

## DISTURBANCE OF STRATOSPHERIC TRACE GAS MIXING RATIOS DURING THE MAP/GLOBUS 1983 CAMPAIGN\*

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**Abstract**—Mixing ratio measurements of several stratospheric trace constituents were performed during the MAP/GLOBUS 1983 campaign by infrared and microwave techniques. The i.r. results indicate a wavelike disturbance in the stratosphere. Similar structures were also found by other experiments as cryo sampling, ion spectrometer, and various O<sub>3</sub> measurements. In total 14 trace gases showed such disturbances. The wavelike structure extended up to the stratopause. Horizontal scale was of the order of 1000 km. The wave was observed during the whole campaign, i.e. for more than 3 weeks. Its period was about 10 days. Induced mixing ratio disturbances are discussed in terms of horizontal and vertical transports. Their magnitudes are correlated with the gradients of the mean vertical mixing ratio profiles for eight trace constituents. Implications of such wave structures for middle atmosphere *in situ* and remote sensing measurements are briefly discussed.

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## 1. INTRODUCTION

Disturbances in vertical profiles of atmospheric temperature and wind speed as well as oscillations in time at a given altitude are well-known features in the middle atmosphere. They are attributed to gravity waves, tidal or planetary waves, or other reasons. Respective modulations of trace constituent mixing ratios are also well known. This is especially true for ozone in the stratosphere where much observational material is available for respective analyses. During the MAP/GLOBUS 1983 campaign a number of trace gases were observed in addition to ozone (see Offermann, 1987). These measurements in Southern France were spread over a time span of about 1 month. One should therefore expect to see effects of waves and other disturbances if they were present during that time period. This was the case, indeed, as will be shown by the preliminary analysis given in this paper.

## 2. DATA

### *Infrared and microwave experiments*

Infrared emission measurements of  $\text{NO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{HNO}_3$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and  $\text{O}_3$  were performed by a newly developed, cryogenically cooled grating spectrometer, which was flown on a balloon to 38 km altitude on 23 September 1983 (Rippel *et al.*, 1987). Due to technical difficulties the measuring time of this instrument was rather short (10 min), and hence the amount of data obtained was limited. The measurements were taken at about 4:30 U.T. The results (except those of  $\text{NO}_2$ ) are given in Table 1. The altitude scale is shown as pressure and height. Pressures were measured, and altitudes were derived from them by means of temperatures that were also measured (see Table 1). Trace gas number densities are the primary information obtained from the i.r. grating spectrometer. They were converted to mixing ratios by means of the measured pressures and temperatures. Measurement errors were estimated for each altitude, and are also given in Table 1. They include calibration errors, uncertainties of spectrometer line shape and atmospheric temperature, and the accuracy of the fit of calculated to measured spectra. The altitude determination is accurate to  $\pm 100$  m at the highest altitude, the error increasing to  $\pm 400$  m at the lowest altitude. For details of the data evaluation see Rippel (1984).

Because of the shortness of time available for the measurements it was not possible to stabilize the azimuth of the view direction. In consequence the gondola slowly rotated, and the measurements were taken at different directions for different trace gases and altitudes. The view directions were as follows: at 26.7

km:  $290^\circ$  for  $\text{H}_2\text{O}$ ,  $330^\circ$  for  $\text{NO}_2$ ,  $160^\circ$  for  $\text{HNO}_3$ ; at 30 km:  $130^\circ$  for  $\text{H}_2\text{O}$  and  $\text{NO}_2$ ,  $190^\circ$  for  $\text{HNO}_3$ ; at 33.3 km:  $330^\circ$  for  $\text{H}_2\text{O}$ ,  $290^\circ$  for  $\text{NO}_2$ ,  $60^\circ$  for  $\text{HNO}_3$ ; at 36.2 km:  $155^\circ$  for  $\text{H}_2\text{O}$ ,  $130^\circ$  for  $\text{NO}_2$ , and  $305^\circ$  for  $\text{HNO}_3$ . At higher altitudes the distance to the tangent point is smaller than 150 km, and the view direction should therefore be unimportant. At the lowest altitude (26.7 km) the distance to the tangent point is about 380 km. At such distances the view direction could influence the measured signal if coherent structures were present in the atmosphere. A respective wave disturbance is discussed below. Its influence on the i.r. signals can be accurately determined only when the final analysis of this structure has been finished. A preliminary estimation shows, however, that the wave analysis of the i.r. data given below is not jeopardized by the variations of view direction. Respective corrections would—if anything—make the wave more pronounced.

Infrared measurements of column densities of  $\text{NO}_2$ ,  $\text{HNO}_3$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and  $\text{O}_3$  were performed on 9 September 1983 by an airplane experiment. Absorption spectra were measured during sunset by a grille spectrometer (Fontanella *et al.*, 1975). The airplane flew at an altitude of 11.9 km from  $45^\circ\text{N}$ ,  $3^\circ\text{W}$  to  $50^\circ\text{N}$ ,  $1^\circ\text{W}$ . The measurement area between the two latitudes is centred at  $5^\circ\text{W}$ . The column densities obtained are given in Table 2. Precision and accuracy of the measurements are also shown. When the tropopause was crossed at  $48.5^\circ\text{N}$  absorptions of  $\text{H}_2\text{O}$  and  $\text{CO}$  showed large variations due to tropospheric contributions. The accuracy of their column densities therefore is poor.

A newly developed microwave radiometer for the determination of  $\text{O}_3$  and  $\text{H}_2\text{O}$  was flown on 28 September 1983 (Dawkins *et al.*, 1983). Daytime measurements were performed during balloon descent from 33 km to 17 km. The concentration profile of  $\text{H}_2\text{O}$  was determined by measuring the signals received in four separate channels centred on the strong  $\text{H}_2\text{O}$  emission line at 183 GHz while the balloon was descending at about  $1\text{ m s}^{-1}$ . The signals were measured along a path inclined upwards at  $10^\circ$  and pointing approximately due North, with the viewing direction stabilized to better than  $\pm 0.1^\circ$  in elevation and  $\pm 15^\circ$  in azimuth. The high vertical resolution obtained in the experiment (1 km) was independent of the beamwidth of the instrument ( $3^\circ$ ) and was only a function of the signal-to-noise ratio of the measurements. The data obtained are given in Table 3. A thorough error analysis has been carried out, the results of which are also shown in Table 3. These error values are the sum of the systematic and random ( $2\sigma$ ) errors. The systematic errors dominate and account

TABLE 1. INFRARED EMISSION MEASUREMENTS BY A BALLOON BORNE, CRYO COOLED GRATING SPECTROMETER OF WUPPERTAL UNIVERSITY ON 23 SEPTEMBER 1983. (TIME OF MEASUREMENT IS 4:30 U.T. DATA ARE PRELIMINARY)

Pressure (mb)	Temperature (K)	Altitude (km)	Trace gas	Number density (cm <sup>-3</sup> )	Volume mixing ratio	Measurement error (%)
20.9	221.5	26.7	H <sub>2</sub> O	2.39 × 10 <sup>12</sup>	3.5 ppmv	+15 -19
12.6	226.4	30.0	H <sub>2</sub> O	1.57 × 10 <sup>12</sup>	3.9 ppmv	+15 -19
7.62	232.0	33.3	H <sub>2</sub> O	0.94 × 10 <sup>12</sup>	3.95 ppmv	+15 -19
5.02	238.0	36.2	H <sub>2</sub> O	0.47 × 10 <sup>12</sup>	3.1 ppmv	±15
4.85	238.3	36.5	H <sub>2</sub> O	0.48 × 10 <sup>12</sup>	3.2 ppmv	±15
3.77	241.0	38.2	H <sub>2</sub> O	0.40 × 10 <sup>12</sup>	3.5 ppmv	(±15)
20.9	221.5	26.7	HNO <sub>3</sub>	2.73 × 10 <sup>9</sup>	4 ppbv	+28 -30
12.6	226.4	30.0	HNO <sub>3</sub>	2.14 × 10 <sup>9</sup>	5.3 ppbv	+26 -27
7.62	232.0	33.3	HNO <sub>3</sub>	0.86 × 10 <sup>9</sup>	3.6 ppbv	+26 -29
4.85	238.3	36.5	HNO <sub>3</sub>	0.34 × 10 <sup>9</sup>	2.3 ppbv	±21
4.85	238.3	36.5	CH <sub>4</sub>	7.9 × 10 <sup>10</sup>	0.54 ppmv	±21
4.85	238.3	36.5	N <sub>2</sub> O	2.64 × 10 <sup>9</sup>	18 ppbv	±11
4.85	238.3	36.5	O <sub>3</sub>	1.25 × 10 <sup>12</sup>	8.5 ppmv	±15

Column density of HNO<sub>3</sub> on 23 September 1983 from 26.7 to 36.5 km: 1.5 × 10<sup>15</sup> cm<sup>-2</sup>.

TABLE 2. INFRARED ABSORPTION MEASUREMENTS BY A GRILLE SPECTROMETER ON AN AIRPLANE (CNRM/TOULOUSE) ON 9 SEPTEMBER 1983 AT SUNSET

Species	Column density above (km)	Value (mol cm <sup>-2</sup> )	Precision (%)	Accuracy (%)
NO <sub>2</sub>	11.9	4.4 × 10 <sup>15</sup>	8	13
HNO <sub>3</sub>	10	1.4 × 10 <sup>16</sup>	12	27
CH <sub>4</sub>	10	7.4 × 10 <sup>18</sup>	5	10
N <sub>2</sub> O	10	1.68 × 10 <sup>18</sup>	10	15
O <sub>3</sub>	10	6.9 × 10 <sup>18</sup>	4	9
H <sub>2</sub> O	11.9	Variable from 4.2 × 10 <sup>19</sup> (50°N) to 9.1 × 10 <sup>19</sup> (45°N)		
CO	11.9	Variable from 2.7 × 10 <sup>17</sup> (50°N) to 4.5 × 10 <sup>17</sup> (45°N)		

for two-thirds of the values given in the table, and although they introduce a possible bias in the results they do not contribute to the structure shown in the concentration profile. These systematic errors arise predominantly from two sources. The first is due to uncertainties in the absolute calibration of the instrument which was performed by two internal black bodies, one at ambient and the other at liquid nitrogen temperature. The second is due to the intrinsic accuracy limitations of the algorithm used to invert the radiometric data. A third source of systematic error should also be mentioned which affects the altitude to which the mixing ratio data are referred. These altitudes were obtained by integrating the hydrostatic

equation using pressure and temperature data measured during the flight. However, the uncertainties in the absolute accuracy of the pressure data could well have introduced a systematic error in the altitudes of at least 1 km.

All NO<sub>2</sub> measurements of the campaign were collected and discussed in detail by Pommereau *et al.* (1987). The respective i.r. results (emission measurements of the Wuppertal instrument) are reproduced in Fig. 1. They show a very peculiar altitude profile, especially when compared to two profiles obtained by the LIMS experiment on the *Nimbus 7* satellite (J. C. Gille and J. M. Russel III, private communication). The lower one of these two (October, 45° N) should

TABLE 3. MICROWAVE EMISSION MEASUREMENTS BY A BALLOON-BORNE RADIOMETER OF RUTHERFORD APPLETON LABORATORY (CHILTON) ON 28 SEPTEMBER 1983

Time (U.T.)	Pressure (mb)	Temperature (K)	Altitude (km)	Trace gas	Number density (cm <sup>-3</sup> )	Volume mixing ratio (ppmv)	Measurement error (ppmv)
13:57	91.2	214.6	17	H <sub>2</sub> O	15.5 × 10 <sup>12</sup>	4.5	± 1.4
13:46	78.3	216.5	18	H <sub>2</sub> O	10.8 × 10 <sup>12</sup>	3.7	± 1.2
13:34	65.3	219.1	19	H <sub>2</sub> O	8.5 × 10 <sup>12</sup>	3.5	± 1.2
13:18	58.2	217.8	20	H <sub>2</sub> O	6.3 × 10 <sup>12</sup>	2.9	± 1.0
13:00	49.7	219.5	21	H <sub>2</sub> O	6.2 × 10 <sup>12</sup>	3.4	± 1.2
12:43	42.4	218.0	22	H <sub>2</sub> O	6.0 × 10 <sup>12</sup>	3.8	± 1.4
12:27	35.6	225.3	23	H <sub>2</sub> O	4.9 × 10 <sup>12</sup>	3.8	± 1.5
12:08	31.2	224.2	24	H <sub>2</sub> O	4.3 × 10 <sup>12</sup>	3.8	± 1.5
11:49	26.7	225.1	25	H <sub>2</sub> O	4.4 × 10 <sup>12</sup>	4.6	± 1.9
11:32	23.1	225.2	26	H <sub>2</sub> O	4.2 × 10 <sup>12</sup>	5.1	± 2.1
11:14	20.2	225.2	27	H <sub>2</sub> O	3.6 × 10 <sup>12</sup>	5.0	± 2.1
11:00	17.1	227.5	28	H <sub>2</sub> O	3.1 × 10 <sup>12</sup>	5.1	± 2.2
10:46	14.7	228.1	29	H <sub>2</sub> O	3.0 × 10 <sup>12</sup>	5.8	± 2.6
10:35	12.7	232.7	30	H <sub>2</sub> O	2.8 × 10 <sup>12</sup>	6.4	± 2.9
10:25	11.2	231.6	31	H <sub>2</sub> O	2.6 × 10 <sup>12</sup>	6.7	± 3.1
10:15	9.4	231.9	32	H <sub>2</sub> O	2.3 × 10 <sup>12</sup>	7.0	± 3.6
10:03	8.3	235.0	33	H <sub>2</sub> O	2.0 × 10 <sup>12</sup>	6.9	± 3.9

be near to what one expects as an undisturbed NO<sub>2</sub> profile for the time of the balloon i.r. measurement. On the other hand, the balloon i.r. results agree quite well (within their error bars) with others obtained by star occultation at the higher altitudes (Pommereau *et al.*, 1987). Star occultation is a well-proven technique (see for instance Naudet *et al.*, 1984), and is completely independent of the i.r. method. Indeed, it is therefore felt that NO<sub>2</sub> densities around 36–38 km were much lower, and at 26–33 km much larger than normal.

If this conclusion is accepted, inspection of Fig. 1 shows that a wavelike disturbance must have been present in the stratosphere on 23 September 1983. This is shown more clearly in Fig. 2, where the quotient of the NO<sub>2</sub> mixing ratio profile of 23 September 1983 and of the LIMS October profile is plotted vs altitude. A strong positive deviation (up to a factor of 1.9) from the undisturbed atmosphere (as given by the abscissa value of 1) is seen below 35 km, whereas the deviation above that altitude level tends towards values which are smaller than 1 by a similar factor.

Nitric acid mixing ratios as derived from the i.r. measurement on 23 September 1983 are given in Fig. 3. LIMS altitude profiles are used for comparison again (Gille *et al.*, 1984). The LIMS data are for 45° N, and the zonal mean profile of 5 November 1978 is considered a reasonable reference representing an undisturbed atmosphere. Again, a remarkable deviation of the i.r. balloon measurements from this reference profile is seen. This time it is, however, opposite to what was found for NO<sub>2</sub>: at high altitudes the

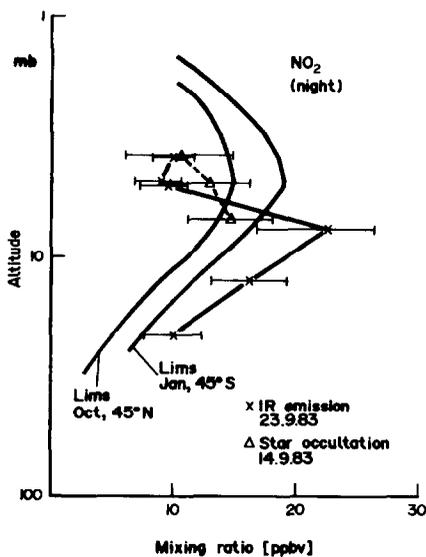


FIG. 1. NO<sub>2</sub> MIXING RATIOS FROM I.R. EMISSION AND STAR OCCULTATION MEASUREMENTS VS ALTITUDE.

(Altitude is given as total pressure.) Reference curves (solid lines) were taken from LIMS measurements on *Nimbus 7*.

balloon data are higher and at low altitudes lower than what one would expect for an undisturbed atmosphere.

The balloon mixing ratios of HNO<sub>3</sub> were again divided by those of the reference profile, and the quotient is given in Fig. 2. The resulting deviation curve reaches the value of 1 at about 30 km, which is some-

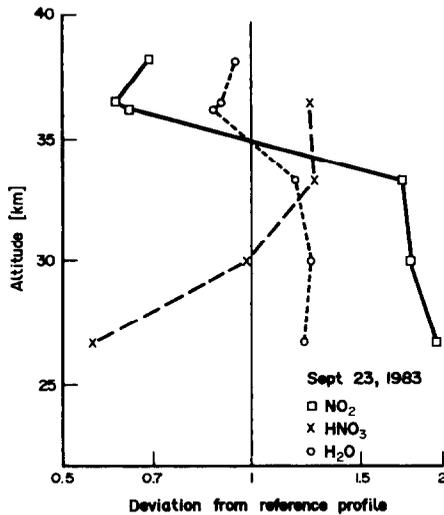


FIG. 2. DEVIATION OF  $\text{NO}_2$ ,  $\text{HNO}_3$  AND  $\text{H}_2\text{O}$  MIXING RATIOS FROM REFERENCE CURVES.

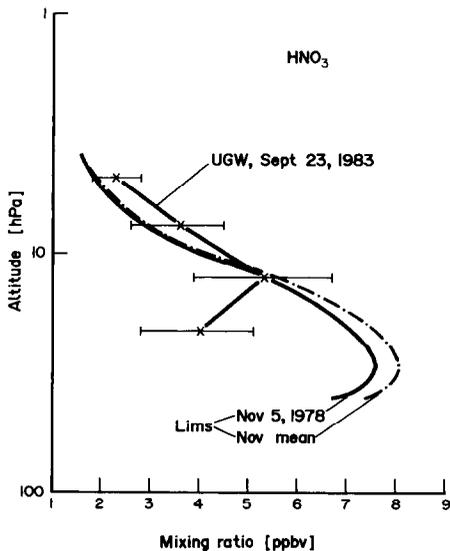


FIG. 3. NITRIC ACID MIXING RATIOS VS ALTITUDE. (Altitude is given as total pressure.) Data are from i.r. emission measurements. Reference curves (solid line and dash-dotted line) are from LIMS measurements on *Nimbus 7*.

what lower than in the case of  $\text{NO}_2$ . This result may possibly be attributed to the preliminary nature of the  $\text{HNO}_3$  data.

Water vapour mixing ratios as measured by the i.r. spectrometer on 23 September 1983 and by the microwave radiometer on 28 September 1983 are shown in Fig. 4. As water vapour is very variable one would not expect a close agreement of two measurements

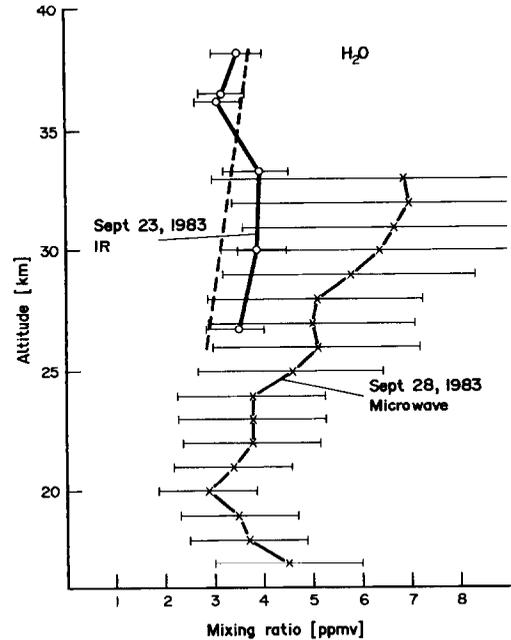


FIG. 4. WATER VAPOUR MIXING RATIOS FROM i.r. (23 SEPTEMBER 1983) AND MICROWAVE (28 SEPTEMBER 1983) EMISSION MEASUREMENTS.

Dashed line has a gradient of  $2\% \text{ km}^{-1}$  and is used as a reference curve.

taken at a time difference of 5 days. On the other hand, the error bars of the microwave data are so large that a real difference of water vapour on the two days cannot be maintained. It was said above that the structures seen in the microwave profile are not affected by the large systematic errors. It is therefore believed that the minimum at 20 km shows the existence of a hypopause at this altitude.

It was checked whether the wavelike structures seen in the i.r. measurements of  $\text{NO}_2$  and  $\text{HNO}_3$  are also present in the  $\text{H}_2\text{O}$  data obtained from that experiment. Because of the large variability of water vapour a reliable reference profile is not available. It is, however, known from balloon experiments that water vapour slowly increases from 20 to 40 km. Respective analyses showed that a gradient of about  $2\% \text{ km}^{-1}$  is a reasonable mean value in this altitude regime (Harries, 1976; Ehhalt and Tönnißen, 1980). Recent balloon measurements tend to confirm this value, with slightly lower numbers at the lower altitudes, and somewhat higher values in the upper regime. It was therefore assumed that an artificial, though not unreasonable, reference profile could be constructed by adopting 3.5 ppmv of water vapour at 35 km altitude and a profile gradient of  $2\% \text{ km}^{-1}$  above and

below that altitude. This profile is shown as a dashed line in Fig. 4. If it is compared to the  $\text{H}_2\text{O}$  data measured by the i.r. experiment a wavelike structure is found, indeed, though its amplitude is smaller than those of the  $\text{NO}_2$  and  $\text{HNO}_3$  curves. This is seen from Fig. 2 where the quotient of the measured  $\text{H}_2\text{O}$  profile and of the reference profile is plotted. The wavelike structure obtained by this analysis is of course dependent on the assumption of the crossing point of measured and mean profiles. For the i.r. measurement an altitude of 35 km was chosen (with 3.5 ppm) in order to be compatible with the  $\text{NO}_2$  results in Fig. 2. Hence the whole procedure is rather coarse. It essentially shows that the  $\text{H}_2\text{O}$  data do not contradict the wave structures obtained from the  $\text{NO}_2$  and  $\text{HNO}_3$  data.

#### Cryosampling experiments

Various source gases were determined during the campaign by two cryosampling experiments flown on 10 September 1983 (KFA Jülich and MPI Lindau). The two flights were performed as closely together as possible to determine the measuring accuracy of this technique. Respective results are discussed in detail by Schmidt *et al.* (1987). Their data points for  $\text{CH}_4$  are reproduced in Fig. 5. A very close agreement of the results of the two experiments is seen. As the measured points of KFA Jülich are more numerous and more evenly distributed with altitude than those of MPI Lindau, they were used to derive an approximate mean curve which is shown as a solid line in Fig. 5. This curve again exhibits a conspicuous wavy structure, which deserves comparison to a mean reference profile. Ehhalt *et al.* (1983) collected a number of measurements of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and  $\text{F12}$  which were performed in Southern France. The data were normalized in June 1979. The authors present also 5 km running means of their data. These mean values were used as a reference profile in Fig. 5 (and in Figs 7 and 8).

A further data point is given in Fig. 5 which was measured on 23 September 1983 by the infrared spectrometer (labelled UGW, see also Table 1). It appears to be considerably above the extrapolation of the reference curve.

The same procedure was used for  $\text{CH}_4$  as was sketched above: the measured solid curve (solid line in Fig. 5) was divided by the reference profile. The result is given in Fig. 6, which shows a maximum above 30 km. At lower altitudes the deviation from the reference profile becomes smaller and reaches a minimum at about 24 km. Below that level the deviation curve again rises and has a second relative maximum at around 20 km.

Two other source gases were measured by the cryo-

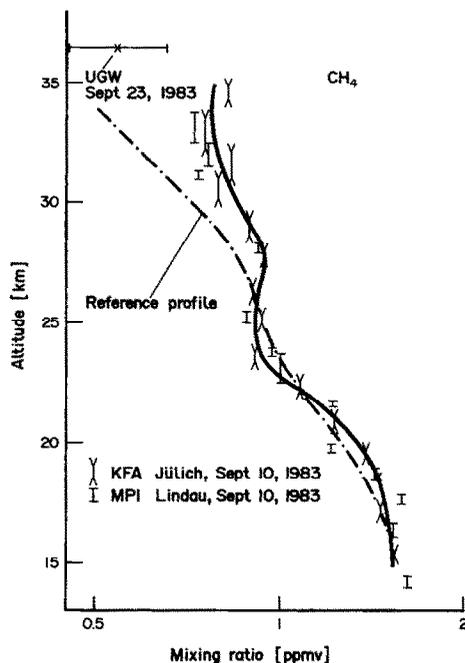


FIG. 5. METHANE MIXING RATIOS MEASURED ALMOST SIMULTANEOUSLY BY TWO CRYOSAMPLING EXPERIMENTS FROM KFA JÜLICH AND MPI LINDAU ON 10 SEPTEMBER 1983. Solid line is an approximate mean curve to the KFA Jülich data. Data point of 23 September 1983 is from an i.r. emission measurement. For explanation of the reference curve see text.

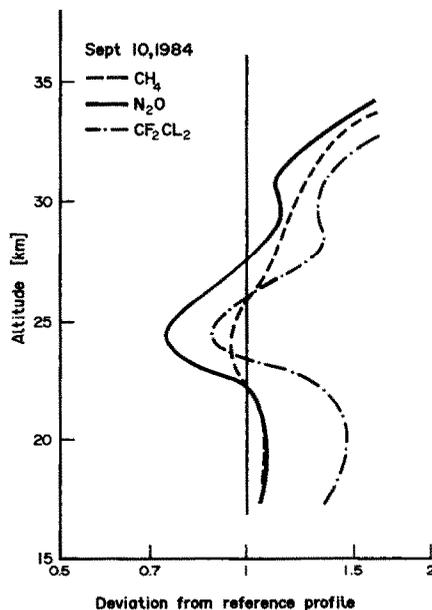
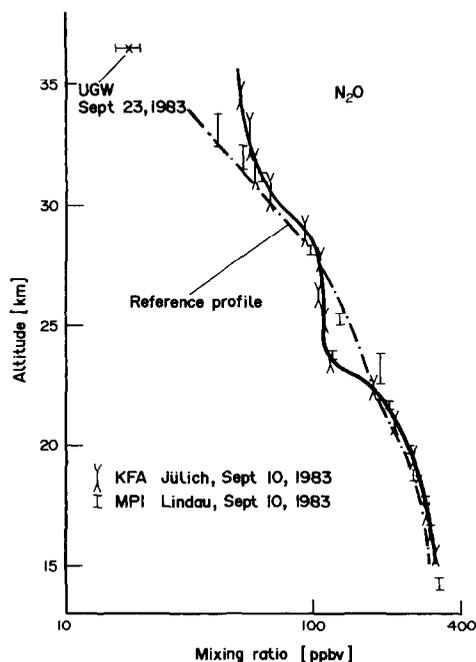


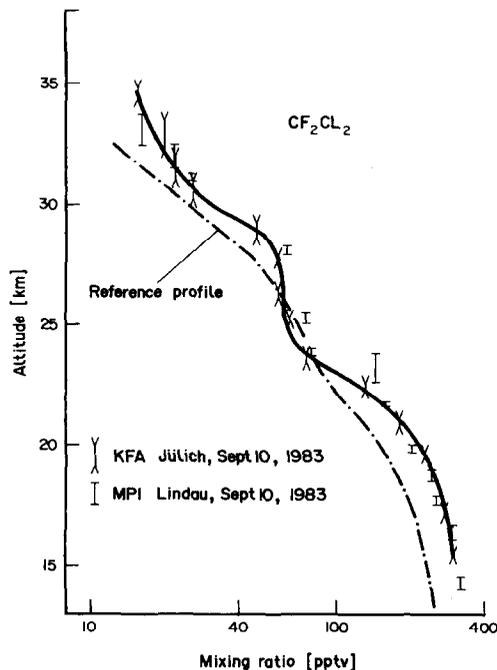
FIG. 6. DEVIATION OF  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  AND  $\text{CF}_2\text{Cl}_2$  MIXING RATIOS AS REPRESENTED BY SOLID LINES IN FIGS 5, 7 AND 8 FROM RESPECTIVE REFERENCE CURVES.

FIG. 7. AS FIG. 5, EXCEPT FOR  $N_2O$ .

samplers, which were analysed in a similar manner to  $CH_4$ : measured data points of  $N_2O$  are shown in Fig. 7, and those of  $CF_2Cl_2$  in Fig. 8. Solid lines again are approximate mean curves to the data points of KFA Jülich. In Fig. 7 a further  $N_2O$  data point from the i.r. measurement of 23 September 1983 is included (labelled UGW; see also Table 1). Reference profiles for  $N_2O$  and F12 were taken from Ehhalt *et al.* (1983) as in the case of  $CH_4$ , and are shown in Figs 7 and 8. The quotients of the mean measured profiles and the reference profiles are given in Fig. 6. The resulting curves show the same features as in the case of  $CH_4$ : high values at and above 30 km, low values at about 24 km, and relatively high values around 20 km.

The  $CH_4$  and F12 profiles in Fig. 6 do not oscillate about the mean undisturbed value of 1, but rather about a value somewhat higher. This indicates that the reference profiles adopted may be somewhat low in these two cases. A possible explanation might be seen in the increase of these two gases from 1979 until 1983.

The wavelike disturbance of the atmosphere is obvious from the pictures presented, as it shows up in the data of two independent experiments (two cryo-samplers on different balloons) and in three different source gases. A number of other trace gases were measured by the cryosamplers during the campaign which also show indications of the wave structure discussed. It is, however, difficult to obtain reliable reference

FIG. 8. AS FIG. 5, EXCEPT FOR FREON 12 ( $CF_2Cl_2$ ).

profiles for these, and hence an analysis like the one given above cannot be performed at present. These gases are  $CFCl_3$  (F11, Schmidt *et al.*, 1987),  $CCl_2F-CClF_2$  and  $CClF_2-CClF_2$  (CFC113 and CFC114, Borchers *et al.*, 1987; Fabian *et al.*, 1985),  $CCl_4$ ,  $CH_3CCl_3$  and  $CHClF_2$  (Fabian, 1985).

#### Ion spectrometers

Measurements of positive and negative ions were performed in the stratosphere by cryo-cooled mass spectrometers on two different days of the campaign (Ingels *et al.*, 1987; Schlager and Arnold, 1987). From positive ion spectra mixing ratios of acetonitrile ( $CH_3CN$ ) were derived by Ingels *et al.* (1987). These authors furthermore suggest two reference profiles for  $CH_3CN$  to compare with. They find large deviations from these profiles that vary with altitude. These deviations are very similar for the two reference profiles. Therefore only the "Model Reference Profile" (see Ingels *et al.*, 1987) will be used in the following. The mixing ratios measured on 18 September 1983 by Ingels *et al.* (1987) are larger than the reference profile up to a factor of 2 at 30 km. At altitudes below 25 km they show large scatter, and are considerably lower than the reference profile. This is in general agreement with the wave structures discussed above.

#### Ozone network

To study the detailed structure of the wave that obviously was present during the campaign, standard

ozone measurements can be used below 30 km. Quite a number of O<sub>3</sub> measurements were performed on many days of the campaign and at various stations, and many of them for the explicit purpose of a dynamical study. These data are discussed elsewhere in detail (Attmannspacher *et al.*, 1987; Muller and Krueger, 1987; de La Noë *et al.*, 1987; Aïmedieu *et al.*, 1987; Simon *et al.*, 1987).

A dynamical analysis of these many data is beyond the scope of the present paper. As an indication of what a future analysis might yield, Fig. 9 shows a series of measurements of ozone profiles taken by the lidar at the Zugspitze station (47.5° N, 11° E; Werner *et al.*, 1985). Nightly mean ozone number densities are plotted (in relative units) vs altitude for 15 days of the campaign. The labels of the profiles denote the day of September 1983 when the measurement started in the evening. It is clearly seen that considerable wave activity was present during all of the campaign, and was especially strong at the beginning and end of September 1983. On 23 September an ozone enhancement appears to have been present around 30 km, with a respective deficiency at lower altitudes. This result is in line with the above findings on this day. It needs, however, proper numerical analysis, which is presently being undertaken. Figure 9 also shows that the wave activity was rather variable, and could be quite different on one day and the next.

A horizontal representation of the ozone field at a fixed altitude and of respective time variations is given in Fig. 10. This picture shows preliminary data of ozone sondes at the 30 mb pressure level, measured at six different stations in Western Europe. The vertical profiles of the Brewer/Mast and ECC sondes have not yet been smoothed for short period waves, and hence the data is quite noisy. (The Garmisch data were smoothed by a 3-day running mean.) Nevertheless, the minima and maxima of the various curves may be interpreted in terms of a wave progressing from West to East. This is seen from the straight lines in Fig. 10 which are lines of constant phase. This is because the scales of O<sub>3</sub> partial pressures at the various stations in Fig. 10 have been arranged in such a way that the distances of the 100 nb marks are proportional to the distances in longitude of the stations. (The same is of course valid for any other selected pressure mark.) The lines of constant phase in Fig. 10 represent a wave with a period of 9.5 days and a zonal wavelength of about 1300 km. It should be noted that the values of El Arenosillo were added to Fig. 10 only after the lines of constant phase had been drawn. These data can therefore be regarded as a check on the validity of the analysis.

A thorough analysis of the ozone data at all height

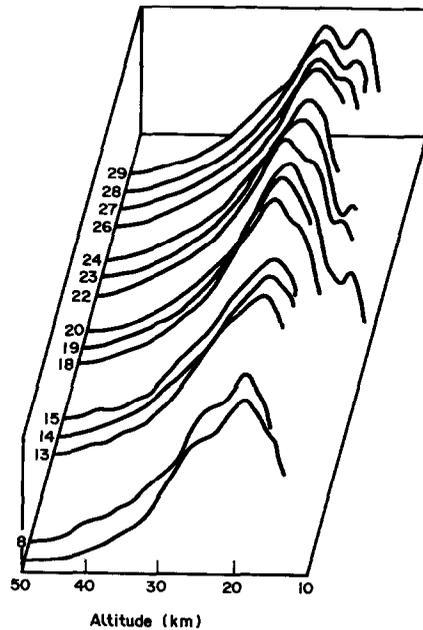


FIG. 9. OZONE DENSITY (IN ARBITRARY UNITS) VS ALTITUDE FOR 15 DAYS OF SEPTEMBER 1983.

Data are mean nightly profiles obtained by a Lidar on the Zugspitze station (Werner *et al.*, 1985). Curves are labelled by the number of the day in September 1983, when the measurement started in the evening.

levels available is presently being undertaken. It will hopefully yield detailed information on the wave behaviour in three dimensions and time. One important question will be as to how far the wave extended upwards in the stratosphere. A first answer is given by the O<sub>3</sub> data shown in Fig. 11. These are data measured by the u.v. experiment on the *Solar Mesospheric Explorer (SME)* at 1 mb altitude (preliminary data). The measurements were taken around 1500 U.T. at 45° N (see Offermann, 1987). The *SME* experiment was turned on above Europe for one pass per day only. The longitudes of these orbits shifted slightly from day to day. They are given in the bottom of Fig. 11. The rate of eastward migration is 3.75° day<sup>-1</sup>. The ozone mixing ratios at 1 mb (about 48 km) show an oscillation, indeed. The period of this oscillation is, however, about 7.5 days and thus much shorter than the one indicated in Fig. 10. This can be explained by a Doppler shift due to the apparent satellite motion relative to the wave motion. From this Doppler shifted period of 7.5 days, the known satellite migration (3.75° day<sup>-1</sup>) and the zonal wavelength, one can derive the true wave period. If tentatively the same wavelength is adopted for 1 mb as was found at 30 mb, a value of 10.6 days is obtained. This is near to

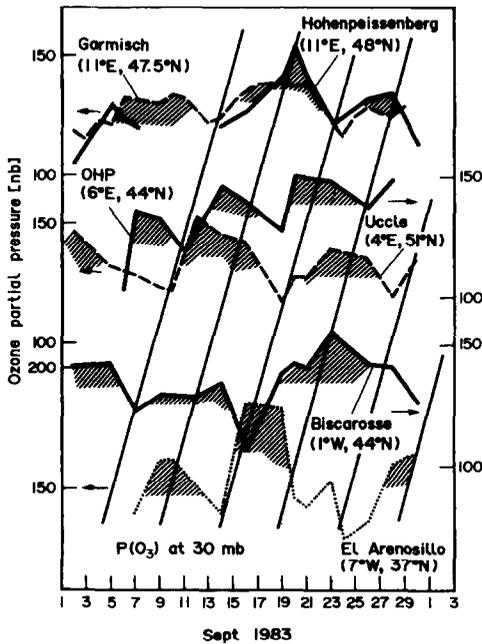


FIG. 10. OZONE PARTIAL PRESSURE MEASUREMENTS BY OZONE SONDES AT SIX STATIONS DURING SEPTEMBER 1983. Ozone data are given for the 30 mb altitude level. Slant solid lines indicate a wave progressing from West to East. For details see text.

the period indicated by Fig. 10 for the lower altitudes, and to the value of 10 days obtained from other data, as discussed below. It strongly suggests that the wave extended up to the 1 mb level at least. These data furthermore show the difficulties that arise in the interpretation of satellite data when the scales of atmospheric waves are much smaller than the satellite footprint. In such cases it may be impossible to determine the true wave period.

### 3. DISCUSSION

The wavelike structures in the vertical profiles of mixing ratios could be attributed to horizontal and/or vertical transports. Horizontal transport of air masses is frequently found to be an explanation for unusual structures in mixing ratios of stratospheric trace gases. As most of the gases discussed above have considerable horizontal gradients in mixing ratio, such an explanation was also tried in the present case. Inspection of the literature on these meridional distributions, and especially of the various results of the *Nimbus 7* satellite (*J. geophys. Res.* **89**, No. 4, 1984) indeed shows that many of the features discussed above (at 30 km) could be explained by a large scale northward transport of trace constituents. There are,

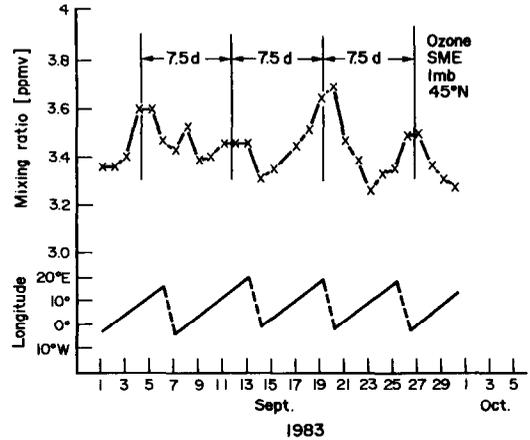


FIG. 11. OZONE MIXING RATIOS MEASURED AT 1 mb BY THE u.v. EXPERIMENT ON *Solar Mesospheric Explorer* OVER EUROPE.

Longitudes of the satellite passes are given at the bottom.

however, several difficulties in this picture as follows. (1) The disturbance of the water vapour profile cannot be explained this way. It would—quite contrary—need a southward transport according to the meridional gradients measured by the LIMS experiment on *Nimbus 7*. (2) The magnitude of the disturbances found is rather large. Meridional gradients in the literature are, on the other hand, quite small for most trace gases. In consequence meridional transports would have to be rather extended and to cover about  $30^\circ$  of latitude or more. In addition, the respective wind system would have to change direction a few kilometers above and below the 30 km level according to the wavelike structures shown above. (3) Trajectory analyses were performed for several occasions during the campaign by Langematz *et al.* (1987). They do not show the required strong meridional transport, but rather weak and small scale motions.

Transport that is mainly horizontal does therefore not appear to be a good candidate to explain the observed disturbances. An easier explanation may be obtained by invoking the vertical gradients of the trace gas mixing ratios, as these are quite steep in most cases. A respective scheme of a slant convection of air parcels with opposite directions in subsequent vertical layers was discussed for instance by Matsuno (1980) in terms of planetary waves. A convection picture of this type predicts an enhancement of mixing ratio (for instance at the 30 km level) if the gradient of the undisturbed mixing ratio profile is negative, and if the movement is directed upwards. If it is directed downwards, or if the gradient is positive, a mixing ratio decrease should result. Hence subsequent layers of upgoing and downgoing convection should pro-

duce a wavelike structure in the mixing ratio profiles. This picture can be checked by means of Figs 2 and 6. On 23 September 1983 a downgoing transport would be inferred from the  $\text{NO}_2$  and  $\text{H}_2\text{O}$  profiles in the 25–30 km regime, and a reversal in direction above this layer. The  $\text{HNO}_3$  profile essentially confirms this conclusion, as its behaviour is opposite to that of  $\text{NO}_2$  and  $\text{H}_2\text{O}$  in Fig. 2. This results from the gradient of the undisturbed  $\text{HNO}_3$  profile (Fig. 3), which—in contrast to those of  $\text{NO}_2$  and  $\text{H}_2\text{O}$ —is negative in this altitude regime. (There is a slight disagreement between the  $\text{NO}_2$  and  $\text{HNO}_3$  profiles as to the altitude where the direction reversal should occur, i.e. where the profiles should reach the value 1 in Fig. 2. This may be due to the preliminary nature of the  $\text{HNO}_3$  curve, as was mentioned above.)

The mean undisturbed profiles of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$  and  $\text{CF}_2\text{Cl}_2$  have negative gradients (Figs 5, 7 and 8). The mixing ratio enhancements in the 30 km area therefore require an upward transport in this regime on the day of this measurement (10 September 1983). This is opposite to the result obtained on 23 September 1983. It would therefore mean that the wave tentatively invoked here for explanation of these disturbances would have changed phase by about one-half cycle (or uneven multiples of it) between these two days.

If the phase of the wave on 23 September 1983 at around 30 km is called “positive”, it was negative on 10 September 1983. The acetonitrile measurements discussed above can also be analyzed in this way. As the  $\text{CH}_3\text{CN}$  reference profile has a negative gradient, the data of 18 September 1983 indicate a negative phase on that day (Ingels *et al.*, 1987). Finally, the star occultation measurements of  $\text{NO}_2$  given in Fig. 1 show a phase for 14 September 1983 similar to that of 23 September 1983, i.e. it was positive.

The above given trace gas data thus allow us to identify the sign of the phase of the hypothetical wave. This is the case for four different days, which are spread over the campaign duration. From this information one can try to estimate a wave period that fits these data. The result of the analysis is shown in Fig. 12. A wave period of 10 days yields a reasonable fit. This is right in between the periods of 9.5 and 10.6 days obtained from the ozone data. To obtain this fit one has to assume that some of the measurements did not take place during the wave extrema, but rather at some intermediate phase (Fig. 12). Some of the wave amplitudes seen in Figs 1–8 and shown by Ingels *et al.* (1987) may therefore represent lower limits to the real amplitudes.

If the vertical gradient of mixing ratio is much more important during slant transport than the horizontal gradient, not only the above discussed information on

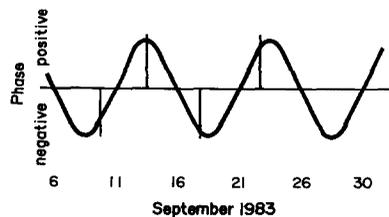


FIG. 12. SIGN OF PHASE OF WAVE AS DETERMINED ON FOUR DAYS BY FIVE BALLOON MEASUREMENTS. Sinusoidal fit curve has a period of 10 days (see text).

the phases of the wave should be obtainable from the measurements. Much more importantly, a correlation should be observed between the measured wave amplitudes and the magnitude of the vertical gradients of the mixing ratio reference profiles: the steeper the gradient of the reference profile, the larger should be the amplitude. This is approximately fulfilled, indeed, as shown by Figs 1–8. The mean profile gradients for  $\text{H}_2\text{O}$  and  $\text{CH}_4$  are small, whereas those of  $\text{CF}_2\text{Cl}_2$ ,  $\text{NO}_2$  and  $\text{HNO}_3$  are large (if the signs are neglected here). In consequence, the magnitudes (amplitudes) of deviations from the reference profiles are relatively small for  $\text{H}_2\text{O}$  and  $\text{CH}_4$ , and much larger for  $\text{CF}_2\text{Cl}_2$ ,  $\text{NO}_2$  and  $\text{HNO}_3$ . The respective values for  $\text{N}_2\text{O}$  are in between. A quantitative analysis yields results that are shown in Fig. 13. Here the deviation amplitudes are given for all gases for which a reference profile is available. They are plotted vs the gradients of the respective reference profiles at 30 km. Amplitudes were obtained by fitting sinusoidal curves through the maximum and minimum deviations (in Figs 2, 6 and Fig. 3 of Ingels *et al.*, 1987).

The fit procedure used for derivation of the amplitudes is rather coarse. It should, however, be noted that even larger errors may result from the fact that for several gases it is not known whether the maximum or minimum deviation is seen at all in the figures quoted. This is illustrated by Fig. 6 where the values at the 33 km level were used as maxima. It is possible that these values are too low, and hence especially the amplitudes for  $\text{N}_2\text{O}$  and F12 given in Fig. 13 may be lower limits.

Difficulties of the analysis are also seen in cases where some of the measured mixing ratios are larger than those of the maximum of the reference curve (Figs 1, 8). This discrepancy can in part be attributed to the error bars. The larger part may, however, be due to problems of the reference profiles.

The  $\text{O}_3$  data analysis is not yet in a state to allow derivation of vertical profile amplitudes. To obtain, nevertheless, a rough indication of the behaviour of  $\text{O}_3$ , the temporal variations of the Biscarosse data

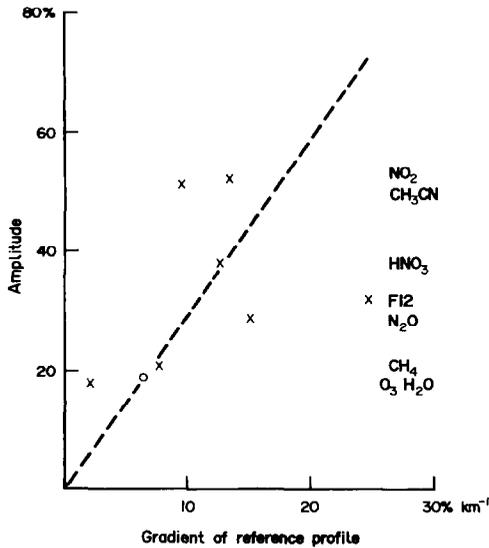


FIG. 13. DEVIATION AMPLITUDES VS GRADIENTS OF REFERENCE PROFILES FOR EIGHT TRACE CONSTITUENTS. Gradients are for 30 km altitude. Dashed line represents an "equivalent displacement height" of 3 km (see text).

shown in Fig. 10 were assumed to be representative of spatial variations. The inferred amplitude is given in Fig. 13 as a circle. The gradient of the long-term mean ozone profile for Biscarosse (September, 30 km) is used as the abscissa value.

Taking into account these limitations, Fig. 13 quite nicely shows the expected correlation of amplitudes and gradients. It should be noted that even if the limitations mentioned were not present, the distribution in Fig. 13 would show considerable scatter. This is because the measurements are not taken during the wave extrema only, but also at intermediate phases, as it was discussed above (see also Fig. 12). Many of the amplitude data shown in Fig. 13 therefore are presumably lower limits to the real amplitudes.

#### 4. CONCLUSIONS

The results shown suggest that considerable wave activity was present during the MAP/GLOBUS 1983 campaign. Periods involved were rather long (10 days), whereas horizontal scales were relatively small (1300 km). Respective results are still preliminary. Analysis of the large amount of ozone data taken during the campaign has not yet been finished, and should yield more accurate results on spatial and time scales in the future.

Such results should enable us to accurately determine the phases at which the various other balloon measurements were taken during the campaign. This

may offer the opportunity to derive time-dependent correction factors for specific trace gas measurements like those of NO<sub>2</sub> from the measured deviations of another gas (like O<sub>3</sub>) and the ratio of the reference profile gradients of the two gases. This could, for instance, facilitate the analysis of the diurnal variations of NO<sub>2</sub> (see Pommereau *et al.*, 1987).

If wave structures like the one described here are frequently present in the atmosphere, they would pose problems for *in situ* as well as for satellite measurements in the middle atmosphere. Single *in situ* measurements would not be able to safely determine the existence of a wave, and the structures possibly contained in the data would be interpreted as noise. Recently, Ehhalt *et al.* (1983) performed a scatter analysis for a number of balloon measurements of the source gases CH<sub>4</sub>, N<sub>2</sub>O, F11, F12 and for O<sub>3</sub>. Using a mixing length hypothesis these authors arrive at the conclusion that most of the observed variance of long-lived trace gases is caused by transport. For this transport they obtain from the scatter data an "equivalent displacement height", which is between 2 and 3 km at 30 km altitude. If in a very rough approximation their standard deviations are identified with the wave amplitudes of the present analysis, a displacement height of 3 km can be represented by the dashed line in Fig. 13. An interesting agreement is obtained, and hence considerable support is given to the idea that a large part of the data scatter is an artifact of the data-taking techniques.

Satellite measurements in the middle atmosphere should experience similar difficulties. These would mostly result from the coarse spatial resolution of the measurements, which frequently is given by the distance from one orbit to the next. This is much larger than the wavelength seen in the data discussed here. (Satellites with better horizontal resolution mostly have a very limited vertical resolution.) In such a case a satellite instrument would measure accidental values depending on where and in what direction its line of sight encounters the wave. Not knowing that a wave is there, this data would be thought to be very noisy. Similar problems may arise if singular ground truth measurements are performed to check on the validity of satellite data.

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