



## Section S1. All supporting data about parameter estimation–optimization and evolution of the PSD

**Table S1.** Fitted parameters of Model A, lower and upper boundaries of the parameters, initial guess, and sum-of-squared residuals *SSR* for different numbers of trials  $N_T$ .

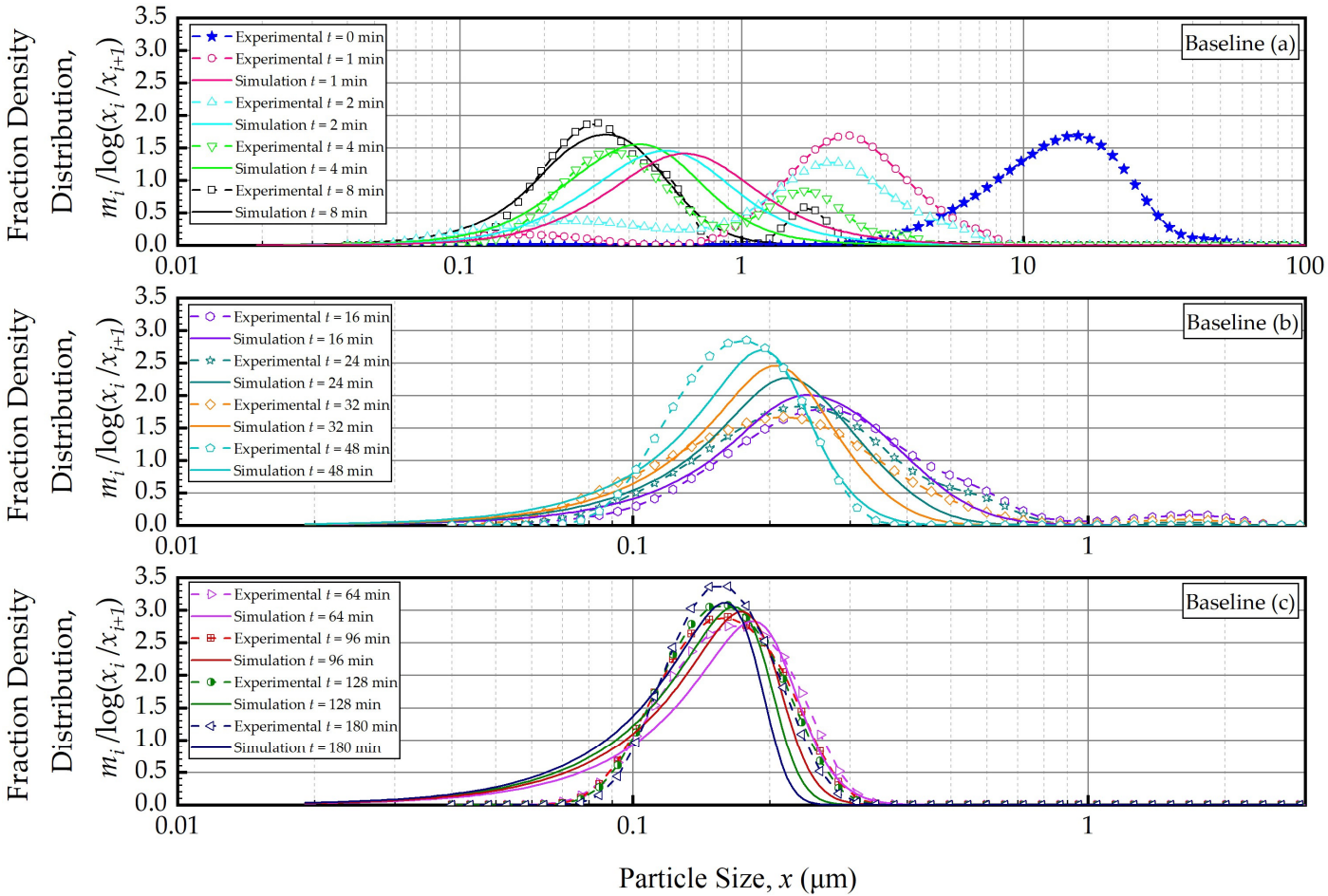
	Model A	$a_1$	$A \text{ (min}^{-1}\text{)}$	$m$
	Lower Boundary	0	500	0
	Upper Boundary	5	$10^{13}$	4
	Initial Guess	2	$5 \times 10^9$	2
$N_T = 200$	Fitted Parameters	2.62	$9.99 \times 10^8$	3.04
	<i>SSR</i>	60.7		
$N_T = 400$	Fitted Parameters	2.45	$4.46 \times 10^{11}$	3.88
	<i>SSR</i>	53.5		
$N_T = 800$	Fitted Parameters	2.46	$3.32 \times 10^{11}$	3.84
	<i>SSR</i>	53.5		
$N_T = 1000$	Fitted Parameters	2.46	$3.32 \times 10^{11}$	3.84
	<i>SSR</i>	53.5		

**Table S2.** Fitted parameters of Model B, lower and upper boundaries of the parameters, initial guess, and sum-of-squared residuals *SSR* for different numbers of trials  $N_T$ .

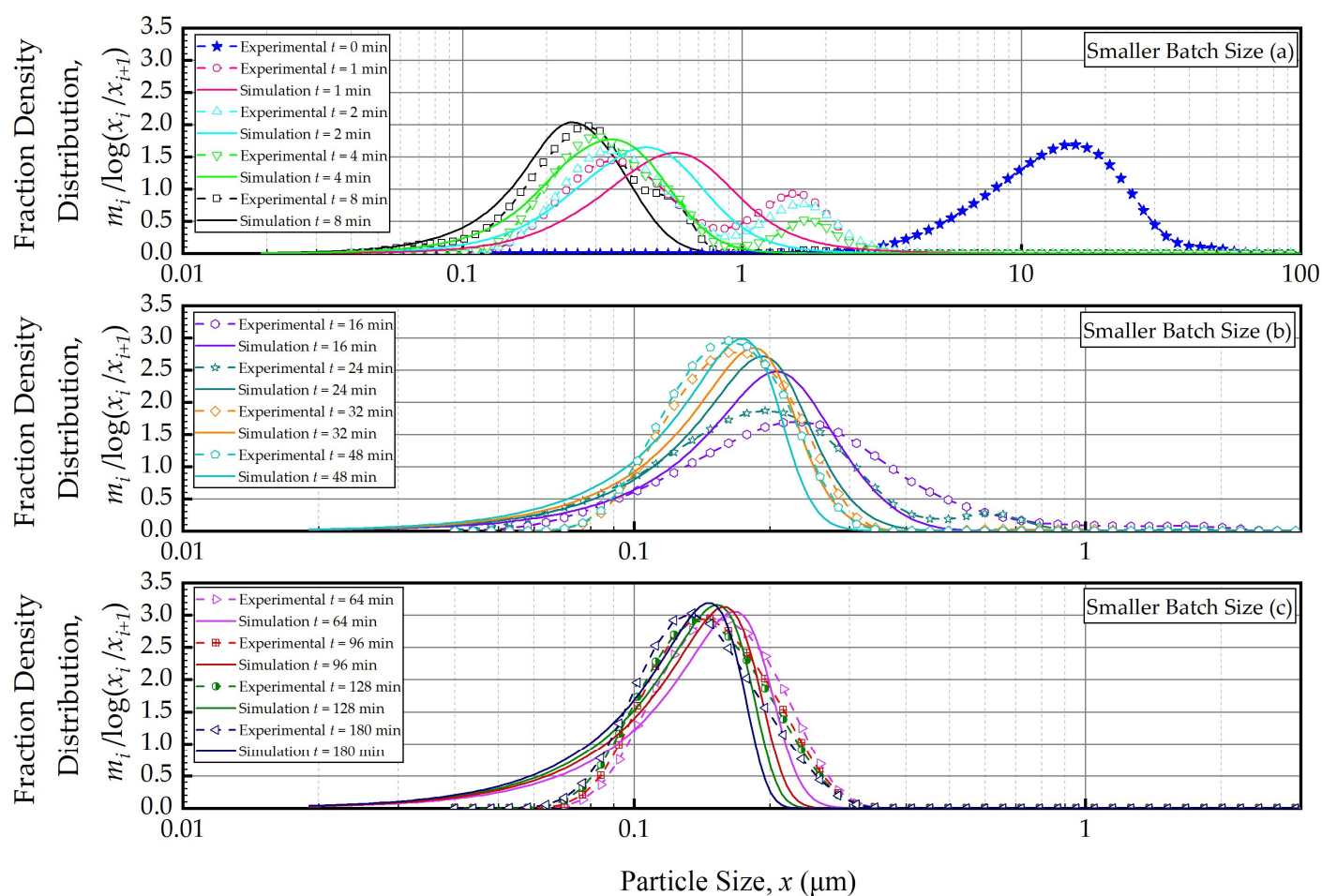
	Model B	$a_1$	$A \text{ (min}^{-1}\text{)}$	$m$	$x^* \text{ (}\mu\text{m)}$
	Lower Boundary	0	500	0	0.038
	Upper Boundary	5	$10^{10}$	4	1
	Initial Guess	2	$5 \times 10^4$	2	0.1
$N_T = 200$	Fitted Parameters	1.95	$2.54 \times 10^8$	2.86	0.195
	<i>SSR</i>	46.2			
$N_T = 400$	Fitted Parameters	2.07	$3.95 \times 10^9$	3.24	0.189
	<i>SSR</i>	43.9			
$N_T = 800$	Fitted Parameters	2.25	$2.92 \times 10^9$	3.19	0.174
	<i>SSR</i>	43.4			
$N_T = 1000$	Fitted Parameters	2.25	$2.92 \times 10^9$	3.19	0.174
	<i>SSR</i>	43.4			

**Table S3.** Fitted parameters of Model D, lower and upper boundaries of the parameters, initial guess, sum-of-squared residuals SSR for different numbers of trials  $N_T$ .

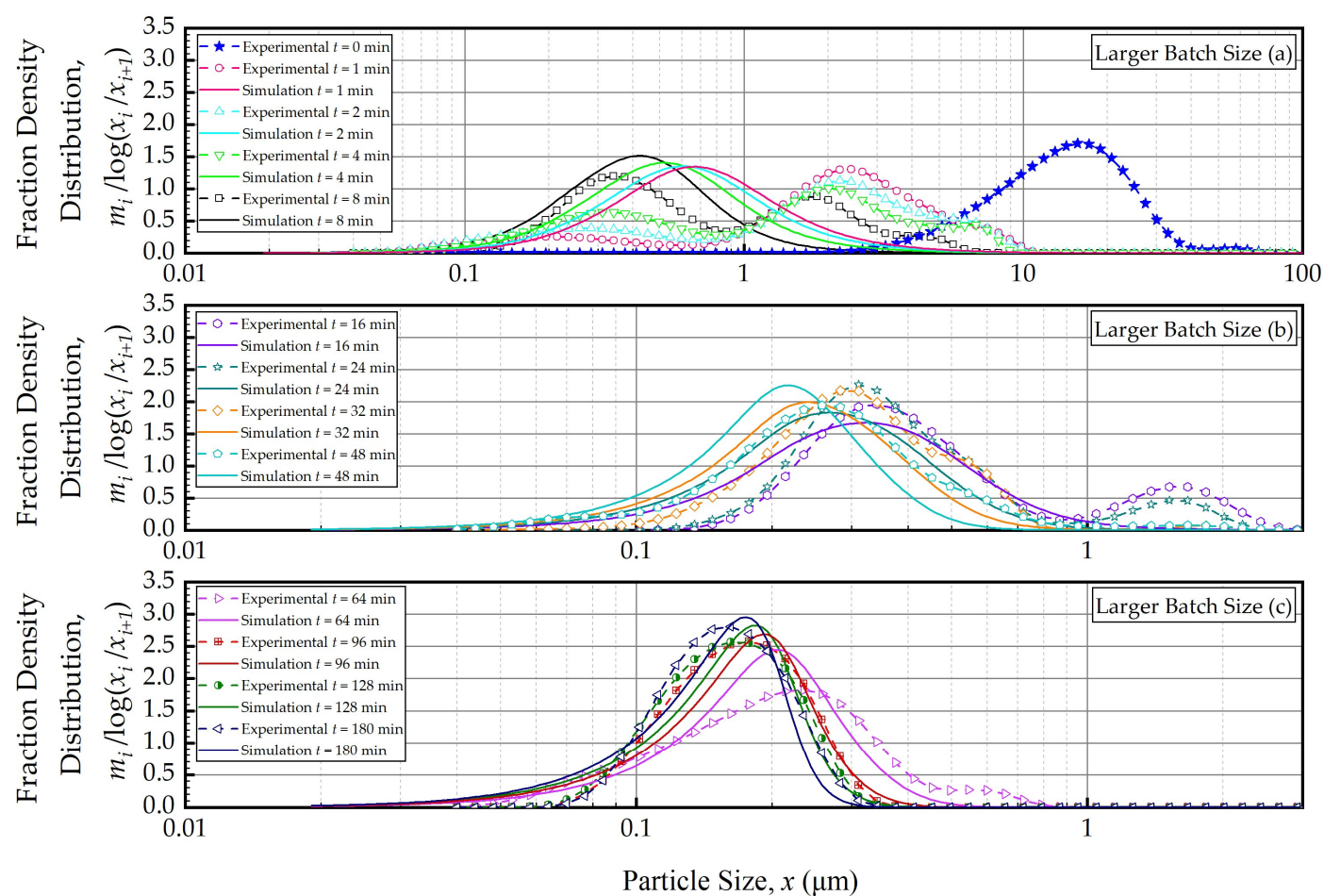
	Model D	$a_1$	$A \text{ (min}^{-1}\text{)}$	$m$	$x^* \text{ (}\mu\text{m)}$	$s_f \text{ (}\mu\text{m}^{-1}\text{)}$
	Lower Boundary	0	50	0	0.038	10
	Upper Boundary	5	$10^8$	4	1	50
	Initial Guess	2	$5 \times 10^4$	2	0.2	30
$N_T = 200$	Fitted Parameters	2.31	$1.15 \times 10^6$	2.03	0.228	36.6
	SSR	38.7				
$N_T = 400$	Fitted Parameters	2.31	$1.03 \times 10^7$	2.36	0.214	41.7
	SSR	38.4				
$N_T = 800$	Fitted Parameters	2.31	$1.03 \times 10^7$	2.36	0.214	41.7
	SSR	38.4				
$N_T = 1000$	Fitted Parameters	2.31	$1.03 \times 10^7$	2.36	0.214	41.7
	SSR	38.4				



**Figure S1.** Temporal variation of the mass fraction density distribution during (a) the first 8 min of milling, (b) between 16 and 48 min of milling, and (c) thereafter for the baseline experiment: volumetric flow rate of 126 mL/min and batch volume of 236 mL (Baseline: Run 1). Simulation: PBM with Model C and its fitted parameters. Note that 1 min and 2 min data were not used in the PBM parameter estimation due to pronounced aggregation and bimodality.

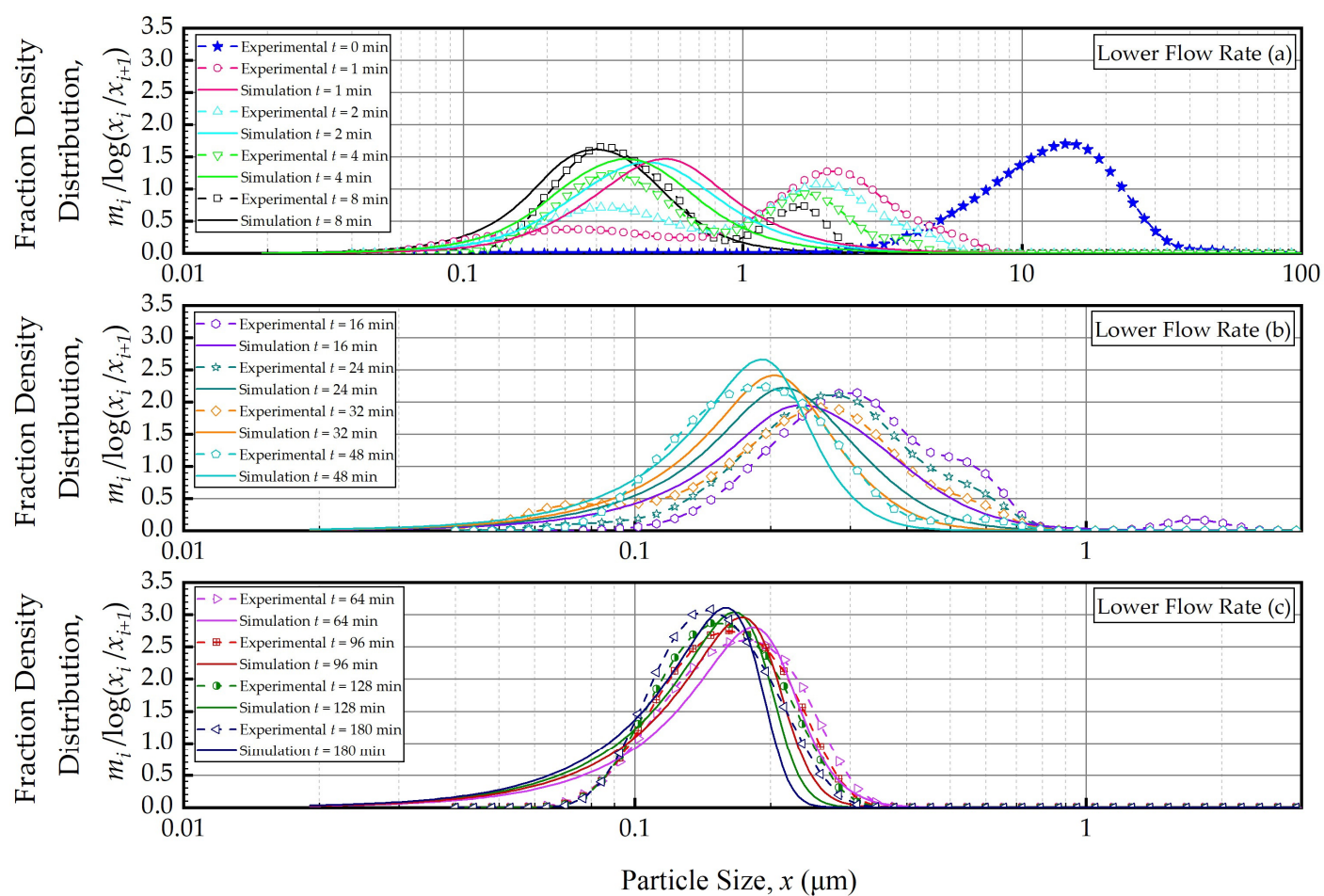


**Figure S2.** Temporal variation of the mass fraction density distribution during (a) the first 8 min of milling, (b) between 16 and 48 min of milling, and (c) thereafter for Run 2: volumetric flow rate of 126 mL/min and batch volume of 118 mL. Simulation: PBM with Model C and its fitted parameters.

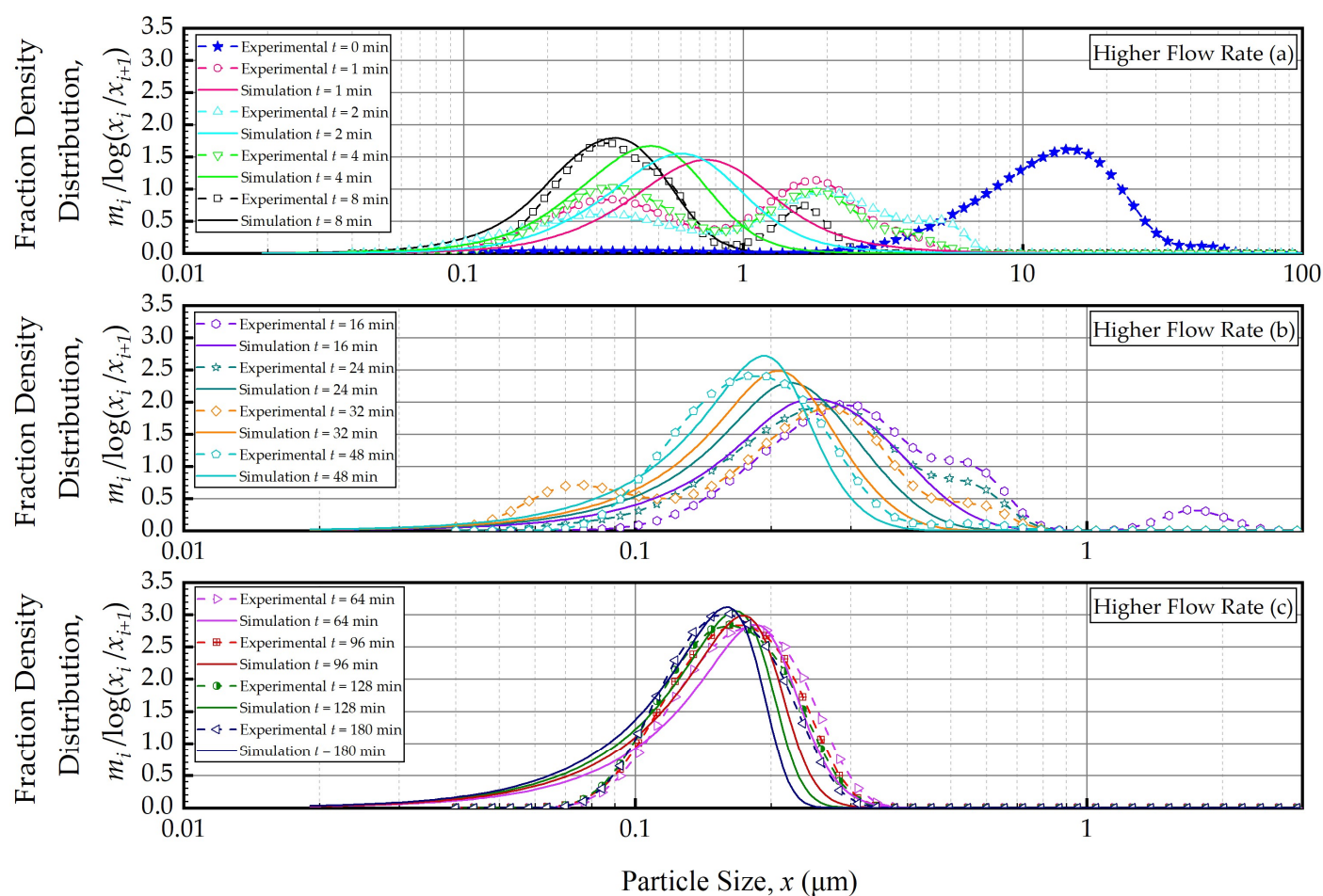


**Figure S3.** Temporal variation of the mass fraction density distribution during (a) the first 8 min of milling, (b) between 16 and 48 min of milling, and (c) thereafter for Run 3: volumetric flow rate of 126 mL/min and batch volume of 472 mL. Simulation: PBM with Model C and its fitted parameters.

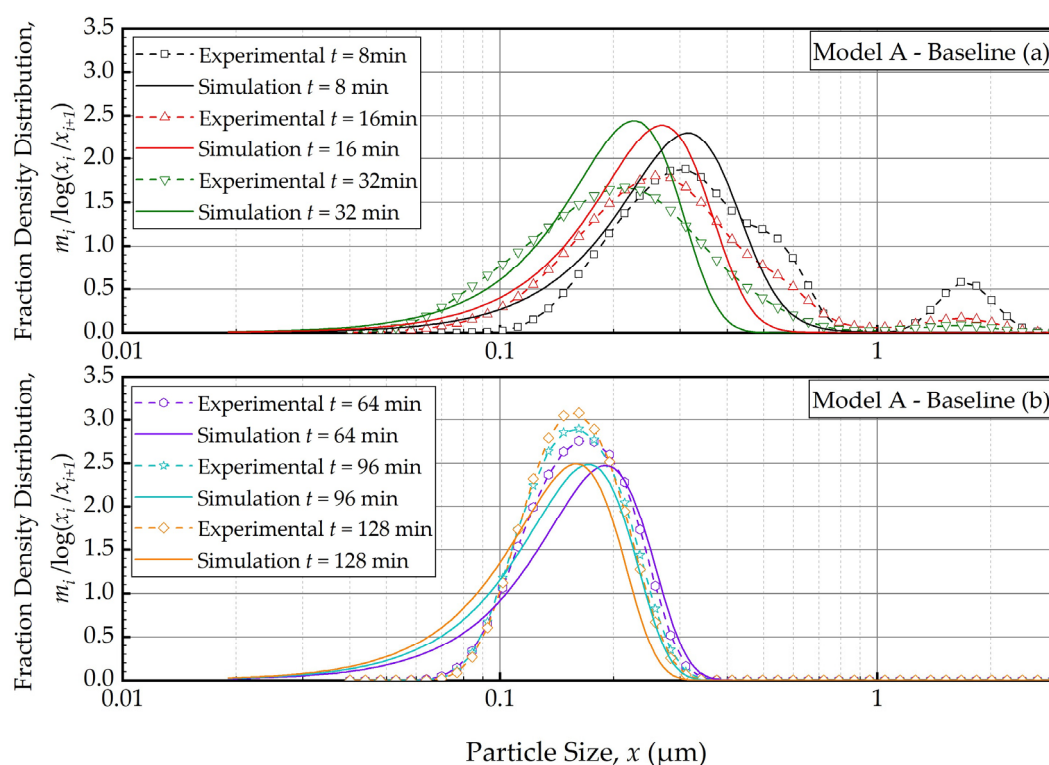




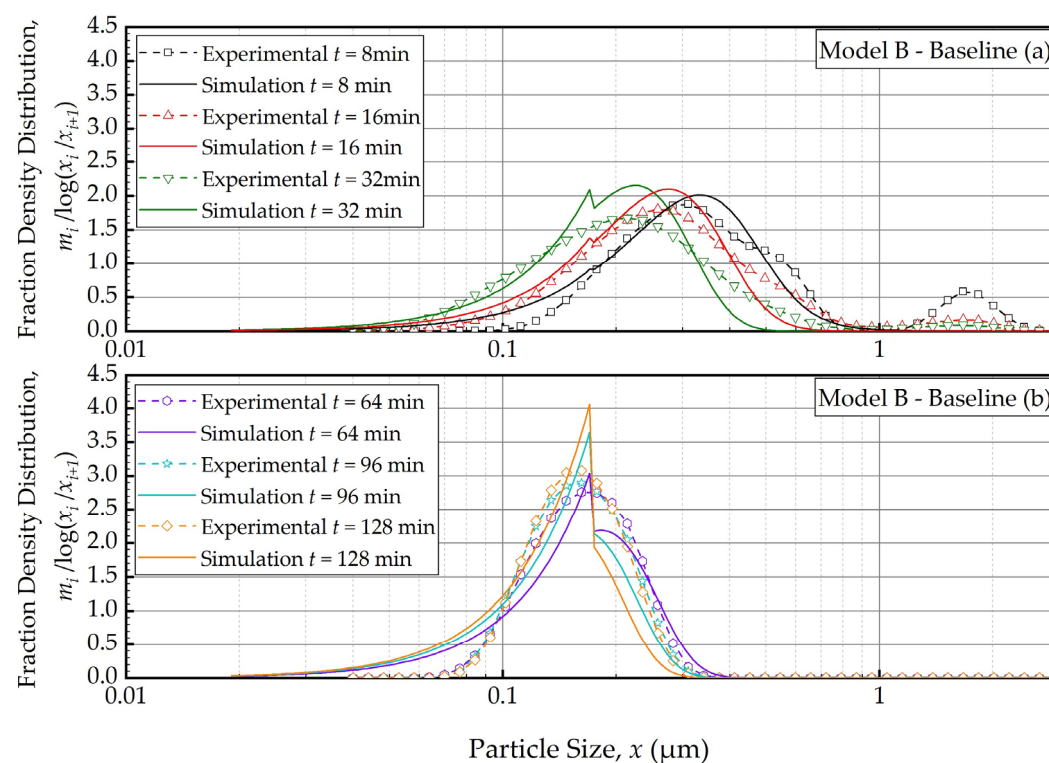
**Figure S4.** Temporal variation of the mass fraction density distribution during (a) the first 8 min of milling, (b) between 16 and 48 min of milling, and (c) thereafter for Run 4: volumetric flow rate of 63 mL/min and batch volume of 236 mL. Simulation: PBM with Model C and its fitted parameters.



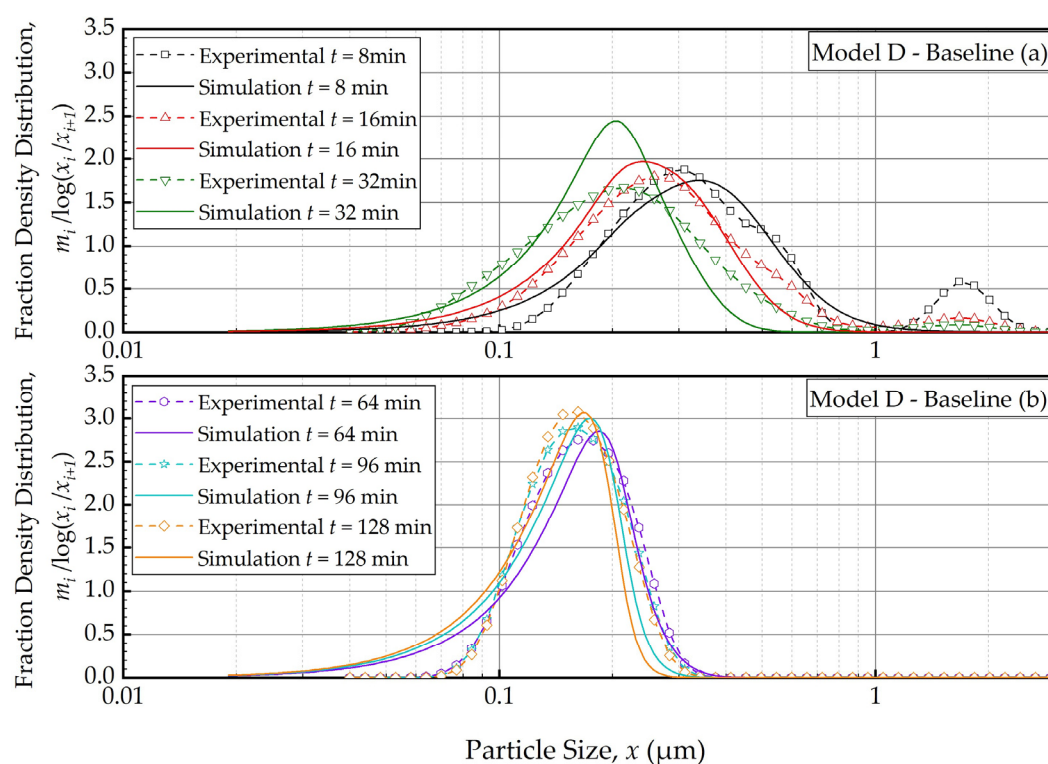
**Figure S5.** Temporal variation of the mass fraction density distribution during (a) the first 8 min of milling, (b) between 16 and 48 min of milling, and (c) thereafter for Run 5: volumetric flow rate of 250 mL/min and batch volume of 236 mL. Simulation: PBM with Model C and its fitted parameters.



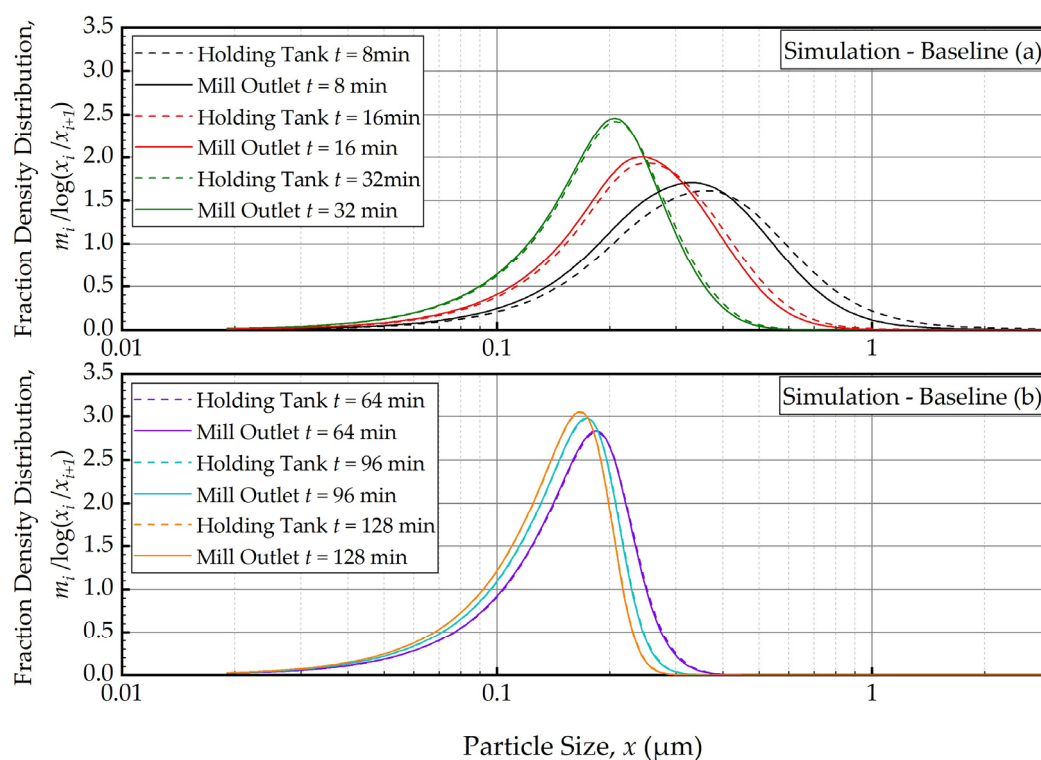
**Figure S6.** Temporal variation of the mass fraction density distribution during (a) the first 32 min of milling and (b) thereafter for the baseline experiment: volumetric flow rate of 126 mL/min and batch volume of 236 mL (Run 1). Simulation: PBM with Model A and its fitted parameters.



**Figure S7.** Temporal variation of the mass fraction density distribution during (a) the first 32 min of milling and (b) thereafter for the baseline experiment: volumetric flow rate of 126 mL/min and batch volume of 236 mL (Run 1). Simulation: PBM with Model B and its fitted parameters.



**Figure S8.** Temporal variation of the mass fraction density distribution during (a) the first 32 min of milling and (b) thereafter for the baseline experiment: volumetric flow rate of 126 mL/min and batch volume of 236 mL (Run 1). Simulation: PBM with Model D and its fitted parameters.



**Figure S9.** Simulated temporal variation of the mass fraction density distribution in the milling chamber and the holding tank during (a) the first 32 min of milling and (b) thereafter for the baseline experiment: volumetric flow rate of 126 mL/min and batch volume of 236 mL (Run 1, Model C).