Recent evolution of atmospheric OCS above the Jungfraujoch station: Implications for the stratospheric aerosol layer

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Introduction

Stratospheric aerosols have important direct and indirect impacts on our environment, i.e., they affect the global radiative balance of the atmosphere by scattering and absorption processes, and they favor heterogeneous chemistry reactions, which threaten the ozone layer. These impacts have been clearly established during recent periods of strong volcanic activity (e.g., El Chichon in 1982 and Mt. Pinatubo in 1991), when large amounts of SO_2 were injected directly in the stratosphere. The latter ultimately end up into H_2SO_4, which takes a particulate form in the lower stratosphere. However, this so-called Junge sulfate aerosol layer, which was discovered nearly 50 years ago (Junge et al., 1961), persists at a background level during periods of low or no volcanic activity.

Crutzen (1976) was the first to point out that carbonyl sulfide (OCS) might be a potential source to maintain the stratospheric background sulfate aerosol layer. With a global average tropospheric mixing ratio of about 500 pptv (parts per trillion by volume, or 10^{-12} ppv) and a lifetime of 4 to 6 years, OCS is by far the most abundant sulfur compound in the remote, non perturbed free troposphere, as compared to other sulfur-bearing, shorter-lived source gases such as SO_2, CS_2, H_2S, DMS (CH_3SCH_3),... Its main sources result from both natural and anthropogenic emission processes at the ground, as well as oxidation in the boundary layer of other sulfur compounds listed above, in particular CS_2. Consequently, substantial amounts of OCS are lifted into the stratosphere, primarily through deep convection events in the tropics, where they progressively transform into SO_2 then into H_2SO_4, with further dispersion to higher latitudes according to the overall atmospheric circulation patterns and the stratospheric aerosol lifecycle proposed by Hamill et al.(1997).

In this work, we report the results of a reanalysis of solar spectra recorded with Fourier transform infra-red (FTIR) spectrometers, operated at the International Scientific Station of the Jungfraujoch (Switzerland, 46.5°N, 8.0°E, 3580 m altitude; http://www.ifjungo.ch) by the University of Liège (ULg), to retrieve, not only total but also distinct partial tropospheric- and stratospheric column abundances of OCS, as well as their relative temporal evolutions, by switching from the SFIT1- to the SFIT2 retrieval algorithm (e.g., Rinsland et al., 1998). The Jungfraujoch station hosts a series of research instruments operated within the frames of the Global Atmospheric Watch (http://www.wmo.ch) and the Network for the Detection of Stratospheric Change (http://www.ndsc.ws). The FTIRs operated there by ULg are affiliated to the latter Network, with the task to monitor changes in the Earth's atmospheric chemical composition at northern mid-latitudes, and to link them to the stratospheric ozone layer depletion and to climate changes.

Database and analysis approach include:

- Nearly 5000 very high-resolution (≤ 0.005 cm^{-1}), wide band-pass FTIR spectra recorded

regularly at the Jungfraujoch since the mid-1980s.

- Simultaneous three-windows spectral analysis (see Figure 1, taken from Mahieu et al., 2003) performed with the SFIT2-Version 3.81 retrieval algorithm, which implements the optimal estimation method of Rodgers (1990).
- Auxiliary inputs adopted in the fitting approach, in particular: (i) the HITRAN-2002+ (http://www.hitran.com) spectroscopic line parameter compilation; (ii) site-specific, daily P-T model atmospheres from the National Centers for Environmental Prediction (http://www.ncep.noaa.gov); (iii) most realistic *a priori* vertical concentration profiles for both OCS and interfering gases, taken from the ATMOS-Version3 (Irion et al., 2002; http://atmos.jpl.nasa.gov./atmos) database.
- Production of daily mean and, subsequently, monthly mean total and partial column abundances and their standard deviations; these latter are displayed in Figure 2.

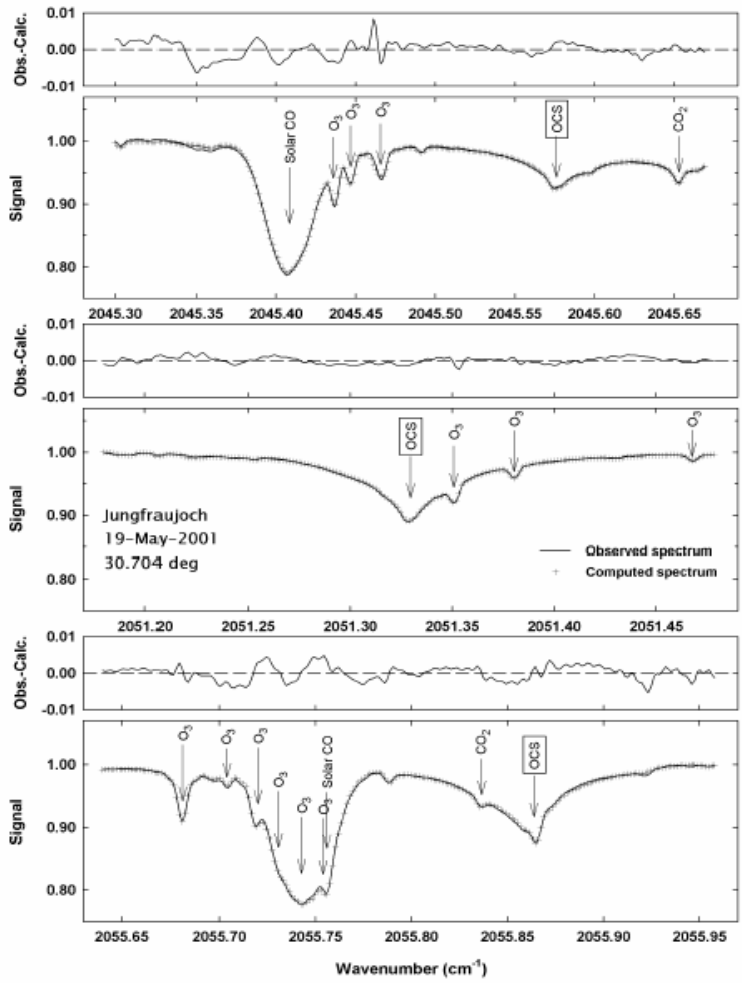

Figure 1. Simultaneous fitting example of the 3 microwindows selected for the OCS analysis with the SFIT2-Version 3.81 retrieval algorithm. The locations of the main target- and interfering gas absorptions are identified by arrows. Residuals (observed minus calculated signals) are displayed on expanded scales above each microwindow.

Main results and conclusions

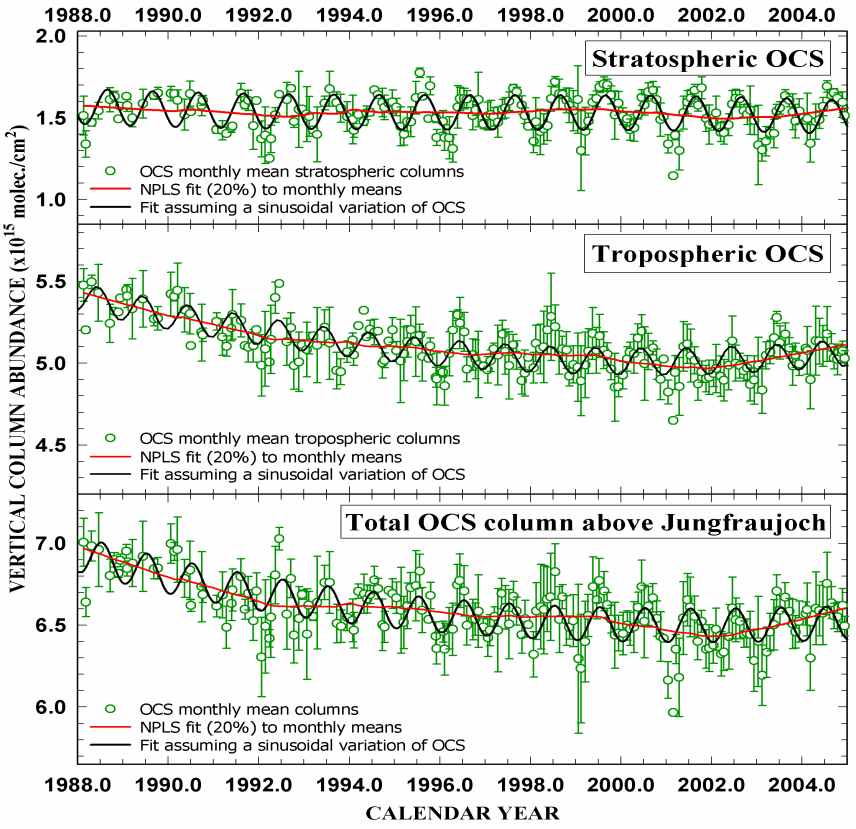

Figure 2. From top to bottom: the temporal evolution of the monthly mean partial stratospheric, partial tropospheric and total OCS column abundances above the Jungfraujoch, as derived with the SFIT2 retrieval algorithm. The mean tropopause altitude was assumed to be 12 km. The standard deviations around the monthly means reflect primarily the variability observed during each month. The continuous red lines in the different frames correspond to nonparametric least-squares fits with sampling proportion of 20% and Gaussian weighting function to the data points. The black lines help visualising the seasonal variations of the datasets.

Striking features emerging from Figure 2 include:

- the overall similarity in the evolution of the long-term trends of the OCS burdens in the stratosphere and in the troposphere, with respective contributions to the total atmospheric loading of about 20% and 80%; considering the altitude of the Jungfraujoch site, this is commensurate with model predictions (e.g., Chin and Davis, 1995) as well as observations of OCS in the southern hemisphere (N. Jones and S. Wood, private communications, 2004).
- the significant total column decreases since the late 1980s till about 2001, followed by a re-increase during the past 3 years; related discrete trends (in %/year) derived from the red curve in the bottom frame are: -1.36%/yr. in 1987-88; -0.82 in 1991-92; -0.46 in 1995-96; -0.67 in 1999-2000, and +1.05%/yr. in 2003-04;
- the seasonal variations simulated by a sinusoidal function through the tropospheric and the stratospheric data points, returning maxima occurring in early June in the troposphere, and in late August in the stratosphere.

Considering the length and the internal consistency of the present OCS database, and because of the statistical significance of the features evoked above, OCS-related chemistry-transport model calculations should be performed to test our understanding of the chemistry affecting sulfate-bearing constituents in the troposphere and in the stratosphere and help improving our current estimates of OCS budgets which still report excess sources over sinks by nearly a factor of 2 (Brasseur et al., 1999). Details regarding the approach for trend evaluation and comparisons with other recent related investigations are documented, e.g., in Mahieu et al. (2003) and in Rinsland et al. (2002).

Acknowledgements

This research has been supported by the Belgian Federal Science Policy Office and the EC-Directorate General, both in Brussels. Thanks are extended to all collaborators having contributed to the observations at the Jungfraujoch. Operation of the site is coordinated by the International Foundation HFJG, Switzerland.

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