Impact of lower stratospheric dynamical variability on total inorganic fluorine derived from ground-based FTIR, satellite and model data

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Background and objectives

The Brewer-Dobson Circulation (BDC, Fig. 1) is projected to speed-up as a result of global warming and thus the mean of age of air (AoA) is expected to decrease. However, AoA studies showed contradicting results. AoA trends exhibit a hemispheric dipole structure for the period 2002-2012 with negative trend values (AoA is lower, i.e., air is younger) in the Southern Hemisphere and positive trend values in the Northern Hemisphere. This structure is likely induced by a southward shift of the BDC global pattern (Stiller et al., 2017). In the end, these recent results demonstrate that the BDC speed is not monotonously increasing over decal time scales and that some variability exists.

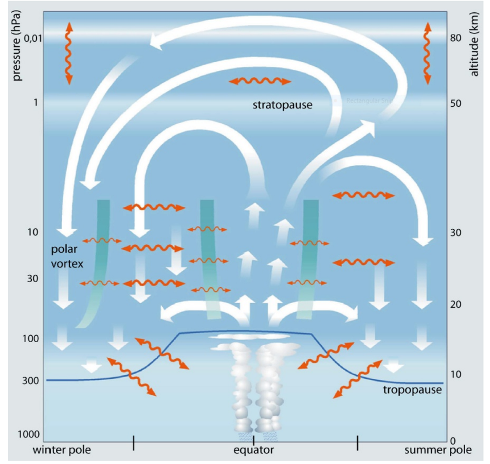

Figure 1: "Schematic of the BDC as the combined effect of residual circulation and mixing in the stratosphere and mesosphere. The thick white arrows depict the residual circulation whereas the wavy orange arrows indicate two-way mixing processes. Both, circulation and mixing are mainly induced by wave activity on different scales (planetary to gravity waves). The thick green lines represent stratospheric transport and mixing barriers." (Bönisch et al., 2011)

The recent stratospheric dynamical variability has been linked with variations in stratospheric chemical tracers time series and the hemispheric asymmetry have also been detected in their trends (e.g., HCl in Mahieu et al., 2014; HF in Harrison et al., 2016 or N2O in Nedoluha et al., 2015a).

In the framework of these results, we investigate on the impacts of the last two decades stratospheric dynamical variability on the chemical composition of the stratosphere using the total inorganic fluorine (Fy, see corresponding frame). Furthermore, BASCOE-CTM is used to evaluate the representation of the investigated circulation changes in state-of-the-art meteorological reanalyses (see frame below). Finally, we also evaluate if WACCM, in its free running mode, is able to reproduce the investigated dynamical impacts on Fy.

Datasets, methods and tools

► FTIR observations:

Very high-resolution infrared solar spectra recorded at the Jungfraujoch scientific station (Switzerland, 46.55°N, 3580 m amsl) are used in order to retrieve total columns of COF_2 and HF. For COF_2, we apply the multi-spectrum approach of Duchatelet et al. (2009), using only the three windows (around ~1936cm^{-1} and ~1952 cm^{-1}) encompassed in the InSb detector spectral range. For HF, we apply the retrieval strategy described in Duchatelet et al. (2010). The retrievals are performed using the SFIT-4 v0.9.4.4 algorithm implementing the Optimal Estimation Method of Rodgers (2000).

► ACE-FTS:

HF, COF_2 and COClF observations from the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer version 3.6 (ACE-FTS; Bernath et al., 2005) are included in this work.

► Chemistry-Transport Models:

-- BASCOE-CTM (Belgian Assimilation System for Chemical ObsErvations; Chabrillat et al., 2018): BASCOE-CTM was recently used to calculate to mean age of air from modern meteorological reanalyses (Chabrillat et al., 2018) in the framework of the Stratosphere–troposphere Processes And their Role in Climate (SPARC) Reanalysis Intercomparison Project (S- RIP). The results show large disagreement between the reanalyses compared. In consequence, in this work, we compare the results of three simulations of BASCOE driven by ERA-Interim, JRA-55 and MERRA-2.

Resolution: 2x2.5° with 60 vertical levels up to ~60 km. Lower boundary conditions: CMIP6 recommendations (Meinshausen et al., 2017). Note that the model has a simplified chemistry of fluorine, i.e., all fluorine sources lead directly to Fy.

-- TOMCAT (Chipperfield et al., 2018). Resolution: 2.8x2.8° with 32 veritacal levels to ~60 km. Lower boundary conditions: Source gas global mean volume mixing ratios from the Scientific Assessment of Ozone Depletion 2018 (WMO)

► Chemistry-Climate Model:

-- WACCM (Whole Atmosphere Community Climate Model version 4; See poster Minganti et al., EGU2019-7472). We include WACCM in a free-running configuration (FR-WACCM). Resolution: 1.9x2.5° with 66 vertical levels up to ~150 km. Same lower boundary conditions as BASCOE-CTM.

► Nonparametric trend smoothing:

Smoothed data in Fig. 3 and 4 are obtained by applying the nonparametric trend smoothing of Friedrich (2019)

Inorganic Fluorine Budget above JungfrauJoch

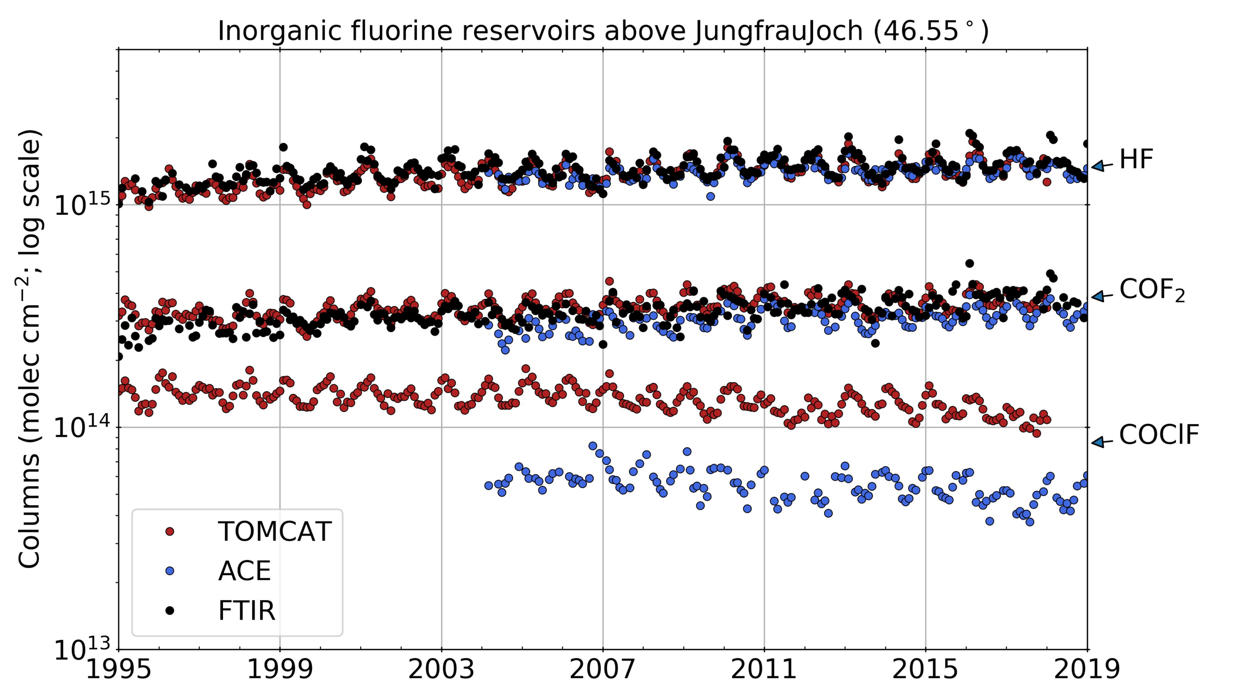

Figure2: Inorganic fluorine reservoirs above Jungfraujoch as observed by ground-based FTIR and ACE (41°N-51°N), and simulated by TOMCAT.

Sources of inorganic fluorine are the manmade chlorofluorocarbons, hydrochlorofluorocarbons and hydrofluorocarbons (CFC, HCFC and HFC, respectively). These long-lived gases reach the stratosphere where they undergo photolysis leading to two main temporary inorganic fluorine reservoirs: carbonyl chloride fluoride (COClF) and carbonyl fluoride (COF_2). COClF and COF_2 in turn undergo photolysis to form the ultimate inorganic fluorine product: hydrogen fluoride (HF). Finally, the weighted sum of the concentrations of these reservoirs defines the total inorganic fluorine Fy:

$$Fy = [HF] + 2x[COF_2] + [COClF]$$
$$= 68\pm2 + 29\pm2 + 3\pm0.5 \quad \text{\% in ACE-FTS (41°N-51°N)}$$
$$= 63\pm2 + 31.5\pm1.5 + 6\pm1 \quad \text{\% in TOMCAT}$$

COF2 and HF FTIR products

Figure 2 depicts the good agreement between the three datasets for COF_2 and HF.

We compute the following fractional differences among the datasets:

-- COF_2

FTIR - ACE = 12.46%

FTIR - TOMCAT = -4.86%

-- HF

FTIR - ACE = 6.71%

FTIR - TOMCAT = 5.84%

Total Inorganic Series (Fy) Above Jungfraujoch

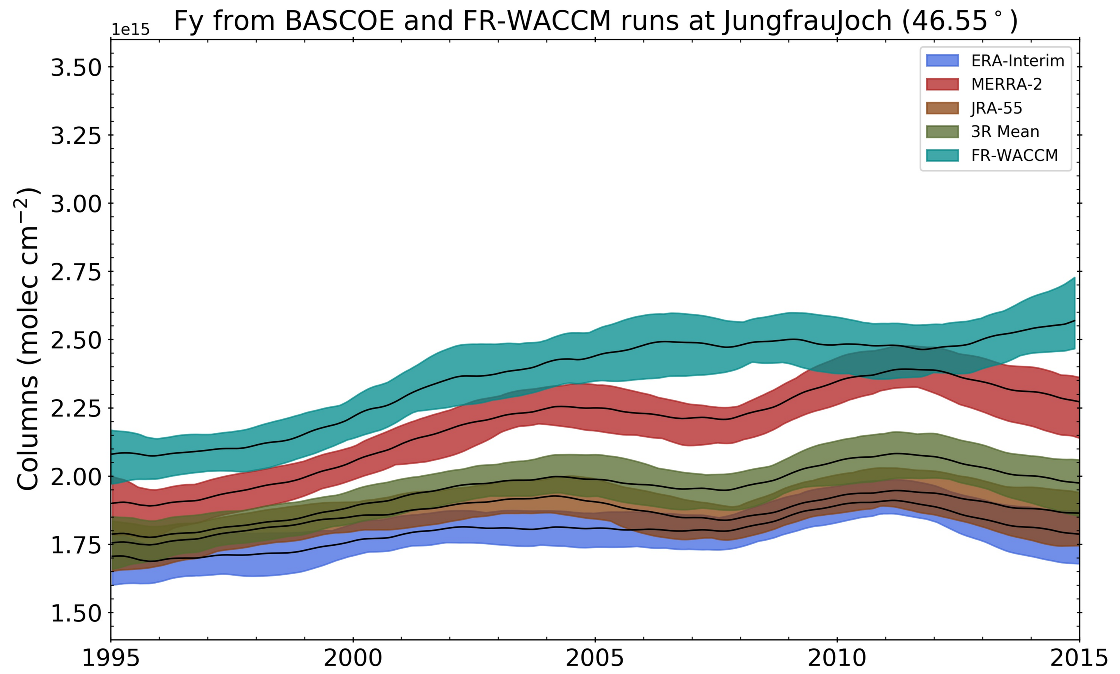

Figure 3: Fy series simulated by BASCOE (driven by ERA-Interim, JRA-55 and MERRA-2) and FR-WACCM. 3R Mean series is the average of the 3 BASCOE simulation results. Lines and shades are the nonparametric estimations performed on the daily mean data. Results are extracted from the station nearest pixel.

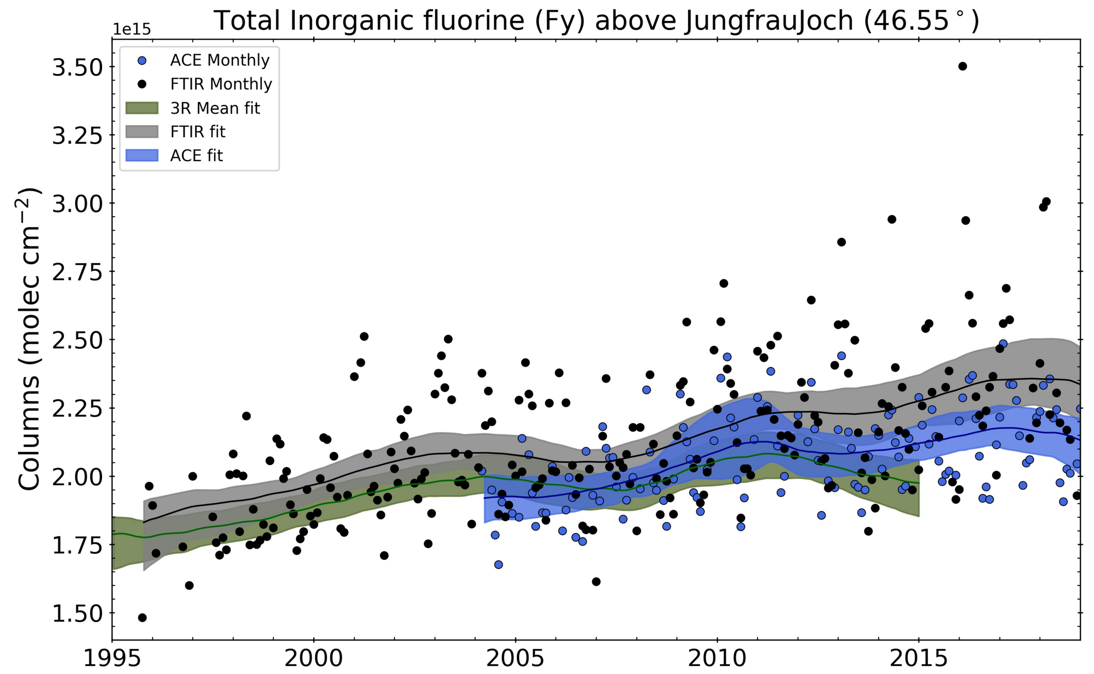

Figure 4: Fy series above Jungfraujoch as observed by ground-based FTIR and ACE (41°N-51°N). Note that the ground-based series is not strictly Fy as only HF and COF_2 are available but that this approximation should correspond to at least 95% of Fy (see frame above). 3R Mean is the average of the 3 BASCOE simulation results (Fig. 4). The gray, blue and green lines and shades are the nonparametric estimations performed on the daily mean data.

Figure 3 and 4 show that WACCM with free dynamics (FR-WACCM) simulates too large Fy columns when compared to observations (FTIR and ACE) and BASCOE-CTM. Fy columns simulated by BASCOE-CTM driven by MERRA-2 are also biased high. Chabrillat et al. (2018) showed that MERRA-2 has the oldest AoA among the three reanalyses compared here, thus explaining the larger columns.

For the comparison with observations (Fig. 4), we use an average of the three BASCOE-CTM simulations (3R mean). The three datasets of Fig. 4 are in an overall good agreement. BASCOE-CTM seems however to simulate a decreasing trend at the end of the simulation period (~2010-2015) that is not observed by ground-based FTIR and ACE.

The minimum in HCl series around 2007 discussed by Mahieu et al. (2014; Fig. 5) seems also detectable on the three datasets of Fig 4. This preliminary result may confirm that the stratospheric dynamical variability of the last two decades has also an impact on Fy series.

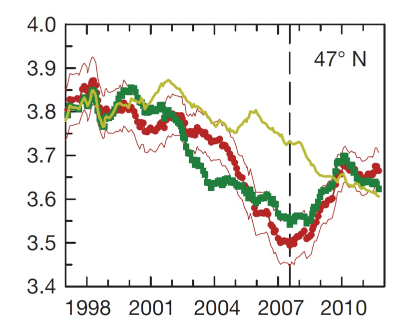

Figure 5: Running average total column time series (1997-2011) of HCl at Jungfraujoch from FTIR observations and SLIMCAT (TOMCAT) simulations. The thin red lines correspond to the ±2 standard error of the mean range. S2000 is a SLIMCAT run with fixed dynamics of 2000, from 2000 onwards. From Mahieu et al. (2014).

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Conclusions and future work

► From the present work, we showed that:

-- From Jungfraujoch FTIR observations, we can build a very good approximation of Fy using only HF and COF2.

-- The three simulations of BASCOE-CTM, using three reanalyses (ERA-Interim, JRA-55 and MERRA-2), show large differences between the simulated Fy columns, highlighting, as many other S-RIP studies, the large discrepancies existing in the stratosphere among modern reanalyses.

-- The impact of stratospheric dynamical variability on tracers depicted in other studies (See Background frame) seems to be also detectable on Fy series.

-- WACCM, with free dynamics, produces too large Fy columns and does not seems to capture the investigated variability.

► Future work:

-- We plan to include Lauder FTIR NDACC station (45°S) HF and COF_2 FTIR series in order to highlight the hemispheric asymmetry. The Lauder COF_2 product is still in development with the help of Dan Smale.

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