

2.2.4 Connection between ε_{eff} and the solubility distribution of the mixture

Let us now assume that the dissolved fraction at the point of activation for each considered mixture can be expressed as a sum of two terms, the contribution from the compounds below a threshold solubility bin i_t and the contribution from the compounds over the threshold:

$$\varepsilon = \frac{\sum_{i=1}^{i_t} m_{i,aqueous} + \sum_{j=i_t+1}^n m_{j,aqueous}}{m_{dry}} = \frac{\sum_{i=1}^{i_t} m_{i,aqueous} + \left(\sum_{j=i_t+1}^n m_{j,dry} - \sum_{j=i_t+1}^n m_{j,insoluble} \right)}{m_{dry}}. \quad (18)$$

We now hypothesize that assuming a single soluble fraction for a given aerosol mixture is in fact equivalent to assuming that everything above i_t is completely dissolved while all the material below this threshold remain undissolved, i.e.

$$\varepsilon_{eff} = \frac{\sum_{j=i_t+1}^n m_{j,dry}}{m_{dry}}. \quad (19)$$

On the other hand, $\varepsilon = \varepsilon_{eff}$ if the following condition is fulfilled (see Eq. 20):

$$\sum_{i=1}^{i_t} m_{i,aqueous} = \sum_{j=i_t+1}^n m_{j,insoluble} = \sum_{j=i_t+1}^n m_{j,dry} - \sum_{j=i_t+1}^n m_{j,aqueous}. \quad (20)$$

Substituting Eq. 5 to Eq. 20 we now have

$$F_w \sum_{i=1}^{i_t} \gamma_i Y_{i,wet} c_{sat,pure,i} = \sum_{j=i_t+1}^n Y_{dry,j} - F_w \sum_{j=i_t+1}^n \gamma_j Y_{j,wet} c_{sat,pure,j}, \quad (21)$$

where

$$F_w = \frac{m_w}{m_{dry}}. \quad (22)$$

At the limit of large n and in the case of a symmetric distribution of material between the insoluble organic and aqueous phases, Eq. 21 is fulfilled by setting the threshold solubility i_t so that

$$\lim_{i \rightarrow i_t} F_w \gamma_i Y_{i,wet} c_{sat,pure,i} = Y_{dry,i_t} - F_w \gamma_{i_t} Y_{i_t,wet} c_{sat,pure,i_t}. \quad (23)$$

In this case threshold solubility c_t is thus found from the bin where

$$c_{sat,pure,i_t} = c_t \approx \frac{Y_{dry,i_t}}{Y_{wet,i_t}} \cdot \frac{1}{\gamma_{i_t}} \cdot \frac{1}{2F_w}. \quad (24)$$

This is also equal to the bin where 50% of the material is partitioned in the insoluble phase, i.e.

$$\chi_{i_t} = \frac{m_{i_t,insoluble}}{m_{i_t,insoluble} + m_{i_t,aqueous}} = \frac{1}{1 + \frac{m_{i_t,aqueous}}{m_{i_t,insoluble}}} = \frac{1}{2}. \quad (25)$$

Finding the solubility threshold c_t requires knowledge on the total amount of organic material as compared with the water at the point of activation, manifested in the ratio F_w . F_w , on the other hand, depends on the ambient supersaturation and the total soluble mass – thus introducing a supersaturation-dependence to the ε given by Eq. 18 as well. The magnitude of F_w as a function of supersaturation can be estimated by plugging Eqs. 15 and 17 into the definition of F_w (Eq. 22) and some rearranging, yielding:

$$F_w = \frac{\rho_w}{\rho_{org}} \left(\frac{2\varepsilon}{s_c} \cdot \frac{\rho_{org}}{\rho_w} \cdot \frac{M_w}{M_{org}} - 1 \right) = \frac{\rho_w}{\rho_{org}} \left(\frac{2\varepsilon\kappa_{max}}{s_c} - 1 \right). \quad (26)$$