Relative Efficiencies of Filters and Impactors for Collecting Stratospheric Particulate Matter

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ABSTRACT

The relative efficiencies of impactors of the type used by Junge and co-workers for collecting stratospheric sulfate particles and of filters used in these laboratories have been determined by flying these collecting devices on the same aircraft in the stratosphere. The impactors have less than 20% of the efficiency of the filters for total sulfate. At most, a small percentage of the stratospheric sulfate was present as the ammonium salt. At least about half of the ammonium ion found on the impactors could have resulted from reaction of the collected samples with ammonia in tropospheric air.

1. Introduction

A layer of particles, several kilometers deep, occurs in the stratosphere worldwide and continuously. The maximum concentration of particles in the layer at mid-latitudes is at an altitude of 16-20 km. Numerous investigations have been made of the composition and concentration of the particles (Chagnon and Junge, 1961; Junge and Manson, 1961; Junge *et al.* 1961; Friend, 1966; Cadle *et al.*, 1970; Cadle, 1972 a, b; Lazrus *et al.*, 1971; 1972; Shedlovsky and Paisley, 1966; Rosen, 1969, 1971.) They are largely in the size range 0.1 to 1.0 μm radius, and usually consist predominately of sulfuric acid droplets (along with smaller amounts of ammonium sulfate, silicates, and numerous trace substances).

Following the early work by Junge and his coworkers, an especially violent volcanic eruption by Gunung Agung in Bali in 1963 injected large amounts of particulate material and sulfur dioxide into the stratosphere. The sulfur dioxide, by oxidation and hydration in the stratosphere, forms dilute sulfuric acid droplets. Measurements of concentrations of stratospheric particles soon after that eruption have indicated concentrations 10–50 times those reported in the paper listed above by Junge *et al.*

The Junge group measured concentrations by means of impactors, mainly flown on aircraft (Junge and Manson, 1961). They estimated, on the basis of theoretical considerations, that the impactors collected particles of size down to about 0.1 μm radius with nearly 100% efficiency. On the basis of these measurements the sulfur mass concentrations were equivalent to 0.003–0.012 $\mu g\ m^{-3}$ sulfate (Junge, 1963). Many measurements of mass concentrations of stratospheric particulate material have been made in recent years by collection on fiber filters and indicate considerably higher sulfate concentrations. Possibly the differences in measured concentrations of sulfate are at least partially the result of differences between the collection efficiencies of the impactors and the filters. The experiments described in this note were designed to resolve this uncertainty by simultaneously flying impactors and filters in the stratosphere on the same aircraft and comparing the amounts of particulate material collected by the two techniques.

2. Experimental

The impactors were essentially identical with those described by Junge and Manson (1961) except for improved seals to decrease the likelihood of contamination of the impactor surface (Fig. 1). Impaction was

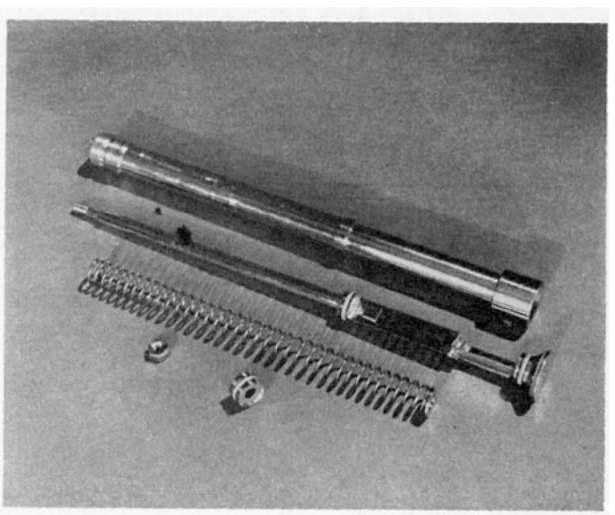

FIG. 1. Disassembled impactor used on aircraft. The motor assembly for pushing the impactor surface into the airstream is not shown. The impactor housing, shown at the top of the photograph, is 28 cm long.

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on the flat, narrow surface facing upward on the far right of the lower of the two "rods" shown in the photograph. The upper "rod" is actually a tube which houses the rod and spring, and is mounted on the outside of the aircraft. When a sample is to be taken, the collecting surface is pushed out of the housing by a motor and into the airstream provided by the speed of the aircraft. Tension on the spring when the collection surface is extended is used to retract the surface at the conclusion of the sampling period and provide a positive seal.

The actual impactor collections were made on strips of heavy platinum foil mounted on the collection regions of the impactor and having the same length and width as those regions. These strips were cleaned by washing repeatedly with absolute ethanol, then with a soap solution, and rinsing ultrasonically with distilled deionized water. They were handled with forceps that had been ultrasonically cleaned in absolute ethanol. Three flights have been made. The platinum strip for one of these (and its control strip which was kept in the laboratory) was used bare. The others were coated with the same grade of silicone oil (Dow-Corning 200 Fluid: 250,000 centistokes) used as a coating by Junge and his co-workers to improve particle retention by the impactors.

The filters were washed IPC filters, prepared, flown and analyzed as described by Lazrus *et al.* (1971). After each flight the platinum strips were removed from the impactors. A few millimeter-length portions were cut from each end of each strip [as done by Junge and Manson (1961)] to eliminate end effects during the sampling, and the collecting surface of the remaining strip was ultrasonically rinsed in distilled water containing 30 ppm BRIJ-35, a non-ionic detergent. The control strips were treated similarly. The rinse water was analyzed for sulfate and ammonium ions in the same way as the extract of the filters. The samples were collected from RB57F U. S. Air Force aircraft at 18–19 km altitude. These planes fly at approximately the same speed as the Lockheed U-2 aircraft used for the research by Junge and Manson, so the collection efficiencies by impaction should be similar with the two types of aircraft. One impactor was carried on each flight.

3. Results

The first two flights, of 4 hr sampling time each in the vicinity of Albuquerque, N. M., were made on 10 and 11 June 1971 with a coated and an uncoated platinum strip. The concentrations of sulfate in the stratospheric air estimated from the filter collections were 0.13 and 0.11 μg m^{-3} ambient, respectively. The amounts collected by the impactors, uncorrected for the controls ("blanks"), corresponded to 22 and 18% of these values, respectively. However, the control surface contained quantities of sulfate sufficiently high to preclude a reliable computation of the actual sulfate samples. The above percentages are thus upper limits of the relative impaction collection efficiencies. Because of the sulfate concentrations observed on the control surfaces in these initial experiments, the techniques for preparing the surfaces and handling the samples were improved and rigorously tested in preparation for a third sampling mission. In this experiment, the sulfate control concentrations were below the limit of detection. The third flight, near Mendoza, Argentina, was made with a silicone-coated strip which had been washed in this manner. The sampling time was about 5 hr. Ambient concentrations of sulfate from the filter and impactor were 0.085 and 0.014 μg m^{-3}, respectively, a relative collection efficiency for the impactor of 16%.

Ammonium ion concentrations determined from the filter collections were 0.98×10^{-9} and 1.3×10^{-9} gm m^{-3} ambient, which indicated that at most 2.0 and 3.1%, respectively, of the observed sulfate could occur as ammonium sulfate. These low values are consistent with results observed on other stratospheric filter samples (Lazrus *et al.*, 1971). Recently reported indirect evidence based on the boiling point of this aerosol suggests it is largely sulfuric acid aerosol (Rosen, 1971).

The control surfaces of the impactor did not contain ammonium ion above our detection limit. The quantities of ammonium ion collected on the filter samples indicated air concentrations approximately 50% less than shown by the impactor results. Possibly, this is due to neutralization of impacted sulfuric acid droplets by ambient ammonia during sample handling in the laboratory. Since there was only two thousandths as much sulfate on the impactor plates as on the filters, ammonia traces would be expected to react with a relatively higher fraction of the impactor sample.

4. Conclusions

These results suggest that either such impactors have much lower efficiencies for collecting stratospheric particles in the size range 0.1–1 μm radius than was assumed by Junge and his co-workers in the early 1960's, or the size distribution of the stratospheric particles has greatly changed since that time, a much larger percentage of the mass now being associated with particles <0.1 μm in radius (Aitken particles). The filters are much more efficient than the impactors for collecting Aitken particles. Even if the first possibility is correct, it would only partially explain the large numerical increase in the stratospheric aerosol concentrations since the early 1960's, which seems to be real and is probably the result of volcanic action.

Observations of ammonium ion on the filters indicate at most a small percentage of the stratospheric sulfate could be present as the ammonium salt. At least about half the ammonium ion found on the impactor samples could have resulted from reaction of the sample with traces of ammonia in tropospheric air. In spite of this possible contamination, the total stratospheric air concentration of ammonium ion determined by impactors

was chemically equivalent to only 5.9% of the stratospheric sulfate concentration between 18 and 19 km.

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