

Supplement S1: Global Box Model

To investigate the overall impact of changes in temperature, [OH] and emissions on an atmospheric gas which is removed by OH a simple global box model can be used. This model permits a first-order understanding of the factors which govern the variation in growth and the relative contribution of emissions and loss to that growth. The model integrates the global mean burden of X (Tg) based on annual mean emissions (E , Tg/yr) and chemical loss (L , Tg/yr) through the reaction $X + OH \rightarrow \text{products}$. The modelled atmospheric burden of X (X_t) can be integrated over a 1-year period (Δt) according to the equation:

$$\frac{1}{\Delta t}(X_{t+\Delta t} - X_t) = E - L = E - k[OH][X] \quad (1)$$

where k ($\text{cm}^3 \text{ molecule}^{-1} \text{ yr}^{-1}$) is the rate constant for the $X + OH$ reaction (e.g. Sander et al., 2011). For CH_4 $k = 2.45 \times 10^{-12} \exp(-1775/T)$ while for CH_3CCl_3 $k = 1.64 \times 10^{-12} \exp(-1520/T)$.

When simulating CH_3CCl_3 we assume emissions from Montzka et al. (2011) or Rigby et al. (2013) and $T=272.9$ K. We then use equation (1) to derive the global mean [OH] which is consistent with the observed variations in CH_3CCl_3 . The same procedure is used to derive global mean [OH] consistent with the observed CH_4 variations assuming $E=553$ Tg/yr.