Relationship of loss, mean age, and distribution of CFCs to model circulation and implications for atmospheric lifetimes

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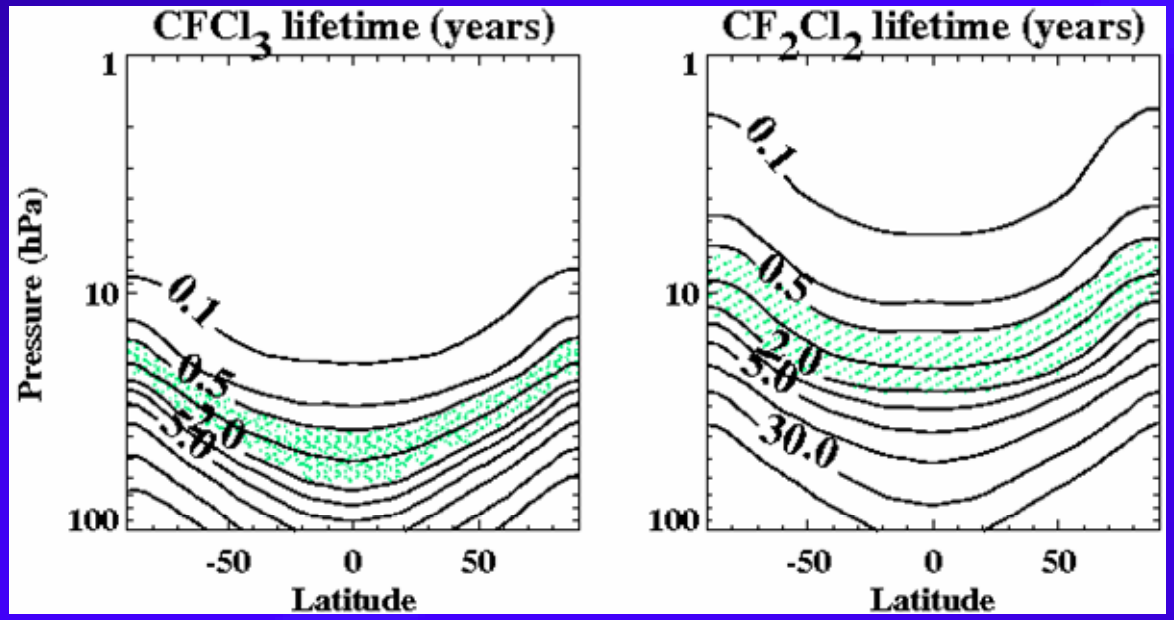
Introduction

- Future boundary conditions are calculated using a presumed atmospheric lifetime.
- Mixing ratio boundary conditions ‘disconnect’ the annual change in atmospheric burden from the input to the atmosphere and the simulated loss
- No evidence that the ‘lifetime’ estimates were changed when models underwent drastic reformulation to produce realistic age of air (c. M&MII, 1999)
- Lifetime also enters when estimating ‘banks’ (i.e., CFCs used in closed

Examine loss, fractional release, age of air and lifetime in several models

MODEL	AGE-of-AIR
• CCM	realistic
• GMI-GCM	realistic
• GMI-DAS	young
• 2D-BASE	realistic
• 2D-FAST	young

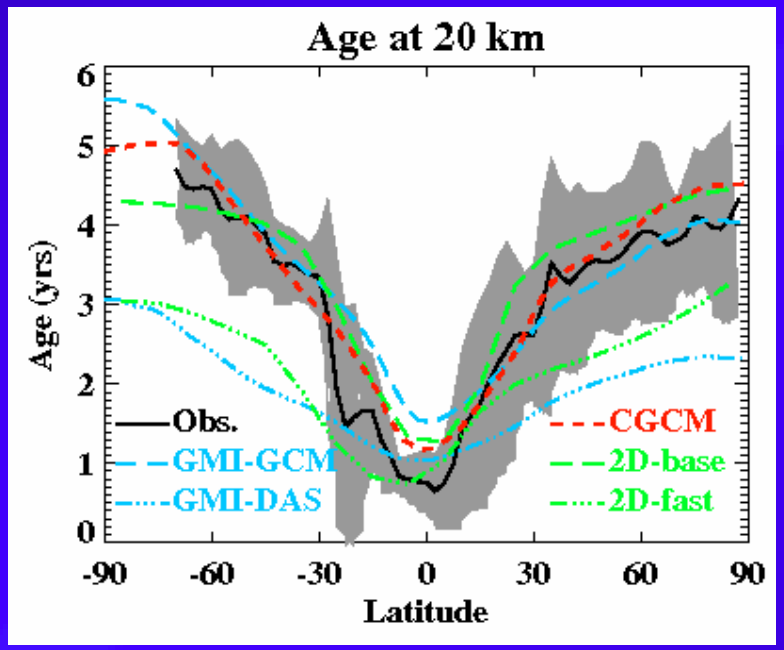
Atmospheric loss is a steep function of altitude



The local lifetime (loss frequency)$^{-1}$ decreases rapidly with altitude; the transition from lifetime greater than 2 years to a few months or less is narrow.

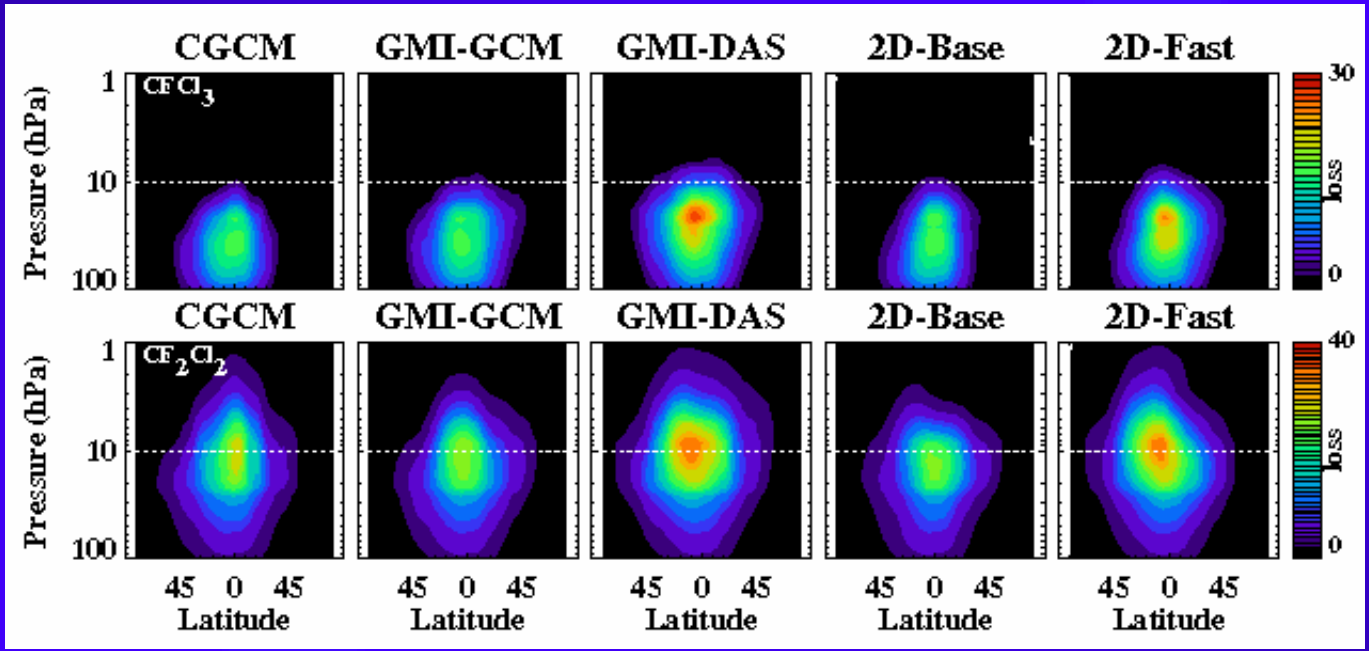
(Parcels that stay in the lower stratosphere (less than 50 hPa) could stay quite a while without experiencing significant loss of CFCs.)

Age distributions look a lot better than they used to . . .



"famous" comparison with age derived from aircraft CO_2 measurements [*Boering et al.*, 1996]. CGCM, 2D-base and GMI-GCM fall w/in 2-sigma limits outside the tropics. GMI-DAS and 2D-fast are typical of results for 1999 M&MII and are 'young'.

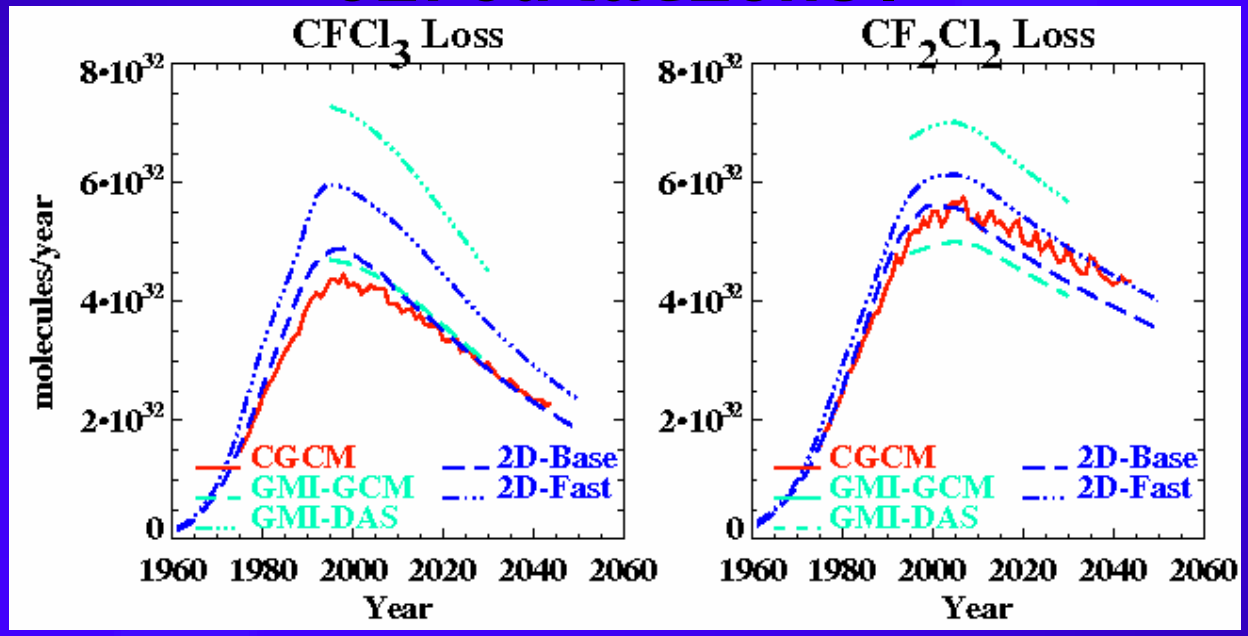
The annual atmospheric loss depends on the circulation



2D-FAST and GMI-DAS have stronger upwelling, leading to higher mixing ratios in the tropics and greater loss.

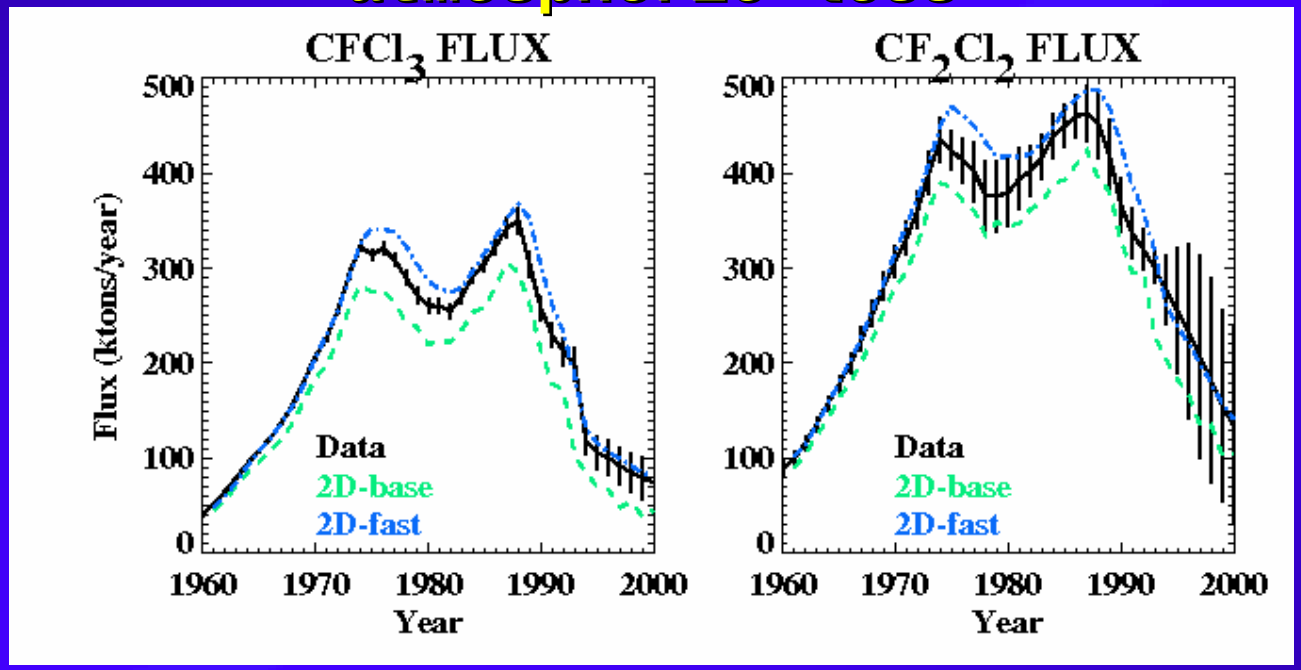
Notice lack of significant loss outside the tropics in general and especially in the lower stratosphere

The annually averaged loss is substantially greater for simulations with faster circulations.



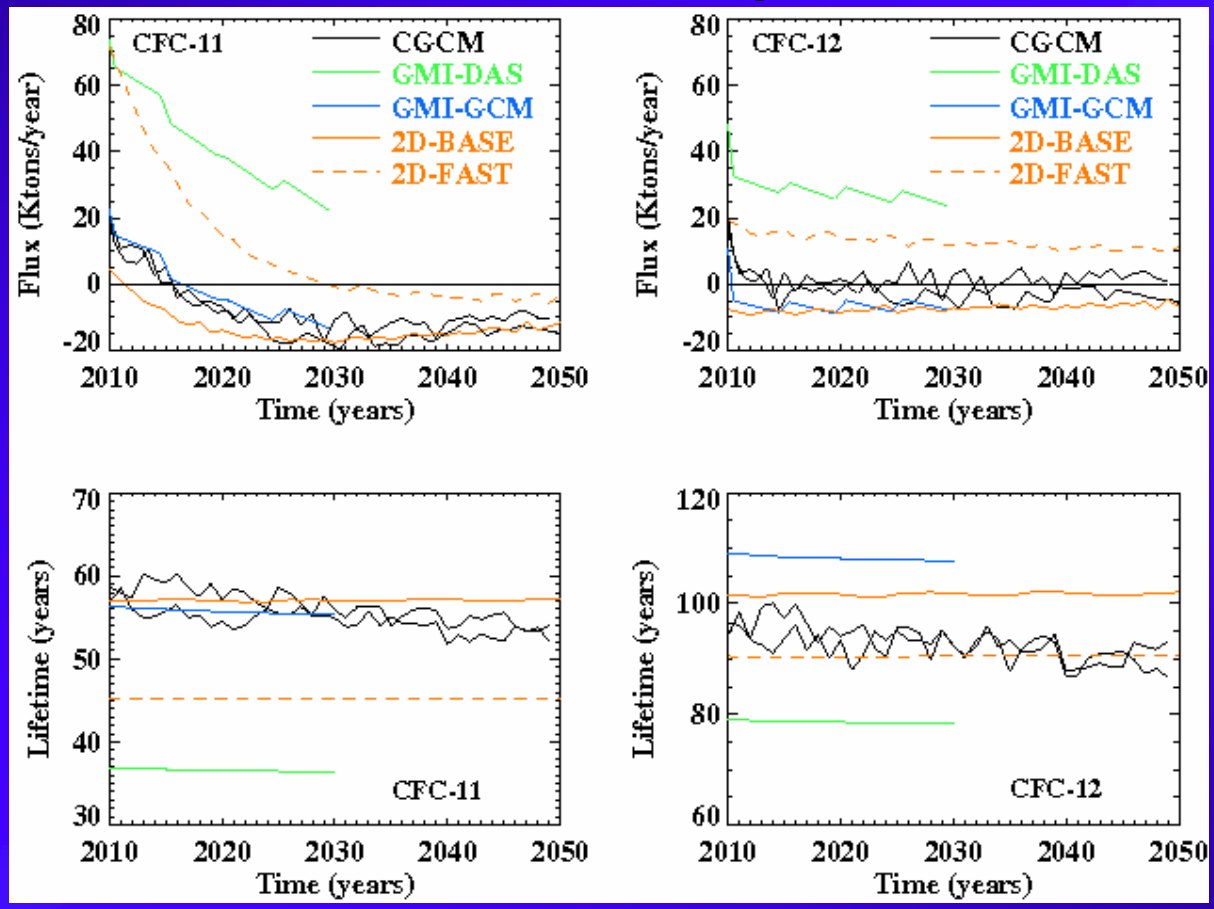
Remember that the BURDEN is controlled by the boundary conditions - time series of 'atmospheric content' of $CFCl_3$ and CF_2Cl_2 are nearly identical for all simulations.

The flux is computed as the amount required to balance a conservation equation given the burden change and atmospheric loss



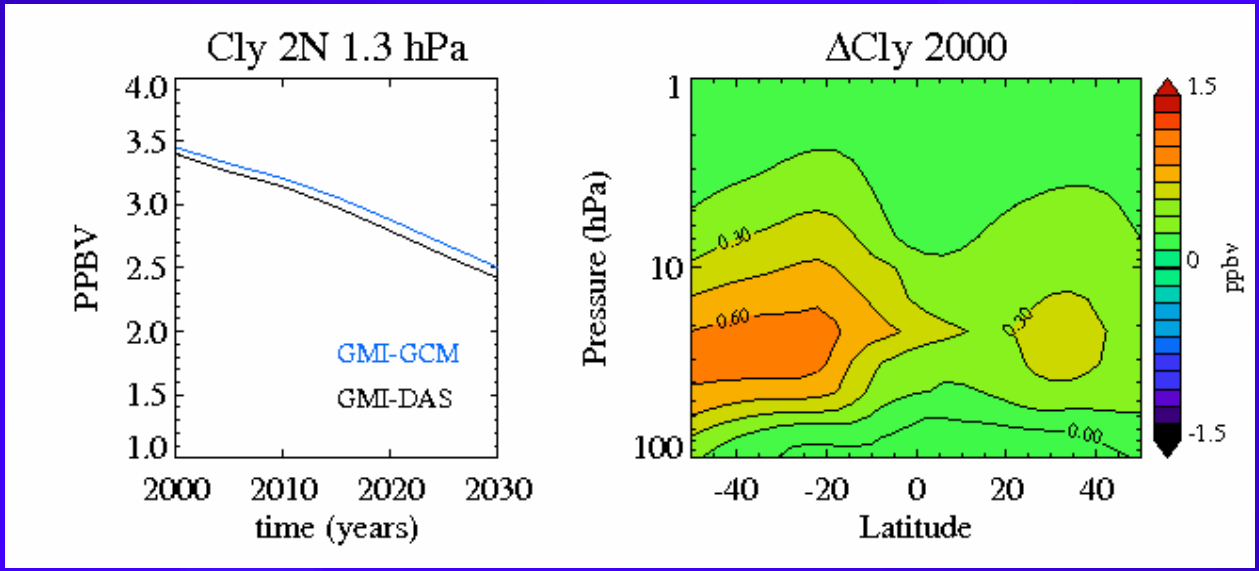
The fluxes for 2D-FAST are substantially larger than those for 2D-Base. For $CFCl_3$ the flux from 2D-base is lower than the estimate of McCullough et al. [2001; 2003]

During later years, the flux is negative because the mixing ratios require more loss than the model produces.



Simulations with 'good' age of air produce a $CFCl_3$ lifetime ~ 56 years rather than 45 years. Discrepancies of CF_2Cl_2 are smaller.

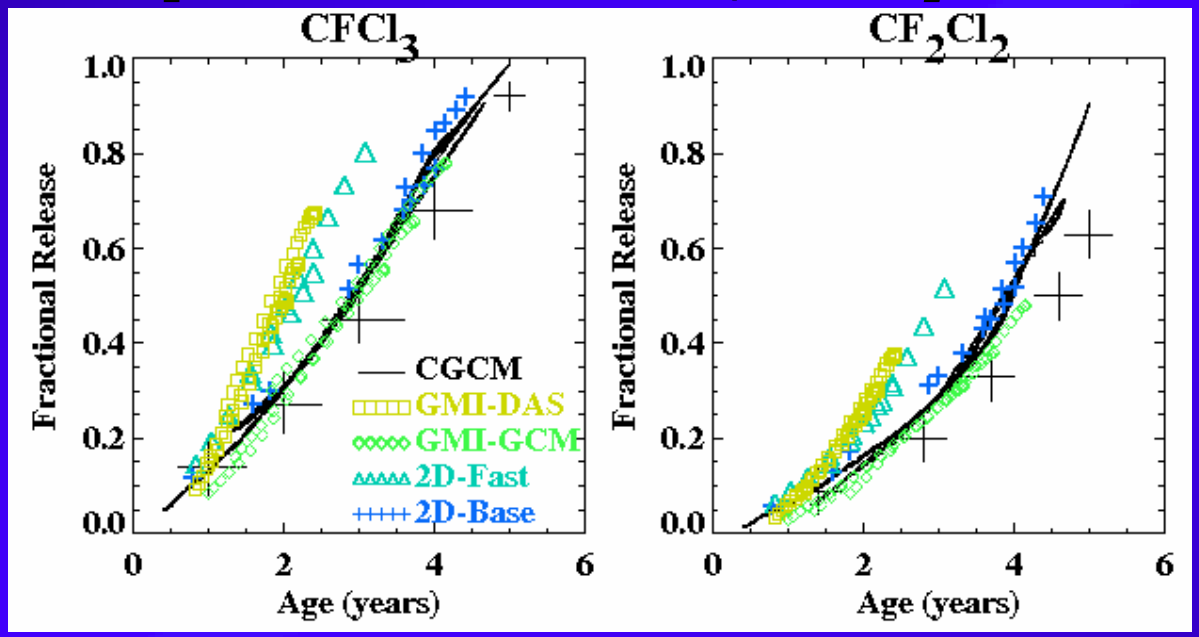
Upper stratospheric Cly doesn't care about the circulation and the loss or the flux



But Cly from GMI-GCM is greater than that from GMI-DAS by as much as 0.6 ppb

We compare the fractional release at 56 hPa with values derived from aircraft observations during SOLVE

[Schauffler et al., 2003]

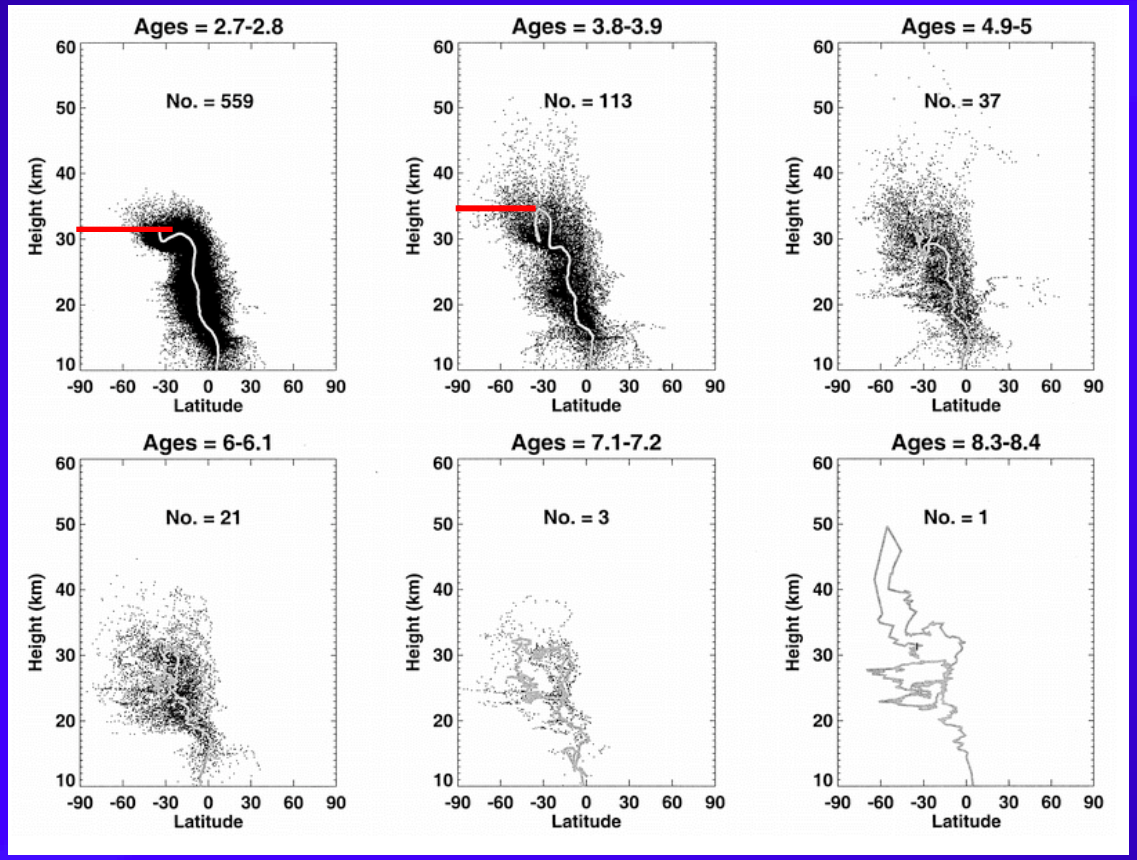


Fractional Release = $(1 - X(\phi,\theta,t)/X_{entry})$

KEY POINT: All simulations produce 'compact relationships'; those with 'realistic' age of air produce values close to observed.

Parcel age can be related to an average parcel height under an average path approximation.

CFC-12 lifetime decreases from > 1 yr to < 0.5 yr

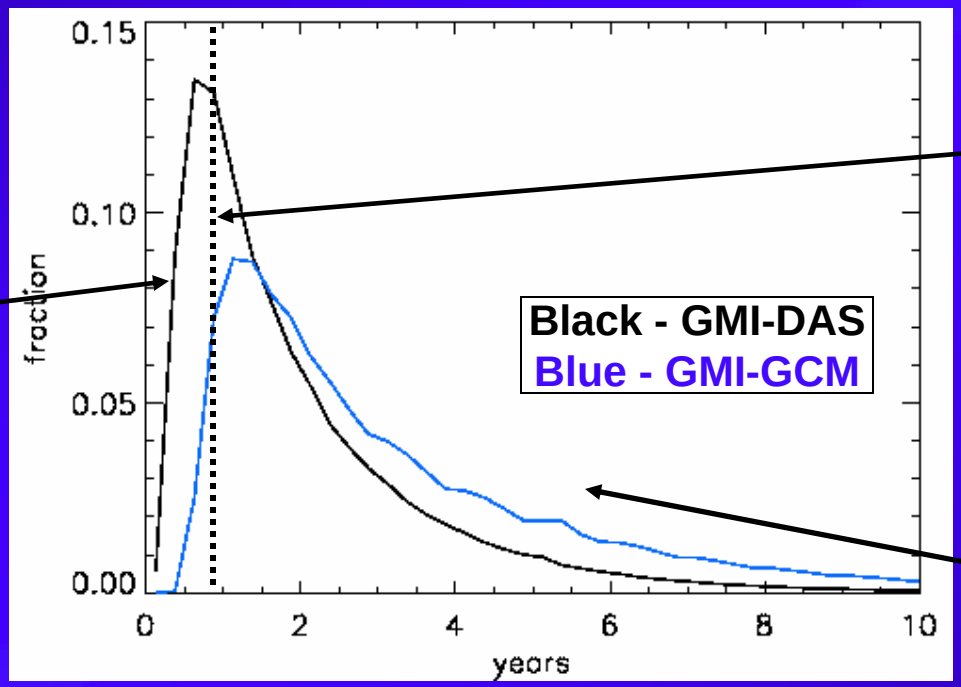


On average, older parcels have been to higher altitudes and experienced more loss. Parcels in the lower stratosphere with significantly less CFC than at entry (calculated using mean age) must have elements that experienced higher altitudes.

Schoeberl et al., A Lagrangian view of stratospheric trace gas distr

The age spectra from the GMI simulations clarify the relationship between fractional release and mean age

Many more GMI-DAS parcels have 'young' ages (and lower maximum heights) therefore the mixing ratio is greater (and the fractional release is smaller)



Parcels with ages younger than this never experience high altitudes and have w_i close to 1.

Old age, high maximum heights, total CFC destruction, w_i=0

Mean age = Σp_i

Mixing ratio = $\Sigma w_i p_i$ where w_i are related to maximum height under the 'average path' approximation.

Conclusions

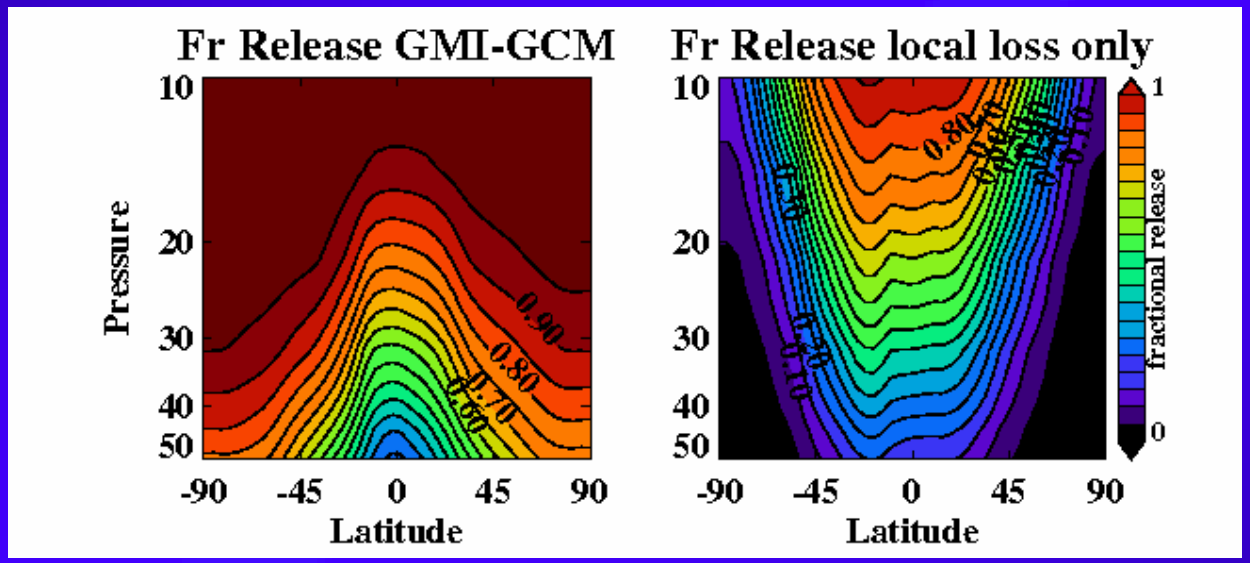
- Comparisons with data support the ‘slower’ circulations
  - Fractional release
  - Mean age
  - Tracers as used by Schoeberl et al. [2003]
- These slower circulations are consistent with a longer lifetime for $CFCl_3$.
- Fractional Release in particular supports the longer lifetime
- A change in lifetime of this magnitude suggests an increase in the ‘banks’ of CFCs similar to the discrepancy between the WMO (top down)* and IPCC (bottom up)** estimates.

*Top down - accumulated difference between estimated production and emission observed from atmospheric observations using a model and an assumed lifetime

**Bottom up - detailed analysis of applications

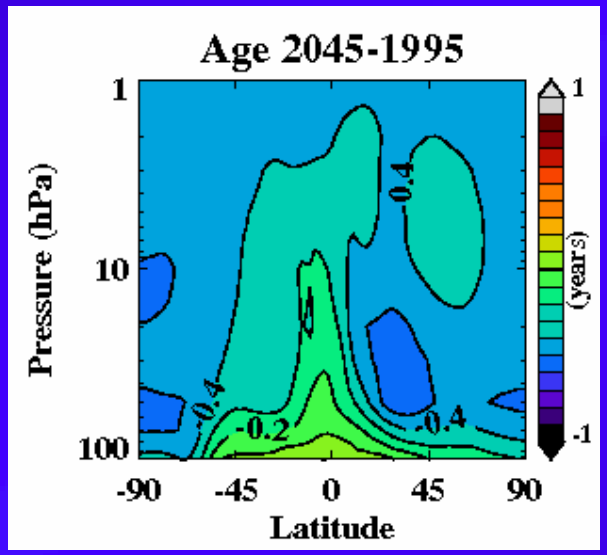
BACKUP

The fractional release would look nothing like observations if the only loss were local loss!



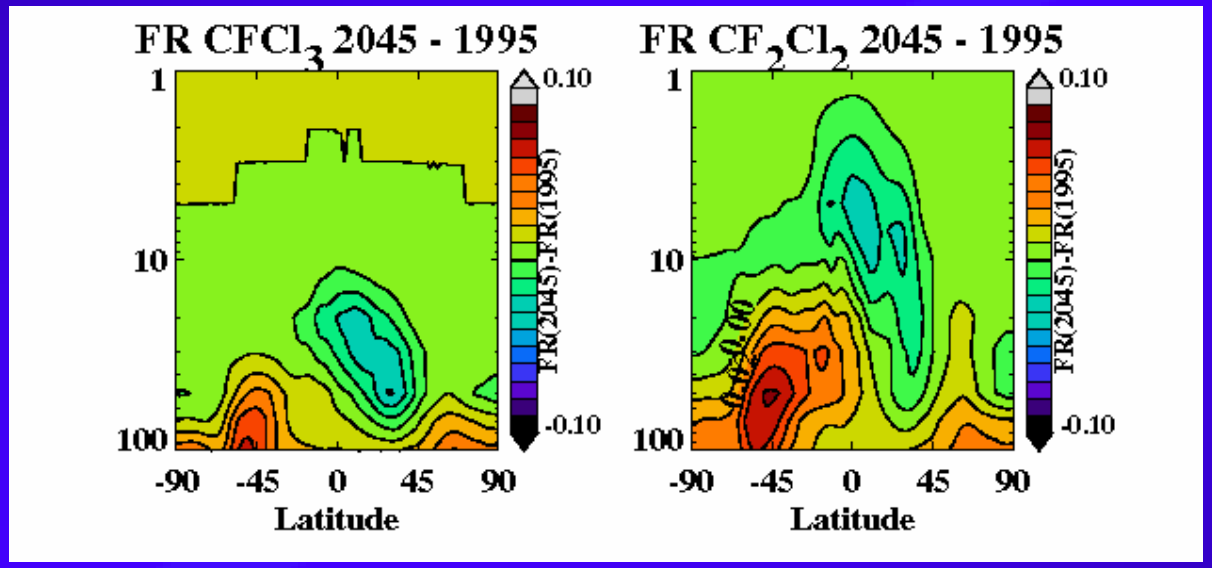
Fractional Release* = $(1 - X_{entry}\, e^{-LT}/X_{entry})$ where L is the local loss rate and T is the mean age

The mean age decreases in CGCM in response to climate change, a common feature of this sort of model.



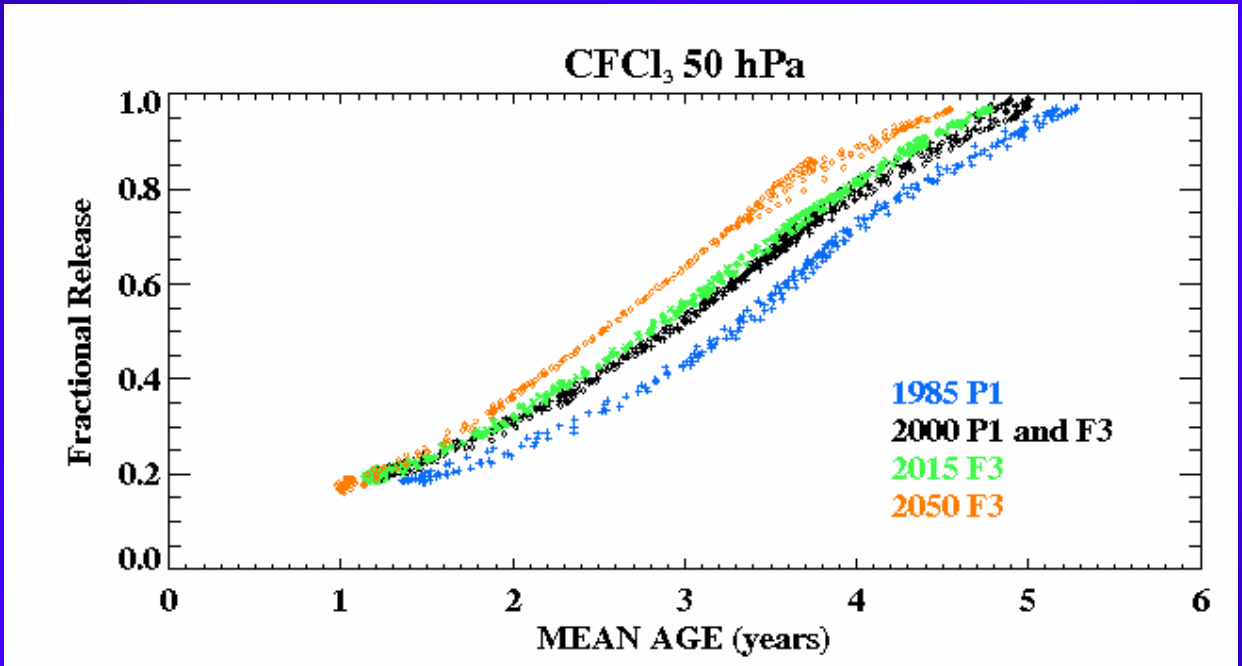
The mean age decreases throughout the model domain, with the largest changes at middle latitudes in the lower stratosphere.

The fractional release changes as the circulation speeds up, but changes are small compared to the change in mean age.



- Fractional release decreases in the tropics
- Fractional release increases in the lower stratosphere middle latitudes
- Mean age decreases everywhere

The compact relationships shift!



The mean age, the age spectrum, and the relationship of various elements in the age spectrum to the maximum height all change as the climate changes. (of course we don't know if this is actually happening ...)