Authors: Thank you for the review.

Anonymous Referee #1
Received and published: 31 March 2015
This study examines the relationship between CCN and light extinction using multiple datasets from short-term aircraft campaigns and long-term ground-based observations in different regions around the world, and proposes a new parameterization for estimating the CCN concentration from the aerosol light extinction measurements. The underlying aerosol processes and the implications for satellite-based CCN estimates related to this study are also discussed in details. The study is very important in that it provides an in-depth analysis of a widely used relationship between CCN and aerosol optical properties which may help reduce the large uncertainties resulting from the use of satellite-based estimates of AOD as a proxy for estimating CCN that is notoriously difficult to measure, let alone on large scales.

While the concept, and some of the datasets used are not new especially in light of a recent similar study by Liu and Li (2014, ACP), use of a bivariate regression method has a merit of reducing or removing a potential bias resulting from uncertainties in both the input and output variables, although the results as shown in Figures 1 and 2 do not reveal much differences. Given the large scattering of the data, such differences may not be statistically significant, which should be tested but was not done.

Authors: The differences brought by the two regression methods are indeed small. Our figures make this clear. We state as much in the second paragraph of Section 3.1 ("A similar result is obtained from the standard least squares regression….”). The similarity is not surprising, as the error bars for our data are short, as we say in Section 3.1.

We suspect that the bivariate method would have produced noticeably different parameterizations in the previous studies. That is because their CCN and AOD data, each after substantial averaging, come with long error bars. To be precise, their bars largely represent variability rather than measurement error. But, all the same, this effect should have been considered in the regression analysis. This is stated in Section 4.2. We say this in a non-definitive tone, because we did not test their data.

As far as this is concerned, more distinctions should be made to make the study original enough to warrant the publication, as the bulk of the measurements used were made at the same sites over similar periods as those used in Liu and Li with a similar objective but somewhat different approaches. If the paper is published, both similarity and distinctions should be highlighted in the abstract. One distinction, for example, lies in the use of the aircraft measurements in ARCTAS Canada.

Authors: We are definitely interested in pointing out the differences from previous studies. The abstract now says “(…) contrary to previous studies based on heavily averaged measurements or a satellite algorithm.” We have added the following sentences in the main text: “$\partial \log CCN / \partial \log \sigma_{sp}$=1.5178 in their parameterization with the 450 nm scattering coefficient, σ_{sp}, for <80% ambient RH and SSA between 0.85 and 0.95“ in Section 1 and “The parameterizations by Gassó and Hegg (2003) and Liu and Li (2014) also have a slope greater than ours (see Section 1)” in Section 4.2. Note also that we have a subsection (Section 2.2, “our study departs from previous ones in two ways….”) specifically to explain minimized data aggregation and the use of bivariate regression.

The introduction isn’t well organized. Most of the text is concerned with two themes: relationship between CCN and aerosol optical prosperities and use of the relationship for ACI studies. However, the discussion mixes up the two themes and elaborate them back and forth without a clear flow of information. The discussion should be rearranged. Besides, no references are given in numerous places where they are apparently warranted such as: Page 2747, Line 22m after "ACI studies" (add such refs as Kaufman et al. 2005; Nakajima et al. 2001;) Page 2748, Line 1 after "CCN-AOD relationships", and Line 23 after "several parameterizations"; such references should be added as Andrea (2009), Liu and Li (2014)

Authors: We have inserted "(see below for examples)" in Page 2747, Line 22. All the references are included in Section 1.

We mix the two themes because they are tightly connected. Each parameterization of the relationship is used in a unique set of ACI studies. For example, we mention Kaufman et al. (2005) immediately after mentioning the use of AOD as a CCN proxy. Since they do not use AI, we avoid mentioning their study after introducing AI.

If the introduction appears lengthy, that reflects the large number of factors related to the CCN-AOD relationship and of previous studies. When drafting the manuscript, some co-authors suggested a longer introduction, perhaps with more details on humidity effect; others suggested a more concise one. We settled for the middle of this spectrum of opinions.

Page 2750, Line 22, It is not true that "hygroscopicity is not directly accounted for". In fact, the parameterization of Liu and Li (2014) includes a term of relative humidity to explicitly account for the hygroscopicity.

Authors: We disagree. Although supersaturation for CCN and relative humidity for extinction are specified, kappa is not. The growth of scattering upon humidity changes, fRH(85%/40%), which they discuss, is not a direct measure of particle hygroscopicity, as it also depends on particle size and refractive index. Particle hygroscopicity is essentially ignored in the existing CCN parameterizations as well as in ours, as we concede in Section 4.1.

Page 2750, Line 29, explain the meaning of "one kilometer horizontal resolutions" for airborne and ground-based observations.

Authors: We aggregate CCN and extinction data over 10–11 s for the airborne data, 240–300 s for the ground-based data. These time periods roughly correspond to one kilometer horizontal distance for the typical P-3 ground speed near the surface (~120ms-1) and for the ground-based observations under the ~4ms-1 winds. We say this in Section 2.2 Resolution and regression.

Page 2751, A brief introduction of the ARCTAS Canada should be given, if there is no pertinent paper available.

Authors: A reference to Jacob et al. (2010) has been inserted.

Page 2754, line 23-26: A two-point ïnˇA˛t to the power law distribution isn't a valid way to analyze the scattering hygroscopic growth as the error is large enough to make the calculated values meaningless. The aircraft data with 2 nephelometers at set RH values weren't scanned over a wide range. Thus further discussions on the uncertainties due to this limitation are necessary here.

Authors: We have inserted "Note that the f(RH) adjustment imposed a negligible effect on our analysis because the ambient humidity was often below 50% over central Canada."

Page 2756, line 18-20: The CCN concentration is
averaged over 11s, which means that the points in figure 1 are the mean values of CCN at different altitude. If so, it is better to give the information on the altitude of the points by using the color map since the CCN-AOD relationship significantly depends on the vertical distribution of aerosol properties, such as concentration, size and composition.

Authors: We attempted to stratify our data and saw only a weak variation in the relationship. The number of data points from our airborne observation is too small to give a definitive analysis. We raise this topic in the 4th paragraph of Section 4.2 and refer to other studies, but stop short of showing our own data stratification.

Page 2757ïijˇN explain the term "deviation within a factor of "

Authors: The explanation is given in the previous sentence, and omitted in this sentence. "(…) which means that the fit estimates CCN concentrations within a factor of 2.3 (…) of the observed value for about two thirds of the cases."

Page 2758, line 10-13: the study seems to suggest that the regression is insensitive to the choice of wavelength of the AOD. One of the challenges to estimate the CCN concentration from aerosol optical quantities lies in that the contributions of aerosol to its CCN and to optical quantities are dominated by different aerosol particle size ranges: larger to the optical extinction than to the CCN concentration. This implies that the optical quantities at short wavelength should be a better proxy for CCN than those at longer wavelengths. Please elaborate more clearly.

Authors: We had the same expectation before conducting this analysis. The sensitivity to wavelength must be smaller than the precision of the regression for this particular data set.

We hesitate to make a more general statement out of the single data set. We need more extinction data that are spectrally wide and coincident with CCN measurements. The nephelometers and PSAPs employed at DOE sites measure at 450-700 nm only. Sunphotometers help, although they do not measure dry extinction.

Page 2770: The new parameterization for estimating the CCN concentration in this study uses the Angstrom exponent (AE) as the indicator of the aerosol size. The implications for satellite-based CCN estimates based on the new parameterization significantly depend on the AE retrieval from satellite. Unfortunately, the retrieval of the AE still has very large uncertainties
(see discussion on Rosenfeld et al., 2014).

Authors: True. We now say "Satellite retrieval uncertainties may be greater, especially over land with passive sensors (Kahn et al., 2009; Levy et al., 2013; Levy et al., 2010) and for small AOD".

Authors: Thank you for the review.

Anonymous Referee #2
Received and published: 31 March 2015
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.