

***Interactive comment on “A simple relationship between cloud drop number concentration and precursor aerosol concentration for the regions of earth’s large marine stratocumulus decks” by D. A. Hegg et al.***

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Response to reviewers’ comments on “A simple relationship . . .” by Hegg et al Ms No. acp-2011-849

1. Reviewer no. 1

1.1 Page 28665, line 24 We agree with the reviewer that it would very interesting and useful to see a substantial comparison of MODIS retrieved AMNC with in situ measurements of the same quantity. However, to our knowledge, this has not been done. We

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find this surprising given that earlier very modest attempts to compare these quantities suggested that more work might yield very useful results (e.g., Gasso and Hegg as cited in the text). Furthermore, there have been several studies relating in situ aerosol concentrations to remotely retrieved CDNC (e.g., Boers et al, J. Geophys. Res., 111, doi: 10.1029/2005JD006838, 2006; Painemal and Zuidema, Atmos. Chem. Phys., 10, 6255-6269, 2010). In our view, this situation has arisen because of the presumed difficulty of relating remotely retrieved AMNC to CDNC, i.e., since actual CCN measurements do not relate particularly well to CDNC, as per Figure 2 of the text, and it is quite difficult to retrieve CCN, as per Kapustin et al (as cited in text), why bother to retrieve the presumably less relevant AMNC, even though it is essentially available (via look-up table) in the MODIS data base? We would hope that the results we have offered here, which suggest that in fact the AMNC is a better indicator of CDNC than CCN (due to the buffer effect we discuss) will encourage such studies. However, we consider it beyond the scope of this study.

1.2 Page 28666, last paragraph We have tried to restrict ourselves to the regions of the main Sc decks of the world, and to select those studies for which we had access to significant data. For stratocumulus decks as a whole, studies in the literature encompass CARMA, VOCALS and SAFARI – the studies we employed – but also ASTEX, ACE-2, DYCOMS and MASE. It is arguable as to the extent to which the Azores Sc deck should be considered of sufficient moment to include as a major factor in global indirect forcing but in any case the Martin et al study that we cite and discuss in the text does cover this area. Indeed, much of the data in it are from ASTEX. For ACE-2, the regional Sc are much less in extent, less stable, and appreciably less “marine.” Several studies of the CDNC-aerosol relationship are extant for the ACE-2 experiment but their focus is largely on CCN closure coupled with the link to CDNC (e.g., Snider and Brenguier, Tellus, 52B, 828-842, 2000; Chuang et al, Tellus, 52B, 843-867, 2000). Chuang et al do in fact report a comparison of CDNC with AMNC with limited data which suggests a sub-linear relationship between AMNC and CDNC but the uncertainties in the data are very large, the fit not particularly good ( $R^2=0.64$ ), and the authors note that such factors as

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entrainment may have obscured the actual relationship. The MASE study also yielded several studies of the relationship between CDNC and precursor aerosol, with the Lu et al 2007 study (as cited in the text) dealing with marine Sc – in fact the California deck. However, the authors chose to compare total below cloud aerosol (essentially CN) to CDNC as noted in our discussion. Other MASE studies, that did compare AMNC with CDNC (e.g., Lu et al, J. Geophys. Res., 113, doi: 10.1029/2007JD009304, 2008), dealt with other cloud types and locals. And, finally, there is the DYCOMS experiment and the specific study by Twohy et al cited by the reviewer. To have neglected this study was a grave oversight on our part. We simply were not aware of it. We now include this study in our discussion since its venue was the southern portion of the California Sc deck and it does compare AMNC with CDNC. Our discussion is summarized here. Following numerous previous studies, Twohy et al chose to fit their data to a non-linear relationship (quadratic polynomial) between CDNC and AMNC, deriving a curve with an  $R^2 = 0.90$ . However, the data also appear able to support a good linear relationship. To explore this, we extracted values of AMNC and CDNC from Figure 3 of Twohy et al, validating the values by reproducing the non-linear fit of the authors within the uncertainty of the regression coefficients and with an  $R^2=0.89$ . We then derived a linear fit of CDNC to AMNC with a slope of  $0.66 \pm 0.7$ , intercept of  $27 \pm 15$ , and  $R^2=0.87$ , i.e., a relationship that does not differ significantly from ours and which in fact provides a fit equally as good as the non-linear, three parameter fit of Twohy et al. Hence, we feel that this study supports our conclusions.

1.3 Page 28667, line 29 We have added the phrase, “. . .and illustrates the close connection between CDNC and AMNC” just after Fig. 1 in line 24, i.e., when the figure is first introduced. Additionally, the sentence, “This also is evident in Figure 1.” Has been appended to the end of line 29.

1.4 Table 1 Yes, Table 1 does include profiles acquired relatively close to the coast during VOCALS. We now note the range of off shore distances for the profiles in the text in the Methodology section.

1.5 Page 28664, line 8 We now give the slope of the regression line in the abstract.

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1.6 Page 28664, line 23 We have added quotes to the expression, “CCN closure studies.”

1.7 Page 28665, line 16 The dash has been deleted.

1.8 Page 28669, line 4 We have added the slope to the text here.

1.9 Figure 3 The fit equation in the figure has been enlarged.

2. Reviewer no. 2

2.1 General comment concerning relevant studies In our view, the issue raised here by the reviewer has two parts. First, is our analysis indeed representative of the regions of the main stratocumulus decks of the earth (the regions adjacent to and offshore of the coasts of California, Chile/Peru and Namibia) or have we neglected other studies that might either re-enforce or alter our conclusions? Second, taking a broader view of available data on the CDNC-aerosol link, irrespective of cloud type or venue, can we specify more explicitly when our derived relationship between CDNC and AMNC can be employed and when it is not appropriate? The first issue is essentially the same as that raised by reviewer 1 in his second comment. Hence, our response in 1.2 is also applicable to this comment. As per that response, we feel that we have indeed encompassed the available appropriate studies in our discussion – with the very noteworthy exception of the study by Twohy et al (more on this below). For example, the studies derived from the MASE experiments cited by the reviewer (the Lu et al studies) are not applicable to our analysis, in the case of Lu et al, 2009, because the comparison of CDNC was to CN rather than AMNC (as was the case with the 2007 paper by Lu et al which we did cite) while Lu et al, 2008, though indeed comparing CDNC to AMNC, did so for polluted cumulus clouds in the Gulf of Mexico. Numerous other fine studies have similar disqualifications (e.g., Raga and Jones Q.J.R. Meteorol. Soc., 119, 1419-1425, 1993; Leitch et al, Tellus, 38B, 328-344, 1986). But then, of course, there is Twohy et al. We reiterate that neglecting this study was a grave (and embarrassing since we know personally several of the authors) oversight on our part.

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As outlined in our response 1.2, we now discuss this important study in the text and feel that it is consistent with our own analysis. The second issue is more complex and, as posed by the reviewer, somewhat open-ended. We have already discussed it in the text to some extent (page 28673, lines 11-29 carrying over to page 28674, line5). A definitive determination of where our proposed relationship is, and is not, applicable would likely be a study in itself and well beyond the scope of our effort here, which focuses on establishing that it is representative of the three major Sc decks of the world. Nevertheless, we are willing to offer further, limited discussion of contrasting results from different venues and offer a tentative rationalization for this. In this regard, the dichotomy in the CDNC-AMNC relationship between continental and maritime clouds observed by Martin et al serves as a useful starting point. Not only do the continental clouds display much more variance in the CDNC-AMNC relationship, it is clear that the slope of any proposed linear fit line would have an appreciably lower slope and larger intercept than would that for the marine clouds, suggesting less efficient activation of aerosol particles. A similar phenomenology is discernible in other studies of polluted clouds. For example, the Lu et al 2008 study suggested by the reviewer had a slope for the CDNC-AMNC relationship still lower than that found by Martin et al for their continental cases, as does the data presented by Chuang et al (op cit) for the ACE-2 study. This low slope in fact typically reflects a decrease in activation efficiency as the AMNC rises beyond a few hundred per cm<sup>3</sup>, with the efficiency at the lower AMNC concentrations similar to those we find in our data. This fall off in efficiency is the proximate cause for the non-linear regressions commonly used to fit CDNC to aerosol precursor concentrations. It is quite reasonably rationalized in terms of vapor depletion as large numbers of aerosol are activated. However, the degree of depletion and thus the fall off in activation efficiency will in principle be a complex function of the aerosol size distribution, composition and updraft velocity, not simply the number concentration (cf., Chuang et al, op cit). Furthermore, it will also be a function of the water vapor available at the lifting condensation level. As pointed out by Brenguier et al (J. Geophys. Res., 108, doi: 10.1029/2002JD002682, 2003), for marine venues,

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the higher aerosol concentrations associated with advection of pollution also bring in drier air and thus one might expect diminished activation of the available aerosol on this basis as well. Indeed, this seems likely to be of at least as much importance as differences on aerosol size distribution and composition. In this regard it is important to note that our Sc deck venues experience a wide range of aerosol size distributions and compositions, reflecting the variety of sources that impact them (e.g., Roberts et al, 2006, as cited in text; Hegg et al, 2010, as cited in text; Chand et al, 2010, as cited in text; Hawkins et al, J. Geophys. Res., 115, doi: 10.1029/2009JD013276, 2010; Haywood et al, J. Geophys. Res., 108, doi:10.1029/2002JD002226, 2003), and yet have the relatively uniform activation efficiency illustrated in Figure 3 of the text. We have already pointed out in the text (page 28674, lines 6-10) that the thermodynamic and dynamic characteristics of our venues is a major factor, perhaps the major factor, that leads to the simple linear relationship between CDNC and AMNC that we find. We will insert additional discussion of this issue, as per the above discussion, in the text.

2.2 Page 28666, Marine Sc and radiative forcing (General Comment) To address the first portion of the reviewer's question, the contribution of marine Sc to the global indirect forcing, at least at present one must rely on studies with a large modeling component. We have selected Kogan et al ( Geophys, Res. Lett., 23, 1937-1940, 1996) as support for an assertion that  $\sim 60\%$  of the indirect aerosol forcing is due to low marine stratiform clouds. The second part of the reviewer's question, why the relationship we give is characteristic of marine Sc, has been addressed in the previous response (2.1).

2.3 Page 28667 (General Comment) The size ranges for the FSSP and CAPS (forward scattering component) probes during the studies were 2-40  $\mu\text{m}$  diameter for the FSSP and 0.5 – 45  $\mu\text{m}$  diameter for the CAPS. These values are now given in the text. For the African data, both the RAF C-130 and the UW Convair 580 used FSSP-100's for the CDNC measurements and PCASP-100'S for the AMNC measurements. We now note this in the text.

2.4 Page 28668, lines 12-14 (General Comment) We are not entirely sure what the

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reviewer is getting at here since we see no distinct change in the data at  $CCN \sim 325$   $cm^{-3}$ . However, if one neglects a single point at  $\sim CCN = 210$   $cm^{-3}$ , which has a corresponding CDNC of 500  $cm^{-3}$ , then the data do display less variance below  $CCN \sim 300$   $cm^{-3}$ . Possibly this is what the reviewer is referring to. If so, then it is certainly conceivable that the enhanced variance at higher CCN concentration is due to a wider range of supersaturations but we see no independent grounds for this and it would simply re-enforce our assertion that CCN alone are not a good prognostic variable for CDNC. Hence, we feel that no further discussion is necessary here.

2.5 Page 28670, 2nd paragraph (General Comment) The contribution of particles larger than 1  $\mu m$  to the AMNC is fairly small. It is typically a few percent and never more than 10%. We would agree that such particles are at least arguably quite important for the microphysics of marine Sc, but this importance arises from their role in modulation of the warm rain flux (as per Jensen and Lee, as cited by the reviewer) and this is not within the scope of this study. Possibly the reviewer is getting at the possibility that an enhanced rain or drizzle flux could obscure the relationship between AMNC and CDNC. This is certainly possible but the drizzle fluxes were rather low, if extant at all, for our data and, more tellingly, the AMNC-CDNC relationship was not obscured.

2.6 Page 28671, 2nd paragraph (General Comment) There seems to be a bit of confusion as to what precisely we are asserting. The citations of Bi et al and of Shao et al are provided to support our assertion that what is meant by an internal mixture is not a uniform composition from particle to particle. The issue of what internal mixing means is a matter of definition and is irrespective of the venue. Similarly, the citation of Pratt and Prather, 2010 here was provided in support of our assertion that differences in particle to particle composition lead to differences in particle to particle hygroscopicity. Again, this would be irrespective of venue. We will revise the text to clarify this. Furthermore, we certainly do not object to citing some of the studies suggested by the reviewer to add weight to our assertion and provide evidence that the assertions we have made really are independent of venue. Hence, we now additionally cite Guazzotti

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et al (2001) and Middlebrook et al (1998).

2.7 Page 28672, line 7 (General Comment) Once again there seems to be confusion as to what use we are making of the citation of Pratt and Prather (2010). We cite it here to provide support for the claim that there is an enhanced organic presence at smaller aerosol sizes. However, we feel that the reviewer has a valid point here in that it is quite conceivable that such a presence would be extant in continental air but not in marine air. The citation of Pratt and Prather alone would thus be somewhat problematic. We have also cited Neususs et al, which is in marine air, but agree that additional references are warranted and we now additionally cite Guazzotti et al (2001) and Chuang et al (2000). Both “new” studies support an enhanced organic presence at smaller size in marine air. We also clarify what precisely we are asserting here.

2.8 Page 28673, lines 11-13 (General Comment) We have in fact utilized all available data and will clarify this in the methodology section, as suggested by the reviewer. Indeed, as discussed in the text after this point (lines 16-line 5, page 28674), all three venues from which we incorporate data have a variety of aerosol sources but relatively uniform dynamics and thermodynamics. As for a differentiation of the CDNC-AMNC relationship based on such variables as air mass type, the whole point of our analysis is to show that the relationship holds for these venues independent of such variance. We feel that Figure 3, which encompasses data from different times, locales, and aerosol characteristics, does this.

2.9 Page 28673, lines 13-14 (General Comment) We presume that the reviewer is questioning why we did not include the Martin et al (1994) continental data in our analysis. Perhaps the responses we have already made to previous comments have clarified this (i.e., responses 1.2 and 2.1). However, to be sure, we note that we have now provided a discussion in the text as to why the continental data of Martin et al, should not be grouped with those from our venues. We further note that the Martin et al data are not from the three main Sc regions with which we are dealing.

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2.10 Page 28674, lines 17-19 (General Comment) We must disagree somewhat with the reviewer here. We noted in the discussion of the venues (page 28666, first paragraph) that the Sc decks in all three venues were impacted by anthropogenic aerosols and provided references in support of this assertion. How else, after all, could they be major factors in aerosol indirect forcing of climate? Possibly the reviewer simply wishes us to explicitly note that there are a variety of aerosol sources impacting these decks. To that end, we have expanded the discussion in the venues section to note this, referencing the various studies that show this (i.e., Hegg et al, 2010, as cited in text; Chand et al, 2010, as cited in text; Keil and Haywood, 2003, as cited in text; Hawkins et al, J. Geophys. Res., 115, doi: 10.1029/2009JD013276, 2010).

2.11 Page 28664, lines 21-22 (Specific Comment) The placement of the dependent clause strikes us as optimal where it is. However, this is something of a matter of taste and we clarify our meaning by converting the clause to a parenthetical phrase.

2.12 Page 28666, line 24 (Specific comment) Keil and Haywood (2003) give the sub-cloud AMNC for only two profiles. We have used both of them. The CARG archive has only three flights in the offshore Sc deck and only 3 profiles available. We used them all. We now indicate this in the text.

2.13 Page 28669, lines 18-19 and 21-22 (Specific Comment) We have re-written the text commencing with, “As a specific . . .”, breaking down the sentence into two new sentences to clarify the nature of the “buffer” discussed here.

2.14 Page 28670, line 4 (Specific Comment) We have altered the sentence to read “A second possible linkage between CDNC, precursor aerosol and in-cloud supersaturation is the one most often discussed in the literature.”

2.15 Page 28670, lines 19-20 (Specific Comment) The discussion after “most likely” is the why. To clarify this, we have appended “for the following reasons” to the end of the sentence that starts the paragraph.

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2.16 Page 28670, lines 25-26 (Specific Comment) We must disagree with the reviewer here. The first sentence states that the excess CDNC are only 14% of the AMNC while the second states that the excess CDNC is 14% of the Aitken particle concentration (on average). These are quite different assertions.

2.17 Page 28673, lines 27-30 (Specific Comment) We are not quite sure what the reviewer is getting at here but assume that it is the “role off” and the “dichotomy” in the Martin et al data that are not clear. Hence, we have reworded the sentence as, “. . . we see neither the decrease in aerosol activation efficiency with size suggested by some earlier studies nor the dichotomy between the efficiency for marine and continental airmasses evident in the Martin et al data.”

2.18 Page 28674, lines 21-22 (Specific Comment) Again, we now cite Kogan et al as per our response 2.2.

2.19 Figures 2,3 and 5 (Specific Comment) The R2 values have been reduced to 2 decimal places and the particle diameters to whole units.

2.20 Figure 4 (Specific Comment) The figure is derived from data presented by Kaku et al, 2006 (as cited in text). We have now included the phrase “Derived from Kaku et al, 2006” in the caption.

2.21 Figure 5, reference (Specific Comment) Measurements of size-resolved hygroscopicity are rare, or perhaps we should say reports of this quantity are rare. It has been implicitly measured in a number of CCN closure experiments but not actually explicitly reported (e.g., Roberts et al, Atmos. Chem. Phys., 10, 6627-6644, 2010). There have also been some reports of size-resolved  $\Delta Z$  in maritime settings that do suggest a somewhat different phenomenology than that shown in, say, Figure 4 (e.g., Bougiatioti et al, Atmos. Chem. Phys., 11, 8791-8808, 2011) but it is not at all clear that the air being examined is truly marine and the size resolution is typically rather limited. On the other hand, a few additional studies have reported hygroscopic growth factors as a function of size for either our venues or ones closely related to them (e.g.,

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Tomlinson et al, J. Geophys. Res., 112, doi: 10.1029/2006JD007771, 2007; Hegg et al, 2008, as cited in text; Swietlicki et al, Tellus, 52B, 201-227, 2000). This parameter is fairly easily converted to  $\text{DZ}$  and the results are consistent with those in the references we do cite. We feel that the phenomenology we present is indeed representative of our venues.

2.22 Figure 6 (Specific Comment) As with Figure 4 (response 2.20), the figure is based on rather than a copy of something presented elsewhere. We now add the statement, “Based on the study of Petters and Kreidenweis, 2007” to the figure caption.

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