Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

Supplementary Information

Improving the performance of portable aerosol size spectrometers for building dense monitoring networks

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1. Measurement of the combined factor of the transfer function, CPC detection efficiency, and penetration efficiency of the modified NanoScan SMPS

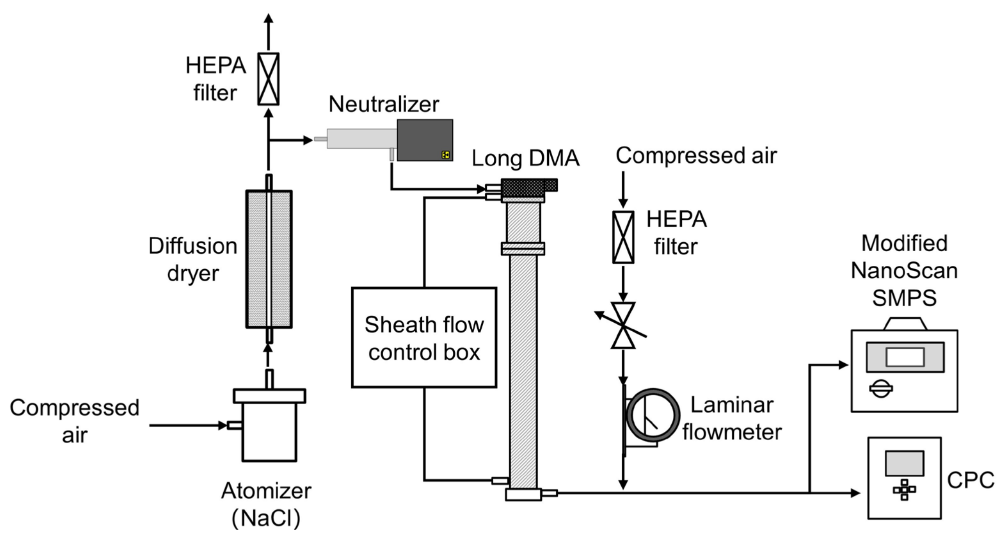

Figure S1. Instrument setup for measuring the combined factor of the transfer function, CPC detection efficiency, and penetration efficiency of the modified NanoScan SMPS

Retrieving the inverted PNSDs measured by the modified NanoScan SMPS requires a data inversion routine similar to those of the SMPS, but the transfer function, CPC detection efficiency, and penetration efficiency need to be measured first. In this study, we used the instrument set-up as shown in Figure S1 to measure the combined factor of them. This is similar to the Tandem DMA method and an iterative deconvolution procedure is used to analyze the data.[1] Polydisperse NaCl aerosols generated by a home-made collision atomizer with a dryer are conditioned by a soft X-ray charger (Model 3087, TSI corp.) and then classified with a long DMA (Model 3081, TSI corp.). The classified monodisperse aerosols are then split and the modified NanoScan SMPS and a CPC (Model 3772, TSI corp.) are measured in parallel. During each measurement period, the voltage of the long DMA was fixed and the voltage of the radial DMA on the modified NanoScan SMPS would scan for 120 seconds. The scan would repeat four times for each selected size. The measured concentrations by CPC are denoted as N_1 and averaged values during each scan were used. The number concentrations from the modified NanoScan SMPS are denoted as N_2. The ratio between N_1 and N_2 can be expressed by the transfer functions of the long DMA and the radial DMA as follows:[2]

$$\frac{N_2}{N_1}=\frac{\int_{Z_{p1}^*-\Delta Z_{p1}}^{Z_{p1}^*+\Delta Z_{p1}}\Omega_1\left(Z_p,Z_{p1}^*\right)\cdot\Omega_2\left(Z_p,Z_{p2}^*\right)\cdot\eta_{CPC}(Z_{p2}^*)\cdot\eta_{pene}(Z_{p2}^*)dZ_p}{\int_{Z_{p1}^*-\Delta Z_{p1}}^{Z_{p1}^*+\Delta Z_{p1}}\Omega_1\left(Z_p,Z_{p1}^*\right)dZ_p} \tag{S1}$$

where $\Omega_1\left(Z_p,Z_{p1}^*\right)$ and $\Omega_2\left(Z_p,Z_{p2}^*\right)$ are the transfer functions of the long DMA and the radial DMA, respectively. Z_{p1}^* is the electrical mobility of the particles classified by long DMA and Z_{p2}^* is the central electrical mobility of the particles measured by radial DMA; ΔZ_{p1} is the half-width of $\Omega_1\left(Z_p,Z_{p1}^*\right)$. $\eta_{CPC}(Z_{p2}^*)$ is the CPC detection efficiency (on the NanoScan SMPS) for particles with electrical mobility of Z_{p2}^*, and $\eta_{pene}(Z_{p2}^*)$ is the penetration efficiency through the modified NanoScan SMPS for particles with electrical mobility of Z_{p2}^*, both of which are assumed to be constant between $Z_{p1}^*-\Delta Z_{p1}$ to $Z_{p1}^*+\Delta Z_{p1}$. Here we use triangular-shaped transfer functions for both DMAs:[1, 3]

$$\Omega_1(Z_p,Z_{p1}^*)=\frac{\alpha_1}{2\beta_1}\left(\left|\frac{Z_{p1}^*}{Z_p}-(1+\beta_1)\right|+\left|\frac{Z_{p1}^*}{Z_p}-(1-\beta_1)\right|-2\left|\frac{Z_{p1}^*}{Z_p}-1\right|\right) \tag{S2}$$

$$\Omega_2(Z_p,Z_{p2}^*)=\frac{\alpha_2}{2\beta_2}\left(\left|\frac{Z_{p2}^*}{Z_p}-(1+\beta_2)\right|+\left|\frac{Z_{p2}^*}{Z_p}-(1-\beta_2)\right|-2\left|\frac{Z_{p2}^*}{Z_p}-1\right|\right) \tag{S3}$$

where α_1 and α_2 are the heights of the transfer functions of long DMA and radial DMA, and β_1 and β_2 are the half-widths of the transfer functions of long DMA and radial DMA. A combined factor of the transfer function, CPC detection efficiency, and penetration efficiency of the modified NanoScan SMPS are defined as:

$$\Omega_2'(Z_p,Z_{p2}^*)=\frac{\alpha_2'}{2\beta_2}\left(\left|\frac{Z_{p2}^*}{Z_p}-(1+\beta_2)\right|+\left|\frac{Z_{p2}^*}{Z_p}-(1-\beta_2)\right|-2\left|\frac{Z_{p2}^*}{Z_p}-1\right|\right) \tag{S4}$$

$$\alpha_2'=\eta_{CPC}(Z_{p2}^*)\cdot\eta_{pene}(Z_{p2}^*) \tag{S5}$$

With the known α_1 and β_1, a deconvolution procedure based on the method of least squares is used to retrieve α_2' and β_2.[1] Besides, using two identical long DMAs one can retrieve the values of α_1 and β_1, which are determined in this study to be 0.91 and 0.11, respectively. The results of $\Omega_2'(Z_p,Z_{p2}^*)$ are shown in Fig. S2, which is incorporated in the data inversion routine described below.

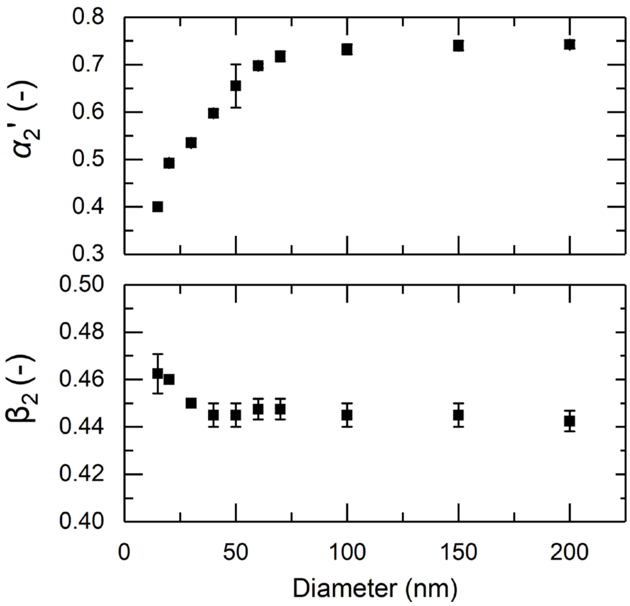

Figure S2. Results of the combined factor of the transfer function, CPC detection efficiency, and penetration efficiency of the modified NanoScan SMPS

2. Calculation of ion mobility ratio and PNSDs for the bipolar data

With the bipolar data from the reference SMPS and the modified NanoScan SMPS, the ion mobility ratio can be calculated with a formula:[4]

$$x = \exp[\ln\left(R^{+}_{d_\mathrm{p}} / R^{-}_{d_\mathrm{p}}\right) / 2] \tag{S6}$$

where $R^{+}_{d_\mathrm{p}}$ and $R^{-}_{d_\mathrm{p}}$ are the raw concentrations of positively and negatively charged particles in the size of d_p ($\mathrm{cm^{-3}}$). d_p is a size with little influence from larger aerosols carrying multiple charges and can be found with an empirical method.[4]

With the ion mobility ratios, charge fractions can be calculated:[4]

$$f(\pm q,\ d_\mathrm{p}) = \frac{e}{\sqrt{4\pi^2\varepsilon_0\alpha d_\mathrm{p}k_\mathrm{B}T}}\exp\frac{-[\pm q - \frac{2\pi\varepsilon_0\alpha d_\mathrm{p}k_\mathrm{B}T}{e^2}\ln(x)]^2}{2\frac{2\pi\varepsilon_0\alpha d_\mathrm{p}k_\mathrm{B}T}{e^2}}$$

$$\alpha = \begin{cases} 0.9630\times\exp(\dfrac{7.6019}{d_\mathrm{p}+2.2476}) & q=1 \\ 0.9826+0.9435\exp(-0.0478 d_\mathrm{p}) & q=2 \\ 1 & q\geq 3 \end{cases} \tag{S7}$$

where $f(\pm q, d_\mathrm{p})$ is the fraction of size d_p particles carrying $\pm q$ elementary charges (dimensionless); e is the elementary charge (C); ε_0 is the permittivity of vacuum ($\mathrm{C^2 \cdot N^{-1} \cdot m^{-2}}$); k_B is the Boltzmann's constant ($\mathrm{J \cdot K^{-1}}$); T is the temperature (K); α is the correction coefficient (dimensionless).

The relationship between measured raw concentrations and the desired concentration of aerosols that are being measured is:

$$R_{d_\mathrm{p}} = \int_0^\infty N(d_\mathrm{p}) \cdot G(i,\ d_\mathrm{p}) \cdot dd_\mathrm{p}\,, \quad i = 1, 2, \ldots 30 \tag{S8}$$

$$G(i,\ d_\mathrm{p}) = Q_\mathrm{a} \sum_{q=1}^{\infty} [f(+q,\ d_\mathrm{p}) + f(-q,\ d_\mathrm{p})] \cdot P\left(q, i, d_\mathrm{p}\right) \cdot dd_\mathrm{p} \tag{S9}$$

Where R_{d_p} is the sum of $R_{d_\mathrm{p}}^{+}$ and $R_{d_\mathrm{p}}^{-}$ (cm^{-3}), $N(d_\mathrm{p})dd_\mathrm{p}$ is the number concentration of all particles of size d_p (cm^{-3}), Q_a is the sample flowrate (cm^{3}·s^{-1}), $P\left(q, i, d_\mathrm{p}\right)$ is a combined factor of the transfer function of the DMA, counting efficiency of the CPC and penetration efficiency through the SMPS systems. For the reference SMPS, the transfer function of the long DMA was calculated by formula from Knutson and Whitby (1975)[5], the counting efficiency of the CPC was calculated with parameters provided by the instrument manual, and penetration efficiency was calculated with the equivalent pipe length method.[6] For the modified NanoScan SMPS, $P\left(q, i, d_\mathrm{p}\right)$ was obtained with the method described above. A linear inversion algorithm was used to obtain $N(d_\mathrm{p})dd_\mathrm{p}$ from R_{d_p}.[7]

3. Calculation of parameters that characterize the PNSDs

For number concentrations in nucleation mode, Aitken mode, accumulation mode, and the whole measured size range, the specific size range for each mode is not unified but set to be different for the original NanoScan SMPS and the modified NanoScan SMPS. The reason is that using a unified size range for two instruments requires interpolation of PNSDs to the same size bins, while the original NanoScan SMPS and the modified NanoScan SMPS have different midpoints of size bins and the bins are wide, meaning that interpolation would cause extra uncertainty. Besides, PNSDs measured by the original NanoScan SMPS and the modified NanoScan SMPS are not directly compared to each other, but both with those measured by the reference SMPS. Consistency with the reference SMPS is then compared to illustrate their performance. For the original NanoScan SMPS, these size ranges are 12 - 27nm (nucleation mode), 27 - 87 nm (Aitken mode), 87 - 274 nm (accumulation mode), and 12 - 274 nm (the whole size range). For the modified NanoScan SMPS, these size ranges are 12 - 25nm (nucleation mode), 25 - 90 nm (Aitken mode), 90 - 297 nm (accumulation mode), and 12 - 297 nm (the whole size range). The PNSDs obtained by the reference SMPS are linearly interpolated to the size bin of either the original NanoScan SMPS or the modified NanoScan SMPS, depending on which one it was being compared to, and number concentrations in different modes are then integrated. The geometric mean diameter and geometric standard deviations are calculated based on the PNSDs without interpolation for the reference SMPS.

4. Indoor aerosols measured by the modified NanoScan SMPS and the reference SMPS

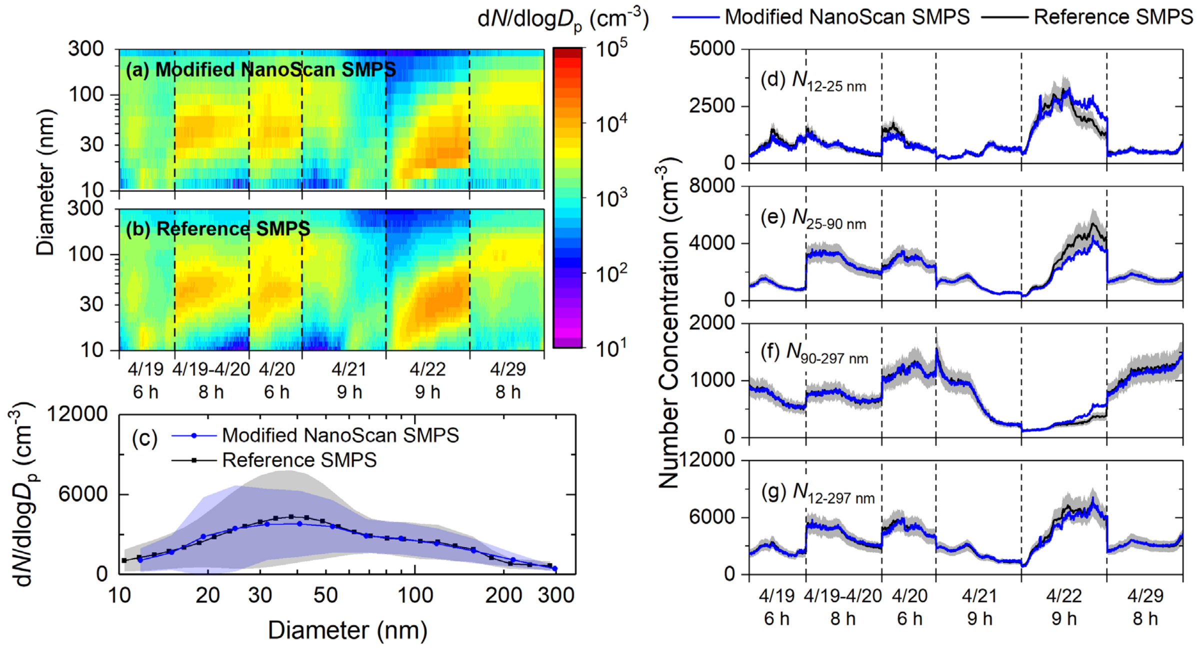

Figure S3. PNSDs of indoor aerosols for a total of 46 hours by (a) the modified NanoScan SMPS and (b) the reference SMPS. (c) The averaged size distributions and standard deviations during the measurement period, as denoted by line and shaded area, respectively. Integrated number concentrations in the size range of (d) 12 - 25 nm, (e) 25 - 90 nm, (f) 90 - 297 nm, (g) 12 - 297 nm of measured size distributions are shown, and the shaded area represent ±20% range of the reference SMPS. The time resolution for the PNSDs and thus the calculated parameters measured by the modified NanoScan SMPS and the reference SMPS are 75 s and 5 min, respectively.

5. Time series of indoor and outdoor aerosols for the original NanoScan SMPS

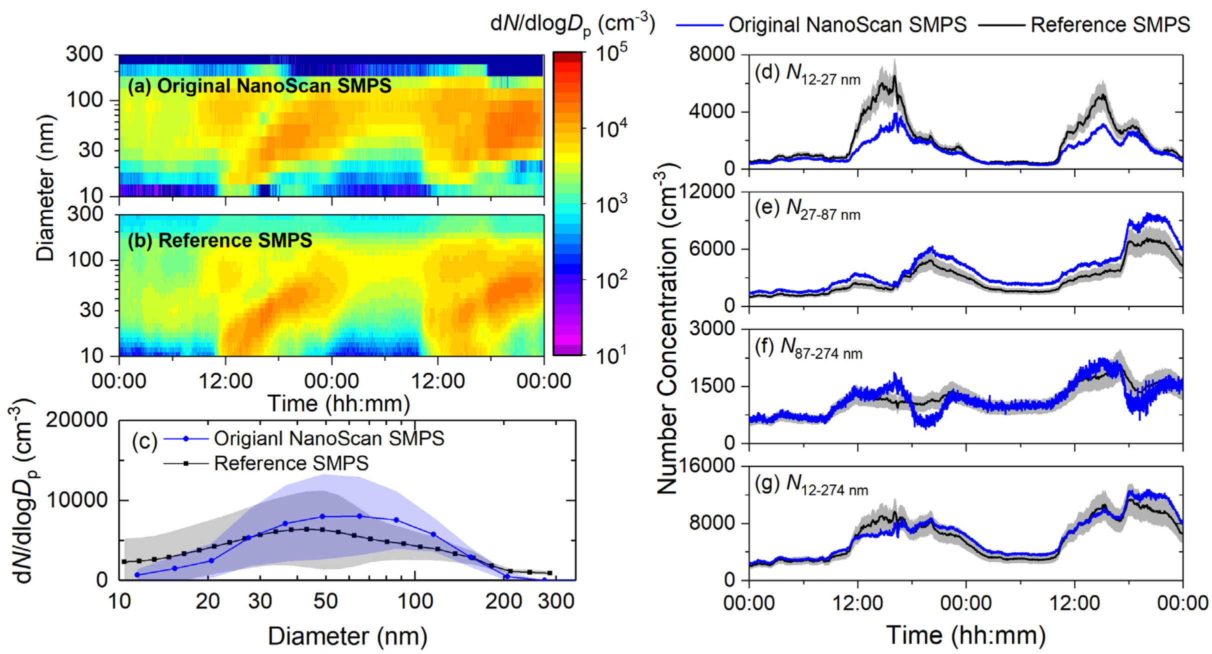

Figure S4. PNSDs of indoor aerosols for a total of 48 hours by (a) the original NanoScan SMPS and (b) the reference SMPS. (c) The averaged size distributions and standard deviations during the measurement period, as denoted by line and shaded area, respectively. Integrated number concentrations in the size range of (d) 12 - 27nm, (e) 27 - 87 nm, (f) 87 - 274 nm, (g) 12 - 274 nm of measured size distributions are shown, and the shaded area represent ±20% range of the reference SMPS. The time resolution for the PNSDs and thus the calculated parameters measured by the original NanoScan SMPS and the reference SMPS are 1 min and 5 min, respectively.

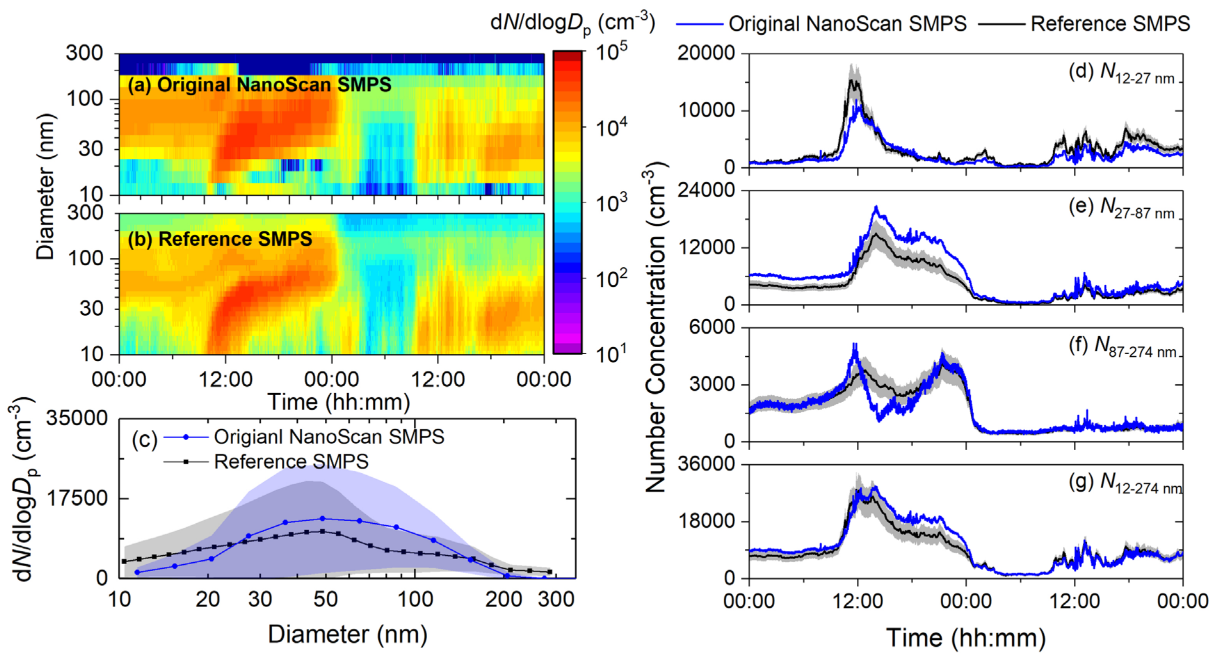

Figure S5. PNSDs of outdoor aerosols for a total of 48 hours by (a) the original NanoScan SMPS and (b) the reference SMPS. (c) The averaged size distributions and standard deviations during the measurement period, as denoted by line and shaded area, respectively. Integrated number concentrations in the size range of (d) 12 - 27nm, (e) 27 - 87 nm, (f) 87 - 274 nm, (g) 12 - 274 nm of measured size distributions are shown, and the shaded area represent ±20% range of the reference SMPS. The time resolution for the PNSDs and thus the calculated parameters measured by the original NanoScan SMPS and the reference SMPS are 1 min and 5 min, respectively.

6. Scatter plots for all measuring periods using indoor and outdoor aerosols

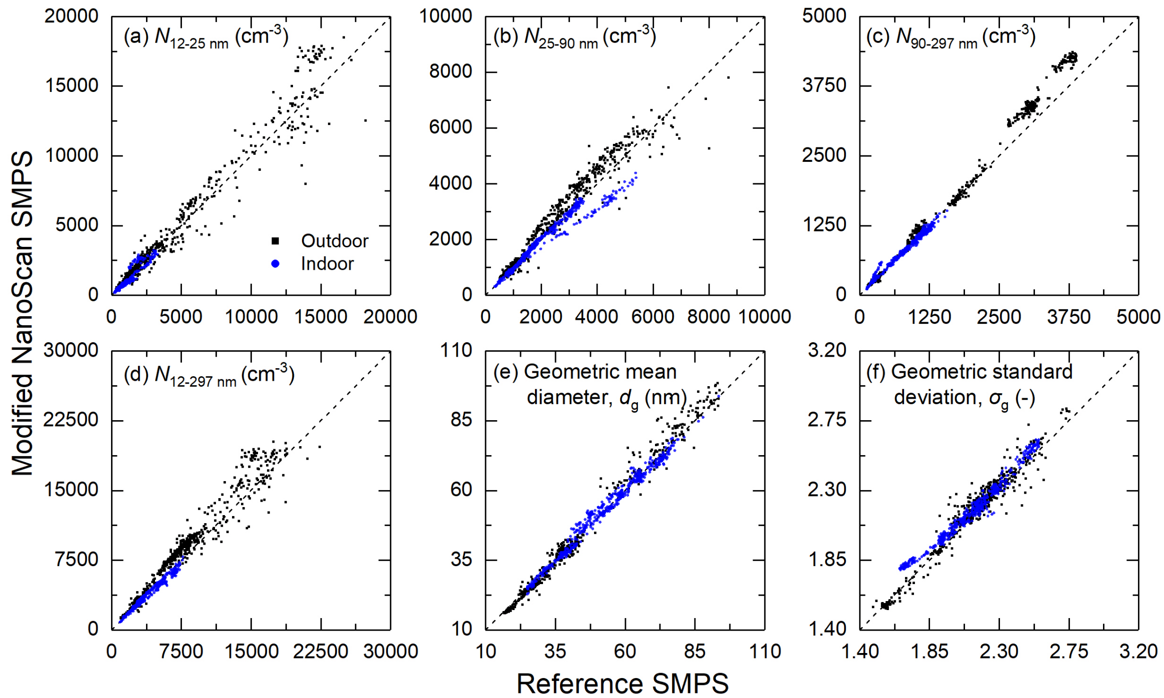

Figure S6. Comparing size distributions of outdoor and indoor aerosols for all measurement periods by the reference SMPS (x-axis) and the modified NanoScan SMPS (y-axis). Integrated number concentrations in the size range of (a) 12 - 25 nm, (b) 25 - 90 nm, (c) 90 - 297 nm, (d) 12 - 297 nm as well as (e) geometric mean diameter and (f) geometric standard deviation of measured size distributions are shown. The time resolution for the data is 5 min. Data points are shown in circles (blue) for indoor aerosols, and in squares (black) for outdoor aerosols. The dash lines are for guiding the eyes.

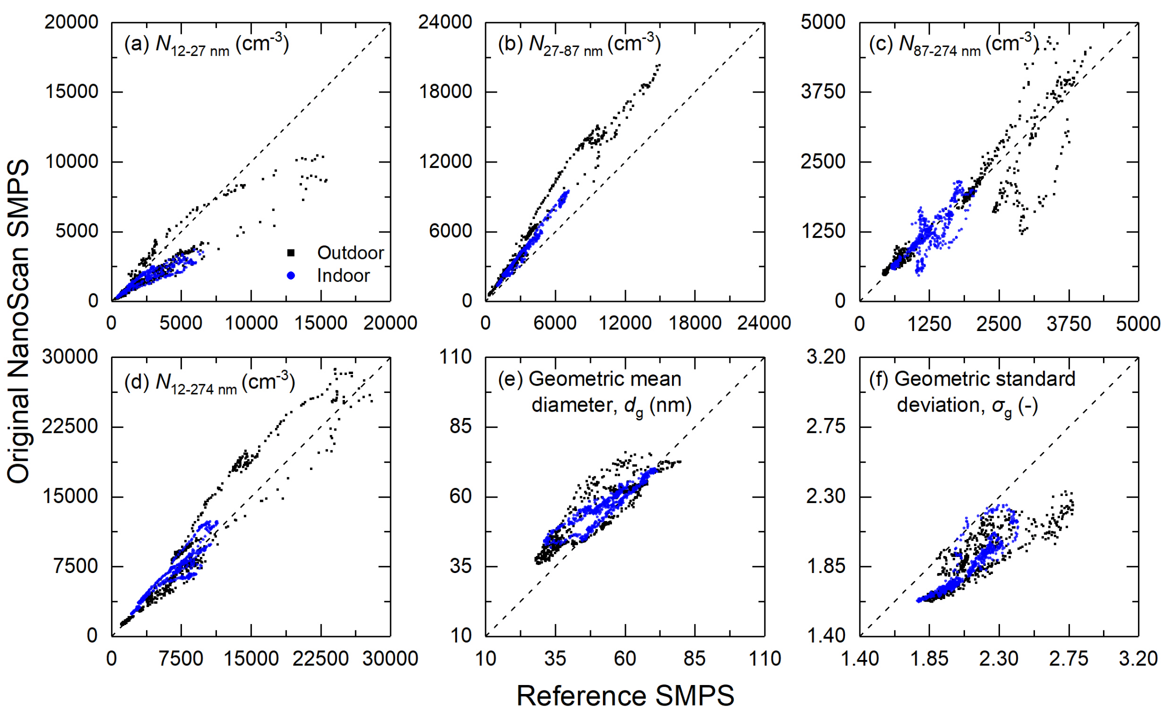

Figure S7. Comparing size distributions of outdoor and indoor aerosols for all measurement periods by the reference SMPS (x-axis) and the original NanoScan SMPS (y-axis). Integrated number concentrations in the size range of (a) 12 - 27nm, (b) 27 - 87 nm, (c) 87 - 274 nm, (d) 12 - 274 nm as well as (e) geometric mean diameter and (f) geometric standard deviation of measured size distributions are shown. The time resolution for the data is 5 min. Data points are shown in circles (blue) for indoor aerosols, and in squares (black) for outdoor aerosols. The dash lines are for guiding the eyes.

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