Evaluation and improvement of CAMS-derived CCN number concentrations using in-situ measurements

Yannick Emanuel Anders, Karoline Block, Mira Pöhlker, and Johannes Quaas

Author's response to comments by referee 2 (Ying Chen, 2026)

In the following, we respond to the referee's comments (**black**) with our statements in **blue** and the adapted text from the revised manuscript in **orange**.

Referee comments

The study "Evaluation and improvement of CAMS-derived CCN number concentrations using in-situ measurements", uses in-situ measurements of CCN concentration in different places over the world to evaluate a model-derived global datasets of CCN number concentration. They performed comprehensive analysis of the bias, and proposed an approach to improve this global CCN dataset. Given CCN is a critical link between aerosol and clouds but with very sparse direct observations in terms of both temporal and spatial coverage, and human activities are an important source of aerosol, a long-term global CCN dataset is of imperative and of great interest to atmospheric and climate communities. The manuscript is well written and structured as well. I would be happy to recommend publishing of this study, and provide my comments below to help further improve the article.
The authors thank Ying Chen for carefully reading the manuscript and acknowledging the significance of our study. We appreciate their valuable suggestions and useful comments.

- Data availability. Given the importance of and of wide interest of this improved global CCN dataset, please be clear about how to access this CCN dataset.
  The improved dataset has not been published yet. Right now, this paper presents merely a suggestion of how this data could be improved based on observations - the exact realization for the entire dataset still needs to be done.

- I would suggest a bit more discussion of the limitations of this study. For example, as shown in Fig.1 that observations in highly polluted megacities, such as Beijing and Delhi, are not included in the evaluation. This would lead to unquantified uncertainty in high pollution regimes.
  Thanks for pointing this out. We have data from one megacity (Seoul, SEO) as part of our evaluation. Datasets from other stations in this regime have not met the requirements for this study or have not been available at the time of the analysis. To address this deficiency, we added a statement to Section 3.1.2 and to the summary.
  "However, examining data from other megacities, such as in South Asia and Africa, might be necessary, as our list of stations only included one (SEO)."
  Further, we acknowledged the overall uneven spatial distribution of available observations, when describing Fig. 1.:
  "As can be seen from Fig. 1, available observational sites are concentrated in Europe and North America, while Africa, Australasia and large parts of central and northern Asia remain particularly underrepresented. Furthermore, effectively only two sites (ENA/GRW, ASI) are available to represent remote oceanic environments. This uneven spatial coverage can

introduce distortions when observations from different sites are combined to derive global estimates. Therefore, oceanic and continental contributions are weighted separately in subsequent analyses, as described in Section 3.2.2."

- When interpolate CCN and supersaturation relationship, linear interpolate applied in L132 but power law in later (L140). Better to keep they consistent.
  In the interval for which we applied the first, linear interpolation ($\pm$ 0.1 %), the deviations to using a power function are small compared to the measurement uncertainty. For the second, wider interpolation more accuracy was required. Hence, we decided to use a power law there. We also added a brief statement clarifying our reasoning behind the two interpolation functions, after introducing Equations 2 and 3:
  "As mentioned, CCN_C are given at 6 specific supersaturations ($s = 0.1$ %, 0.2 %, 0.4 %, 0.6 %, 0.8 % and 1.0 %). Observed Nccn are usually not measured at the same supersaturations. Therefore, CCN_O are directly taken (or averaged if several exist) within a range of $\pm$ 0.05 %. In case there is no data in the $\pm$ 0.05 % range, we interpolated linearly in the narrow range of $\pm$ 0.1 %. The resulting uncertainties from this interpolation are small in comparison to the measurement uncertainties. However, not all supersaturations are covered at each measurement site, not even in a $\pm$ 0.1 % range. To be able to interpolate across a wider range of supersaturations, we preferred a power law to a linear function. Nevertheless, we limit the inter-/extrapolation interval $[s_1, s_2]$ to a maximum range of $\pm$ 0.4 % around the desired value s_x, to reduce the inaccuracies, which are introduced by using a constant slope k across a range of supersaturations ..."

- The study has clearly stated that volcanic aerosols are not considered and organic matters are an important source of CCN. Given wildfire is one of the major natural source of aerosol, particularly for OM, could authors please elaborate that how wildfire emissions/aerosols are considered in this CCN estimate?
  Block et al. (2024) gives information on why volcanic aerosols are not included in CAMS-derived Nccn and on the representation of wildfire and biomass-burning emissions. The reviewer will find more details to the aerosol composition in the CAMS reanalysis in Inness et al. (2019). We also extended our statement here as follows:
  "Wildfires and biomass burning events are well captured by CAMS and have large impacts on total Nccn via BC and OM, whereas aerosols related to volcanic eruptions, nitrate aerosols or stratospheric aerosols are not represented at all (Block et al., 2024)."

- I feel L247-251 is unclear, please rephrase it. How does the regime category defined by the bias? When there is a conflict between bias and observation defined category, how should a site be classified?
  As part of our revision, we simplified our classification (clean/polluted, marine/continental). The new clustering is entirely based on CAMS-derived Nccn, as this is necessary for the application of the improved parametrization. We also added a comparison of the regimes as defined by a) observations and b) CAMS-derived Nccn to Tab. A2. Lastly, we rephrased the paragraph as part of the necessary update:
  "For most stations, the pollution regime as identified by CCN_C ($s = 0.4$ %) matches the regime it would have if regimes were defined by the observational CCN_O ($s = 0.4$ %). A comparison of both definitions is part of Tab. A2. Only 4 stations deviate as a result of their bias, and thus would fall into a different category. Due to their negative bias, SMR, VAV and COR are classified in CCN_C as clean regimes instead of polluted ones. Meanwhile, PYE is classified as polluted instead of clean due its positive bias. A decrease in absolute bias would lead to a correction of pollution regime identification."

- Fig.6 Please provide units.
  Thanks for noticing. We added the unit to the x axis. The slope parameter k is without unit.

- Since the Nccn is estimated based on aerosol species, I wonder if k(s) is also species dependent, and how sensitive (or uncertainty) your results is to this dependence?
  In the preprint, we discuss the species dependence of a single-value k (e.g., in the classical Twomey power law). In Fig. 5 we show this correlation for the ratio of CAMS species organic matter (OM) and sulfate (SU). Furthermore, in Section 2.4, we refer to previous literature that investigates k for different environments and thus, aerosol compositions:
  "Instead, k has been shown to decrease with increasing supersaturation (Cohard et al., 1998; Khvorostyanov and Curry, 2006). The curvature of the activation spectrum depends on the size distribution of aerosol particles and thus, the environment of the respective air mass. Marine air masses with hygroscopic and bigger aerosol tend to have lower k values than continental environments, where small, hydrophobic particles are present (Jayachandran et al., 2020; Pruppacher and Klett, 1997)."

- L380 point-1. Although authors refer to some of previous studies about the evaluation of performance in time-variability, the discussion of time-variability (eg. seasonal variation) is superficial in this study itself. I would suggest not to call point-1 (L380) as specific focus of this study.
  We agree that we did not examine the temporal variability for every site individually. Nevertheless, we found new, valuable results with important implications when using CAMS-derived Nccn as a timeseries:
  1. Using daily 0-UTC medians, only 5 out of 25 stations show R values above 0.5 indicating at least moderate correlation of day-to-day fluctuations. This is indeed a poor result considering that the CCN data are derived from a reanalysis.
  2. We note that diurnal variations cannot be captured adequately using 0-UTC data and that there is potential for using 3-hourly resolved CAMS data to derive CCN from.
  3. The correlation coefficients of a majority of 17 stations increase for longer averaging periods, spanning over synoptic time scales to months. Still, only about half of the stations show R values above 0.5.
  4. We added Fig. A3 to support our statement that dry and wet seasons are captured in CAMS-derived Nccn, and we elaborated on the ability of CAMS to reproduce yearly cycles of CCN based on Fig. 2 and Fig. A2. Since CAMS agrees on a general yearly distribution of CCN for 13 out of 18 representative stations, we concluded that CAMS-derived Nccn mostly reproduces the observed variability on seasonal and annual time scales and is therefore suitable to be used as a climatology, despite its' deficiencies at shorter timescales.
  Thus, we value the temporal analysis at least equally as important as the investigation of different environments and the dependence of the bias to the supersaturation. We have however rephrased this in the conclusion to be more specific:
  "The evaluation focuses on three aspects of the CAMS-derived total Nccn: (1) its ability to reproduce observed temporal variability, from day-to-day fluctuations over synoptic-scale variations to monthly timescales; (2) its suitability across different environments (marine, continental) and pollution regimes (clean and polluted); and (3) its sensitivity to supersaturation."

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