A Steady-State Stratospheric Aerosol Model

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ABSTRACT

This paper deals with the development of a one-dimensional steady-state stratospheric aerosol model and the subsequent perturbations caused by including the expected space shuttle particulate effluents in the model. Two approaches to the basic modeling effort have been made: in one, enough simplifying assumptions were introduced so that a more or less exact solution to the descriptive equations could be obtained; in the other, very few simplifications were made and a computer technique was used to solve the equations. The most complete form of the model contains the effects of sedimentation, diffusion, particle growth and coagulation. The results indicate that the model is capable of describing many aspects of the stratospheric aerosol layer, such as size distribution and the vertical profile of particles >0.3 μm diameter.

1. Introduction

It is well known that a relative maximum exists on a global scale in the aerosol mixing ratio for particles having a diameter $d>0.3$ μm at $\sim$10 km above the tropopause. The particles are apparently composed of H_2SO_4 with many of them containing smaller solid inclusions. Following a large volcanic eruption the aerosol layer may experience a significant increase in concentration; but during quiet periods of volcanic activity the layer could approach a quasi-steady-state distribution. It is the purpose of this paper to present a one-dimensional model describing this steady-state situation and to investigate perturbations of this state.

2. The model

At the heart of the model is the assumption that a supersaturated layer of H_2SO_4 exists about 10 km above the tropopause. Although the model itself is not concerned with the chemistry of this layer's source, it could be formed from sulfur-bearing gases such as SO_2 or CSO diffusing up through the tropopause and after a series of chemical reactions eventually forming H_2SO_4; or the source could be continual small volcanic eruptions with the required effective injection altitude. Since H_2SO_4 has a very low vapor pressure at stratospheric temperatures and water vapor concentrations (Gmitro and Vermeulen, 1963), even a very modest production rate of H_2SO_4 could produce a large supersaturation.

The model further assumes that this saturated vapor condenses on any particles that are present at a rate governed by the thermal flux of H_2SO_4 molecules onto the particles' surface. Replenishment of the particles comes from diffusion of tropospheric aerosol upward, and as an option other sources can also be included. The effect of coagulation is also taken into account by the model.

In the present work, evaporation of particles has been neglected for what we feel are justifiable reasons. The expected saturation vapor pressure profile of H_2SO_4, illustrated by Hamill *et al.* (1977b) using extrapolated data from Gmitro and Vermeulen (1963), shows that with the known H_2SO_4 mass loading, only a very small amount of evaporation of the aerosol present is required to maintain a saturated atmosphere below 30 km. At $\sim$35 km and above, there is not enough available mass in H_2SO_4 to maintain 100% saturation and particles will tend to evaporate down to their core size. This transition region from saturated to unsaturated conditions is quite sharp, because the saturation vapor pressure of H_2SO_4 is increasing quite rapidly with altitude. It is apparent that this region will act as an effective sink for particles that have grown in the 20 km layer. However, this sink has essentially no effect on the aerosol profile in the vicinity of 20 km, because it is so far away that only a small percentage of these particles could even reach 35 km under the influence of sedimentation and diffusion. The shape of the aerosol profile in the vicinity of 35 km would be affected by the presence of the effective sink and for that reason the model presented here should not be considered entirely realistic above 30–35 km. On the other hand, this is above the main region of interest and should have little effect on the major results presented. The inclusion of evaporation effects would increase to a considerable extent the complexity and uncertainties in the computer model. The basic reason for this is that the particles cannot evaporate to a size smaller than that of their original

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core. Since individual particle identities are usually lost in the mathematical description of the diffusion process, the original core size is unknown.

As noted above, the H_2SO_4 profile is treated as an adjustable parameter rather than deriving it from an appropriate chemical reaction model. Although this approach at first may seem unrealistic and questionable, the assumptions involved in present sulfur chemistry models (including the magnitude and type of sources) are simply too uncertain to produce a reliable H_2SO_4 vapor profile. For this reason, we have chosen to make the H_2SO_4 vapor profile a parameter that can be adjusted to obtain a good model fit to the observed aerosol profiles. It should be expected that similar results would be obtained from a more elaborate model containing the sulfur chemistry provided the H_2SO_4 vapor profiles in each case are similar. Such a comparison has been made and will be discussed in a later section.

3. Philosophy of one-dimensional models

In general the atmosphere should be treated as a three-dimensional system and the applicability of one-dimensional models is open to considerable debate. Under present circumstances the limited amount of detail that can be included in three-dimensional models severely detracts from their credibility. Thus, regardless of the number of dimensions used in a model, the applicability of the results will be open to a certain amount of justified criticism. In order to overcome this dilemma, it seems reasonable to make a working assumption that one-dimensional models can presently be used as a testing or proving ground for new ideas and concepts. If such ideas show promise they will naturally (and eventually) evolve into meaningful multi-dimensional models. Another useful working assumption is that a good one-dimensional model will capture some of the main general features of the constituents of interest. In the case of stratospheric aerosol this assumption is at least plausible, because it is known from direct measurement that the aerosol profile has the same general characteristic over the entire globe. Obviously, in comparing one-dimensional model predictions with actual field data, agreement between the general character of the profiles is more important than an exact absolute quantitative agreement in a limited region. This view will be adopted here in comparing model predictions with typical field measurements.

4. Basic model equation

The basic equation describing the time rate of change of the differential size distribution $n(r,z)$ at altitude z is

$$\frac{\partial n}{\partial t} = -\frac{\partial F}{\partial z} - \frac{\partial}{\partial r}\{Gn\} + C,$$

where r is the radius, F the particle flux (due to sedimentation and eddy diffusion), G the growth rate, C the coagulation term and n the particle number concentration per unit radius interval.

5. The simplified approach

Under certain simplifications the above equation can be solved by elementary analytical methods. This is done by using the equilibrium condition ($\partial n/\partial t=0$), taking the eddy diffusion coefficient D to be constant (5000 $cm^2\ s^{-1}$), letting $C=0$, using a Dirac δ function with respect to altitude for G, approximating the gravitational settling speed by a function that is proportional to particle radius and inversely proportional to air density, and requiring an isothermal stratosphere. Although some of these simplifications may seem quite crude, it will later be seen, after comparison with solutions containing more elaborate detail, that this simple approach captures the essential characteristics of the model both qualitatively and quantitatively. The boundary conditions are specified by a source of single-size "seed particles," corresponding to condensation nuclei (CN), at the lower boundary (tropopause) which diffuse up to the δ function growth layer, a sink for these particles. For simplicity the specific gravity of the seed particles is taken to be the same as that of the particles after growth. Larger particles are formed at the growth layer and distribute themselves under the influence of eddy diffusion, sedimentation and a sink at the tropopause. A schematic diagram summarizing these processes can be found in the Appendix.

In this paper it is assumed that every molecule of H_2SO_4 that strikes an aerosol particle will stick and immediately two water vapor molecules are taken on by the particle. This will result in about a 75% solution of H_2SO_4 for the stratospheric aerosol, which is in agreement with measurement (Rosen, 1971). Growth due to collisions between H_2SO_4 molecules has been shown to be negligible in the stratosphere (Hamill *et al.*, 1977a) and has been neglected.

With the above simplifications the solution is mathematically very similar to that obtained by simply requiring a δ function source at the desired aerosol maximum, but with one important addition: it provides the mechanism for generating a size distribution for the stratospheric aerosol from the single-size seed particles diffusing up from the tropopause.

The absolute concentration at the aerosol maximum is in part determined by the total number of H_2SO_4 molecules in the δ function growth layer and the concentration of seed particles at the tropopause. In this model the flux of seed particles into the stratosphere is balanced by the flux of all the larger particles out of the stratosphere. More detail concerning the derivation of the analytical form of the solution is presented in the Appendix.

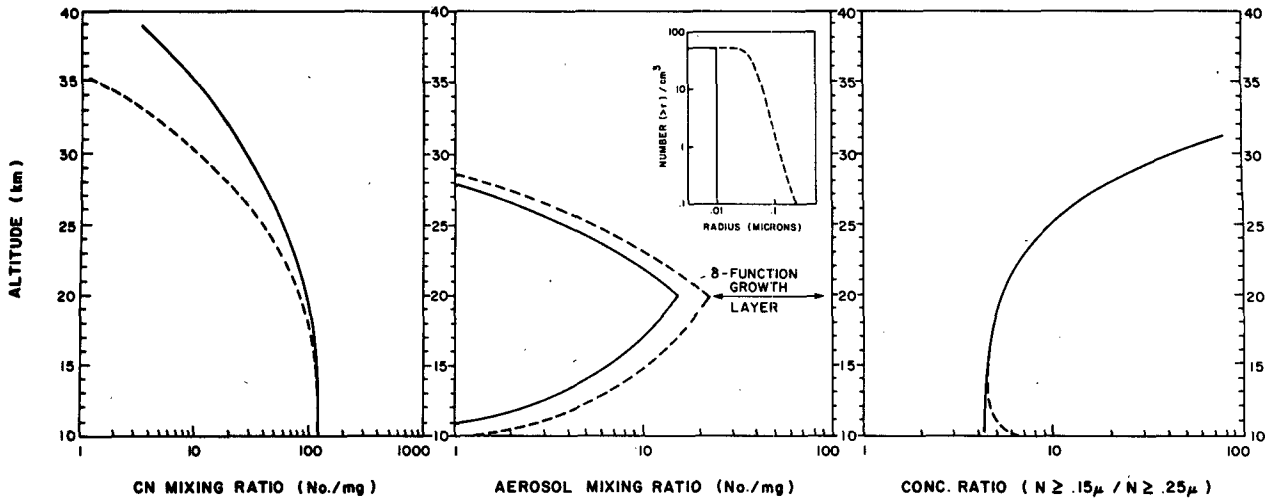

FIG. 1. A comparison of profiles using two types of size distributions. The insert shows the actual cumulative size distributions employed. The CN profile refers to particles >0.02 μm diameter and the aerosol profile refers to particles >0.3 μm diameter. The ratio profile refers to the ratio of the concentration of particles with diameters >0.3 μm to the concentration of particles with diameters >0.5 μm.

One minor and correctable shortcoming of this solution is the requirement of only one size particle at the tropopause when in reality an entire size distribution should exist. This fault can be alleviated by summing the solutions generated by a series of values for the seed particle sizes that approximate the desired size distribution at the tropopause. The result of such an approach will here be referred to as the quasi-analytic solution and can be made as accurate as desired by increasing the number of points needed to approximate the size distribution at the tropopause.

6. Results

A comparison of solutions using one size for the seed particles at the tropopause and using a complete size distribution at the tropopause is shown in Fig. 1. The insert shows the actual two integral size distributions used. Appropriate parameters have been chosen (see Appendix) to reproduce the general character of the observed aerosol profiles (as shown in Fig. 5). The particles referred to as CN are actually the total number of particles present above a diameter of 0.02 μm and the profiles that refer to aerosol are the total number greater than 0.3 μm diameter or those generally associated with the stratospheric sulfate layer. The remaining profile is the ratio of the concentration of particles with diameters >0.3 μm to the concentration of particles with diameters >0.5 μm. As can be seen, there is barely a significant difference between the two examples, and most of this can be attributed to the difference in average size of the two classes of seed particles. In the CN profile the particles associated with the smooth size distribution cannot diffuse to as high an altitude due to their larger average size and the consequently greater influence of sedimentation. In the aerosol profile, particles associated with the smooth size distribution are more numerous because they start out at a larger average size; that is, for a given amount of growth more of them will reach 0.3 μm diameter than the other class of particles. It can be concluded that the results are generally not very dependent on the exact seed particle size distribution used at the tropopause (lower boundary), and that using a size distribution rather than a single size is, in most cases, not worth the considerable extra effort and complexity.

The complete size distribution at the aerosol maximum generated by using single-size 0.02 μm diameter seed particles at the tropopause (solid line model from Fig. 1) is shown in Fig. 2. For comparison, results of experimental measurements are also shown in the figure. These experimental data have been critically discussed in some detail by Harris and Rosen (1976). It is apparent that the agreement between the simple model and the data is surprisingly good and would tend to support the credibility of the modeling approach. In addition, the mass flux of sulfur needed to sustain the H_2SO_4 vapor layer as derived from the model is about 3.3×10^4 tons year^{-1} and is reasonably consistent with other estimates (4–17×10^4 tons year^{-1}) as discussed by Crutzen (1976). A sulfur flux of 5×10^4 tons year^{-1} was used in Crutzen's CSO model of stratospheric aerosols.

7. Perturbations

Assuming that this simple approach to a solution captures the essence of the real stratospheric aerosol, it is of some interest to examine the consequence of several types of perturbations. Fig. 3 shows the result of a perturbation caused by a meteoritic dust source

and a space shuttle Al_2O_3 particle source. Only the CN profile is shown because the change in the $d \geqslant 0.3$ μm aerosol and ratio profiles was found to be only of the order of 10%. These perturbations were calculated under the conditions of a constant number of H_2SO_4 molecules in the growth layer. It would also be of interest to perform the calculations with a constant source strength of the H_2SO_4 vapor in the growth layer. Under this latter condition the resulting perturbations, due to growth, would not be as large, because the number of H_2SO_4 molecules in the growth layer would not be as great. Thus, the results shown in Fig. 3 were calculated under assumptions which would produce the largest effect.

Mathematically the meteoritic dust source is treated similarly to the tropospheric source of seed particles. The upper boundary is taken high enough so that only sedimentation and diffusion are important processes. The meteoritic particle concentration at the upper boundary is chosen to be consistent with a conservative meteoritic flux estimate (in this case, 10^4 tons year^{-1} with an average radius of 0.04 μm and an average density of 2 g cm^{-3}). Thus, the growth layer acts as a sink for both the tropospheric seed particles and the meteoritic dust particles, and the net flux from both of these sources is the relevant quantity to be used in the balance equations discussed in the Appendix.

TABLE 1. Reference code for Fig. 2.

Code	Reference	Type of measurement/Comments
B	Bigg (1975)	Impactor
BFT	Brownlee *et al.* (1976)	Impactor for large particles
F	Friend (1966)	Impactor
FL	Ferry and Lem (1974)	Impactor
I	Ivlev (1976)	Impactor/Data from August 1975
JCM	Junge *et al.* (1961)	Impactor/Data from 26 August 1958 at 18.4 km
M	Mossop (1964)	Impactor/Data from 7 April 1964
MD	Miranda and Dulchinos (1975)	Photoelectric particle counter
MDM	Miranda *et al.* (1973)	Photoelectric particle counter
W	Wyoming results (Pinnick *et al.*, 1976)	Photoelectric particle counter

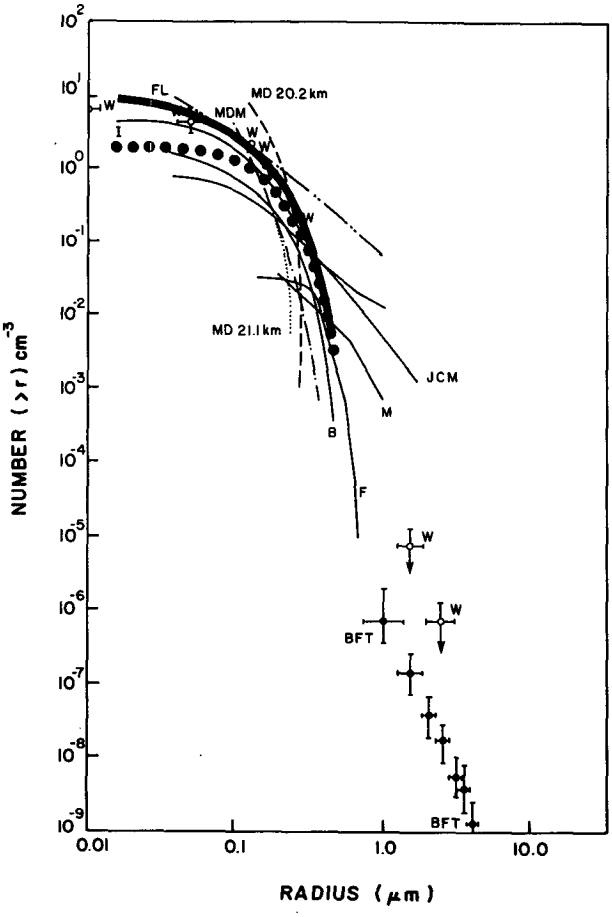

FIG. 2. A comparison of the size distributions at the aerosol maximum derived from the single-size seed particle solution shown in Fig. 2 (thick solid line) and the computer solution shown in Fig. 5 (heavy dotted line) with the actual measurements. The difference in the two calculated curves at small radii is due to the different choice of seed particle concentrations (see Figs. 2 and 5 for numerical values) assumed at the tropopause. See Table 1 for key to references.

The space shuttle perturbation is dealt with as a superposition of solutions. Each individual solution is that for a point source at the desired altitude increment. Thus, the net profile is a sum of a large number of exact solutions. The absolute values of the injection rates are the same as those used by Hofmann *et al.* (1975).

For reference, the expected CN profile for the space shuttle only (no growth layer, tropospheric source or meteoritic source) is also shown in Fig. 3. It is interesting to note that the space-shuttle-perturbed profile is practically the sum of the profile obtained for the space shuttle alone and the natural CN profile predicted by the model. This result, along with the previously mentioned fact that the $d \geqslant 0.3$ μm aerosol profile is practically unchanged under the illustrated perturbations, suggests that the equilibrium distribution of the space shuttle particulate effluents is practically independent of the aerosol growth model used here. Thus, it would appear that simple calculations neglecting the growth mechanics of the natural aerosol may be reasonably valid.

It should also be noted that the basic parameters used in the model to generate Fig. 3 are not exactly the same as those used to generate Fig. 1. The principal difference is a reduction in the concentration of particles at the tropopause by a factor of 0.4 in the latter

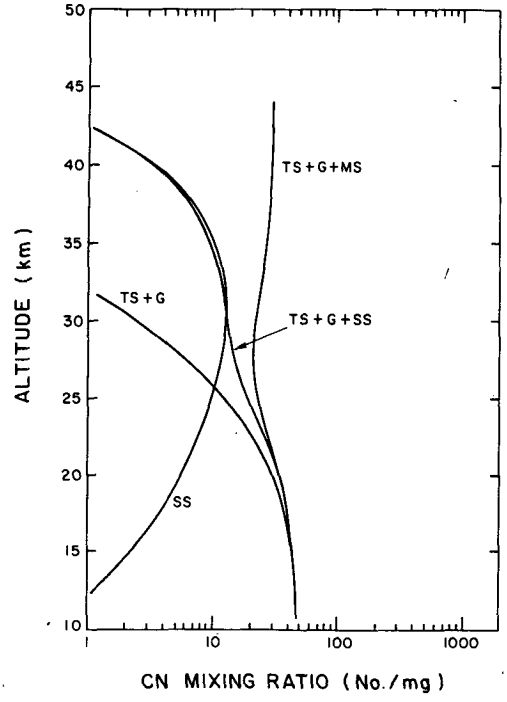

FIG. 3. The CN profile from the quasi-analytic solution using various assumptions: TS, tropospheric source (of seed particles); G, δ function growth layer at 20 km present; MS, meteoritic source (of seed particles); SS, space shuttle source (of seed particles).

figure. This change tends to bring the absolute aerosol concentration at the maximum into better agreement with the measurements (shown in Fig. 5), but the general character of the profiles remains unchanged.

The result of this simple perturbation study indicates that the space shuttle could change the high-altitude CN concentration by a large factor if meteoritic sources are not important. However, even a small meteoritic source could change this conclusion. In addition, higher values of the coefficient of eddy diffusivity than those used here (5000 $cm^2\ s^{-1}$) would also reduce the effect of the space shuttle at high altitude. At present, good experimental measurements are needed above 25 km to determine typical CN profiles, and until these measurements are made it will not be possible to determine the extent to which the space shuttle activities will increase the CN concentration in the upper stratosphere. According to the prediction of this simple model the effect of the space shuttle on the $d \gtrsim 0.3$ μm aerosol profile appears to be quite negligible.

8. Computer solution approach

Of all the approximations used to obtain a quasi-analytic solution, only two stand out as possibly being quite unrealistic: the assumption of a δ function growth layer and the neglect of coagulation. From the work of Hofmann *et al.* (1976), it is clear that a fairly narrow source region would be required to successfully explain the surprisingly narrow stratospheric aerosol layer. The use of a δ function to describe the source region is therefore not entirely unrealistic in terms of the results it produces. Thus the principal shortcoming of the quasi-analytic solution is the complete neglect of coagulation. In what follows, a more general and more complete solution to the basic equation will be obtained by employing computer methods.

A finite-difference method has been used to obtain a computer solution to the basic equation. The altitude grid size for most cases was taken to be 1 km and the radius grid points differed by factors of $2^{\frac{1}{4}}$. The method of solution involved letting the initial conditions evolve in time as dictated by the computer algorithm of the differential equation until a steady-state solution was reached. In most cases, time intervals of 1 day were used. Errors that develop in this method due to finite grid size were investigated by decreasing the grid sizes and comparing the resulting successive steady state solutions.

The eddy diffusion profile used is shown in Fig. 4. The upper portion is similar to that suggested by Hunten (Johnston *et al.*, 1976), but the lower portion has been modified to that of Chang (Johnston *et al.*, 1976), which we feel is more consistent with a mid-latitude stratosphere and a tropopause at 10 km. We have observed that the resulting particle profiles are not very sensitive to the exact nature of the eddy diffusion profile, and the use of a constant value of 5000 $cm^2\ s^{-1}$ would not change the character of the solutions which will be illustrated.

The gravitational settling velocities have been

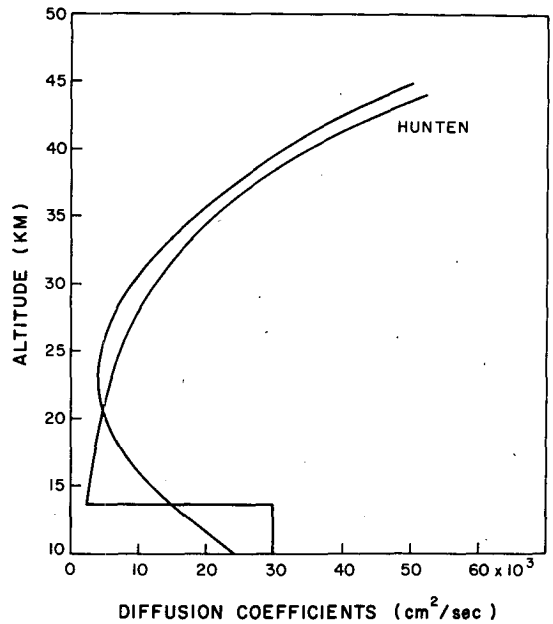

FIG. 4. The eddy diffusivity profile used in the computer solution compared with a profile suggested by Hunten.

adopted from Kasten (1968) by fitting his tabulated data to a curve that is proportional to the particle radius and specific gravity and inversely proportional to the ambient air density. An effective particle specific gravity of 1.65 has been used which is consistent with a composition of about 75% H_2SO_4 and 25% H_2O. The vapor pressure profile of H_2SO_4 was taken to be a Gaussian function centered at 20 km with a width at half maximum of 2.4 km. A peak H_2SO_4 partial pressure of 2.75×10^{-11} torr was chosen, a value in considerable excess of 100% saturation which the work of Hamill *et al.* (1977b) indicates is about 5×10^{-14} torr.

The cumulative size distribution at the tropopause was taken to be inversely proportional to the 3.5 power of the radius and specified by an absolute value that was consistent with field measurements of the aerosol. A smooth lower cutoff in the size distribution near 0.05 μm radius was found necessary in order to obtain agreement in the absolute values of the calculated and measured aerosol profiles. As will be seen, this cutoff leads to using smaller values of the CN concentration at the tropopause than are actually observed. The significance of this problem will be discussed in a later section.

The upper boundary was high enough that the flux of particles across it could be taken as zero. (If a meteoritic source was included, then the flux was derived from the corresponding concentration at the level of the upper boundary.)

Due to limited computer facilities an approximation to the treatment of coagulation was developed. A constant value of the coagulation coefficient K was used (36×10^{-10} cm^3 s^{-1}) that was obtained by averaging over typical size distributions encountered. It should be pointed out that an inconsistency of a factor of 2 in the definition of K has developed in the literature. We use the notation and formulation of Walter (1973). The values reported by Fuchs (1964), for instance, are about a factor of 2 lower, which can be attributed to a slightly different definition of K.

An expression for the time rate of change of the size distribution due to coagulation (which is required in the basic model equation) has been given by Walter (1973). It was necessary to develop an approximation to this expression consistent with the radius grid size. Since the details of the required approximation are cumbersome and tedious, they will not be presented here. A complete documentation of every detail of the model would require an unreasonably large addition to this paper and would undoubtedly be of little interest to the reader. In its place, the overall model consistency checks will be discussed.

9. Solution checks

Since there are many opportunities for serious hidden errors to develop in the computer solution (e.g., those due to approximations, cumulative errors, inappropriate behavior of solution due to the numerical technique and programming mistakes), it is essential to have some independent means of verifying the overall results. Conservation of mass was checked by comparing the mass loss of H_2SO_4 vapor to the net loss of the particles diffusing out of the stratosphere. (It is necessary to make a correction to the latter mass flux due to its partial water content, as previously described.) This test is very sensitive to the accuracy of the treatment of coagulation. In the limit of no coagulation the solution can be checked for particle conservation because the flux of seed particles into the stratosphere must be balanced by the flux of larger particles out of the stratosphere when equilibrium is established. Also in this limit, it is possible to judiciously choose the parameters so that a direct comparison between the computer solution and the accurate quasi-analytic solution can be made. In this case, however, good altitude resolution is needed to reasonably approximate the δ function H_2SO_4 vapor profile with a narrow Gaussian curve required by the computer method. Results of these tests indicate that the overall accuracy of our computer solution is $\sim$10%.

10. Results

Typical computer-generated profiles are shown in Fig. 5, along with the range of actual measured values for a tropopause near 10 km. The experimental data were taken from Hofmann *et al.* (1975), Rosen *et al.* (1975), Pinnick *et al.* (1976) and Rosen and Hofmann (1977).

The size distribution at the aerosol maximum associated with the profiles shown in Fig. 5 is presented in Fig. 2 and is very similar to that of the quasi-analytic solution. The mass flux of sulfur required to sustain the H_2SO_4 vapor layer, used in calculating the profiles of Fig. 5, is about 8×10^4 tons year^{-1} and is in reasonable agreement with that obtained from the simple model illustrated in Fig. 1.

Even though the computer model contains the effects of coagulation and the quasi-analytic model does not, the two are in reasonable agreement. This result can be attributed to the low particle concentration used at the tropopause, which subsequently yields only slight coagulation effects. The influence of coagulation can be most easily detected in the CN profile near the tropopause. The shape of the CN profile in which coagulation does not play a role is shown in Fig. 1. It will be noted that the computer-generated CN profile in Fig. 5 is very similar to this, indicating that, for the choice of parameters, the computer calculation is not very sensitive to coagulation. Fig. 6 shows a case where coagulation does play a role in the CN profile. The general shape of this so-called coagulation profile was analytically described

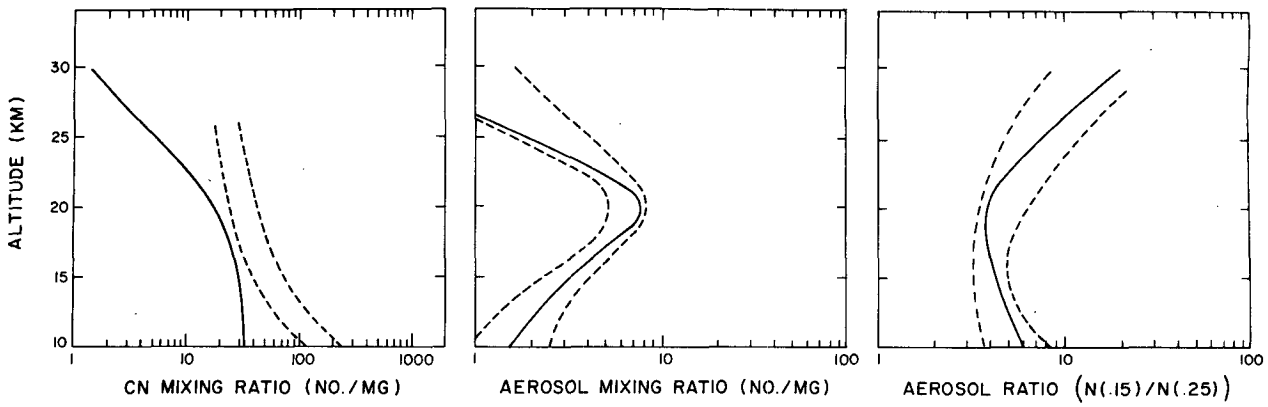

FIG. 5. A typical computer solution (solid lines) compared to the range of actual measurements (dashed lines). The CN profile refers to particles >0.2 μm diameter. The aerosol ratio refers to the ratio of the concentration of particles with diameters >0.3 μm to the concentration of particles with diameters >0.5 μm.

(in an approximate sense) by Junge *et al.* (1961). The latter investigators used a coagulation profile to explain their observed CN concentrations above the tropopause.

The computer model along with the optimum parameters was used to examine perturbations of the stratospheric aerosol. However, the results were similar to the prediction of the quasi-analytic solution shown in Fig. 3, and for that reason will not be discussed further.

11. Comparison with other models

The computer model presented here has been compared to the one described by Turco *et al.* (1976), in which an effort was made to include the chemistry of the H_2SO_4 vapor formation as well as the effect of particle evaporation. This latter effort will be referred to as the Ames model. The results of the comparison are shown in Fig. 6. The parameters used were not necessarily those that optimally describe the observed stratospheric aerosol, and it is clear from the measurements shown in Fig. 5 that they do not. In order to make this comparison it was necessary to use the H_2SO_4 vapor profile predicted by the Ames model in the present (Wyoming) model.

The only significant discrepancy between the two models occurs at higher altitudes, and this difference can in part be attributed to the evaporation effects contained in the Ames model. At high-altitude evaporation would tend to make the average particle size decrease; consequently, gravitational settling will have a smaller effect and the particles will be able to diffuse to a higher altitude. This explains the higher values of CN in the Ames model at high altitude. On the other hand, evaporation would act as a sink for large

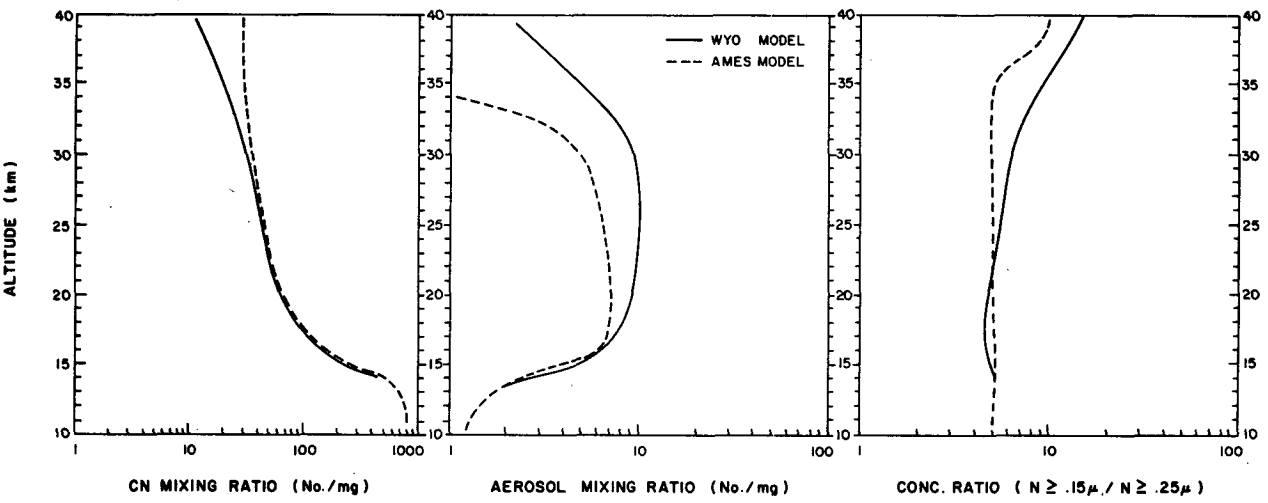

FIG. 6. A comparison of the Ames model (dashed lines) and the Wyoming model (solid lines) using similar input parameters. See text for explanation of differences. The CN profile refers to particles >0.02 μm diameter and the aerosol profile refers to particles >0.3 μm diameter. The ratio profile refers to the ratio of the concentration of particles with diameters >0.3 μm to the concentration of particles with diameters >0.5 μm.

particles, which explains the lower values of aerosol concentration in the Ames model at high altitude.

It should be pointed out that a rigorous comparison between the two models is not strictly possible because they do not require identical input parameters. Some judgment is required as to what exactly would constitute a mutually consistent set of parameters. Thus, the results shown in Fig. 6 are not entirely objective. Within the range of possible mutually consistent parameters, however, it is our experience that the two models produce profiles that are very similar in character.

12. Conclusion

Although many aspects of the stratospheric aerosol are consistent with this model, the predicted CN profile is not in good agreement with observation. It is possible to change the input parameters in such a way as to bring the CN profile into better agreement with measurements (see, e.g., Fig. 6), but this would result in other unacceptable profiles (i.e., the $d \geqslant 0.3\ \mu$m aerosol profile). We did not find a reasonable set of parameters that would bring the model into essentially complete agreement with the measurements.

The disagreement between the predicted and measured CN profiles at high altitude (Figs. 1 and 5) is probably not serious. This discrepancy could be corrected by using larger values of eddy diffusion at high altitude (as was done in Fig. 6) or by adding a meteoritic source, the effects of which are illustrated in Fig. 3. The problem with the CN profile near the tropopause seems to be more basic: the model simply does not require as many CN as are measured. One possible explanation could be that only a certain relatively small class of seed particles have the ability to act as effective nucleation sites and experience growth. In this case the CN profile would be determined mainly by the non-nucleating seed particles and would look very similar to the measured curves shown in Fig. 5. Another reason could be that the natural variability of atmospheric parameters and processes are so great that a one-dimensional model is simply not applicable. If this were the case, however, it is hard to understand why the model does so well in describing certain aspects of the aerosol. Furthermore, the consistency and regularities of the measurements themselves suggest that a simple modeling effort might well be successful. Finally, it could be suggested that the above shortcomings arise from applying the wrong physical model, and the partial success obtained was fortuitous. In conclusion we feel that the model presented in this paper is capable of describing many of the features of the stratospheric aerosol but certain basic shortcomings indicate that the model is not entirely correct or complete.

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APPENDIX

Analytic Solution

The method of obtaining a solution to the simplified sedimentation-diffusion-growth equation as discussed in the main text, can be easily understood by referring to Fig. A1. The δ function growth layer acts as a sink for the seed particles that originate at the tropopause. The flux of these particles to the growth layer which acts as a sink for them can be calculated from the well-known sedimentation-diffusion equation (see, e.g., Junge *et al.*, 1961). This equation states that for a given particle radius r and altitude z the net differential particle flux $\phi(z,r)$ is equal to the sum

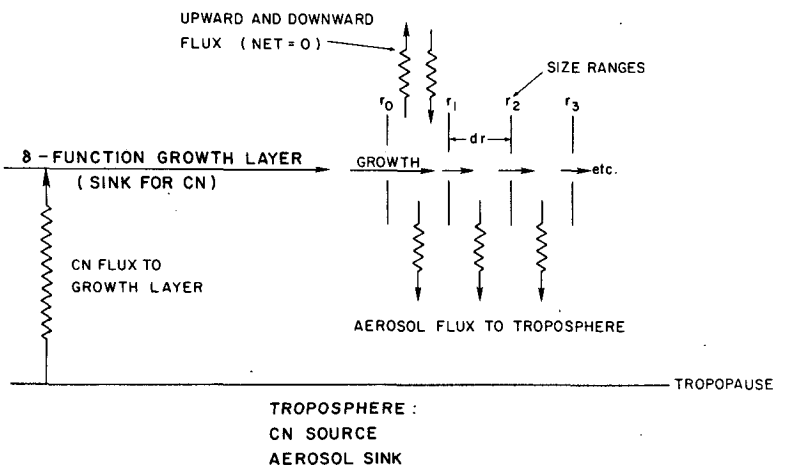

FIG. A1. A schematic diagram of the stratospheric aerosol model.

of the sedimentation and diffusive flux. That is

$$\phi(z,r) = -\rho(z)v(z,r)\gamma(z,r) - \rho(z)D(z)\partial\gamma(z,r)/\partial z$$

and

$$\frac{\partial\gamma}{\partial t} = -\frac{\partial\phi}{\partial z} \text{ at equilibrium,}$$

where z is the altitude (positive direction upward), $\rho(z)$ air density, $v(z,r)$ fall speed, $\gamma(z,r)$ the differential mixing ratio (number per unit air mass and radius interval) and $D(z)$ the eddy diffusivity. Under certain simplifications stated below, the net equilibrium flux of seed particles to the growth layer is found to be

$$\Phi_{\text{in}} = \rho(z_0)ur_s\Gamma_s \exp(-r_s/g_m)[1-\exp(-r_s/g_m)]^{-1}.$$

Here

- $\rho(z_0)$ density of air at the tropopause
- u constant in the sedimentation equation $v_s = ur\exp(z/H)$
- Γ_s mixing ratio of seed particles at the tropopause (number per unit air mass)
- $1/g_m$ $[\exp(z_m/H)-1]uH/D$
- H scale height of atmosphere
- D eddy diffusivity, now a constant
- z_m height of δ function growth layer above tropopause
- r_s radius of seed particles.

Consider the particles in the growth layer itself. In each size interval there will be a gain in concentration due to smaller particles growing larger, a loss due to particles growing out of the size range, and a loss due to sedimentation-diffusion to the tropopause. The growth can be treated as a "flux" of particles through the size distribution given by Gn, where G is the growth rate [in radius units (μm) per time] and n the differential number concentration (cm^{-3} μm^{-1}). The "divergence" of this flux must, therefore, be the loss by sedimentation-diffusion to the troposphere; that is, this divergence is the source of aerosol (at radius r) and the tropopause is the sink. Using the well-known sedimentation-diffusion equation previously mentioned, the differential flux of particles to the tropopause is found to be

$$\phi_{\text{out}} = \gamma(z_m,r)\rho_0 ur[1-\exp(-r/g_m)]^{-1},$$

where $\gamma(z_m,r)$ is the equilibrium differential mixing ratio in the growth layer. On the other hand, the loss term from the divergence of the flux of particles through the size distribution is $-\partial(Gn)/\partial r$. Under present circumstances, G is due essentially to the thermal flux of H_2SO_4 molecules onto the particles. This is given by $G = \frac{1}{4}Nv_tV_e$, where N is the concentration of H_2SO_4 molecules, v_t the effective velocity of H_2SO_4 molecules, V_e the effective volume of one H_2SO_4 and two H_2O molecules. The units of $\partial(Gn)/\partial r$ are number of particles per unit volume in size interval dr being lost per second. Thus, the net stratospheric loss (due to growth) of a given size particle can be found by integrating over the altitude range of the layer itself. Since the layer (N) here is a δ function the result is

$$\phi_g = -\tfrac{1}{4}\rho(z_m)v_tV_eA\,\partial\gamma(z,r)/\partial r,$$

where A is the total number of H_2SO_4 molecules per square centimeter of the layer.

The total flux of particles out of the stratosphere can be found by integrating the expression for ϕ_{out} over all size ranges; this quantity will be referred to as Φ_{out}.

By equating ϕ_{out} to ϕ_g, a condition required for equilibrium in all size ranges, a simple differential equation is obtained which yields a relative size distribution in the growth layer (i.e., at z_m). By equating Φ_{out} to Φ_{in}, a condition requiring the net flux of particles into the stratosphere to equal the net flux of particles out of the stratosphere, the absolute size distribution is obtained, *viz.*, $\gamma(z_m,r)$. The value of γ at other altitudes is obtained by simply treating the growth layer as a source [with a fixed mixing ratio $\gamma(z_m,r)$], treating the tropopause as a sink, and using the well-known sedimentation-diffusion equation to determine γ at other values of z. The results of this calculation are as follows:

At the aerosol maximum the mixing ratio is given by

$$\gamma(z_m,r) = f\Gamma_s \exp[-ar-br^2-cr^3-\cdots],$$

where

$$a = 4D/(v_tV_eAH)[1-\exp(-z_m/H)]^{-1}$$

$$b = \tfrac{1}{4}a(uH/D)[\exp(z_m/H)-1]$$

$$c^* = 0.4b^2/a$$

$$f = 4r_sb\exp(-4r_sb/a)[1-\exp(-4r_sb/a)]^{-1} \times\exp(1.12^{**}r_sa) \approx a.$$

* Optimum value of c for truncated series.

** Approximate value for range of values of r_s, a and b expected in the stratosphere.

Γ_s is the mixing ratio of seed particles at the tropopause and r_s the radius of seed particles.

Only a few terms are needed in the exponent describing $\gamma(z_m,r)$, and, in fact, including only the first term is generally adequate for the size ranges in which the concentrations are high enough to measure easily. This result shows that a stratospheric aerosol generated by a growth process is probably better described by an exponential size distribution than by a power law size distribution. A log-normal curve will also make an excellent approximation to $\gamma(z_m,r)$ for radii larger than the seed particle size.

A sample calculation using parameters involved in

generating the profiles shown in Fig. 1 is given below.

$v_t = (8kT/\pi m)^{\frac{1}{2}} = 2.15 \times 10^4$ cm s^{-1} ($T = 216$ K and m is the mass of H_2SO_4 molecule)

$V_e = 1.3 \times 10^{-22}$ cm^3 ($\sim 75\%$ $H_2SO_4 + 25\%$ H_2O)

$A = 1.47 \times 10^{11}$ H_2SO_4 molecules cm^{-2} (compare this value to 3×10^{11} derived from the computer model results shown in Fig. 5)

$v_s = 8.9 \times 10^{-3} r \exp(z/H)$ (r in μm)

$H = 6.3$ km $\quad z_m = 10$ km

$D = 5000$ cm^2 s^{-1} $\quad r_s = 0.01$ μm

$\rho(z) = 4.127 \times 10^{-3} \exp(-z/H)$ (z is the distance above tropopause at 10 km)

$\Gamma_s = 1.21 \times 10^5$ gm^{-1} (or 50 cm^{-3}).

With these parameters the following results were obtained:

Function	Value
a	9.71 μm^{-1}
b	10.59 μm^{-2}
c	4.62 μm^{-3}
f	10.63
$N(0.15)$ at z_m	1.32 cm^{-3}
$N(0.15)/N(0.25)$	5.0

The complete profiles are shown in Fig. 1. $N(0.15)$ and $N(0.25)$ refer to the concentration of particles larger than 0.15 μm radius and 0.25 μm radius, respectively. Note also that $f \approx a$, as earlier indicated.

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