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Investigating stratospheric changes between 2009 and 2018 with aircraft, AirCores, a global model and a focus on CFC-11

Johannes Laube[1], **Felix Ploeger**[1], and the Stratospheric CFC-11 team*
[1]Forschungszentrum Jülich GmbH, Institut für Energie- und Klimaforschung - Stratosphäre (IEK-7), Jülich, Germany (f.ploeger@fz-juelich.de)
*A full list of authors appears at the end of the abstract

We present new observations of trace gases in the stratosphere based on a cost-effective sampling technique that can access much higher altitudes than aircraft. The further development of this method now provides detection of species with abundances in the parts per trillion (ppt) range and less. We focus on mixing ratios for CFC-11 and CFC-12 which are important for understanding stratospheric ozone depletion and circulation. After demonstrating the quality of the data through comparisons with ground-based records and aircraft-based observations we combine them with the latter to demonstrate their potential. We first compare them with results from a global model driven by three widely used meteorological reanalyses (ERA-Interim, JRA-55, MERRA-2). Secondly, we focus on CFC-11 as recent evidence has indicated renewed atmospheric emissions of that species relevant on a global scale. Because the stratosphere represents the main sink region for CFC-11, potential changes in stratospheric circulation and troposphere-stratosphere exchange fluxes have been identified as the largest source of uncertainty for the accurate quantification of such emissions. Our observations span over a decade (up until 2018) and therefore cover the period of the slowdown of CFC-11 global mixing ratio decreases measured at the Earth's surface. The spatial and temporal coverage of the observations is insufficient for a global quantitative analysis, but we do find some trends that are in contrast with expectations; indicating that the stratosphere may have contributed to tropospheric changes. Further investigating the model data we find that the required dynamical changes in the stratosphere required to explain the apparent change in tropospheric CFC-11 emissions after 2013 are possible, but with a very high uncertainty range in the change of stratosphere-to-troposphere flux of CFC-11. This is partly caused by the high variability of mass flux from the stratosphere to the troposphere, especially at time scales of a few years, and partly by large differences between runs driven by different reanalysis products, none of which agree with our observations well enough for such a quantitative analysis.

Stratospheric CFC-11 team: Emma C. Leedham Elvidge (2,3) Karina E. Adcock (2), Bianca Baier (11,4), Carl A.M. Brenninkmeijer (5), Huilin Chen (6), Elise Droste (2), Jens-Uwe Grooß (1), Pauli Heikkinen (7), Andrew J. Hind (2), Rigel Kivi (7), Alex Lojko (2,8), Stephen A. Montzka (4), David E. Oram (2), Steve Randall (9), Thomas Röckmann (10), William T. Sturges (2), Colm Sweeney (4), Max

Thomas (2), Elinor Tuffnell (2)