Chapter 5

Summary and Scope for future work

This chapter holds in it the summary of the thesis with emphasis on the important results obtained. Suggestions for where work in the future direction should lead to, as an extension of the present work are also enlisted.

5.1 Summary of results obtained

5.1.1 Tropospheric aerosols

An automatic Sun-tracking photometer was developed and used to measure the columnar aerosol optical depths over Ahmedabad. Aerosol optical depths are found to exhibit diurnal, day to day and seasonal variations. Also, an increase in the values from January to May are found, indicating that the increase in surface temperature and winds induce more soil derived particles into the atmosphere. Results show that there are no appreciable changes in the aerosol optical depths which could be attributed to the Gulf oil fires indicating that the natural sources of aerosols over Ahmedabad, mainly wind blown dust dominated over the oil fire effluents. Other results [*Browning et al.,* 1991; *Bakan et al.,* 1991] using model calculations predicted that most of the smoke from the oil fires in Kuwait would remain in the lowest few kilometres of the troposphere. Airborne observations by *Johnson et al.* [1991] also confirmed the above findings. *Johnson et al.* [1991] concluded that although the effects may be significant on a regional scale, those on global scale including the Asian summer monsoon are likely to be insignificant. Also, according to *Bakan et al.* [1991] there was no indication of a weakening of the Indian summer mon-

soon. However, the spectral dependences show that the aerosol size distribution changed from bimodal in 1991 to monomodal in 1992, indicating that there are two sources (smoke particles from oil fires and local wind derived mineral particles) in 1991 when compared to one dominating source (wind blown dust) in 1992. Though the 1992 data are influenced by the Mt. Pinatubo volcanic eruption in June 1991, the aged volcanic aerosols are found to exhibit a monomodal size distribution with mode radius around 0.6 μm [*Asano et al.*, 1993], which could have been superimposed on the derived total columnar aerosol size distribution, with mode radius around 0.5 μm in the present work. The ozone column density values from Sun photometry measurements are consistently lower than the Dobson spectrophotometer values, by about 40-50 Dobson units. This observed difference could be due to the effects of high aerosol optical depths and their diurnal variations found over Ahmedabad. Hence, reliable estimation of ozone column amounts from UV extinction measurements is possible only in locations of low turbidity, unlike Ahmedabad. The increase in aerosol optical depth with decreasing relative humidity and increasing temperature, confirms that the increase in surface temperature reduces the soil moisture content, thus inducing more soil derived particles into the atmosphere. Secondly, since aerosols over Ahmedabad are mainly mineral, local wind derived particles which are non-hygroscopic, the aerosol optical depth is not directly related to relative humidity, unlike at a coastal station where the aerosol optical depth increases with an increase in relative humidity, because of the hygroscopic nature of maritime particles.

5.1.2 Stratospheric aerosols

a) Balloon-borne optical studies of Pinatubo aerosol layer over Hyderabad

An analysis of the direct and scattered radiation intensity measurements made over Hyderabad using Sun-tracking and Sun-scanning photometer systems onboard the balloons during October 1991 and April 1992 indicates a dense Pinatubo volcanic aerosol layer between 16 and 30 km. The aerosol extinction coefficients and number density values over Hyderabad are found to be the highest ever obtained during a decade of stratospheric aerosol measurements. The aerosol extinction coefficients obtained in 1991 and 1992 at all wavelengths are about 2 orders higher in magnitude at the peak altitude when compared with that of 1985 (background, volcanically quiescent) values over Hyderabad.

The recent results showing the highest values ever obtained over the same site indicate the magnitude of Pinatubo eruption which is the strongest eruption in this century. The aerosol size distribution parameter derived from the scattered radiation intensities also shows layered structures. About 4 months after the eruption, in October 1991, the region between 17 and 23 km, is found to have ν values around 1.8, indicating the presence of larger aerosol particles. The size parameter obtained during April 1992, about 10 months after the eruption, indicates formation of larger aerosols at higher altitudes by coagulation with a subsequent reduction in the aerosol number density. The aerosol layer peak was found to occur at 23 km in October 1991 with about 40 particles per cm^3. In April 1992 the aerosol number densities have shown a decline, with a peak aerosol number density of about 20 particles per cm^3 at an altitude of about 20 km. Also, the October 1991 measurement revealed the existence of larger aerosol particles in the 17 and 26 km altitude range, with a mode radius of 0.2-0.3 μm, formed due to coagulation of sulphate aerosols. The derived asymmetry factor g also revealed the existence of larger particles which are qualitatively in agreement with the results on the size distribution parameter ν, the slope of Junge power law. The computed mass of the Pinatubo layer in October 1991, after 4 months of the eruption, is about 0.053 gm^{-2}, which is 3.75 times higher than the earlier reported value for El Chichon layer, at the same time of evolution. The obtained aerosol extinction profiles are found to agree well with that of an independent lidar measurement made over Ahmedabad and with SAGE II results.

b) Lidar studies of Pinatubo aerosol layer over Ahmedabad

A state-of-the-art Nd:YAG lidar commissioned in Ahmedabad (23°N) during April 1992 operating at 532 nm was employed to study the Pinatubo volcanic aerosol layer and its decay. 36 vertical profiles of scattering ratio obtained during April 1992 to May 1994, a period corresponding to 10 months after the Pinatubo eruption to about 3 years, every fortnight, have been used for the study. The volcanic aerosol layer is seen very prominently as an increase in the scattering ratios from about 17 km (1 km above the local tropopause) to about 30 km. Also, the scattering ratios which were about 10 during April-May 1992 have become about 1.5 in May 1994. The results obtained on the integrated mass densities and aerosol backscatter from 17 to 30 km give a 1/e folding time of 9 months, assuming

a typical mode radius of aerosols to be in the range of 0.1 to 0.3 μm. Calculations show that if the layer decays at the same rate, then it may take about 4.5 years to attain the background aerosol backscattering coefficient for the tropical region. However, the peak scattering ratio value shows a longer decay time of 12.5 months as has been reported earlier in the case of Fuego and El Chichon eruptions. As the removal mechanisms of aerosol are faster below the peak, the integrated aerosol mass for the whole layer shows a faster decay than the decay time shown by the peak value alone. The average aerosol size distribution for the layer has not undergone any considerable change during the decay phase from 1 year after the eruption to about 3 years. After about 2 years, the integrated mass and the backscattering coefficient in the case of Mt. Pinatubo are found to be in the same range as that of the El Chichon reported by *Jäger and Hofmann* [1991], as also shown by the model studies of *Pinto et al.* [1989] that the residual aerosol masses at stratospheric altitudes become comparable after about 2 years for eruptions of wide range of magnitudes.

c) Modeling studies of Pinatubo aerosol layer

Earlier results using balloon-borne Sun-tracking photometers for the El Chichon layer over Hyderabad, showed that the layer produced after the eruption decayed in about 3 years over the tropics [*Jayaraman,* 1991]. Since Mt. Pinatubo had injected as much as 2 to 3 times the material compared to El Chichon, it was not clear whether the lifetime of these aerosols will also be longer because of the larger number density of aerosols or, perhaps, the physical processes such as growth, coagulation and sedimentation will take place at a much faster rate to give more or less the same decay time and whether the residual aerosol masses would be similar, as shown through model calculations [*Pinto et al.,* 1989].

To understand better the relative roles of these processes in influencing the formation and decay of the stratospheric aerosol layer after major volcanic eruptions, a simple model was developed to study the time evolution of a volcanic aerosol layer (17-30 km) and aerosol size distribution using the microphysical processes of growth, coagulation and sedimentation. The results show that the Pinatubo aerosol layer has decayed to the background aerosol level in about 3 years, over the tropics. The experimental lidar data

obtained continuously on the Pinatubo layer also gives this result. The latitudinal dependence of the aerosol layer is important only in the first few months after the eruption, as later on, low latitudes become the source of aerosols to higher latitudes. The data obtained in the case of El Chichon shows that the layer decayed in about 3 years. The latitudinal dependence in the case of El Chichon also is clearly seen only in the first few months after the eruption.

d) Conclusions

In conclusion, the balloon-borne, lidar and modeling studies have revealed the processes that are responsible for the formation of volcanic aerosol layer and its decay. Immediately after the eruption, within a day, homogeneous nucleation takes place giving rise to high concentrations of aerosol droplets. After nucleation the sulphuric acid-water solution droplets can grow by heterogeneous condensation of H_2SO_4 and H_2O vapours, the condensation occurring first on large aerosols and then progressively on smaller aerosols. Condensation on the preexisting sulphate aerosols would increase the extinction and decrease the extinction ratio as the extinction becomes more and more dominated by large aerosols. If the amount of SO_2 injected is large, as in the case of Pinatubo, conversion of H_2SO_4 produces higher number of new small H_2SO_4/H_2O aerosols and coagulation proceeds at a faster rate. Thus the condensation of H_2SO_4 on the already existing H_2SO_4/H_2O droplets and subsequent coagulation is one of the principal mechanisms for the formation of larger aerosol particles. Once the aerosol grows to a few microns they are removed from the stratosphere by gravitational sedimentation. Sensitivity studies conducted using the model developed show that the processes of growth and coagulation are important, in the first year or so after the eruption in maintaining the aerosol layer, whereafter when the particles grow sufficiently larger, of the order of a few microns, sedimentation becomes important. The lidar obtained aerosol mass decay results show that the aerosol size distribution has not undergone any noticeable change throughout the decay phase from 1 year to 3 years after the eruption. It is also seen that aerosol mass decayed at various rates at different altitude regimes, indicating that dynamical removal processes such as gravitational settling and horizontal dispersal act differently depending on the particle size. The faster decay time for the mass of the layer in the 25-30 km region (Figure 4.28)

further corroborates this fact. Also it is observed that the integrated backscattering coefficient (IBC) of the volcanic aerosol layer decays at a faster rate than the peak scattering ratio. The faster decay rate of IBC shows that due to sedimentation the particles at low altitudes below the aerosol peak are more effectively removed from the layer while the major contribution to IBC comes from these altitudes. Though the amount of SO_2 injected by Pinatubo is more than 2 times that of El Chichon, the model study showed that the aerosol microphysical processes of growth and coagulation, have taken place faster giving rise to larger particles and hence the removal rates of these large aerosols have been faster, giving the same decay times of 3 years to both the volcanic aerosol layers.

5.2 Scope for future work

5.2.1 Tropospheric aerosols

Tropospheric aerosols are much more variable on time and space scales because of the great diversity and wide distribution of sources and their short residence times, in many ways it is more difficult to study tropospheric aerosols than stratospheric aerosols and hence the characterisation of tropospheric aerosols continues to pose a tougher challenge. Though certain characteristics have emerged recently the global trend on tropospheric aerosols remains highly uncertain. The studies on the tropospheric aerosol started with rejuvenated zeal after the recent work of *Charlson et al.* [1992] who suggested that the anthropogenic sulphate aerosol may be forcing climate in opposition to greenhouse warming with a comparable magnitude. Long term tropospheric aerosol measurements are limited to a few stations, to suggest any systematic trend on the climatic effects of aerosols. So in order to fully understand the climatic effects of aerosols, both due to natural and anthropogenic sources and to make model assessments it is very essential to have long term data on aerosol radiative properties, for example, optical depth and a better understanding of the physical and chemical processes going on at tropospheric levels. The emphasis should be given for the determination of physical and chemical properties of aerosols, by conducting coordinated measurements. As Sun photometers are versatile and easy to use devices to determine the aerosol optical depths, these measurements can be combined with collection of aerosol particles near simultaneously and can be used to determine

the mass and the chemical composition of particles for different seasons to study the seasonal dependence of aerosols and the variation of aerosol mass with respect to seasons. These measurements when combined with multiwavelength lidar vertical profiles of aerosol extinction can give the size distribution of aerosols and the dominance of the aerosol particles of different sizes at different altitudes. All these information when combined can give an excellent data set of information on the physico-chemical processes that are responsible for the formation of aerosols and their settling times.

5.2.2 Stratospheric aerosols

Studies made after major volcanic eruptions such as Mt. St. Helens, El Chichon and Mt. Pinatubo have increased our scientific awareness on various processes which are responsible for the formation of aerosol layer at stratospheric altitudes, evolution and its decay and the radiative-chemical- dynamical influences, combined with technological advancements, in terms of techniques available to determine aerosol characteristics and fast computational facilities. This gives a confidence to say that now the volcanic aerosol layer formation and decay seems to be well understood. However, certain outstanding scientific issues regarding the properties of stratospheric aerosols and their roles in global change problems can be pointed out. First and foremost is regarding the background sulphate aerosol mass increase due to anthropogenic emissions, particularly due to high altitude commercial aircrafts [*Hofmann,* 1990]. Regarding volcanic aerosols, the precise composition of them is not known for evaluating heterogeneous chemistry in the stratosphere and do they act as nuclei for the formation of cirrus clouds in the upper troposphere, is also not clear.

Finally as has been commented upon earlier, large gaps exist in the knowledge of anthropogenic aerosols due to lack of data, which prevents quantification of their influence for use in climate models. Need of the hour therefore, is a study of coupling of the physical-chemical processes that produce them and the meteorological processes that distribute and remove them with the physical and optical characteristics that determine the radiative transfer and cloud microphysical effects.

- Items which are important but not studied in this thesis include:

  1. Heterophase chemical reactions involving aerosols, which are important in the ozone depletion problem and the possibility of such reactions occurring in the tropical stratosphere.

  2. The perturbations in the temperature profile which is very important to draw a conclusion on the radiative effects of aerosols. An aerosol-induced radiative heating can cause changes in the local energy balance and in atmospheric dynamics besides causing changes in the atmospheric temperature structure [*Ramaswamy*, 1988a]. Recent model studies of *Ramaswamy and Bowen* [1994] show that the lower stratospheric thermal profile is sensitive to the tropospheric aerosols and the rate of change of temperature is found to vary as a function of altitude and optical depth. The model calculations demonstrated that changes in the concentrations of radiatively active species including aerosols can contribute to a cooling of the lower stratospheric region. Also it is clear that in addition to being a significant climate change issue in its own right, the cooling of the lower stratosphere due to changes in radiatively active species has important climatic implications involving the stratospheric hydrological cycle [*Toon et al.*, 1986; *Ramaswamy*, 1988b] and the chemical and dynamical processes leading to ozone depletion [*Austin et al.*, 1992; *Mahlman et al.*, 1993].

  3. Role of cirrus clouds in altering aerosol properties, which in turn determine cooling or warming of the Earth's surface depending on optical depth.