

## Response to Anonymous reviewer 1

We thank both reviewers for their in-depth reviews and their thoughtful comments. In response to the reviewers' main criticism we have added another section and figure dealing with uncertainty estimates and outlining our argument for why we think the standard deviation is the better uncertainty estimate. Below please find our point-by-point response to the reviewer's concerns.

### Major comments

1. The authors introduce the notion of the idealized stratiform boundary layer cloud model. But they leave it unclear what the difference is to the previous adiabatic assumption. Do they include a profile of sub-adiabaticity as Boers et al. (2006)? Is CDNC in this model not vertically uniform and/or the liquid water content not linearly increasing with height? What is the assumption on sub-adiabaticity?

We have clarified this more now in Section 2.2. We use a linear profile, with condensation rate calculated from cloud top temperature at 80 % of its adiabatic value. Please note, that we introduce the terminology ISBLC in order to avoid the confusion we were facing over the last ten years when using the term 'adiabatic cloud model'. We are not defining a new cloud model. Rather, we are trying to avoid ambiguities that are embedded in the word 'adiabatic'. Our experience was that half of the community accepted the term and the other half very strongly rejected it because the model is actually sub-adiabatic. This led to long and in our view possibly pointless discussions about terminology, which we hope to avoid.

2. The uncertainty analysis is highly superficial. Fundamentally, the authors just write that in a single case (VOCALS, 20 flights) the "error" as propagated from the MODIS-retrieved reff and tau uncertainty assessed by Platnick et al. (2015) is similar in magnitude as the spatial variability at the scale considered for these cases (up to 51x51 km<sup>2</sup>). As such, it is highly astonishing that the authors take the spatial variability at face value as the uncertainty for any other cloud regime as well. The result of course is foreseeable: the variability is small for stratiform, and large for broken clouds. Although it is not unlikely that the actual error behaves like this, it cannot be concluded from the analysis by Bennartz and Rausch. I suggest the authors either perform a rigorous uncertainty analysis, or else abstain from calling the variability "uncertainty", but actually call it, e.g., "sub-scale variability".

We agree with the reviewer's comment about the lack of detail on uncertainty analysis in the first version of the manuscript. We have added more detail and an additional figure on uncertainty analysis now.

Title: clarify this is only over ice-free oceans (maybe in the abstract is also sufficient)

We added this to the abstract.

p215: Studies preceding the Bennartz (2007) satellite climatology should be acknowledged, such as Han et al. Geophys. Res. Lett. 1998 (AVHRR) and Quaas et al. Atmos. Chem. Phys. 2006 (MODIS).

Fixed.

P3 I14: what is “maximum” adiabatic value here?

The maximum condensation rate of an airparcel undergoing saturated adiabatic ascent, which is a weak function of temperature and pressure. We have clarified this in the text.

P3 I16: probably something like “...a growing body of work has been devoted to understanding...”?

Fixed

P3 I20: what is the distinction of CDNC at cloud base and the one “observed”? This should be clarified, since for the assumption of vertically constant CDNC, there seems to be no difference.

We believe this difference adequately captured by the following sentence, highlighted in italics in the following excerpt (Page 3 Line 18-21, original manuscript): “Furthermore, the cloud microphysical interpretation of retrieved CDNC is also not straight forward, as ultimately one would be interested in the number of cloud droplets activated at cloud base and not the number of cloud droplets observed. *Entrainment mixing processes, precipitation formation, and additional activation above cloud base can lead to differences between these two properties.*”

P4 I6: should read  $\text{km}^2\text{s}^{-1}$

Fixed

p4 I8: specify what type of observations

Fixed.

p4 I18: \citep[e.g.,][Brenguier} and later cite{Bennartz}

Fixed

p5 I3: it would be good to give the reader some insight about the aircraft-observed range in  $k$ . E.g. Freud and Rosenfeld (J. Geophys. Res. 2012) find  $k = 0.93$ , while values from Martin et al. 1994 and Pawlowska and Brenguier (Tellus 2003) go down to 0.67. This introduces an uncertainty of about 20%.

We have mentioned this uncertainty now (and 20% is also what we have been using in our error propagation).

p5 I16: “for the 13 years”

FIXED

p6 I1: better clarify: “liquid water cloud”

FIXED

p6 I28: some more detail is required how this series of reflectances was constructed. Which thermodynamic profiles, cloud-base heights and cloud-base updraft speeds / CCN concentrations were sampled?

We have added detail to this discussion. However, since we did not start with CCN concentration, updraft speeds and CCN concentration were not considered.

P7 I21: “horizontal and vertical” ?

FIXED

P7 126: black cross? And CDNC = 500 cm<sup>-3</sup> / H = 175 m?

FIXED

P8 15: It would be useful if the authors discussed how often the MODIS algorithm fails to diagnose partly cloudy pixels, i.e. how important this problem remains after condition (4) on p6 is enforced.

This issue is addressed in Section 4 in much detail both in terms of its impact on mean CDNC as well as in term of its impact on data density.

P8 112: correct color and numbers p8 113: “an optical depth”

FIXED

p8 115: the assumed single scattering albedo should be reported

The single scattering albedo at 0.55 micron (SSA) reported by Haywood is 0.89-0.91. The cont. poll. SSA is 0.892. These values of course depend on wavelengths. The full information can be looked up in the referenced publications, which we believe is sufficient for the study presented here.

p8 116: why “not shown”? It is represented in Fig. 1 as well, as far as I understand.

Good point. FIXED.

P9 16: “angle increases” or “angles increase”

FIXED

p9 116: the screening criteria should be explained (reference to Tab. 3)

FIXED

P9 120: “from”

FIXED

p9 122: “in a manner that”

FIXED

p10 12: “biases” - it is not a priori clear that these are biases, or do I miss a point?

We have formulated this more carefully now.

P10 115: citet

FIXED

p11 12: The authors should report how many datapoints are sorted out by the stratification criterion.

We added this information.

P11 17: From Fig. 4, it seems the problem of reduction in amount of data is mainly due to the sunglint angle > 35 criterion. However, compared to the scattering angle criterion, this one seems to be of minor importance. I suggest to split the two issues and investigate them separately.

While the sunglint angle criterion does remove a large amount of data, we could not detect any significant sensitivity of the results toward including or excluding this data. We have tried to make this

argument clearer now in the paper. It really is the scattering angle that determines systematic artifacts we are seeing. This would also make sense from the standpoint of the retrievals. Both, solar zenith angle and sunglint angle are partly correlated with scattering angle, so they do show some of the same effects because of this correlation.

P12 I20: This is a very brief uncertainty quantification discussion. The notion that spatial variability could fully represent the error seems implausible.

We agree the uncertainty quantification was too short. See our general comments further up.

P13 I22: the trend should have a per time unit.

FIXED

P13 I28: aerosol indirect effect? Or rather “radiative forcing due to aerosol-cloud interactions”?

We were thinking of the first indirect aerosol effect here

P14 I25: traceability

FIXED

p20, Fig. 2: It would be desirable to add smaller CDNC to the plot, since  $CDNC < 20 \text{ cm}^{-3}$  are frequently retrieved.

We do not believe this will add value to the analysis presented here, since we are exemplarily showing results for one observation geometry and under highly idealized conditions. If a similar method were used in a true retrieval, we agree the lower limit should be changed.

P21, Fig. 3: The caption should explain the label “fraction of open water”: is this  $1 - \text{cloud fraction}$ ? Also the scale is unclear, is this at the  $1 \times 1 \text{ km}^2$  scale?

We have added a sentence to clarify this.

P22, Fig. 4: Are the curves averaged over the year and globe for given scattering/sunglint angle?

Correct. We have revised the caption slightly in order to make this clearer. See also additional discussion further up.

P23 Fig. 5: title top panel “Difference”

FIXED

p26 I8: this is the central result of the climatology, so it deserves more attention. I suggest to move the boxes from the top panel to the (less important) bottom panel.

FIXED

Rather than reporting the number of missing months, I suggest to report the fraction of missing days in each  $1 \times 1$  grid box. For the climatology (top panel) a color should be chosen that better allows to distinguish CDNC at lower concentrations, possibly a non-linear color scale would be very helpful in this regard.

We re-plotted CDNC on a log-scale as suggested. We kept the missing months for the third panel as we feel this information is more useful to the user of the climatology, since it more directly relates to the actual content of the climatology.



p29 references: journal names should be abbreviated. Some titles are in upper cases.

FIXED

## Response to Anonymous reviewer 2

We thank both reviewers for their in-depth reviews and their thoughtful comments. In response to the reviewers' main criticism we have added another section and figure dealing with uncertainty estimates and outlining our argument for why we think the standard deviation is the better uncertainty estimate. Below please find our point-by-point response to the reviewer's concerns.

This paper describes a climatology of monthly mean cloud droplet concentrations (CDNCs) using the MODIS-Aqua satellite instrument. Some of the potential retrieval issues that can affect CDNC retrievals are discussed along with some examples of maps and seasonal cycles from the final climatology. This data set is likely to be a very useful commodity to many researchers and I commend the authors for making it available online. Thus I recommend the publication of the paper once certain issues have been addressed and additional discussion points added. The major issues are summarized next with more specific comments and typos below that.

### Major issues

One key point on which I disagree with the authors is their restriction to situations where  $re_{3.7} > re_{2.1} > re_{1.6}$ . Painemal and Zuidema (2011) showed that this criteria was often violated for MODIS retrievals of stratocumulus (generally  $re_{1.6}$ ,  $re_{2.1}$  and  $re_{3.7}$  were very close) and yet the aircraft observed LWC was found to increase with height. This is likely due to other errors in the retrieval of  $re_{eff}$  that affect the different MODIS channels in different ways. E.g. sub-pixel heterogeneity can increase  $re_{2.1}$  more than  $re_{3.7}$  even in relatively homogeneous stratocumulus (Zhang, 2012), and Grosvenor (2014) showed that resolved heterogeneity is likely to affect the  $re_{3.7}$  more than  $re_{2.1}$ . Also, King (2012) show that the information content from the different MODIS channels is not enough to be able to retrieve vertical height variation in  $re_{eff}$  and so it seems to me that this restriction might be causing some spurious filtering of the data. It is also not clear from Fig. 3 that  $re_{3.7} > re_{2.1}$  would select the cloud covered part of the curve as stated since  $re_{3.7}$  and  $re_{2.1}$  are very close, as was also seen in the Painemal and Zuidema (2012) aircraft observations. Differences between the stratified and non-stratified CDNC values are reported in Section 4.2.1, but can we be sure which is more realistic? I think that there should be more discussion about and acknowledgement of these issues.

Thanks for pointing us to this. We agree and we have addressed this issue in the paper as well as in the conclusions now.

Consideration of scattering angle effects and the effect of sunglint is made. However, the applied filtering does not take into account the likelihood of retrieval biases at high solar zenith angles. A little more detail should be given on this issue along with consideration on how this bias will affect your data set. For example, this could potentially cause large-scale biases in the winter months at high latitudes.

It can't be said for sure whether some of the scattering angle and sunglint effects in Section 4.1 are due to variations in region – certain regions or seasons may be sampled more often at certain scattering angles and with certain sunglint angle. And there can be strong natural regional changes in  $re_{eff}$ ,  $\tau$ , CDNC, etc. Is there a way to rule this out? E.g. can you look at the differences from within the same swath relative to the swath mean and then average these, so that the locations of samples are roughly the same? This is possible for sensor angle (as in Maddux, 2010) and sunglint angle, but I'm not sure

about scattering angle. There is also no direct examination of the effect of at sensor angle. This would be useful or at least you could directly refer to this from the results of Maddux (2010)? Also, the swath edges may also sample a different range of scattering angles than elsewhere in the swath, which may also cause artifacts. Have you looked into this?

Yes. We have looked into the issue regionally and the scattering angle dependency comes out very clearly regardless of where you look. We believe this is an inherent issue related to the actual retrieval code rather than something related to natural variability or unfavorable observer or solar zenith angles. We have tried to clarify this in the paper but we do not think that it is necessary to add further plots to it as the effect is clear even on a global scale.

In this regard we also feel that an expansion of the section to cover every possible situation of observer and solar zenith angles will not add to what we already know from the work of e.g. Maddux and Grosvenor. We believe the issue we are concerned with much more an algorithmic that could be remedies also in a plane-parallel world if the retrieval algorithm accounted correctly for single scattering. Further, if the angular dependencies in itself are of interest, it would make more sense to study these effects in the context of the actually retrieved effective radius and optical depth, rather than in the context of CDNC.

RE “uncertainties” – using the variability of the daily CDNC values means that a lot of what is termed “uncertainty” will actually be natural variation in CDNC, which, even with perfect retrievals, would not be expected to be constant throughout a cloud field, nor over the course of a month, nor over the climatology. I understand that given the fact that the uncertainties are not well characterized it is hard to separate the real variability from the noise introduced by retrieval errors. However, perhaps the quoted term should just be renamed “standard deviation” with it being made clear that some of this will be actual variability and some uncertainty. Also, since you can calculate the propagated CDNC uncertainty from the pixel level uncertainties in tau and reff and I think it would be useful if these were provided to get a sense of how much they contribute. However, it should also be made clear that these would not account for the entirety of the uncertainty since there will be a lot of error introduced by the forward model plane parallel independent pixel approximation that will not be captured.

We agree with the reviewers that this was discussed too superficially in the original version of the paper. We have added a figure and a section discussing uncertainty in more detail. We do believe that the term ‘uncertainty’ is the correct term though, as we are concerned with the monthly mean CDNC values.

Fig. 5 - it looks like there is very little data in box X12 – the numbers of samples going into each box should be quoted for this figure. There is also overlap between box R03 and X12, yet box X12 shows quite different results, perhaps indicating that the statistics for this box are too poor to be robust?

Your notion about X12 and R03 is correct but it really is caused by differences in the annual cycle. If you look at Fig 10. Bottom panel, you can see how different the annual cycles are in this area. Since R03 is larger and further south, it sees a much different annual cycle than X12. In retrospect, R03 is probably an ill-defined and too wide grid box.

We are not too concerned with the data density. Recall that only grid-boxes with at least ten days per month with at least 10 observations each make it into the climatology. Those were then averaged to get the average values for each of the larger boxes like X12. Thus each box has virtually hundreds of observations in it. Also, for each of the boxes, the annual cycle is relatively smooth. If we had a random noise problem, this would also show in very noisy annual cycles.

## Specific comments

**Abstract – “Resulting CDNC uncertainties for the climatology are in the order of 30% in the stratocumulus regions and 60% to 80% elsewhere”** – as discussed in the other comments is it appropriate to call this “uncertainty”. A more careful description is needed here.

We hope to have addressed the issue by providing more in-depth discussing. We do believe the term uncertainty to be appropriate though.

**p.2, L27 – “1. The cloud is assumed to be horizontally homogeneous.”** – This seems a little vague. It would be good to mention over what scales the cloud needs to be horizontally homogeneous and why. E.g. 1km, 1x1 degree or both? Or larger even? Perhaps it would be worth mentioning the application of the independent pixel approximation for reff and optical depth retrievals such that inhomogeneity over scales larger than the 1km pixel size is likely to violate this assumption with the scale being set by the degree to which net horizontal photon transport occurs (the scale of any shadows/bright spots, etc., or rather deviations from the plane-parallel reflectances, as defined in Marshak, 2006). This could be separated from the requirement of homogeneity within the 1km pixel to satisfy the plane-parallel retrieval assumption. In terms of the CDNC derivation itself only the sub-pixel homogeneity is explicitly required once we have the correct 1km reff and optical depth since 1km resolution CDNC values can be calculated.

Thanks for pointing this out. This refers to clouds at Level 2 at a scale of about 1x1 km. We have clarified this now.

**p.3, L16 – “The true three-dimensional variability of clouds poses significant challenges to any remote sensing algorithm and a growing body work has been devoted toward understand the impact of this variability on remote sensing estimates of CDNC.”** – these works should be cited here, or else you should refer to them in another section in the paper.

FIXED

**p.3, L19 – “as ultimately one would be interested in the number of cloud droplets activated at cloud base and not the number of cloud droplets observed”** – I think this would depend on the application. Some studies may be interested in how the cloud top CDNC might change due to lateral mixing, evaporation, etc., or removal of CDNC by precipitation and not necessarily just the cloud base CDNC. I can see that the cloud base CDNC would be of interest for comparing to model processes, but I think that the statement here generalizes too much.

Agreed. FIXED

**p. 4, Eqns. 2 & 4** – You should describe how  $c_w$  (condensation rate) was calculated – did you use the MODIS cloud top temperature? What pressure did you use? Did you use the full adiabatic value, or 80% of this as mentioned on p.3. An issue with the calculation used in Bennartz (2007) is mentioned in the acknowledgments - it would be good to add more detail in the main part of the paper about this issue along with a reference for the correct formula – e.g. Ahmad (2013). A discussion of the compensation of errors in  $k$ , fraction of adiabatic condensation rate,  $\text{reff}$  errors found by Painemal and Zuidema (2012) should also be included.

We have added some discussion and a reference.

**p.6 – “3. The cloud mask had to indicate the observation to be cloudy but not over ice or land.”** – does this include the filtering of sea-ice covered regions? How robust is the detection of sea-ice? This is important since this is likely to lead to a poor retrieval.

Agreed. The ice mask would be important near the ice edge, but we do not know the answer to this question. We have clarified in the text that we are only concerned with ice-free ocean.

**p.6 – “6. Observations were only considered, if the three MODIS-retrieved effective radii stacked up as  $\text{re}_{3.7} > \text{re}_{2.1} > \text{re}_{1.6}$ , as observations violating this criterion will also violate the key assumption of a vertically increasing LWC in the ISBLC.”** – this is a key point on which I disagree with the authors – Painemal and Zuidema (2011) showed that this criteria was often violated for MODIS retrievals of stratocumulus (generally  $\text{re}_{1.6}$ ,  $\text{re}_{2.1}$  and  $\text{re}_{3.7}$  were very close) and yet the aircraft observed LWC was found to increase with height. This is likely due to other errors in the retrieval of  $\text{reff}$  that affect the different MODIS channels in different ways. E.g. sub-pixel heterogeneity can increase  $\text{re}_{2.1}$  more than  $\text{re}_{3.7}$  even in relatively homogeneous stratocumulus (Zhang, 2012), and Grosvenor (2014) showed that resolved heterogeneity is likely to affect the  $\text{re}_{3.7}$  more than  $\text{re}_{2.1}$ . Also, King (2012) show that the information content from the different MODIS channels is not enough to be able to retrieve vertical height variation in  $\text{reff}$  and so it seems to me that this restriction might be causing some spurious filtering of the data.

Thanks for pointing us to this. We agree and we have addressed this issue in the conclusions now.

**p.6, L24 – “a solar zenith angle of 56 degrees. The resulting scattering angle of 124 degrees”** – it would be good to define what you mean by the scattering angle. I.e. that it is the angle between the sun and the satellite within the plane of the sun and the satellite and that 180 degrees is backscatter.

We added this information.

**p.7, L23 – “Typically, most of these issues affect the effective radius at 1.6  $\mu\text{m}$  and 2.1  $\mu\text{m}$  more strongly than the effective radius at 3.7  $\mu\text{m}$  (Zhang et al., 2012b).”** – this was true for the sub-pixel optical depth heterogeneity effect, but are there other examples? Grosvenor (2014) showed that the 3.7 $\mu\text{m}$  channel is likely to be more strongly affected by resolved 3D radiative effects, so this is not always the case. Although the latter effect is more likely at higher solar zenith angles.

We added this information and reference.

**p.7, L33 – “observations (i.e. if cloud thickness and liquid water path varies within the field-of-view).”**  
– Strictly, Zhang (2012) talk about the effect of having sub-pixel heterogeneity in optical depth (which cloud arise through combinations of LWC, CDNC, etc.) changes, rather than cloud thickness and LWP. We were not sure how this statement is different from what we wrote.

**p. 8, L 4 – “so that in the example shown in Figure 3 (right panels) the largest part of the retrievals would be (correctly) rejected.”** – it is not clear from Fig. 3 that  $re_{3.7} > re_{2.1}$  would select the cloud covered part of the curve since  $re_{3.7}$  and  $re_{2.1}$  are very close, as was also seen in the Painemal and Zuidema (2012) aircraft observations. Why not just filter based on the cloud fraction?

Sub-scale cloud fraction is not available for the MODIS observations. This is a theoretical experiment how the (in reality unknown) sub-scale cloud fraction affects the retrievals.

**p.9, L4 – “Grosvenor and Wood (2014) address issues the dependency of cloud microphysical retrievals on solar zenith angle.”** – However, it seems that the filtering applied does not take into account the likelihood of retrieval biases at high solar zenith angles and this issue is not brought up again. A little more detail should be given on this issue along with consideration on how this bias will affect your data set.

Agreed. We added this disclaimer in the conclusions.

**p.9, L14 & Fig. 4 – sunglint angle is not defined.** I’m guessing that you mean the absolute difference between the sensor zenith angle of a given pixel and the sensor zenith angle of the middle of the sunglint band within the swath, but it is not obvious.

We added the definition of sunglint angle.

**Section 4.1 –** It can’t be said for sure whether some of the effects seen are not due to variations in region – certain regions or seasons may be sampled more often at certain scattering angles and with certain sunglint angle. And there can be strong natural regional changes in  $re_{eff}$ ,  $\tau$ , CDNC, etc. Is there a way to rule this out? E.g. can you look at the differences from within the same swath relative to the swath mean and then average these, so that the locations of samples are roughly the same? This is possible for sensor angle (as in Maddux, 2010) and sunglint angle, but I’m not sure about scattering angle.

Regarding angular dependencies: Please see our answer at the very top.

**p.10, L10 –** presumably high sunglint angles (if my definition of this is correct) will be associated with high sensor angles since they will have to be near the edges of swaths. So, this could be contribute to the artifact. Why not also look at sensor angle directly? Or at least directly refer to it from the results of Maddux (2010)? However, the above interpretation seem inconsistent with Maddux (2010). They showed that the swath edge liquid optical depth values were lower and the  $re$  higher than those near nadir. Are the results consistent with Maddux (2010) as stated at line 15? Also, the swath edges may also sample a different range of scattering angles than elsewhere in the swath, which may also cause artifacts. Have you looked into this?

Regarding angular dependencies: Please see our answer at the very top.

**Section 4.2.1 –** differences between the stratified and non-stratified CDNC values are reported, but can we be sure which is more realistic? Also, it looks like there is very little data in box X12 – the numbers of samples going into each box should be quoted for Fig. 5. There is also overlap between box R03 and

X12, yet box X12 shows quite different results, perhaps indicating that the statistics for this box are too poor to be robust?

See our answer to identical point above under Major comments.



**Fig. 6** – the title uses the term “unflagged”, but “stratified” would be more consistent with the rest of the paper. Also, the frequent positive values seem inconsistent with that expected from Fig. 4 where the main effect appears to be a decrease in CDNC at high scattering angles. Or is the increase at low sunglint angles dominating for these regions? Why not also exclude the very high sunglint angles for the flagged pixels?

We have corrected the terminology. It is very hard to find the expected value for difference from Fig 4., because of the large differences in frequency of occurrence of different angular combinations. High sunglint angles are not excluded because they do not pose a retrieval challenge (virtually no direct backscatter from the ocean surface).

**p.12, L 5 – “PZ11. Various effects, including cloud top entrainment or the representativeness of the chosen condensation rate, can potentially cause the somewhat larger bias for the stratified cases.”** – It’s hard to see how this might be the case? Can you explain? Could it also be that the stratified cases might be further from the aircraft observation?

You are right. That was a weak explanation. We tentatively agree with the point you made in the very beginning and have revised the explanation accordingly.

**p.12, L10 – “However, for both 51x51 and 21x21 neighbourhoods, the mean uncertainty (size of error bars in Figure 7 and last two columns in Table 1) is reduced for stratified cases over the un-stratified cases. ... Effective radius stacking is likely decreasing the selection of pixels of inhomogeneous clouds or those subject to sub-pixel effects, which, as discussed in Section 3, can result in retrievals with a wide spread of unphysical effective radii.”** - Could this also be due to there being fewer samples in each region due to being more selective (since this would reduce the std. deviation)? Or were all boxes chosen to have all possible pixels present?

It might just be because of the suppression of noise due to the selection criterion.

**p.13, L3 – RE “uncertainties”** – using the variability of the daily CDNC values means that a lot of what is termed “uncertainty” will actually be natural variation in CDNC, which, even with perfect retrievals, would not be expected to be constant throughout a cloud field, nor over the course of a month, nor over the climatology. I understand that given the fact that the uncertainties are not well characterized it is hard to separate the real variability from the noise introduced by retrieval errors. However, perhaps the quoted term should just be renamed “standard deviation” with it being made clear that some of this will be actual variability and some uncertainty. Also, since you can calculate the propagated CDNC uncertainty from the pixel level uncertainties in tau and reff and I think it would be useful if these were provided to get a sense of how much they contribute. However, it should also be made clear that these would not account for the entirety of the uncertainty since there will be a lot of error introduced by the forward model plane parallel independent pixel approximation that will not be captured.

We have added significant discussion and clarification on the issue of uncertainty. We have also, as suggested, provided in Figure 8 quantitative analysis on the error-propagation results.

**p. 13 & Fig. 9 – “magnitude of the annual cycle”** – can you state somewhere what you actually mean by this? Is it the peak-to-peak amplitude of the annual cycle (i.e. max monthly mean minus min monthly mean), or is it calculated based on the cosine fits? “Amplitude” would be a better word than “magnitude” since it relates more directly to the definition.

Good point. We have revised this to amplitude and specified we mean the amplitude of the cosine fit.

**Fig. 10** – the bottom right panel needs the y-axis range changing to show the negative values.  
FIXED

**p. 14, L31** – “We further show that neglecting this screening does not only lead to moderate biases in the annually averaged CDNC” – Are these biases or just differences? How do we know which one is correct due to the issues raised earlier in this review?

Agreed. We changed this to read differences.

p.15, L32 – “We found some remaining retrieval artefacts in the MODIS-retrieved effective radius and optical depth that propagate through into artefacts of the CDNC climatology as well.” – it would be useful to mention the issues that you are talking about here.

Done

p.15, L6 – “However, feel that it might be beneficial in future work to re-create retrievals working directly on the Level-1 reflectances that address some of the issues we have identified.” – although, perhaps it should be mentioned that it may not be possible to resolve many of these issues even if re-processing Level-1 data – e.g. it would be hard to correct biases for 3D radiative effects, sunglint effects, etc.

Done.

**Tables** – it would make sense to alter the order of these to align with the order of mention in the paper – i.e. first Table 3, then Table 2 and then Table 1.

FIXED

## Typos

p.9, L4 – “Grosvenor and Wood (2014) address issues the dependency of” – remove “issues”.

FIXED

p.9, L20 – “Fro Figure 4”

FIXED

p.9, L22 – “vary in a manner the roughly resembles”

FIXED

p.10, L3 – “Since Aqua’s local equator crossing time around 14:30,”

FIXED

p.10, L13 - “with an strong increase retrieved optical depth” -> “with a strong increase in retrieved optical depth”

FIXED

p. 10, L19 – “4.2. Impact Stratification and retrieval artefacts on climatology” -> “Impact of Stratification...”

FIXED

p.10, L24 - “boxes are shown were all 12 months”

FIXED

p.12, L2 – “in the order of” -> “on the order of”

FIXED

p.12, L4 – “For stratified results, magnitude of the RMSE and bias increase” -> “For the stratified results, the magnitude ...”

FIXED

p.14, L25 – “a better traceable of results” -> “a better traceability of results”

FIXED

p.15, L6 – “However, feel that”

FIXED

p.20, L20 – “cover have discussed” -> “have been discussed”

FIXED

## References (only ones not cited in the paper under review are listed)

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# Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations

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**Abstract.** We present and evaluate a climatology of cloud droplet number concentration (CDNC) based on 13 years of Aqua-MODIS observations. The climatology provides monthly mean 1x1 degree CDNC values plus associated uncertainties over the global ice-free oceans. All values are in-cloud values, i.e. the reported CDNC value will be valid for the cloudy part of the grid-box. Here, we provide an overview on how the climatology was generated and assess and quantify potential systematic error sources including effects of broken clouds, and remaining artefacts caused by the retrieval process or related to observation geometry. Retrievals and evaluations were performed at the scale of initial MODIS observations (in contrast to some earlier climatologies, which were created based on already gridded data). This allowed us to implement additional screening criteria, so that observations inconsistent with key assumptions made in the CDNC retrieval could be rejected. Application of these additional screening criteria led to significant changes in the annual cycle of CDNC both in terms of its phase and magnitude. After an optimal screening was established a final CDNC climatology was generated. Resulting CDNC uncertainties are reported as monthly-mean standard deviations of CDNC over each 1x1 degree grid box. These uncertainties are in the order of 30% in the stratocumulus regions and 60% to 80% elsewhere. The climatology is available in Network Common Data Format (netCDF) and adheres to the Climate and Forecast (CF) convention. The climatology is available via Digital Object Identifier (Bennartz and Rausch, 2016, 10.15695/vudata.ees.1).

## 1 Introduction

Presently one of the largest sources of uncertainty in predicting future climate is the degree to which clouds will alter the Earth's radiative balance (Stocker et al., 2013). In particular stratiform boundary layer clouds play an important role in modulating earth albedo and are 'at the heart of tropical feedback uncertainties in climate models' (Bony and Dufresne, 2005). Aerosol-cloud interactions contribute to these uncertainties, e.g. through the First Indirect Aerosol Effect (Twomey, 1974), in which an aerosol-perturbed cloud reflects solar radiation more efficiently than an unperturbed cloud under otherwise identical conditions.

Satellite observations play a critical role in understanding current-day variability of clouds, in validating and constraining climate models, and in furthering our understanding of cloud processes. Based on earlier work (Brenguier et al.,

2000;Schuller et al., 2005;Schuller et al., 2003;Han et al., 1998). Bennartz (2007) published an initial version of a Cloud Droplet Number Concentration (CDNC) climatology for liquid boundary layer clouds using cloud parameters derived from NASA's Aqua Moderate Resolution Imaging SpectroRadiometer (MODIS). Similar efforts were reported by Quaas et al. (2006). Revised and extended versions of this climatology have been used for climate model validation and evaluation (Storelvmo et al., 2009;Hoose et al., 2008;Hoose et al., 2009;Makkonen et al., 2009;Zhang et al., 2012a;He et al., 2015;Wang et al., 2015;Ban-Weiss et al., 2014). A variety of observational case studies and process studies were also published using similar approaches for deriving CDNC (Boers et al., 2006;George and Wood, 2010;Painemal and Zuidema, 2010;Rausch et al., 2010;Bennartz et al., 2011). Various authors have addressed shortcomings and issues related to CDNC climatologies (Merk et al., 2016;Grosvenor and Wood, 2014) as well as issues related to the cloud-retrievals underlying the CDNC climatologies (Zhang and Platnick, 2011;Nakajima et al., 2010;Maddux et al., 2010;Horvath et al., 2014). Painemal and Zuidema (2011) validate MODIS-derived CDNC against in-situ observations taken in the South Pacific during VOCALS-Rex (Wood et al., 2011). Wood et al. (2012) point out important other factors, namely precipitation generation, that hamper the interpretation of CDNC results in correlative observational studies on the context of the first indirect aerosol effect. Several studies also propose different and potentially more elaborate approaches for deriving CDNC. For example, Rosenfeld et al. (2012) propose a spatially highly-resolving satellite mission dedicated to the retrieval of cloud condensation nuclei for convective clouds and also point to limitations of the CDNC retrieval approach pursued here when applied to convective clouds. Zeng et al. (2014) propose a combination of Cloud-Aerosol Lidar with Orthogonal Polarization (CALiOP) observations with MODIS observations to retrieve CDNC.

In light of these developments, the objective of the present paper is threefold. Firstly, we wish to update older versions of the CDNC climatology by Bennartz (2007) accounting for the 13 full years Aqua-MODIS observations available by now, taking advantage of the revised and improved NASA MODIS Collection 6 cloud retrievals as outlined in Platnick et al. (2015), with better traceability via DOI-assignment. Secondly, we wish to provide readers and users of the climatology with a comprehensive overview on issues and limitations of the climatology. Thirdly, wish to quantitatively assess possible systematic error sources, quantify uncertainties, and provide validation of the climatology.

Systematic errors and uncertainties in the CDNC climatology stem largely from assumptions made in the retrieval process and in the cloud model that underlies the CDNC retrievals. In particular three assumptions are critical throughout the retrieval process:

1. The cloud is assumed to be horizontally homogeneous at the scale of an individual MODIS pixel of about 1x1 km. This is an assumption frequently made in various cloud retrievals, not only CDNC retrievals. It potentially leads to systematic errors for situations, where the observed scene deviates from horizontal homogeneity.
2. Cloud liquid water content (LWC) is assumed to increase linearly from cloud base to cloud top. The rate of increase of LWC with height (in kg/m<sup>4</sup>) is often called the 'condensation rate' similar to the use of the term 'lapse rate' for the change of temperature with height. The assumption of linearity has been a matter of some confusion in the context of CDNC retrievals. We point out that it is not necessary to hold the condensation rate constant at its

maximum adiabatic value, which is a weak function of temperature and pressure. A strict linear increase in LWC is also not necessary as long as reasonable assumptions about the vertical LWC profile are made. For example, Boers et al. (2006) relax the strict linear assumption near cloud top to better account for cloud top entrainment.

3. CDNC is typically assumed constant throughout the cloud's vertical extent. This assumption can also be relaxed somewhat as long as a clear dependency of CDNC on height above cloud base exists.

In the past, we have labelled these three assumptions collectively as the 'adiabatic cloud model'. This labelling appeared justified because the above-described assumptions are inspired by an idealized air parcel rising under moist adiabatic conditions. However, here we refrain from using the term 'adiabatic cloud model' as it has led to misunderstandings mostly revolving around the usage of the word 'adiabatic'. Indeed, the cloud model described above is not strictly adiabatic as it allows for modification and deviations from strict adiabaticity in terms of the profile of both, LWC and CDNC. A term that probably better, albeit less attractively, describes the intent of the above assumptions would be the 'Idealized Stratiform Boundary Layer Cloud' (ISBLC) model, which we will use throughout this paper. In fact, most authors use sub-adiabatic profiles and condensation rates around 80% of their maximum value are often found in experimental studies (Wood, 2012). A recent study by Merk et al. (2016) finds LWC at about 75 % of its adiabatic value in updrafts and about 60 % of its maximum adiabatic value in downdrafts.

While the ISBLC captures important aspects of actual stratiform boundary layer clouds, none of its assumptions will ever be fully valid for any observed cloud. The true three-dimensional variability of clouds poses significant challenges to any remote sensing algorithm and recent studies have made progress toward understanding and possibly correcting the impact of this variability on cloud remote sensing (Zhang et al., (2012), Zhang et al. (2016) and references therein). Furthermore, the cloud microphysical interpretation of retrieved CDNC is also not straight forward, as ultimately one would be interested in the number of cloud droplets activated at cloud base and not the number of cloud droplets observed. Entrainment mixing processes, precipitation formation, and additional activation above cloud base can lead to differences between these two properties. We will elaborate more on issues related to three-dimensional cloud structure as well as cloud microphysical assumptions in Section 3.

The remainder of this paper is structured as follows. In Section 2 we briefly describe the datasets as well as the principal methods used here to derive CDNC from satellite observations. Section 3 summarizes issues related to the use of the ISBLC as well as other assumptions made in the retrieval process, thereby providing interpretational context for the use of satellite-derived CDNC retrievals as well as guidance on the expected magnitude and relative importance of uncertainties introduced by the various assumptions. In Section 4 we address actual uncertainties and possible biases in CDNC retrievals. Potential biases are for example caused by remaining artefacts in the underlying MODIS retrievals caused by unresolved dependencies on observation geometry or underlying assumptions on the width of the droplet spectrum. In addition, in Section 4 we also validate our CDNC retrievals against in-situ observations of CDNC taken during the VOCALS-Rex campaign and summarized by Painemal and Zuidema (2011). In Section 5 we evaluate the 13-year climatology of MODIS observations. In



our analysis we put particular emphasis on the phase and amplitude of the observed annual cycle of CDNC over various regions of the globe. We further identify areas where trends in CDNC are observed. In Section 6 we provide concluding remarks and discussion of remaining issues that could help improve future satellite-based CDNC estimates.

## 2. Datasets and Methods

### 5 2.1. MODIS Collection 6 cloud parameters

Observational data are from NASA’s MODIS Collection 6 (C6) Level-2 Cloud Product (Platnick et al., 2015; Platnick et al., 2017). The term ‘Level-2’ refers to individual MODIS cloud retrievals at a resolution of 1x1 km at nadir. The Level-2 Cloud Product provides retrievals of cloud optical thickness, cloud top temperature, and three droplet effective radii retrievals using radiances observed at 1.6, 2.1 and 3.7  $\mu\text{m}$ . These cloud parameter retrievals form the basis of our CDNC retrievals. Relative to the earlier MODIS Collection 5, there are several improvements in retrievals of parameters necessary to determine CDNC. These improvements include a better co-registration of the visible and near-infrared focal planes of the Aqua-MODIS instrument as well as significant improvements in the forward radiative transfer models used in the retrieval framework (Platnick et al., 2015). The impact of these changes on CDNC retrievals and gridded climatologies is assessed in detail in Rausch et al. (2016). For the current study, Level-2 cloud retrievals are from Aqua MODIS spanning the years 2003 through 2015. Additionally, for some comparisons with in-situ observations, selected Terra-MODIS granules were used also.

### 2.2. Derivation of CDNC

Under the ISBLC assumption, closed formulas can be derived that relate between two different pairs of cloud physical variables. The first pair of variables are cloud optical depth and effective radius at cloud top, and the second pair of variables are CDNC and cloud geometrical thickness (Brenguier et al., 2000). Following the notation of Bennartz (2007), these relations are:

$$W = \frac{5}{9} \rho_l \tau r_{e,top} = \frac{1}{2} c_w H^2 \quad (1)$$

where  $W$  is the liquid water path,  $\rho_l$  the density of liquid water,  $\tau$  the optical depth,  $r_{e,top}$  the effective radius at the top of the idealized ISBLC,  $c_w$  the condensation rate, and  $H$  the ISBLC’s geometrical thickness. The second relation provides an estimate for CDNC:

$$N = \frac{\tau^3}{k} [2W]^{-5/2} \left[ \frac{3}{5} \pi Q \right]^{-3} \left[ \frac{3}{4\pi\rho_l} \right]^{-2} c_w^{1/2} \quad (2)$$

where  $N$  represents CDNC,  $k$  is a factor related to the dispersion of the assumed cloud droplet size distribution, and  $Q$  is the scattering efficiency of the cloud droplets. The variable  $k$  exhibits some variability (Brenguier et al., 2011; Martin et al., 1994) but is set constant at a value of  $k=0.8$  here. A realistic uncertainty estimate for  $k$  between different studies is about 20%. Similarly,  $Q$  is set to its geometric optics limit value of  $Q=2$ . Bennartz (2007) shows that uncertainties in the representation of  $Q$  and  $k$  are only a minor contributor to the total uncertainty in  $N$  and  $H$ . The condensation rate  $c_w$  is calculated as 80% of its maximum adiabatic value for the MODIS-derived cloud top temperature (CTT). In summary, our particular implementation of the ISBLC for the climatology presented here is  $k=0.8$ ,  $Q=2$ ,  $c_w=f(\text{CTT})$ , and CDNC constant vertically.

The above equations allow for a conversion between  $(\tau, r_{e, \text{top}})$  and  $(N, H)$  thereby enabling the use of NASA's retrieved effective radius and optical depth for calculating CDNC without the need for dedicated retrieval algorithms that would have to be build on Level-1 reflectances. An important assumption made in this conversion between  $(\tau, r_{e, \text{top}})$  and  $(N, H)$  is that the retrieved effective radius is valid at the top of the ISBLC, as indicated by the subscript 'top' in the above equations. However, satellite-derived effective radii are typically valid at some penetration below cloud top that depends on observation wavelength and geometry. Thus, if the satellite-derived effective radius is used directly in the above conversion, one assumes that  $r_{e, \text{top}} = r_{e, \text{retrieved}}$ . Throughout this paper we employ this assumption and use the effective radius retrieved at  $3.7 \mu\text{m}$  to calculate CDNC. The implications and limitations of this assumption are discussed further in in Section 3.

### 2.3 Gridded Climatology

The individual CDNC retrievals discussed above were used to calculate global fields of monthly averages of CDNC on a regular  $1^\circ \times 1^\circ$  latitude-longitude grid for the 13 years 2003 - 2015. First, daily averages were calculated as the arithmetic mean of all Level-2 MODIS-retrieved CDNC values within each  $1^\circ \times 1^\circ$  grid box within one day. In order to provide a valid daily  $1 \times 1$  degree average at least ten valid Level-2 retrievals had to be within that grid-box. Screening criteria performed on individual pixels are outlined further below. If, for a given month and grid-box, more than ten days had valid daily values, the arithmetic mean of those was assigned to be the monthly mean value. The so-derived monthly mean fields, along with uncertainty estimates, are part of the published dataset.

Uncertainty estimates were generated as follows: In addition to the daily mean value of CDNC for each  $1^\circ \times 1^\circ$  grid box, the variance of Level-2 CDNC observations within this grid-box was also calculated. The monthly uncertainty for this grid-box was then calculated as the square-root of the mean of the daily variances over the course of each month, thereby assuming uncertainties between days to be uncorrelated. This approach of creating monthly uncertainties is similar to the approach used in NASA's Level-3 gridded MODIS cloud product (Hubanks et al., 2016). The difference between our approach and that of NASA is that we calculate the underlying daily grid-box-level uncertainties from spatial standard deviations of retrieved CDNC whereas NASA uses a-posteriori retrieval errors as basis for their daily grid-box-level uncertainties. We address the difference between these two choices with respect to CDNC in Section 4.

In order to ensure that only valid cloud observations were accumulated, a series of screening criteria were used on each Level-2 data point. The impact of different screening choices is discussed in Section 4. Here we list the final set of screening choices made in the climatology:

1. Both the infrared-derived and the visible/near-infrared-derived cloud phases had to indicate a liquid water cloud.
2. The retrieved cloud top temperature had to be between 268 and 300 K.
3. The cloud mask had to indicate the observation to be cloudy but not over ice or land.
4. Clear-sky-restoral and pixels identified in the MODIS Level-2 cloud product as partly cloudy pixels were not included.
5. All three effective radii retrieved at 1.6, 2.1, and 3.7  $\mu\text{m}$ , respectively, as well as the three corresponding retrieved optical thicknesses had to be valid.
6. Observations were only considered, if the three MODIS-retrieved effective radii stacked up as  $r_{e,3.7} > r_{e,2.1} > r_{e,1.6}$ , as observations violating this criterion will also violate the key assumption of a vertically increasing LWC in the ISBLC. More details on the motivation of this screening criterion are provided subsequently in Section 3.

### 3. Assumptions and limitations of the ISBLC

NASA’s MODIS effective radius and optical thickness retrievals build on the use of ‘Nakajima-King Diagrams’ (Nakajima and King, 1990) similar to the red mesh shown in Figure 1. The retrieval algorithms effectively map between the observed reflectances, on the x- and y-axis, and the retrieved ( $\tau, r_{e,retreived}$ ) parameters shown in the red mesh. Details on the actual retrievals used in MODIS C6 can be found in Platnick et al. (2015). Note, that NASA’s operational retrievals, as most other retrievals, are performed using a cloud model in which effective radius and liquid water content are constant vertically, thereby replacing assumptions 2 and 3 of the ISBLC while maintaining its assumption 1. This cloud model is sometimes tagged the Vertically Homogeneous Cloud (VHC) model. The ISBLC-based relation between ( $N, H$ ) and the observed reflectances is also depicted in Figure 1 (blue mesh).

Subsequently, we discuss several issues related to the retrieval of cloud optical properties in general and CDNC in particular. All radiative transfer simulations underlying the discussions in this section were performed using the Spherical Harmonics Discrete Ordinate Method (SHDOM) model (Evans, 1998). Spectral optical properties for all aerosols were taken from the Optical Properties of Aerosols and Clouds (OPAC) database (Hess et al., 1998). Simulations were performed for idealized situations over a black surface, nadir observations and a solar zenith angle of 56 degrees. The resulting scattering angle of 124 degrees samples the phase function in a relatively smooth region outside of the backscatter or rainbow peaks. Retrievals on simulated data were performed using Levenberg-Marquardt optimization. Scattering angle is defined as the angle between the sun, the point of observation and the satellite. A value of 180 degrees defines the backscatter direction. Clouds were

assumed to be at 288 K. CDNC was varied within the ISBLC between 10 and 2000  $\text{cm}^{-3}$  and geometrical thickness H was varied between 20 m and 2000 m.

### 3.2. Penetration depth

Figure 2 shows the penetration depth of radiation into ISBLCs as relevant for effective radius retrievals. Firstly, a series of reflectances for ISBLCs with different CDNC and geometrical thickness were calculated using SHDOM as described above. The so-derived reflectances were used as ‘observations’. Secondly, these ‘observations’ were input to retrieval algorithms using the VHC model (red meshes in Figure 1). We keep the quotation marks around ‘observations’ here to indicate that this is a simulation experiment. The simulated retrieval process is analogue to the actual MODIS effective radius retrieval where VHC retrievals are applied to stratiform boundary layer clouds that are typically vertically stratified. The so-retrieved effective radius is then compared to the effective radius profile of the ISBLC that underlie the ‘observations’. The penetration depth is then defined as the height below cloud top, where the retrieved effective radius equals the actual effective radius of the underlying ISBLC. The definition of penetration depth used here cannot be considered directly as the radiative penetration depth at a given wavelength because it uses two-channel effective radius retrievals to determine where vertically the retrieved effective radius is representative. For a full discussion of the radiative penetration depth we refer to Platnick (2000), who also discusses in detail the difference between our definition and the radiative penetration depth as well as angular dependencies.

From Figure 2 one can identify that at 1.6  $\mu\text{m}$  the penetration depth is about two to three times higher than that at 3.7  $\mu\text{m}$ . The results are in good general agreement with the results shown in Platnick (2000). See e.g. Table 4 therein. The resulting retrieved effective radii at the three wavelengths are typically within about 1-2  $\mu\text{m}$  of each other with the 3.7  $\mu\text{m}$  effective radius being the largest. Actual clouds of course deviate often substantially from ISBLCs. Some issues related to cloud inhomogeneities are discussed next.

### 3.3. Cloud inhomogeneities

The liquid water content profile does often not increase linearly with height above cloud base and CDNC as well as effective radius might be reduced near cloud top. In particular inhomogeneous mixing processes reduce CDNC and are observed frequently in boundary layer clouds (Burnet and Brenguier, 2007). Furthermore, horizontal inhomogeneities have a critical impact on cloud optical property retrievals. Such artefacts in cloud retrievals associated with inhomogeneous sub-pixel cloud cover have been discussed in detail for example in Zhang and Platnick (2011) and Horvath and Gentemann (2007) among others. Drizzle processes also significantly alter cloud horizontal and vertical structure, and droplet number concentration (Wood, 2012, 2005). Drizzle might also contribute directly to the observed reflectances thereby increasing the retrieved effective radius at 1.6  $\mu\text{m}$  (Suzuki et al., 2011). Typically, most of these issues affect the effective radius at 1.6  $\mu\text{m}$  and 2.1  $\mu\text{m}$  more strongly than the effective radius at 3.7  $\mu\text{m}$  (Zhang et al., 2012b). Conversely, at high solar zenith angle also the the

effective radius at 3.7  $\mu\text{m}$  might also be biased leading to possible increases in CDNC by 40% to 70% at solar zenith angles higher than about 70 degrees (Grosvenor and Wood, 2014).

The blue line depicted in Figure 1 highlights the issue. Even without accounting for the three-dimensional radiative transfer, partly cloudy scenes will exhibit positively biased effective radii. The blue cross in Figure 1 corresponds to an ISBLC with  $N=500 \text{ cm}^{-3}$  and  $H=175 \text{ m}$ . The blue line gives the resulting observed reflectance, if this cloud covers between 0 and 100 % of the sensor's field of view. For all three wavelengths the blue line cuts through the retrieval grid toward larger effective radii and smaller CDNC. Figure 3 (right panels) shows the impact on retrieved effective radii as one moves from completely cloudy to nearly cloud-free. For sub-pixel fractions of open water above 10% the retrieved effective radius at 1.6  $\mu\text{m}$  starts to exceed 2.1 and 3.7  $\mu\text{m}$  but all three effective radii increase significantly as the pixel becomes less cloud-filled. Zhang et al. (2012b) and Hayes et al. (2010) discuss this effect in more detail, which is also present in case of sub-scale inhomogeneities of fully cloud covered observations (i.e. if cloud thickness and liquid water path varies within the field-of-view). Shading and side-illumination of broken clouds as well as true three-dimensional radiative transfer effects modify this picture somewhat but at the same time the inherent averaging performed over a  $1 \times 1 \text{ km}^2$  MODIS field-of-view averages out some of these higher-order effects (Zhang et al., 2012b). Because of the strong impact of sub-scale inhomogeneity we have limited the climatology to cases where  $r_{e,3.7} > r_{e,2.1} > r_{e,1.6}$ , so that in the example shown in Figure 3 (right panels) the largest part of the retrievals would be (correctly) rejected. We discuss the impact of this criterion on the actual climatology in Section 4.

### 3.4. Aerosol above clouds

Another important topic related to effective radius and CDNC retrievals is the impact of aerosol above clouds. This has received a significant amount of attention in the literature as especially off South Africa biomass burning aerosols, consisting of black carbon (soot), organic compounds, ammonium, nitrate, and sulfate, are often vertically displaced above the stratocumulus clouds by several kilometres (Wilcox et al., 2009; Painemal et al., 2014; Wilcox, 2012; Haywood et al., 2004). We illustrate the effect of aerosol layers on retrievals in Figure 1 (see golden curve with large dot at the end). We start from the black cross in Figure 1, which corresponds to an ISBLC with  $N=500 \text{ cm}^{-3}$  and  $H=175 \text{ m}$ . From there we increase aerosol load above the cloud from zero to an optical depth at 550 nm of one toward the golden dot. The aerosol type we choose is 'continental polluted' from the OPAC database (Hess et al., 1998), which has roughly similar optical properties as the aged biomass burning aerosol reported in Haywood et al. (2003). The corresponding retrievals are shown in Figure 3 (left panels). As aerosol optical depth increases, the retrieved effective radius increases and the retrieved optical depth decreases. This results in a decrease in CDNC with increasing aerosol optical depth. The impact of aerosol above clouds depends highly on its single scattering albedo. The more absorbing the aerosol is, the stronger the effect. Note that, unlike for broken clouds, the retrieved effective radius at 3.7  $\mu\text{m}$  remains larger than the 2.1 and 1.6  $\mu\text{m}$  effective radii. Therefore, the criterion  $r_{e,3.7} > r_{e,2.1} > r_{e,1.6}$  does not help reject potentially aerosol-affected observations. Note further, that the effect of aerosol on

effective radius can also lead to a decrease in effective radius with increasing aerosol optical depth, depending on observation geometry and where exactly in the Nakajima-King diagrams the observation lies.

#### 4. Sensitivity studies

In this section we study the sensitivity of the climatology to choices made in the underlying retrievals as well as data screening choices, such as the aforementioned criterion  $r_{e,3.7} > r_{e,2.1} > r_{e,1.6}$ . We further investigate dependencies of the retrieved CDNC and effective radii on scattering angle and sunglint angle and provide a discussion of uncertainties derived alongside with the CDNC climatology. The sunglint angle is defined as the angle between the satellite, the point of observation and the direct specular reflection of sunlight off the ocean surface. Lastly, we evaluate the climatology against the limited set of in-situ CDNC observations that were readily available to us. Except for the comparisons to in-situ observations all sensitivity studies were done by re-deriving and stratifying one full year (2008) of MODIS data by different quantities, such as the scan angle or position within the scan.

##### 4.1. Impact of observation geometry

Various studies have already shown that scan dependent biases existed in the older Collection 5 version of MODIS. For example, Maddux et al. (2010) find significantly larger effective radius retrievals along with lower optical depth retrievals near the scan edge than in the centre of the scan. Grosvenor and Wood (2014) address the dependency of cloud microphysical retrievals on solar zenith angle. Issues related to the observation geometry can in principle stem from two different sources. Firstly, as the solar or observation zenith angle increases, the potential effects of side-illumination and shading become more pronounced and, for increasing observation zenith angle, also the field-of-view gets larger. Secondly, assumptions and constraints of the retrieval itself might cause artefacts. For example, the choices made on whether to include sun-glint affected areas might cause differences or the discretization of the retrieval to particular combinations of observation geometries can cause artefacts in the retrieval. When creating climatologies based on a large number of observations some of these artefacts might average out, whereas others might cause systematic biases also in the climatologies. Subsequently, we first stratify results by various quantities to identify and discuss potential artefacts. Then, we study the impact of these artefacts on the climatology.

Figure 4 shows the dependency of effective radius, optical depth, and CDNC retrievals on both scattering angle and sunglint angle. Results indicate averages over all valid retrievals for the year 2008 applying the screening criteria corresponding to ‘Stratified’ and ‘Non-Stratified’ (see Table 1 for details on screening criteria).. One can identify a variety of different artefacts in the dataset. In all cases the ‘Stratified’ results (blue) show the artefacts more strongly than the ‘Non-stratified’ cases. We suspect this is caused by the generally larger variability seen in the ‘Non-stratified’ cases caused by the issues discussed in Section 3 (e.g. horizontal inhomogeneity). Observations affected by e.g. inhomogeneity are at least partly

screened out in the ‘Stratified’ dataset, which therefore shows additional artefacts more strongly. From Figure 4 the following issues can be identified:

- Near the rainbow peak (scattering angle around 145 degrees) and the backscatter peak (Scattering angle 180 degrees) the average retrieved effective radii vary in a manner that roughly resembles the phase function of typical droplet spectra. For these two regions the optical depth is comparably flat with much less variation than the effective radius. We speculate these artefacts could be caused either by the treatment of single scattering in the MODIS C6 retrievals or by deviations between the spectral widths of the droplet spectra used in the retrievals and the real world droplet spectra. The “hybrid discretization scheme” used in MODIS C6 retrieval is outlined in Platnick et al. (2015), see Table 2.9-2 therein. While the single scattering contribution is calculated exactly at the angles provided in the LUT, larger interpolation errors appear to occur in the rainbow and backscattering region (Platnick et al. (2015), see Figure 2.9-3, lower right panel), potentially consistent with the features found here. The second possible source of deviations, potential differences between observed and simulated droplet spectral width, could potentially lead to a similar behaviour as it would affect the scattering phase function also most strongly near the rainbow and backscatter peak. We note here that a similar behaviour in retrieved effective radii is also apparent at 1.6 and 2.1  $\mu\text{m}$  (not shown).
- At scattering angles between 70 and about 100 degrees, both effective radius and optical depth show above-average values, which likely are biases in the retrieval. These low scattering angles occur near the edge of the MODIS swath that is oriented toward sun. Since Aqua’s local equator crossing time is around 14:30, the sun is toward the west of the satellite; therefore the western edge of the scan is most strongly affected. Note, there are only relatively few observations at such low scattering angles. This region also loosely corresponds to the region with low sunglint angle, discussed next.
- In terms of sunglint angle (Figure 4, right panels), dependencies can be observed at the low end of sunglint angle and, more strongly, at the high end of sunglint angles. The low end of sunglint angles occurs to the west of the swath, where the solar zenith angle equals the sensor zenith angle. High sunglint angles occur at the eastern scan edge. While some dependence on sunglint angle can be observed for low sunglint angles, the dominant variability on this (western) side of the swath appears to lie in the scattering angle dependency, which is dominated toward the scan edge by variations in observer zenith angle. On the eastern side of the swath a similar picture emerges with an strong increase in retrieved optical depth and, consequently, CDNC toward the (eastern) scan edge. For high sunglint angles the data density however is also relatively low.

To summarize, view angle dependencies are found both toward the eastern and western scan edge. Results here are largely consistent with earlier results reported in (Maddux et al., 2010) and are likely caused by increasing field-of-view size and three-dimensional radiative transfer effects toward the edge of the scan. In addition, dependencies of retrievals on scattering angle are also found in the rainbow and backscattering of the phase function. These are likely caused by assumptions made in the retrieval process. From hereon forward we refer to all of these effects combined as ‘retrieval artefacts’.

## 4.2. Impact Stratification and retrieval artefacts on climatology

Before we discuss the impact of retrieval artefacts on the climatology, we discuss the impact of the stratification on the CDNC climatology.

### 4.2.1. Impact of stratification

5 Figure 5 shows the mean relative difference ‘Stratified’ versus ‘Non-Stratified’ for the year 2008. Only 1x1 degree grid boxes are shown where all 12 months had valid values for both, ‘Stratified’ and ‘Non-Stratified’. In particular in areas where broken clouds are frequently observed, the stratification criterion removes a significant number of observations, sometimes in the order of 90 %. In the stratocumulus areas, a much lower percentage of observations are removed. Resulting differences in climatology are in general positive with the exception of a narrow band in the tropical Pacific. Typically relative differences in mean climatology are smaller than 10 % with a few exceptions off the east coasts of Asia and North America. Figure 5 also shows the (2008) annual cycle for four selected areas. We selected areas where the difference between ‘Stratified’ and ‘Non-Stratified’ are particularly large. For example, in area X12 one can identify a secondary peak in the annual cycle in October that does not exist, if only stratified observations enter the climatology. The area BC1 shows an annual cycle offset by two months between the two climatologies. The area R05 shows in general much lower CDNC and also a decreased annual cycle if the stratification criterion is not applied. In summary, we find that deviations in mean value between ‘Stratified’ and ‘Non-Stratified’ are small but largely systematic globally. Further, the annual cycle of CDNC can be strongly affected, depending on the area observed.

### 4.2.2. Impact of retrieval artefacts

Figure 6 shows the impact of the above discussed retrieval artefacts on the CDNC climatology. The figure compares the ‘stratified’ results used in the final climatology with ‘flagged results’ in which sampling was only performed for observation geometries that did not show large variations in retrieved effective radius and optical depth (see Section 4.1 and Table 1). The additional constraints on observation geometry significantly reduce spatial coverage as can be seen in the upper panel of Figure 6. The relative difference in CDNC between ‘flagged’ and ‘stratified’ is less than 10 %. The annual cycles for the four selected areas also discussed under 4.2.1. are very similar between ‘flagged’ and ‘stratified’. An exception to this is R03 where the ‘flagged’ results show an enhanced annual cycle over the ‘stratified’ results but does not show a shift in annual cycle. Other selected areas (not shown) substantiate these findings, i.e. the relative differences between ‘flagged’ and ‘stratified’ are small, the annual cycle appears to be not affected, but the magnitude of the annual cycle is somewhat muted in the ‘stratified’ cases. Clearly, while some of the retrieval artefacts propagate through into the climatology, their impact is somewhat reduced by averaging out strong the observation geometry dependencies seen in the Level 2 data (Section 4.1.) when the monthly 1x1 degree mean values are aggregated. Recall also that the retrieval artefacts cannot easily be fixed without potentially re-deriving new Level-2 cloud optical properties retrievals. While a restriction of the climatology to



‘flagged’ results would ameliorate some of the impact of the retrieval artefacts, it would at the same time severely limit the climatology spatially. Weighing these different aspects, we opted in favour of increased spatial coverage.

#### 4.3. Assessment versus Vocals-Rex in-situ observations

To assess the robustness of the CDNC retrievals in this study, the results were compared to the in situ CDNC observations of (Painemal and Zuidema, 2011), hereafter referred to as PZ11. Twenty profile-averaged CDNC measurements were collected in 2008 during the VAMOS Ocean-Cloud-Atmosphere-Land Study Regional Experiment (VOCALS-Rex) coincident with Aqua and Terra overpasses. The PZ11 CDNC estimates were derived from the profile average of the onboard cloud droplet probe. Details on the sampling strategy are given in PZ11. Here we compare individual MODIS CDNC retrievals as well as areal averages of MODIS CDNC retrievals to the in-situ-derived CDNC values from PZ11. Nearest neighbour (NN) MODIS observation were found spatially corresponding to each profile’s latitude and longitude as reported in PZ11. Temporally, either Aqua-MODIS or Terra-MODIS was used depending on the observation time reported in PZ11. In addition to the NNs also spatial averages of MODIS observations over 21x21 and 51x51 pixels were used, corresponding to areal extents of 0.2x0.2 degree and 0.5x0.5 degree respectively at nadir. The approach was performed independently for both stratified and non-stratified retrieval selections. For the non-stratified NN cases, the nearest pixel was generally within 2 km of the profile location. When considering stratified nearest neighbours, the distance tends to increase which may decrease coherence, but most stratified observations were within 10 km of the profiled cloud. Results are presented in Figure 7 and Table 2.

Regardless of grid-box size and whether or not the stratification criterion was applied, the comparisons between in-situ and remotely sensed CDNC show high correlations and biases on the order of at maximum 10% of the mean retrieved CDNC values. The non-stratified results show a generally good agreement with PZ11, with almost negligible bias and root mean square errors less than  $25 \text{ cm}^{-3}$ . For the stratified results, magnitude of the RMSE and bias increase, but are generally also in agreement with PZ11. One possible explanation of the larger bias observed in the stratified cases relates to the fact that cases observed by PZ11 are very thin and hence the three effective radii are very close to each other. Random noise on top of the retrievals might in some cases lead to the  $3.7 \mu\text{m}$  effective radius being smaller than the  $2.1 \mu\text{m}$  effective radius. These cases would be excluded, thereby favouring cases where the  $3.7 \mu\text{m}$  is positively biased. This would lead to a negative bias in CDNC as observed caused by the combination of random noise and the selection criterion chosen. While the dataset compiled by PZ11 is extremely helpful, it is not globally representative. More in-situ comparisons, covering a wider variety of clouds and observation geometries are needed to further elucidate such effects on retrievals globally.

Results for the average 51x51 and 21x21 domain cases show results similar to the single pixel cases in terms of biases and RMSE values. However, for both 51x51 and 21x21 neighbourhoods, the mean uncertainty (size of error bars in Figure 7 and last two columns in Table 2) is reduced for the stratified cases over the un-stratified cases. Recall that the mean uncertainty reported for these two neighbourhoods is the standard deviation of all valid CDNC retrievals in the respective averaging box. Effective radius stacking is likely decreasing the selection of pixels of inhomogeneous clouds or those subject to sub-pixel effects, which, as discussed in Section 3, can result in retrievals with a wide spread of unphysical effective radii.

Finally, we note that the uncertainties of the NN approach were calculated using Gaussian error propagation using the reported effective radius and optical thickness errors in the MODIS Collection 6 data following the approach in Bennartz (2007). One can see that the so-derived uncertainties are of about the same magnitude as the uncertainties derived from the standard deviations of the 21x21 and 51x51 boxes. This analysis holds true also for larger box sizes and a wider variety of cases, as shown below.

#### 4.4 Uncertainty estimates

As outlined in Section 2.3, two main pathways exist for the definition of uncertainties associated with the climatology. Firstly, uncertainties in retrieved effective radius and optical depth reported in the MODIS Collection 6 Level 2 product can be propagated forward to yield Level 2 uncertainties in CDNC (see Bennartz, 2007 for details). Secondly, the standard deviation of all Level 2 observations within a given 1x1 degree box can serve as a measure for the product's uncertainty. The latter would include the synoptic variability of CDNC as well as all actual retrieval uncertainties and should in any case be larger than the uncertainty derived from error propagation. Here, we compare these two measures. Recall further from Section 2.3 that the monthly uncertainties for each grid-box were calculated as the square-root of the mean of the daily uncertainties over the course of each month, thereby following the approach used in NASA's Level-3 gridded MODIS cloud product (Hubanks et al., 2016). Figure 8 compares the two different uncertainty estimates in terms of the relative uncertainty averaged over the entire year 2008. Except for some stratocumulus regions the uncertainty estimate based on the standard deviation of the individual Level 2 observations is significantly larger than the uncertainty based on error propagation. This would be consistent with monthly uncertainties driven by synoptic variability. However, for some of the stratocumulus areas, outlined with black isolines in the bottom panel of Figure 8, the uncertainty by standard deviation is smaller (by up to 50%) than the uncertainty by forward propagation of retrieval errors. Hypothetically, even if the actual CDNC in those areas were perfectly constant spatially and temporarily, the uncertainty by standard deviation would be identical to the uncertainty by forward-propagated retrieval error. The only scenario under which this situation could be reversed would be, if estimated uncertainties feeding into the theoretical forward propagation are too large. For example, if the reported uncertainty of the effective radius in the MODIS Level 2 product were too large, this would lead to too large uncertainties in the propagated CDNC uncertainty. Thus, from the bottom panel of Figure 8, it would appear that the forward propagated uncertainties for individual CDNC retrievals are slightly too large. That is, we put slightly less trust into the Level 2 observations than appears to be warranted by this comparison.

For the practical matter of characterizing uncertainty associated with CDNC retrievals for the gridded product, the reported uncertainty estimates should include the impact of day-to-day variability in order to allow for a realistic estimate of the true variability of CDNC. In the final CDNC climatology we therefore report the standard deviation of the retrieved CDNC values within each 1x1 degree grid box as an uncertainty estimate.

## 5. Climatological results

### 5.1. Global overview

Figure 9 and Figure 10 show a global overview of the CDNC climatology. The upper two panels of Figure 9 show mean CDNC and relative uncertainty for the entire time period. The lowermost panel of Figure 9 shows the fraction of months with missing data. One can see that mean CDNC is typically high near the coasts, in particular downwind of the major continents, and lower over the remote oceans. In particular in the tropics certain areas exhibit very low data coverage. Areas in the northern Indian Ocean and the western Pacific exhibit no valid data at all. These areas are typically associated with convective regions where the ISBLC assumption frequently breaks down because either isolated cumuli or deep convection are observed. Near the fringe of these regions also the largest relative uncertainties are observed. Over the mid-latitude storm tracks as well as over the traditional stratocumulus areas west of the major continents, data coverage is higher and various regions show full coverage for all months. Relative uncertainty is in the order of 60% to 80% in the storm tracks and increases polewards. In the stratocumulus regions the relative uncertainty is about 30%. Recall that uncertainties are calculated as the square-root of the mean of the daily 1x1 degree variances over the course of each month, thereby assuming uncertainties between days to be uncorrelated (see Section 2.3). The reported uncertainties are however very similar to the a-posteriori uncertainties derived from error propagation (see Section 4.3 and 4.4).

Figure 10 shows information on the annual cycle of CDNC. The data density in Figure 10 is lower than in Figure 9 because we only included grid boxes where a full annual cycle was available. For some areas, in particular in the tropics, data was only available for certain months and these grid boxes were excluded from the annual cycle analysis. The upper panel of Figure 10 shows the amplitude of the annual cycle both color-coded and as isolines. Amplitude here is defined as the amplitude of a cosine-fit of the annual cycle of CDNC. These same isolines are over-plotted also over the other two panels of Figure 10. The strongest annual cycle is found off the coast of China and off the east coast of the North- and South America. Strong annual cycles are also found in the stratocumulus regions off the west coast of South America and Australia and, to a lesser, extent, off the west coast of Africa. In all of these regions with strong annual cycle a large fraction of the CDNC variability is explained by simple cosine fits. In contrast, certain areas in the North Atlantic and off southern Africa show a more complicated behaviour although the annual variability there is lower (for examples, see Figure 11). The lowermost panel of Figure 10 shows the month of the peak CDNC. The month of peak CDNC varies greatly depending on region. Subsequently we discuss and highlight some of the regions in more detail.

### 5.2. Regional results

We present regional results for a variety of different areas that have been discussed in the literature (see Figure 9 for details). We present results in terms mean CDNC as well as trends in Table 3. In addition we show the time series as well as the average annual cycle for selected areas in Figure 11.

### 5.2.1. Remote Areas

We categorize as remote areas the areas R01, R06, K08, X20, and X21, the last three of which are also shown in Figure 11. The areas are sufficiently remote to be not strongly affected by anthropogenic changes in aerosol conditions to serve as a check for the temporal stability of the dataset. Indeed, for none of these areas trends in CDNC are larger than a  $\pm 3 \text{ cm}^3/10\text{yrs}$

and none of trends are even remotely statistically significantly different from zero. We therefore conclude that the baseline of the climatology appears to be robust and the underlying MODIS observations do not exhibit any serious trends. Such trends could for example have been caused by slight changes in absolute calibration or other sensor-specific issues.

### 5.2.2. Southeast Atlantic (off southern Africa) and south of Madagascar

The Atlantic region off southern Africa has been widely studied in the context of biomass burning and the potential impact of biomass burning aerosols on clouds via the first aerosol effect (Bennartz, 2007; Wilcox et al., 2009; Painemal et al., 2014). Notably, the annual cycle of CDNC peaking in July appears to be a persistent feature in CDNC climatologies (see Figure 10 and also area X12 in Figure 11) that is consistent with southern Africa biomass burning. Peak anthropogenic biomass burning does not coincide with the height of the dry season but does coincide with the middle of the burning season over the northern parts of Angola, the Democratic Republic of Congo, and Zambia, where often Savannah fires are lit in July/August well before the peak of the dry season (Le Page et al., 2010). This picture is substantiated by individual fire counts based on geostationary satellite observations, which also shows peak fire activity around July/August with peak burning of about 6Tg per day in July (Roberts et al., 2009). In contrast, in the eastern part of South Africa and Mozambique as well as northern Namibia, the middle of the fire season is shifted toward September. Depending on large-scale weather patterns biomass burning aerosol can be transported westwards over the southern Atlantic or eastwards over the southern Indian Ocean (see e.g. Sinha et al. (2003)). With the biomass burning season in south-eastern Africa and Madagascar peaking in September, one would expect to find the annual cycle in CDNC also peaking around that time and indeed the climatology does show exactly this (see Figure 10 and also area BC1 in Figure 11)

### 5.2.3. East China Sea

Various authors have studied the East China Sea in one form or another (see Figure 9, R05, BA5, BB1, X15 (shown in Figure 11), X16, X17). It is interesting to see that over the observation period of the current study all of these areas exhibit a negative trend, often statistically significant, in CDNC for the period 2003-2013, which is opposite the positive trend in CDNC reported for the area BB1 and the period 1982 - 2010 (Bennartz et al., 2011). In that earlier publication we have also established a causal link between the Chinese  $\text{SO}_2$  emissions and wintertime CDNC over the East China Sea. We speculate that decreases in Chinese  $\text{SO}_2$  emissions over the last decade or so have partly reversed the effect seen in the original study (Bennartz et al. (2011)). This speculation is substantiated by other studies that report a slight decrease in Chinese  $\text{SO}_2$  emissions from its peak value in the 2004 to 2006 time range, the decrease in  $\text{SO}_2$  emissions largely caused by flue gas

desulfurization technology installed in coal power plants (Lu et al., 2011). Satellite observations suggest an even larger decrease in Chinese SO<sub>2</sub> emissions by 50 % for the recent time period 2012-2015 compared to the year 2005 (Krotkov et al., 2016).

## 6. Conclusions

5 The climatology described in this publication provides a number of incremental improvements over earlier climatologies published by us. Importantly, by making the climatology static and providing a DOI, we hope to be able to contribute to a better **traceability** of results through different studies that might use this climatology. We believe this is a particularly significant issue, as design choices in the generation of the climatology, such as data screening, can have a major influence on scientific results. To this extent we have aimed at clarifying as accurately as possible the screening choices as well as any

10 other decisions that went into the generation of this climatology. For example, we decided to screen out any observation where the three MODIS-derived effective radii are inconsistent with the underlying assumption of the ISBLC. While this additional screening reduces data coverage, it at least partly guards against the misinterpretation as variability in CDNC of three-dimensional radiative transfer effects in broken clouds. We further show that neglecting this screening does not only lead to moderate differences in the annually averaged CDNC but also to sometimes large shifts in phase and amplitude of the

15 annual cycle of CDNC in various regions. **Potentially adverse effects of this screening could occur in situations where the three effective radii are very close to each other, as might be the case for thin stratocumulus clouds. In such cases random noise in the observations might remove valid observations and potentially bias CDNC slightly low as preferably higher effective radii at 3.7  $\mu\text{m}$  would be selected. This hypothesis would be consistent with our finding comparing to the PZ11 cases. However, we believe this issue secondary compared to the large number of situations where the radiation field is**

20 **affected by either broken clouds or precipitation and the three effective radii clearly indicate that key assumptions in the CDNC retrieval are violated. Further retrieval issue might arise at high zenith angles and/or near the ice edge. At high zenith angles, earlier work by Grosvenor and Wood (2014) showed the effective radius at 3.7  $\mu\text{m}$  to be more strongly biased, potentially leading to retrieval issues. Further, cloud mask classification errors near the ice edge might lead to increased noise and our artefacts in those regions.**

25 We found some remaining retrieval artefacts in the MODIS-retrieved effective radius and optical depth that propagate through into artefacts of the CDNC climatology as well. **These artefacts manifest itself in a strong dependency of retrieved effective radius on scattering angle and could potentially be related to the treatment of first order scattering in the MODIS Level 2 retrievals of effective radius and optical depth.** Our assessment of those effects on the climatology shows however that effects clearly visible in the Level-2 data are averaged out to some degree in the aggregated climatology. We therefore

30 did not in the final climatology include any additional screening for those effects. **However, we feel** that it might be beneficial in future work to re-create retrievals working directly on the Level-1 reflectances that address some of the issues we have identified. Such retrievals could be specific to stratiform boundary layer clouds and might include more appropriate

assumptions for example about the width of the droplet spectrum and the stratification of the cloud as well as a finer retrieval grid or a direct calculation of the first one or two orders of scattering that corresponds to the actual observation geometry for each pixel. However, other error sources, for example related to observation geometry or broken clouds, cannot be resolved by directly retrievals of CDNC from Level 1.

Another important issue not addressed here is the consistency between Terra-MODIS and Aqua-MODIS as well as transfer of the methods derived here to geostationary satellites, in particular the recently launched GOES-16 and Himawari satellites that both provide the necessary channel settings to derive three effective radii and in addition allow to capture the diurnal cycle of clouds with very high resolution.

Beyond these natural extensions of the current work, it will be important to devise new strategies to better derive CDNC from satellite using different types of satellite observations. Various approaches have been proposed and partially tested by other authors as outlined in the introduction. Only innovative approaches and new observational strategies will allow overcoming some of the more difficult issues that affect CDNC observations. Examples of such issues include overlying aerosol layers as well as the effects of entrainment on the vertical profile of clouds, where assumptions made in the simple ISBLC could be replaced either by direct observations or by better assumptions that would make CDNC estimates more robust. Another important aspect is the need for more airborne in-situ observations of CDNC, collocated with satellite observations, also in regions outside of the traditional stratocumulus areas. While satellite observations and modelling studies based on high-resolution numerical models can help develop and improve CDNC estimates, only in-situ observations can be regarded as independent validation.

## Acknowledgements

The authors are indebted to Rob Wood and Daniel Grosvenor for identifying and iteratively helping fix an error in the formulation of condensation rate in Bennartz (2007). The authors would further like to thank Veronica Ikeshoji-Orlati for her help with publishing the dataset via the Vanderbilt Library.

## Tables

**Table 1: Terminology used to differentiate various screening tests performed on the Level-2 data.**

Term used	Explanation
Stratified	All six screening criteria listed in Section 2.3 are used. This is the screening used for the final published climatology.
Non-stratified	Only screening criteria 1-5 listed in Section 2.3 are used. Therefore, in contrast to ‘stratified’ the effective radii are not required to increase monotonically between the 1.6 and 3.7 $\mu\text{m}$ retrievals.
Flagged	In addition to screening criteria 1-6 in Section 2.3, the aggregation process excluded observations with a sun glint angle smaller than 35 degrees and scattering angles smaller than 95 degrees or larger than 165 degrees.

5 **Table 2: Statistics of comparison between PZ11-reported airborne and satellite-based derived CDNC estimates, corresponding to the scatter plots shown in Figure 7. The columns marked ‘N’ refer to ‘Non-stratified’ case, whereas the columns marked ‘S’ refer to the ‘Stratified’ case. The reported uncertainty is the mean value of the error bars seen in Figure 7.**

Domain	RMSE [ $\text{cm}^{-3}$ ]		Bias [ $\text{cm}^{-3}$ ]		Correlation		Uncertainty [ $\text{cm}^{-3}$ ]	
	N	S	N	S	N	S	N	S
Nearest Neighbor	22.1	29.7	-2.7	-17.3	0.97	0.97	36.7	35.6
21x21	21.6	31.7	3.8	-16.9	0.97	0.96	28.3	19.8
51x51	23.1	37.4	1.1	-18.7	0.96	0.94	42.1	31.5

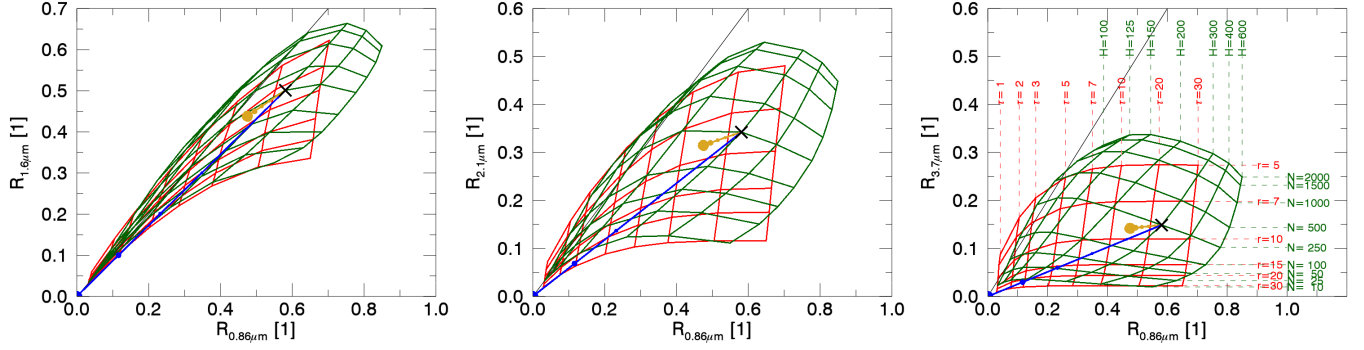


5 Table 3: For all areas shown in Figure 9, this table lists the latitude/longitude boundaries, mean CDNC, standard deviation of CDNC, regression slope of CDNC against time, and the statistical significance of the slope. Rows highlighted green have a significance level higher than 95%. The regression slope was calculated on monthly CDNC anomalies. The monthly CDNC anomalies were calculated by subtracting from each individual month the 13-year mean value of that same month.

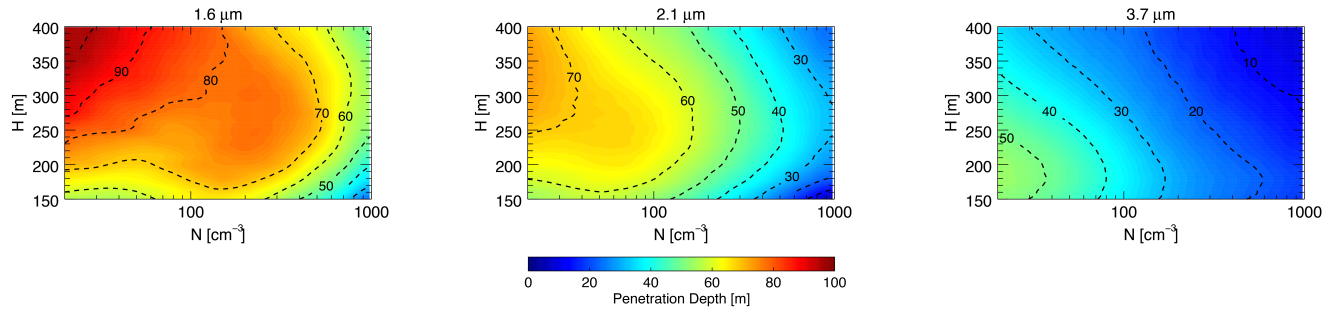
Area	Left Lon	Right Lon	Lower Lat	Upper Lat	Mean	Stddev	Slope	Stat. Sig.
	deg E	deg E	deg N	deg N	cm <sup>-3</sup>	cm <sup>-3</sup>	cm <sup>-3</sup> /10yr	%
X01	-130	-120	30	40	98.3	16.4	-8.1	69.3
X02	-70	-60	35	45	140.5	25.2	-50.5	100.0
X03	-80	-70	20	30	91.5	20.1	-31.7	98.6
X04	-95	-85	20	30	127.8	20.2	-28.7	95.0
X05	-100	-90	5	15	63.6	7.4	2.9	55.8
X06	-90	-80	0	10	72.4	10.1	-0.4	5.3
X07	-90	-80	-10	0	92.0	8.8	-10.9	93.7
X08	-85	-75	-20	-10	97.1	9.9	-9.9	95.5
X09	-45	-35	-30	-20	93.1	15.8	-5.5	56.1
X10	-20	-10	35	45	101.2	21.5	-17.9	89.5
X11	-30	-20	10	20	70.7	7.5	-4.1	76.0
X12	0	10	-10	0	98.9	10.4	-5.3	65.3
X13	60	70	15	25	94.1	12.2	0.9	9.1
X14	82	92	10	20	83.9	13.4	-6.4	39.2
X15	120	130	30	40	240.0	21.3	-19.5	96.7
X16	120	130	20	30	165.6	20.3	-16.6	87.1
X17	110	120	10	20	98.4	19.0	-20.9	92.2
X18	120	130	-10	0	67.1	11.1	-2.5	18.1
X19	90	100	-15	-5	48.0	8.4	-1.4	23.8
X20	0	120	-50	-40	74.2	5.0	-3.1	74.0
X21	-170	-90	-50	-40	64.7	4.7	-1.8	51.3
R01	88	103	-40	-30	53.8	5.0	-3.1	76.7
R02	-140	-115	15	35	85.0	10.1	-8.6	86.8
R03	-10	15	-25	-5	89.5	9.8	-6.8	87.1
R04	-90	-70	-28	-8	90.3	8.1	-6.9	93.9
R05	105	150	10	40	136.9	11.1	-15.6	99.0
R06	-175	-135	-35	-20	45.4	4.2	-1.7	59.3
BA1	-157	-108	20	39	74.6	7.6	-7.4	92.0
BA2	-43	-9	14	43	74.6	8.0	-8.8	95.4
BA3	-101	-62	-37	-8	69.2	5.5	-4.1	89.2

BA4	-20	18	-34	0	76.3	6.8	-4.2	84.0
BA5	109	148	11	30	107.7	12.5	-9.3	82.0
BB6	120	132	18	34	169.6	17.0	-17.1	96.0
BC1	30	50	-35	-25	95.9	11.1	0.6	10.0
K01	-90	-80	-20	-10	68.5	8.4	-4.7	75.2
K02	0	10	-20	-10	98.7	12.3	-7.8	79.7
K03	-130	-120	20	30	91.5	13.1	-7.7	70.9
K04	95	105	-35	-25	52.0	5.7	-0.4	11.0
K05	-90	-80	10	20	78.5	12.1	-9.9	80.0
K06	170	180	40	50	92.0	13.0	-3.6	43.2
K07	-45	-35	50	60	98.3	13.3	-12.4	90.6
K08	-180	180	-59	-50	81.5	4.1	0.9	23.5
K09	105	120	20	30	188.8	25.2	-25.5	90.1

## Figures



**Figure 1:** Nakajima-King plots showing idealized relations between satellite-observed reflectances and retrieved cloud variables over a black surface for all three MODIS bands used for retrievals. The x-axis shows the reflectances at 0.86  $\mu\text{m}$ , the y-axis, the reflectance at 1.6, 2.1, and 3.7  $\mu\text{m}$ , respectively. The red mesh shows the dependency of retrieved optical depth ( $\tau$ , dimensionless, given in the rightmost panel) and retrieved effective radius ( $r$ , in  $\mu\text{m}$ ) on reflectance. The green mesh shows the dependency of retrieved CDNC ( $N$ , in  $\text{cm}^{-3}$ ) and cloud geometrical thickness ( $H$ , in  $\text{m}$ ) on the reflectance. The thin black line is the one-to-one line for reference. The thick blue and golden lines indicate the impact of partly cloudy scenes (blue) and absorbing aerosol above the clouds (golden). More details are provided in the text.



**Figure 2: Average penetration depth relevant for effective radius retrievals of CDNC in the ISBLC. For the purpose of this study, we define the penetration depths as the vertical distance below cloud top at which the retrieved effective radius equals the effective radius predicted by the ISBLC.**

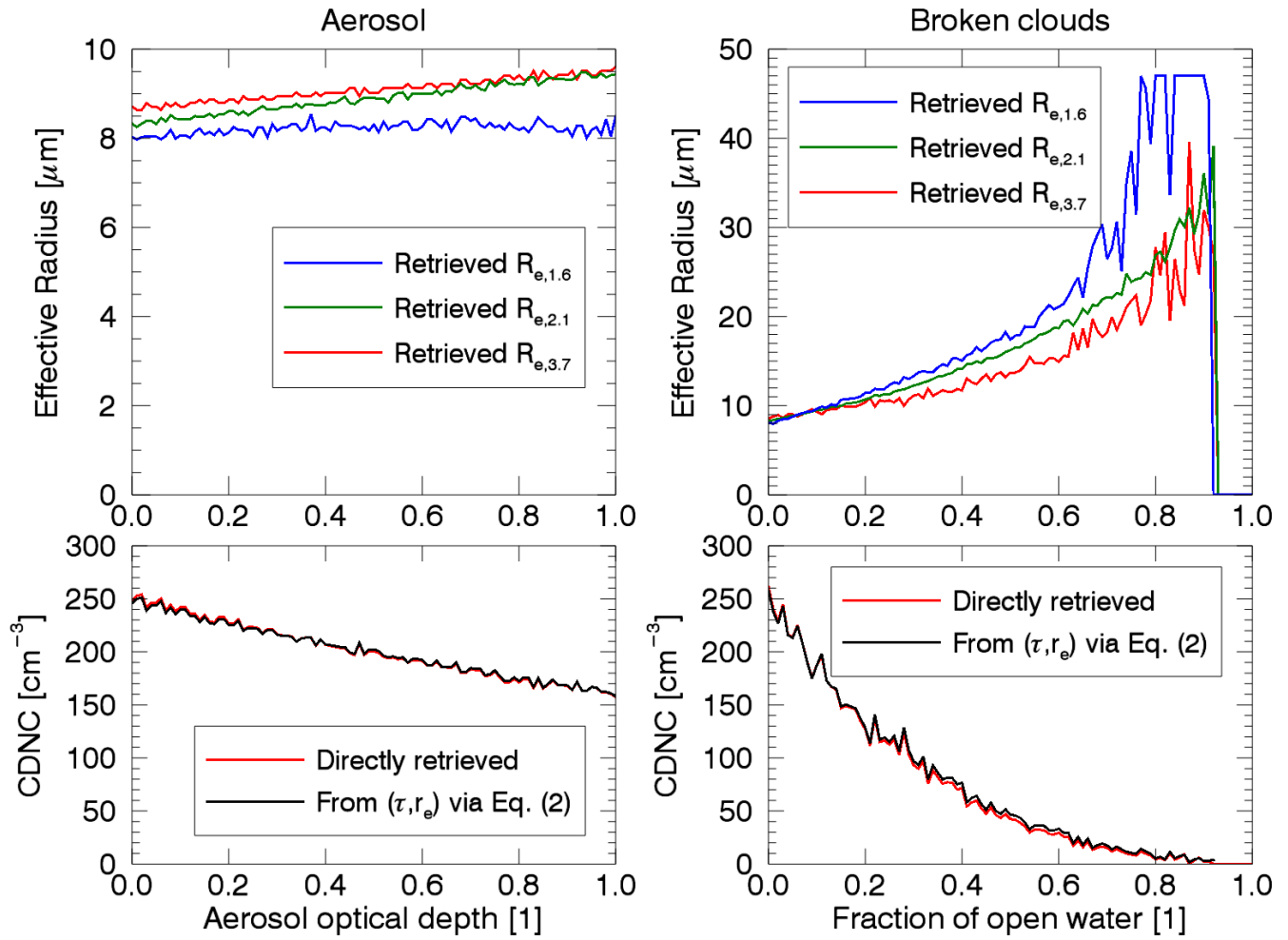
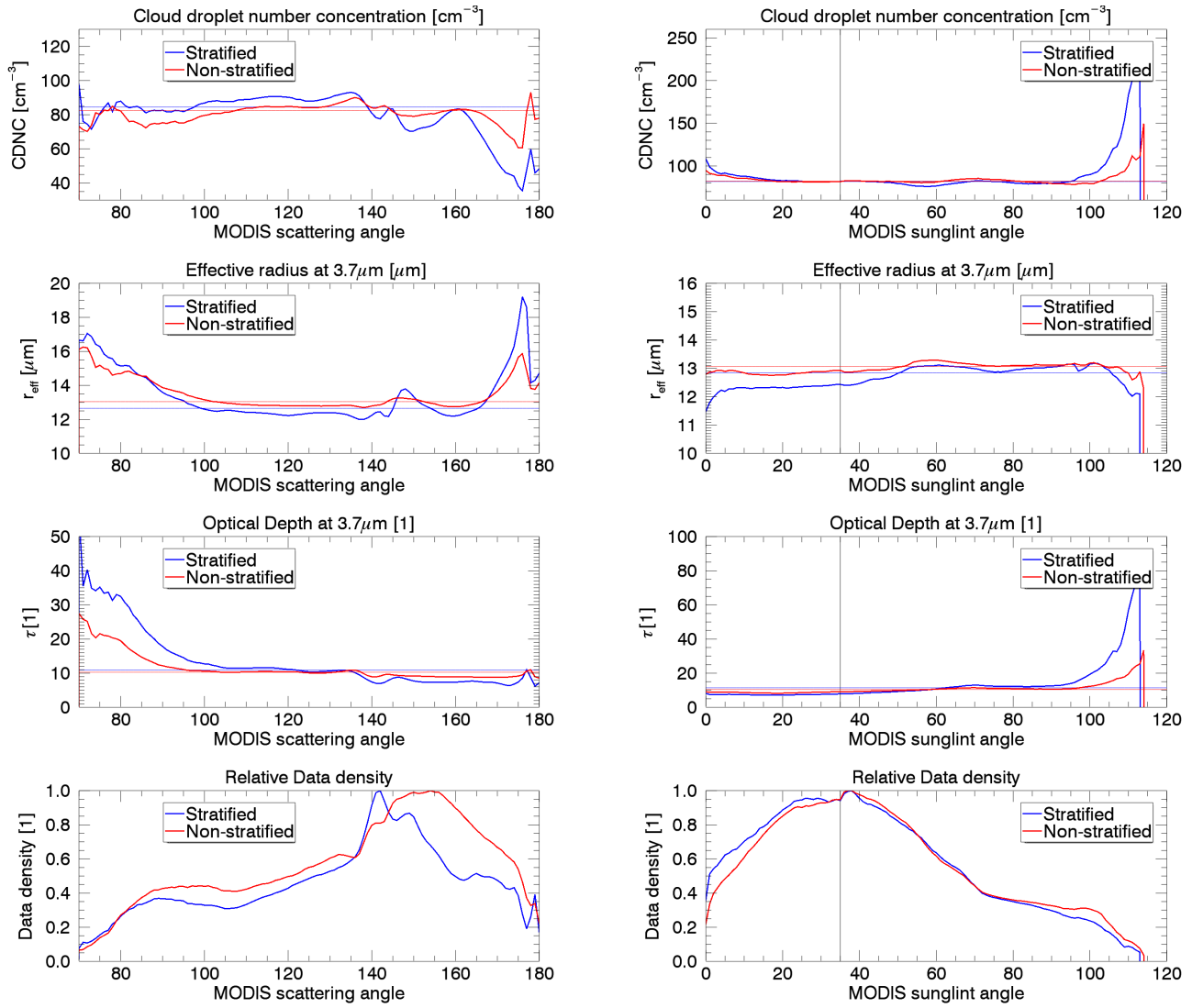


Figure 3: Simulated retrieval for effective radius (upper panels) and CDNC (lower panels) for the aerosol-affected (left) and partly cloudy reflectances (right), respectively, shown in Figure 1. CDNC retrievals tagged ‘directly retrieved’ are performed using the green meshes in Figure 1, rightmost panel, whereas CDNC-retrievals marked as ‘from Eq. (2)’ are derived by first using the 3.7  $\mu\text{m}$  optical depth and effective radius retrieval and subsequently converting it to CDNC using Eq. (2). This latter approach is similar to the actual way we retrieve CDNC from MODIS cloud optical properties. The ‘Fraction of Open Water’ refers to the fraction of a typical inhomogeneous MODIS-pixel that is not covered by cloud.



**Figure 4: Dependency of retrieved CDNC, effective radius and optical depth, all at  $3.7\mu\text{m}$ , on scattering angle (left panels) and sunglint angle (right panels) of Aqua MODIS observations. The curves are showing global annual averages for the year 2008. See Table 1 for terminology regarding ‘Stratified’ and ‘Non-Stratified’. The vertical line in the right panels shows the sunglint angle below which the MODIS cloud masks flags a sunglint contamination. The horizontal red and blue lines are the average retrieved values over the entire dataset.**

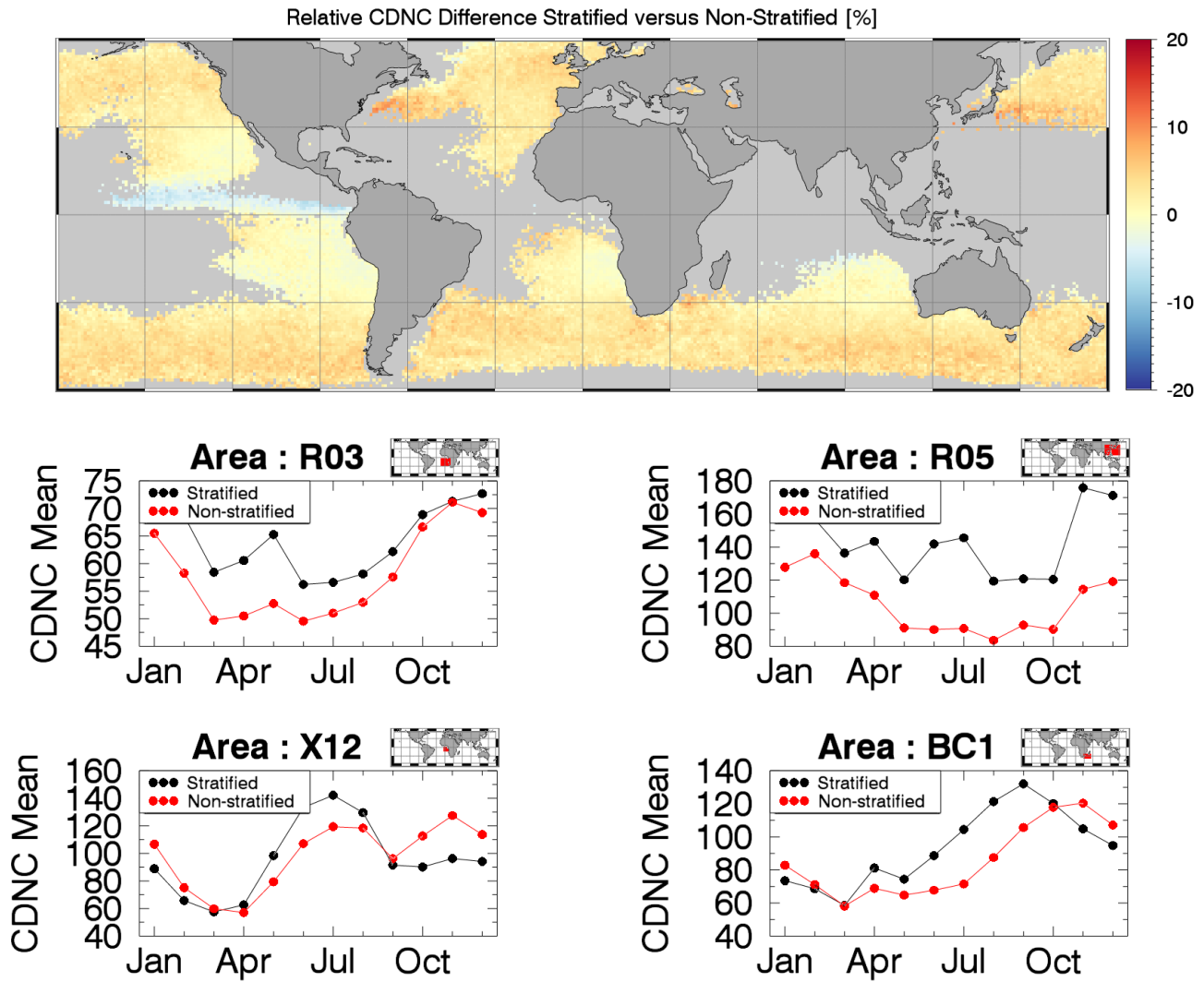


Figure 5: Comparison of results for 2008 for ‘Stratified’ versus ‘Non-stratified’ climatologies; see Table 1 for terminology. The upper panel shows the relative difference in 2008 mean CDNC between stratified and non-stratified. The four lower panels show the annual cycle of CDNC for four selected regions where differences between stratified and non-stratified were particularly strong. For details on the selected regions, see also Table 3.

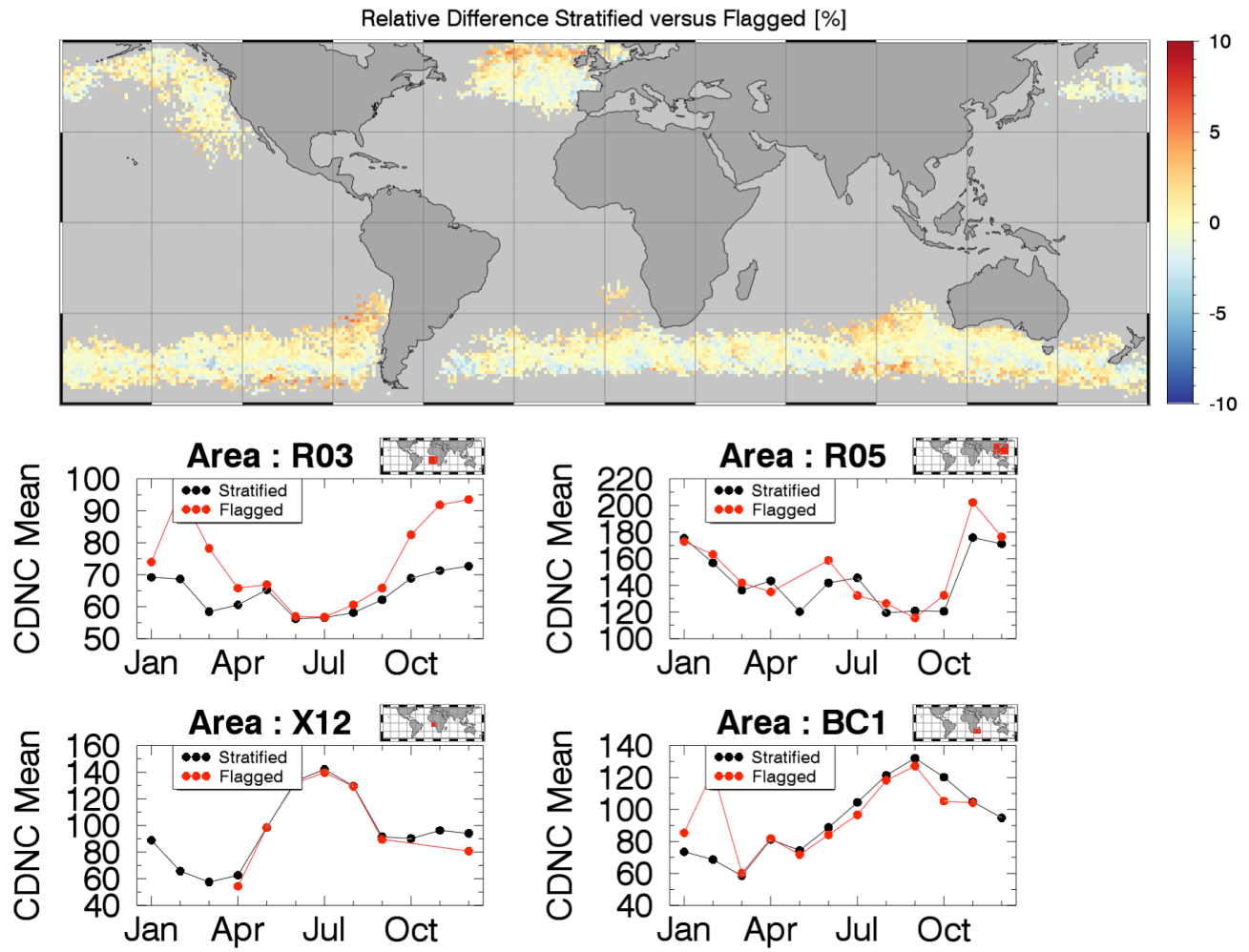


Figure 6: Same as Figure 5 but for ‘Stratified’ versus ‘Flagged’; see Table 1 for terminology. For details on the selected regions, see also Table 3.



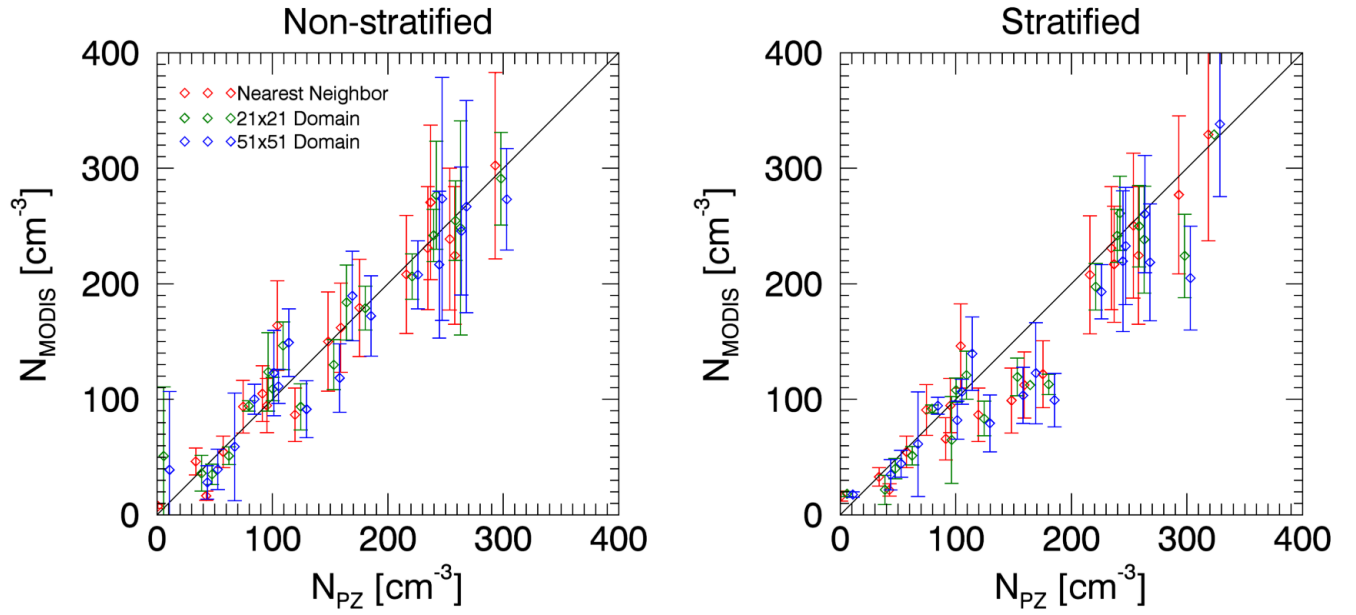


Figure 7: Scatter plot between PZ11-reported airborne and satellite-based derived CDNC estimates, corresponding to the statistics shown in Table 2. Uncertainty for nearest neighbor is estimated using Gaussian error propagation of uncertainties of effective radius and optical depth reported in MODIS C6. Uncertainties for the 21x21 and 51x51 domains are estimates as the standard deviation of all valid CDNC retrievals within the domains. Note, that for each data point the x-axis values of the NN, 21x21, and 51,x51 are slightly offset around the (green) center value, in order to be able to better visually discriminate the error bars. See Table 1 for terminology regarding ‘Stratified’ and ‘Non-Stratified’.

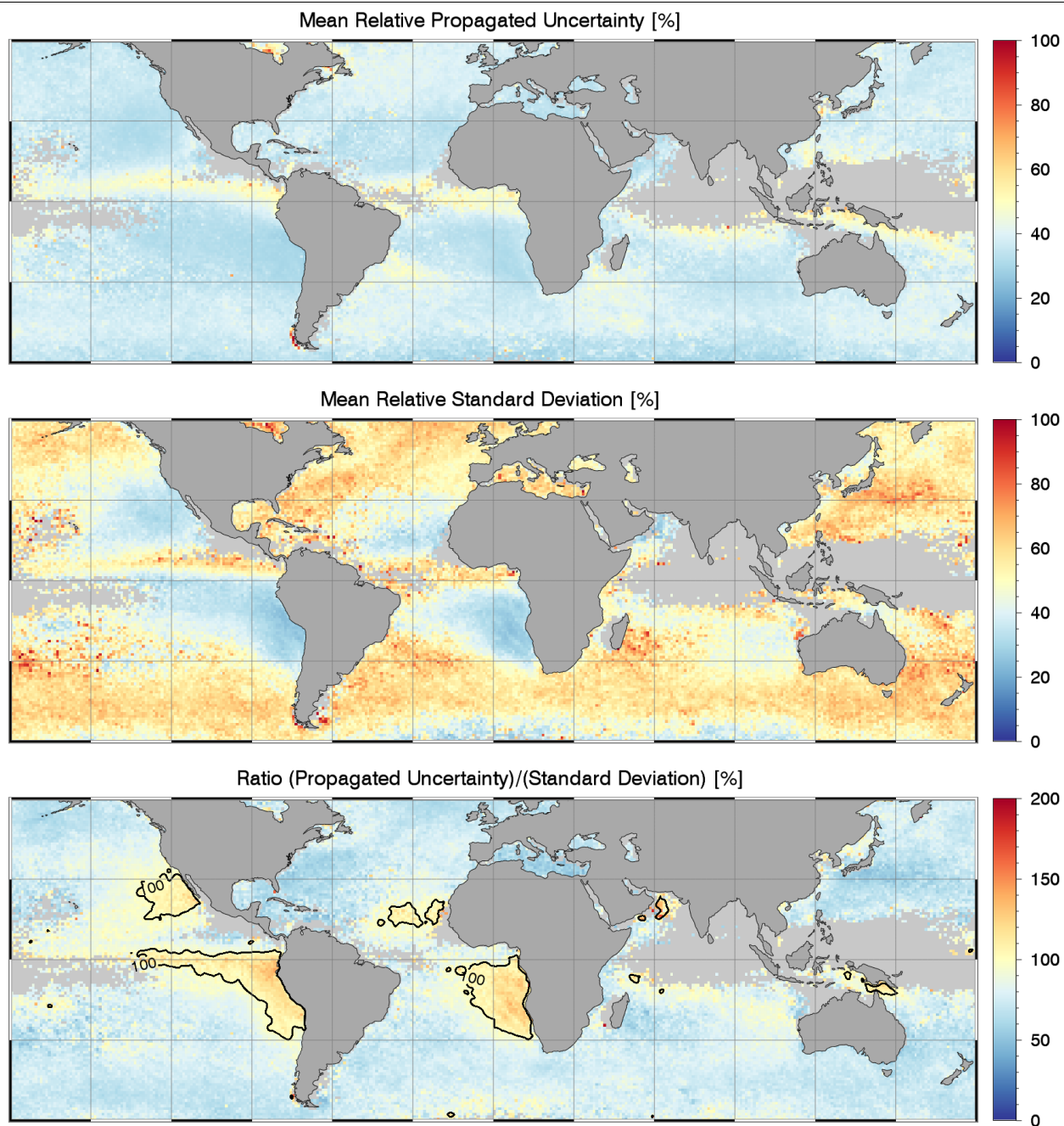
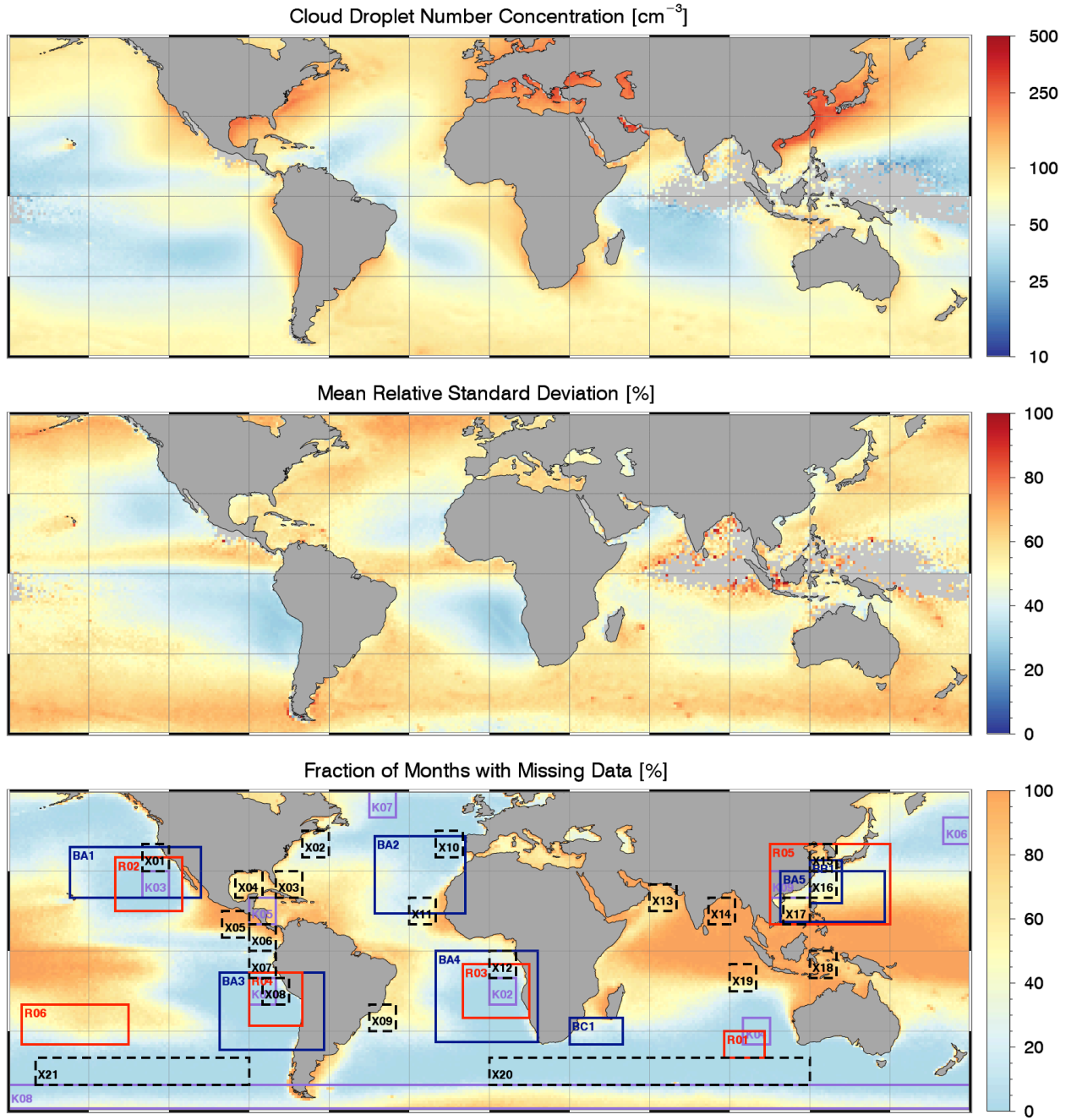


Figure 8:



**Figure 9:** The upper panel shows the mean CDNC over the period 2003-2013. The middle panel shows the mean relative spatial standard deviation of the CDNC estimates and the lower shows the fraction of months with missing data in the climatology. Areas BA1-BA7 are defined in Bennartz (2007), BB1 in Bennartz et al. (2011), BC1 in this publication, K01-K09 in Klein and Hartmann (1993), R01-R06 in Rausch et al. (2010), and X01-X21 in Zhao et al. (2016). The corresponding latitude/longitude boundaries are also listed in Table 3.

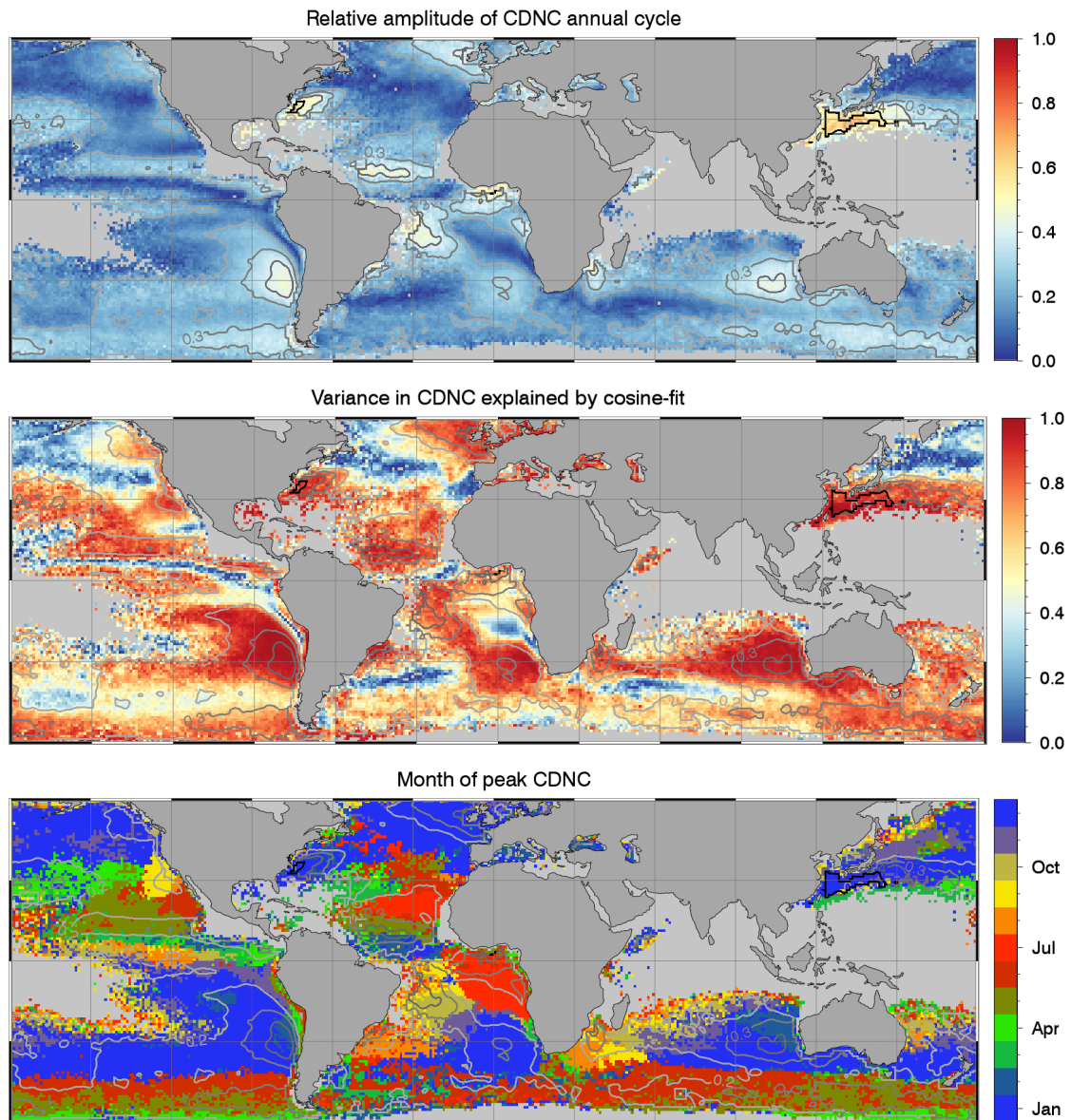


Figure 10: The upper plot shows the relative magnitude of the 13-year mean annual cycle of CDNC. The middle plot shows the variance explained by a simple cosine-fit of the 13-year mean annual cycle. The lower plot shows the months of the occurrence of the maximum CDNC. The isolines in all three plots show the relative magnitude of the CDNC annual cycle (same as upper plot).

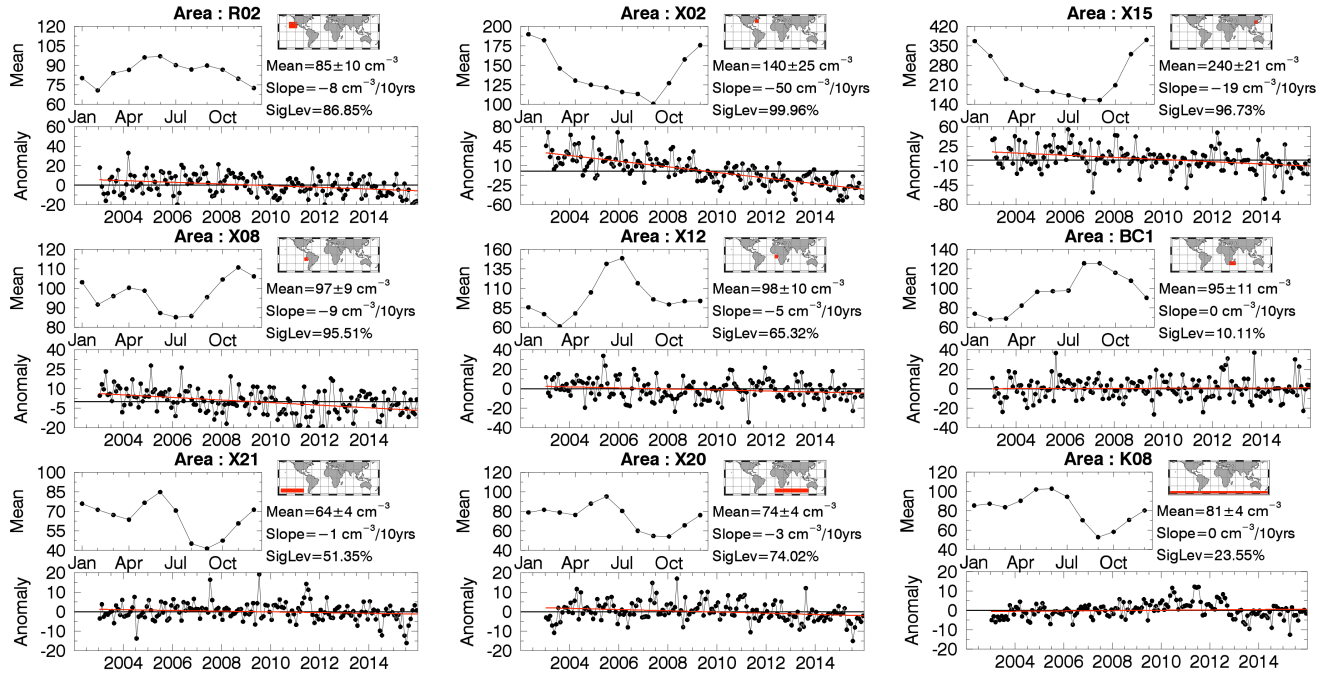


Figure 11: Mean CDNC annual cycle, monthly CDNC anomalies, and statistics for selected areas as shown in Figure 9 and summarized in Table 3. The red curve in the anomaly plots is the regression line.



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