Appendix

A

We can compare the relative standard deviations of the metrics we investigated as stated in Eq. (8) in terms of magnitude. It is obvious that for the same conditions the particle number concentration has the lowest relative uncertainty since for PS and PMx additional terms add to the total uncertainty.

The comparison of surface concentration and mass concentration is more complex.

We can state that $\sigma_{PS,rel}$ is proportional to $\frac{Var(S)}{<S>^2}$ while $\sigma_{PMx,rel}$ is proportional to $\frac{Var(V)}{<V>^2}$. If $\frac{Var(V)}{<V>^2}$ is always greater than $\frac{Var(S)}{<S>^2}$, $\sigma_{PMx,rel}$ is necessarily greater than $\sigma_{PS,rel}$. In terms of the size distribution p(D) we can interpret the expectation value of the volume of a single particle as the second moment of the distribution times a constant by assuming spherical particles, as can be seen in Eq. (9). The variance is proportional to the sixth moment of the distribution as shown in Eq. (10).

$$< V > = \frac{1}{6}\pi \int_0^\infty p(D)\, D^3 dD \qquad (9)$$

$$Var(V) = < V^2 > - < V >^2 = \left(\frac{1}{6}\pi\right)^2 \int_0^\infty p(D)\, D^6\, dD - \left(\frac{1}{6}\pi \int_0^\infty p(D)\, D^3 dD\right)^2$$

(10)

In a similar way, we can state that the expectation value of the surface of a single particle is proportional to the second moment of the distribution, while the variance is proportional to the fourth moment.

If we assume a lognormal distribution of the particle diameter, we can use Eq. (11) to calculate the n-th moment of the distribution.

$$E(X^n) = e^{n\mu + n^2\sigma^2/2} \qquad (11)$$

We can use Eq. (12) to postulate Eq. (13) .

$$\frac{Var(A)}{<A>^2} = \frac{<A^2> - <A>^2}{<A>^2} = \frac{<A^2>}{<A>^2} - 1 \qquad (12)$$

$$\frac{<V^2>}{<V>^2} > \frac{<S^2>}{<S>^2} \qquad (13)$$

If we can show that Eq. (13) is valid for any lognormal distribution, then the relative uncertainty of the volume and therefore the mass concentration is greater than the uncertainty of the surface concentration for any lognormal distribution. We use Eq. (11) to proof Eq. (13).

$$\frac{e^{6\mu+36\,\sigma^2/2}}{e^{6\mu+18\,\sigma^2/2}} \geq \frac{e^{4\mu+16\,\sigma^2/2}}{e^{4\mu+8\,\sigma^2/2}}$$

$$e^{9\,\sigma^2} \geq e^{4\,\sigma^2}$$

$$e^{5\,\sigma^2} \geq 1$$

Since $\sigma^2 \geq 0$ we have shown that the relative variability of the surface concentration is necessarily higher than the variability of the mass distribution for spherical particles from a lognormal distribution. Only for cases where $\mathrm{Var}(\rho) = 0$ and σ of the log-normal distribution approaches zero, leading to a monodisperse particle size distribution, the variability of the surface concentration and the variability of the mass concentration are equal to each other and are equal to the relative variability of the particle number concentration which can easily be seen in Eq. (8).

Therefore, we can state Eq. (14) which is valid for spherical lognormal distributed particles.

$$\sigma_{PMx,rel} \geq \sigma_{PS,rel} \geq \sigma_{PN,rel} \quad \text{.....(14)}$$