Response to Reviewer 1:

We would like to thank the reviewer for the constructive feedback on our manuscript and for aiding our progress towards publication. These comments were very useful, and we appreciate the time taken to help improve the paper. Each comment is repeated here, and our responses are given below each one in blue text. Excerpts from the text of the paper are given in *italics*, where ***new additions are bolded*** and text removed is noted using ~~strikethrough~~. All line numbers mentioned in our responses correspond to the line numbers in the updated version of the manuscript.

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.
Thank you for the constructive and positive feedback on our study!

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.
Thank you for this suggestion! We looked at the range of CCN_{obs} corresponding to the peak of the BSC_{obs} (0.0006-0.0008 km-1sr-1) and RH (80-90%) distributions and added the following sentences to Lines 144-148:
"If we consider, as an example, the subset of SFS CCN_{obs} with BSC_{obs} between 0.0006-0.0008 $km^{-1}sr^{-1}$ and RH between 80-90%, both small ranges that capture the peak of observed conditions for SFS aerosols, CCN_{obs} ranges from 25 to 2128 cm^{-3}. While this range captures the maximum observed variability, similar magnitudes can also be seen for MPM and URB aerosols within similar small ranges of BSC_{obs} and RH."

Additionally, to reiterate this point in the next paragraph when motivating the rest of the study, the following sentence has been added to Lines 160-161:

"Additionally, we find that within individual aerosol types and for small ranges of BSC_{obs} and ambient RH that the magnitude of CCN_{obs} can vary by nearly two orders of magnitude."

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

Since the Aerosol ID defines aerosol types using arbitrary numbers, we exclude cases where more than one numeric value (aerosol type) is present. Therefore, we left this as σ(Aerosol ID) > 0 to signify that there is no variation in HSRL-2 defined aerosol types for a given data point.

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.

To clarify this significant filtering, the Figure 4 caption now reads:

"Flowchart describing the data filtering steps applied to data for all analyses and filtering steps applied to data for specific analyses. The number of points remaining after each step (n) is given in parentheses. ***Therefore, approximately 25% of the original number of collocated samples remain for the observational analysis in Sect. 2, 5% for analyses comparing observations and theoretical values in Sect. 4.1, and 7% for purely theoretical analyses in Sect. 4.2 and 4.3."***

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.

To clarify and acknowledge the role of approximations in the BSC theory calculations, the following sentence was added to Lines 410-412:

"Other discrepancies in the BSC_{theory} calculation may come from approximations including the Mie theory assumption of spherical particles and our use of literature average refractive index values for different aerosol types."

Sections 4.2 and 5.1: The predictors used in the analysis are not fully independent. For instance, RH affects the effective radius (Reff) depending on the aerosol's hygroscopicity. Therefore, the influence of RH on the CCN–BSC relationship may already be captured via Reff. Moreover, Reff 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, Reff = GMR $\cdot\ ex\ p(5\ 2\ \ln 2\sigma)$, where σ is the geometric standard deviation. I suggest incorporating such relationships when interpreting the relative importance of the predictors.

Thank you for pointing this out. We have added some discussion of these relationships in Sect. 4.2 (Lines 452-461), which now reads as:

"It is important to note that the predictors used in this analysis are not fully independent. For example, RH impacts R_{eff} depending on the corresponding kappa value, meaning that the influence of RH on the CCN_{theory} – BSC_{theory} relationship may be captured through R_{eff}. However, we include both parameters separately to investigate if one of these variables is more important than the other in constraining the CCN_{theory} – BSC_{theory} relationship. *Additionally, both R_{eff} and GMR capture the shape of the size distribution* ***and can be related through functional relationships.*** *We use R_{eff} and GMR separately because of their different ~~magnitudes and varying~~ information content. The weighting of R_{eff} toward larger particles increases its relevance for AOPs, while GMR tends to fall within the fine mode of the size distribution closer to D_{crit} and aerosol sizes relevant for CCN activation. Therefore, based on this combination of input variables we train the RF models to predict the ratio of CCN_{theory}:BSC_{theory}."*

An additional discussion of these relationships in terms of interpreting the predictor importance estimates was added in Sect. 5.1 (Lines 544-548), which reads as:

"As previously mentioned, RH also has an impact on R_{eff} that depends on kappa. The indication that R_{eff} is the most important predictor suggests that understanding the CCN_{theory}:BSC_{theory} relationship as based on ACTIVATE observations is not as straightforward as simply constraining RH, as could be done in L23. Rather, the impact of RH on the aerosol size distribution is more important in determining how CCN_{theory} and BSC_{theory} are related."

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.
The following sentence was added at the end of Sect. 4.2 (Lines 473-475):
"The relatively low importance of Aerosol ID and kappa in these models is expected, considering BSC_{theory} is primarily determined by aerosol size and CCN activation is also more sensitive to size than to aerosol chemical composition (Dusek et al., 2006)."

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

The ERFaci abbreviation was removed from the abstract for clarity.

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.

Thank you for making us aware of this additional paper! Choudhury et al. (2025) was added to the list of citations in Line 68 and to the reference list.

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

The following sentence was added to Lines 126-127:

"In this study, we combine smoke with fresh smoke (SFS) and marine with polluted marine (MPM) due to similarity in their optical properties. We also consider the urban/pollution (URB) aerosol type."

Additionally, the sentence in Lines 206-207 was adjusted to read as:

"As in Sect. 2, we *combine smoke with fresh smoke (SFS) and marine with polluted marine (MPM) due to similarity in their optical properties. We also consider the urban/pollution (URB) aerosol type."*

This is done to clarify that we use the same aerosol type abbreviations and combinations in the observational analysis and the theoretical calculation analysis.

Line 339: Consider replacing "unnecessary" with "anomalous" for clarity.

"Unnecessary" was changed to "anomalous."

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

This sentence was revised to include "with R_{eff}."

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

The subscript was corrected and "with R_{eff}" was added.