1 Supplement

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3	The Supplement contains the following sections:
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5	S1. Amount of data used for the analyses, fractions of accepted data and criteria for data removal
6	S2. Application of Reduced Major Axis (RMA) regression
7	S3. Derivation of Equation (8)
8	S4. Derivation of Equation (9)
9	S5. Analysis of the uncertainty related to the number of samples
0.1	S6. N _{CCN} (AOP) calculated by using the site-specific median SAE
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1 2 3	S1. Amount of data used for the analyses, fractions of accepted data and criteria for data removal Suspicious data within the whole dataset were removed according to the following criteria:
4 5	1) For the size distribution data, all the data with unexplainable spikes were removed manually;
6 7 8 9	There were 7587 available hourly-averaged-PNSD in MAO with 104 bins of each. A total of 5234 spikes were removed. This accounts for ~0.7% of the total number of bins. 423 out of 7587 (~5.6%) distributions had at least 1 bin(s) removed. A distribution with few missing bins are still usable if treated properly. Only 55 (~0.7%) distributions had more than 10 spikes removed.
11 12 13 14	Besides for MAO, other data sets rarely suffered from such spikes. 32 out of 11502 (~0.3%) distributions were removed for ASI. For SORPES and SMEAR2, less than 1% of distributions were removed. We didn't remove anything from PNSD of PVC and PNSD is not available in PGH.
15 16 17 18	2) for CCN measurements, insufficient water supply may cause underestimation of CCN, especially at lower supersaturation ratios (DMT, 2009). Nocon reading at lower SS% has a sudden drop a few hours before the similar sudden drop for higher SS% under such conditions, so data from such periods were removed;
19 20 21 22 23	Besides from the QC flag within MAO dataset, additional 55, 112,120 and 123 data points were removed at SS=0.25%,0.4%, 0.6% and 0.8% respectively, which accounts for ~0.7%-1.6% of total available data. For SORPES and SMEAR2 ~1% of total available data were removed. For ASI, PVC and PGH, no further treatment was applied besides the original QC flag.
242526	3) if any obvious inconsistencies between the AOPs and PNSD or between the Nccn and PNSD were found on closure study, all the data in the same hour were removed.
27 28 29 30	51 successive hours of data from PVC were removed before analysis, which account for ~3% of the data we used in this study. 84 sparse data points were removed from the ASI data set, which account for ~0.7% of total available data. For SORPES and SMEAR2 less than 1% of data were removed.
31 32 33 34 35	In total, additional quality control removes ~2%, ~3%, ~1% and 0% of the total available data in MAO, PVC, ASI and PGH respectively. The exact number for SORPES and SMEAR2 is not applicable since those 3 criteria are within the original data process procedures. However, a rough estimation of fractional data removed by such criteria are 0.5%~2%.
36 37	The total number of available hourly-averaged data, accepted data and removed data and the fractions of these are presented in Table TS1.

Table TS1. Number of data and fractions of removed data from all stations

			AOPs		CCN			Size distribution			
	Period		from	Addtional	finalized	from	Addtional		from	Addtional	finalized
	(hours)		Dataset	QC	data	Dataset	QC	finalized data	Dataset	QC	data
SMEAR2	8784	N_total			8626			6973-6994			8461
SMEARZ	6/64	Percentage			98.2%			79.3-79.6%			96.3%
SORPES	8760	N_total			5266			4825~4906			5440
SORPES	8700	Percentage			60.1%			55.1~56%			62.1%
ASI	12144	N_total	11851	84	11767	9894-10343		9894-10343	10931	32	10899
ASI	12144	Percentage	97.6%	0.7%	96.9%	81.5-85.2%		81.5-85.2%	90.0%	0.3%	89.7%
PVC	1800	N_total	1637		1637	1495		1495	1730	0	1730
PVC	1800	Percentage	90.9%		90.9%	83.1%		83.1%	96.1%	0.0%	96.1%
MAO	8160	N_total	7532		7532	7574-7653	55-123	7507-7541	7587	56	7541
MAO		Percentage	92.3%		92.3%	92.8~93.8%	0.7-1.5%	92-92.4%	93.0%	0.7%	92.4%
PGH	3498	N_total	3453		3453	3380-3420		3380-3420			
run	3498	Percentage	98.7%		98.7%	96.6-97.8%		96.6-97.8%			

S2. Application of Reduced Major Axis (RMA) regression

The Matlab code of Trujillo-Ortiz and Hernandez-Walls (2010) was applied to calculate the reduced major axis (RMA) regressions of $R_{\text{CCN/}\sigma}$ vs. BSF to get the slope and offset (a and b, respectively) of $R_{\text{CCN/}\sigma} = a$ BSF + b at the supersaturations (SS) of the CCN counters at the six stations. The results are shown in Table TS2. The values of Table TS2 were plotted as a function of SS in Fig. SF1 where also the fittings to the data are shown.

Table TS2. Slopes (a) and offsets (b) of $R_{CCN/\sigma} = a$ BSF + b obtained with RMA. The unit of the coefficients is $[N_{CCN}]/[\sigma_{sn}] = cm^{-3}/Mm^{-1}$.

	ie unit of the e	••••••	to 12 ft (CCN1/fosp	,	
Station	SS(%)	а	(a _{LOW} - анібн)	b	(b _{LOW} - b _{HIGH})
SMEAR II	0.1	175	(170 - 181)	-15.0	(-15.814.3)
	0.2	511	(502 - 521)	-49.8	(-51.248.5)
	0.5	1031	(1011 - 1050)	-110.1	(-112.9107.3)
	1	1492	(1459 - 1525)	-164.4	(-169.1159.7)
SORPES	0.1	121	(117 - 125)	-9.1	(-9.58.7)
	0.2	333	(326 - 341)	-25.8	(-26.625.0)
	0.4	657	(643 - 671)	-53.0	(-54.651.5)
	0.8	926	(905 - 946)	-76.6	(-78.974.4)
PGH	0.12	-53	(-54.651)	5.1	(5.0 - 5.2)
	0.22	161	(156 - 167)	-6.9	(-7.36.5)
	0.48	712	(689 - 734)	-37.6	(-39.236.0)
	0.78	849	(823 - 876)	-44.1	(-46.042.3)
PVC	0.15	517	(500 - 534)	-42.4	(-44.540.3)
	0.25	989	(956 - 1023)	-85.8	(-89.981.7)
	0.4	1465	(1416 - 1514)	-130.7	(-136.7124.7)
	1	2452	(2369 - 2536)	-223.5	(-233.7213.3)
MAO	0.25	472	(462 - 481)	-46.7	(-48.145.4)
	0.4	833	(817 - 849)	-83.4	(-85.681.1)
	0.6	1188	(1163 - 1213)	-122.1	(-125.6118.7)
	1.1	2128	(2065 - 2190)	-226.5	(-234.9218.2)
ASI	0.1	150	(147 - 153)	-15.9	(-16.315.4)
	0.2	319	(312 - 325)	-34.0	(-34.933.1)
	0.4	372	(365 - 380)	-39.8	(-40.938.7)
	0.8	406	(397 - 414)	-42.4	(-43.641.1)

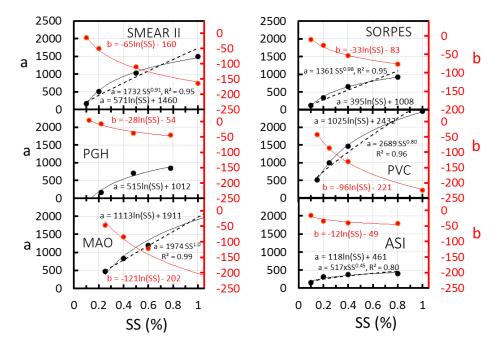


Figure SF1. The RMA-derived coefficients a and b of each station (Table ST2) as a function of supersaturation. Two types of functions, a logarithmic and a power fuction were fitted to the coefficient a, to coefficient b only a logarithmic function. The squared correlation coefficients R^2 are shown only for the power function fittings, for the logarithmic fittings they were all > 0.99. The unit of the coefficients is $[N_{CCN}]/[\sigma_{sn}] = cm^{-3}/Mm^{-1}$.

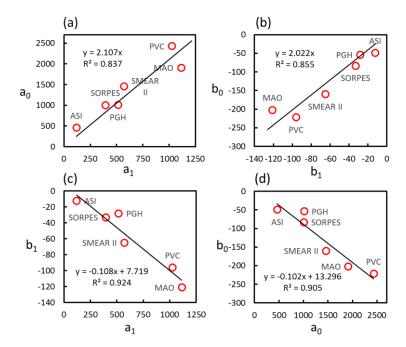


Figure SF2. Relationship between the coefficients a_0 , a_1 , b_0 and b_1 shown in Fig. SF1 that were obtained from the fitting of $a = a_1 ln(SS) + a_0$ and $b = b_1 ln(SS) + b_0$ with the data in Table TS2. SF1. a) a_0 vs. a_1 , b) b_0 vs. b_1 , c) b_1 vs. a_1 , d) b_0 vs. a_0 . The unit of the coefficients is $[N_{CCN}]/[\sigma_{sp}] = cm^{-3}/Mm^{-1}$.

When using RMA-derived slopes and offsets of $R_{CCN/\sigma} = a$ BSF + b the relationship between the factor a_1 and SAE became $a_1 \approx 391 \cdot SAE_{10}$ (Fig. SF3).

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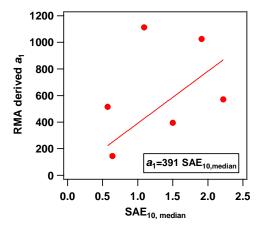


Figure SF3: Relationship between RMA-derived a_1 and SAE_{10,median}.

This was further used to estimate CCN number concentration in the formula

$$N_{CCN}(RMA) \approx \left(\ln \left(\frac{SS}{0.12 \pm 0.02} \right) a_1(BSF - BSF_{min}) + R_{min} \right) \sigma_{sp}$$
 (ES1)

The derivation of (ES1) is presented in supplement S4. $N_{CCN}(RMA)$ is in general in agreement with the $N_{CCN}(AOP_2)$ and $N_{CCN}(meas)$. However for SS \sim 0.1% the performance of RMA method is poor. At SS \sim 0.1%, R^2 between $N_{CCN}(RMA)$ and $N_{CCN}(meas)$ is much lower than between $N_{CCN}(AOP_2)$ and $N_{CCN}(meas)$ which indicates using RMA gives very uncertain results ast lowest SS. Nevertheless, for SS>0.15%, OLS-derived $N_{CCN}(AOP_2)$ and RMA-derived $N_{CCN}(RMA)$ agree well. Figure SF4 shows the scatter plots for $N_{CCN}(RMA)$ vs. $N_{CCN}(meas)$ and R^2 and bias. The R^2 are between 0.5 \sim 0.85 and bias are within 0.5 \sim 2 when SS>0.15% for $N_{CCN}(AOP_2)$.

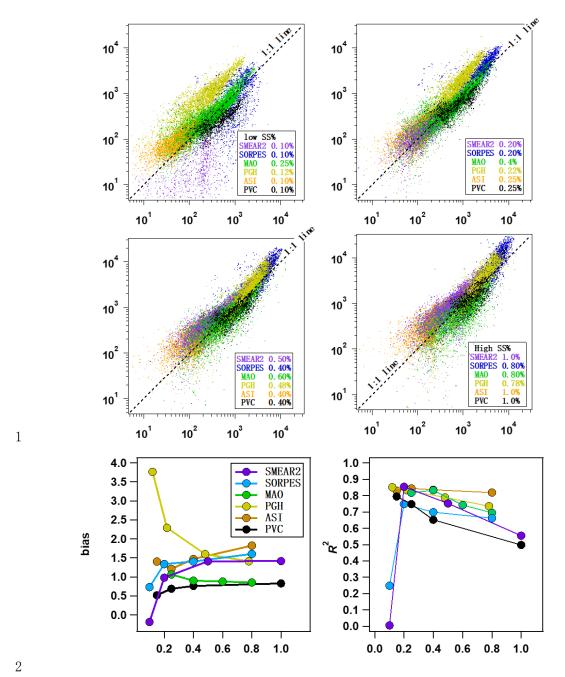


Figure SF4. Statistics of $N_{\rm CCN}({\rm RMA})$ from parameterization in Eq. (ES1). $N_{\rm CCN}({\rm RMA})$ vs. $N_{\rm CCN}({\rm meas})$ at different sites at different supersaturations, bias = $N_{\rm CCN}({\rm RMA})/N_{\rm CCN}$ (meas) at different sites and supersaturations, and R^2 of the linear regression of $N_{\rm CCN}({\rm RMA})$ vs. $N_{\rm CCN}$ (meas) at different sites and supersaturations. same as Figure 8, but for $N_{\rm CCN}({\rm RMA})$.

The choice between OLS and RMA

- 2 Many studies use the reduced major axis (RMA) method instead of ordinary least squares (OLS)
- 3 method to define a line of best fit for a bivariate relationship when variable represented on the
- 4 X-axis contains measurement error. Smith (2009) point out that the major difference RMA and
- 5 OLS is not in the difference in the assumption made about the distribution of error, but in their
- 6 symmetry/asymmetry property. The reduced major axis regression is to describe the symmetric
- 7 relationship between two variables and not for predictive use of the variable x with respect to
- 8 y or y with respect to x (Smith, 2009). For predictive use OLS is preferred.

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References

- 12 Smith, R. J.: Use and Misuse of the Reduced Major Axis for Line-Fitting, Am. J. Phys.
- 13 Anthropol., 140, 476–486, doi:10.1002/ajpa.21090, 2009

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- 15 Trujillo-Ortiz, A. and Hernandez-Walls, R.: gmregress: Geometric Mean Regression (Reduced
- 16 Major Axis Regression), a MATLAB file available at:
- 17 http://www.mathworks.com/matlabcentral/fileexchange/27918-gmregress, 2010.

1 S3. Derivation of Equation (8)

2 a) Using slopes and offsets from ordinary linear regressions

$$\begin{split} &N_{CCN}(AOP) = \left(a_{SS}BSF + b_{ss}\right)\sigma_{sp} = \left(\left(a_{1}\ln(SS) + a_{0}\right)BSF + b_{1}\ln(SS) + b_{0}\right)\sigma_{sp} \\ &R_{CCN/\sigma} = \frac{N_{CCN}(AOP)}{\sigma_{sp}} = a_{SS}BSF + b_{ss} = \left(a_{1}\ln(SS) + a_{0}\right)BSF + b_{1}\ln(SS) + b_{0} \\ &\text{Linear regressions of the coefficients in Table 2 yield} \\ &a_{0} \approx (2.38 \pm 0.06)a_{1}, b_{0} \approx (2.33 \pm 0.03)b_{1}, b_{1} \approx -(0.096 \pm 0.013)a_{1} + (6.0 \pm 5.9) \\ \Rightarrow \\ &a_{1}\ln(SS) + a_{0} \approx a_{1}\ln(SS) + (2.38 \pm 0.06)a_{1} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06)) \\ &b_{1}\ln(SS) + b_{0} \approx b_{1}\ln(SS) + (2.33 \pm 0.03)b_{1} = b_{1}(\ln(SS) + (2.33 \pm 0.03)) \\ \approx \left(-(0.096 \pm 0.013)a_{1} + (6.0 \pm 5.9)\right)(\ln(SS) + (2.33 \pm 0.04)) \\ \Rightarrow \\ &R_{CCN/\sigma} = \left(a_{1}\ln(SS) + a_{0}\right)BSF + b_{1}\ln(SS) + b_{0} \\ \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF + \left(-(0.096 \pm 0.013)a_{1} + (6.0 \pm 5.9)\right)(\ln(SS) + (2.33 \pm 0.03)) \\ &\text{Approximation, since } (2.33 \pm 0.03) \approx (2.38 \pm 0.06) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.07)) + (6.0 \pm 5.09)(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.07)) + (6.0 \pm 5.09)(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.07)) + (6.0 \pm 5.09)(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.07)) + (6.0 \pm 5.09)(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.07)) + (6.0 \pm 5.09)(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.06)) \\ \Rightarrow \\ &R_{CCN/\sigma} \approx a_{1}(\ln(SS) + (2.38 \pm 0.06))BSF - (0.096 \pm 0.013)a_{1}(\ln(SS) + (2.38 \pm 0.06)) \\ \end{cases}$$

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$$\kappa_{CCN/\sigma} \approx a_1 \left(\ln(SS) + (2.38 \pm 0.06) \right) BSF - (0.096 \pm 0.013) a_1 \left(\ln(SS) + (2.38 \pm 0.07) \right) + (6.0 \pm 5.09) \left(\ln(SS) + (2.38 \pm 0.06) \right)$$

$$\approx a_1 \left(\ln(SS) + (2.38 \pm 0.06) \right) \left(a_1 (BSF - (0.096 \pm 0.013)) + (6.0 \pm 5.9) \right)$$

$$\approx \left(\ln(SS) - \ln(0.093 \pm 0.006) \right) \left(a_1 (BSF - (0.097 \pm 0.013)) + (6.0 \pm 5.9) \right)$$

$$\approx \ln \left(\frac{SS}{0.093 \pm 0.006}\right) \left(a_1(BSF - (0.096 \pm 0.013)) + (6.0 \pm 5.9)\right)$$

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b) Using slopes and offsets from reduced major axis regressions

$$\begin{split} N_{CCN}(RMA) &= \left(a_{SS}BSF + b_{ss}\right)\sigma_{sp} = \left(\left(a_{1}\ln(SS) + a_{0}\right)BSF + b_{1}\ln(SS) + b_{0}\right)\sigma_{sp} \\ R_{CCN/\sigma} &= \frac{N_{CCN}(RMA)}{\sigma_{sp}} = a_{SS}BSF + b_{ss} = \left(a_{1}\ln(SS) + a_{0}\right)BSF + b_{1}\ln(SS) + b_{0} \end{split}$$

6 RMA regressions $\Rightarrow a_0 \approx (2.11 \pm 0.16) a_1, b_0 \approx (2.02 \pm 0.16) b_1, b_1 \approx -(0.108 \pm 0.016) a_1 + (7.7 \pm 11.0)$

The same steps as above in (a) \Rightarrow

$$\begin{split} &R_{CCN/\sigma} \approx \left(\ln(SS) + (2.11 \pm 0.16)\right) \left(a_1(BSF - (0.108 \pm 0.016)) + (7.7 \pm 11.0)\right) \\ &\approx \left(\ln(SS) - \ln(0.12 \pm 0.02)\right) \left(a_1(BSF - (0.108 \pm 0.016)) + (7.7 \pm 11.0)\right) \\ &\approx \ln \left(\frac{SS}{0.12 \pm 0.02}\right) \left(a_1(BSF - (0.11 \pm 0.02)) + (8 \pm 11)\right) \end{split}$$

- 1 S4. Derivation of Equation (9)
- 2 If the original slopes and offsets were calculated using ordinary linear regressions

$$N_{CCN}(AOP) \approx \ln\left(\frac{SS}{0.093 \pm 0.006}\right) \left(a_1(BSF - BSF_{min}) + C\right)\sigma_{sp}$$
,

where C is an unknown constant.

If
$$BSF = BSF_{min}$$

$$\Rightarrow a_1(BSF - BSF_{min}) = 0$$

$$\Rightarrow N_{CCN}(AOP, BSF_{min}) \approx \ln\left(\frac{SS}{0.093 \pm 0.006}\right) C \cdot \sigma_{sp}$$

$$\Leftrightarrow C \approx \frac{1}{\ln\!\left(\frac{SS}{0.093 \pm 0.006}\right)} \frac{N_{CCN}(AOP,BSF_{\min})}{\sigma_{sp}} \approx \frac{1}{\ln\!\left(\frac{SS}{0.093 \pm 0.006}\right)} R_{\min}$$

 \rightrightarrows

$$N_{CCN}(AOP) \approx \ln\left(\frac{SS}{0.093 \pm 0.006}\right) \left(a_1(BSF - BSF_{\min}) + \frac{1}{\ln\left(\frac{SS}{0.093 \pm 0.006}\right)}R_{\min}\right)\sigma_{sp}$$

$$3 \qquad \approx \left(\ln \left(\frac{SS}{0.093 \pm 0.006} \right) a_1 (BSF - BSF_{\min}) + R_{\min} \right) \sigma_{sp}$$

- 4 If the original slopes and offsets were calculated using reduced major axis regressions
- 5 $N_{CCN}(RMA) \approx \left(\ln \left(\frac{SS}{0.12 \pm 0.02} \right) a_1(BSF BSF_{min}) + R_{min} \right) \sigma_{sp}$

S5. Analysis of the uncertainty related to the number of samples

- The following procedure was used for testing how different values would be change if the number of samples decrease.
 - 1. For each site 2%,3%,5%,10%,20%,30%,50% and 100% of samples were taken from the whole period.
- 6 2. The slope and offset a, b, BSF_{min} (calculated as the 1st percentile of the BSF data) and SAE₁₀,median were calculated from the randomly chose subsets.
 - 3. The a, b, BSF_{min} and SAE₁₀,median should be slightly different if the sub-set is different. Therefore the random sampling was repeated 100 times resulting in 100 different results
 - 4. The averages and standard deviations of the 100 results were calculated and plotted below for all the sites. The average are the reds circles and the stds the error bars in the plots.

Results of the analysis

The averages of a,b, BSF_{min} and SAE_{10} ,median don't have clear dependence on the number of samples. However, the uncertainty is very large at low number of samples and decreases with increasing number of samples. The uncertainties depend on parameter and site. The plots suggest that if the number of samples is larger than 1000 the uncertainty is low enough. For example, the std of BSF_{min} is ~0.0005-0.005 and the std of SAE_{10} ,median is ~0.01-0.02. For a and b, std is ~10% of the a average value.

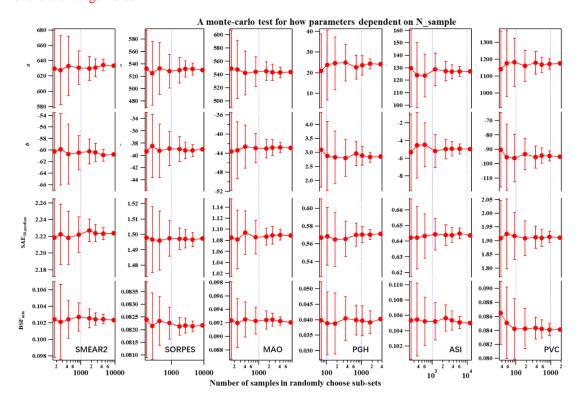


Figure SF5. A monte-carlo test on the dependence of the parameters a, b, $SAE_{10,median}$ and BSF_{min} on the number of hourly-averaged samples. The average are the reds circles and the stds the error bars.

S6. N_{CCN}(AOP) calculated by using the site-specific median SAE

3 The general combined parameterization was presented in the main test as Eq.10:

$$4 \qquad N_{CCN}(AOP_2) \approx \left(a_1 \ln \left(\frac{SS}{0.093 \pm 0.006}\right) (BSF - BSF_{min}) + R_{min}\right) \sigma_{sp}$$

$$\approx \left((286 \pm 46) \text{SAE} \cdot \ln \left(\frac{SS}{0.093 \pm 0.006}\right) (BSF - BSF_{min}) + (5.2 \pm 3.3)\right) \sigma_{sp}$$

In the main text, we used SAE of hourly-averaged σ_{sp} to estimate $N_{CCN}(AOP_2)$. Here we give another alternative for using this formula by using the site-specific median SAE values (Table 4 in the main text). The N_{CCN}(AOP) calculated by using the site-specific median SAE is compared with N_{CCN}(meas) in Figure SF6. When compared with N_{CCN}(AOP) calculated by using the hourlyvarying SAE (Fig. 8 in the main text), it is obvious that the two approaches are competitive with each other. A comparison of the biases and correlation coefficients is presented in Table TS3 below. For some combinations of SS and sites, the site-specific median SAE gives a smaller R² and a higher bias than the hourly SAE especially for ASI.

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However, site-specific median SAE is very probably always positive, while the hourly SAE is sometimes negative which may yield negative N_{CCN}(AOP). For the 6 sites of this study, the fraction of negative SAE of all hourly data varied between 0-6%. To estimate N_{CCN} for a site with a large fraction of negative SAE, we recommend to use site-specific median SAE.

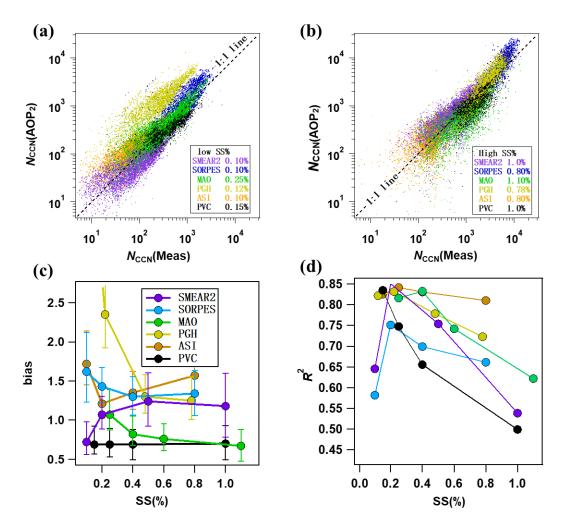


Figure SF6. Same as Figure 8 in the main text, but $N_{CCN}(AOP)$ calculated by using the site-specific median SAE. For details see the caption of Fig. 8 in the main text

-	Fraction of			N _{CCN} (AOP) calculated using				
	hourly		hourly-va	arying SAE	median SA	median SAE		
Station	SAE < 0	SS	R^2	bias	R^2	bias		
П		0.10%	0.675	0.72	0.657	0.72		
SMEAR	0.0%	0.20%	0.832	1.09	0.850	1.07		
ÆΕ	0.0%	0.50%	0.719	1.26	0.754	1.24		
S		1.00%	0.504	1.20	0.554	1.18		
Š		0.10%	0.595	1.61	0.587	1.62		
SORPES	0.0%	0.20%	0.773	1.36	0.751	1.43		
OR	0.070	0.40%	0.650	1.22	0.699	1.30		
		0.80%	0.636	1.27	0.687	1.34		
		0.25%	0.840	1.24	0.816	1.07		
MAO	6.0%	0.40%	0.834	0.97	0.832	0.82		
Ξ	0.070	0.60%	0.725	0.91	0.742	0.76		
		1.10%	0.583	0.71	0.622	0.67		
		0.12%	0.852	4.53	0.821	4.71		
PGH	4.4%	0.22%	0.871	2.13	0.832	2.35		
Ъ		0.48%	0.784	1.13	0.779	1.30		
		0.78%	0.703	1.07	0.723	1.25		
	0.04%	0.10%	0.872	1.92	0.828	1.72		
ASI		0.20%	0.923	1.41	0.844	1.21		
⋖		0.40%	0.900	1.61	0.836	1.35		
		0.80%	0.857	1.90	0.818	1.57		
	0.3%	0.15%	0.880	0.71	0.835	0.69		
PVC		0.25%	0.780	0.70	0.747	0.69		
P		0.40%	0.687	0.71	0.655	0.69		
		1.00%	0.519	0.71	0.499	0.70		

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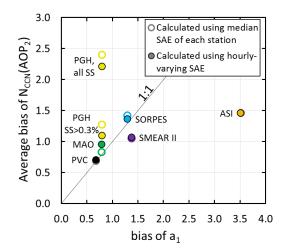
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The bias of N_{CCN}(AOP₂) presented in Table TS3 was calculated from the ratio $N_{CCN}(AOP_2)/N_{CCN}(meas)$. Since $N_{CCN}(AOP_2) \approx (a_1 ln(SS/0.093)(BSF - BSF_{min}) + R_{min})\sigma_{sp}$ it is obvious that biases of a_1 affect the bias of $N_{CCN}(AOP_2)$. If we consider the a_1 values in the main text Table 4 as the accurate station-specific values then the fitted line $a_1 = 286 \cdot SAE$ overestimates or underestimates a, by +37%, +30%, -20%, -32%, -20% and +251% for SMEAR II, SORPES, PGH, PVC, MAO and ASI, respectively. These values were calculated from 100% (286 SAE – a_1)/ a_1 . The biases of a_1 calculated from 286·SAE/ a_1 are therefore 1.373, 1.295,0.796, 0.675, 0.792, 3.509 for the respective stations. The average biases of $N_{CCN}(AOP_2)$ at all supersaturations of each station presented in Table TS3 are compared with the biases of a₁ in Figure SF7. For each station two values are shown: the average bias of N_{CCN}(AOP₂) calculated by using the median SAE of each station and the average bias of N_{CCN}(AOP₂) calculated by using the hourly-varying SAE. For PGH the average bias of N_{CCN}(AOP₂) at all supersaturations and at SS> 0.3% are shown because the biases at the lowest supersaturations are anomalously high. The plot shows that for most stations the bias of N_{CCN}(AOP₂) can be explained by the bias of a₁: when a₁ is underestimated so is N_{CCN}(AOP₂) and when a₁ is overestimated so is N_{CCN}(AOP₂). PGH is the only exception to this, especially at the lowest two supersaturations (SS = 0.12% and 0.22%) and we cannot explain why. For ASI the bias of N_{CCN}(AOP₂) is clearly smaller than the bias of a₁. This would happen when in the formula $N_{CCN}(AOP_2) \approx (a_1 \cdot ln(SS/0.093)(BSF - BSF_{min}) + R_{min})\sigma_{sp}$ both SAE and BSF are very small and especially when BSF is close to BSF_{min}. Both of these would take place when aerosol is dominated by large aerosols. This is true especially for ASI, a site dominated by marine aerosols.



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Figure SF 7. Biases of $N_{CCN}(AOP_2)$ vs the bias of a_1 calculated from $a_1 = 286 \cdot SAE$. The biases of $N_{CCN}(AOP_2)$ are the averages of biases at all supersaturations presented in Table TS3. For each station two values are shown: the average bias of $N_{CCN}(AOP_2)$ calculated by using the median SAE of each station (open circles) and the hourly-varying SAE (filled circles). For PGH the average bias of $N_{CCN}(AOP_2)$ at all supersaturations and at SS> 0.3.