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 \(\Delta t\) according to the equation:

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

where \(k\) (cm\(^3\) molecule\(^{-1}\) 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.