



Post-peak trend of upper stratospheric hydrogen chloride derived from ground-based FTIR solar spectra and model simulations

Emmanuel Mahieu (1), Maxime Prignon (1), Christian Servais (1), Simon Chabrillat (2), Quentin Errera (2), Marina Friedrich (3), Stephan Smeekes (4), Lucien Froidevaux (5), Ross J. Salawitch (6), Pamela Wales (7), Justus Notholt (8), and Martyn P. Chipperfield (9)

(1) Institute of Astrophysics and Geophysics, University of Liège, Liège, Belgium (emmanuel.mahieu@uliege.be), (2) Royal Belgian Institute for Space Aeronomy, Brussels, Belgium, (3) Potsdam Institute for Climate Impact Research, Potsdam, Germany, (4) Department of Quantitative Economics, Maastricht University, Maastricht, The Netherlands, (5) Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, (6) Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD, (7) Department of Chemistry and Biochemistry, University of Maryland, College Park, MD, (8) Institute for Environmental Physics, University of Bremen, Bremen, Germany, (9) School of Earth and Environment, University of Leeds, Leeds, United Kingdom

After several decades of sustained increase, hydrogen chloride (HCl, the main reservoir for stratospheric chlorine) reached a maximum abundance around 1997. Since then, its decrease has been documented, characterized by short-term variability which was attributed to atmospheric circulation changes, affecting mainly the lower stratosphere (Mahieu et al., 2014). This notably led to a temporary increase of HCl over 2007-2011, complicating the determination of the long-term HCl trend and the accurate verification of the success of the Montreal Protocol for the protection of the stratospheric ozone layer. Studies have used other long-lived tracers to remove the effects of dynamical variability in the lower stratosphere (e.g., Stolarski et al., 2018), while other investigations have suggested that trends in the upper stratosphere were potentially more appropriate for the long-term characterization of the HCl decrease (e.g., Froidevaux et al., 2015; Bernath and Fernando, 2018), especially when dealing with satellite height-resolved data.

In this contribution, we use FTIR (Fourier Transform InfraRed) data from the Jungfraujoch station (Swiss Alps, 3580 m a.s.l.), a site of the NDACC network (<http://www.ndacc.org>), to study the evolution of HCl in some detail. The SFIT-4 retrieval algorithm implementing the Optimal Estimation Method of Rodgers (2000) is employed, providing HCl columns with good sensitivity from the tropopause up to about 40 km altitude. Moreover, the vertical resolution is sufficient to determine independent partial columns for the lower and upper stratosphere.

With the support of model simulations performed with the 3D-Chemistry Transport Model of the Belgian Assimilation System for Chemical Observations (BASCOE; Chabrillat et al., 2018), driven by the ERA-Interim meteorological reanalysis, we investigate the post-peak trend of HCl in the lower and upper stratosphere. We also determine the magnitude of the uncertainties affecting the various trends, using bootstrap tools which are specifically developed to take into account the auto-correlation present in our geophysical data sets.

Acknowledgments

This research has been primarily supported by the F.R.S. – FNRS (Brussels), under Grants T.0040.16 and J.0147.18. The vital supports from the Fédération Wallonie Bruxelles and the Swiss GAW-CH programme are further acknowledged. We thank the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG, Bern) for supporting the facilities needed to perform the observations. We gratefully acknowledge support by the Deutsche Forschungsgemeinschaft DFG within the SFB/Transregio 172 (AC)3.

References

- Bernath, P. and Fernando, A. M.: Trends in stratospheric HCl from the ACE satellite mission, *J. Quant. Spectrosc. Radiat. Transf.*, 217, 126–129, doi:10.1016/j.jqsrt.2018.05.027, 2018.
- Chabrillat, S., Vigouroux, C., Christophe, Y., et al.: Comparison of mean age of air in five reanalyses using the BASCOE transport model, *Atmos. Chem. Phys.*, 18(19), 14715–14735, doi:10.5194/acp-18-14715-2018, 2018.
- Froidevaux, L., Anderson, J., Wang, H. J., et al.: Global Ozone Chemistry and Related trace gas Data records for the Stratosphere (GOZCARDS): Methodology and sample results with a focus on HCl, H₂O, and O₃, *Atmos. Chem. Phys.*, 15(18), 10471–10507, doi:10.5194/acp-15-10471-2015, 2015.

Mahieu, E., Chipperfield, M. P., Notholt, J., et al.: Recent Northern Hemisphere stratospheric HCl increase due to atmospheric circulation changes, *Nature*, 515(7525), 104–107, doi:10.1038/nature13857, 2014.

Rodgers, C.D.: Inverse methods for atmospheric sounding: theory and practice (Series on atmospheric, oceanic and planetary physics), Vol. 2. Singapore, 2000.

Stolarski, R. S., Douglass, A. R. and Strahan, S. E.: Using satellite measurements of N₂O to remove dynamical variability from HCl measurements, *Atmos. Chem. Phys.*, 18(8), 5691–5697, doi:10.5194/acp-18-5691-2018, 2018.