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STRATOSPHERIC IMPACT OF EL CHICHON

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RESUMEN

Se colectaron aerosoles y gases estratosféricos mediante aviones de elevada altitud y balones para determinar el efecto de El Chichón sobre la estratosfera. El Chichón inyectó compuestos sulfurosos que resultaron en la producción de alrededor de 7.6 ± 2.3 gT de sulfatos en la estratosfera global. El hecho de que el muestreo por balón se hizo a una latitud única arroja considerable incertidumbre. No se encontraron cantidades importantes de sulfuro de carbonilo en la porción de la pluma estratosférica del volcán en las altitudes accesibles al aeroplano. Esta erupción tuvo sobre la estratosfera un impacto mayor que cualquier otra erupción volcánica desde 1971.

ABSTRACT

Stratospheric aerosols and gases were collected by high-altitude aircraft and balloons to assess the effect of El Chichón on the stratosphere. El Chichón injected sulfurous compounds that resulted in the production of about 7.6 ± 2.3 Tg sulfate in the global stratosphere. Considerable uncertainty arises from balloon sampling being available at only one latitude. No significant amounts of carbonyl sulfide were detected in portion of the stratospheric plume of the volcano which was at aircraft altitudes. This eruption had the largest impact on the stratosphere of any volcanic eruption since 1971.

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INTRODUCTION

The eruptions of El Chichón on 28 March, 3 April, and 4 April 1982 sent large amounts of sulfurous gases and dust into the stratosphere. The US Department of Energy's High Altitude Sampling Program (HASP) has measured the distribution of sulfate resulting from this eruption. We also measured a number of trace gases in the stratospheric plume of the eruption. We include, in this paper, concentrations of carbonyl sulfide as this has been shown to be an important precursor gas for the formation of stratospheric sulfate aerosols (Crutzen, 1976).

HASP consists of Project Airstream and Project Ashcan. Project Airstream samples the northern hemisphere stratosphere with a specially equipped WB-57F high-altitude aircraft. The aircraft samples from the equator to 75°N at four altitudes between 12.2 and 20.7 km. The sampling route (Fig. 1) is flown each April, July

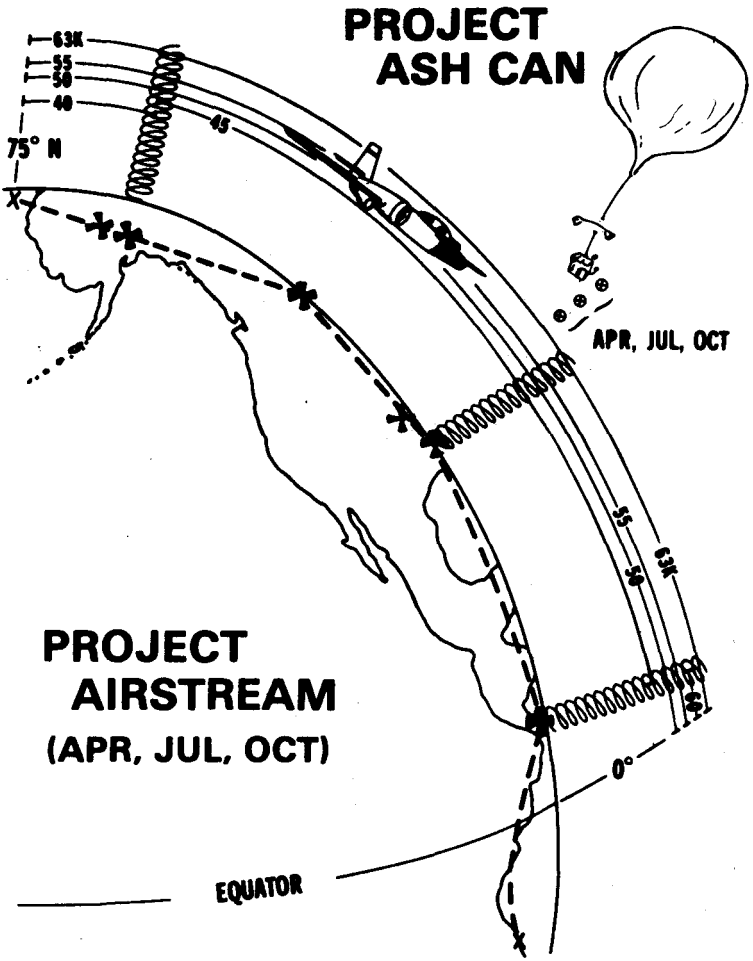

Fig. 1. HASP sampling routes and locations.

and October. Operating bases are located at Houston (Texas), Tacoma (Washington), Anchorage (Alaska), and Panama, Rep. of Panama. Honolulu (Hawaii) was used in place of Panama during a November, 1983 flight series. Vertical profiles (indicated by spirals) are flown over each location except Tacoma. Project Ashcan samples the stratosphere during the same periods with devices carried by helium-filled balloons. The balloons are launched from Holloman AFB, New Mexico (33°N, 106°W). Samples are collected at altitudes between 20 and 30 km. Several reports based on LIDAR observations (Hirono and Shibata, 1983; Labitzke *et al.*, 1983; Clemesha and Simonich, 1983) have shown that large amounts of aerosol were detected at about 25 km altitude, and thus accessible only to balloons.

Both the aircraft and balloons collect stratospheric aerosols on IPC 1478 filter paper. The aircraft uses ram air, whereas the balloons use a highspeed blower or an air ejector to move air through the filter paper. Trace gases are collected on the aircraft through a nose probe and compressed into stainless steel bottles (Leifer *et al.*, 1981). Trace gases are not collected aboard the balloon platforms.

Before sampling the filters are washed to reduce the sulfate blank levels (Gandrud and Lazrus, 1972). The exposed filters and blanks are analyzed for sulfate by colorimetric (Lazrus *et al.*, 1968) and ion chromatographic techniques. The compressed air samples are analyzed by gas chromatographic and mass spectrometric techniques (Rasmussen and Khalil, 1980) for a number of trace gases.

RESULTS

October-November 1981. Measurements made during this period reflect the condition of the stratosphere before the eruption of El Chichón. Most of the aerosol is residual debris from the eruptions of Alaid and Pagan that had occurred in the spring of 1981 (Sedlacek *et al.*, 1983). As shown in Fig. 2a, the sulfate is uniformly distributed throughout the stratosphere. Data from the fall Ashcan flights (Fig. 3) are consistent with the concentrations measured by the Airstream flights.

April-May 1982. Fig. 2b clearly shows that this Airstream cycle intercepted the stratospheric plume of El Chichón on 20-21 April at 19.2 km between the equator and about 25°N. Robock and Matson (1983) show that the leading edge of the main cloud was passing over Africa at this time. However, they also show that the trailing edge of the cloud was still in the vicinity of the Yucatan Peninsula and it is possible that this is the portion of the cloud that we sampled. It may also be possible that we intercepted the leading edge of the 28 March eruption; a smaller eruption than that on 4 April and possibly not tracked by Robock and Matson *(ibid.)*.

On 21 April 1982 the WB-57F collected three aerosol samples at 19.2 km between 7°N and 16°N that had sulfate mass mixing ratios of 127, 167, and 163

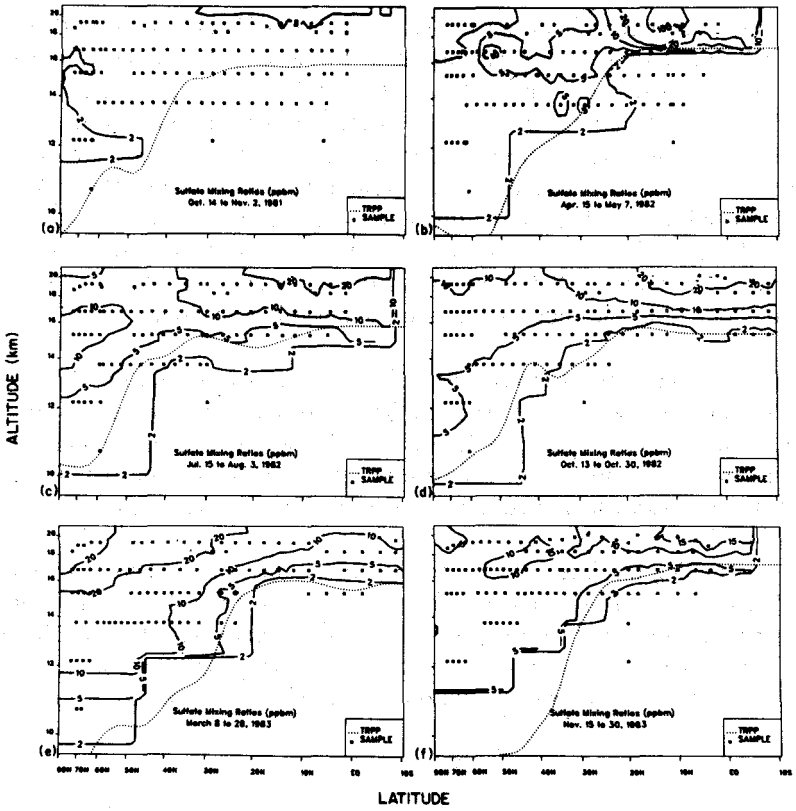

Fig. 2. Contour plots of Project Airstream sulfate mixing ratios for the periods indicated. TRPP denotes the calculated tropopause as discussed in the text.

ppbm. These extraordinarily high concentrations of sulfate (the highest ever measured by Project Airstream in over 13 years of stratospheric sampling) greatly exceeded the aerosol concentration simultaneously measured by a quartz crystal microbalance cascade impactor (QCM). This is only an apparent contradiction because the QCM is less than 50% efficient for particles smaller than 0.05 μm whereas the filter paper is greater than 99% efficient for particles as small as 0.03 μm diameter (Stern *et al.*, 1960; Institute of Paper Chemistry, 1960). We suspect that most of the sulfate on the filter paper consisted of particles generally smaller than 0.05 μm diameter. The implication is that the aircraft was sampling in a portion of the plume that was actively producing new sulfate aerosols. If so then the importance of COS as a sulfate aerosol precursor can be ascertained from trace gas measurements made simultaneously with the sulfate aerosol measurements. If El

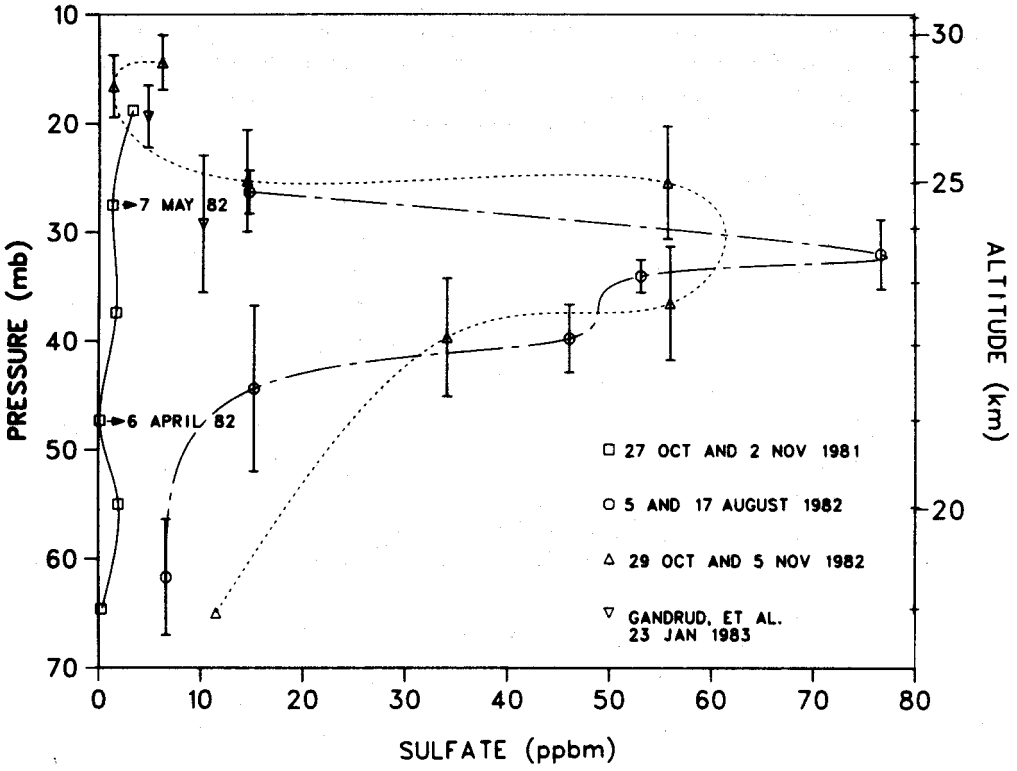

Fig. 3. Vertical profiles of sulfate mixing ratios from Project Ashcan and Gandrud *et al.* (1983).

Chichón had injected significant quantities of COS into the stratosphere, it should be detectable in this region of extraordinarily high sulfate concentrations. Fig. 4 indicates that COS concentrations in this region were no higher than was observed in October-November and, furthermore, show no enhancement over concentrations measured at similar heights above the tropopause at latitudes that could not have been affected by El Chichón. Based on these data we conclude that the sampled plume did not contain elevated levels of COS. This does not preclude COS having been injected above 20 km. Additional work on the COS burden will appear in Leifer *et al.*, 1984.

Rasmussen *et al.* (1982) suggested that volcanic ash is an effective sink for COS. Measurements of volcanic ash in the equatorial region during the same time have been made by ourselves and Gooding *et al.* (1982). It could be argued that the slight reduction of COS observed at 17°N and at 19.2 km might be due to adsorption on the ash. However, measurements of N_2O, shown in Fig. 4, show a similar decrease in concentration. Measurements of F-11, F-12, CO_2, and SF_6 and other

gases shown in Fig. 4 which were made at the same time show an identical reduction in concentration. We believe the small decrease in COS and other trace gas concentrations is associated with dynamical processes such as transport of polar air into the equatorial region, or stratospheric subsidence.

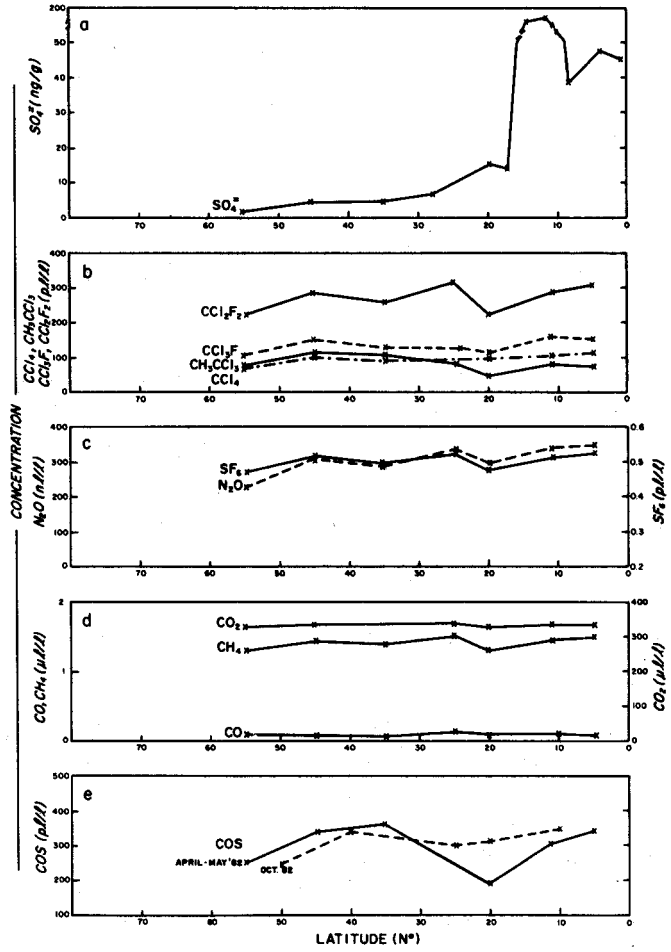

Fig. 4. Trace gas and sulfate concentration data. The data selected are ~2 km above the tropopause. The tropopause height at the equator is 17 km: (a) sulfate data from April-May, 1982 (Mroz *et al.*, 1983); (b) CCl_3F, CCl_2F_2, CH_3CCl_3, CCl_4 (April-May, 1982); (c) SF_6 and N_2O (April-May, 1982); (d) CO, CO_2, CH_4 (April-May, 1982); (e) COS (April-May and October, 1982).

The slightly elevated values (as compared to the October-November 1981 measurements) of sulfate found between 25° and 75°N are probably the result of the

dispersion of debris from the so-called Mystery Cloud that came from an unidentified volcano in January 1982 (SEAN, 1982). This eruption had only a small effect on the inventory of sulfate in the stratosphere when compared to the impact of El Chichón (Mroz *et al.*, 1983).

Ashcan data from this period (Fig. 3) indicate that the debris above 20 km from El Chichón had not yet reached 24.5 km at 33°N by 7 May. Yet on 12 May a very high concentration (133 ppbm) was measured at 22.5 km. This point is the only sample obtained on the flight of that date. It is not shown on Fig. 3 as it far exceeds the concentration scale used.

July-August 1982. Fig. 2c shows that by this period the debris from El Chichón has spread from the equator to at least 75°N at altitudes between the tropopause and 20 km. Ashcan data for this period (Fig. 3) show that the debris at altitudes above 20 km had also reached 33°N by early August.

October-November 1982. The sulfate contours for this period (Fig. 2d) are similar to those for summer 1982. The sampling route for the aircraft was modified to extend into the southern hemisphere to 10°S. The highest concentrations were measured between 10°S and 20°N between 18 and 20 km. Likewise, the concentrations of the trace gases discussed above showed no unusual behavior. Ashcan data for this period show sulfate profiles that are similar to those measured in August, as seen in Fig. 3.

March 1983. The concentrations of sulfate measured in March 1983 (Fig. 2e) are somewhat higher than were measured in the fall of 1982. Records of volcanic activity during the intervening period do not contain any tropopause-penetrating eruptions; however, fresh material was found in April 1983 at 18-19 km in mid-latitudes (SEAN, 1983). The absence of any areas of high mixing ratios in March leads to the conclusion that the early 1983 eruption was a minor contributor to the March inventory. We attribute the increased spring burden to transport of El Chichón aerosol from the region above 20 km to the region below 20 km and thus accessible by the aircraft. This transport process could be either sedimentation of the aerosol or subsidence of air from the stratosphere above 20 km during the winter of 1982-83. We know from a variety of sources (McCormick and Swissler, 1983; Hofmann and Rosen, 1983; this work) that the concentration of aerosols above 20 km was substantially greater than the concentration below 20 km throughout 1982. No balloon flights were made in March, but balloon and aircraft data obtained in January by Gandrud *et al.* (1983) are consistent with the present work in showing mixing ratios in the range 20-30 ppbm between 16 and 18 km from 45° to 50°N and also show the descent of the height of the peak mixing ratio from about 24 km to 20 km during the winter. The balloon data of Gandrud *et al.* (ibid) are shown in Fig. 3.

November 1983. The profiles measured in Nov. 1983 showed a continuing subsidence of the sulfate from higher levels, including a significant penetration of the equatorial tropopause. A 15 ppbm contour is seen in the equatorial region where such levels were absent in the spring. By comparison, the 20+ region at mid-latitudes in the spring appears to have been transported northward and downward along isentropic surfaces and ultimately removed across the tropopause. Fig. *5*, which will be discussed further below, shows that about half of the sulfate present in March had been removed by November.

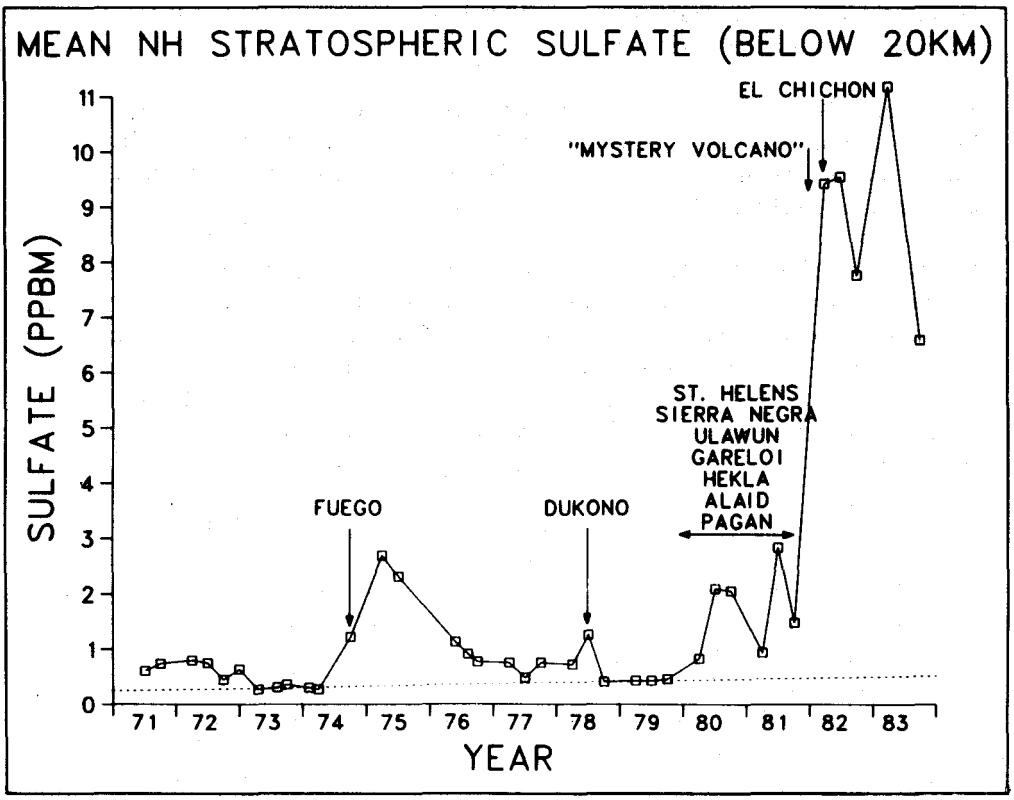

Fig. 5. Historical trend of mean stratospheric sulfate mixing ratio from Project Airstream measurements.

DISCUSSION

Because of Airstream's extensive latitudinal coverage of the stratosphere, the data lend themselves to inventory techniques (Leifer *et al.*, 1982; Mason *et al.*, 1982; Mroz *et al.*, 1983; Telegadas, 1979). Burdens for this report were calculated between the tropopause and 21.3 km (70,000 ft) for the entire hemisphere. Extrapolation was involved above the maximum flight altitude (19.2-20.7 km), and from 75°N to the North Pole. The zone north of 75°N contains only about 3% of the hemispheric atmosphere, rendering the effect of that extrapolation negligible. The region between the maximum flight altitude and 21.3 km comprises at most 12%

of the stratosphere, and this extrapolation is also considered to have a negligible effect on the accuracy of the calculated burden. Details of the burden calculation are discussed below. Reported tropopause height data for the actual flight days were obtained for upper-air stations lying along the flight paths, and smoothed to provide a continuous estimate of the tropopause over the latitude span.

Calculations were done using matrix interpolation and contouring routines from the DISSPLA Version 9.0 (Integrated Software Systems Corporation) library. The interpolation consists of calculation of sulfate mixing ratios at the points of a regular grid of atmospheric pressure and sine of latitude. This coordinate system has the property that each cell of the grid represents a constant zonal air mass. A 48 by 35 grid was used, extending from the North Pole to 10°S. The south boundary was selected to permit use of data collected on the Oct. 1982 and Mar. 1983 deployments in the contouring procedure, and thus estimation of the mass of sulfate transported across the inter-tropical convergence zone. The contouring routine examines the neighborhood of each grid point for data lying within a specified range, in this case 7 cells horizontally by 5 cells vertically, and calculates a sulfate value for that grid point by weighting the data according to a specified function of the distance from the grid point, here the inverse cube. The search is modified near the grid boundaries to include only data lying within the grid, avoiding use of false zero values. The integration is performed by taking the arithmetic mean of the four grid point sulfate values as the mixing ratio within the cell, and multiplying that value by the constant air mass of the cells. The resulting burdens are summed to calculate the hemispheric burden. Cells lying entirely below the tropopause or south of the equator are not included in the summation, except for the separate calculation between the equator and 10°S described above.

Estimation of the injection above aircraft altitude is far less certain. The burdens calculated by Mroz *et al.* (1983) and summarized here were done by considering the Project Ashcan balloon data, which were taken at 33°N, as representative of the zone from 20°S to 40°N and from 20 to 30 km. Balloon series occurred in August and again in October-November 1982. The two burdens were in general agreement, and Table 1 shows that from the second series. This value is also in general agreement with that of Hofmann and Rosen (1983).

Using much of the same data presented here we have estimated the amount of sulfate introduced into the stratosphere by El Chichón (Mroz *et al.*, 1983). This inventory is summarized in Table 1. We estimate that El Chichón introduced 7.6 ± 2.3 Tg of sulfate into the stratosphere. We do not intend to imply that sulfate aerosol was directly injected into the stratosphere by El Chichón. Rather the values expressed in Table 1 represent the estimated amount of sulfur injected expressed as sulfate. Hofmann and Rosen (1983) estimated that the eruption of El Chichón introduced 10 to 20 Tg of aerosol. McCormick and Swissler (1983) estimated 12 Tg of aerosol. The stratospheric aerosol consists of a solution of sulfuric acid in water

Table 1
Stratospheric sulfate attributed to El Chichón

	Tropopause to 20 km (Tg)	20 km to 30 km (Tg)	Tropopause to 30 km (Tg)
Amount injected April 1982	3.1 ± 0.8	4.5 ± 1.5	7.6 ± 2.3
Amount remaining Northern hemisphere October 1982	1.8 ± 0.4 $0^o - 90^oN$	2.2 ± 1.1 $0^o - 40^oN$	4.0 ± 1.5
Amount remaining Southern hemisphere October 1982	0.8 ± 0.3 $0^o - 20^oS$	1.2 ± 0.6 $0^o - 20^oS$	2.0 ± 0.9
Amount remaining Global October 1982	2.6 ± 0.7	3.4 ± 1.7	6.0 ± 2.4

that is about 60% to 80% sulfate by weight (Hofmann and Rosen, *ibid.*). Hence our estimated inventory of 7.6 Tg of sulfate is in good agreement with other estimates made by independent means, especially McCormick and Swissler (1983) which was derived from a latitudinal data set.

The fraction of the sulfur emitted as sulfate or as sulfurous precursor gases at the time of the injection is unknown. However, as noted above, there is evidence that COS is not an important precursor from El Chichón. This leaves unresolved an important question as to the nature of the precursor gases. Vedder *et al.* (1983) measured sulfur dioxide and condensation nuclei simultaneously on 19 April 1982 at 24.6^oN, 28.5^oN and 31.9^oN. They observed very high concentrations of condensation nuclei indicating the occurrence of gas-to-particle conversion but the sulfur dioxide mixing ratios were at background levels. Observers of the quiescent plume reported very high concentrations of hydrogen sulfide (Kotra *et al.*, 1983) emanating from the lake that had formed in the crater. Whether the volcano was emitting hydrogen sulfide during the eruption is unknown.

The magnitude of the impact of El Chichón on the stratosphere can best be seen within the perspective of the 13-year "climatological" record of the stratospheric sulfate gathered by HASP. In Fig. 5 we present a summary of that 13-year history by plotting the average concentration of stratospheric sulfate as function of time. During that period we can see the impact of a number of volcanic eruptions on the sulfate concentrations (Sedlacek *et al.*, 1983). In the 70's the biggest impact on the

stratosphere was from the eruption of Fuego in 1974. Since 1980 there have been increased impacts on the stratosphere from a number of volcanoes. But all of these have been dwarfed by the impact of El Chichón.

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