

## Stratospheric Aerosol Measurements IV: Global Time Variations of the Aerosol Burden and Source Considerations

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### ABSTRACT

Balloon-borne measurements of the stratospheric sulfate aerosol from late 1971 to mid-1974, a quiescent period in terms of large volcanic eruptions at stations ranging from 85°N to 90°S, are utilized in a study of the global spatial and temporal variations and for sulfur budget and aerosol source considerations. Similarities in the aerosol loading in the two hemispheres, both spatial and temporal, are evident. An apparent long-term decay in total aerosol appears to have occurred globally during the period suggesting a transient source. SO<sub>2</sub> budgetary considerations and model calculations favor a larger injection of SO<sub>2</sub> than would be expected from a quasi-static natural exchange of tropospheric air.

### 1. Introduction

In Part II of this series of papers (Rosen *et al.*, 1975) global measurements of the vertical profile of stratospheric aerosols were used in constructing latitudinal distributions of aerosol mixing ratio profiles. Details on the locations of the stations were given in Part II and on the instrumentation in Part I (Hofmann *et al.*, 1975). In Part III (Pinnick *et al.*, 1976) the aerosol size distribution was discussed. It is the purpose of this paper to present the global time variations of the total stratospheric aerosol loading for particles with diameter  $>0.3 \mu\text{m}$  during 1972–74, prior to the eruption of Fuego in Guatemala in October 1974. These time variations are then utilized in aerosol budget calculations and an attempt at characterizing the aerosol source, applicable for that period, is made.

### 2. Observations

Detailed time variations at northern mid-latitudes (Laramie, 41°N) were presented in Part I and only data on the total aerosol loading for this station will be used in this work. Early measurements in polar regions were reported previously (Hofmann *et al.*, 1973) and will be updated here.

Fig. 1 shows three soundings taken approximately one year apart from the floating ice island T-3. In each case the island was near 85°N. Above the tropopause, which was observed at about 10 km in each sounding, a general decrease with time is observed. Such a decrease, with superimposed seasonal variations, was also seen at the Laramie station during this period (see

Part I). As in the Laramie soundings, little variation above about 17 km is apparent.

Fig. 2 shows similar data for the South Pole soundings. Once again the annual measurements suggest a decrease trend in the total stratospheric aerosol loading (area under the curve above the tropopause observed at about 9 km), although a considerably larger decrease occurred between the 1973 and 1974 soundings than between the 1972 and 1973 soundings.

The total stratospheric aerosol loading, as computed from the area under the vertical concentration distributions above the tropopause (in units of  $10^6$  particles  $\text{cm}^{-2}$ -column), is shown in Fig. 3. Data are included for all stations which were utilized long enough to give a representation of a temporal study. Apparently only the monthly Laramie data, an updated version of those presented in Part I, are statistically significant. However, the sparser points from other stations, when viewed in conjunction with the Laramie data, present what we consider a valid global temporal study. In Fig. 3 the time scales for stations in different hemispheres are shifted by 6 months so that if a simple seasonal variation as observed at Laramie (see Part I for details) exists globally, all curves should be in phase. A number of features are noteworthy.

1) There appears to be a general latitudinal gradient in particle loading with a minimum occurring in equatorial regions. This may be contrasted with our observation that the maximum particle *mixing ratio* occurs in equatorial regions (Part II) but since the maximum occurs at high altitude ( $\sim 25$  km), it does not contribute

to the total particle loading to the extent that lower lying aerosol of lesser mixing ratio does at middle and high latitude.

2) Although the data from other stations are not nearly as comprehensive as the Laramie data, there is an indication of a slow decay in aerosol loading, similar to that clearly visible in the Laramie data. This is most noticeable in the Panama and Antarctic data.

3) Stations at comparable latitudes in opposite hemispheres (e.g., Palestine, Tex., and Mildura, Australia) suggest that there is not a great hemispheric difference in aerosol loading. This similarity is in fact enhanced when one takes into account that a slight bias is introduced by the time-scale shifting procedure when a long-term variation is present. The bias is such that the relative magnitude of the Southern Hemisphere data is about 10% less than it should be. Latitudinal surveys of mixing ratio in the two hemispheres, reported in Part II, also suggested a general similarity. Filter collections of stratospheric sulfate aerosol in the Northern and Southern Hemisphere are in agreement with this observation (Lazrus and Gandrud, 1974).

4) The seasonal variation in the Northern Hemisphere appears to be in phase for all stations except Panama (9°N) which shows a variation 6 months out of phase. This apparent discrepancy was explained in terms of the seasonal variation of the height of the tropopause (Hofmann *et al.*, 1974).

5) The seasonal variation in the Southern Hemisphere appears to have a phase as expected for a simple seasonal variation except possibly for the Antarctic stations (McMurdo and South Pole), where, although measurements are lacking during more than a single season, the

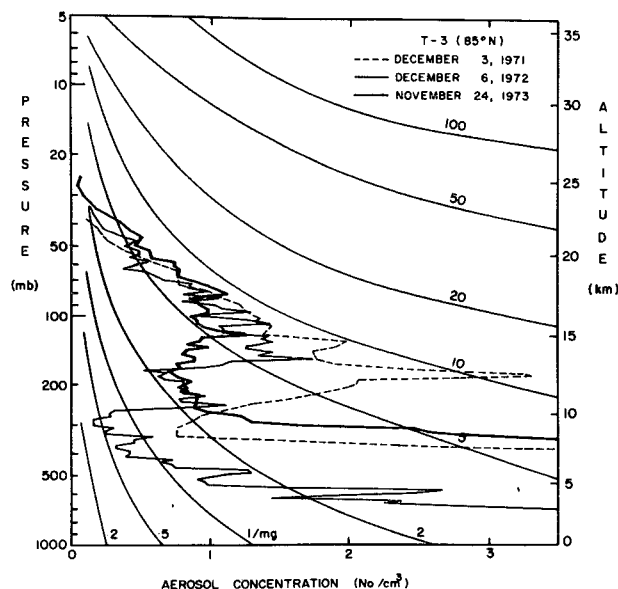


FIG. 1. Vertical distribution of aerosol particles having diameters  $>0.3 \mu\text{m}$  over ice island T-3 (85°N) on three occasions a year apart. Smooth curves are lines of constant mixing ratio in units of particles per mg of air.

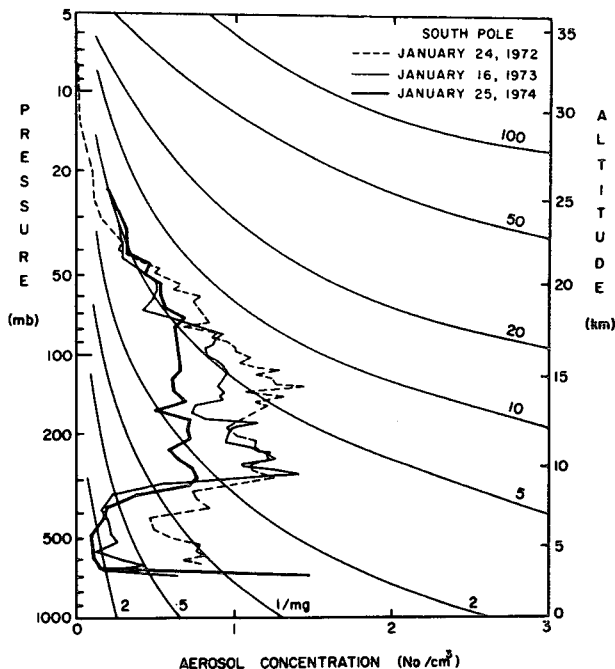


FIG. 2. As in Fig. 1 except over the South Pole on three occasions a year apart.

total aerosol is very similar to that observed in the north polar region at approximately the same time of year (opposite seasons), suggesting either an asymmetric seasonal variation or a considerably higher annual average aerosol burden in the south polar stratosphere.

### 3. Discussion

Having observed an excellent correlation between total stratospheric aerosol loading and tropopause height at Laramie (Part I), we investigated whether such a relation was valid at other latitudes. In Fig. 4 we show the integrated aerosol loading as a function of the observed tropopause pressure for all the stations. A considerably more linear relationship with pressure is found here for all stations as compared with the data reported earlier for Laramie only (Part I). Such a linear relation might be expected if the aerosol mixing ratio profile moves uniformly up and down with the tropopause as has been observed at Panama and Barrow but not at Laramie (Part II).

The observation that Panama and possibly the Antarctic stations do not fit into the simple global seasonal variation picture was explained by noting that the seasonal variations of tropopause height at these locations also do not conform to what is generally considered "standard", i.e., a low and high tropopause in January and July, respectively, in the Northern Hemisphere and vice versa in the Southern Hemisphere (Hofmann *et al.*, 1974). Thus the fact that Panama has a seasonal variation of the tropopause as one would expect to occur in the Southern Hemisphere and the



diffusing into the stratosphere and forming sulfate aerosol following oxidation by a simple first-order chemical reaction. He used the theory to estimate the magnitude of the first-order rate coefficient. An equilibrium aerosol particle sulfur mixing ratio profile is obtained by balancing eddy diffusion and chemical reaction of the  $\text{SO}_2$ -sulfur with eddy diffusion and sedimentation of the aerosol-sulfur.

Since the calculation is based at the tropopause, the calculated profiles are a function of chosen tropopause height and will obviously move up and down with the tropopause as observed. A useful parameter to test the model is the height above the tropopause of the maximum in the mixing ratio profile. As shown in Part II, the observed value is about 8–10 km, somewhat dependent on latitude although more variable at mid-latitude in the tropopause gap region than in equatorial or polar regions. In Junge's model the maximum occurs at about 6 km above the tropopause, assumed to be at 10 km, for his best choice of variables, i.e., an eddy diffusion coefficient of  $5 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ , an  $\text{SO}_2$  oxidation rate coefficient of  $7 \times 10^{-9} \text{ s}^{-1}$ , an  $\text{SO}_2$ -sulfur mixing ratio at the tropopause of 0.1 ppbm,<sup>1</sup> and aerosol parameters suggested by measurements in the pre-Agung (pre-1963) era, i.e., a mean particle radius of  $0.3 \mu\text{m}$  and an aerosol-sulfur mixing ratio at the tropopause of 0.0016 ppbm.

The present measurements suggest somewhat different aerosol parameters; for example, the size distribution reported in Part III suggests a mean aerosol particle radius somewhat smaller than the  $0.3 \mu\text{m}$  used by Junge and the seasonally averaged mixing ratio profiles indicate an aerosol-sulfur mixing ratio at the tropopause considerably higher than that used by Junge. It is to be expected that using knowledge of the actual size distribution will alter the calculated vertical aerosol profiles and, similarly, the considerably larger observed aerosol mixing ratios will alter, for example, the amount of  $\text{SO}_2$  required at the tropopause. In order to make the comparison of Junge's theory and our data more valid, we reevaluated Junge's model (Hofmann and Rosen, 1975) using observed aerosol parameters at Panama ( $9^\circ\text{N}$ , tropopause height 16 km) and Barrow, Alaska ( $71^\circ\text{N}$ , tropopause height 10 km). The observed size distribution was used to take gravitational settling properly into account. The same values of vertical eddy diffusion coefficient and  $\text{SO}_2$  oxidation rate constant as proposed by Junge were used. The  $\text{SO}_2$  mixing ratio at the tropopause was adjusted to give the proper aerosol mixing ratios at the stratospheric maximum for both stations.

The results indicated that an  $\text{SO}_2$ -sulfur mixing ratio at the tropopause of about 1 ppbm was required for both stations. An example for the Barrow station is shown in Fig. 5. The calculated aerosol mixing ratio

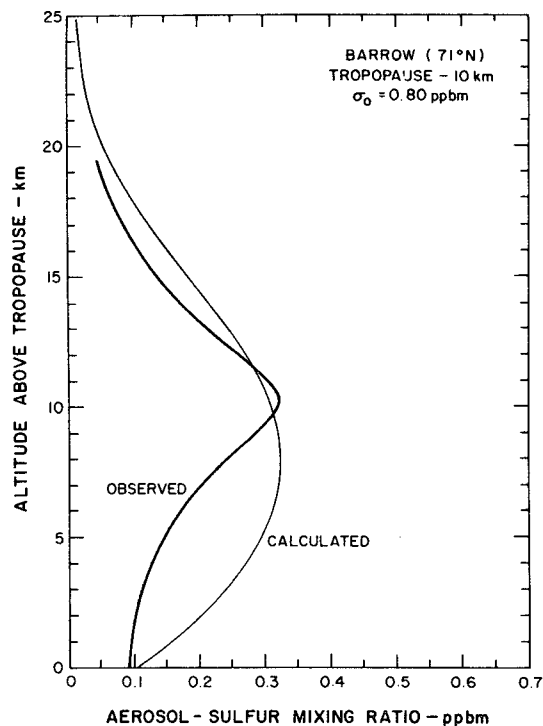


FIG. 5. Comparison of observed and theoretical aerosol mixing ratio profiles for a diffusing, reacting gas of tropospheric origin, as an aerosol source at Barrow.

profiles again peaked about 2 km lower in the stratosphere than observed. Increasing the diffusion coefficient moved the maximum up but broadened the distribution to the point where the calculated mixing ratio was much too high above and below the maximum. Decreasing the oxidation rate coefficient will cause the peak to occur at higher altitudes, e.g., a factor of 10 decrease ( $7 \times 10^{-10} \text{ s}^{-1}$ ) moved the peak up about 1 km. However, the rate is already too low considering the probably rapid oxidation of  $\text{SO}_2$  by OH as discussed by Harrison and Larson (1974). The latter authors also tried to calculate stratospheric sulfate profiles by including the oxidation of  $\text{SO}_2$  by hydroxyl and hydroperoxyl radicals. Although their work showed the importance of the OH radical in the oxidation of  $\text{SO}_2$ , it lacked proper treatment of aerosol formation and consequent gravitational settling. Thus although their calculated sulfate concentration profile, due to oxidation of  $\text{SO}_2$  by OH, showed a maximum about 4 km above the tropopause, mixing ratio profiles generated from these data would show no maximum at all since the mixing ratio is conserved in eddy diffusive processes with no other loss mechanisms. Since the observed mixing ratio profiles show a definite maximum in the stratosphere (see Parts I and II), loss processes such as sedimentation must be important. In addition, since the gravitational settling velocity is equal to the vertical diffusive velocity for a  $0.2 \mu\text{m}$  radius particle at 20 km, sedimentation cannot be ignored. Adding sedimentation to the Harri-

<sup>1</sup> Parts per billion by mass.

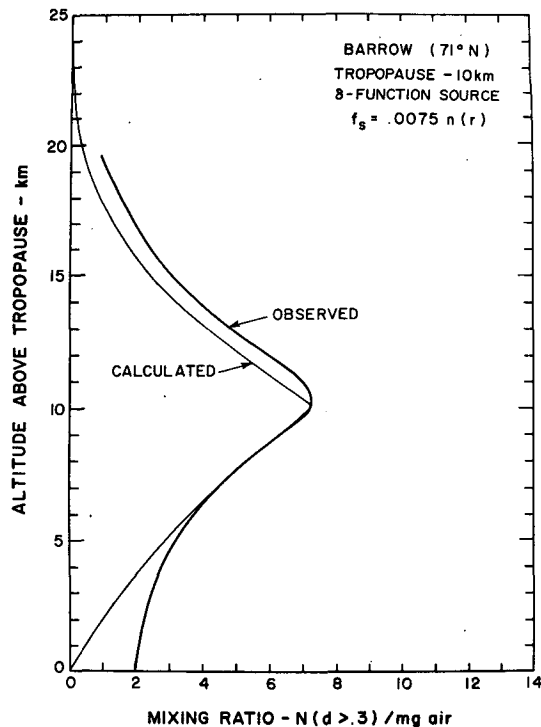


FIG. 6. As in Fig. 5 except profiles for a stratospheric point aerosol source at a height of 10 km above the tropopause at Barrow.

son-Larson model moves the maximum even lower in the stratosphere and one must conclude that a simple diffusing, reacting gas cannot account for the observed aerosol profile.

A further disturbing result of such models is the amount of  $\text{SO}_2$  required at the tropopause—1 ppm sulfur translates into about 2 ppm  $\text{SO}_2$ . This is probably considerably more than can be expected at stratospheric heights under normal conditions. We can also look at the amount of  $\text{SO}_2$  required by seasonal variations in the aerosol burden since motion of the tropopause apparently causes the loss of substantial amounts of aerosol, at least in mid-latitudes in spring (see Part I). As indicated in Fig. 3, a sizeable annual cyclic variation with an average global amplitude of about  $7.5 \times 10^5$  particles  $\text{cm}^{-2}$ -column of stratosphere has been observed. Integrating this amount over the earth and over the size distribution results in the production of  $5 \times 10^5$  tons of aerosol per year, or about  $3.3 \times 10^5$  tons of  $\text{SO}_2$  per year are required for aerosol replenishment. For a total stratospheric air mass of  $10^{15}$  tons and a 75% annual stratospheric-tropospheric mass exchange (Danielsen, 1968), an  $\text{SO}_2$  mixing ratio of about 0.4 ppm would be required at the tropopause. This may be compared with an estimate of 0.2 ppm by Lazrus and Gandrud (1974) from airborne filter collections of sulfates and again assuming that the entire stratosphere takes part in the exchange process. If only upwelling tropospheric air in equatorial regions

is considered for the  $\text{SO}_2$  source, then  $\text{SO}_2$  concentrations about 10 times larger are required (Lazrus and Gandrud, 1974). The latter  $\text{SO}_2$  concentrations would be as large or larger than observed in measurements of clean tropospheric air (Georgii, 1970) and one must conclude that if steady-state tropospheric  $\text{SO}_2$  is the sole aerosol source, then entrance into the stratosphere in other regions, in addition to tropical, is probably necessary. With no measurements of  $\text{SO}_2$  in the upper troposphere or stratosphere, it is difficult to rule out a purely steady-state, tropospheric  $\text{SO}_2$  source. However, difficulties with maintaining tropospheric levels of  $\text{SO}_2$  up to the tropopause make other  $\text{SO}_2$  sources such as volcanic eruptions more plausible.

Another difficulty with the quasi-static tropospheric  $\text{SO}_2$  model is the general shape of the calculated curves in the lower stratosphere (see Fig. 5). In this region the theoretical mixing ratio gradient decreases with height, whereas the observed gradient increases. This part of the theoretical curve is influenced by the exponential falloff of  $\text{SO}_2$  above the tropopause due to chemical reaction and represents too strong a source in this region suggesting that this theory is basically in doubt at least insofar as explaining the aerosol observed during the 1972–1974 period.

To investigate the other extreme, i.e., no sources in the lower stratosphere, we have considered a stratospheric aerosol point source model (Hofmann and Rosen, 1975). Although physically unreal, it is the source distribution which resulted in the best reproduction of the observed aerosol profiles. A comparison for the Barrow station is shown in Fig. 6. Thus the aerosol source must be very inefficient in the lower stratosphere to explain the observations.

The observation of a long-term decrease in total aerosol and in aerosol mixing ratio at the stratospheric maximum at Laramie (Part I) and suggested at other stations in this work supports the idea of a large injection of  $\text{SO}_2$  or a direct injection of sulfate from a volcanic eruption. The volcanic material would have to be injected in equatorial regions or above 20 km in mid-latitudes as seasonal fluctuations of the tropopause at mid-latitudes would presumably lead to annual loss below this altitude. There are some difficulties concerning particle lifetimes with a source of this nature.

As indicated in Part I, the mixing ratio at the maximum is perhaps the best parameter to use for long-term time variation studies as it is apparently not affected by seasonal variations to the extent that the total aerosol burden is. Fig. 7 shows the time variations of this parameter at Barrow, Ft. McMurray (Canada), Laramie, Palestine, Tex., and Panama. Also shown are  $e$  folding decay times in years.

If one is to assume that the time variation in Fig. 7 is due to a single  $\text{SO}_2$  injection which occurred in late 1971, as suggested by the time variation in the size distribution (see Part I), then a relatively long lifetime

of over 3 years would be implied. Decay of radioactive particles generally suggest shorter lifetimes than this, averaging about 11 months over the year (Machta and Telegadas, 1972); however, the size of the radioactive particles are not known, and in the case of the sulfate aerosol, a chemical process which may have a long lifetime is involved. Thus, with current knowledge of the chemistry involved and the dynamic transport effects, it is difficult to determine if the observed time variation in maximum mixing ratio is due to a single event or to multiple events of varying sizes. For example, the small increase in early 1973 seems to be present to some degree in all the data in Fig. 7. Fig. 8 compares soundings at Laramie before and after this increase for the two particle-size ranges measured. The increase occurred mainly above 18 km and was concentrated in the larger particles. Some possible candidate volcanic eruptions and first date of eruption are Helgafell in Iceland, 23 January 1973; Asamayama in Japan, 1 February 1973; and Fuego in Guatemala, 22 February 1973 (Smithsonian, 1973). The fact that the increase was already observed at Laramie on 26 January 1973 and that the Barrow station showed the largest increase in mixing ratio while the Panama station was the last to show an increase, indicates that the Icelandic eruption is the more tenable of the three. It is likely, however, that all could have contributed to some degree although

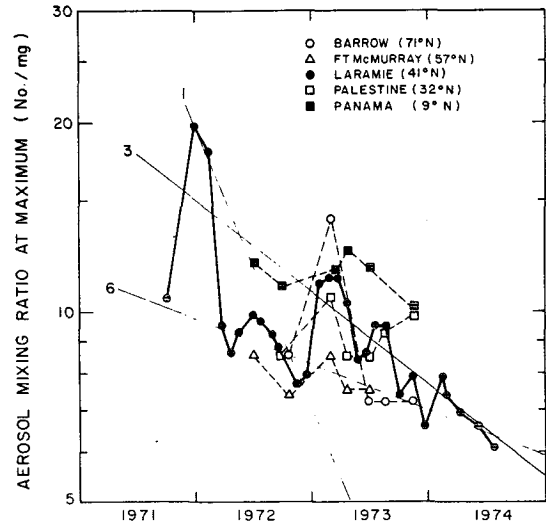


FIG. 7. Maximum observed stratospheric aerosol mixing ratio as a function of time at several stations in the Northern Hemisphere. Straight lines correspond to exponential decay with characteristic decay times marked in years.

the slow time variation in the size distribution would favor a less sporadic source. A sizeable aerosol increase, probably due to the Fuego eruption which occurred in October 1974 and is currently under study, may shed

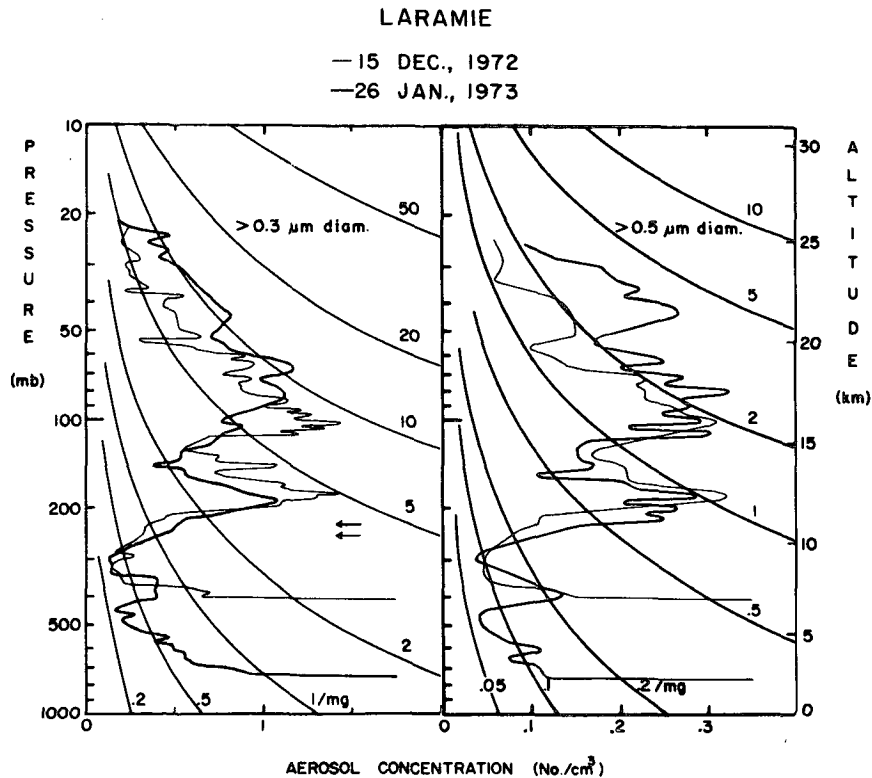


FIG. 8. Comparison of aerosol soundings for two size ranges before and after an apparent high-altitude increase.

more light on this problem, as a greater range of measurements by many different groups is being focussed on it.

#### 4. Conclusions

Measurements of stratospheric aerosol in both hemispheres during 1972-74, a period of minimal large volcanic activity, suggest that there is not a great difference in the two hemispheres in terms of total aerosol loading. Asymmetries in the seasonal variation of the total loading could be explained in terms of corresponding seasonal asymmetries in tropopause height.

A slow decay in the total aerosol loading during 1972 and 1973, following an apparent injection in late 1971, appears to be global in nature suggesting that at least for this time period, a transient phenomenon such as a volcanic eruption is probably the primary source of sulfate aerosol or of SO<sub>2</sub> for the stratospheric aerosol formation process. There may, in addition, have been other small volcanic injections, further complicating the picture.

Sulfur budget considerations and model calculations suggest that a non-volcanic, quasi-static tropospheric SO<sub>2</sub> source for the aerosol layer is doubtful. In addition, the aerosol formation process appears inhibited in the lower stratosphere and is probably associated with volcanic injections of SO<sub>2</sub>.

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