processes

MDPI

*Supplementary Material*

# Development of a Semi-Empirical Model for Droplet Size Determination of a Three-Channel Spray Nozzle for Pellet Coating Based on the Optical Method Concept

*S1. Model Construction*

In our study, the final model for average droplet size was established for the arithmetic average diameter ($d_{10}$) or number-based diameter, for the De Brouckere average diameter ($d_{43}$) or volume-based diameter, and for the Sauter average diameter ($d_{32}$) (SMD) or surface-based diameter [23], which provided a lower coefficient of determination ($R^2$) and lower cross-validation coefficient of determination ($Q^2$) in the models constructed. Lower values of the coefficient of determination are presumably a consequence of greater experimental error propagation into the calculation of $d_{43}$ and $d_{32}$ in comparison to $d_{10}$. This can be attributed to the measuring technique, which produced $d_{10}$ values, and all other values had to be derived from the basic data. With calculation to other diameters, where multiplication of $d_{10}$ is needed, the measurement error is increased, which results in models with lower fidelity.

Furthermore, in the model, better linearity was achieved by using natural logarithms of factors and the average of the natural logarithm of droplet size, E[ln($d_{10}/D$)], where $d_{10}/D$ denotes the arithmetic average diameter in reduced units. In the case of modeling the distribution scale, SD[ln($d_{10}/D$)], a better model fit was achieved when the values of SD[ln($d_{10}/D$)] were transformed with a logarithmic function, and therefore the following quantity is predicted by the model equation obtained: ln(SD[ln($d_{10}/D$)]).

Potential outliers were identified with the aid of a studentized residuals quantile plot, resulting in the exclusion of one of the data points (C19) from a total of 28 experiments. The final model properties are shown in Figure S1 for average droplet size, E[ln($d_{10}/D$)], which provides information on the strength and robustness of the model. The $R^2$ of 0.90 indicates a low measurement variability in the response (average droplet size) and strong fit between the data and the model. The $Q^2$ value of 0.73 demonstrates high predictive power, allowing for confident prediction of the effect of changing the reduced quantities / process parameters on average droplet size and process optimization. The model constructed also demonstrates an acceptable score for validity (0.46). Moreover, the value of reproducibility is also high (0.96), indicating good experimental control and low residual variability.

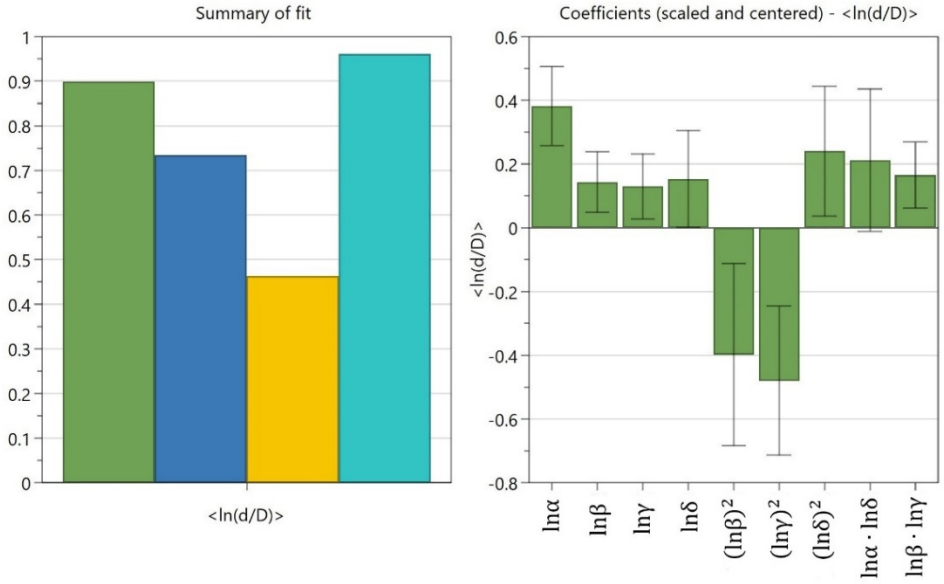



**Figure S1.** Overview of the model for average droplet size. (**a**) summary of regression analysis: Column 1: $R^2$ = model fit, Column 2: $Q^2$ = model predictability, Column 3: model validity, and Column 4: model reproducibility; (**b**) factor effect plots.

As demonstrated in Figure S1, all reduced factors (ln$\alpha$, ln$\beta$, ln$\gamma$, and ln$\delta$) contribute to the average droplet size. Based on these factors and their relation to the underlying physical quantities, we confirmed that the process of droplet formation is complex and dependent on various mechanisms, as discussed in published literature [10,12,16,22,40]. The magnitude of model coefficients can be used for a general overview of the importance of process parameters for droplet formation. The greatest influence on droplet size can be attributed to the quadratic term of ln$\gamma$ and ln$\beta$ (negative impact), followed by the main term of ln$\alpha$ and the quadratic term of ln$\delta$ (positive impact). Figure S2 can aid in translating the reduced quantities into process parameters. It is well known that increasing atomizing pressure will reduce droplet size [7,10,13,21,22,43], which was confirmed in this study. It is interesting to see that the maximum droplet size can be observed at $\ln\gamma = \ln(p_m/p_a) = -1$. In addition, the model confirms that both higher dispersion flow (increasing ln$\alpha$ when the atomizing pressure is kept constant) and higher surface tension (increasing ln$\delta$ at constant ln$\beta$), increases the droplet size.

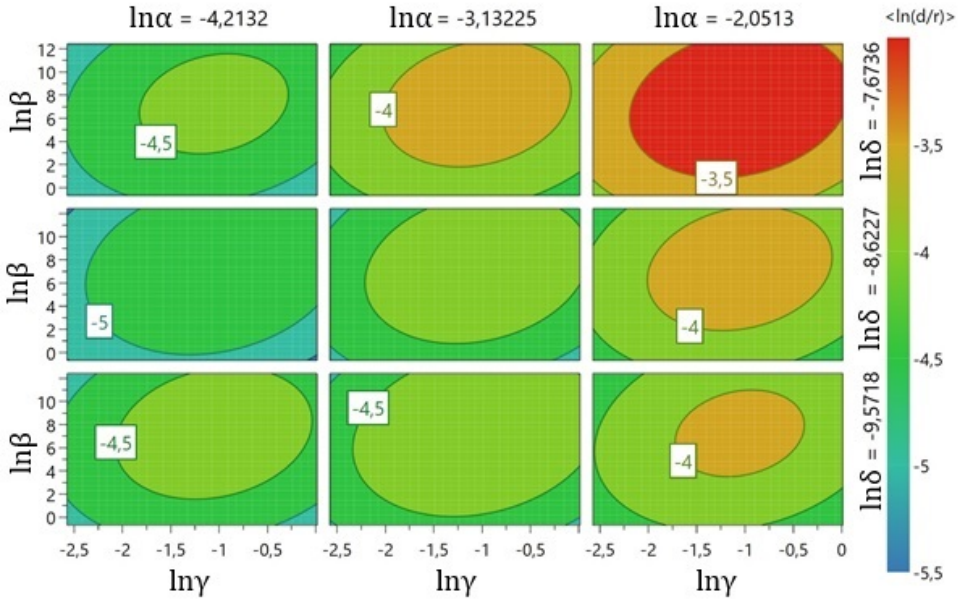



**Figure S2.** 4D response contour plots for $E[\ln\left(\frac{d_{10}}{D}\right)]$ in correlation with reduced quantities ln$\alpha$, ln$\beta$, ln$\gamma$, and ln$\delta$. ln$\alpha$ and ln$\delta$ are presented at minimum, middle, and maximum value, and ln$\beta$ and ln$\gamma$ are presented throughout the entire range tested.

As mentioned above, in order to capture the distribution of droplets, standard deviation of droplet size, SD[ln($d_{10}/D$], was also modeled. The properties of the model constructed were estimated as good and are shown in Figure S3. ($R^2$ = 0.84, $Q^2$ = 0.61; model validity = 0.64 and reproducibility = 0.90).

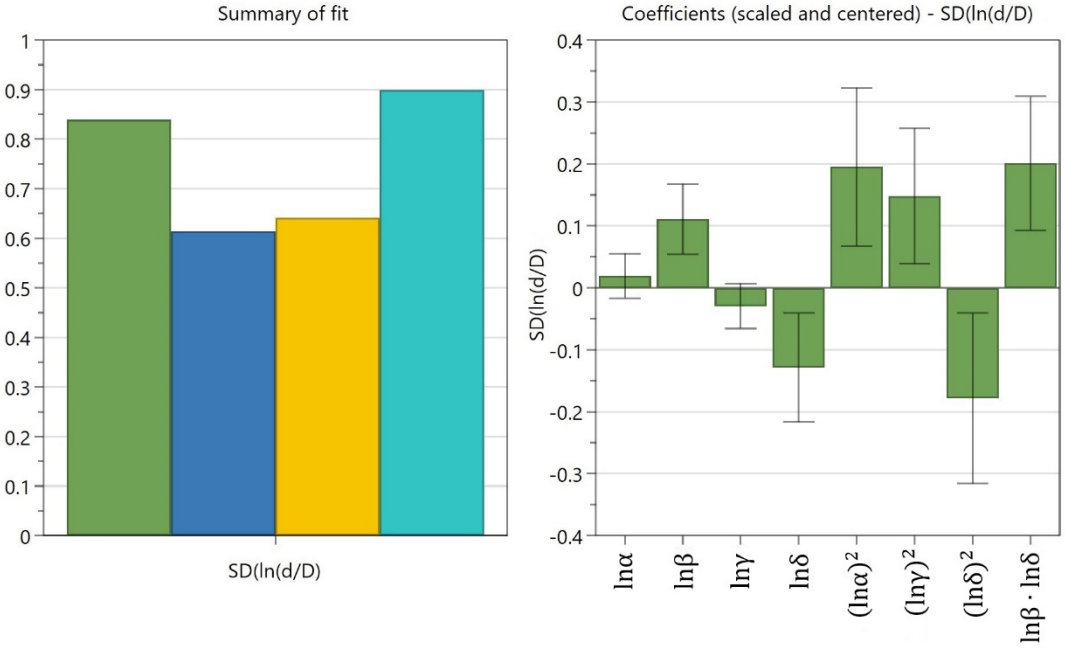



**Figure S3.** Overview of the model for standard deviation of diameter of droplet size. (**a**) summary of regression analysis: Column 1: $R^2$ = model fit, Column 2: $Q^2$ = model predictability, Column 3: model validity, and Column 4: model reproducibility; (**b**) factor effect plots.

The 4D response contour plot (Figure S4) shows that increasing surface tension and decreasing viscosity results in a wider distribution (increasing ln$\beta$), which is in line with the theory and published literature [19–23,32,40,48,49].

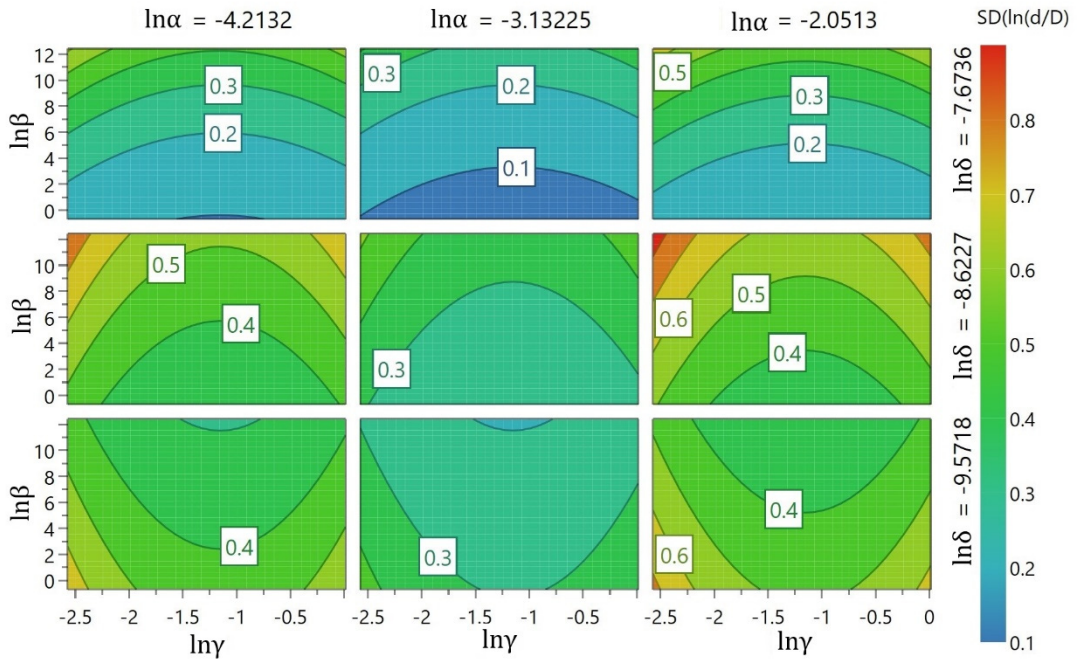



**Figure S4.** 4D response contour plots for the standard deviation of average diameter of droplet size in correlation with reduced quantities ln$\alpha$, ln$\beta$, ln$\gamma$, and ln$\delta$. ln$\alpha$ and ln$\delta$ are presented at minimum, middle, and maximum value, and ln$\beta$ and ln$\gamma$ are presented throughout the entire range tested.

*S2. Droplet Size Distribution*

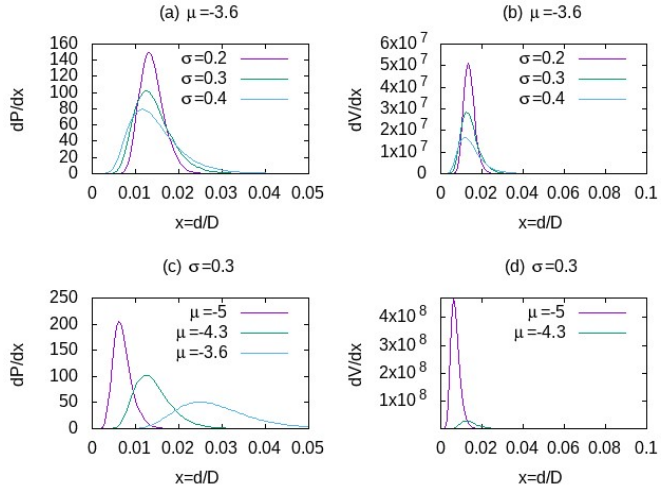



**Figure S5.** Size and volume distribution of droplets at different averages (μ= E[ln(x)]) and standard deviations (σ=SD[ln(x)]). Droplets in the top row have the same log-scale mean size but different spread of sizes. Droplets in the bottom row have the same log-scale spread of sizes, but different mean size.

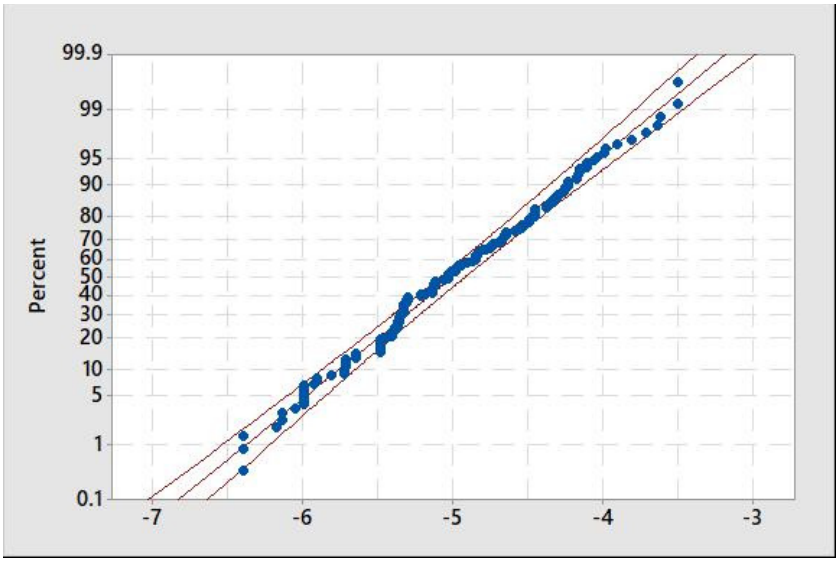



**Figure S6.** Representation of the empirical droplet size distribution of Experiment N2 after logarithmic transformation as a normal probability plot. The transformed distribution follows the normal distribution (red line with confidence bands).