Assessment of CALIOP-Derived CCN Concentrations by In Situ Surface Measurements

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Abstract: The satellite-based cloud condensation nuclei (CCN) proxies used to quantify the aerosol-cloud interactions (ACIs) are column integrated and do not guarantee the vertical co-location of aerosols and clouds. This has encouraged the use of height-resolved measurements of spaceborne lidars for ACI studies and led to advancements in lidar-based CCN retrieval algorithms. In this study, we present a comparison between the number concentration of CCN ($n_{\text{CCN}}$) derived from ground-based in situ and spaceborne lidar cloud-aerosol lidar with orthogonal polarization (CALIOP) measurements. On analysing their monthly time series, we found that about 88% of CALIOP $n_{\text{CCN}}$ estimates remained within a factor of 1.5 of the in situ measurements. Overall, the CALIOP estimates of monthly $n_{\text{CCN}}$ were in good agreement with the in situ measurements with a normalized mean error of 71%, normalized mean bias of 39% and correlation coefficient of 0.68. Based on our comparison results, we point out the necessary measures that should be considered for global $n_{\text{CCN}}$ retrieval. Our results show the competence of CALIOP in compiling a global height- and type-resolved $n_{\text{CCN}}$ dataset for use in ACI studies.

Keywords: CCN validation; CALIPSO validation; OMCAM; POLIPHON; aerosol-cloud interactions

1. Introduction

Aerosol particles form an important component of Earth’s radiative budget by either interacting directly with short- and longwave radiation, or indirectly by acting as cloud condensation nuclei (CCN), which affect cloud properties. Under most atmospheric conditions, aerosols are required for water vapor to condense into cloud droplets. Thus, changes in aerosol concentration may alter the number of cloud droplets formed within a cloud [1] and adjust the cloud’s extent and lifetime [2]. These aerosol-cloud interactions (ACIs) are the major contributor to the total aerosol effective radiative forcing and still remain the most uncertain component of anthropogenic radiative forcing [3].

The impact of changes in aerosol concentration on cloud droplets is non-linear. It depends not only on the aerosol’s physical (size and shape) and chemical properties (hygroscopicity) but also on ambient meteorological parameters such as water vapor content and atmospheric stability [4–6]. The aerosol concentration may vary regionally by several orders of magnitude (10 to $10^5$ cm$^{-3}$) depending on the type, strength, and proximity of sources and sinks. Today, in situ aerosol observatories provide continuous and temporally highly resolved long-term measurements of cloud-relevant aerosol properties such as aerosol-sized distribution and chemical composition and CCN concentrations at different supersaturations (ss). However, they are limited to selected geographical locations. In contrast, satellite instruments can provide global observations of aerosols and clouds and, thus, are used extensively for studying ACIs with constrained meteorology and selected cloud regimes [7–9].

The fundamental aerosol information needed to study ACIs for liquid clouds is the number of available CCN close to the cloud base as only those will interact with the cloud droplets. Satellite retrievals, however, give aerosol optical properties, which are
used either directly as proxies for the number concentration of CCN ($n_{CCN}$) or to derive information on the cloud-relevant aerosol fraction. Most satellite-based ACI studies use aerosol optical depth (AOD) [10–12] or an aerosol index (AI) [13–16] as CCN proxies. The AOD may not be an accurate proxy for CCN as it does not include any information about the size of the observed aerosol particles. For instance, a large number of small particles can result in the same AOD as a small number of large particles. Furthermore, hydrophobic particles that are less efficient CCN compared to hygroscopic particles may contribute significantly to the AOD. Conversely, the AI, as the product of the AOD, and the Ångström exponent form a slightly better qualitative CCN proxy than the AOD as it is weighted more towards fine aerosols [17,18]. To better quantify the radiative forcing associated with ACI, Hasekamp et al. [19] used polarimetric observations over oceans to infer column-integrated, aerosol-sized distributions. They further used the aerosol number concentration with a wet radius $>$150 nm as the CCN proxy and found the forcing estimates to be almost 50% higher than those where the AOD or AI were used. One of the intrinsic limitations of using any of the three CCN proxies is that they are all column-integrated parameters, i.e., they may not necessarily represent aerosols close to the cloud base, which are the ones relevant for ACI [20,21]. Moreover, the AI and polarimetric retrievals are not reliable over land [18,22], where most of the anthropogenic aerosols are generated and the concentrations are the highest. A way to overcome the shortcomings associated with CCN products inferred from observations with passive sensors is to shift towards height-resolved aerosol and cloud observations using spaceborne lidar, which is available over both land and ocean [18].

Shinozuka et al. [23] used in situ measurements to report a linear relation between $n_{CCN}$ and the aerosol extinction coefficient on a log–log scale. Following their work, Mamouri and Ansmann [24] present the first CCN retrieval algorithm for ground-based lidar, where specific aerosol-type extinction-to-number-concentration conversion factors are used to infer the number concentration of particles larger than a set radius. This particle concentration is subsequently used in CCN parametrizations to estimate $n_{CCN}$ at multiple supersaturations. The application to spaceborne Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) observations was found to give aerosol number concentrations that were in reasonable agreement with in situ measurements [25–27]. In a recent study, Choudhury and Tesche [28] presented a CCN retrieval algorithm that had been developed specifically for CALIOP application. The algorithm used normalized size distributions in the CALIOP aerosol model [29] and scaled them to reproduce the CALIOP-derived extinction coefficient. These inferred aerosol-type specific-size distributions were integrated to obtain aerosol number concentrations that were found to be in reasonable agreement with airborne in situ measurements [27]. However, a direct comparison of CALIOP-derived CCN concentrations with in situ CCN measurements is still missing in the validation of both algorithms.

Schmale et al. [30] presented multi-year continuous co-located in situ measurements of $n_{CCN}$ and aerosol size distributions at 11 ground stations that covered a variety of environments with varying aerosol signatures. Here, we compare the $n_{CCN}$ estimated from spaceborne CALIOP data using the aforementioned methodologies with the ground-based in situ measurements of Schmale et al. [30]. The concurrent in situ measurements of $n_{CCN}$ and aerosol-sized distribution were furthermore used to assess the applicability of CCN parameterizations related to different aerosol types and size ranges. Based on our results, we also suggest necessary measures for compiling a global CCN climatology. The article is structured as follows. We describe the datasets, retrieval algorithms, and comparison methodology in Section 2. The comparison between the in situ and satellite derived $n_{CCN}$ is given in Section 3. The findings and possible steps forward are summarized in the final section.
2. Data and Methods

2.1. In Situ Observations

The in situ observations used in this study were obtained from Schmale et al. [30]. The dataset consists of simultaneous measurements of aerosol-sized distributions and \( n_{\text{CCN}} \) at multiple supersaturations for 11 ground-based stations. The data have a temporal resolution of one hour and include a total of 98,677 h and 157,880 h of \( n_{\text{CCN}} \) and size distribution measurements, respectively. Of the 11 stations, we used measurements from 7 stations: Barrow, Cabauw, Finokalia, Melpitz, Vavihill, Puy de Dôme, and Seoul. Station selection was based on the availability, location, and proximity of the CALIOP overpasses relative to the station location. Other factors, such as very low aerosol concentrations (Mace Head) and presence of clouds (Jungfraujoch) close to the surface, hindered CALIOP retrievals; thus, such stations were not considered in our comparison. Among the selected stations, Puy de Dôme is a high-altitude station that represents the continental background and free-tropospheric air masses. Barrow and Finokalia are coastal stations covering Arctic and Mediterranean conditions. Cabauw, Melpitz, and Vavihill represent continental background conditions, while Seoul characterizes the polluted urban environment. The geographical location of these stations is shown in Figure 1. Details about the altitude, environment, and temporal coverage of each site are listed in Table 1. A comprehensive description of the instruments, inlet system, sampling procedure, and quality control measures used in the data collection at each station is given in Schmale et al. [30].

![Figure 1. Location of the in situ sites used in this study. The large inset gives a closer look of Europe. The small insets present daytime (blue) and nighttime (black) ground tracks of CALIOP that fall within a 3° × 3° latitude–longitude grid box centered at Melpitz and Barrow, respectively, for a randomly selected month. The world map in the background is taken from http://www.shadedrelief.com/natural3/pages/textures.html (accessed on 7 July 2022).](image)

| Station                | Environment              | Location                | Elevation | Temporal Coverage              |
|------------------------|--------------------------|-------------------------|-----------|---------------------------------|
| Cabauw, The Netherlands | near coast, continental background | 51°58′N, 4°56′E         | −1 m      | 1 January 2012–31 December 2014 |
| Melpitz, Germany       | continental background   | 51°32′N, 12°56′E        | 86 m      | 1 January 2012–31 December 2014 |
| Vavihill, Sweden       | rural background         | 56°01′N, 13°09′E        | 172 m     | 20 December 2012–11 November 2014 |
| Seoul, South Korea     | urban, monsoon-influenced | 37°34′N 126°58′E        | 38 m      | 1 January 2006–31 December 2010 |

Table 1. Details of the in situ stations considered in this study.
Table 1. Cont.

| Station                | Environment                         | Location                  | Elevation | Temporal Coverage               |
|------------------------|-------------------------------------|---------------------------|-----------|----------------------------------|
| Puy de Dôme, France    | mountain, continental background     | 45°46′N, 02°57′E         | 1465 m    | 1 January 2014–1 January 2015   |
| Barrow, USA            | Arctic maritime                      | 71°19′N, 156°37′W        | 11 m      | 20 July 2007–25 June 2008       |
| Finokalia, Greece      | coastal background, Mediterranean    | 35°20′N, 25°40′E         | 250 m     | 1 January 2014–31 December 2015 |

2.2. CALIOP

CALIOP is a two-wavelength polarization-sensitive lidar on the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite, which has been observing the vertical distribution and occurrence of aerosols and clouds since June 2006 [31]. CALIPSO aerosol products include vertical profiles of the aerosol extinction coefficient, aerosol backscatter coefficient, particle–linear depolarization ratio, and aerosol subtype. CALIPSO aerosol subtypes defined in the most recent version 4 data products include marine, dust, dusty marine, polluted dust, clean continental, polluted continental/smoke, and elevated smoke [32]. In the present work, we used the CALIPSO level 2 version 4.20 aerosol profile product [33], which included aerosol optical properties and subtype information at a uniform horizontal resolution of 5 km and a vertical resolution of 60 m within the troposphere. We also used the relative humidity profiles included in the CALIPSO product obtained from the Global Modelling and Assimilation Office Data Assimilation System [34].

CCN Concentrations from CALIOP

The Optical Modelling of CALIPSO Aerosol Microphysics (OMCAM) [28] and the Polarization Lidar Photometer Networking (POLIPHON) [24,35] are the two techniques for estimating CCN concentrations from CALIOP measurements. Prior to the application of the CALIPSO aerosol parameters, we applied all the quality control measures suggested by Tackett et al. [36] and only selected high-quality cloud-free retrievals. We also separated the dust mixtures into dust and non-dust contributions following Tesche et al. [37]. In both methods, we first needed to convert the CALIOP extinction coefficient to dry number concentrations of aerosols within a size range where they were likely to act as CCN. The number concentration was then used in aerosol-type specific CCN parametrizations to compute \( n_{j,\text{dry}} \) at defined supersaturations. POLIPHON uses a set of equations to convert the CALIOP extinction coefficient (\( \alpha \)) to a dry aerosol number concentration with radius greater than \( j \) nm (\( n_{j,\text{dry}} \)) as

\[
n_{j,\text{dry}} = C \alpha^x,
\]

where \( j \) is 50 nm for continental and marine aerosols and 100 nm for dust aerosols; \( C \) is the conversion factor; and \( x \) is the extinction exponent obtained from the regression analysis of long-term AERONET measurements of AOD and size distributions [24,38,39]. The values of \( C \) and \( x \) used in this work are listed in Table 2. Similar to Choudhury et al. [27], we used regression constants for continental (clean continental and polluted continental) and marine aerosols from Mamouri and Ansmann [24], desert dust from Ansmann et al. [38], and smoke aerosols from Ansmann et al. [39].

In contrast to POLIPHON, OMCAM uses the aerosol microphysical properties (normalized size distributions and refractive indices) in the CALIPSO aerosol model [29] and scales the size distribution to reproduce the CALIOP extinction coefficient [28] using the MOPSMAP modelling package [40]. The scaled size distribution is then used to compute the required aerosol number concentration to be used in the corresponding CCN parametrization. While evaluating the OMCAM estimated aerosol number concentrations with airborne in situ measurements, Choudhury et al. [27] found that the marine model
from Omar et al. [29] resulted in an underestimation of $n_{50,\text{dry}}$ and suggested using the AERONET-based marine model from Sayer et al. [41]. We thus used the OMCAM algorithm with an updated marine model in our validation study. To correct the ambient CALIOP extinction coefficient for the hygroscopicity of hydrophilic aerosols, we used the kappa parametrization [42] included in the MOPSMAP package with globally averaged kappa values of 0.3 for continental aerosols (clean continental, polluted continental, elevated smoke), and 0.7 for marine aerosols [43]. Schmale et al. [44] also found similar kappa values using the in situ data considered in this study. Dust is treated as hydrophobic, so no hygroscopicity correction was applied for dust retrievals. To apply the hygroscopicity correction, following Choudhury et al. [27], we first estimated the growth factors at different relative humidity (RH) values for different aerosol subtypes using the microphysical properties from the CALIPSO aerosol model and Sayer et al. [41] (for marine subtype). We then correct the CALIOP extinction coefficient by using these growth factors. Previous studies found this method to yield reasonable results even under highly humid conditions such as within the marine boundary layer [27,28]. Choudhury et al. [27] parametrized the dry aerosol number concentrations linearly ($x = 1$ in Equation (1)) for the dry aerosol extinction coefficient. The corresponding values are given in Table 2. It is worth noting that the linear relationship in OMCAM held only for the dry extinction coefficient. In contrast, the POLIPHON technique was originally formulated for ambient conditions assuming a constant RH of 80 and 60% for marine and continental aerosols, respectively [24]. Thus, for POLIPHON, we only applied the hygroscopicity correction when the ambient RH exceeded these values for the corresponding aerosol types.

### Table 2. Conversion factor ($C$ in Mm cm$^{-3}$) and extinction exponent ($x$) values for POLIPHON and OMCAM algorithms used to estimate $n_{j,\text{dry}}$ from Equation (1). Note that $x = 1$ for OMCAM.

| Type            | POLIPHON (Ambient) | OMCAM (Dry) |
|-----------------|---------------------|-------------|
|                 | $C$                | $x$         | $C$         |
| Dust            | 8.855              | 0.7925      | 11.085      |
| Clean continental | 25.3               | 0.94        |            |
| Marine          | 7.2                | 0.85        | 3.6         |
| Elevated smoke  | 17                 | 0.79        | 22          |
| Polluted continental | 25.3              | 0.94        | 24.93       |

The fraction of aerosols that can act as CCN depends not only on the particles’ physical and chemical properties but also on the atmospheric water vapor supersaturation, depends on meteorological parameters such as temperature, pressure, water content, vertical wind velocity and the resulting cooling rate. Given the complexities in measuring atmospheric $n_{\text{CCN}}$, Mamouri and Ansmann [24] defined $n_{j,\text{dry}}$ in Equation (1) as representing the $n_{\text{CCN}}$ at $ss = 0.15$–0.20%. The $n_{\text{CCN}}$ at higher supersaturations are expressed as a multiple of $n_{j,\text{dry}}$. In the present work, we considered CALIOP-derived $n_{\text{CCN}}$ at $ss = 0.2\%$ (i.e., $n_{j,\text{dry}}$) in the comparison study because this parameter was provided by all of the in situ stations. The in situ data included simultaneous measurements of hourly $n_{\text{CCN}}$ and aerosol-sized distributions [30]. To assess the CCN parameterizations used in our retrievals, we compared the $n_{j,\text{dry}}$ as estimated from the size distributions with measurements of the $n_{\text{CCN}}$ at 0.2% supersaturation. Figure 2 shows a comparison of the in situ $n_{\text{CCN}}$ from direct measurements and $n_{j,\text{dry}}$ inferred from the concurrent in situ size-distribution measurements for the sites listed in Table 1. We considered $n_{50,\text{dry}}$ for all stations except Finokalia, for which we compared $n_{100,\text{dry}}$ as this particular site is frequently influenced by dust aerosols [30,44]. This approach was also supported by the CALIOP profiles within a 3° by 3° grid box surrounding the station in which 70–90% of the monthly extinction coefficients were classified as related to dust, polluted dust, and dusty marine aerosol subtypes (not shown).
Figure 2 shows very good agreement between in situ $n_{j,\text{dry}}$ and $n_{\text{CCN}}$ with a Spearman correlation coefficient ($R$) of 0.9, a normalized mean bias (NMB) of 20%, and normalized mean error (NME) of 34%. We therefore concluded that the use of aerosol-size-based CCN parametrizations as suggested by Mamouri and Ansmann [24] provided reasonable estimates of $n_{\text{CCN}}$.

2.3. Comparison Methodology

Compared to passive sensors and the resulting column-integrated parameters, CALIOP measures height-resolved aerosol properties that can be aggregated to obtain their spatial and vertical distribution. However, CALIOP has a very small footprint on the order of tens of meters compared to the hundreds of kilometers of passive sensors. Hence, monthly CALIPSO level 3 data products are reported at a coarse $2^\circ \times 5^\circ$ latitude-longitude grid [36,45]. To compare CALIOP observations with those at the in situ stations considered in this study, we defined a $3^\circ \times 3^\circ$ latitude-longitude grid box centered at the geographical location of a station and then considered all the CALIPSO level 2 profiles within that domain. Thus, the selected profiles were used to compute the $n_{\text{CCN}}$ as discussed in Section 2.2. These individual profiles were then averaged to obtain monthly mean profiles of $n_{\text{CCN}}$ for each grid box that was compared to the in situ data. Monthly averaging was used to compensate for (i) the relatively large area in the satellite-to-surface comparison that might include scenarios in which CALIOP and an in situ site observed different air masses and (ii) the local extremes in the in situ time series that were unlikely to be covered in the satellite observations. Note that our grid was finer than the $5^\circ \times 5^\circ$ in Fanourgakis et al. [46] for comparing multi-model simulations of $n_{\text{CCN}}$ with surface in situ measurements. For comparison to the in situ data at ground, CALIOP-derived profiles of CCN concentration were averaged from the surface to 1 km in height (to capture boundary-layer aerosols) except for the alpine site (Puy de Dôme), for which the averaging was extended to a height of 2 km. The comparison method used to determine the absolute error between the satellite and in situ measurement was the NME and to assess the relative bias of the satellite retrieval it was the NMB. The Spearman correlation coefficient ($R$) was used to assess the ability of satellite retrievals to represent the variability in the in situ measurements. As the CALIOP-derived $n_{\text{CCN}}$ represented $n_{j,\text{dry}}$ ($j = 50$ nm for continental and marine, and 100 nm for dust aerosols) at 0.2% supersaturation, we also compared them with the in situ measurements of $n_{j,\text{dry}}$ ($j = 100$ nm for dust influenced Finokalia station and 50 nm for other stations). This approach enabled us to consider in situ measurements also for months that were lacking CCN measurements and to increases the number of data points to be considered in the statistical analysis.
3. Comparison of CCN Concentrations

The comparison of the monthly mean $n_{CCN}$ (at ss = 0.2%) at the in situ stations and inferred from CALIOP measurements is presented in Figure 3 and Table 3. At all sites, monthly in situ $n_{j,\text{dry}}$ are either comparable or larger than the directly measured in situ $n_{CCN}$ with average NMB and NME values of 20.7 and 39.8%, respectively and nearly identical monthly variations. Overall, the CALIOP estimates of monthly $n_{CCN}$ using OMCAM algorithm were larger than the in situ observations with a mean NMB and NME of 49% (31%) and 76% (93%) for nighttime (daytime) retrievals, respectively. The POLIPHON algorithm resulted in even larger CCN values with NMB and NME values of 129% (89%) and 138.5% (133.2%) for nighttime (daytime) retrievals, respectively. A fraction of this overestimation comes from the consideration of pure size-based CCN parameterization (Equation (1)), which considers all aerosols within the selected size limit to be CCN active. This is also seen in Figures 2 and 3, where $n_{j,\text{dry}}$ overestimated $n_{CCN}$ with a positive bias of about 20%. The statistics improve somewhat for the comparison of $n_{j,\text{dry}}$ (Table 3). The best $n_{CCN}$ absolute error agreement was found at Puy de Dôme with nighttime OMCAM estimates resulting in an absolute error of about 43%. Overall, worst agreement between CALIOP and in situ measurements was found at dust-influenced Finokalia with NME values as high as a factor of 1 for OMCAM and 1.5 for POLIPHON retrievals. Such disagreement was also reported in Choudhury et al. [27] for dust and marine aerosol mixtures and may be because of changes in the microphysical properties of the aerosol types caused by either chemical or cloud processing. Assuming dust aerosols to be hydrophobic may also have contributed to the disagreement. Overall, about 88% (91%) and 77% (88%) of either of the daytime or nighttime monthly CALIOP $n_{CCN}$ estimates from OMCAM and POLIPHON algorithms, respectively, stayed within a factor of 1.5 of the monthly in situ $n_{CCN}$ ($n_{j,\text{dry}}$) measurements. In some cases, the findings from CALIOP daytime and nighttime retrievals differed by several orders of magnitude. On closer inspection, we found that the daytime and nighttime tracks covered different geographical locations within a grid box. Furthermore, in any given month, the number of days with a daytime CALIPSO track within the considered domain was not always the same as that for nighttime overpasses. Also, the sensitivity of CALIOP to aerosols was different during day and night. These factors were likely to have caused the differences observed between the daytime and nighttime $n_{CCN}$ retrievals.

Table 3. Comparison statistics of monthly $n_{CCN}$ at ss = 0.2% (in cm$^{-3}$) and $n_{j,\text{dry}}$ (in cm$^{-3}$) derived from in situ and CALIOP measurements. The values enclosed within brackets are for daytime CALIOP retrievals and the unbracketed values represent nighttime retrievals. The normalized mean bias (NMB) and normalized mean error (NME) are given along with their averages weighted by the number of observation months for each station.

| Stations     | OMCAM NMB (%) | POLIPHON NMB (%) | OMCAM NME (%) | POLIPHON NME (%) |
|--------------|---------------|------------------|---------------|------------------|
|              | $n_{CCN}$     | $n_{j,\text{dry}}$ | $n_{CCN}$     | $n_{j,\text{dry}}$ | $n_{CCN}$     | $n_{j,\text{dry}}$ | $n_{CCN}$     | $n_{j,\text{dry}}$ |
| Cabauw       | 44 (−9.9)     | 14.2 (−30.3)     | 123.3 (36.7)  | 79.8 (6.13)      | 72 (53.2)     | 66.6 (40.6)       | 134.8 (84.6)  | 102.6 (47.7)     |
| Melpitz      | 47 (7.8)      | 43 (53.1)        | 134.9 (62.5)  | 128.5 (135)      | 82.7 (70)     | 78 (97.8)         | 151.6 (104.6) | 140.3 (154.1)    |
| Vavilhill    | 17 (54.3)     | 12.1 (33.7)      | 99.6 (147.6)  | 83.6 (115)       | 44.2 (82.2)   | 47.8 (66.5)       | 105.4 (159.2) | 92.1 (129)       |
| Seoul        | 31.5 (19.6)   | 2.7 (−10.6)      | 88.3 (62.9)   | 48.9 (23.5)      | 75.3 (80)     | 39.7 (49)         | 106.5 (97.9)  | 63.5 (59)        |
| Puy de Dôme  | 29.5 (100.8)  | 23.5 (122.2)     | 125.7 (219.2) | 115.2 (254.6)    | 43 (157.3)    | 41.4 (172.3)      | 125.7 (254.1) | 115.2 (281.1)    |
| Barrow       | 113.6 (−41.7) | 1 (−15)          | 241.7 (−15)   | 61.6 (56.7)      | 117.5 (84.9)  | 72.5 (55.8)       | 241.7 (94.2)  | 80.7 (70.9)      |
| Finokalia    | 93.9 (127.3)  | 109.8 (126.3)    | 151.9 (146.1) | 167.8 (150)      | 106.8 (189.3) | 109.8 (170.4)     | 154.7 (196)  | 167.8 (174.1)    |
| Average      | 48.8 (31.1)   | 26.7 (25.3)      | 128.6 (89)    | 91.2 (78.4)      | 75.9 (93.3)   | 62.6 (79.4)       | 138.5 (133.2) | 104.3 (107.9)    |
Along with the aerosol load, the number of days in a month observed (DMO) by CALIOP may fail to detect aerosol layers with a lower aerosol load [18,47,48]. On analyzing the number of CALIOP data bins with valid aerosol retrieval ($N_{\text{bins}}$) used to compute the monthly $n_{\text{CCN}}$ time series in Figure 3, we identified several cases where $N_{\text{bins}} < 100$ (25th percentile) coincided with outliers (semi-transparent points in Figure 3). Figure 4 shows how the comparison of CALIOP-derived CCN concentrations with the in situ measurements improved when only months with $N_{\text{bins}} > 100$ are considered in the analysis. In that case, the CALIOP estimates of $n_{\text{CCN}}$ using POLIPHON are in reasonable agreement with the in situ $n_{j,dry}$ and $n_{\text{CCN}}$ with NME values of 83 and 123%, NMB values of 62 and 108%, and R values of 0.61 and 0.7, respectively. The OMCAM estimates were in even better agreement with the in situ $n_{j,dry}$ and $n_{\text{CCN}}$ with NME values of 54 and 71%, NMB values of 9 and 39%, and R values of 0.63 and 0.68, respectively.

It is known that CALIOP may fail to detect aerosol layers with a lower aerosol load [18,47,48]. On analyzing the number of CALIOP data bins with valid aerosol retrieval ($N_{\text{bins}}$) used to compute the monthly $n_{\text{CCN}}$ inferred from CALIOP observations using the OMCAM (gray dots for nighttime data and blue for daytime data) and POLIPHON (gray diamonds for nighttime and blue for daytime data) algorithms at Cawbauw (a), Melpitz (b), Vavihill (c), Seoul (d), Puy de Dôme (e), Barrow (f), and Finokalia (g) stations. The semi-transparent data points are for the cases where the number of CALIOP data bins with finite aerosol retrievals used to produce the monthly average are less than 100.

Figure 3. Monthly time series of $n_{j,dry}$ (green) and $n_{\text{CCN}}$ (red) as derived from ground-based in situ measurements and $n_{\text{CCN}}$ inferred from CALIOP observations using the OMCAM (gray dots for nighttime data and blue for daytime data) and POLIPHON (gray diamonds for nighttime and blue for daytime data) algorithms at Cawbauw (a), Melpitz (b), Vavihill (c), Seoul (d), Puy de Dôme (e), Barrow (f), and Finokalia (g) stations. The semi-transparent data points are for the cases where the number of CALIOP data bins with finite aerosol retrievals used to produce the monthly average are less than 100.
result. Even so, CALIOP has the potential to provide height- and type-resolved \( n_{\text{CCN}} \) over both land and oceans. With regards to the ACI study, the height-resolved measurements can be used to estimate the \( n_{\text{CCN}} \) close to cloud base and the type-resolved measurements to quantify the anthropogenic component. The availability of more than a decade of CALIOP measurements provides a unique opportunity to study the global and seasonal distribution of CCN concentrations for different aerosol types. However, such a study is not within the scope of the present work and will be presented in future studies.

4. Conclusions

We presented a comparison of monthly in situ CCN concentrations \( (n_{\text{CCN}}) \) and dry aerosol number concentrations \( (n_{j,\text{dry}}) \) with the spaceborne lidar CALIOP retrievals. POLIPHON and OMCAM algorithms were used to estimate \( n_{j,\text{dry}} \) and \( n_{\text{CCN}} \) from CALIOP measurements. Both techniques rely on size-based CCN parametrizations. A comparison of the concurrent in situ measurements of \( n_{\text{CCN}} \) and \( n_{j,\text{dry}} \) at all stations supported the applicability of the size-based CCN parametrizations. We found that the CALIOP estimates of monthly \( n_{\text{CCN}} \) at 0.2% supersaturation were generally in good agreement with the in situ measurements: about 88% (91%) and 77% (88%) of \( n_{\text{CCN}} \) \((n_{j,\text{dry}}) \) estimates from OMCAM and POLIPHON algorithms remaining within a factor of 1.5 of the in situ measurements, respectively. Disagreement was primarily found for the monthly retrievals where the number of aerosol samples detected by CALIOP was less than 100 (25th percentile). Excluding such retrievals, we found the OMCAM \( n_{\text{CCN}} \) estimates to have better agreement with the in situ measurements with a normalized mean error of 71%, normalized mean bias of 39%, and correlation coefficient of 0.68.

The in situ stations considered in this validation study cover different continental environments. Future studies involving a direct comparison of CALIOP retrievals with measurements over oceans, (e.g., from Hudson et al. [49]) will provide better insight into the ability of CALIOP to estimate marine \( n_{\text{CCN}} \). Having said that, our findings along
with previous comparison studies [25–28] support the feasibility of constructing a global height-resolved \( n_{CCN} \) climatology from CALIOP measurements. Such a dataset would be invaluable not only for studying aerosol-cloud interactions [8,18] but also serve as a benchmark for regional and global climate models.

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**Data Availability Statement:** The CALIPSO level 2 v4.20 aerosol profile data product used in this work is available at https://doi.org/10.5067/CALIOP/CALIPSO/LID_L2_05KMAPRO-STANDARD-V4-20 (accessed on 7 July 2022). The in situ data is available at http://actris.nilu.no/Content/products (accessed on 7 July 2022).

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