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FINAL REPORT

NASA-Ames Grant No. NSG-2126

"Observed and Theoretical Variations of Atmospheric Ozone"

1 January - 30 April 1976

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Final Report

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OBSERVED AND THEORETICAL VARIATIONS OF ATMOSPHERIC OZONE

Causes for the variations of atmospheric ozone, both total amount and i ' vertical distribution, have been a focal point for our studies during the past few years. In the four month period covered by this final report we have continued to establish a data base of ozone observations and analysis of the observed ozone variations. Our studies covered three subphases:

a) continued analysis of the global distribution of total ozone with the aim of extending our global ozone Atlas to summarize 15 years (1957-72) of ground based observations;

b) analysis of balloon borne ozonesonde observations for Arosa, Switzerland, and Hohenpeissenberg, Germany (GFR);

c) continued processing of the OGO-IV satellite data to complete the analysis of the stratospheric ozone distribution from the available OGO-IV data.

Results of the analysis of the total ozone observations indicated that the long term ozone variation that have been documented for the 1960's have marked regional patterns and tend to alternate with season and hemisphere. It is becoming increasingly clear that these long period changes are associated with large scale variations in the general upper atmosphere circulation patterns.

As discussed in previous reports, the vertical distribution of ozone shows a very pronounced seasonal variation and the amplitude of this seasonal variation has a maximum in the lower stratosphere at levels of about 50-100 mb. Long period changes in total ozone are seen to be influenced by long period changes occuring at the levels of maximum ozone. Curiously, ozone variation, up to the top of the ozonesonde ceiling (about 10 mb) show no definite response to solar variability as described by monthly sunspot data. However, at the level of ozone maximum there is a significant 26 month periodicity that is clearly associated with the

tropical quasibiennial oscillation. This is another strong indication of ozone variability as a response to stratospheric circulation patterns. Comparison of the data from both stations (Arosa and Hohenpeissenberg), which are about 250 air kilometers apart, show near identical average results for the same approximate periods suggesting that these are representative results.

The research papers were, in part, prepared during the grant period for presentation at the International Ozone Symposium that was held in Dresden, Germany (GDR) 9-17 August 1976. [A trip report with a summary of the two papers was submitted to NASA Headquarters dated 10 November 1976. A copy that that trip report is attached.] Reprints of the papers presented are included with this Final Report. The papers will be published by the German (East) Academy of Sciences as the Proceedings of the Symposium.

The research conducted under Grant No. NSG-2126 and reported here was continued under a follow-up NASA Grant NSG-7224.

UNIVERSITY OF COLORADO
BOULDER, COLORADO 80309

Department of
Astro-Geophysics

10 November 1976

NASA Headquarters
Code I
Washington, D. C. 20546

Attention: Personnel Exchanges Division

Subject: Trip Report: Joint Symposium by the International Ozone Commission of Atmospheric Chemistry and Global Pollution, Dresden, German Democratic Republic, 9-17 August 1976.

The following report briefly summarizes the scientific program of the ozone symposium. The symposium was sponsored locally by the Academy of Sciences of the GDR and the National Commission of Geodesy and Geophysics. All sessions were held in the Andreas-Schubert House of the Technical University of Dresden.

The main topics covered at the symposium were:

a) New developments in ozone measurements;

b) observations and distribution of ozone and other atmospheric trace constituents;

c) photochemistry and transport of ozone and other trace constituents;

d) anthropogenic influences on ozone.

During the discussion on instruments and observations it was clearly shown that whereas great advances have been made in ground based observational techniques for measuring ozone, there is still a need for an improved, compact, well calibrated standard instrument for the international network. Efforts are underway to make the Dobson Spectrophotometer fully automatic and to improve the M-83 filter instrument used in the network in the Soviet Union and other Eastern European countries (there are about 45 stations in the Soviet Union taking daily total ozone observations).

Results of balloon borne ozonesonde and satellite and rocketsonde measurements provide an excellent data base for model calculations of the ozone distribution and its variation. The observations indicate the importance of meteorological interaction with ozone photochemistry up through the middle stratosphere--particularly at high latitudes during the winter. Stratospheric ozone observations were shown to be closely related to other meteorological parameters such as the jet stream and the statistical characteristics of the large scale circulation in the stratosphere.

An excellent review paper (by Dr. J. S. Chang of LLL) discussed the important strengths and weaknesses of 1-D, 2-D and 3-D photochemical-dynamic models of the ozone stratospheric distribution. It was pointed out that each model can be useful providing that the model limitations are clearly realized and specified.

Recent data and calculations of anthropogenic influences on the ozone concentration were presented. These summarized the conclusions that have since been reported in the panel report of the Climate Impact Committee of the NAS. Namely, continued use of chemicals such as CCl_2F_2 and $CClF_3$ as serosol propellants represented a clear and serious potential for reduction of ozone concentrations.

Two of the papers presented at the symposium covered research done under our NASA Grant NSG 7224. These papers are described below:

1. "Satellite Observations of the Global Distribution of Stratospheric Ozone" by Julius London, John E. Frederick and Gail P. Anderson

   Measurements made by the Orbiting Geophysical Satellite (OGO-IV) contained, in part, observations of backscattered ultraviolet radiation at wavelengths from 1100 to 3400Å for the period mid July 1967 to 19 January 1969. We were able to reduce a portion of these observations--for the period September 1967 to February 1968--to determine the geographic distribution of ozone at different levels in the middle and upper stratosphere (30-55 km). The derived distributions, averaged to give monthly mean values, show significant seasonal and geographic variation with important differences indicated between winter and summer Hemisphere distributions. In particular, the latitudinal gradient of ozone mixing ratio is very flat in the summer Hemisphere but shows marked increases with latitude in the winter Hemisphere.

   The OGO derived distributions are consistent with observations made from rocketsondes and from Nimbus-4 (BUV). It was found that ozone in the middle and upper stratosphere is in near photochemical equilibrium during the summer but not during the winter where transport processes are effective in redistributing the ozone at levels up to about 45 km. At the top of the stratosphere the ozone concentrations are affected by the temperature sensitive chemistry at these levels.

   It should be noted that plans are currently underway to process the remaining OGO-IV backscatter data for the period March 1968 to January 1969.

2. "Variations including Possible Solar Cycle Variations of Stratospheric Ozone over Central Switzerland" by Julius London and Hans-Ulrich Dütsch

   Balloon borne observations of the ozone partial pressure have become available at a number of stations during the past ten years or so. These observations are taken 2 to 3 times a week and measure the ozone partial pressure generally up to heights of about 30 km. They therefore represent an important bridge between daily routine observations of total ozone and satellite observations of the vertical ozone distribution above 30 km. We presented an analysis of the ozone distribution derived from balloon observations over Central Switzerland for the eight year

period 1966-1974. The analysis showed that:

a) the upward trend of ozone in the late 1960's which ended abruptly in early 1970 was produced by long term variations in the ozone partial pressure in the lower stratosphere. No such trend variation was noted in the middle stratosphere above about 30 mb;

b) a pronounced autocorrelation with a period of 26 months was evident for the layer 62.5-31.3 mb but not apparent in the data for other layers;

c) correlation between monthly relative sunspot number and ozone concentrations at different levels shows a marginally significant (2σ) correlation with a lag of about 12 months (similar to that between sunspots and total ozone over the same period).

However, no significant correlation between sunspots and ozone concentration in the middle stratosphere are found. The theoretical implications of these results were discussed.

A separate analysis of the data from the Hohenperssenberg Observatory (just west of Munich) for the same approximate period showed a completely consistent set of results.

Incidentally, we are now planning to utilize the balloon observations from W. Berlin and Brussels together with those from Switzerland and Southern Germany to find what geographic patterns (if any) are apparent over Western Germany.

In all about 125 papers were presented. These will be published in the Proceedings of the Symposium to appear early in 1977.

Julius London
Project Director

JL/rlg

cc: Code SU, Attention: L. R. Greenwood

VARIATIONS INCLUDING POSSIBLE SOLAR CYCLE VARIATIONS OF STRATOSPHERIC OZONE OVER CENTRAL SWITZERLAND

N78-12

Julius London and Hans-Ulrich Dütsch

1. Introduction

The meteorological importance and reasons for current societal interests in variations of atmospheric ozone have been outlined in a recent report of the U. S. National Academy of Sciences, NAS (1975)[1]. Although the overall distribution of ozone, both geographic and vertical, is reasonably well known and is even partially understood (e.g. Dütsch, 1974)[2], as in the general case of changes in climate, there is not yet available an acceptable theory for time variations of the ozone distribution for periods of more than one year.

There are indications of how some observed climatic parameters are associated with ozone changes at particular levels in the atmosphere, but a number of fundamental questions are still to be answered (e.g. Why is the observed ozone Quasi-Biennial Oscillation (QBO) stronger in the Southern than Northern Hemisphere?) (See, for instance, Angell and Korshover, 1973)[3].

From time to time different variations of ozone amounts have been reported involving stratospheric sudden warmings, quasi-biennial oscillations, solar cycle variations, long period trends, etc. For instance, strong horizontal and vertical transport associated with the breakdown of the winter/spring polar circulation vortex is accompanied by a large increase in the ozone concentration - particularly at levels near the ozone maximum. Also, the variation of total ozone in the tropics and the ozone concentration in the layer 30-60 mb at other latitudes seems to be associated with the quasi-biennial wind oscillation in the tropical stratosphere such that there is a higher ozone concentration during the time of easterly stratospheric winds at 50 mb. (Angell and Korshover, 1973)[3].

Long term trends in ozone during the 1960's have been reported (Komhyr et al, 1973[4], London and Kelley (1974)[5], and others but are not explained. Of all the suggested observed period in ozone variations, perhaps the most intriguing is that related to the solar cycle. These suggestions date back at least to Humphreys, 1910 [6], and were claimed by Willett (1962)[7] on the basis of data from the ozone network up to 1959.

Establishment of a clear association between ozone variations and some manifestation of solar activity has, in addition to its intrinsic value, the possibility of providing some clues of physical mechanisms for varied suggestions of solar-weather relations. It is not at all certain, however, that the observations show a pattern linking ozone variations to anomalous solar activity. The search for such a pattern has, in general, involved total ozone amount and sunspots as the solar parameter. As more stratospheric ozone data become available, through ozonesonde and/or satellite observations, the search for a solar-ozone relationship needs to make use of stratospheric concentrations as the ozone parameter.

If the observations clearly show the likelihood of an ozone variation with solar activity - it is possible that the physical mechanism is a more or less direct one. In this case the solar induced variations should be most pronounced at the levels where photochemistry can have a significant influence on the ozone concentration. There should then be a relatively short phase lag between solar activity and middle and upper stratospheric ozone variations. The phase lag should then increase downward.

Alternatively, the mechanisms by which such an association comes about could be through variations in the tropospheric and/or stratospheric circulation patterns - as, for instance, by modulation of the strength of the Hadley circulation. The significant variations should then be found in the lower stratosphere at the level of ozone maximum and below since at these levels the ozone distribution is largely determined by

meteorological processes. As mentioned above, evidence of the QBO of lower stratospheric ozone is probably a good example of such a meteorological influence.

Convincing theoretical arguments (Frederick, 1976 [8]; Crutzen et al, 1975)[9] and evidence from satellite observations (Heath et al, 1976)[10] indicate that at the time of a strong solar proton storm there should be a large ozone decrease in the mesosphere and upper stratosphere at high geomagnetic latitudes. It has also been suggested (Ruderman and Chamberlain, 1975)[11] that a significant increase in ozone at the height of ozone maximum should occur at high geomagnetic latitudes after solar maximum and that this increase would be propogated to mid-latitudes in about 1 - 3 years. It is suggested that decreased Galactic Cosmic Rays penetrating to the polar middle stratosphere would result in smaller NO concentrations and therefore decrease the destruction rate of stratospheric ozone.

Information concerning the ozone distribution and its variations,has been derived mostly from the world network of total ozone observations. The level of ozone maximum, however, is in the lower and middle stratosphere and it is at these levels that most of the significant variations should be found.

The vertical ozone distribution has been observed from ground based - Umkehr - measurements at about 15 stations having Dobson spectrophotometers. In addition, there are about 10 ozonesonde stations taking in-situ observations of the ozone concentration up to about 30 km. These latter observations have been taken more or less regularly with a frequency that varies from year to year and station to station. In general efforts have been made to take observations from 1 to 3 times per week. Most of the observations have been made with the Brewer-Mast ozonesonde (see, for instance Dütsch and Ling, 1973 [12], DeMuer, 1976 [13], Attmanspacher, 1976)[14]. One of the regular ozonesonde stations is that operated by the Federal Institute of

Technology at Zürich in cooperation with the Swiss Meteorological Office. The observations were taken at Thalwil (near Zurich) until August 1968 at which time the observation site was changed to Payerne (about 140 km SW of Zurich). Details of the observational program and discussions of analysis of some of the data are contained in Dütsch and Ling (1973)[12], Dütsch, (1974b)[15].

2. Data

The data discussed here are in the form of monthly mean partial pressure of ozone in 7 different layers - from the surface to 7.8 mb. Each layer is about 4.5 km thick. The period of observations used here was September 1966 to May 1976. The long term monthly means for each layer were determined from the approximately 10 years of observations.

It is well known that there is a pronounced seasonal variation of the ozone concentration in the troposphere and stratosphere (e.g. Dütsch, 1974)[2]. An harmonic analysis was performed on the long term monthly ozone variation for each layer. The results are shown in Fig. 1. For comparison purposes, a similar analysis for Hohenpeissenberg (about 300 km ENE of Payerne) was computed for about the same period and is also shown in Fig. 1. Fig. 1a and 1b give the height distribution of the phase and amplitude of the first harmonic (annual variation) of the ozone partial pressure for the two stations. The percent variance of the total variation that is accounted for by the first harmonic is shown in Fig. 1c. It is clear from the similarity of the two curves in Fig. 1a,b and c that the computed values are quite representative of the seasonal variation of ozone in this general locality. The results shown here are analogous to those presented and discussed by Dütsch (1974)[2], DeMuer (1976)[13] and Attmanspacher (1976)[14].

Ozone in central Europe is a maximum in the lower and middle stratosphere during March and April. There is a phase lag of the ozone variation in the troposphere so that tropospheric ozone increases through

the spring as ozone is sporadically transported downward from the polar stratosphere. In the middle stratosphere, the ozone concentration tends to increase towards the summer as photochemical processes governing ozone production become dominant. There are indications, however, that above ten mb the combination of photochemistry and transport act to produce a winter maximum in the ozone concentration (e.g. London et al, 1976)[15]. It is clear from Fig. 1b that the largest amplitude of the annual ozone variation is found in the lower stratosphere and this is responsible for determining the phase and amplitude of the annual variation of total ozone. Note, for instance, that the lower amplitude of stratospheric ozone at Hohenpeissenberg is reflected in the lower amplitude of total ozone. Fig. 1c shows that the annual variation is quite pronounced up to 30 mb but is very weak just above the level of maximum ozone. This is because of a phase change in the middle stratosphere above 10 mb (e.g. DeMuer 1976)[13].

The long term monthly mean ozone amount was subtracted from the individual monthly values to give a time series of monthly ozone anomalies for each layer and for total ozone (at Arosa). This time series with a 1-2-1 smoothing function is shown in Fig. 2 for the period September 1966 to May 1976 for total ozone and two layers in the lower and middle stratosphere.

Total ozone at Arosa shows an increase to a maximum in early 1970 and then a general decrease to its lowest value in 10 years. This variation is best reflected in the variation in the lower stratospheric layer 125-62.5 mb. However, there is a large difference between the ozone changes in the layer 125-62.5 mb and that immediately above. No particular long period trend is evident for the ozone partial pressure in the top layer 15.6 to 7.8 mb. There is a general decrease starting in late fall 1972 and reaching its lowest value (6.9 nb) in February, 1975. It may be that this decrease was associated with the strong solar proton event that

occurred in early August 1972. However, satellite observations failed to show any strong decrease at 50° latitude in the total ozone above 7 mb. Nor was this decrease evident from the Hohenpeissenberg observations at 10 mb.

In order to check on the possibility of various periodicities in the ozone variation, we calculated the autocorrelation of the filtered monthly ozone values for each of the layers considered. The largest value for the autocorrelation occurred at a lag of 25/26 months (0.40) at a level of 62.5-31.3 mb. A similar value for the autocorrelation was found for Hohenpeissenberg at 40 mb (0.43) at a lag of 26 months. No such significant value for the autocorrelation in the temperature variation was found. This periodicity in ozone has already been noted by Dütsch (1974)[1] and others and is clearly related to the tropical Quasi-Biennial Oscillation. The fact that the oscillation is evident in the ozone variation in the layer of maximum ozone and is not apparent in the temperature variation at this or other lower stratospheric levels suggests that it is the variation in the strength of the ozone transport from tropical regions that is responsible for the observed Quasi-Biennial Oscillation in ozone over Switzerland and Southern Germany. This suggestion, however, needs documentation.

3. Possible Solar Activity Related Variations

There are many studies reported in the literature of suggested relations between some form of anomalous solar activity and variations of ozone. The results of these studies tend to indicate that there may be a connect on between anomalous solar activity, characterized by relative sunspot number, and atmospheric ozone, usually represented by total ozone in an atmospheric column. Whereas the relative sunspot number is a fairly good indicator of various forms of solar activity, variations of total ozone best reflect variations in the ozone concentration at and just below the level of maximum ozone. It is expected as discussed earlier in this

paper that if there does exist a solar-ozone relationship, it would most likely be realized in a stratospheric ozone response. We have made use of the ozonesonde observations at Thalwil/Payerne to study such a possible relation. We have computed the cross correlation between the monthly relative sunspot number and monthly ozone partial pressure anomalies as described above. Again each series was subjected to a 1-2-1 smoothing function in order to minimize some of the high frequency noise in the data. Computations were made for total ozone and for each of the 7 layers for which we had data. Correlations were computed for time lags (month of ozone - month of sunspots) of 0 to 57 months. The results of the computated correlation coefficients ($\pm 2\sigma$ as indicated) are shown in Fig. 3 for some representative layers.

The correlation is barely significant - at the 2σ level - for total ozone with a lag of 14 months. Note that this is different from the long term correlation of sunspots and total ozone which has a reported maximum at 3-4 years (e.g. London and Oltmans, 1973)[17]. In the troposphere the correlation coefficient between ozone and sunspots for the period studied is about 0.5 with a lag of 36 months. However, in the lower stratosphere (150-62.5 mb) the correlation reaches a maximum again at a lag of 14 months. In the layer of maximum ozone (62.5-31.3 mb) the correlation coefficient is largest with about a 2 month lag. There is no significant correlation indicated in the middle stratosphere (up to 7.8 mb).

The computed correlations shown here for the different levels at Thalwil/Payerne are very similar to those derived for the data from the same period for Hohenpeissenberg. That is, relatively large correlation with a lag in the troposphere decreasing in the stratosphere to near simultaneous patterns at the level of ozone maximum and then insignificant correlation above.

The fact that there are significant - at the 2σ level - correlations between the two series studied does not mean, of course, that there is a real physical relationship between the two. Since the correlations could arise if the two phenomena were periodic with a similar period but each phenomenon having a different physical origin. Indeed, a careful statistical analysis of the entire record of the Arosa total ozone series suggests that there is probably no direct relationship between total ozone at Arosa and relative sunspot number (Hill and Sheldon, 1976 [18]. But, if there were a physical relation between solar activity and ozone variations - we would expect that the phase lag for such a relation would decrease with height - at least up to the level of maximum ozone. It may be, as suggested by Zerefos and Crutzen (1975)[19] that the ozone response to energetic solar proton events changes sign at about 35 km. If so, this would explain the absence of any noticeable relationship in the layer 31.3-7.8 mb in the Thalwil/Payerne data. (It should be noted, however, that this latter would be inconsistent with the Umkehr observations above 40 km at Arosa discussed by Dütsch (1969)[20]. Zerefos and Crutzen, 1975[18] also state that high latitude stratospheric temperature variations follow the solar cycle. We find no evidence of this in the stratospheric temperature data for the period studied here.

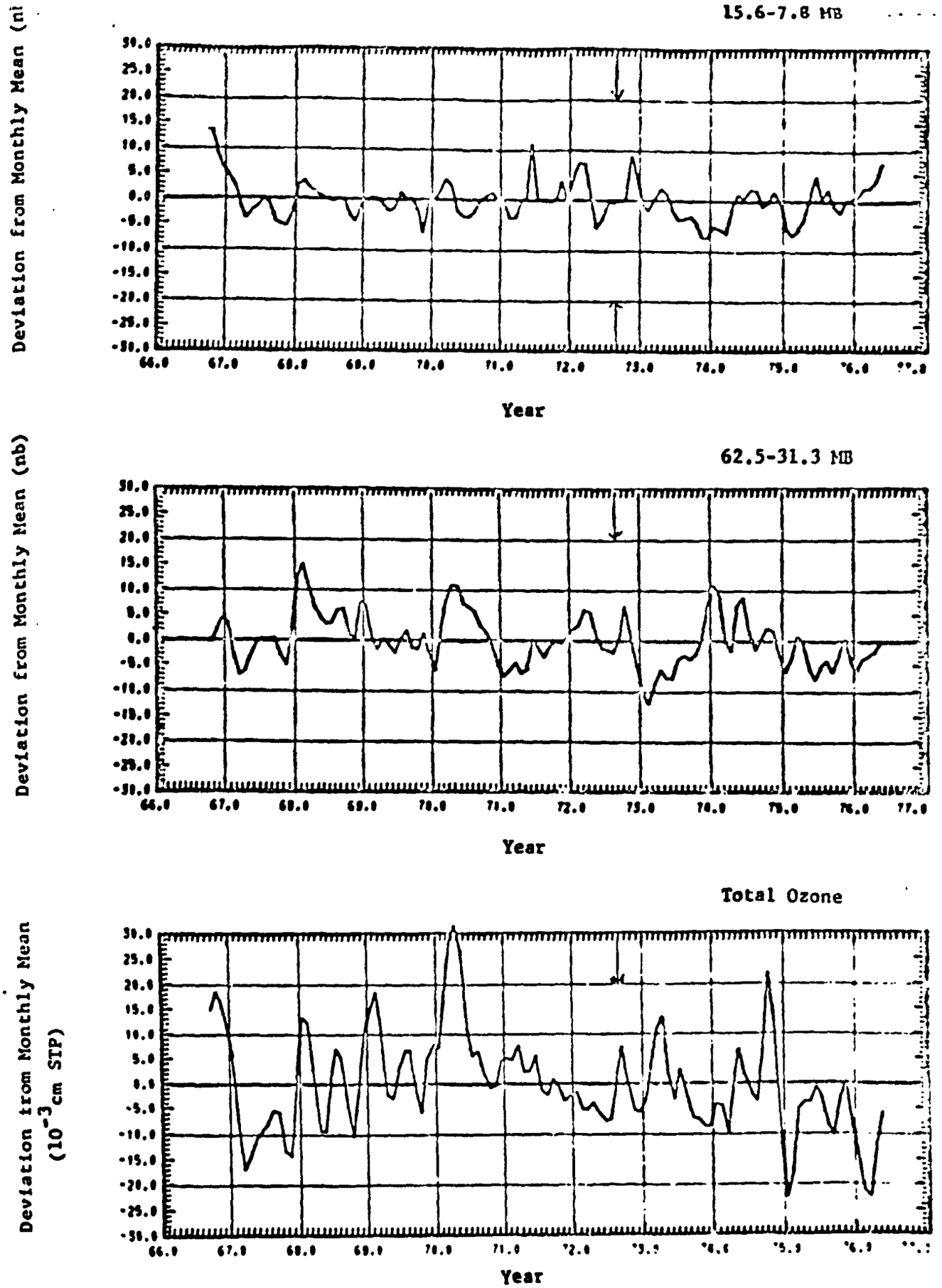
4. Summary

We have analyzed the ozone data derived from ozonesonde observations taken over Central Switzerland for the period September, 1966 to May 1976. The annual variation of the total amount of ozone and the partial pressures in each of 7 layers from the surface to 7.8 mb shows the pattern representative of mid-latitude ozone: Summer maximum in the troposphere, spring maximum in the lower stratosphere up to 30 mb and summer maximum in the middle stratosphere up to ~8 mb. It was also shown

that at the level of the ozone maximum the ozone partial pressure has a pronounced period of about 26 months. Total ozone and the ozone concentration in the different layers have marginally significant (at $\pm 2\sigma$) correlations with relative sunspot number. The time lag for these correlations is long in the troposphere (about 36 months) but decreases with height to the level of ozone maximum (to about 2 months). A similar analysis of the data derived from the ozonesonde program at Hohenpeissenberg gave completely consistent results with those discussed above. A more extensive ozonesonde program than presently exists and utilization of complementary satellite observations could provide considerable help in developing a data base for unraveling the intriguing and important problem of the existence of solar cycle induced ozone variations.

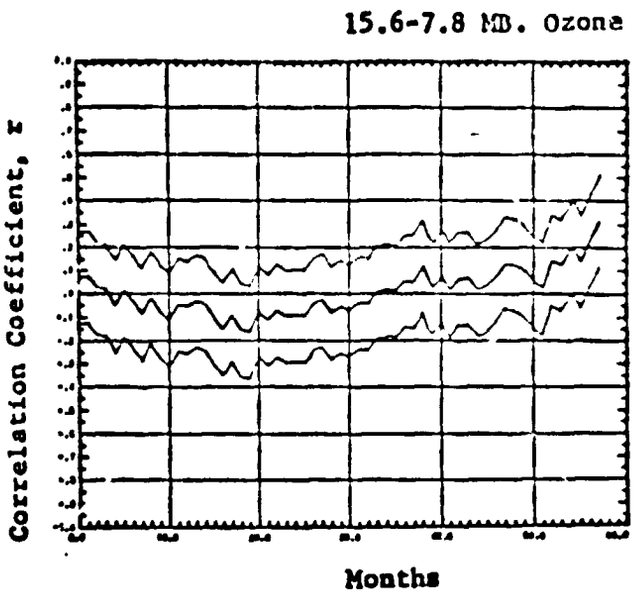
Acknowledgments

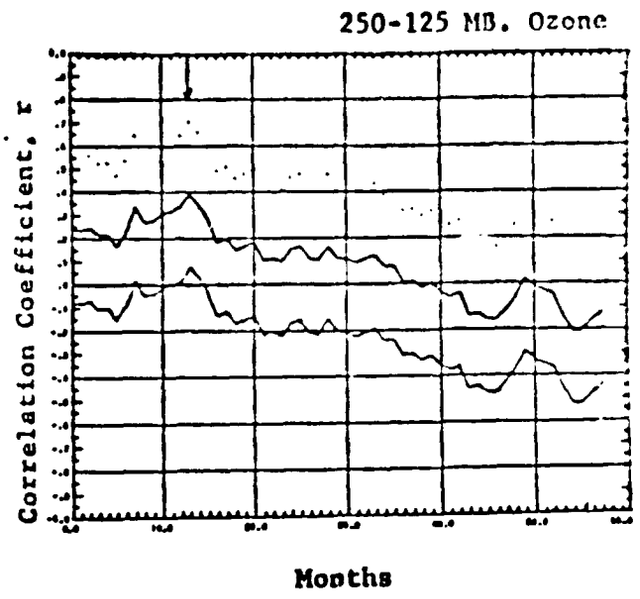
A portion of this study was when the first author was visiting professor at the Federal Institute of Technology (ETH) in Zürich, Switzerland. We would like to thank Mark Clayson for programming the statistical evaluation and graphics used in this paper. The work discussed here was supported, in part, by NASA-Ames Grant No. NSG-2126 and NASA NGR-06-003-127.

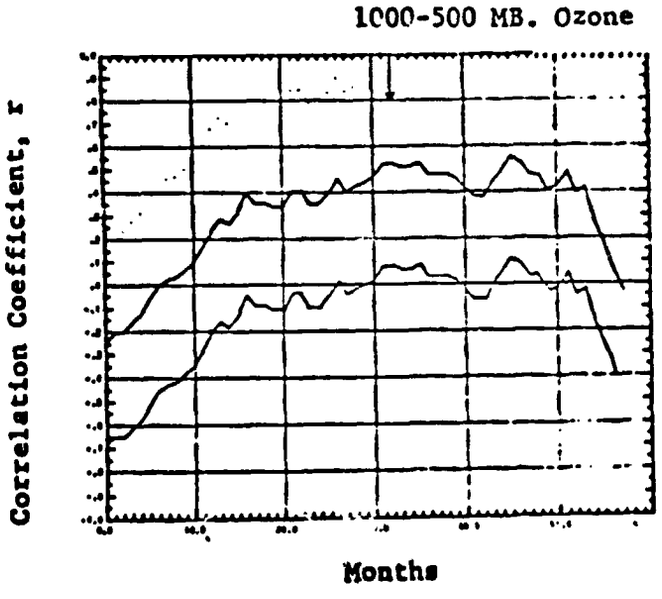


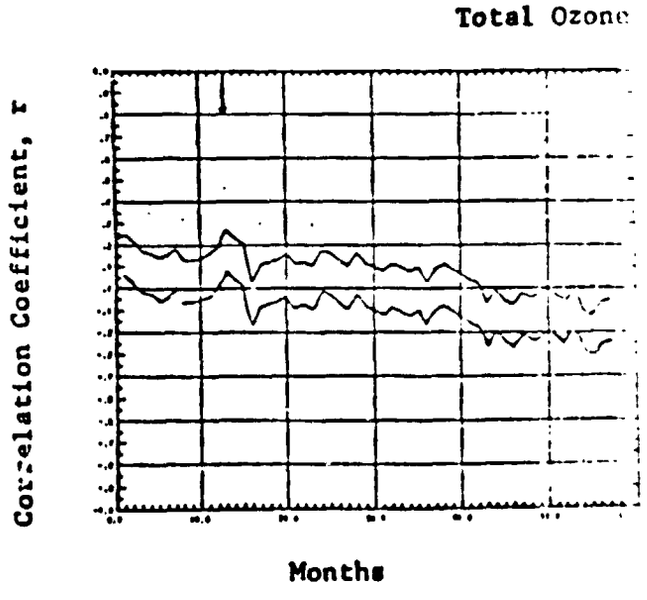
Filtered Deviation of Ozone from Long Term Monthly Mean for Arosa and Thalwil/Payerne.

Fig. 2









Lag Correlation of Ozone vs Sunspots
(Ozone mo. minus SS mo.)

Fig. 3

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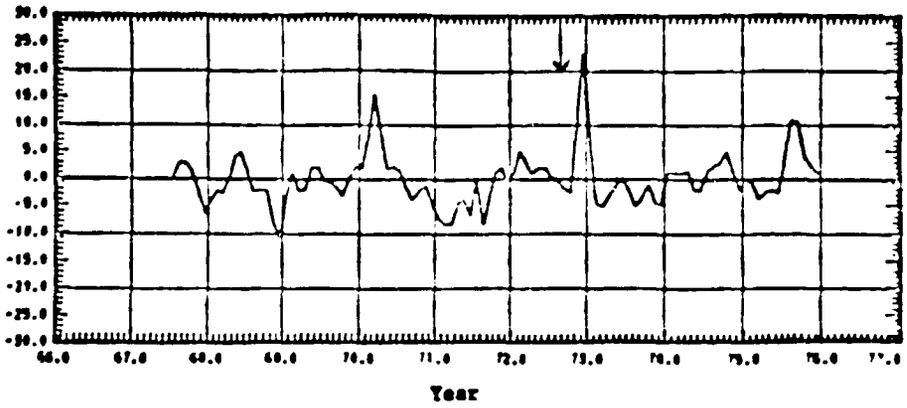

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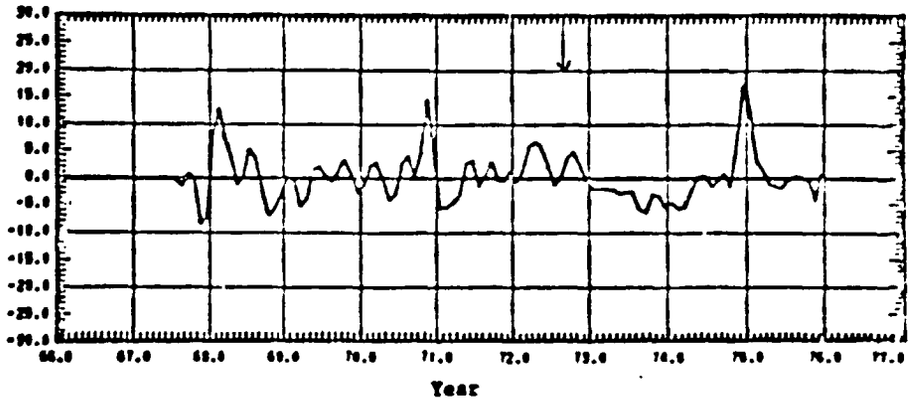

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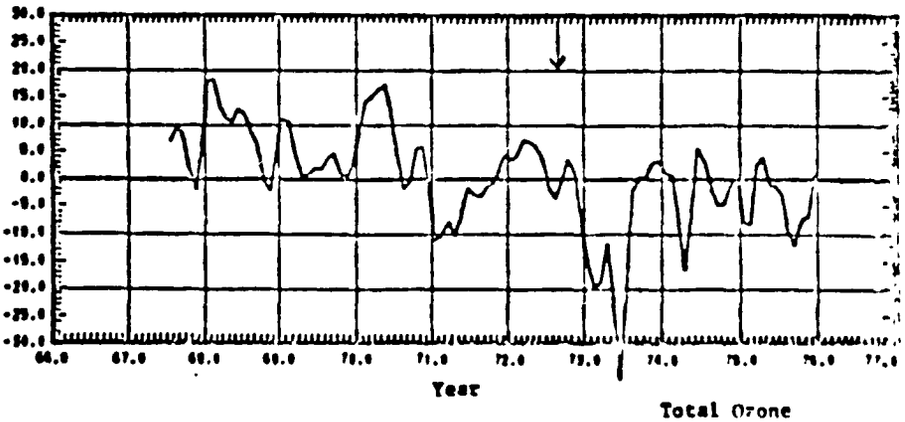
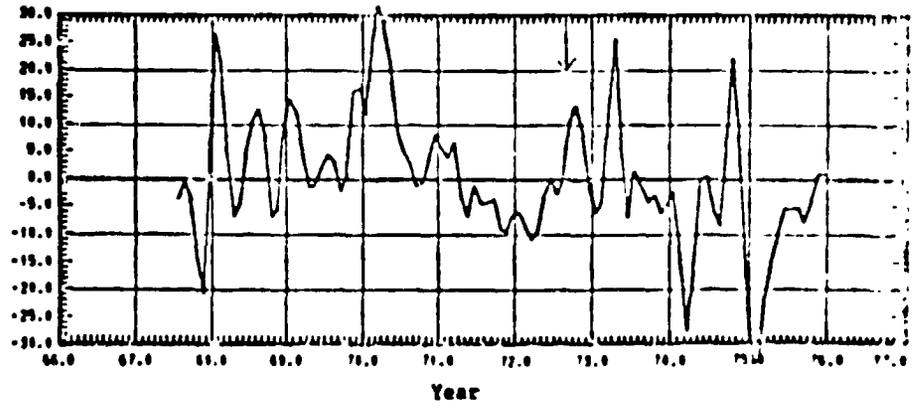


Total Ozone



Filtered Deviation of Ozone from Long Term Monthly Mean for Hohenpeissenberg.

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SATELLITE OBSERVATIONS OF THE GLOBAL DISTRIBUTION OF STRATOSPHERIC OZONE*

Julius London, John E. Frederick and Gail P. Anderson

ABSTRACT

Observations of backscattered radiation from an Orbiting Geophysical Observatory (OGO) Satellite for the period September 1967 to February 1968 have been used to determine the global distribution of ozone in different layers in the middle and upper stratosphere. The derived distributions show significant seasonal and geographic variations with important differences indicated between winter and summer hemisphere distributions. The OGO derived distributions are compared with other observations (rocket and satellite) and with photochemical calculations. It is suggested that the increased ozone mixing ratio in the high latitude winter hemisphere can be accounted for by transport processes up to about 40-45 km and by the effects of seasonal variations of NO_x, HO_x and temperature in the region above.

1. Introduction

Successful efforts to measure the vertical distribution of stratospheric ozone were first made by Götz et al (1934)[1] from ground based observations and E. and V. Regener (1934) [2] from in-situ optical measurements. A number of other techniques have since been developed and a review of methods currently used for ozone observations is given by Dütsch (1974)[3].

The ground based technique used by Götz et al [1] (Umkehr technique) is based on measurements of solar radiation scattered vertically downward from the zenith sky in two different wave lengths of the Huggins bands as

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*Modified and condensed from a paper submitted for publication in the Journal of Geophysical Research.

$$I_\lambda = I_{o\lambda} \frac{3(1+\cos^2\theta)}{16\pi} \int_o^\infty e^{-\tau_\lambda(1+\sec\theta)} \omega(\tau_\lambda) d\tau_\lambda \qquad (1)$$

where $I_{o\lambda}$ is the incident solar intensity, θ is the solar zenith angle, τ_λ is an optical depth variable and $\tau_\lambda^*(p)$ is the total vertical optical depth of the atmosphere at wave length λ down to a level of atmospheric pressure p. τ_λ and $\omega(\tau_\lambda)$ are given by:

$$\tau_\lambda = k_\lambda X(p) + \beta_\lambda p$$

$$\omega(\tau_\lambda) = \frac{\beta_\lambda p}{\beta_\lambda p + k_\lambda X(p)}$$

where X(p) is the total ozone (cm STP) in a vertical column above pressure p, k_λ is the ozone absorption coefficient (cm^{-1}) and β_λ is the Rayleigh scattering coefficient (atm^{-1}).

Equation (1) can be written as

$$S_\lambda = \int_0^1 y^{(k_\lambda/k_o)(1+\sec\theta)} \frac{dp}{dy} dy \qquad (2)$$

where $S_\lambda \quad \frac{I_\lambda}{I_{o\lambda}} \quad \frac{16\pi}{3\beta_\lambda(1+\cos^2\theta)}$

It is assumed that $k_\lambda X(p) \gg \beta_\lambda p$ and $y = e^{-k_o X(p)}$, and k_o is a normalizing function of solar zenith angle. The lower bound assumes that the total amount of ozone in a vertical column above the ground is large enough so that $e^{-k_\lambda X_o} \approx 0$.

Equation 2 is solved for dp/dy at a series of pressure levels defined by the condition that level i has an ozone amount $X(p_i)$ above it. dp/dy can then be converted in a straightforward way to the ozone mixing ratio (see, for instance, Anderson (1969) [6].

In the evaluation procedure an initial profile was assumed and a first calculated estimate of the vertical distribution was made using Equation (2). A second iteration to the solution was then performed by including the Rayleigh scattering term in the integrand of Equation 2.

The assumption of single scattering and the limit of the instrumental signal to noise ratio defined the regions of useful data retrieval. Usable information was obtained from levels of about 10 to 0.4 mb and for solar zenith angles less than 70°. It should be noted that the information content of the parameters and the inversion technique used in the present evaluation could yield only four independent pieces of information on the vertical ozone distribution for each sounding.

Satellite derived distributions were compared with a set of five rocketsonde observations taken over the same area (Hawaii) and at approximately the same time as the satellite crossings. There was very good agreement (generally within about ten percent) between the mixing ratios derived from the satellite and the rocketsonde observations (London et al, 1976)[7].

3. Results

Vertical ozone distributions were determined from observations taken during September, October and November 1967 and January and February 1968. No data were available during December 1967 because the precessing satellite was generally in a twilight orbit during that month. The availability and distribution of data was better during September, October and November than during January and February. In general, the amount of ozone information acquired by the spectrometer was less than ten percent of that which could have been obtained under suitable solar observing conditions. A typical day on which the spectrometer operated had only one to three orbits of data. These were widely spaced in longitude but for each orbit the data were usually continuously distributed over a large range of latitude.

As mentioned earlier, ozone distributions were derived for the stratospheric layer 10 mb - 0.4 mb. The global distribution of average monthly ozone concentration was determined for each of the five months

the solar elevation angle decreases during the day. A modification of this technique was suggested by Singer and Wentworth (1957) [4] for a method of inferring the vertical ozone distribution in the upper atmosphere by measuring the backscattered solar radiation at a number of wave lengths in the Hartley-Huggins bands from instrumented satellite platforms. Various satellite programs for observing stratospheric and mesospheric ozone, based on vertical backscatter, occultation (absorption) and limb scanning (scattering and emission) techniques, have been developed since about 1965 (see, for instance, Heath et al, 1973)[5].

Satellite observations of stratospheric ozone have many obvious advantages over other observational methods (i.e. ground based, balloon ozonesonde, or rocketsonde) since they can be continuous in time and global in geographic coverage. Satellite observations are, of course, expensive and they cannot provide detailed vertical distribution as is available with balloon ozonesonde or rocketsonde measurements. Also satellite observations are less accurate for total ozone measurements than ground based Dobson observations.

The OGO-IV satellite was launched in July 1967 and took measurements from which stratospheric ozone data could be derived for the period up to mid-January 1969. We here report on an analysis of part of this data set from September 1967 to February 1968. A spectrometer was used in OGO-IV to monitor backscattered ultraviolet radiation. The vertical distribution of ozone in the region 30-55 km was determined from analysis of the data measured in the spectral interval 2550Å to 3010Å. The data set and the methodology involved in evaluation of the data has been discussed by Anderson (1969)[6] and London et al (1976)[7] and is briefly outlined below.

2. Data Evaluation

For a plane parallel atmosphere, the vertically emergent intensity, I_λ, with the assumption of single scattering is given by

for which we processed the data. The ozone equivalent depth (cm STP) was calculated for three different layers 10-4 mb, 4-1.6 mb and 1.6-0.6 mb representing regions where atmospheric transport, photochemistry and transport, and photochemistry respectively are generally assumed to be dominant in determining the ozone distribution. In addition, the ozone mixing ratio at various levels in the middle and upper stratosphere were calculated for each month. We present here the global distribution of average equivalent depth for the two layers 10-4 mb and 1.6-0.6 mb for the months September and January (Figs. 1a,b,c,d).

The equivalent depth of ozone in the layer 1.6-0.6 mb (approximately 43-51 km) varies from about 2.5 to 6.5 x 10^{-3} cm STP. As indicated in Fig. 1a,b the ozone concentration in this layer is a minimum in equatorial regions and generally increases poleward during September and January. In September 1967, there was significant geographic variation of the ozone amount in the upper stratosphere in the Southern Hemisphere (spring) with a belt of maximum ozone shown around 50-60° S. During January 1968 the poleward increase in the Northern Hemisphere (winter) is quite strong but there is no significant latitudinal gradient in the Southern Hemisphere (summer).

The amount of ozone in the layer 10-4 mb (approximately 30-36 km) varies from about 25 to 55 (10^{-3} cm STP) and on the average is about 15 percent of the total equivalent ozone in a vertical column down to the ground. During September 1967 the amount of ozone in the middle stratosphere (Fig. 1c) was quite symmetrical with respect to the Equator. Maxima were observed at about 35° N and S with a weak minimum at the Equator. As in the upper layer, the Southern Hemisphere maximum in the layer 10-4 mb was slightly stronger than for the Northern Hemisphere. In both hemispheres the ozone values decreased poleward from their mid-latitude

maxima. In January 1968 (Fig. 1d) the mid-latitude maximum is more pronounced in the Northern than Southern Hemisphere. Also, the January 1968 equatorial minimum was slightly lower than that observed during September 1967.

Analogous data taken from BUV observations for total ozone above 10 mb and 2.8 mb have been shown for a single day 5 July 1970 by Heath et al (1973)[5] and for the layer 10 - 1 mb for January 1971 by Ghazi (1976)[8]. The global patterns shown by Heath et al [5] are similar to those given here although their amounts as shown are unrealistically high. The distribution discussed by Ghazi [8], on the other hand, give ozone amounts almost equivalent to those shown here but the latitudinal gradients are reversed from those shown in Fig. 1d. We do not know the reason for this difference.

Average height-latitude cross sections of ozone mixing ratio ($\mu g/g$) are given in Figs. 2a,b,c for September and November 1967 and January 1968. The hatched areas shown in Figs. 2a,b,c are regions where the data were limited because of large optical depths or low solar elevation angles. It should be noted that the usual assumption made in calibrating balloon ozonesondes - that the ozone mixing ratio is constant above the level of maximum - could result in an overestimate of the ozone concentration by about 5 percent. The ozone mixing ratio is a maximum between 10 and 3 mb (depending on latitude and season) and the strongest vertical gradient occurs between 3 mb and 1 mb during all seasons.

Observations for the three months listed above indicated that in the middle stratosphere (30-40 km) there are fairly pronounced latitudinal variations of ozone mixing ratio with values generally increasing to maxima in mid-latitudes. At levels higher than 1 mb (about 48 km) the latitudinal gradient seemed to be very small. During September the mixing

ratio distribution was rather symmetrical about the Equator but was slightly higher at all levels in the Southern Hemisphere (spring) than the Northern Hemisphere (fall). This slight asymmetry persisted during October (not shown here) only up to about 2 mb. In November 1967 the ozone distribution showed a middle stratosphere maximum at about 5 mb (36 km) at 30° S. The latitudinal variation in the Southern Hemisphere is quite small but the poleward increase in the Northern Hemisphere has become rather pronounced.

During January 1968 the high latitude maximum was quite pronounced in the winter (Northern) Hemisphere and the axis of maximum ozone tilted northward with height.

The mixing ratio distribution in the stratosphere as discussed by Dütsch (1974)[3], Krueger (1973)[9], Watanabe and Tohmatsu (1976)[10] and others, although derived from balloon borne ozonesondes, Umkehr observations and rocketsondes show essentially the same latitude-height-seasonal variation as given in Figs. 2a,b,c. Heights-latitude cross sections calculated from the BUV experiment on Nimbus IV have been discussed by Krueger et al (1973)[11]. Although the mixing ratio cross sections discussed by Krueger et al (1973)[11] were for sample individual days during 1970, comparison shows that the overall patterns are surprisingly similar. In both cases the patterns during September are nearly symmetrical about the Equator whereas the pattern for the cross section corresponding to Northern Hemisphere winter shows much higher mixing ratio values in north polar latitudes and very flat latitudinal gradient in the Southern Hemisphere.

Theoretical models of the height-latitude ozone distribution have been discussed by Park and London (1974)[12], Cunnold et al (1975)[13] and others. Comparisons show that the theoretical distribution agrees better

with the observations up to about 40 km when horizontal and vertical transport is included (Cunnold et al, 1975)[13] but requires more complete photochemistry above 40 km (Park and London, 1974)[12]. Indeed, above 40 km except at high latitudes during the winter, the distribution calculated by Park and London [12] is very close to that observed during January (London et al, 1976)[7].

4. Discussion

The photochemical relaxation times for ozone depends strongly on height and solar zenith angle (therefore latitude and season). The relaxation time varies from a few days to a few months at 35 km and decreases to a few hours at 50 km except at middle and high latitudes during the winter when it can be of the order of a few days. In the upper stratosphere the ozone distribution is expected to be determined mainly by photochemical processes during the summer. Below 40 km (about 50 km at high latitudes during the winter) the ozone distribution results from a combination of photochemical and dynamical processes.

Ozone production depends on the available solar ultraviolet radiation. In the middle and upper stratosphere, this is balanced by destructive processes due to atomic oxygen and to catalytic recombination cycles involving NO_x ($NO + NO_2$) and HO_x ($H + OH + HO_2$) species. Ozone losses are higher with increased NO_x and HO_x abundance. In addition, the photochemical concentrations of ozone is temperature sensitive so that increased temperature results in a decreased ozone amount.

At times of low solar elevation (high latitude during winter) model calculations show minimum values of HO_x in the stratosphere (see, for instance, Nicolet, 1975)[14]. Also, airborne observations by Noxon (1975)[15] have shown a strong wintertime decrease of stratospheric NO_2 poleward of 45° N. The decrease of odd nitrogen and odd hydrogen coupled with the

well known stratospheric temperature decrease during the winter at polar and sub-polar latitudes would result in a relative increase with latitude in the mixing ratio of ozone in the upper stratosphere (about 50 km) during the winter.

A large part of the high latitude ozone increase in the middle stratosphere (30 to 40 km) probably results from the same process as that responsible for the buildup of ozone at high latitudes in the lower stratosphere -- i.e. slantwise eddy transport. In the upper troposphere and lower stratosphere the isentropic surfaces slope downward with increasing latitude and in mid-latitudes there is a net poleward and downward ozone transport at these levels. In the middle and upper stratosphere there is an upward slope of the isentropic surfaces during all seasons except summer, the maximum slope being found at about 35-40 km in mid-latitudes. Upward and poleward eddy transport of ozone in these regions would contribute to the observed ozone increase.

5. Summary

The stratospheric ozone concentrations as derived from OGO-IV satellite observations show amounts and patterns of geographic distributions that are consistent with those reported by rocketsonde and other satellite observations. The results discussed in this paper show that the ozone mixing ratio in the middle and upper stratosphere has significant vertical, geographic and temporal variations. In general, the ozone mixing ratio increases poleward in both hemispheres but especially in the winter hemisphere. During the summer the mixing ratio decreases with height from a maximum at about 30 km with the largest decrease in the layer of about 40-50 km. However, during the winter, the level of maximum ozone mixing ratio seems to tilt upward with latitude. The wintertime ozone mixing ratios in the middle and upper stratosphere are higher than the summer values. The

mixing ratio distributions show strong geographic variations at about 35 km and these variations persist even up to about 50 km, although at the upper levels the variations are very small. Thus even at those levels where photochemistry is dominant, inhomogeneities in temperature and concentrations of important ozone-related species (e.g. N_x, HO_x), and their interaction with motions at these levels certainly significantly affect the ozone distribution.

The observed ozone distribution reported here provides an information base for input and testing of photochemical-transport models of the stratosphere. It should be noted that these data represent average distributions for a particular time period. Some interannual variations are to be expected particularly during the winter at high latitudes. However, the general patterns shown here are likely to be representative of the average seasonal variation of ozone in the middle and upper stratosphere.

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Legends

Fig. 1 The global distribution of the equivalent depth of ozone (10^{-3} cm STP)

a) 10 - 4 mb September 1967

b) 10 - 4 mb January 1968

c) 1.6 - 0.6 mb September 1967

d) 1.6 - 0.6 mb January 1968

Fig. 2 Height-latitude cross section of ozone mixing ratio ($\mu g/g$)

a) September 1967

b) November 1967

c) January 1968

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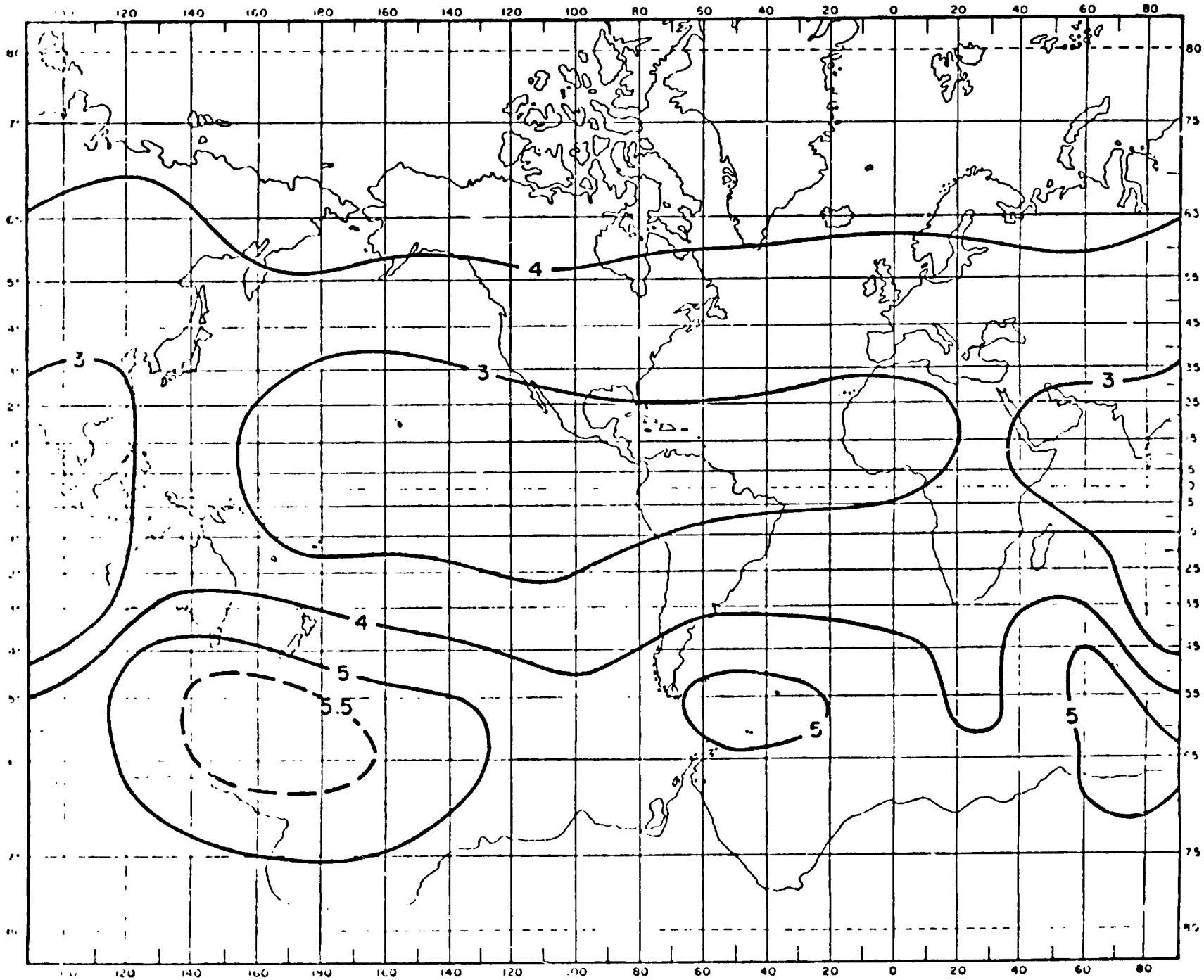

(a) 0.6 - 1.6 mb SEPT 1963

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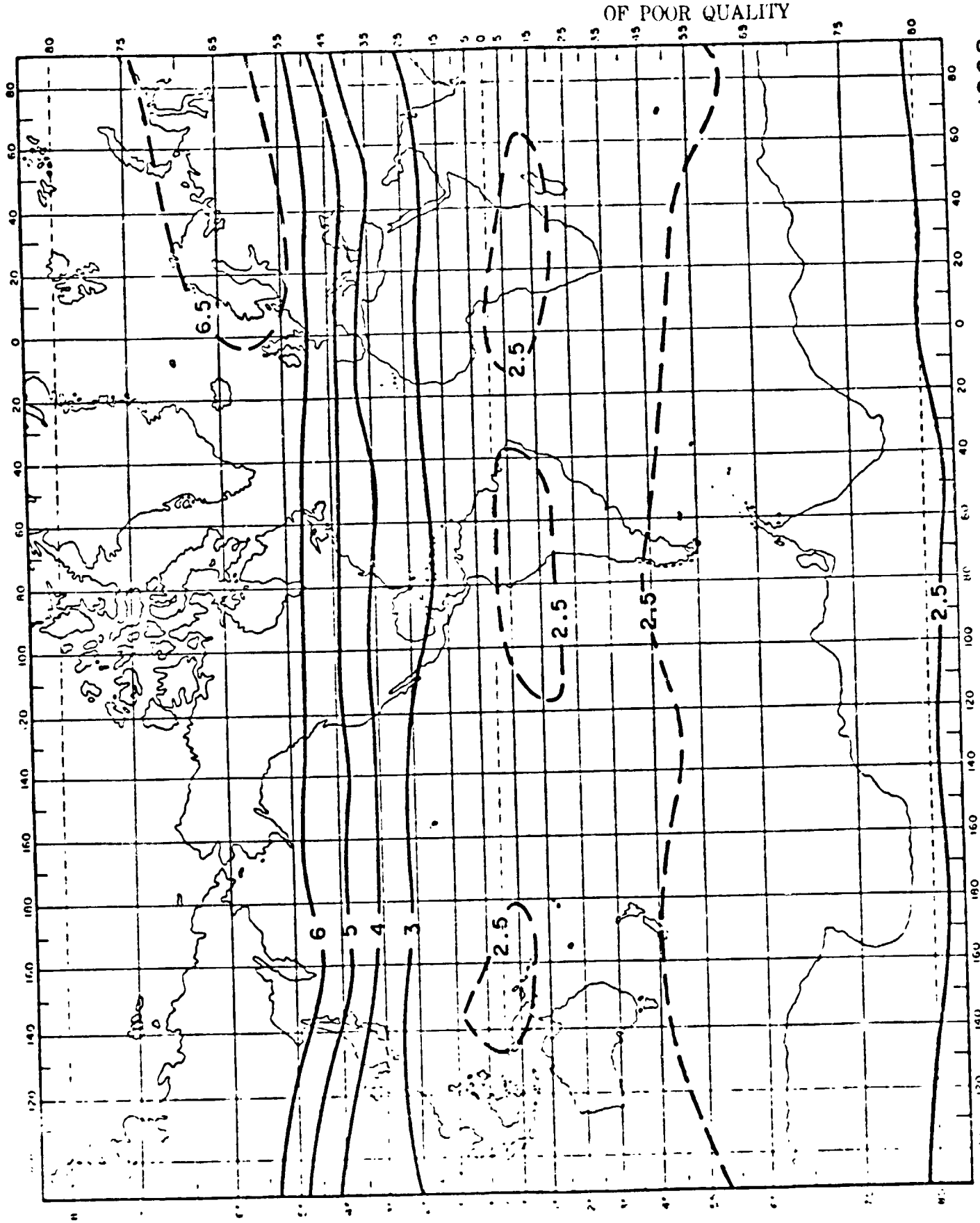
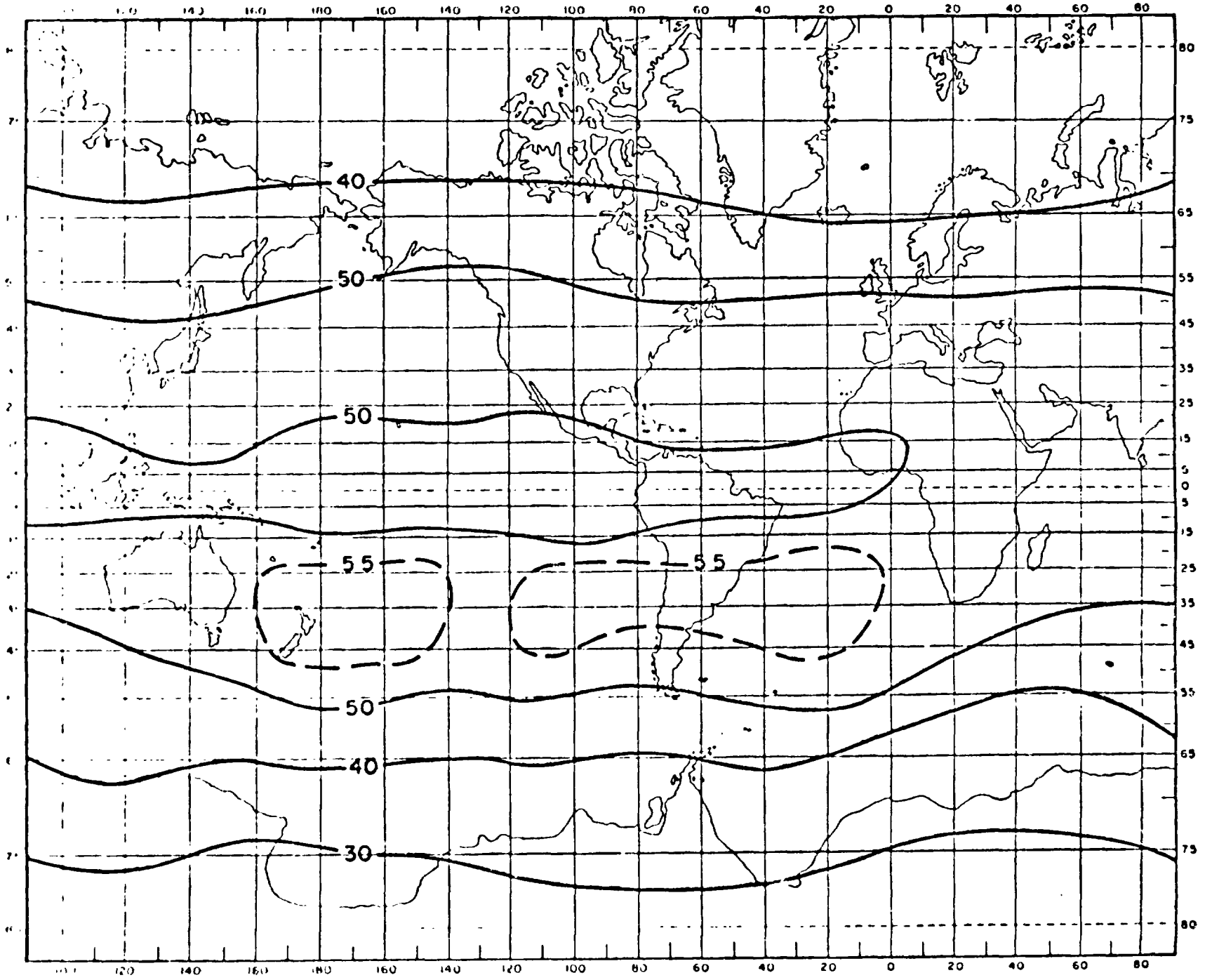
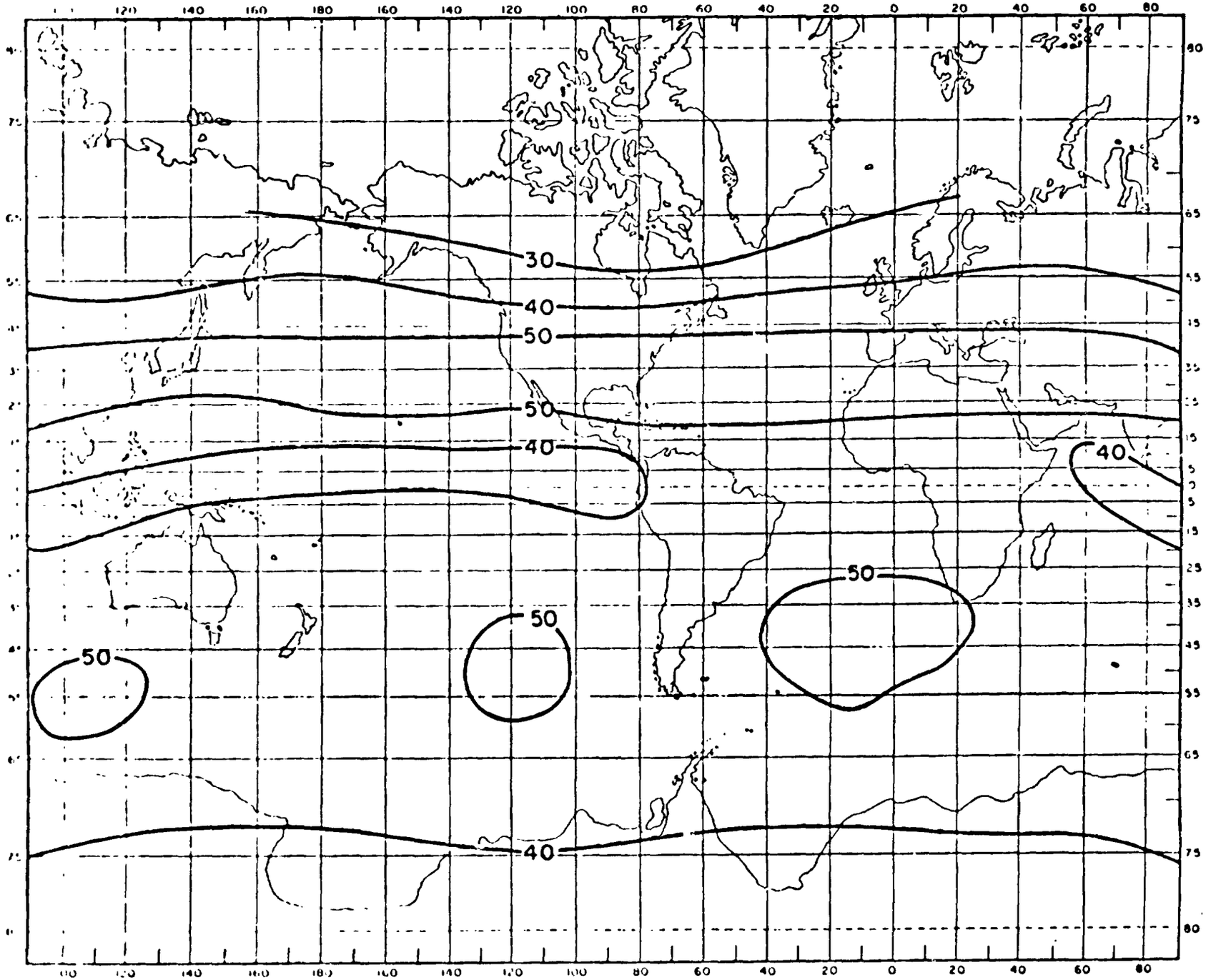


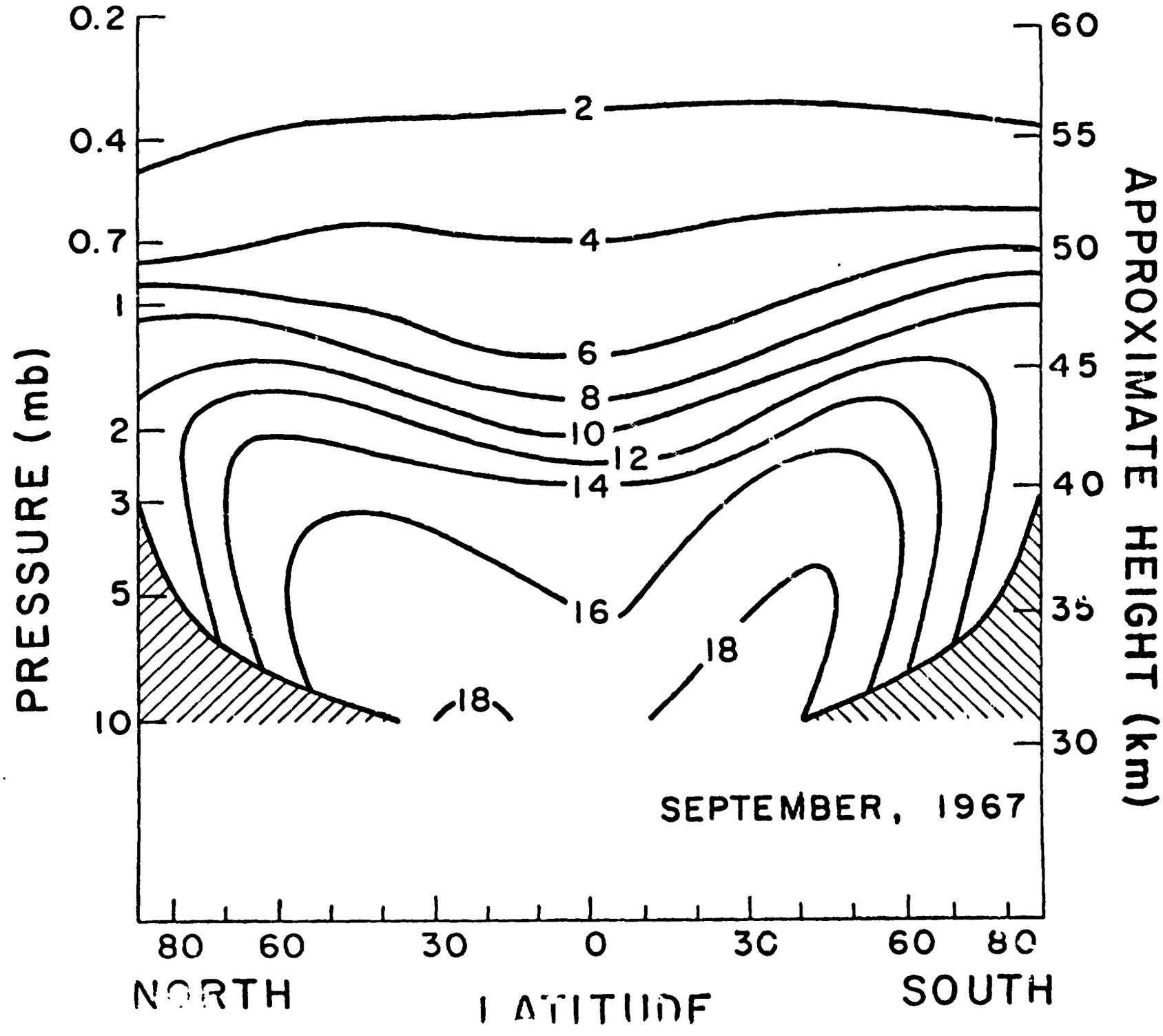
(b) 0.6-1.6mb JAN 1968



(c) 4.0 - 10 mb SEPT 1967



(d) 4.0 - 10 mb JAN 1968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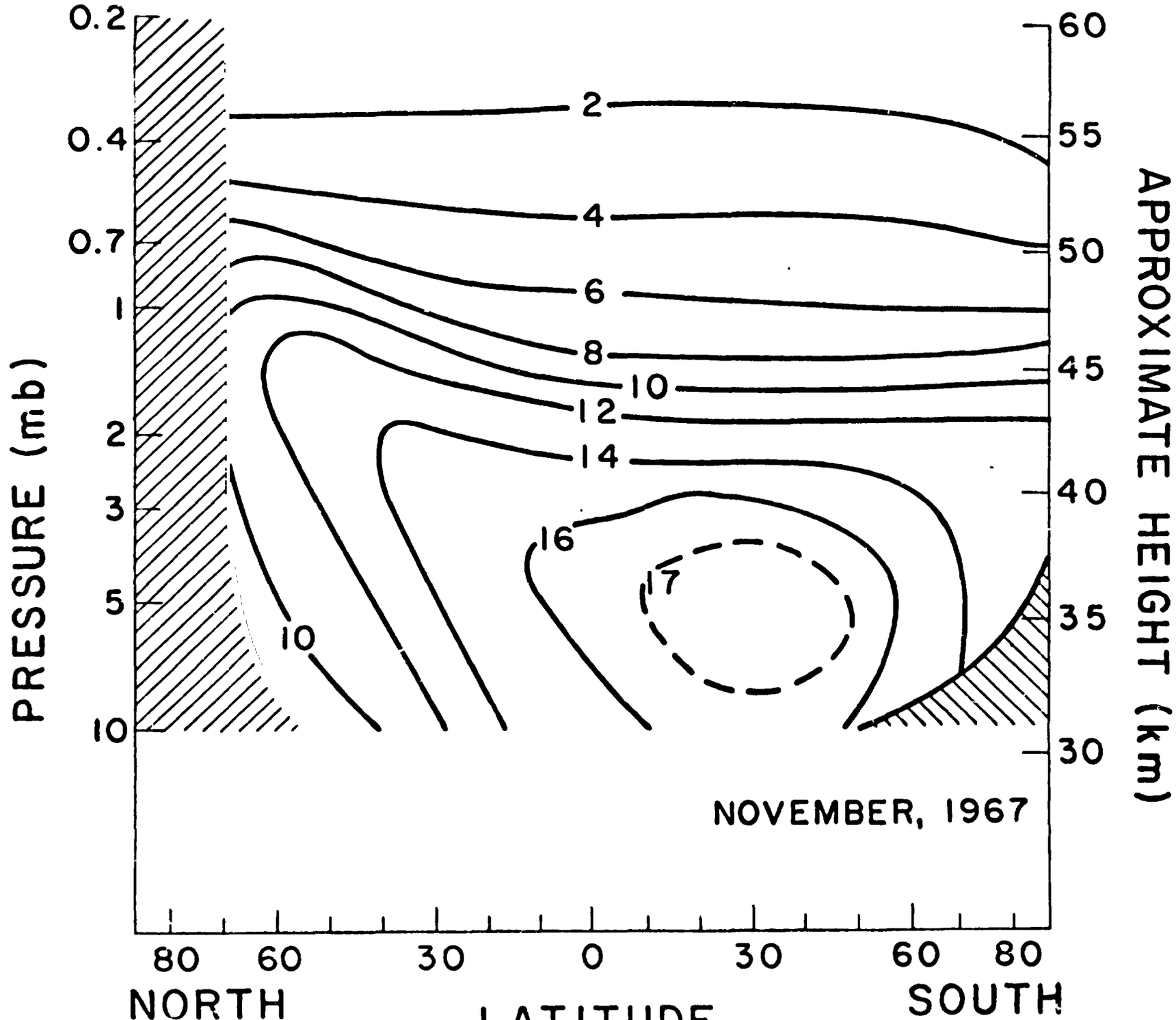
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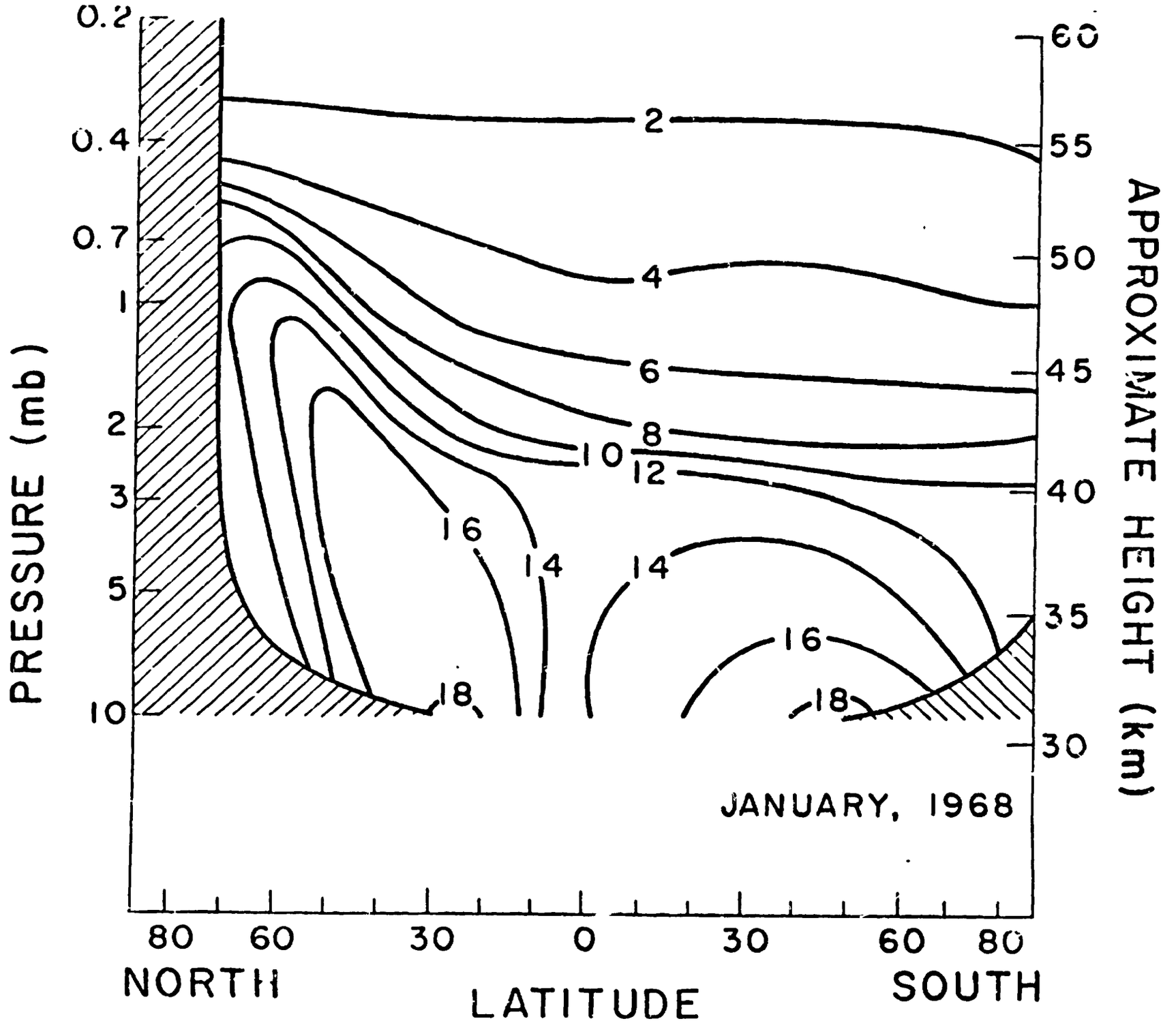
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