

Review of: **Observation-based assessment of stratospheric fractional release, lifetimes, and Ozone Depletion Potentials of ten important source gases**, by Laube et al. (ACPD 28525-28557, 2012)

General remarks:

This paper uses aircraft measurements of chlorine and bromine halocarbons in the mid- and high-latitude stratosphere to derive Fractional Release Factors (FRFs), stratospheric lifetimes relative to CFC-11, and Ozone Depletion Potentials (ODPs). All of these quantities are critically important to understanding stratospheric ozone depletion and for predicting the evolution of any future ozone recovery. The study presents an impressive amount of information derived from measurements of the ten source gases – CFC-11, CFC-12, CFC-113, CCl₄, CH₃CCl₃, HCFC-22, HCFC-141b, HCFC-142b, H1211, and H-1301 - plus SF₆ used for stratospheric mean ages. It will be an important contribution to the growing body of knowledge on this topic and should be published in ACP, provided that the concerns noted below can be addressed. Overall the paper is well written, with a detailed account of methodology and a clear presentation of results in tables and figures.

Specific comments/suggestions:

1. Age of air. There is a very good discussion of the overall data analysis, quality checks, and specific numbers for the amount of useful measurements, number of excluded measurements, etc., but the fraction of measurements with insufficient precision for SF₆ was not clear (I estimated about half based on all of Ufra data plus all 2011 data). If it is a significant fraction of the data, then the use of a 2010 high-latitude correlation between age of air and CFC-12 could be explained further. Although a similar methodology was used in a previous paper, the correlation from that paper was based on tropical measurements and it was used for deriving tropical FRFs, whereas the correlation in the present paper is for high latitudes and it is used for mid latitude FRFs. Clearly the age-CFC-12 correlation is different between the tropics and extra-tropics but it is not so clear that it would be the same for mid and high latitudes. Since there is a discussion on pp. 9-10 about FRF differences between mid and high latitudes, one might wonder whether eq (1) would be appropriate to use for mid-latitude data.

It would be desirable to show the age-CFC-12 correlation plot in the Supplement and include more discussion of the possible impact on FRFs in the text. One reason why this is important is because eq (1) could play a large role in the FRF vs. age relationship for CFC-12. For those data where CFC-12 is used to infer age, the CFC-12 FRF is based entirely on measured CFC-12 since the same measurement is used directly for $X(x,y,z,t)$ and indirectly for X_{entry} . When the CFC-12 FRF is plotted vs. Γ , those data points using CFC-12 for age appear to be just expressing some form of the relationship set by eq (1).

There seems to be an inconsistency between equation (1), the slope in Figure 1, and the correlation slope in Figure S5. It may not be that significant to the overall results, though it

should be looked into. If eq (1) is used to evaluate $d\Gamma/dX_{12}$ at $X_{12} = 540$ ppt, it would be -0.03482 yr/ppt. Combined with the result $dX_{11}/d\Gamma = -20.7$ ppt/yr at $X_{11}=241$ ppt from Figure 1, one would find

$$\frac{dX_{12}}{dX_{11}} = \frac{\frac{dX_{12}}{d\Gamma}}{\frac{dX_{11}}{d\Gamma}} = 1.387$$

The value of dX_{12}/dX_{11} based on the correlation slope in Figure S5 is 1.339, so it is only a few percent smaller. Perhaps is this because eq (1) is based on a subset of the data whereas Figure S5 includes all of the data, but it would be worth checking to make sure that eq (1) is consistent with the rest of the analysis.

2. Figure 1, showing the correlation of CFC-11 mixing ratios and mean age of air, shows an interesting pattern. It is not immediately obvious from the raw data that the slope is changing by all that much, in order to extrapolate a slope at age=0 yr. The linear relationship between age and slope as shown in the figure (intercept=-20.7 ppt/yr, slope=-6.9 ppt/yr²) implies a quadratic polynomial between X_{11} and Γ ,

$$X_{11} = 241 - 20.7\Gamma - 3.45\Gamma^2$$

and it might be useful to overplot such a curve on the figure, in order to show that there is sufficient curvature to justify a linear change in slope.

There is a question of why there is such a sharp break where the slope is approximately constant for $\Gamma > 2.5$ yr. If there is a physical explanation then it would be desirable to include a discussion of this behavior. Otherwise, how can one be sure that there is not another sharp break where the slope is constant for $\Gamma < 0.7$ yr, so that the intercept should be -25 ppt/yr rather than -20.7 ppt/yr?

3. The correlation slopes of the mixing ratios versus CFC-11 that are presented in the Supplement, which are fit to polynomials and extrapolated to the slope at the tropopause, also show interesting behavior. Most (but not all) show an upward concavity. If there a physical explanation for why some gases have this curvature, then it might be useful to include a discussion since this curvature does seem to be important for extrapolating the slope at the tropopause.

For example, Volk et al 1994 identified two key factors contributing to curvature: tropospheric growth and fluxes across the tropical/mid-latitude barrier (the "leaky pipe"). Can anything be updated from that discussion using this newer data? The curvature here for CFC-12 vs. CFC-11 appears to be a little larger than seen in the 1994 data from the Volk et al paper, even though effective linear growth rates for the 2010 data are negative as opposite to positive for 1994, and the growth magnitude for CFC-12 is now much smaller than what is was in 1994. This might

imply that the leaky pipe has more of an influence on curvature than growth, or is there another conclusion that could be drawn?

Finally, it seems that the extrapolation will be sensitive to the behavior of the correlation slope for CFC-11 mixing ratios less than 170 ppt, and yet this region is not considered when assessing the slope of CFC-11 vs. age in Figure 1 (see comment 2 above). For example if data for CFC-11 < 170 ppt were excluded from the fit in Figures S4 and S5, then the extrapolated slopes and lifetimes for CFC-113 and CFC-12 could be different. On the other hand, if data for CFC-11 < 170 were included in a quadratic fit to the slope in Figure 1, consistent with the way the other correlations are treated, then the derived CFC-11 vs. age slope, which is important for all lifetimes, would probably be different. Either way there should be a consistent treatment of fitting and data ranges, or sound reasons given for changing things up.

Minor suggestions/corrections:

p2 line 3: “we calculate lower ODPs than recommended by WMO”. Otherwise the implication is that WMO calculated the ODPs.

p2 line 25 “study” should be “studies”

p3 line 15 “GCMS” (no hyphen)

p4 line 13 Levin and Hesshaimer (1996) is not in the ref list

p5 line 23 I suggest adding mean in this sentence: “We then calculated the mean mixing ratios...” and similarly suggest adding an overbar to $X_{i,entry}$ in eq (2) to make it clear this is a mean entry mixing ratio

p6 eq (4) It might be useful to remind the reader after this equation that $\Lambda = \Delta^2 / \Gamma$, rather than waiting until halfway down the next page (lines 18-20). Also, it is mentioned that three different values of Λ were used for lifetimes. I could not find where it was discussed how this impacted the lifetime values (I suspect not very much), though it should be noted that the value of 1.25 used for the best estimate lifetime is different from the value of 0.7 that is used for the FRFs. It seems odd to use different values of a supposedly invariant quantity in the same paper.

p7 lines 3-10: It was not clear to me why expanding the slope expression into two terms offers an advantage. Even though CFC-11 has one of the best precisions in the dataset and the best-constrained slope against mean age, it would seem that the addition of any random error from CFC-11 should make the expansion of the slope expression less accurate than a direct correlation of that source gas with age. On simple terms, one might expect that if there is a good correlation of any tracer with CFC-11, then there ought to be a reasonable correlation of that tracer with mean age.

p10 line 10 Is it that the FRFs are too small, or that the uncertainties are too large for a meaningful comparison? On line 11, “differences with Newman et al”

p11 line 10. Statement is unclear. I interpreted Engel et al (2009) to have observed no change in mean age.

p12 line 7 “differences with the correlations of”

p12 lines 16-17 may be an extra “CFC” in this sentence

p13 lines 1-3 This is a really interesting discussion. There should be some comment on the implications for stratospheric chemistry as well, since presumably there is a larger perturbation to stratospheric OH implied by the CH₃CCl₃ lifetime. There is a small but caring community that still finds stratospheric OH interesting!

p14 line 18 Grammar “for; here: 60”

p15 line 1 Why choose mean age of 3 years to evaluate FRF? I thought WMO used 3.5 years as recommended by Schauffler et al 2003. Similarly for lines 25-26 on p. 15

p15 line 21 Suggest adding “as our H-1301 FRF” to make it clear. Also, might add “and our $\tau(1301)/\tau(F11)$ ” since this ratio also is involved in setting the ODP.

p. 24 Table 1 header “Differences with NOAA-ESRL”