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A Stratospheric Aerosol Model
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ABSTRACT

This report 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 approach very few simplifications were made and a computer technique was used to solve the equations. The most complex form of the model contains the effects of sedimentation, diffusion, particle growth and coagulation. Results of the perturbation calculations show that there will probably be an immeasurably small increase in the stratospheric aerosol concentration for particles larger than about 0.15 μm radius. The increase in very small particulates (greater than .01 μm diameter) is potentially large but cannot be adequately evaluated until the true natural background of these small particles is determined.

Stratospheric Aerosol Model with Perturbations Induced by the Space Shuttle Effluents

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Introduction

It is well known that a relative maximum exists on a global scale in the aerosol mixing ratio for particles having a diameter $>0.3\mu m$ approximately 10 km above the tropopause. The particles are composed of H_2SO_4 (for the last decade at least) 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 in principle could approach a quasi steady state distribution. It is the purpose of this paper to present a one dimensional model describing this quasi steady state situation and to investigate perturbations of this state.

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 & Vermeulen, 1963) even a very modest production rate of H_2SO_4 could produce a large super saturation.

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 this present work evaporation of the particles is neglected. This is more or less justifiable because such a process would eventually saturate the entire stratosphere with H_2SO_4 vapor providing there were no sinks for the vapor. Under saturated conditions neither growth nor evaporation would take place. However at high altitude there may be a sink for H_2SO_4 due to photodissociation by ultraviolet light. Under this circumstance the model presented here should not be considered entirely realistic above about 30 to 35 km. On the other hand this is above the main region of interest and should have little affect on the major results presented. The inclusion of evaporation effects would increase to a considerable extent the complexity and uncertainties in the model. The basic reason for this is that the particles cannot evaporate to a smaller size than that of their original core. Since individual particle identities are lost in 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. Our interpretation of a reasonable profile is one which has a relative maximum in supersaturation near the observed aerosol maximum, and a concentration consistant with the over all sulfur budget and transport properties of the atmosphere. 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 the same. Such a comparison has been made and will be discussed in a later section. Again it should be emphasized that in our opinion the present uncertainties involved in the sulfur chemistry make a more elaborate model (one containing sulfur chemistry) less fundamental than the approach presented here.

Philosophy of one Dimensional Models

In general the atmosphere should be treated as a 3 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 3 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 for a testing or a proving ground of new ideas and concepts. If such ideas show promise they will naturally (and eventually) evolve into accurate 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 does in fact have recognizable features on a global scale. 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.

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 is the particle flux (due to sedimentation and eddy diffusion), G is the growth rate, C is the coagulation term and n is the particle number concentration.

The Simplified Approach

Under certain conditions the above equation can be solved by simple 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/sec in most cases), letting C=0, using a δ - function 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 realistic detail, that this simple approach captures the essential characteristics of the model both qualitatively and quantitively. 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 H_2SO_4. 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 Appendix 1.

In this paper it is assumed that every molecule of H_2SO_4 that strikes an aerosol particle will stick and immediately 2 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, 1977) 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 of 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 Appendix I.

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.

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 figure 1. The insert shows the actual two integral size distributions used. Appropriate parameters have been chosen to reproduce the general character of the observed aerosol profiles (as shown in figure 4). The particles referred to as cn are actually the total number of particles present above a diameter of .01 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 diameter greater than 0.3 μm to the concentration of particles greater than 0.5 μ diameter. 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 consequent 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 generally concluded that the results are 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 .01 micron diameter seed particles at the tropopause is shown in figure 2. For comparison, results of experimental measurements are also shown in the figure. This experimental data has 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 is about 3.3×10^4 tons/yr. and is reasonably consistent with other estimates (4 to 17×10^4 tons/yr) as discussed by Crutzen (1976). A sulfur flux of 5×10^4 tons/yr was used in Crutzen's CSO model of stratospheric aerosols.

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. Figure 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 $d \geq 0.3$ 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 figure 3 were calculated under assumptions which would produce the largest effect.

Mathematically the meteoritic dust source is treated similarly as 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 is chosen to be consistent with a conservative meteoritic flux (in this case, 10^4 tons/yr with an average radius of .04 microns and an average density of 2 gm/cm^3). Thus the growth layer acts as a sink for both the tropospheric seed particles as well as meteoritic particles and the net flux from both of these sources is the relevant quantity to be used in the balance equations discussed in Appendix I.

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 figure 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 $\geq$ 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 [illegible] here. Thus it would appear that simple calculations neglecting the g[illegible]anics of the natural aerosol may be reasonably valid.

It should also be noted that the basic parameters used in the model to generate figure 3 are not exactly the same as those used to generate figure 1. The principle difference is a reduction in the concentration of particles at the tropopause by a factor of 0.4 in the latter figure. This change tends to bring the absolute aerosol concentration at the maximum into better agreement with the measurements (shown in figure 4) 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 ($5000\ cm^2/sec$) 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 $d \geq 0.3\ \mu m$ aerosol profile appears to be quite negligible.

Computer Solution Approach

Of all the approximations used to obtain a quasi analytic solution, only two may be questioned as not being realistic: 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 observed stratospheric aerosol layer. The use of a δ-function to describe the source region is therefore not entirely unrealistic. Thus the principle short coming 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 essentially $2^{1/3}$. The method of solution involved starting with initial conditions and letting the differential equation develop in time until a steady state solution was reached. In most cases, time intervals of one 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 figure 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 consistent with a tropopause at 10 km. Recently other modelers have adopted a profile similar to the one used here (Luther 1977). 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/sec would not change the character of the solutions which will be illustrated.

The gravitational settling velocities have been 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. A particle specific gravity has been used that 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 and several kilometers wide. A peak H_2SO_4 concentration of 2.75×10^{-11} mm Hg was chosen, a value in considerable excess of 100% saturation.

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 cut off in the size distribution near .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 cut off 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 so 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/sec) that was obtained by averaging over typical size distributions encountered. It should be pointed out that an inconsistancy of a factor of two 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 two lower which can be attributed to a 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. In its place, a discussion of the overall model consistency checks will be made.

Solution Checks

Since there are many opportunities for serious errors to develop in the computer solution (i.e. those due to approximations, cumulative errors and programming mistakes) it is essential to have some independent means of verifying the over-all results. Conservation of mass was checked by comparing the mass loss of H_2SO_4 vapor to the net mass 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. However, in this case 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 about 10%.

Results

Typical computer generated profiles are shown in figure 4 along with the range of actual measured values for a tropopause near 10 km. The experimental data was 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 figure 5 is presented in figure 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 figure 5, is about 8×10^4 tons per year and is in reasonable agreement with that obtained from the simple model illustrated in figure 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 affects. 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 figure 1. It will be noted that the computer generated cn profile in figure 5 is very similar to this indicating that, for the choice of parameters, the computer calculation is not very sensitive to coagulation. Figure 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 (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 figure 3 and for that reason will not be discussed further.

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 figure 6. The parameters used were not necessarily those that optimally describe the observed stratospheric aerosol. 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 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 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 judgement is required as to what exactly would constitute a mutually consistent set of parameters. Thus the results shown in figure 6 are not entirely objective. However, within the range of possible mutually consistent parameters, it is our experience that the two models produce profiles that are very similar in character.

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 so as to bring the cn profile into better agreement with measurements (see figure 6 for example) but this would result in other unacceptable profiles (i.e. the $d \geq 0.3$ µm 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 (figure 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 figure 6) or adding a meteoritic source, the effects of which are illustrated in figure 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.

In conclusion we feel that the model presented in this paper is capable of describing most of the features of the stratospheric aerosol but certain shortcomings indicate that the model is not entirely correct or complete. If these shortcomings are overlooked and the model is used to predict the change in particulate content of the stratosphere due to the space shuttle Al_2O_3 effluents, then only a small and probably immeasurable change in the concentration of particles larger than 0.3 μm diameter would be expected to occur. Although a potentially significant amount of smaller particles ($d \geq .01$ μm) will be added to the stratosphere above 30 km it is not possible to determine if a measurable increase above natural levels will occur simply beacuse the natural background in this size range and altitude is unknown.

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FIGURE CAPTIONS

Figure 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 larger than 0.01 μm diameter and the aerosol profile refers to particles larger than 0.3 μm diameter. The ratio profile refers to the ratio of the concentration of particles with diameter greater than 0.3 μm to the concentration of particles with diameter greater than 0.5 μm.

Figure 2. A comparison of the size distributions at the aerosol maximum derived from the quasi-analytic solution (thick solid line) and the computer solution (heavy dotted line) with the actual measurements. The difference in the two types of solutions at small radii is due to the different choice of seed particle concentration assumed at the tropopause. See table for key to references.

Figure 3. The cn profile obtained 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) and SS = space shuttle source (of seed particles).

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

Figure 5. A typical computer solution (solid lines) compared to the range of actual measurements (dashed lines). The cn profile refers to particles larger than .01 μm diameter and the aerosol profile refers to particles larger than 0.3 μm diameter. The aerosol ratio refers to the ratio of the concentration of particles with diameter greater than 0.3 μm to the concentration of particles with diameter greater than 0.5 μm.

Figure 6. A comparison of the Ames Model (dashed lines) and the Wyoming Model (solid lines) using similar input parameters. See text for explanations of differences. The cn profile refers to particles larger than .01 μm diameter and the aerosol profile refers to particles larger than 0.3 μm diameter. The ratio profile refers to ratio of the concentration of particles with diameter greater than 0.3 μm to the concentration of particles with diameter greater than 0.5 μm.

TABLE

Figure 2 Reference Code

Code	Reference	Type of Measurement/Comments
B	Bigg (1975)	Impactor
BFT	Brownlee, Ferry, & Tomendl (1976)	Impactor for large particles
F	Friend (1966)	Impactor
FL	Ferry & Lem (1974)	Impactor
I	Ivlev (1976)	Impactor/Data from Aug. 1975
JCM	Junge, Chagnon & Manson (1961)	Impactor/Data from 26 Aug. 1958 @18.4 km
M	Mossop (1964)	Impactor/Data from 7 April 1964
MD	Miranda & Dulchinos (1975)	Photoelectric Particle Counter
MDM	Miranda, Dulchinos & Miranda (1973)	Photoelectric Particle Counter
W	Wyoming Results (Pinnick et al 1976)	Photoelectric Particle Counter

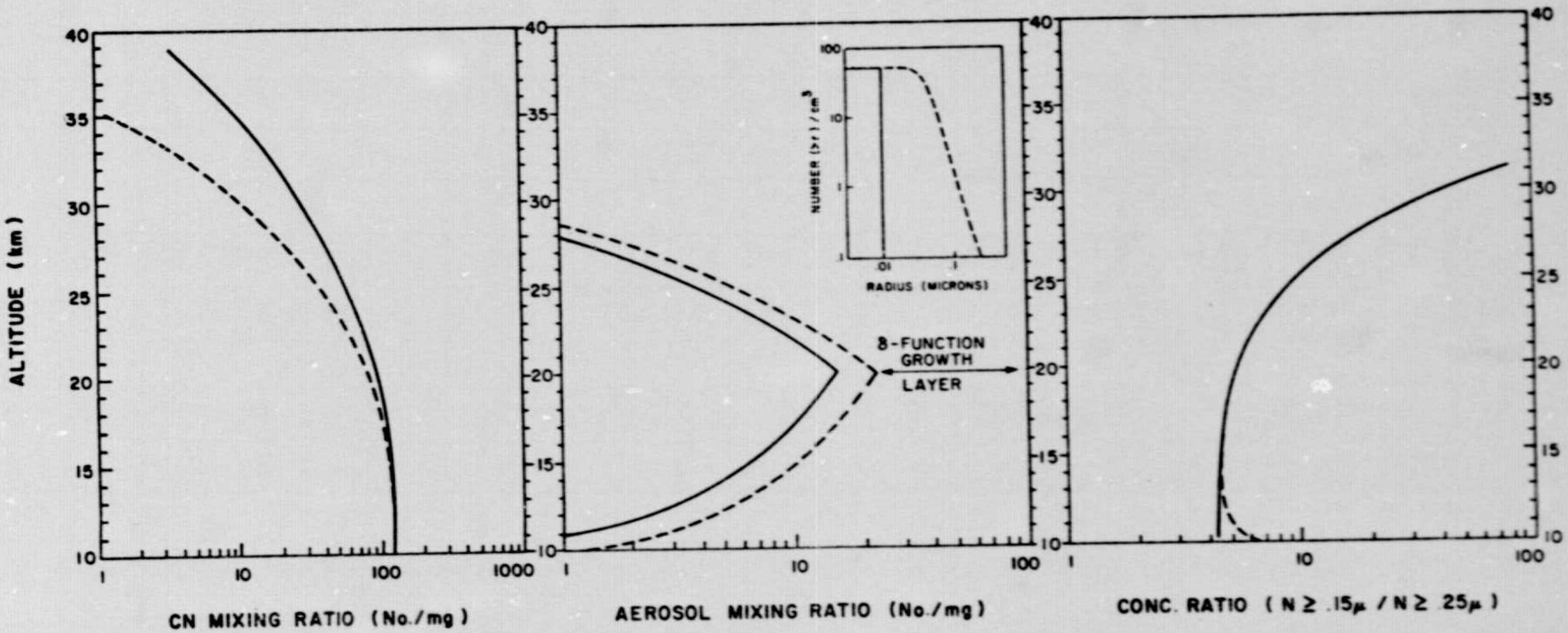
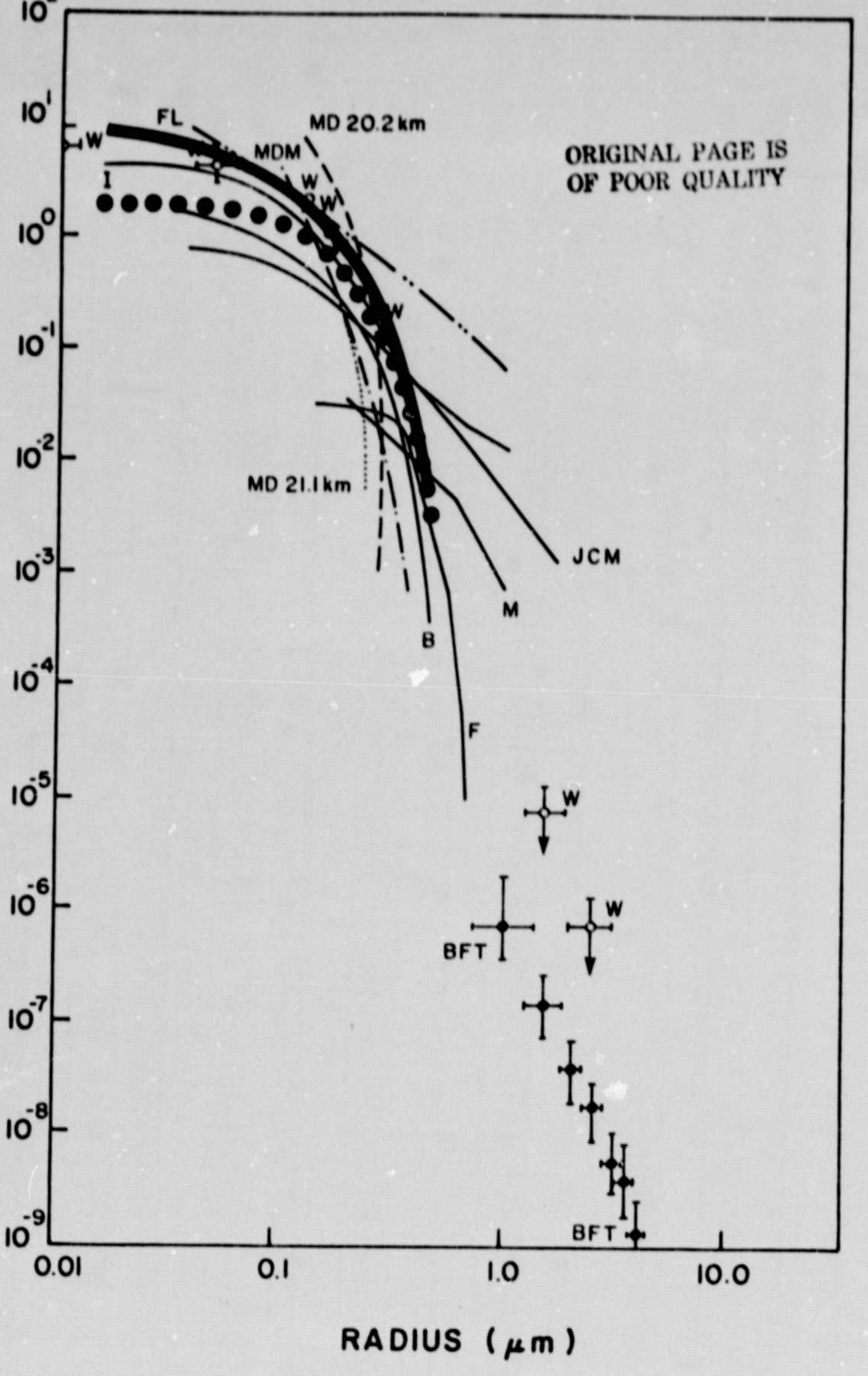

ALTITUDE (km)
40
35
30
25
20
15
10
1
10
100
1000
CN MIXING RATIO (No./mg)
1
10
100
AEROSOL MIXING RATIO (No./mg)
δ-FUNCTION GROWTH LAYER
NUMBER (>r) / cm^3
100
10
1
.1
.01
.1
RADIUS (MICRONS)
1
10
100
CONC. RATIO (N ≥ .15μ / N ≥ .25μ)

Figure 1


10^{2}
10^{1}
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10^{-6}
10^{-7}
10^{-8}
10^{-9}
FL
MD 20.2 km
W
MDM
I
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W
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W
MD 21.1 km
JCM
M
B
F
W
W
BFT
BFT
0.01
0.1
1.0
10.0
RADIUS (μm)

Figure 2

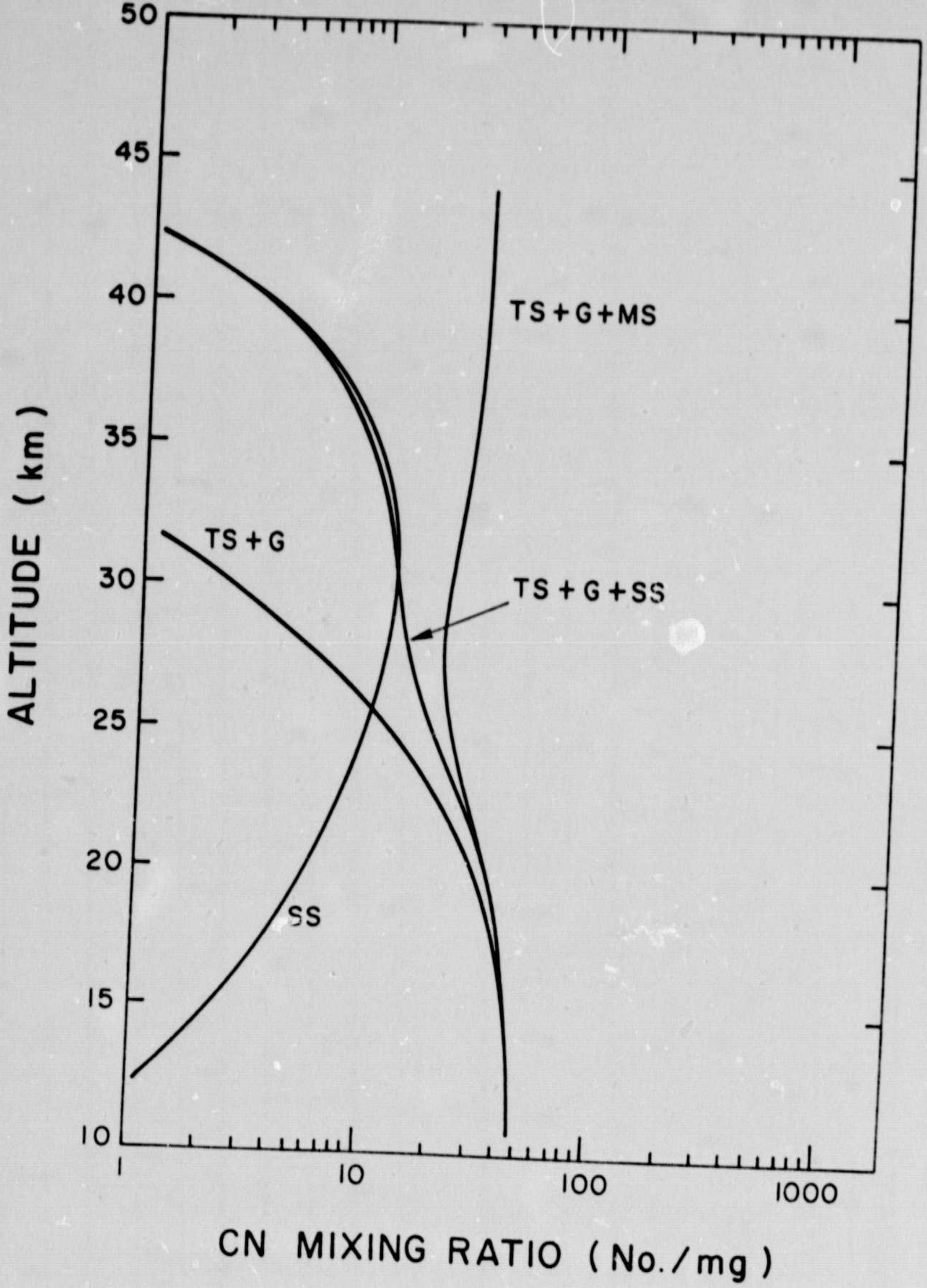

50
45
40
35
30
25
20
15
10
1
10
100
1000
ALTITUDE (km)
CN MIXING RATIO (No./mg)
TS + G + MS
TS + G
TS + G + SS
SS

Figure 3

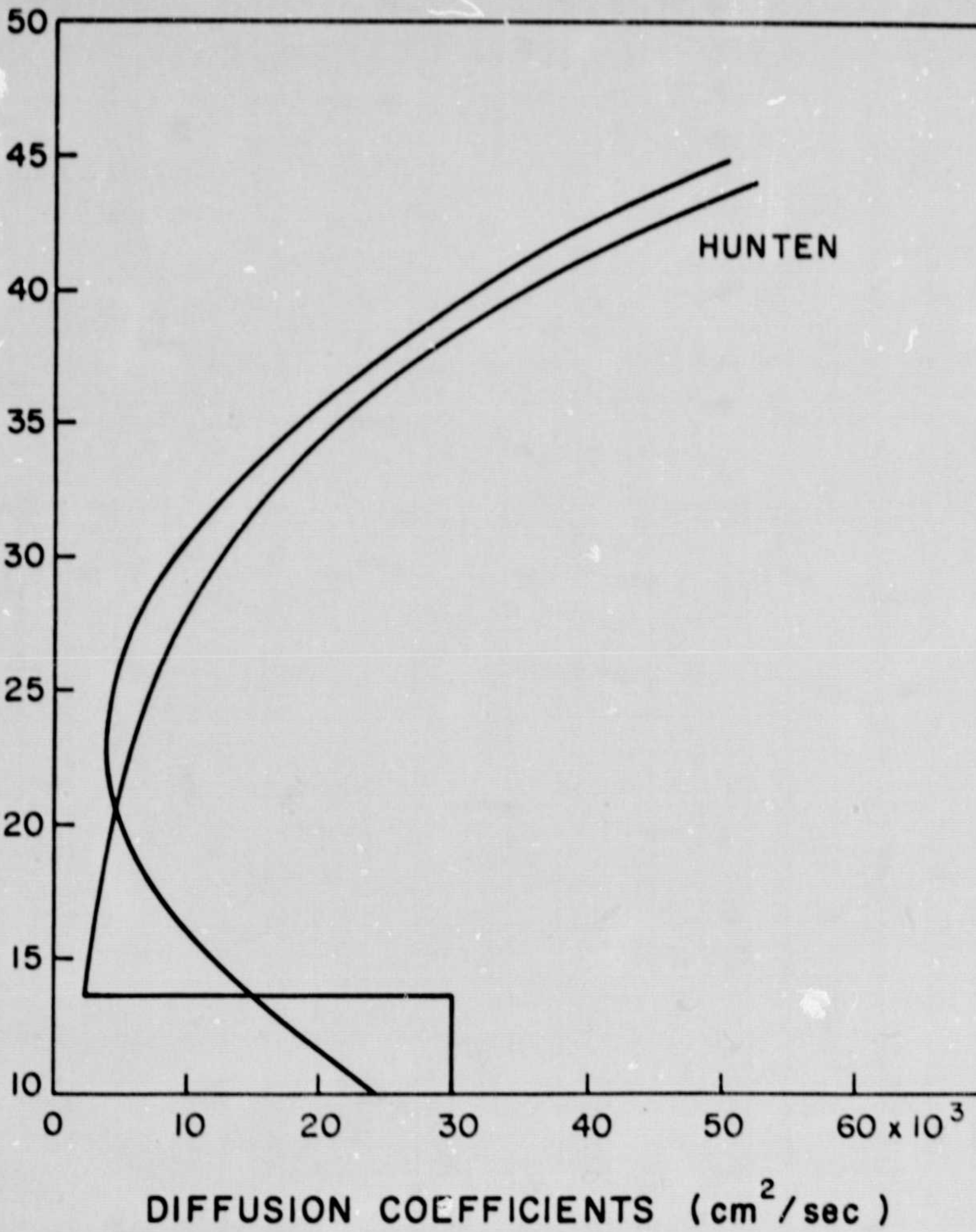

50
45
40
35
30
25
20
15
10
HUNTEN
0
10
20
30
40
50
60 x 10^3
DIFFUSION COEFFICIENTS (cm^2/sec)

Figure 4

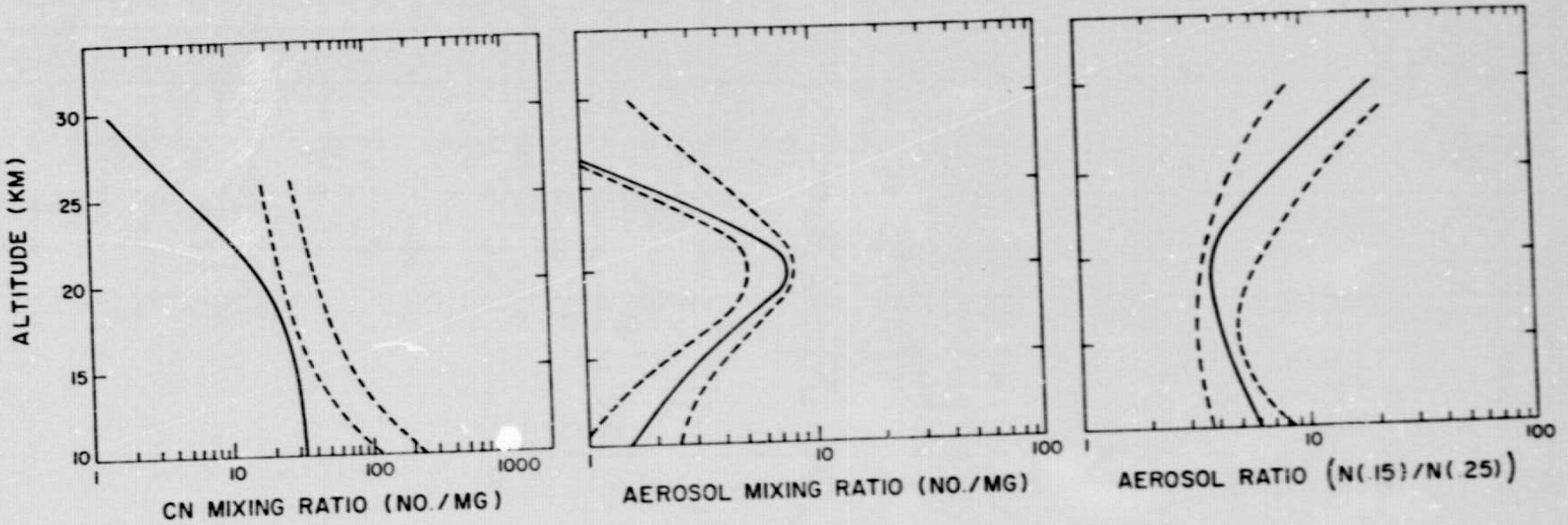

ALTITUDE (KM)
30
25
20
15
10
1
10
100
1000
CN MIXING RATIO (NO./MG)
1
10
100
AEROSOL MIXING RATIO (NO./MG)
1
10
100
AEROSOL RATIO (N(.15)/N(.25))

Figure 5

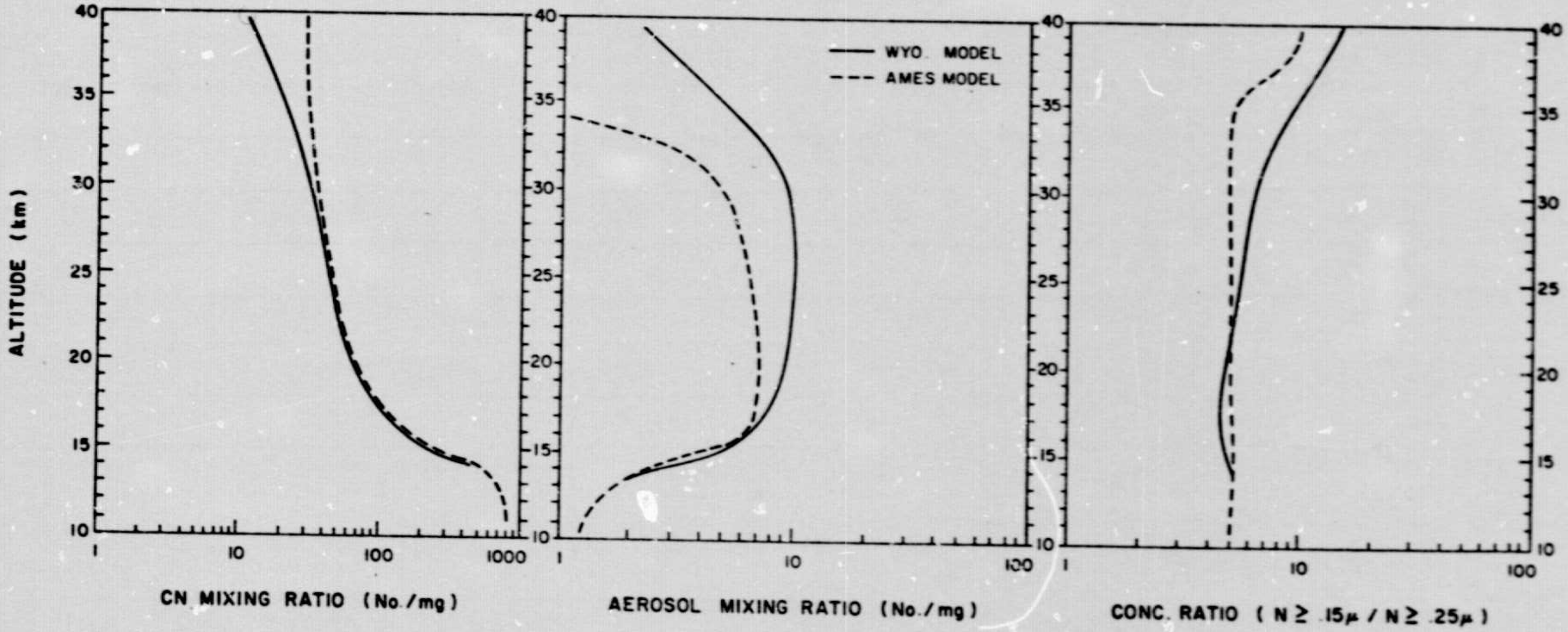

ALTITUDE (km)
40
35
30
25
20
15
10
1
10
100
1000
CN MIXING RATIO (No./mg)
1
10
100
AEROSOL MIXING RATIO (No./mg)
WYO. MODEL
AMES MODEL
1
10
100
CONC. RATIO (N ≥ .15μ / N ≥ .25μ)

Figure 6

Appendix I

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 figure A-1. 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 is therefore easily found to be (see example 1, Appendix II):

$$\Phi_{in} = \rho(z_o)\, ur\, \Gamma_s\, e^{-r_s/g_m}\ (1-e^{-r_s/g_m})^{-1}$$

where

$\rho\ (z_o)$ = density of air at the tropopause.

u = constant in the sedimentation velocity equation: $v = ure^{z/H}$

r = radius of particles

Γ_s = mixing ratio at tropopause of seed particles.

$1/g_m$ = $(e^{z_m} - 1)\ uH/D$

H = scale height of atmosphere.

D = eddy diffusivity (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 and 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/time) and n is the differential number concentration

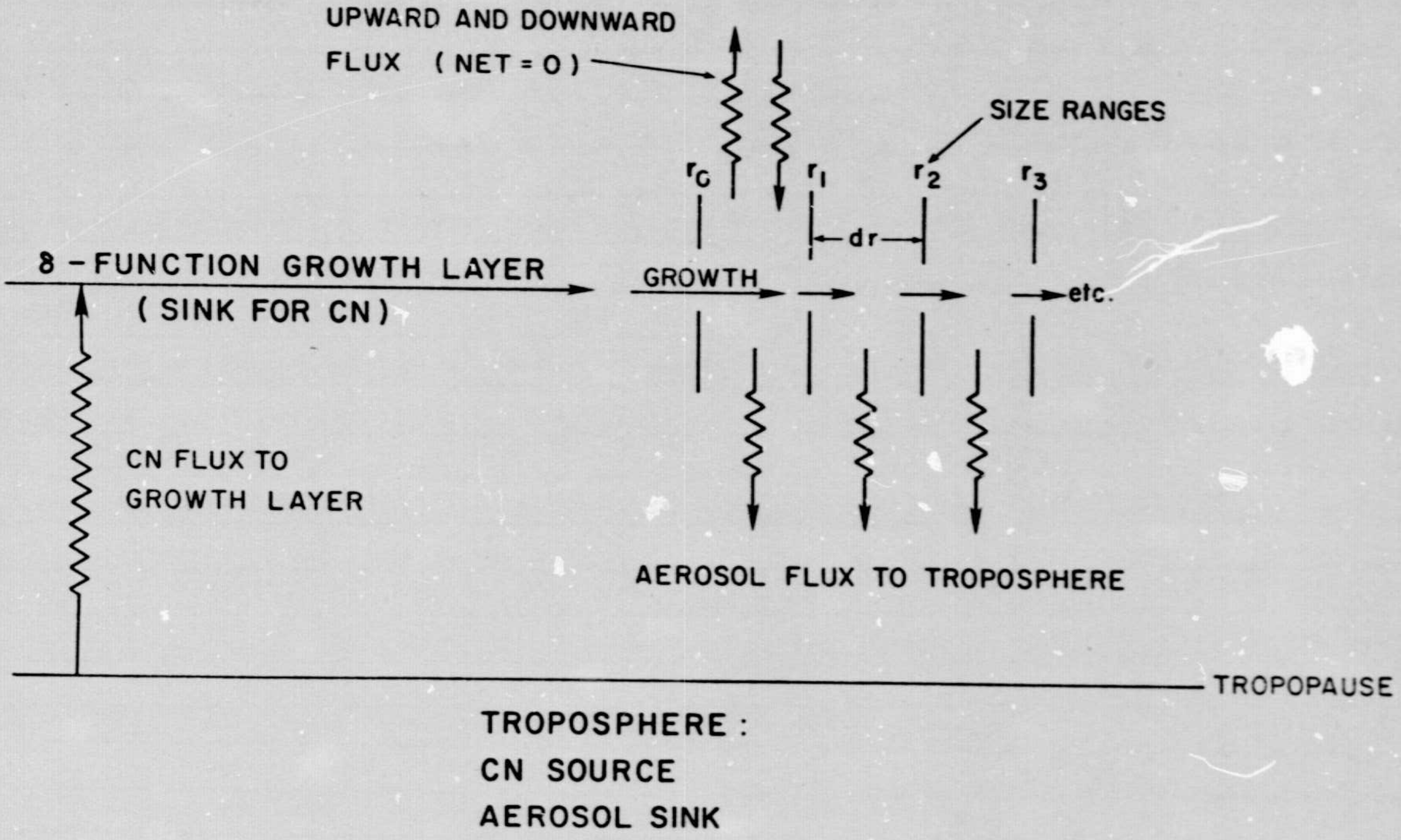


Figure A-1. A schematic diagram of the stratospheric aerosol model.

($\text{cm}^{-3}\ \mu\text{m}^{-1}$). The "divergence" of this "flux" must therefore be the loss by sedimentation and diffusion to the troposphere. From the arguments given in Appendix II (see example 4) this loss term is found to be:

$$d\Phi_{out} = \gamma(z_m, r)\ \rho_o u r\ (1 - e^{-r/g_m})^{-1}$$

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

$$\Phi_g = \frac{\partial}{\partial r}\ (Gn)$$

Under present circumstances G is due essentially to the thermal flux of H_2SO_4 onto the particles. This is given by $G = \frac{1}{4} N\, v_{th}\, V_e$ where N = the concentration of H_2SO_4 molecules, v_{th} = the effective velocity of H_2SO_4 molecules and V_e is the effective volume of 1 H_2SO_4 and 2 H_2O molecules. The units of Φ_g are number of particles per unit volume in size interval dr being lost per second. Thus the losses of a given size particle can be found by integrating over the altitude range of the layer itself. Since the layer here is a δ-function the result is

$$\Phi_g = \frac{1}{4}\rho(z_m)\ v_{th}\ V_e\ A\ \frac{\partial}{\partial r}\ \gamma(z_m, r)$$

where A = total number of H_2SO_4 molecules in the layer.
The total flux of particles out of the stratosphere can be found by integrating the expression for $d\Phi_{out}$ over all size ranges; this quantity will be referred to as Φ_{out}.

By equating Φ_{out} to Φ_g 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} the absolute size distribution is obtained (that is, $\gamma(z_m,r)$). The value of γ at other altitudes is obtained by simply treating the growth layer as a source (with a fixed mixing ratio $\gamma(z_m,r)$) and using sedimentation-diffusion equations to determine the spreading (see Appendix II).

The results of this calculation are as follows:

At the aerosol maximum the mixing ratio is given by

$$\gamma(z_m,r) = f\Gamma_s\, e^{-[ar + br^2 + cr^3 + \ldots\ldots]}$$

where

$$a = 4D/(v_t\, V_e\, AH)\, [1-e^{-z_m/H}]^{-1}$$

$$b = \frac{a}{4}\,\frac{uH}{D}\,[e^{z_m/H} - 1]$$

$$c = \frac{0.4b^2}{a}$$

$$f = 4r_s b\, e^{-4r_s b/a}\,(1-e^{-4r_s b/a})^{-1}\, e^{1.12^{*}\, r_s a}$$

Γ_s = mixing ratio of seed particles at tropopause.

r_s = radius of seed particles.

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

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 easily measure. 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 sample calculation using parameters involved in generating the profiles shown in figure 1 is given below.

$v_{th} = \sqrt{8KT/\pi m} = 2.15 \times 10^4$ cm/sec (T = 216° K and m = mass of H_2SO_4 molecule)

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

$A = 1.47 \times 10^{11}$ H_2SO_4 moluecules/cm^2 (Compare this value to 3×10^{11} used in the Computer Model results in figure 5)

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

$H = 6.3$ km

$D = 5000$ cm^2/sec

$z_m = 10$ km

$r_s = .01$ μm

$\rho(z) = 4.127 \times 10^{-3}\, e^{z/H}$ (z = distance above tropopause)

$\Gamma_s = 1.21 \times 10^5$/gm (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(.15) at z_m	1.32 cm^{-3}
N(.15)/N(.25)	5.0

The complete profiles are shown in figure 1. N(.15) and N(.25) refer to the concentration of particles larger than .15 μm radius and .25 μm radius respectively.

Appendix II

Library of Profiles for Simple Diffusion-Sedimentation Equilibrium.

It is often quite informative to refer to simple one-dimensional eddy diffusion-sedimentation profiles of aerosol in order to gain an approximate understanding of the spatial distribution of this contituent. In addition, complex profiles can often be viewed as a combination of several simple profiles.

Under equilibrium conditions the flux of particles of radii between r and r+dr and at altitude z is given by

$$\Phi(z,r) = -\rho(z)\ v(z,r)\ \gamma(z,r) - \rho(z)\ D(z)\ \frac{\partial\gamma(z,r)}{\partial z}$$

$$\text{and}\quad \frac{\partial\gamma}{\partial t} = -\frac{\partial}{\partial z}\ \Phi = 0$$

where

$\rho(z)$ = air density

$v(z,r)$ = fall velocity

$\gamma(z,r)$ = mixing ratio

$D(z)$ = eddy diffusivity.

These equations can be solved exactly for the following conditions which are fairly realistic.

$\rho(z) = \rho_0\ e^{-z/H}$ (H = atmospheric scale height)

$v(z,r) = ure^{z/H}$ (u = constant)

$D(z)$ = constant

Under these conditions a general solution is:

$$\gamma(z,r) = \gamma_a\ f(z,r) + \gamma_b$$

where γ_a and γ_b are constants determined by the boundary conditions and

$$f(z,r) = \exp[-(e^{z/H}-1)\ Hur/D] \equiv e^{-r/g\ (z)}$$

when the lower boundary is at $z=0$,

and

$$f(z,r) = \exp[-(e^{z/H}-e^{z_m/H})\ Hur/D \equiv e^{-r/g'(z)}$$

when the lower boundary is at $z=z_m > 0$.

By definition

$$1/g(z) \equiv (e^{z/H}-1)\ uH/D$$

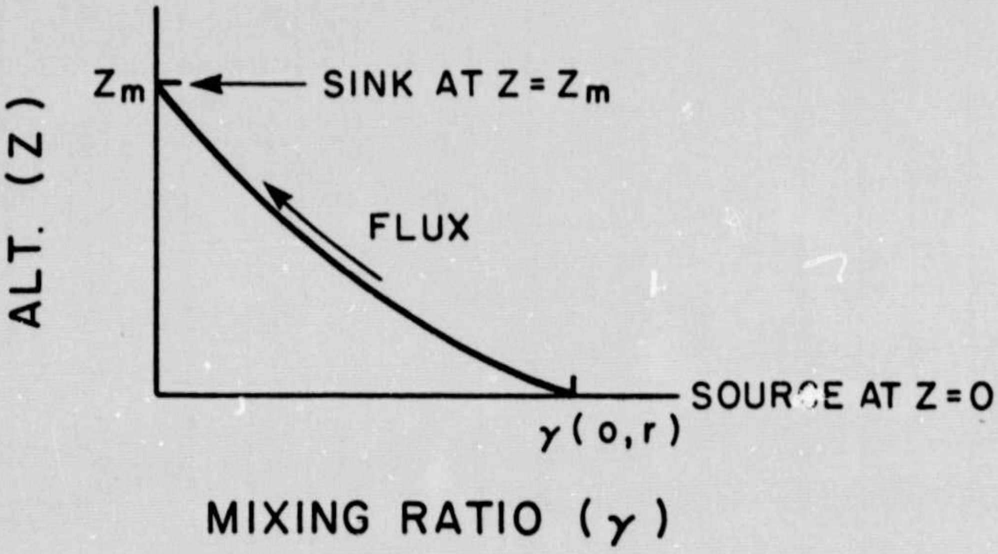
$$1/g'(z) \equiv (e^{z/H} - e^{z_m/H})\ uH/D$$

Some useful examples of solutions follow.

1. Source at $z=0$ and sink at $z=z_m > 0$.

$$\gamma(z,r) = \gamma(o,r)\ (e^{-r/g(z)} - e^{-r/g(z_m)})\ (1-e^{-r/g(z_m)})^{-1}$$

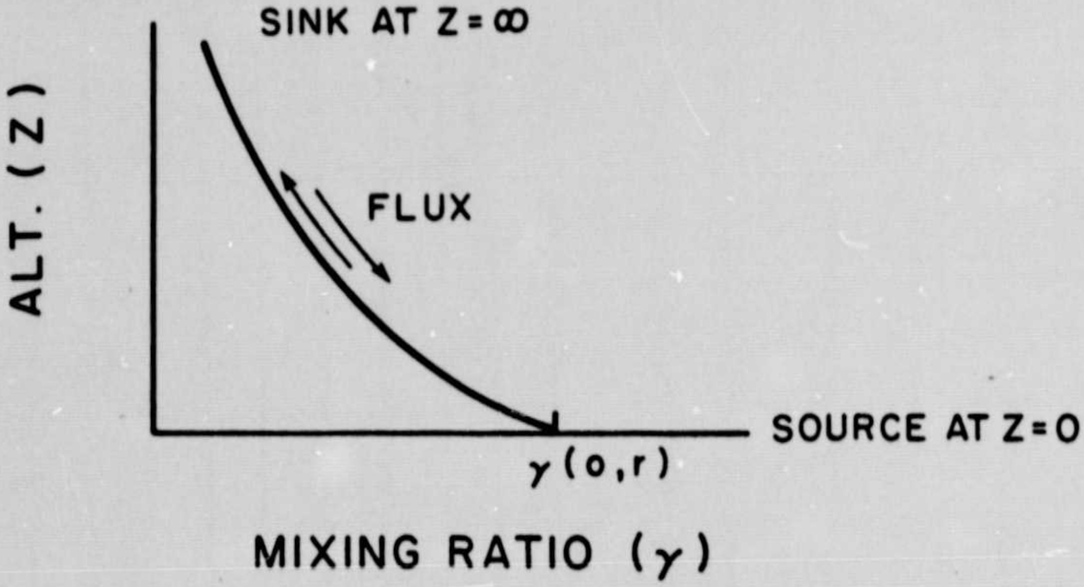
$$\phi = \text{constant} = \rho(z_o)ur\ \gamma\ (o,r)\ e^{-r/g(z_m)}\ (1-e^{-r/g(z_m)})^{-1}$$



2. Source at $z=0$ and sink at $z=\infty$.

$\gamma(z,r) = \gamma(o,r)e^{-r/g(z)}$

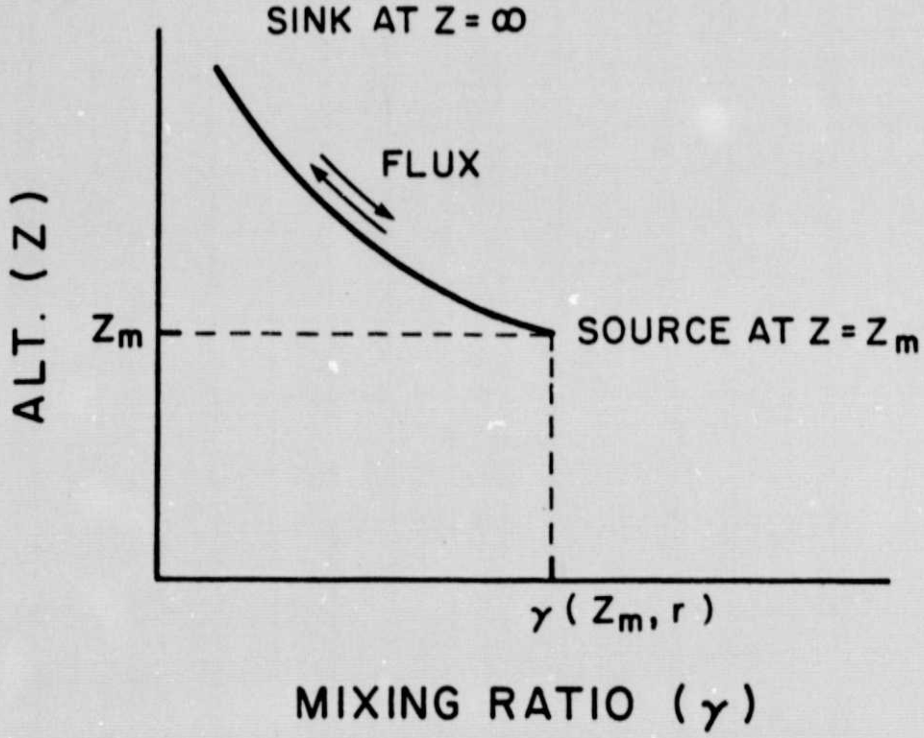
$\phi=0$



3. Source at $z=z_m$ and sink at $z=\infty$.

$\gamma(z,r) = \gamma(z_m,r)\ e^{-r/g'(z)}$; $\gamma(z_m,r)$ given

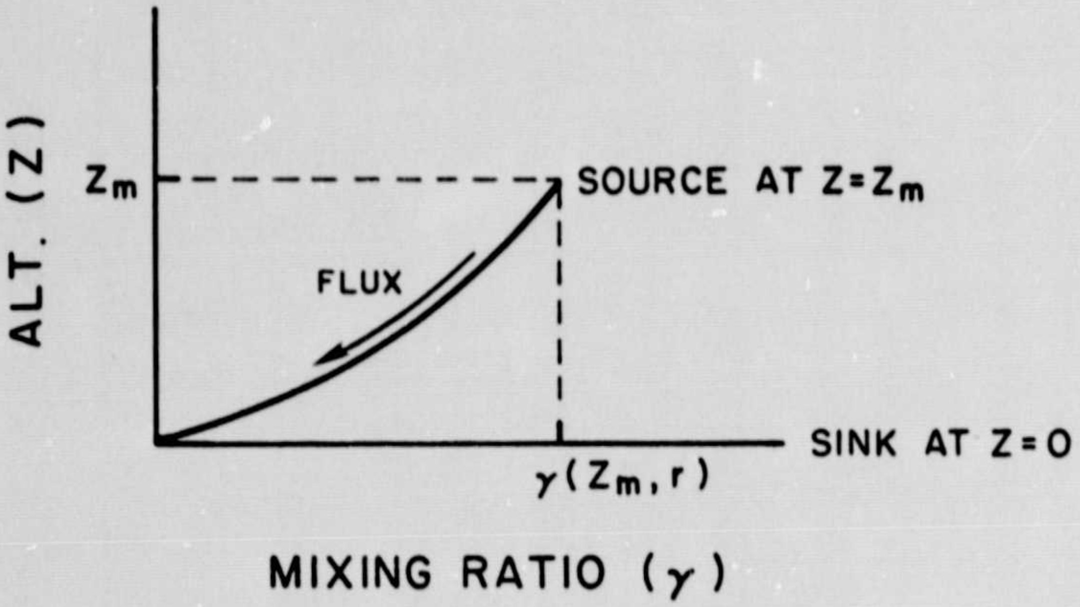
$\phi = 0$



4. Source at $z=z_m$ and sink at $z=0$.

$$\gamma(z,r) = \gamma(z_m,r)\,(1-e^{-r/g(z)})\,(1-e^{-r/g(z_m)})^{-1}$$

$$\phi = -\ \gamma(z_m,r)\,\rho_o ur\,(1-e^{-r/g(z_m)})^{-1}$$

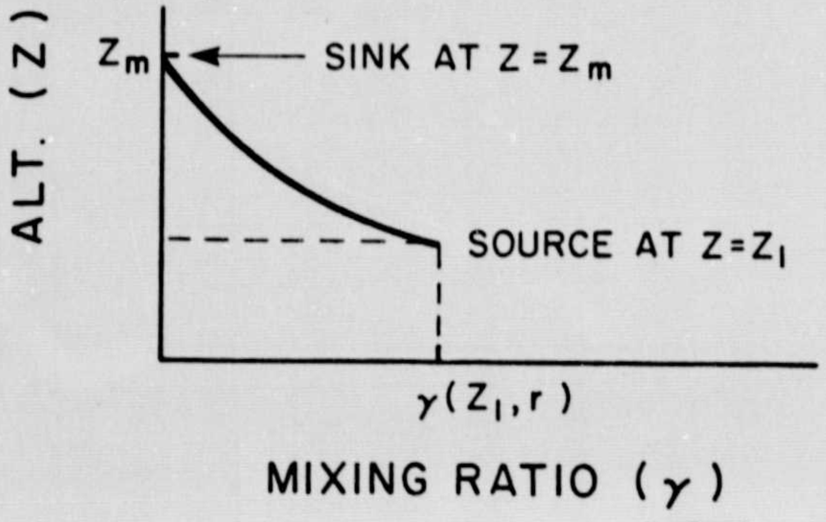


5. Source at z_1 and sink at $z=z_m$.

$(0<z_1<z_m)$

$$\gamma(z,r) = \gamma(z_1,r)\,(e^{-r/g'(z)} - e^{-r/g'(z_m)})\,(1-e^{-r/g'(z_m)})^{-1}$$

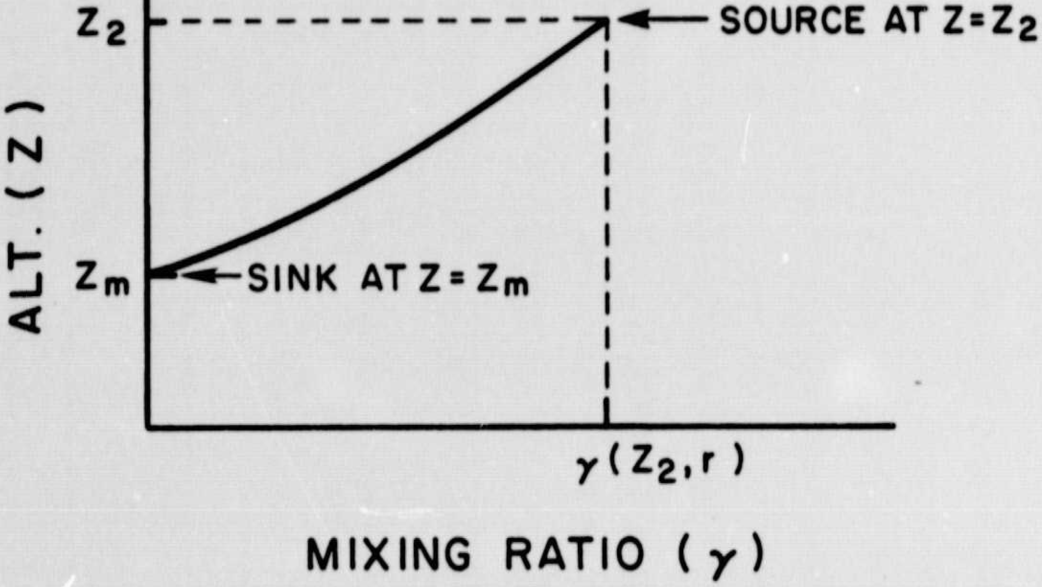
$$\phi = \gamma(z_1,r)\,\rho_o ur\,e^{-r/g'(z_m)}\,(1-e^{-r/g'(z_m)})^{-1}$$



6. Source at z_2 and sink at $z=z_m$. $(z_2 > z_m > 0)$

$$\gamma(z,r) = \gamma(z_2,r)\,(1-e^{-r/g'(z)})\,(1-e^{-r/g'(z_2)})^{-1}$$

$$\Phi = -\,\gamma(z_2,r)\,\rho_o ur(1-e^{-r/g'(z_2)})^{-1}$$



7. Source at $z = +\infty$ and sink at z_m.

$\gamma(z,r) = \gamma(\infty,r)\,(1 - e^{-r/g'(z)})$; $\gamma(\infty,r)$ given

$\Phi = \text{constant} = -\gamma(\infty,r)\,\rho_o ur$