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OBSERVATIONS OF MIDDLE ATMOSPHERIC CH_4 AND N_2O VERTICAL DISTRIBUTIONS BY THE SPACELAB 1 GRILLE SPECTROMETER

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Abstract. Methane and nitrous oxide have been observed by limb absorption spectrometry using the Spacelab One grille spectrometer. The CH_4 ν_3 band is obsserved with a 0.1 cm^{-1} resolution up to 80 km while N_2O can be seen up to 52 km in strong lines of its ν_3 band. The values are compared, where possible, with previous observations and are in agreement with rocket determinations.

Introduction

Methane and nitrous oxide are known to be atmospheric minor constituents for about forty years [Adel, 1938; Migeotte, 1948]. The interest in their observation has been driven mainly in the last years by their role as tracers of stratospheric and mesospheric transport, they have no presently known production above ground level; also they are important in themselves as sources of nitric oxide and hydrogen in the upper atmosphere [Brasseur and Solomon, 1984] Moreover, the recent evidence of their increase in the last centuries [Rasmussen and Khalil, 1984] has led to speculate on their effects on radiative transfer and photochemistry.

CH_4 has been previously observed from aircrafts and balloons [Ackerman et al., 1977, 1978; Farmer et al., 1979; Borghi et al., 1983; Ehhalt et al., 1983; Rinsland et al., 1984] as well as N_2O [Ehhalt et al., 1983; Borghi et al., 1983; Rinsland et al., 1982; Farmer et al., 1980 and references therein]. Methane has also been observed at higher altitudes by means of sampling rockets [Ehhalt et al., 1972, 1975]; these results being completed by the satellite data of Jones and Pyle (1984).

Space observation of nitrous oxide was performed too using the Nimbus 7 pressure modulated cell SAMS radiometer [Jones and Pyle, 1984] up to the stratopause levels; the purpose of the spacelab grille spectrometer was to extend these observations up to the altitude where these species become undetectable at the 0.1 cm^{-1} instrumental resolution.

Observation

The grille spectrometer and its operation from November 28 to December 4, 1983 have already been described [Lemaître et al., 1984]. It is a solar absorption limb sounding spectrometer and was first flown during the Spacelab One flight. Due to the launch date and orbit inclination the sunset occultations were all performed at middle Northern latitudes while the sunrises were observed at high Southern latitudes. Two observations of CH_4 and one of N_2O were successfully performed, all in sunset mode, thus taking place at Northern latitudes: 34°N for N_2O and 27°N for both CH_4 observations.

The two CH_4 observations were performed in two wavelengths intervals : the ν_3 Q branch spanning 3010. to 3020 cm^{-1} covering the most intense CH_4 features and the ν_3 band manifold around 2978.8 cm^{-1}. For N_2O, the choosen interval was from 2205 to 2212 cm^{-1} and was including the strong R_2, R_1, P_2, P_3, P_4 and P_5 ν_3 N_2O lines. The choice of these intervals responded to a need to detect these two constituents at the highest possible altitude. Typical spectra are shown on Figures 1 and 2, showing the growth of the lines with decreasing limb altitudes. CH_4 was observed in its Q branch from 80 to 25 km and in the 2978.8 cm^{-1} manifold from 80 TO 35 km; N_2O was seen from about 52 to 28 km. Both observations being performed at a resolution close to 0.1 cm^{-1}.

Inversion of spectra

The spectroscopic data were taken from the AFGL line parameters table, 1982 edition [Rothman et al., 1983] while the atmospheric model used was the mid-latitude spring-fall U.S. standard atmosphere which was found in reasonable agreement (around 5% in temperature) with the corresponding meteorological lower stratospheric satel-

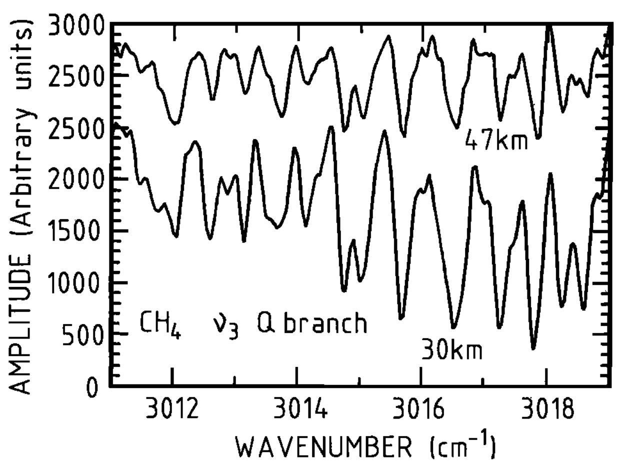

Fig. 1. Solar spectra showing the telluric CH_4 Q branch obtained at two different limb altitude during sunset.

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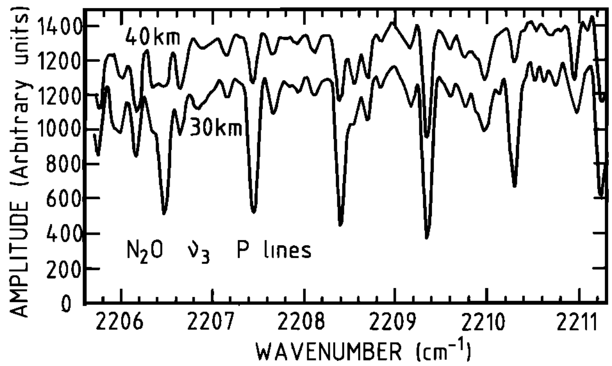

Fig. 2. Solar spectra showing telluric N_2O lines obtained at two different limb altitudes during sunset;

lite soundings [Planet, personal communication, 1984].

The accuracy of the molecular parameters is very high for N_2O of which single lines are observed but still could be improved for CH_4 [Toth et al., 1981] as some lines remain unresolved blends even at the spectral quality of fundamental laboratory work; in the worst CH_4 cases, a 20% error on absolute line strengths remains possible.

The inversion was made using an onion peeling technique fitting calculated equivalent line widths and observed ones by an iterative process using a Voigt line shape, the altitude resolution being 2 km. The results were simultaneously checked by simulating the observed spectra by synthetic ones adjusted by trial and errors. The obtained vertical distributions are shown on Figures 3 for CH_4 and 4 for N_2O while numerical results and observational conditions are presented in Tables 1 and 2.

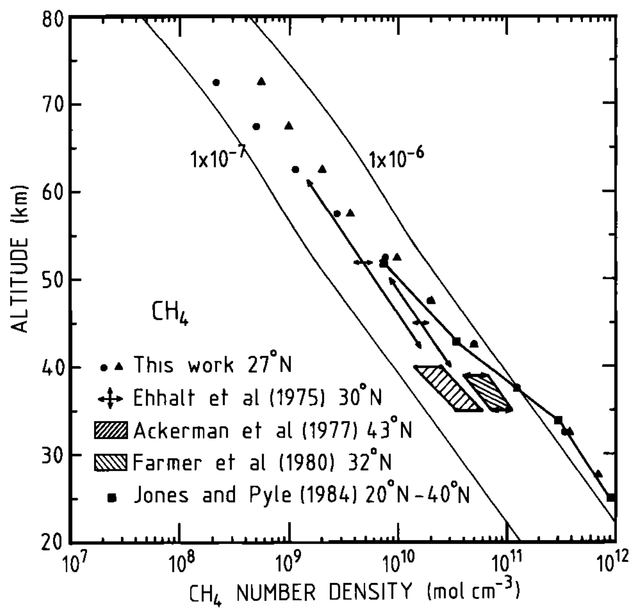

Fig. 3. Obtained CH_4 vertical distributions. Triangles and circles correspond respectively to the CH_4 Q branch and the 2978.8 cm^{-1} CH_4 manifold. Error bars are ± 15% of the indicated values. These data are compared with the upper stratospheric and mesospheric average observations of Jones and Pyle (1984) and Ehhalt et al. (1972, 1975).

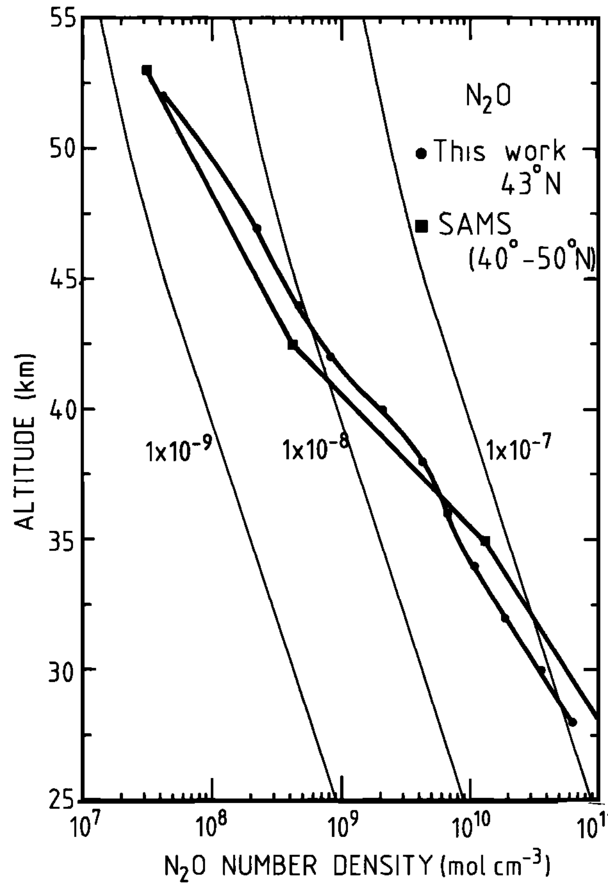

Fig. 4. Obtained N_2O vertical distribution. Errorbars are ± 15% of the indicated values. The results are compared with the upper stratospheric average observations of Jones and Pyle (1984).

The results are indicated in concentrations which are the measured quantities because for absorption measurements, the model dependence of the derived number densities is much lower than the dependence of the conversion to mixing ratios. In the case where these data should be used in comparison with theoretical models, we would suggest that the modelers use the pressures and temperatures actually derived in their theoretical computations, as long as they correspond to the time and location of the observations.

The error on the measurements of equivalent widths range from 20% on the top of the distribution to lower values in the botton where the absorptions are the strongest; however, due to strong non-linearities in the absorption, the uncertainties on the quoted densities are higher. In order to determine a precise error bar, the synthetic spectrum program was used and it was shown that concentrations differing by 15% from those indicated on figure 3 and 4 would have led to significantly different spectra in all the observation altitude range and thus ± 15% becomes our tentative error bar. These data could also be slightly modified, especially in their bottom part, if more refinement was to alter signifi-

TABLE 1 : CH_4 vertical distribution.

Altitude	CH_4 Q branch	CH_4 2978.8 cm^{-1}
27.5	7×10^{11}	
32.5	2.8 -	3.36×10^{11}
37.5	1 -	1.11 -
42.5	5×10^{10}	5×10^{10}
47.5	2 -	2 -
52.5	1 -	7.5×10^{9}
57.5	3.7×10^{9}	2.85 -
62.5	2 -	1.25 -
67.5	1 -	5×10^{8}
72.5	5.4×10^{8}	2.1 -
Latitude	27° N	27° N
Longitude	176° W	161° E
Time (GMT) (1983, Dec. 3)	3h 47m	5h 16m

* Error bars are ± 15% of the indicated concentrations.

cantly the orbit parameters of the first Spacelab mission, parameters used in these determinations are still those communicated directly by NASA orbit engineers during the flight and immediately after landing; their correctness was already checked in the case of CO_2 determination from the grille spectrometer spectra obtained together with the CH_4 data [Lippens et al., 1984] where a constant CO_2 mixing ratio was observed up to 98 km and has now been confirmed by the latest release of flight data tapes.

Discussion and conclusion

The two CH_4 vertical distributions obtained at the same latitude but at 23° longitude difference and separated by 1h 30m give the same values below 50 km but at higher altitudes they show a divergence reaching a factor of 2 at 75 km. The origin of this difference cannot be explained now, the discrepancy is too high to be attributed to errors in the AFGL line parameters. Two possibilities are left: first, deviations from a rigorous Voigt profile cannot be completely excluded in the mesosphere and secondly, a short term spatial and temporal variability of mesospheric methane cannot a priori be ruled out. This problem will be addressed during the next flight of the grille spectrometer, in late 1986, where more CH_4 runs will be scheduled in several spectral intervals.

Few other CH_4 and N_2O observations have been performed at altitudes higher than 35 km and those are compared on Figure 3 and 4 with our observations. Until now, no models spanning the entire middle atmosphere have been published and no comparison with model data will be attempted. For CH_4, observations are up to about 55 km the SAMS data of Jones and Pyle (1984), which are represented for the corresponding latitudes on Figure 3 and the 32°N rocket data of [Ehhalt et al., 1972, 1975]. These three data sets show satisfactory agreement with the data presented here above 40 km, the Jones and Pyle (1984) vertical distribution agreeing down to 27 km altitude. It should be stated here that the SAMS data are zonal averages and annual means. Between 35 and 40 km altitude, two balloon data sets have been published by Ackerman et al. (1977) and Farmer et al. (1980) using infrared spectrometry in the ν_3 band, respectively at 43°N and 30°S; Farmer et al. (1980) publish also a 32°N profile limited to 37 km which is lower by 25% than the 30° S profile but still higher than the upper limit of Ackerman et al. (1977). The latitude gradients suggested by these balloon data sets are in contradiction with the Jones and Pyle (1984) results where the higher latitudes mixing ratio are actually higher than the mid-latitudes values. SAMS data [Jones and Pyle, 1984] show in this respect a very complex structure with marked seasonal variations and an equatorial maximum coupled with a tropical minimum followed by again an increase toward the higher latitudes.

In the nitrous case, there is a good agreement in the whole range of our observation with the corresponding Jones and Pyle (1984) average, other measurements are limited to the 35 km altitude with the exception of two values reported by Ehhalt et al. (1975) : (3 ± 7) ppb for the 40-50 km altitude range and (0.3 ± 7) ppb for the 42-62 km altitude range; the upper limits include the present determinations. There is agreement in the 28-35 km range between the here reported values and the in situ or infrared measurements [Schmeltekopf et al., 1977; Farmer et al., 1980]; the Jones and Pyle (1984) data being,

TABLE 2 : N_2O vertical distribution

Altitude (km)	Concentration (cm^{-3})
28	6.3×10^{10}
30	3.8 -
32	1.9 -
34	1.05 -
36	6.8×10^{9}
38	4.2 -
40	2.1 -
42	8.1×10^{8}
44	4.7 -
47	2.3 -
52	4.25×10^{7}
Latitude	43° N
Longitude	154° N
Time (GMT) (183, Nov. 30)	2H 42m

* Error bars are ± 15% of the indicated concentrations.

on their lower altitude values at the upper limit of other measurements.

A second flight of the instrument is scheduled on the Spacelab Earth Observations Mission 1/2 in late 1986, while the CH_4 ν_3 Q branch will again be observed, other P manifold will be included; N_2O will be observed in the 2240 cm^{-1} spectral interval in order to enhance the instrumental signal to noise ratio and to be able to detect it at higher altitudes.

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