A recent slowdown in the decline of CFC-11 concentrations in the upper troposphere

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Intro/highlights

- Recent studies based solely on surface-level measurements have shown strong evidence for new CFC-11 production, leading to an increase in CFC-11 emissions over the past decade (*Montzka et al*., 2018; *Prinn et al*., 2018; *Rigby et al*., 2019).
- The Atmospheric Chemistry Experiment – Fourier Transform Spectrometer (ACE-FTS) is the only space-based instrument that measures height-resolved concentrations of CFC-11 in the UTLS
- ACE-FTS measurements show that, between ~6-9 km, the rate of decrease of global CFC-11 concentrations was slower during 2013-2018 (-1.2 pptv/year) than during 2004-2012 (-2.0 pptv/year). Similar to surface measurements.
- These trends are consistent with TOMCAT 3-D model simulations constrained by observed surface CFC-11 values. The measured trends below ~10 km are significantly slower for 2013-2018 than those from a model simulation without increased emissions (-2.4 pptv/year).

ACE-FTS

Atmospheric Chemistry Experiment – Fourier Transform Spectrometer

- Canadian satellite SciSat was launched in August 2003
  - Data starts in Feb 2004, and is still operational
- ACE-FTS is a solar occultation, limb viewing instrument
  - High spectral resolution FTS in the 2.2 to 13.3 μm spectral range
  - Vertical resolution of 3-4 km, ~6-120 km altitude range
  - Majority of measurements are at high latitudes
- 74 trace species are retrieved, including:
  - CFC-11, CFC-12, CFC-113, HCFC-22, HCFC-142b, HCFC-141b, HFC-134a, HFC-23
- Level 2 version 4.0 data were used in this study
  - Feb 2004 to December 2018

TOMCAT/SLIMCAT

- 3-D offline chemical transport model
  - Tropospheric and stratospheric chemistry using prescribed source-gas surface boundary conditions
  - Concentrations of multiple trace species in O_x, NO_y, HO_x, Br_y and Cl_y families
  - The model is forced using wind and temperature fields from ERA-Interim meteorological reanalysis data
    - See Dhomse et al., (2019)

- Model output data was sampled at ACE-FTS locations

- Two different runs, with different surface CFC-11 inputs:
  - Control run: input is global average surface measurements (up to 2017)
  - Banks run: input is global average minus “extra” emissions, so that the only source of emissions is from estimated bank sources

Methodology

- Data were averaged to make 30-day mean time series at each altitude level and were fit using multivariate linear regression to the model

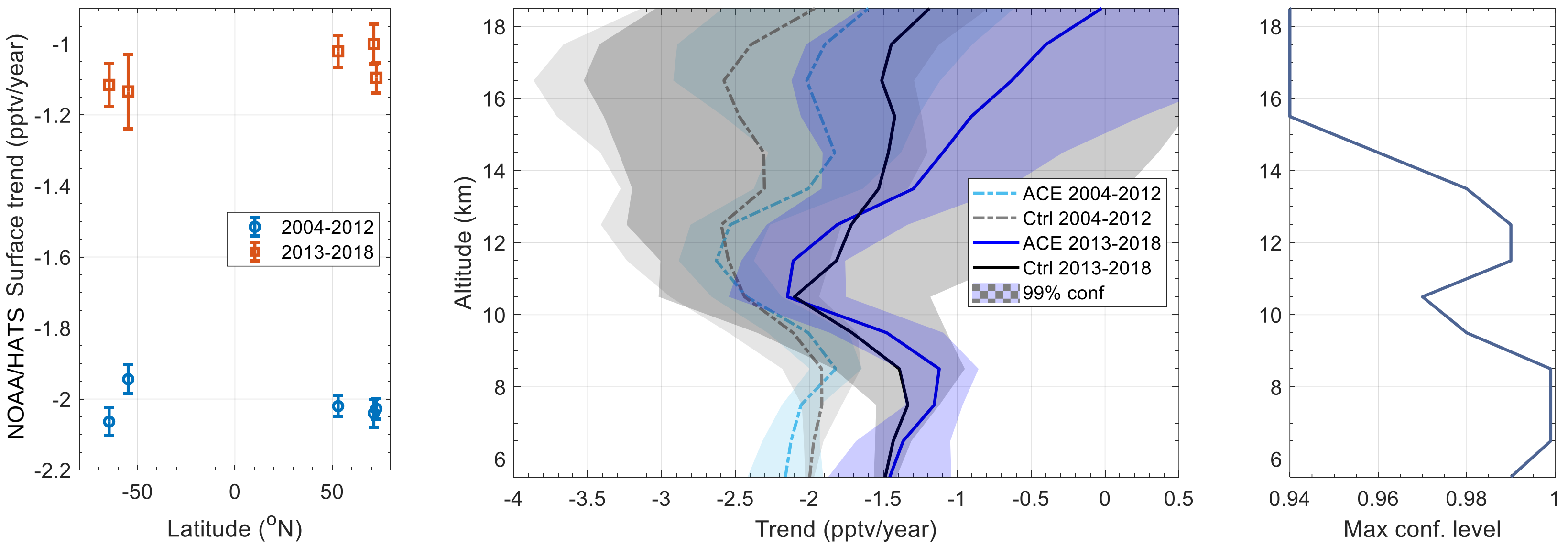
$$f(t) = a + b_1 t + b_2 \sin(2\pi\phi_1) + b_3 \sin(4\pi\phi_2) + b_4 DT,$$

  - t is time in years,
  - a is the offset,
  - b_i are the resulting regression coefficients,
  - first sine term is an annual cycle with phase ϕ_1,
  - second sine term is a semi-annual cycle with phase ϕ_2.
  - DT is ACE-FTS 30-day mean N_2O data, a dynamical tracer,
    - instead of using multiple indices such as QBO, ENSO, F10.7, etc. as proxies.

Global, 30-day mean results

Similar changes in trend are observed at the surface as those in the in the upper troposphere

- 2004-2012: ~2.0 pptv/year
- 2013-2018: ~1.2 pptv/year

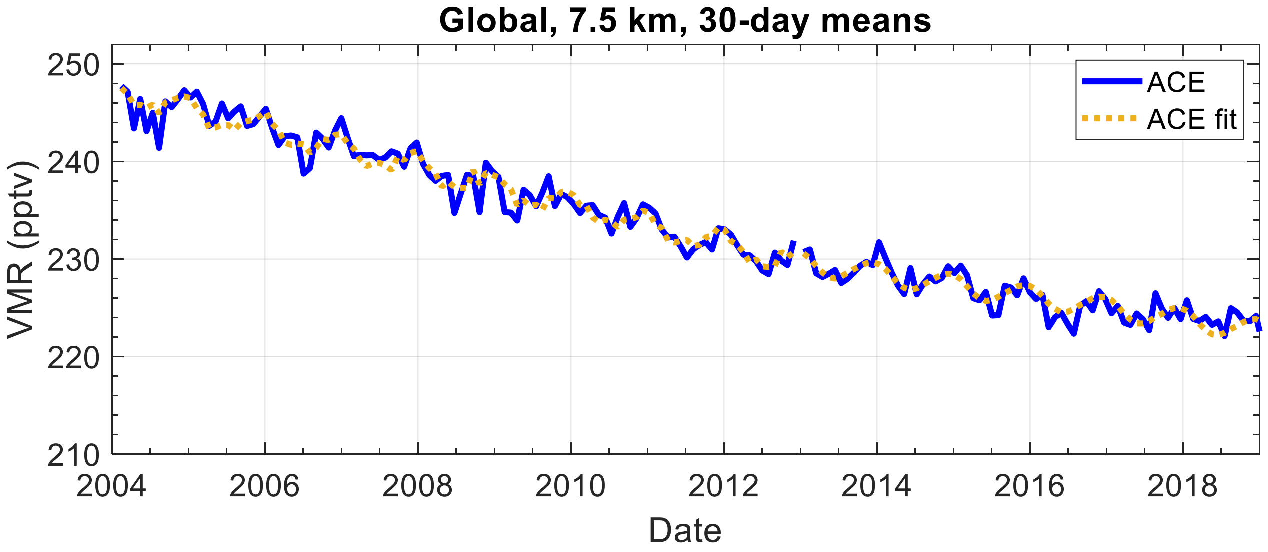


ACE-FTS trends were calculated for a range of confidence levels between 0.9 and 0.999. Figure shows max conf level where there is still a significant difference between 2004-2012 and 2013-2018 trends

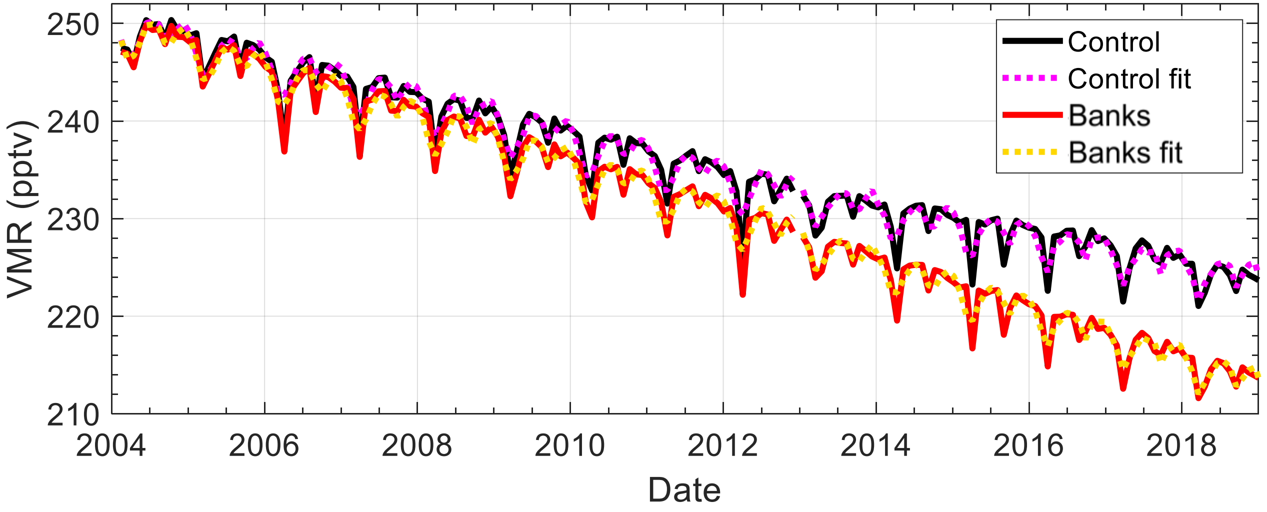
Linear trend values were similarly calculated for monthly averaged surface CFC-11 measurements from the NOAA/HATS network. Showing data from high latitude stations

Global, 30-day mean results

ACE-FTS values and corresponding linear regression fit

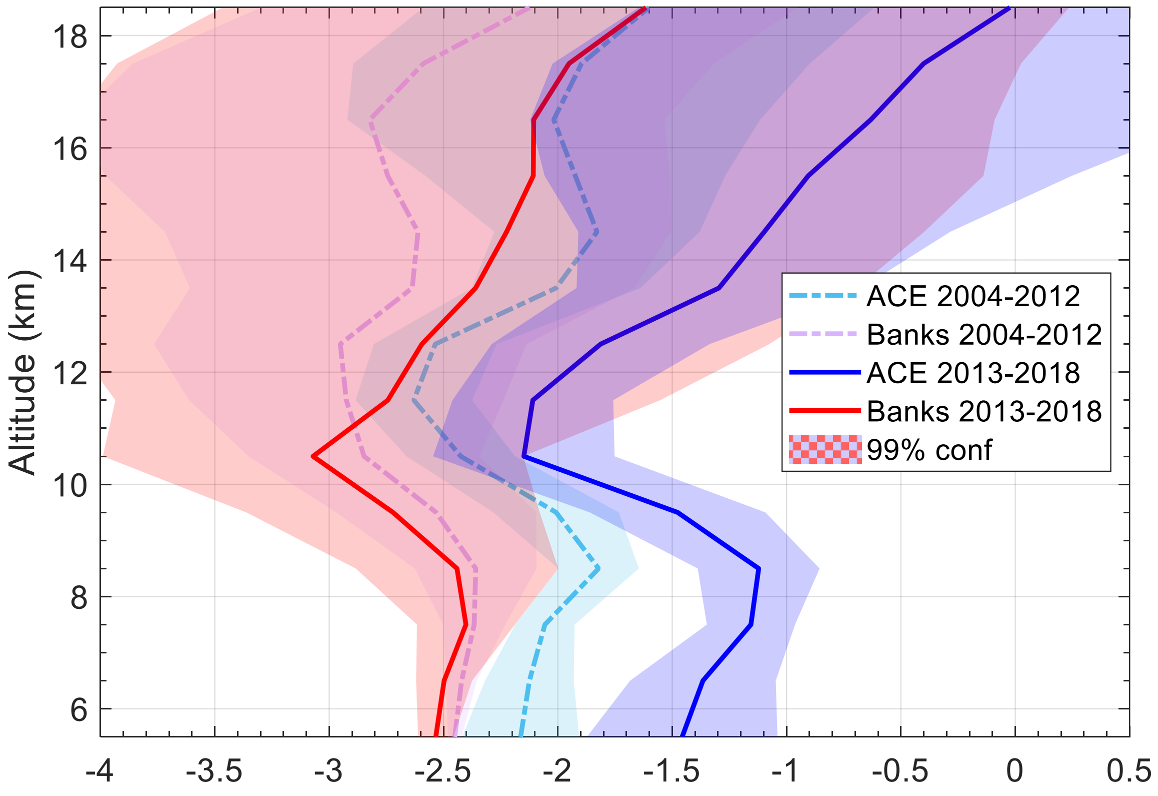


TOMCAT 3-D model values for the Control and Banks runs and their corresponding fits.



ACE-FTS Comparisons with “banks” run

- In the upper troposphere, both 2004-2012 and 2013-2018 show significant differences in trend between what ACE-FTS measures and what the model Banks run simulates
  - In the scenario of only banks emissions, the model suggest a CFC-11 trend on the order of 2.4 pptv/year in upper trop



Summary

- Throughout the UTLS, CFC-11 trend profiles from ACE-FTS and the TOMCAT Control (standard) run (forced by observed surface mixing ratios) agree within the uncertainties for all time periods and locations that were analyzed
- ACE-FTS measured a significant change in global CFC-11 concentration trends between 2004-2012 (-2.0 pptv/year) and 2013-2018 (-1.3 pptv/year) within the altitude range of 6.5-9.5 km. These are similar to corresponding surface trends.
- The 2013-2018 ACE-FTS and Control trends below 10 km were also significantly slower (less negative) than the Banks scenario ("extra" emissions removed) trends (-2.4 pptv/year) for the same time period
- Future work will include analyzing data using dynamic linear modelling in order to examine time dependent trends

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