

SUPPLEMENTARY MATERIAL

SENSITIVITY ANALYSIS OF CCN PARAMETERIZATION

ACTIVATION CURVE DATA REFINEMENT CRITERIA

A first set of sensitivity calculations were conducted to investigate how the median D_{50} values, and hence, the predicted CCN concentrations, depend on the numerical limits used in the four activated fraction curve cleaning criteria as first introduced in section 2.3.1. The calculations were done by using one-factor-at-a-time analysis, where one of the four criteria limits was altered by varying but appropriate amounts whilst retaining physically reasonable behaviour for cloud activation, and the rest kept constant. The sensitivity analysis for the predicted CCN concentrations was conducted on a one-year sample analysis period (2009).

A final calculation was run, changing the limit of all four criteria at once, aiming to retain only activation curves that were closest to ideality, whilst still considering the influence of instrumental noise. For this calculation, the mean change in median D_{50} value was 3.95% and the mean percent change in predicted CCN concentration was 5.16%, of which both values represent the largest changes out of all sensitivity runs. These results demonstrate D_{50} having little sensitivity to the activation curve filtering criteria initially selected. Predicted CCN concentrations also exhibit minimal sensitivity to the activation curve filtering criteria initially selected as a possible result of predicted CCN concentrations having low sensitivity to changes in D_{50} and/or D_{50} changes being so minimal their effect fails to be visibly notable. Confidence is provided in the parameterization robustness with respect to the initial activation curve filtering criteria.

Table S1: Values used in the curve filtering criteria as outlined in section 2.3.1, and their respective influence on percentage changes in median D_{50} value and predicted CCN concentrations. Those values highlighted in red indicate a change from the original filtering criteria values used. Percentage changes are reported to 2 d.p.

Run	Criteria 1	Criteria 2	Criteria 3	Criteria 4	Mean percent change in median D_{50} value / %	Mean percent change in predicted CCN concentrations / %
Base Run	0.7	1.2	-1	-2	0.00	0.00
1	0.6	1.2	-1	-2	2.02	2.28
2	0.8	1.2	-1	-2	0.99	1.15
3	0.9	1.2	-1	-2	3.45	4.27
4	0.7	1	-1	-2	2.12	2.41
5	0.7	1.1	-1	-2	0.11	0.04
6	0.7	1.3	-1	-2	0.03	0.00

7	0.7	1.2	-0.5	-2	0.02	0.04
8	0.7	1.2	-1.5	-2	0.00	0.00
9	0.7	1.2	-1	-1	1.01	1.09
10	0.7	1.2	-1	-3	0.08	0.14
11	0.9	1.1	-0.5	-1	3.95	5.16

AIR MASS SECTOR DEFINITION

The second sensitivity analysis performed in this study, focused on the definition of the air mass sectors for classification of the D_{50} values, as first introduced in section 2.3.1. The study investigated the impact of the air mass sector definition on the sector classification of each period identified as cloudy, in turn, influencing the median D_{50} values used as the input of the parameterization, and consequently the impact of this on the predicted CCN concentrations as the parameterization output. The sensitivity runs involved changing the angles from the central point, taken as Pallas research station ($67^{\circ} 58' N$, $24^{\circ} 07' E$), used in the sector definition.

Table S2. Sensitivity analysis for airmass sector selection. Base Run is the sector definition used in the study and Runs 1-6 are for the sensitivity analyses.

Run	Arctic sector angle north of Pallas / °	East sector angle north of Pallas / °	West sector angle north of Pallas / °	Mean percent change in median D_{50} value / %	Mean percent change in predicted CCN concentrations / %
Base Run	0 – 90, 270 – 360	90 – 195	195 – 270	0.00	0.00
1	0 – 60, 270 – 360	60 – 195	195 – 270	1.91	2.50
2	0 – 90, 300 – 360	90 – 195	195 – 300	0.86	1.49
3	0 – 90, 270 – 360	90 – 185	185 – 270	1.66	3.08
4	0 – 90, 250 – 360	90 – 195	195 – 250	2.06	2.24
5	0 – 90, 270 – 360	90 – 205	205 – 270	2.24	4.23
6	0 – 75, 255 – 360	75 – 195	195 – 255	2.10	2.70

Further work on the sensitivity of the parameterization and the consequent outputs, on the air mass sector definition may include testing requirements for the trajectories to spend a given fraction of transport time in an air mass sector before being giving an overall assignment.

Table S3: Statistics of the D_{50} values obtained from the parameterization based on classifications by air temperature and air mass sector (1 d.p). D_{50} values in nm.

Temperature Range / °C	Airmass Sector	Median D_{50} Value	25 th Percentile D_{50} Value	75 th Percentile D_{50} Value	Mean D_{50} Value	No. of Datapoints
-15 - -10	Arctic	88.0	76.3	138.5	104.7	31
-15 - -10	East	96.0	82.5	109.3	97.2	61
-15 - -10	West	99.0	90.0	108.0	101.1	28
-10 - -5	Arctic	98.0	83.0	131.5	111.6	144
-10 - -5	East	104.0	86.0	125.0	109.1	404
-10 - -5	West	110.5	91.0	129.0	112.8	130
-5 - 0	Arctic	89.0	72.0	112.0	100.7	229
-5 - 0	East	108.0	93.0	135.0	118.5	680
-5 - 0	West	112.0	93.0	138.0	120.5	401
0 - 5	Arctic	90.0	75.0	115.0	100.5	203
0 - 5	East	105.0	86.0	125.0	110.3	327
0 - 5	West	115.5	99.0	134.0	121.6	298
5 - 10	Arctic	105.0	87.0	126.3	109.9	329
5 - 10	East	122.0	104.3	152.0	131.1	323
5 - 10	West	141.0	107.5	163.5	144.2	99
10 - 15	Arctic	127.5	99.0	182.0	143.3	130
10 - 15	East	144.0	113.0	171.5	141.0	172
10 - 15	West	187.0	112.0	209.0	156.5	54

Table S4. Seasonal variation of median aerosol concentrations (in $\#/\text{cm}^3$) in the nucleation mode ($D_p < 20 \text{ nm}$), the Aitken mode ($20 \text{ nm} < D_p < 100 \text{ nm}$), and the accumulation mode ($D_p > 100 \text{ nm}$) at Pallas for 2005 – 2020, and Värriö for 1997-2021.

Pallas	Annual	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
Nucl	31	15	21	47	88	53	33	17	24	47	44	19	15
Ait	250	66	119	132	287	450	519	409	435	386	158	58	50
Acc	113	47	66	82	107	174	207	345	212	109	44	28	31
Tot	588	155	275	335	724	961	1043	1037	915	728	328	131	118

Värriö	Annual	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
Nucl	27	12	20	47	100	81	47	21	28	40	25	11	12
Ait	208	85	138	206	328	452	570	465	425	356	144	69	68
Acc	145	127	156	150	172	188	262	398	226	132	68	70	82
Tot	511	260	356	547	849	941	1151	1121	871	682	305	187	187

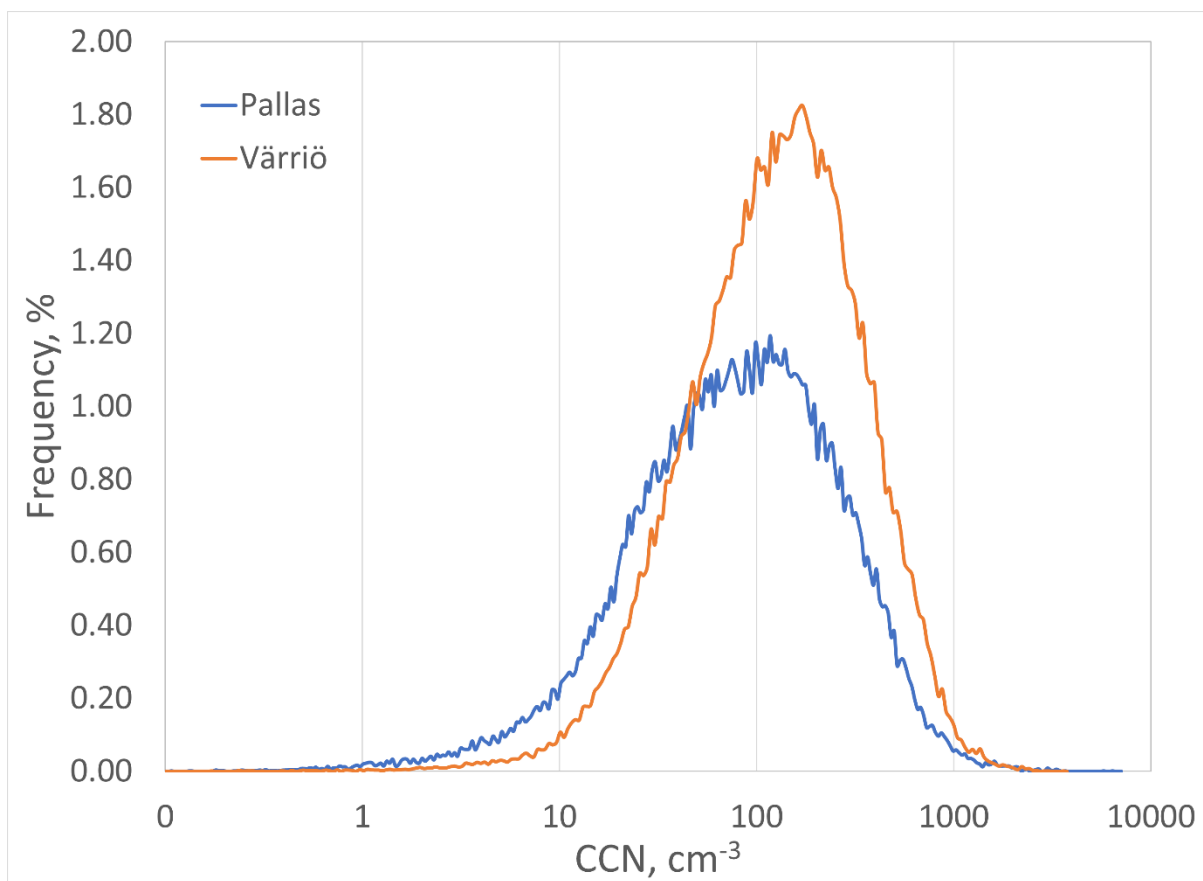


Figure S1: Histogram of CCN concentrations calculated at Värriö from December 1997 to November 2021 (red line), and at Pallas from January 2005 to December 2020 (blue line).

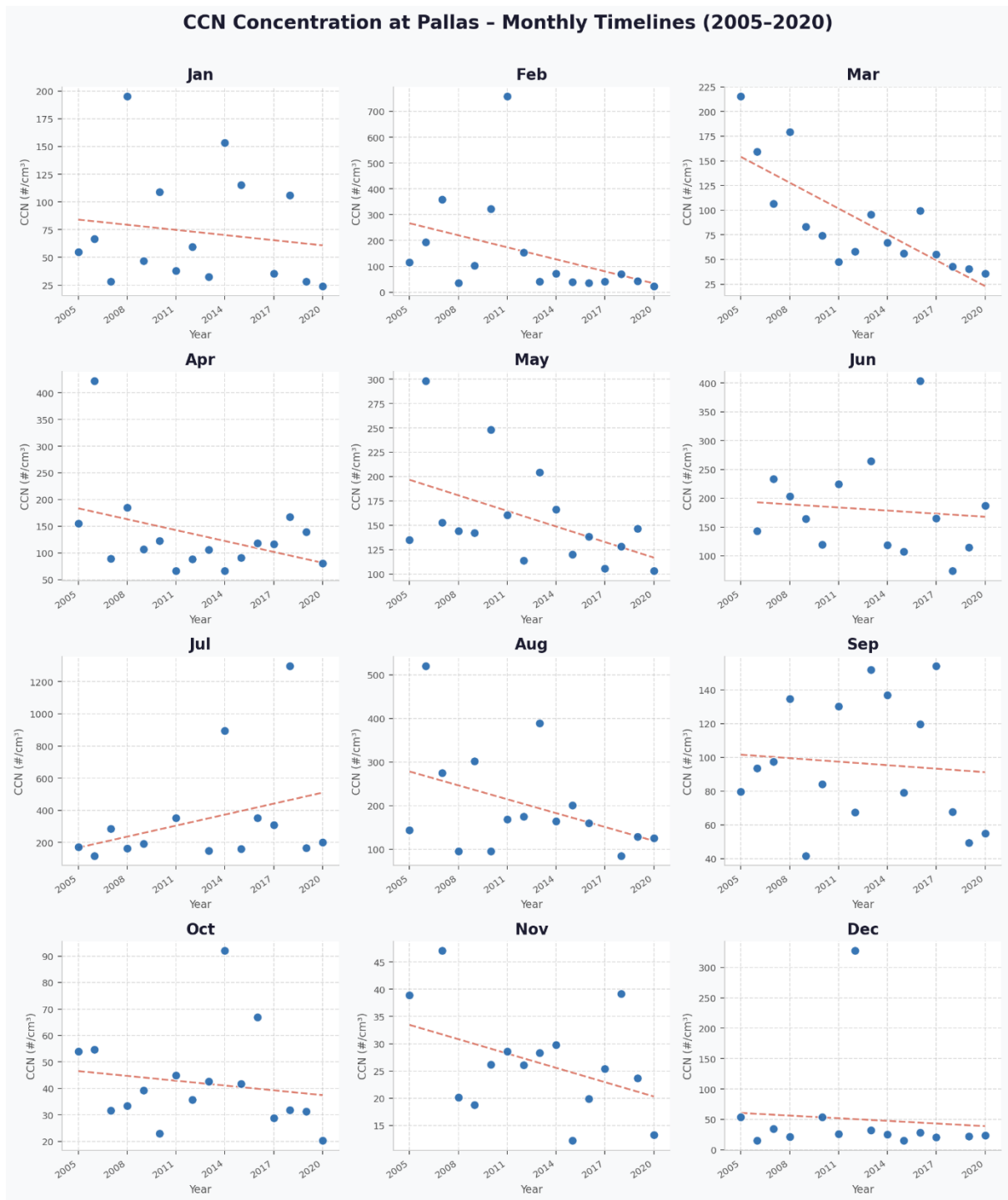


Figure S2: Long-term monthly timeline of CCN concentrations from the parameterization at Pallas from 2005 to 2020. Circles are monthly averages and red lines denote the trend.

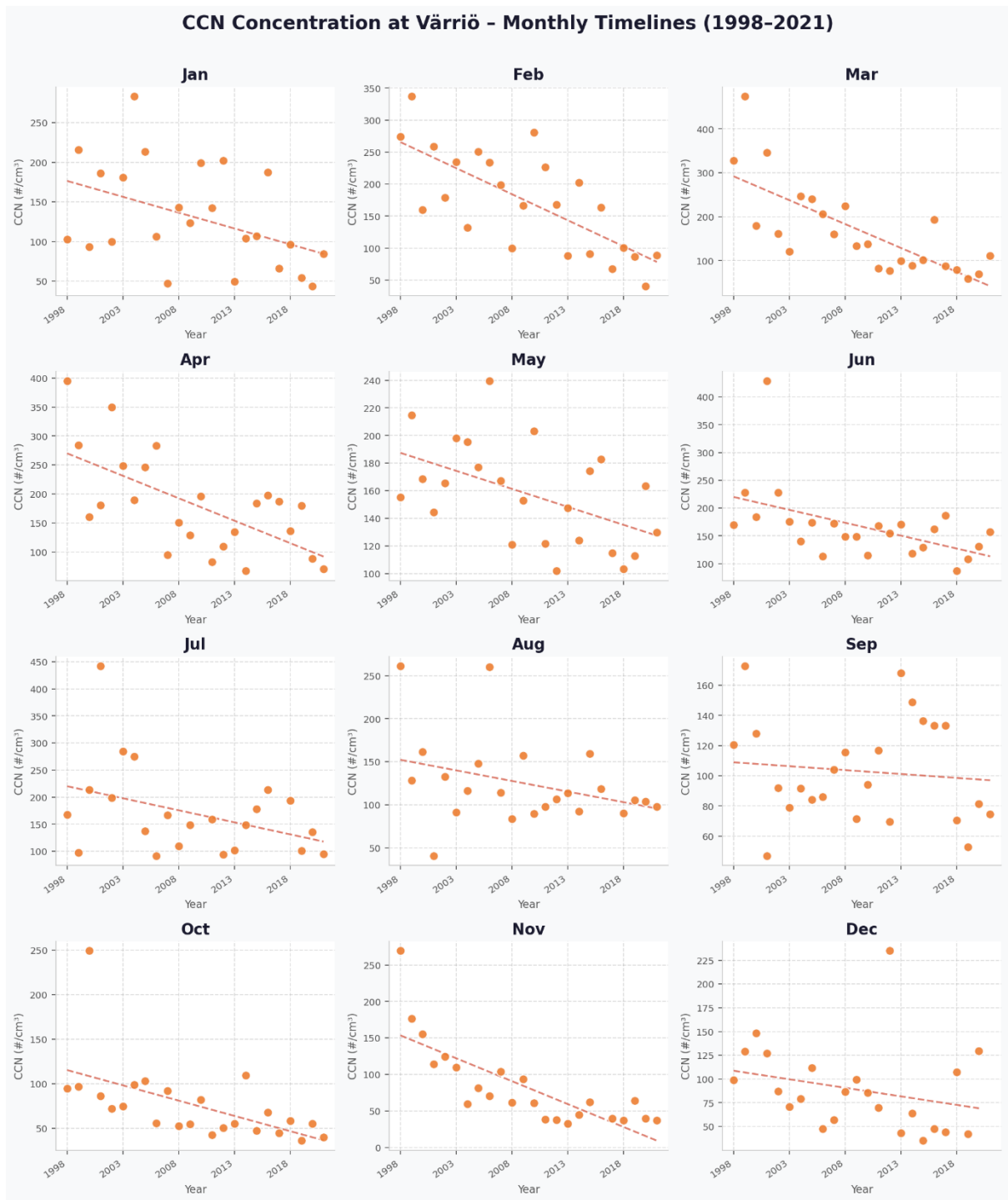
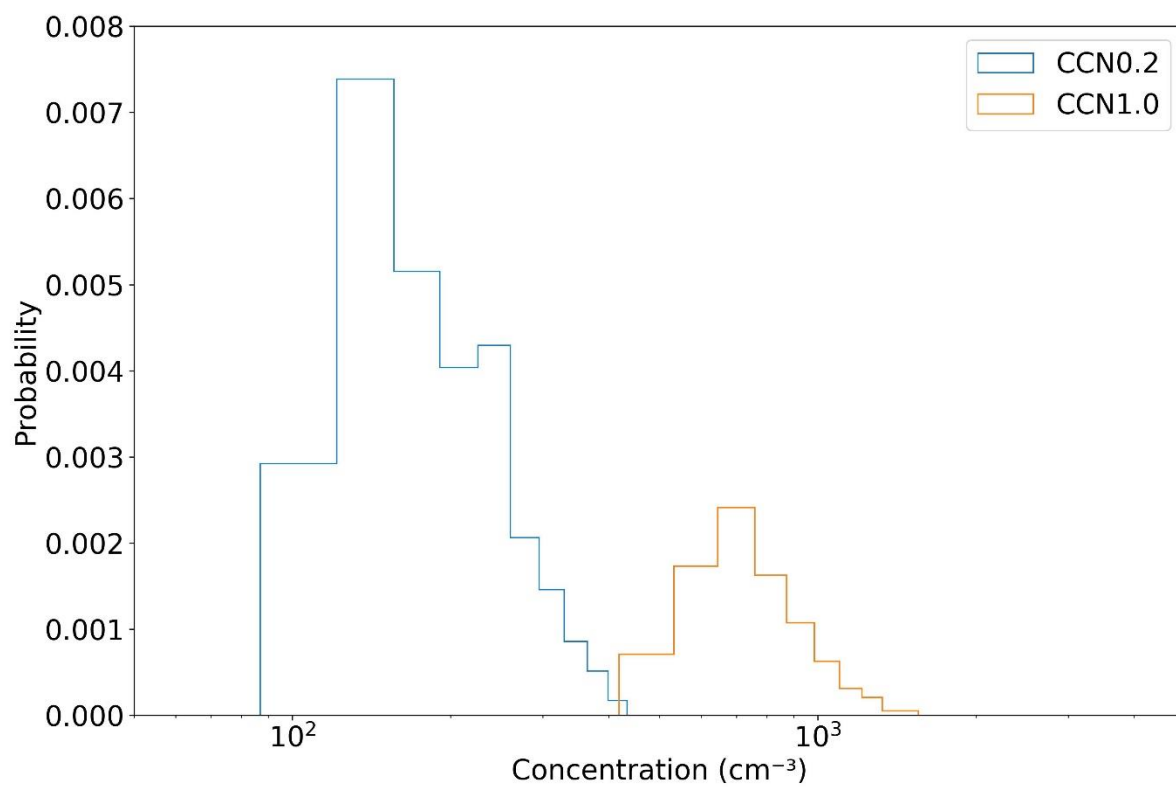


Figure S3: Long-term monthly timeline of CCN concentrations from the parameterization at Värriö from 1998 to 2021. Circles are monthly averages and red lines denote the trend.

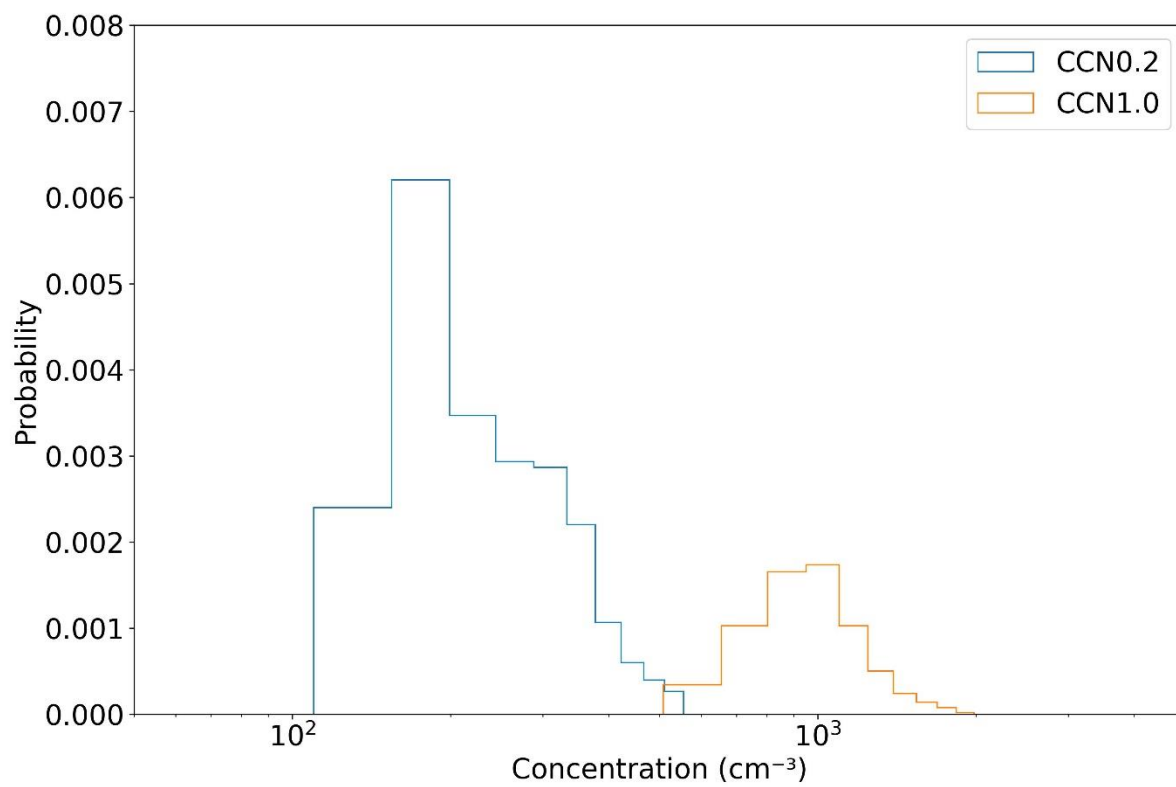
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70 Figure S4 : Probability distribution functions of CCN at 0.2% (blue) and 1.0% (red)
71 supersaturation over Pallas(a) and Värriö(b) for the period January 1987 to December 2014,
72 simulated by the EC-Earth3.