Lenhardt et al. use in situ and HSRL measurements collected during the ACTIVATE campaign to investigate the factors controlling the relationship between cloud condensation nuclei (CCN) concentrations and aerosol backscatter coefficient (BSC). Given the current demand for a reliable global dataset of CCN concentrations, which have been attempted using satellite retrievals of aerosol optical properties such as BSC, their findings offer a valuable new perspective on remote sensing-based CCN retrievals. Particularly noteworthy is their demonstration of how aerosol size distribution (and thus effective radius) can vary significantly even within a given aerosol species, leading to a non-linear relationship between CCN and BSC. I found their methodology to be accurate and their interpretation of the results convincing. Uncertainties related to the study are also documented in detail. I recommend publication in *Atmospheric Chemistry and Physics* after the authors address the following (mostly minor) comments.

Comments:

Line 140: Consider discussing the observed order-of-magnitude variation in CCN concentrations for nearly constant BSC within a given aerosol type and relative humidity (RH). This would help frame the manuscript's central question regarding the drivers of such variability.

Figure 4 (second column): The label should read *o(Aerosol ID > 1)* since you exclude cases where more than one aerosol type is present.

Figure 4 caption: From the figure, it appears that only 3,939 out of 83,678 co-located samples are used in subsequent analyses. This significant filtering (about 95%) should be noted in the caption for clarity.

Lines 380–382: Please acknowledge that BSC_{theory} is also derived using approximations. For example, assuming spherical particles in Mie theory and using climatological refractive indices for different aerosol types. These assumptions may contribute to the observed discrepancies.

Sections 4.2 and 5.1: The predictors used in the analysis are not fully independent. For instance, RH affects the effective radius (R_{eff}) depending on the aerosol's hygroscopicity. Therefore, the influence of RH on the CCN–BSC relationship may already be captured via R_{eff}. Moreover, R_{eff} and geometric mean radius (*GMR*) are related through a well-defined expression if the aerosol size distribution follows a known functional form. For example, under a lognormal distribution, $\mathrm{R_{eff}} = \mathrm{GMR} \cdot exp\left(\frac{5}{2}\ln^2\sigma\right)$, where σ is the geometric standard deviation. I suggest incorporating such relationships when interpreting the relative importance of the predictors.

Section 4.2: The low impact of Aerosol ID and κ on the CCN–BSC relationship is expected, since (i) BSC is primarily determined by aerosol size, and (ii) CCN activation is more sensitive to size than chemical composition (Dusek et al., 2006). I recommend including this discussion when presenting the relative importance of predictors.

Line 12: The abbreviation "ERFaci" is not used later in the abstract; consider removing or defining it where relevant.

Line 68: More recently, Choudhury et al. (2025) also reported a similar disagreement between aerosol extinction coefficient and CCN concentrations for marine aerosols across the globe.

Line 133: The abbreviation “URB” should be defined upon first use.

Line 339: Consider replacing “unnecessary” with “anomalous” for clarity.

Line 495: Revise to: “…steeper decrease in CCN_{theory}:BSC_{theory} with R_{eff}”

Line 496: Correct the subscript format of BSC_{theory}; add “with R_{eff}” after CCN_{theory}

References:

Choudhury, G., Block, K., Haghighatnasab, M., Quaas, J., Goren, T., and Tesche, M.: Pristine oceans are a significant source of uncertainty in quantifying global cloud condensation nuclei, Atmos. Chem. Phys., 25, 3841–3856, https://doi.org/10.5194/acp-25-3841-2025, 2025.

Dusek, U., Frank, G.P., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S., Jung, D. and Borrmann, S.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, *312*(5778), 1375-1378, https://doi.org/10.1126/science.1125261, 2006.