



Impacts of stratospheric dynamical variability on total inorganic fluorine from observations and models constrained by state-of-the-art reanalyses

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Man-made halogenated compounds emitted from the Earth's surface ultimately reach the stratosphere where they undergo photolysis, leading to three main fluorine reservoirs: hydrogen fluoride (HF), carbonyl fluoride (COF₂) and carbonyl chloride fluoride (COCIF). This process is directly influenced by the strength of the mean meridional circulation of the stratosphere, the Brewer-Dobson Circulation (BDC). The BDC is projected to speed-up with the greenhouse gases induced global warming. However, studies have highlighted a multiyear variability in the strength of the BDC resulting in hemispheric asymmetries in observed and modelled trends of age of air and long-lived tracers.

Total inorganic fluorine (F_y , the fluorine weighted sum of HF, COF₂ and COCIF) is used here as a tracer of the stratospheric circulation changes. We perform an analysis and interpretation of Fourier transform infrared (FTIR) multidecadal time-series of HF and COF₂ from the Jungfraujoch (Switzerland, 46.55°N) and Lauder (New-Zealand, 45.03°S) stations and from the space-borne Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS). Indeed, the summation of HF and COF₂ is a very good proxy of F_y as we determine, from ACE-FTS and the chemical-transport model (CTM) TOMCAT, that COCIF is only accounting for less than 5% of the total F_y budget.

The kinematic CTM BASCOE (Belgian assimilation system for chemical observations) is used here to assess the representation of the investigated circulation changes in four state-of-the-art meteorological reanalyses, i.e., ERA-Interim, JRA-55, MERRA and MERRA-2. We also investigate if WACCM4 (Whole Atmosphere Community Climate Model version 4) is able to reproduce these changes through a free-running simulation.

The ground-based and satellite FTIR time-series of COF₂ show contrasting results over their

common time period (2004-2019), with a positive total column trend above the Jungfrauoch, and a non-significant (ground-based) or decreasing trend (ACE-FTS) above Lauder. We find large discrepancies between the BASCOE-CTM simulations, with MERRA-2 inducing overly large simulated F_y total columns which could confirm the weaker tropical upwelling highlighted in previous age of air studies.