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BALANCE OF THE TROPOSPHERIC OZONE AND ITS RELATION TO STRATOSPHERIC INTRUSIONS INDICATED BY COSMOGENIC RADIONUCLIDES

Technical Progress Report

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Abstract

The balance of the tropospheric ozone is investigated considering the ozone sources with emphasis on tropospheric pollutants and stratospheric-tropospheric exchange processes.

The measuring series of ozone concentration from the years 1977 - 1979 obtained at three different levels of the boundary layer (700, 1800, and 3000 m a.s.l.) have been analyzed. In the course of this work the data have been evaluated in correlation with relevant meteorological parameters, for instance solar radiation. It became evident that for the different levels various types of ozone sources must be assumed. At the mountain stations prevails influx of stratospheric ozone. In the valley, however, photochemical production must be regarded as main source.

Experiences with a New Zealand filter photospectrometer are discussed.

A systematic study of ozone profiles obtained by balloon sondes revealed that as a rule after solar flares associated with Forbush effect drastic changes of the ozone profile take place in the lower stratosphere. Then, extremely high maxima of the ozone partial pressure are observed immediately above the tropopause and also intensive influxes of tropospheric air into the stratosphere between 200 and 100 mb.

At mountain stations just above the timberline the amplitude of the CO_2 daily variation due to vegetation is now balanced to such an extent that these measurements can be regarded as representative of the free atmosphere and thus seem to be suited for trend analyses.

Effects of a modified lidar system on measurements of stratospheric aerosol layers and necessary corrections in evaluating the backscatter profiles are discussed and most recent measuring results presented.

1. GENERAL REMARKS

1.1. Scope of Work, Expenditure of Time and Funds

The research performed under DOE-Contract was fully in keeping with the proposed research activities of Modifications A001 - A003.

The entire scope of work was accomplished. Time used by our staff and expenditure of funds correspond to the cost estimate in our proposal of 23 July 1979.

1.2. Essential Features

Activities under contract can be subdivided into several main subjects which will now be described:

I. Continuous recording of ozone concentration at three levels (Zugspitze, 3000 m; Wank, 1800 m; Garmisch-Partenkirchen, 700 m a.s.l.).

II. Continuous aerosol sampling at the station Zugspitze, chemical separation of cosmogenic radionuclides Be7, P32, P33 with simultaneous measurement of concentration.

III. Evaluation of 3-years' measuring series aimed at identifying the sources of tropospheric ozone. Parameterization according to various meteorological aspects.

IV. Meteorological evaluation of radiosonde ascents from Eastern USA to Eastern Europe for selected long-term periods, calculation of isentropic trajectories to the measuring station, statistical evaluation of tropospheric flow conditions, scientific evaluation of the results.

V. Study of the effects of solar events on the fine structure of the ozone profile up to 35 km altitude.

VI. Remote sensing of stratospheric aerosol layers.

VII. Recording of the CO_2-concentration at the stations Wank and Garmisch-Partenkirchen.

Points I and II furnish the data designed to resolve the problem of the CO_2 budget. By comparing the mean diurnal and seasonal variations of tropospheric ozone at three different levels in the lower troposphere the question of its sources is discussed under point III.

Results from IV shall deepen the basic understanding of the stratospheric-tropospheric exchange and provide the means for a large-scale handling, also from the climatological point of view.

The influence of solar events on layers and transport of stratospheric ozone is discussed under point V.

Effects of anthropogenic pollution on stratosphere and troposphere are studied under VI and VII.

2. ROUTINE WORK

2.1. Measuring the Tropospheric Ozone

Measurements of tropospheric ozone at the stations Zugspitze (3000 m a.s.l.), Wank (1800 m a.s.l.), and Garmisch-Partenkirchen (740 m a.s.l.) have been continued without interruption. Comparability has been ensured by regular calibration of the measuring devices.

2.2 Aerosol Sampling at the Zugspitze to Determine Stratospheric Radionuclides

Aerosol sampling at the station Zugspitze to determine tracers for stratospheric air (Be7, P32) was continued without any problems. No difficulties arose with the chemical separation of radionuclides. The radiochemical efficiency, however, could not be exactly determined in the recent past because P32 is hard to get and Be7 no longer obtainable as standard solution.

2.3 Measuring the Total Ozone

Measurement of total ozone with a New Zealand filter photospectrometer was continued. Available are however only data from the CD-wavelength pair inasmuch as the difficulties (which we mentioned in the preceding Technical Report) could not be overcome. A comparison between the measured values and data from the neighboring Dobson stations Arosa and Hohenpeißenberg has shown that in principle the New Zealand filter photospectrometer functions properly (see 4.2).

2.4 Measurement of the Ozone Partial Pressure up to 35 km Altitude

During the reporting period 39 ozone radiosonde ascents have been carried out. These were distributed over 5 series with daily successive flights (8,11,9,4 and 7 ascents). The method described in the preceding Technical Report remained unchanged. A first systematic evaluation is given in this report (see 4.3). The results point a way to the elucidation of the observed intensive variation of the ozone profile in the lower stratosphere.

2.5 CO_2 Recordings

Measurements of the CO_2-concentration at the stations Wank and Garmisch-Partenkirchen have been carried out. The measuring sequences have been calibrated in accordance with the Keeling standard. Thus, they are also comparable on an international basis. A first comprehensive evaluation will be given in section 4.4.

2.6 Remote Sensing of Stratospheric Aerosol

Lidar observations of stratospheric aerosol were continued. The technique used in this work (see Tech. Rep. 79) proved a full success. The evaluation method could be improved. New results are presented in section 4.5.

3. CURRENT EVALUATIONS

3.1 Climatology of Stratospheric Intrusions

The isentropic trajectory analysis of 3 long-term periods from the year 1974 will be concluded in near future. It serves to disclose transport processes in the troposphere. After stratospheric intrusions it is possible to trace the route of transport of stratospheric air to the measuring station. From a comparison with the tropospheric flow conditions during periods without intrusions information is expected about the coupling between stratospheric-tropospheric exchange and large-scale transport between tropopause and boundary layer. Seasonal variations can be explained by the above material. A final account hereon will be given in Annual Report Part IX.

3.2 Calculation of the Stratospheric Residence Time

Measuring series of radionuclides Be7 and P32 are now continuously available for a period of 10 years. Using concentration ratio P32/Be7 we have begun to recalculate the residence time of stratospheric air in the lower stratosphere.

3.3 Correlation between Ozone and Be7 Concentration

Analysis of concentration ratio Be7/O_3 was carried on. More recent results will follow in the next reports.

4. RESULTS

4.1 Daily and Annual Variation of Tropospheric Ozone under Pure Air Conditions at 740, 1780, and 2964 m a.s.l. and its Possible Causes

4.1.1 Introduction

Since 1977, continuous recordings of the local ozone concentration have been taken at mountain stations of small horizontal distance (valley station 740 m, Wank peak 1780 m, and Zugspitze peak nearly 2964 m a.s.l.). The stations are located at the northern edge of the Bavarian Alps and in relatively to extremely pure air. Special care has always been placed on the calibration of instruments.

The following sources of ozone are effective:

i. Influx from higher levels (stratospheric source) down to the lower troposphere by intrusions (R. Reiter et al., 1977; R. Reiter et al., 1977; R. Reiter et al., 1978)

ii. Photochemical production in the boundary layer, even under pure air conditions.

iii. Influx in the boundary layer from remote smog areas into the mountain valley and - possibly - to the mountain stations through vertical convective transport.

4.1.2 Annual Variations Based on Monthly Mean Values

Fig. 1 gives the annual variation based on mean monthly O_3-values from the years 1977 through 1979 for the 3 stations (altitudes indicated above) and for all days of the year irrespective of weather conditions. We find a slightly asymmetrical annual variation with maximum in May but almost sustained high values until July followed by a steep decline until October with values remaining nearly constant till January.

The monthly mean O_3-conc. in the valley (740 m) remains always clearly below that at the mountain stations where the difference between 1780 and 2964 m is insignificant. This means that, on an average, there are no marked additional O_3 sources at an altitude of nearly 3 km which may lead to an enhancement of concentration with height from 2000 m on.

If we select days on which relative sunshine duration exceeded 80% at all stations (Fig. 2, almost cloudless days, no precipitation) we find other conditions:

In the valley (740 m) O_3-conc. are essentially higher from April to June reaching in part the mean values at 2964 m. It is remarkable that from March to July O_3-conc. are higher at 1780 m than at 2964 m. Thus, here exist special conditions obviously connected with increased solar radiation.

4.1.3 Mean Diurnal Variations at Different Solar Radiation

Fig. 3 shows the mean monthly diurnal variations for all days from the years 1977 to 1979 for the 3 mountain stations. Here we notice at once that a strongly pronounced variation appears at 740 m peaking shortly after noon. From January on, the daily maximum value increases currently from month to month and reaches the peak in June/ July. Even from March on until September, O_3-conc. are after noon at 740 m higher than at the 2 mountain stations. Hence it is certain that we have here either an O_3 influx from sources at some distance of the measuring site, which is unlikely, or local O_3 production in the nearground air layer. Daily variations at the mountain stations are very weak pronounced and run oppositely to the daily variations in the valley in summer.

If we view for a better insight in Fig. 4 first days with high relative sunshine duration without precipitation, separately for spring and summer (left half of the fig.), and otherwise days with extremely short duration of sunshine despite light precipitation (right on the fig.), we observe a marked difference. The amplitude of the daily variation in the valley is essentially greater on sunny days (mainly in midsummer) and the O_3 concentration quite considerably exceeds the values at the mountain stations (in summer nearly 60 ppb are reached in the valley). In contrast, we find at noon a weak minimum at the mountain stations.

We have indeed also on days with sparse sunshine an O_3 maximum in the valley (right half, Fig. 4), however, the values are always lower than at the station Zugspitze.

Hence we can conclude the following:

1. The "ozone supply" in the middle and upper troposphere,

which is tapped by stronger convection so that O_3 is transported to the valley, cannot be regarded as source for the high afternoon values in the valley.

2. Stratospheric sources can likewise be ruled out as explanation. With and without sunshine, the O_3 concentration at the Zugspitze remains approximately constant.

At 1780 m it is remarkable that we observe on sunny days, mainly in spring, also in the afternoon increased concentrations of O_3 which lie above the level at 2964 m; this is not the case on days with less intensive sunshine (Fig. 4). Hence it is beyond doubt that in the nearground air layer (thickness of the responsible air layer is presently explored by vertical profile measurements) we have to envisage special processes, namely, as indicated above:

1. Under certain meteorological conditions possibly an influx into the valley from areas with polluted air (to clarify this another special study is being conducted).

2. Photochemical production of O_3 in the nearground air layer in the presence of reactive trace gases (which are now being investigated just as well).

Here it should be interpolated that under certain meteorological conditions at the station Zugspitze we nevertheless can demonstrate an impulsive influx of O_3 when a stratospheric intrusion occurs. Studies are carried out by means of stratospheric radionuclides (see R. Reiter et al., 1977a; R. Reiter et al., 1977b; R. Reiter etal., 1978).

Finally, we look at Figs. 5 and 6. They show the diurnal variations respectively for Zugspitze and valley for all days from 1977 to 1979 with relative sunshine duration in excess of

80% and without precipitation. At the Zugspitze, no daily variation can be perceived, evident is however the initially mentioned annual variation increasing in concentration from Jan on to reach a maximum in Jul followed by a decline. This variation may be explained by the mean frequency of stratospheric intrusions as established also by other investigations at our Institute (with the aid of Be7).

Fig. 6 shows in contrast the extreme variability of O_3 during the day as a function of the month. In July little more than 60 ppb O_3 are reached on such days as a mean value, on individual days the values approach nearly 100 ppb. Since we have here a pure air region such values are remarkable.

4.1.4 Conclusions

Simultaneous recordings at 740, 1780, and 2964 m a.s.l. at neighboring mountain stations provide informative results on different sources of O_3 in pure air regions. At higher levels, the impulsive influx of O_3 from the stratosphere is no doubt determinant but reveals itself only in a distinct annual variation.

At valley level we find both an extremely pronounced diurnal variation whose amplitude is tied to the intensity of solar irradiation and a seasonal influence. Maximum daily amplitudes are found in spring and summer, minimum values correspondingly in the winter months.

4.2. Measurement of Total Ozone with a New Zealand Filter Photospectrometer

As is known from previous reports, a New Zealand filter-photospectrometer is used in Garmisch-Partenkirchen for measuring the total ozone. The acquired data are currently checked by comparions with the neighboring Dobson stations Hohenpeißenberg and Arosa.

4.2.1 Measuring Technique

The filter photometer functions in 6 wavelength ranges with which the X_{AD}, X_{AC}, and X_{CD} values are calculated through ratio and subtraction of the individual channels. Since in February 1978 a drift of the 305,5 nm filter was observed to longer wavelengths, only the X_{CD} value could be determined from this time on. The defective filter was replaced by a new one in October 1979 and the complete instrument was calibrated on 4 consecutive days from sunrise to culmination or sunset in October 1979. First evaluations of data obtained with the new filter showed however a significant trend in the X_{AD} and X_{AC} values. Through a long-term comparison of the counting rates it became evident that the error was due again to a drift and transmission change of the new 305,5 nm filter. Since the beginning of this year the filter is removed and the manufacturer promised to replace it shortly. Hence again, merely the X_{CD} values of the total ozone are available for the entire period since October 1978.

4.2.2 Comparison of X_{CD}-Filter Values with Dobson Values

Due to the low wintertime solar zenith angle, the calibration of October 1979 could be repeated only in March 1980. Further calibrations in March and April confirmed an excellent calibration constant of the X_{CD} wavelength pairs.
The comparison of our measured values and Dobson values from Arosa yielded in the period from October through December 1979, with total ozone values in the range from 200 - 300 DU (Dobson Units) and small time variability in the total ozone content very good agreement. From January 1980 through April 1980 with total ozone values from 300 - 430 DU at high temporal and hence also spatial variability, the total ozone content in Garmisch-Partenkirchen is systematically higher by about 10 DU than at Arosa.

The comparison with the Dobson station Hohenpeißenberg just 38 km away from Garmisch-Partenkirchen supplies in the period from January 1980 through April 1980 on 44 identical days the excellent regression of

$$X_{CD\ GAPA} = X_{AD\ Dobson}$$

at a correlation coefficient of 0,97. During this period the mean difference Garmisch-Partenkirchen/Hohenpeißenberg is 0, the standard deviation of the differences of total ozone values lies at 8,9 DU. From October through December 1979, with a smooth trend of the total ozone content, a mean difference of -1,3 DU emerged at a standard deviation of 5,5 DU.

These results stand for the very good quality of total ozone measurements in the CD range with the filter photospectrometer. By using the 3 total ozone values X_{AD}, X_{AC}, and X_{CD}, results will surely become still more reliable provided that the technical problems with the 305,5 nm filter can be overcome.

4.3 On Periods of Extremely Slight and Extremely Strong Fluctuating Ozone Profiles in the Lower Stratosphere Based on Daily Balloon Radiosonde Soundings

4.3.1 Introduction

For two years we have periodically been conducting daily ozone rawin-radiosonde soundings (5-14 days each, total 146 flights, 19 periods) to at least 30 - 35 km altitude at our Institute in the northern Alps. For the numerical evaluation we use - as far as possible - own measurements of total ozone or such of nearby stations (Hohenpeißenberg, Arosa, for which we are most grateful). All synchronous meteorological rawinsonde data are just as well determined.

An observation during the course of our solar-terrestrial studies (R. Reiter, 1979 a,b,c) prompted us to start this type of ozone flights: After violent solar flares with Forbush effects (R. Reiter, 1979 a,c,d) we found sharp fluctuations of the O_3 concentration in the lower stratosphere, often just above the tropopause. Frequently such patterns persist over several days.

Now we performed specific measurements of that kind. Announcements in URSIGRAM and from European solar and geomagnetic observatories constituted the basis for starting such daily flight series during <u>maximum</u> and <u>minimum</u> solar activity, respectively.

The O_3 profile structures are first analyzed quite irrespective of solar activity from the meteorological point of view considering the following parameters:

Structure of temperature, wind velocity and wind direction in the total atmospheric layer under study and large-scale maps of the topography of the 100, 200, and 300 mb level.

It is attempted to deduce the meteorological preconditions under which the two above mentioned striking patterns of the stratospheric O_3 will occur. Then, we compare the O_3 behavior with solar-geophysical data.

4.3.2 Earlier Observations

R. Hartmannsgruber (1973) has already been able to show that secondary O_3 maxima appear occasionally above the tropopause. The material consisted of one ascent each per week over several years. The author observed 5 days <u>after</u> the appearance of the secondary O_3 maximum in the troposphere a significant warming. This is likely to be a direct consequence of the changes shown in the relative topography of the areas considered which are suggestive of corresponding subsidence motions and hence of temperature increases. These observations have no causal connection with our findings to be described later.

W.F.J. Evans et al. (1979) found during daily to more than once-daily radiosonde ascents regularly a persistent O_3 dip at about 30 mb. Also this observation, interesting though it is, has no relation to the O_3 layers described by us below. We have here by no means an influx of stratospheric air to this level but a horizontal shifting through eddy and transport processes.

4.3.3 Some Examples as Representative Results from the Recent Past

We can present here just a few examples from 146 balloon flights.

1. General references to the figures

Figs. 7 - 10 give in the upper line daily O_3 profiles along with temperature profile (T) and height of the tropopause (TR). The second line indicates the profile of wind speed (V) and wind direction (D).
The last line shows contours of the topography of the 200 mb or 100 mb weather map where our measuring site is marked by a solid dot. Dates for the weather maps begin mostly several days before the radiosonde flights to facilitate presentation of the initial meteorological conditions. Numbers indicate geographic latitude and longitude.
Figs. 11 - 14 give characteristic solar-geophysical data, i.e. total O_3 in Dobson units (TO_3), rel. sunspot number (R, Zürich scale), geomagnetic activity (AK), number of slow neutrons as a measure of galactic cosmic radiation, and solar flux at 10,7 cm wavelength (FL).
The time scale indicates solar flares with respective intensity 1 - 2, where B means bright and N normal.

2. Days with largely normal O_3 profile but superposed fine structure

Figs. 7 and 8 present the more or less normal O_3 profile. In addition, it is evident from Figs. 11 and 12 that the plotted solar-geophysical parameters show no marked changes. Especially N (dashed curve) shows no Forbush effect (displacement of galactic neutrons through plasma clouds from the sun). The same holds for Fig. 12 although in this period various solar flares have been observed.

Concerning these two periods (Jul 79 and Aug 79) the following can be said, meteorologically: Although the O_3 profiles are, in essence, of normal character, some interesting fine structures can be identified which we will come to know as well-defined effects in section 3.

July 1979 (Fig. 7): Initially, we have a typical zonal flow in the stratosphere. However, on the 20th appears in outlines an O_3 peak at about 200 mb with a minimum above it. The 100 mb level (not plotted) shows a slightly anticyclonic flow pattern with wind maximum north of our measuring area. Thus, with persisting vorticity, an influx - even though weak - of tropospheric air is possible at 100 mb. Following is a distinct stabilization, on some days discernible through a temperature rise in the lowermost stratosphere. A process indicated on July 20 happens again on July 24. Note on July 20 a marked reversal of wind direction from SW to NW at 100 - 250 mb. By that, an influx of stratospheric air from England may well lead to a temporary O_3 enhancement just above the tropopause. From July 20 to July 23 it comes over France to a formation of convergence and thus to an approach of tropospheric air which leads temporarily to a reduction of the O_3 concentration in the lower stratosphere.
Be that as it may, all these are insignificant effects on the background of a largely "normal" O_3 profile.

August 1979 (Fig. 8): The O_3 profile reveals from 23rd to 26th an increase in the O_3 conc. just above the tropopause. Aloft we find the normal profile.
The weather situation is characterized by a cut-off low over southern France extending to the Alpine region. On Aug. 24, SW-flow largely prevails at 200 mb where our range of operation lies on the cyclonic side and within a well-defined jet stream through which an influx of O_3 right above the tropopause associated with subsidence seems plausible (see temperature rise above the tropopause).

Yet, it is remarkable that at roughly 30 mb the otherwise normal O_3 profile shows a distinct, though small but stationary, minimum (c.f.W.F.J. Evans et al.). Our wind recordings at this level (changes of wind direction) are in reasonable agreement with those in the above mentioned publication. Also a look at the 100 mb map (not plotted) makes it likely that ozone-poor air in the upper stratosphere was brought in from Northwest Africa which may have led to this O_3 depression. Certainly we have here to do with large-scale transport processes.
In all, we can regard these two examples as typical, normal cases.

January 1980 (Fig. 9): O_3 profiles are characterized as follows: On Jan 8 and 13, profiles are about normal with relatively high values at 50 mb corresponding to the season. Between these days we find however high O_3 values above the tropopause and minimum values at about 100 mb. The meteorological conditions can be defined as follows:
At first prevails pronounced NW-current and hence a well marked regular O_3 peak (50 mb). On Jan 7 develops the required meteorological situation to be expected for explaining the future O_3 profiles. On Jan 8 forms an influx of tropospheric air from SW via Spain. On Jan 9, the O_3 profile shows two marked peaks: A positive peak with increased stratospheric O_3 conc. above the tropopause and above it a distinct minimum through tropospheric air flown in from SW. On this

day happens at about 200 mb, induced by a turbulence over western Germany and France, an influx of stratospheric air in a very flat layer from the north. Above that we find a wind reversal and hence a zone providing the means for a massive influx of tropospheric air between 150 and 100 mb. Characteristic of this period from 9 Jan to 11 is that first an O_3 peak occurs at 200 mb which can however be explained by an extrusion in the vicinity of our observational area while the overlying tropospheric air with low O_3 conc. moves advectively to our region from the Atlantic. On Jan 11, the O_3 maximum formed by extrusion has disappeared and only the minimum - due to tropospheric air - is left.

The conditions apparent from the 200 mb map on Jan 11 are still effective in a remaining O_3 minimum of tropospheric origin with a light O_3 maximum below it. The transition from Jan 11 to Jan 13, 1980 in the structure of the O_3 profile can be deduced directly from the meteorological situation in the lower stratosphere and is also logically related to the observed wind speeds and wind directions. The O_3 profile normalized again.

The utmost complex O_3 distribution from 9 - 11 Jan is associated in time with a drastic Forsbush effect (dashed curve N in Fig. 13).

Based on our former experiences we may assume that the complex and extraordinary flow conditions described here are correlated with solar activity.

Feburary 1979 (Fig. 10): In Fig. 10 we have to do again with exceptionally extreme O_3 profiles. At first, on Feb. 21, we find merely a profile which is about normal but already marked by an O_3 peak at roughly 150 mb. This peak becomes more and more pronounced until Feb. 25, but at the same time develops above it at 100 mb a deep depression in the O_3 conc. Both events, peak and minimum, disappear all of a sudden from 25 to 26 February.

The meteorological course of events in the lower stratosphere is as follows: Still on Feb 18, we find no suggestion of any changes in the lower stratosphere. The same holds still for the 19th although formation of a high-level ridge is indicated. The O_3 peak on the 21st and 22nd at 150 mb results from a subsidence process of stratospheric air above the tropopause (see temperature rise above the tropopause) due to a cut-off low over Germany and Italy, across the Alps. On account of a high-level wedge over the North Sea develops an influx of tropospheric air from southern latitudes of the Atlantic west of Spain. The result is a drastic formation of an O_3 minimum from the 23rd to the 24th at 100 mb due to tropospheric air with a travel time of 1 1/2 days. On the 24th forms all over Europe an anticyclonic zone with distinctly tropospheric air at 100 mb providing simulteneously the means for an influx of stratospheric air over eastern to middle Europe at 200 mb. Temperature shows on the 24th a significant decline between 200 and 100 mb which is likewise indicative of tropospheric air. From the extent of this temperature change, the soil temperature may be estimated at +25^{o} (in February!). Temperature behavior between 200 and 100 mb on the 24th and just as well temperature rise over the remaining O_3 maximum on the 25th are in logical relation to the O_3 profile. According to the large-scale weather situation, the tropospheric ozone-poor air between 100 and 150 mb might well be subtropical air from the Bermuda region. Towards the 25th, the tropospheric air is gradually supplanted, nevertheless happens a vigorous stratospheric intrusion with high O_3 conc. corresponding to the conditions on the 200 mb map. This is also in keeping with a drastic cooling at tropopause level (invasion of fresh polar air - see weather map 200 mb, 25. 02). However, this O_3 influx from high latitudes is blocked already on the 26th and 27th (weather map) so that a normal profile appears again.

Finally, it follows from Fig. 14 that we have also in this period a well pronounced Forbush-effect, apart from a plenty of solar flares.

4.3.4 Conclusions

O_3 balloon flights undertaken on a day-to-day basis have shown that a) the stratospheric O_3 profile may drastically change within 24 hrs, b) but may also maintain its normal profile over a prolonged period of time. For a study of a) daily O_3 profile data are the minimum requirement.

Frequently we find not only high enrichment of O_3 just above the tropopause (as a consequence of subsidence motions or influx from higher latitudes) which exceeds by far the "normal peak" at about 50 mb but also injections of tropospheric air between 100 and 200 mb from lower latitudes corresponding to the flow conditions. Drastic changes in the profile caused by often abrupt modifications in the large-scale flow conditions between 300 and 100 mb appear as a rule in connection with solar events (violent solar flares with simultaneous Forbush effects).

4.4 Results of CO_2-Recordings Obtained in the Year 1978 at our Stations Garmisch-Partenkirchen and Wank Peak

Fig. 15 shows the diurnal variations (hourly means) obtained at the valley station during the months of January-December 1978.
One clearly recognizes at the first sight that during the winter months (December-March) when the soil in the valley is completely or to large extent covered with snow, trees are leafless and temperatures comparatively low, the daily variation of the CO_2 is but weakly pronounced. Nevertheless one observes a striking maximum accentuated in the figure through hatching.
From spring (April) to midsummer (August) the CO_2-concentration

forms a "trough-shaped" depression beginning at sunrise and followed by a minimum at late noon. After sunset the values increase steeply again. In transition from summer to fall and winter this "trough" disappears more and more but nevertheless it is clearly perceived still in November. The absolutely highest values are found during the night hours and the absolutely lowest values in the afternoon.
There is no doubt that this "trough" (Fig. 15) in the daily course of the CO_2 which develops conspicuously in summer between spring and fall results from the alteration between CO_2-formation by respiration at night and netphotosynthesis during daytime. It is remarkable that we perceive at sunrise, especially in the months from May to August, a sharp kink and that - at the same time - the photosynthetic CO_2-decomposition does not occur at the highest position of the sun but in late afternoon. Here it seems obvious that both solar radiation and daily variation of the atmospheric temperature play a determinant role.
In Fig. 16 we have selected 3 typical daily variations (January, April and August). These again, give clear evidence that in August the maximum of the CO_2-concentration (355 ppm) is reached until daybreak. At sunrise the values drop rapidly. The strongest activity of photosynthesis and thus reduction of CO_2 is perceived at 14,00 with a minimum of 313 ppm; the daily mean value in August comes up to 330,6 ppm, the daily amplitude to 43 ppm.
An appreciably smoother course is found in case of the daily variation of the CO_2 in April: There is a long, constant plateau with a CO_2 maximum persisting until sunrise after 05.00, followed by a weak yet clearly distinct minimum in the early afternoon.
In winter, the daily variation is completely different. Fig. 16 shows this in detail for the month of January. The nighttime production of CO_2 by respiration and decay of humus declines practically to zero from summer to winter. Also an effect of

net-photosynthesis can hardly be perceived in January meaning that due to the low temperature, mainly frost and snow, all biological processes come to a standstill. The maximum between 07.00 and 11.00 deserves specific interpretation. It is found at about the same time, as mentioned before, uniformly in all cold months (Fig. 15). Here we resort to Fig. 17. We recognize maxima, emphasized through hatching, also in the SO_2-concentration. These are easy to explain meteorologically. During the respective period the aerosol plume enriched with waste gases from oil and coal burning is transported from the town of Garmisch-Partenkirchen across the measuring station by the developing valley wind. This gives rise to a temporary peak in the SO_2-concentration and, consequently, the CO_2-concentration peaks at the same time. However, while the SO_2-maxima are significant (amplitude in the order of 50 - 100%), the anthropogenic contribution to the CO_2-concentration is extremely small (~ 1% only).

This observation is of value because it shows that in our area the anthropogenic production of CO_2 in the cold season, considered in the absolute, is pratically to be neglected and plays no role whatever from the moderately warm to the hot season. Here - at the valley station - the biological processes prevail exclusively as factors influencing the CO_2-concentration.

Fig. 18 shows the diurnal CO_2 variations at our mountain station Wank. Although this station lies only 1000 m higher and is at times even in the exchange layer, we notice there just an extremely weak variation and that only during the warm season from June to September. Then, however, it corresponds in type to the daily course in the valley but the amplitude is much smaller.

Despite the sharp daily variations in the valley during the warm season, the annual mean value of the CO_2 (337 ppm) at the valley station does not differ essentially from the annual mean (331 ppm) at the mountain station. Now, whereas due to

the position of the station relative to altitude and vegetation at 1780 m we lack in Fig. 18 a marked daily variation (an anthropogenically-induced effect is absent altogether,we clearly perceive at the same level - as evident from Fig.19 - the annual variation. It should be mentioned here that a representative annual variation could not be derived so far from the recordings in the valley. This is easily understood since processes like respiration and photosynthesis, as well as decay of humus are subject to drastic seasonal and weather-dependent fluctuations so that one single year of recordings could merely yield a more or less fortuitous variation. In contrast - because of the absence of a diurnal variation - we have at the station Wank a well-pronounced annual variation. The maximum values are found in the cold season, the lowest values in late summer. This trend can be explained as follows: In the cold season the station is mostly above the exchange layer and is therefore coupled to global conditions.
Thus, for instance, the winter value of 337 ppm corresponds positively to that which presently in winter is measured also on Mt. Mauna Loa.
The stronger the warming, and the more frequent the station Wank is in the convection layer - i.e. progressively from March to about August - the lower become the concentrations there. Since already in October due to frequent inversions the valley station is largely separated from the mountain station as regards the vertical exchange, the CO_2 values on the Wank jump steeply just between the two months September and October. Because of the negligible importance of the activity of vegetation in the vicinity of our measuring station (consider also the prolonged periods of winter character there) it is possible to derive a distinct annual variation and arrive just as well at global CO_2 conditions.
In conclusion, it appears expedient to us to gather experiences at alpine stations through simultaneous recordings of the CO_2 concentration at different altitudes and under varying

climatological and meteorological conditions in order to facilitate recognition of sources, sinks, and fluxes, as well as trends of the CO_2 and thus make a contribution to the current, very crucial question: Is it likely that our climate will alter on account of a change in the CO_2 concentration?
In view of the low population density and the almost entire absence of CO_2 producing industry we believe that recordings of the CO_2 at measuring stations in the Alps are, in addition to the advantages mentioned, of particular benefit.

4.5 Lidar Monitoring of the Stratospheric Aerosol Layer

4.5.1 Noise Produced by Strong Backscattering Signals

The 64-channel photon counter which had been installed in our lidar system in May 1979 not only produced better aerosol backscattering profiles but also raised new problems. These problems became apparent after the increase of measuring precision of the new counting system. Effects known as non-linear response of gated photomultipliers or signal-induced noise had to be investigated with the detector system in use. The tests performed were utilizing the complete lidar system under naturally occurring measuring situations.

The detector system is provided with a gated photomultiplier, i.e. the photomultiplier high voltage is switched to its normal value only during a period when the counting channels are wanted for recording a signal. By delaying this gate after the laser pulse emission the strong close-range signal can be suppressed and the backscattered signal can be observed from defined altitudes. The tests performed were to investigate the influence of very high backscattering signals on the subsequent signal during the gate period and during the delay period. In the case of such effects present the possibilities of their reduction and correction were to be examined.

4.5.1.1 Strong Backscattering Signals During the Gate Period

The influence of a very strong backscattering signal at the beginning of the gate (measuring) period on the subsequent signal could be studied during the following good test situations: night periods with clear sky and periods with (very transparent) cirrus layers relieving each other. (Due to the high background radiation by the blue sky stratospheric aerosol measurements can be performed only during night time). The delay time was chosen so that the cirrus layer was measured shortly after the opening of the gate. The comparison of measurement series with and without cirrus signals shows that the high signal caused by the cirrus layers results in a noise superposed on the subsequent signal. This noise (about 5% of the total signal) is declining until about 40 µsec after the cirrus signal and is then increasing again (to 5 to 15% of the underlaying signal, depending on the strength of the causing signal). Considering that the aerosol signal is about 5 to 10% of the total signal, this error can amount to more than 100%.

4.5.1.2 Strong Signals During the Delay Period

This test was concerned with a very high signal caused shortly after the laser pulse emission: The scattering and reflection of the outgoing laser pulse at the exit window of the lidar housing during the delay time. Mesurements with the receiver totally covered leaving only a small aperture for the emitted laser pulse, revealed that the signal reflected at the exit window caused a noise signal during the gate period which was initiated at least 40 µsec after the pulse emission. This noise depends on the strength of the causing signal and decreases during the gate period. Removing the exit window reduces this noise to about 1/4 and the remaining noise may be caused by atmospheric scattering in the vicinity of the exit aperture.

A further noise decrease could be achieved by decreasing the photomultiplier high voltage from 2300 to 2200 V. The residual noise is in the order of fractions of photons per channel time (4 µsec).

4.5.1.3 Improvements and Corrections

The tests described show that signal induced noise can be caused by very strong backscattering signals during both the gate period and the delay period. As a consequence the exit window of the lidar housing has been omitted, the photomultiplier high voltage has been reduced to 2200 V, and cirrus clouds, if present, have to be cut off by a properly chosen delay time. The influence of very strong cirrus signals during the delay period is still questionable.
The residual noise caused by the close range signal during the delay period has been determined with sufficient accuracy by normalizing backscattering signals from altitudes with negligible aerosol concentrations to the expected Rayleigh signal. The Rayleigh or molecular signal has been determined from actual radiosonde data. Correction functions obtained in this way have been incorporated in the data evaluation program.

4.5.2. Matching of the Measured Backscattering Profile with the Calculated Molecular Profile

Another consequence of the introduction of the 64-channel photon counter was a reconsideration of the matching level, where measured lidar backscattering profile and calculated molecular return are set equal. Previously this normalization was done at about 15 km, where between the tropopause and the aerosol layer an aerosol minimum had been observed. A great number of investigations by other groups and techniques during the last years indicate that this minimum of the aerosol concentration is not zero and that there are seasonal and geographic variations.

The height level around 25 km proved to be suitable to normalize our lidar profiles, and all measurements since the introduction of the 64-channel photon counter are matched around this level.

4.5.3. Error Calculation

The error calculation has been improved to meet the measuring conditions of the 64-channel counter. The error calculation comprises lidar inherent uncertainties in the

counting rate,
time resolution,
noise correction,
matching,

and uncertainties in the determination of the expected molecular backscattering signal caused by the

radiosonde signal,
transmission changes by aerosols and ozone,
use of a standard atmosphere instead of actual radiosonde data.

4.5.4 Lidar Observations of the Stratospheric Aerosol Layer

During the period reported here 21 profiles of the stratospheric aerosol layer have been recorded. Each profile is composed of three individual profiles received through attenuation filters and unattenuated. These profiles cover the height intervals 9 to 20 km, 14 to 25 km, and 19 to 35 km. Each of the individual profiles is determined by about 150 laser shots.

The vertically integrated backscattering coefficient as a measure of the total load of particulate matter in the stratosphere has been determined as described in the last report. This value shows an increase from about 1.5×10^{-5} [sr^{-1}] before the eruption of the volcano La Soufrière on St. Vincent in April 1979 to values around 3×10^{-5} in July and since then a less steep increase to about 4.5×10^{-5} in April this year.

These values are significantly above the minimum aerosol values in early 1977, 78, and 79. It is likely that these minima are not caused by seasonal fluctuations of the aerosol production and transport in the stratosphere but rather by an input of volcanic material after occasional minor eruptions with a subsequent increase of the backscattering properties of the aerosol layer through particle growth and transport. The eruption of La Soufrière obviously was more violent and more effective in the stratosphere than the volcanism since 1976, although a defined eruption cloud of this event could not be traced.

On May 18 this year the situation most probably has changed and the termination of the period of stratospheric background aerosol can be expected by the eruption of the Mt. St. Helens in Washington state, which is reported to have penetrated into the stratosphere with a substantial eruption cloud. Since then our lidar system is monitoring the stratosphere, the only interuptions caused by overcast weather conditions. A very time constant backscattering peak at 11-12 km appearing first on May 26 can be the forerunner of an intense increase of the aerosol concentration in the stratosphere.

Future Plans

1. Continuation of ozone recording at three different levels of the boundary layer (700 m, 1800 m, and 3000 m a.s.l.), start of measuring the ozone profile between 1000 m and 3000 m a.s.l. by means of a cable car-borne ozonesonde.

2. Continuation of aerosol sampling at the Zugspitze, analysis of cosmogenic radionuclides Be7, P32, P33 to identify stratospheric intrusions, study of the influx of stratospheric ozone after intrusions, clarification of the sources of tropospheric ozone.

3. Continuation of CO_2 measurements at the stations Wank and Zugspitze.

4. Monitoring of the variation of the ozone profile in the lower stratosphere for selected periods of time by balloonsondes, comparison with values of total ozone measured with a filter photospectrometer, investigations as to a possible control of the ozone layer by solar events.

5. Lidar-observation of the stratospheric aerosol layer after the eruption of volcano Mt. St. Helens, comparison with results from the pre-volcanic layer.

Garmisch-Partenkirchen
July 1, 1980

(Dr. R. Reiter)
Director
Principal Investigator

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Reiter. R.: Influences of solar activity on the exchange intensity between stratosphere and troposphere.
B.M. McCormac and T.A. Seliga (Eds.): Solar-terrestrial influences on weather and climate, 289. D. Reidel Publishing Company, Dordrecht, Holland (1979c)

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Legendes of Figures

Fig. 1: Mean monthly values of ozone concentration measured at the stations Zugspitze (2964 m a.s.l.), Wank (1780 m a.s.l.), and Garmisch-Partenkirchen (740 m a,s.l.). Measuring period 1977 - 1979. To form the mean all days have been used.

Fig. 2: Same as Fig. 1, for forming the mean only days with high solar irradiation have been used (rel. sunshine duration at all stations > 80%, no precipitation).

Fig. 3: Monthly averaged daily variation of ozone concentration for stations Zugspitze, Wank, and Garmisch-Partenkirchen. Measuring period 1977 - 1979, mean formed over all days.

Fig. 4: Daily varitations of ozone concentration at stations Zugspitze, Wank, Garmisch-Partenkirchen.

a. Season: Spring (15 March - 31 May) intense solar irradiation (rel. sunshine duration >80%, no precipitation)

b. Season: Summer (1 June - 30 September) intense solar irradiation (rel. sunshine duration > 80%, no precipitation)

c. Season: Spring (15 March - 31 May) low solar irradiation (sunshine duration < 1 hr, precipitation amount below 2 mm)

d. Season: Summer (1 June - 30 September) low solar irradiation (sunshine duration < 1 hr, precipitation amount below 2 mm).

Fig. 5: Monthly averaged daily variations of ozone concentration at the station Zugspitze for days with intense solar irradiation (rel. sunshine duration > 80%, no precipitation).

Fig. 6: Analogous to Fig. 5 at the station Garmisch-Partenkirchen.

Fig. 7÷10: upper row: Profile of the ozone partial pressure O_3 (nbar) solid line, temperature profile T (^oC) dashed line, and height of the tropopause TR,

mean row: Profile of wind direction D and wind speed V ($msec^{-1}$),

bottom row: Topography of the 200 and 100 mb level, resp., of 50^o W to 60^o E, heavy dot indicates location of the measuring station Zugspitze. Dates are given respectively above the individual profiles.

Fig. 11÷14: Time variation of solar-geophysical parameters. Total ozone TO_3 (Dobson units), rel. sunspot number R (Zürich scale), geomagnetic activity AK, number of slow neutrons N, solar Flux FL at 10.7 cm wavelength . Arrows at the time scale indicate occurrence of flares (B : bright, N : normal).

Fig. 15: Mean CO_2 diurnal variations during the months from January (1) to December (12) at the valley station Garmisch-Partenkirchen in the year 1978.

Fig. 16: Specific view of the diurnal variation in January, April and August. In the warm season extremely strong production at night; by day decomposition

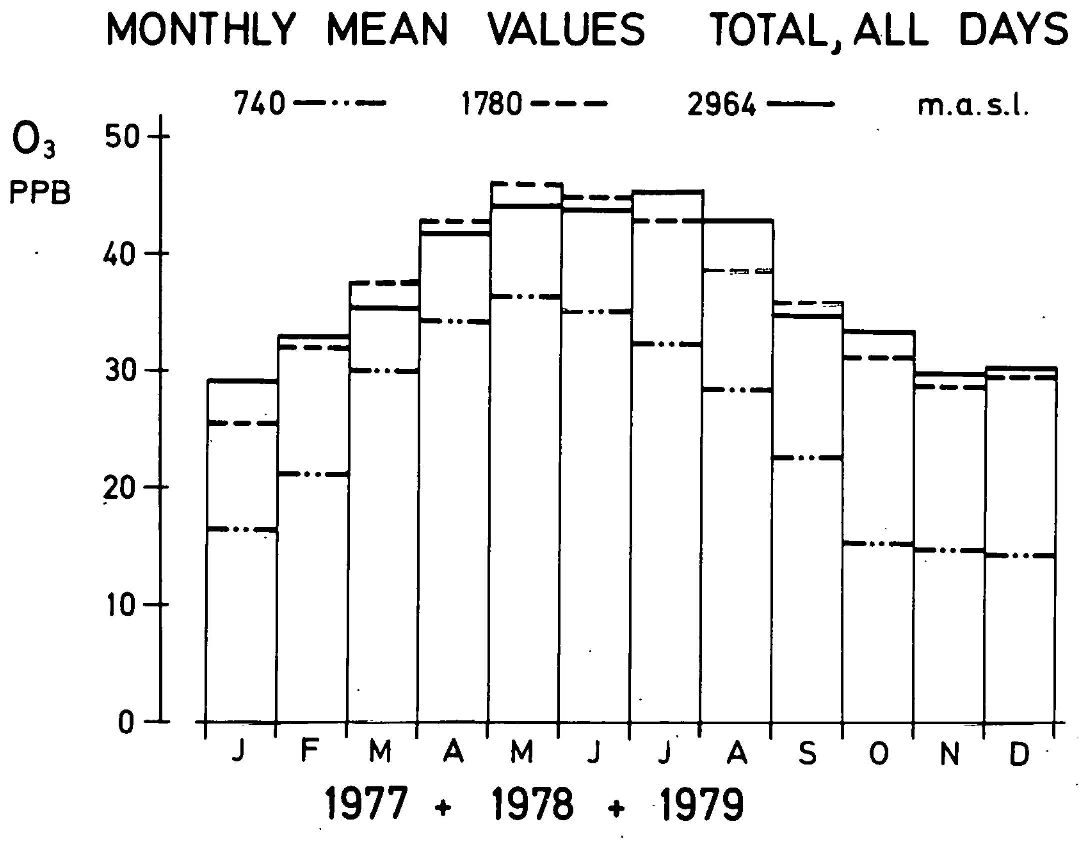
through photosynthesis. In January, biologically-induced variations as good as absent. April-values approx. intermediate.

Fig. 17: Mean diurnal variation of the SO_2-concentration at our station Garmisch-Partenkirchen with maximum in the forenoon during the cold season resulting from influx of the aerosol plume caused by domestic heating and carried from the town to the station through the valley wind.

Fig. 18: Mean diurnal variation of the CO_2-concentration at our mountain station Wank; a summerly daily variation weakly defined from June to September only.

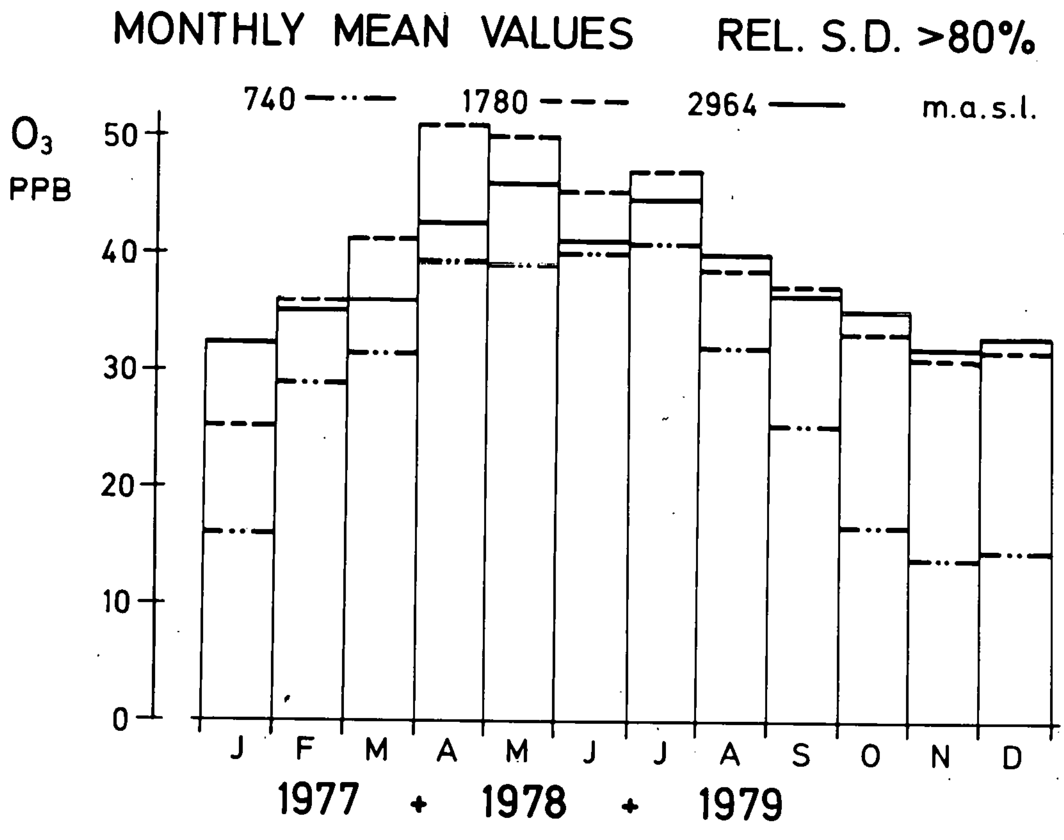
Fig. 19: Annual variation of the CO_2-concentration at Wank. Due to the station's getting progressively into the exchange layer from spring to September, the CO_2-values decline. When the station is separated from the exchange layer (winter and spring) one finds the maximum, globally representative values.

FIGURES



MONTHLY MEAN VALUES TOTAL, ALL DAYS
740 —··— 1780 – – – 2964 —— m.a.s.l.
O_3
PPB
50
40
30
20
10
0
J F M A M J J A S O N D
1977 + 1978 + 1979

FIG. 1



MONTHLY MEAN VALUES REL. S.D. >80%
740
1780
2964
m.a.s.l.
O_3
PPB
50
40
30
20
10
0
J
F
M
A
M
J
J
A
S
O
N
D
1977 + 1978 + 1979

FIG. 2

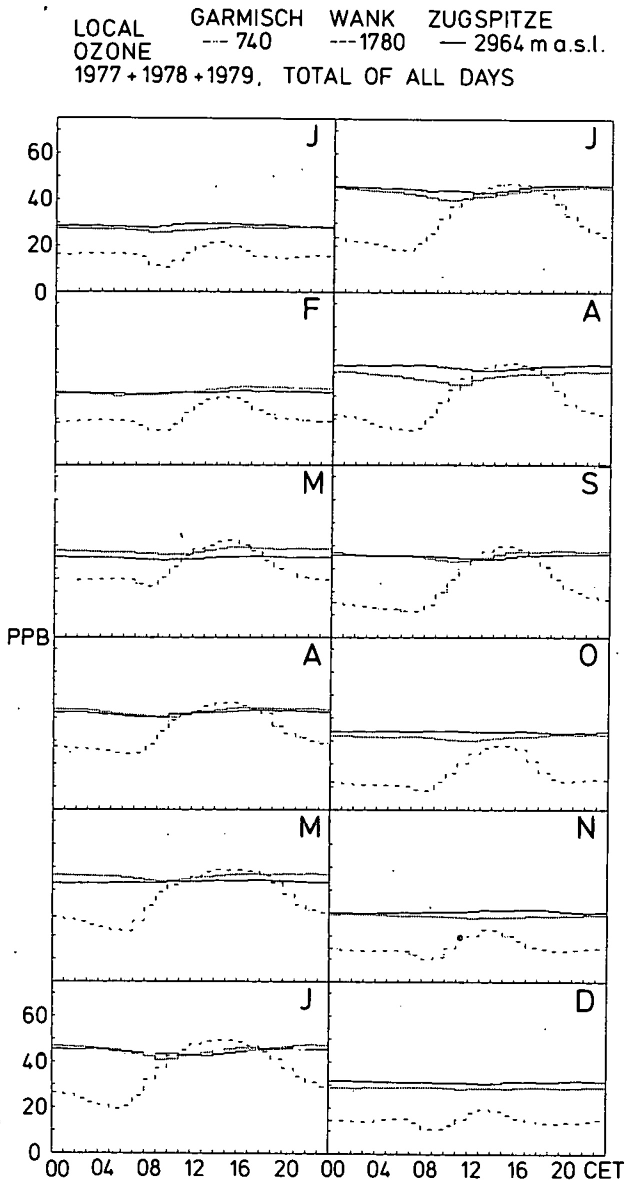

LOCAL OZONE
GARMISCH --- 740
WANK ---1780
ZUGSPITZE — 2964 m a.s.l.
1977 + 1978 + 1979, TOTAL OF ALL DAYS
J
F
M
A
M
J
J
A
S
O
N
D
PPB
0
20
40
60
00 04 08 12 16 20
CET

FIG. 3

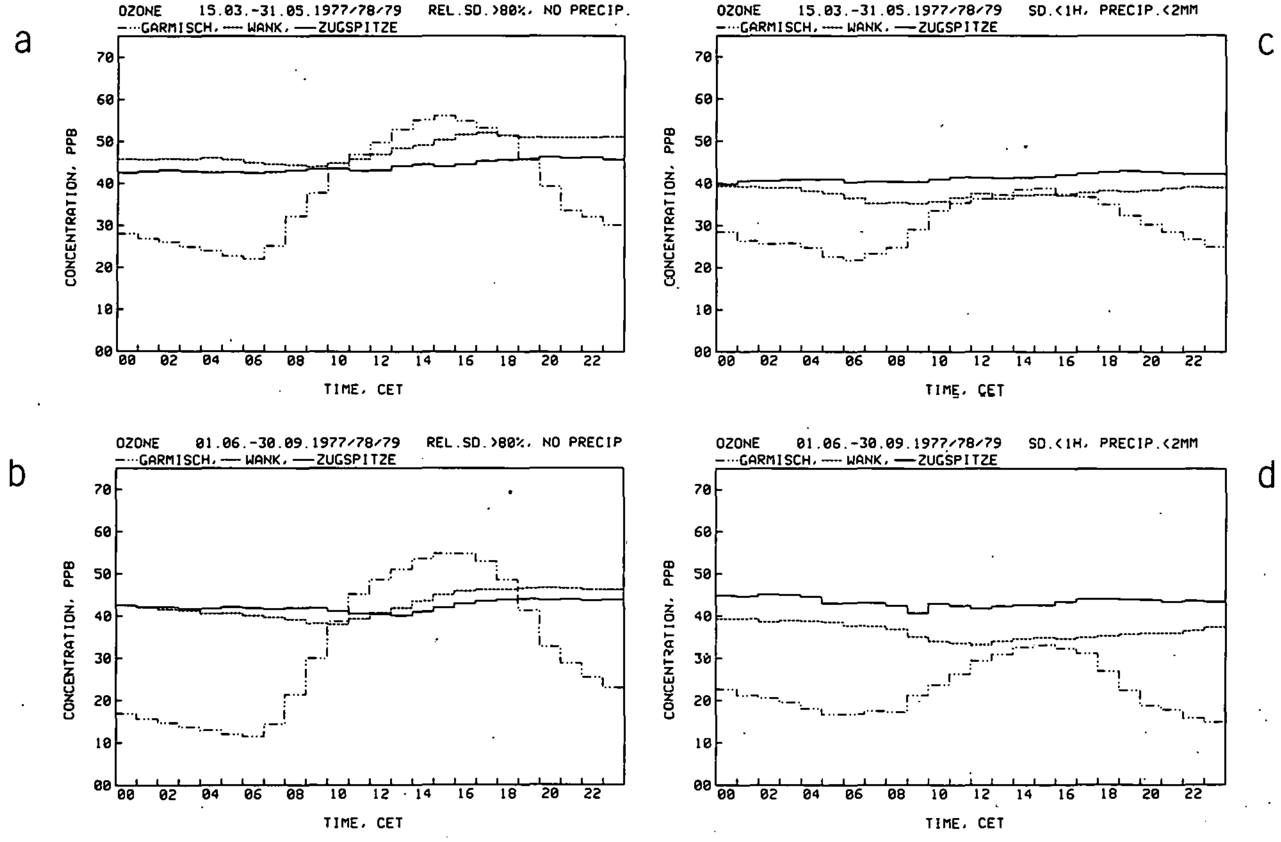

a
OZONE 15.03.-31.05.1977/78/79 REL.SD.>80%, NO PRECIP.
GARMISCH, WANK, ZUGSPITZE
b
OZONE 01.06.-30.09.1977/78/79 REL.SD.>80%, NO PRECIP
GARMISCH, WANK, ZUGSPITZE
c
OZONE 15.03.-31.05.1977/78/79 SD.<1H, PRECIP.<2MM
GARMISCH, WANK, ZUGSPITZE
d
OZONE 01.06.-30.09.1977/78/79 SD.<1H, PRECIP.<2MM
GARMISCH, WANK, ZUGSPITZE
CONCENTRATION, PPB
TIME, CET

FIG. 4

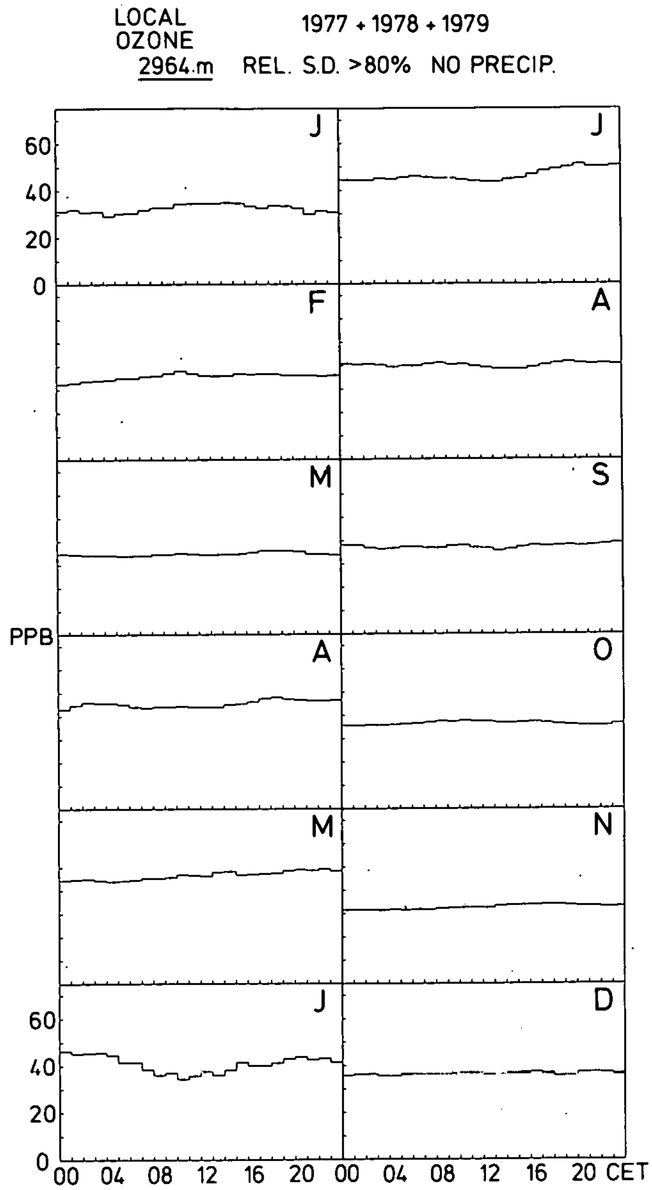

LOCAL
OZONE
2964 m
1977 + 1978 + 1979
REL. S.D. >80% NO PRECIP.
PPB
0
20
40
60
J
F
M
A
M
J
J
A
S
O
N
D
00 04 08 12 16 20 00 04 08 12 16 20 CET

FIG. 5

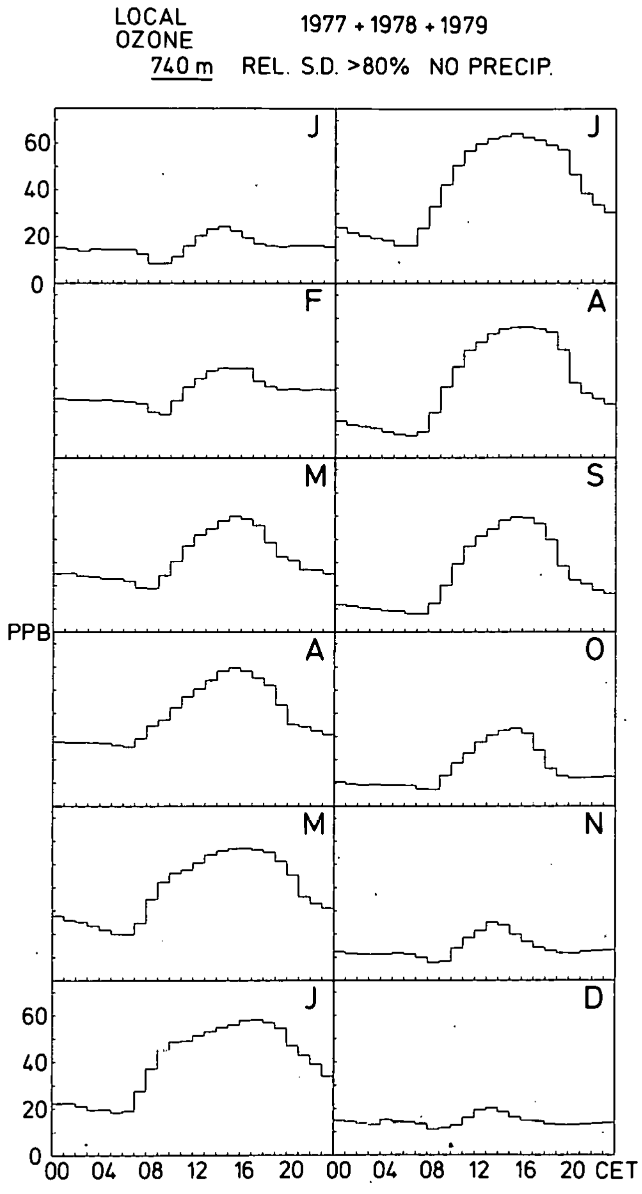
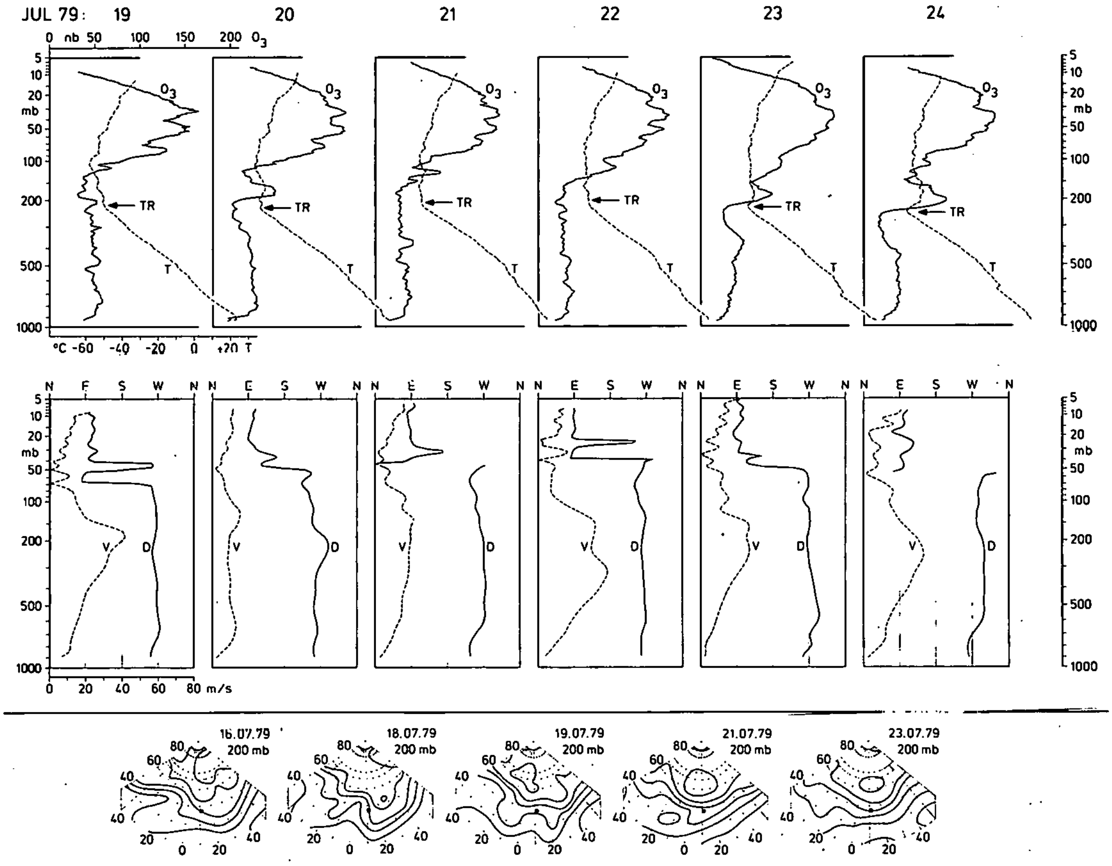

LOCAL
OZONE
740 m
1977 + 1978 + 1979
REL. S.D. >80% NO PRECIP.
J
F
M
A
M
J
J
A
S
O
N
D
PPB
0
20
40
60
00 04 08 12 16 20 00 04 08 12 16 20 CET

FIG. 6



JUL 79: 19
20
21
22
23
24
0 nb 50 100 150 200 O_3
TR
T
°C -60 -40 -20 0 +20 T
N E S W N
V
D
0 20 40 60 80 m/s
mb
16.07.79 200 mb
18.07.79 200 mb
19.07.79 200 mb
21.07.79 200 mb
23.07.79 200 mb

FIG. 7

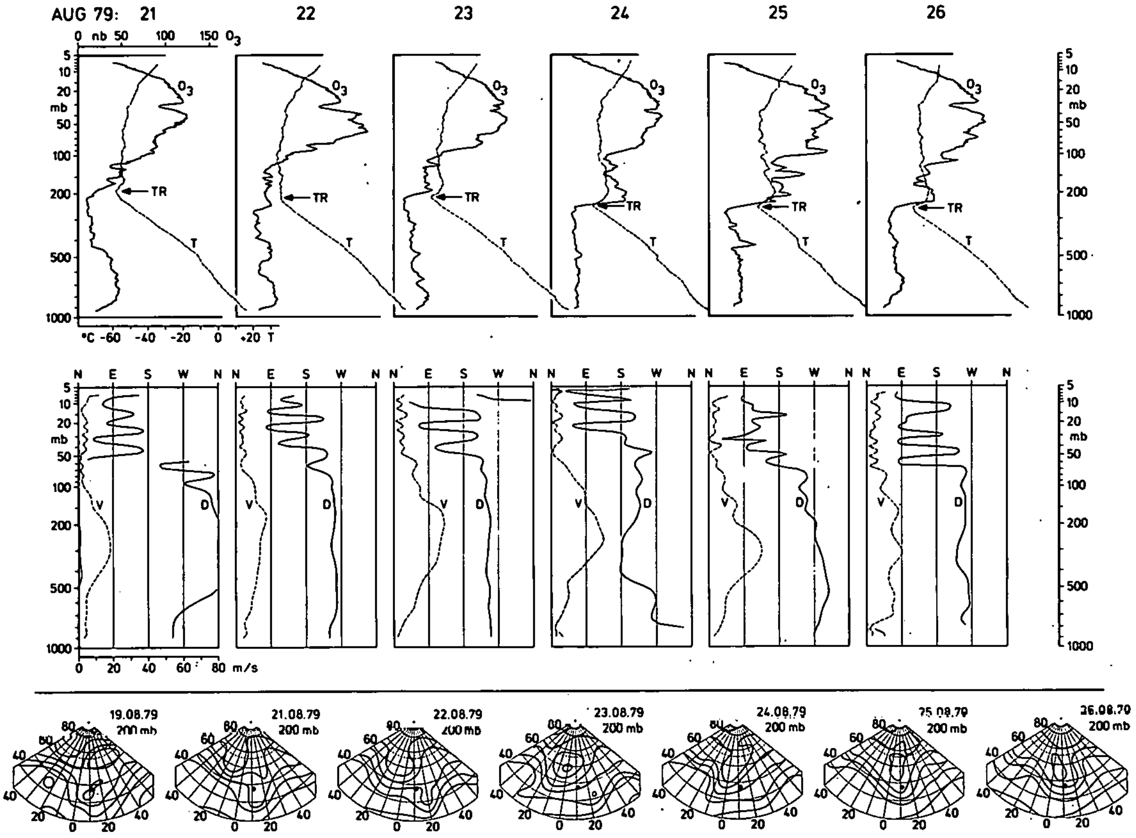
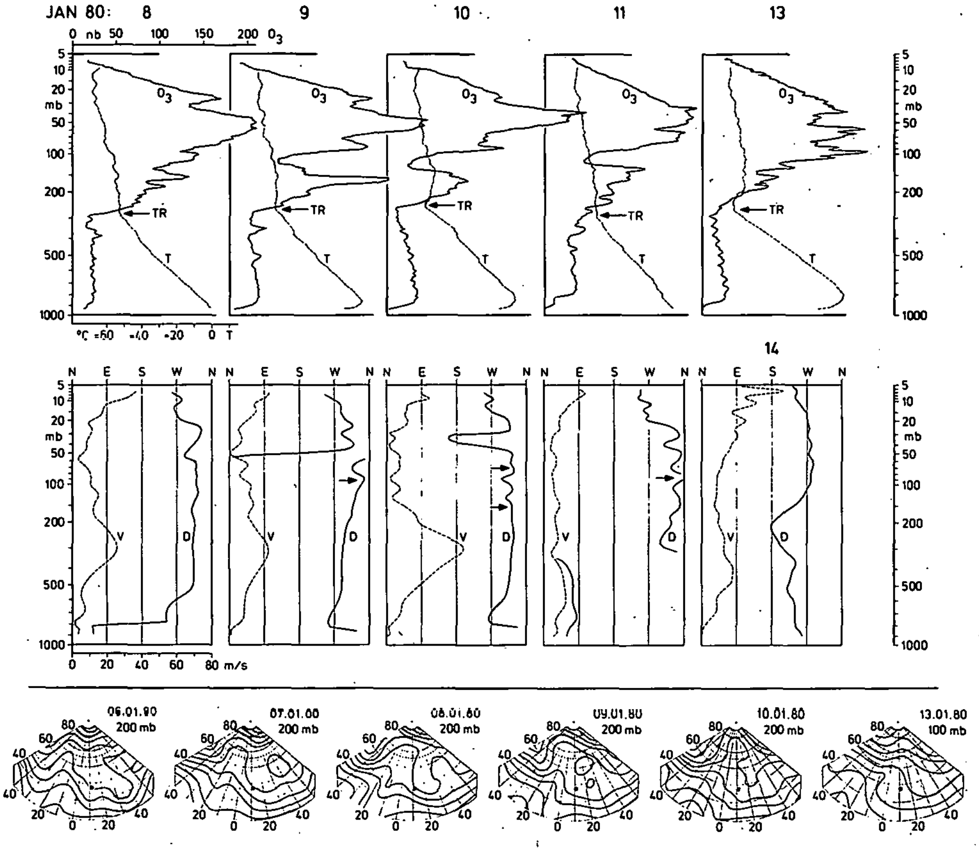

AUG 79: 21
22
23
24
25
26
0 nb 50 100 150 O3
5
10
20
mb
50
100
200
500
1000
TR
T
°C -60 -40 -20 0 +20 T
N E S W N
V
D
0 20 40 60 80 m/s
19.08.79
200 mb
21.08.79
200 mb
22.08.79
200 mb
23.08.79
200 mb
24.08.79
200 mb
25.08.79
200 mb
26.08.79
200 mb

FIG. 8



JAN 80: 8
9
10
11
13
14
0 nb 50 100 150 200 O3
mb
TR
T
°C -60 -40 -20 0 T
N E S W N
V
D
0 20 40 60 80 m/s
06.01.80 200 mb
07.01.80 200 mb
08.01.80 200 mb
09.01.80 200 mb
10.01.80 200 mb
13.01.80 100 mb

FIG. 9

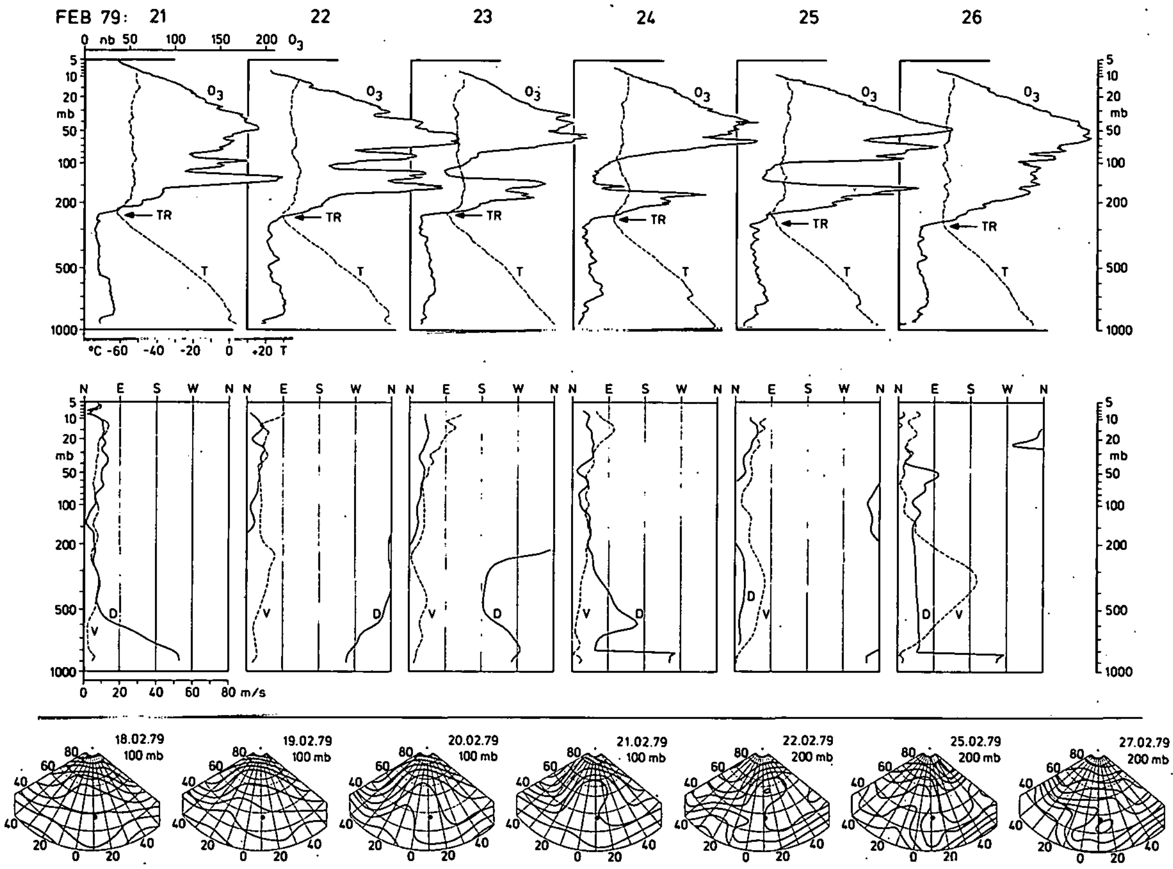

FEB 79: 21 22 23 24 25 26
nb 50 100 150 200 O3
mb
TR
T
°C -60 -40 -20 0 +20 T
N E S W N
D
V
0 20 40 60 80 m/s
18.02.79 100 mb
19.02.79 100 mb
20.02.79 100 mb
21.02.79 100 mb
22.02.79 200 mb
25.02.79 200 mb
27.02.79 200 mb

FIG. 10

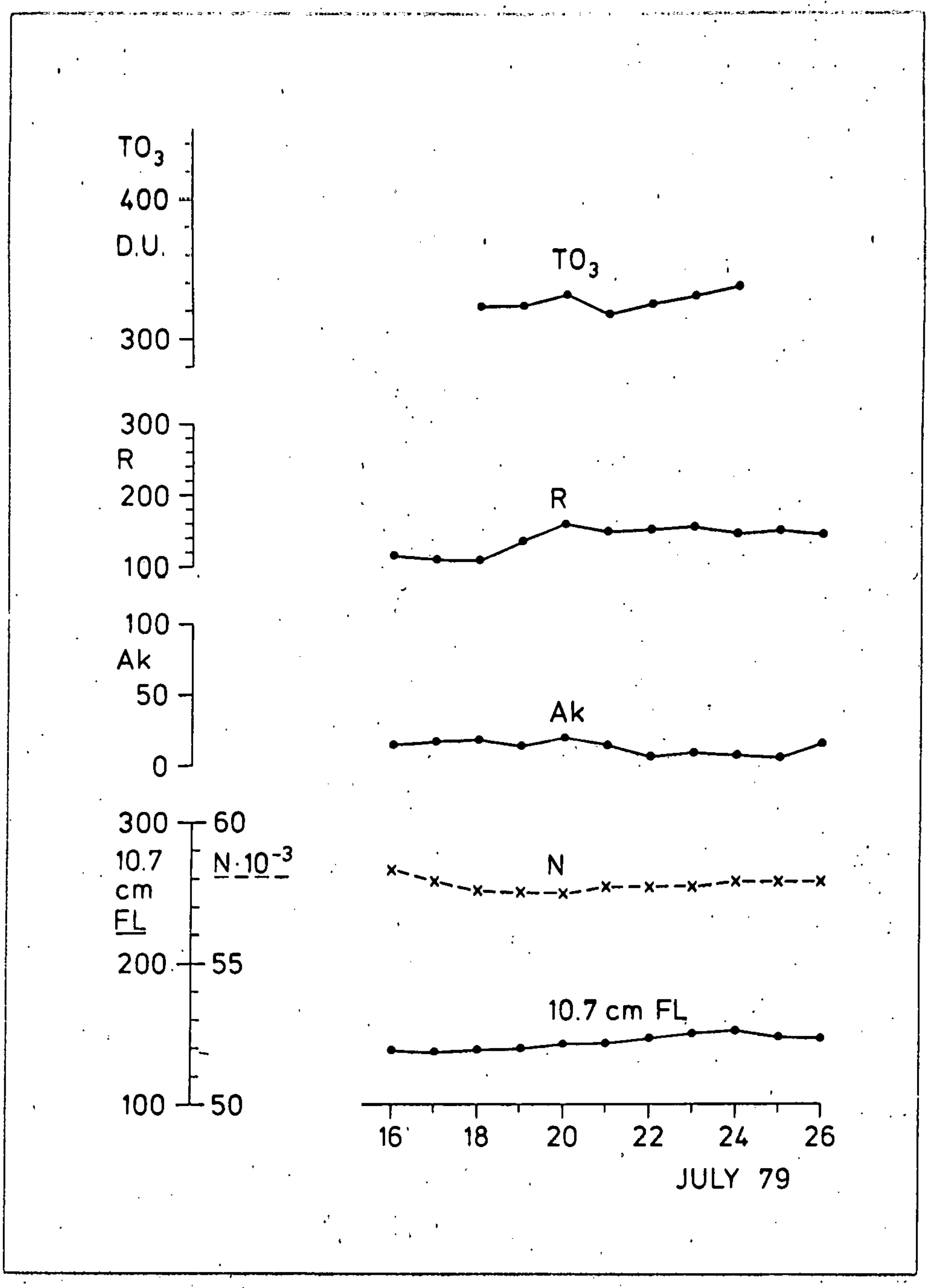

TO3
400
D.U.
300
TO3
300
R
200
100
R
100
Ak
50
0
Ak
300
10.7
cm
FL
200
100
60
N·10^-3
55
50
N
10.7 cm FL
16
18
20
22
24
26
JULY 79

FIG. 11

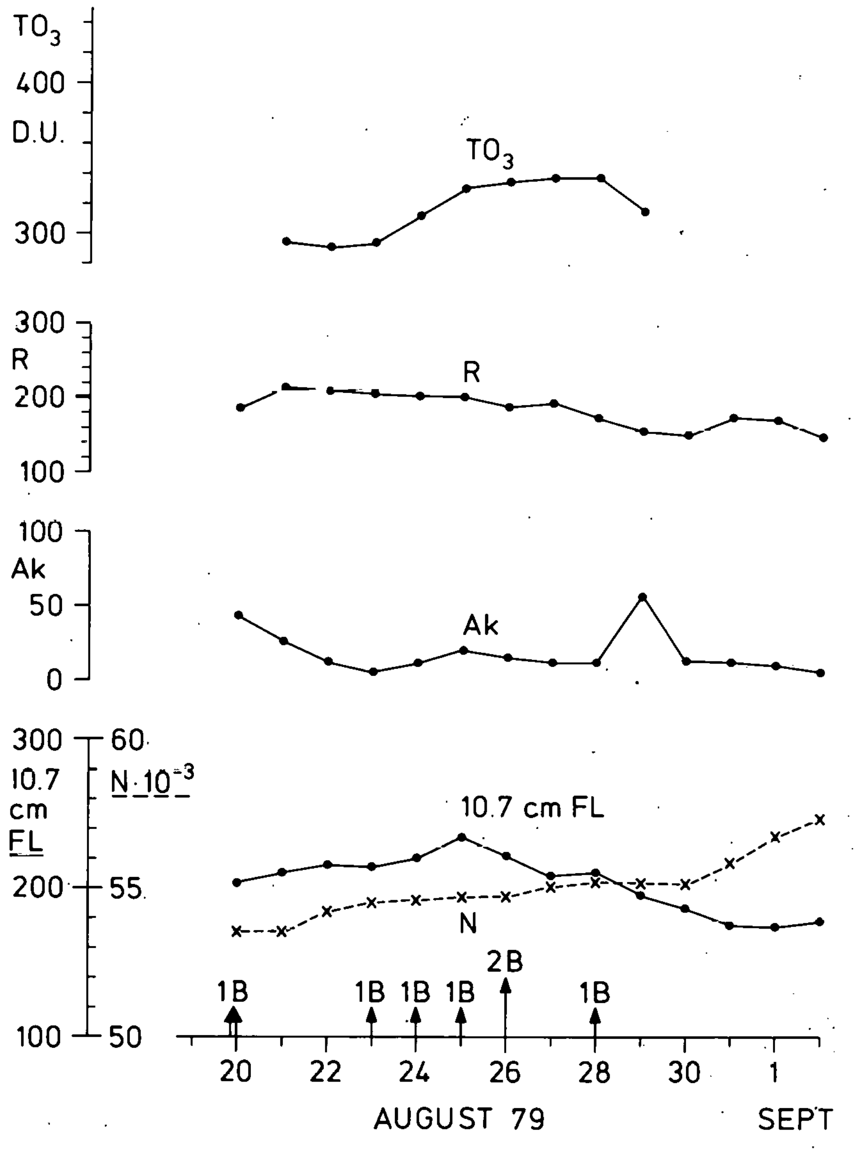
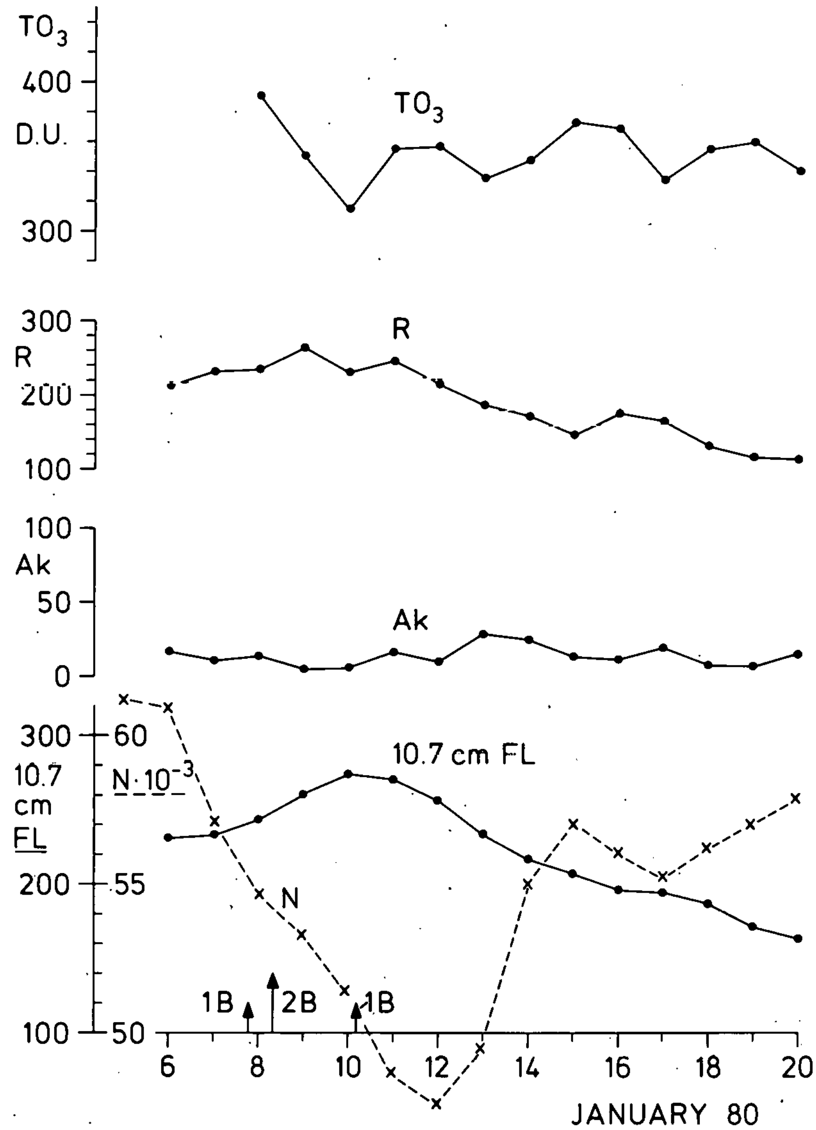
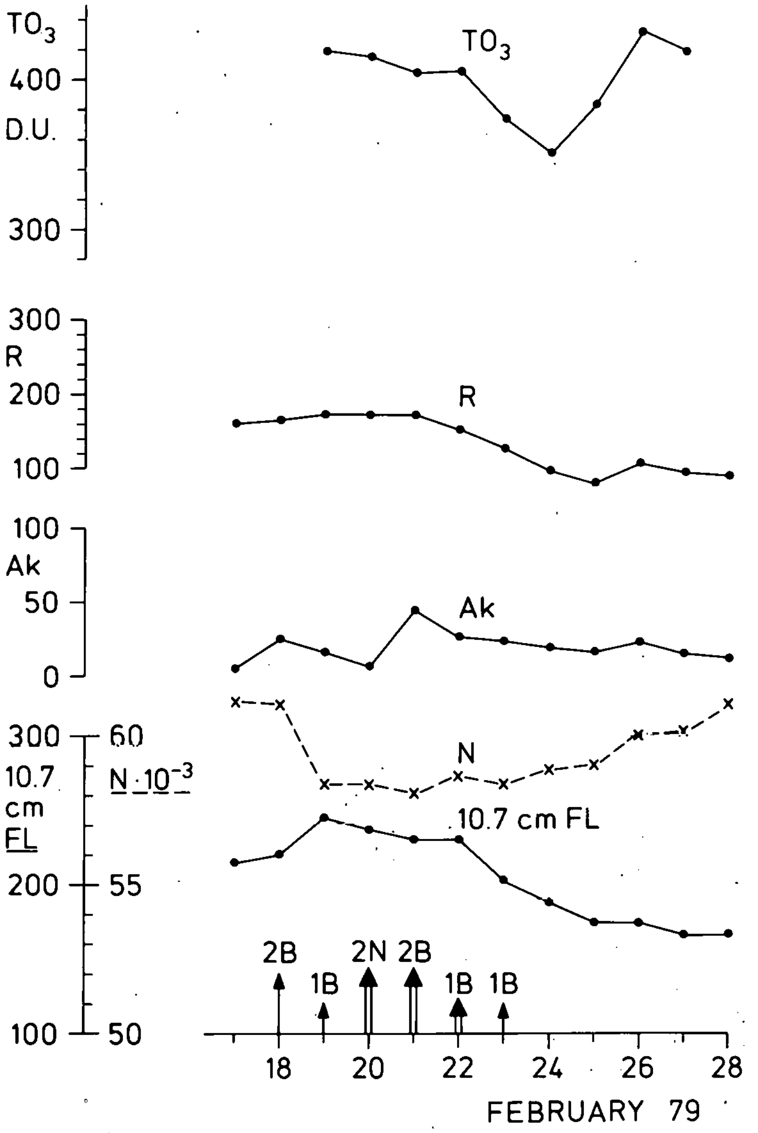

TO3
400
D.U.
300
TO3
300
R
200
100
R
100
Ak
50
0
Ak
300
10.7
cm
FL
200
100
60
N·10-3
55
50
10.7 cm FL
N
1B
1B
1B
1B
2B
1B
20
22
24
26
28
30
1
AUGUST 79
SEPT

FIG. 12



TO3
400
D.U.
300
TO3
300
R
200
100
R
100
Ak
50
0
Ak
300
60
10.7
cm
FL
N·10^-3
200
55
100
50
10.7 cm FL
N
1B
2B
1B
6
8
10
12
14
16
18
20
JANUARY 80

FIG. 13



TO3
400
D.U.
300
TO3
300
R
200
100
R
100
Ak
50
0
Ak
300
10.7
cm
FL
200
100
60
N·10-3
55
50
N
10.7 cm FL
2B
1B
2N
2B
1B
1B
18
20
22
24
26
28
FEBRUARY 79

FIG. 14

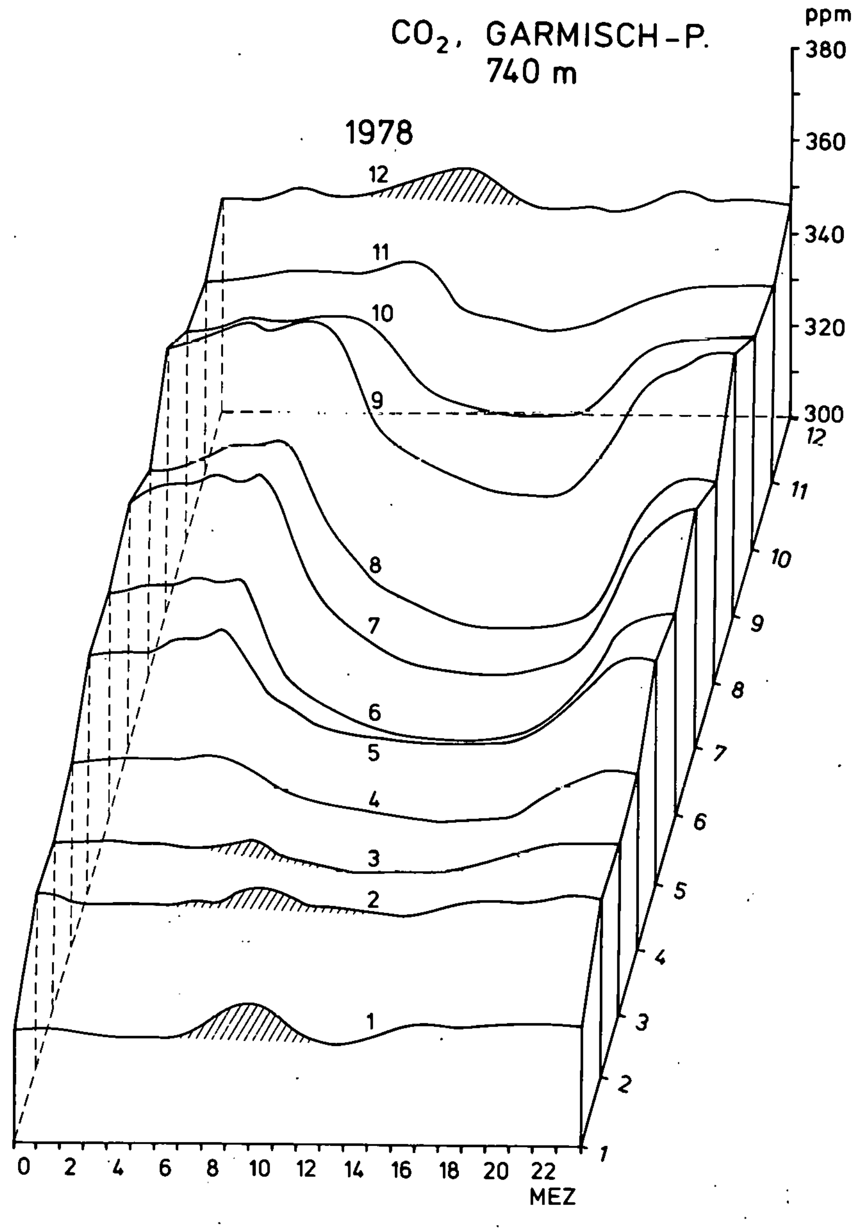

CO2, GARMISCH-P.
740 m
1978
ppm
380
360
340
320
300
12
11
10
9
8
7
6
5
4
3
2
1
0 2 4 6 8 10 12 14 16 18 20 22
MEZ

FIG. 15

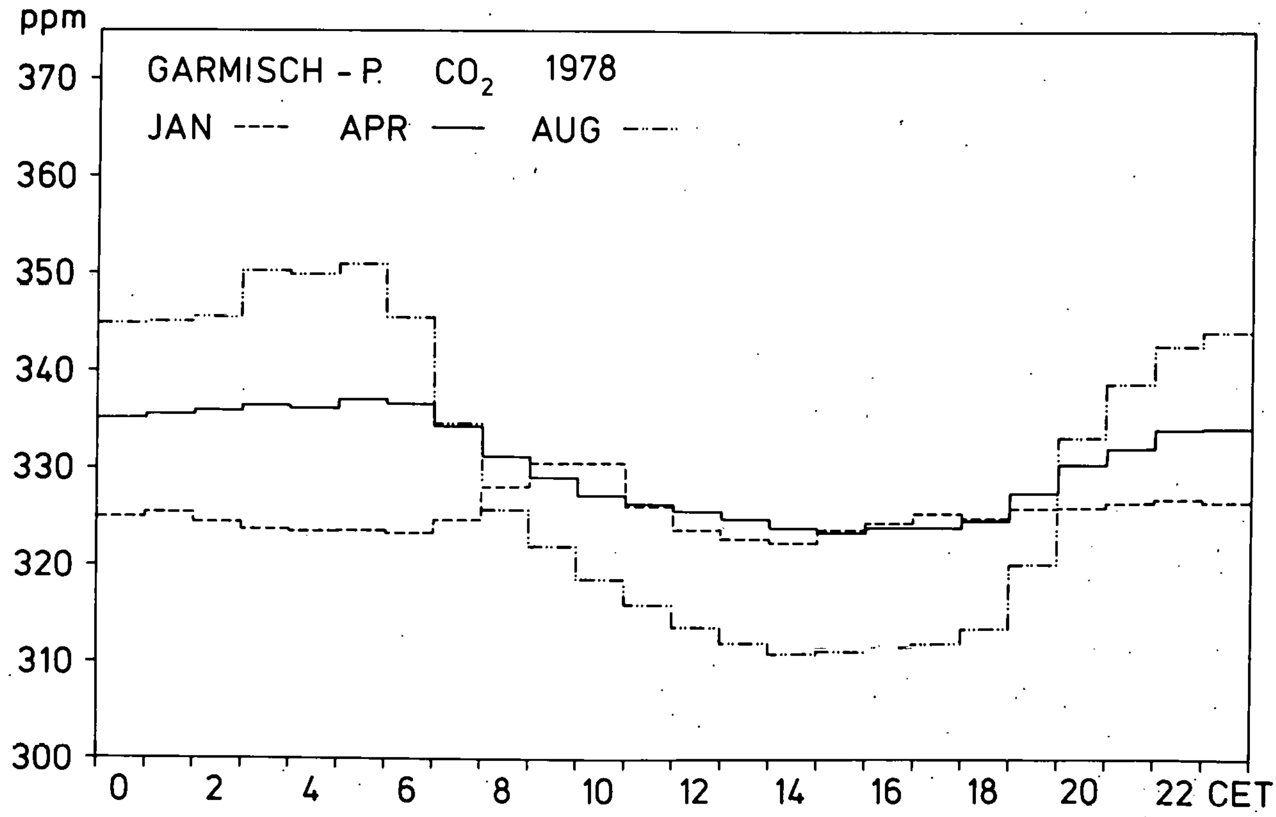

ppm
GARMISCH - P. CO2 1978
JAN ---- APR —— AUG -·-·-
370
360
350
340
330
320
310
300
0
2
4
6
8
10
12
14
16
18
20
22 CET

FIG. 16

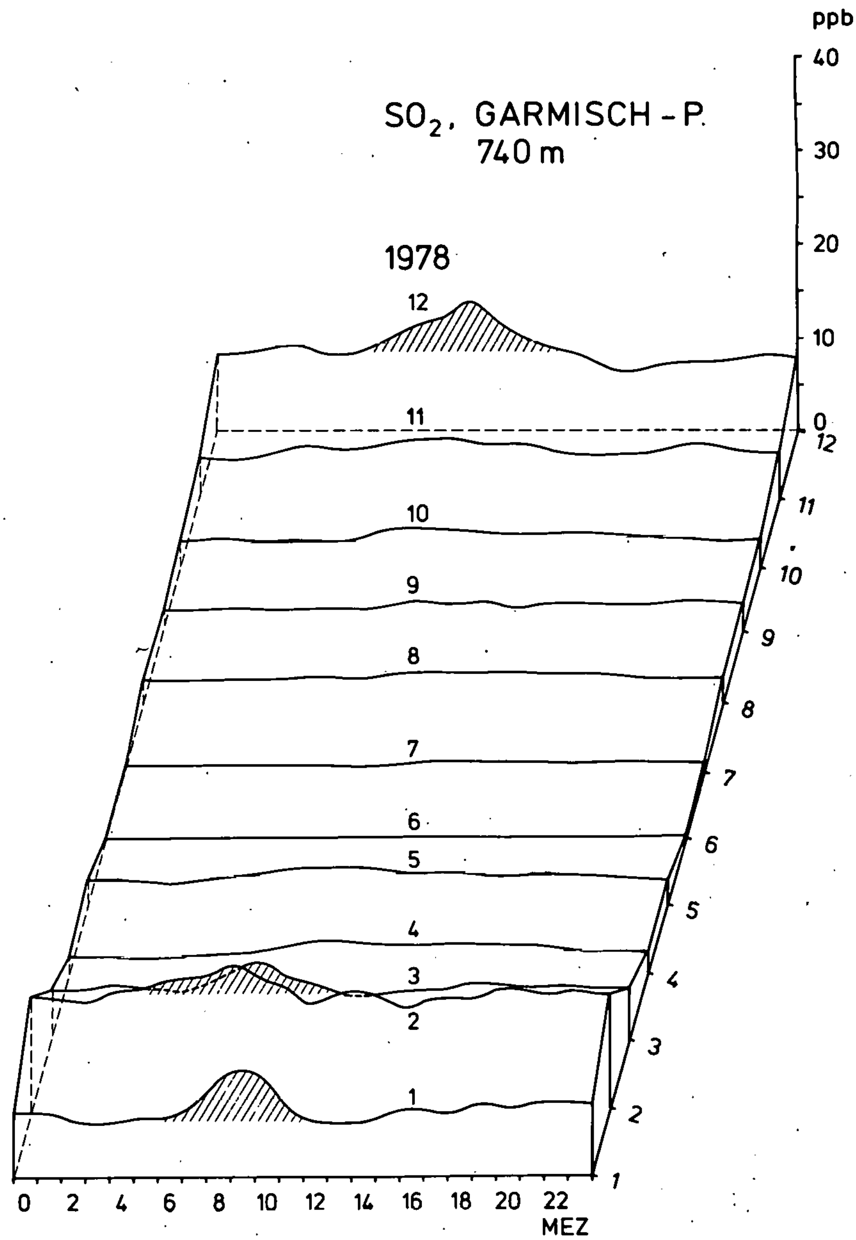

SO2, GARMISCH-P.
740 m
1978
ppb
40
30
20
10
0
12
11
10
9
8
7
6
5
4
3
2
1
0 2 4 6 8 10 12 14 16 18 20 22
MEZ

FIG. 17

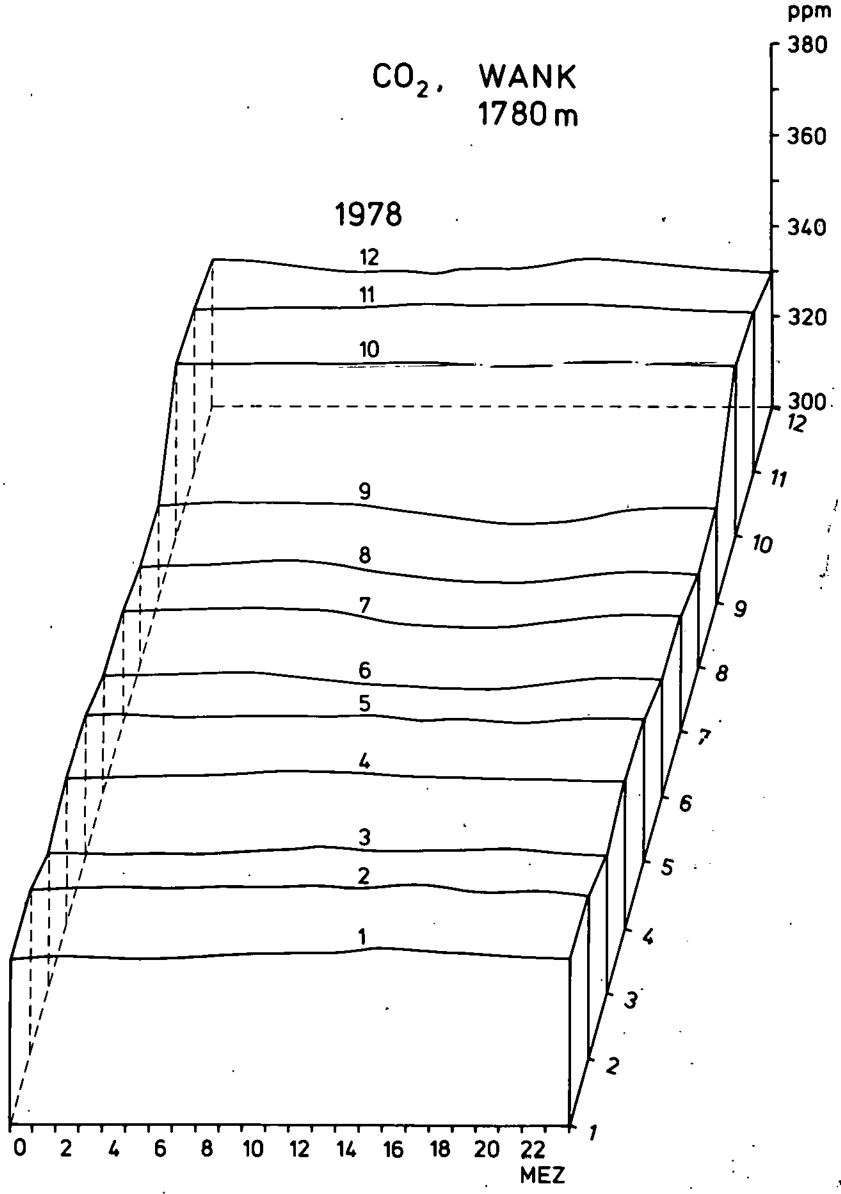
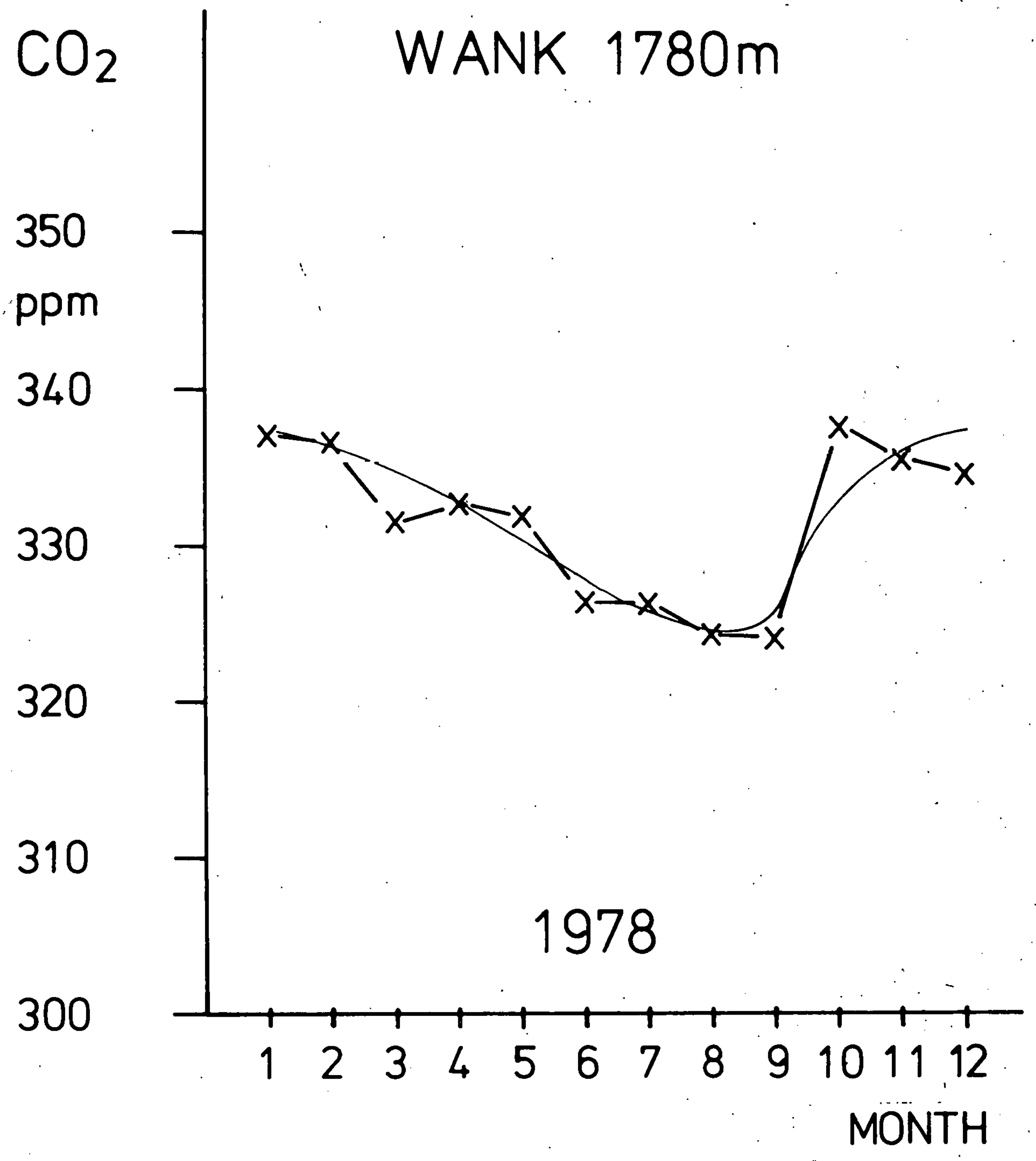

CO2, WANK
1780 m
1978
ppm
380
360
340
320
300
12
11
10
9
8
7
6
5
4
3
2
1
0 2 4 6 8 10 12 14 16 18 20 22
MEZ

FIG. 18



CO2
WANK 1780m
350
ppm
340
330
320
310
300
1978
1 2 3 4 5 6 7 8 9 10 11 12
MONTH

FIG. 19