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SECOND FLIGHT OF THE SPACELAB GRILLE SPECTROMETER DURING THE ATLAS-1 MISSION

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Abstract. The SPACELAB grille spectrometer on its second space flight during the ATLAS-1 mission (March 24 - April 2, 1992) took advantage of the favorable timeline and of the extra day to perform more than 65 successful solar occultation runs. It succeeded in obtaining spectra pertinent to its ten target molecules in the full range of altitudes available to the solar infrared occultation technique. These ten molecules are H_2O, CO, CO_2, CH_4, NO, NO_2, N_2O, HCl, HF and O_3. The preliminary analysis of the sunset observation presented here adds new information to the available database on HCl vertical profiles, for assessing long-term trends of this important stratospheric species.

Introduction

The purpose of the ATLAS missions, onboard the American Space Shuttle, is to study the evolution of atmospheric properties over an entire solar cycle. The grille spectrometer, which already flew onboard Spacelab-1 and was used for several balloon and airplane observations since 1972, measures vertical concentration profiles of trace gases. It uses the method of infrared absorption spectrometry during sunrise or sunset periods, with the sun as the source of light. The largest amount of absorbing molecules is observed along the optical path through the Earth atmosphere at various tangent altitudes from the stratosphere up to the lower thermosphere, leading to concentration profiles with an instrumental vertical resolution of about 4 km.

This paper gives an overview of the grille spectrometer observations performed during the ATLAS-1 flight (March 24 - April 2, 1992), and presents an example of data retrieval. Because of the strong interaction between chlorine and stratospheric ozone, a preliminary analysis of a vertical profile of hydrogen chloride has been chosen.

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Experiment

The grille spectrometer flew for the first time in space (Lemaître et al, 1984) onboard SPACELAB-1, with a launch on November 28, 1983. The flight came up to its objectives (see final review by Girard et al., 1988) despite an orbit entirely sunlit during the last six days of the ten days mission and numerous technical difficulties encountered for this spacelab test flight. Still, this mission provided an early database very useful for comparisons with later flights.

On the basis of the SPACELAB-1 grille experience, the scientific program (spectral windows, altitude range for each window) was optimized for the ATLAS-1 mission (Camy-Peyret et al., 1992). The use of a NASA provided high pressure nitrogen vessel (GN2) for cooling the detectors by Joule-Thomson expansion, and exploitation of radiative cooling through the open heliostat for avoiding overheating of the instrument internal electronics, allowed to run more occultation observations than during the SPACELAB-1 mission. The calibration, performed about four hours after launch, confirmed correct operation of the instrument. After correction of a few target errors due to the launch delay, the observations followed nominally except for the sunrises that were affected by late sun acquisition due to the fact that a wrong positioning of its multi-layer insulation hindered the heliostat from correct pointing. The real-time data quality was good, permitting optimization of the scientific program through uplink of some new spectral windows and new altitude ranges for each window. Emission observations were attempted, but the signal did not allow any reliable analysis : after two trials it was decided to cancel all emission runs in order to save resources.

Figure 1 shows the geographical distribution of the observations for each species.

Sample analysis

The occultation performed during the sunset in orbit 124 (54°S, 153°E) is chosen as an example of an absorption run because of the importance of hydrogen chloride measurements in the middle atmosphere. In particular, HCl, in the 40 to 50 km altitude range is a good indicator of the total active chlorine in the stratosphere (W.M.O., 1985).

The spectral window covers the $H^{35}Cl$ line at 2944.9 cm^{-1}, some lines of CH_4, and a few

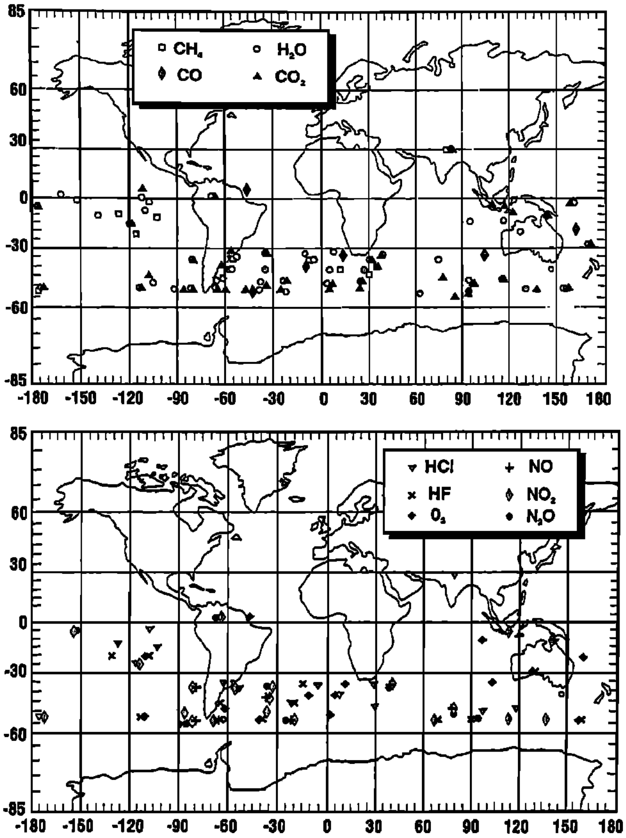

Fig. 1. Geographical distribution of trace species observations : (a) CH_4, H_2O, CO and CO_2; (b) HCl, HF, O_3, NO, NO_2 and N_2O.

solar lines. The HCl profile has been derived between 30 and 55 km, using 8 consecutive spectra (numbered 33 to 40) just before loss of sun. The measurements below 30 km were perturbed by the Pinatubo aerosols obscuring the lower part of the atmosphere. Figure 2 shows spectra obtained in this altitude range. Two independent retrieval methods were used in this work. The first one is based on an algorithm proposed by Mill, starting from the lower level and iterating to the highest tangent height spectrum. The second one uses a least squares global fit method : all the spectra are included simultaneously in order to retrieve the vertical concentration profiles together with the background. Identical parameters of the instrument function are used in both computations. The concentration values and uncertainty limits resulting from both methods are in good mutual agreement. The tangent heights used in the computation are derived from the orbital analysis performed by Marshall Space Flight Center orbit engineers during and at the end of the flight and are still preliminary.

Figure 3 shows the agreement obtained between observed and computed spectra around the HCl line. The final retrieved data reported in Table 1 and on Figure 4 are the averages and combined statistical uncertainties (black arrows of Figure 4) of both methods at any given level. These uncertainties take account of the 68% statistical confidence intervals from both methods. Because of uncertainties remaining on the orbit parameters, retrievals have been made

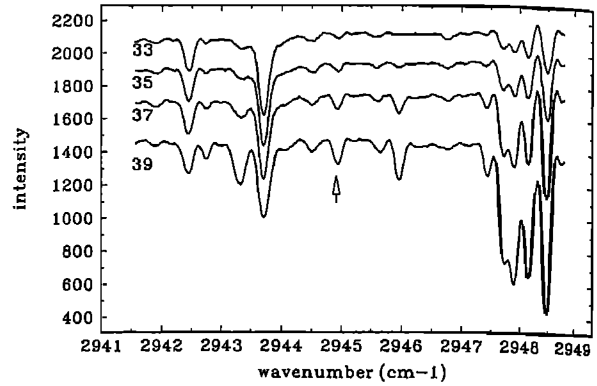

Fig. 2. Spectra recorded at sunset 124. The amplitude of the signal is shown versus wavenumber (cm^{-1}), nominally for spectrum 33; the others are successively shifted downwards by 200 amplitude units. Several solar absorption features appear in the spectral range. The HCl line at 2944.9 cm^{-1} is indicated by the arrow. The tangent altitudes of the line of sight to the sun's center of this absorption line are 55.7, 48.5, 41.2 and 33.8 km for spectra 33, 35, 37 and 39, respectively.

for the nominal values of the tangent heights, and for tangent heights equal to the nominal one plus or minus 1 km. The inclusion of the $\pm$ 1 km tangent height uncertainty increases the uncertainty on the retrieved values as indicated on Figure 4 (extended uncertainty intervals) and in Table 1.

Also shown in Figure 4 for comparison are the HCl vertical concentration profiles derived by ATMOS during the SL-3 flight in April-May 1985 (Zander et al., 1990) and during the BIC-2 balloon campaign in the second half of June 1983 (W.M.O, 1985). At the lowest altitude level, the value retrieved from the present data can be compared with data taken during a balloon flight of a similar grille spectrometer in 1975 (De Mazière et al., 1989).

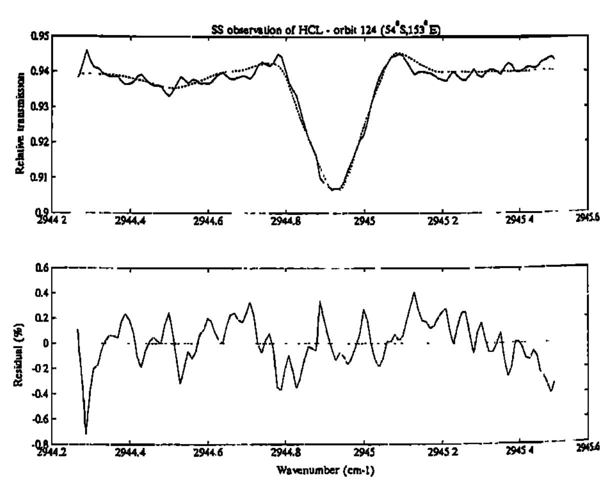

Fig. 3. The upper graph shows part of spectrum 37 versus wavenumber (in cm^{-1}) as the continuous line, the dashed line representing the computed spectrum. The lower graph shows the relative difference between computed and observed values (residual in %).

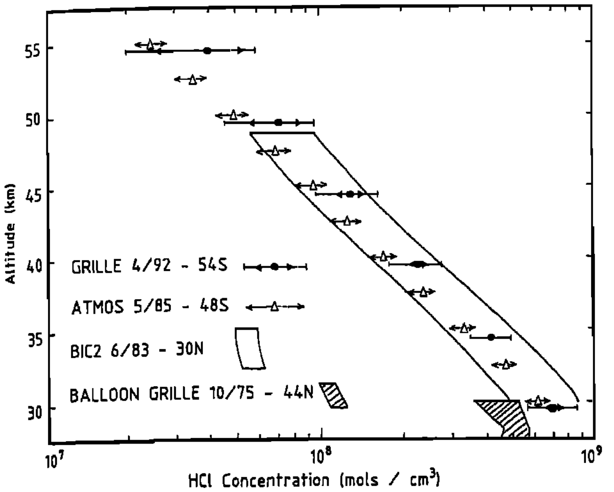

Fig. 4. Concentration of hydrogen chloride versus altitude.
The ATLAS-1 Grille values are represented by the closed circles and corresponding uncertainty limits, with the arrows excluding the effect of the + 1 km altitude uncertainty. Other data are shown for comparison : open triangles for Spacelab 3 - ATMOS 1985 (Zander et al. 1990), open envelope for BIC 1983 (W.M.O., 1985) and striped envelope for 1975 balloon data (De Mazière et al., 1989).

The overall data set does not give a clear indication of any steady long-term trend in the HCl stratospheric concentration. However, HCl is known to vary with season and latitude and the BIC-2 results have large uncertainties. Therefore, restricting the comparison to the ATMOS '85 and Grille '92 data, an increase of the HCl concentration becomes apparent, which is of order 18 to 28% between 35 and 45 km at which altitudes the relative uncertainties of both data sets are smallest and non-overlapping.

The same data set allows to estimate the column abundance of HCl between 30 km and 55 km to be of order 5.60×10^{14} mols/cm^2 based on the actual 1992 Grille data, in comparison with 4.70×10^{14} mols/cm^2 based on the 1985 Atmos data, which represents an increase of order 19%. Keeping in mind the uncertainties, this figure is of the same order of magnitude as the total column trends observed for the same period of time at the International Scientific Station of the Jungfraujoch (50%) (R. Zander, private communication), or at Kitt Peak Observatory (30%-35%) (Rinsland et al., 1991; Wallace and Livingston, 1991). In making such comparisons, the following points should be noticed. Figures for long-term trends of HCl are altitude dependent, since there is a time-lag of order 5 years between the source gas emissions (CH_3Cl, CFC, etc.) in the troposphere and the appearance of HCl in the stratosphere (Zander et al., 1992). The increases of most source gas emissions have flattened out since 1987 or even reversed after 1988, after steep rises between 1982 and 1987 (Weisenstein et al., 1992; AFEAS, 1992). Therefore one expects trends to be higher at higher altitudes in the stratosphere, as confirmed by our data. The fact that the ground-based column measurements show a stronger increase than the stratospheric column above 30 km may be related to the fact that the former ones include the tropospheric and lower stratospheric contributions that amount to 75% of the total column. Moreover, seasonal and latitudinal variations should be taken into account : the ground-based measurements cited above are northern hemisphere measurements, while the data set presented here relates to the southern hemisphere.

TABLE 1 : Results from the sunset in orbit 124.

z(km)	T(K)	n_{AIR} (10^{16}cm^{-3})	n_{HCl} (10^8cm^{-3})	VMR_{HCl} (ppbv)
55	250	1.10	0.37 ± 0.14	3.4 ± 1.3
50	263	2.01	0.67 ± 0.15	3.34 ± 0.75
45	258	3.90	1.22 ± 0.15	3.12 ± 0.38
40	240	8.25	2.15 ± 0.18	2.61 ± 0.22
35	230	17.7	3.95 ± 0.10	2.22 ± 0.06
30	224	38.2	6.99 ± 0.46	1.83 ± 0.12

z = geometric altitude, T = temperature, n = molecular number densities.
The error resulting from the ± 1 km tangent height altitude uncertainty is of order ± 14% and is not included in the above uncertainty limits that only represent the combined statistical uncertainties.

It has been verified by us that the slight difference between the HITRAN '92 parameters used by us and the ones used in the ATMOS '85 retrieval for the HCl R2 line cannot account for the observed increase. One must keep in mind however that erroneous estimation of the tangent altitudes of the spectra by only ± 1 km may alter our conclusion significantly, as shown in Fig. 4. So care should be exercised and more ATLAS-1 HCl observations analysed at various northern and southern latitudes before generalizing this early conclusion and investigating in detail the comparison with ground-based observations.

Conclusions

During the ATLAS-1 mission, the grille spectrometer has performed more than 65 solar occultation runs. The analysis of the recorded spectra has just started. It appears that no data retrieval will be possible below 25 to 30 km altitude due to volcanic aerosols.

The first preliminary results on HCl at 54°S seem to indicate an increase of 25% at 40 km altitude in comparison with 1985. This conclusion needs to be confirmed by analysis of more ATLAS-1 HCl observations at various latitudes and better determination of the orbital parameters.

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References

AFEAS, "Alternative Fluorocarbons Environmental Acceptability Study", Washington, D.C 20005, 1992.

Camy-Peyret, C., N. Papineau, R. Armante, V. Achard, J. Besson, C. Muller, M. De Mazière, C. Lippens, J. Vercheval, M. Ackerman and D. Frimout, "Grille" spectrometer scientific programs for the ATLAS-1 Space Shuttle flight, La Recherche Aérospatiale, 3, 67-77, 1992.

De Mazière, M., C. Lippens and C. Muller, Observations of stratospheric HCl : 1975-1985, in "Our Changing Atmosphere", Crutzen, P.J., Gerard, J.-C. and Zander, R., Ed. University of Liège, 61-68, 1989.

Girard, A., J. Besson, D. Brard, J. Laurent, M.P. Lemaître, C. Lippens, C. Muller, J. Vercheval and M. Ackerman, Global results of grille spectrometer onboard Spacelab 1, Planet. Space Sci., 36, 291-299, 1988.

Lemaître, M.P., J. Laurent, J. Besson, A. Girard, C. Lippens, C. Muller, J. Vercheval and M. Ackerman, Sample performance of the grille spectrometer, Science, 225, 171-172, 1984.

Rinsland, C.P., J.S. Levine, A. Goldman, N.D. Sze, M.K.W. Ko and D.W. Johnson, Infrared measurements of HF and HCl total column abundances above Kitt Peak, 1977-1990 : Seasonal cycles, long-term increases, and comparisons with model calculations, J. Geophys. Res., 96, 15523-15540, 1991.

Wallace, L. and W. Livingston, Spectroscopic observations of atmospheric trace gases over Kitt Peak 3. Long-term trends of hydrogen chloride and hydrogen fluoride from 1978 to 1990, J. Geophys. Res., 96, 15513-15521, 1991.

Weisenstein, D.K., M.K.W. Ko and N. Sze, The chlorine budget of the present-day atmosphere : a modeling study, J. Geophys. Res., 97, 2547-2559, 1992.

W.M.O., World Meteorological Organization, Global Ozone Research and Monitoring Project Report no. 16, 1985.

Zander, R., M.R. Gunson, J.C. Foster, C.P. Rinsland and Z. Namkung, Stratospheric $ClONO_2$, HCl and HF concentration profiles derived from atmospheric trace molecule spectroscopy experiment Spacelab 3 observations : an update, J. Geophys. Res., 95, 20519-20525, 1990.

Zander, R., M.R. Gunson, C.B. Farmer, C.P. Rinsland, F.W. Irion and E. Mahieu, The 1985 chlorine and fluorine inventories in the stratosphere based on ATMOS observations at 30° North latitude, J. Atmos. Chem., 15, 171-186, 1992.

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