

Stratospheric Aerosol Modification by Supersonic Transport and Space Shuttle Operations—Climate Implications

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ABSTRACT

We have estimated the potential effects on stratospheric aerosols of supersonic transport emissions of sulfur dioxide gas and submicron soot granules, and space shuttle rocket emissions of aluminum oxide particulates. Recently, exhaust particles from large aircraft and rocket engines have been characterized experimentally, and we have adopted new data where appropriate. We use an interactive particle-gas model of the stratospheric aerosol layer to calculate changes due to exhaust emissions. We also employ an accurate radiation transport model to compute the effect of aerosol changes on the earth's average surface temperature. Our major conclusions are as follows. The release of large numbers of small particles (soot or aluminum oxide) into the stratosphere should *not* lead to a corresponding significant increase in the concentration of large, optically active aerosols. On the contrary, the increase in large particles is severely limited by the total mass of sulfate available to make large particles *in situ*, and by the rapid loss of small seed particles via coagulation. We find that a fleet of several hundred advanced supersonic aircraft operating daily at 20 km, or the launch of one space shuttle rocket per week, could produce roughly a 20% increase in the large-particle concentration of the stratosphere. We find, in addition, that aerosol increases of this magnitude would reduce the global surface temperature by less than 0.01 K, a negligible climate change.

1. Introduction

Interest has been growing in the role of terrestrial aerosols in the radiation budget and climate of earth, and in the possibility that civilization may be disturbing the natural balance of airborne particulates (Kellogg, 1977). For this reason, the effects on stratospheric aerosols of aircraft emissions of sulfur gases and soot particles were considered during the Climatic Impact Assessment Program (CIAP, 1975); it was estimated that a fleet of advanced supersonic transports emitting 3×10^7 kg SO_2 year⁻¹ between 18 and 21 km might reduce the earth's surface temperature by roughly 0.05 K. However, the CIAP conclusions were based on a simple residence time model for an aerosol of fixed size dispersion and, accordingly, were somewhat uncertain. The effects on stratospheric aerosols of space shuttle rocket motor emissions of aluminum oxide (Al_2O_3) particles were studied qualitatively by Hofmann *et al.* (1975a) who suggested, but could not prove, that such particles might cause a large enhancement in the aerosol layer.

Pollack *et al.* (1976a,b) also investigated the climatic impact of sulfur dioxide gas released by supersonic transports and aluminum oxide particles emitted by space shuttle engines. They found that the injection of 3×10^7 kg SO_2 year⁻¹ between 18 and 21 km might decrease the average global surface temperature by ~ 0.006 K. This temperature change is much smaller than that estimated by CIAP (1975) for several reasons. The ambient aerosol layer assumed by Pollack *et al.* consisted of smaller (more transparent) particles on the average, and the related temperature calculations accounted for infrared radiation trapping by aerosols, which was neglected by CIAP. In addition, Pollack *et al.* omitted certain feedback mechanisms, such as increased ice formation leading to enhanced surface albedo, from their model. Finally, Pollack *et al.* adopted a residence time for particles in the altitude range from 18 to 21 km that was about half the CIAP value. With regard to the space shuttle, Pollack *et al.* (1976a,b) determined that 50 launches per year could lead to a surface temperature reduction of the order

of 10^{-4} K. The accuracy of the space shuttle and aircraft calculations made by Pollack *et al.* (1976a,b), however, were limited by an incomplete treatment of aerosol physics and the neglect of soot emissions.

In the present study, we expand on these earlier investigations. Specifically, we consider the impact on the stratospheric aerosol layer of exhaust emissions from a hypothetical fleet of advanced supersonic transports (SST's), and from planned launches of space shuttle vehicles (SSV's). For calculating aerosol changes, we utilize a new and detailed model of the aerosol layer (Turco *et al.*, 1979; Toon *et al.*, 1979). For determining the related impact on global surface temperatures, we employ an improved radiation balance model (Pollack *et al.*, 1976a,b,c). We have chosen to study the stratospheric aerosol layer because it is believed to be quite sensitive to small anthropogenic perturbations (Hofmann *et al.*, 1975a; Rosen *et al.*, 1978; Turco *et al.*, 1979), and is thought to influence the global climate (Pollack *et al.*, 1976b,c). Moreover, the ambient characteristics of the layer are relatively well-established observationally (Junge, 1961; Lazrus and Gandrud, 1974, 1977; Hofmann *et al.*, 1975b; Rosen *et al.*, 1975; Toon and Pollack, 1976; Farlow *et al.*, 1977).

2. Engine emissions of gases and particles

Supersonic transport engines emit gaseous sulfur oxides and carbonaceous particulates as exhaust constituents. The sulfur oxides are generated thermochemically from residual organic sulfur compounds found in most aviation fuels, while the soot is produced at high temperature in the engine combustors. Soot particles add directly to the natural aerosol loading of the atmosphere, while sulfur gases add indirectly through condensation on preexisting particles.

For the gaseous sulfur component, we adopt an emission index of $1 \text{ g SO}_2 \text{ kg}^{-1}$ fuel from CIAP (1975). This is only a rough value based on an average amount of residual sulfur found in a wide variety of fuels. Inasmuch as sulfur is a recoverable component of such fuels, sulfur emissions to the stratosphere are readily controllable, although removal adds to the cost of the fuel. For our calculations, we assume an SST fleet of 300 aircraft of advanced engineering design. It has been estimated that each advanced SST would consume roughly $38\,000 \text{ kg fuel h}^{-1}$ and be in operation 7 h day^{-1} (e.g., see Poppoff *et al.*, 1978). Accordingly, our assumed fleet of 300 aircraft would release $\sim 2.9 \times 10^7 \text{ kg SO}_2 \text{ year}^{-1}$ worldwide.

We average the SO_2 emissions over the globe, because advanced SST's are expected to fly in both hemispheres, and we inject the SO_2 at 20 km, the likely cruise altitude for future supersonic aircraft (Poppoff *et al.*, 1978). Our horizontal averaging

of exhaust emissions is necessary because our analysis is made with a one-dimensional model. Normally, the zonal dispersion of pollutants injected into the stratosphere occurs within a few days, while meridional spreading may take a few months. By comparison, stratospheric residence times for gases and aerosols are a year or more (CIAP, 1975). Hence, global-scale averaging of continuously or frequently injected materials is a reasonable assumption in a study of global responses.

The soot emission of a J79-GE-10B jet engine operating at military power in a test chamber has been measured by the Naval Environmental Protection Support Service (NEPSS, 1977); the observed size distribution is shown in Fig. 1. The corresponding emission index ($\sim 0.3 \text{ g soot kg}^{-1}$ fuel) is a factor of 3 larger than that previously estimated for advanced aircraft by Grobman and Ingebo (1974).

The rate of mass addition to the aerosol layer due to soot emission ($\sim 0.3 \text{ g kg}^{-1}$ fuel) is much less than that due to sulfur emission ($\sim 2 \text{ g kg}^{-1}$ fuel) once the SO_2 is converted into a sulfuric acid-water solution with a 75% H_2SO_4 weight fraction [corresponding roughly to the observed composition of stratospheric aerosols (Rosen, 1971)]. Despite their small mass, it is conceivable that many of the soot particles released in the stratosphere could serve as condensation nuclei and eventually grow into large aerosol droplets. However, using our model calculations, we later demonstrate that soot coagulates rapidly enough with existing particles to restrict its role as seed for new aerosols.

Space shuttle launch vehicles will be propelled by rocket motors burning a solid fuel mixture of ammonium perchlorate and elemental aluminum with traces of other additives (Potter, 1977). About 30% of the exhaust mass is aluminum oxide, and almost all of this condenses into an Al_2O_3 particulate.

Based on theoretical estimates of space shuttle fuel consumption rates during launch, Hofmann *et al.* (1975a) have determined the rate of Al_2O_3 mass deposition as a function of altitude for 50 shuttle launches per year. Their deposition rate is expressed in terms of the mass of Al_2O_3 which is added to each cubic kilometer of air at a given height per year, averaged over the Northern Hemisphere. Because the residence time for particles in the stratosphere is only about a year or two, slow interhemispheric transfer of shuttle exhaust particles can be ignored (Hofmann *et al.*, 1975a). The exhaust mass deposition profile derived by Hofmann *et al.* is approximately the same as that derived from revised projections of space shuttle fuel consumption rates (Potter, 1977). Consequently, we adopt the average Al_2O_3 mass deposition rates of Hofmann *et al.* for the present assessment.

Aluminum oxide particles have been sampled in the wakes of Titan rockets burning a solid fuel similar

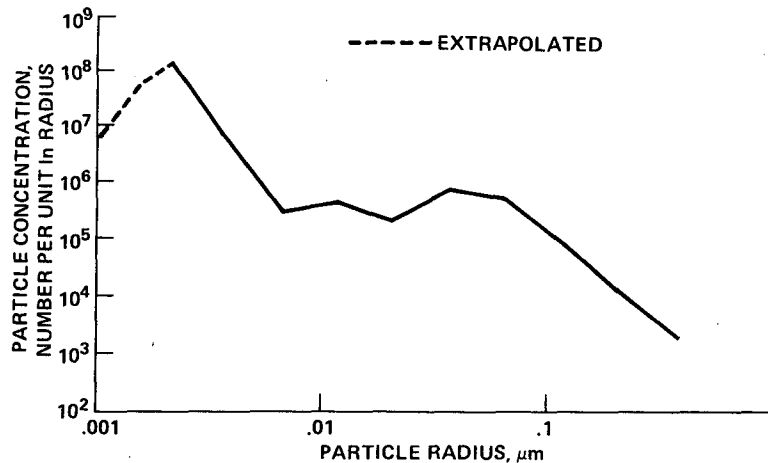


FIG. 1. The differential size distribution (the number of particles per unit in radius) of soot particles found in 22 700 m³ of exhaust (and entrained air) generated by a J79-GE-10B jet engine burning 227 kg of fuel at military power in a sea-level test chamber (NEPSS, 1977). Our extrapolation of the measured particle abundances to smaller sizes is also indicated.

to that proposed for the space shuttle. The observed particle characteristics have been summarized by Potter (1977). The particulate matter was found to consist mainly of smooth, relatively pure aluminum oxide spherules. Mass densities determined for a few individual particles were between 1.5 and 3.5 g cm⁻³ (compared to ~4 g cm⁻³ for pure aluminum oxide) even though most of the collected particles appeared to be solid. For this assessment, we choose an average mass density of 2 g cm⁻³ for Al₂O₃ particles (this leads to the release of twice as many particles per unit mass of fuel burned as when 4 g cm⁻³ is used).

The average particle size distribution (particles cm⁻³ μm⁻¹ radius) measured by wire and tape impactors and a mobility analyzer in several Titan wakes varied approximately as $r^{-4.5}$ in the size range 0.035 ≤ r ≤ 5 μm, and decreased rapidly for sizes < 0.035 μm radius. However, the rapid decrease below 0.035 μm may be incorrect because a condensation (Aitken) nuclei counter simultaneously detected at least 2–3 times as many particles in the wakes as were implied by the number of collected particles, and at least 50% more than were consistent with the rocket engine mass deposition rate and the observed size distribution, assuming a mass density corresponding to pure alumina (Potter, 1977). Impactor collection of small particles is normally quite inefficient, which could explain the greater number of small particles seen by the nuclei counter. Moreover, a lower average particle mass density (2 g cm⁻³, as is assumed in this study) would imply more particles released per unit mass of exhaust, which would bring into line the observed size distribution, total particle number and mass deposition rate. Thus, we believe that the apparent dis-

crepancy in observed total particles is largely due to the unresolved difficulties in collecting and sizing a highly dispersed particulate.

Hofmann *et al.* (1975a) have analyzed some of this same Titan exhaust data and derived an exponential size distribution [$\exp(-r/0.012 \mu\text{m})$] for the particulate; the distribution is similar to the $r^{-4.5}$ distribution in the size range 0.04 < r < 0.1 μm, but is somewhat lower for $r > 0.1 \mu\text{m}$. We adopt the exponential size distribution of Hofmann *et al.* for particle radii, 0.001 ≤ r ≤ 2.6 μm (the total size range treated in our model); the concomitant underestimate for $r > 0.1 \mu\text{m}$ is unimportant in this work. The exponential size distribution also emphasizes the number of condensation nuclei below 0.035 μm; thus, we can test the suggestion of Hofmann *et al.* that small particles might dominate the shuttle impact. We calculate the number of particles of each size released by rocket engines by normalizing the exponential size distribution to the exhaust mass deposited assuming a particle mass density of 2 g cm⁻³.

3. The aerosol model

We employ a one-dimensional model of gaseous and particulate atmospheric constituents to calculate the effects of SST and SSV effluents on the stratospheric aerosol layer. The numerical techniques utilized in the model are discussed in detail by Turco *et al.* (1979). Briefly, the particles are cataloged into "bins" according to their radius and altitude. The model has 35 radius categories ranging from 0.001 to 2.56 μm with particle volume doubling between sizes, and 30 altitude levels from zero to 58 km at 2 km intervals. The number of particles in each size-

altitude bin is evaluated at each time step during a calculation. Two types of particles are distinguished: aerosol droplets and condensation nuclei. The sizes of inclusions (cores) within the aerosol droplets are also computed; in the present model, these cores consist of condensation nuclei which have been nucleated into new droplets, or have coagulated with existing droplets. Cores can accumulate and coalesce within droplets, and are released when droplets evaporate.

The dominant source of sulfur for the (model) aerosol layer is OCS, which diffuses upward from the troposphere to the stratosphere where it is photolyzed into S and CO (Crutzen, 1976); the existence of OCS in the stratosphere has recently been confirmed by the measurements of Inn *et al.* (1979). The subsequent rate of formation of H₂SO₄ vapor, which is a precursor of sulfate aerosol particles, is limited principally by the association reaction of SO₂ with OH (Castleman *et al.*, 1975).

Some new laboratory evidence suggests that, in the atmosphere, sulfur radicals such as HSO₃·O₂·H₂O (or H₃SO₆) will be formed following the reaction of SO₂ with OH. Moreover, clusters of only two of these radicals can apparently act as nuclei for water vapor condensation in a supersaturated "cloud" chamber (J. P. Friend, private communication, 1978). The sulfur radicals, on entering a solution droplet or possibly even in the molecular phase, should react rapidly to form H₂SO₄ and other sulfates. We are led to this conclusion by the fact that SO₄⁻ is found to be the dominant oxidation state of sulfur in collected aerosol samples, and H₂SO₄/H₂O solutions have most of the observed spectroscopic and physical properties of natural aerosols (Lazrus and Gandrud, 1974, 1977; Toon and Pollack, 1976; Rosen, 1971). We have studied the changes in our model predictions caused by allowing sulfur radicals (HSO₃) to condense directly onto aerosols. We conclude that, as long as H₂SO₄ is the end product of sulfur radical chemistry, our approach assuming H₂SO₄ as the precursor of aerosol droplet formation and growth is appropriate for calculating the properties of aerosols > 0.01 μm. However, in order to be able to predict the concentrations of sulfur radicals in air as well as their precise effects on aerosol size dispersion and composition, we will have to incorporate species such as H₃SO₆ in our model when additional information about their photochemistry and solution properties becomes available.

Suitable theories are available to describe the nucleation, growth and coagulation of aqueous sulfuric acid solution droplets under stratospheric conditions. In our model, nucleation of supersaturated H₂SO₄ vapor occurs on the surfaces of condensation nuclei (Hamill *et al.*, 1977a,b); the condensation nuclei are transported by eddy diffusion

from the troposphere to the stratosphere. Whenever these nuclei are mixed into a supersaturated stratospheric environment or are deposited in such a region by aircraft or rocket engines, they are assumed to be nucleated in 10⁶ s. We are currently evaluating the influence on natural aerosol formation of H₂SO₄ homogeneous nucleation and nucleation onto ions, and sulfur radical nucleation; preliminary results suggest that these processes are more critical in the troposphere than in the stratosphere.

Aerosol droplets can grow by heteromolecular condensation of sulfuric acid and water vapors. We use a growth equation that is based on the formulation of molecular diffusion to particles developed by Fuchs and Sutugin (1971), suitably modified for heteromolecular condensation (Hamill *et al.*, 1977b). Coagulation is a growth mechanism which, for small droplets, can be more important than condensation. Accordingly, we have developed a detailed coagulation algorithm for our model, utilizing the coagulation kernels derived by Fuchs (1964). Particle gravitational sedimentation rates are calculated using the Stokes-Cunningham equation for the terminal velocity of spherical droplets in air (Kasten, 1968). Vertical particle diffusion rates are computed with an "eddy" diffusion coefficient (which is illustrated in Turco *et al.*, 1979).

The lifetime of an aerosol droplet against washout in the troposphere is assumed to be 1 day near the ground, with the inverse lifetime decreasing linearly to zero at the tropopause level (situated near 13 km in our model). For aerosol droplets in the upper troposphere, this leads to an effective washout lifetime of about one week.

Most of the physical and chemical processes that are treated in our aerosol model are summarized schematically in Fig. 2.

4. Aerosol model predictions: The ambient and perturbed stratosphere

Model simulations of the ambient stratospheric aerosol layer, and detailed tests of model sensitivity to a large number of physical parameters, are presented by Toon *et al.* (1979). The model predictions are in good agreement with observational data for the following aerosol parameters: the height distribution and magnitude of the total particle mass mixing ratio; the vertical concentration profile of total particulates; the aerosol size distribution; the mixing ratio of large aerosol particles (>0.15 μm radius); the number ratio of particles with radii >0.15 μm to those >0.25 μm; and the aerosol droplet sulfuric acid weight fraction.

In Fig. 3, for example, we compare calculated and observed large aerosol particle mixing ratios in the stratosphere. As the figure shows, the model

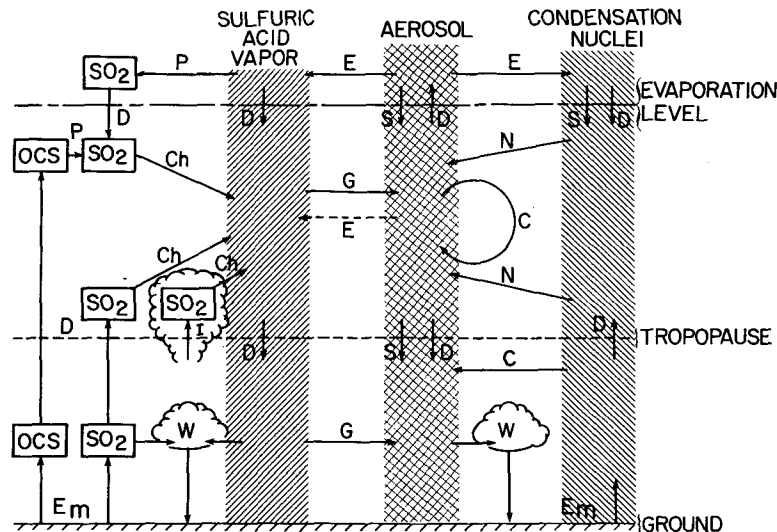


FIG. 2. A partial schematic outline of the physical and chemical interactions included in our one-dimensional model: C, coagulation; Ch, chemistry; D, diffusion; E, evaporation; Em, emission; G, condensation and growth; I, injection; N, nucleation; P, photolysis; S, sedimentation; W, washout and rainout.

prediction appears to represent a reasonable average of the observational values between 15 and 25 km [see Toon *et al.* (1979) for a complete discussion of model comparisons with data]. Incidentally, it is the large particles which dominate the radiation transport properties of stratospheric aerosols.

The effect of SST emissions on the large-particle mixing ratio is also shown in Fig. 3. (Note that all of the perturbation calculations reported here correspond to five years of continuous contaminant injection; for aerosols, these are essentially steady-state calculations because we verified that extension of simulation times to 10 years causes less than a 10% adjustment in the calculated *perturbations*.) The most pronounced effect of SST's is due to sulfur gas emission, not soot emission. The soot particles, which are quite small on the average, coagulate rapidly with aerosols of all sizes; likewise, sulfur gas is absorbed by existing particles of all sizes. Therefore, the overall effects of soot and SO_2 on existing aerosols are alike in that both pollutants cause the existing particles to grow in size, with their relative importance being largely determined by the relative mass injected. Interestingly, the predicted changes in the aerosol layer lie within the range of variability (or uncertainty) in current measurements (see Fig. 3), and might therefore be difficult to detect. Also, note that SST emissions affect the entire aerosol layer because of vertical dispersion by diffusion from the height of injection (20 km in this case).

The addition of very small soot particles to the stratosphere could, in theory, increase the abundance of 0.1–0.3 μm aerosols despite the small total mass

added. This is so because small particles with large effective surface area efficiently absorb injected sulfur gas as they grow and coagulate into the middle size range ($r \approx 0.1\text{--}0.3 \mu\text{m}$). The number of large particles produced by soot emission would be maximized if coagulation did not occur and if the sulfur gas supply were sufficient to allow every newly formed droplet to grow at a rate as fast as $0.5 \mu\text{m year}^{-1}$. However, the coagulation lifetime of added soot particles and the small aerosols that are nucleated on them (~ 1 month) is much shorter than the time required to grow a large particle (~ 1 year). Moreover, as Toon *et al.* (1979) have pointed out, the natural supply rate of sulfur to the stratosphere via transport from the lower atmosphere is quite restricted, as is the supply rate of injected sulfur for the pollution cases treated here, and this also limits the number of new particles that can grow to large size in the model. It is unlikely that the natural supply rate of sulfur to the aerosol layer is much larger than that simulated by our model (equivalent to $\sim 1\text{--}2 \times 10^5$ metric tons of $\text{SO}_2 \text{ year}^{-1}$) because of the implications for the observable properties of the ambient aerosols. Accordingly, the release of soot particles in the stratosphere can only result in a small increase in large particles, by the mechanism of combined growth and coagulation described above.

Novakov *et al.* (1974) have demonstrated that soot particles can efficiently oxidize SO_2 into sulfate via a catalytic process involving oxygen molecules adsorbed on the particle surfaces: Soot released in the stratosphere could therefore act as centers for heterogeneous nucleation and heterogeneous chemi-

cal formation of sulfates. Nevertheless, the end result is the same, as these particles (whether they are dry soot or soot particles coated with sulfuric acid) will quickly coagulate with each other and with pre-existing aerosol particles. Because aerosol production is limited by the availability of sulfur-bearing gases in the stratosphere, the total aerosol mass is largely independent of the nucleation mechanisms operating. Further, as implied earlier, an increased rate of SO₂-to-sulfate conversion would not greatly affect the model predictions made here (precluding the possibility of rapid particle formation in the near wake of the aircraft). In the upper troposphere, on the other hand, where large quantities of soot are continually supplied by commercial air traffic, and where SO₂ is much more abundant, a substantial

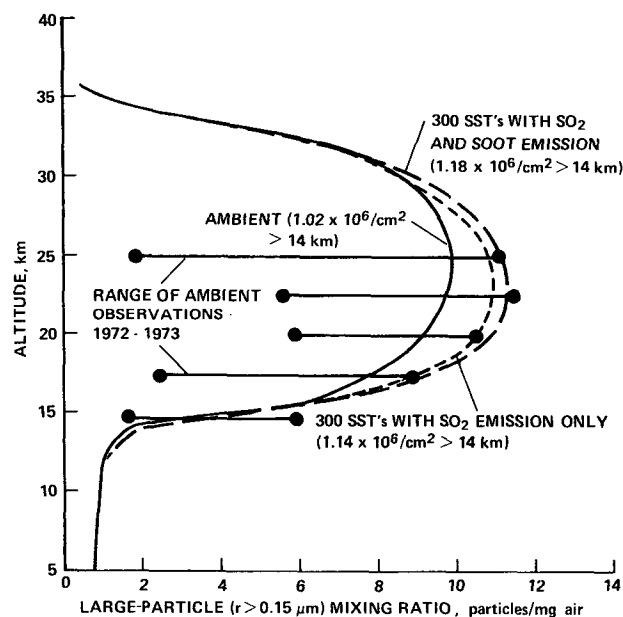


FIG. 3. Supersonic transport effects on large aerosol particles. Calculated steady-state large particle ($r > 0.15 \mu\text{m}$) mixing ratios in the stratosphere are shown for 20 km SST flight, both with and without soot emission. The global exhaust injection rates are: SO_x ($2.9 \times 10^7 \text{ kg SO}_2 \text{ year}^{-1}$); soot ($8.7 \times 10^6 \text{ kg soot year}^{-1}$) with the soot size distribution of Fig. 1. The ambient model large particle mixing ratio profile and some observational data are included for comparison. The measurements were made at northern midlatitudes during 1972-73 with balloonborne optical particle counters that discriminate against particles $< 0.15 \mu\text{m}$ (Hofmann *et al.*, 1975b; Rosen *et al.*, 1975). Shown is the envelope of all measurements. Individual measurements reveal that the layer is highly stratified with 50% variations in mixing ratio over 1 km vertical distances. The stratospheric data indicate little seasonal trend. A mixing ratio maximum generally occurs around 24 km at the equator and 17 km over the poles. The maximum mixing ratio is 50% greater at the equator than at the poles. We chose to illustrate data from 1972-73 since the total number of particles was near a minimum and the data were thus the least volcanically perturbed data available. For each calculated profile, the total stratospheric column concentration of large particles is given in brackets.

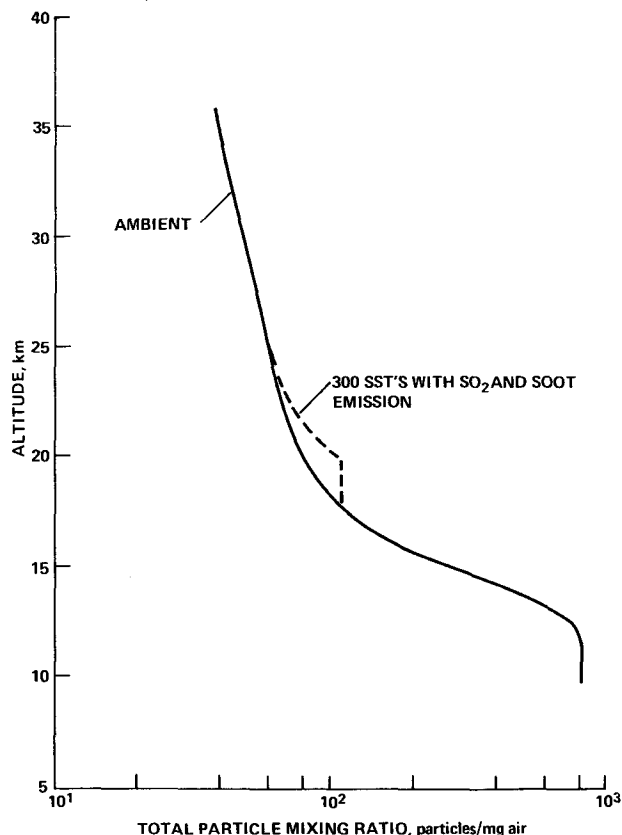


FIG. 4. Supersonic transport effects on the total mixing ratio of stratospheric particles.

number of new aerosol particles might be formed around soot. This problem is beyond the scope of the present work, however.

With regard to SST's, then, it appears that sulfur gas emissions cause a much larger increase in the total aerosol mass and large-particle concentration than soot emissions. Note, by comparing the results in Fig. 3, that if the sulfur component of aviation fuel were eliminated, the net effect of soot emission alone on large aerosol particles would be quite small (the soot effect is roughly independent of SO₂ emission for the injection levels considered).

Further insight into the effects of SST's on aerosol particles can be gained from Figs. 4 and 5. Fig. 4 shows that the total number of stratospheric particles is increased only slightly by SST's, despite the large number of small particles injected. Fig. 5 compares the total number of soot particles of each size that are injected into a cubic centimeter of air at 20 km during one year of flight activity to the ambient and perturbed steady-state particle size distributions. The comparison illustrates that, because the soot particles are injected near 20 km altitude where fairly large numbers of acid droplets exist, the soot is rapidly depleted by coagulation. Note in Figs. 1 and 5 that the soot size distribution is some-

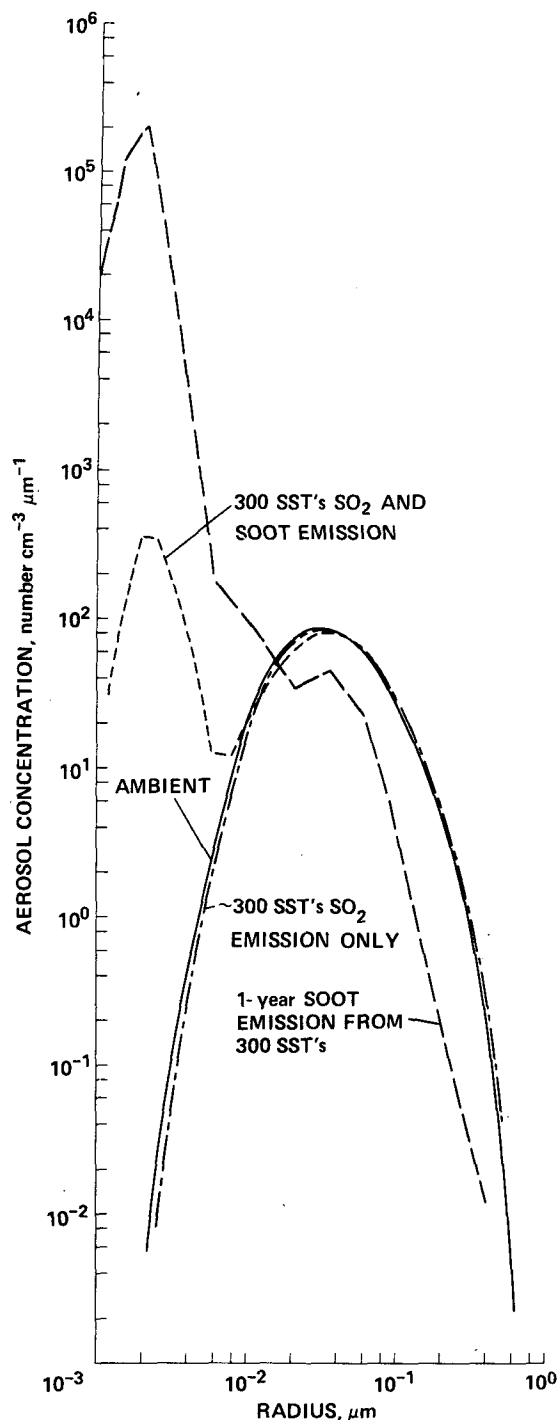


FIG. 5. Predicted aerosol particle size distributions at 20 km for the same SST injection cases as in Fig. 3. Also shown is the quantity of soot released at 20 km during one year by the assumed SST fleet.

what deficient in the range between 0.01 and 0.03 μm . Particles in this range would be quite efficient (per unit mass injected) in producing additional large stratospheric particles. The increase in aerosol

mass caused by SST emissions is difficult to discern in Fig. 5 because it is only $\sim 20\%$ (similar to the increase in large particles shown in Fig. 3), which implies only $\sim 5\%$ increase in the average particle radius.

The effect of regular space shuttle traffic on the large aerosol particle concentration of the stratosphere is illustrated in Fig. 6. Apparently, planned space shuttle activity could produce an effect as large as 300 SST's. It was originally proposed by Hofmann *et al.* (1975a) that the numerous small particles emitted by space shuttle engines might act as seeds from which new large particles could be grown in a supersaturated environment. In fact, the total particle mixing ratio profiles in Fig. 7 show a marked increase due to space shuttle emissions, particularly at high altitudes. However, the effect on large particles is much smaller. The tiny Al_2O_3 seed particles coagulate quite rapidly with one another and with preexisting aerosols; in this way, the number of new stratospheric particles which survive to grow to large size is severely restricted. Occasional space shuttle launches could, therefore, cause only a limited change in the natural sulfate layer. For example, our calculations indicate that 50 shuttle launches per year might increase the total number of large particles by about 20%. As in the case of SST's, the predicted aerosol changes in Figs. 6 and 7 would be practically indiscernible against the natural variations of the layer.

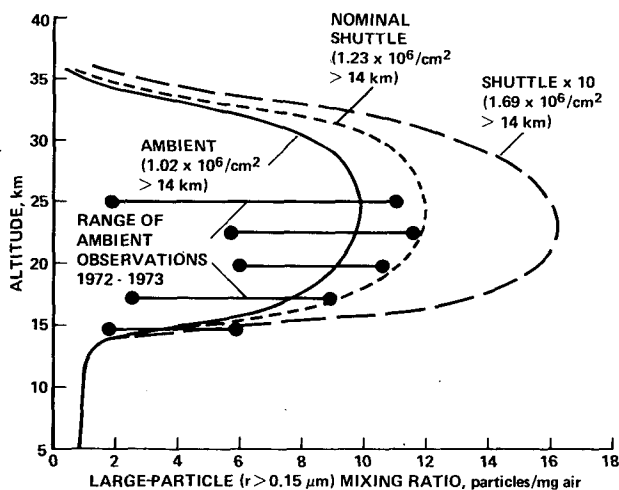


FIG. 6. Space shuttle effects on large aerosol particles. Calculated steady-state large particle ($r > 0.15 \mu\text{m}$) mixing ratios in the stratosphere are shown for nominal space shuttle operations, assuming the particulate deposition rate described in the text [basically corresponding to 50 shuttle launches per year with the mass emission averaged over the Northern Hemisphere, an $\exp(-r/0.012 \mu\text{m})$ particle size spectrum, and a particle mass density of 2 g cm^{-3}]. Also shown are results for 10 times the nominal shuttle effluent. The ambient model large particle mixing ratio profile and some observational data are included for comparison.

If we assume 10 times as many space shuttle launches per year as are planned (i.e., roughly 500 launches per year), the effect on the large aerosol particle concentration is much greater, but not nearly 10 times as great (see Fig. 6). Again, coagulation rapidly depletes the population of newly injected particles and, moreover, enhanced particle surface area decreases the concentration of sulfur vapor needed to grow new large particles. A secondary effect caused by the injection of numerous small particles is the reduction of the average particle size and thus the mass loss rate due to gravitational sedimentation.

The total number of particles injected into the stratosphere each year by 50 space shuttle launches ($\sim 3.2 \times 10^7 \text{ cm}^{-2}$) is roughly the same as the number injected by a fleet of 300 SST's ($\sim 4.9 \times 10^7 \text{ cm}^{-2}$). However, comparing Figs. 4 and 7, we see that the space shuttle causes a much larger enhancement in total particles. This occurs in part because the shuttle injects Al_2O_3 particles at high altitudes where there are few aerosol droplets to remove them by coagulation. Comparison of condensation nuclei (CN) measurements (which are essentially measurements of the total particle concentration, because both nuclei and aerosol droplets are detected by a CN counter) with the predicted increase in total particles at high altitude indicates that the shuttle could have a significant impact on the total particle concentration. However, existing CN measurements are subject to considerable uncertainty. For example, it is possible that meteors ablating in the mesosphere regularly generate large numbers of CN which can settle into the stratosphere, causing the ambient particle concentration to correspond more closely to the upper-limit measurements shown in Fig. 7. If this is true, the shuttle injection would cause only a small perturbation of the background particle concentration.

It is also interesting to compare the effects of space shuttles and SST's on the aerosol size distribution (Figs. 5 and 8). Shuttles inject more particles in the range from 0.01 to 0.1 μm than SST's, which increases the number of acid droplets near 0.1 μm by redistributing the available acid mass from larger particles ($r > 0.3 \mu\text{m}$) to smaller particles ($r < 0.3 \mu\text{m}$). This is indicated by the crossover of the ambient and perturbed size distributions near 0.3 μm in Fig. 8. Nonetheless, this shift in particle sizes still results in a small net increase in total particles above 0.15 μm . SST's inject many more particles below 0.01 μm than space shuttles, but as mentioned earlier, the fate of most of these is coagulation with larger aerosols. The uptake of very fine soot granules causes all of the aerosol particles to grow more or less uniformly.

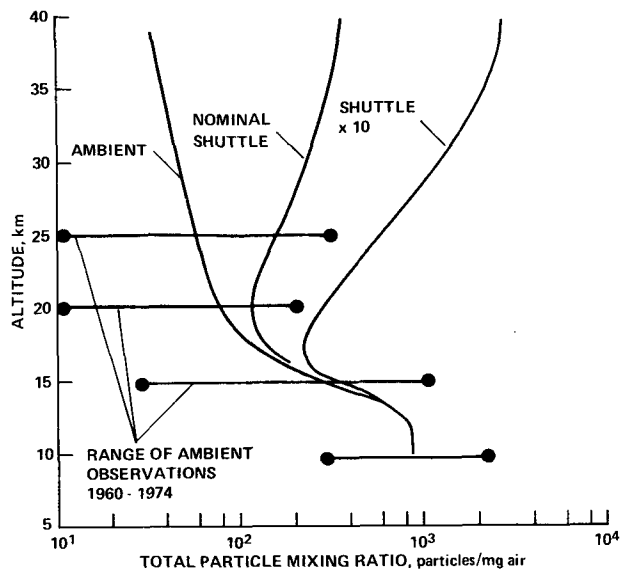


FIG. 7. Space shuttle effects on the total mixing ratio of stratospheric particles. Computational results are given for the same cases as in Fig. 6. The ambient model prediction and the range of observational data (Junge *et al.*, 1961; Käselau *et al.*, 1974) are also indicated.

5. Radiative transfer calculations of climate effects due to aerosol perturbations

In previous studies, Pollack *et al.* (1976a,b) considered the climatic impact of Al_2O_3 particles injected into the stratosphere by 50 space shuttle launches per year, as well as the impact of SO_2 injected by various numbers of SST's flying in the stratosphere. The present study represents an improvement over this earlier work in several respects.

For one thing, Pollack *et al.* (1976a,b) were not able to model the stratospheric aerosols. Therefore, they could not consider the interaction of aluminum oxide particles with ambient stratospheric aerosols. Pollack *et al.* also neglected soot emission by SST engines and its interaction with aerosol droplets. Moreover, Pollack *et al.* had to assume a fixed stratospheric particle size distribution, and could only roughly estimate optical depth changes due to exhaust emissions because of particle residence time variations with altitude. Here we include both the gaseous and particulate emissions of SST's and SSV's, and explicitly treat gas-particle and particle-particle interactions. The present model, in addition, predicts most of the physical parameters needed to make an accurate calculation of aerosol optical properties.

The second major improvement in this work is that we use a fully convergent radiative-convective solution for the vertical temperature profile. In our earlier work we assumed that the shape of the tem-

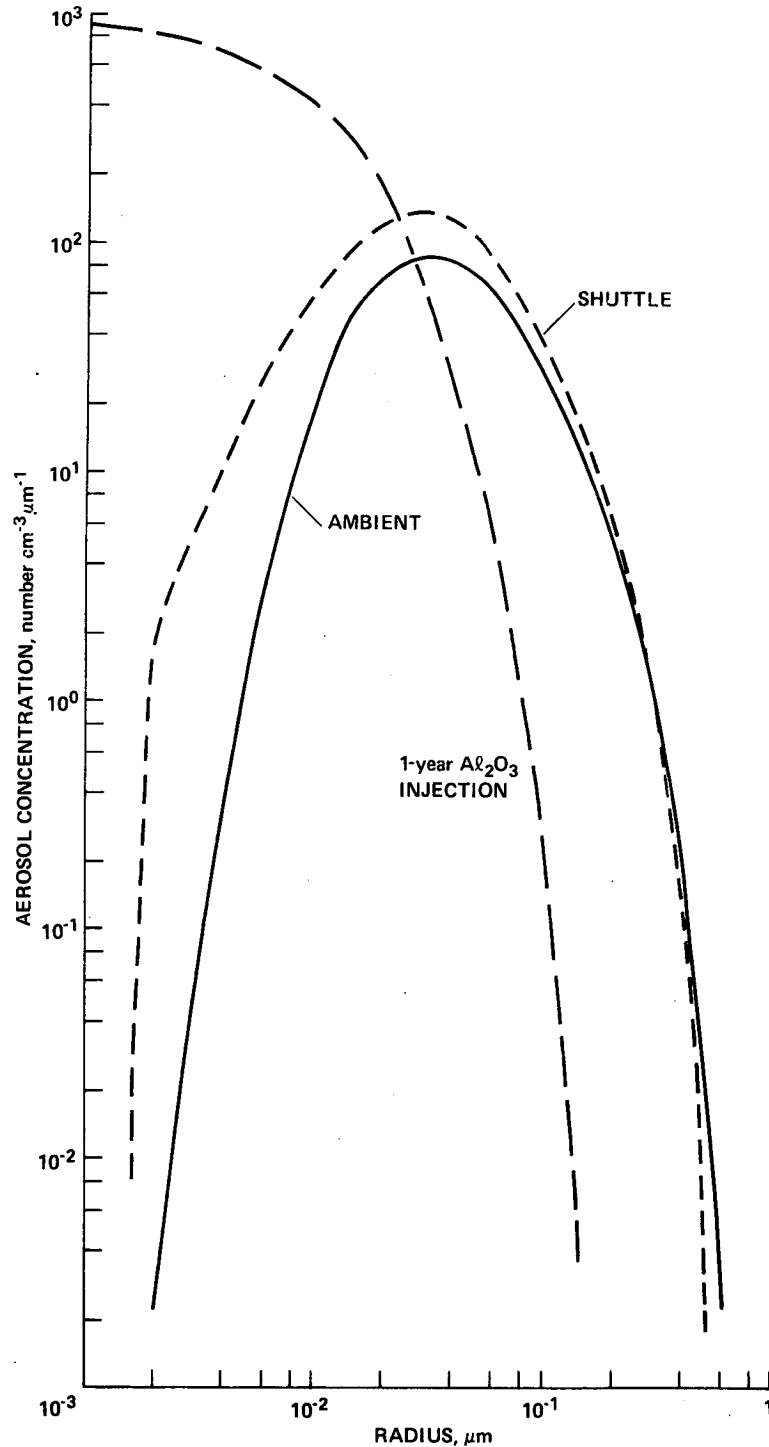


FIG. 8. Predicted aerosol particle size distributions at 20 km for nominal shuttle operations. Also shown is the quantity of aluminum oxide particulate deposited at 20 km during one year of shuttle flights.

perature profile was invariant, and calculated the change in surface temperature by achieving a flux balance between incoming solar energy and emitted infrared radiation at the top of the atmosphere.

To calculate the climatic effects of aerosols injected by SST's and space shuttles, we utilize the altitude-dependent size distributions predicted by our model in the doubling routine described by

Pollack *et al.* (1976a). The doubling calculations are highly accurate multiple-scattering computations that explicitly account for solar energy absorption by CO_2 , O_3 , O_2 and H_2O , and absorption and scattering by aerosols in the stratosphere and troposphere. In this way, we obtain the altitude-dependent solar energy deposition rate for perturbed and unperturbed conditions. For stratospheric aerosols we use the optical constants of a 75% sulfuric acid aqueous solution, and for condensation nuclei we use the optical constants of $(\text{NH}_4)_2\text{SO}_4$. Of course, the soot grains from SST's and the Al_2O_3 particles from SSV's have optical constants that differ from natural condensation nuclei. However, most atmospheric CN are found in the troposphere; the fraction residing in the stratosphere, either by number or mass, is extremely small, both for ambient and perturbed conditions. Accordingly, our results are not sensitive to the optical constants assumed for soot and Al_2O_3 particles. For tropospheric particles, which are held fixed, we use the model of Toon and Pollack (1976).

Once the solar energy deposition rate profile is determined from the doubling calculations, an infrared radiation transport calculation is performed to achieve radiative-convective equilibrium. This routine uses the same numerical techniques described by Pollack *et al.* (1976a) to calculate the infrared radiation emitted and absorbed by H_2O , CO_2 , O_3 and aerosols. Scattering by aerosols is ignored because of the small single-scattering albedo. The routine iterates to obtain radiative equilibrium in the stratosphere with relative humidity, cloudiness (50% cloud cover) and cloud top height held fixed. Convergence is assumed when the difference between the net upward infrared heat flux and the total solar flux absorbed below a given level is less than 10^{-6} of the solar flux absorbed below that level. The surface temperature change between the next to last and the final iteration is always much less than 10^{-3} K, although at high altitudes (above 50 km) temperature changes are sometimes several tenths of a degree; the slower temperature convergence above 50 km has a negligible effect on convergence at the ground.

We find that the global surface temperature decrease to be expected between current conditions and those with a fleet of 300 SST's flying 7 h day⁻¹ at 20 km is on the order of 3×10^{-3} K. (Between the ambient and perturbed solutions, at the final iteration no differences in computed temperatures $>10^{-2}$ K were found at higher altitudes, and even these might have been reduced if the convergence had been carried out further.) By comparison, the data of Pollack *et al.* (1976a,b) suggest that the same number of aircraft could lower the surface temperature by $\sim 6 \times 10^{-3}$ K. Evidently, our more sophis-

ted calculations confirm that the earlier estimates of Pollack *et al.* were quite reasonable.

We have also performed aerosol model, doubling routine and infrared transfer calculations for the case of 500 space shuttle launches per year. Again, we find a possible temperature decrease of only $\sim 3 \times 10^{-3}$ K. Because of the extremely small magnitude of this change, we did not perform explicit calculations for the temperature changes due to the nominal space shuttle launch rate. However, scaling from the calculated optical depth changes suggests that a nominal launch rate of 50 shuttles year⁻¹ might decrease the surface temperature by $\sim 3 \times 10^{-4}$ K. Our results are similar to the findings of Pollack *et al.* (1976a), who reported that the surface temperature changes due to space shuttle operations would amount to $\sim 1 \times 10^{-4}$ K. Plainly, such temperature changes are so small as to be completely undetectable. We may conclude, therefore, that the release of moderately large numbers of Al_2O_3 particles by rocket engines should not dramatically alter the properties of stratospheric aerosols.

6. Uncertainties in the predictions

From the preceding discussion, it should be apparent that there are numerous sources of uncertainty in our calculations. Due to a lack of detailed information, it is difficult to quantify such uncertainties in a statistical sense. Accordingly, we will only outline the major sources of uncertainty and make some subjective estimates of their magnitudes to guide the reader. It should be kept in mind that, in many areas of atmospheric science, calculations bearing an uncertainty factor as large as 2 or 3 are still considered to be quite accurate.

Aerospace engine emissions are not very well defined (see our previous discussion of the data). For SST's, the rate of soot ejection is uncertain by at least a factor of 2. Moreover, the number of small soot particles released is undetermined to within a factor of 2 or 3. The SO_2 emission index varies widely between aviation fuel lots and could differ by a factor of 2 from our adopted value of 1 g SO_2 kg⁻¹ fuel. The number of Al_2O_3 particles emitted by space shuttle rocket motors is unknown by a factor of 2 or 3, although the mass deposition rate is fairly well established by design specifications. The size distributions of particles released by large rocket and aircraft engines are also poorly known. Careful measurements of these exhaust parameters using prototype aircraft and shuttle engines would help to reduce some of the related assessment uncertainties.

The aerosol model introduces additional uncertainty into the calculations; most of the uncertainty sources are discussed by Turco *et al.* (1979) and Toon *et al.* (1979). Because the model has a one-

dimensional structure and incorporates physical parameters corresponding to middle latitudes, it presumably represents average global or hemispherical conditions. While this interpretation of the model is only partially valid, Turco *et al.* (1979) and Toon *et al.* (1979) argue that the global uniformity of the stratospheric aerosol layer allows an accurate description with a one-dimensional model. Even so, many of the components of normal temporal and geographical variability cannot be treated with such a model and, as a result, only globally averaged steady-state predictions are given here. In addition, there is the question of the significance of *in situ* aerosol nucleation from injected sulfur gases, which we have not yet dealt with exhaustively but which seems unimportant based on our simulations of large stratospheric injections of soot and Al_2O_3 particles (which act as condensation nuclei). Despite such reservations, however, we feel that the aerosol model reasonably simulates the natural sources and sinks of stratospheric sulfate particles, which is a critical factor in this assessment (see the discussion in Section 4). Therefore, our computed aerosol changes are probably uncertain by no more than a factor of 3.

Uncertainties in the radiation transport calculations are carefully outlined in the papers by Pollack *et al.* (1976a,b,c). The current improved radiation calculations are thought to be accurate to within a factor of 2.

We estimate an overall uncertainty range of roughly a factor of 5 for our predicted global temperature reductions caused by aerospace activity, although unidentified sources of systematic uncertainty outside this range are not precluded. While the overall uncertainty is large, it is not unusual in atmospheric science. Inasmuch as the predicted climate changes are far below the threshold for detection, the large residual uncertainties in the magnitude of the changes should have little significance.

7. Summary and conclusions

We have made some new, and more detailed, calculations of the effects of supersonic transport and space shuttle exhaust emissions on the stratospheric aerosol layer and on the surface temperature of earth. For this purpose, we have employed an interactive gas-particle model of the sulfate aerosol layer and a comprehensive simulation of terrestrial radiation transport processes. We consider both gaseous and particulate emissions of aircraft and rocket engines which might affect the natural aerosols. Basically, we conclude that, although likely future levels of aerospace activity could exert a moderate influence on the stratospheric aerosol layer, such activity would have a negligible impact

on the climate of earth. For example, 300 SST's used in daily service, or 50 space shuttle launches per year, are estimated to increase the large particle concentration of the stratosphere by only $\sim 20\%$, and to decrease the planet's average surface temperature by < 0.01 K. These estimates are based on computer model simulations and must, accordingly, be considered as tentative. Interestingly, despite the greater sophistication of the models used here, our new predictions are very close to those made several years ago by Pollack and co-workers (1976a,b).

With our more complete models, however, we have been able to resolve several issues related to the effects of large-scale injections of very fine particles into the stratosphere. We find that the number of small particles which can grow freely to large size in the stratosphere is quite limited. Indeed, the total number of large particles is restricted by the supply rate of sulfur to the aerosol layer. This rate is presently controlled by natural processes, and amounts to $\sim 1-2 \times 10^5$ metric tons of SO_2 year⁻¹ (Lazrus and Gandrud, 1974; Hofmann *et al.*, 1975b); by contrast, only about 0.29×10^5 metric tons of SO_2 year⁻¹ would be released by several hundred advanced SST's. Because of the dominance of natural processes, the total mass of the sulfate layer and the number of large particles comprising it are relatively invariant for the conditions studied. The fate of small soot and Al_2O_3 particles added to the aerosol layer is primarily coagulation with larger particles. Thus, it appears that SST emissions of SO_2 , not soot, would have the greatest impact on stratospheric aerosols, and SO_2 emissions can be regulated if necessary.

Perhaps a more important issue than the direct modification of the aerosol layer by *in situ* contamination is the possibility of indirect modification due to man's industrial activity. For example, increases in the background concentrations of SO_2 and OCS caused by expanding rates of fuel combustion and coal gasification could enhance the quantity of particulate sulfur in the stratosphere. We are presently looking into some of these possibilities.

There are other questions related to climate modification by aerosols that should be addressed in the near future. For example, it is possible that changes in condensation nuclei abundances caused by heavy aircraft traffic in the upper troposphere and lower stratosphere might alter the frequency, persistence and extent of cirrus cloud formations? Atmospheric scientists are understandably concerned about such problems.

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