Atmospheric Aerosols and Climate Change

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Introduction

Aerosols are particles found suspended in the atmosphere. The particles such as mineral dust, sea-salt, soot are directly injected into the atmosphere and are known as primary particles where as particles such as sulphates are produced from precursor gases released into the atmosphere and are known as secondary particles. Both natural and anthropogenic activities are responsible for the production of particles. In the recent years there is an increase in the concentration of aerosols in the atmosphere caused mainly by anthropogenic activities (IPCC 2001, Andreae and Crutzen, 1997). Aerosols play major roles in modifying the weather by altering the energy budget of the earth atmosphere system through scattering and absorbing the solar radiation (Charlson et al., 1991) as well as by altering the cloud parameters (Twomey 1977, Rosenfeld et al., 2000) such as cloud droplet size and cloud lifetime (Hansen et al., 1997, Ackermann et al., 2000). Indian subcontinent is a rich source of a variety of aerosol particles into the atmosphere. Fine crushed mineral (sand) particles from the arid and semi-arid regions, soot (carbon) particles from automobiles, industries, biomass burning and sulphates from fossil fuel burning are injected into the atmosphere in varying proportions and reside in the atmosphere from few hours to few days depending on the size of the particles. During winter months, prior to the onset of the summer monsoon, wind over India is predominantly from northeast, which transport these particles from the subcontinent to the tropical Indian Ocean region. Taking the average wind speed as 10 m/s we can show that the wind can carry particles to a distance of about 6000 km in 7 days. The sub-micron size particles coagulate to form bigger particles and the bigger particles are removed from the atmosphere by gravitational settling and rain-wash. Unlike over land surfaces, where the settled particles can be airlifted again, over ocean, the particles settled are lost and they form the ocean sediments. We are concerned with the interactions of these particles when they are airborne with the incoming solar radiation as well as with clouds.

Aerosol Sources and Sinks

The main components of aerosols are dust particles produced by winds, sea salt particles produced over ocean surface and a variety of sub-micron size particles produced in the atmosphere from gas to particle conversion mechanism. The amount of particles present over any location is region specific. For example, over urban region the secondary particles (particles produced from gaseous pollutants) are found in large quantities that are responsible for the fog we experience during winter mornings in cities like Delhi. Atmospheric aerosol content decreases as one goes away from a polluted urban region to remote continental regions. The particles found over rural regions are mainly materials from the nearby surface sources somewhat modified by the coagulation of particles of different origin and by condensation products resulting from gas phase reactions. Atmospheric gas phase reactions lead to formation of condensable products and subsequently to aerosols. The best known example of production of aerosols in the atmosphere is oxidation of SO$_2$ to H$_2$SO$_4$ and its neutralization by ammonia to form sulfate aerosol. Sulfur containing gases are released mainly from the biosphere and about 10 to 20% from volcanoes. In the troposphere, oxidation of SO$_2$ to sulfate can occur with radicals such as OH and HO$_2$. Sulfate particles are found to be concentrated mainly in the size range 0.1 to 1µm in the troposphere. In the stratosphere, however, photo-oxidation can be a prominent process where SO$_2$ is dissociated by UV radiation of wavelength less than 0.24 µm. Explosive volcanoes are the major source
of SO$_2$ in the stratosphere. Similar to sulfate particles, nitrogen-containing compounds are also found in the atmosphere. Nitrous oxide (N$_2$O) is produced mainly by microbiological processes. Though N$_2$O is stable in the troposphere, it decomposes chemically into nitrogen and nitric oxide (NO) in the stratosphere. Nitric oxide is oxidized to nitrogen dioxide (NO$_2$) by reacting with ozone. Gaseous HNO$_3$ is formed from NO$_2$ and OH. HNO$_3$/H$_2$O condense to form aerosol. Organic and carbonaceous particles are produced from condensable gaseous material released into the atmosphere from the biosphere and from biomass burning. Volatile hydrocarbon can exist simultaneously in gaseous and particulate phases. Fires mainly produce carbonaceous particles and wildfires are the major natural source of these particles. Anthropogenic activities such as fossil fuel and biomass burning also lead to the emission of carbonaceous particles into the atmosphere. The residence time of aerosols in the atmosphere can be estimated by means of radioisotopes that are attached to aerosol particles and removed with them due to rain wash or gravitational fallout. Using this method the earlier estimated residence time for aerosol particles was about one month on an average. With more detailed measurements we now know that one month is the upper limit for the residence time for particles in the upper troposphere and the same value becomes the lower limit for particles in the stratosphere. In the planetary boundary layer, the region that is in contact with the Earth’s surface, the residence time is typically a week. Aitken particles (0.001<r<0.1µm) are mainly removed by coagulation with other particles. The residence time for these particles can be estimated from the decline of their concentration to 1/e of the initial value, in the absence of other sources. The rate of coagulation depends on the mobility of the Aitken particles as well as the number density of the entire population. It can be shown that the lifetime of Aitken particles increases proportional to the square of the radius. The main removal mechanism for particles in the size range 0.01-10 µm is wet precipitation (rain-wash). For particles of size greater than 10 µm, sedimentation is the most important removal mechanism.

**Aerosol Size Distribution**

Aerosol size distribution may refer to number density, volume or mass that varies with particle size. For size classification, particles are assumed to be spherical, and their radius or diameter defines the size. Aerosol number density distribution describes the number of particles in each given radii interval. Since particle size extends over several orders of magnitudes, it is convenient to describe the size (radius) in logarithmic scale. This leads to the log-radius distribution such that there are n(log r) Δlog r particles with log radii between log r and log r + Δlog r. The commonly used size distribution is log normal distribution. A normal (Gaussian) distribution is symmetric about its mean. If the argument in the normal distribution is taken as log r where r is the radius of the particles, we get the lognormal distribution. The log normal distribution function lays emphasis on the individual components in a mixture of particles originating from different sources. Each component has its specific mode radius, standard deviation and refractive index value. For many purposes such as deposition calculations, radiative transfer model calculation, up to three log normal size distributions have been used to describe atmospheric aerosol size distribution. Fig. 1 shows the typical aerosol number distribution fitted with three individual lognormal curves for the nucleation, accumulation and coarse modes. The sum of the three curves describes the size distribution of the total ensemble.
Fig. 1: Typical aerosol size distribution for a polluted region. The measured points (diamonds with vertical bar showing the standard deviation of the mean) could be fitted using a sum of three individual log-normal curves

Optical Properties of Aerosol

The important parameter that determine the efficiency of scattering or absorption of light by a spherical particle is the size parameter ($\alpha$), which is the ratio of the circumference of the particle to the incident wavelength ($\alpha = \pi D_p/\lambda$). Based on the $\alpha$ value the scattering process can be divided into three domains viz., Rayleigh scattering, when particle size is very small compared to the incident light wavelength; Mie scattering, when particle size is comparable to the wavelength and Geometric scattering, when particle size is much larger than the wavelength. The other important parameter is the complex refractive index of the particle, $n + ik$. The real part $n$ represents scattering and the imaginary part $k$ represents absorption and both are functions of $\lambda$. The elemental carbon (soot) is the most significant absorbing component in atmospheric aerosols and has a high value for the imaginary part of the refractive index. 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 aerosols 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 (Jayaraman and Koepke, 1992).

For aerosol radiative forcing studies we are concerned with the total column concentration of aerosols, distributed from surface to top of the atmosphere. This can be inferred by measuring the attenuation of the incoming solar radiation at selected wavelength bands in and around the visible region of the electromagnetic spectrum. The aerosol optical depth, $\delta$, at a wavelength $\lambda$ is defined such that, the surface reaching direct solar radiation intensity decreases by $\exp(-\delta/\cos \theta)$ due to aerosol scattering and absorption, where $\theta$ is the solar zenith angle measured from the vertical. Fig. 2 shows average aerosol optical depth values for 500 nm wavelength measured at different regions during January and February. The values are high over the peninsular India and over the Bay of Bengal region. The value is comparatively less over the Arabian Sea and decreases still further as we go over the tropical Indian Ocean down to Antarctic. The large latitude variation shows the significance of man-
made pollutants over the main land which get diluted as they are transported to long distances over the ocean surface.

![Aerosol Optical Depth](image)

**Fig. 2:** Aerosol optical depth measured at different regions (BOB: Bay of Bengal; TIO: Tropical Indian Ocean). The measurements are made using sun-photometer at wavelength 500 nm. Average values for the period January - February are shown. The vertical bars represent plus and minus one standard deviation of the mean values.

### Vertical Profiles of Aerosols

The vertical distribution of aerosols is determined by the amount of particles present in the boundary layer, in the free troposphere and in the stratosphere. Particles present in the boundary layer, close to the earth’s surface is highly region specific, whereas in the free troposphere particles could be transported to distances of few thousand kilometers (Krishnamurti, 1998). In the stratosphere submicron size particles reside for two to three years and during this time they are more or less uniformly mixed all around the globe. Study of the vertical distribution of aerosols was first initiated at PRL using rocket-borne photometers (Subbaraya and Jayaraman, 1982, Jayaraman and Subbaraya, 1988). The rocket experiments were done in early eighties from the Thumba Equatorial Rocket Launching Station (TERLS) situated near Thiruvananthapuram. Later a balloon-borne sun-tracking photometer was developed and flown on high altitude balloons from TIFR Balloon Facility situated at Hyderabad to obtain the vertical distribution of aerosol concentration and size distribution up to stratospheric altitudes (Jayaraman et al., 1987). Detailed studies are made on the volcanic aerosol layer formed after the Pinatubo volcanic eruption in 1991 (Ramachandran et al., 1994 a,b). The effects of volcanic aerosols are many. Mainly they perturb the chemical, optical and thermal structure of the atmosphere, which produce significant meteorological effects that affect global climate.

In order to have a regular monitoring of volcanic aerosol layer in the stratosphere a Laser Radar or LIDAR (Light Detection And Ranging) was developed at PRL in early nineties (Jayaraman et al., 1995 a,b). The lidar system uses Nd:YAG laser operated at 532 nm (second harmonic) and measures backscattered light from the atmosphere up to mesospheric altitudes. **Fig. 3** shows the total (air molecules + aerosols) backscattering coefficient derived from the measured lidar return signal during different years. A large increase in scattering coefficient above the tropopause (17 km) is seen extending up to about 28 km in the April 1992 measurement which is about 10 months after the Pinatubo eruption. Both the layer thickness and the altitude of maximum aerosol concentration are found to decrease with time. Depending on the material put into the stratosphere it takes few years for the particles to settle
down completely. Using lidar data as well as model computations (Ramachandran et al., 1997) it is shown that though the injected amount is more in case of Pinatubo it took about three years for the stratosphere to revert to background condition.

Fig. 3: Vertical profiles of backscattering coefficient measured by PRL lidar showing the stratospheric aerosol layer formed after the Mount Pinatubo eruption in June 1991. For comparison typical data for the year 1999 is shown which is volcanically a quiescent period and the profile represent the background stratospheric condition.

Aerosol Radiative Forcing

The instantaneous radiative forcing is defined as the ratio of the change in the surface reaching solar radiation intensity, $\Delta F_{\downarrow}$ in W/m$^2$ or the upscattered solar radiation flux $\Delta F_{\uparrow}$, leaving the atmosphere to the change in column aerosol amount, usually the columnar aerosol optical depth, $\Delta \delta$ (Jayaraman et al., 1998). The forcing efficiencies at the surface ($\Delta F_{\downarrow}/\Delta \delta$) and at the top of the atmosphere ($\Delta F_{\uparrow}/\Delta \delta$) are estimated separately and the difference between the two terms is defined as the net aerosol absorption efficiency within the atmosphere. The net forcing is obtained by multiplying the forcing efficiency and the average aerosol optical depth over the study area. This estimation helps in quantifying the role of aerosols in altering the radiation budget at the surface and top of the atmosphere (TOA) separately and the radiation loss due to absorption in the atmosphere. Simultaneously measured surface reaching solar radiation intensity and the columnar aerosol optical depth during the recently conducted Indian Ocean Experiment (Ramanathan et al., 2001) are used to estimate the aerosol radiative forcing for different regions. The magnitude of the aerosol
radiative forcing at any location and time depends on the amount of aerosols present, their optical properties and the underlying surface albedo.

In Fig. 4 the diurnally averaged aerosol radiative forcing values are presented individually for the surface, at the top of the atmosphere for the Arabian Sea, coastal India (west coast) and the Bay of Bengal regions. In the absence of \textit{in situ} ship cruise measurements over the Bay of Bengal region, the NOAA satellite data are used to obtain the aerosol optical depth and the radiative forcing (Coakley et al 2002). Radiative forcing detected over the ocean regions surrounding the Indian sub-continent is about an order of magnitude high compared to the pristine ocean region south of the ITCZ. This is of great concern and its effect on the heat budget of the atmosphere and the impact on dynamics need to be studied in greater detail and depth.

![Fig. 4: The aerosol radiative forcing in W/m². The radiative forcing is defined as the change in radiation flux because of the presence of aerosols. TOA stands for the change in flux at the top of the atmosphere, Absorption is the amount of radiation absorbed in the atmosphere due to aerosols, and Surface stands for the net reduction in the surface reaching solar flux due to the presence of aerosols.](image)

**Aerosols and Climate Change**

The importance of aerosol radiative forcing in the climate system is well recognized as an important factor in interpreting both the past climate variations as well as in predicting the potential future climate change. We have fairly good theoretical knowledge on how aerosols affect directly the earth’s radiation budget and hence the forcing of climate. Also, experimentally the eruption of Mount Pinatubo in June 1991 gave us an opportunity to observe the impact of the volcanic aerosols on the atmospheric temperature structure. Compared to the volcanic aerosols in the stratosphere, the tropospheric aerosols are far more complex in terms of their chemical composition, particle size and shape, and temporal and spatial variations (Jayaraman, 2001). The radiative forcing due to tropospheric aerosols could be computed only if we know the optical parameters of the individual chemical species and the knowledge on the proportion of aerosols that are internally and externally mixed. Considerable gap exists in our knowledge on these parameters. It is important to have
‘closure’ experiments to bridge the gap in our knowledge between observations and theoretical predictions. The combined model and observation approach is very useful for evaluating the uncertainties and limitations of both the models and observations. A preliminary assessment of the observed aerosol forcing over the INDOEX region shows (Ramanathan et al., 2001) a low level air temperature increase by about 0.5 to 1.5 K over the region where the aerosol forcing was imposed. In addition the aerosols also found to perturb the inter-hemispheric heating gradients. The gradient that is one of the driving forces of the thermohaline circulation is found to increase by as about 50%. Another important impact could be on the ecosystem as about 70% of the forcing is concentrated in the photosynthetic part of the solar spectrum.

While the aerosol transport in the horizontal direction is controlled by winds (Krishnamurti et al., 1998) and is a mesoscale process, the vertical transport and scavenging is determined by the convective motions and takes place in convective scales. The aerosol particles caught in the convective updrafts serve as cloud condensation nuclei (CCN) and help in the cloud droplet formation. The efficiency of an aerosol particle to serve as a CCN depends on its size and the amount of water adsorbing material in it. The cloud development is different for different air masses such as over ocean or continent. The precipitation rate, vertical distribution of latent heat and other cloud related phenomenon in clear and polluted environment could significantly be different. An increase in the aerosol amount can suppress precipitation at least from low clouds. The rain formation within the clouds depends on the cloud drop size distribution. For a given liquid water content, precipitation will be accelerated in the case of less number of larger drops than with a larger number of smaller drops. The presence of soot, due to its absorption of solar radiation can also burn off the low clouds. A reduced rainfall leads to a reduced rain scavenging of aerosols that induces a positive feedback, with a possibility of an increase in the haze amount and extend.

Concluding Remarks

It has been undoubtedly shown that aerosols play a major role in modifying the earth’s radiation budget over the polluted region. During clear sky (cloud free) conditions the aerosol direct forcing at the surface is shown to be about three times higher than the top of the atmosphere forcing and the difference is the forcing caused within the atmosphere by absorbing aerosols such as soot particles. More work needs to be done to establish aerosol radiative forcing in the presence of clouds (indirect radiative forcing). Large inter annual variability can exists in the aerosol loading over the ocean surfaces surrounding the peninsular India, caused mainly due to varying meteorological conditions. The consequences of the increased aerosol amount and the large solar radiation heating are many. For example the increased aerosol amount decreases the surface reaching solar flux by as much as 15%. This would result in a decreased evaporation, which can result in a decreased rainfall. Since most of the reduction in the surface reaching solar radiation is concentrated in the photo synthetically active visible and UV part of the solar spectrum, the aerosols could have an effect on the marine and terrestrial biological productivity. While the increased aerosol concentration can suppress the precipitation from low clouds the very large atmospheric heating by aerosol absorption obtained near the Indian coast can also burn off the low clouds.

In the Indian context there is an urgent need to characterize both the naturally occurring and anthropogenically produced aerosols in different geographical regions. There is also an urgent need to do size resolved chemical analysis of aerosols to identify the relative importance of different sources such as fossil fuel, biomass burning etc. Role of large-scale atmospheric heating by aerosol absorption needs to be examined through coordinated modeling efforts. Observations on land surface processes (soil moisture, surface winds etc.) need to be looked into for studying regional impacts of the atmospheric heating induced by aerosol absorption. The estimated population increase in India and China will make these
regions more susceptible to the associated effects of pollution. A national/international level coordinated study and establishment of observatories for baseline monitoring of aerosol, radiation and atmospheric trace gases should get the highest priority.

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