant sulfur gas. Krauskopf [1964] has shown that changes in  $\text{PO}_2$  in magma causes large increases in the volatility of Cu and Zn as chlorides. Thus increasing Cu and Zn abundance is likely in gases from more oxidized magma. The Cu and Zn may be volatilized from the magma as a chloride vapor, which cools and reacts with the atmosphere to form oxide. Although the volatility of both elements are very sensitive to a change in oxidation, Cu is much more sensitive [Krauskopf, 1964], and this may explain why tiny amounts of pure Zn-oxide is found in early Mount St. Helens eruption clouds, while later clouds contain larger amounts of Cu-Zn oxide.

#### Conclusions

Significant differences in the size distribution and compositions of small (less than 25 micrometer) particles in the Mount St. Helens volcanic plumes have been measured. Passive plumes show trimodal particle size distributions with peaks at 10, 3, and 0.1 micrometer, low total particle concentrations, and only rare silicate particles. Active and precursive plumes show higher total concentrations, a predominance of silicate particles and unimodal size distributions, with peaks at 2 micrometers. The data

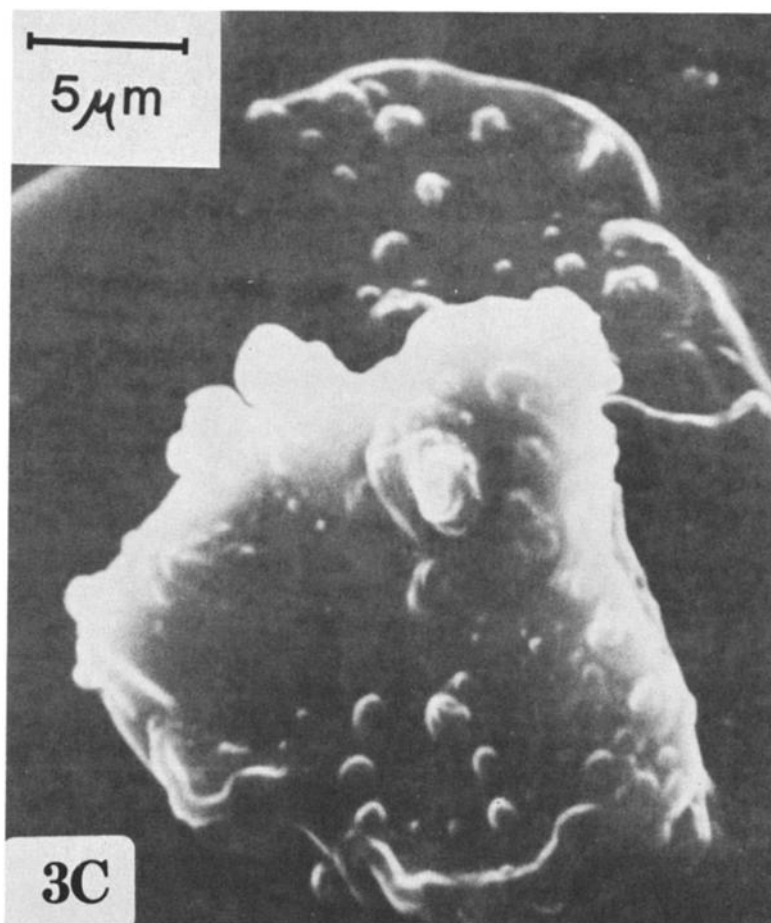


Fig. 3c

TABLE 3. List of Nonsilicate Solid and Liquid Phases Likely, Inferred or Identified in Volcanic Plumes

CaSO <sub>4</sub> (n H <sub>2</sub> O?)*	CaF <sub>2</sub>	H <sub>2</sub> SO <sub>4</sub> *
NaCl	CaSiF <sub>6</sub>	H <sub>2</sub> O*
KCl	NaF	ZnO*
AlF <sub>3</sub>	CaCl <sub>2</sub>	S*
AlCl <sub>3</sub> (n H <sub>2</sub> O?)*	CaSiF <sub>6</sub>	Na <sub>2</sub> SO <sub>4</sub> *
Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> (n H <sub>2</sub> O?)*	MgCl <sub>2</sub>	K <sub>3</sub> Na (SO <sub>4</sub> ) <sub>2</sub>
CuO*	MgSO <sub>4</sub>	NH <sub>4</sub> Cl
Al <sub>2</sub> O <sub>3</sub>	FeCl <sub>3</sub> (n H <sub>2</sub> O?)*	(NH <sub>4</sub> ) <sub>2</sub> SiF <sub>6</sub>
Na <sub>2</sub> SiF <sub>6</sub>	Na <sub>6</sub> ClF(SO <sub>4</sub> ) <sub>2</sub>	NaAl <sub>4</sub> O <sub>4</sub> Cl <sub>5</sub>
NaMgAl(F,OH)H <sub>2</sub> O*	Cu <sub>2</sub> ZnO <sub>3</sub> (?)*	

Sources: Rose [1977]; Naughton et al. [1974]; Stoiber and Rose [1974]; Rose et al. [1980]; Oskarsson [1980, 1981]; Thomas et al. [1981]; Keys [1980].

\*Directly observed or strongly inferred.

suggest that particle monitoring in plumes can provide warning of impending activity.

The list of species of nonsilicate particles described or inferred in volcanic clouds is long,

and getting longer. In August - September 1980 the appearance of Cu-Zn oxide suggests an oxidizing magma body at Mount St. Helens. Continued monitoring of particles may show significant changes in the magma body and gaseous emissions from it.

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