

Supplementary Material for
Evaluation of stratospheric age-of-air from CF₄, C₂F₆, C₃F₈, CHF₃, HFC-125, HFC-227ea and SF₆; implications for the calculations of halocarbon lifetimes, fractional release factors and ozone depletion potentials

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S1. A comparison of UEA and NOAA SF₆ time series

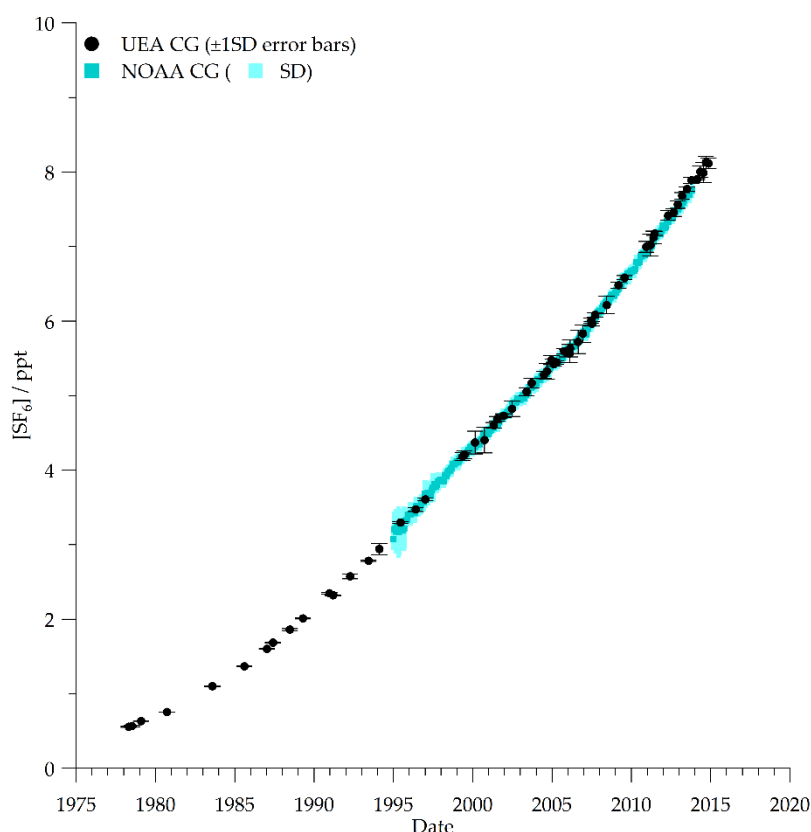


Figure S1. A comparison of Cape Grim (CG) time series data from UEA and NOAA (see inset legend) taken from flask measurements. The UEA CG time series analytical uncertainties are outlined in Section 2 of the main manuscript. The NOAA CG time series is a monthly average of approximately weekly flask samples with one standard deviation (SD) errors shown as the lighter shaded area. UEA SF₆ data is reported on the NOAA calibration scale. NOAA data was retrieved from the ‘Combined Sulfur hexafluoride data from the NOAA/ESRL Global Monitoring Division’ file on the NOAA server on 12th July 2017 and is accessible here: <https://www.esrl.noaa.gov/gmd/hats/combined/SF6.html>.

S2. Mean age uncertainty analysis – a summary

Table S1 provides an overview of all the tests performed as part of the mean age uncertainty analysis described in Section 3 of the main manuscript.

An introduction to our terminology used in Table S1:

- ‘Raw’: UEA’s Cape Grim time series used as is (grey points in main manuscript Fig. 1)
 - ‘Raw_min’ and ‘raw_max’ tropospheric trends derived from the mean mixing ratio values \pm the analytical uncertainties outlined in Section 2.
- ‘Fit’ is the fit-interpolated dataset derived from UEA’s Cape Grim time series (blue line in main manuscript Fig. 1)
 - ‘Fit_min’ and ‘fit_max’ are derived from the fit \pm the uncertainties from the bootstrapping procedure outlined in Section 3a.
- ‘NOAA’: UEA stratospheric data used with a tropospheric trend derived from the NOAA SF₆ time series available from <https://www.esrl.noaa.gov/gmd/hats/combined/SF6.html>
- ‘SIO’: mean ages calculated using an independent analysis of the stratospheric samples performed by the Scripps Institution of Oceanography, SIO and tropospheric trends provided by SIO.
 - ‘SIO_min’ and ‘SIO_max’: as for ‘raw_min’ and ‘raw_max’ but derived from the SIO trends \pm their analytical uncertainties.
- MRs = mixing ratio
- The difference between ‘quadratic’ and ‘convolution’ AoA routines is described in the main manuscript Section 3c.
- The ‘base case’ column shows the scenario we compared the ages in question to when calculating the mean age difference, e.g. the differences shown in the residual plots (Fig. 2 and SI-3).

Note: Where ‘symmetrical’ tests were performed, namely ‘min’ and ‘max’ analyses where the same uncertainty was either added or subtracted, the average of both of these results combined is presented to simplify the table.

Table S1. A summary of all tests performed as part of the uncertainty analysis described in Section 3 of the main manuscript. Abbreviations and other terminology described on previous page.

| Experimenting with... | Case # | Tropospheric trend input | Stratospheric data | AoA routine | Parameterisation of width of age spectrum | Average absolute difference / months | | | | | | | | Manuscript section |
|---|--------|--------------------------|--------------------|-------------|---|--------------------------------------|-----------------|-------------------------------|-------------------------------|------------------|---------|-----------|-----------------|------------------------|
| | | | | | | Base case | SF ₆ | C ₂ F ₆ | C ₃ F ₈ | CHF ₃ | HFC-125 | HFC-227ea | CF ₄ | |
| Tropospheric trend inputs | 1 | Fit | Mean MRs | Quadratic | 0.7 | | | | | | | | | Base case |
| | 2 | Raw | Mean MRs | Quadratic | 0.7 | 1 | 0.5 | 2.2 | 0.8 | 0.9 | 1.1 | 1.8 | | - |
| | 3 | NOAA | Mean MRs | Quadratic | 0.7 | 1 | 1.9 | | | | | | | - |
| | 4 | Fit_min | Mean MRs | Quadratic | 0.7 | 1 | 1.1 | 1.8 | 2.5 | 1.5 | 0.6 | 2.4 | | 3a |
| | 5 | Fit_max | Mean MRs | Quadratic | 0.7 | 1 | | | | | | | | |
| | 6 | Raw_min | Mean MRs | Quadratic | 0.7 | 2 | 3.2 | 3.8 | 2.2 | 4.8 | 0.9 | 3.2 | | - |
| | 7 | Raw_max | Mean MRs | Quadratic | 0.7 | 2 | | | | | | | | |
| Stratospheric sample inputs | 8 | Fit | Min MRs | Quadratic | 0.7 | 1 | 2.5 | 5.8 | 3.2 | 4.5 | 0.6 | 2.9 | | 3b |
| | 9 | Fit | Max MRs | Quadratic | 0.7 | 1 | | | | | | | | |
| | 10 | Raw | Min MRs | Quadratic | 0.7 | 2 | 2.5 | 5.9 | 3.3 | 4.5 | 0.7 | 2.7 | | - |
| | 11 | Raw | Max MRs | Quadratic | 0.7 | 2 | | | | | | | | |
| Parameterisation of width of age spectrum | 12 | Fit | Mean MRs | Quadratic | 0.5 | 1 | 0.2 | 0.7 | 0.7 | 0.2 | 0.6 | 0.5 | | 3d |
| | 13 | Fit | Mean MRs | Quadratic | 1 | 1 | 0.3 | 0.8 | 0.7 | 0.4 | 0.4 | 0.4 | | 3d |
| | 14 | Raw | Mean MRs | Quadratic | 0.5 | 2 | | 0.7 | 0.8 | 0.4 | 0.9 | 0.9 | | - |
| | 15 | Raw | Mean MRs | Quadratic | 1 | 2 | | 0.8 | 0.8 | 0.3 | 0.8 | 0.8 | | - |
| | 16 | NOAA | Mean MRs | Quadratic | 0.5 | 3 | 0.3 | | | | | | | - |
| | 17 | NOAA | Mean MRs | Quadratic | 1 | 3 | 0.5 | | | | | | | - |
| AoA routine (samples >1 yr mean age only) | 18 | Fit | Mean MRs | Convolution | 0.7 | 1 | 0.2 | 0.7 | 1.2 | 0.1 | 0.6 | 0.2 | | 3c |
| | 19 | Raw | Mean MRs | Convolution | 0.7 | 2 | 1.1 | 3.4 | 1.4 | 2.3 | 2.9 | 10.4 | | - |
| | 20 | NOAA | Mean MRs | Convolution | 0.7 | 3 | 0.4 | | | | | | | - |
| Tropospheric trend inputs using the 'convolution' AoA routine | 21 | Fit_min | Mean MRs | Convolution | 0.7 | 18 | 2.8 | 1.8 | 2.2 | 3.2 | 0.6 | 2.2 | | - |
| | 22 | Fit_max | Mean MRs | Convolution | 0.7 | 18 | | | | | | | | |
| | 23 | Raw_min | Mean MRs | Convolution | 0.7 | 19 | 3.4 | 3.6 | 3.1 | 4.2 | 0.9 | 13.3 | | - |
| | 27 | Raw_max | Mean MRs | Convolution | 0.7 | 19 | | | | | | | | |
| Stratospheric sample inputs using the 'convolution' AoA routine | 28 | Fit | Min MRs | Convolution | 0.7 | 18 | 2.4 | 5.9 | 2.5 | 4.3 | 0.6 | 2.8 | | - |
| | 29 | Fit | Max MRs | Convolution | 0.7 | 18 | | | | | | | | |
| | 30 | Raw | Min MRs | Convolution | 0.7 | 19 | 2.4 | 6.5 | 2.8 | 4.2 | 0.6 | 4.8 | | - |
| | 31 | Raw | Max MRs | Convolution | 0.7 | 19 | | | | | | | | |
| Independent verification using SIO data | 32 | SIO | Mean SIO | Quadratic | 0.7 | | | | | | | | | Base case for SIO data |
| | 33 | SIO | Min SIO | Quadratic | 0.7 | 32 | 2.8 | 11.1 | | | | | 4.2 | 3b |
| | 34 | SIO | Max SIO | Quadratic | 0.7 | 32 | | | | | | | | |
| | 35 | SIO_min | Mean SIO | Quadratic | 0.7 | 32 | 2.6 | 4.2 | | | | | 2.1 | 3a |
| | 36 | SIO_max | Mean SIO | Quadratic | 0.7 | 32 | | | | | | | | |

S3. Residual plots showing the uncertainties associated with Sections 3c and 3d of the main manuscript

‘Residual plots’ are plots showing the difference between the mean age derived from the base case and those derived from variations on this used to test the impact of measurement (and other) uncertainties on derived mean ages.

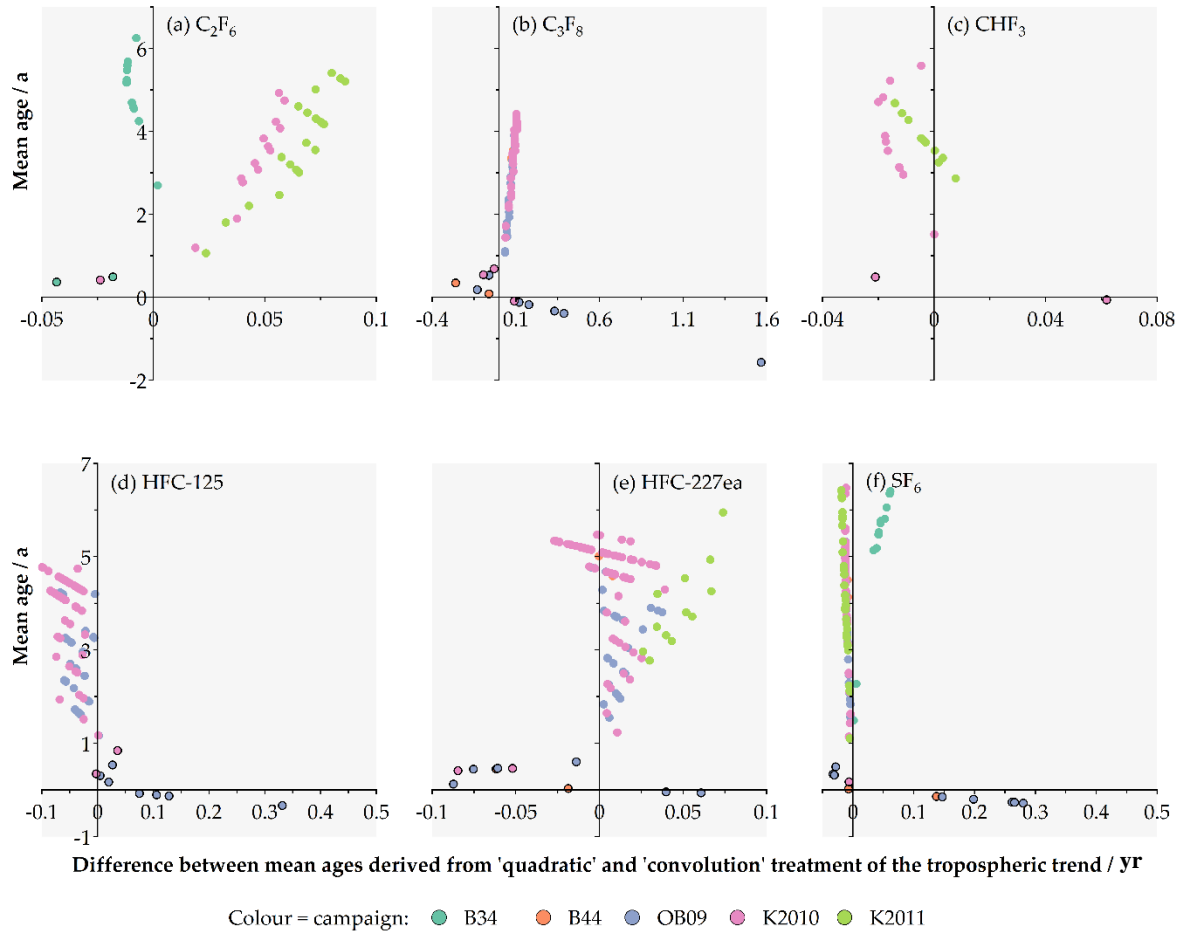


Figure S2. Residual plots showing the uncertainties associated with varying the treatment of the tropospheric trend within the age of air routine. The base case uses the standard quadratic fit and this is compared to a convolution approach. See main manuscript Section 3c for full details. The x-axis shows the difference between these two approaches, plotted against the mean age from the quadratic approach. Marker colour denotes the stratospheric campaign, see inset legend. Values for mean ages less than 1 year were removed from further analysis, see details in Section 3d, these are highlighted by a black outline in this figure. Vertical axis labels for each row are in the left panel.

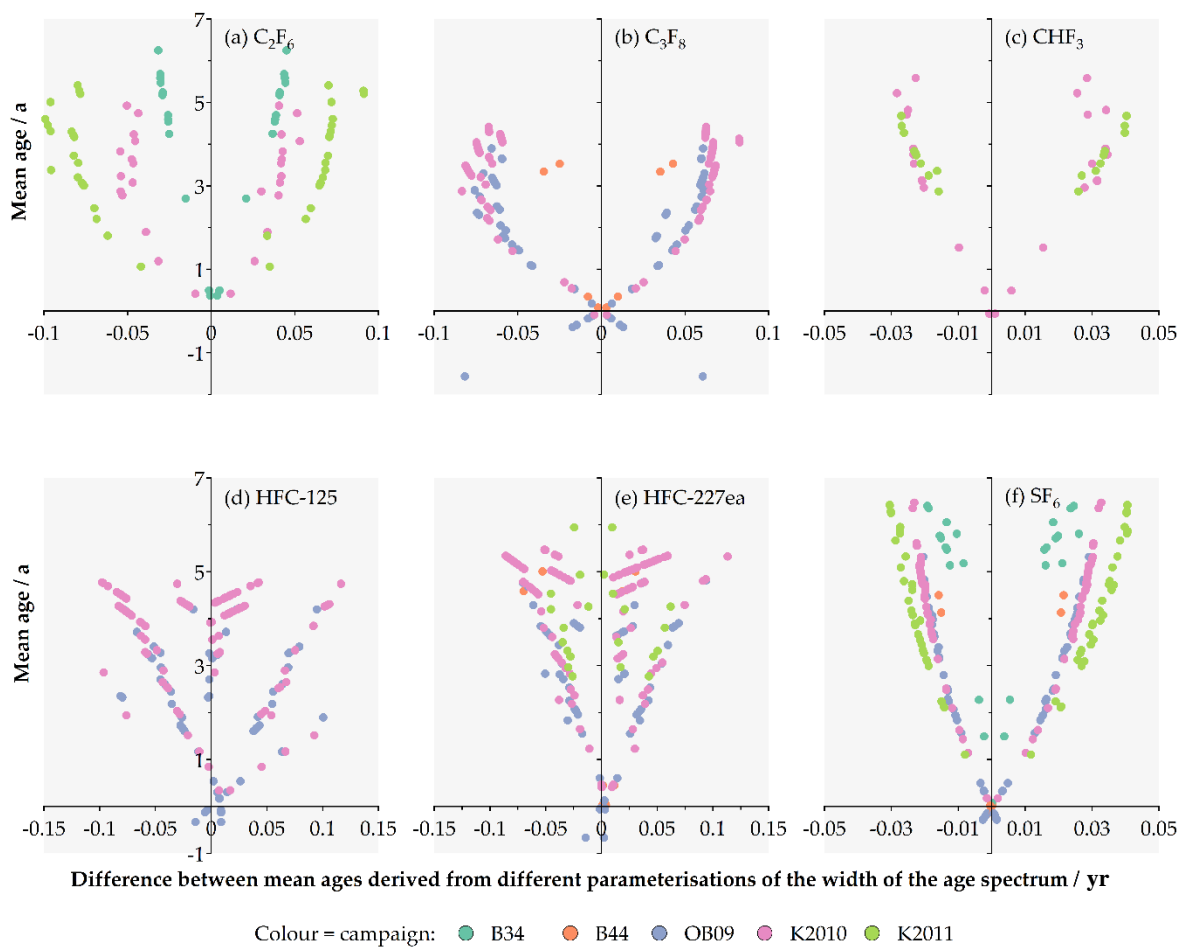


Figure S3. Residual plots showing the uncertainties associated with varying the parameterisation of the width of the age spectrum, see main manuscript Section 3d for further details. The x-axis shows the difference between the base case (width = 0.7) and the variants (widths = 0.5 and 1), plotted against the mean age derived from the base case. Marker colour denotes the stratospheric campaign, see inset legend. Vertical axis labels for each row are in the left panel.