SOME STUDIES ON MINOR NEUTRAL CONSTITUENTS OF THE MIDDLE ATMOSPHERE OF LOW LATITUDE

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AND

PHYSICAL RESEARCH LABORATORY

CERTIFICATE

I hereby declare that the work presented in this thesis is original and has not formed the basis for the award of any degree or diploma by any University or Institution.

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Dedicated to my parents

Contents

Preface

This thesis is a presentation of work carried out at Physical Research Laboratory, Ahmedabad, India, on minor neutral constituents of the middle atmosphere. Both experimental and theoretical studies have been made. Experimental studies have been conducted by ground-based technique at Ahmedabad (23°N, 73°E), India and Gurushikhar (24°N, 73°E), India. The experimental work has three parts viz: (a) NO_2 study, (b) NO_3 study and (c) Volcanic aerosol study and theoretical study consists of the explanation of the first two parts of the experimental work. These are described in different Chapters whose outline is given below :

The Chapter 1 is the introduction to this thesis. The scope of the thesis has also been given in this chapter.

Chapter 2 deals with the technique used, instrumental setup, and observational procedure.

Chapter 3 describes the results of NO_2 measurements over Ahmedabad and the effect of Mt. Pinatubo volcanic aerosols on NO_2 variability.

Another important species of NO_X group is NO_3. Results obtained from NO_3 measurements are presented in Chapter 4. Variability of NO_3 during nighttime and its disappearance during dawn have been studied over low latitude northern hemisphere.

The observational results of Chapter 3 and Chapter 4 are explained by theory in Chapter 5. This theoretical study has been made using one - dimensional model. The details of the scheme and comparison between model results and observational values are described in the same Chapter.

Chapter 6 deals with the study of volcanic aerosols produced in the stratosphere after Mt. Pinatubo erupted in June 1991. For this purpose zenith sky intensity

during twilight period has been measured using ground-based photometric technique. The detailed description of the technique and the results thus obtained are given in the same Chapter.

The concluding remarks and suggestions for the further study are given in Chapter 7.

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I would like to thank Dr. Susan Solomon, NOAA Aeronomy Laboratory, Boulder, Colorado, for discussions on NO_2 and NO_3 results; Prof. W. B. Grant, NASA Langley Research Center, Hampton, Viriginia for providing recent SAGE data and valuable discussion on the effect of Pinatubo aerosols on different minor constituents; Dr. R. L. McKenzie, NIWAR, Lauder, New Zealand, for discussion on NO_2; Prof. S. Chandra, GSFC, NASA, for discussions on ozone variability due to volcanic eruption; Prof. Y. Kondo, STELab, Nagoya Univ., Nagoya, Japan, for discussions on NO_2 results; Prof. G. Brasseur, NCAR, USA, for discussions on NO_2 and NO_3 chemistry. I record my gratitude to them.

I express my thanks to the personnel of the PRL workshop and in particular to Mr. P. S. Panchal who has made lot of efforts in designing and fabricating different accessories of the monochromator system. I am grateful to the staff of the computer center and the Library for their cooperation and help. I thank Mr. D. R. Ranpura of the photographic and documentation section for his meticulous work and timely delivery of photographic prints.I am also thankful to the draftsman Mr. S. C. Bhavsar for making figures and diagrams.

NO_3 measurements have also been taken from hill station, Gurushikhar. I am indebted to all the Gurushikhar staff for the success of the nighttime observations and in particular to Mr. R. Shah and Mr. Kothari. I express my gratitude to them.

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List of Publications

(1) A. Jayaraman, S. Lal, **M. Lal**, B.H. Subbaraya, S.C. Garg, T. John, K.S. Zalpuri, N. Seshadri, C.R. Sreedharan, R. Vijaykumar, V.M. Ignatov, G.A.Kokin, S.P. Perov, O.V. Shtrikov, S.V. Tishin, and A.F. Chizov.
The Indo-Soviet collaborative experiment at Thumba to study diurnal variations in the ozone vertical distributions over the tropics.
in Ozone in the Atmosphere Eds. R.D. Bojkov and P. Fabian, A. Deepak Publishing, 1989, pp. 113 - 116.

(2) **M. Lal**, D.K. Chakrabarty, J.S. Sidhu, S.R. Das, and S.D. Verma.
Some results of ground-based measurements of atmospheric NO_2 at Ahmedabad by visible absorption spectroscopy.
Indian J. Radio Space Phys., 22, 108 - 113, 1993.

(3) D.K. Chakrabarty, **M. Lal**, G. Beig, J.S. Sidhu, and S.R. Das.
Balloon measurements of ionic conductivities of the middle atmosphere in India during MAP.
In press of **J. Atmos. Terr. Phys.** (UK).

(4) **M. Lal**, D.K. Chakrabarty, J.S. Sidhu, and S.R. Das.
Atmospheric NO_3 observations over low latitude northern hemisphere during night.
In press of **J. Geophys. Res.** (USA).

(5) **M. Lal**, D.K. Chakrabarty, J.S. Sidhu, and S.R. Das.

Near simultaneous measurements of NO_2 and NO_3 over tropics by ground-based absorption spectroscopy.

In press of proceeding of Quad. ozone symposium, Charlottesville USA, 1992.

(6) **M. Lal**, and D.K. Chakrabarty.

Derivation of water vapour absorption cross - sections in the red region.

In press of proceeding of Quad. ozone symposium, Charlottesville USA, 1992.

Chapter 1

Introduction

1.1 General Introduction

Man has always been interested in the atmosphere around him - in its changing weather patterns, glorious twilight phenomena, rainbows, aurora etc. The daily variation between day and night is one of the processes of nature which exerts a striking impact on the structure of organism and the way of life of every inhabitant of our planet. This atmosphere is also a vital natural resource which man has taken for granted throughout the history. To fulfill his own requirements, he has been dumping millions of tons of chemical materials into this atmosphere. Nature has its own method to remove these pollutants from the atmosphere. Often these chemicals offer resistance to nature's removal mechanism. Nature makes them interact with air and sunlight and transform them to substances which are susceptible to nature's removal mechanism. Often these are transported to suitable regions from where they are finally removed by rain. There is now evidence that these natural capabilities of the earth system are being overtaxed. The air, the ocean and the land of our earth are no longer able to keep pace with our discharges. As a result, worldwide, the amount of numerous key chemicals are increasing above their natural background amount in

this atmosphere. CO_2, CH_4, N_2O, lower atmosphere ozone, CFCs are some among them. Convincing data from the gases in polar ice cores tell us that the present CO_2, CH_4 and N_2O concentrations are unprecedented, at least for the earth of the last 160,000 years. It is time that we make correct assessment of this trend, predict their impact on global change and take corrective measures before a permanent damage is done to our environment. It is in this greater perspective, that this thesis presents some work on the minor constituents of the middle atmosphere at low latitude.

The earth's atmosphere has been divided into different layers viz. troposphere, stratosphere, mesosphere, and thermosphere. The region approximately below 15 km is called troposphere, between 15 and 50 km is called stratosphere, between 50 and 95 km is called mesosphere and above mesosphere is thermosphere. Fig. 1.1 shows these regions along with their temperature nomenclature. The region between 10 and 100 km is called the middle atmosphere. In Fig. 1.2 temperature distribution below 60 km has been shown in a magnified way for tropical and polar region. Note the low tropopause temperature and high tropopause level in the equatorial region compared to mid and high latitude region. Our studies are confined to the region below 50 km.

The major constituents of the earth's atmosphere below about 100 km are N_2 (78%) and O_2 (21%), rest are minor constituents such as CO_2, CH_4, N_2O, H_2O, NO_2, O_3, NO_3 etc. and aerosol particles of size varying from 0.2 μ to 20 μ. But they control the climate and the way of life on earth's surface. Fig. 1.3 illustrates the height variation in number density for various chemical compounds in the middle atmosphere [*Ackerman*, 1979]. Although these were compiled from observational data obtained at different times and places by different methods, it shows the general trend in the height variation for each constituent. The curves 10^{-1}, 10^{-2} etc. represent the variations in number density for the case of a constant mixing ratio. Thus these minor constituents are at ppmbv and ppbbv levels.

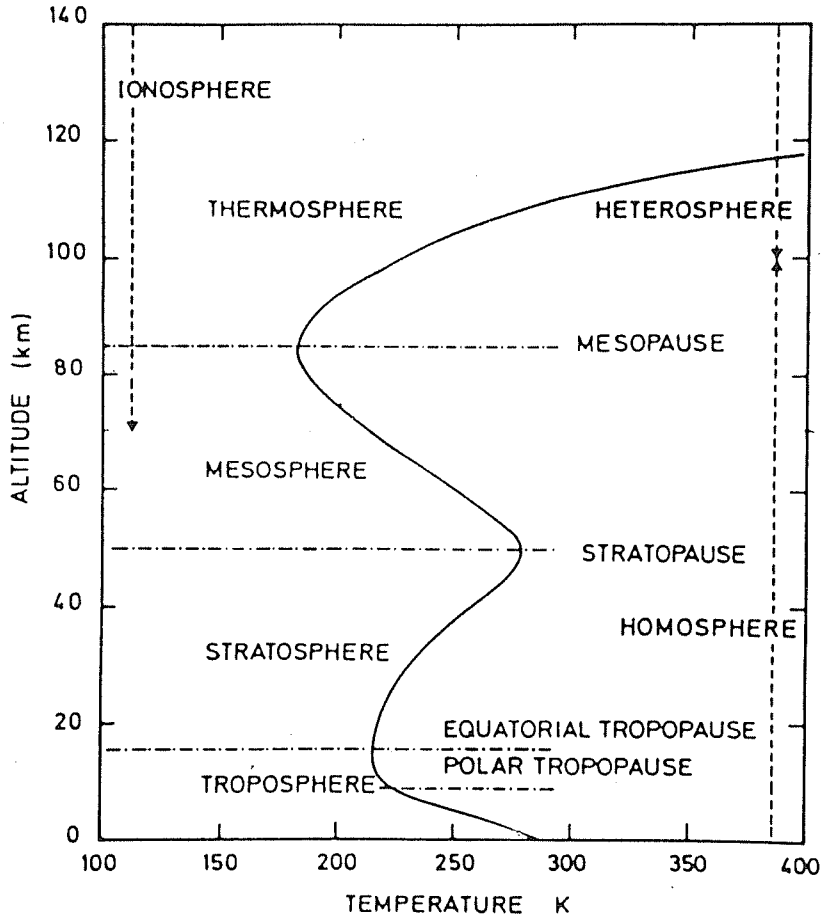

Figure 1.1 Thermal structure of atmospheric layers (Brasseur and Solomon, 1986).

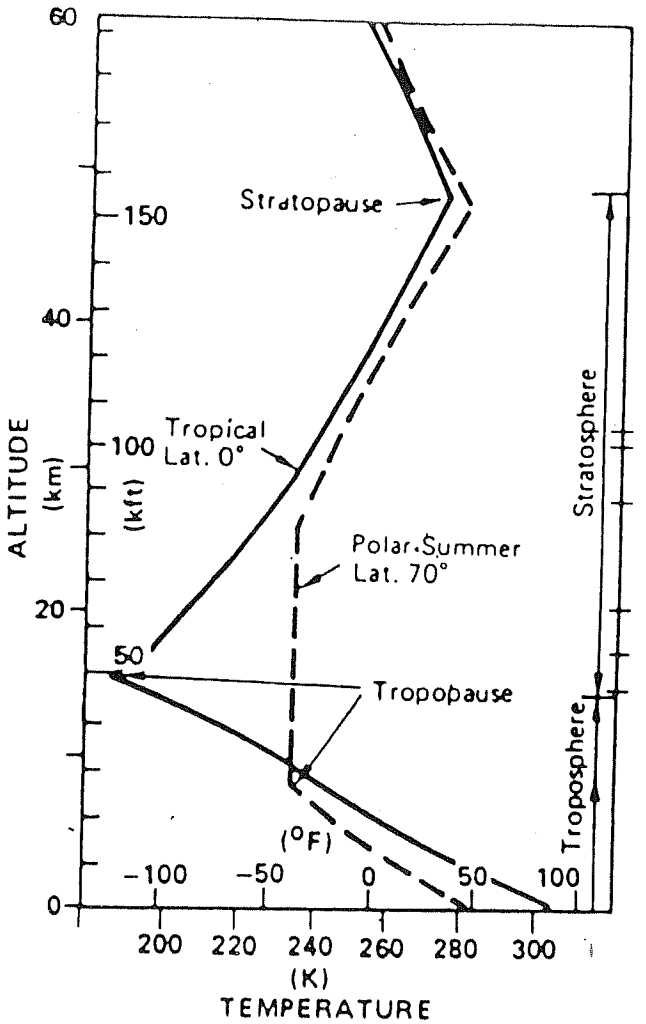

Figure 1.2 The temperature of the atmosphere below 60 km as a function of altitude.

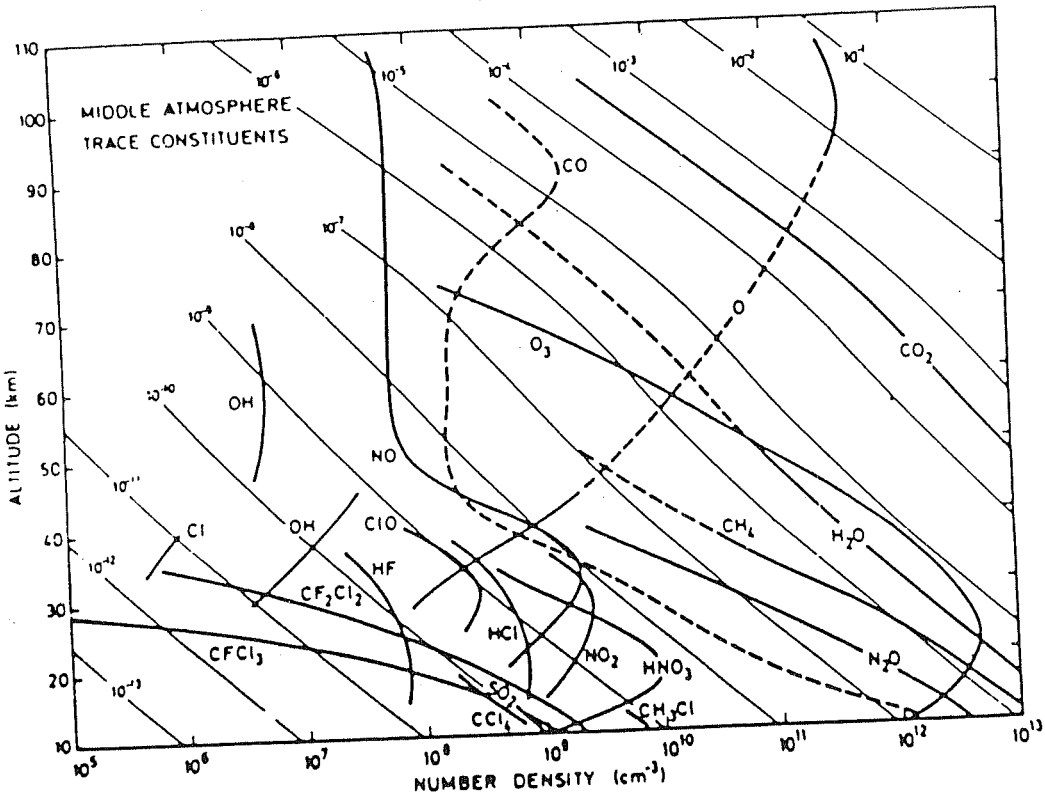

Figure 1.3 Vertical profiles of observed number densities of the typical trace constituents in the middle atmosphere (Ackerman, 1979).

The most important among the species mentioned above is O_3 which absorbs UV radiation coming from the Sun and thus prevents the harmful radiations from reaching the earth's surface. Fig. 1.4 shows the variation in the penetration height of solar radiation with wavelength. It can be seen that the radiation between $\sim$ 200 and $\sim$ 300 nm can penetrate to the stratosphere, and most of the energy is absorbed there. The radiation between $\sim$ 130 and $\sim$ 200 nm is absorbed mainly in the mesosphere, and below $\sim$ 100 nm the radiation is absorbed mainly in the thermosphere. Fig. 1.4 also shows the main absorbing molecules over various wavelength ranges. O_3 is the main absorber at $\sim$ 240 - 320 nm, O_2 at $\sim$ 100 - 200 nm, and both O_3 and O_2 absorb radiation at $\sim$ 200 - 240 nm. Absorption below $\sim$ 100 nm can occur by various molecules and atoms (N_2, O_2, N, O). Without the protection of this ozone shield, even a lower form of animal life could not have survived on the earth's surface, as the harmful solar UV radiation would have destroyed the chromosomes of the cell nucleus, thus prohibiting cellular multiplication. The production and loss processes of O_3 were first given by Sydney Chapman in 1936. According to him, O_2 molecules get photodissociated and form O_3 molecules as follows-

$$O_2 + h\nu \quad \rightarrow \quad O + O$$

$$O + O_2 + M \quad \rightarrow \quad O_3 + M$$

At the same time O_3 absorbs the radiation and gets dissociated into O and O_2.

$$O_3 + h\nu \quad \rightarrow \quad O(^1D, ^3P) + O_2$$

But the above simple chemistry of ozone is not able to balance the ozone budget, some processes are missing. It was pointed out in 1970 by *Crutzen* [1970] and *Johnston* [1975] that the ozone concentration is greatly affected by chemical reactions with minor constituents, belonging to three groups viz. HO_X, NO_X, and ClO_X. Although concentrations of these species are very small compared to O_2 and N_2, yet they catalytically reduce ozone by a large scale as follows:

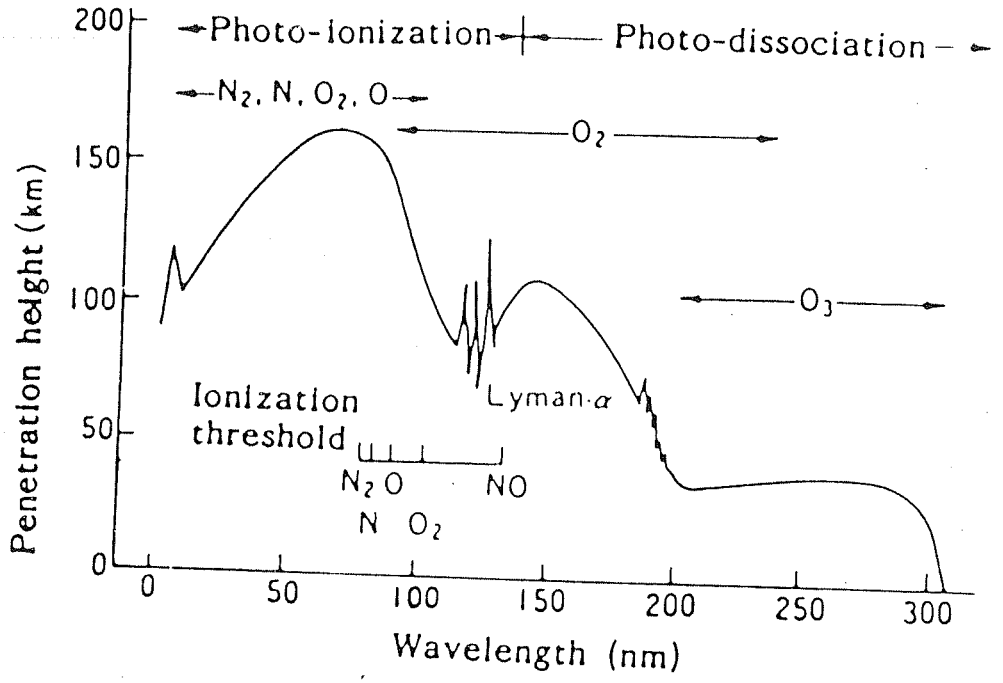

Figure 1.4 Penetration height for vertical incidence of solar radiation at various wavelengths (Shimazaki, 1985).

$$
\begin{array}{lcl}
X + O_3 & \rightarrow & XO + O_2 \\
XO + O & \rightarrow & X + O_2 \\
\hline
\text{Net} : O_3 + O & \rightarrow & O_2 + O_2
\end{array}
$$

where, $(X = NO_X, HO_X, ClO_X)$

In the present work, characteristics of two species belonging to NO_X group viz. NO_2 and NO_3 have been experimentally and theoretically studied at a low latitude station Ahmedabad (23°N, 72°E), India.

As mentioned above aerosols play a significant role in the radiation budget of the earth's atmosphere. The aerosol content of the atmosphere is disturbed by various processes; volcanoes are one among them. In June 1991, eruption of Mt. Pinatubo (15°N, 122°E) volcano took place. It produced a huge amount of aerosols into the stratospheric region. In the present study, characteristics of these volcanic aerosols and their effect on the NO_2 distribution have been studied at low latitude station Ahmedabad by a ground-based technique.

1.2 Aim and Scope of the Present Study

- It has been mentioned above that NO_X group of species can catalytically destroy ozone. NO_2 is one of the important species belonging to NO_X group. Variabilities of this species are known at middle and high latitude zone. At low latitude, these have not been properly studied. This is mainly because the data at low latitude is sparse. Satellite observations are available at low latitude but these are not fully validated. We have measured NO_2 column density at low

latitude station, Ahmedabad from winter 1989 to summer 1992. One area of current interest is whether volcanic eruption affects NO_2. A volcanic eruption which took place at Mt. Pinatubo, Philippines in June 1991 has provided us an opportunity to study its effect on NO_2 over Ahmedabad.

- NO_3 is another important species which belongs to the NO_X group. NO_3 variability has been studied at higher and mid latitude regions, but at low latitude region it has not been properly studied. Also NO_3 observational data at low latitude is very meagre. We have, therefore, studied the characteristics of this species at a low latitude station, Ahmedabad by a ground-based technique. Since NO_3 density is low during daytime, its density has been measured at night. Here, we have used direct moon as a source of light. This might introduce some tropospheric contamination in our measurements. To study this aspect, observations were taken at Gurushikhar, a hill station, 1.8 km above the mean sea level and 200 km away from the north of Ahmedabad where tropospheric pollution is less. In addition, the effect of Pinatubo volcanic aerosols on NO_3 has also been studied.

  Since NO_3 gets dissociated in the presence of solar radiation, it gives us an opportunity to study the disappearance of NO_3 during twilight period. Sunrise disappearance rate of NO_3 has been observed at sunrise time by collecting scattered photons from the west horizon, opposite to the direction of sunrise.

- An attempt has been made to reproduce the features of NO_2 and NO_3 observed by us. Chemical equations are formed with production and loss terms of these species. These are solved simultaneously for steady state conditions. How far the observational results match with the theoretical model, has been studied.

- Occasionally stratospheric region is loaded by volcanic aerosols. As mentioned

earlier, a volcanic eruption took place in June 1991 at Mt. Pinatubo, Philippines. In this eruption, a huge amount of sulphur dioxide was injected into the tropical stratosphere. These SO_2 gases were converted into H_2SO_4/H_2O aerosols. Characteristics of these aerosols have been studied by a ground-based technique at Ahmedabad.

This thesis has been subdivided into seven chapters, In the following sections, a brief summary and important results of the subsequent Chapters are given. These are characteristics of NO_2 and NO_3 species and volcanic aerosols. These results will contribute greatly to the global data set which will be useful not only for a better understanding of each of the basic processes, but also for obtaining an overall picture of the phenomena in the middle atmosphere.

Chapter 2 deals with the instrumental setup, observational procedure and data reduction method. Measurements of absorption spectrum in the region 436 to 448 nm for NO_2, 655 to 667 nm for NO_3, and 800 nm for volcanic aerosols by a ground-based technique is our aim. For that purpose we have used a McPherson UV-Visible monochromator ,model-270. The details of the instrument has been described. A chopper in conjunction with a Lock-in amplifier was used to improve the signal to noise ratio. The chopper was placed in front of the entrance slit of the monochromator and Lock-in amplifier was attached to the detector system. These details are given. Photomultiplier tubes were used as detector. For NO_2, R-372 Hamamatsu and for NO_3 and aerosol studies R-955 Hamamatsu photomultipliers were used. A cooling system was provided to R-955 tube. Scanning of the spectra was done by a computer. Data was digitized and stored in a floppy. For that purpose a software was developed by us. Besides operation of the instrument, various steps involved in the data reduction are also discussed in detail.

In Chapter 3, NO_2 and O_3 results obtained by us have been presented. Observations have been taken during twilight period. Scattered light from the zenith sky has been measured in the wavelength range 448 to 436 nm. A spectrum has also been taken near the noon time. This spectrum has been used as the back-ground spectrum. The path length covered by the solar radiation during twilight period is 20 to 30 times more than that during the noon time. The 436-448 nm wavelength region is full of Fraunhofer structure. By dividing the twilight spectrum by the background spectrum, Fraunhofer component has been removed. The ratio spectrum thus obtained contains the contribution due to NO_2, O_3, aerosol, air, H_2O and O_4. Using Beer Lambert's law slant column abundances of NO_2 and O_3 have been determined for sunrise and sunset times by a matrix inversion method. These slant column abundances have been converted into vertical column abundances by dividing them by the appropriate air mass factors. Diurnal and seasonal variations of NO_2 over Ahmedabad have been studied. During monsoon period observations have not been taken. NO_2 total column abundance is found to be minimum in the winter. The decay factor of NO_2 during night has also been determined. The decay factor is the ratio of $[NO_2]$ density at preceding evening to the morning $[NO_2]$ density. Correlation between NO_2 and O_3 has been examined for winter period. A weak positive correlation has been found. In addition, effect of volcanic eruption on NO_2 has also been investigated.

Chapter 4 deals with the total NO_3 column density measured by us. For these measurements we have used moon as a light source. NO_3 has strong absorption at 662 nm. Direct lunar spectrum between 655 and 667 nm region has been taken near the full moon period. Background spectrum has been taken during daytime at higher solar zenith angle with direct sun as light source. Background spectrum has also been taken with moon as source at lower lunar zenith angle. Besides NO_3, water vapour also has absorption in this wavelength region. The contribution of water vapour has been removed by taking the ratio of the nighttime to the daytime spectra.

The nighttime spectrum is further divided by the background spectrum. The ratio spectrum thus obtained has been used to derive the NO_3 slant column abundance by using the matrix inversion technique. NO_3 slant column abundance is then converted to vertical column abundance by dividing it by the appropriate airmass factor.

Ahmedabad being an urban area, the contribution of tropospheric NO_3 is likely to be significant. Therefore, some observations have been taken on a hill station Gurushikhar (24°N, 73°E), 1.8 km above the sea level and about 200 km north of Ahmedabad. Here tropospheric pollution is negligible. Signal to noise ratio at lunar zenith angle 85° and more becomes very small in the urban area due to the presence of dust particles in the lower atmosphere. Therefore, at Ahmedabad, observations could be taken upto $\sim 85^o$ lunar zenith angle only and at Gurushikhar, observations were possible up to $\sim 90^o$ lunar zenith angle. Tropospheric and stratospheric contributions can be distinguished in slant column abundance for lunar zenith angle greater than 83°. Slant column abundance of NO_3 obtained at Gurushikhar shows stratospheric contribution. NO_3 values obtained at Ahmedabad appears to have some tropospheric component. A large scatter in the values of NO_3 is noticed. These could be because of highly temperature dependence of NO_3 production rate.

Vertical column abundance of NO_3 remains constant throughout the night. After sunrise, NO_3 starts getting photodissociated. The decreasing rate of NO_3 after sunrise has been studied by measuring the scattered photons coming from the west horizon in the morning. As the solar depression angle decreases, the amount of atmospheric illumination increases and the NO_3 value starts decreasing. Using this feature, lower limiting value of NO_3 has been determined for Gurushikhar.

An attempt has been made to reproduce theoretically the features of NO_2 and NO_3 observed by us. Chemical equations are formed with production and loss terms of these species. These are solved simultaneously for steady state conditions and

the results of this study has been presented in Chapter 5. Calculations have been made at an interval of 5 km for day and nighttime conditions. Calculated vertical distribution of these species have been converted into total column abundance and compared with the values of NO_2 and NO_3 observed by us. It appears that nighttime NO density plays a crucial role in the distributions of NO_2 and hence NO_3 at least in 1 D model. Calculation of decay factor has been carried out for winter at 200 K and 220 K stratospheric temperatures. Values obtained for 220 K and O_3 abundance observed along with NO_2 show very good correlation with the observed decay factor. Similarly observed and calculated diurnal variations of NO_2 agree with 220 K.

Occasionally the stratospheric region is loaded by volcanic aerosols. A volcanic eruption took place in June 1991 at Mt. Pinatubo, Philippines and injected huge amount of SO_2 into the tropical stratosphere. These SO_2 gases were converted into H_2SO_4/H_2O aerosols through condensation. Physical characteristics of these aerosols have been studied by a ground-based technique at Ahmedabad and has been discussed in detail in Chapter 6. Normally, after sunset the twilight intensity decreases exponentially with the increase of solar depression angle. However, due to an additional aerosol loading after volcanic eruption, the twilight radiation intensity after sunset again increases and then decreases. Using this anomalous twilight intensity variation, lower altitude of aerosol layer, layer thickness, aerosols strength (which is proportional to the total aerosol mass in the stratospheric region) have been determined in the same way as done by *Ashok et al.* [1982]. Most of the observations have been taken at 800 nm wavelength by a photometer looking in the zenith direction. Scattered intensity has been measured on the ground from 0° to $\sim 8^o$ solar depression angle.

Aerosols strength Q is proportional to the total stratospheric air mass of aerosols. Observations have been taken since November 1991. A monotonic increase

in the Q values have been obtained upto December 1991. This agrees with the optical depth results of SAGE-II. Height of the aerosols layer has been found to vary between 22 and 28 km which agrees with lidar observations taken near the equator by *Grant et al.* [1992]. Sometimes multiple layers have also been observed. Thickness of the single aerosol layer shows seasonal variation, decreasing as winter progresses and increasing as summer approaches.

Chapter 7 gives the conclusions of the present study and some suggestions for future study.