



*Supplement of*

## **Fast and sensitive measurements of sub-3 nm particles using Condensation Particle Counters For Atmospheric Rapid Measurements (CPC FARM)**

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## S1. CPC FARM Single Channel Schematic

Figure S1 shows a schematic of a single channel of the Condensation Particle Counters For Atmospheric Rapid Measurements (CPC FARM). Each channel samples at  $3.3 \text{ L min}^{-1}$ . Prior to entering the first stage of the growth tube,  $3 \text{ L min}^{-1}$  of transport flow is removed. The remaining  $0.3 \text{ L min}^{-1}$  continues into the  $7.1 \text{ cm}$  long x  $4.7 \text{ mm}$  ID, cold conditioner stage. In this study, the conditioner stage was set between  $1 - 5 \text{ }^\circ\text{C}$ . Next, the flow enters the  $2 \text{ cm}$  long, hot initiator stage which was set between  $35 - 99 \text{ }^\circ\text{C}$ . The final cold moderator stage is  $7.1 \text{ cm}$  long and set at  $1 \text{ }^\circ\text{C}$ . The flow is then directed through a nozzle before entering the optics which counts particles by collecting the light diffracted by particles passing through a laser beam.

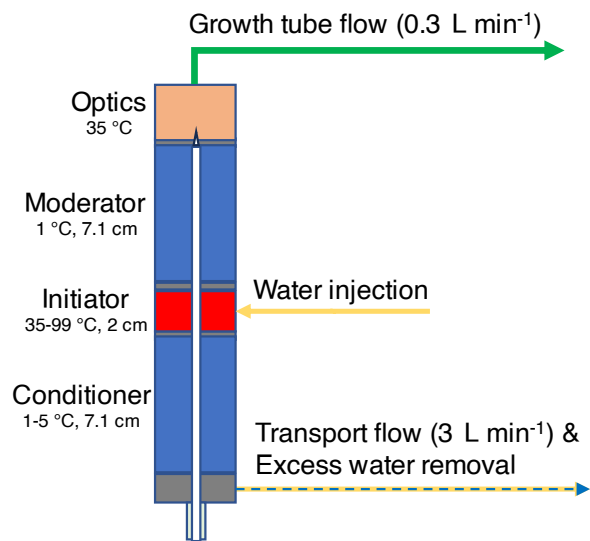


Figure S1. Schematic of a single channel of the CPC FARM showing the dimensions and operating temperature ranges of each stage. The green arrow indicates the sample growth tube flow, blue arrow is the transport flow, and the yellow arrows are the injection and ejection of water.

## S2. CPC FARM detection efficiency curves

Figure S2 displays the measured detection efficiency curves of each channel of the CPC FARM for positively charged ammonium sulfate particles. To avoid electrometer noise, detection efficiency measurements were set to 0 where the measured electrometer concentration was  $<50 \text{ cm}^{-3}$ . For almost all temperature settings, the detection efficiency reaches a plateau maximum which is not 100%. This is due to diffusional wall loss inside the CPC FARM. Note, the plateau in the detection efficiency was not reached when the conditioner and initiator temperatures were set to 5 and 35 °C, respectively (and subsequently noted as 5/35 °C), due to the maximum size range of the Half Mini DMA as configured. Figure S2 shows that the five channels behave similarly. The lowest  $d_{50}$  was found at the maximum difference between conditioner and initiator stages and ranged between 1.56 and 1.74 nm between the 5 channels. On all five channels, as the difference between the conditioner and initiator temperatures decreased, the  $d_{50}$  increased at a higher rate.

A fitted model, given in Eq. (S1), is shown as solid lines in Figure S2.

$$\eta(x) = \eta_{\max} \left[ 1 - \exp \left( -\ln(2) \left( \frac{x - d_0}{d_{50,\text{cal}} - d_0} \right) \right) \right] \eta_{\text{GK}}(x, L_{\text{eff}}, q_a) \quad \text{Eq. (S1)}$$

$x$  describes the particle diameter,  $\eta_{\max}$  describes the maximum efficiency (plateau) at  $x \gg d_{50}$ ,  $d_0$  describes the diameter where the detection efficiency begins to increase from 0%,  $d_{50,\text{cal}}$  describes the diameter where the detection efficiency reaches 50% assuming no diffusion losses, and  $\eta_{\text{GK}}$  is the (size-dependent) particle transmission efficiency in laminar flow through a tube (Gormley and Kennedy, 1948) which is parameterized with an effective tube length ( $L_{\text{eff}}$ ) for given aerosol flowrate ( $q_a$ ). The parameters used in Eq. (S1) for each channel are summarized in Table S1. For the 5/35 °C curve, a point was artificially added at 100 nm at the max detection efficiency of the 1/41 °C conditioner/initiator settings for the curve fit. The 5/35 °C curve fit is for visual reference only and was not used to extrapolate setpoint temperatures.

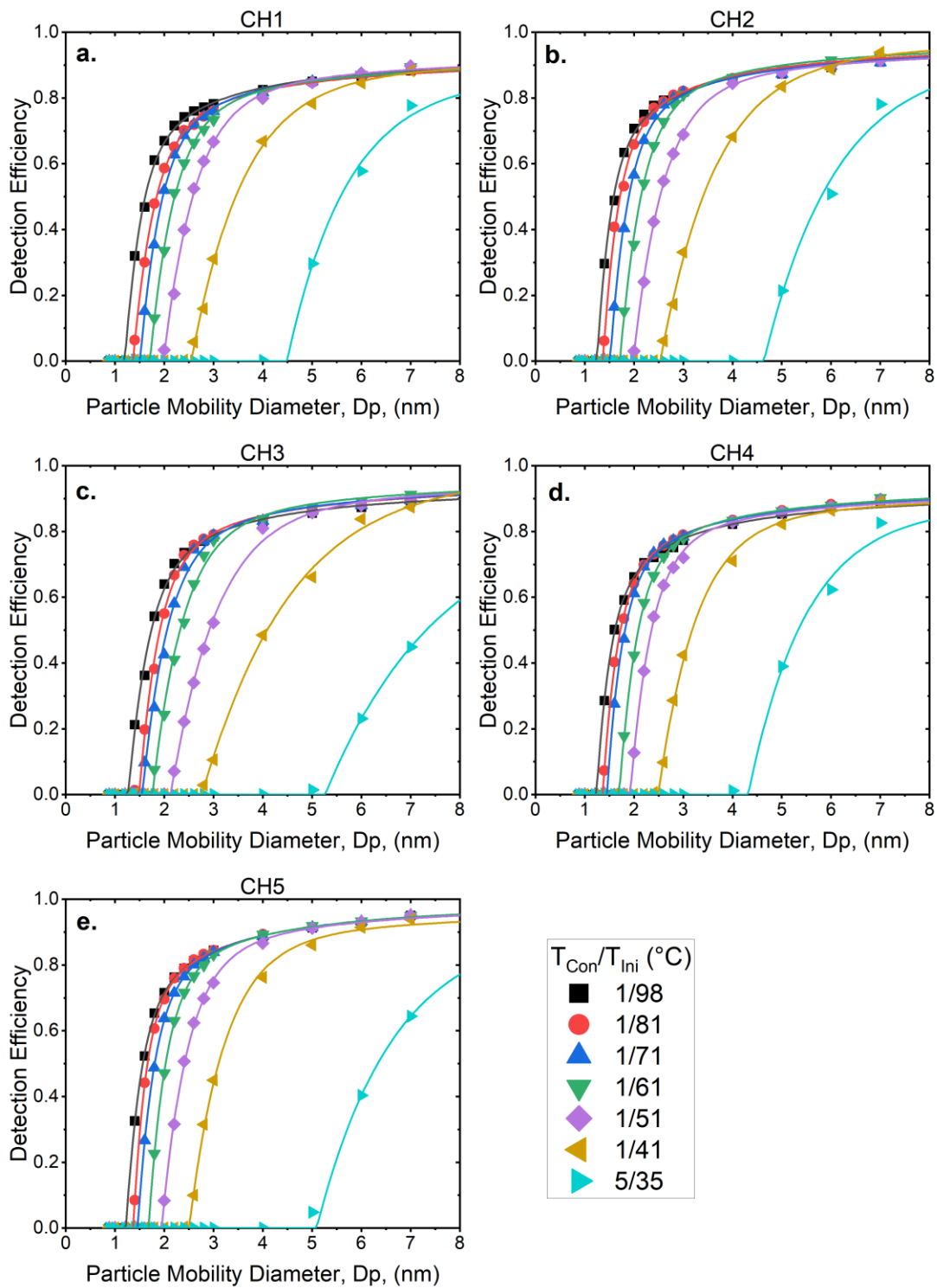


Figure S2. Detection efficiency curves for each of the five channels sorted by cut-point used during the Pittsburgh campaign with Channel 1 set to the lowest cut-point. Each color/symbol represents a different set of conditioner and initiator temperatures. Solid lines represent the fitted model.

Table S1. CPC FARM fitted particle detection efficiency parameters

	$\eta_{\max}$	$d_0$ (nm)	$d_{50,\text{cal}}$ (nm)	$L_{\text{eff}}$ (m)	$q_a$ (L min <sup>-1</sup> )
Ch. 1	0.934	1.201	1.378	0.050	0.300
Ch. 2	0.972	1.426	1.616	0.050	0.300
Ch. 3	0.958	1.604	1.913	0.050	0.300
Ch. 4	0.934	2.038	2.346	0.050	0.300
Ch. 5	0.979	2.421	2.851	0.050	0.300

### S3. CPC FARM Live time fraction

Figure S3 shows the relationship between live time fraction,  $f_L$ , and the calculated number concentration reported by each channel,  $N_i$ , of the CPC FARM. The data shown were taken in Pittsburgh, PA between October 11<sup>th</sup> and 14<sup>th</sup> 2023. The live time fraction describes the amount of time within each CPC sampling interval where the detector can detect a particle. As the particle counts increase, the fraction live time decreases. The fraction of real particles detected by the CPC detector over the sampling interval decreases and a correction factor is needed to resolve the true particle concentration. A fitted model, given in Eq. (S2), is shown as solid lines in Figure S3.

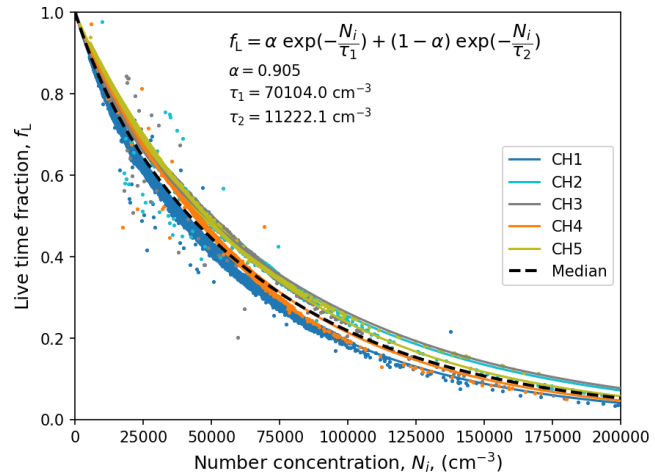


Figure S3: Plot showing the relationship between live time fraction and the calculated number concentration measured by each channel of the CPC FARM. Points represent the raw data points. Solid lines represent the curve fitted to the displayed equation. The constants are provided for the median fit. Data were taken between October 11<sup>th</sup>, 2023 and October 14<sup>th</sup> 2023.

$$f_L(N_i) = \alpha \exp\left(-\frac{N_i}{\tau_1}\right) + (1 - \alpha) \exp\left(-\frac{N_i}{\tau_2}\right) \quad \text{Eq. (S2)}$$

where  $N_i$  describes the number concentration, and  $\alpha$ ,  $\tau_1$  and  $\tau_2$  are fitting parameters. In the future, the live time fraction of the CPC-FARM could easily be increased by reducing the nozzle diameter.

#### S4. $\eta_{max}$ variation over time

Figure S4 shows the  $\eta_{max}$  calculated daily over from June 15<sup>th</sup>, 2024 to June 28<sup>th</sup>, 2024. The study was conducted at the Department of Energy Atmospheric Radiation Measurement Southern Great Plains (SGP) field site in Billings, OK. The CPC FARM was set up as described in Fig. 1. The study was conducted at SGP because, unlike Pittsburgh in Fall 2023, there were weeks where each day had significant periods without sub-10 nm particles in the size distribution.

The  $\eta_{max}$  represents the overall transmission efficiency of each channel on the CPC FARM.

The  $\eta_{max}$  was found by taking the ratio of  $N_i$  on each channel to the average of  $N_i$  across all channels over the 10-minute span where both  $\Delta N$  and the change in sum of  $\Delta N$  across all channels is the lowest. The plot shows that  $\eta_{max}$  only varies  $\pm 2\%$  of the starting value within these two weeks and does not consistently drift over time.

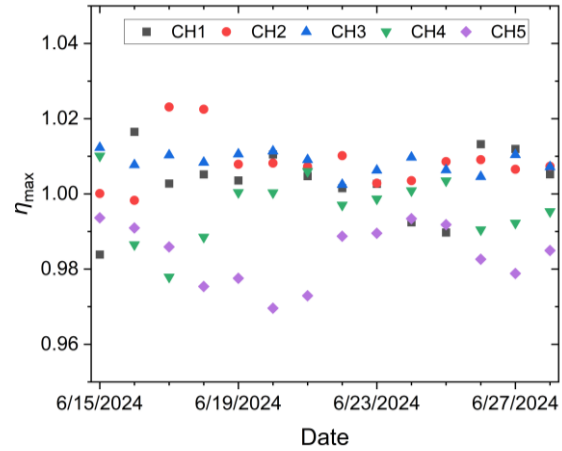


Figure S4: Plot showing the variation of  $\eta_{max}$  for the CPC FARM. Data were taken between June 15<sup>th</sup>, 2024 and June 28<sup>th</sup>, 2024 at the Department of Energy Atmospheric Radiation Measurement Southern Great Plains field site.

## S5. Simulated data inversion with low-concentration input

Figure S5 shows a simulated data inversion example similar to that shown in Fig. 5. The input distribution here has the same modal parameters as the original example, but the particle number in the  $<5$  nm mode is reduced from  $5,000$  to  $500$   $\text{cm}^{-3}$  to demonstrate the inversion performance and expected error at low differential signal levels. The numerical inversion still shows good agreement with the true input, and the approximate inversion is still lower than the true input.

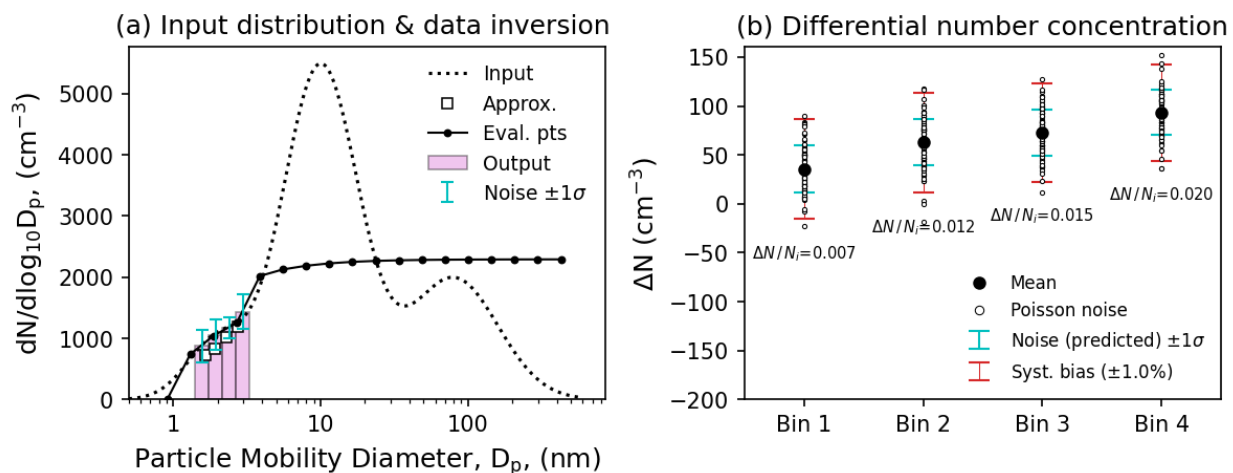


Figure S5. Simulated data inversion example with a 3-mode lognormal distribution. a) Output (numerical data inversion, bars) and approx. (approximate data inversion, square points) vs. input size distribution (dots) for noisy CPC FARM signals. Blue error bars represent  $1\sigma$  of instrument noise. Black points are the 20 evaluation points used in the particle size vector for the kernel matrix Eq. (5). b) The resulting differential number across the instrument channels (mean values and noise due to random Poisson error), as well as systematic error due to  $\pm 1\%$  bias in the signal of the first channel of each bin.  $\Delta N/N_i$  values indicate the relative differential number concentration in each bin. A sample flow rate of  $0.3$   $\text{L min}^{-1}$  and a sampling time interval of  $5$  s were used in this example.

## S6. CPC FARM numerical inversion compared to approximate inversion

Figure S6 displays the particle size distribution measured in Pittsburgh on October 12<sup>th</sup>, 2023 by the CPC FARM inverted using the numerical and approximate methods. In the approximate inversion, the second and third largest size bins are lower and higher, respectively, compared to the numerical inversion. These discrepancies can be attributed to the incorrect assumption of step-function detection efficiency curves as the approximate inversion cannot account for overlap in the detection efficiency curves between neighboring channels nor the true shape of the sigmoidal detection efficiency curve.

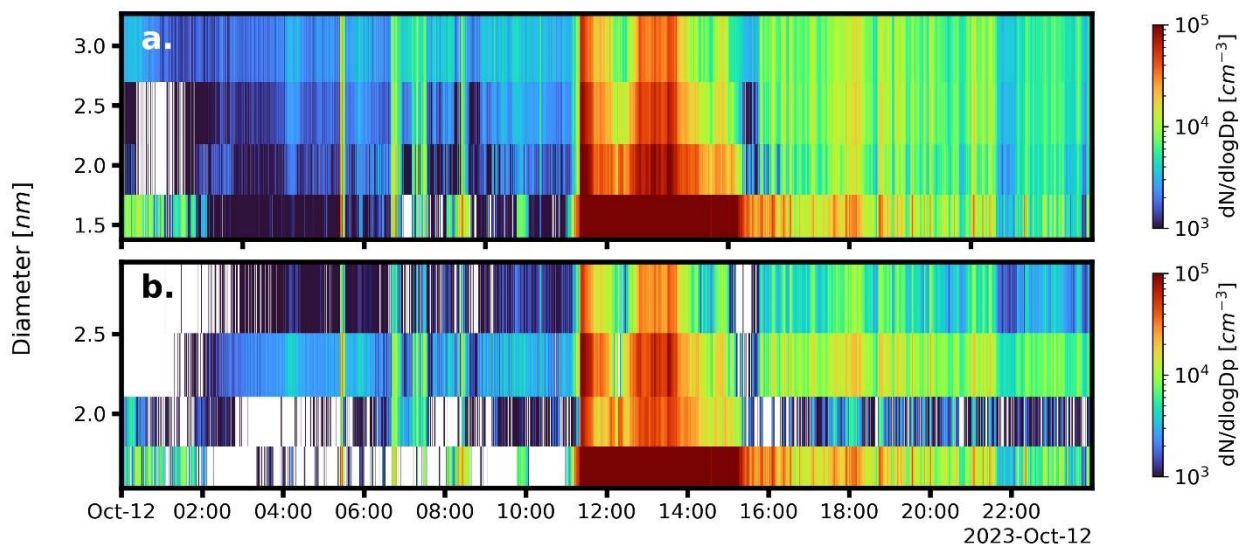


Figure S6: 1.6 – 2.9 nm size distributions taken on October 12<sup>th</sup>, 2023. (a) Contour plot showing the numerically inverted CPC FARM data recorded at 1 Hz with a 10-second average applied. The midpoints of the four size bins are 1.6, 1.9, 2.3, and 2.9 nm. (b) Contour plot showing the CPC FARM data inverted with the approximate method. The midpoints of the four size bins are 1.7, 1.9, 2.3, and 2.7 nm.



## S7. Comparison of CPC FARM and PSD

Figure S7 shows parity plots of the PSD and CPC FARM at the midpoint of each size bin of the CPC FARM and the closest corresponding size bin midpoint of the PSD. For each data point, the CPC FARM observations were averaged over the five minutes of the corresponding PSD scan. The figure shows that at 1.6 and 1.9 nm, PSD measured concentration does not increase with increasing concentration measured by the CPC FARM. Between 2 and 3 nm, the PSD appears to undercount particle concentration by 50%. This figure also shows that the noise of the PSD decreases from 500,000  $\text{cm}^{-3}$  to 20,000  $\text{cm}^{-3}$  as the size increases from 1.6 to 2.4 nm. This is not surprising as the SMPS correction factors are larger for smaller sizes to due to lower charging efficiency and higher diffusion losses.

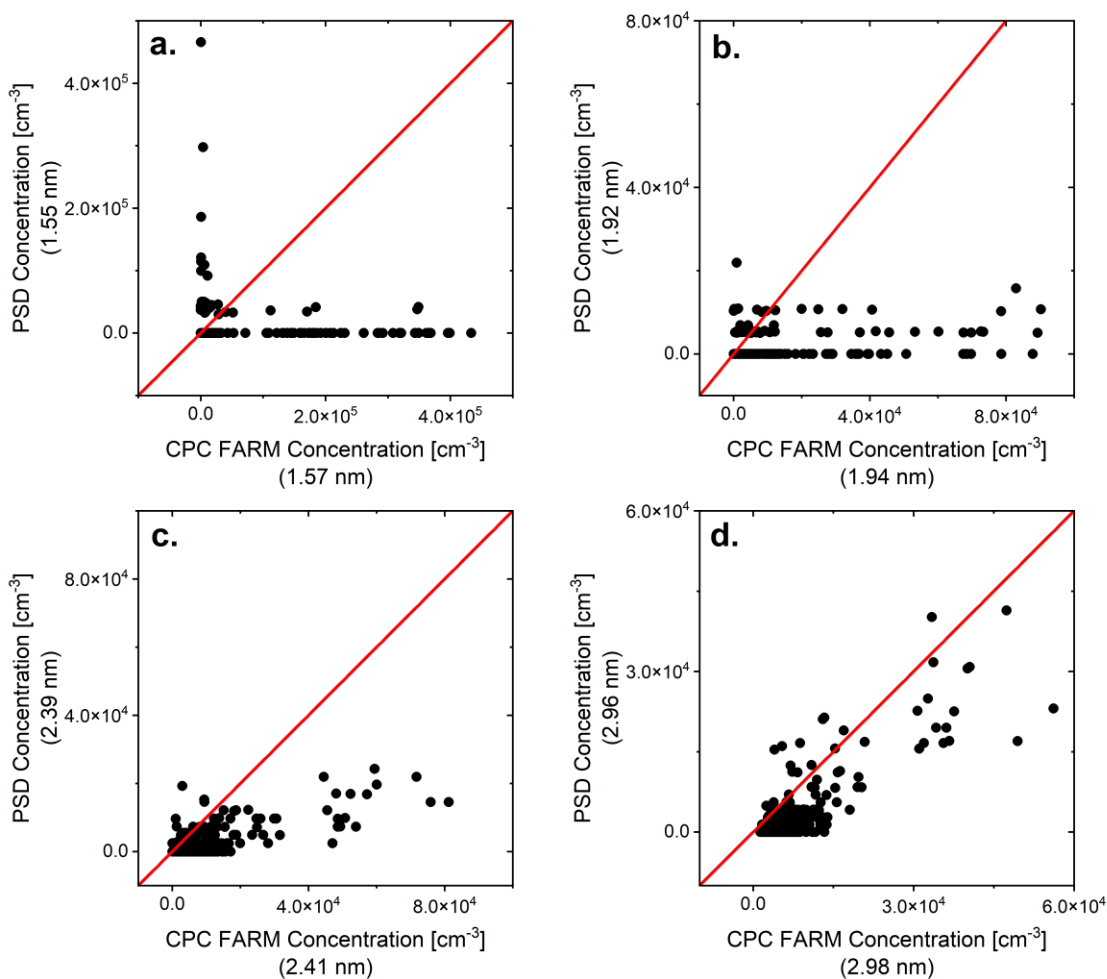


Figure S7 Parity plots comparing the PSD concentration at four size bins to their corresponding CPC FARM concentrations. The size-bin midpoints for each are displayed within the brackets. The solid red line is the parity line.

## References

- Riipinen, I., H. E. Manninen, T. Yli-Juuti, M. Boy, M. Sipilä, M. Ehn, H. Junninen, T. Petäjä, and M. Kulmala. 2009. “Applying the Condensation Particle Counter Battery (CPCB) to Study the Water-Affinity of Freshly-Formed 2–9 Nm Particles in Boreal Forest.” *Atmospheric Chemistry and Physics* 9 (10): 3317–30. <https://doi.org/10.5194/acp-9-3317-2009>.