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Optimized approach to retrieve information on the tropospheric and stratospheric carbonyl sulfide (OCS) vertical distributions above Jungfraujoch from high-resolution FTIR solar spectra.

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Carbonyl sulfide (OCS), which is produced in the troposphere from both biogenic and anthropogenic sources, is the most abundant gaseous sulfur species in the unpolluted atmosphere. Due to its low chemical reactivity and water solubility, a significant fraction of OCS is able to reach the stratosphere where it is converted to SO_2 and ultimately to H_2SO_4 aerosols (Junge layer). These aerosols have the potential to amplify stratospheric ozone destruction on a global scale and may influence Earth's radiation budget and climate through increasing solar scattering. The transport of OCS from troposphere to stratosphere is thought to be the primary mechanism by which the Junge layer is sustained during nonvolcanic periods. Because of this, long-term trends in atmospheric OCS concentration, not only in the troposphere but also in the stratosphere, are of great interest.

A new approach has been developed and optimized to retrieve atmospheric abundance of OCS from high-resolution ground-based infrared solar spectra by using the SFIT-2 (v3.91) algorithm, including a new model for solar lines simulation (solar lines often produce significant interferences in the OCS microwindows). The strongest lines of the ν_3 fundamental band of OCS at 2062 cm^{-1} have been systematically evaluated with objective criteria to select a new set of microwindows, assuming the HITRAN 2004 spectroscopic parameters with an increase in the OCS line intensities of the $\nu_3$band main isotopologue $^{16}O^{12}C^{32}S$ by 15.79% as compared to HITRAN 2000 (Rothman et al., 2008, and references therein). Two regularization schemes have further been compared (deducted from ATMOS and ACE-FTS measurements or based on a Tikhonov approach), in order to select the one which optimizes the information content while minimizing the error budget.

The selected approach has allowed us to determine updated OCS long-term trend from 1988 to 2009 in both the troposphere and the stratosphere, using spectra recorded on a regular basis with Fourier Transform Infrared spectrometers (FTIRs), under clear-sky conditions, at the NDACC site (Network for the Detection of Atmospheric Composition Change, visit http://www.ndacc.org) of the International Scientific Station of the Jungfraujoch (Swiss Alps, 46.5˚N, 8.0˚E, 3580m asl). Trends and seasonal cycles deduced from our results will be compared to values published in the literature and critically discussed. In particular, we will confirm the recent change in the OCS total column trend, which has become positive since 2002 before undergoing a slowing down over the last years.

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