Diagnosing CFC-11's Emissions in a Chemistry-Climate Model

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Trichlorofluoromethane (CFC-11) is one of the major stratospheric ozone-depleting substances for which production has been strictly controlled by the Montreal Protocol. As demonstrated in a recent paper (Montzka et al. 2018), the decline of global CFC-11 concentration slowed by 50% after 2012, and the hemispheric difference of CFC-11 increased by 50% concurrently. Neither the observed global growth rate nor the hemispheric difference of CFC-11 after 2012 can be simulated in our Chemistry-Climate Model (CCM) simulations without an emission increase. However, large uncertainties still remain on how much and where the additional emissions occur. Much of this uncertainty comes from poor constraints on interannual variability in stratosphere-troposphere exchange of CFC-11. Thus, to quantify the emissions from global surface concentration changes we need a better understanding of the stratospheric influence on the global growth rate and hemispheric difference. We use a CCM to show that some of the seasonal and inter-annual variability observed by the ground-based measurements include stratospheric dynamical components. This dynamical variability is connected to changes in the Brewer-Dobson circulation and the quasi-biennial oscillation (QBO) in the stratosphere. We explore uncertainties in the modeled surface concentrations of CFC-11 and other trace gases induced by choices in the modeling configuration, such as the reanalysis input, model resolution, and nudging methods. We demonstrate that the ESRL/GMD measurements of long-lived gases provide an excellent observational constraint to diagnose and improve the CCM's capability to model dynamical variability. In addition, the surface measurements provide one of the few means by which interannual changes in stratosphere-troposphere exchange can be assessed.

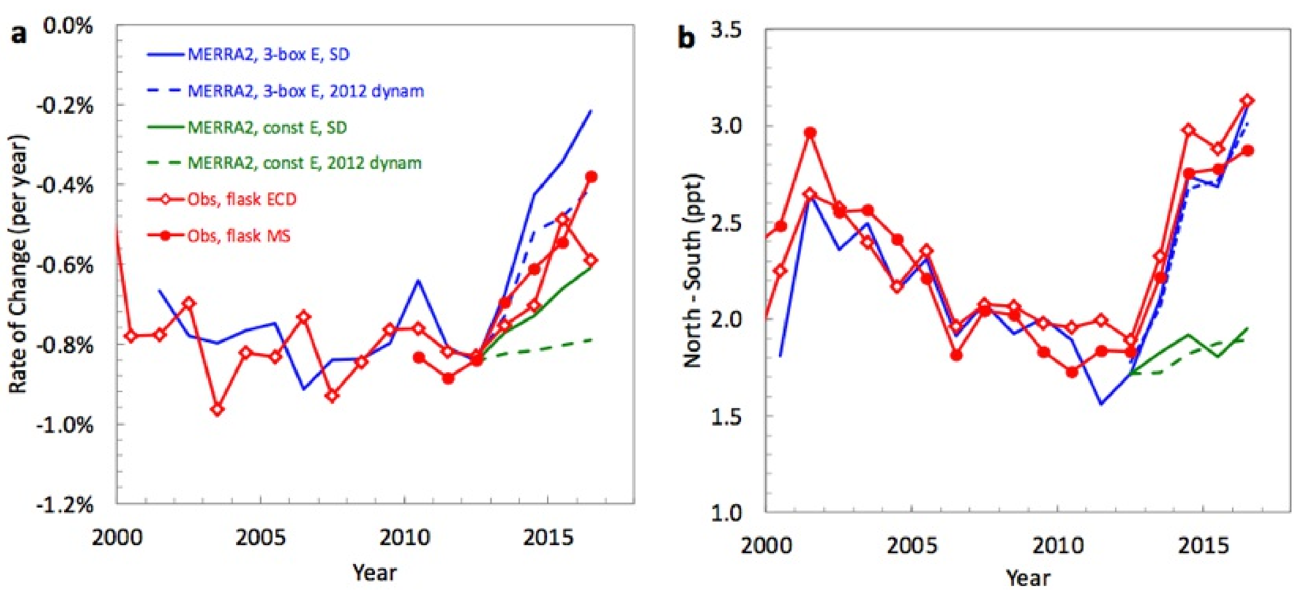

Figure 1. Global annual rates of change and hemispheric differences from 3-D simulations and observations. **a**, Global rates for CFC-11 mole fractions simulated in a 3-D model (CAM) with Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) reanalysis meteorology (SD) and emissions from 1) the 3-box model (blue lines), or 2) same as 1) but with constant emissions after 2012 at the 2012 rate (green lines). Simulations were also performed with 2012 dynamics for years after 2012 (dashed lines). **b**, simulated and observed hemispheric differences. In **a** and **b**, observed annual means are from flasks analyzed by GCECD and GCMS (red lines and symbols).