ATMOSPHERIC OZONE AND THE GENERAL CIRCULATION OF THE ATMOSPHERE

K. R. RAMANATHAN (India)

INTRODUCTION

I WISH to express my cordial thanks for the honour you have done me by inviting me to be the President of the Association of Meteorology for the three-year period culminating in this meeting in the great city of Rome. I consider that this is symbolic of the significance you attach to the Meteorology of the Tropics in the general Meteorology of the globe.

At the Lisbon meeting of the Assembly in 1933, one of the themes for discussion in the Association of Meteorology was 'Geophysical Knowledge of the Stratosphere[1]'—Dr. F. J. W. WHIPPLE opened a discussion on the determination of the temperature of the upper atmosphere by the study of air-waves. Prof. CHARLES FABRY the great Master of Optics and the founder, with BUISSON, of the modern study of atmospheric ozone presented a report on the progress of studies in that subject. He spoke of the absorption spectrum of ozone and the laboratory determinations of the coefficients of absorption, done mainly by himself and his co-workers in France, fundamental studies which have been the mainstay of most of the later experimental work on atmospheric ozone. He spoke of the different sources which had been utilized for studying atmospheric ozone, direct light from the sun, diffused sunlight from the sky or the moon and the light from the stars. He welcomed the successful employment by DOBSON of the photoelectric cell in place of the photographic plate for the study of these spectra. Prof. Fabry briefly discussed the important problem of the distribution of ozone in the vertical and of the great advantage of the use of light from the zenith sky for this purpose. He also referred to the development of chemical and optical methods for the measurement of ozone in the layers near the ground.

At the same meeting, Dobson and GÖTZ* reported the progress that had been made on the study of the height-determination of ozone from the *umkehr effect*, an effect that had been discovered by Götz during his observations in Spitzbergen. Using light from the clear zenith sky and comparing the intensities of light of two wavelengths in the Huggins band, Götz found that, as the sun went down, the shorter wavelength was relatively less and less absorbed than the longer wavelength. A study of this effect led to the conclusion that the height of the centre of gravity of ozone in the atmosphere was in the neighbourhood of 20 km instead of 35–50 km as had been thought before and that there was some ozone present in lower layers also.

This immediately made it easier to understand the connection that had

* Ozone studies have suffered a great loss by the recent death of Prof. F. W. P. Götz. His contributions to the study of atmospheric ozone have been of outstanding importance.

Sci. Proc. Int. Assn. Met., IUGG 10th General Assembly, Rome 1954, Butterworths, London, 3–24, 1956.

been observed between ozone changes and weather and brought ozone within the realm of interest of the synoptic meteorologist.

Since the Lisbon meeting, the subject has advanced in many directions. Shortly after the discovery of the Götz effect, Drs. E. and V. H. REGENER[2] sent up spectrographs in sounding balloons and determined the amount of ozone lying above different levels in the atmosphere and confirmed the substantial correctness of the findings of GÖTZ, MEETHAM and DOBSON.[3] Our American colleagues have used rockets and recorded the solar spectrum up to 70 km, above which there is very little ozone left. Physical chemists and mathematicians have steadily improved CHAPMAN's theory of the photochemical equilibrium of atmospheric ozone and given us more precise information regarding the equilibrium distribution of ozone which may be expected to exist if atmospheric ozone were formed and destroyed by the action of sunlight on our atmosphere.

Improvements in the technique of the chemical method of measuring ozone effected by Dr. V. H. Regener and Dr. EHMERT[4] have made easier the study of ozone changes at ground level and at heights accessible to aeroplanes and opened up a new way of studying the transport processes from the stratosphere downward.

In the meantime, the studies of our colleagues in the field of dynamical meteorology and synoptic aerology have improved our general understanding of atmospheric processes and given an insight into the mechanism of the day-to-day changes in ozone observed in extra-tropical latitudes.

The International Ozone Commission of our Association with Prof. DOBSON as Chairman and Sir CHARLES NORMAND as Secretary has kept up vigorous activity in the study of 'Ozone and Weather' by organizing a network of ozone measuring stations in Europe, distributing calibrated spectrophotometers to interested workers in different countries and by holding periodical conferences, both in the Association of Meteorology and elsewhere, to discuss new results.

BRIEF SUMMARY OF FACTS RELATING TO ATMOSPHERIC OZONE

As a result of many years of work, we know the following facts about atmospheric ozone.

Ozone is present in the earth's atmosphere over all places, but its distribution is not uniform. At the same place, the total amount is variable from day to day, season to season and even year to year. The broad features of the height distribution of ozone at a few places are known; it is present in measurable quantities up to 70 km and more, and variations in distribution are common. In spite of these variabilities, a considerable portion of atmospheric ozone behaves over short periods as if it were conservative and it is possible to associate certain types of ozone changes with specifiable features of moving disturbances.

The usual way of expressing the total ozone amount present over a place is as x cm of ozone at standard temperature and pressure. The amount varies from 0·15 cm to 0·45 cm. In tropical latitudes, the average amount is about 0·180 cm and the day-to-day variations are generally small. At higher latitudes in the region of the westerlies, the average amount is larger

and also the day-to-day and seasonal variations. The seasonal changes at a few selected places are shown in *Fig. 1.* It includes the data of Tromsö (70° N), Dombås or Oslo (61° N), Arosa (47° N), Azores (37° N), Zi-ka-wei (31° N), Delhi (28½° N), Abu (24° N) and Kodaikanal (10° N). The monthly mean values of the first three stations and of Zi-ka-wei are based on

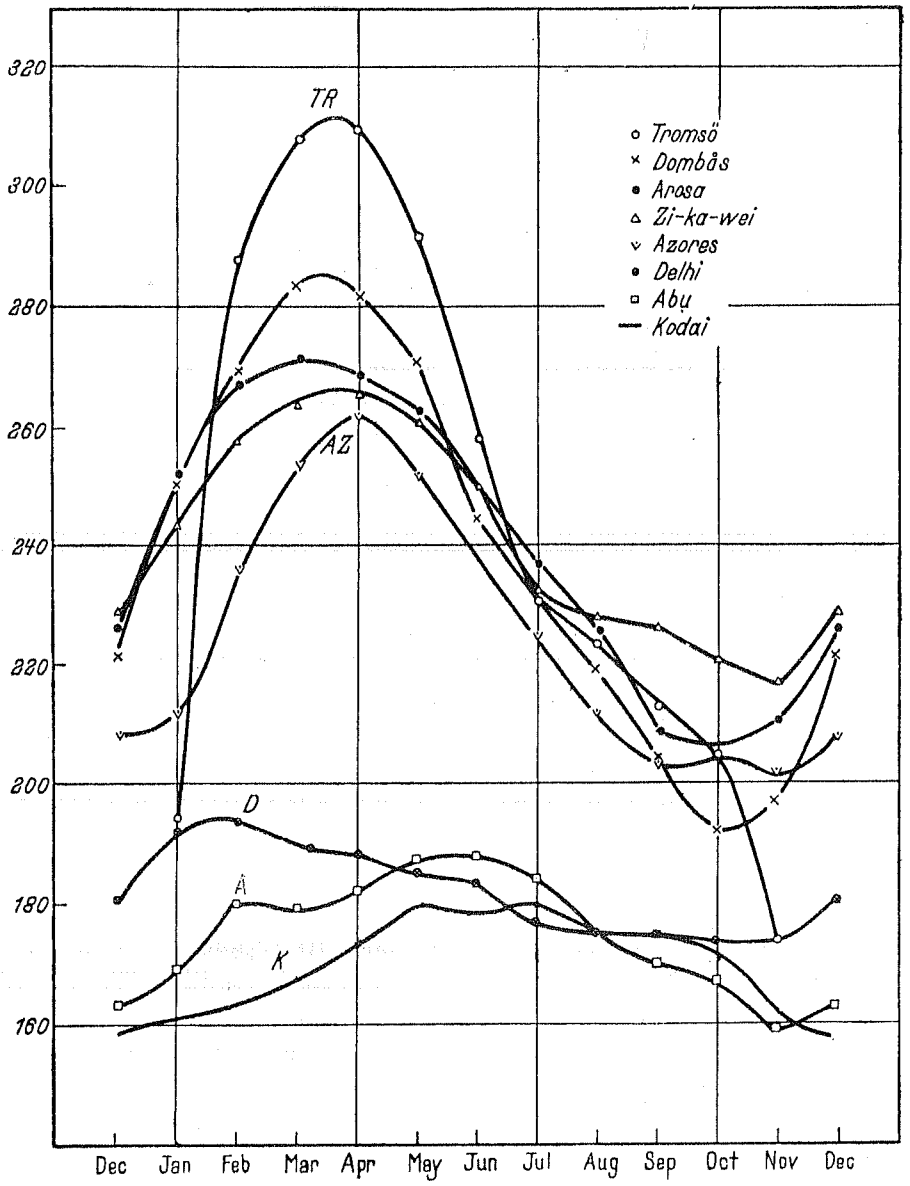

Figure 1. Mean monthly ozone amounts

9 to 10 years of observations (taken from LANGLO[5]) while the other stations have much shorter periods of observation. That the maximum ozone amount occurs in the spring months March–April and the minimum in October–November has remained a curious and interesting fact ever since its discovery by Dobson and his co-workers in 1926–29. Another fact is now clear; namely, that the ozone amounts at stations south of 30° N stand in a class apart. It is interesting to note that the maximum at Kodaikanal,

feeble though it is, occurs in June–July and that at Abu is shifted to May–June. In the southern hemisphere, the maxima and minima occur in the respective spring and autumn seasons. We shall return to this question of zonal distribution and its seasonal changes later.

THE HEIGHT-DISTRIBUTION OF OZONE

In general, the ozone amount increases with height, reaching a maximum at some level between 20 and 30 km and decreases thereafter with further increase in height. The ozone concentration at the level of its maximum varies from 0·008 to 0·020 cm/km. There is detectable ozone even at a height of 70 km. In tropical and subtropical latitudes there is only a small amount of ozone in the troposphere. With increasing ozone more and more of it is present at levels below 25 km.

The methods of determining the height-distribution of ozone are based on:

1. The Götz effect in zenith-scattered light;
2. Direct sun spectra taken at different levels in the atmosphere with spectrographs carried in balloons or rockets;
3. Spectrophotometry of the light of the eclipsed moon in the Chappuis band region;
4. Chemical determination from aeroplanes of ozone content at different levels.

Each method has its advantages and disadvantages. For obtaining information about ozone at great heights, the rocket-carried spectrograph has unique advantages. But it is expensive and difficult and cannot be adopted for daily use at a number of places. REGENER's balloon-carried spectrograph or the simpler modification of it with photoelectric cell and filters as used by COBLENZ and STAIR and telemetering similar to that used by STRANZ will probably find more extensive use in the future. The Weissenau group under Prof. E. Regener are specializing in this field. For a general over-all survey, the Götz umkehr method has proved extremely effective, and for following day-to-day changes of ozone, it will be hard to replace. WALTON[6] has worked out a method for making an approximate correction for secondary scattering. A defect of the umkehr method is that it does not give a sharp enough answer about the distribution of ozone in the lower levels. One can get an average value over a large depth, but if an attempt is made to subdivide the layers, many differing solutions can be obtained. Dr. Ehmert, and recently Dr. KAY[7] have used the chemical method for measuring ozone from aeroplanes up to a height of 12 km and information collected by this method can be very helpful to improve the height-distributions computed from the Götz effect. Dr. CHALONGE and Dr. VIGROUX in France and Dr. PAETZOLD[8] in Germany have made studies of the spectrum of the light of the eclipsed moon across the shadow frontier and since that light has passed through the earth's atmosphere in a nearly horizontal direction, it shows pronounced absorption in the Chappuis bands. They find that the ozone distributions obtained by this method agree reasonably well with those obtained by other methods. This method promises to give information about the differing distributions of ozone in different latitudes on the same day.

A summary of the mean height distributions obtained by the umkehr method at Tromsö, Arosa, Delhi and Poona for different ozone amounts is given in *Fig. 2.* A few selected distribution curves obtained by Dr. REGENER

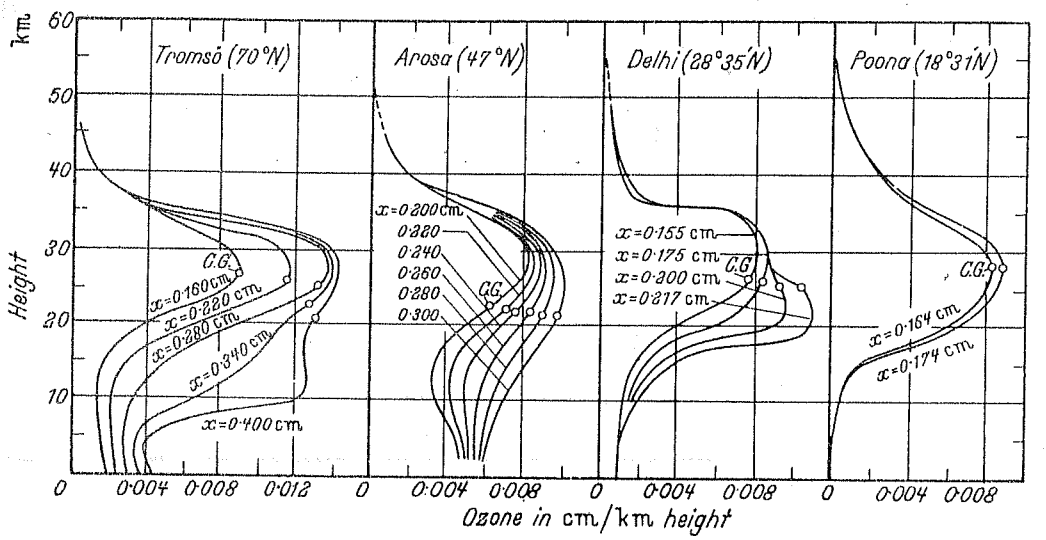

Figure 2. Height distributions of atmospheric ozone in different latitudes

and his associates[9] by the sounding balloon and aeroplane methods are given in *Fig. 3.* F. S. JOHNSON, J. D. PURCELL and R. TOUSEY[10] have compared a distribution curve obtained at New Mexico by a rocket spectrograph with a

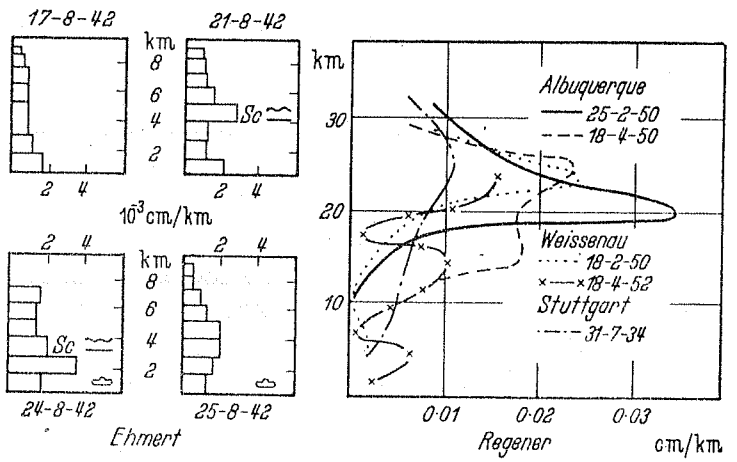

Figure 3. Distribution of ozone in the vertical from aeroplane and balloon ascents

curve obtained from umkehr observations on the same day. While the rocket spectrum gives the same total ozone amount, the maximum shown by it is more concentrated than the flat maximum given by the umkehr method.

OZONE AND WEATHER

FABRY[11] and BUISSON in their first series of measurements at Marseilles noted that there were large variations of ozone amount between one day and another, but it is to DOBSON and his collaborators[12] that we owe the

opening out of this fruitful subject. The changes associated with weather are not only in the total ozone amount but are also in its vertical distribution. We have a large body of data relating to the former, but factual information regarding day-to-day changes in vertical distribution is still very scanty.

It has been established that in middle and high latitudes:

1. Low ozone is associated with high pressures and high ozone with low pressures;
2. The approach of a warm front causes a fall in ozone even before any frontal clouds are seen. The surface front may then be more than 1000 km away;
3. The lowest ozone amount is observed generally before the passage of the surface warm front. The ozone amount remains low in the warm sector of a cyclone;
4. With the passage of the cold front, the ozone amount rises and the highest values are found in the rear of the cyclone at a distance of 100 to 300 km;
5. Occlusion of warm air in upper warm air tongues causes a fall in ozone and occluded slow-moving cold air type cyclones cause a comparatively large rise in ozone;
6. Ozone changes associated with highs and lows are most marked in winter and spring and least marked from June to October.

From a study of the upper air data obtained at Kew or Sealand together with the ozone measurements at Oxford, MEETHAM[13] was able to show that there was high positive correlation between ozone amount and potential temperature at 18 km and negative correlation with densities of air at 12 to 18 km and with heights of tropopause.

The studies of trajectories of air at high levels with ozone amounts measured at Arosa, Potsdam and other stations by MOSER[14] and PENNDORF[15] have established that, in Europe, the advection of air from polar regions particularly at heights of 11 to 16 km is associated with high ozone values in winter and spring and that advection from subtropical and mid-Atlantic regions goes with falls in ozone amount. Moser emphasizes the importance of large-scale air movements near the tropopause level in determining the ozone changes. During the last three years, Sir CHARLES NORMAND[16] has produced convincing diagrams showing the close association of low ozone at Oxford with passages of ridges in tropopause level and high ozone with troughs (slide). The 500–300 mb thickness goes in unison with height of tropopause, thus associating cold upper troposphere with low tropopause and high ozone. Normand has shown that the whole cycle of changes from high ozone to high ozone through an interval of low ozone has a quasi-period of 6 to 14 days. In lower latitudes the day-to-day changes of ozone are much smaller and it is therefore more difficult to analyse the effect due to weather disturbances. Observations at Delhi made by KARANDIKAR in 1945–47 and recently by MURTHY, KULKARNI, SANYAL and DEGAONKAR at different places in India show however that there are significant variations of ozone in December to April, the period when western disturbances are active. The ozone changes at Abu and Delhi during the winter and spring of 1953 and 1954 have been recently studied with the upper wind charts of the India Meteorological

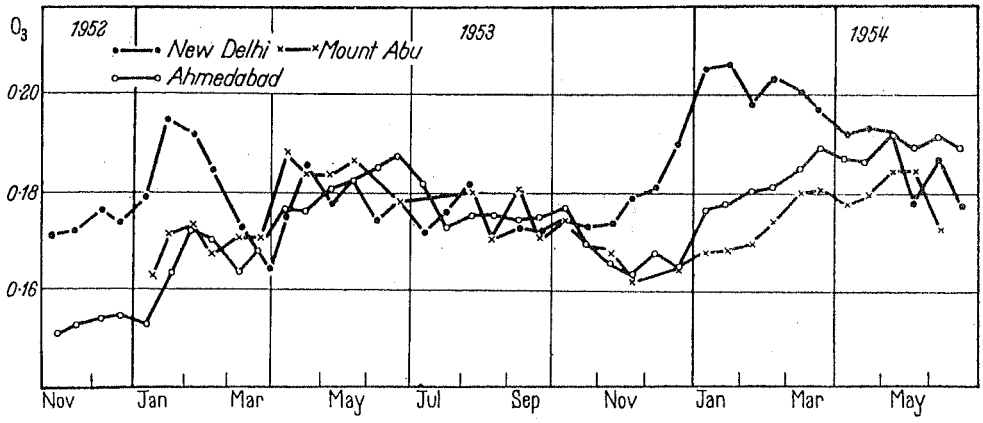

Figure 4. Fifteen-day mean ozone values at New Delhi, Mount Abu and Ahmedabad (Veraval)

Department. The general conclusions are:

The ozone increases come in a series of surges commencing in November or December. Some pronounced surges at Delhi, Abu and Ahmedabad are associated with the passage of deep troughs of low pressure at 6 and 9 km. The rise of ozone takes place when the north-westerly winds are replacing the south-westerlies, but once the north-westerlies get settled, the ozone amount begins to fall. Quite often, the ozone variations at Delhi and Abu

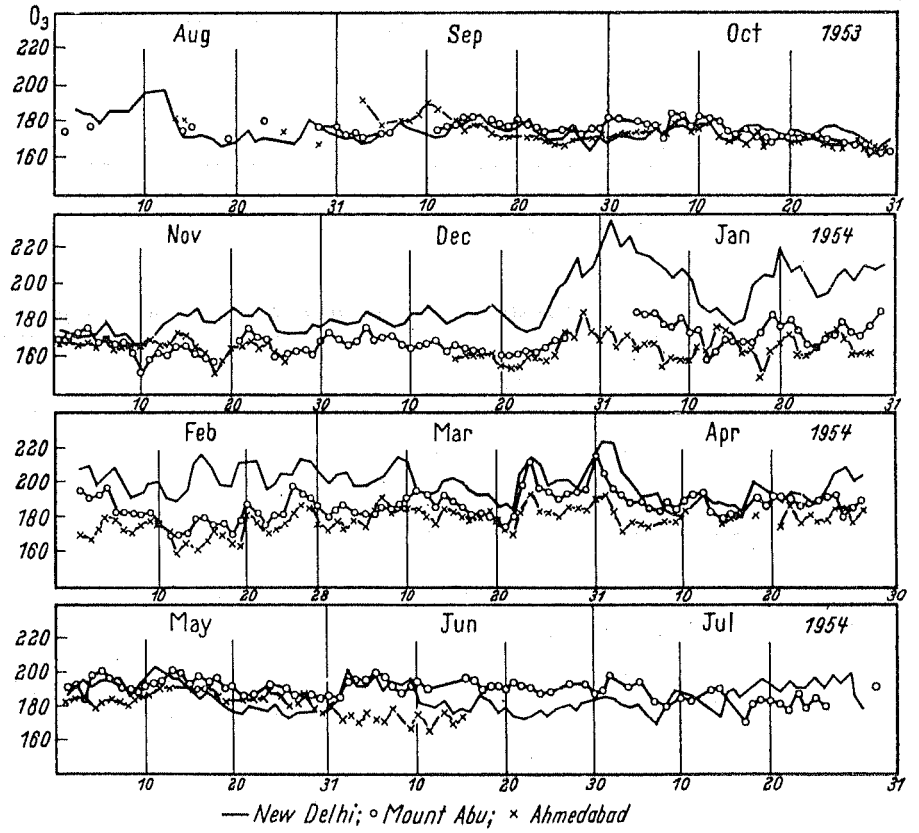

Figure 5. Daily ozone values at New Delhi, Mount Abu and Ahmedabad or Veraval

run parallel to each other, but the amplitude at Abu is invariably smaller than that at Delhi. At Ahmedabad or Veraval, the amplitude is still smaller.

The observations in north-west India have made it clear that the middle latitude variations of ozone associated with waves in the upper atmosphere are observable at latitudes down to 20° N in winter and spring, though less frequently and in a less pronounced manner.

Fig. 4 shows the simultaneous half-monthly mean values of ozone at Delhi, Abu and Ahmedabad or Veraval. There is a conspicuous increase in ozone at Delhi in winter and spring. One recalls the quick change from a higher to a lower tropopause over Habbaniya[17] which occurs towards the end of October. From April to October, the ozone amounts at the three places do not differ materially from each other.

Fig. 5 shows the simultaneous daily values of ozone at the three places during the period September 1953 to July 1954. The sudden increase of ozone at Delhi on November 10, 1953 and the large differences between the values at Delhi and Abu in January to March may be noted. There is another feebler increase in the spatial variation of ozone early in June when the upper easterly jet begins to be active. It is worthy of note that there are periods during the monsoon when the ozone amount at Abu exceeds both the amount at Delhi and that at Veraval.

CHANGES IN HEIGHT-DISTRIBUTION OF OZONE DURING WEATHER CHANGES

We have still too little direct data regarding the changes in the vertical distribution of ozone corresponding to the day-to-day variations of total ozone. But knowing the manner in which (1) these variations depend on the position of a station with respect to moving disturbances and (2) the vertical distribution corresponding to different ozone amounts at each station, we can build up synthetically a rough picture of the vertical distributions in the field of the disturbances. The basis for such a picture is provided by the composite diagrams of height distribution such as are given in *Fig. 2*. With increasing amounts of ozone, we may expect to find more and more ozone at levels below the level of maximum concentration. With large amounts of ozone like 0·340 or 0·400 cm at a place like Tromsö, the lower unit of high ozone concentration seems to come down to 8–10 km, while at a place like Delhi where the ozone amount rarely goes above 0·220 cm, only small quantities come down into the troposphere. As a companion to Götz's diagram of seasonal and latitudinal distribution of ozone, I have shown in *Fig. 6*, a diagram of the vertical distribution of ozone corresponding to different ozone amounts.

At Abu with its favourable skies for making ozone measurements, we have been able to take umkehr observations on many successive days, but since the day-to-day changes are small, there are generally no large differences in vertical distribution. There was, however, one instance in which umkehr observations were obtained on two alternate days with ozone amounts 0·188 cm and 0·209 cm. The distributions calculated by the usual method without correcting for secondary scattering are shown in *Fig. 7*. With the higher ozone content, there is more ozone between 18 and 27 km and in the lower atmosphere below 9 km.

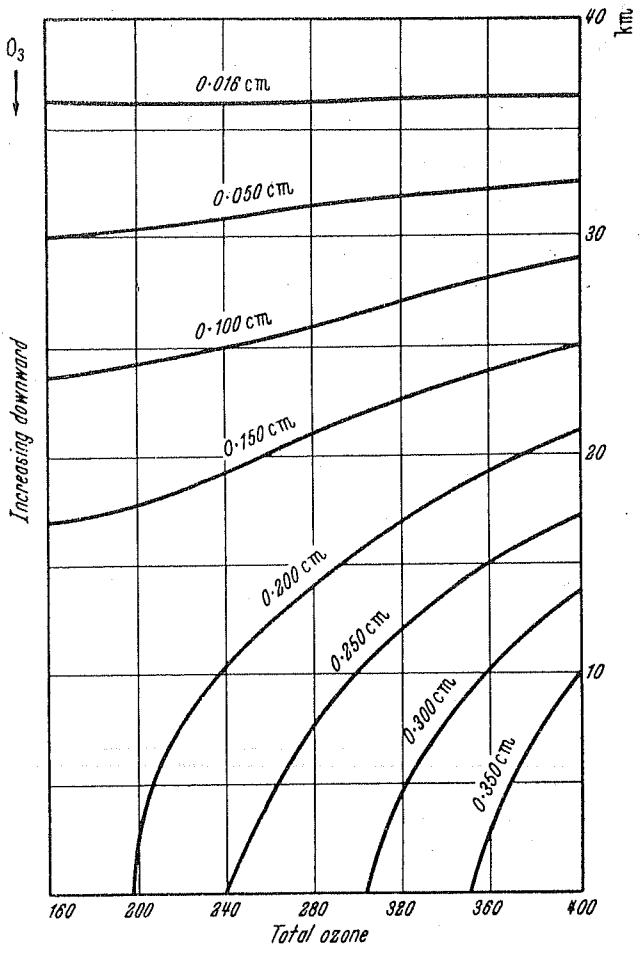

Figure 6. Generalized diagram showing vertical distribution of ozone for differing ozone amounts (from umkehr observations)

Observations on a series of days made by Mr. KULKARNI at Mount Abu in which there were gradual changes of ozone, either decreasing or increasing, have been used to find out how the height distribution depends on the total ozone amount. *Table 1* gives the results of grouping the observations according to the total amount (see also *Fig. 8*).

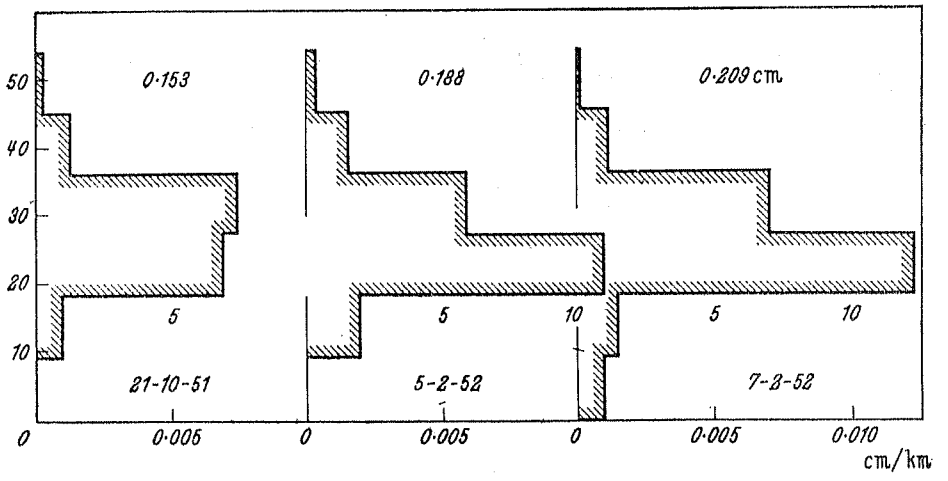

Figure 7. Mount Abu, vertical distribution of ozone for different ozone amounts

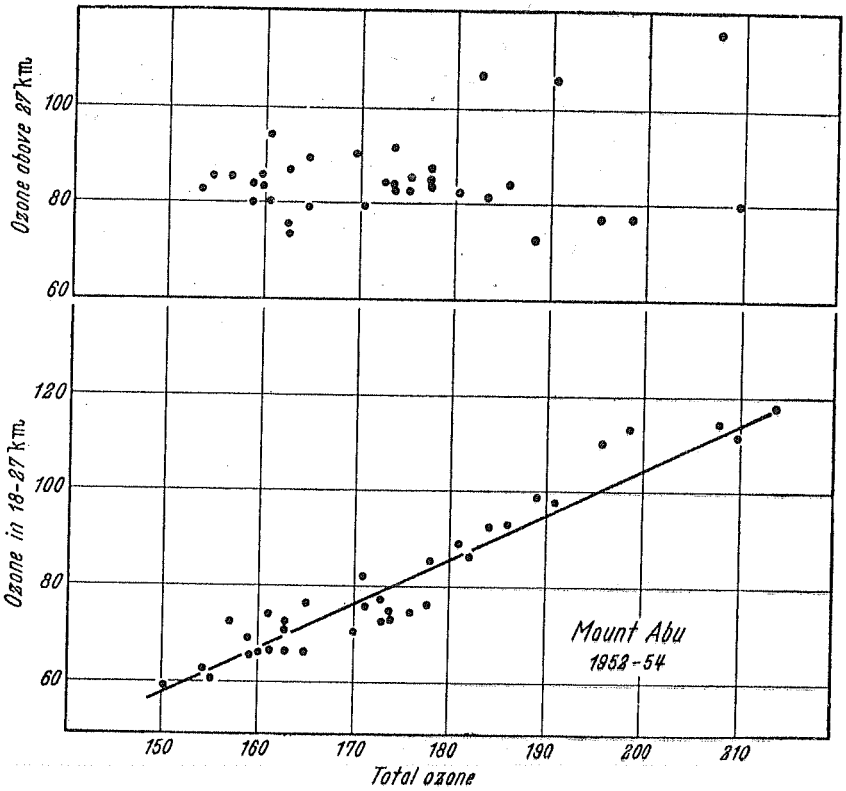

Figure 8

Table 1

Ozone Amount Group average cm	No. of days of observation	Average ozone amount in different layers in 10^{-3} cm/km				
		0–18 km	18–27 km	27–36 km	36–45 km	45–54 km
0·153	4	0·5	6·7	7·6	1·4	0·3
0·159	11	0·5	7·6	7·5	1·5	0·3
0·172	8	0·6	8·4	7·8	1·6	0·4
0·180	6	0·6	9·5	7·4	1·5	0·4
0·195	4	0·6	11·8	6·3	1·5	0·5
0·209	1	1·3	12·3	7·1	1·2	0·5

Most of the increase in ozone took place in the layer 18–27 km. This is also the conclusion to which KARANDIKAR and I[18] came after examining the Delhi observations. There was however a slightly greater amount of ozone at Delhi in 9–18 km. The division of the atmosphere into 9-km layers in the manner in which we have done is artificial and it is possible to find slightly alternative solutions which will fit the observed umkehr curves within the limits of experimental error. But the basic feature that, in the subtropical region, an increase of ozone, when it takes place, occurs mainly in the lower layers of the stratosphere below the level of ozone maximum will, I believe, not be changed.

OZONE IN THE LOWER LAYERS OF THE ATMOSPHERE

It has been known for a long time that there is a very small percentage of ozone present in the air near the ground. In recent years, this has been confirmed by optical methods by measuring the absorption of ultraviolet radiation by long paths of surface air. The mean monthly values of surface ozone obtained from a long series of observations at Montsouris, near Paris,

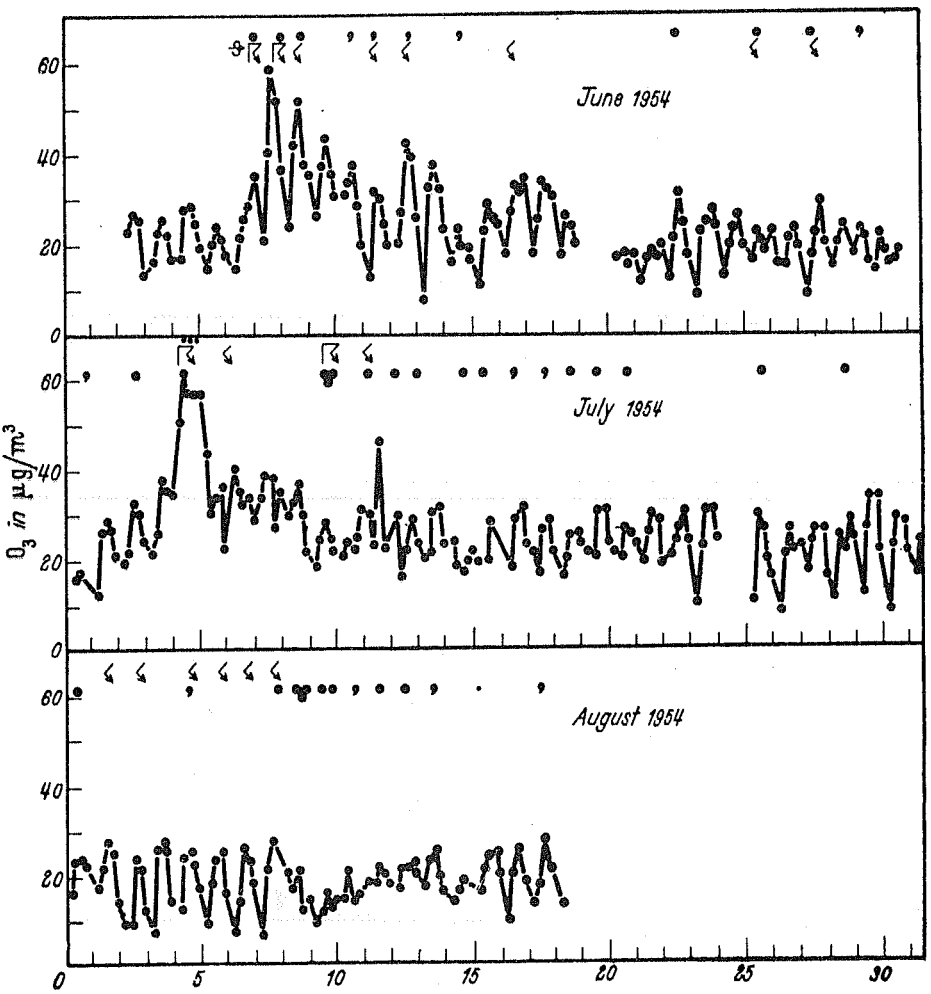

Figure 9. Surface ozone at Ahmedabad

collected by COLANGE and LEPAPE are given in Prof. FABRY's book on 'L'ozone atmosphérique'. It varies from 13$\mu g/m^3$ in November to 22$\mu g/m^3$ in May–June. In recent years, Dr. V. H. REGENER and Dr. EHMERT have greatly improved the chemical method and regular observations have been made at many places in Germany, at Arosa and in New Mexico. Prof. E. REGENER and Dr. Ehmert summarized the work done in recent years in the Brussels symposium on ozone in 1951. The following are the main facts:

In stagnant or slowly moving air, the ozone amount decreases to a low value in a few hours. In conditions of strong turbulence, there is a rapid increase in ozone. Usually, the surface ozone reaches a maximum at

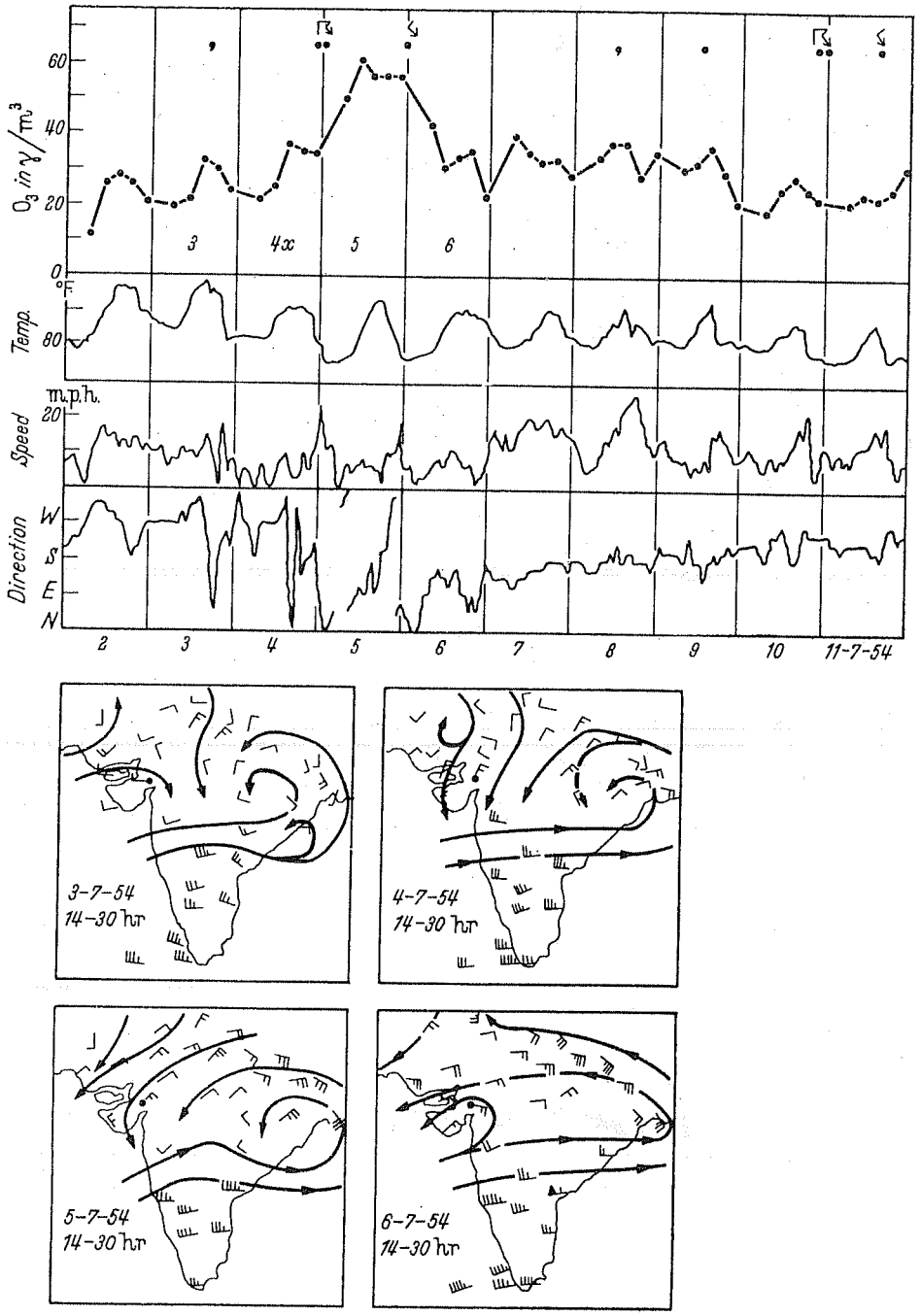

Figure 10. Surface ozone at Ahmedabad during 2–11.7.54. Also temperature (° F), wind speed (m.p.h.) and wind direction. Winds at 5,000 ft

14 to 15 h and a minimum in the early hours of the morning. At a mountain station like Arosa or Capillo Peak,[15] the ozone amount is larger than in a neighbouring lower-level station and the daily variation is smaller.

All this suggests that there is a downward transport of ozone at times of strong turbulence and that when the turbulence dies down, the ozone is destroyed by chemical or catalytic action.

In view of the small amount of ozone found in the lower atmosphere in the tropics, it was thought worth-while to obtain some observations in India and through the kind offices of the International Ozone Commission, an Ehmert apparatus was made available to us. Some of the results of observations made at Ahmedabad during the last three months by Mr. J. V. DAVE are shown in *Figs. 9* and *10*. The results are extremely interesting and

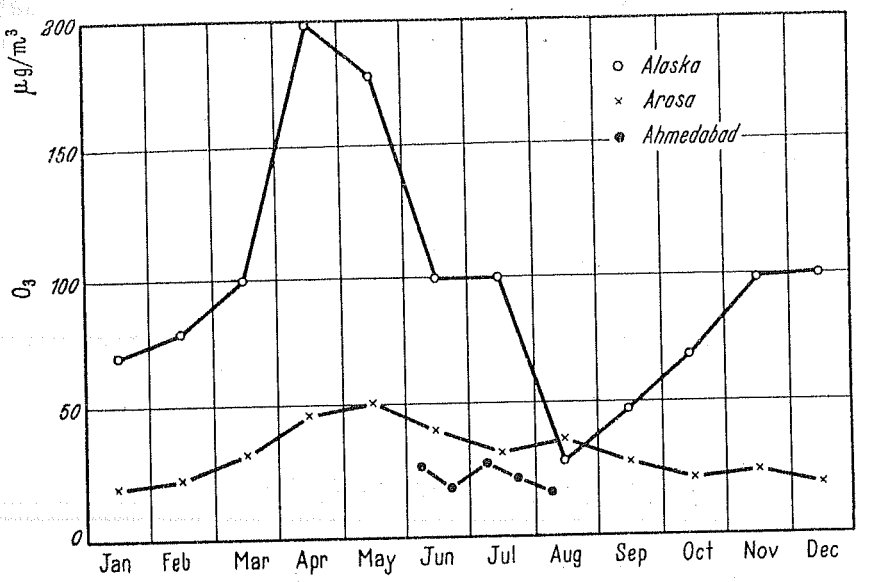

Figure 11. Monthly mean surface ozone at Alaska, Arosa and Ahmedabad

show that there is a substantial amount of ozone even in the lower atmosphere over the tropics of the order of 0·001 cm/km, that the surface ozone undergoes a regular daily variation, and there are occasions when owing to special weather conditions, substantially larger amounts of ozone are brought down. For comparison I have shown in *Fig. 11* the mean monthly values of surface ozone measured at Alaska by W. S. WILSON[19] and others, at Arosa by GÖTZ and VOLZ[20] and at Ahmedabad by Mr. Dave. The annual variation and the decrease in the amounts as we go from polar regions to the sub-tropics are very marked. The increase of ozone from August to November has been explained as being due to the fact that Alaska is snow-covered after September and Volz has shown that ozone undergoes comparatively little decomposition by coming in contact with ice.

MECHANISM OF OZONE CHANGES AND ATMOSPHERIC CIRCULATION

Day-to-day variations

A photochemical theory of atomic oxygen and atmospheric ozone was outlined by CHAPMAN in 1930. Since then, many authors have worked on the photochemistry of atmospheric ozone, MECKE, WULF and DEMING, SCHRÖER, DÜTSCH[21] and CRAIG.[22]

Oxygen atoms are produced by the absorption of sunlight of wavelengths shorter than 2420 Å; these atoms combine with oxygen molecules to form ozone. Ozone is destroyed by absorption of sunlight below 11,800 Å.

Under these two contrary actions, a certain equilibrium distribution of ozone will be established even in a stagnant atmosphere depending on the zenith distance of the sun. In *Fig. 12* are shown the equilibrium vertical distributions of ozone for a few different latitudes corresponding to summer and winter according to the calculations of Dütsch. The total equilibrium ozone amount decreases with increasing zenith distance of the sun. According to a pure photochemical theory, there should be more ozone over equatorial regions in all seasons, and in middle and high latitudes, more ozone in

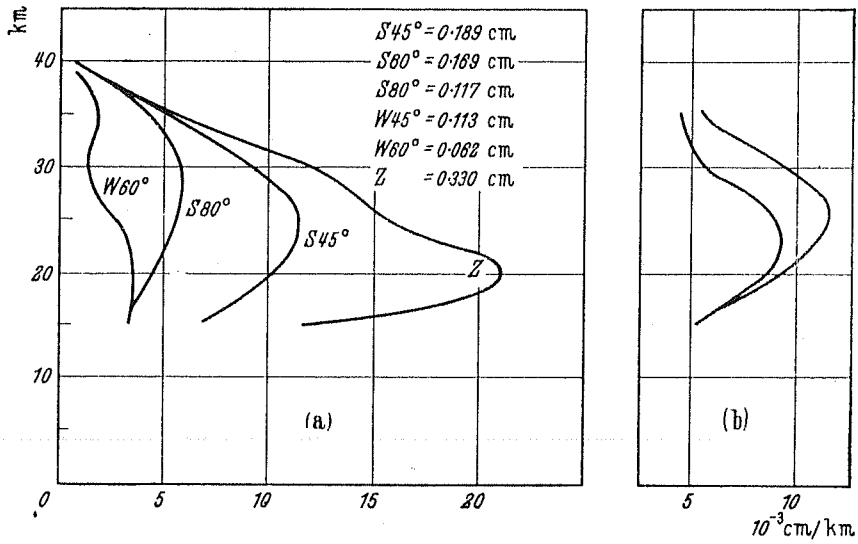

Figure 12 (a). Photochemical equilibrium ozone (Dütsch) (Z = Zenith Sun; S45 = Summer at lat. 45°; W60 = Winter lat. 60°)
(b). Non-equilibrium max. and min. ozone at 45°

summer than in winter. The equatorial maximum should also lie at a lower level. In the actual atmospheric problem, not only is the ultimate equilibrium value of ozone important, but also the rate at which the equilibrium is approached if it has not been attained or has been disturbed. It appears that the time required to restore half-equilibrium when it is disturbed is of the order of a few hours at 35 km, a few days at 30 km and a few weeks at 20–25 km. At levels below 30 km, the actual ozone amount present at any time cannot therefore correspond to the amount in equilibrium with the radiation existing at the time. On the right-hand side of *Fig. 12*, the maximum and minimum non-equilibrium values of ozone during the year as calculated by Dütsch are also given for the latitude of 45°. While photochemical action by itself cannot explain the observed distribution of ozone and its short period changes, it is sufficient to provide all the ozone that we observe, provided there is some agency capable of transporting the ozone with sufficient rapidity from where it can be quickly formed to places where it can be preserved. All those who have worked on the vertical distribution of ozone are agreed that the ozone changes which take place from day to day occur mainly below 25 km, that is, in the region where photochemical formation of ozone is far too slow to account for the rapidity of the changes.

Dobson's composite diagrams of ozone distribution in the field of atmospheric disturbances, Meetham's correlations and Normand's curve parallels are all consistent with our present ideas of the aerology of extra-tropical disturbances. The ozone amount increases behind upper air troughs, because there is converging air flow in the upper troposphere and lower stratosphere from regions where there is a larger amount of ozone. This flow is accompanied by a descending component of motion with a lowered tropopause and higher stratospheric temperatures. Sometimes, the lowering of the tropopause is so great that the old tropopause is obliterated and a new one forms at a lower level. Stratospheric air can then stream into the troposphere and become incorporated in it. Since, in general, the ozone mixing ratio increases with height up to 30 km, the descent of the tropopause means increased ozone amount. Similarly in the ridge between two troughs, the tropopause is high and the air flow in the upper troposphere is from lower latitudes where the ozone concentration is lower. As shown by Mr. SAWYER[23] in his valuable studies of tropopause variations, the tropical tropopause often folds over in the advancing part of the high-level ridge, descends, and gradually disappears.

W. J. REED[24] has shown that advection together with vertical motion, as first suggested by PALMEN, can account in a reasonable manner for the observed day-to-day variations of ozone in middle latitudes. There is general agreement that, without downward vertical movement, it would not be possible with advection alone to explain either the larger variations of ozone in winter and spring or the variations in summer and autumn. Langlo[5] has quoted an instance in March 1948 in which, associated with a deep closed depression with a low tropopause at about 6 km, the ozone amount increased from 0·220 cm to 0·330 cm.

The essential part of the story is that, as clearly brought out by BJERKNES and PALMEN[25], the moving upper waves of extra-tropical latitudes, with which lower level fronts are often coupled, not only exchange air between subtropical and polar regions, but have associated with them a definite scheme of vertical air movements, bringing stratospheric air into the troposphere and injecting tropical tropospheric air into the middle latitude stratosphere. FLEAGLE'S[26] diagrams of horizontal convergence and vertical movements associated with travelling troughs and ridges are well known. I reproduce a diagram (*Fig. 13*) by PALMEN and NAGLER[27] in which they have shown the vertical sections along the 37,800-ft contour of the 200-mb surface associated with a deep polar trough in the U.S.A. in February and the two adjacent high-level ridges. The upper boundaries of the polar air, the tropopauses, and a few isentropes are drawn. The isentropes are approximate stream-lines in the lower stratosphere and show clearly the interpenetration of stratospheric and tropospheric airs. The 37,800-ft contour meanders between 55° N and 30° N. The slopes of the isentropes near the 200-mb surface give an idea of the vertical displacement of the air in moving from ridge to trough and from trough to ridge. In 12 hours, the vertical displacement was about 70 mb or 2 km and the average vertical velocity about 4 cm/sec.

Palmen and Nagler point out that in addition to the general subsidence on the west side of a trough line shown in Fleagle's diagrams, 'there must be

another motion which intensifies the subsidence in the lower stratosphere and weakens it somewhat in certain regions of the troposphere'. When in an eastward moving disturbance, the polar front is sloping upward from SE to

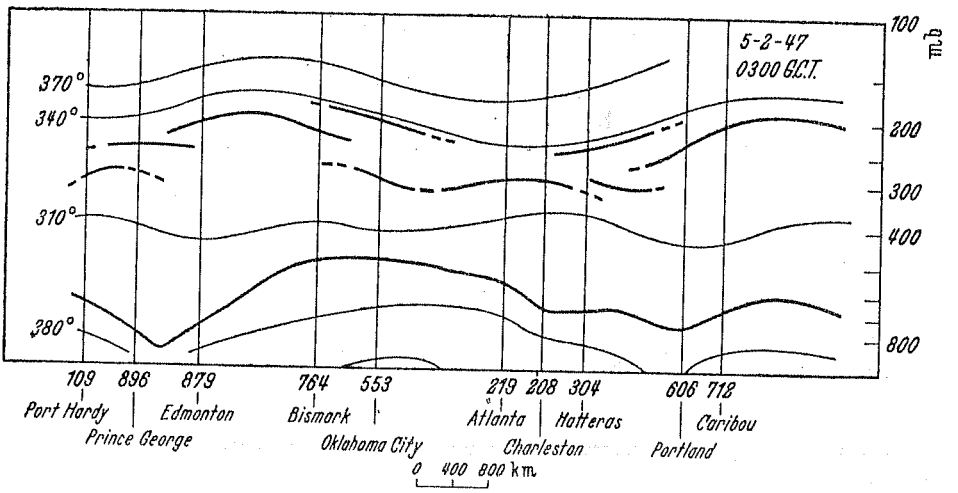

Figure 13. Cross-section along 37,800 ft contour of 200 mb surface showing isentropes, tropopauses and upper boundary of frontal layer—(Palmen and Nagler)

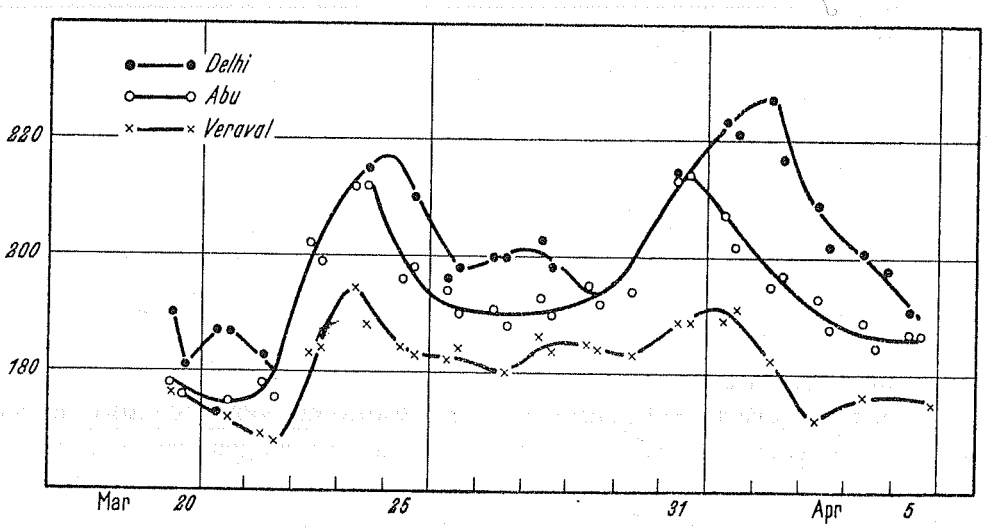

Figure 14 (a). Ozone variations at Delhi, Abu and Veraval, March 20–April 5, 1954

NW, the tropical upper tropospheric air ascends and gets into the stratosphere above the lower tropopause of the polar air. Similarly when the polar front is pointing from SW to NE, there is a sinking movement of the tropical substratosphere and some of the polar stratosphere air gets into the upper troposphere on the southern side of the polar front. There is little doubt that the upper vertical circulations which are normally active from 400 mb to 150 mb in the region of extra-tropical cyclones play a very important role in bringing down ozone from the lower stratosphere into the troposphere.

The ozone variations in N. India associated with winter disturbances suggest that similar exchanges of air between stratosphere and troposphere must be taking place in the subtropical jet-stream region between the tropical tropopause and the lower middle latitude tropopause. See *Figs 14 (a)* and *14 (b)* showing ozone variations at Delhi, Abu and Veraval on March 20 to

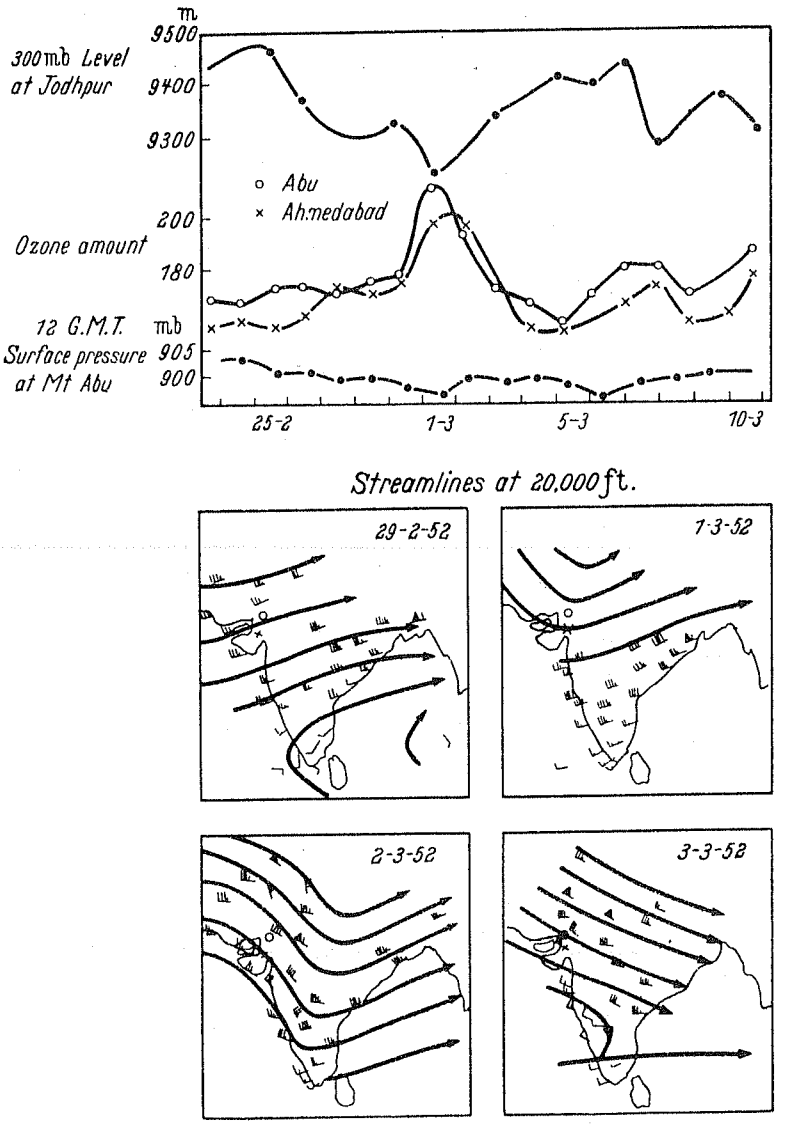

Figure 14 (b). Ozone at Mount Abu and Ahmedabad, surface pressure at Abu, and 300 mb level at Jodhpur

April 5, 1954 and at Abu and Ahmedabad on February 29, to March 3, 1952 respectively. We do not have sufficient upper air data at high levels to work out tropopause contours or stream lines above 300 mb. But from the seasonal and day-to-day variations of ozone at Delhi, Abu and Ahmedabad, it is clear that so long as the subtropical jet-stream remains well to the north of Delhi, there is very little change in the ozone amount and that the increase of ozone in N. India in winter is associated with the formation of double

tropopause over N. India and a leakage of middle latitude stratospheric air into the N. Indian troposphere. The much larger ozone amounts observed by Mr. FOURNIER D'ALBE and his collaborators at Quetta in Pakistan and Mr. ABDUL KHALEK[28] in Afghanistan may be partly explicable by the more frequent occurrence of lower tropopause at those stations. Calibrated, inter-compared instruments are, however, necessary before decisive conclusions can be drawn.

Latitudinal and seasonal variations

The outstanding features of the latitudinal and seasonal variations of ozone are shown in an expressive way in *Fig. 15.* This diagram differs somewhat

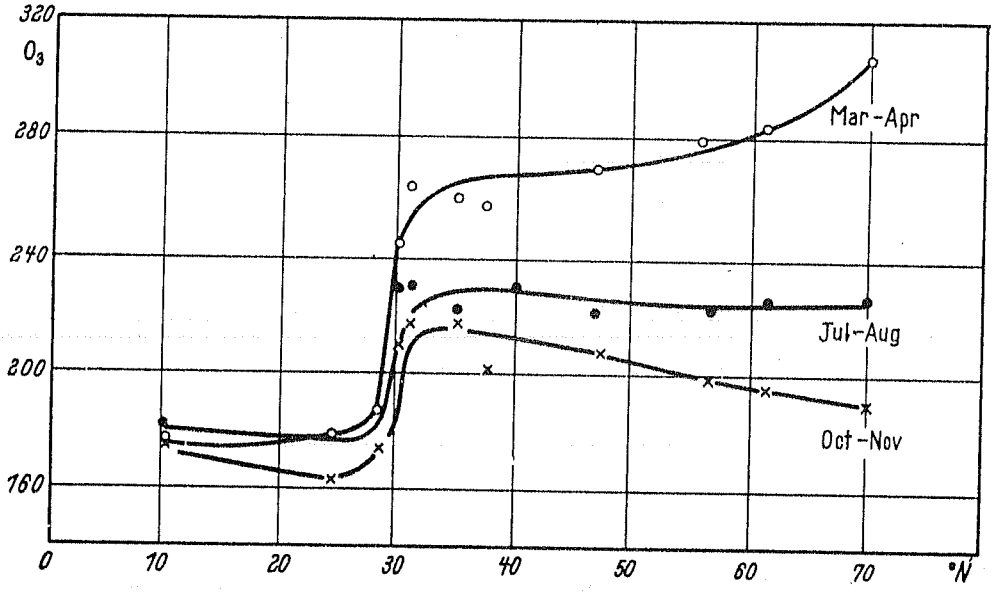

Figure 15. Latitudinal distribution of mean ozone amounts in different seasons

from Dobson's classical diagrams, but the difference is due to the inclusion of Indian data for latitudes up to 28½° N and the data of Helwan, Zi-ka-wei and Azores. Long-period averages of the European stations have been used. The ozone data are no doubt non-homogeneous and for very unequal periods but there does seem to be a discontinuity in ozone amount at about 30°, a fact on which Langlo has already commented. This diagram should be compared with a diagram showing the tropopause levels at different latitudes. I have reproduced in *Fig. 16* a diagram due to the London Meteorological Office. Comparing these two and bearing in mind the variations of ozone at Delhi during winter it seems difficult to avoid the conclusion that the steep increase in ozone amount to the north of 30° is associated with the steep lowering of the tropopause in the same direction. We have seen that, in middle latitudes, a lowering of the tropopause goes with increase in ozone amount; this association is also apparently true for the tropopause funnels investigated by Palmen and others as well as for the transition from the tropical to the temperate latitude tropopause. The thicker the stratospheric layer between the level of maximum ozone concentration and the tropopause the greater is its ozone-holding capacity.[29]

How much ozone is collected in that reservoir in a certain time depends on the rate at which it is filled and the rate at which ozone leaks out or is decomposed. In the cold dry dust-free air of the lower stratosphere, decomposition is very slow. We are therefore concerned with finding out how this reservoir can be filled or discharged.

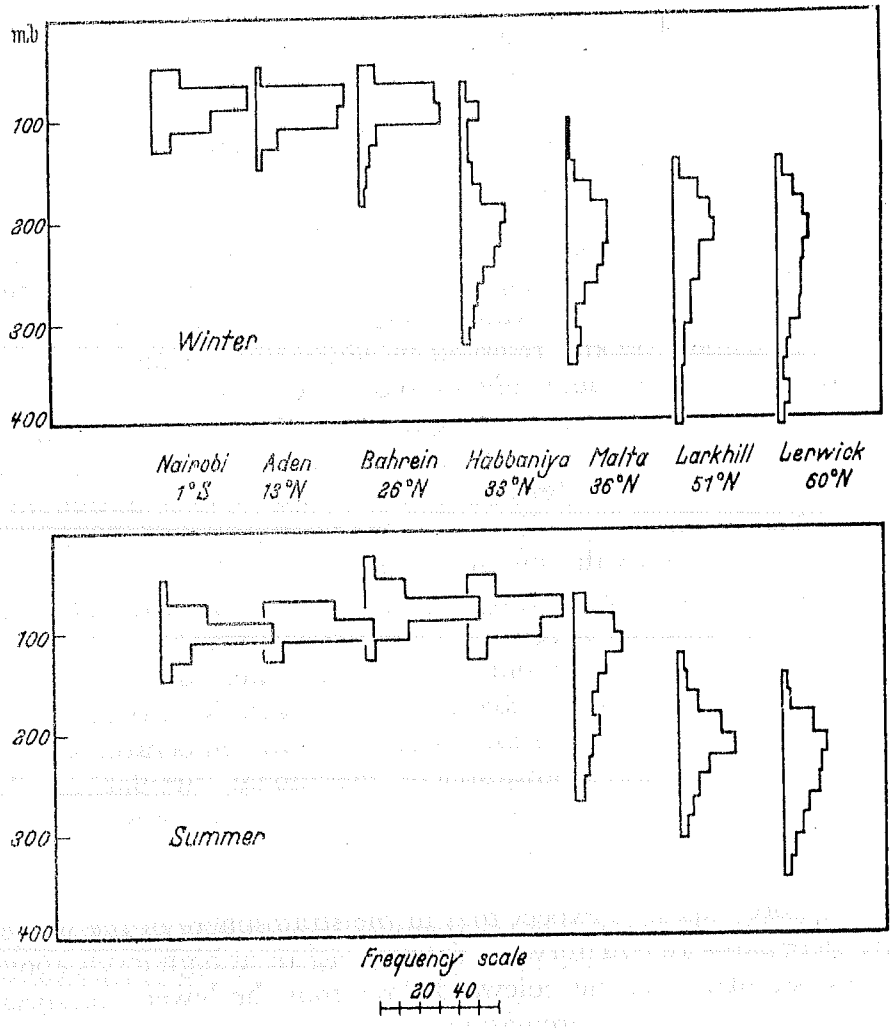

Figure 16. Percentage frequencies of tropopause pressures (London M.O.)

The average tropopause has a height varying from 17 km to 9 km with a steep drop between 16 km and 11 km and yet other breaks near polar fronts. Although the air in the stratosphere between 9 and 17 km is highly stratified, the existence of the tropical upper troposphere with its upper pool of cold air and of the subtropical jet-stream at about 12 km causes large-scale horizontal movements with pronounced vertical components. The tropical air streaming out into the stable stratosphere of higher latitudes will retain its individuality for much longer distances and times than the middle latitude stratospheric air which descends into the tropical troposphere. That means the tropical tropopause will carry with it the low frost point characteristic of its temperature[30] and high ozone content of the lower tropical stratosphere.

It is difficult to say anything precise about the depth of the stratospheric layer which will participate in this south–north movement but from the known temperature distribution in the stratosphere, the outflow for the tropics must extend to a height of 22–24 km when it starts. This must carry to higher latitudes the richer ozone content of the lower stratosphere of the tropics.

Thus, there are features in the circulation of the upper troposphere and lower stratosphere which give opportunities for stratospheric air to get rapidly into the troposphere. Weak spots often occur in the tropopause near the upper parts of polar fronts and also on the polar side of subtropical jets. Such opportunities are fewer and feebler in summer.

We know, however, that the vertical movements connected with the vorticity concentrations in the upper troposphere are rapidly enfeebled as we ascend the stratosphere, and so we cannot expect tropospheric activity to affect the atmosphere in the ozone regeneration region above 30 km. There must be another upper circulation which brings down ozone from the ozone regeneration region above 30 km. reservoir in winter and spring.

In the winter stratosphere, there are two cold-air sources, one immediately above the tropical tropopause up to a height of 20–22 km and a second one in the polar stratosphere well above 20 km. Upper winds at Lerwick (60° N), Larkhill (51° N) [31] and New Jersey (40° N) [32] show minimum strength of westerlies at about 20 km and steadily increasing westerlies up to 30 km and more. This is consistent with the supposition that the air in the polar stratosphere is colder than the air at the same levels over middle latitudes.

Many years ago, WHIPPLE suggested a meridional distribution of temperatures in the stratosphere with cold air over the winter polar region and warm air over the summer pole. Recently, Dr. A. H. R. GOLDIE[33] has used the later measurements and estimates of upper air temperatures up to 48 km to sketch a tentative stratosphere meridional circulation. Its essential features are a strong west wind circulation round the winter pole increasing with height above 20 km and weaker easterly circulation in the summer hemisphere.

There is therefore reason to expect that in the stratosphere of the winter hemisphere, there exists an equatorward flow of cold air at high levels above 25 km. This, together with the poleward flow from the lower equatorial sub-stratosphere must lead to a circulation of the type shown in *Fig. 17* with upper stratospheric air descending over middle and lower latitudes.

As shown by the upper wind measurements over Lerwick and Larkhill, the upper stratospheric westerly circulation persists from November till April and is strongest from January to March. There must be associated meridional movements of a wave-like character.

The effect of a high-level polar circulation, the upper arm of which extends into the region of photochemical ozone formation and the lower arm into the region of ozone conservation, would be to enrich the ozone content of the air. It is not surprising that the effect of this enrichment is most clearly visible in March–April in high and middle latitudes when the enrichment process has led to an accumulation of ozone in the protected region.

The equatorial storage reservoir of ozone above the tropical tropopause is wider in area but of shallower depth. Its replenishment and emptying

goes on throughout the year, but presumably at a faster rate in winter when the subtropical jet-stream is strongest. The seasonal increase of ozone in winter and spring is thus largely an effect of (1) more effective ozone preservation in the lower stratosphere and troposphere and (2) the existence of an upper high-level meridional circulation between high and middle latitudes. The warming up of the summer stratosphere over polar regions can have only the effect of weakening the upper circulation. The replenishment from above will fail and the ozone amount will gradually decrease due to the distinctive action of radiation and of chemical activity in the troposphere.

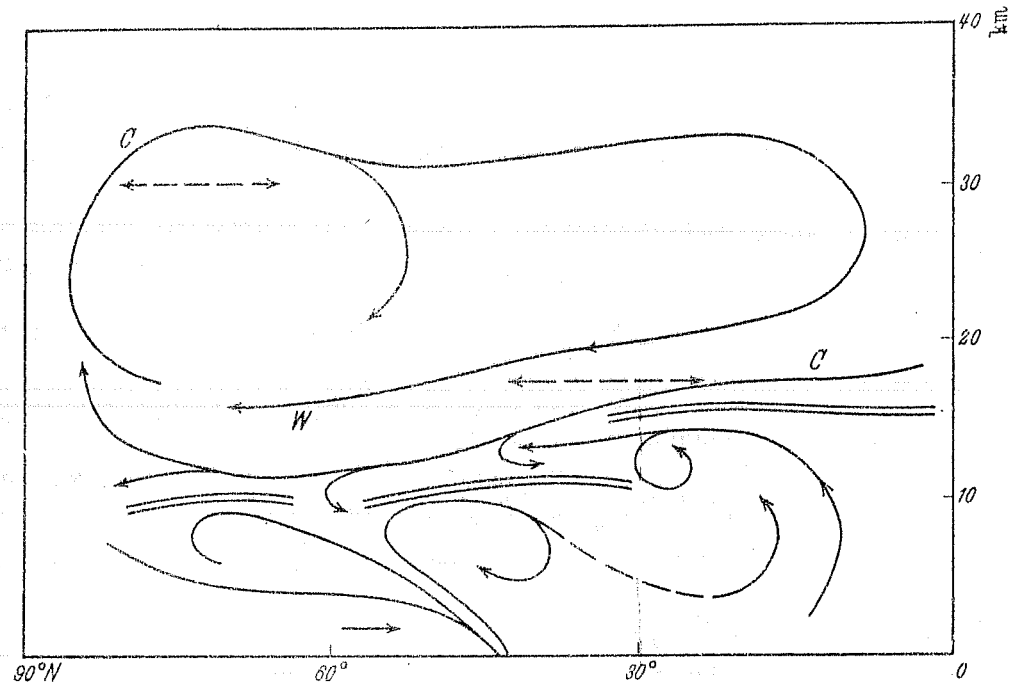

Figure 17. Idealized winter meridional circulation over northern hemisphere

Götz and Tonsberg and Langlo have commented on the year-to-year variations of the ozone amounts over Arosa and Tromsö. Similar behaviour has been noted at other places. When we have standardized ozone measuring stations at selected places over the earth, the data will no doubt provide a valuable index of the activity of the general circulation in the upper troposphere and lower stratosphere.

The study of atmospheric ozone has opened up a new means not only of following atmospheric movements in the upper atmosphere, but also of testing schemes of general circulation in the stratosphere up to 40 km. A more extensive regional network now seems to be called for with special attention to observations on either side of the subtropical *jet-stream* with inter-compared instruments. Vertical distribution data on individual days are still too few. Side by side with them, efforts have to continue to obtain quantitative information about other possible agencies which may produce or destroy ozone in the atmosphere.

I have not touched, or touched very briefly, on many questions of importance. As you are aware, we are going to have a discussion on the

16th on 'Atmospheric Ozone and the Circulation in the Stratosphere' under the chairmanship of Prof. Dobson, our leader in ozone studies. We hope to have some questions elucidated there by workers from different parts of the world.

Ozone studies in India have been made possible by the whole-hearted co-operation of the India Meteorological Department and of the Council of Scientific and Industrial Research in India, to both of whom I express my heartfelt thanks.

REFERENCES

1 U.G.G.I. Association of Meteorology, Memoirs and Discussions, Lisbon (1933)
2 REGENER, E. and V. H., *Phys. Z.* 35 (1934) 788
3 GÖTZ, F. W. P., MEETHAM, A. R. and DOBSON, G. M. B., *Proc. roy. Soc.* A 185 (1934) 144
4 EHMERT, A., *Z. Naturf.* 4b: 321
5 LANGLO, K., *Geofys. Publ., Oslo* 18, No. 6 (1951)
6 WALTON, G. F., U.G.G.I. Association of Meteorology, Memoirs and Discussions, Brussels (1951)
7 KAY, R. H., *Rocket Exploration of the Upper Atmosphere*, 202, Pergamon Press, London, 1954
8 PAETZOLD, H. K., *J. atmosph. terr. phys* 2 (1952) 183
9 REGENER, E., PAETZOLD, H. K. and EHMERT, A., *Rocket Exploration of the Upper Atmosphere*, 202, Pergamon Press, London, 1954
10 JOHNSON, F. S., PURCELL, J. D. and TOUSEY, R., *Rocket Exploration of the Upper Atmosphere*, 189, Pergamon Press, London, 1954
11 FABRY, C., *L'Ozone Atmosphérique*, C.N.R.S., Paris, 1950
12 For a summary, see DOBSON, G. M. B., BREWER, A. W. and CWILONG, B. M., *Proc. roy. Soc.* A 185 (1934)
13 MEETHAM, A. R., *Quart. J. R. met. Soc.* 63 (1937) 289
14 MOSER, H., *Ber. dtsch. Wetterdienstes U.S. Zone*, 11 (1949)
15 PENNDORF, R., *Ber. dtsch. Wetterdienstes U.S. Zone* 11 (1949)
16 NORMAND, C. W. B., *Quart. J. R. met. Soc.* 79 (1953) 39
17 DEWAR, D., *Met. Mag. Lond.* 80 (1951) 823; also FROST, R., *Prof. Notes met. Off. Lond.* (1953) 107
18 RAMANATHAN, K. R. and KARANDIKAR, R. V., *Proc. Indian Acad. Sci.*, 29 (1949) 330
19 WILSON, W. S., *et al.*, *Trans. Amer. geophys. Un.* 33 (1952) 361
20 GÖTZ, F. W. P. and VOLZ, F., *Z. Naturf.* 6a (1951) 634
21 DÜTSCH, H. W., Inaugural Dissertation, Zurich (1946); also 1953; U.G.G.I. Association of Meteorology, Memoirs and Discussions, Brussels (1951)
22 CRAIG, R. A., *Amer. met. Soc. Monographs*, I. 2 (1953)
23 SAWYER, J. S., *Geofys. Mem. Lond.* 92 (1954)
24 REED, W. J., *J. Met.* 7 (1950) 263; 10 (1953) 296
25 BJERKNES, J. and PALMEN, E., *Geophys. Publ., Oslo*, 12, No. 2 (1937)
26 FLEAGLE, R. G., 1947, *J. Met.* 4 (1947) 165
27 PALMEN, E. and NAGLER, K. M., *J. Met.* 5 (1948) 58
28 ABDUL KHALEK, *C.R. Acad. Sci., Paris* 236 (1953) 2424
29 RAMANATHAN, K. R. and KULKARNI, R. N., *Proc. Indian Acad. Sci.* A 37 (1953) 321
30 BREWER, A. W., 1953, U.G.G.I. Association of Meteorology, Memoirs and Discussions, Brussels (1951)
31 SCRASE, F. J., *L. J. Roy. met. Soc. Lond.* (1951)
32 BRASEFIELD, C. J., *J. geophys. Res. Washington* (1954)
33 GOLDIE, A. H. R., *Centenary Proc. R. met. Soc.* 175 (1950)