Ozone studies in India

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India has a long history of ozone measurements which goes back to 1928-29, and is one of the few countries in the world where a serious study of atmospheric ozone and its spatial and temporal distribution has been systematically made for over forty years. The paper describes the pioneering work done by Ramanathan and his colleagues in the India Meteorological Department (IMD), Poona, and later at the Physical Research Laboratory (PRL), Ahmedabad, before and after the International Geophysical Year. The excellent tradition established in the fifties is being continued at a number of laboratories, such as IMD, PRL and the National Physical Laboratory (NPL), New Delhi. The major findings of the various study groups are presented.

1 Introduction

Ozone measurements in India were first made over sixty years ago, to be exact during 1928-29, as part of Dobson's programme of world-wide total ozone measurements with his photographic ozone spectrophotometer. Those classical measurements, organized to study the variations of ozone with latitude, seasons and weather, formed the basis of all discussion of ozone on a world-wide scale till the International Geophysical Year (IGY) when regular ozone observations were organized on a global basis.

The measurements of total ozone in India were first made at Kodaikanal (10°N) during 1928-29 (Ref.1), followed by measurements at Bombay (19°N) and Poona (18°N) by Chiplonkar2 during 1936-38, using the same photographic instrument. Dobson's measurements at Kodaikanal had established the existence of low values of total ozone over the tropics with very little variation throughout the year and the absence of any correlation between ozone and weather. Chiplonkar's measurements2 at Bombay and Poona confirmed the earlier observations at Kodaikanal.

The first Dobson ozone spectrophotometer used for regular measurements of ozone was acquired in 1940 by the India Meteorological Department (IMD), Poona. Preliminary measurements were made by Ramanathan and his colleagues at Poona soon after, but war-time work prevented their continuance till 1945, when observations were resumed. Daily observations were made at New Delhi (29°N) during 1945-47 and at Simla (31°N), Poona and Kodaikanal during 1948-49.

Systematic measurements were started by Ramanathan at one station, Ahmedabad (24°N) in 1951, followed by three more stations, namely, Kodaikanal, Srinagar (34°N) and New Delhi in the late fifties. Regular measurements at a network of six stations in India were organized by him during the International Geophysical Year (IGY). The instruments have regularly been checked and periodically calibrated and the data from the network have regularly been published since then by the World Ozone Data Centre at Toronto. Poona is the National Ozone Centre for India and the Regional Ozone Centre for the Regional Association II (Asia) of the World Meteorological Organization.

It was Ramanathan and his associates who were responsible for the pioneering investigations of atmospheric ozone in India and who carried out detailed measurements of total ozone and its vertical distribution in the atmosphere using Umkehr techniques during the forties and fifties and established the main features of the horizontal and vertical distribution of ozone over the tropics. They confirmed the fact that the day-to-day variations in the ozone amounts in the tropics are small and showed that the level of maximum ozone is higher (25-28 km) in the tropics than at higher latitudes3,4. They also showed that in the tropics, unlike in other places, the vertical distribution does not change with the seasons. Studying the seasonal and latitudinal variations of total ozone, they pointed out that ozone amounts in the tropics south of 30°N stand in a class apart.

They were also the first to study the vertical distribution of atmospheric ozone on a global scale5. The extremely important relationship between atmospheric ozone and the general circulation of the atmosphere, announced by Ramanathan6 in his Presidential address to the International Association
of Meteorology at Rome in 1954, was, in a sense, responsible for the establishment of a large number of ozone measuring stations in the world during the IGY and the theoretical studies, that followed, of the behaviour and transport of ozone in the atmosphere. Another important contribution by Ramanathan to the study of the geographical distribution of ozone was his work on the mean meridional distribution of ozone in different seasons and the probable transport mechanisms. In his classical analysis of the data collected during IGY and IGC on the global distribution of ozone, Ramanathan pointed out the pronounced longitudinal differences in ozone that exist, particularly, in late winter and spring, and showed that the spring rise in ozone in high and middle latitudes was associated with the break-up and warming of the polar stratospheric vortex. He also pointed out the fundamental similarity between the Arctic and Antarctic stratospheric circulations, but with a difference in timing. He also discovered the quasi-biennial oscillation of total ozone, associated with the 26-month oscillation in stratospheric winds and temperatures.

2 General features of ozone in the tropics

World-wide measurements of atmospheric ozone during the last sixty years, both from ground-based and satellite-borne instruments, have enabled us to understand the broad features of the distribution of ozone over the globe. The mean annual distribution of total ozone over the globe, based on 23 years' data, is shown in Fig.1 (Ref. 12).

While the total ozone and its vertical distribution over the tropics are more or less constant throughout

the year, large seasonal variations are observed in both total ozone and its vertical distribution in extra-tropical latitudes. This is illustrated in Fig.2 which shows the mean vertical distribution of ozone in the northern hemisphere in three typical months. Fig.3, based on IGY and IGC data, shows the distribution of total ozone in the two hemispheres. At high and middle latitudes, pronounced maxima and minima in total ozone occur in spring and autumn, respectively, while in the tropics, the
maximum, though feeble, occurs in summer, and the minimum in winter. In marked contrast with the situation in the middle and high latitudes, the total ozone in tropical region is subject to much smaller inter-diurnal, seasonal and inter-annual fluctuations. This is primarily because of the very weak coupling between the troposphere and the stratosphere in the tropical regions. A study of the monthly mean total ozone at a few tropical stations showed that certain long-period variations occur simultaneously over large areas\textsuperscript{13,14}. While a part could be attributed to the quasi-biennial oscillation (QBO), the remainder could only be attributed to volcanic activity, solar activity or to the destruction of ozone in the stratosphere\textsuperscript{15}.

In their study of the global ozone budget, Brewer and Wilson\textsuperscript{16} had arrived at a net production of ozone in the tropics, with a maximum at about 30 km, and net destruction at high latitudes, particularly, in the winter hemisphere and below 25 km. To balance the budget, ozone is transported from the region of primary production in the equatorial middle and upper stratospheres, polewards and downwards to the lower stratosphere, mainly by large-scale atmospheric motions, and then transferred to the upper troposphere in the middle and high latitudes, and eventually destroyed in the lower troposphere or at the surface of the earth. This broad picture of the ozone cycle emerges from most studies, but quantitative evaluation of the photochemical processes differs among the various workers. Details of the transport processes are also not yet settled.

The main features of the spatial and temporal distribution of ozone over the globe were known and the observed variations explained many years ago. More sophisticated measurements with ozone sondes and satellite-borne sensors in the last three decade have not basically changed the earlier picture. The stress has now shifted from the study of ozone as a natural tracer in the study of atmospheric motions in the lower stratosphere to its study as a factor in producing climate change.

3 Recent work on atmospheric ozone in India

In addition to the regular measurements of total ozone and its vertical distribution using Dobson ozone spectrophotometers at a network of six stations, regular surface ozone measurements, using electrochemical sensors at five stations and vertical profile measurements using electrochemical balloon-borne ozone sondes at three stations, are made by IMD. Fig.4 shows the network of ozone measuring stations in India. Measurements with rocket-borne optical sensors are periodically made from one station, namely, the Thumba Equatorial Rocket Launching Station near Trivandrum (8°N).

The first Indian balloon-borne electrochemical ozone sonde was developed in 1962 (Refs 17,18) and the first surface ozone recorder in 1970 (Ref.19) at the Instruments Division of the Meteorological Office at Poona. Regular ozone soundings at three stations, Delhi, Poona and Trivandrum, have been made since 1971 and surface ozone measurements made since 1973 and results from both have been analysed\textsuperscript{20-34}. Fig.5 shows five typical ozone profiles obtained at 5 stations in India. The ozone maximum, which is at about 27-28 km at Trivandrum, lowers to 23-24 km at New Delhi. The lowest ozone amounts are found just below the tropopause at about 15 km over all the stations and the largest fluctuations are found over New Delhi in the lower stratosphere. These large variations are caused by the influx of ozone-rich air from higher latitudes through breaks in the tropopause which occur during February-April over New Delhi. Above 30 km, the ozone remains nearly constant with mixing ratios of the order of 15-20 µg/g at 8°N decreasing to 10-15 µg/g at 29°N. Significant amounts of ozone, about 8 to 15 per cent of the total, are found below the tropopause and the average ozone mixing ratio increases from about 0.05 µg/g near the equator to about 0.1 µg/g at 29°N.

Fig.6 shows the mean ozone profiles during the four main seasons, namely, winter (November-February), pre-monsoon (March-May), monsoon
ANNA MANI: OZONE STUDIES IN INDIA

(6Ue-August) and post-monsoon (September-October) at Poona. The vertical distribution of ozone over the equatorial regions remains the same in all seasons and a change in total ozone is brought about by a proportionate change at all levels up to 30 km. Both the total ozone and the level of maximum ozone are higher in the pre-monsoon and monsoon months (March-August, 280 DU) and low in the post-monsoon and winter months (September-February, 261 DU). The level of maximum ozone rises from 24 km in the winter to 26 km in the monsoon season. The ozone mixing ratio above the maximum also increases from 12 µg/g in the winter to 15 µg/g in the pre-monsoon and monsoon seasons.

The first measurements of surface ozone in the tropics were made by Ramanathan and Dave at Ahmedabad in 1954 using Ehmert's apparatus. They showed that substantial amounts of ozone are present in the lower atmosphere, and that surface ozone undergoes regular daily variations with a maximum in the afternoon and a minimum early in the morning, with a rapid increase in conditions of strong turbulence. Recent measurements confirm these earlier observations. They also show that the concentration is lowest during the pre-monsoon months and highest during the winter months (40-60 µg⁻³). The monsoon months are characterized by low average ozone densities and the smallest diurnal variation.

Fig. 7 shows the diurnal variation of surface ozone at Trivandrum during four typical months. An analysis of the ozone soundings data at New Delhi, Poona and Trivandrum had shown the ozone mixing ratio at all the 3 stations, at levels below about 3 km, to be remarkably constant with values ranging from 0.03 to 0.07 µg/g. Within this range, relatively higher values tend to occur during the pre-monsoon months, when mixing with higher levels due to turbulence is more active. Sreedharan and Rangarajan also found the ozone mixing ratio to be increasing slowly, with the highest values from about 3 km to about 12-16 km, thus providing a small vertical gradient necessary to maintain the long term ozone fluxes from the stratosphere to the earth's surface. At Poona and Trivandrum, representing the tropics, the maximum mixing ratio occurs during April-August, whereas at Delhi, it occurs during December-February when troughs in the mid-latitude westerlies are active.

The first rocket-borne multichannel UV photometer ozone sonde was developed in 1976 at the Physical Research Laboratory (PRL), Ahmedabad, using the absorption of solar ultraviolet radiation in the wavelength bands 250, 280 and 310 nm. Flown on Centaure and M-100 rockets, a number of ozone profiles have been obtained over Thumba. High-altitude balloon measurements up to altitudes of 35 km have also been made over Hyderabad, using a sun-tracking multichannel radiometer, which yields absolute values of ozone number densities and which has much better altitude resolution than rocket sondes. A 1.27 µm photometer which can determine ozone concentrations in the 50-90 km region has also been developed at PRL.

Using microwave radiometers operating at 110-836 GHz, Vivekanand and Arora have obtained, from measurements of the emission spectra of ozone in the mm-wave region, vertical profiles of ozone over Bangalore. They expect more accurate results with an acousto-optic spectrometer, when installed.

Investigations of the ozone layer and other trace gases, particularly over the low latitudes, their variations and chemistry, formed one of the important components under the Indian Middle Atmosphere Programme (IMAP) during 1982-86 and are currently being continued under IMAP-Continuation (IMAP-C). The National Physical Laboratory (NPL), New Delhi, has a major programme involving ground-based rocket and satellite systems. A two-channel rocket-borne solar ultraviolet photometer has been developed at NPL and flown from Thumba. An infrared laser heterodyne system to monitor trace species in the
Fig. 6—Seasonal variations in ozone profiles at Poona
Two major ozone campaigns, using a number of sounding rockets carrying different types of experimental payloads to measure ozone profiles, were carried out in 1983 and 1987 (Refs 45, 46). Based on available data, a mean reference ozone profile over the tropics has been constructed47-48.

A number of national and international comparisons were conducted to verify the accuracy of the measurements made with balloon-borne and rocket-borne ozone sondes and Dobson ozone spectrophotometers49,50. A number of national and international workshops on ozone have also been held, where the current status of research on ozone was examined and priority areas for future research were identified51-52.

Measurements of both total ozone and its vertical distribution at Dakshin Gangotri in the Antarctic as well as surface ozone measurements over the Indian Ocean have also been made53-55.

A recent analysis of long-term series of total ozone in the tropics shows no significant trend56. However, there are large variations on many time scales which would make a small trend of, say, less than 1% per decade, difficult to identify. Ozone in the upper

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stratosphere and troposphere, using the 9-11 μm window, has been set up at NPL, New Delhi43. The ozone profiles obtained with this system during 1987-88 are shown in Fig.9. There has also been some efforts in postulating simple 1-D photochemical-diffusive models to study the latitudinal sensitivity of climate to ozone perturbations44.

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Fig. 7—Diurnal variation of surface ozone in 4 typical months at Trivandrum

Fig. 8—Ozone concentration measured with PRL optical ozone sonde (after Subbaraya37)
stratosphere appears to have decreased by about 2-3% since 1978, although measurement of this quantity is particularly error-prone.

In addition to studies of atmospheric ozone measurements have also been made of other trace gases in the atmosphere\textsuperscript{48,57}. A number of balloon ascents, using cryogenic air samplers and PRL's multichannel sun-tracking photometers, have been made over Hyderabad (17°N) by PRL in collaboration with the Max Planck Institute for Aeronomy (MPAE), West Germany, to measure the vertical distribution of various trace species in the atmosphere. Results show that while the abundance of these gases are the same in both tropical and middle latitudes, the decreases in mixing ratio are lower over Hyderabad than over mid-latitudes\textsuperscript{38,39,58}.

A number of rocket ascents have also been made by NPL in collaboration with Japanese scientists over Thumba (8°N) to measure the vertical distribution of various trace gases in the atmosphere, apart from ozone.

The UV-B radiation (280-320 nm) is also being monitored at seven stations (New Delhi, Shillong, Jodhpur, Mysore, Waltair, Poona and Trivandrum), using the NPL's automatic, integrating type UV-B radiometers\textsuperscript{59}.

To conclude, considerable effort has been made in India in recent years to measure atmospheric ozone and other minor constituents in the atmosphere, using a variety of sensors and techniques. The approach of Indian scientists in studying the entire atmosphere as an integrated whole should accelerate future developments in this area in the country.

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ANNA MANI: OZONE STUDIES IN INDIA


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