

Authors: Thank you for the review.

Anonymous Referee #1

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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: " $\partial \log \text{CCN} / \partial \log \sigma_{\text{sp}} = 1.5178$ 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, $f_{RH}(85\%/40\%)$, 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 fit 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 2757: 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”.