

Radiative Properties of the Middle Atmosphere

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ABSTRACT In this paper a brief outlines of the atmospheric formation, destruction and distribution processes and size distributions. Also recent measurement of aerosol is presented. Some important aspects of the radiative effects of the aerosols are discussed.

KEY WORDS aerosol, radiative budget, climate

1. Definition and Concepts of Aerosol

1.1. Introduction

Atmospheric aerosols play a very important role in many atmospheric processes some of which may have in turn an impact on the climate.

however, our ability to assess the role of aerosols in atmospheric processes is greatly disabled by the very limited information available on the properties of aerosols. The inputs to atmospheric aerosols come from natural and manmade sources.

The recent thrust to aerosols studies came from the concern about the possible effects of manmade aerosols on atmospheric processes and climate.

However, a realistic assessment of this cannot be made because of our incomplete knowledge about the characteristics of natural aerosols and their role in various atmospheric processes.

The radiation budget of the middle atmosphere is influenced primarily by ozone, carbon dioxide and water vapour.

Aerosols affect the radiation budget through scattering and absorption processes. For a proper evaluation of this effect, the data required on aerosol characteristics are rather scanty.

The size range can again be subdivided into three ranges based on atmospheric effects of aerosols. The smallest particles of size less than $0.1\mu\text{m}$ are called Aitken particles and are important in atmospheric electricity. Particles in the size range $0.1\mu\text{m}$ to $1.0\mu\text{m}$ are referred to as large particles are effective in atmospheric optics and in radiation budget. These are also defective as condensation nuclei. Particles of size range $>1.0\mu\text{m}$ play an important role in cloud physics and are effective as condensation nuclei.

The large particles ($0.1\mu\text{m}$ to $1.0\mu\text{m}$) sometimes called haze particles. Aerosol particles are either directly produced or formed by various gas-to-particle conversion processes.

In either case, the 'original' aerosol particle

undergoes a variety of chemical processes and physical modifications in the atmosphere.

These processes and modifications result in a continually changing chemical composition and particle size distribution and hence physical properties. Particles from different sources are mixed by Brownian diffusion and coagulation processes on a microscale and by atmospheric turbulence and circulation on a macroscale. The principle sources of aerosols are listed in Table 1 with approximate estimates of their input levels.

The wide diversity in the different estimates is an indication of the uncertainty involved in making the estimates and the need for more relevant information. Some of the characteristics of the size distribution of aerosols depend upon the various formation and transformation processes of aerosols.

Particles in the size range below about $1\mu\text{m}$ (transient/Aitken nuclei range) are produced by gas-to-particle conversion processes whereas particles of size greater than $1\mu\text{m}$ (coarse range) are formed directly by mechanical processes such as wind blown dust and sea salt spray.

Particles at the lower end of the size range have relatively short atmospheric residence times because of chemical reactivity and greater physical mobility. These particles will eventually be incorporated in the $0.1\mu\text{m}$ to $1.0\mu\text{m}$ range which is generally referred to as the accumulation mode.

Table 1 Estimates of particle production in 10^{+6} tons/year

source	After Peterson and Brook	After Hidy and Jungle
Man-made		
(i) Direct particle production		
Transportation	1.8	
Stationary fuel sources	9.6	
Industrial processes	12.4	
Solid waste disposal	0.4	
Miscellaneous	5.4	
Subtotal	29.6	37-110

(ii) Particles formed from gases		
Converted sulphates	200	110
Converted nitrates	35	23
Converted hydrocarbons	15	27
Subtotal	250	160
Total man-made sources	280	197-270
Natural		
(i) Direct particle production		
Sea salt	500	1095
Wind blown dust	250	60-360
Volcanic emissions	25	4
Meteoroid debris	0	0.02-0.2
Forest fires	5	146
Subtotal	780	1610
(ii) Particles formed from gases		
Converted sulphates	335	37-365
Converted nitrates	60	600-620
Converted hydrocarbons	75	182-1095
Subtotal	470	2080
Total of natural sources	1250	3690
Grand Total	1530	3960

Conversion of particles in the transient and accumulation range to coarse range and vice versa is not a very efficient process.

Thus, particles in the lower two range are quite distinct from those in the coarse range.

1.2. Gas-to-Particle Conversion Processes

It is seen from Table 1 that gas-to-particle conversion processes is a major contribution to the aerosol particle production. It contributes more than half of the total particle production due to natural sources as well as man-made sources. Atmospheric gases can interact with each other and with existing particles to form new particles or to modify the existing particles by different processes.

The main processes are (1) homogeneous homomolecular nucleation, (2) homogeneous heteromolecular nucleation, and (3) heterogeneous heteromolecular nucleation.

The first process involves formation of new, liquid or solid ultrafine particles from a gas phase consisting of a single gas species only.

The second process involves formation of new particles from a gas phase consisting of two or more gaseous species. In this, the most common one of the species is water vapour.

In the third process, i.e. heterogeneous, heteromolecular nucleation growth of pre-existing particles takes place due to condensation of gaseous species. Atmospheric trace gases undergo chemical reactions to produce reaction products with a low vapour pressure.

As more and more of these products are formed, a state of supersaturation will be reached with respect to these molecules. The degree of supersaturation will determine the degree of nucleation (formation of new particles) and condensation (deposition on pre-existing particles). From the thermodynamic point of view, more energy is required to form a new particle (nucleation) than to enlarge the surface of the existing particle (condensation).

The following reactions are considered to be important for the gas-to-particle conversion: (1) sulphur dioxide reacts with hydroxyl radicals to form eventually sulphuric acid molecules, (2) non-methane hydrocarbons react with ozone and/or hydroxyl radicals to form aldehydes, alcohols, carboxylic acids and dicarboxylic acids, and (3) most secondary reaction products of non-methane hydrocarbons react with oxides of nitrogen to form organic nitrates.

The particles formed by these mechanisms are in the fine particle size range ($<0.5\mu\text{m}$). Gas-to-particle conversion can also take place through direct reaction of gases with particles on the surface or in the interior in case of liquid particles. Gas-to-particle conversion plays a major role in the aerosol production in the stratosphere. Sulphur dioxide is converted through various reactions to sulphate aerosols in the stratosphere. In fact it is this process of gas-to-particle conversion which is primarily responsible for the Junge layer in the stratosphere.

Ever since its discovery Junge the stratospheric aerosol layer has been a subject of great interest. It has been well established that the stratospheric particles are primarily composed of liquid solutions of sulphuric acid and water. Other substances like ammonium sulphate are also generally present.

These sulphate particles are produced locally in the stratosphere mainly from precursor sulphur-bearing vapours. Sulphur dioxide (SO_2) and carbonyl sulphide (OCS) are the most important precursors SO_2 , which is an important component of volcanic effluents, is injected directly into the stratosphere in large quantities by major volcanic eruptions.

During periods of quiescent volcanic activity, the major precursor gas is carbonyl sulphide (OCS) with carbon disulphide (CS_2) as a possible secondary source (Sze and Ko; Crutzen). Unlike most of the other sulphur gases, OCS is expected to have a long tropospheric lifetime of several years. OCS originates in biological volcanic and industrial processes (mainly from the surface). CS_2 is less inert than OCS but it has agronomic significance because of its possible oxidation to OCS.

Apart from OCS and CS₂, SO₂ from surface emission is also transported to lower stratosphere even during periods of quiescent volcanic activity.

Detailed modelling studies have been made of the tropospheric OCS cycle and the stratospheric sulphur balance during volcanically quiescent times .

These studies show that OCS is the dominant sulphur source compared to CS₂ and SO₂ for the stratospheric aerosol layer.

The precursor gases from troposphere are transported to stratosphere through eddy diffusion and direct injection. The direct injection can occur during thundercloud penetration, tropopause folding and vertical convection especially in the tropical zone. Horizontal advection subsequently distributes injected material zonally and meridionally.

In the lower stratosphere, OCS breaks up into sulphur and carbon monoxide by photo-decomposition due to absorption in the extreme ultraviolet, 200-400nm. The sulphur atoms thus produced react rapidly with molecular oxygen to form SO which in turn is rapidly oxidized to SO₂. SO₂ is further oxidized mainly through reactions with OH(three-body reaction) to H₂SO₄.

Depending the saturation levels, the H₂SO₄ and H₂O vapours nucleate by heterogeneous heteromolecular or homogeneous heteromolecular processes .

It is generally believed that the former process is much more efficient than the latter one.

However, recent assessment in this regard brought out that at temperatures around -75°C which are not unusual in the stratosphere, the homogeneous nucleation mechanism can be a significant contributor to the overall nucleation process.

Yue and Deepak studied the relative contribution of the two nucleation processes at different temperatures and vapour pressures(of H₂SO₄ and H₂O) and arrived at the above result.

Other processes of nucleation include ion nucleation and ion-ion precondensation nucleation. Ion nucleation involves condensation of gaseous vapours(e.g., H₂SO₄ and H₂O) on charged molecular clusters .

The electrostatic energy of the core ion stabilizes the nucleation embryo, effectively lowering the barrier to nucleation. The ion-ion precondensation nuclei may be formed when large cluster ions opposite charges recombine.

The contribution of these processes to the total nucleation is expected to be not significant compared to the other processes described above.

After nucleation, an aerosol continues to grow by vapour condensation .

Both evaporation and attachment processes take place at any given instant and the net effect of these two processes determines the rate of growth by condensation.

In the troposphere, growth of aerosol particles by condensation of water vapour plays an important role.

1.3. Role of Meteoric Debris

The meteoric influx to the earth amounts to 16 mega tones per year. The meteor input constitutes only a few percent. Hunten and Truco made detailed theoretical calculations on the effects of meteoric debris on stratospheric aerosols.

Truco concluded from their model calculations that meteoric debris is an important natural aerosol constituent for particles large than 1µm radius throughout the stratosphere and may be dominant for particles smaller than 0.01µm radius above 20 km.

For other sizes, the meteoric debris generally constitutes less than 10 percent of the total aerosol mass. Their calculations show that very few meteoric particles reach the upper troposphere in their natural state and participate as cloud ice condensation nuclei.

This is in contrast to the Brown's hypothesis of relation between precipitation and meteoric dust influxes.

1.4. Sedimentation

The fall velocity and the diffusion coefficient are basic parameters to determine the sedimentation rate. These parameters are particle size dependent.

At sizes comparable to molecular dimensions, kinetic theory of gases is applicable and for larger particles Stokes-Cunningham formula is applicable.

For large particles, the fall velocity is given by

$$v = msgBs \quad (1)$$

and the diffusion coefficient by

$$dcs = kTBs$$

where

ms Mass of the particle

g Acceleration due to gravity

k Boltzmann's constant

Bs Particle mobility, defined as the velocity per unit driving force

The approximate values of fall velocity for different sizes at different heights are given in Table2.

Table 2. Fall Velocities of Particles of Different Sizes

Altitude Km	Velocity(cm/s)at r=			
	5µm	2µm	1µm	0.5µm
30	0.5	0.24	0.06	0.015

20	0.17	0.08	0.02	0.005
15	0.12	0.055	0.014	0.0035
10	0.10	0.045	0.011	0.003

1.5. Coagulation

Coagulation is controlled by the diffusion coefficient and is thus important for smaller particles. Coagulation controls the smaller particle end of the aerosol size distribution whereas sedimentation controls the larger particle end.

The rate of coagulation between two groups of particles of different sizes within a population $n(r)$ is given by

$$Dn(r_1, r_2)/dt = -4\pi(D_1 + D_2)(r_1 + r_2) \times n(r_1)n(r_2)dr_1dr_2 \quad (2)$$

where $D(=kTB)$ is the diffusion coefficient and $n(r)$ is the concentration of particles.

Substituting for D ,

then it can be written as

$$dn(r_1, r_2)/dt = K_0 f(r_1, r_2)n(r_1)n(r_2)dr_1dr_2 \quad (3)$$

The combination of r_1 and r_2 result in a particle with

$$r = (r_1^3 + r_2^3)^{1/3}$$

The largest particles which can thus participate in such formation of particle with radius r are of size, $r_1 = r/2^{1/3}$.

The total number of particles with radii r formed during dt is then

$$I_1 = \int K_0 f\{r_1, (r^3 - r_1^3)^{1/3}\} n(r_1)n((r^3 - r_1^3)^{1/3}) dr_1 dr dt \quad (4)$$

The number of particles with radii decreases by coagulation with smaller and larger particles by

$$I_2 = \int -K_0 f(r_1, r)n(r_1)n(r)dr_1 dr dt \quad (5)$$

So, the total change in particles with radii is given by

$$\Delta n(r) = \int (I_1 + I_2) dt \quad (6)$$

This results in a displacement of the maximum towards larger particles .

1.6. Aerosol Removal Processes

There are two processes of direct removal of particles from the atmosphere .

These are (1) dry deposition, i.e. gravitational sedimentation, and (2) wet removal. Wet removal is of two types, namely, rain out and washout. In rain out the particles are incorporated in precipitation nuclei during processes occurring within the cloud. Washout involves incorporation of a material into precipitation as a consequence of processes occurring below the cloud.

These removal processes are highly size dependent. Furthermore, the removal rate of

hygroscopic and non-hygroscopic particles will be different because hygroscopic aerosols will increase greatly in size with increasing relative humidity. The wet removal processes are important mainly in the lower troposphere where cloud formation exist.

However, in tropical latitudes where the vertical extent of the clouds (thunderclouds) often extends into the stratosphere, the wet removal processes can become important even at altitudes of upper troposphere and lower stratosphere.

Study of precipitation chemistry is important in understanding the effect of wet removal processes.

1.7. Aerosol Size Distributions

The relationship between aerosol producing processes and their contribution to different aerosol size ranges has been discussed earlier.

The size distribution of aerosols is very important information in the study of effect of aerosols on the atmospheric processes. The aerosol size distribution can be expressed in terms of number density distribution or surface area and volume distribution.

The surface area and volume distributions are very important in the study of aerosol chemistry.

Junge found that the number density size (radius r) distribution can be approximated by a power law in the size rang 0.04-10 μ m in the case of continental aerosols.

The log radius-surface(s) and log radius-volume (V) distributions are written as

$$ds(r)/d(\log r) = 4\pi r^2 n(r)/d(\log r) \quad (7)$$

and

$$dV(r)/d(\log r) = 4/3\pi r^3 n(r)/d(\log r) \quad (8)$$

The power law index (v) is typically 3 but could vary from 2 to 6. This type of distribution has been very useful as it is a simple function .

However, it is generally found to be more representative, if the size distribution (especially of troposphere aerosols) is expressed as a sum of log normal distributions with different mode radii. It appears that each particle source will have particles with their own log normal distributions.

So, with multiple sources, which is in general the case, a multi modal log normal distribution with each mode specified by a total number density, a spread parameter and a mode radius will represent the size distribution.

Two modes are generally adequate to characterize most aerosol distributions.

A third mode may be necessary to represent the Aitken nuclei especially near sources of combustion particulates. Stratospheric aerosols can be represented by a single mode as these are

dominated mainly by SO₄ aerosols.

The numbers on each distribution denote the height above the tropopause. There is remarkable similarity in the distributions at $r > 0.1$ m. Turco et al. explained this as due to the constant input rate of gaseous sulphur to the stratosphere and to the dependence of aerosol removal rate on particle size.

The removal rate varies as r^4 because of an r^3 dependence of particle mass and an r^3 dependence of sedimentation rate.

These factors constrain the size distribution to a relatively small variation.

At sizes less than $0.1 \mu\text{m}$, the small particle concentration decreases with increasing separation from tropopause.

This can probably be attributed to increasing aerosol maturity with altitude in the stratosphere.

1.8. Aerosol Continuity Equation

The diffusive flux of the particles is usually derived from the relationship

$$\phi_0 = -D_e n_v \partial(n/n_v) / \partial z \quad (9)$$

where ϕ_0 is the eddy diffusion coefficient and n is the air number density.

The eddy diffusion coefficient is often derived from measurements of long-lived tracers such as N₂O and CH₄ (species which are not produced photochemically in the atmosphere). The aerosol continuity equation can be written by including the rates of production, destruction (removal) and transport.

This is written as

$$\partial n / \partial t = Q - un - y - \partial(ny) / \partial z - \partial(\phi_0) / \partial z \quad (10)$$

In above equation, Q can be taken represent the rate of aerosol production due to different processes, u ($=1/t$, t is the residence time) is the wet removal rate, and y is the coagulation rate. Recourse has to be taken to numerical methods to solve the continuity equation. One-dimensional modelling using the continuity equation has been carried out for stratospheric aerosols and the results have been reviewed by Turco et al. The model predictions compare fairly well with the observations.

However, realistic three-dimensional modelling is needed to understand the global variation of stratospheric aerosols. Modelling of tropospheric aerosols is still in its infant stage mainly because of the plethora of sources of different nature.

2. Aerosol Radiation interaction

The scattering of electromagnetic radiation in the atmosphere involves molecules and particles both smaller and larger than the wavelength of the

radiation. If the scattering is dominated by particles small compared to the wavelength it is referred to as 'Rayleigh Scattering'.

When the particle dimension is not negligible compared to the wavelength, a more complicated scattering pattern is found and this type of scattering is known as 'Mie scattering'.

The complete scattering theory for a spherical scatterer of arbitrary size was first given by the German mathematician Gustav Mie in 1908. Mie scattering includes Rayleigh scattering as a simpler limiting case.

2.1. Rayleigh Scattering

In 1890's Lord Rayleigh showed that the scattering of light by air molecules is responsible for the blue colour of the sky. He showed that, when the size of the scatterer is small compared to the wavelength of the incident radiation a dipole is induced within the scatterer.

The induced dipole is in the same direction as the incident electrical vector, its moment proportional to the field and its phase same as that of the incident field.

Electromagnetic theory shows that the dipole radiates a polarized wave with one component of the amplitude falls off as r^2 (r being the distance from the dipole) and the other falls off as r^1 .

The r^2 dependence is valid only near the immediate vicinity of the dipole.

We will now consider the problem of obtaining the electromagnetic field at a point away from the immediate vicinity of the dipole (known as far field

$$\iint (1 + \cos^2 \theta) \sin \theta d\theta d\phi = 16\pi / 3$$

solution, where the field falls off as r^1).

To obtain the scattered electromagnetic field at a point p , it is convenient to resolve the field into two components, one lying in the scattering plane (the plane containing the incident radiation, the scatterer and the scattered radiation) and the other perpendicular to the scattering plane.

Following the notation introduced by Chandrasekhar in 1950, subscript l is used to denote the parallel component and subscript r is used to denote the perpendicular component.

The arbitrary incident electric vector can be written as

$$(E_{0r} + E_{0l}) \exp(i\omega t) \quad (11)$$

At p the dipole E_{0r} gives a field component,

$$E_r(r) = \omega^2 / C^2 r \exp[i\omega(t-r/c)] E_{0r} \quad (12)$$

and the dipole E_{0l} gives a field component,

$$E_{0l}(r) = \omega^2 / C^2 r \exp[i\omega(t-r/c)] E_{0l} \cos \theta \quad (13)$$

The intensity of the scattered radiation becomes,

$$I_{\theta} = |E|^2 = (\omega^4 / C^4 r^4) \alpha^2 (1 + \cos^2 \theta) I_0 \quad (14)$$

where θ is the scattering angle, ω is the frequency of the incident radiation, C is the velocity of light and r is the distance between the scatterer and the point under consideration. α is the polarizability of the scatterer.

For a spherical Rayleigh scatterer we can rewrite the above expression in terms of refractive index m , and radius a of the scatterer as,

$$I_{\theta} = (\omega^4 a^6 / C^4 r^2) (m^2 - 1/m^2 + 2) (1 + \cos^2 \theta) I_0 \quad (15)$$

Thus the scattered intensity from a single very small particle varies as the sixth power of its radius. The error in applying Rayleigh theory to rather than Mie theory to small particles is less than 1% when $r \leq 0.03\lambda$. This is a useful rule to remember.

In radiative transfer work it is often necessary to consider the normalized phase function.

The phase function describes the portion of the scattered radiation that appears per unit solid angle in the direction θ .

For Rayleigh scattering it is obtained by multiplying $(1 + \cos^2 \theta)$ by normalizing factor obtained by integration over all solid angles,

So, the Rayleigh scattering phase function becomes,

$$Y(\theta) = 3/16\pi (1 + \cos^2 \theta) \quad (16)$$

The characteristics of Rayleigh scattering are:

(i) The Scattered intensity is proportional to ω^4 , i.e., to the -4th power of the wavelength.

(ii) The scattered intensity is proportional to the square of the polarizability, and hence for a spherical particle the sixth power of radius.

(iii) The angular distribution of the scattered intensity is given by $(1 + \cos^2 \theta)$.

The distribution is symmetric in the forward and backward directions.

At 90° the parallel component vanishes and hence the scattered radiation is plane polarized and perpendicular to the scattering plane.

Note that the intensity of Rayleigh scattering is proportional to the sixth power of the radius of the scatterer. The absorption on the other hand is proportional only to the cube of the radius, i.e., to the volume.

This has the consequence that, with decreasing size, scattering falls off more rapidly compared to absorption leading to a decrease in single scattering albedo with decreasing size.

2.2. Mie Scattering

When the size of the scatterer becomes comparable to the wavelength (of the incident radiation, it is no longer sufficient to consider merely

an induced dipole. In such cases, a three dimensional charge distribution is set up within the scatterer with non-vanishing quadruple and higher moments and the scattering processes is known as 'Mie scattering'. We will not derive the Mie solution here.

In Mie scattering neither the absolute value of λ nor the absolute value of particle size is important, rather the ratio of the two values become the governing factor.

The ratio of the particle radius to wavelength is expressed by the dimensionless size parameter, x .

$$x = 2\pi r / \lambda \quad (17)$$

The problem is to obtain three dimensional solution for Maxwell's equations for the electric and magnetic fields inside and outside a spherical region of radius a , subject to certain boundary conditions at $r=a$.

The Mie solution process is to find a set of complex numbers a and b , called Mie coefficients, which give vectors E and H that satisfy the boundary conditions at the surface of the sphere. The solution is obtained in term of two complex functions $S_1(\theta)$ and $S_2(\theta)$ which are the complex amplitudes of the scattered wave.

$$S_1(\theta) = \sum (2n+1) / n(n+1) (a_n \pi_n + b_n T_n) \quad (18)$$

$$S_2(\theta) = \sum (2n+1) / n(n+1) (b_n \pi_n + a_n T_n) \quad (19)$$

π_n and T_n are directives of legendre polynomial of order n and argument $\cos \theta$.

Theoretically speaking, the summation is done from $n=1$ to infinity.

However, in practice the solution converges for n values between 30 to 50 depending on value of α . The polarized components of the scattered intensity I_r and I_l are obtained by squaring the amplitudes

$$I_r(x, m, \theta) = S_1^2$$

$$I_l(x, m, \theta) = S_2^2 \quad (20)$$

and the net scattered intensity becomes

$$I(\theta) = (I_r + I_l) / 2 \quad (21)$$

The important characteristics of Mie scattering are:

(i) The angular distribution of the scattered radiation intensity is complicated and the complexity increases with increasing particle size.

(ii) The ratio of the energy scattered in the forward direction increases with increasing particle size.

(iii) The dependence of scattering on wavelength decreases with increasing particle size.

The most notable feature of Mie scattering theory is the forward peak.

If we start with a very small scatterer the scattering diagram would be found to be symmetric,

equal amounts of energy being scattered in the backward and forward directions.

If the particle size is increased, the smooth $(1 + \cos 2\theta)$ shape of the Rayleigh scattering diagram would develop asymmetry in the backward direction.

When the particle size parameter, x exceeds unity the scattering diagram begins to develop peak and troughs.

There is a rough correspondence between x and the number of peaks and trough between 0° and 180° with always a strong peak in the forward direction at $\theta=0^\circ$.

If the particle size is held constant and the imaginary part of the refractive index (which is proportional to the absorption coefficient) is increased the forward lobe is not generally affected but the radiation scattered in the backward direction is most affected.

As far as polarization is concerned, for size parameters up to about 2, the polarization is similar to that for Rayleigh scattering.

For size parameter between 4 and 10 the polarization is opposite to that for Rayleigh scattering at near-forward and near-backward scattering angles.

For larger size parameters and in scattering directions other than near forward or near backward,

$$\sigma_a = 2\pi \int (r^2 / x^2) i(x, \theta, m) \sin \theta d\theta \quad (22)$$

a rapid variation of polarization with both size and angle is found.

2.3. Integrated Optical Properties of Aerosols

In the atmospheric point of view, we are concerned not with the properties of single particles but with integrated properties relating to a volume element

$$\beta_a = 2\pi \iint dn(r)(r^2 / x^2) i(x, \theta, m,) \sin \theta d\theta \quad (23)$$

of the atmosphere. This volume element may contain many particles of varying size, shape and composition in addition to air molecules and other absorbing gases. To calculate integrated effects for a volume of aerosols, the intensities contributed by various particles have to be added.

The scattering coefficient σ_a of an aerosol particle is the sum of the radiation scattered by the particle in all 4π direction

when there are $do(r)$ particles per cubic centimeter, all particles with radii r contribute to scattering

The extinction (total reduction in the energy of the incident beam due to scattering and absorption) of the solar beam as it traverses the atmosphere is primarily determined by the aerosol content of the air. If the air is very clean, the extinction by aerosol

may be comparable or less than the air molecule scattering.

In such case the air molecule scattering can be computed theoretically and subtracted from the total to obtain the loss due to aerosol scattering.

However, when the aerosol amount is high, there will be interactions between molecular and particulate scattering and multiple scattering becomes applicable. By multiple scattering we mean that the radiation received by a detector is scattered more than once by both particles and molecules.

As it has been mentioned earlier, there are number of peaks and troughs in the angular distribution of the scattered intensity scattered by a single particle.

However, when the intensity patterns of the many members of a distributed population of particles are combined, maxima for one size tend to be offset by minima for another size and the overall scattering diagram is smoother than a single particle.

For a normally incident solar beam with initial intensity $I_0(\lambda)$ the attenuation while passing through the atmosphere is $\exp(-T\lambda)$.

For all other solar zenith angles X the attenuation is $\exp(-T\lambda \sec X)$, where $T\lambda$ is defined as the optical thickness of the atmosphere.

If $T\lambda$ remains constant in time while solar zenith angle changes, a simple logarithmic plot of radiation flux against $\sec X$ enables $I_0(\lambda)$ and $T\lambda$ to be determined because

$$\log I_0(\lambda) = \log I(\lambda) - T\lambda \sec X \quad (24)$$

The negative of the slope of $\log I(\lambda)$ versus $\sec X=0$ gives $T\lambda$ and the extrapolation of the plot to $\sec X = 0$ gives $I_0(\lambda)$, the intensity of the radiation at the top of the atmosphere.

The aerosol optical thickness T_a (evaluated by subtracting Rayleigh optical thickness from the total) when plotted against wavelength, often exhibits a power law dependence of the form $\lambda^{-\alpha}$.

The parameter $-\alpha$ is first introduced by Angstrom in 1931 and is referred to as the Angstrom turbidity coefficient. So far we have been considering only single scattering. But, a photon once scattered can be scattered again and depending on the density of the medium.

Therefore the radiation emerging in any direction will be composed of photons which have been scattered once, twice or more often. It can be shown that the optical thickness T is the quantity which determines whether secondary and higher order scattering are important in a given scattering volume.

The optical thickness is given by $\int k ds$ where k being the attenuation coefficient. From this

definition it follows that the fraction of the incident radiation which emerges without being scattered is e^{-T} , which is also defined as transmission.

Therefore the fraction of incident radiation which has been scattered once is $(1-e^{-T})$ and that part which is scattered again is of order $(1-e^{-T})^2$. If $(1-e^{-T})$ is small, that part is when is small, secondary and higher order scattering can be neglected.

It can be said that if the scattering volume is optically thin ($T \ll 1$) only single scattering need to be considered. For optically thick layer ($r \gg 1$) multiple scattering effect should be taken into account.

3. Aerosol Measurement

3.1. Experimental Methods for Aerosol Studies

Experimental methods for the study of atmospheric aerosols can be classified into two categories: direct methods and indirect methods. The direct methods involve actual collection of aerosol samples from the atmosphere using impactors and analysing these to determine the characteristics such as chemical composition and size distribution by laboratory methods.

The impactors are carried by balloons and the collected samples are recorded for later analysis.

Another direct methods is the filter method in which aerosols of different size range are collected by using different filters and are analysed for their mass distribution and chemical composition.

Indirect methods use the aerosol property to scatter and absorb the incident light and thereby causing extinction. The scattered light signal strength or the attenuation of the light signal as it traverses a known path in the atmosphere is recorded. This can be analysed to determine aerosol characteristics such as number density and size distribution.

The indirect methods can be carried out in site using balloon or rocket platforms. Balloon-borne dustsondes are among the indirect in site experiments that have been widely conducted. In the balloon-borne dustsondes the air containing aerosols is pumped into an illuminated cell.

The light scattered by the pumped-in air is recorded. As aerosols pass through the cell they give rise to scattered light pulses. The number of pulses and the pulse width give information on the aerosol number density and the size when analysed using calibration data.

Another indirect method using balloon-borne/rocket-borne payloads is the light scattering method. In this method, the scattered sunlight by air molecules and aerosols is recorded

at different scattering angles at different altitudes. The recorded signal strength as a function of scattering angle is analysed to derive the aerosol characteristics.

The ground-based techniques for the measurement of aerosol characteristics employ indirect methods. Among these are the turbidity measurement technique and the multi-wavelength solar radiometer. Turbidity measurements are carried out by a sun photometer using wide band filters. These measurements can be used to obtain the Angstrom's turbidity coefficient and the wavelength exponent using power law size distribution for aerosols.

As the size distribution differs significantly from power law distribution, especially for the lower tropospheric aerosols which contribute most to the total atmospheric turbidity, it is necessary to make measurement at a number of wavelengths using narrow band filter to derive the size distribution function. Multi-wavelength radiometers (MWR) use this method.

In MWR, the aerosol optical depth is obtained as a function of wavelength by making measurements of direct solar flux at a number of wavelengths in the visible at different solar zenith angles.

By adopting inversion methods the columnar aerosol size distribution function can be derived from MWR observations.

A very powerful ground-based method for the study of aerosol characteristics with good altitude resolution is the lidar (acronym for Light Detection and Ranging) method. In lidar (operated in monostatic mode) a high power pulsed laser is used as a transmitter. The laser beam is directed into the atmosphere by suitable optics. The backscattered light from the atmosphere is received by a suitable telescope and is detected by a photomultiplier tube whose output is further amplified for recording. The backscattered signal strength is analysed to obtain information on aerosol characteristics.

3.2. Aerosol measurement

We have measured the aerosols during three months from March to April in 1999. The measurement was done by means of balloon-borne impactor.

Fig1 Aerosol number density(m⁻³)

Altitude profiles of aerosol extinction/number density have been obtained. The balloon-borne impactor responds to the particle size range 0.1-1.0µm with maximum sensitivity at 0.15µm.

The obtained profile is shown Fig.1 and also the variations of total mass of aerosols are shown Fig.1. The profile indicates that the total mass of aerosols is greater in March than April due to strong winds.

4. Effect of Aerosols on Radiation in the Middle Atmosphere

We are to present a few important aspects of the effects of aerosols on the radiation in the middle atmosphere.

The solar radiation spectra at the top of the atmosphere and at the sea level are well known in many literatures. The difference between the top of the atmosphere and the sea level is due to scattering.

The radiation field should be defined at each point in space and for each wavelength for the study of interaction of radiation with the atmosphere and resulting agronomic and thermal effects.

The analysis of the radiation field can be done from the equation of radiative transfer. The radiative transfer equation expresses the energy balance in each unit volume of the atmosphere including absorption, scattering and emission. The equation can be solved by analytical methods only for the most simple cases.

To obtain quantitative solutions numerical methods are generally used. The most important elements contributing to the middle atmosphere radiation budget are of cooling and heating rates.

There are large variations in the vertical distribution of solar short wave heating rates by O₃, O₂, NO₂, H₂O and CO₂ and terrestrial long wave cooling rates by CO₂, O₃ and H₂O. The mean heating due to O₃ is balanced by the corresponding cooling due to infrared radiation from CO₂(15µm), O₃(9.6µm) and water vapour(15µm).

Near the stratosphere the 15µm band of CO₂ yields a radiative temperature change(net cooling) about twice as large as the 9.6µm band of the O₃ and about 10 times larger than that from the 18µm

band of water vapour.

4.1. Effect of Aerosol on the Atmospheric Heating

Aerosol absorb energy from the solar and planetary radiation fields, exchange energy by collisions with the ambient gas and emit thermal radiation.

In case any phase change is possible, they acquire or lose latent heat.

The interaction with the radiation field is a function of the size distribution and the refractive index of the particles and the spectral density of the radiation.

The collisional bet exchange with the ambient gas depends upon the temperature of the particles. Thus the equilibrium conditions for a given particle size distribution are a function of the attitude, time, of the day, season and planetary albedo.

Based on the equilibrium condition for a spherical aerosol particle of radius *r*, the energetic equilibrium of aerosols in the atmosphere is expressed by

$$P_{as} + P_{a\,pla} - P_e - P_c = 0 \quad (25)$$

where P_{as} is the power absorbed from solar radiation by the particle, $P_{a\,pla}$ the power absorbed from planetary radiation, P_e the thermal power radiated by the particle, and P_c the power lost by the particle due to collisions with the ambient gas.

Based on the model of Ivelve and Popova, it is known that in the particle size interval 0.1 to 0.3µm, daily average heating rates for summer are 0.15-0.1k/day and for winter these are 0.07-0.1 k/day in the stratosphere.

Mugnai et al. used the zeroth order logarithmic distribution function(Zold) suggested by spenscheid for the aerosol size distribution.

This is given by

$$dN/dr = C2 \exp[-\exp(\ln r - \ln r_m)^2 / 2(\ln \sigma)^2] \quad (27)$$

where $C2$ is a constant whose value depends on the aerosol concentration, r_m is the mode radius, and σ is related to the width and the skewness of the distribution.

From the values of P_e for each particle size, Mugnai obtained the heating (k/day) from the expression

$$dT/dt = 3x Pe/r3\rho_s C_p \quad (28)$$

where T is the air temperature, x the aerosol mass mixing ratio, ρ_s the particle density assumed (=2.3g/cm³ for volcanic ash particles and 1.6g/cm³ for sulphuric acid droplets), and C_p the specific heat at constant pressure.

Aerosol mass mixing ratio x is taken to be constant with respect to altitude with a value of 10-9g/g

of air.

The daily average heating rate for the polydisperse aerosol given by the Zold distribution is calculated from the daily average heating rate for each particle size in the range 0.01-10 μ m.

There are large variations of heating rate with height and aerosol composition. The heating rates are always positive indicating that aerosols add heat to the ambient gas by conduction throughout the stratospheric aerosol layer region. These heating rates of aerosol when compared to other heating rates in the same region.

However, during volcanic eruption when stratospheric aerosol loading increases enormously, the aerosol heating rates can become significant.

It is evident that during major volcanic eruptions, the aerosol heating rate becomes quite significant to effect the ambient thermal structure.

There was a large temperature increase at the two higher levels of 16.5km and 19.5km (stratosphere) associated with the Mt. Agung eruption in 1963. There is no significant effect noticeable at 9.5 km. However, an indication of a decrease is seen. An increase in stratospheric temperature is reported due to the El Chichon volcanic eruption in 1982.

Aerosols in the stratosphere reduce the solar radiation flux by absorption that is incident in the troposphere. Thus an increase in the aerosol loading in the stratosphere after major volcanic eruption could cause tropospheric cooling. The extent of tropospheric cooling is proportional to the solar energy lost by the troposphere.

In general, the effect of aerosols is to decrease the solar radiation reaching the earth's surface and increase the global albedo of the earth-atmosphere depending upon the absorption and scattering properties of aerosols.

4.2. Climatic Effects

The effect of aerosols on earth's climate has been investigated by a number of works. The investigations on this are based on analytical methods or numerical methods involving solution of radiation transfer equation. In the analytical models of climate change due to aerosols, the change in the earth-atmosphere system albedo from unperturbed (without aerosols) condition (A) to perturbed (by introduction of an aerosol layer in the atmosphere) condition (A') is obtained. If A'>A the aerosol layer will reduce the energy of the earth-atmosphere system (causing a cooling) and if A'<A it will increase the energy of the system (causing a heating). The aerosol layer is

characterized by the parameters a and b, where a is the fraction of incident radiation absorbed by the layer and b is the fraction of incident radiation backscattered by the layer. For an 'optically thin' aerosol layer, the critical value δ_c of δ ($=a/b$) is approximated by

$$\delta_c = (1-A)^2 / 2A \quad (29)$$

The models predict that an aerosol layer with $\delta > \delta_c$ will cause a decrease in system albedo A and a layer with $\delta < \delta_c$ will cause an increase.

Table 3 gives the value of δ_c for different surface types.

Table 3. Values of δ_c for Different Surface Types

Surface Type	A	δ_c
Urban areas	0.20	1.6
Desert	0.30	0.82
Farm lands	0.20	1.6
Forest	0.16	2.2
Oceans	0.08	5.3
Snow fields	0.70	0.064

The model predicts a decrease in surface temperature due to an aerosol layer (stratosphere). This means that the increased solar albedo effect dominates over the increase in trapping or 'greenhouse' effect due to a stratospheric aerosol layer.

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