Towards improved evaluations of total ozone at the Jungfraujoch, using vertical profile estimations based on auxiliary data.

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Abstract The currently available database of total ozone amounts observed at the Jungfraujoch site in the Swiss Alps starts in 1984, based on high-resolution infrared solar absorption spectroscopic measurements, and has been complemented by daily SAOZ data since mid-1990. The latter instrument (Système d'Analyse par Observations Zénithales) measures the ozone column by application of the DOAS (Differential Optical Absorption Spectroscopy) method to zenith-sky scattered-light spectra in the ultraviolet-visible range, taken at twilight. The actual intercomparison for the overlapping period reveals a negative systematic offset of the FTIR with respect to the SAOZ data of 3.8% on average, but showing a seasonal variation in the difference. Part of the observed difference probably originates in the model atmospheres that influence the retrieved column differently for each technique. This work shows the development of a climatological ozone vertical distribution model that is more representative of the real atmosphere, exploiting daily meteorological data that are commonly available at the Jungfraujoch site. It argues that the use of this climatological model may improve the accuracy of the retrieved total columns, and hence the agreement between the SAOZ and FTIR datasets; some possible improvements to the model are suggested. Application of this concept to a re-analysis of past observations, making use of existing meteorological long-term records, will enhance the validity of the ozone database at the Jungfraujoch.

Introduction

The International Scientific Station of the Jungfraujoch (ISSJ), located at 3580 m altitude in the Swiss Alps, at 46.55°N and 7.98°E, is one of the alpine sites that constitute the primary NDSC (Network for Detection of Stratospheric Changes) station for the northern mid-latitude hemisphere. The NDSC station is equipped with various instruments for atmospheric measurements, most of them validated in this framework, and complementary ones mostly integrated in Swiss and/or international networks; here we will focus the experiments dealing with ozone. Among the former category are the SAOZ (Système d'Analyse par Observations Zénithales) and Fourier Transform Infrared (FTIR) spectrometers located at ISSJ, and the O_3 microwave (μwave) radiometer located at Bern; among the latter are the in-situ O_3 measurements based on a UV (254 nm) absorption measurement that are taken at ISSJ as part of the NABEL network (EMPA, 1994), the O_3 soundings performed periodically at Payerne (46.5°N, 6.6°E) (Staehelin *et al.*, 1991 and 1992), close to Bern, and the Dobson instruments at Arosa (46.8°N, 9.7°E) (Staehelin *et al.*, 1995), some 130 km to the NE of ISSJ. An essential contribution to datavalidation can be acquired through intercomparison of datasets obtained from different techniques at the same or nearby sites or from correlative observations, as has been proven lately at various occasions for several instruments/techniques, often in the frame of NDSC and European collaborations (EC contracts/projects as, e.g., SESAME). FTIR total ozone column data have been compared recently at some locations with data from collocated Dobson spectrometers (David *et al.*, 1993, Rinsland *et al.*, 1996). At ISSJ, a preliminary intercomparison between limited FTIR and SAOZ O_3 datasets has been discussed by De Mazière *et al.*, 1996: apart from a systematic negative offset of the order of 3 to 5% between the FTIR and SAOZ data, random differences were observed for which explanations have been proposed in terms of geophysical and/or algorithm-related parameters (dynamics, geometry, O_3 vertical distribution and spectroscopic parameters,...). The intercomparison presented in this work has been expanded and updated, and a systematic seasonal variation in the mutual difference appears: we attribute it to the different treatments of the O_3 vertical distribution in the SAOZ and FTIR analyses. Therefore to eliminate this seasonal dependence and thereby improving the mutual agreement among both datasets, we propose to introduce in the column retrieval procedures an ozone vertical profile climatology that has been established as a function of tropopause altitude. Here, a first result as to FTIR retrievals is shown; its application to SAOZ air-mass factor (AMF) evaluations will be tested readily. The idea is based on the observed and well-known correlation between ozone total amount and tropopause pressure (Meetham, 1937; De Mazière *et al.*, 1996) and the vertical motions associated with it (Reed, 1950). The approach is presented and some preliminary results are discussed.

Instrumentation, analysis and datasets intercomparison

Some experimental details about the SAOZ and FTIR spectral data are summarised in Table I. The SAOZ

spectral window	instrument /spectral resolution	fitted signatures
1127.50 - 1128.10 cm^{-1}	FTS1: 6.1 mk B: 4.0 or 6. 1 mk	O_3, N_2O
1128.50 - 1129.65 cm^{-1}	FTS1: 6.1 mk B: 4.0 or 6. 1 mk	O_3, N_2O
2084.00 - 2085.28 cm^{-1}	FTS1: 5.0 mk B: 2.9 mk	O_3, CO, CO_2, solar lines
3039.18 - 3040.05 cm^{-1}	FTS1: 4.9 mk B: 2.9, 4.0 or 5.0 mk	O_3, CH_4
470 - 540 nm	SAOZ: 0.6 nm	O_3, NO_2, H_2O, O_4, ring-effect

Table I. Specifications of spectra from which daily mean O_3 vertical columns have been retrieved. B and FTS1 = Bruker and home-made FTS, resp.; FTIR resolution = 1/2L, with L the maximum optical pathlength.

observations of the O_3 total column are made daily at sunrise and sunset, according to the DOAS technique for zenith-sky observations in the UV/visible spectral range; unreliable data due to tropospheric pollution events or enhanced multiple scattering have been removed (Van Roozendael *et al.*, 1994, and references therein). The reported data are morning (am) and evening (pm) mean vertical columns based on retrieved slant columns for solar zenith angles (SZA) between 87° and 91°; the daily mean is the average of am and pm values. The retrieval is done with the common SAOZ algorithm (Goutail *et al.*, 1994), apart from the conversion from slant to vertical columns for which seasonally varying instead of standard AMFs have been used, as explained in Lambert *et al.*, 1996. The FTIR O_3 data have been retrieved from direct-sun absorption spectra in various spectral windows by two different Fourier transform spectrometers (FTS) that are of equal data-validity: a commercial Bruker IFS HR-120 spectrometer (B) and a home-made FTS, identified here as FTS1 (see Table I). Spectra are recorded throughout the day, under clear-sky conditions; the actual daily mean O_3 columns are derived from individual retrievals from spectra limited to 15° to 75° SZA. We use the SFIT 1.09c non-linear least-squares fitting programme (C.P. Rinsland, LaRC), with the SFIT standard O_3 vertical profile but daily NCEP (National Center for Environmental Prediction) pressure-temperature fields, and the HITRAN-92 spectroscopy database (Rothman *et al.*, 1992). No significant systematic biases (< 1%) have been found among the columns retrieved in different FTIR windows.

The intercomparison of both datasets is shown in Figure 1. We know that in the period between mid-1991 and end-of-1992, the SAOZ O_3 observations were perturbed by the heavy aerosol load in the stratosphere: the disagreement between both datasets increases to ≥ +45%, and this period will be omitted from any further discussion. It appears that the FTIR results are systematically lower than the SAOZ results, by about 3.8%, and that there is a seasonal variation in the difference of about 7.4% peak-to-peak amplitude, if modelled as a sine; random differences

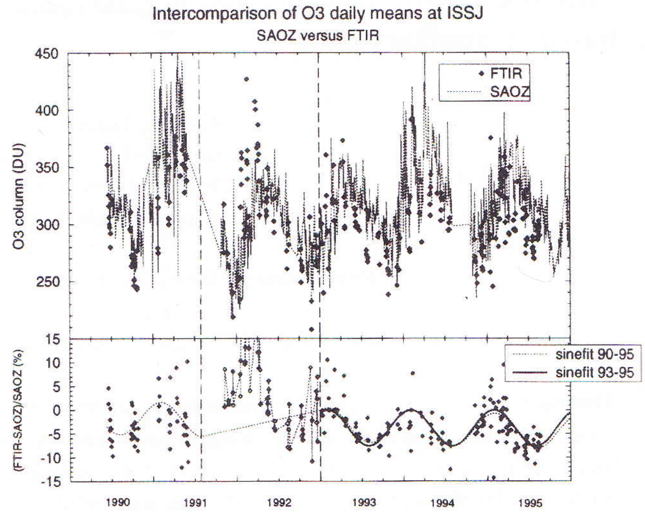

Fig. 1 Intercomparison of SAOZ and FTIR daily mean O_3 vertical columns for the common period of operation at ISSJ. Lower plot: 'sinefit 93-95' is a pure sinefit covering 1993 to 1995; 'sinefit 90-95' superposes the sine on a linear trend covering the whole period, except for the mid-1991 to end-of-1992 period.

are more important in winter-spring, probably due to the more variable dynamics in that period of the year. We believe that the seasonal variation originates in the FTIR analysis, for the following reasons: (i) including nearby Dobson data in the intercomparison reveals no more seasonal variation in the Dobson to SAOZ differences, thanks to the use of the seasonally varying AMF as a parametrisation of variations in the O_3 vertical profile (Lambert *et al.*, 1996), and (ii), one single O_3 vertical profile is used in the actual FTIR analysis the choice of which is known to have a non-negligible impact on the retrieved total column, as demonstrated also hereafter.

Ozone vertical profile climatology at ISSJ

Development/validation

An O_3 vertical profile climatology has been established as a function of tropopause height or pressure, from averages over 10 years (1985-1995) of O_3 soundings at Uccle (50.8°N, 4.4°E). Nine classes have been distinguished corresponding to tropopause-altitudes ranging from (7±0.5) km to (15± 0.5) km altitude. The most frequently occurring tropopause altitudes at ISSJ are between 9 and 12 km (cf. Figure 3, upper plot): the corresponding climatological profiles are shown in Figure 2. This climatology has been verified mainly against microwave O_3 profiles at Bern, most reliable above 15 km; for the lower part, some comparisons with O_3 sonde profiles at Payerne, up to 30 to 35 km, have been done (Staehelin *et al.*, 1991 and 1992); hereto, the experimental data have been averaged according to the same classes of tropopause altitudes. Fig. 2 summarises the conclusions of the former verification. Except for the highest and lowest values of tropopause height that occur only sporadically, the experimental data and the climatological model agree as to the relationship between the tropopause height and the altitude at which the ozone concentration peaks, indicated

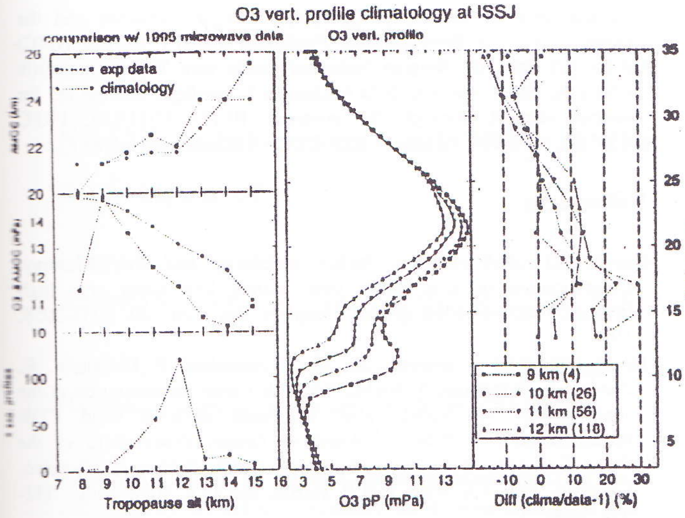

Fig. 2 Characteristics of the climatological ozone vertical profile model ('climatology') in comparison with O_3 μwave observations at ISSJ ('exp. data'). Left: Altitude of maximum O_3 concentration (AMOC), O_3 partial pressure (pP) at AMOC, and number of profiles per experimental dataset, as a function of tropopause altitude. Right: model vertical profiles and their percentage altitude-dependent relative differences ('clima/data-1') with respect to the μwave observations in the overlapping altitude range, for the most common tropopause altitudes (number of exp. data in brackets).

hereafter as AMOC for altitude of maximum ozone concentration: the AMOC rises with tropopause altitude. Concurrently, the value of the ozone partial pressure (pP) at AMOC decreases, which is associated with the total ozone content decrease as the tropopause is rising (London, 1985). The systematic positive offset of the climatological model based on Uccle soundings with respect to the microwave data obtained at ISSJ is in qualitative agreement with the ozone amount increase with latitude. Fig. 2 also illustrates that a better agreement between the climatological model and the experimental data is obtained in the altitude range between 18 and 28 km, which contains the ozone maximum, and that the upper and lowest parts (not shown) are less dependent on tropopause height, as is excepted from dynamical considerations (Meetham, 1937; London, 1985).

Results as to total O_3 climatology and FTIR retrievals

In Fig. 3, second plot, we contrast the climatology of the ozone total column amount observed by SAOZ with the one calculated from the above O_3 vertical profile climatology. Hereto, the climatological vertical profiles have been extrapolated above their upper altitude up to 90 km according to the standard SFIT ozone profile, without any further scaling; daily tropopause pressures are used for determining the corresponding column amounts from which then a monthly average has been calculated. For illustration, the upper and third plots show the ISSJ tropopause pressure climatology and its daily residuals (actual minus climatological pressure), respectively. It appears that the calculated seasonal variation amplitude is too small and is out-of-phase with respect to the observed one. One should keep in mind that (i) the stratospheric part

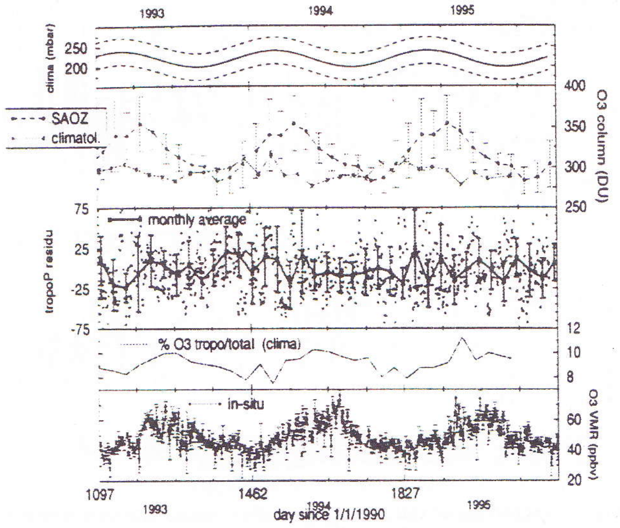

Fig. 3 ISSJ climatologies of total and tropospheric O_3 in comparison with tropopause pressure (tropoP) including 1σ limits (upper plot); residuals of actual tropoP with respect to the latter climatology, and their monthly averages (thick line) are shown in the third plot. Second plot: monthly mean TOC, from SAOZ (1990-1995 averages, dashed line) , in comparison with those calculated from the raw climatological model corresponding to the daily tropopause (solid line). Lower 2 plots: relative contribution of the tropospheric part of calculated TOC, and daily mean in-situ O_3 concentrations with 1σ 'error bar'.

of the total ozone content (TOC) is dominant in the SAOZ observations (Van Roozendael *et al.*, 1994), (ii), that 10% of the total O_3 column is of tropospheric origin (cf. plot 4 in Fig. 3), and (iii), that the seasonal variation's amplitude and phase depend on altitude (London, 1985; Staehelin *et al.*, 1991). The latter point is illustrated in the lowest plot of Fig. 3 that shows the in-situ O_3 concentrations at ISSJ (Filliger, 1996) of which the seasonal variation has a phase-lag in comparison with that of TOC. Therefore, we believe that the actual climatological model is already an improvement over the standard model but cannot yet represent the variability of ozone completely: we believe that exploitation of the in-situ measurements, that we found to be representative of the free-tropospheric amount at 3.6 km, will improve the representation of the lower part of the troposphere up to about 7 km, that is not or much less dependent on the tropopause altitude (in progress).

To demonstrate the impact of the a-priori vertical profile in the inversion procedure, Figure 4 compares the real O_3 columns observed by the Payerne soundings for some periods in 1991, 1994 and 1995, with the ones retrieved from the corresponding simulated spectra (3040 cm^{-1} spectral window, SZA = 75°, spectral resolution = 2.85mk) if in the retrieval procedure one adopts (a), the standard ozone vertical profile, and (b), the vertical profile climatology. For this test we used the SFSP code which was validated in the NDSC framework (Zander *et al.*, 1993), as was also SFIT. Although in both cases errors up to ±15% occur, the average error goes down from -1.83% in case (a) to 0.013% in (b); the standard deviation however does not decrease significantly (from 4.9% to 4.4%). It turns out that the error decreases mostly in cases

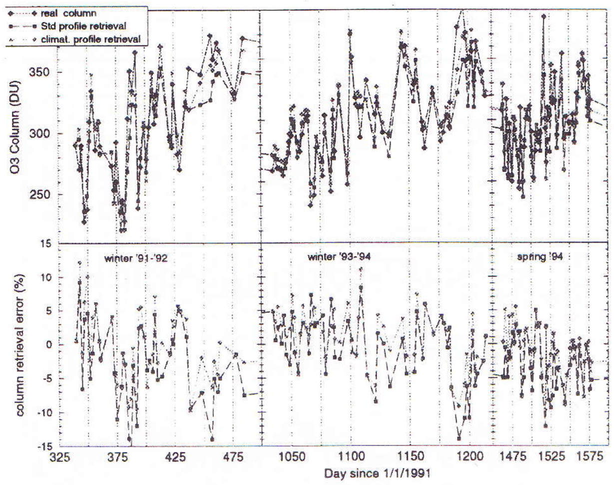

Fig. 4 Comparison of real and retrieved O_3 columns from a simulation of FTIR spectra in the 3040 cm^{-1} window, and retrieval by SFSP using several O_3 vertical profiles.

of exceptionally high or low tropopauses, for which the standard ozone vertical profile is quite different from the actual and climatological one, e.g., showing a secondary maximum just above the tropopause (see Fig. 2). The current simulations cover only winter-spring periods in which the variability of ozone is largest: the work will be extended to all seasons and to real observations.

Conclusions and perspectives

The agreement between the currently available total ozone timeseries from SAOZ and FTIR measurements at ISSJ is of the same order of magnitude as reported recently by David *et al.,* 1993 and Rinsland *et al.,* 1996 between FTIR and Dobson data. Contrary to this latter report that is based on a 10-days study, the present work covers 5 years of common measurements, from which a seasonal variation in the differences appears. The climatological O_3 vertical profile model that is presented in this paper should help resolve this latter discrepancy. The model up to now relates the vertical profile to the tropopause height only. Additional correlations between ozone concentration and meteorological parameters are known, as, e.g., temperature and potential vorticity. The latter one is an even better tracer of the ozone concentration, yet it has the disadvantage that it is not a purely local parameter, requiring ancillary data over some spatial extent. Integration of such knowledge in the model is envisaged in a near future. The ultimate goal is to establish a set of vertical profiles that will be used as a valid a-priori input for any vertical profile retrieval method, and for improving the accuracy of the retrieved ozone total columns.

Acknowledgements

Thanks are due to all organisations that support the Hochalpine Forschungsstation of the Jungfraujoch; to the Swiss Meteorological Institute for the meteorological data, in particular Dr. P. Viatte of the Station Aérologique at Payerne for the O_3 sonde data at Payerne and the Dobson data at Arosa; to Dr. P. Filliger of BUWAL (Bern) for communication of the in-situ O_3 data; to Dr. P. Newman and the National Center for Environmental Prediction (NCEP) (former NMC) for the p,T data; the Belgian Nationaal Fonds voor Wetenschappelijk Onderzoek, Fonds National de la Recherche Scientifique and the EC for financial support, through the contracts FKFO 2.0115.93, FRFC 2.4533.93, and ESMOS/Alps (EV5V-CT93-0348), respectively.

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