Authors: Thank you for the review.

Anonymous Referee #2
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In this manuscript, the authors examined the relationship between the number concentration of boundary-layer cloud condensation nuclei (CCN) and light extinction by dried particles to analyze underlying aerosol processes influencing CCN-AOD relationship and satellite-based CCN estimates. Using airborne and ground-based observations of aerosols at about one kilometer horizontal resolutions, the authors derived a new parameterization between the CCN concentration and light extinction of dried particles.

They found that the slope of the logCCN–log(ExtCoef) relationship, to be 0.75_0.25, is smaller than existing parameterizations. This is a well-written paper with useful in-depth analysis of processes and uncertainties associated with CCN-AOD relationship. The content of this manuscript is within the scope of ACP. I recommend the publication of this manuscript after the following comments are addressed.

1. Abstract. I think that the abstract should be enhanced by providing more information. It will be helpful to summarize the data analyzed under this study. The authors should also describe briefly the main difference of their approach to those of previous studies. In the last sentence, please spell out what is the "common assumptions".

Authors: To provide more information on the data, we now say "not dominated by dust", in addition to the brief description of the variables and resolution used in the study. We have stopped short of describing all nine deployments and the aerosol species encountered there, for conciseness. To describe briefly the main difference, we now say "This (…) associates a doubling of aerosol optical depth with less than a doubling of CCN, contrary to previous studies based on heavily averaged measurements or a satellite algorithm". Here, the part after "contrary to" spells out what we meant by "common assumptions".

2. The major finding of this study is that CCN-AOD slope is smaller than those derived in previous studies. Fig. 6a provides a good illustration of possible reason – data aggregation over space. I think that it will be useful to see what the slope (of CCN vs dry ext. coef.) you can get to aggregate together all data in eight panels of Fig. 2.

Authors: We have done this and produced a new figure. This exercise has improved the discussion.

Section 4.1 now says: "In principle, the discussion above would be less relevant if data were extensively aggregated. The aerosol physicochemical processes and transport phenomena would be less traceable in data averaged over, say, 1000 km or a year. Figure 6 shows the arithmetic mean and standard deviation of the CCN and dry extinction for each of the eight deployments with supersaturation between 0.3% and 0.5% and with no limit on Angstrom exponent. This figure lacks the spread of data points that is present in Figure 2 and Table 3. This figure hides the general trend that the CCN almost triples as the Angstrom exponent is increased from 0.5 to 2.0 in the finer resolution.
In practice, regression results do not change drastically upon aggregating the CCN and dry extinction. The slope through the deployment averages excluding Niamey is 0.90±0.19 with the bivariate regression (Section 2.2) when one over the standard deviation squared is used as weights for both *x* and *y*. This largely falls in the 0.75±0.25 range, though the one-sigma (the square root of the variance) value, 0.19, is greater than the values for the sorted fine-resolution data (Figure 2 and Table 3). Figure 6 also demonstrates that the standard least-squares method is sensitive to the

choice of dependent and independent variables, to reiterate our remark in Section 2.2. ∂logCCN/∂logσ is 0.80 when *x* is logσ and *y* is logCCN, 0.94 when *x* is logCCN and *y* is logσ."

As a result of these findings, the discussion in Section 4.2 between CCN concentration and AOD more accurately reflects their convoluted relationship as follows:

"Our simulation here explicitly accounts for the vertical profile and the humidity effect on extinction. Data aggregation seems to influence regression in the logCCN-logAOD space, in a manner not possible in the logCCN-logσ space (Section 4.1, Figure 6): Indifference to the humidity effect and vertical profile seems to invite the logCCN-logAOD slope to appear greater than it actually is in finer scales within aerosol types. If so, this could mislead satellite-based estimates of ACI."

3. Figure 6a is interesting but the number of data points is small. Any way to add more data points to make it more convincing?

Authors: We consider that for a future article. With the data markers from Andreae (2009), Figure 6a (now 7a) does its job of contrasting the two studies. A separate figure showing more data (e.g., AOD at the DOE sites compared with CCN) without extensive aggregation would address aerosol vertical profile and humidity response in addition to the in situ dry extinction-CCN relationship. These additional topics are important and complex, and warrant a separate paper.

4. Page 2749, line 15. Please use one sentence or two to summarize what Quaas et al. found out with regard to the magnitude of "effects".

Authors: We have inserted "Their results indicate that global climate models generally overestimate the cloud albedo effect, though this, along with the effects on cloud droplet number concentration, liquid water path and other cloud properties, varies with the location and model".

5. Page 2753, last sentence. Why? Please illustrate. Is this true for specific conditions?

Authors: This paragraph explains the adjustment of ARCTAS CCN counts for the difference between the instrument supersaturation (0.3-0.5%) and the reference (0.4%). An assumption that 20% of the particles in each SMPS size bin are hydrophobic (i.e., external mixing), instead of internal mixing, makes a negligible difference in the supersaturation adjustment, according to our simulation. This sentence has been inserted in Section 2.1.

6. Page 2754, first paragraph. What are the kapa values for other sites listed in Table 1?

Authors: We do not know. No measurement of aerosol size distribution or chemical composition is available for the ground-based data used in our study. These properties accompany more recent Azores data (starting in 2013), so a future paper may be able to better address particle hygroscopicity.

7. Page 2757, lines 16-17. What about in other locations?

Authors: "in this environment" has been inserted. The humidity varies more widely in general.

8. Page 2771, line 25. Please provide a few references to this. Satellite-based estimation of aerosol indirect radiative forcing appears to be smaller than those derived from

model (e.g., Ma et al., JGR, 2014JD021670, 2014). It would be useful to discuss in more specific the implications of your findings.

Authors: We agree that it would be useful. Because satellite-based estimates and general circulation models are complicated and because our parameterization is simple, their developers are better positioned than we are to discuss its implications. We hope that our remarks about a hypothetical simplistic model invite such discussion.

Authors: We voluntarily dropped the reference to Stier et al. More minor voluntary edits have been made in the text. They include an article ("a doubling" instead of "doubling" in several places), hyphenation ("one-kilometer", "least-squares"), an acronym ("relative humidities (RH)"), a renumbering (Figure 6 turned Figure 7), phrases for a better flow of discussion ("While our observation…" in the tenth paragraph of Section 4.1, "in this context" in the 13th paragraph of Section 4.2 and "The fact that…" in the last paragraph of Section 4.2) and the acknowledgments.