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Trace constituents measurements deduced from spectrometric observations on board Spacelab

by

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FOREWORD

The text "Trace constituents measurements deduced from spectrometric observations on board Spacelab" will be presented at the A1 topical meeting on Measurement of Minor Species, XXV COSPAR Assembly. It will be published in Advances in Space Research.

AVANT-PROPOS

Le texte "Trace constituents measurements deduced from spectrometric observations on board Spacelab" sera présenté à la session A1 sur la mesure d'espèces en traces, XXV Assemblée du COSPAR. Il sera publié dans Advances in Space Research.

VOORWOORD

De tekst "Trace constituents measurements deduced from spectrometric observations on board Spacelab" zal voorgesteld worden op de A1 vergadering over de meting van minderheidsbestanddelen, XXV COSPAR Bijeenkomst. Hij zal gepubliceerd worden in Advances in Space Research.

VORWORT

Der Text "Trace constituents measurements deduced from spectrometric observations on board Spacelab" wird präsentiert werden an der A1 Sitzung über der Messung von Minoritätsbestandteile, XXV COSPAR Tagung. Er wird in Advances in Space Research herausgegeben werden.

TRACE CONSTITUENTS MEASUREMENTS DEDUCED FROM SPECTROMETRIC OBSERVATIONS ON BOARD SPACELAB

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Abstract

The observation of infrared absorption lines by means of a grille spectrometer on board Spacelab 1 allows the determination of CO_2 and CO in the low thermosphere and in the middle atmosphere. Equal abundances of CO and CO_2 are found at 112 ± 2 km altitude. CO_2 is observed to depart from its homospheric volume mixing ratio near 100 km, dropping by a factor of 10,15 km higher. The CO largest number density is observed near 70 km altitude, close to the H Lyman alpha photoproduction peak.

The analysis of one run dedicated to the observation of water vapor shows a middle atmospheric mixing ratio of this species within the limits : 2 to 5 ppmv up to 70 km altitude, with the indication of a slight increase from 30 to 50 km altitude. The H_2O mixing ratio drops very rapidly above 70 km.

The comparison of the results from strong and weak H_2O and CO_2 lines shows the need to introduce a natural Lorentz line width in the line profiles.

Résumé

L'observation de raies d'absorption infrarouge au moyen d'un spectromètre à grille à bord de Spacelab 1 permet la détermination de CO et CO_2 dans la basse thermosphère et dans l'atmosphère moyenne. CO et CO_2 sont observés en concentration égale à 112 ± 2 kilomètres d'altitude. La concentration volumique homosphérique de CO_2 est maintenue jusqu'à 100 km pour décroître à plus haute altitude d'un facteur 10 en 15 kilomètres. La concentration la plus élevée de CO est observée aux environs de 70 km d'altitude, près du maximum de photo-production par la raie H Lyman alpha.

L'analyse d'une observation de la vapeur d'eau montre une concentration volumique dans les limites 2×10^{-6} à 5×10^{-6} jusqu'à 70 kilomètres d'altitude avec indication d'un léger accroissement de 30 à 50 kilomètres. La concentration volumique de H_2O décroît abruptement au-dessus de 70 km.

La comparaison des résultats obtenus à partir de raies fortes et faibles de H_2O et de CO_2 montre la nécessité de tenir compte lors du traitement des données d'une largeur naturelle de Lorentz dans le profil des fortes raies d'absorption utilisées.

Samenvatting

De waarneming van infrarode absorptielijnen met een raster-spectrometer aan boord van Spacelab 1 laat toe CO en CO_2 te bepalen in de lage thermosfeer en de middenatmosfeer. CO en CO_2 worden in gelijke concentratie waargenomen op 112 ± 2 km hoogte. De homosferische volumeconcentratie van CO_2 wordt tot op 100 km in stand gehouden om daarna op grotere hoogte, 15 km, te dalen met factor 10. De grootste CO concentratie wordt waargenomen op ± 70 km hoogte, dicht bij het maximum van de fotoproduktie van de H Lyman alpha straal.

De analyse van een waarneming van de waterdamp toont een concentratie binnen de limieten 2×10^{-6} en 5×10^{-6} tot op 70 km hoogte, met aanduiding van een lichte verhoging van 30 tot 50 km. Boven 70 km hoogte daalt de concentratie van H_2O bijzonder snel.

De vergelijking van de resultaten bekomen uit sterke en zwakke H_2O en CO_2 lijnen toont aan dat het noodzakelijk is, bij de gegevensverwerking, rekening te houden met een natuurlijke Lorentz lijnbreedte in het profiel van de gebruikte sterke absorptielijnen.

Zusammenfassung

Die Beobachtung von infraroten Absorptionslinien mit einem GrilleSpektrometer an Bord Spacelab 1 erlaubt CO und CO_2 zu bestimmen in der Niederthermosphäre und der Mittelatmosphäre. CO und CO_2 werden in gleicher Konzentration beobachtet auf 112 ± 2 Km Höhe. Die homosphärische CO_2 Volumenkonzentration wird bis zu 100 Km bewahrt um danach auf grösserer Höhe, 15 Km, ab zu nehmen mit einem Faktor 10. Die grösste CO Konzentration wird beobachtet auf ± 70 Km Höhe, dicht beim Maximum der Photoproduktion der H Lyman alpha Linie.

Die Analyse einer Beobachtung der Wasserdampf zeigt eine Konzentration in den Limiten 2×10^{-6} und 5×10^{-6} bis zu 70 Km Höhe, mit Andeutung einer kleinen Erhöhung von 30 bis zu 50 Km. Über 70 km Höhe nehmt die Konzentration von H_2O sehr schnell ab.

Die Vergleichung der Resultaten bekommen aus starke und schwache H_2O und CO_2 Linien zeigt die Notwendigkeit, bei die Datenverwertung, zu rechnen mit einer naturlichen Lorentz Linienbreite im Profil der starke benützte Absorptionslinien.

INTRODUCTION

Infrared absorption spectrometry of the atmosphere, using the sun as a light source at sunrise or sunset, has for the past 15 years proved to be a powerful method of studying vertical distributions of trace species. The largest possible amount of light-absorbing molecules is observed on the optical path tangent to the earth's surface at various altitudes, allowing the deconvolution of very low concentrations as a function of altitude. A great deal of information has already been gathered with this method from high-altitude platforms such as aircraft and balloons. An orbiting platform provides access to higher altitudes and to nearly global coverage. Whereas with high-altitude platforms the earth's rotation provides altitude scanning at sunrise or sunset, scanning is achieved at a much higher rate from an orbiting spacecraft. Such fast spectral scanning requires a high-throughput instrument.

One of the main goals of the grille spectrometer operating on the Spacelab pallet was to observe trace species in the low thermosphere and in the mesosphere such as CO_2, CO, H_2O and CH_4. Preliminary data on this latter molecule have already been published (1). A part of the results on the three others will be presented here. They confirm previous mass spectrometric measurements on CO_2 in the transition atmospheric region, the turbopause, where mixing is replaced by diffusion. They show the large increase of the CO mixing ratio at altitudes where the CO_2 photodissociation by the solar H Lyman α radiation takes place and where the H_2O abundance vanishes abruptly.

INSTRUMENTATION

The optics consists of a two-axis steerable frontal plane mirror which tracks the sun in front of a Cassegrain telescope with aperture 30 cm and focal length 6 m. The sun is imaged on the grille, which intercepts a square portion of the solar image (8 arc minutes). The spectrometer has a grating of 59 grooves per millimeter, which is illuminated by a parabolic mirror oscillating at 436 Hz with an amplitude of ± 20 arc seconds; the position of the mirror is controlled within 5 arc seconds. The exit light flux, split into two beams, passes through interference filters to two detectors (InSb, 2.5 to 5.5 μm, and HgCdTe, 2.5 to 10.5 μm). The spectral resolving power is at least 1.3×10^4 (instrumental line width at half peak height).

The electronics in the Spacelab modules links the pallet instrument to the Command and Data Management System (CDMS) and the high-rate multiplexer (HRM). Using data originating from the orbiter (time, attitude, and orbit parameters) and from Spacelab (time-line, onboard and ground commands, sun ephemeris), it manages the execution of the stored measurement programs, including inflight up-dating. The electronics on the pallet instrument provides the functions of electromechanical control and signal detection and formatting. The main role of the crew was to check the instrument wavelength calibration, spectral resolution, and sensitivity by monitoring the display of a calibration spectrum generated inside the spectrometer with a calibration lamp shining through a gas cell. The mission specialist in charge of this task performed a wavelength alignment 12 hours after launch.

The pallet instrumentation weighed 122.8 kg, was 1.8 m high, and occupied 0.7 m^2. The weight of the module equipment was 15 kg. The data rate in operation was 51.6 kilobits per second.

TREATMENT OF THE DATA

Narrow spectral intervals of the central part of the solar disk are rapidly scanned while the sun is rising or setting. Telluric absorption features are then analysed using the onion pealing method on the basis of the absorption lines parameters (2) adapted for the atmo-spheric temperature and pressure conditions of the Mid Latitude - Spring Fall model. A Voigt line shape is used. The observed equivalent widths are inverted by successive iteration every 2 km from high to low altitudes.

RESULTS

Carbon dioxide

Several portions of absorption runs were scheduled during the mission to observe CO_2 at the highest possible altitudes where its mixing ratio has been already shown to depart from its homospheric values (3,4). The region of the strongest absorption lines was chosen (near 4.24 μm). Measurements for two runs are shown in Fig. 1. Event 13 has been studied in detail and the results from two lines are shown as obtained in the linear part of their curve of growth where the equivalent width is small but very reliable since the line width is practically only due to Doppler broadening. The line parameters are reproduced in Table 1.

Up to 100 km altitude, the CO_2 volume mixing ratio follows closely the 3×10^{-4} value and drops abruptly above by a factor of 10 over 15 kilometers. These findings are in good agreement with the mass spectrometric data (3,4). These results give great confidence in the data used both from the instrument and from the shuttle (state vector and altitude versus time).

Carbon monoxide

It is known that carbon dioxide must be most strongly photo-dissociated in the upper mesosphere and thermosphere by solar U.V. radiation. In fact, the H Lyman α radiation is mostly responsible for the CO production as shown in figure 2 where the production of CO is shown as well as the contribution of H Ly α and of the rest of the U.V. solar spectrum. The values shown here have been computed for

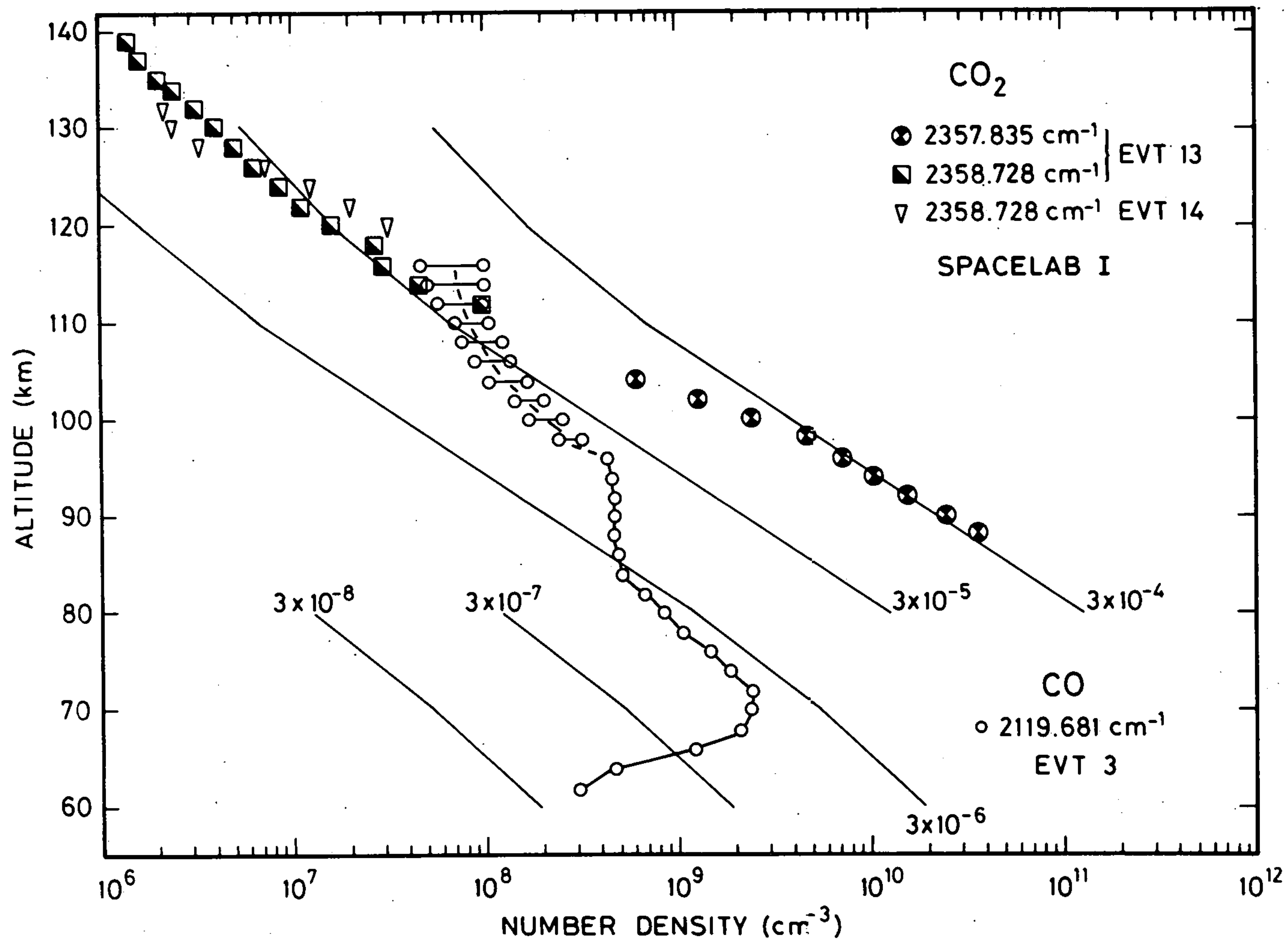

Figure 1. Number density of CO_2 and CO in molecules per cm^3 versus altitude using two different CO_2 absorption lines and the P_6 CO line. The nearly parallel lines represent constant volume mixing ratios from 3×10^{-8} to 3×10^{-4}.

Table 1.- Line parameters used : integrated absorption cross section at 286°K, (S), in cm; energy of the upper state (E) in cm^{-1}; pressure half width at half peak height, (W_p), in cm^{-1} and one atmosphere; wavenumber, (ν), in cm^{-1}.

Species	ν	S	E	W_P
CO_2	2357.835	7.77×10^{-20}	1031.129	.069
	2358.728	3.53×10^{-18}	60.871	.074
CO	2119.681	3.65×10^{-19}	80.735	.072
	2154.596	2.60×10^{-19}	11.535	.080
H_2O	3816.092	2.10×10^{-19}	79.496	.0942
	3819.905	9.30×10^{-21}	136.762	.0940
	3824.281	3.47×10^{-21}	142.274	.0923

two solar zenith angles (0° and 78°) using Nicolet's study of the absorption of H Ly α by oxygen (5). Few ground based mesospheric CO measurements have been published (6,7), all show a large mixing ratio increase from the stratopause to the mesopause.

Two absorption runs provided data on CO during the Spacelab One Mission. The spectral lines used are listed in Table 1 and the inverted abundances are shown in figure 3. The P_6 line was observed at sunset in the northern hemisphere yielding equivalent widths at 70 km which are at the upper limit of low linear part of the curve of growth leading to some uncertainty on the maximum value observed at that altitude. In addition this line is contaminated by O_3 absorptions below 60 km precluding CO determinations at lower altitude. The other observation took place at sunrise in the southern hemisphere. In this case the weaker R_2 line was used which provided data down to the low stratosphere where a mixing ratio value equal to 10^{-8} is reached at 30 km altitude in agreement with balloon borne measurements (8,9).

In both cases the thermospheric upper values are in good agreement indicating (figure 1) an equal abundance of CO and CO_2 at 112 ± 2 km altitude. At lower altitudes the vertical profiles exhibit similar shapes with a maximum abundance near the production peak shown in figure 2. Figure 1 also shows a well marked change in the CO profile at the turbopause level which occurs at higher altitude in the northern hemisphere observation than in the southern hemisphere run. The results indicate also a large variability of the mesospheric CO.

At the wavelengths of the P_6 and R_2 lines solar lines have small equivalent widths : respectively 5×10^{-3} and 10^{-2} cm^{-1}. In solar atmospheric models these lines exhibit a broad shape leading to a small influence on the solar continuum intensity (10). Their effect has hence been neglected. A more refined treatment will be undertaken which should not alter the results presented here more than the scatter of the observed equivalent widths.

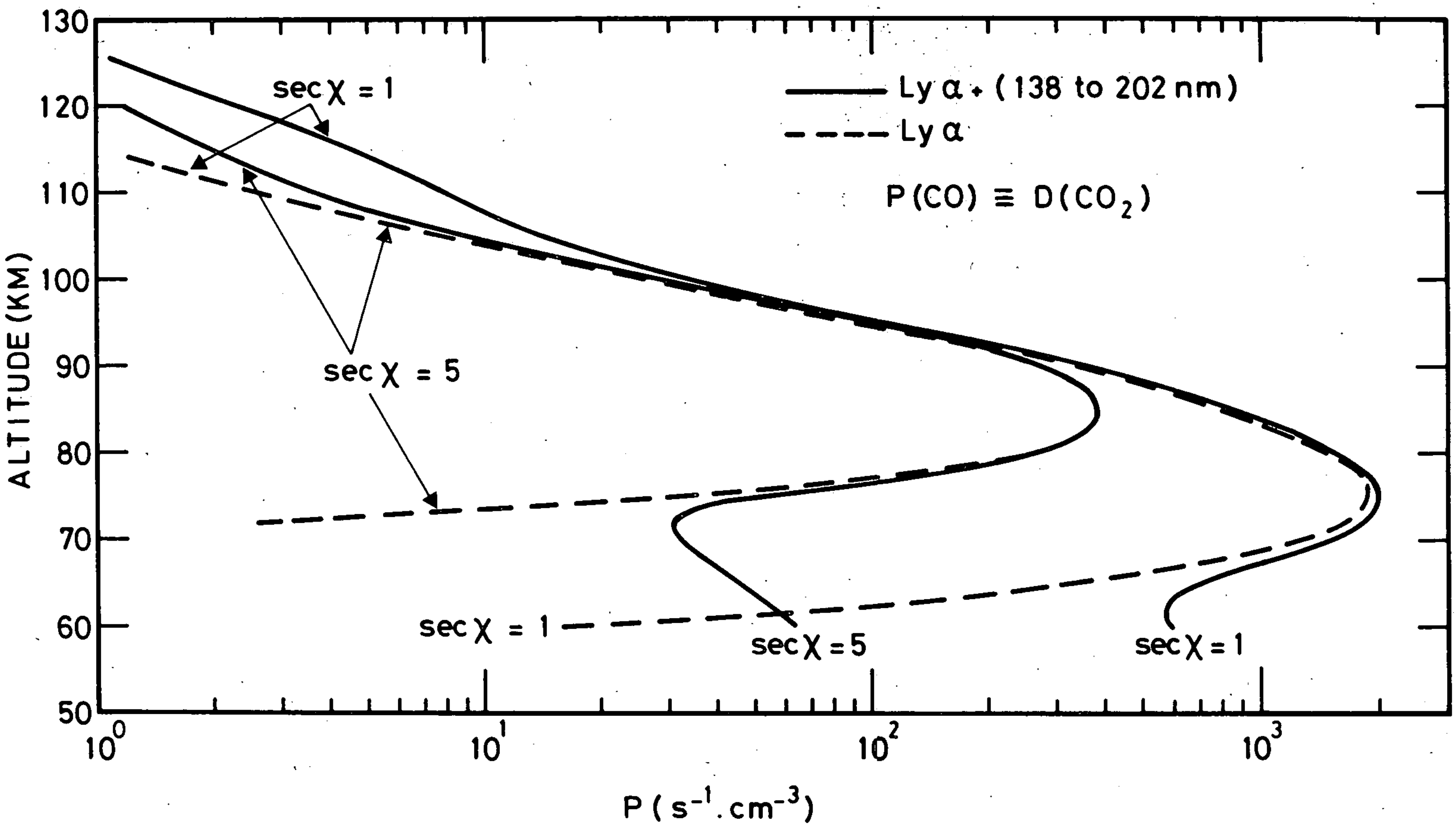

Figure 2. Photodissociation rate, P, of CO_2 versus altitude for two solar zenith angle ≡. The dashed curves correspond to the effect of the H Lyman α line alone while the total rate is represented by the solid curves. This process is the only known source of CO in the upper atmosphere.

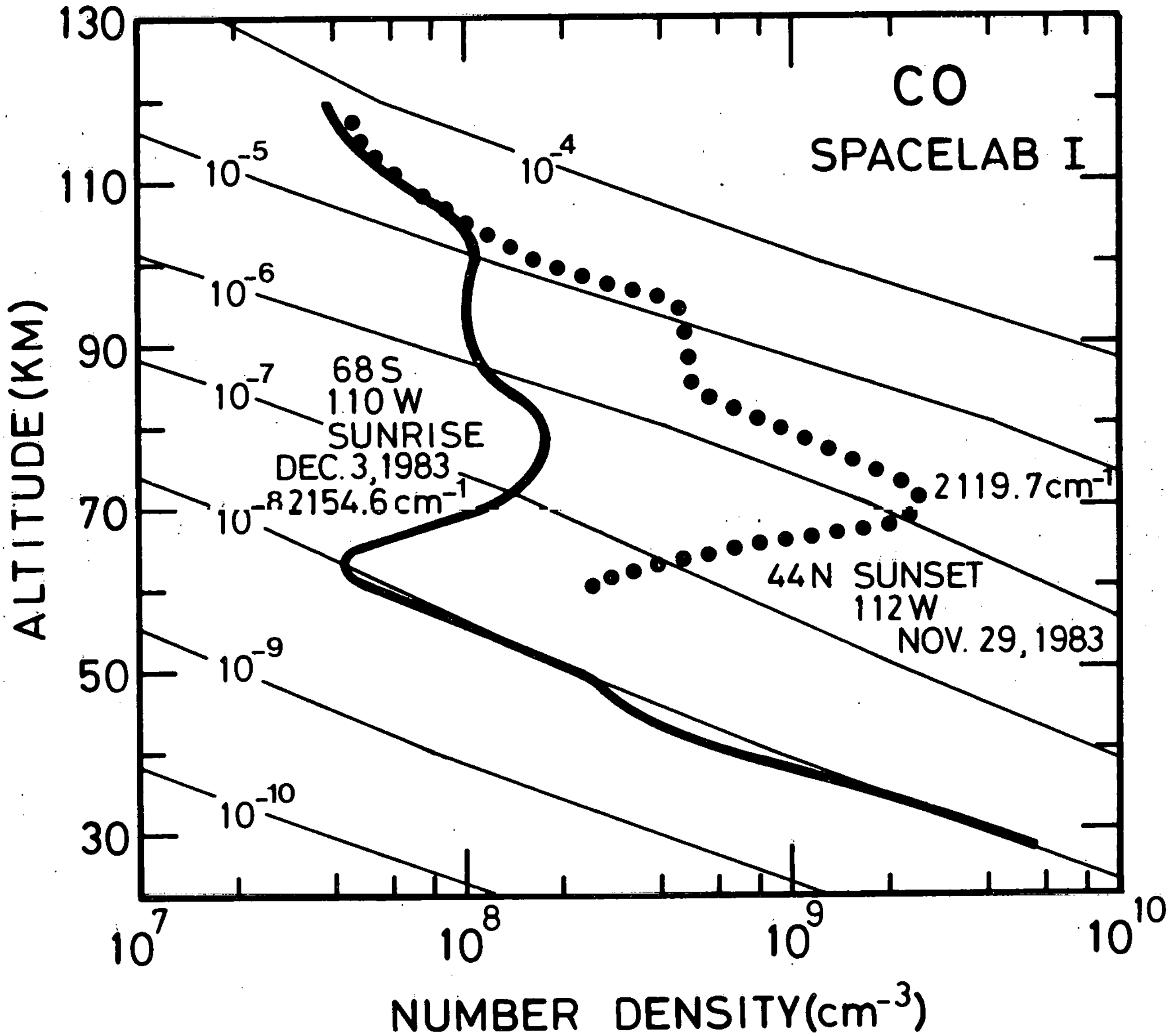

Figure 3. Carbon monoxide number density versus altitude retrieved from two solar occultation runs, one in the southern hemisphere at sunrise and the other one in the northern hemisphere at sunset. The geographic coordinates of the solar rays tangent points at 50 km altitude are indicated. Two different absorption lines were used of which the wavenumbers are indicated. The nearly parallel lines represent constant volume mixing ratios from 10^{-10} to 10^{-4}. The dates of data capture are also given.

Water vapor

The most recently published summary of water vapor determinations from 20 to 90 km altitude (11) indicates a wide range of mixing ratio values : from 0.2 to 15 parts per million in volume depending on altitudes and techniques. Several of our Spacelab runs were dedicated to the measurement of this trace constituent so important for the middle atmosphere aeronomy. Two wavelength regions were selected : 6.3 µm and 2.7 µm. Due to a better signal to noise ratio at the short wavelength, the results of a sample run near 3820 cm^{-1} are shown here.

Figure 4 shows the retrieved abundance of H_2O versus altitude using three absorption lines. The strong line at 3816.092 cm^{-1} leads to a broad maximum mixing ratio reaching 20 ppmv near 60 km altitude.

However the study of the curve of growth of this line indicates that the equivalent width observed at altitudes lower than 72 km altitude are beyond its low linear part making the data retrieval strongly dependent of the line shape. This is not the case for the two other lines shown in Table 1 for H_2O which lead to different H_2O abundances (fig. 4). The two sets of data can be reconciled as shown in figure 5 by adding a constant Lorentz half width at half peak equal to 2×10^{-4} cm^{-1} to the pressure dependent width, W_p, shown in table 1. This does not practically affect the results from the two weaker lines but reduces considerably the values deduced from the strong line.

This finding supports partially the introduction in the interpretation of rocket borne observations (11) of a super-Lorentzian line shape. It is of decisive importance for the interpretation of broad band and pressure modulated satellite measurements. The use in this case (12) of a super-Lorentzian line shape was found to only partially improve the results which may show to be further improved by the introduction of a pressure independent constant Lorentz width, at least at the low middle atmosphere pressures.

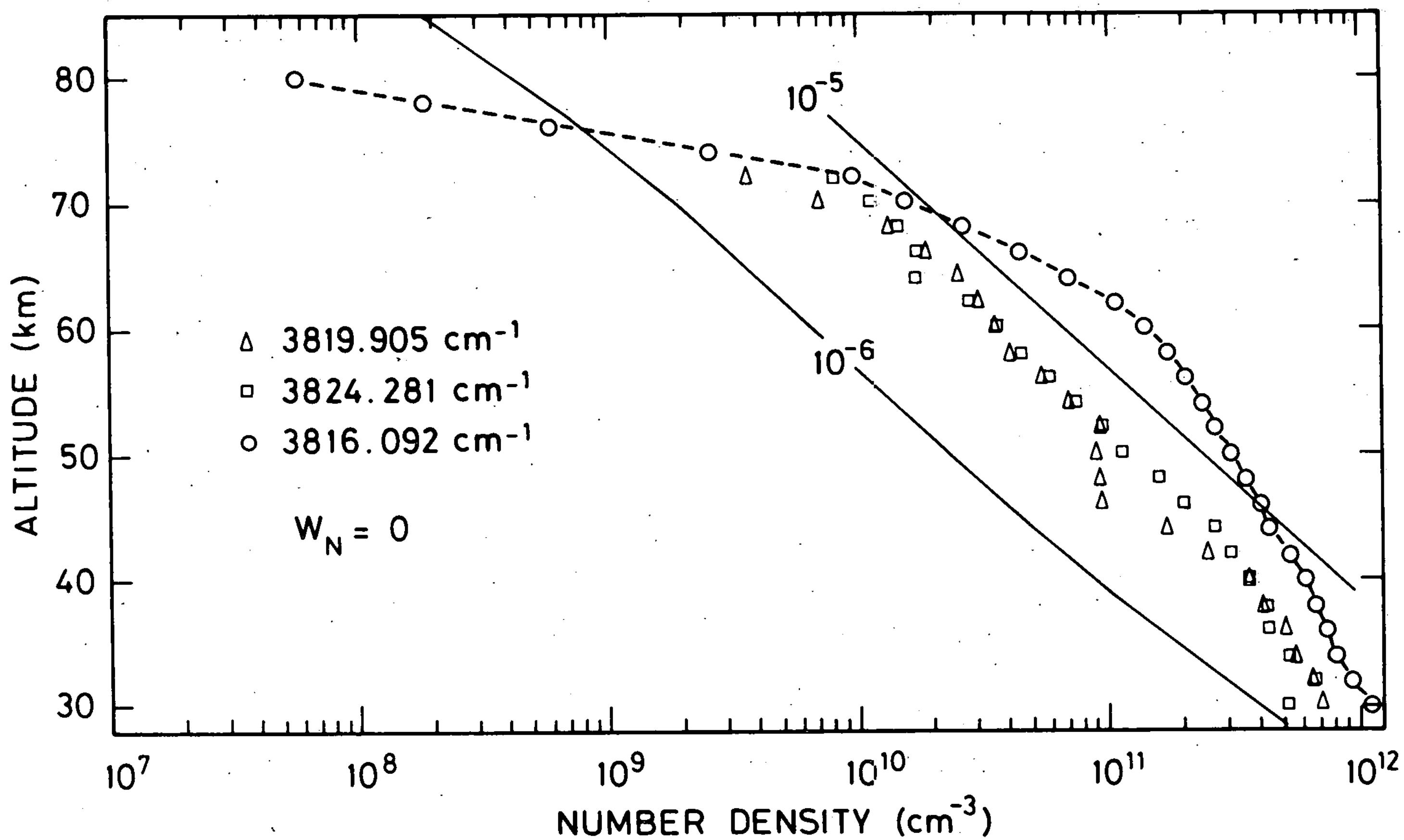

Figure 4. Number density of H_2O versus altitude. These profiles have been retrieved using the collision induced Lorentz half line width shown in table 1. The contradiction been the results from the strong line (3916.092 cm^{-1}) and from the two weaker lines is obvious.

The water vapor abundances shown in figure 5 can be summarized as follows. The water vapor mixing ratio from 30 to 70 km altitude is 3.5 ± 1.5 ppmv with a slight tendency to increase from 30 to 50 km. A decrease of the mixing ratio by a factor of at least 20 occurs from 72 to 80 km altitude.

DISCUSSION AND CONCLUSION

Vertical distributions of carbon monoxide in the mesosphere and in the thermosphere combined with distributions of carbon dioxide show for the first time where these two species present equal abundances. They also show a large CO variability indicating the need for more measurements. The water vapor vertical distribution in the middle atmosphere exhibit, for the case studied, a small variation with altitude up to about 70 km where the volume concentration drops abruptly. More observations performed during the mission will allow to refine this picture.

Previous observations from rockets and satellites had indicated the need to take into account an infrared rotation-vibration line shape different from the traditional convolution of a Doppler component and of a collision induced Lorentz component. We confirm this need. For H_2O the addition of a constant Lorentzian component brings in agreement the results obtained from weak absorption lines in their Doppler linear part of the curve of growth with those obtained from strong lines in the non linear part of the curve of growth at all altitudes and hence pressures. This indicates that an unexpected (11) natural broadening of the line occurs.

A similar phenomenon occurs in the 4.2 μm band of CO_2 which has forced us in this preliminary report to take into account results from weak absorptions only, using lines of decreasing strength to retrieve CO_2 abundances at lower and lower altitudes. As an example, in this band of CO_2 the line at 2345.985 cm^{-1} exhibits a value of W_N

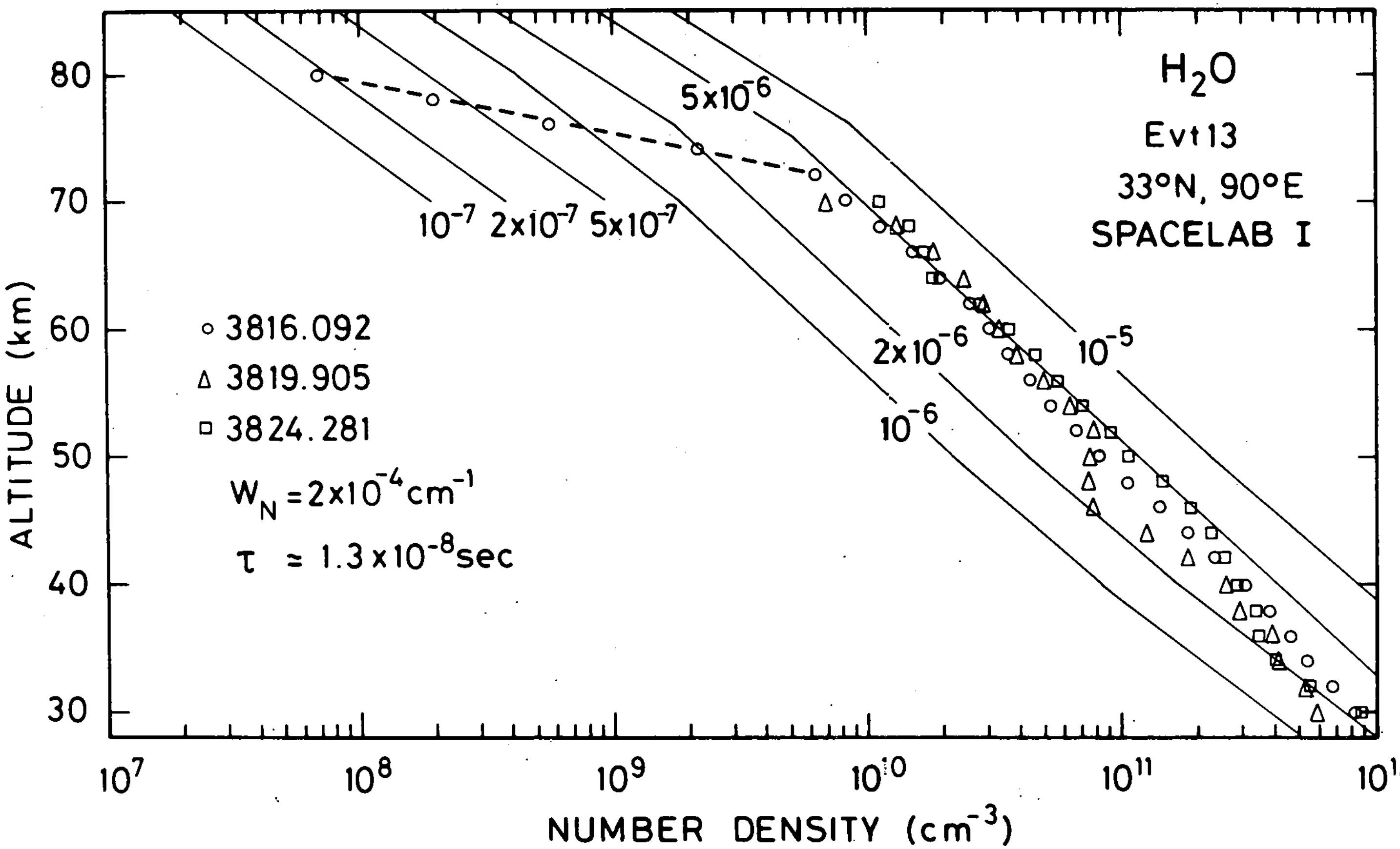

Figure 5. Number density of H_2O versus altitude retrieved after addition of a constant Lorentz half width at half peak height, W_N, equal to 2×10^{-4} cm^{-1} to the pressure dependent Lorentz width W_P shown in table 1. The three profiles show a good agreement. The nearly parallel lines represent constant volume mixing ratios from 10^{-7} to 10^{-5}.

equal to about ($1.5 \pm 0.5 \times 10^{-4}$ cm^{-1}). These aspects are of fundamental importance for molecular physics and for its application in atmospheric remote sensing of number densities and of temperatures. When it will be fully understood, it will benefit to broad spectral band measurements in atmospheres, by extending their dynamic range. It may also affect the atmospheric cooling rates evaluations (13).

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