Stratospheric ozone concentration profiles from Spacelab–1 solar occultation infrared absorption spectra.

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ABSTRACT.

A Grille Spectrometer has been operated on board Spacelab–1 launched on the 28th of November 1983 for a 9–days mission. Solar occultation absorption spectra that show ozone specific absorption features have been taken in the infrared range between 1039.6 and 1081.3cm–1, with a spectral resolution of about 0.055 cm^{-1}. Northern as well as Southern hemisphere locations have been covered. The determination of ozone vertical concentration profiles from these spectra has required the development of an improved inversion program based on Mill's algorithm. The most important ameliorations are the more accurate treatment of the molecular line parameters and the introduction of Fourier filter techniques for minimizing the influence of noise that severely affects the ozone spectra. The resulting ozone vertical profiles, between about 25 and 65 km altitude, are discussed and compared with data taken at the same time and location by other instruments (e.g., SME).

In the future, these results will be compared with data taken with the same instrument during the ATLAS–1 mission and with a slightly adapted version of the spectrometer onboard the soviet space station MIR, in order to detect global changes.

1. INTRODUCTION.

The Grille Spectrometer is an infrared spectrometer with a scanning grating, an oscillating parabolic collimator and a grille mask in place of the conventional entrance slit. It has been designed and operated in a close collaboration between the French "Office National d'Etudes et de Recherches Aérospatiales" in Paris and the "Belgian Institute for Space Aeronomy" in Brussels.[1] Its purpose is to study the vertical structure of the earth's atmosphere composition between approximately 15 and 150 km altitude, which limits depend on the observation conditions and the atmospheric minor constituent of interest. The technique used is solar occultation absorption spectrometry, in the wavelength region between 2.5 and 10.5 μm, and subsequent inversion of the series of spectra obtained during one sunset or sunrise. Several balloon and aircraft flights have been performed in the past with initial versions of this instrument; a space–qualified version of the Grille Spectrometer has last been operated onboard the Space Shuttle, during the Spacelab–1 mission from November 28 to December 8, 1983. Descriptions of the instrument and of the scientific results of these previous missions have been published earlier;[2] only some characteristics of the spectrometer that are relevant to the understanding of the present paper and a brief summary of the results obtained previously will be repeated hereafter (Section 2.). The inversion algorithms used earlier for the determination of vertical concentration profiles were based either on the measurements of equivalent widths and an onion peeling algorithm, or on a quasi–automatic retrieval based on Mill's method.[3]

Ozone spectra recorded during that Spacelab–1 mission contain many absorption lines and moreover are contaminated by noise; therefore at first they seemed difficult to analyze by these same methods. This paper will dwell on the problems associated with these data and the modifications we applied to the inversion program in order to retrieve the O_3 vertical profiles.

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The comparison with simultaneous data from the Solar Mesosphere Explorer (SME), in Section 3.2.3. , will show that the program as it stands enables to retrieve O_3 vertical profiles up to about 60 km altitude from the Grille Spectrometer Spacelab–1 spectra.

2. INSTRUMENTATION, AND PREVIOUS SPACELAB–1 RESULTS.

The Grille Spectrometer is a 80 cm focal length infrared spectrometer equipped with a square plate grille as its entrance aperture and a 58 lines/mm grating. This grille consisting of alternating hyperbolic transparant and opaque zones lends the spectrometer an enhanced luminosity as compared to one that is equipped with a conventional entrance slit providing the same spectral resolution.[4] For the Spacelab–1 Grille spectrometer the spectral resolution varies between 1 x 10^4 and 2 x 10^4, depending on the wavelength region, and the luminosity enhancement factor is of the order of 40. The surface of the grille (15x15 mm^2) limits the field of view of the instrument to about 10' on the solar disk; the solar tracking points to the central portion of the disk. Two detectors are used to cover the spectral range from 2.5 to 10.5 µm: an InSb detector covering the range 2.5 to 5.5 µm and a HgCdTe detector covering the 2.5 to 10.5 µm region; both are cooled to liquid N_2 temperatures (77K). The Grille spectrometer operations use both detection channels simultaneously, in two different wavelength intervals corresponding to different diffraction orders of the grating.

An overview of the observations made by the Grille spectrometer during the Spacelab–1 mission is given in Table 1 of Ref. 2. Ten different trace species have been measured: vertical profiles have been retrieved and published for H_2O, CO, CO_2, CH_4, NO, NO_2,[2] and for HCl.[5] Telemetry problems during the mission disturbed most of the HF spectra. The analysis of the O_3 spectra required the inversion program to be improved: this will be discussed in the next sections, giving special attention to the filtering technique that has been developed to cope with the problems associated with these particular spectra.

3. OZONE DATA RETRIEVAL.

3.1. Ozone data.

Table 1 lists dates and locations of the occultations during which O_3 has been succesfully monitored; it includes the corresponding measurement spectral windows, which are acquired in diffraction order 3, and that are covered by the HgCdTe detector; in these windows, the resolving power is of the order of 18600. According to the orbital parameters prevailing during these measurements, the geographical displacement of the tangent points during one occultation (20 –> 100 km) is between 3.6° and 8° (combined latitude and longitude), as indicated in the table.

It has turned out that the signal/noise ratio in this second detection channel is more than a factor 10 lower than in the other one, in particular at the higher tangent point (TGP) altitudes (> 40 km). Two distinct interferences are distinguished in the high–altitude spectra. A first one shows up systematically as a low–frequency modulation and is associated with the spectral bandpass characteristics of the interference filter in front of the detector. The second one is of more random nature: it is present most often but not always if high incident light intensities are met; it appears to have a rather constant frequency of ~17 Hz, without showing up any mutual phase relationships among different spectra. Up to now, the origin of this second source of noise could not be identified unambiguously in the laboratory. A typical series of recorded O_3 spectra, in the 1060 cm^{-1} spectral window, is shown in Fig. 1 These spectra are recorded at a grating rotation speed of ~465"/s, corresponding to ~1.64 cm^{-1}/s in the spectral window considered. Hence in Fig. 1, the noise frequency of 17 Hz corresponds to a period of ~96.5 x 10^{-3} cm^{-1}, which can be distinguished visually, e.g., in the spectra at tangent heights between 73 and 60 km. Probably the interference is present also in, at least parts of, the lower altitude spectra, but here it is confounded with the many (>100) O_3 absorptions present. Analogous comments apply to both other O_3 spectral windows.

Moreover, single spurious points have been discovered in the raw measurements: they have been corrected manually by interpolation before starting the inversion process.

3.2. Vertical profile retrieval.

3.2.1. General characteristics of the inversion program.

The inversion program is based on the Mill's algorithm, as described earlier in Ref. 3. It has been rewritten into an improved and fully portable (Fortran + GKS) version. It has been designed in a modular way and calls for auxiliary input data files and conversion programs and preparatory routines.

In a first step, it calculates the synthetic spectra corresponding to the various observation geometries during the occultation under study, including optical refraction in the earth's atmosphere. The inversion program automatically calculates the observation geometry associated with each spectrum from the orbital data recorded continuously during the mission, including Spacelab attitude and altitude data, and sun ephemeris. The atmosphere is divided into spherical concentric layers based on the algorithm used in the AFGL FASCOD2 software.[6] Each layer is characterized by a constant pressure (molecular density), temperature, and initial concentrations of minor species; these values are taken as desired either from one of the available reference atmospheres, or from a user–defined atmosphere which may be real experimental values corresponding to the actual measurement conditions. The refraction routine was developed by us[3] and has about the same precision as the routine used in FASCOD2;[6,7] because ours is somewhat slower however, it will be replaced by the latter in the near future. Moreover, slight changes to the FASCOD routine have proven necessary for better compatibility with our inversion program.[7] The difference between the refraction coefficient n and 1 is taken proportional to the local atmospheric density, $qmol$ (molecules/cm^3), according to: $n-1 = 1.0752 x 10^{-23}\ qmol$.

The absorptions are calculated taking into consideration all presently available molecular line parameters as are they are published in the HITRAN86 database.[8] In particular, the temperature dependences of the linewidths and of the absorption intensities, including the temperature dependence of the rotational partition function, are taken into account. For O_3, apart from the linewidths and their temperature coefficients that have been taken from the HITRAN86 compilation, the most recently published lineparameters[9] are used. For the new O_3 lines, we have set the linewidth equal to the average over the neighbouring lines, and the temperature coefficient equal to 0.76. A Voigt line shape is assumed, approximated by the Whiting[3] profile, that is convolved subsequently with an empirically modeled instrument function of which the parameters may be optimized by the user. Since for the Grille spectrometer it is the instrument profile that governs the resulting width and shape of the O_3 absorption lines at the altitudes of interest, the inaccuracies of the Whiting approximation play a minor role in the retrieval accuracy; nevertheless it shall be replaced by a better approximation in the near future.

In a second step, calibration and/or baseline corrections, derived from a linear regression analysis, are applied to the calculated spectrum for improving its agreement with the corresponding experimental one. Then in an iterative procedure, the rms differences between corresponding experimental and synthetic spectra are minimized starting from below, i.e., considering the spectra in the order of increasing TGP heights, by adjusting the adopted concentrations of the absorbers by a constant factor in all layers above and including the one that contains the TGP. This procedure has been made more efficient than it was previously. It is worth mentioning that the user can decide himself which part of the spectral window and which molecules he wants to be incorporated in the fit, i.e., the program has an option for adjusting simultaneously the concentrations of several of the absorbing molecules.

Although the inversion program could be run quasi–automatically, the analysis process can be continuously monitored and controlled by the user through graphical displays of experimental and calculated spectra and of the retrieved profiles –including listings of calculated parameters–, already in the intermediate phases of the inversion process.

3.2.2. Peculiarities of the O_3 retrieval.

The time duration for scanning the O_3 spectral windows is of the order of two seconds, by which the tangent point height altitude may have changed by about 1.5 km (orbit at 250 km altitude). In comparison with the uncertainty on the TGP height due to the 10' field–of–view of the instrument, which is of the order of 5 km and which has not been taken into account up to now, this change with time of the line of sight is a minor effect: the effective line of sight associated with a spectrum is that corresponding to the timing of its central wavenumber.

The atmosphere model has been set as close as possible to the real conditions by integrating into it measured temperature and pressure data. The required experimental data are derived through data conversion and interpolation –linear as regards temperature, exponential as regards pressure– from the NMC database,[10] which contains temperature and geopotential height measurements at discrete pressure levels, interpolated to a fixed latitude – longitude grid (latitudinal resolution: 2.5°, longitudinal ~: 5°); we decided to determine the NMC data at the latitude and longitude of thatTGP of which the altitude approximately coincides with that of the maximum in the O_3 vertical profile (30–35 km). The lowest pressure level covered by the NMC database is 0.4 mbar, corresponding to about 55 km altitude. SME temperature[11] data are used in the higher altitude layers; however here, only monthly averaged, zonal mean values at a 5° latitudinal resolution are currently available. As regards the molecular densities in the present analysis, the ratios found between the NMC and the standard US '76 model[12] values at 50 km altitude have been kept over the higher altitude range for extrapolating the standard model densities. As such, no discontinuities are introduced. Some comments as to the effects of temperature uncertainties on the retrieved profiles are given below (Sec. 3.2.3.).

Of most concern is the influence of the noise components in the raw O_3 spectra on the retrieved profile. For each of them, a different solution has been developed.

(i) The slow modulation can rather easily be identified in the so–called out–of–atmosphere (OA) spectra. As it appears to have a constant phase over the whole occultation series, several OA spectra are averaged in order to obtain a clean reference spectrum to which all the other ones can be normalized. Comparing the residuals between experimental and synthetic spectra with and without incorporation of this normalization procedure gives evidence of the usefulness of this method. At the same time, this technique ensures that the thus normalized spectrum contains only telluric features.
(ii) As regards the 17 Hz interference, we noticed that it appears most perceptibly in the so–called residuals, i.e., the difference spectra between measurements and calculations. Therefore we adopted the following procedure: a first analysis of the spectra is made, as described above. Then, for each individual spectrum (situation) of an occultation series, a discrete fast Fourier transform[13] of its residual is performed, and the exact frequency position of the interfering noise around 17 Hz is estimated from the residual's power spectrum plot. Then for each situation of the occultation a numerical (digital) notch filter centered at the determined noise frequency is applied, both to the experimental and the synthetic spectrum, and the iteration sequence for minimizing the rms difference between both spectra is started over. Indeed, since the frequency of the spurious noise is located well inside the frequency span of the real absorption features, filtering tends to eliminate part of the latter. Identically eliminating them from the calculated spectrum allows us to compare two equivalent quantities in the iteration procedure. The power spectrum is calculated according to Eq. (12.7.10) in Ref. 14. Before passing to the Fast Fourier Transform routine, the data were padded by zeroes at both ends, and Parzen windowing was applied.[14] In order to have a smaller spectral variance per data point in the power spectrum, the technique of K overlapping segments (by one half of their length) has been applied, with K equal to 3.[14] The notch filter has been designed according to Ref. 15. The determination of the filter tolerance parameters is based on how the power spectrum looks like, and even more, on the number of available spectral data points limiting the number of filter coefficients that can be allowed. Before proceeding in the inversion procedure, a number of data points at both the beginning and the end of the spectrum are discarded in order to avoid edge effects: we chose this number to be equal to one third of the number of filter coefficient pairs.

We have chosen one specific event, namely, event 16 in the 1060 cm^{-1} window (see Table 1), as a test case for validating the correction and inversion procedures outlined above; Fig. 2 illustrates the situation at a TGP height of 41 km. In Fig. 2 (a), both normalization and filtering were applied before starting the inversion procedure, whereas none of them was in Fig. 2 (b): the slow modulation and high (~17 Hz) frequency component that are clearly apparent in the residual in the latter case are almost absent in the former one. In going from (b) to (a), the overall rms error over the spectrum has decreased from about 3.8% to about 2.4%. Fig. 3 illustrates, for the same situation of event 16, the power spectra corresponding to the cases of Fig. 2 (a) (full line) and (b) (dashed line), resp; the x–axis units (ν_N) are normalized frequency units, relative to the Nyquist frequency (which is equal to one half the spectrum sample frequency). The location where the spectral power has seriously decreased ($\nu_N = .21$) corresponds to the ~17 Hz frequency and was chosen as the notch filter's central position. Some spectral power decrease is also observed at the very low frequency end, corresponding to the elimination of the slow modulation by the normalization procedure. These interpretations have been confirmed by applying the normalization and filtering separately.

3.2.3. Spacelab–1 O_3 profiles: discussion.

The most important results of the correction procedure (normalization + filtering) outlined in this paper are to establish that the Spacelab–1 data permit O_3 profiles to be retrieved with good confidence, contrary to our initial beliefs, and to permit the association of a realistic error bar with the inverted results. The highest altitude up to which the profile can be retrieved from our dataset is about 60 to 65 km.

To demonstrate this, Fig. 4 shows the O_3 profile retrieved from the event 16 spectra discussed above (1060 cm^{-1} –window) in comparison with, on the one hand, the MAP[16] model O_3 profiles between 20° N and 30° N for the month of December and the US'76 standard atmosphere model[12] profile, and, on the other hand, the profile measured by SME[17] on the same day (Dec 3, 1983) at 25° N (latitude) and 71° E (longitude); no SME data are available at the longitude of the Grille measurement, but the SME dataset shows that the longitudinal variation is less important. The SME profile drawn in Fig. 4 combines the data from the UV and the near–infrared (airglow) spectrometers; also included are the 15% error bars that hold for them. In the overlapping altitude region, the Grille spectrometer result is in very good agreement with the SME data. At the lower altitudes (< 50 km), no simultaneously measured data are known to us. In comparison with the model data (MAP and US '76 standard[12]), discrepancies of the order of 10–20% are observed. They might be associated with the differences in temperature we observe between the models (US '76 standard[12] and MAP[18] (December)) temperature profiles and the one recorded in the NMC and SME databases for the particular time and location of the Grille measurement in question, as shown in Fig. 5. For this purpose, as has been discussed above (Sec. 3.2.2.), the Grille event has been fixed at 24° N, 155° E, Dec. 3, 1983; the SME temperature data are the monthly average for December, at 25° N.[19] Tests have indicated that temperature uncertainties of the order of 5K may induce changes in the retrieved O_3 profile of the order of up to 10%.

This study is being continued, together with the retrieval of all available Grille Spacelab–1 O_3 profiles (cf. Table 1) and a more profound examination of the error bars pertinent to them. Also the inversion program is being improved constantly; some of the modifications envisaged have already been mentioned above.

4. CONCLUSIONS AND PROSPECTS FOR THE FUTURE.

It has been demonstrated in Sec. 3. that the Grille spectrometer Spacelab–1 spectra provide valuable measurements of the O_3 vertical distribution in the earth atmosphere, between about 25 and 65 km, in November–December 1983; they constitute a dataset that is in part comparable, in part complimentary to the SME O_3 data.[17] The data will be further analysed and interpreted, taking into account the simultaneously measured distributions of other trace species (e.g., Ref. 2) intervening in the O_3 photochemistry. In addition, the next flight of the Grille spectrometer onboard the Space Shuttle during the ATLAS–1 mission,[20] will allow some study of the time evolution of these species, including O_3, on a global scale. An ameliorated version of the Grille spectrometer is scheduled for 1995 to fly on the soviet space station MIR for a nominal duration of one year, with specific attention to the monitoring of O_3. In particular, the use of coolers of higher performance will help optimizing the detection signal/noise ratios.

5. ACKNOWLEDGEMENTS.

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Table 1. Specifications of Grille Spectrometer successful O_3 observations (Spacelab–1), limited to the data below 70 km altitude. SR: sunrise; SS: sunset; the spectral windows are identified by about their central wavenumber, as follows: 1080 cm^{-1} stands for 1077.2 –1081.4 cm^{-1} · 1060 cm^{-1} for 1058.8–1062.0 cm^{-1}, and 1040 cm^{-1} for 1039.7–1043.2 cm^{-1}.

Event	date/ GMT (hrs)	SR/SS	TGP latitude N	TGP longitude E	TGP altitudes (< 70 km)	spectral window
17	337/ 7: –> 8:	SR	–67.6° –> –68.1°	236.7° –> 252.3°	11 km –> 60 km	1080 cm^{-1}
20	337/ 10: –> 11:	SR	–67.6° –> –68.1°	191.8° –> 204.1°	22 km –> 60 km	1080 cm^{-1}
22	337/ 11: –> 12:	SR	–67.4° –> –67.9°	167.3° –> 174.6°	15 km –> 46 km	1080 cm^{-1}
21	337/ 11: –> 12:	SS	25.9° –> 21.1°	87.6° –> 89.6 °	61 km –> 20 km	1040 cm^{-1}
16	337/ 6: –> 7:	SS	27.8° –> 24.7°	153.7° –> 155.1°	69 km –> 35 km	1060 cm^{-1}
16	337/ 6: –> 7:	SS	24.6° –> 23.6°	155.2° –> 155.5°	34 km –> 20 km	1080 cm^{-1}
13	336/ 12: –> 13:	SS	33.7° –> 30.6°	58.9° –> 60.5°	61 km –> 19 km	1040 cm^{-1}
3	333/ 23: –> 24:	SS	42.6° –> 44.5°	250.1° –> 248.8°	69 km –> 17 km	1060 cm^{-1}

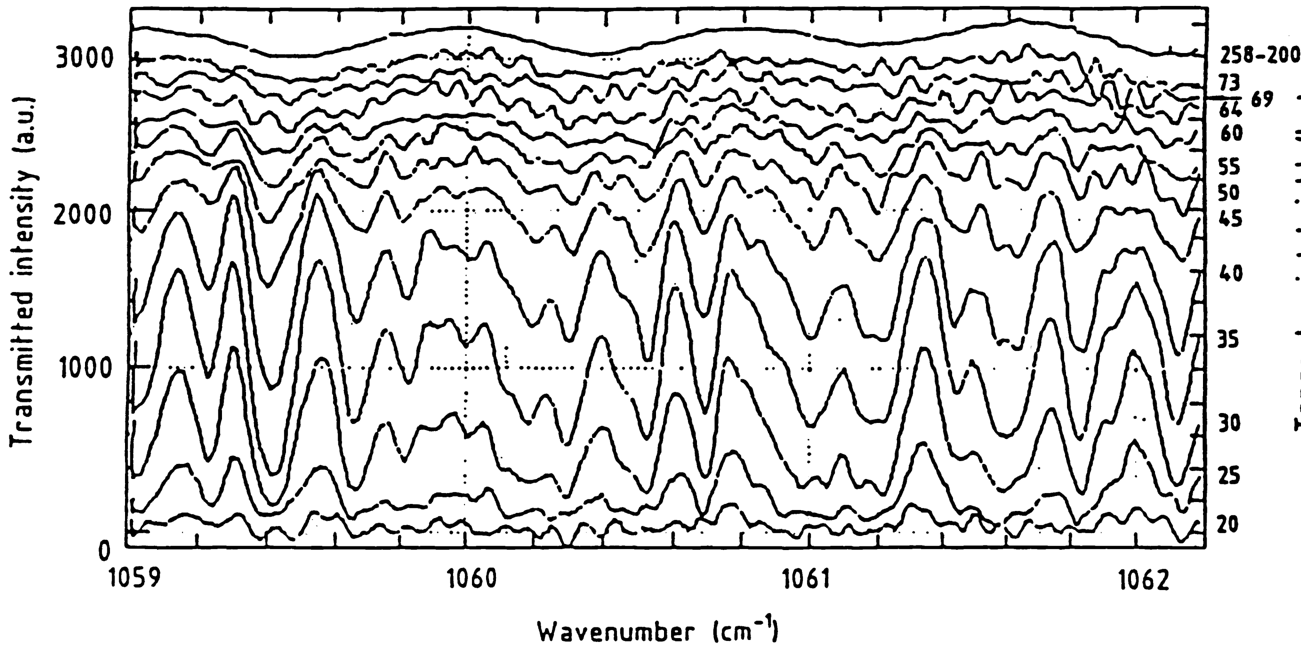

Fig. 1. Typical O_3 occultation series (event 3). The vertical scale holds for the lowest spectrum; each subsequent spectrum is shifted upwards by 100 arbitray units (a.u.). On the right hand side the tangent point (TGP) altitudes corresponding with each spectrum are indicated. The top spectrum is an average over 36 out–of–atmosphere spectra, at TGP altitudes between 200 and 258 km.

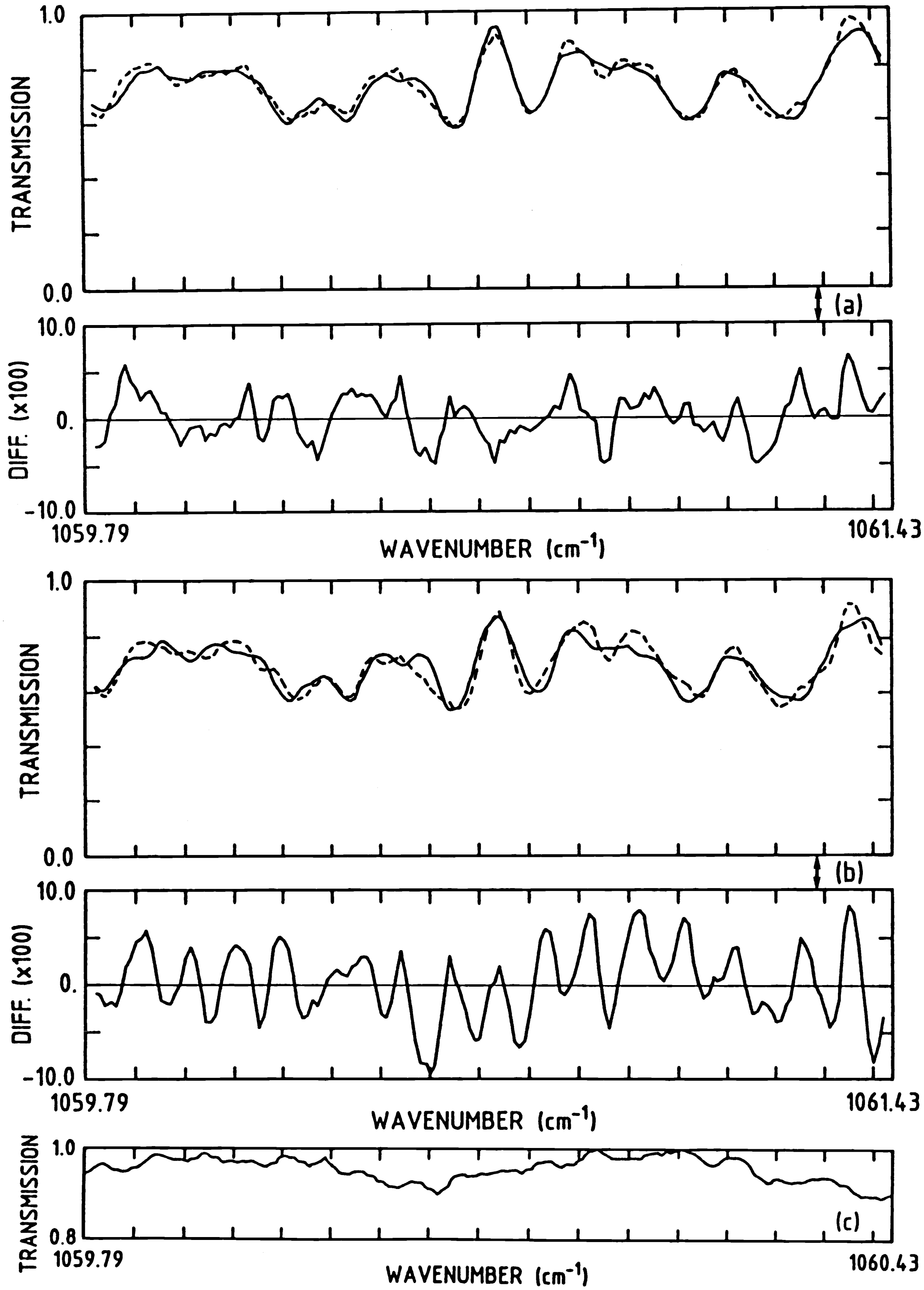

Fig. 2. Example of fit (event 16, TGP at 41 km), based on the present inversion program, with (a) and without (b) application of the correction procedure, outlined in Sec. 3.2.2. (c) shows the used reference out–of–atmosphere spectrum. – – – measured spectrum, —— calculated spectrum, lower curve: difference (residual) between measured and calculated spectrum.

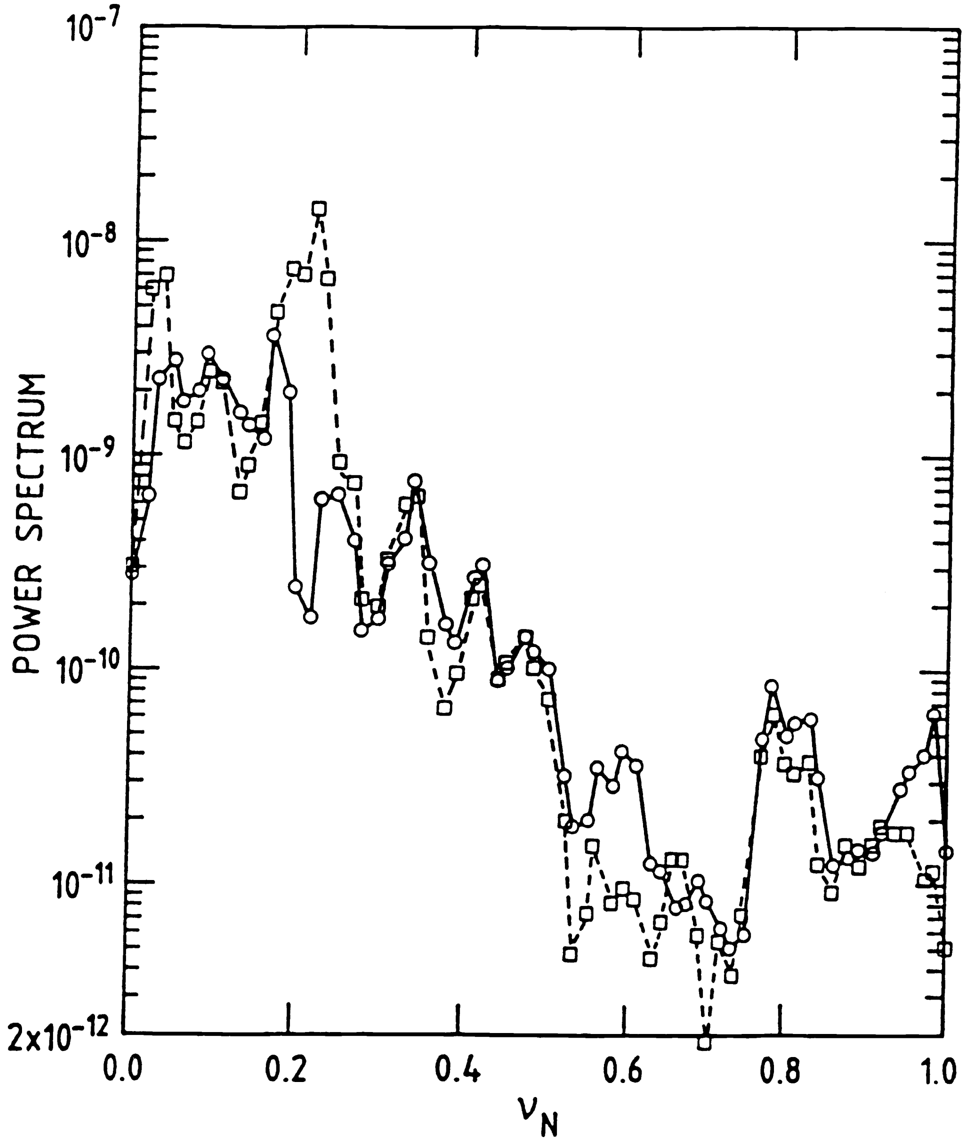

Fig. 3. Typical power spectral distribution of the residual spectrum, corresponding to the situations in Fig. 2 (a) ——— and (b) – – – – . $\nu_N = f/f_N$, f_N being the Nyquist frequency. The filter parameters used are: pass–to–stop and stop–to–pass transition locations: $\nu_N = .19$ and .23, resp, pass– and stopband tolerance: 25 dB, transition bandwidth: 0.01 .

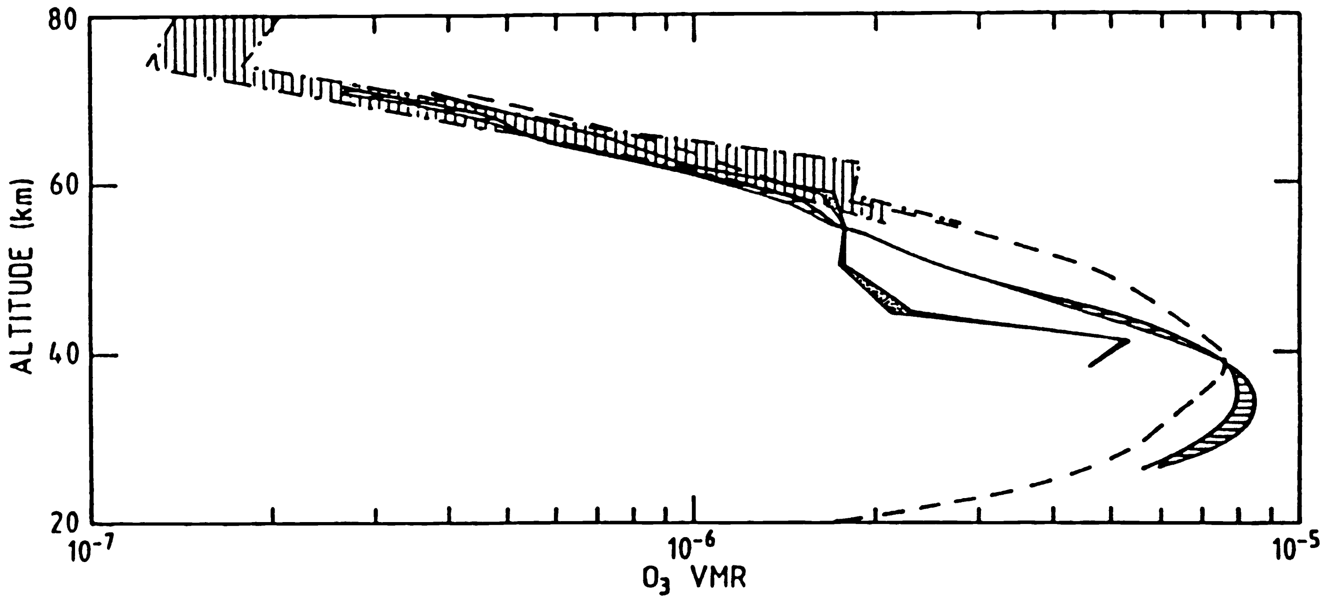

Fig. 4. Comparison between O_3 profiles, according to: — — — US'76 standard atmosphere model, ▤ MAP model (20° N–30° N, December) , ▥ SME (UV + near–infrared) measurements (25° N; Dec 3, 1983), ▩ Grille spectrometer data (event 16).

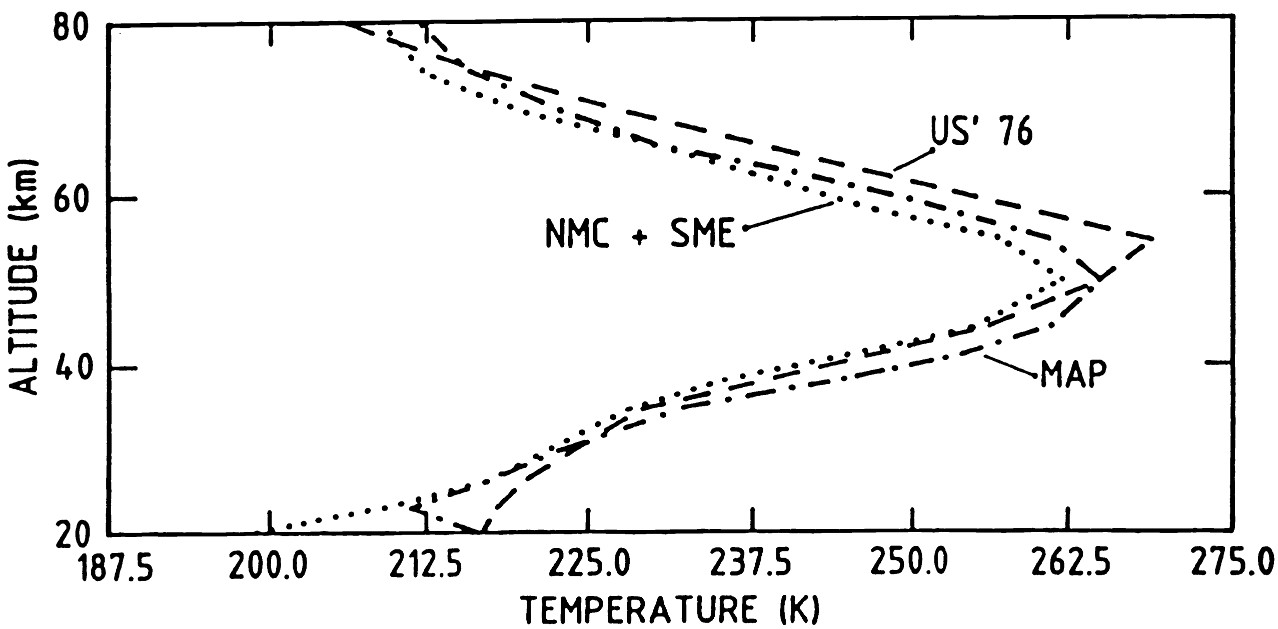

Fig. 5: Comparison between temperature profiles: .... measured data (NMC+SME) pertinent to the Grille data (event 16). – – –US'76 standard atmosphere, – . – . –MAP (December) model.