May the plume was well defined, and the water vapor mixing ratio in the plume was at least an order of magnitude above background concentrations.

Data obtained on the other three flights through the volcanic plume with the frost point hygrometer show larger variations in water vapor mixing ratios than observed during the 15 May flight, but the higher mixing ratios are generally only a factor of 2 or so above background. These increases are not as easily correlated with the time when the aircraft was in the plume. On the 27 May flight the range in mixing ratio by mass was from 1.5×10^{-6} to 4.5×10^{-6}; the highest readings were obtained between 0200 and 0215, the time during which the pilot was in the area where the cloud was predicted to be. High readings were also observed around 0245 when the aircraft was over Washington state but thought to be out of the plume.

The data obtained during the 14 June flight also showed great variability in comparison with the 15 May flight. Water vapor mixing ratios by mass ranged from 1.5×10^{-6} to 5.2×10^{-6}. There seemed to be no extensive areas where the mixing ratio was higher than that in surrounding areas; rather the regions appeared to be patchy.

The data obtained during the 17 June flight show practically no increase in water vapor mixing ratio anywhere along the flight path except for one brief period around 2135. At that time the mixing ratio reached 6.0×10^{-6}. During the rest of the flight the mixing ratio was between 2.0×10^{-6} and 2.5×10^{-6}.

From the data obtained with the frost point hygrometer it is evident that a large amount of water vapor was injected into the stratosphere by the 18 May eruption. It is less evident that the later eruptions carried much water vapor into the stratosphere. During the flight of 22 May, the enhanced water vapor mixing ratios were well correlated with the plume, and the plume size determined by Danielson (*4*) can be used to estimate the total amount of water injected by the eruption. Danielson gives the volume of the plume as $\simeq 2 \times 10^6$ km^3. From an average mixing ratio in the plume of 2×10^{-5}, it appears that a total mass of 3.2×10^9 kg of water was injected by the eruption. The injected water could come either from the volcano or from entrained tropospheric air. If the only source of water is tropospheric air, with a mixing ratio of 2 g/m^3, it would require the entrainment of 1.6×10^3 km^3 of tropospheric air to account for the water vapor measured in the plume on 22 May. This appears to be too large a volume, and thus some of the water vapor probably had to come from the volcano. If the source is totally volcanic, 3.2×10^{-6} km of liquid water would be required which, in view of the size of the 18 May eruption, appears reasonable. The water vapor mixing ratios measured after the other eruptions were not enhanced enough to argue for major injections of water from the weaker eruptions.

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Measurements of Cloud Condensation Nuclei in the Stratosphere Around the Plume of Mount St. Helens

Abstract. *Measurements of cloud condensation nuclei were made from small samples of stratospheric air taken from a U-2 aircraft at altitudes ranging from 13 to 19 kilometers. The measured concentrations of nuclei both in and outside the plume from the May and June 1980 eruptions of Mount St. Helens were higher than expected, ranging from about 100 to about 1000 per cubic centimeter active at 1 percent supersaturation.*

The eruptions of Mount St. Helens in May and June 1980 injected significant amounts of gases and particles into the stratosphere. We measured cloud condensation nuclei (CCN), the part of the aerosol capable of nucleating water vapor condensation at supersaturations of the order of 1 percent (relative humidity of 101 percent).

Rather elaborate projections of the effects of volcanic aerosol on the earth's climate have been made in recent years, such as that of Pollack *et al.* (*1*). Whether stratospheric CCN are important in the total picture of global weather depends on (i) the numbers of CCN available in the stratosphere, (ii) the rate at which they enter the troposphere, and (iii) the way in which they affect weather systems in the troposphere. Our measurements relate to the first of these three aspects. The second may involve any of eight mechanisms described by Shapiro (*2*). Once in the troposphere, CCN of stratospheric origin could modify cloud microstructure, leading to two possible effects upon climate: alteration of precipitation processes and alteration of the scattering and absorption of solar radiation by clouds (*3*).

To the best of our knowledge, our CCN data are the first reported from altitudes above the local tropopause. An indication of the CCN count, however, can be gained from the measurements of Rosen and Hofmann (*4*) taken between 10 and 20 percent supersaturation before June 1980, and more recently at 200 percent supersaturation, in numerous balloon ascents over Laramie, Wyoming. These investigators reported evidence of both anthropogenic and volcanic increases in stratospheric sulfates (*5*). Our measurements are taken as a function of two to three supersaturations within the range of those found in actual clouds; if compared to tropospheric counts, they should help to resolve questions of whether or not the stratosphere can ever be a significant source of CCN.

We analyzed four 1-liter samples of stratospheric aerosol collected by a NASA U-2 aircraft. These samples, although well suited for their original purpose of trace gas analysis, presented a serious concern with respect to our CCN measurements. It was expected that losses due to Brownian diffusion to the walls might cause unacceptably rapid depletion of the CCN present in the small sample containers, which were stainless steel cylinders with rounded ends (radius, 5 cm; length, 20 cm). Laboratory simulations of the experiment with similar containers showed that the loss of CCN active at 1 percent supersaturation was a rather consistent 35 ± 5 percent per hour. (No attempt was made to establish stable thermal stratification of the container contents.)

The 1-liter sample containers were cleaned and evacuated before each flight and were opened by pilot activation of motor-driven valves at the specified horizontal and vertical coordinates. All samples were obtained from a sample entry system designed for gas analysis. Diffusion losses of CCN were probably neg-

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ligible, but bends and restrictions in the sample entry plumbing may have allowed impaction losses of some of the largest CCN. Both kinds of losses would cause our final results to be underestimates of the actual stratospheric concentrations of CCN. A range of 1.2 to 5.3 hours elapsed between the time the sample bottles were filled and their CCN counts were measured.

Sample 1 (Fig. 1A) was taken over western central Montana (46°18′N, 112°25′W) at an altitude of 13.6 km. Although trajectory analyses (*6*) place this sample slightly outside the northern boundary of the plume from the 13 June eruption, it is difficult to attribute the very high CCN count of this sample to a source other than the volcano. We suggest that the boundary of the plume injected by this eruption may be diffuse enough so that our sample 1 was actually volcanic material. Samples 2 (Fig. 1A; 38°00′N, 120°30′W) and 3 (Fig. 1B; 39°00′N, 106°30′W) were both taken between 18 and 19 km, over central California and central Colorado, respectively. Sample 2 was apparently taken within the widely scattered debris of the 18 May eruption, but trajectory analyses and other data are unable to confirm this. The coordinates of sample 2 were chosen to intercept the 18 May plume after it had passed once around the world. Sample 3 was taken within a portion of the 18 May plume, as judged by supporting lidar and aerosol data. Sample 4 (Fig. 1B; 37°50′N, 120°35′W), which was taken at an altitude of 13.6 km over central California, provides a CCN background count near, but above, the tropopause; volcanic aerosol was not involved.

A continuous-flow diffusion chamber (*7*) was used as the detection apparatus in all experiments. Sample containers were unloaded as soon as possible after each U-2 flight, and were immediately connected to the diffusion chamber. Precautions, including test runs on blank and mock-up samples, were taken to avoid contaminating the samples with room air. Each sample container was backfilled with particle-free air to bring its internal pressure up to the ambient value required for operation by the continuous-flow diffusion chamber; backfilling continued as each sample was withdrawn from its container through a stainless steel probe at the container's geometric center, at a flow rate of 1 cm^3 sec^{-1}. By varying the operating settings of the diffusion chamber, spectra of CCN active at various supersaturations were taken for each sample.

For the CCN spectra obtained from these four samples (Fig. 1), the primary source of error is the uncertainty in the aerosol depletion rate. The background count of the diffusion chamber and residual amounts of room air in the very short lengths of connecting tubing contributed a basic sensitivity level, or background count, conservatively estimated to be 20 to 30 CCN cm^{-3} or less. This background has been subtracted from the results shown in Fig. 1.

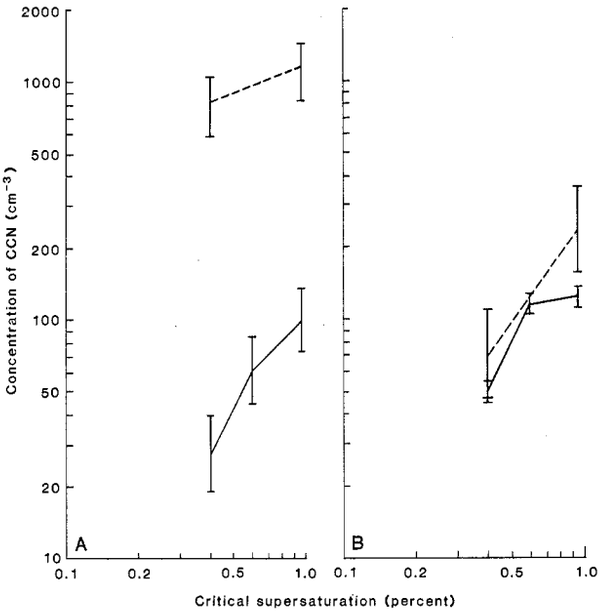

Fig. 1. (A) Measurements of CCN active at critical supersaturation from samples taken on 14 June 1980. The dashed line is sample 1 from 13.6 km; the solid line is sample 2 from 18.6 km. (B) Measurements of CCN active at critical supersaturation from samples taken on 17 June 1980. The dashed line is sample 3 from 18.8 km; the solid line is sample 4 from 13.6 km.

The results from samples 1 and 4, both taken at 13.6 km, suggest that volcanic eruptions may temporarily cause the CCN count near, but above, the tropopause to be in the range of 100 to 1000 cm^{-3} active at 1 percent supersaturation, a higher value than that measured in samples from below the tropopause (*8*). Samples 2 and 3, taken above the tropopause at 18.6 and 18.8 km, respectively, appear to be consistent with data of Rosen and Hofmann (*9*). The results of samples 2 and 3 differ qualitatively from those of sample 1 because they were taken in volcanic plumes whose aerosols have had a chance to age, and perhaps to coagulate. The CCN counts of samples 2 and 3 are about one order of magnitude above the counts obtained by Rosen and Hofmann (*4*) after the eruption of Volcan de Fuego in Guatemala in 1974. Our measurements are, however, about equal in magnitude to the tropospheric CCN counts thought typical of maritime conditions below cloud level by Twomey and Wojciechowski (*10*). Sample 4 represents what may be the background count at altitudes much lower than those of samples 2 and 3.

More measurements are needed to establish the normal CCN background count at these altitudes and the spatial and temporal dependence of the volcanically caused CCN count. Aerosol losses in the sample entry lines are only one mechanism that would cause our results to be an underestimate of true stratospheric CCN counts; further underestimation would result if some of the stratospheric CCN are sulfuric acid particles, which partially evaporate when the sample bottles are warmed as they are brought to the laboratory. Our results, higher by an order of magnitude than anticipated, suggest that volcanoes may be an important source of CCN in the lower stratosphere, both by direct injection of CCN and by contributing precursors for homogeneous nucleation of CCN.

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