The size distribution and interaction of radioactive and natural aerosols in the stratosphere

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ABSTRACT

Artificial radioactivity which persists in the stratosphere on a time scale of years is shown to be associated with particles below 0.02 μ radius above 27 km and with particles very nearly 0.1 μ radius between 21 km and the tropopause. Assuming the artificial radioactivity to be associated with natural aerosols at each level, the radioactive particle size data provide insight on the size distribution of micrometeorites and other particulates in the upper stratosphere and their interaction with sulfate particles in the lower stratosphere.

The decrease in specific radioactivity with increased sulfate particle radius near 20 km suggests that the photochemical oxidation of SO_2 may not be the important mechanism for large particle formation at this level. Chemical factors and residence time considerations support the view that Aitken nuclei in the upper troposphere are sulfate particles of sufficient size and population to account for the production of large sulfate particles near and above the tropopause by Aitken nuclei coagulation. Approximate estimation of the total sulfate mixing ratio with altitude above 5 km indicates a broad maximum in the upper troposphere. Photochemical and radiochemical evidence for the rapid oxidation of SO_2 in the stratosphere reinforces these views. It is tentatively concluded that Aitken particles in the troposphere account for most of the sulfate in the atmosphere and that there is no stratospheric sulfate layer but only a stratospheric "large particle" layer.

Introduction

Knowledge of the size distribution of radioactive aerosols at various levels of the atmosphere is fundamental to our understanding of the mechanisms of formation, transport and removal of radioactive aerosols and to their suitability as atmospheric tracers. Such information is also essential to the design of efficient particle collection systems for use in radioactive aerosol tracer and fallout studies. It is the purpose of this paper to discuss the limited available data on the size distribution of artificial radioactive aerosols in the stratosphere and their interaction with natural aerosols in the upper and lower stratosphere, together with some interesting implications with respect to the distribution of micrometeorites, the mechanism of the stratospheric large particle sulfate layer formation, the composition of Aitken nuclei and the atmospheric distribution of sulfate.

Published information on the physical and chemical properties of nuclear explosion debris in the atmosphere is limited. ADAMS, FARLOW & SCHELL (1959) have discussed the formation of local fallout particles from surface shots which involve the association of radioactive bomb products with unvaporized particles of surface materials, mainly in the size range exceeding tens of microns in radius. The occurrence and characteristics of very high specific radioactivity particles ranging from about 1 to 10 μ radius in early fallout following large yield air shots has been reported by SITTKUS (1959), EDVARSON *et al.* (1959), SISEFSKY (1961) and others. It is suggested that these particles may be formed by condensation of radioactive bomb products on tropospheric aerosols carried up in the convective system associated with the rising fireball and its outgrowth cloud. Fallout particles in this 1 to 10 μ radius size range will settle rapidly out of the stratosphere and exhibit atmospheric half-residence times on the order of weeks to months.

A large percentage of the radioactive debris from high yield surface shots and most of the radioactivity from large air shots was observed

TABLE 1. *Size distribution of radioactive aerosols in the stratosphere (Drevinsky & Martell, 1962).*

Flight date	Altitude interval (km)	Fraction of total β^- activity		
		Stage 1 (> 0.15 μ rad.)	Stage 2 (0.15 to 0.02 μ rad.)	Stage 3 (< 0.02 μ rad.)
Minneapolis, Minnesota				
7 June 1960	9–15	0.90	0.02	0.08
	15–21	0.03	0.86	0.11
	21–27	0.01	0.64	0.35
	27–30	0.01	0.42	0.57
11 Oct. 1960	9–15	0.16	0.74	0.11
	15–21	0.10	0.85	0.05
	21–27	0.05	0.71	0.24
	27–30	0.13	0.24	0.63
15 Feb. 1961	~20	(0.29)[a]	(0.57)[a]	(0.14)[a]
16 June 1961	~20	(0.86)[b]	(0.13)[b]	(0.01)[b]
Hyderabad, India				
13 April 1961	15–21	0.15	0.75	0.10
	21–27	0.54	0.04	0.42
	27–28	~0.15	~0.15	~0.70
20 April 1961	15–21	0.13	0.72	0.15
	21–27	0.09	0.59	0.32
	27–30	0.05	0.17	0.78

[a] Impactor cutoffs adjusted to 0.18 and 0.0075 μ radius.
[b] Impactor cutoffs adjusted to 0.05 and 0.025 μ radius.

to persist in the stratosphere on a time scale of years. Thus it has been evident that the bulk of stratospheric radioactivity resides in particles below a few tenths of a micron in radius. Data on the size distribution of radioactive aerosols at several levels in the lower stratosphere have been reported by DREVINSKY & MARTELL (1962), DREVINSKY, MARTELL & LAL (1962) and DREVINSKY & PECCI (1965). Some of these results and their implications are discussed below.

Radioactive aerosol size distribution

The approximate size range of radioactive fallout aerosols at several stratospheric levels was investigated by DREVINSKY & MARTELL (1962) in a series of balloon flights over Minneapolis, Minnesota and Hyderabad, India, during the period June 1960 to April 1961. The balloon borne sampler consisted of a two stage impactor backed up by a high efficiency polystyrene microfiber filter. Four collectors were used on each balloon flight, programmed for obtaining 4 samples during ascent over the altitude intervals 9 to 15, 15 to 21, 21 to 27 and 27 to 30 kilometers. For the profile collections, the impactors were calibrated to give a 50 per cent collection efficiency at 0.15 μ and 0.02 μ radius for the first and second stages, respectively, at mid-altitude interval for a particle density of 2. The design, calibration and operation of the impactors has been reported by ZELLER (1960). The polystyrene filter, with fibers about 0.1 to 1.0 μ diameter, provided for the collection of the submicron aerosols which pass the second stage of the impactor with nearly 100 per cent efficiency. The results, in terms of the activity fraction in each size range for each altitude interval, are summarized in Table 1. Details of the radioactivity measurements are reported elsewhere (DREVINSKY & MARTELL, 1962).

The results, Table 1, show a number of interesting features for the radioactive aerosols which persisted in the stratosphere a minimum of 1.5 to 2.5 years following their production

in nuclear tests (i.e., tests in October 1958 or earlier). At this time most of the artificial radioactivity above 27 km was associated with particles below 0.02 μ radius. By comparison, radioactive debris only several months old in this altitude range shows most of the radioactivity on particles between 0.02 μ and 0.15 μ radius (DREVINSKY & PECCI, 1965). The fall speeds for spherical particles in still air can be estimated using the Stokes-Cunningham formula (DAVIS, 1945). By such calculation, a particle of density of 2.5 g cm^{-3} and radius 0.1 μ requires about one year to settle from 30 to 20 km altitude and nearly a decade to fall from 20 to 10 km. Thus the role of sedimentation in the downward transport of artificial radioactivity within the lower stratosphere diminishes with time and should be unimportant for old debris.

In the lowest layers of the stratosphere in the region of the large-particle sulfate layer, most of the radioactivity, Table 1, was associated with particles of about 0.1 μ radius. That the radioactivity was confined to a very narrow size range near 0.1 μ is clearly indicated by the results from the 16 June 1961 flight for which a small downward adjustment of the first impactor stage cutoff shifted most of the radioactivity from the second stage to the first. The results for the two lowest altitude intervals on 7 June 1960 gives striking evidence of a very narrow size distribution. In this case the apparent shift in size distribution with altitude may be due to a small drift in the effective cutoff radius for the first stage of the impactor.

Particles below 0.02 μ radius would be expected to undergo fairly rapid depletion by attachment to each other and to larger particles (ZEBEL, 1958; JUNGE, 1963). The persistence of artificial radioactivity above 27 km on particles below 0.02 μ radius on a time scale of years testifies to the low concentration and the small size of both the radioactive particles and the natural aerosols to which they become attached above this altitude. The gradual shift to larger sizes for old radioactive debris as it mixes down into the lower stratosphere, Table 1, must be due to attachment of the small radioactive particles to the natural aerosols at these levels. The remarkable concentration of the radioactivity in particles of a narrow size range near 0.1 μ radius in the lowest levels of the stratosphere indicates a sharp peak in the population of natural aerosol near 0.1 μ radius, a result not inconsistent with observations for sulfate particles in the lower stratosphere. The association of the radioactivity with natural stratospheric aerosols on the basis of population would also occur if the radioactive particles served as nuclei for sulfate aerosol growth. However, such a process might be expected to yield a broader size distribution of radioactive particles below 0.1 μ radius in the lower stratosphere than observed, especially for the 7 June 1960 and 16 June 1961 flights, Table 1. Thus it is likely that the smaller radioactive particles mixing down from above become associated with the natural aerosol population in the lower stratosphere by direct attachment. The consequences of this result to the question of the mechanism of the stratospheric large particle sulfate layer formation will be considered following a brief review of the distribution of natural aerosols. The possible association of artificial radioactivity with micrometeorites at altitudes above 25 km is also considered later in this paper.

Natural aerosol distribution

Results of comprehensive experimental study of the properties and distribution of natural aerosols in the upper troposphere and lower stratosphere up to 30 km were reported by JUNGE, CHAGNON & MANSON (1961). Vertical profiles obtained with impactors showed that large particles (i.e., particles in the size range, 0.1 to 1.0 μ radius) increased in concentration with altitude in the upper troposphere and lower stratosphere, with a broad maximum between 3 and 9 km above the tropopause and reduced concentrations at higher levels. Vertical profiles of Aitken nuclei (particles below 0.1 μ radius) showed a marked decrease in concentration above the tropopause up to about 20 km (JUNGE, 1961). An extension of these studies by use of rod impactors on aircraft near 20 km altitude from 72° N to 63° S (JUNGE & MANSON, 1961) indicated that the large particle layer was fairly stable and constant in time and space and was composed mainly of sulfate particles. Additional data on the vertical distribution of large particles and Aitken nuclei were obtained over Hyderabad, India (CHAGNON & JUNGE, 1962; JUNGE, 1962). The approximate mean vertical distribution of Aitken nuclei

and large particles, based on the results of JUNGE (1961) and CHAGNON & JUNGE (1961), are summarized in Figure 1.

The size distribution of natural aerosols in the lower stratosphere is not very well established. JUNGE (1963) indicates that the log radius number distribution is approximately inversely proportional to the square of the radius for particles between 0.1 and 1 μ radius. The sharp drop observed in the particle population between 1 and 2 μ indicates the approximate upper limit in the size distribution. In his discussion of a model size distribution for stratospheric aerosols at 1 and 4 km above the tropopause JUNGE (1963) assumes an average radius of 0.03 μ for the Aitken nuclei. Mossop (1965) shows a considerably different size distribution for the large particles at 30 km, with a maximum in the number distribution near 0.15 μ radius and a decrease in particle concentration above 0.4 μ radius by about the fourth power of the radius. Other recent studies by FRIEND *et al.* (Defense Atomic Support Agency, 1964) show that the size spectrum of stratospheric aerosols is somewhat variable from sample to sample. These authors indicate about 0.3 μ radius as the maximum in the large particle number distribution.

Consideration of mechanisms of large sulfate particle formation, below, indicates that Aitken nuclei coagulation may be the primary process. On this basis the maximum in the large particle number distribution would increase with atmospheric residence time, and, thus, with altitude. Therefore, complex and variable large particle size spectra would be expected in regions and periods of rapid vertical exchange.

Stratospheric large particle formation

JUNGE, CHAGNON & MANSON (1961) MANSON *et al.* (1961) and JUNGE (1963) propose that the large particle sulphate layer in the lower stratosphere must be due mainly to the photochemical oxidation of SO_2 and H_2S in the stratosphere. They conclude that the shape of the vertical profile of large particles excludes the possibility that the stratospheric large particle layer can be due to direct upward mixing from the troposphere, except for the unlikely process of equatorial injection and spread by a Brewer-Dobson type circulation. Thus they attributed the increase in large sulfate particles to the in

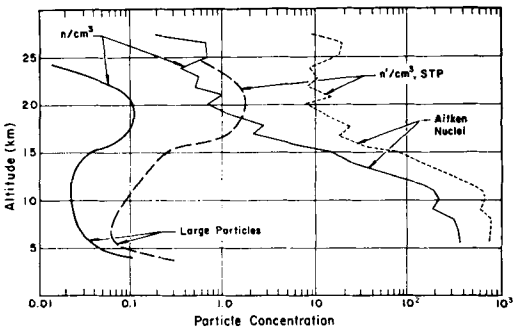

FIG. 1. The average vertical distribution of Aitken nuclei and large particles in the troposphere and lower stratosphere (JUNGE 1961; CHAGNON & JUNGE, 1961).

situ photoxidation of SO_2 and H_2S, followed by attachment of molecular SO_3 and H_2SO_4 to Aitken particles of about 0.03 μ radius.

The possibility that attachment to natural aerosol accounts for the observed change in the size spectrum of the radioactive particles mixing down into the lower stratosphere, Table 1, imposes some difficulties for the mechanism of stratospheric sulfate particle formation proposed by Junge *et al.*, as described above. If SO_3 or H_2SO_4 molecules, formed by the photochemical oxidation of SO_2 at stratospheric levels, give rise to the continuous growth of large sulfate particles by attachment to Aitken particles, the process should be very similar to that for attachment of the minute radioactive particles which are continuously mixing down into the layer from above. Thus in the steady state transport of radioactivity down through the lower stratosphere, there should be a nearly constant specific radioactivity throughout the stratospheric sulfate aerosol if the latter grew in situ by SO_2 photochemical oxidation. The result that the specific radioactivity of sulfate particles decreases markedly with increasing radius between 0.1 to 1.0 μ (MANSON, 1963) suggests a different mechanism or different region for production of the sulfate aerosol.

Some characteristics of the stratospheric aerosol distribution are also difficult to explain on the basis of the in situ stratospheric production proposed by Junge *et al.* The concentration of sulfate in the stratospheric large particle layer near 20 km shows little variation with latitude and season (MANSON, JUNGE & CHAGNON, 1961). The persistence of the characteristic large sulfate particle layer concentration in the

lower stratosphere at high latitudes during periods of rapid vertical exchange in late winter seems difficult to account for by the proposed photochemical oxidation of H_2S and SO_2 at altitude. Furthermore, the vertical distribution of large particles in the lower stratosphere at middle latitudes, Figure 1, is not unlike that observed for radioactive aerosols for sources injected into the stratosphere at various levels at both higher and lower latitudes (FEELY & SPAR, 1963; Defense Atomic Support Agency, 1959). Thus the vertical distribution of the stratospheric sulfate may very well be the consequence of atmospheric mixing processes with some contributing effect from particle coagulation and sedimentation, as is the case for radioactive aerosols at similar stratospheric levels.

These considerations indicate the possibility that the sulfate aerosol is produced in the troposphere and is introduced into the stratosphere by mixing. Thus it was suggested (MARTELL, 1964) that the stratospheric large particle layer may be due to upward transport of tropospheric aerosols by convective mixing both in large convective storms and in locations and periods with a quite different vertical profile of large particles than that found by CHAGNON & JUNGE (1961, 1962). JUNGE (1965) considered the possible role of convective storms in the formation of the stratospheric layer. He pointed out a number of difficulties due to cloud washout, chemical considerations and the volume and height distribution of convective storms, and concluded that this process cannot be important for stratospheric aerosol production.

In this paper it is proposed that a likely mechanism for the formation of the stratospheric large particle layer is the coagulation of Aitken nuclei in the upper troposphere and lower stratosphere. Chemical considerations discussed below indicate that the Aitken nuclei in the troposphere above 5 km may have essentially the same composition as stratospheric sulfate aerosol and thus must be produced by SO_2 and H_2S oxidation at tropospheric levels. On this basis the Aitken nuclei and large particles comprise one broad distribution of sulfate particles and the increase in the large particle population with altitude above 10 km can be explained by coagulation of the Aitken particles. This possibility is considered in further detail below. It will also be shown that the total sulfate mixing ratio must exhibit a broad maximum in the troposphere. If these tentative results are correct, there would be no stratospheric sulfate layer but only a stratospheric "large particle" layer.

Aitken nuclei coagulation

For uncharged particles of uniform size, the approximate rate of decrease in number can be estimated using the SMOLUCHOWSKI (1917) relation for aerosol coagulation in still air:

$$1/n - 1/n_0 = Kt, \tag{1}$$

where n_0 and n are the initial and final number concentrations in particles per cm^3, t is the time in seconds and K is the coagulation coefficient in units of cm^3 sec^{-1}. K is defined by equation (2):

$$K = 4kT\beta/3\mu, \tag{2}$$

where k is the Boltzmann constant (1.38×10^{-16} erg deg^{-1}), T is the absolute temperature, μ is the coefficient of viscosity and β is the Fuchs correction term (FUCHS, 1964; HIDY & BROCK, 1965).

For a steady state population, n_0, of Aitken particles in the atmosphere, the differential form of equation (1) gives the approximate coagulation rate:

$$-dn/dt = Kn_0^2. \tag{3}$$

The corresponding population turnover time, or average particle lifetime, τ_c, due to coagulation is given by the relation:

$$\tau_c = 1/Kn_0 = 3\mu/4kT\beta n_0. \tag{4}$$

Using this equation, the average coagulation times for Aitken nuclei in the atmosphere were estimated and are given in Table 2. The Aitken particle population, n_0, was taken from Figure 1. The arbitrary values assumed for the Aitken particle radii, Table 2, increase with aerosol residence times, τ_r, and therefore with altitude. The magnitude and trend of the assumed radii are qualitatively justified by coagulation considerations and by the results. The corresponding ratios of mean free path to particle radius, λ/R, are in the range 5 to 10 for which present theories indicate maximum, but uncertain, coagulation rates (HIDY & BROCK, 1965).

The approximate mean atmospheric residence

time τ_r for aerosols at various atmospheric levels based on radioactive aerosol studies is given in the last column Table 2 and also in Figure 2. This atmospheric residence time, τ_r, takes into account all atmospheric removal processes and thus indicates the average time available for particle coagulation at each level. Based on radioactive aerosol studies of BLIFFORD *et al.* (1952), HAXEL & SCHUMANN (1955) and BURTON & STEWART (1960), the mean residence times for atmospheric aerosols are taken to be ~0.5 weeks in surface air and ~4.5 weeks at the tropopause for purposes of discussion in this paper. Although there is less agreement about residence times for stratospheric aerosols (MARTELL & DREVINSKY, 1960), a value of 1 year for the 20 km altitude level is assumed.

The estimated coagulation times, τ_c, are longer than the residence times, τ_r, at corresponding altitudes, Table 2. However, the coagulation times will decrease somewhat due to turbulence, can be influenced by particle charging effects, and would be reduced considerably by large non-uniformities in the aerosol concentration distribution. For particles of a given size and altitude the mean time of coagulation varies inversely with particle population, as indicated by equation (4). Thus if the Aitken nuclei are irregularly distributed, the coagulation times, τ_c, will be reduced and may approach the magnitude of the residence times, τ_r, near the tropopause. NEWKIRK & EDDY (1963) observed that natural aerosols in the lower stratosphere are distributed in extremely thin laminae of particles. Also it has been suggested that Aitken nuclei in the troposphere are produced by the evaporation of small cloud droplets. Thus their production distribution is likely to result in a highly irregular distribution of Aitken particles at tropospheric levels. Noting that the population of Aitken particles per unit volume, STP, decreases by a factor of only 50 between 10 and 20 km altitude, it becomes apparent that the coagulation of Aitken nuclei can be an important mechanism for the production of large particles in the upper troposphere and lower stratosphere.

TABLE 2. *Estimated mean coagulation life for Aitken nuclei vs. altitude.*

Altitude (km)	Average population (n/cm³)	Assumed radii (cm)	τ_c [a] (wks)	τ_r [b] (wks)
5	350	2×10^{-6}	2.8	0.75
10	250	3×10^{-6}	4.2	1.0
12.5	100	4×10^{-6}	11.5	2.5
15	14	5×10^{-6}	83	5.0
20	1	6×10^{-6}	1620	50

([a]) $\tau_c = 1/n_0 K = 3\mu/4kT\beta n_0$, mean coagulation life in seconds.

([b]) τ_r is the approximate atmospheric residence time.

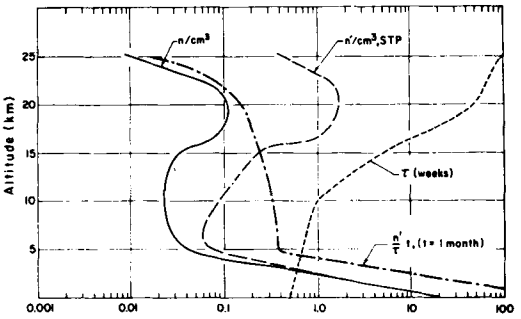

FIG. 2. The approximate number distribution and residence time of large particles vs. altitude. Since τ is the average atmospheric life of a particle at each level, the curve $n't/\tau$ represents the total number of large particles mixed into and produced at each level, or the total number removed, in the time interval t.

Sulfate in the troposphere

In the absence of representative data on the vertical distribution of H_2S and SO_2 in the troposphere and lower stratosphere the vertical distribution of sulfate aerosol production is not readily assessed. However, published information on the tropospheric distribution of sulfate aerosol provides some insight on the possibilities for the sulfate production distribution at lower atmospheric levels.

JUNGE (1963) shows that both NH_4^+ and $SO_4^=$ are prominent constituents of large particles (i.e., 0.1 to 1.0 μ particles) in continental surface air but that they contribute on the average only a fraction of one per cent of the total dust load of continental surface air in both urban and rural areas. In sea salt, $SO_4^=$ is present at a concentration of 7.7 per cent by weight. Because sea salt particles are present in the atmosphere in relatively low number concentra-

tion and mainly as giant particles (i.e., 1 to 10 μ radius and larger) they should decrease rapidly with altitude relative to Aitken nuclei and large particles, due to the sedimentation of giant particles and to the continuous atmospheric production of Aitken nuclei and large particles by chemical and coagulation processes.

PODZIMEK *et al.* (1963), JUNGE (1963), GEORGII & WEBER (1964) and others have discussed many aspects of the chemistry of precipitation. This work indicates the presence of $SO_4^=$ to the extent of from 5 to 50 per cent of the total non-volatile inorganic constituents in precipitation. Georgii and Weber show a marked enrichment of $SO_4^=$ relative to other constituents for continuous rains in comparison to light showers, for rains collected at higher elevations and for snows in comparison with rains. The $SO_4^=/Cl^-$ ratio for continental rains ranges from about 1 to 10, compared to only 0.14 for sea salt, indicating the minor contribution of marine $SO_4^=$ to aerosols in the lower troposphere. Na^+/Cl^- ratios indicate that the loss of chloride as gaseous HCl or Cl_2 cannot explain the $SO_4^=$ enrichment. Because an appreciable fraction of the precipitation constituents are accumulated below cloud level from continental surface aerosols of low sulfate content, the enrichment of $SO_4^=$ in aerosols at cloud level must exceed that observed in rains. Further, the sedimentation of the giant sea salt particles during the longer residence times in the upper troposphere contributes to the further enrichment of $SO_4^=$ at these levels. These considerations indicate a continuous enrichment of $SO_4^=$ relative to surface generated aerosols with increasing altitude in the troposphere.

Some aspects of the chemical production of $SO_4^=$ in the lower atmosphere and in cloud droplets have been studied by several investigators. GERHARD & JOHNSTON (1955) indicate a fairly rapid photochemical oxidation of SO_2 in sunlight which may proceed by the suggested reactions:

$$SO_2 + h\nu \rightarrow SO_2^*$$

$$SO_2^* + O_2 + M \rightarrow SO_4 + M$$

$$SO_4 + O_2 \rightarrow SO_3 + O_3$$

LEIGHTON (1961) points out that, while the absorption coefficients over the range 3200–3900 Å are higher for SO_2 in water solution than for gaseous SO_2, the fraction of atmospheric SO_2 in fog and cloud droplets is negligibly small, thus indicating that the gas phase reactions must play the decisive role with respect to production of SO_3 by photochemical oxidation. Once formed, the SO_3 molecules are rapidly hydrolyzed to H_2SO_4 molecules which attach rapidly to aerosols and cloud droplets. JUNGE & RYAN (1958) and VAN DEN HEUVEL & MASON (1963) have proposed that the catalytic oxidation of SO_2 adsorbed in cloud and fog droplets may account for most of the production of $SO_4^=$ in the lower atmosphere. MEETHAM (1950) and JUNGE (1960) have suggested half-lives for atmospheric SO_2 of 11 hours and 4 days, respectively, but these estimates are very uncertain. The remarkable enrichment of $SO_4^=$ in sea fog, with $SO_4^=/Cl^-$ ratios ranging up to 14 (JUNGE, 1963), and the comparably high enrichment of $SO_4^=$ in precipitation (LODGE, 1960; ERIKSSON, 1959) testify to the rapid oxidation of H_2S and SO_2. Thus the $SO_4^=$ distribution in the lower troposphere may be very dependent on the H_2S and SO_2 source distribution. It also seems clear that the production of $SO_4^=$ must be rapid and continuous (in daylight) at all levels of the troposphere, decreasing with altitude as H_2S and SO_2 are consumed or removed by chemical and physical processes.

The approximate vertical distribution of large particles and Aitken nuclei, Figure 1, and the estimated atmospheric residence times, τ_r, vs. altitude, Figure 2, provide some additional insight on the vertical distribution of sulfate production. In Figure 2, the monthly integral of large particles vs. altitude is given by the curve labeled $n't/\tau_r$ and constructed with t equal to 1 month. This curve represents the approximate number of large particles mixed into or produced at each level, or alternatively, the number removed by all processes at each level within one month. Because the surface of the earth is the only sink for large particles, the integral of large particles must decrease with altitude. Intermediate values of τ_r were adjusted accordingly. Undoubtedly the actual vertical distribution of large particles and their residence time profile will differ appreciably from values given in Figure 2. The large particle population is increasingly uncertain below 10 km and may vary widely with season, latitude and other factors at levels below 5 km. It is shown below that Aitken nuclei, not large

TABLE 3. *Approximate aerosol mixing ratio vs. altitude.*

Altitude (km)	Estimated Approximate Aitken particle volume([a,b]) ($cm^3 \times 10^{15}$)	large particle volume([b]) ($cm^3 \times 10^{15}$)	Estimated total aerosol mixing ratio ($\varrho_s = 2.5$) (g/g dry air)
5	21.8	0.9	4.1×10^{-11}
10	109	1.2	17.3×10^{-11}
12.5	153	2.0	23.7×10^{-11}
15	63	3.5	10.1×10^{-11}
20	18.2	26	6.7×10^{-11}

[a] Based on Aitken particle populations and radii given in Table 2.
[b] Particle volume per cm^3 of air, STP.

particles, must account for most of the sulfate at all tropospheric levels.

It is obvious that a very substantial amount of sulfate aerosol must be produced in the troposphere to explain the enrichment of $SO_4^=$ relative to surface generated aerosols with increased altitude in the lower atmosphere. As SO_2 is oxidized in the troposphere the molecular SO_3 and its hydrolysis product, H_2SO_4, quickly attach to fog and cloud droplets or to the large population of Aitken nuclei at all tropospheric levels. Thus it seems reasonable to assume that Aitken nuclei undergo changes in composition with altitude and above 5 km are composed mainly of sulfate. On this basis, the volume concentration of sulfate vs. altitude between 5 and 20 km would be determined by the combined volume distribution of Aitken nuclei and large particles. For Aitken particles with the radii listed in Table 2 and large particles with a mean radius of 0.15×10^{-4} cm (MOSSOP, 1965), both of density 2.5, the approximate total aerosol mixing ratio at levels between 5 and 20 km can be estimated and are given in Table 3. The combined mixing ratio of Aitken particles plus large particles, Table 3, indicates a broad maximum in the upper troposphere. Assuming that the Aitken particles are sulfate and that their assumed radii, Table 2, are not seriously overestimated, they can account for about one to two orders of magnitude more sulfate than the large particle population.

The only available data on the SO_2 concentration above surface levels is that of GEORGII & JOST (1964) who find about 8 μg SO_2 per m^3 at 5 km over Germany, corresponding to an SO_2-sulfur mixing ratio of about 5.5×10^{-9} g/g. The complex distribution of surface sources and the short atmospheric lifetimes for H_2S and SO_2 should result in their highly variable distribution in the lower troposphere. If the estimated atmospheric half-life of about 4 days or less for SO_2 is correct, the SO_2 concentration at 5 km over Germany is unlikely to provide a representative 5 km value for the northern hemisphere. The expected rapid oxidation of any SO_2 that mixes up into the lower stratosphere (see below) suggests that the average SO_2–S mixing ratio must fall rapidly below that of the total $SO_4^=$–S mixing ratio in the lower stratosphere (Table 3). It is clear that the global average vertical distributions of SO_2 and $SO_4^=$ must be determined experimentally before we can assess the relative importance of SO_2 oxidation or Aitken particle coagulation as mechanisms for large-particle production in the lower stratosphere.

SO_2 oxidation in the stratosphere

In addition to the photoxidation of SO_2 in the presence of O_2 which can occur at all atmospheric levels, any SO_2 and H_2S which mixes up into the lower stratosphere should be even more rapidly destroyed by reaction with ozone and atomic oxygen. For the reaction, $SO_2 + O + M \rightarrow SO_3 + M$, KAUFMAN (1958) reports a fast reaction with a rate constant $\sim 3 \times 10^{16}$ cm^6 $mole^{-2}$ sec^{-1}. In a more recent study of the same reaction, POWERS & CADLE (1965) found it to proceed with nearly zero activation energy, with a rate constant about an order of magnitude smaller than reported by Kaufman. Thus the reaction is fast and nearly independent of temperature. Although the average concentration of atomic oxygen in the troposphere and lower stratosphere is negligibly low, the continuing production of atomic oxygen by ozone dissociation may contribute significantly to the oxidation of SO_2 in regions of relatively high ozone concentration. CADLE (1965) suggests an atmospheric mean life for SO_2 as low as 2 weeks and 2.4 days at 15 and 20 km respectively due to reaction with ground state atomic oxygen. The distribution and reactions of atmospheric H_2S are less well understood (JUNGE, 1963). In a recent study of the reaction, $H_2S + O_3 \rightarrow H_2O + SO_2$, CADLE & LEDFORD (1965) obtain

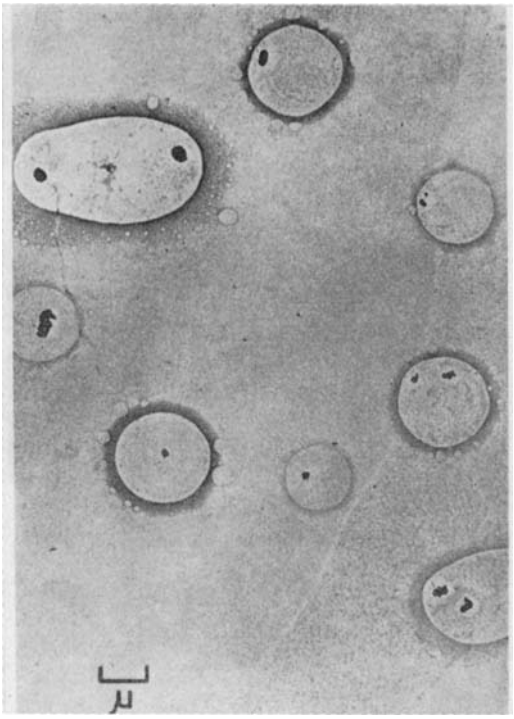

FIG. 3. Residue after dialysis of water-soluble material; collection of 31 January 1963. (S. C. MOSSOP, 1965, Stratospheric particles at 20 km altitude, *Geochim. et Cosmochim. Acta*, 29, p. 201.)

results which set upper limits on the rate of reaction of H_2S with O_3 in the atmosphere.

The "hot atom" chemistry of sulfur and the chemical fate of the 87 day half-life sulfur-35 radioisotope produced by cosmic-ray spallation of argon in the atmosphere provides further insight on the atmospheric chemistry of sulfur. However, there is incomplete understanding of the initial chemical distribution for S^{35} resulting from its hot atom reactions in the atmosphere. HYDER & MARKOWITZ (1964) find that the $S^{34}(n,\gamma)S^{35}$ reaction on H_2S, SO_2 and other gases of sulfur in the presence of oxygen, argon, or other additive gases gives $S^{35}O_2$ as the main reaction product. However, CHIOTAN *et al.* (1964) indicate that $Cl^{35}(n,p)S^{35}$ on KCl crystals yields a very reactive form of atomic sulfur which apparently goes directly to $S^{35}O_4^=$ in the presence of water. It is suggested that $S^{35}O_2$ will be the main product of hot atom sulfur reactions in the atmosphere by the proposed primary reaction process:

$$S^* + O_2 \rightarrow SO_2^* \rightarrow SO + O$$
$$SO + O_2 \rightarrow SO_2 + O$$
$$\downarrow \quad \uparrow$$
$$\longrightarrow SO_3^*$$

Undoubtedly some $S^{35}O_3$ will also be formed in the hot atom reactions.

Direct measurements of S^{35} activities collected on IPC filters by aircraft (BHANDARI & RAMA, 1963; RAMA & HONDA, 1961) showed nearly secular equilibrium production amounts of S^{35} to be present on the filters at altitudes of 18 to 20 km. Since $S^{35}O_2$ would pass through the filters, the initially formed $S^{35}O_2$ must have been almost completely oxidized to sulfate. This result indicates a lifetime for SO_2 in the lower stratosphere that is short compared to the 87 day half-life of S^{35}. However, the S^{35} data are inconclusive because the fraction initially in the form of $S^{35}O_2$ is uncertain and because retention of $S^{35}O_2$ by the cellulose fibers of IPC filters may be important. Clarification of the hot atom chemistry of sulfur in the atmosphere and further data on the chemical distribution of S^{35} in the stratosphere is needed.

Aerosols above 25 km

An indication of the size distribution of natural aerosols above 25 km is provided by the radioactive particle size range data, Table 1. It is obvious that debris from thermonuclear explosions which persists in the stratosphere must contribute only a negligible fraction of the total stratospheric aerosol volume. Therefore it is possible that most of the radioactivity in the 27–30 km range, Table 1, has become attached to natural stratospheric aerosols as it mixed down from higher levels. On this basis it is indicated that the natural aerosol population above 25 km is made up mainly of particles below 0.02 μ radius, the size range in which most of the radioactivity is found, Table 1.

MOSSOP (1965) has identified insoluble inclusions in stratospheric sulfate particles collected by aircraft near 20 km altitude between 16° and 45° South, during the period 29 January to 14 March 1963. The samples were collected on electron-microscope grids coated with nitrocellulose and carbon films. Dialysis of the coated grid specimens by floating them on purified water left an enlarged ring of undialyzed mate-

rial plus one or more insoluble particles. Typical results, illustrating the appearance of the majority of such particles are shown in Figure 3. Particles at higher magnification are shown in Figure 4. Because nearly all stratospheric sulfate particles contained at least one insoluble inclusion, Mossop concluded that the insoluble particles must serve as nuclei upon which sulfate is deposited in the stratosphere.

A different interpretation of Mossop's observations is more consistent with the radioactive size data, Table 1. The radioactivity mixing down from above 27 km is associated with aerosols below 0.02 μ radius, presumably insoluble micrometeoritic particles. The insoluble inclusions in sulfate particles at 20 km, Figures 3 and 4, appear to be clusters of particles both larger and smaller than 0.02 μ in radius. It is suggested that the insoluble inclusions are clusters of small particles which were dispersed at higher stratospheric levels and which became attached to the surface of the large sulfate particles in the lower stratosphere. During the process of melting, hydrolysis and dialysis of the stratospheric sulfate particles, the numerous small insoluble particles became attached to each other due to surface tension, either in the surface layer or within the solution droplet to form the observed irregular aggregates. This process can explain why nearly every stratospheric sulfate particle holds one or more insoluble inclusions.

Although little is known of the composition and origin of these insoluble inclusions it is reasonable to assume that they are mainly meteoritic, with a small admixture of nuclear explosion debris and other contaminants and natural aerosol present in the atmosphere above 25 km. The possibility that the insoluble material originates at the surface of the earth can be ruled out as unlikely using an argument similar to that applied to sodium (JUNGE, OLDENBERG & WASSON, 1962). The particles studied by MOSSOP (1965) were collected before the Mt. Agung volcanic explosion on 17 March 1963, making volcanic dust an unlikely source for the observed insoluble inclusions. SHEDLOVSKY & PAISLEY (1965) have investigated the composition of stratospheric aerosols at 20 km by analysis of selected neutron activation products of particles collected on high-purity polystyrene filters. Their results give some preliminary information on the composition of the insoluble constituents and allow better estimation of the meteoritic component of stratospheric aerosols.

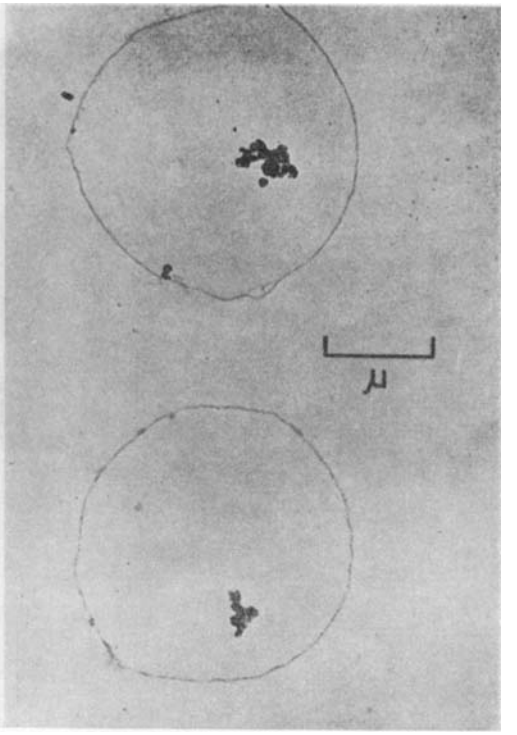

FIG. 4. Residue after dialysis of water-soluble material. Collection of 14 February 1963, showing highly irregular "fluffy" particles of low electron density. (S. C. MOSSOP, 1965, Stratospheric particles at 20 km altitude, *Geochim. et Cosmochim. Acta*, **29**, p. 201.)

Dispersed as individual particles of $< 0.02\ \mu$ radius above 25 km, the insoluble material present in aerosols at 20 km suggests an Aitken particle population which increases with altitude above 25 km due to the longer atmospheric residence times and the less complete coagulation of the meteoritic material and other aerosols at higher stratospheric levels. Thus it is indicated that Aitken particles above the stratospheric large particle layer are probably meteoritic of the order of 0.01 μ radius while those below the layer may be mainly sulfate particles of nearly 0.1 μ radius.

Concluding remarks

This paper began in an attempt to explain the few available experimental data on the size

distribution of radioactive debris in the stratosphere in terms of the interaction of the radioactive particles with natural aerosols at stratospheric levels. As it turned out, the radioactivity is useful to trace the behavior of the natural aerosols to which the radioactivity is attached. In addition, atmospheric residence times based on radioactive aerosol studies provide an index to the time available for coagulation and for other physical and chemical processes at each atmospheric level. Cosmic ray produced S^{35}, an 87 day half-life radioisotope, has special application to the atmospheric sulfur distribution. Some of the tentative conclusions arrived at from such considerations are at odds with widely accepted views, particularly with respect to the formation mechanism and the very existence of the stratospheric sulfate aerosol layer. Due to the lack of adequate experimental data, the conclusions reached are far from final but should prove useful as a guide to further experimental work. A brief summary of these preliminary findings follow:

Artificial radioactivity which persists above 25 km is associated with particles, presumably micrometeorites, mainly below 0.02 μ radius. Upon mixing down into the lower stratosphere these small particles become attached to sulfate particles of very nearly 0.15 μ radius. Gravitational sedimentation rates, estimated by the Stokes-Cunningham formula, are slow compared to atmospheric exchange times for aerosols of these sizes at these altitudes.

The decrease in specific radioactivity with increasing sulfate particle size near 20 km suggests that the photoxidation of SO_2 in the stratosphere may not be the important mechanism of formation for the large particles in the lower stratosphere. The coagulation of Aitken nuclei in the upper troposphere and lower stratosphere provides a good alternative formation mechanism. Chemical considerations support the view that both Aitken nuclei and large particles in the troposphere above 5 km are sulfate aerosols of nearly the same composition as large particles in the lower stratosphere. On this basis the total sulfate mixing ratio was estimated, Table 3, and indicates a broad maximum in the troposphere. This result raises doubts about the existence of a stratospheric sulfate layer.

Evidence from the chemistry of SO_2 and from the chemical distribution and hot atom chemistry of cosmic ray produced S^{35} suggests that SO_2 is rapidly oxidized in the lower stratosphere. On this basis, the $SO_4^=$–sulfur mixing ratio is indicated as an upper limit for the SO_2–sulfur mixing ratio in the lower stratosphere.

Much experimental work remains to be done to confirm these conclusions or to discover better alternatives. The total sulfate mixing ratio from surface levels to 20 km will be investigated using high purity filter collections and neutron activation analysis methods described by SHEDLOVSKY *et al.* (1965). SO_2 and H_2S measurements should be extended to the upper troposphere and lower stratosphere in order to assess the vertical distribution of sulfate aerosol production. Stable isotope studies of atmospheric H_2S, SO_2 and $SO_4^=$ are necessary to an improved understanding of the source distribution and other features of the atmospheric sulfur cycle. Further work on the hot atom chemistry of sulfur in air and on the chemical distribution of S^{35} in the atmosphere should provide supplemental information on the rate of SO_2 oxidation and its variation in time and space.

It is clear that we are only on the threshold of understanding the origin, distribution, reactions and role of sulfur compounds in the atmosphere. In this connection it is noted that the proposed concentrated layer of sulfate aerosol over the broad altitude range from 5 to 20 km may play an important role in the physics and chemistry of clouds and in the chemistry and removal of gaseous and particulate trace constituents of the troposphere and lower stratosphere.

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ВЗАИМОДЕЙСТВИЕ И РАСПРЕДЕЛЕНИЕ ПО РАЗМЕРАМ РАДИОАКТИВНЫХ И ЕСТЕСТВЕННЫХ АЭРОЗОЛЕЙ В СТРАТОСФЕРЕ

Показано, что искусственная радиоактивность с характерным временем нахождения в стратосфере порядка нескольких лет связана на высоте большей 27 км с частицами радиусом меньшим, чем 0,02 μ и на высоте ниже 21 км, включая тропопаузу с частицами радиусом около 0,1 μ. Предположив, что в каждом уровне искусственная радиоактивность связана с естественными аэрозолями, мы можем по данным о размерах радиоактивных частиц представить распределение микрометеоритов и других частиц по размерам в верхней стратосфере и их взаимодействие с частицами сульфата в нижней стратосфере.

Уменьшение удельной радиоактивности с ростом радиуса частиц сульфата на высоте около 20 км наводит на мысль, что фотохимическое окисление SO_2 не может быть важным механизмом для образования больших частиц на этом уровне. Рассмотрение химических факторов и времени нахождения частиц подтверждает, что ядрами Aitken'a в верхней тропосфере являются частицы сульфата, которые имеются в достаточном количестве и необходимого размера, чтобы объяснить образование больших частиц сульфата в окрестности и выше тропопаузы с помощью соединения ядер Aitken'a. Приближенные оценки распределения полной концентрации сульфатов с высотой, начиная с 5 км, дают широкий максимум в верхней тропосфере. Фотохимические и радиохимические данные о скорости окисления SO_2 в стратосфере подтверждают эту точку зрения. На основании экспериментов заключаем, что основная часть сульфата в атмосфере объясняется наличием частиц Aitken'a в тропосфере, и что нет стратосферного сульфатного слоя, а есть только стратосферный слой «больших частиц».