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Water Vapour and Ozone in the Atmosphere and Stratospheric Circulation*

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1. Water vapour in the stratosphere

Most of what we know about water vapour in the stratosphere has come from the development of the frost-point hygrometer by Dobson, Brewer and Cwilong (1946) and its regular use in the Meteorological Research Flights of the U.K. Meteorological Service. Additional information is available for the study of mother-of-pearl clouds in Scandinavia and Russia, some measurements made in U.S.A. with balloon-borne frost-point hygrometer, and a few aircraft measurements of infra-red absorption by water vapour in U.K. and U.S.A.

The existing knowledge is summarised in the following two diagrams—

Fig. 1. A histogram of frost-points measured at 48,000 ft over U.K. Taken from a paper on "Further observations of humidity up to 50,000 ft made from an aircraft" by N. C. Helliwell *et al.* in *Quart J. R. met. Soc.*, **83**, 1957, p. 257.

Fig. 2. A diagram showing temperatures and frost-points at 34,000 ft and 46,000 ft in July 1956 obtained by Meteorological Research Flights extending from 40° to 65°N at about the longitude of U.K. (*Weather*, Sep. 1958).

Reference may also be made to a paper on "Humidity in the Stratosphere at 27 km" by F.R. Barclay *et al.* in the *Quart. J. R. met. Soc.*, **86**, 1960 where they found a frost-point of —76°C.

The Meteorological Research Flights of U.K. have established that while the frost-point *at the tropopause* is variable over the U.K. from —50° to —78°C, it decreases rapidly with height above the tropopause, and becomes practically constant at about —83°C at a level of 15 km (Mixing ratio 2×10^{-3} gm/kgm). They have also shown that at the same height (15 km) over Idris in Morocco, a similar value of the frost-point is observed. The special meridional flights undertaken by them have shown that while at low stratospheric levels, the frost-point depends on the synoptic weather situation, at 15 km, the latitudinal variation of humidity becomes small, although there are marked discontinuities of frost-point on crossing fronts with lower frost-points towards the north.

The infra-red observations made by Houghton over U.K. show an average mixing ratio of 3×10^{-3} gm of H_2O/kgm of air above 13·5 km. This is in reasonable agreement with the frost-point hygrometer findings.

On the other hand, a few American soundings made in 1949-50 and the indirect evidence of mother-of-pearl clouds over Norway indicate higher frost-points at 23-27 km. The question is : Is the very low frost-point of 190°K confined to a restricted depth in the lower stratosphere or can it be considered to extend throughout the stratosphere?

The conclusion drawn by Brewer and Dobson (1946) from the persistent extreme

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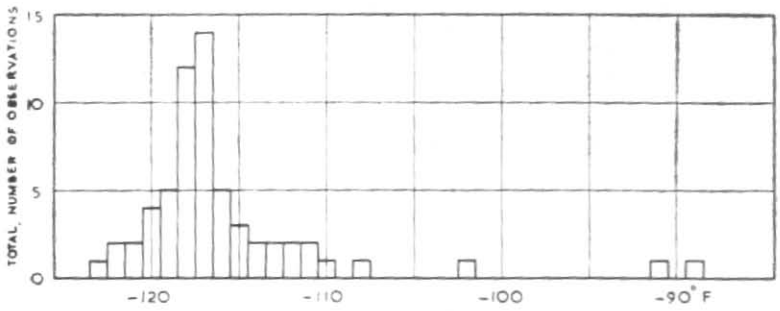
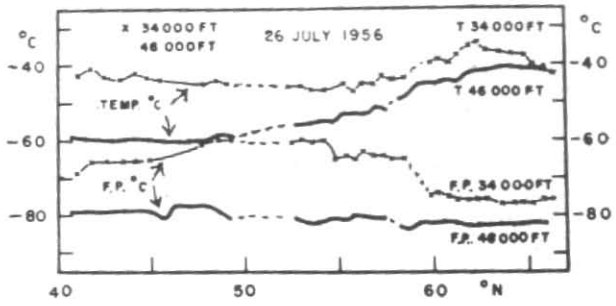


FIG. 1 HISTOGRAM: FROST POINTS AT 48,000 FT OVER U.K

(HELLIWELL & OTHERS)

Q.J. ROY. MET. SOC. 1957



TEMPERATURES AND FROST POINTS AT 34000 AND 46000 FT. ON 26 JUL. 1956, ROUGHLY ALONG U.K. MERIDIAN

FIG. 2 (a)

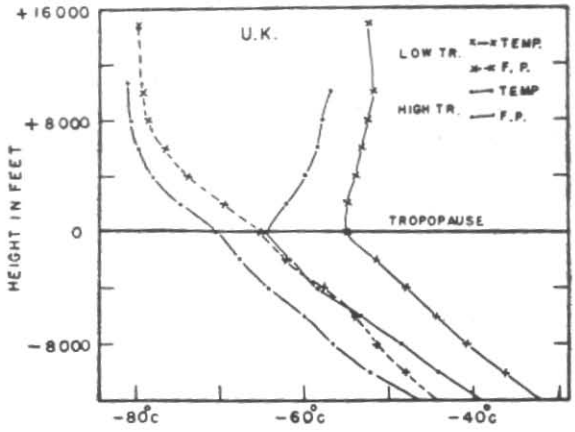


FIG. 2(b). TEMPERATURES AND FROST POINTS IN LOWER STRATOSPHERE OVER U.K. (AFTER HELLIWELL)

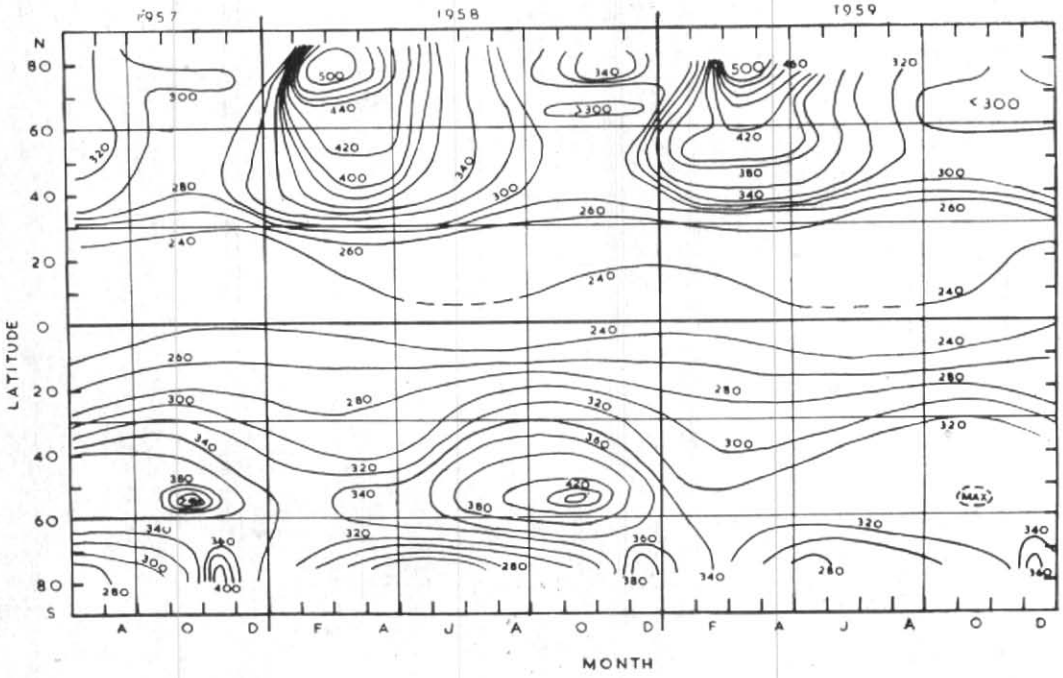


FIG. 3. OZONE DISTRIBUTION IN DIFFERENT MONTHS FROM 1957 TO 1959

dryness of air at 15 km over U.K. is well-known, namely, it provides evidence of a meridional circulation involving ascent of air over equatorial latitudes across the tropopause and slow sinking of the dried air in the lower levels of the stratosphere over middle and polar latitudes.

Umkehr studies and recent ozone observations made by Brewer and Milford (1960) over Malta, Arosa, Liverpool and Tromso in summer provide supporting evidence for the transport of stratospheric air in the indicated direction.

2. Atmospheric Ozone and its variations

Before the IGY, 55 ozone stations had been equipped with Dobson instruments and 13 others with instruments of other types. Professor Dobson and Sir Charles Normand undertook the task of preparing detailed instructions for the use of the Dobson instrument and monitoring the ozone observations during the IGY. Nearly 40 stations actually took ozone observations, and through the kindness of various authorities, the author has been able to get the provisional daily ozone data of about 36 stations. Umkehr observations are also available from a few stations.

Diagrams illustrating some important points about the distribution of ozone and its variations are given below—

(1) Latitudinal variations of total ozone (1957—1959) in different months (Fig. 3).

It may be noted that the mean monthy ozone amount is not a function of the latitude only, but that there are regional differences. For example, in the Indian monsoon region, the ozone amounts are abnormally low, while according to London, there are three ozone ridges in the northern hemisphere, one over eastern N. America, one over Central Europe and a third over W. Pacific. The large and rapid increase in ozone amount over the

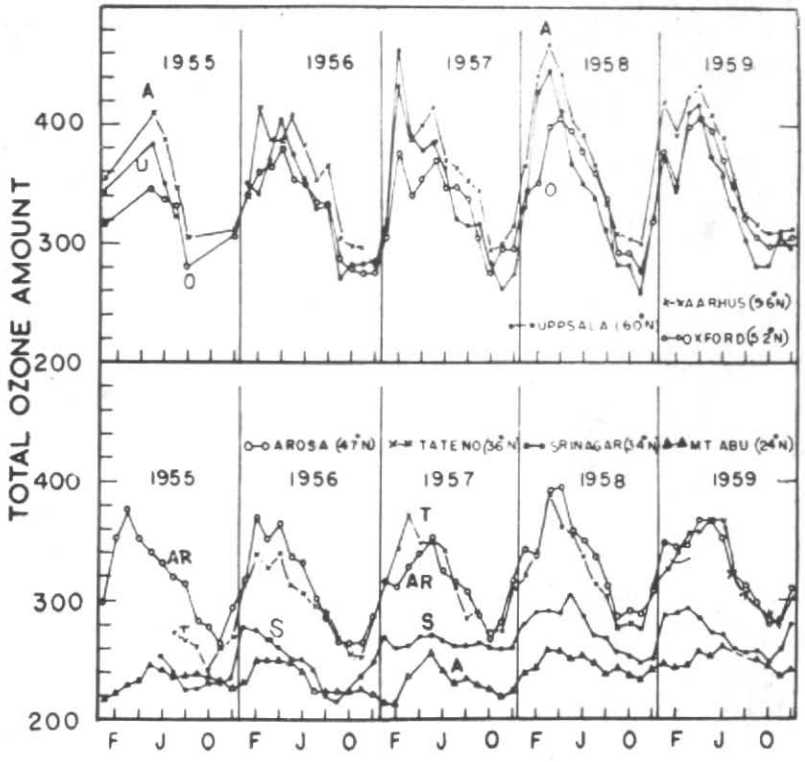


FIG. 4. MONTHLY MEAN OZONE AMOUNTS AT SELECTED STATIONS (1955-1959)

Arctic at the end of the polar night is now a subject of intensive study.

(2) Monthly mean ozone amounts at selected stations—

Uppsala (60°N), Aarhus (56°N), Oxford (52°N), Arosa (47°N), Tateno (36°N), Srinagar (34°N) and Mt. Abu (24°N) (1955—59)—Fig. 4.

Please note the unusually large values at Aarhus and at Tateno (Japan).

(3) Percentage frequencies of occurrence of total ozone amounts at selected stations in March and October—(Tromso, Oxford, Arosa, Tateno, Srinagar and Mt. Abu)—Fig. 5.

The day-to-day variations of ozone amount are largest in Tromso and least in Mt. Abu. Note the large variations in Tateno in March compared to Srinagar.

(4) Rapid rise of ozone is second half of winter in the Arctic (Fig. 6).

(5) Comparison of monthly mean amounts at Resolute (75°N) and at Halley Bay (75°S) (MacDowall 1960) (Fig. 7).

The observations are centered round the *respective* winter solstices.

In the Antarctic, the post-winter rise of ozone is delayed till summer, and the rise is markedly weaker.

Before and during the IGY, special attention was devoted by a few investigators to observations and study of the vertical distribution of ozone. In addition to the Umkehr

method which involves the measurement of zenith sky intensities in the ultra-violet between 3324 and 3055 Å at low altitudes of the sun, other methods have been developed. Of these, the method of infra-red spectroscopy which requires the measurement from the ground of the absorption caused by the atmosphere of the solar radiation in the band 9·6 μ and of the emission of the same radiation from the sky at different zenith distances of the sun has been developed by Adel and Epstein, Goody, Walshaw and Roach, and by Migeotte and Vigroux. According to Vigroux, the method is capable of fixing with some precision the level of maximum ozone concentration. Methods using an optical filter in the Hartley region of the spectrum with a photocell carried in a balloon has been developed by Paetzold and by Madame Vassey and Rasool. Paetzold's instrument has been used in Germany, Switzerland and in Congo. But perhaps the most significant new work in this field is the development of a light balloon-borne chemical ozone sounde by Dr. Brewer. The instrument measures and signals the current developed between platinum electrodes or between a platinum electrode and a silver electrode in a solution of KI over which ozonised air has been passed. The method can operate continuously and can measure concentrations of ozone of the order of 1 in 10^9 in 20 seconds. Brewer's instrument with its quick response to changes, has been found capable of showing up details of ozone distribution with height. For example, there is, almost invariably, a sharp increase of ozone observed as the balloon enters the stratosphere. But more work seems to be needed to obtain reliable absolute values by this method. The type of results obtained from Brewer sondes is shown in Mr. MacDowall's diagram showing temperature and ozone distribution over Halley Bay (75° S) in summer and winter (Fig. 8).

In July-August 1958, an inter-comparison of the following four methods of determining the vertical distribution of ozone was carried out at Arosa (Brewer, Dutsch *et al.* 1960).

(*a*) Umkehr method (Perl and Dutsch),

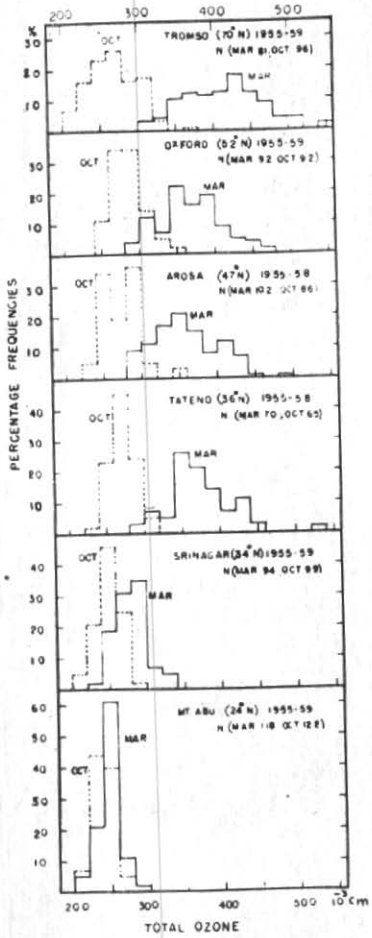


FIG. 5. PERCENTAGE FREQUENCIES OF OCCURRENCE OF TOTAL OZONE AMOUNTS AT SELECTED STATIONS. (MAR. & OCT.)

(*b*) Infra-red spectroscopy (Migeotte-Vigroux-Pastiels),

(*c*) Optical Filter-method (H.K. Paetzold), and

(*d*) Balloon-borne chemical radiosonde (A.W. Brewer).

Examples of the comparison are shown in Fig. 9. From an examination of these results, Dr. Dutsch concluded that the Umkehr method over-estimates the height of the maximum ozone layer and under-estimates its value. In other words, it evens out the

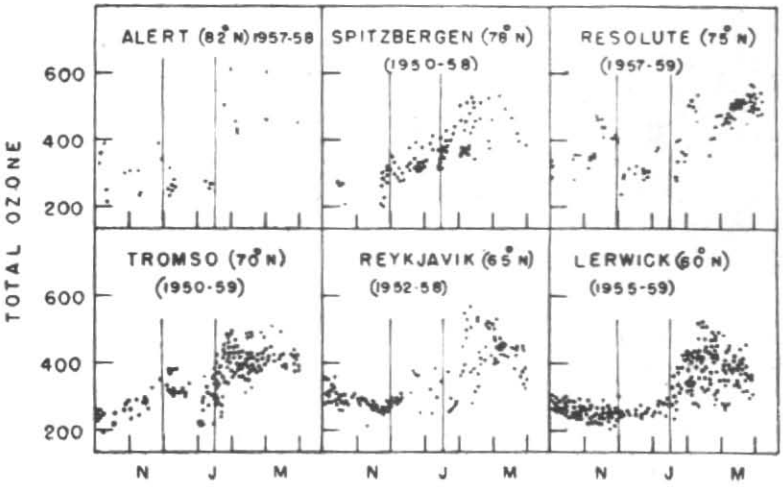


FIG. 6. LATE WINTER RISE IN OZONE IN THE ARCTIC

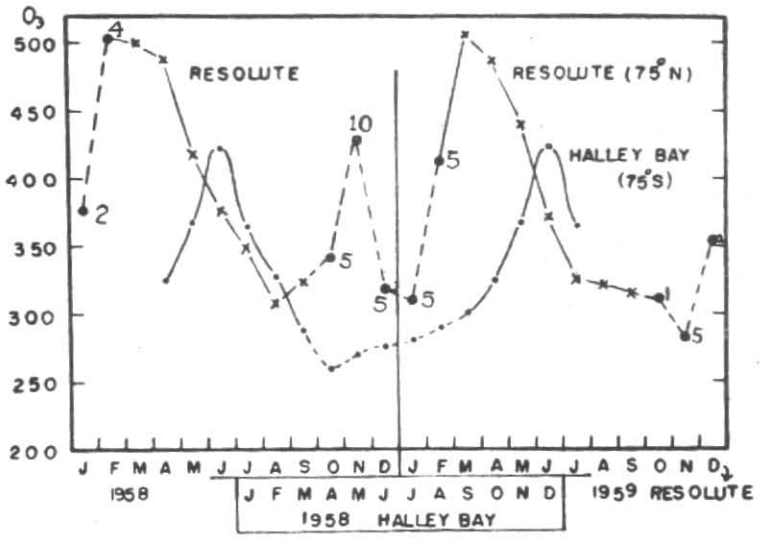


FIG. 7. MONTHLY MEAN OZONE AMOUNTS (PROVISIONAL) AT RESOLUTE (75°N) AND HALLEY BAY (75°S)

distribution too much. It is also insensitive to changes in ozone distribution in the lowest 12 km.

In spite of its defects, the Umkehr method has been used extensively in India, Switzerland, Japan and Canada and has yielded results of substantial value and importance. It enables an estimate to be made of the changes in ozone amount in the higher *ozone production* layer.

3. Transport of ozone from the stratosphere to the troposphere

From an analysis of representative Umkehr curves obtained at places in different latitudes and in different months, it is possible to build up an over-all general picture of the vertical

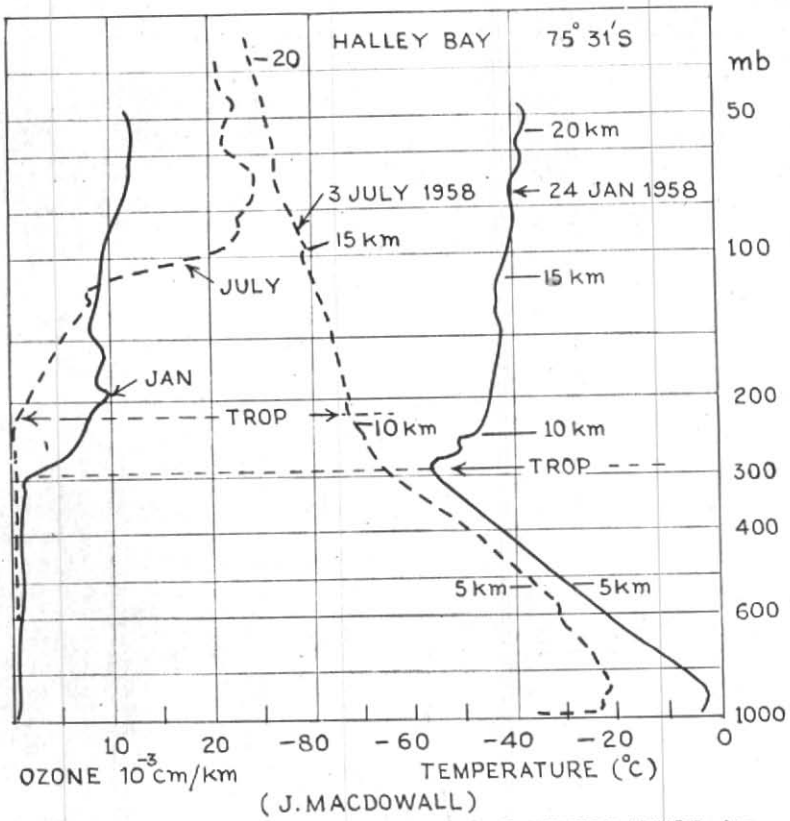


FIG. 8. TWO SOUNDINGS OF OZONE AND TEMPERATURE AT HALLEY BAY. THE CONSIDERABLE CHANGE IN THE FORM OF THE OZONE LAYER WHICH OCCURS FROM WINTER TO SUMMER IS SHOWN. NOTE THE SIGNIFICANCE OF THE TROPOPAUSE

distribution of ozone in different months of the year (Ramanathan and Kulkarni 1960). Such a picture for the months March, July and November is presented in Fig. 10.

As the ozone amount increases from November to March, the increase takes place mainly at levels below 25 km. In March at 18 km, the mean ozone amount using Vigroux' coefficients changes from 5×10^{-3} cm km^{-1} at the equator to 15×10^{-3} cm km^{-1} at 70°. In the same season, there is a tendency for the ozone amounts above 30 km to be higher than over tropical latitudes.

In the latitude range 25° to 75°N, the ozone amount in March from ground to 12 km is 60 to 70 per cent more than that in October-November. An amount larger than this should have been transported down from above during the interval as some ozone would have been destroyed in the troposphere. The excess ozone in 0-12 km in March over that in November corresponds to about 30 per cent of the mean ozone load between 12 and 24 km in the period November to March. Allowing for loss at the ground and in the troposphere it is estimated that about 40 per cent of the ozone load in 12 to 24 km would come down below 12 km in winter and early spring. This result may be compared with that obtained from measurements of stratospheric fallout.

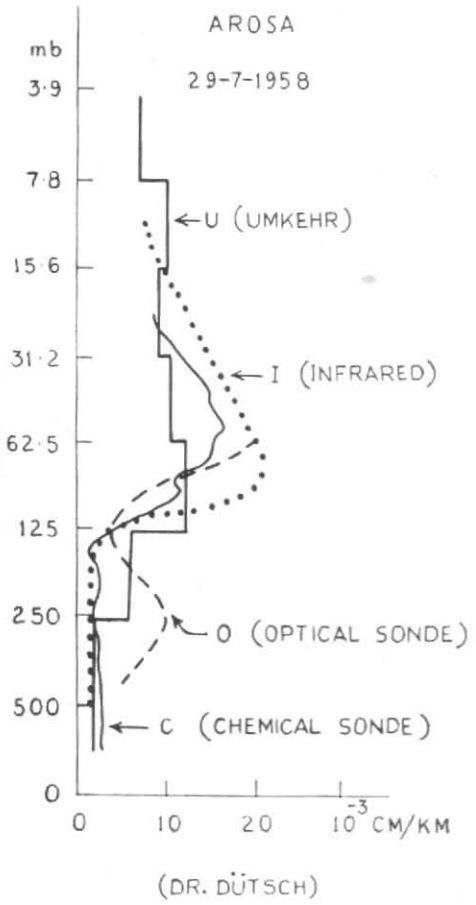


FIG. 9. VERTICAL OZONE DISTRIBUTIONS FROM BREWER-SONDE AND OTHER METHODS

THE TOTAL OZONE AMOUNT WAS 0·310 cm FROM DIRECT SUN OBSERVATION AND 0·289 cm COMPUTED FROM THE VERTICAL DISTRIBUTION OBTAINED FROM UMKEHR OBSERVATIONS

In period April to October owing to the much greater stability of the stratosphere, weakness of the circulation and the gradualness of the tropopause transition between tropical and middle latitudes, a much smaller percentage of ozone may be expected to come down into the troposphere.

4. Stratospheric Circulation

Studies of water vapour and ozone in the atmosphere raise the following questions—

(1) How is the persistent dryness of the stratosphere observed at 45,000 to 50,000 ft over U.K. maintained?

The persistence of the dryness in *all* ascents and the limiting value of the frost-point at 190°K demand a meridional circulation in the lower stratosphere between tropical and higher latitudes as envisaged by Brewer and Dobson. A large-scale exchange of air will not do.

It is accepted that practically all the ozone in the earth's atmosphere is created by the photo-chemical action of sunlight above 24 km. The time required to reach photo-chemical equilibrium when the equilibrium is disturbed is of the order of 4 months at 25 km, a month at 30 km, a week at 35 km and a day at 45 km. Above 35 km, ozone can re-form quickly under the influence of sunlight. Between about 24 km and the tropopause, the ozone can be preserved for long periods of time. In the troposphere, the ozone is destroyed after variable intervals of time.

All studies of vertical distributions have shown that the greater the total amount of ozone, the greater is the amount below 24 km. The additional amount is stored primarily above the tropopause, but substantial amounts come down into the troposphere in high and middle latitudes in winter and spring. There, it is destroyed after varying intervals of time.

(2) What is the reason for the rapid increase in the total ozone content of the atmosphere in high and middle latitudes in the second half of winter and spring? (Godson 1960)

This cannot be due to an increase in production rate, because the largest increase takes place in regions where there is little ozone-producing radiation. As pointed out many years ago by Dutsch (1956), the phenomenon requires a northward transport of

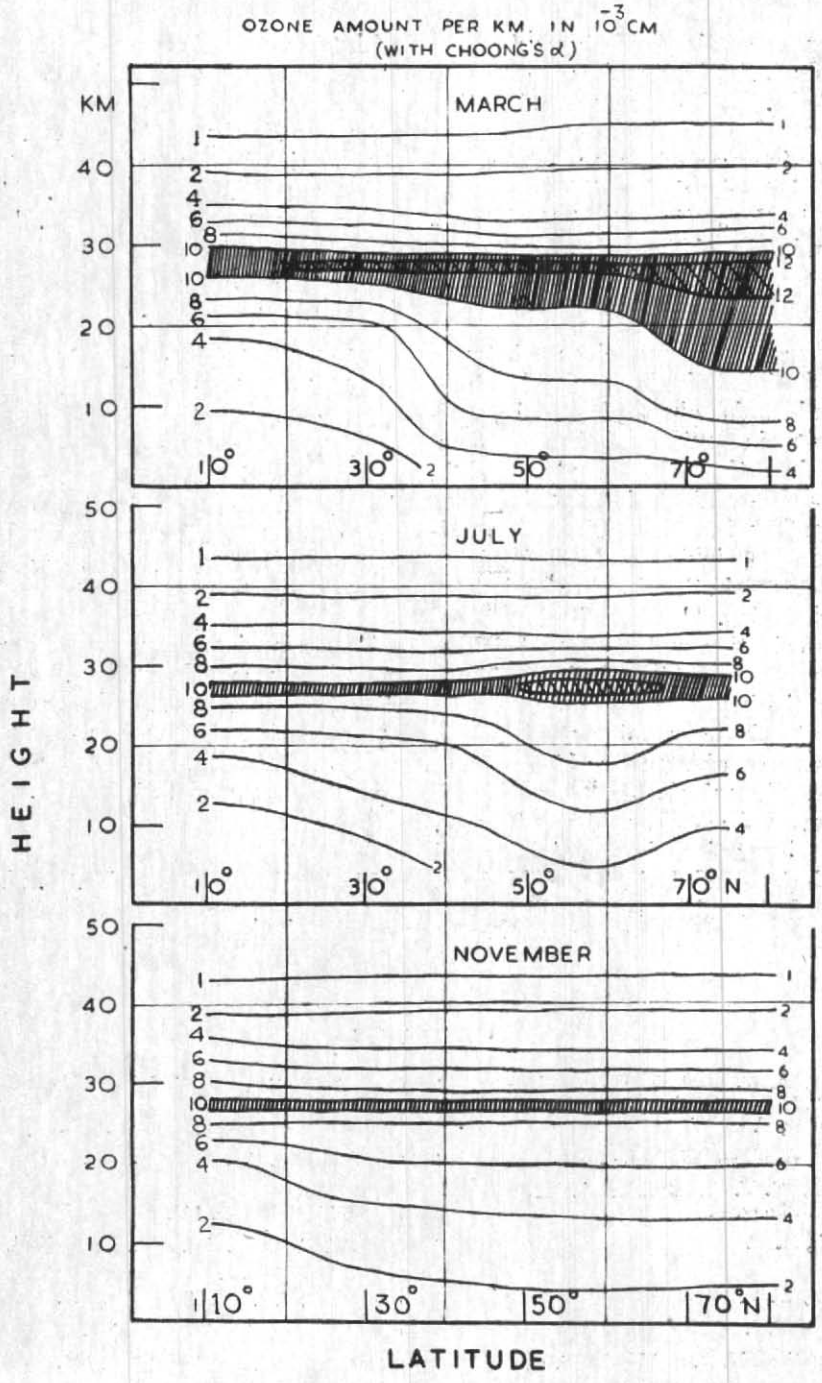


FIG. 10. MEAN VERTICAL DISTRIBUTION OF OZONE

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ozone at ozone-creating levels from middle to high latitudes and a *downward transport* at the *higher latitudes*. It should be emphasised that over regions of polar night, ozone can be accumulated in excess of the amounts corresponding to photo-chemical equilibrium at the same levels in the sunlit part of the atmosphere, even in the middle and upper stratosphere. The de-ozonising radiations are absent.

A large scale mixing scheme between middle and high latitudes at levels above 25—30 km in which air ozonised to its equilibrium value in middle latitudes flows northward into the dark atmosphere and in its place, under-ozonised air from the polar night region flows into the sunlit area, coupled with a gradual sinking of the air in higher latitudes can explain the rise of ozone in later winter and spring. The amplitude of the meridional oscillation and the rate of sinking have however to be sufficiently large. When large scale sinkings of air associated with sudden stratospheric warmings took place, we may expect abnormally high ozone amounts in the middle stratosphere over polar regions and also large rises of ozone. Upper air observations in the Arctic and Antarctic, before and during the IGY, have demonstrated the existence of cold stratospheric lows near the poles during winter with rings of circumpolar strong winds. This ozone of strong winds has probably a much larger amplitude in the northern hemisphere than in the southern, due no doubt to the distributions of continents and oceans (Wexler and Moreland 1958). The vortex breaks down earlier in the Arctic. These breakdowns are associated with explosive warmings of the stratosphere and the warming extends from above downwards.

We have thus to envisage two stratospheric circulations: One a persistent meridional lower stratosphere circulation associated with the equatorial cold tropopause. This is strongest in late winter and early spring. Another, a shortlived winter circulation in the middle stratosphere between polar regions and middle latitudes associated with the cold stratospheric low over the winter pole.

The former is mainly responsible for accumulating ozone and dry air in the lower part of the stratosphere and the latter for the rapid transportation of ozone from the upper ozone-creation layer to the ozone-preservation layer in the second half of winter.

5. Acknowledgement

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