



1 Cloud condensation nuclei at two Finnish sub-Arctic sites: long-term observations of aerosol
 2 activation, predictions based on a simple semi-empirical parameterization, and comparison to global
 3 climate model simulations

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18 **ABSTRACT**

19 In this study, we present long-term observations of cloud condensation nuclei (CCN) at two locations
 20 in the Finnish sub-Arctic: Pallastunturi and Värriö. Both stations belong to the ACTRIS research
 21 infrastructure. At Pallastunturi, the interactions between aerosols and clouds have been measured
 22 in situ at a fjell top, 565 m a.s.l. since 2005. From Pallastunturi data, a simple parameterisation was
 23 developed to predict the activation diameter D_{50} and subsequent CCN- concentrations, based on air
 24 mass origin and temperature. The parameterization was applied to aerosol size distribution
 25 measurements conducted at Värriö from 1998 to 2021. The average annual CCN concentrations
 26 were $140 \pm 40 \text{ # / cm}^3$ at Pallas and $170 \pm 50 \text{ # / cm}^3$ at Värriö. The CCN- concentrations exhibited
 27 statistically significant decreasing trends at both stations: for Pallas $-2.5 \text{ CCN / cm}^3 / \text{ year}$, and for
 28 Värriö $-4.2 \text{ CCN / cm}^3 / \text{ year}$. The decrease was most pronounced during February and March, i.e.
 29 during the Arctic Haze period. We also compared the results with EC-Earth3 global climate model
 30 simulations. Using two measurement sites separated by $\sim 230 \text{ km}$ reduces the uncertainty in the
 31 spatial representativeness of observations and provides a robust basis for climate model evaluation.
 32 EC-Earth3 predicted the measured CCN- concentrations fairly well at 0.2% supersaturation,
 33 reproducing the observed long-term trend. However, the modelled seasonal variation was slightly
 34 different, predicting higher concentrations during winter time (November to April). The study



35 demonstrates the value of long term in-cloud aerosol measurements in validating and improving
 36 climate predictions.

37

38 **1 INTRODUCTION**

39 The interactions between atmospheric aerosols and clouds serve as the largest uncertainty in
 40 estimating the Earth's radiative forcing (IPCC, 2021), and hence, in predictions for climate change
 41 (Forster et al., 2007). A critical process affecting aerosol-cloud interactions is the ability of aerosol
 42 particles to act as cloud condensation nuclei (CCN), and the way these CCNs affect cloud properties.
 43 For example, high CCN- concentrations lead to smaller cloud droplets and higher albedo (Twomey,
 44 1977); and increase cloud lifetime by suppressing precipitation (Albrecht, 1989). The ability of an
 45 aerosol particle to act as a CCN depends heavily on its size, and to a secondary degree, its chemistry
 46 (e.g. Dusek et al., 2006). Moreover, meteorology, through updraft velocity and the availability of water
 47 vapour, affect the maximum supersaturation and thus the activation efficiency of the aerosol
 48 population. This leads to aerosol-cloud interactions that are 'regime'-dependent, meaning that they
 49 can vary under different meteorological conditions (Chang et al., 2015). Based on updraft velocities
 50 and particle concentrations, Reutter et al. (2009) segregated cloud droplet formation into three
 51 regimes: an aerosol-limited regime, an updraft- limited regime, and a transitional regime. This affects
 52 for example the susceptibility of cloud droplet number concentration (N_d) to CCN concentration
 53 (Virtanen et al. 2025). A high susceptibility of N_d to CCN is expected in the aerosol limited regime,
 54 while a low susceptibility can be expected in the updraft limited regime. As the cloud droplet
 55 formation regimes can be highly localized, high-quality observations are critical in understanding
 56 spatial differences of cloud formation, and providing observational constraints for global climate
 57 models (Wang et al., 2026).

58 The Arctic is one of the regions where clouds play a vital role in modulating the climate, and there is a
 59 need for more long-term comprehensive observations of aerosols and clouds (such as described in
 60 Platt et al. 2022, Koike et al., 2019 and Motos et al., 2023) in the region. As the Arctic is warming
 61 rapidly (Rantanen et al. 2022), clouds in the region can also be expected to undergo changes,
 62 highlighting the value of long-term observations.

63 Direct measurements of CCN- concentrations typically involve CCN- counters, which utilize pre-
 64 defined supersaturation levels for the aerosol activation to occur (Roberts and Nenes 2005). Such
 65 measurements have been conducted in several places around the world (e.g. Schmale et al., 2017),
 66 and provide gold-standard data for development and validation of climate models. The caveat of these
 67 measurements is that the predefined supersaturations do not represent real cloud base conditions



68 (where activation takes place) where supersaturation is not constant and may fluctuate substantially
 69 (Yang et al., 2019). Therefore, CCN- measurements should preferably be complemented by
 70 measurements of updraft velocity (or by other means to estimate the true supersaturation), to be able
 71 to estimate the number of cloud droplets formed.

72 An alternative way to include the effect of supersaturation for aerosol-cloud interaction studies, is to
 73 make the measurements directly inside a cloud. Apart from aircraft measurements, this can be
 74 conducted at ground stations (mostly mountain locations or towers) that experience clouds *in situ*.
 75 Here, sampling of aerosol particles is performed through a cloud residual inlet (Karlsson et al., 2021),
 76 or with inlets sampling the total (including cloud droplets) and interstitial (excluding cloud droplets)
 77 aerosol (Kivekäs et al., 2009). This enables quantification of the aerosol particles that acted as a real
 78 CCN, implicitly taking the cloud supersaturation effect into account. Only a few such ground stations
 79 exist worldwide (see e.g. Laj et al. 2024).

80 As direct CCN- measurements are scarce, parameterisation approaches based on observations have
 81 been developed to predict CCN concentrations. The parameterizations range from using simple
 82 regression proxies of CCN such as satellite-derived aerosol optical depth, AOD (e.g. Ahn et al. 2021),
 83 to employing theory-based activation schemes for measured aerosol properties; in particular the
 84 Köhler theory (Köhler, 1936) and its derivatives (e.g. Petters and Kreidenweis, 2007). Recently, Fang
 85 et al. (2016) compared seven methods for CCN parameterization based on measured total particle
 86 concentration, particle number size distribution and cloud condensation nuclei concentrations on Mt.
 87 Huang, China. They concluded that a parameterization using an average critical diameter above which
 88 particles are assumed to activate to cloud droplets, provides the best estimate of CCN. Similarly, Deng
 89 et al. (2013) recommended predicting CCN concentrations by using particle number size distributions
 90 with determined critical diameters or size-resolved activation ratios.

91 The impact of aerosols and their interactions with clouds are one of the main sources of uncertainty
 92 in climate models (Boucher et al. 2013). The causes of uncertainties include incorrect modelling of
 93 aerosol precursor emissions, nucleation events, as well as incorrect representation of aerosol size
 94 distributions in models (Pierce et al. 2009; Lee et al. 2013). Among the uncertainties noted in Lee et
 95 al. (2013), high latitudes during wintertime are noted as a region of high uncertainty. This long-term
 96 CCN dataset provides an opportunity to evaluate the modelled CCN along with the size distribution
 97 data.

98 The overarching aim of this study was to investigate the long-term CCN- concentrations in the Finnish
 99 sub-arctic, based on directly measured data from two ground-based stations. At the first station,
 100 which is regularly inside low-level clouds, aerosol activation into cloud droplets has been measured



101 routinely; while the second station is equipped with aerosol size distribution measurements that can
 102 be used as a proxy for CCN. In this study, we demonstrate how the data can be combined to predict
 103 CCN concentrations at both stations.

104 To be explicit, the main goals of this study were to: 1) develop a simple cloud activation
 105 parameterisation based on long-term measurements at the Pallastunturi research station (from
 106 hereafter: Pallas), enabling the derivation of the aerosol activation diameter based on prevailing
 107 atmospheric conditions, 2) apply the parameterization to predict CCN concentrations at the Värriö
 108 research station, 3) present the long-term CCN concentrations from both stations and 4) compare
 109 them against global climate model results. To the best of our knowledge, this study presents the
 110 longest (> 10 year) time series of CCN concentrations in the Arctic region.

111

112 **2 MATERIALS AND METHODS**

113 **2.1 MEASUREMENT SITES**

114 Measurements relevant to this study were conducted at Pallas Atmosphere-Ecosystem Supersite
 115 (Hatakka et al., 2003, Lohila et al. 2015), and Värriö SMEAR I (Station for Measuring Ecosystem-
 116 Atmosphere Relations) sub-arctic research station (Hari et al., 1994). Both stations belong to the
 117 European ‘Aerosols, Clouds and Trace Gases Research Infrastructure’, ACTRIS (Laj et al., 2024). The
 118 stations are located in Finnish Lapland (Figure 1), and their surrounding environments are very similar,
 119 being that of boreal forest with no significant local air pollution sources. The closest major sources of
 120 pollution are the smelters Montshegorsk and Nikel in the Kola Peninsula region, North-West Russia,
 121 located 140 km East and 200 km North of Värriö station (67° 45′ N, 29° 36′ E), respectively.

122 Pallas station (67° 58′ N, 24° 07′ E) consists of several measurement stations, of which data from the
 123 main station, Sammaltunturi, is used in this work. The station resides on the top of a hill (565 m a.s.l.,
 124 about 100-250 m higher than the surrounding landscape), and is regularly inside low-level clouds,
 125 making it suitable for studies on aerosol-cloud interactions (Doulgeris et al. 2023). The distance
 126 between Pallas and Värriö is about 230 km. As there are no major local emission sources in the region,
 127 we assume that aerosol properties measured at both stations are similar and governed by long-range-
 128 transported air masses.



Figure 1: Map of the study area, highlighting specifically the relative locations of Pallas and Värriö research station

2.2 INSTRUMENTATION

At Pallas, two differential mobility particle sizers (DMPS) measured the aerosol number size distribution in the size range 7 – 500 nm, divided into 30 logarithmically spaced size bins. A complete size distribution was recorded approximately every 5.5 minutes. One DMPS system, referred to as the total DMPS, measures the concentration of all aerosol particles including the cloud droplet nuclei by evaporating the condensed water vapour, as well as the non-activated particles. The second, interstitial DMPS measures only the interstitial (non-activated) aerosol particles using an inlet cut-off (c.a. 7 μm), preventing larger, cloud activated particles from entering the instrument (Komppula et al., 2005). When the station is inside a cloud, the difference between the two recorded particle size distributions provides the size distribution of particles activated into cloud droplets, yielding the CCN-concentration, N_{CCN} . At the prevailing updraft velocity and supersaturation of water vapour, this can be estimated to be equal to the cloud droplet number concentration N_d , i.e. $N_{CCN} = N_d$. For this study, the period of 2005 – 2020 of quality assured data from both instruments simultaneously was used. On



occasions where there are no clouds present at the station, the size distributions recorded by the total and interstitial DMPS instruments were within instrumental uncertainties. Various meteorological parameters, out of which specifically relevant to this study is air temperature, were recorded at Pallas by the Vaisala Milos 500 automatic weather station. Visibility was recorded by the Vaisala FD12P weather sensor. The above data were recorded as one minute means. At Värriö, the aerosol size distribution was recorded by a DMPS instrument approximately every 10 minutes (Aalto et al., 2001) during 1998-2021. Only out-of-cloud measurements were conducted at Värriö, thus the aerosol data recorded is comparable to the total air size distribution measurements made at Pallas. The lower and upper limits of the size bins varied over the time period of DMPS measurements, so for consistency, the diameter range 8 – 450 nm, separated into 28 logarithmically spaced size bins was selected for this study. Temperature was recorded every minute, with Pt100 sensors.

Table 1: Measurement resolution and availability of data recorded at Pallas and Värriö research stations, used in this study

Site	Measurement	Time resolution	Time frame used in study
Pallas	Interstitial aerosol size distribution	5.5 min	01/2005 – 12/2020
Pallas	Total Air aerosol size distribution	5.5 min	08/2005 – 12/2020
Pallas	Visibility	1 min	01/2005 – 12/2020
Pallas	Temperature	1 min	01/2005 – 12/2020
Värriö	Total Air aerosol size distribution	10 min	01/1998 – 12/2021
Värriö	Temperature	1 min	01/1998 – 12/2021

160

2.3 CLOUD ACTIVATION PARAMETERIZATION

2.3.1 DATA PROCESSING

Hourly median values of both the total and interstitial DMPS data at Pallas were calculated. The period of 2008 – 2014 was selected as the training period, and this dataset was used in the development of the parameterization. Hours when the visibility was less than 500 m were classified as cloudy, and this corresponded to 17% of all available data.

Such periods were used in the calculation of the size dependant activated fraction ($A_F(Dp)$), defining the number fraction of aerosol particles that have formed a cloud droplet (Eq. 1).

$$A_F(Dp) = \frac{\frac{dN_{total}(Dp)}{d\log Dp} - \frac{dN_{interstitial}(Dp)}{d\log Dp}}{\frac{dN_{total}(Dp)}{d\log Dp}}$$

169



(1)

170 The calculation of the activated fraction at each size bin diameter measured by the DMPS system
 171 enabled the assembly of activated fraction curves, that overall, demonstrate expected shapes (Fig. 2);
 172 following a sigmoidal shape starting with approximately null activation at small diameters and
 173 reaching a plateau generally approaching one at larger diameters. However, some curves appeared
 174 anomalous, and data refinement was conducted, based on values that are physically reasonable but
 175 still account for measurement noise. The activated fraction curves defined by the following criteria
 176 were removed:

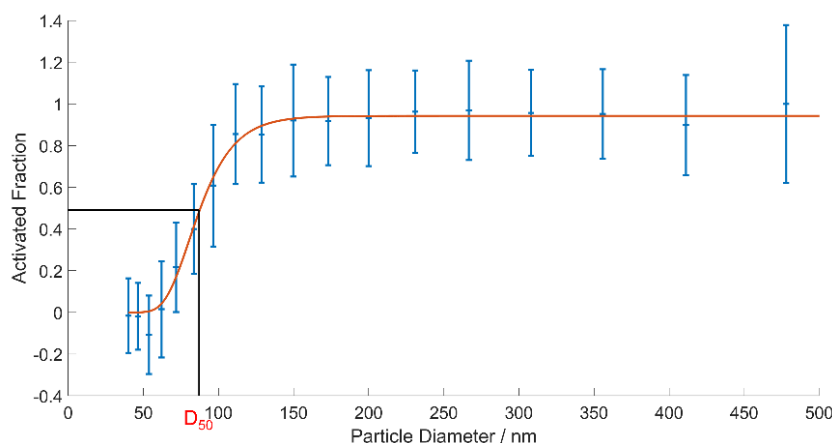
- 177 Criteria 1: The mean activated fraction value of the four largest size channels was less than
- 178 0.7
- 179 Criteria 2: Any activated fraction value exceeded 1.2
- 180 Criteria 3: Any activated fraction value **above** 200 nm was below -1
- 181 Criteria 4: Any activated fraction value **below** 200 nm was below -2

182 A sensitivity analysis for the selection of activation curve limits used in each of the above-stated
 183 criteria is presented in the Supplementary material.

184 Activated fraction curves carried forward for the development of the cloud activation
 185 parameterization were fitted using the Gompertz function (Eq. 2) (Gompertz, 1825) using the
 186 minimization of the sum of squared errors approach. Standard errors used in the fitting were
 187 calculated from the standard deviation of the hourly averaged DMPS data using error propagation
 188 methodology. The diameter at which 50% of aerosol particles are activated into cloud droplets is called
 189 D_{50} . This is a central variable utilized in this work. From the Gompertz fit to the activated fraction data
 190 (See example in Figure 2), hourly D_{50} values were obtained from determining the diameter
 191 corresponding to 50% of the activation curve plateau height.

(2)

$$192 \quad A_f(Dp) = a \cdot e^{(-e^{(-b(Dp-c))})}$$



193

194 Figure 2: Example Gompertz curve function fitted to activation fraction data. Blue lines represent the
 195 activated fraction value with standard error. The red line represents the Gompertz fit. D_{50} = diameter
 196 at which 50% of aerosol particles are activated.

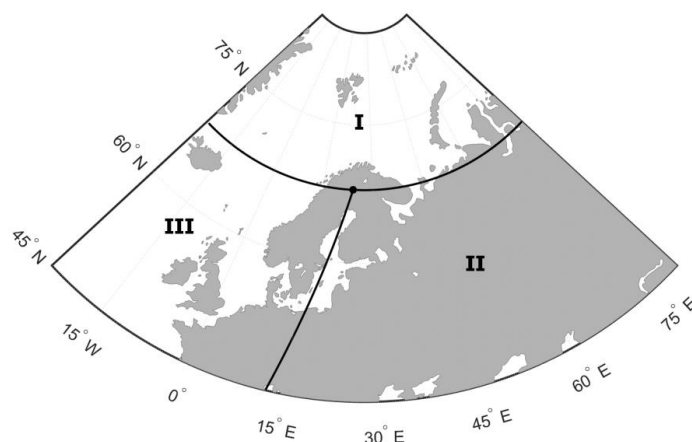
197

198 The determined D_{50} values were classified by two selected variables that can be considered as drivers
 199 of aerosol chemical composition: air temperature and air mass origin. Classification based on air
 200 temperature was chosen due to the direct relationship between temperature and aerosol organic
 201 loading (Yli-Juuti et al., 2021). Air mass origin provides an indication of the type and strength of natural
 202 and anthropogenic aerosol emissions, that the air arriving at the measurement station has been
 203 exposed to. This has been suggested to correlate with the chemical composition of the particles
 204 (Ovadnevaite et al., 2014; Kivekäs et al., 2009; Petäjä et al., 2022). Consequently, the variability in the
 205 chemical composition affects aerosol hygroscopicity (Hong et al., 2014), and hence the ability of the
 206 particles to activate into a cloud droplet (D_{50} value).

207 The determined D_{50} values were first grouped into six different temperature classes ranging from -15
 208 to 15 °C separated by 5 °C intervals. The D_{50} values separated by temperature class were further
 209 classified by air mass sectors based on a backward trajectory calculation. The trajectories were
 210 calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et
 211 al., 2015). Seventy-two-hour backward trajectories were calculated, arriving at 100 m a.g.l. at Pallas
 212 every 1 hr for the period 2008–2014. Three different air mass sectors were selected to be applied to
 213 the CCN activation parameterization: Arctic, East and West as shown in Figure 3. The trajectories were
 214 assigned one of the three overall sectors based on the sector that the trajectory spent the most time
 215 in. The sector selection was based on typical air masses which both Pallas and Värriö experience (Asmi
 216 et al., 2011), and simplicity to support the creation of a robust cloud droplet activation scheme.



217 Separation of the D_{50} values by six temperature classes and three air mass sectors forms a look-up
 218 table of 18 median D_{50} values (Table S3) that forms the basis of the parameterization. This
 219 classification approach enables simple and logical applicability of the cloud activation
 220 parameterization. A sensitivity analysis for the choice of the sector bounds is presented in the
 221 Supplementary material.



222
 223 Figure 3: Map of air mass sectors used to classify trajectories arriving at Pallas (location shown as
 224 central point): I (Arctic), II (East), and III (West)

225

226 2.3.2 APPLICATION OF THE CLOUD ACTIVATION PARAMETERIZATION

227 The concentration of CCN (N_{CCN}) in the parameterization was obtained from the integration of hourly
 228 averaged total particle number size distribution of particles larger than the D_{50} value (See Fig.4 and
 229 Eq. 3). The relevant D_{50} value was found in the look-up table (Table S3) based on the combination of
 230 temperature and air mass origin classification. Interpolation of the size distribution data was necessary
 231 to provide increased resolution and enable more accurate integration around the desired D_{50} value.

$$232 \quad N_{CCN} = \int_{D_{50}}^{\infty} n(\log D_p) d\log D_p \quad (3)$$

233

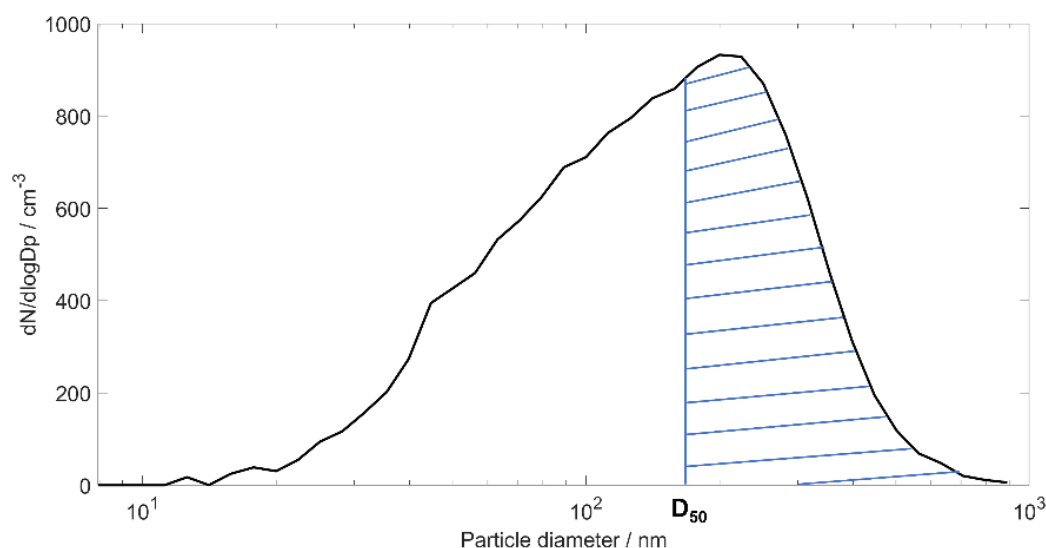


Figure 4: Integration of particle number size distribution data to obtain predicted N_{CCN}

At Pallas, the parameterization was applied to the aerosol size distribution data from 2005 to 2007 and from 2015 to 2020, i.e. outside of the parameterization training period. This provided a continuous CCN dataset for the years 2005-2020, that would be consistent with that from Värriö, see below.

For Värriö, the same temperature classification system was used as developed at Pallas. Trajectory analysis was conducted in a similar way to that at Pallas, but Värriö was now selected as the central point for trajectory arrival. The most significant difference to Pallas is that central Finland is now included in the West sector. Eq. 3 was then utilized for the Värriö aerosol size distribution data, resulting in CCN- concentrations for the period 1997 – 2021. The approach rests on the assumption that both Pallas and Värriö reside in a similar sub-Arctic environment, and that the air mass sector definitions are robust enough to assume similar chemistry in the defined incoming air mass sectors at both sites.

2.4 GLOBAL CLIMATE MODEL

The long-term observational data from this study provide a chance to evaluate how well a model captures the aerosol processes at a station over the years. EC-Earth3 AerChem is an earth system model which couples the Integrated Forecasting System (IFS) meteorological model with Tracer Model Version 5, Massively Parallel version (TM5), a component to simulate aerosols and atmospheric



255 chemistry (van Noije et al. 2021). The aerosol components represented in TM5 include sulphate (SO₄),
 256 black carbon (BC), organic aerosols (OA), sea salt, and mineral dust. These are described by the modal
 257 aerosol microphysical scheme M7 (Vignati et al. 2004) as described in Stier et al. 2005, which consists
 258 of four water-soluble modes (nucleation, Aitken, accumulation, and coarse) and three insoluble
 259 modes (Aitken, accumulation, and coarse). In Bergman et al. 2022 the TM5 model was further
 260 developed to improve the representation of Secondary Organic Aerosols (SOA). For the determination
 261 of CCN- concentrations, EC-Earth3 calculates the critical dry particle diameter using Köhler theory.

262 Using the model, 6 ensemble runs were created, producing output from 1987 to 2015 as monthly
 263 average values. The simulations were run at horizontal resolution of 0.678 degree by 0.678 degree
 264 and a vertical resolution of 92 sigma levels. An ensemble mean of the dataset was obtained using the
 265 ensemble mean tool from Climate Data Operators (Schulzweida 2023) and the point corresponding to
 266 the Pallas and Värriö stations were obtained using nearest neighbour interpolation. A comparable
 267 dataset from the observations was created by calculating the monthly mean from the available
 268 datapoints. We used CCN at 0.2% and 1.0% supersaturation for comparison with the measured values.
 269 In an earlier case study at the Pallas station during autumn (Anttila et al. 2009), the critical
 270 supersaturation based on observations was estimated to vary between 0.18 - 0.26.

271 For additional calculations of representative aerosol size distributions, we ran simulations using TM5
 272 only, from January 2019 to September 2020, with 9 months of the run considered as spin-up.

273 **3 RESULTS AND DISCUSSION**

274 **3.1 AEROSOL SIZE DISTRIBUTIONS AND TRENDS**

275 The aerosol size distribution is a governing factor for cloud condensation nuclei concentrations (Dusek
 276 et al., 2006). Therefore, we first look at the basic features of aerosol size distributions observed at
 277 Pallas and Värriö during the study period. For simplicity, we have segregated the size distributions into
 278 three modes, namely the nucleation mode ($D_p < 20$ nm), the Aitken mode ($20 \text{ nm} < D_p < 100$ nm), and
 279 the accumulation mode ($D_p > 100$ nm). The size ranges for these modes are the same as for an earlier
 280 trend analysis for both stations (Leinonen et al. 2022).

281 The aerosol concentrations at Pallas and Värriö were very similar, with average annual concentrations
 282 about 860 # /cm³ at Pallas and 790 # /cm³ at Värriö (Table 2). In the context of cloud formation, the
 283 Aitken- and accumulation mode particles are more relevant, although the nucleation mode particles
 284 have the potential to grow into CCN- active sizes as well. At both locations, the Aitken mode dominates
 285 the average annual aerosol size distribution, with over 50 % of particles residing in this size range.

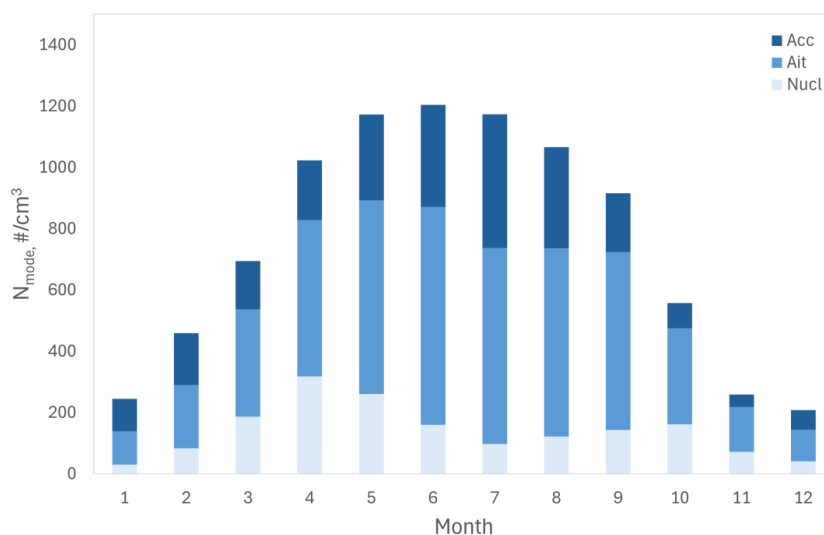
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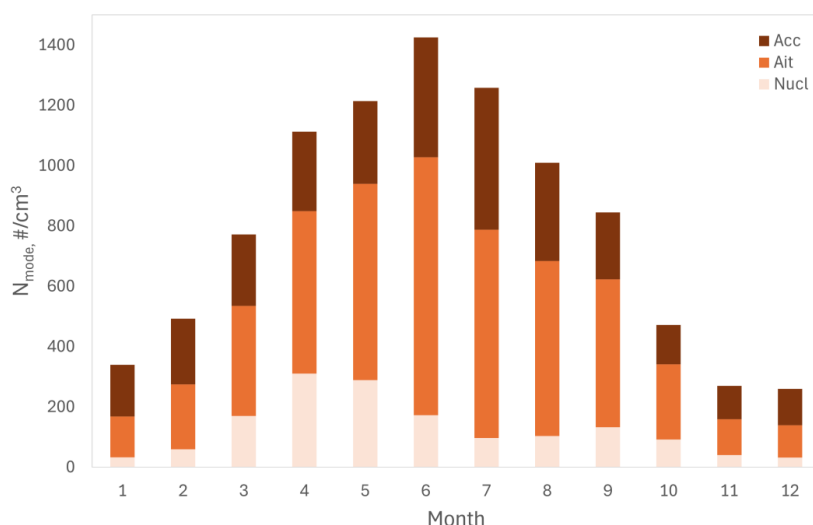
Table 2. Annual average (N) and standard deviation, with annual trend (dN), for the nucleation mode ($D_p < 20$ nm), the Aitken mode ($20 \text{ nm} < D_p < 100$ nm), and the accumulation mode ($D_p > 100$ nm) for Pallas 2005-2020 and Värriö 1998-2021. Statistically significant trends ($p < 0.05$) have been marked with an asterisk.

	N_{Nuc} #/cm ³	dN_{Nuc} #/cm ³ /year	N_{Ait} #/cm ³	dN_{Ait} #/cm ³ /year	N_{Acc} #/cm ³	dN_{Acc} #/cm ³ /year	N_{Tot} #/cm ³	dN_{Tot} #/cm ³ /year
Pallas avg.	158	3.9	468	-9.1*	228	-8.8*	863	-12.7*
st.dev.	42		60		64		103	
Värriö avg.	130	0.6	413	-4.3*	244	-5.4*	791	-7.6
st.dev.	19		61		51		108	

The long-term concentration measurements reveal a decreasing trend at both stations. At Pallas, total concentrations have decreased at a rate of about $13 \text{ \#/cm}^3 / \text{year}$ ($p = 0.0045$), and at Värriö at a rate of about $8 \text{ \#/cm}^3 / \text{year}$ ($p = 0.087$). This decrease is most pronounced, and statistically significant, in the Aitken and accumulation modes, with the nucleation mode showing an increasing, albeit statistically insignificant, trend.



a)



299 b)

300 Figure 5. Monthly variation of average accumulation ($D_p > 100$ nm)-, Aitken- ($20 \text{ nm} < D_p < 100$), and
 301 nucleation- ($D_p < 20$ nm) mode particle concentrations at a) Pallas during 2005-2020 and b) Värriö
 302 during 1998-2021.

303

304 Both stations have a pronounced seasonal variation in their aerosol size distribution (Figure 5). The
 305 highest total concentrations were found in summer, with a maximum monthly average concentration
 306 around 1220 $\#/\text{cm}^3$ at Pallas and 1420 $\#/\text{cm}^3$ at Värriö in June. The lowest monthly concentrations
 307 occurred in December, when the monthly average particle concentration was 210 $\#/\text{cm}^3$ at Pallas
 308 and 260 $\#/\text{cm}^3$ at Värriö. In the autumn, the concentrations at Pallas were slightly higher than at
 309 Värriö. The highly variable seasonal size distributions reflect the remoteness of both stations from
 310 anthropogenic aerosol sources. During summer, natural emissions dominate, while during winter,
 311 when natural activity is low, aerosol is mostly long-range-transported.

312

313 3.2 AEROSOL ACTIVATION AND D_{50} VALUES

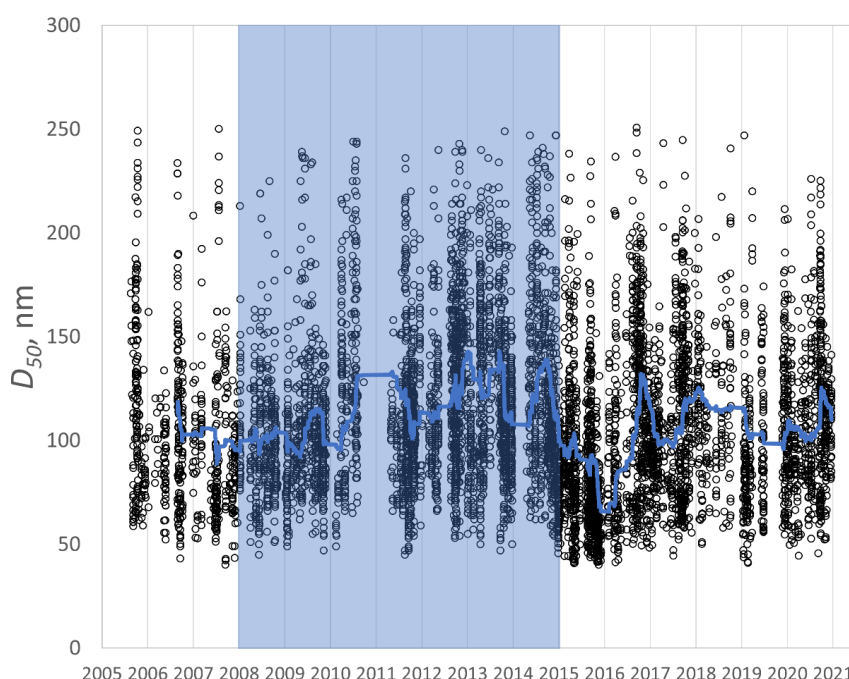
314 3.2.1 OBSERVED D_{50} VALUES AT PALLAS

315 The aerosol activated fractions for occasions when the station was inside a cloud at Pallas, were
 316 calculated using Eq. 1 as described in section 2.3.1. Figure 6 presents a time-series of D_{50} values as
 317 obtained from the fitting procedure (Eq. 2). The median D_{50} value was 105 nm, and the lower and
 318 upper quartile D_{50} values were 84 and 133 nm, respectively. Overall, there is considerable variability



319 in the D_{50} values, particularly at the upper range. This may be attributed to the non-constant
 320 supersaturation of the clouds as well as variation in the CCN chemical composition.

321 Previous studies at Pallas that had derived D_{50} values for shorter time periods, are in reasonable
 322 agreement with the presented observations. Komppula et al. (2005) reported D_{50} values in the range
 323 of 50 – 128 nm, while Anttila et al. (2009) reported a range of 88 – 215 nm. Both of these observation
 324 periods were limited to autumn. Reported D_{50} values from other stations tend to lie in the range of
 325 100 – 200 nm (Väisänen et al., 2016, Henning et al., 2002, and references therein).

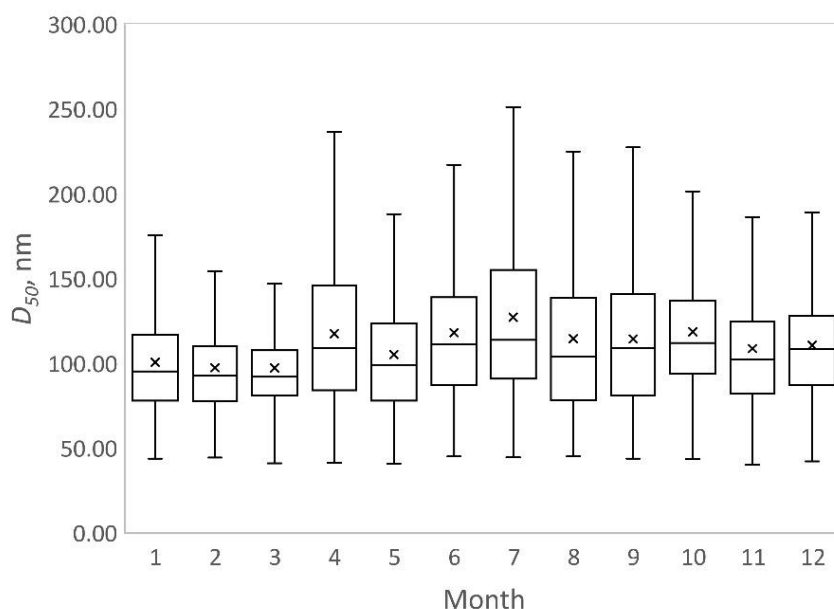


326

327 Figure 6: 1h- average diameter of the 50% cloud activated aerosol population (D_{50}) derived from the
 328 difference between the total and interstitial aerosol populations at Pallas. The solid line is 10-day
 329 running mean, and the training period for CCN- parameterization is shaded in blue.

330

331 In Figure 7, the seasonal variation of D_{50} is shown as monthly distributions. Overall, the median and
 332 mean D_{50} values are in the 100 – 125 nm range with only a weak seasonal variation. In general, low
 333 D_{50} values (the 10th percentile value) can be seen throughout the year, while high values ($D_{50} > 175$
 334 nm) were not observed during winter.



335

336 Figure 7. Monthly diameter of the 50% cloud activated aerosol population (D_{50}) at Pallas during 2005
 337 – 2020. Box plot shows the 10th, 25th, median, 7th and 90th percentiles, while crosses denote the
 338 monthly average.

339 The greatest fraction of the cloud periods occurred during autumn and continued through to winter,
 340 concurrent with previous studies at Pallas based on shorter study periods (Jaatinen et al., 2014; Anttila
 341 et al., 2012). During this time of the year in the Northern Hemisphere, the sun's radiation hits the
 342 surface of the Earth, 1) at a shallower angle resulting in less surface heating; 2) for shorter periods of
 343 time leading to colder air temperature. Subsequently, this increases the likelihood of supersaturated
 344 water vapour in the air.

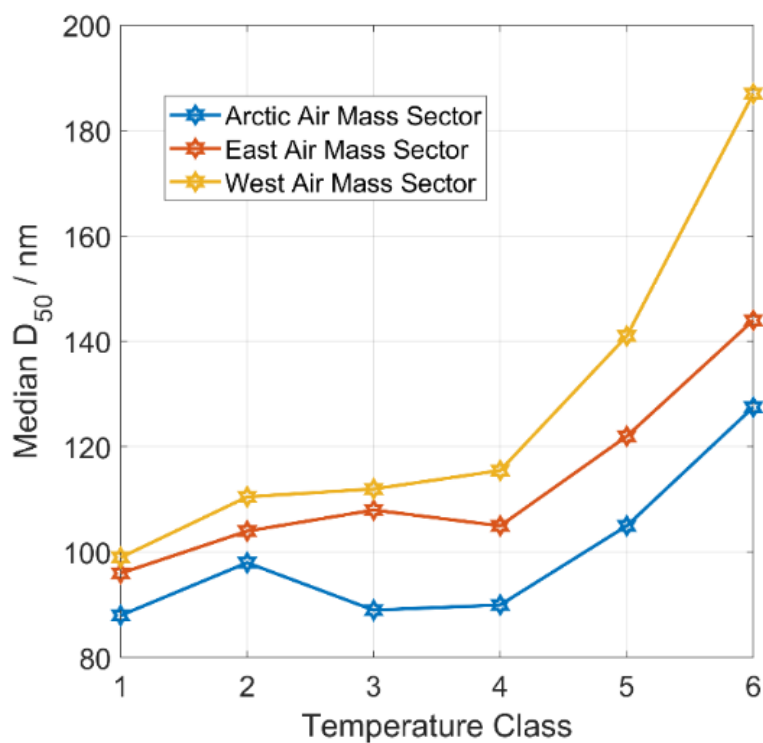
345

346 3.2.2 D_{50} PARAMETERIZATION

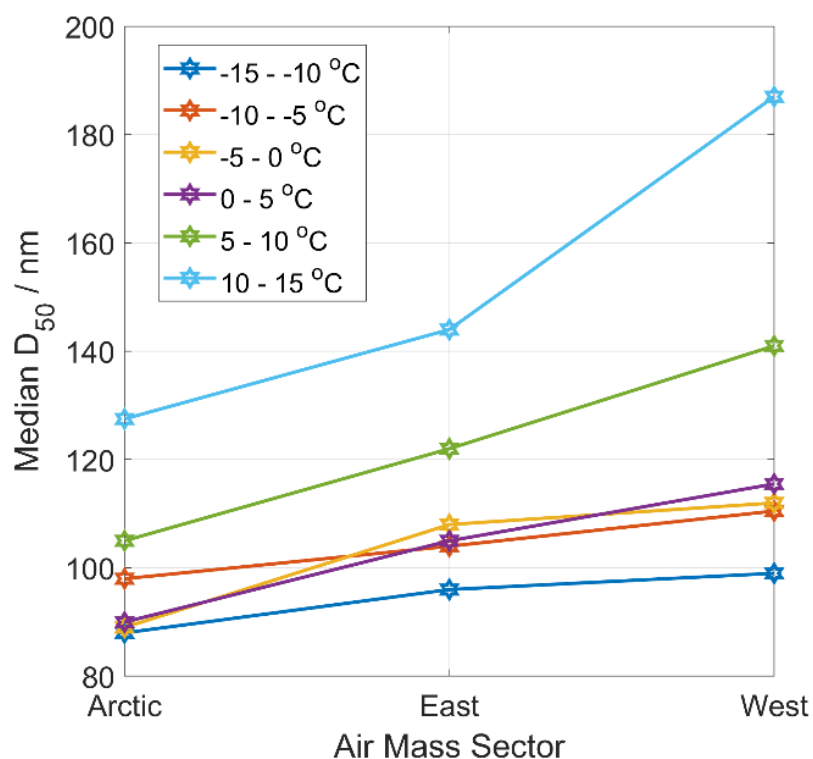
347 The D_{50} values presented in section 3.2.1 were the basis for the CCN parameterization. The
 348 parameterization training set comprised of data measured from 01/2008 to 12/2014 and was
 349 classified by two variables: air temperature and air mass origin, as first introduced in section 2.3.1.
 350 The remaining data, from 08/2005 to 12/2007, and from 01/2015 to 12/2020 was used to evaluate
 351 the parameterization performance at Pallas (See section 3.4.1).



352 Figures 8a and 8b display the median D_{50} values based on the 6 temperature and 3 air mass origin
 353 classifications, respectively. The look-up table of these values, alongside a more detailed statistical
 354 distribution can be found in the Supplementary data, Table S3. The median D_{50} values form the centre
 355 of the cloud activation parameterization.



356 a)



357 b)

358 Figure 8. Median D_{50} values obtained from the fitted activated fraction data derived from the total and
 359 interstitial aerosol concentrations during cloudy periods at Pallas (2008 – 2014), calculated based on
 360 air temperature and air mass origin classifications. a) as a function of temperature classification: Class
 361 1(-15 - -10 °C), 2(-10 - -5 °C), 3(-5 – 0 °C), 4(0 – 5 °C), 5(5 – 10 °C), 6(10 – 15 °C).; b) as a function of air
 362 mass sector.

363

364 Almost consistently, the median D_{50} values increase as the temperature class increases in each air
 365 mass sector (Figure 8a). This increase may be attributed to potential increases in the relative
 366 contribution of organics to the aerosol composition. With increased temperatures, it has been shown,
 367 that emissions of biogenic volatile organic compounds (BVOCs) from vegetation increase (Kesselmeier
 368 and Staudt, 1999). Recent investigations at Hyytiälä SMEAR II station, a site with surroundings
 369 relatively comparable to that of Pallas, demonstrated that the organic loading of an aerosol particle
 370 increased directly with an increase in temperature (Yli-Juuti et al., 2021). This, in turn, would be
 371 expected to decrease the hygroscopicity of the aerosol particle (Hong et al., 2014), hence, increasing
 372 the diameter at which the particle can activate into a cloud droplet. Temperature has also been found



373 to correlate well with BVOC emissions also at Pallas (Hellén et al., 2020). It is notable that during
 374 wintertime, the temperature dependence is nearly flat, supporting the hypothesis that BVOC-
 375 emissions are the main driver of the observed temperature dependence.

376 Observing the D_{50} trends by air mass origin in Figure 8b, consistently for each temperature class, D_{50}
 377 values in the Arctic sector are the lowest, they increase in the East sector, and the largest values are
 378 observed in the West sector. This is likely to be attributed to differences in the chemical composition
 379 of the aerosol from different source regions. Air masses arriving at Pallas from the Arctic come directly
 380 over the marine environment where aerosol sources are mainly composed of primary sea spray and
 381 secondary sulphate (Leck et al., 2002; Sanchez et al., 2018). The subsequent hygroscopic nature of
 382 marine aerosols provides plausible rationalisation for the lowest observed D_{50} values in the Arctic
 383 sector. For the Western and Eastern sector, the most notable difference is the potential for the air
 384 masses to spend time over land. Studies have demonstrated that as an air mass spends time over
 385 boreal forest regions, the organic aerosol mass concentration increases linearly with time over land
 386 (Petäjä et al., 2022). This would reduce the hygroscopicity of the aerosol. Comparing the East and
 387 West sector D_{50} values, the Eastern sector hosts large industries in Eastern Continental Europe and
 388 the Kola peninsula, emitting sulphate-based, and therefore more hygroscopic, aerosol particles
 389 (Virkkula et al., 1997, Sipilä et al., 2021).

390

391 **3.3 CCN- CONCENTRATIONS AT PALLAS AND VÄRRÖ**

392 CCN concentrations over time were obtained by applying the CCN- parameterization (Eq. 3 with D_{50} -
 393 values from the look-up Table S3) to the long-term DMPS- measurements at both sites. At Pallas, to
 394 maximize the time series length while ensuring the homogeneity of the dataset, the cloud screened,
 395 interstitial aerosol size distribution data were used. The data covered 01/2005 – 12/2020 for Pallas
 396 and 12/1997-11/2021 for Värriö, making these one of the longest CCN- time series reported from sub-
 397 Arctic regions.

398

399 **3.3.1 LONG TERM VARIATION**

400 The average annual CCN concentrations at Pallas and Värriö were approximately $140 \pm 40 \text{ # / cm}^3$
 401 (median 90 # / cm^3) and $170 \pm 50 \text{ # / cm}^3$ (median 130 # / cm^3), respectively. The seasonal variation of
 402 the CCN concentrations was high at both stations, with 1-h average CCN concentrations spanning from
 403 less than 1 to more than 1000 # / cm^3 , as seen in Figure 9. Supplementary Figure S1 shows the
 404 statistical distribution of CCN concentrations at both stations. It is notable that even though total



aerosol concentrations at Pallas were slightly higher than at Värriö (see Table 2), the CCN concentrations were lower at Pallas. This is because at Värriö, the accumulation mode concentrations were higher, and these particles activate more readily. However, the differences are within statistical variation.

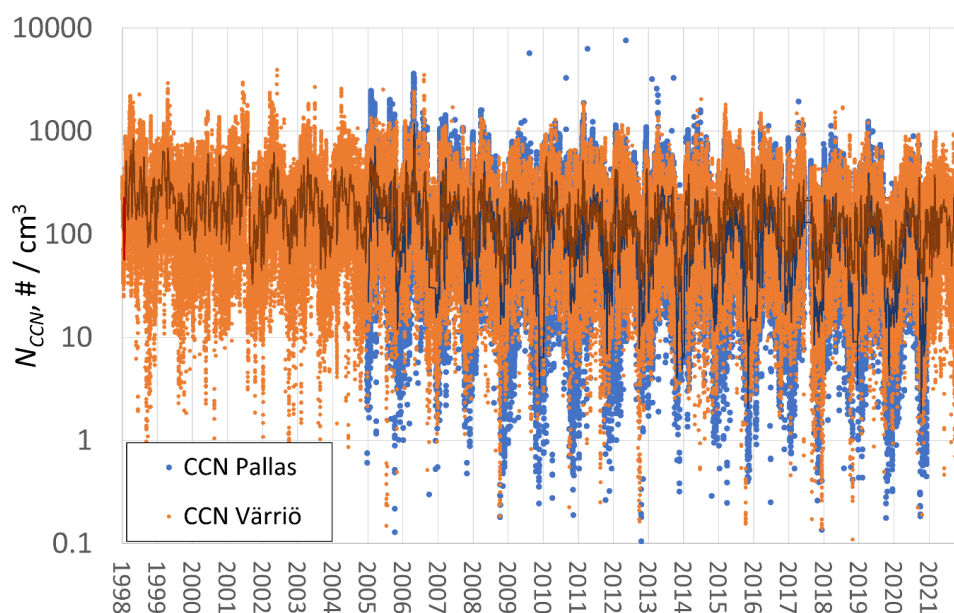
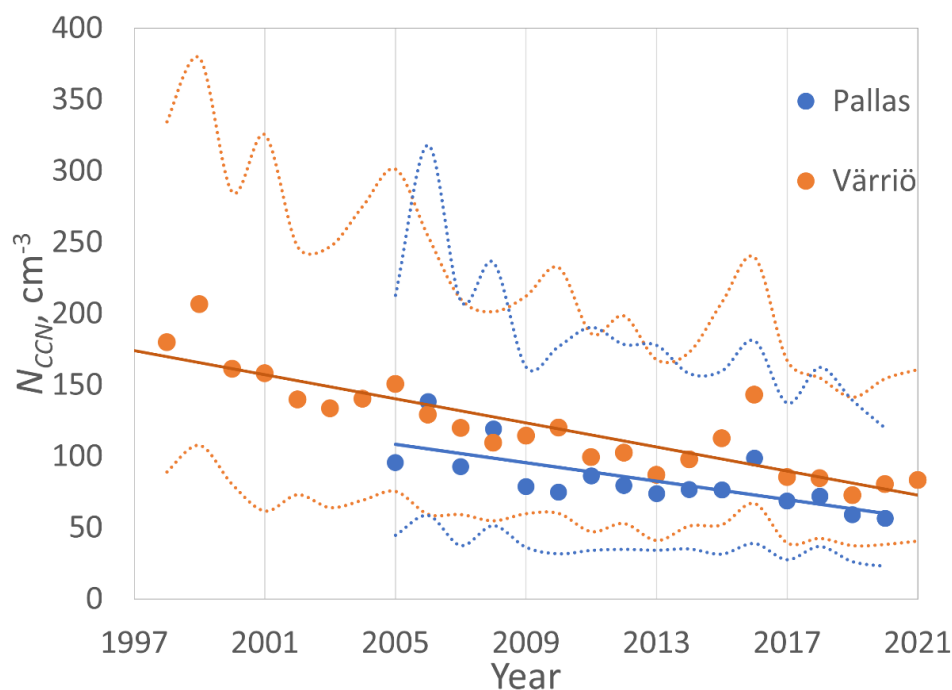


Figure 9. Timeline of 1h average CCN concentrations with a 10-day running mean in Pallas (blue) and Värriö (red), based on CCN parameterization.

A trend analysis of the long-term CCN concentration reveals a clear, statistically significant decreasing trend for both sites, see Figure 10. At Pallas, the median annual concentration has decreased steadily at a rate of $-2.5 \text{ CCN} / \text{cm}^3 / \text{year}$, and at Värriö at a rate of $-4.2 \text{ CCN} / \text{cm}^3 / \text{year}$. For the same period as in Pallas, the trend in Värriö is about $-3.3 \text{ CCN} / \text{cm}^3 / \text{year}$.



418

419 Figure 10. Annual trend of CCN- concentrations at Pallas during 2005 – 2020 (blue) and Värriö during
 420 1998 – 2021 (red). Markers are annual median concentrations, solid lines are linear fits to the medians,
 421 and dashed lines indicate the annual upper and lower quartiles.

422

423 Monthly long-term data were also analyzed to find out how different months contribute to the
 424 observed long-term trend. At Värriö, all months except July, August and September show a statistically
 425 significant decreasing long-term trend, with strongest trends in February, March and April with slopes
 426 of -8.3, -8.8 and -6.8 CCN / cm³ / year, respectively. Meanwhile at Pallas, February and March are the
 427 only months to show statistically decreasing trends at -8.0 and -7.6 CCN / cm³ / year, respectively; and
 428 contrarily, the summer month of July shows a positive, however a non-significant trend of +5.3 CCN /
 429 cm³ / year. This highlights that the decreasing trend was mostly due to a decrease in anthropogenic
 430 aerosols (Arctic Haze), while natural aerosols can play even an opposite role.

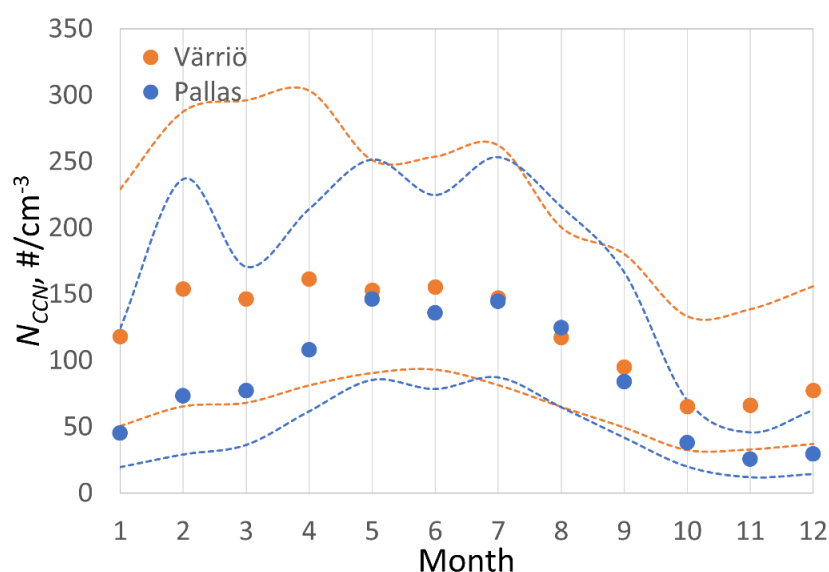
431

432 3.3.2 SEASONAL VARIATION

433 The CCN concentrations show a distinctive seasonal variation for both sites, see Figure 11. At Pallas
 434 high CCN concentrations were observed from May to August, and low concentrations from October



435 to January. At Värriö, highest CCN concentrations were additionally observed from February to April,
 436 and concentrations were lowest from October to December. At Värriö, the Arctic Haze phenomenon
 437 appeared stronger (most likely due to emissions from the Kola region), leading to the highest CCN
 438 concentrations in the first quarter of the year.



439

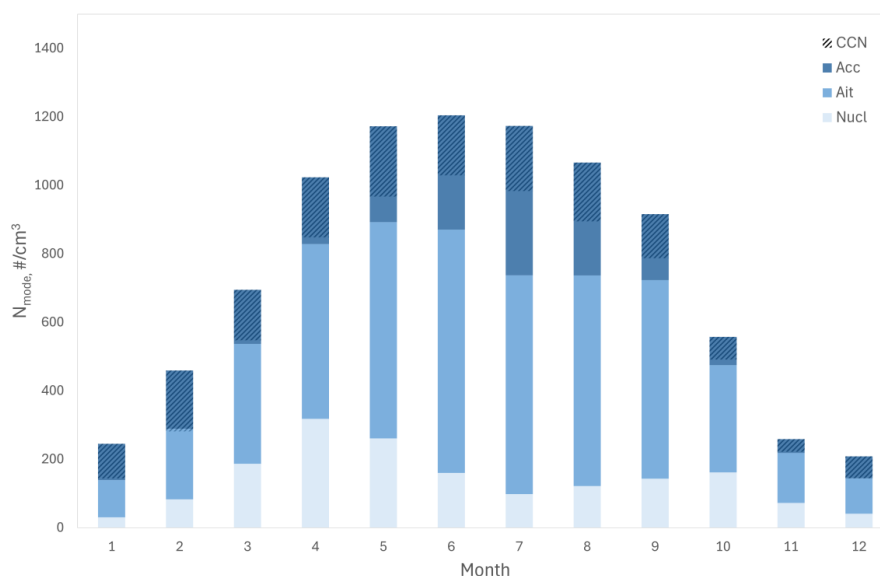
440 Figure 11. Seasonal variation of CCN- concentration at Värriö during 1998 – 2021 (red) and Pallas
 441 during 2005 – 2020 (blue). Solid lines with markers are median values, while dashed lines indicate the
 442 upper and lower quartiles.

443

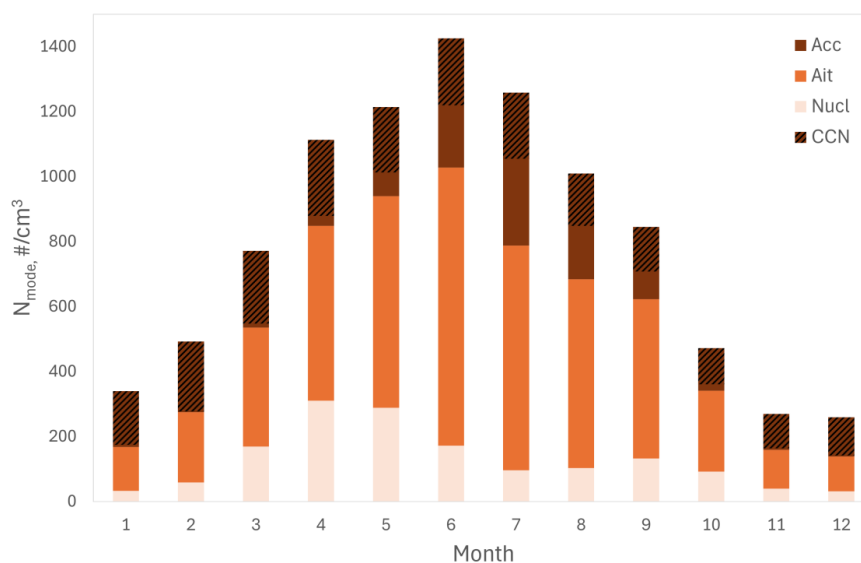
444 Figure 12 illustrates how particles in different size modes activated into cloud droplets during different
 445 seasons. Assuming that largest particles activate first, the shaded area of bars in Figure 13 act as CCN.
 446 At both sites during winter (from November to February), the whole accumulation mode, comprising
 447 30 to 50 % of all particles, activated into cloud droplets. However, in March, April and May, the fraction
 448 of accumulation mode particles acting as CCN decreased. This reaches a minimum in June and July,
 449 when only about 15 % of all particles activated into cloud droplets. From August to October a larger
 450 fraction of accumulation mode particles began to act as CCN again. The seasonal variation in the
 451 fraction of aerosol particles forming cloud droplets illustrates the role of organic aerosol in cloud
 452 formation. Even though the number of aerosol particles increases in the summer due to secondary
 453 organic aerosol formation, the aerosol hygroscopicity decreases, which in turn increases the D_{50}
 454 activation diameter.



455 Another possible explanation for the summertime behavior is that the aerosol activation conditions
 456 shift from the aerosol-limited cloud activation regime to the transitional regime, and a decreasing
 457 supersaturation hinders cloud formation. To inspect this, we utilized the same updraft velocity (σ_w)
 458 measurements at Pallas as presented by Virtanen et al. (2025). While monthly averages correspond
 459 to conditions being in aerosol-limited regime ($\sigma_w/N_{tot} < 10^{-3}$ m/s cm³) (Reutter et al., 2009) throughout
 460 the year, we note that the monthly 25th percentile values of σ_w are in the range of 0.1 – 0.3 m/s during
 461 April – September. Considering the elevated aerosol concentrations at Pallas and Värriö during these
 462 months, these values correspond to conditions shifting just to the transitional regime. Therefore, it is
 463 plausible that limited supersaturation had partly caused the observed ineffective aerosol activation
 464 during summer.



465 a)



466 b)

467 Figure 12. Monthly average CCN- forming potential of particle size distribution modes. Color bars
 468 denote the fractional contribution of the different particle modes to the total particle concentration
 469 (See Figure 5.). The shaded bar areas indicate particles acting as CCN. a) Pallas, b) Värriö.

470

471 What should also be kept in mind is that the D_{50^-} activation diameter has a wide statistical distribution.
 472 It would be a misinterpretation to conclude that Aitken mode particles did not activate into cloud
 473 droplets. In fact, particles from the Aitken mode activated throughout the year (Fig. 7). However, it is
 474 clear that particle size distribution plays a significant role in cloud formation on average, and that an
 475 increase in larger particles will have a more effective cloud formation potential compared to smaller
 476 particles.

477

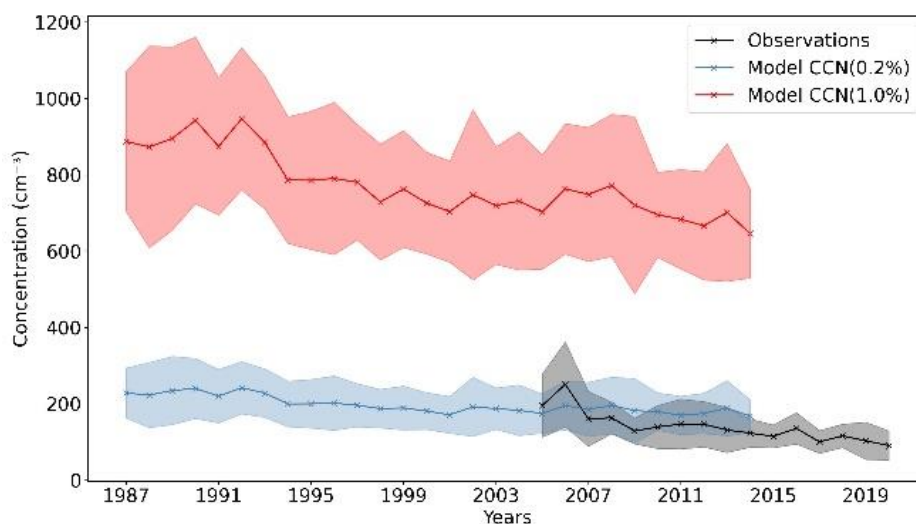
478 3.4. COMPARISON TO GLOBAL CLIMATE MODEL RESULTS

479 One of the motivations of the study was to provide observational constraints to global climate models.
 480 Utilizing data from two stations with a distance similar to global climate model grid cell size mitigates
 481 the uncertainty of observation representativeness. Here, we compare the observed seasonal- and
 482 long-term trends for aerosols and CCN at Pallas and Värriö to the TM5 (for aerosol size distribution)
 483 and the EC-Earth3 AerChem (for CCN) model results.

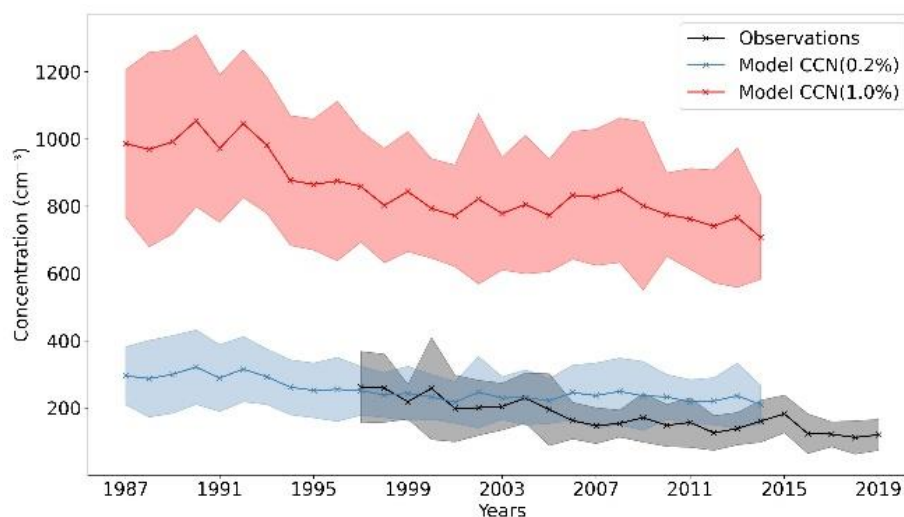


484 Figure 13 illustrates the comparison of long-term CCN concentration trends between EC-Earth3 and
 485 the observations. We utilized CCN- concentrations at pre-determined 0.2% and 1.0%
 486 supersaturations as outputs from EC-Earth3. This was chosen despite the availability of CDNC
 487 concentrations as the model takes into account the cloud cover resulting in a dataset that is not
 488 directly comparable to the observations. What can consistently be observed from both stations' data
 489 is that the modelled long term CCN concentration at 0.2% supersaturation follows closely the
 490 observed decreasing N_d trend, with concentration within the variance range. This can be considered
 491 a very good estimation, given that the critical supersaturation inside the real clouds was estimated
 492 earlier (Anttila et al. 2009) to vary between 0.18 – 0.26. The probability density functions of CCN
 493 from EC-Earth3 are provided in Supplementary figure S4.

494



495 a)



496 b)

497 Figure 13: Yearly average values of measured CCN values, modelled CCN at 0.2% and 1.0%
 498 supersaturation and modelled CDNC values. a) Pallas; b) Värriö. The shaded area represent variance.

499

500 To pinpoint reasons for possible discrepancies between the model and observations, aerosol size
 501 distributions were modelled by the TM5 model for a representative period of 10/2019-09/2020. The
 502 measured and modelled aerosol concentrations are quite similar to each other. However, there some
 503 differences in the modal concentrations, with the model overestimating nucleation mode
 504 concentrations while underestimating accumulation mode concentrations, see Table 3. As the CCN
 505 concentration is sensitive to the concentration of the accumulation mode particles, this may also
 506 decrease the amount of modelled CCN's. This is in contradiction with the slight overestimation that
 507 can be observed in Figure 13.

508

509 Table 3. The average $\pm$ standard deviation of monthly data for nucleation (N_{Nuc}), Aitken (N_{Ait}) and
 510 accumulation (N_{Acc}) mode particle concentrations from the measurements and TM5 from 10/2019 to
 511 09/2020.

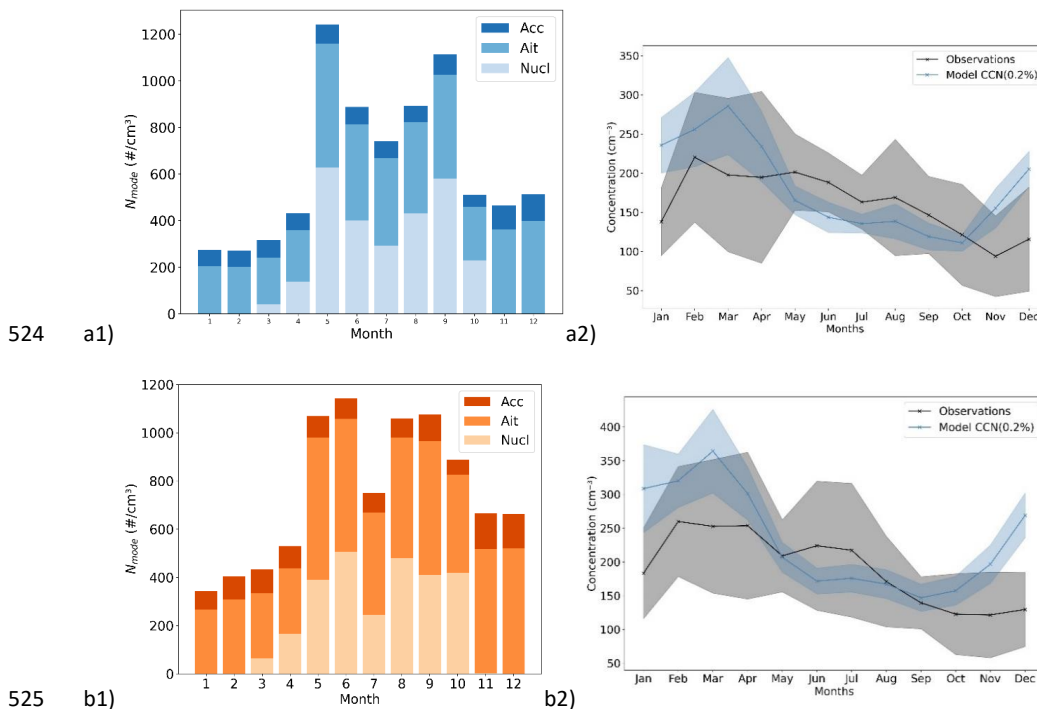
	N_{Nuc} #/cm ³	N_{Ait} #/cm ³	N_{Acc} #/cm ³	N_{Tot} #/cm ³
Pallas				
Measured	163 $\pm$ 145	413 $\pm$ 325	144 $\pm$ 139	729 $\pm$ 528
TM5	229 $\pm$ 235	331 $\pm$ 115	79 $\pm$ 17	638 $\pm$ 331
Värriö				



Measured	151±132	424±337	189±153	783±559
TM5	224±208	432±124	97±25	752±290

512

513 The climate-model-predicted seasonal variation of aerosol concentrations (Figures 14a1 and 14b1)
 514 are similar to observations (Figure 11 and 12), however with TM5 predicting maximum concentrations
 515 already during May–June, rather than in July as seen in the observations. We can speculate that this
 516 is caused by an underestimation of secondary aerosol formation, and/or missing growth to
 517 accumulation sized particles. Furthermore, TM5 predicts relatively higher aerosol concentrations in
 518 November and December compared to the measurements especially in Värriö. This propagates to the
 519 estimation of CCN concentrations, which rise sharply from October to January, unlike seen in the
 520 observations. The aerosol population during this period is dominated by particles transported from
 521 surrounding regions. Prank et al. (2010) shows that emissions from Nikel have partly been attributed
 522 to Murmansk, which is closer to the two stations considered in this study. A similar misattribution
 523 could partially explain the overestimation of aerosol- and CCN concentrations.



526 Figure 14 : Monthly average values from TM5 at a) Pallas and b) Värriö for 1) nucleation-, aiten- and
 527 accumulation mode particle concentrations; and from EC-Earth3 2) for N_{CCN} at $SS=0.2\%$ compared to
 528 observations.

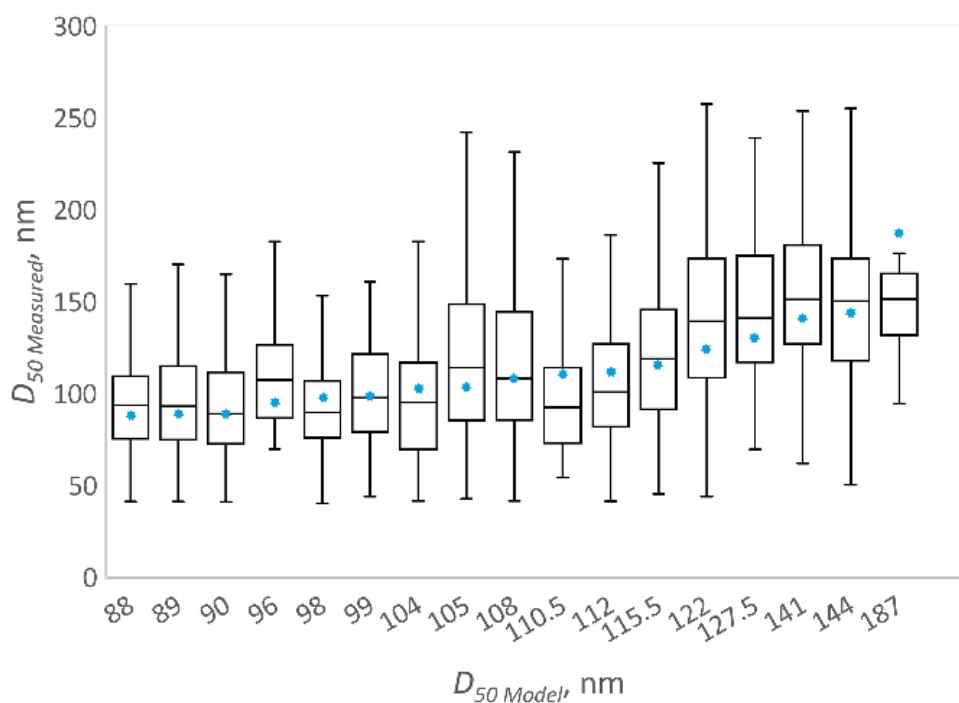


529

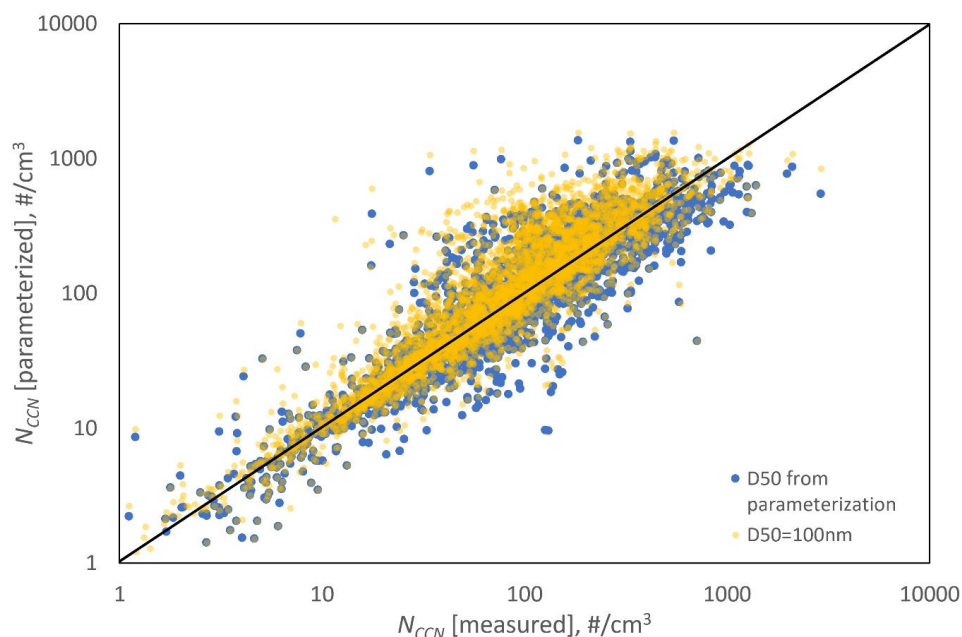
530 3.5 DIAGNOSTICS OF THE CCN-PARAMETERIZATION

531 3.5.1 PARAMETERIZATION PERFORMANCE EVALUATION

532 The CCN-parameterization performance was evaluated by first investigating the agreement of the D_{50}
533 diameters predicted by the parameterization from each trajectory/temperature-bin pairs (Table S3)
534 against D_{50} determined at Pallas during in-cloud events outside of the parameterization training
535 period, i.e. from 2005 to 2007, and from 2015 to 2020. For the measurements, the total- and
536 interstitial size distribution data was used, and equations 1 and 2, including the data quality criteria,
537 were used to deduct the D_{50} -diameters. Figure 15 illustrates that there is reasonable agreement
538 between the measured median D_{50} -values and the D_{50} -values from the parameterization. However,
539 the distribution of the measured D_{50} values was relatively broad for all individually parameterized D_{50} -
540 values, with the observed 25th percentiles being on average 23 nm lower, and the 75th percentiles 25
541 nm higher than the parameterized D_{50} -diameter. Furthermore, for the very highest D_{50} class (for
542 Western airmass and $10\text{ }^{\circ}\text{C} < T < 15\text{ }^{\circ}\text{C}$), the training dataset gives a clearly higher value compared to the
543 evaluation dataset. This is likely due to the small training sample size (28 data points) for this individual
544 data bin.



545
 546 Figure 15. Statistical evaluation of the D_{50} diameter median diameters derived from the
 547 parameterization. The blue stars denote to parametrized bin median values, and the box plot
 548 illustrates the corresponding measured values (10th, 25th, 50th, 75th and 90th percentiles) during in-
 549 cloud events from 2005 to 2007, and from 2015 to 2020 at Pallas.
 550



551

552 Figure 16. Comparison of the measured (using the total and interstitial DMPS set-up) vs.
 553 parameterized hourly N_{CCN} during in-cloud events from 2005 to 2007, and from 2015 to 2020 at Pallas.

554

555 Figure 16 displays the comparison between the measured CCN- concentrations and those from the
 556 parameterization during the evaluation period. To evaluate the parameterization against in-cloud
 557 measurements, now the total inlet size distribution, with eq. 3, was used as base data for the
 558 parameterization. Overall, the parameterization predictions agree well with the measurements, with
 559 $R^2 = 0.83$ and a slope of 0.95. We also tested how a conventional method of assuming a fixed D_{50} -
 560 diameter compares with the parameterization approach in estimating N_{CCN} . Assuming a $D_{50} = 100$ nm
 561 resulted in $R^2 = 0.82$, and a slope of 0.90, i.e. nearly identical to the parameterization. This highlights
 562 the role of the broad statistical distribution of the individual D_{50} - diameter values from the
 563 parameterization. While the approach is suitable for statistical tasks such as comparing against climate
 564 simulations, it is less suitable for investigating individual cloud cases, as discussed in the following
 565 “Parameterization limitations”- section. The largest benefit of using the parameterization approach
 566 compared to using a fixed D_{50} value in our study, occurs when the temperature is between 5 °C and 15
 567 °C, and the trajectories arrive from the East and West sectors. For these combinations $D_{50} = 100$ nm
 568 would clearly be too low a value, leading to considerable overestimation of CCN concentrations.

569



570 3.5.2 PARAMETERIZATION LIMITATIONS

571 There are several sources for uncertainties when determining CCN- concentrations with the method
 572 used here. First, as the measurements are made inside a real cloud, there are fluctuations in the
 573 supersaturation. Second, the relationship of the aerosol chemical composition with air mass
 574 classification and temperature is naturally not constant. Both of these cause a distribution of D_{50} -
 575 diameter values (see Table S3), leading to an uncertainty in the parameterized N_{CCN} . Third, there are
 576 measurement errors and instrumental noise in both DMPS instruments which become amplified when
 577 being used in calculations of the activated fraction. Affirmatively, using direct measurements of the
 578 activation ratios often resulted in activation curves that were not sigmoidal, especially at low
 579 concentrations.

580 More strict definitions of air mass trajectory source regions, as well as utilizing updraft velocity
 581 information, could help constrain the difference between the parameterization and measured N_{CCN} .

582

583 4. SUMMARY AND CONCLUSIONS

584 In this study, we presented long-term CCN concentrations in the Finnish sub-Arctic, based on surface
 585 measurements at Pallastunturi- and Värriö- research stations. At Pallas, the research station is
 586 regularly inside low-level clouds, representative of large sub-Arctic stratocumulus cloud fields. Here,
 587 aerosol activation measurements between 2005 and 2020 were utilized to develop a simple
 588 parameterization of cloud activation, and to assess the performance of the parameterization. The
 589 parameterization is based on derivation of the aerosol activation diameter (D_{50}) during different
 590 meteorological conditions (air mass origin and temperature) and estimating the CCN- concentration
 591 from aerosol size distribution measurements. As the parameterization is based on actual aerosol
 592 activation measurements inside real clouds, supersaturation as a variable is implicitly embedded in it.
 593 This parameterization was then applied at Värriö, where aerosol size distribution measurements have
 594 been made during 1998-2021. This provided a time series of CCN concentrations which, to our
 595 knowledge, is the longest reported from the Arctic region.

596 The median D_{50} value measured at Pallas was 105 nm, with lower and upper quartiles of 84 and 133
 597 nm, respectively. The average annual CCN concentrations were $140 \pm 40 \text{ # /cm}^3$ and $170 \pm 50 \text{ # /cm}^3$
 598 at Pallas and Värriö, respectively. According to our data, the annual average CCN- concentrations ($\approx N_d$)
 599 have decreased by about 55 % during 1998-2021 at Värriö, and by about 45 % during 2005-2020 at
 600 Pallas. This was mostly due to decreases in anthropogenic aerosols, as statistically significant decrease
 601 occurred during the seasonal Arctic Haze period. McCoy et al. (2017) estimated the change of N_d from
 602 the pre-industrial era to present day. They utilized a combination of remote-sensing estimates within



603 stratocumulus regions from 2001- 2013, aerosol reanalysis and AeroCom Phase II models, and found
 604 an about 2.5-3.0 times increase of N_d in our study region from the pre-industrial era to present day
 605 due to human emissions (See Figure 9 in McCoy et al., 2017). If the annual decrease of N_d observed in
 606 this study remains similar, it can be estimated that N_d concentrations in the Finnish sub-Arctic will
 607 resemble pre-industrial levels already by the end of the current decade.

608 The seasonal variation of the CCN concentrations was high at both stations, and 1h- averaged CCN-
 609 concentrations spanned from less than 1 to more than 1000 $\#/cm^3$. At Pallas, CCN concentrations were
 610 high from May to August, and at Värriö, the highest CCN concentrations were additionally observed
 611 from February to April. For both sites, concentrations were lowest from October to December.

612 The determined D_{50-} values imply that mostly the accumulation mode particles act as CCN, and that
 613 the particle chemistry of the accumulation mode is in a key position to affect aerosol activation. This
 614 was particularly evident in the winter and spring data, when anthropogenic aerosols dominate and
 615 most of the accumulation mode has the potential to activate into cloud droplets. However, during
 616 summer, when natural, organic aerosol dominates, on average less than 50% of the accumulation
 617 mode particles activate into cloud droplets. The low susceptibility of cloud droplet formation to
 618 aerosol concentrations during summer may partly be also due to aerosol activation moving from an
 619 aerosol limited regime to the transitional regime.

620 Comparison measurements to EC-Earth3 global climate model results revealed that the model was
 621 able to capture the general decreasing trend in CCN concentration, and CCN modelled at 0.2%
 622 supersaturation are close to the concentration levels of the measured CCN in real clouds. However,
 623 we find that the model is unable to capture seasonal cycle of aerosol size distributions and CCN values.
 624 We can speculate that this is due to incorrect emissions of aerosol precursors resulting in missing
 625 secondary aerosol formation, and possible misattribution of emissions from the Kola- region. The long
 626 term observations provides an opportunity to understand the long-term behaviour of CCN in the
 627 model.

628 Assessment of the parameterization performance provides additional insights into aerosol-cloud
 629 interactions and their measurements inside real clouds. We found that similar meteorological
 630 conditions (classified by air mass origin and temperature) resulted in a broad distribution of D_{50-} -
 631 values. This is likely due to a fluctuating supersaturation inside real clouds and/or considerably
 632 different particle chemistry inside a single parameter class. This implies that apart from specific
 633 meteorological conditions, a pre-defined constant D_{50-} diameter can be an adequate choice when
 634 estimating CCN- concentrations from aerosol size distribution measurements.



635 Finally, a few words about the limitations of the study and of the developed cloud activation
 636 parameterization. When developing the parameterization, we did not exclude precipitating clouds or
 637 take the cloud phase into account. Obviously the exact CCN- parameterization cannot be extended
 638 very far beyond the presented study regions, as air mass classes and the temperature dependence of
 639 aerosol activation varies with the location. However, the principle and approach can be used at other
 640 locations as well. Given the above, the parameterization is applicable as a statistical approach for N_{CCN}
 641 determination, rather than for individual cloud cases. However, our study provides a simple method
 642 for extending the estimation of CCN concentrations, if, for example, campaign-type measurements
 643 have been first been conducted to determine representative D_{50} diameters.

644

645 **Author contributions**

646 MS and APH designed the study. CGS performed the experimental CCN analysis under supervision of
 647 KN and completed the first version of the manuscript. TN performed the trajectory analysis. AC, JM
 648 and RM performed the global model simulations and analysis. CGS, APH, AV, RM and JJ discussed the
 649 results. APH edited the manuscript. All authors reviewed and approved the final version of the
 650 manuscript

651

652 **Competing interests**

653 The authors declare that they have no known competing financial interests or personal relationships
 654 that could have appeared to influence the work reported in this paper.

655

656 **Data and code availability**

657 The interstitial inlet aerosol number size distribution from Pallas, and aerosol size distribution from
 658 Värriö are available through the EBAS database (<https://ebas.nilu.no>). The meteorological data sets
 659 from Pallas (Sammaltunturi) are available at <https://en.ilmatieteenlaitos.fi/download-observations>,
 660 and from Värriö at <https://smear.avaa.csc.fi/>. The total inlet aerosol number size distribution from
 661 Pallas, the derived D_{50} and CCN time series from Pallas and Värriö, and the results from the EC-Earth3
 662 and TM5 are available at the METIS data portal (DOI:10.57707/fmi-b2share.kg4ej-9sy11). The TM5-
 663 MP is available in GitLab repository hosted by KNMI. Access can be obtained by emailing Philippe Le
 664 Sager (sager@knmi.nl). The TM5-MP used is the version developed in FORCeS project. The HYSPLIT
 665 model is available at <http://ready.arl.noaa.gov/HYSPLIT.php>.

666



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 669 Ecosystem Supersite and the Värriö SMEAR I station for maintaining the long-term measurements.

670

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 675 Institute through funding from the Ministry of Transport and Communications for ACTRIS.

676

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