On the uneven decline of atmospheric CFC-11: bumps in the road to ozone recovery or variations in atmospheric transport and/or loss?

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CFC-11 (CCI₃F) in the atmosphere today:

→ Accounts for 20-25% of ozone-depleting chlorine reaching the stratosphere (second largest contributor)

→ Was the largest contributor to atmospheric Cl declines from 2007-2012

→ Near-zero global production *for all uses* since 2007 (as reported to UNEP's Ozone Secretariat)

But... Significant emissions persist... a large "bank" of chemical remains in existing equipment (~1200 Gg in 2012).

The NOAA CFC-11 atmospheric record



Derived with data from 5 to 12 sites with multiple instruments

The result of a concerted effort by many dedicated people over multiple decades!











The slowdown in the atmospheric decline of CFC-11

 \rightarrow observed by 3 different methods at NOAA**:

- 1) in situ GC-ECDs at 6 sites
- 2) flasks, by GC-ECD, from 12 sites
- 3) flasks, by GC-MS, from 12 sites (since 2009)



**Calibration scale for GCMS is maintained independently from ECD instruments

Derived global emissions vs. reported production

(assuming a constant loss frequency)



Fairly constant and invariant (+/-4%) emissions for over a decade, then a 30% increase.

Deriving emissions from global atmospheric changes:

for G = global mass τ = global lifetime

dG/dt = Emission – Loss

where loss is derived from annually repeating loss frequencies and transport.

...stratospheric loss signals propagate to the troposphere (Nevison *et al.*, 2014)

- How significant are variations in the loss signal?
- Can we avoid aliasing loss variability in derived emissions?

Time-varying anomalies in the strat \rightarrow trop flux of CFC-11

→ Derived from a model of stratospheric circulation with reanalysis meteorology (E. Ray et al., 2014), photolytic fluxes, and trace gas photolysis cross sections:



Transport anomalies should affect other trace gases CFC-11 CFC-113



Conclusions:

- 1) The atmospheric decline of CFC-11 slowed significantly during 2013-2016, and has been slower than expected for over a decade. Total CI decline has significantly slowed too.
- 2) Derived CFC-11 emissions have not decreased since 2005 and <u>appear to</u> <u>have increased by 30%</u> since 2012 (~18 Gg/yr) despite a phase-out of global production in ~2007. Such a large emission increase seems very difficult to understand given past compliance with the Montreal Protocol.
- 3) Inter-annual variations in CFC-11 transport from the stratosphere likely influence the magnitude of emissions derived from remote surface measurements. Although these variations seem unlikely to entirely account for the observed anomalies derived for CFC-11 during 2013-2016,
- 4) Emissions derived from surface observations should be more reliable with an accurate accounting of loss variability.