How reliably can we estimate inter-annual changes in global emissions of long-lived trace gases from atmospheric measurements?

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CFC-11 global emission derived from remote atmospheric measurements

Hourly measurements at 5 sites
Weekly measurement at 12 sites

Emissions derived with simple mass balance considerations (3-box-model analysis):

\[ \frac{dG_{F11}}{dt} = \text{Emission} - k \times G \]

Are these inter-annual changes real?
Is the 2017-2018 difference robust?

Uncertainties (2 to 4 Gg yr\(^{-1}\)) include measurement precision & consistency, atmospheric variability, & an estimate of network representation of the true global surface mean
Uncertainties (2 to 4 Gg yr\(^{-1}\)) don’t explicitly include:

* **calibration consistency:**
  0.1% error in annual mole fraction
  \(\rightarrow\) 5 Gg yr\(^{-1}\) emission error
  - NOAA inter-annual calibration consistency is \(~0.03\%\)
  - Annual global mean variability (NOAA vs AGAGE) is also \(~0.03\%\)

or

* **Variability in atmospheric transport and dynamics**
  particularly between loss region and measurement locations at Earth’s surface
Investigating the influence of variability in dynamics and air transport on derived emissions (e.g., see Ray et al., 2020*)

From the observations,
- derive a smoothed emission history
- use the smoothed emission history as input to:

1) a simple 3-box model
2) two 3-D global models using different meteorology

Then:
Assess measured vs. simulated mole fraction rates of change

Using the smoothed emission history as input:
Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)
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From 3-box model
From 3-D model WACCM using specified dynamics from MERRA2
Using the smoothed emission history as input:
Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)

1) Inter-annual variability in model is similar to what is measured
2) Phasing of variations in model often captures what is measured

From 3-box model
From 3-D model WACCM using specified dynamics from MERRA2
From measurements, 8 sites in NH, 4 sites in SH
Using the smoothed emission history as input:
Simulated hemispheric mean mole fraction rates, CFC-11 (12-month smoothed)

And with a different 3-D model:

Next:
Derive emissions from 3-D model-simulated mole fractions, to estimate dynamics-induced biases on box-model emissions.

From 3-box model
From 3-D chemical transport model TOMCAT with ERA5
From measurements, 8 sites in NH, 4 sites in SH
Dynamics-related biases on inferred CFC-11 emissions obtained from the difference between:

* Smoothed input emissions &
* Emissions derived from 3-D model-simulated mole fractions

From both models:

Inter-annual changes
- typically have the same sign,
- often a similar magnitude:
  - mean inter-annual bias: 5 Gg yr\(^{-1}\),
  - as high as 15 Gg yr\(^{-1}\)
  (compared to 2 - 4 Gg yr\(^{-1}\) uncertainty)

WACCM suggests a significant shift in 2000, reflecting a known perturbation in the stratospheric circulation (Randel et al., 2006)
Inferred global CFC-11 emissions including dynamics-related biases derived from 3-D models

From WACCM

From TOMCAT

→ Smoother emission changes implied after 2010, perhaps to be expected

Pre-2010 variability is sometimes enhanced
→ real?
Enhanced errors in observations or models?
Summary:

Improvements in measurement capabilities (precision, consistency, global coverage) yield uncertainties in derived annual emissions of 2 to 4 Gg yr\(^{-1}\) are implied.

3-D models with reanalysis meteorology suggest that larger biases in year-to-year emission changes can stem from variability in dynamics.

→ some dynamics-related biases can persist for multiple years (post 2000)

Models do a good job of simulating measured interannual variability in mole fraction trends in some years, not all.

Assessing emission changes on a year-to-year basis, (e.g., for rapid feedback to policymakers) requires an accurate estimate of these non-emissive influences on derived global emissions.
NH vs SH rates from 3-D models:

Much of the variability has similar phasing in the two hemispheres

→ variability out of phase less often (N–S exchange?)

→ implying source of variability as begin the BDC or strat-trop exchange (e.g., QBO as in Ray et al., 2020).
Looking at uncertainties: measurement precision at ppt-levels.

→ mean replicate injection precision vs. mole fraction:

![Graph showing replicate injection precision vs. ambient mole fraction (ppt) for various compounds: CH₃I, CH₃CCl₃, CH₂Cl₂, CFC-11, HCFC-22. The graph indicates precision percentages ranging from 0.1% to 10.0%.](image)
Estimating uncertainty in global mean mole fraction from 12 measurement sites:

a) Annual site means ($X_j$) are derived from a random draw of monthly mole fractions given the measured s.d. ($\sigma$).

b) Sites used in estimating a global mean ($G$) and randomly chosen.

c) Multiple network representations give an estimate of $G$ and $\sigma$

Use $G \pm \sigma$ in simple box model to estimate emission uncertainty.

Bootstrap analysis with replacement; Dlugokencky et al., 1994
Looking at uncertainties: atmospheric variability.

**CFC-11**

- Individual sites

**Answer:** ~0.03% at 1 s.d.

**NOAA / AGAGE monthly ratio**

- NOAA: 4 – 5 samples/month
- 8-12 sites
- AGAGE: 300 samples/month
- 5 sites
- 2010-2015

Which is similar to our (NOAA) estimate of inter-annual calibration consistency.

Errors of ± 0.03% → ± 1.5 Gg on annual emission