MULTIPLE OZONE MEASUREMENTS AT THE JUNGFRAUJOCH: AN UPDATE AFTER OPTIMISED RETRIEVAL FROM FTIR MEASUREMENTS.

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INTRODUCTION

The permanent co-location of several instruments at the International Scientific Station of the Jungfraujoch (ISSJ) enables the intercomparison of measurements over a period spanning several seasons, eliminating some time-specific biases. This paper addresses the validation of ozone measurements made at ISSJ by a zenith-sky SAOZ spectrometer on the one hand, and two Fourier-Transform spectrometers (FTS) operating in the infrared (IR) on the other hand. Previous analyses of the FTIR data led to O_3 values systematically lower than the SAOZ results by more than 10%. The actual re-analyses, based on a careful selection of the O_3 absorption lines in the FTIR spectra for the inversion, demonstrate that the systematic offset is smaller than 5%. In order to fully understand the remaining discrepancies, some other issues such as spatial overlap of the intercompared data, adopted model parameters, etc, must be considered. Taking into account that the SAOZ data are recorded automatically at each twilight opportunity, independent of common meteorological parameters, whereas the FTSs can be operated only manually and need clear-sky conditions, the final objective is the exploitation of all available O_3 measurements for an improved assessment of the O_3 climatology at the Jungfraujoch and the evaluation of its long-term trends.

INSTRUMENTATION AND MEASUREMENT TECHNIQUES

The International Scientific Station of the Jungfraujoch (ISSJ) located in the Swiss Alps (altit. 3580 m; latit. 46.55N; longit. 7.98E) forms the European alpine station within NDSC (Network for Detection of Stratospheric Changes), together with Observatoire de Haute Provence and Plateau de Bure. It also plays a key role in the European ESMOS/Alps and SCUVS networks, and has participated to the EASOE and SESAME campaigns.

Since mid-1990, a SAOZ (Système d'Analyse par Observations Zénithales) has been operated quasi-permanently at this site, providing continuous measurements of NO_2 and O_3 column amounts at morning and evening twilight. The O_3 data used here are extracted from the zenith-sky observations in the 470-550 nm spectral range, taken at solar zenith angles (SZA) between 87 and 91 degrees which is the most reliable range when considering the signal-to-noise ratio and the airmass factor uncertainties (Sarkissian et al, 1995). The spectral analysis relies on the DOAS (differential optical absorption) technique, in this case using the SAOZ least-squares fit algorithm (Service d'Aéronomie, CNRS).

FTIR O_3 data from ISSJ are available since 1984. The high-resolution Fourier Transform Spectrometers (FTS) actually used are a Bruker 120HR and a home-made high-resolution instrument, both operated in direct sun, solar absorption mode in the mid-infrared range (2 - 15 µm; spectral resolution of order 0.002 cm^{-1}; InSb and HgCdTe detectors). Measurements can be taken throughout the whole day, provided the sky is clear; for the present work they are limited to SZA values lower than 86 degrees. The spectra-inversion algorithms used for the actual data-analysis are SFIT, developed by C.P. Rinsland et al., and SFSP, developed at BISA. Both rely on a least-squares minimization technique and have been validated in the so-called "ESMOSII/NDSC IR algorithms intercomparison exercise" in 1993/1994 (Zander et al., 1993).

Some direct-sun Dobson data from the nearby Arosa station (altit. 1850m; latit. 46.5N; longit. 9.5E) have been included in the intercomparison. Likewise as for the FTIR data, they are taken under clear-sky conditions, preferably around noon-time; anyway the SZA at observation time must not exceed 70°.

The intercomparison has been limited to the 1993-1995 timeframe: the perturbed post-Pinatubo period has been excluded because at this moment no general consensus exists as to the optimal airmass factors to be applied to the SAOZ measurements. It has been assessed generally that the aerosol perturbations are negligible from early 1993 on.

ANALYSIS METHODS AND DISCUSSION

Ground-based FTIR spectra offer many possible spectral lines for the study of O_3, of which the spectroscopic characteristics are not all known with the same accuracy. Previous laboratory work (Barbe et al, 1993) has identified the least T-dependent lines in the bands $3\nu_3$ (~3040 cm^{-1}), $\nu_1+\nu_2+\nu_3$ (~2850 cm^{-1}), and $\nu_1+\nu_3$ (~ 2150 cm^{-1}), taking into account the combined effect of the lower energy level of the transition and the T-dependence of the Lorentz halfwidth. Progress has been made recently as to the selection of the most suitable lines, by several approaches presented hereafter.

The analysis of the IR spectra from both FTS instruments at ISSJ in the selected period (1993-1995) has been performed in 4 different spectral windows: (i) 1128.50 - 1129.65 cm^{-1}, (ii) 2084.00 - 2085.28 cm^{-1}, (iii) 2112.75 - 2112.86 cm^{-1}, and (iv) 3039.18 - 3040.05 cm^{-1}. The resulting daily average O_3 columns have been compared among each other and also with the data series (mean of sunrise and sunset values) obtained by the SAOZ instrument (Figs. 1a and 1b).

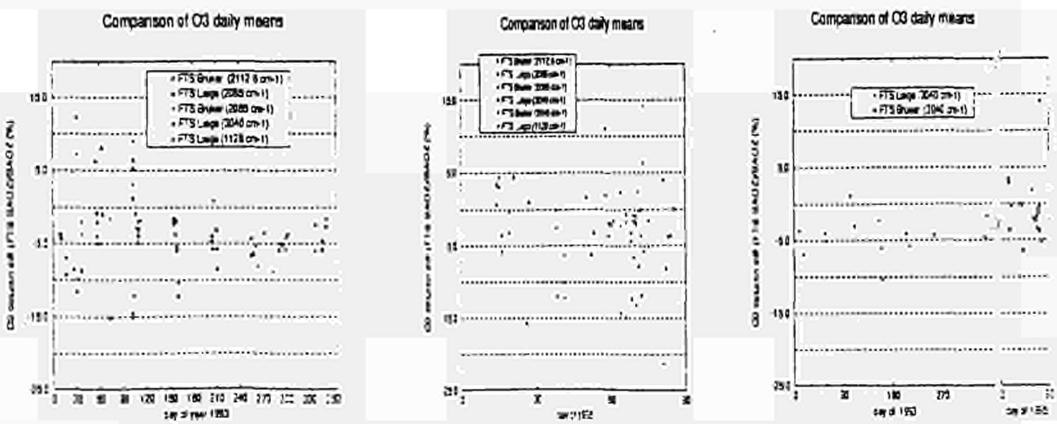

Fig. 1 Relative differences between SAOZ and FTIR O_3 results at ISSJ: (a) and (b) for all µwindows, (c) restricted to the best (3040 cm^{-1}) one

One observes that the degree of random variability of the results depends on the choice of the μwindow, being largest for the third (2112.75 - 2112.86 cm^{-1}) μwindow. The latter choice also gives rise to a more significant negative offset with respect to the SAOZ data. Increasing the extent of this window leads to better stability among the individual results throughout one day. The most stable results are obtained using the fourth μwindow (3039.18 - 3040.05 cm^{-1}) in the $3\nu_3$ band. Among the reasons for this is the more appropriate linestrength as compared to the $\nu_1+\nu_2+\nu_3$ band in which absorptions are too strong, and the $\nu_1+\nu_3$ band in which they are too weak. Therefore in Fig. 1c the intercomparison has been limited to this last window.

Although a small negative systematic offset not exceeding 5% may still exist, with the FTIR O_3 values being lower than the SAOZ ones, the agreement between FTIR and SAOZ data comes very close to the one obtained in the most recent intercomparison among various UV/Vis spectrometers including SAOZ (Vaughan et al, 1995), of order 5% also.

A somewhat larger random dispersion among the data is observed in the first trimester of 1993, in which large day-to-day ozone variations occurred. Dynamical effects are suspected to be the cause, and therefore we compared the ISSJ data to O_3 measurements from the Dobson instrument in Arosa; the relative positions of both stations are shown in Fig. 2. On short-term timescales the FTIR data appear to be in closer agreement with the Dobson data than with the local SAOZ data.

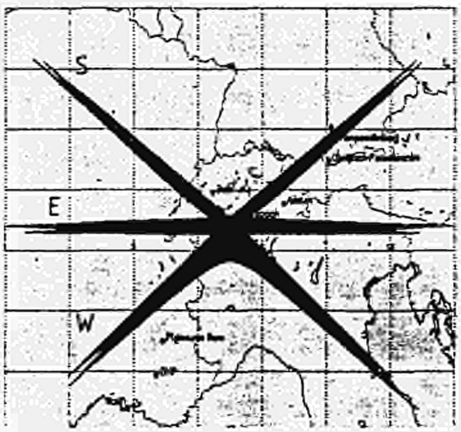

Fig. 2 Location of the Jungfraujoch Observatory, indicating the spatial extent of SAOZ (grey) and FTIR (black) ozone measurements (see text) at the equinoxes (E) and summer (S) and winter (W) solstices.

A first impact of the dynamics is explained with the help of Fig.2, in which the geographical regions for sampling of the O_3 columns by the different techniques are shown: the line one can imagine connecting the observatory with the border of one of the shaded areas is the projection on the earth's surface of the line-of-sight (LOS) at the azimuth corresponding to the local time of the measurement, and with a length representative of the distance along the LOS that integrates 90% of the total observed ozone amount. The upper and lower shaded areas are representative of summer and winter solstices resp., the middle one of equinox conditions. The spatial overlap between SAOZ and FTS sampled regions is high if data are taken at about the same time (same SZA), but can be very different if one is comparing SAOZ data taken at twilight with FTIR data taken at about noon. In such circumstances, better spatial overlap is possibly obtained with the airmass sampled by the Dobson instrument at Arosa, although it is located some 180 km away from ISSJ. This effect should be confirmed by inspection of the individual measurements from both FTIR and Dobson instruments.

A second impact concerns the atmospheric model used in the data retrieval: up to now, the SAOZ analysis uses one single standard model for deriving its airmass factor for all measurements; the FTIR analyses have used several distinct models for the (p,T) profiles. The actual data for 1993 clearly reveal a correlation between the tropopause pressure and the O_3 column determined by SAOZ, especially as to the short-term fluctuations: the degree of linear correlation r increases from .49 to .58 after removal of slow (seasonal) variations. A similar analysis comparing Dobson data and tropopause pressure (assuming the ISSJ tropopause data are representative of Arosa as well), shows an even stronger correlation: $r = .64$. Thus especially in the case of a dynamically perturbed period, it should be verified to what extent the adopted models are representative of the true atmospheric conditions. This study will be pursued.

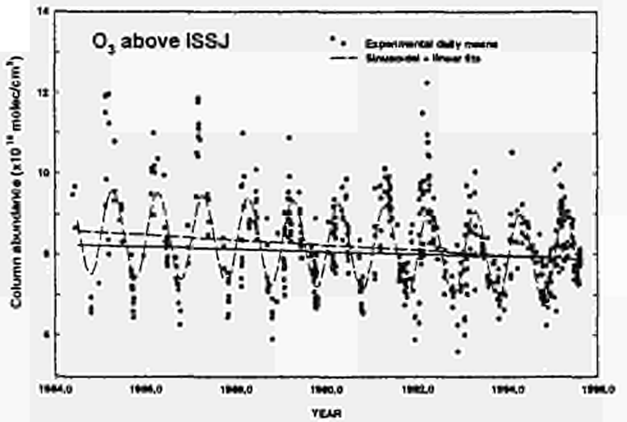

Fig. 3 Ozone long-term trend observed at ISSJ over the last 10 years; superimposed on it is a sinusoidal seasonal variation. For different symbols: see text

The above results have led us to estimate the long-term O_3 trend based on FTIR data from the spectral windows cited above, except the 2112.8 cm^{-1} μwindow. The fit (Fig. 3) superimposes a sinusoid for representing the seasonal variation, and a linear trend. The trend has been derived over 10 years, (i) using data all-year round, and (ii) excluding the first trimester of each year (data points represented by open squares), in which larger O_3 variations occur; the 1984-1995 time-span has been covered, the reference value taken at the beginning of 1990. The values obtained for the sinusoidal variation are quite similar in both cases: 22.3% pp in case (ii) as compared to 25.6% pp in case (i). However the derived rate of O_3 decrease changes from (-0.76 ±0.12) %/year with the 1990 reference value equal to 8.23E18 mol/cm^2 in case (i) (dashed line), to (-0.38±0.11) %/year in (ii) with the 1990 value equal to 8.06E18 mol/cm^2 (solid line). The difference between both trend-values is indicative of the fact that the maxima in the seasonal variation were more pronounced in the first 5 years of the considered period. These results are close to the ones published in WMO (Harris et al, 1995) under similar conditions. It should be noted that these trends include the perturbed post-Pinatubo period in which O_3 values were particularly low.

CONCLUSIONS

The analysis of O_3 measurements carried out at the International Scientific Station of the Jungfraujoch together with spectroscopic arguments have led to the identification of the (3039.18 - 3040.05) cm-1 range in the ozone $3\nu_3$-band as an optimal spectral μwindow for inversion of O_3 data from high-resolution FTIR ground-based spectra. As such, the overall agreement between FTIR and SAOZ O_3 data at ISSJ is in the -5% to +5% range, close to the mutual agreement obtained between UV/Visible spectrometers, SAOZ, Dobson and Brewer instruments in the latest campaign held in Camborne in 1994 (Vaughan et al, 1995).

It has been pointed out how dynamical perturbations and associated atmospheric parameters may influence the data retrieved from various instruments to different extent; this work is still in progress.

The results obtained here are important for ongoing studies regarding the O_3 climatology and long-term trends. Actually, we estimate an O_3 decrease above ISSJ at a rate of (- 0.38 ± 0.11) % per year over the 1984-1995 time period, when excluding the first trimester of each year.

The all-year round downward trend over the same period is found to be (-0.76±0.12) %/year. More refined evaluations will be made in a near future, including SAOZ and other additional data.

ACKNOWLEDGEMENTS

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