Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks

— RESPONSE TO REFEREE 1 —

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We would like to thank referee 1 for her/his review of the manuscript and her/his constructive criticism. Comments by the referee are colored in blue, our replies are colored in black.

We have thoroughly considered and discussed your input and after careful analysis of each review point concur with you that we have indeed not sufficiently 'articulate[d] what the new thing is that [we] bring to the table', as you state as your 'overarching concern'. The work in this manuscript has a history of several years, over which we have discussed ideas and results with peers and internally many times, so that in writing the manuscript we may have taken several points for granted that are in fact new to a reader confronted with the study for the first time. In this spirit, we have now attempted, guided by your suggestions, to more carefully explain the whats, hows and whys of our research, as well as what is new, and what is not.

I appreciate that the authors have attempted to diversify the ACI investigation field with the use of neural networks. It is often difficult with studies such as this that attempt a new analysis method to create a coherent message. However, I do not think this paper can be published in its current form. My overarching concern in this paper is that the authors do not articulate what the new thing is that they bring to the table besides the black box of a neural network.

General response: See above. Figure 1 included in this document is intended to illustrate the concept of our study schematically: Frequently, aerosol-cloud interactions are studied in a rather isolated manner (in red). At the same time, it is commonly acknowledged that the influence of aerosols is modulated by many environmental factors. With this study, we aim at analyzing the aerosol-cloud-climate system in its entirety. This includes all variations in the environmental conditions, including the seasonal cycle (and its variability) of clouds and meteorology. Our first aim therefore is to find a way to statistically capture this system as completely as possible, including seasonality. Then, in a second step, we focus on and try to separate the effects of aerosols on cloud occurrence and properties from everything else. Our work is not intended to refute previous work done in this field. On the contrary: We would argue that most of the results presented within the study confirm many known aspects of the aerosol-cloud-climate system. But the fact that we were able to find these relationships in a statistical approach considering much more than only aerosol and cloud properties adds an additional line of independent evidence that strengthens the confidence in the existing system understanding. However, this is achieved without isolating specific processes of interest but rather by viewing the system in its entirety. Accordingly, these are the main new things we 'bring to the table': Confidence that the observation data sets considered in a multivariate statistical approach capture the natural variability, and that aerosol effects similar to those found in other studies can be identified in this system. No more, no less.

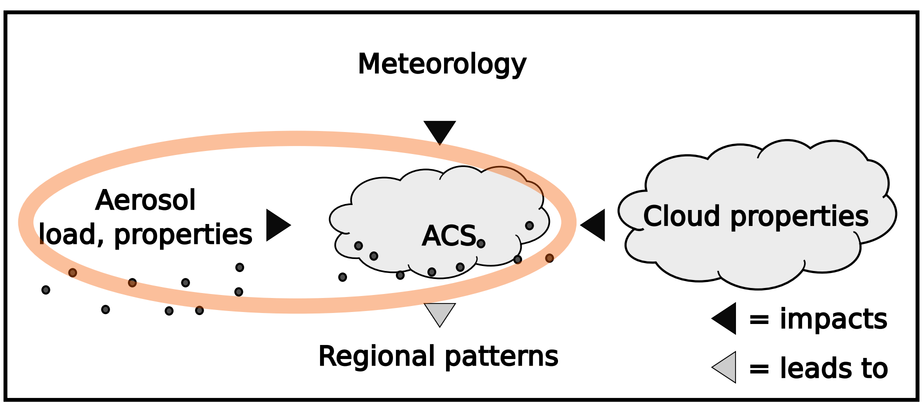

Figure 1: A schematic illustration of the concept of this study (ACS: aerosol-cloud sensitivity).

I have grouped my concerns about this paper into the following categories:

Statistical evaluation

1) It is unclear to me why doing multiple neural networks on sub regions on monthly data tells us anything useful about what is going on. I need some sort of confidence that a high R2 model cannot be created by a large neural network using a collection of meteorological predictors picked at random. Monthly data has the issue of being driven by the seasonal cycle, which will drive almost everything else, and making it regional will mean that the neural network doesn't need to tell us anything particularly meaningful about how the clouds are driven by their environment. The authors should consider using anomalies relative to the seasonal mean, or simply using annual means. Either of these options would be better than the approach taken in this paper. Admittedly the authors talk about this on page 2 line 25, but they don't provide any convincing proof that they haven't just created a regional seasonal cycle simulator.

This is related to what we argue above: We intend to model liquid-water clouds including their seasonal cycle by using information on aerosol loading and a set of meteorological drivers that were identified as main drivers of liquid-water clouds after careful study of current literature. One could probably create a relatively high R^2 model with a very large array of randomly selected predictors due to spurious covariation of seasonal cycles between predictors and predictands. However, in this study, we avoid this by capturing the aerosol-cloud-climate system with a small number of the known main drivers of cloud occurrence and properties. Within this modeled system we then try to understand the effects of each driver and its regional patterns. We argue that regionally specific neural networks are needed to capture the regional variability of liquid-water clouds. Regional patterns exist due to regional differences in cloud type, aerosol composition, meteorology and the respective seasonal cycles.

2) On page 4/line 10 the authors note that they throw out models that have a low R2. I'm not sure why this is ok to do.

We have identified R^2 and the root mean square error relative to the mean as good indicators for model skill. We are interested in understanding predictor-predictand relationships by analyzing their respective sensitivities, however, we choose to trust only models that can adequately represent the observed cloud patterns. We prefer to err on the side of caution to avoid reaching conclusions based on inadequate statistical relationships; thus we exclude models that in our opinion are not capable of representing the system well enough. We are open to other ideas regarding alternative ways to ensure adequate model skill.

3) On page 6 I find this something of a straw man. A better test would be to compare multiple linear regression of all the predictors to the ANN, as opposed to a regression on AOD alone. Or to compare the ANN trained using only AOD. I think that the paper would actually be vastly improved by just repeating the analysis with a multiple linear regression to demonstrate to skeptical readers why their paper brings anything new to the table as compared to the numerous previous papers that have looked at ACI and low cloud variability in the past.

We probably did not communicate the intention of this figure with sufficient clarity: This figure is intended to show how well a combination of aerosol and meteorological conditions can explain the variance of cloud properties (multi-variate statistics) as opposed to a simple bivariate approach. We have added results of a multiple linear regression using all the ANN predictors to the figure (2). The comparison of the results of the multiple linear regression and the ANN

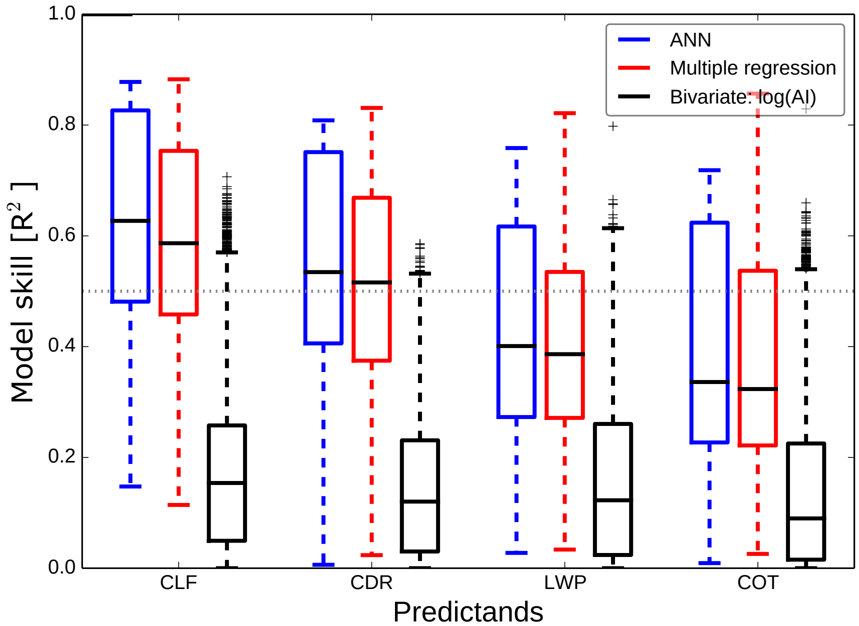

Figure 2: Predictand correlation with ANN (multivariate) test output, multiple linear regression (multivariate) and log(AI) (bivariate). The median is represented by the black horizontal line, framed by the interquartile range (boxes), whiskers expand the boxes by 1.5 interquartile ranges.

suggest that the ANN is an appropriate method to be used in this context.

Neural networks were our statistical method of choice, as they have the advantage of not being reliant on statistical assumptions on predictor and predictand distributions and they are capable of modeling nonlinear relationships. That being said, we agree that other multivariate methods (e.g. multiple linear regression) could also have been used.

4) Figure 5- If the error bars give the range in sensitivity does that mean that nothing except LTS and AOD have a robust relationship with cloud properties that holds outside of a few regions? Didn't we already know this very well from

simple regression models that were easy to interpret (Klein & Hartmann, 1993; Nakajima, Higurashi, Kawamoto, & Penner, 2001)?

We agree with the referee that many of the results of this study confirm what previous studies have already shown. Since we have reached these conclusions using a different methodology, we add another line of evidence. The lack of other relevant relationships would not have been obvious without such an analysis. In our opinion the value of our study is that the results were produced by looking at the entire system at once rather than at isolated relationships. Using this method, we can compare the relevance of each predictor to each predictand including spatial patterns.

5) Choice of predictor/predictands: The choice of predictors by the authors is not appropriate for a paper in the last decade. Why have the authors chosen AOD to be a CCN proxy? AOD is not equivalent to CCN since it has a large contribution from larger, non-CCN relevant aerosols. Why don't the authors use AI, which is far more relevant and typical of more recent studies (Patel, Quaas, & Kumar, 2017)? The authors acknowledge this, but then shrug this off because papers from almost a decade ago do it. In a similar vein, why do the authors use effective radius instead of CDNC? Effective radius for a fixed CCN increases with increasing LWC, making it sensitive to meteorological drivers. The authors do acknowledge this in page 10, section 25 noting that the interaction between inversion strength and effective radius is most likely driven by variations in LWC. This makes the interpretation of the CDR as a proxy for aerosol-cloud effects muddied. Further, the authors use LTS. Why not use EIS, which is used by every study investigating low cloud in the last decade (Myers & Norris, 2015; Qu, Hall, Klein, & Caldwell, 2014; Seethala, Norris, & Myers, 2015; Webb, Lambert, & Gregory, 2013)? Finally, I am concerned with the use

of RH. Clouds and RH are a semi-equivalent quantity, which may just mean that they are comparing ECMWF-interim's cloud cover to MODIS, further aliasing in the seasonal cycle to their prediction model.

AOD vs. AI: For this study, we used the newest version of MODIS products available, collection 6 (C6). In C6, the MODIS Ångström exponent (needed for the computation of the aerosol index as it is the product of AOD and the Ångström exponent) has been discontinued in level 3 (L3) data (p. 3018 Levy et al., 2013). We believe that for this and for other reasons, other recent studies also use the AOD as a proxy for CCN (see: Gryspeerdt and Stier, 2012; Tang et al., 2014; Chakraborty et al., 2016; Stathopoulos et al., 2017; Patel et al., 2017). We agree with referee 1 though, that the aerosol index is an appropriate measure for CCN and have chosen to use it in the ANN. The following figures 3 and 4 are the new results of the ANN when using AI instead of AOD. The spatial patterns in the ANN skill, as well as the mean global sensitivities are nearly identical (compare with figures 3 and 5 in the original ACPD manuscript).

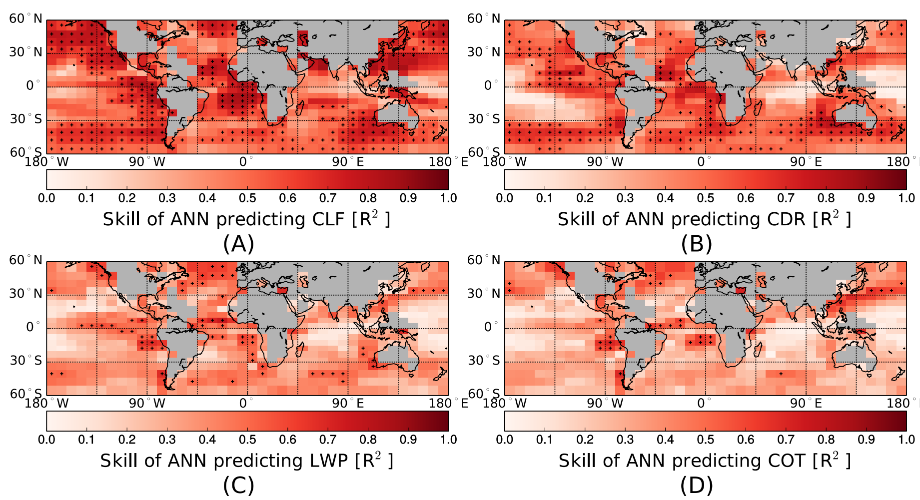

Figure 3: Global patterns of ANN skill as in the manuscript; AI has been used instead of AOD.

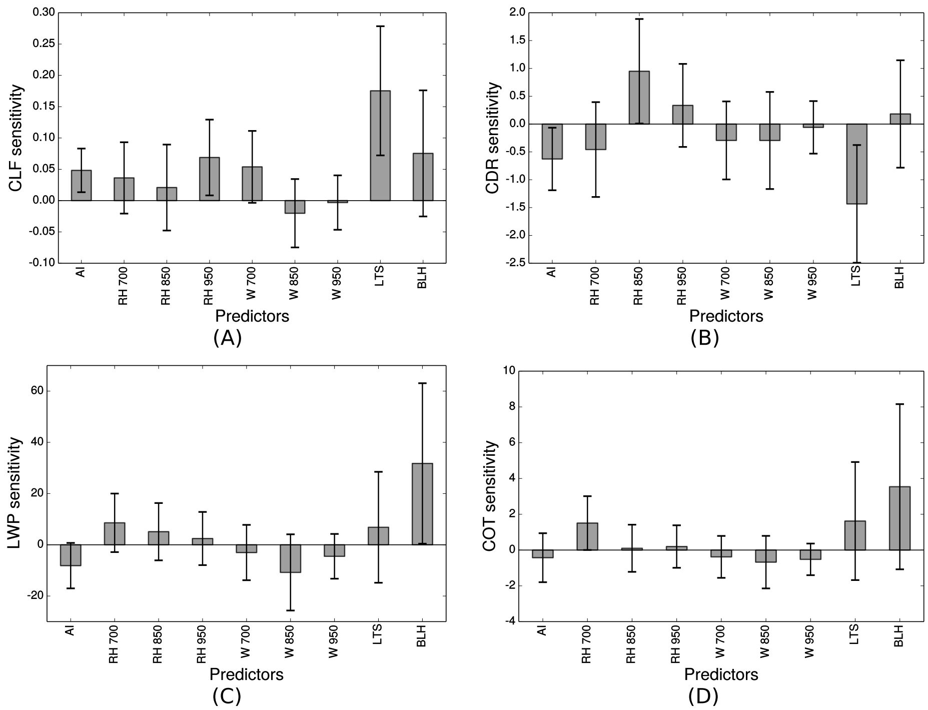

Figure 4: Global mean relative sensitivities as in the manuscript; AI has been used instead of AOD.

Small differences can be observed in the regional patterns of ANN sensitivities (fig. 5 on the following page). The CLF sensitivity to AI is higher in the Southeast Atlantic than its sensitivity to AOD in that specific region. The Southeast Atlantic is of course dominated by biomass burning aerosol, which are mostly in the fine mode and thus feature a relatively larger AI than AOD. The sensitivity of CDR to AI differs from its sensitivity to AOD in regions that are dominated by desert dust. Dust is relatively coarse, so that the AI would be underproportional to the AOD in these regions which might explain the differences between the sensitivities of the two.

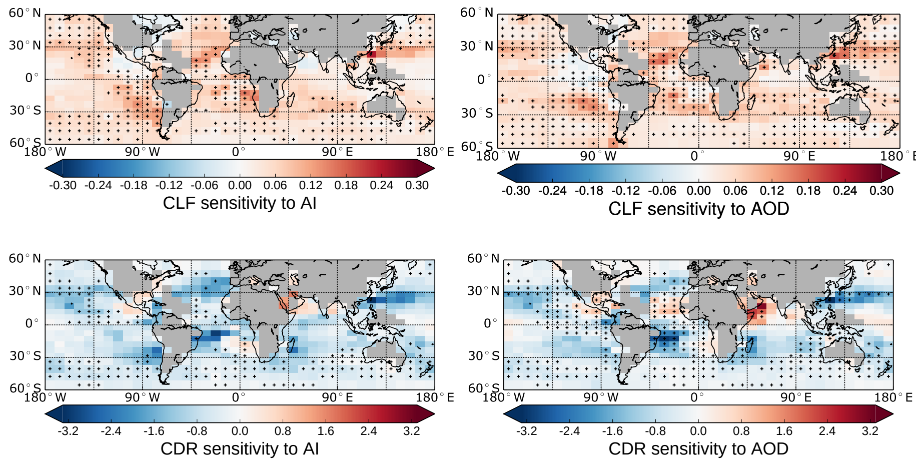

Figure 5: Difference in sensitivities of CLF and CDR to AI (left-hand column) vs. AOD (right-hand column).

CDR vs. CDNC: We agree with the referee that CDNC is a better quantity for the direct analysis of the first aerosol indirect effect, however, its retrieval from satellite is quite problematic, as the retrieval of CDNC requires additional assumptions on the cloud water profile. The commonly-applied adiabatic assumption might be a good proxy for many regions and cloud types (i.e. stratocumulus clouds), however, we are investigating all liquid-water clouds on a global scale. Bennartz and Rausch (2017) showed that the uncertainties in the CDNC retrievals are significantly increased in non-stratocumulus regions. As we are investigating global patterns for various liquid-water cloud types, we came to the conclusion that the uncertainty related to the CDNC retrievals outweighs the theoretical advantages of using CDNC rather than CDR.

LTS vs. EIS: We do not see a specific advantage of using EIS over LTS, as e.g. Lacagnina and Selten (2013) found that for the Californian stratus, LTS is a better predictor than EIS. Some other recent studies that use LTS are e.g. George and Wood (2010); Chen et al. (2014); Gryspeerdt et al. (2014, 2016); Painemal et al. (2014a,b); Adebiyi et al. (2015); Adebiyi and Zuidema (2016); Coopman et al. (2016); Eastman et al. (2016); Ghan et al. (2016). That being said, we would agree that EIS is an appropriate alternative measure for large-scale thermodynamics.

RH: As pointed out above, our intention is to capture the entire aerosol-cloud-climate system and in our opinion, relative humidity has a key role within this system. Thus, the inclusion of RH in the model was a necessity.

Writing:

The writing is rushed and hard to follow. Clearly expressing why the methodology is valid is crucial for this study and as such the writing needs to be tightened up substantially to clarify their ideas.

See our comment at the beginning of this letter. We will attempt to describe the reason for the methodology, the hypotheses and the relevance of our work more clearly in the revised manuscript.

Summary:

The authors articulate their guiding hypotheses, which I think is a good thing to do. I am not sure why (1) is a hypothesis. It seems to be more of a statement about neural networks and is worrisome since I am still concerned that the neural network is just looking at the seasonal cycle and is guaranteed to get a high R2. (2) is odd. Why would we have regional patterns? I could see it if this was a regime-dependent analysis (eg stratus vs convection), but the use of w and LTS as predictors in the neural network should mean that the authors can create a single neural network that effectively does this for them. Why is this not the case? What makes a specific lat-lon box a natural choice.

(3) seems to imply that meteorology plays a secondary role to aerosols, which is not true. We don't expect aerosol to tell us where convection and stratus are, for instance.

1) Neural networks have not been used in this context before, so their capabilities in this context were not quite clear. This is also the case for the separation of aerosol and meteorological effects.

2) While this study does not contrast e.g. stratus vs. convection, we analyze all liquid-water clouds globally. It is clear that these feature different cloud types in different regions and that different processes drive these different clouds. This is shown in figure 6. Regional patterns in aerosol-cloud sensitivity exist. They have been shown to be dependent on meteorology and aerosol species composition (e.g. Andersen et al., 2016). If we created a single neural network, all of the regional characteristics and regionally specific sensitivities (c.f. figure 6) would be blurred or missed completely.

3) Our third hypothesis is certainly not intended to imply that meteorology plays a secondary role to aerosols. We will change the wording for clarity in the revised manuscript.

References

Adebiyi, A. A. and Zuidema, P. (2016). The role of the southern African easterly jet in modifying the southeast Atlantic aerosol and cloud environments. *Quarterly Journal of the Royal Meteorological Society*, 142(697):1574–1589.

Adebiyi, A. A., Zuidema, P., and Abel, S. J. (2015). The Convolution of Dynamics and Moisture with the Presence of Shortwave Absorbing Aerosols over the Southeast Atlantic. *Journal of Climate*, 28(5):1997–2024.

Andersen, H., Cermak, J., Fuchs, J., and Schwarz, K. (2016). Global observations of cloud-sensitive aerosol loadings in low-level marine clouds. *Journal of Geophysical Research: Atmospheres*, 121(21):12936–12946.

Bennartz, R. and Rausch, J. (2017). Global and regional estimates of warm cloud droplet number concentration based on 13 years of aqua-modis observations. *Atmospheric Chemistry and Physics Discussions*, 2017:1–32.

Chakraborty, S., Fu, R., Massie, S. T., and Stephens, G. (2016). Relative influence of meteorological conditions and aerosols on the lifetime of mesoscale convective systems. *Proceedings of the National Academy of Sciences of the United States of America*, 113(27):7426–7431.

Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H. (2014). Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds. *Nature Geoscience*, 7(9):643–646.

Coopman, Q., Garrett, T. J., Riedi, J., Eckhardt, S., and Stohl, A. (2016). Effects of long-range aerosol transport on the microphysical properties of low-level liquid clouds in the Arctic. *Atmospheric Chemistry and Physics*, 16(7):4661–4674.

Eastman, R., Wood, R., Eastman, R., and Wood, R. (2016). Factors Controlling Low-Cloud Evolution over the Eastern Subtropical Oceans: A Lagrangian Perspective Using the A-Train Satellites. *Journal of the Atmospheric Sciences*, 73(1):331–351.

George, R. C. and Wood, R. (2010). Subseasonal variability of low cloud radiative properties over the southeast Pacific Ocean. *Atmospheric Chemistry and Physics*, 10(8):4047–4063.

Ghan, S., Wang, M., Zhang, S., Ferrachat, S., Gettelman, A., Griesfeller, J., Kipling, Z., Lohmann, U., Morrison, H., Neubauer, D., Partridge, D. G., Stier, P., Takemura, T., Wang, H., and Zhang, K. (2016). Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability. *Proceedings of the National Academy of Sciences of the United States of America*, 113(21):5804–5811.

Gryspeerdt, E., Quaas, J., and Bellouin, N. (2016). Constraining the aerosol influence on cloud fraction. *Journal of Geophysical Research: Atmospheres*, 121:3566–3583.

Gryspeerdt, E. and Stier, P. (2012). Regime-based analysis of aerosol-cloud interactions. *Geophysical Research Letters*, 39(21):L21802.

Gryspeerdt, E., Stier, P., and Partridge, D. G. (2014). Satellite observations of cloud regime development: the role of aerosol processes. *Atmospheric Chemistry and Physics*, 14(3):1141–1158.

Lacagnina, C. and Selten, F. (2013). A novel diagnostic technique to investigate cloud-controlling factors. *Journal of Geophysical Research: Atmospheres*, 118(12):5979–5991.

Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C. (2013). The Collection 6 MODIS aerosol products over land and ocean. *Atmospheric Measurement Techniques*, 6(11):2989–3034.

Painemal, D., Kato, S., and Minnis, P. (2014a). Boundary layer regulation in the southeast Atlantic cloud microphysics during the biomass burning season as seen by the A-train satellite constellation. *Journal of Geophysical Research: Atmospheres*, 119(19):11288–11302.

Painemal, D., Road, N., and Stop, M. (2014b). Mean Structure and diurnal cycle of Southeast Atlantic boundary layer clouds: Insights from satellite observations and multiscale modeling framework simulations. *journal of climate*, pages 1–51.

Patel, P. N., Quaas, J., and Kumar, R. (2017). A new statistical approach to improve the satellite based estimation of the radiative forcing by aerosol–cloud interactions. *Atmospheric Chemistry and Physics Discussions*, 17:3687–3698.

Stathopoulos, S., Georgoulias, A., and Kourtidis, K. (2017). Space-borne observations of aerosol - cloud relations for cloud systems of different heights. *Atmospheric Research*, 183:191–201.

Tang, J., Wang, P., Mickley, L. J., Xia, X., Liao, H., Yue, X., Sun, L., and Xia, J. (2014). Positive relationship between liquid cloud droplet effective radius and aerosol optical depth over Eastern China from satellite data. *Atmospheric Environment*, 84:244–253.

Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks

— RESPONSE TO REFEREE 2 —

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We would like to thank referee 2 for her/his review of the manuscript and her/his constructive criticism. Comments by the referee are colored in blue, our replies are colored in black.

This paper addresses a topic of significant current research, namely quantifying the effect of aerosols on cloud properties. The authors note the importance of local meteorology in determining the properties of clouds and that as meteorological factors are also correlated to aerosol properties, this can obscure the influence of aerosols on cloud properties. To explore the role of meteorology and aerosols, they make use of an artificial neural network (ANN) to examine the sensitivity of cloud properties to different predictors. Similar to previous studies, they show that meteorology is a strong control on the cloud properties, such that the cloud properties can be accurately predicted on a monthly timescale using reanalysis data and observed aerosol properties.

I think that this paper is a good addition to the literature on this topic, presenting a new way to investigate the drivers of cloud properties. However, there are a couple of points, listed below, that I think should be clarified before publication. I particular, I think that using monthly data rather than daily/instantaneous data must be better justified. It would also make the paper stronger if the ANN method was compared to a more comparable statistical technique, such as a multiple linear regression across meteorological parameters. This might help to highlight the benefits of using an ANN, especially if it results in a different sensitivity of cloud properties to aerosol. Following these changes, I feel that this article would be suitable for publication in Atmospheric Chemistry and Physics.

We respond to each point individually below.

Main points

1) While some previous studies have used monthly data for investigations into aerosol-cloud interactions, this disguises a lot of the variability in the cloud field and focuses on very large scale changes in cloud properties. The effect of seasonal variations can generate non-causal relationships between cloud properties and meteorological factors that might be accounted for if the study was done on a sub-seasonal scale using higher temporal resolution data. Can the authors explain why monthly data is used in this case and why daily data is unsuitable?

With this study, we specifically aim at analyzing the aerosol-cloud-climate system at a very large scale ('system scale'). The monthly time scale is used here, as a) this enables a focus on the large-scale patterns and relationships and b) GCM output is also at a monthly time scale, so that future comparisons between our observationally-based results and GCMs can be conducted. We acknowledge the 'non-causal relationship' argument by referee 2 by using only a very limited number predictors in ANNs that have previously been shown to be the main drivers of liquid-water clouds. The results of the ANNs are physically plausible (signs, magnitudes and regional patterns of the sensitivities) and give another line of independent evidence that strengthens the confidence in our current system understanding. That being said, we cannot exclude the possibility that some of the observed relationships might be in part non-causal (which is true for other averaging time scales as well).

2) The use of an ANN seems to give a large improvement over just using AOD as a predictive variable for cloud properties. However, I am not sure this is a suitable comparison, as AOD is rarely assumed to be a good predictive variable for cloud properties on its own. As better comparison would be the predictive

ability of (log) AOD on its own using a linear regression and from the ANN. Alternatively a comparison of a multiple linear regression and an ANN for predicting the cloud properties could show the added utility of using an ANN over existing methods. This might then highlight further useful properties of the ANN - for example, does it show a stronger (or weaker) sensitivity of cloud properties to aerosols when compared to current methods?

We probably did not communicate the intention of this figure with sufficient clarity: This figure was simply intended to show how well a combination of aerosol and meteorological conditions can explain the variance of cloud properties (multi-variate statistics) as opposed to a simple bivariate approach. We have added results of a multiple linear regression using all the ANN predictors to the figure as suggested to illustrate the skill of the ANN vs. another multivariate method. The comparison of the results of the multiple linear regression and the ANN suggests that the ANN is an appropriate method to be used in this context. As suggested, we have switched from using the AOD to the AI and used log(AI) for this figure.

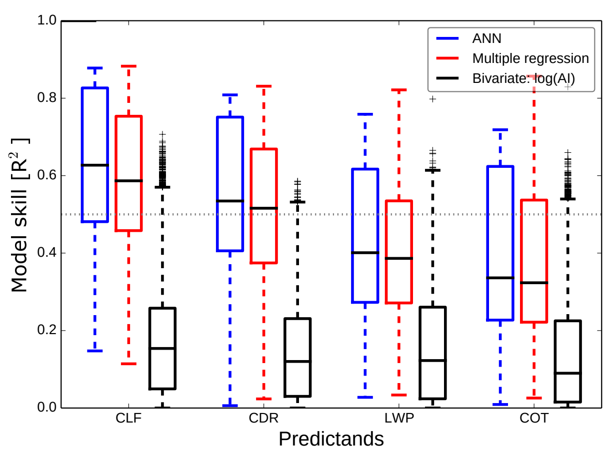

Figure 1: Predictand correlation with ANN (multivariate) test output, multiple linear regression (multivariate) and log(AI) (bivariate).

3) How do regional ANNs compare to a single global model? Presumably if enough meteorological parameters can be included, a single global model should be able to predict cloud properties everywhere. Requiring different models in different locations would then indicate that some meteorological parameter is missing from the ANN. A global pattern of the accuracy of the ANN might then give an indicator as to which parameters should be included. The ANN might be expected to differ as a function of cloud type, but perhaps a separate model for each cloud type (e.g. Gryspeerdt and Stier, 2012 or Oreopoulos et al., 2016) might be useful.

If one trains a single global model to predict CLF, using the same predictors and model setup as for the regional ANNs, it cannot predict CLF as well as most regional ANNs (R^2 of global model $\approx$ 0.45; median of regional ANNs $>$ 0.60). While adding additional predictors to the global ANN could still improve the skill of the model, it is unrealistic to think that a single model could represent clouds as well as regional models can (it would also increase the probability of non-causal relationships). Regional ANNs are superior, as they are able to reproduce the regionally varying predictor-predictand relationships (c.f. fig. 6 in the manuscript). These regional differences would be blurred or missed completely when using a single global ANN. Regional ANNs also have the advantage that knowledge on typical regional characteristics (e.g. aerosol species composition) can be included in the interpretation of the results (as in Andersen et al., 2016). That being said, cloud type-specific ANNs seem to be an interesting idea for future work.

Minor points

P2L9: Perhaps only e.g. is necessary

We agree and have changed the manuscript accordingly.

P2L24: Why is the 2.1um effective radius used with the 3.7um LWP retrieval?

We have changed the cloud products used (see our response below).

P2L29: Is the liquid fraction a suitable measure of cloud fraction, as it depends on the overlying ice cloud fraction? The authors could consider using cases where only liquid cloud exists in a gridbox, as this would remove this source of uncertainty.

After internal and peer discussions, we have decided to run the ANN with monthly means of single layer clouds only. While the results are nearly identical, the argument is valid, so that we only use single layer cloud products the current version of the manuscript.

P3L4: AOD is proportional to CCN (at least at some scales, see Andreae, 2009), it is just not a direct measurement (the same as with mass, as it also depends on aerosol optical properties)

Yes, we agree. We have corrected this in the revised manuscript.

P3L7: Many recent studies have used aerosol index (AOD times angstrom exponent) or a reanalysis aerosol parameter (e.g. Lebsock et al., 2008; McCoy et al., 2016). As these have been shown to more accurately predict cloud properties, they might further improve the skill of the ANN. Although MODIS AI is not necessarily accurate over land (Levy et al., 2013), it could be used over ocean in this study.

For this study, we used the newest version of MODIS products available, collection 6 (C6). In C6, the MODIS Ångström exponent (needed for the computation of the aerosol index as it is the product of AOD and the Ångström exponent) has been discontinued in level 3 (L3) data (p. 3018 Levy et al., 2013). We believe that for this and for other reasons, other recent studies also use the AOD as a proxy for CCN (e.g. Chakraborty et al., 2016; Stathopoulos et al., 2017; Patel et al., 2017). We agree with the referee though that the aerosol index might be a more appropriate measure for CCN and have thus chosen to compute the Ångström exponent (550 and 867nm) ourselves to use aerosol index instead of AOD in the ANN. The following figures 2 and 3 are the new results of the ANN when using AI instead of AOD. The spatial patterns in ANN skill, as well as the mean global sensitivities are nearly identical (cf. figures 3 and 5 in the original ACPD manuscript).

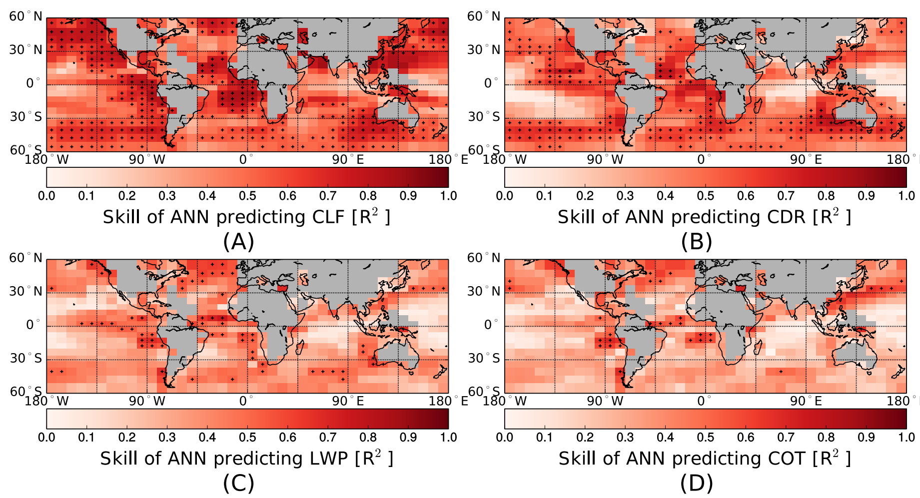

Figure 2: Global patterns of ANN skill as in the manuscript; AI has been used instead of AOD.

Small differences can be observed in the regional patterns of ANN sensitivities (fig. 4) to AI vs. AOD. The CLF sensitivity to AI is higher in the Southeast Atlantic than its sensitivity to AOD in that specific region. The Southeast Atlantic is of course dominated by biomass-burning aerosols, which are mostly fine mode and thus feature a relatively larger AI than AOD. The sensitivity of CDR

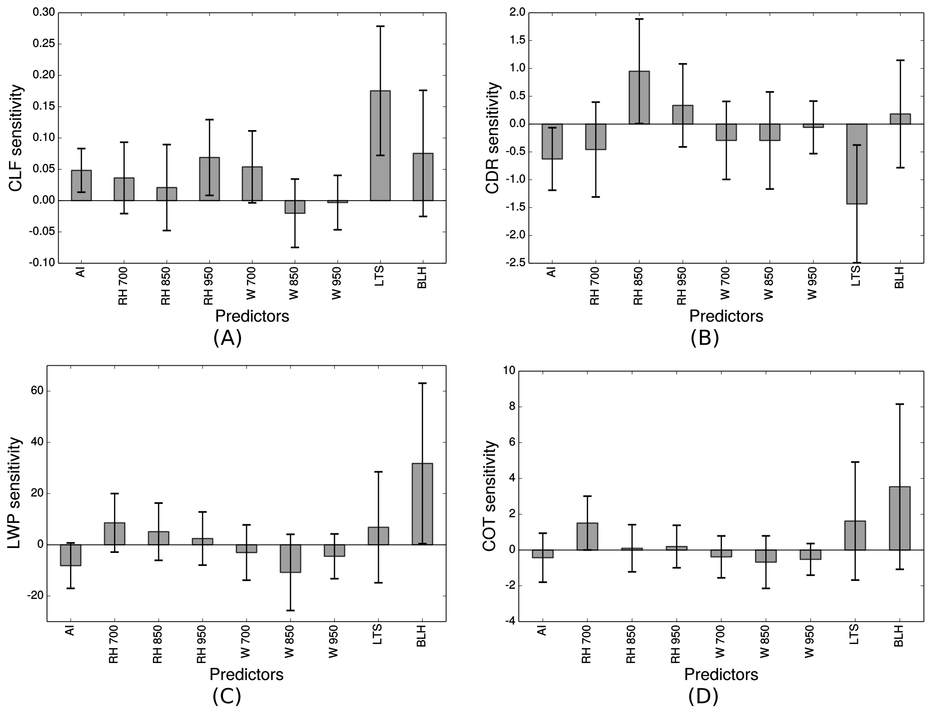

Figure 3: Global mean relative sensitivities as in the manuscript; AI has been used instead of AOD.

to AI differs from its sensitivity to AOD in regions that are dominated by desert dust. Dust is relatively coarse, so that the AI would be disproportionally lower than the AOD in these regions, which might explain the differences between the sensitivities of the two.

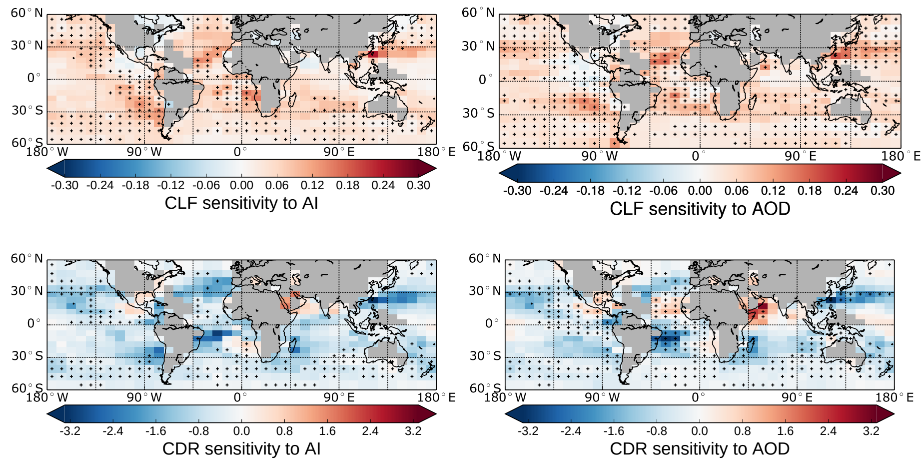

Figure 4: Differences in sensitivities of CLF and CDR to AI (left-hand column) vs. AOD (right-hand column).

P3L13: It is definitely a good idea to investigate variables that have been previously used in aerosol-cloud studies. Koren et al., (2010) might also provide some useful guidance here. Although it was focussed on looking at convective clouds, some of the results (e.g. Figs. 8,9) might help decide which variables should be included in the ANN).

We agree that additional variables (e.g. geopotential height, horizontal winds) might improve the ANN performance in some regions. Our goal in predictor selection was to minimize the number of predictors to a few key variables, in order to prevent covaration between the predictors. Also, additional predictors increase the probability of highlighting non-causal relationships.

P4L33: Is there any significance behind using five hidden nodes?

After thorough testing, five hidden nodes appeared to be a good global number. In general, the optimum number of nodes is dependent on the problem at hand. The number of nodes needed is connected to the complexity of the relationships, the amount of noise in the data and the amount of training data available. Too many nodes can lead to overfitting and poor generalization, whereas the ANN may not converge to a global minimum when too few nodes are used (Gardner and Dorling, 1998). We found that while regional ANNs may differ, five nodes where a reasonable choice, as additional nodes typically only marginally, if at all, increased model skill. To illustrate this, figure 5 is an example of the effect of the number of hidden nodes on ANN skill in the Southeast Atlantic region. This figure is obviously not the basis for our decision to use 5 nodes, but is intended to illustrate a typical example for the dependence of a regional ANN skill on the number of hidden nodes.

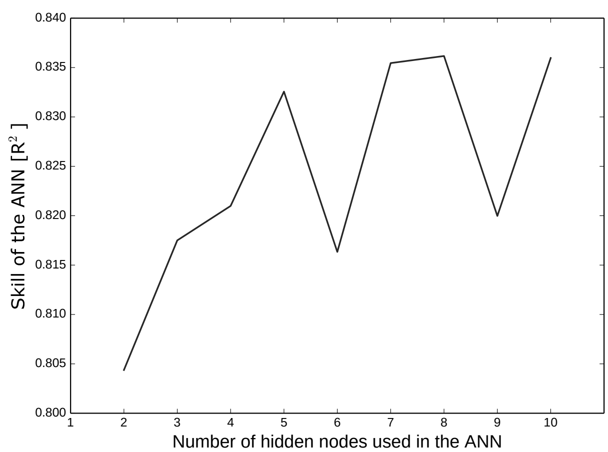

Figure 5: Example (Southeast Atlantic) for the effect of the number of hidden nodes in the ANN.

P5L7: Are the sensitivities calculated using the local variation of meteorological values, or the same artificial values globally? If the relationship is non-linear and the mean values of the meteorological variables vary across the globe, this could strongly affect the calculated sensitivity.

This sentence was intended to describe how sensitivities can generally be computed with an ANN. In the text passage further down (P5L14), we describe how sensitivities are computed in this study. To answer your question: Yes, the sensitivities are calculated using the local variation of meteorological values ('grid cell specific mean values'). In the revised version of the manuscript, we will attempt to describe both text passages more clearly.

P5L14: I am not sure I understand this sentence (which might explain my previous query?)

We compute ANN-predicted outputs for two groups of input data:

- All grid-cell specific retrievals of a specific predictor smaller than its 25th percentile.
- All grid-cell specific retrievals of a specific predictor greater than its 75th percentile.

In all cases, all other predictors are held constant at their grid-cell specific mean values. We then compute the average of both groups of ANN-predicted outputs. The difference between the two averages is defined as the sensitivity of the predictand to the specific predictor that was varied. We will try to more clearly describe this in the revised version of the manuscript.

P5L20: If the other meteorological factors in the ANN are held constant, does this produce a different result for the simple sensitivity? (see main point)

We have tested this for the sensitivity of CLF to AI. As above, we have also used data from the the Southeast Atlantic for this example. We found that the sensitivity (linear slope of AI-CLF relationship) of CLF to AI is $\approx 40\,\%$ lower in the ANN than in the observations. This is, of course, because in the sensitivity of the ANN, the other predictors are held constant, constraining their effect on CLF. This corresponds rather well to Gryspeerdt et al. (2016) who found that the sensitivity of CLF to AOD is reduced even further ($80\,\%$) when including information on CDNC along the causal pathway of the AOD-CLF relationship.

P6L7: As I understand it previous work focusses on the sensitivity as this is related to the strength of the cloud response to aerosol. It is not often assumed that aerosols can explain much of the variability in cloud properties which might explain the low skill here.

Yes, we agree. This figure is not intended to illustrate sensitivities, but that we are in a space of large uncertainty when we derive sensitivities using bivariate methods. Using a multivariate approach (also the case for multiple regression, as outlined above) we are capturing more of the aerosol-cloud climate system. The derived sensitivities might thus be more reliable.

P7L1: Perhaps another measure of skill might be useful in addition to the R^2? It could be argued that the skill in the shallow cumulus regions is quite good, in that the ANN (presumably) gets the cloud properties roughly right (the rms error might be small)?

Yes, indeed, we also looked at the relative RMSE. Actually, the a combination of relative RMSE and R^2 thresholds (P7L4) are used to select the regions that are used for the computation of sensitivities (marked with a '+' in the maps). The relative RMSE and R^2 are basically invertly related.

P7L4: Does this removal of the poor skill models bias the results, perhaps as a function of meteorology (as would appear to be the case from the maps in Fig. 3)

The computed sensitivities are only valid for the regions and are not intended to be "global" in that sense.

P7L9: How does these sensitivities compare to previous results? Several studies have calculated AOD-CF or AOD-droplet number concentration sensitivities which could be compared here (e.g. Quaas et al (2008), Grandey et al. (2012), Gryspeerdt et al. (2016))

We compute the sensitivity a slightly different way, so a straight-forward comparison is not possible. However, in a similar way that Gryspeerdt et al. (2016) constrain the aerosol-CLF relationship with CDNC, the ANN constrains the aerosol-cloud relationships by meteorology. In the updated version of the manuscript, we will include comparisons to sensitivities found by other recent studies.

P12L3: Are the covariations really spurious? The argument here is not that the covariations don't exist, but that they are not representative of the causal relationship. I would suggest that if 'direct physical relationship' was replaced with 'causal relationship', this could instead mention the issue of confounding variables, similar to Gryspeerdt et al.,(2016).

We will restructure this text passage in the updated version of the manuscript.

P12L4: To what extent has using RH in the ANN accounted for this effect?

As shown in figure 6 within this document, the sensitivity of CLF to AI is weakened in the ANN, probably due to the meteorological constrains of the model. These are hard to track down to a single predictor, though (e.g. RH). It is likely that the main confounding factor for this relationship is RH and that most of the change in AI-CLF sensitivity is due to constraining RH.

References

Andersen, H., Cermak, J., Fuchs, J., and Schwarz, K. (2016). Global observations of cloud-sensitive aerosol loadings in low-level marine clouds. *Journal of Geophysical Research: Atmospheres*, 121(21):12936–12946.

Chakraborty, S., Fu, R., Massie, S. T., and Stephens, G. (2016). Relative influence of meteorological conditions and aerosols on the lifetime of mesoscale convective systems. *Proceedings of the National Academy of Sciences of the United States of America*, 113(27):7426–7431.

Gardner, M. and Dorling, S. (1998). Artificial neural networks (the multilayer perceptron) – a review of applications in the atmospheric sciences. *Atmospheric Environment*, 32(14):2627–2636.

Gryspeerdt, E., Quaas, J., and Bellouin, N. (2016). Constraining the aerosol influence on cloud fraction. *Journal of Geophysical Research: Atmospheres*, 121:3566–3583.

Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C. (2013). The Collection 6 MODIS aerosol products over land and ocean. *Atmospheric Measurement Techniques*, 6(11):2989–3034.

Patel, P. N., Quaas, J., and Kumar, R. (2017). A new statistical approach to improve the satellite based estimation of the radiative forcing by aerosol–cloud interactions. *Atmospheric Chemistry and Physics Discussions*, 17:3687–3698.

Stathopoulos, S., Georgoulias, A., and Kourtidis, K. (2017). Space-borne observations of aerosol - cloud relations for cloud systems of different heights. *Atmospheric Research*, 183:191–201.

Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks

— RESPONSE TO REFEREE 3 —

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We would like to thank referee 3 for her/his review of the manuscript and her/his constructive criticism. Comments by the referee are colored in blue, our replies are colored in black.

This paper pursues a promising approach to study the sensitivity of marine liquid-water cloud properties on a set of meteorological and aerosol predictors, using an artificial neural network approach. It steers clear of correlative approaches for studying aerosol-cloud interactions and instead considers the meteorological context, segregated by region / meteorological regime. In essence, this amounts to a multi-variate analysis based on an optimal combination of satellite and re-analysis data. The paper is very well written, clearly represents new ideas, and has the potential to lead to major improvements in our assessment of ACI, regionally and globally. It is rare to see such a high-quality paper. I only have minor comments, which don't necessarily have to be addressed in this manuscript, but could be considered in future work. The most important ones are probably #1 regarding scale, and regarding the quality (reliability) of the data. Also, follow-up papers might consider using the co-sensitivity of some predictors (details below).

In a separate comment to the editor, I recommended that the paper be highlighted because it seems highly innovative in its approach and deviates from the traditional correlative aerosol-cloud interaction studies. I believe that it has potential to change the direction of this field of research.

Thank you very much for this kind assessment. We respond to each point individually below.

General comments:

p5,L18: In the spirit of the McComiskey and Feingold ACI papers, it would have been interesting to also consider the impact of scale on ACI relationships. Here, one specific scale has been used (dictated by the analysis grid) - but it may not be straightforward to generalize these relationships.

This is a good point and we agree that the scale of the data sets used to study aerosol-cloud interactions influences the derived sensitivities (McComiskey et al., 2009; McComiskey and Feingold, 2012). Here, we use temporally and spatially highly aggregated data sets (monthly means in the defined equal-area regions), as with this study, we are specifically interested in the very large scale mechanisms and patterns of the aerosol-cloud-climate system. This is certainly not the scale at which the processes occur, so that our derived sensitivities may not match the magnitude of the sensitivities at the process scale. An analysis of the impact of the extent of spatial aggregation of the 1°x1° data on the derived sensitivities would be interesting; however, the spatial aggregation we chose was needed for sampling reasons (sufficient number of observations for the statistical model). In the revised version of the manuscript, we discuss this on P6L1–3. ("*As the temporal and spatial scales considered in this study are not on the same scale as the actual processes, so that the calculated sensitivities may not match the magnitude of the sensitivities at the process scale (McComiskey et al., 2009; McComiskey and Feingold, 2012).*")

p6,L4: "skill of simple correlation between AOD & cloud properties": It is a bit unclear, which "simple correlations" specifically have been used for this study. This statement calls for elaboration. The statement on p6,L6/7 shows the intent - the "simple correlations" are used as a baseline to show the improved predictive skill of ANN. The quantitative results would be more useful by including more information about that baseline.

Here, with "simple correlation" we referred to a "simple" Pearson correlation between AOD and either CLF/CDR/LWP/COT in each equal area region. In the revised version of the manuscript, we describe this at P6L8, however, in the current version of the manuscript, the results of Pearson correlations between log(AI) and the respective cloud properties is illustrated in figure 2.

p6,L11 (fig 4): How/where are the equal-area regions defined? Are those just pixel aggregated that meet the selection criteria for the sensitivity analysis?

This is explained in the manuscript on P4L33-P5L3. The equal-area regions are defined by dividing the space between 60°N and 60°S (and all longitudes) into 20x40 equally sized areas. The original 1°x1° data is aggregated in these regions at their original spatial resolution. The selection criteria for the sensitivity analysis is checked for each equal-area region (but only for the sensitivity analysis - in figure 4, all equal-area regions are shown). In the revised version of the manuscript, we added some information to the caption of figure 4 for clarity.

p9, Fig 5. How is the CF and LWP sensitivity to AOD compatible? Is it a fair statement to say that we get more clouds with lower LWP for higher aerosol loading, while COD stays the same (perhaps because the "classical" indirect effect kicks in) - or can we not make such a blanket statement?

The CLF sensitivity to AOD/AI is probably the sensitivity that is the most uncertain, due to cloud contamination of the satellite aerosol retrievals and the influence of confounding variables on both CLF and the satellite retrieved aerosol quantity. While we weaken the influence of confounding variables by including them in the ANN, we are not able to reduce effects related to data quality (this is discussed on P13L4–6 in the revised version of the manuscript:

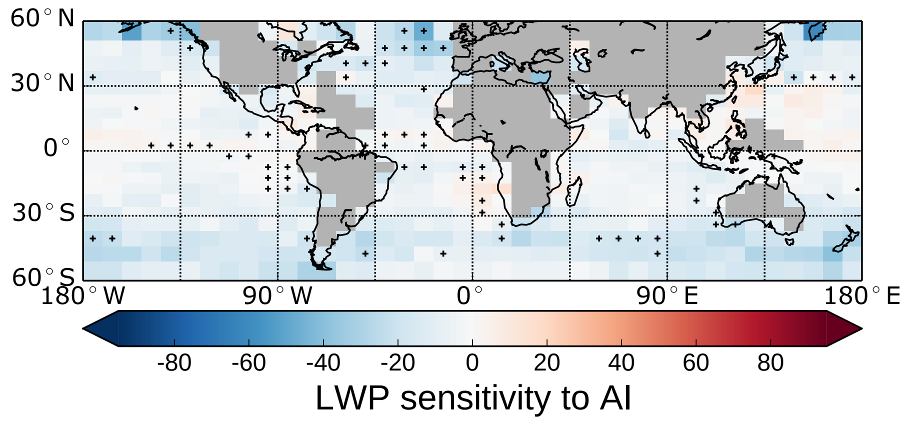

Figure 1: Global map of LWP sensitivity to AI: The globally averaged sensitivities are based on the regions marked with a '+'.

"*While the influence of confounding factors is limited by the multivariate approach, effects concerning data quality (e.g. cloud contamination) are not accounted for and need to be considered when interpreting the CLF sensitivity to AI.*"). One should also note that the averaged LWP sensitivities rely on very few regions (due to the selection criteria) and should thus not be considered global. In most regions, the sensitivity of LWP to AI is relatively low.

While it makes sense to combine the sensitivities as proposed by you, one needs to remember that these are derived from separate ANNs. While LWP and CLF in the respective ANNs respond to AI/AOD in the way that you point out, changes in LWP might also affect CLF and vice versa, which would not be accounted for. Therefore, we are somewhat cautious in the interpretation of combined sensitivities.

p10, L5: Would it make sense to plot co-sensitivity maps, considering that many predictands co-vary with predictors. In the inverse theory equivalent, one would consider the off-diagonal elements of the covariance matrices. After all, one of the attractive features of this analysis is that it allows multi-variate analysis of ACI, fully considering the meteorologic conditions - but then the plots / analysis do not reap the full benefits of this approach. The authors do explain some of the co-variabilities/co-sensitivities, but then again it would be even better to have some graphical representation for some of these connections.

Yes, this is a good idea - and an idea which we discussed internally, as well. Ultimately, this level of detail exceeds the scope of this study, as one would have to create co-sensitivity plots for each grid-cell-specific ANN individually and would thus not be able to produce summarized global co-sensitivities easily. This is an idea we are currently pursuing in a more detailed regional study.

p10,L28: Does the CDR - AOD relationship for the SE Atlantic region make sense? For the outflow from the Arabian peninsula and the Sahara, it does, and the manuscript explains this with dust - but on the West coast of Namibia and Angola the dust is confined to the coast. It is possible that the identified relationships here points to limitations of the data set(s) that serve as the basis. Perhaps dust is overrepresented in the data? Overall, it would be good to see a discussion in which regions we would trust the correlations (given the uncertainties in the data).

This is a good question - in a regional study some years ago, we found that in certain conditions (stable/humid), AI and CDR are positively related in the Southeast Atlantic (Andersen and Cermak, 2015). However, in most cases, the AI-CDR relationship was found to be negative as in (e.g. Costantino and Bréon, 2013). This specific regional sensitivity may be affected by retrieval or sampling issues, as now discussed in the revised version of the manuscript on P10L5–8 ("*Issues of sampling (few aerosol retrievals in high CLF-regions) or scale (highly aggregated data) or their combination might affect the observed CDR sensitivity to AI in this region.*").

p12, L15: So, cloud radiative effect sensitivities are actually not (yet) addressed in the manuscript. Instead, cloud properties are analyzed. Earlier in the manuscript (p4,L24), it is stated that cloud radiative effects are analyzed. This should be fixed (minor comment).

Yes, you are correct. We have deleted the mentioned text passage in the revised manuscript.

References

Andersen, H. and Cermak, J. (2015). How thermodynamic environments control stratocumulus microphysics and interactions with aerosols. *Environmental Research Letters*, 10(2):24004.

Costantino, L. and Bréon, F.-M. (2013). Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations. *Atmospheric Chemistry and Physics*, 13(1):69–88.

McComiskey, A. and Feingold, G. (2012). The scale problem in quantifying aerosol indirect effects. *Atmospheric Chemistry and Physics*, 12(2):1031–1049.

McComiskey, A., Feingold, G., Frisch, a. S., Turner, D. D., Miller, M. a., Chiu, J. C., Min, Q., and Ogren, J. a. (2009). An assessment of aerosol-cloud interactions in marine stratus clouds based on surface remote sensing. *Journal of Geophysical Research: Atmospheres*, 114(D9):D09203.

List of important changes

P2	Wording of hypotheses modified for clarity (Referee 1)
P2–P3	Single-layer cloud products are now used (Referee 2)
P3	Aerosol index is now used instead of aerosol optical depth (Referees 1, 2)
P3	Discussion of the usage of cloud droplet effective radius vs. cloud droplet number concentration (Referee 1)
P3–P4	Discussion of predictor selection (Referee 2)
P5	Discussion of regional ANNs (Referee 1)
P5	Discussion of quality criteria for sensitivity studies (Referee 1)
P6	Discussion on importance of temporal and spatial scales (Referee 3)
P6–P7	Multiple linear regression now included for comparison (Referees 1, 2)
P11	Discussion of data quality in certain regions (Referee 3)
P13	Comparisons with results of other studies (Referee 2)
P13	Discussion of contributions of this study (Referee 1)
P13	Outlook is expanded (Referee 2)

Understanding the drivers of marine liquid-water cloud occurrence and properties with global observations using neural networks

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Abstract. The role of aerosols, clouds and their interactions with radiation remain among the largest unknowns in the climate system. Even though the processes involved are complex, aerosol-cloud interactions are often analyzed by means of bivariate relationships. In this study, 15 years (2001–2015) of monthly satellite-retrieved nearly-global aerosol products are combined with reanalysis data of various meteorological parameters to predict satellite-derived marine liquid-water cloud occurrence and properties by means of regionally-specific artificial neural networks. The statistical models used are shown to be capable of predicting clouds, especially in regions of high cloud variability. At this monthly scale, lower tropospheric stability is shown to be the main determinant of cloud fraction and droplet size, especially in stratocumulus regions, while boundary layer height controls the liquid-water amount and thus the optical thickness of clouds. While aerosols show the expected impact on clouds, at this scale they are less relevant than some meteorological factors. Global patterns of the derived sensitivities point to regional characteristics of aerosol and cloud processes.

1 Motivation and aim

Clouds and their microphysical properties play a central role in the Earth's radiative budget by increasing the albedo but also by interacting with outgoing thermal radiation, leading to a net cooling effect (Boucher et al., 2013). Low-level marine liquid-water clouds are the cloud type with the biggest net cooling effect; their shortwave signal by far exceeds their longwave signal (Hartmann et al., 1992; Wood, 2012; Russell et al., 2013; Chen et al., 2014). A global increase in the occurrence frequency or cooling properties of marine low-level liquid-water clouds could thus offset some of the greenhouse gas warming (Latham et al., 2008). Thus, a complete understanding of the physical processes that determine marine liquid-water clouds and their properties is critical.

Atmospheric aerosols are essential for the formation of clouds, influencing cloud properties as cloud condensation nuclei. An increase in aerosol particles leads to a higher cloud droplet number concentration, and, assuming a constant cloud water content, to smaller droplet radii. This changes the cloud's radiative properties, as the larger overall droplet surface area increases cloud reflectivity (Twomey, 1977). These changes in droplet number concentration and size are also thought to have ramifications on cloud lifetime (Albrecht, 1989) and cloud vertical extent (Pincus and Baker, 1994). However, these processes are nonlinear (Bréon et al., 2002; Koren et al., 2014; Andersen et al., 2016; Glassmeier and Lohmann, 2016) and dependent on various environmental conditions that all feature different patterns in time and space (e.g. Loeb and Schuster, 2008; Stevens and Feingold, 2009; Su et al., 2010; Andersen and Cermak, 2015; Andersen et al., 2016).

Even though there have been significant efforts and advances in understanding aerosol-cloud interactions (ACI) over the last decades, the overall scientific understanding is still considered as low (Boucher et al., 2013). This springs from the complexity of ACI and cloud processes themselves, the temporal and spatial scales at which these processes occur, as well as challenges in observing them.

In the satellite observational community, a typical investigative approach to analyze ACI is to directly relate aerosol and cloud observations quantitatively using bivariate statistics, often explicitly considering one or two meteorological variables ~~(c.f. e.g. Matsui et al., 2004, 2006; Chen et al., 2014; Andersen and Cermak, 2015)~~(e.g. Matsui et al., 2004, 2006; Chen et al., 2014; Ande Even though important process inferences have been made on this basis, the limitation of said method set is clearly that the complexity of the processes is not mirrored by the complexity of the statistical method: only selected aspects of the aerosol-cloud system can be analyzed at one time. A multivariate analysis of the relationships between cloud properties and various predictors, including aerosol and meteorological conditions, might be more appropriate for an adequate representation of these atmospheric interactions. In this spirit, this study combines near-global observational and reanalysis data sets as predictors in a multilayer perceptron artificial neural network (ANN) to model near-global marine water cloud occurrence and properties. The main goal of this study is to identify the main drivers of marine liquid-water cloud occurrence as well as physical and optical properties on a global scale, estimate sensitivities for each predictor, and determine regional patterns therein.

The guiding hypotheses are:

1. Neural networks are capable of skillfully modeling cloud patterns on monthly time scales, and allow for a separation and estimates of the relative importance of aerosol and various meteorological factors.

2. Global aerosol and cloud patterns are not only related at a global scale, but regional patterns exist as well.

3. ~~While aerosols are a key determinant for cloud occurrence and properties, other factors are at least equally relevant at~~ At the spatial and temporal scales considered here, meteorological conditions are more important for cloud occurrence and properties than aerosols.

2 Data and methods

2.1 Data sets

The analysis uses 15 years (2001–2015) of nearly global (60°N–60°S) satellite retrievals and reanalysis fields. Monthly averages of level 3 collection 6 products based on measurements by the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor on the Terra platform (Levy et al., 2013) are used for information on cloud fraction (CLF; data set: Cloud_Retrieval_Fraction_1L_Liquid_FMean), cloud-top droplet effective radius (CDR; data set: Cloud_Effective_Radius-_1L_Liquid_Mean_Mean), cloud liquid water path (LWP; data set: Cloud_Water_Path_~~37~~1L_Liquid_Mean_Mean) and cloud optical thickness (COT; data set: Cloud_Optical_Thickness_1L_Liquid_Mean_Mean). While cloud microphysics may also be represented by cloud droplet number concentration, its retrieval requires additional assumptions on vertical cloud water distribution, leading to increased uncertainty (Brenguier et al., 2000), especially in non-stratocumulus cloud regions (Bennartz and Rausch, As this study investigates liquid-water cloud properties globally, CDR is thus used as a more robust proxy, even though it is also dependent on cloud liquid-water content to some extent (Brenguier et al., 2000). To confine the analysis to liquid-water clouds ~~, only~~ and to reduce measurement uncertainties due to overlying ice clouds, only single-layer liquid-water cloud products are used. Information on aerosol loading as a proxy for cloud condensation nuclei is provided by aerosol ~~optical depth~~ index (AI; computed as a product of the aerosol optical depth (AOD; at 0.55 μm~~(AOD; data set: Aerosol_Optical_Depth_Land_Ocean_Mean_Mean).~~) and the Ångström exponent (0.55 and 0.867 μm)). While many studies use the AOD as a proxy for cloud condensation nuclei (e.g. Andreae, 2009; Quaas et al., 2009, 2010; Peters et al., 2012; Koren et al., 2012), the AI has often been found to be a superior measure for this quantity (Stier, 2016), as it weights the fine mode stronger than AOD alone (Nakajima et al., 2001). Some constraints of ~~AOD~~ AI are that it can be affected by aerosol swelling due to hydration in humid environments ~~, that it is proportional to aerosol mass and not CCN concentration~~(Loeb and Schuster, 2008), and that the retrieval describes vertically integrated information and not specifically aerosol at cloud base height where cloud condensation nuclei are typically activated (Shinozuka et al., 2015). ~~Stier (2016) discusses that in 71 % of the ocean area, AOD only explains 25 % of the cloud condensation nuclei (CCN) variance at cloud base. Still, it is commonly used as a proxy for the columnar aerosol concentration or CCN in ACI studies (Andreae, 2009; Quaas et al., 2009, 2010; Peters et al., 2012; Koren et al., 2012).~~

Satellite retrievals are combined with reanalysis data sets from the European Centre for Medium-Range Weather Forecasts (ECMWF) for information on meteorological predictors. The ERA-Interim reanalysis provides data for the time since 1979 and is still continued (Dee et al., 2011). Monthly means of mean daily reanalysis data are used for information on various meteorological predictors at selected atmospheric pressure levels. Meteorological determinants may be grouped into information on relative humidity (RH - at pressure levels 950 hPa (Andersen and Cermak, 2015), 850 hPa (Chen et al., 2014) and 700 hPa (Engström and Ekman, 2010)), vertical velocity (W - at pressure levels 950 hPa, 850 hPa (Kaufman et al., 2005; Engström and Ekman, 2010) and 700 hPa (Engström and Ekman, 2010)), boundary layer height (BLH (Painemal et al., 2014)) and lower tropospheric stability (LTS - computed as the difference in potential temperature between 700 hPa and the surface (Klein and Hartmann, 1993; Chen et al., 2014; Andersen and Cermak, 2015; Andersen et al., 2016)). The reanalysis data used features an original spatial resolution of 0.5°x0.5° and is subsequently resampled to fit the MODIS 1°x1° grid.

Typically, clouds form when air cools off, increasing RH. Once supersaturation is reached, water vapor can condense on the CCN. Predictors are selected that are thought to capture this very basic concept well: Vertical velocity and relative humidity are selected as indicators of cloud dynamics and stratification at various pressure levels. CCN are represented by ~~AOD~~AI, and BLH and LTS describe the large-scale setting. All predictors have been shown to be relevant determinants of liquid-water clouds or their interactions with aerosols in the studies named above. When available, vertically resolved information is preferred to column integrated (e.g. RH at three different pressure levels is preferred to total columnar water vapor), in order to trace processes at various atmospheric levels. While a higher number of reasonable predictors (e.g. geopotential height or horizontal winds as in Koren et al. (2010)) is likely to marginally increase the skill of the ANN, it would increase model complexity and make interpretation more difficult.

By design the data sets applied in this study average over time and space to specifically study the large-scale changes within the aerosol-cloud-climate system and to allow for future comparisons with global climate models. While on these scales, the causal sequence of cloud processes may not be intact and the processes themselves cannot be observed, their overall ramifications are thought to be represented adequately, in that temporal averaging is intended as a proxy for process relationships.

2.2 Artificial neural networks and study design

Basics of artificial neural networks

Machine learning systems consist of a set of numerical operators designed to compute a designated output on given input data. The basic principles, such as the number of numerical links between parameters, are fixed. Artificial neural networks can be described as a branch of machine learning systems. Multilayer perceptrons are a specific type of neural network that are commonly used in the atmospheric sciences and environmental sciences in general, as they are able to model highly nonlinear functions. This type of ANN consists of several layers of interconnected neurons. In general, the architecture of multilayer perceptron ANNs is variable but a typical ANN may consist of an input layer, at least one hidden layer and an output layer. The information from an input pattern is strictly passed from the input layer via the hidden layer(s) to the output layer that yields the desired output pattern (feed-forward ANN). Multilayer perceptron ANNs are fully connected, i.e. each neuron is connected to every neuron in the neighboring layer(s). All connections between neurons in the ANN are specifically weighted so that the information passed to a neuron is the sum of the weighted outputs from the previous layer (net input). The neuron modifies the information by multiplication with a nonlinear transfer function and passes this information through specific weights to all neurons of the following layer (Gardner and Dorling, 1998).

In general, these types of ANNs learn through training. During the training period a subset of the input and output data sets are fed into the ANN. Using this training data, a learning algorithm adjusts the individual weights of each neuron in the network to minimize the error of the output (e.g. the difference between the modeled and observed outputs). The speed of the learning process is adjusted by a learning rate that determines the step size taken during the iterative learning process. While a high learning rate leads to faster convergence, it may miss a global optimum. An additional momentum term adds a fraction of the previous weight change to the current weight change to assist the optimization algorithm out of local minima (Gardner and Dorling, 1998). After the learning algorithm has reached convergence, the predicted output of the network can be compared to the original output for an estimate of model skill. To ensure that the ANN does not only represent the particular data used in the training (overfitting) and is able to generalize the functional relationships underlying the training data, the model is validated using a second independent subset of the input data. If the ANN is able to generalize the relationships between the data sets, the difference between training and validation errors and the overall error are small. The ANN is tested on a third set of independent data to ensure that the model is not overfit to the validation data.

Design of the study and application of the neural network

The ability of the ANN to predict cloud occurrence ~~, properties and radiative effects~~ and properties is dependent not only on an informed choice of predictors, ANN also require sufficient data that fully represent all cases that the ANN is required to generalize, as ANNs perform well for interpolation but poorly for extrapolation (Gardner and Dorling, 1998). In order to circumvent sampling issues and to enable a direct comparability of results in different regions, the near-global data sets are summarized in 40x20 equal area grid cells by aggregating grid cells at the original spatial resolution of 1°x1°. This leads to an increase from the original 180 data points (15 years, 12 months) for each input/output to between 8,000 and 14,000, depending on the number of 1°x1° pixels that fall into a specific region. Region-specific neural networks are needed to capture regionally varying relationships between cloud properties and their determinants. These relationships feature regional patterns as they depend on liquid-water cloud type, aerosol composition, meteorology and the respective seasonal cycles, all of which exhibit regional patterns (e.g. Stevens and Feingold, 2009; Andersen et al., 2016). These regional characteristics would be blurred or missed completely when using a single global ANN.

The ANN is only applied in grid cells where a minimum of 2000 valid observations exist. In each equal area, an independent ANN is trained over 500 epochs (i.e. number of times the network iterates over the training data) with 60 % of the data, validated and tested on 20 % of the data each. A simple network topology with one hidden layer consisting of five hidden neurons is applied a) for a more comprehensible model and b) to reduce potential overfitting (Gardner and Dorling, 1998). Multilayer perceptrons with just one hidden layer are frequently used in ecological studies (e.g. Hartmann et al., 2008; Cermak and Knutti, 2009) as they have been shown by several independent studies to be able to approximate any continuous function (Cybenko, 1989; Funahashi, 1989; Hornik et al., 1989; Kecman and Vojislav, 2001; Olden and Jackson, 2002; Di Noia et al., 2013). A hyperbolic tangent is used as the activation function, the weights are initialized randomly from a uniform distribution between -0.1 and 0.1. Gradient descent (Werbos, 1990; Le et al., 2017) is used as the optimization algorithm, with a learning rate of 0.003 and a momentum of 0.01. In-depth testing was undertaken to adjust the details of the model's settings by comparing model skill for a wide number of model setups as in Hartmann et al. (2016). Once the ANN is trained and able to generalize the relationships between the data sets adequately, sensitivity analyses ~~are conducted. Sensitivities~~ can be conducted. In general, sensitivities are systematically tested by varying each input variable while holding all other input variables constant, e.g., at their average (details see below). In this way the individual contributions of each variable can be analyzed (Olden and Jackson, 2002). A schematic view on the general architecture of the ANN and the training, validation and sensitivity steps is given in Fig. 1.

The ANN skill in modeling the desired outputs is evaluated with the correlation (R^2) between ANN testing output and the corresponding observation data. Sensitivities are only computed for grid cells, where the ANN $R^2 > 0.5$ and the root mean square error relative to the mean (rel. RMSE) $<$ its global average in order to ~~exclude unreasonable models~~ only investigate sensitivities of models that are capable of adequately representing the observed cloud patterns. The derived average sensitivities are only valid for the considered regions and should thus not be interpreted as 'global'. In order to derive a representative and meaningful sensitivity, the mean of ANN-predicted outputs are compared for two groups of input data: all retrievals of a specific predictor smaller than its 25th percentile and ~~those~~ all retrievals greater than its 75th percentile;

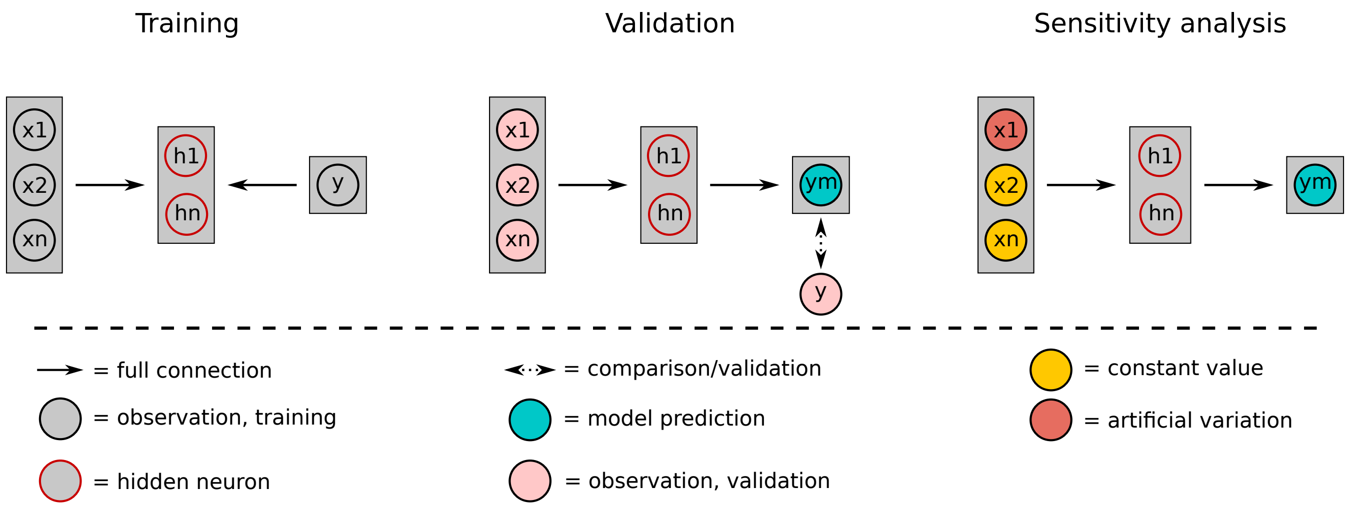

Figure 1. A schematic view on the general architecture and design of multilayer perceptron artificial neural networks. In this study, the ANN features a single hidden layer with 5 neurons.

in all cases, the other predictors are held constant at their grid-cell specific mean values. In comparison to a stepwise increase of one specific predictor, a more relevant measure of a typical sensitivity can be derived, as the predictor distribution is considered. Thus, in the context of this study, the sensitivity is defined as the mean difference between the predicted cloud property in the groups of low and high predictor values. Typically, the aerosol effect on e.g. CDR is described by the $\delta log(CDR)/\delta log(aerosol)$ relationship, where aerosol can be either AOD or ~~aerosol index~~ AI (e.g. Costantino and Bréon, 2013). While this gives a regionally comparable estimate of the aerosol-cloud sensitivity, it does not explicitly consider the meteorological framework. As the temporal and spatial scales considered in this study are much larger than the actual processes, the calculated sensitivities represent the system scale, and may not match the magnitude of the sensitivities at the process scale (McComiskey et al., 2009; McComiskey and Feingold, 2012).

3 Results and discussion

3.1 Skill of the ANN in predicting cloud occurrence and properties

The skill of the ANNs to predict marine liquid-water cloud occurrence, as well as physical and optical properties is shown in Fig. 2 (blue boxes) and contrasted with the skill of a ~~simple correlation between AOD~~ multiple linear regression using the identical set of predictors (red boxes) and a simple Pearson correlation between log(AI) and the cloud properties (~~red~~ black boxes). In the ANN, CLF is predicted with the highest accuracy~~(mean R^2 of 0.55)~~. While for CDR the skill of the ANN is also $>$ 0.5 for many regions~~(mean R^2 of 0.45)~~, LWP and COT are predicted less accurately~~(mean R^2 of 0.35)~~. The skill of the multiple linear regression is close to the skill of the ANN, but typically explains a few percent less of the cloud variability, possibly indicating a small contribution in model skill by nonlinear representations of relationships within the ANN. It is shown that ~~AOD~~ log(AI) alone typically explains less than ~~10~~20 % of the cloud property variability. As a much higher fraction of the cloud variability is explained in the multivariate approaches, the sensitivities derived from the ANN are likely to be more reliable.

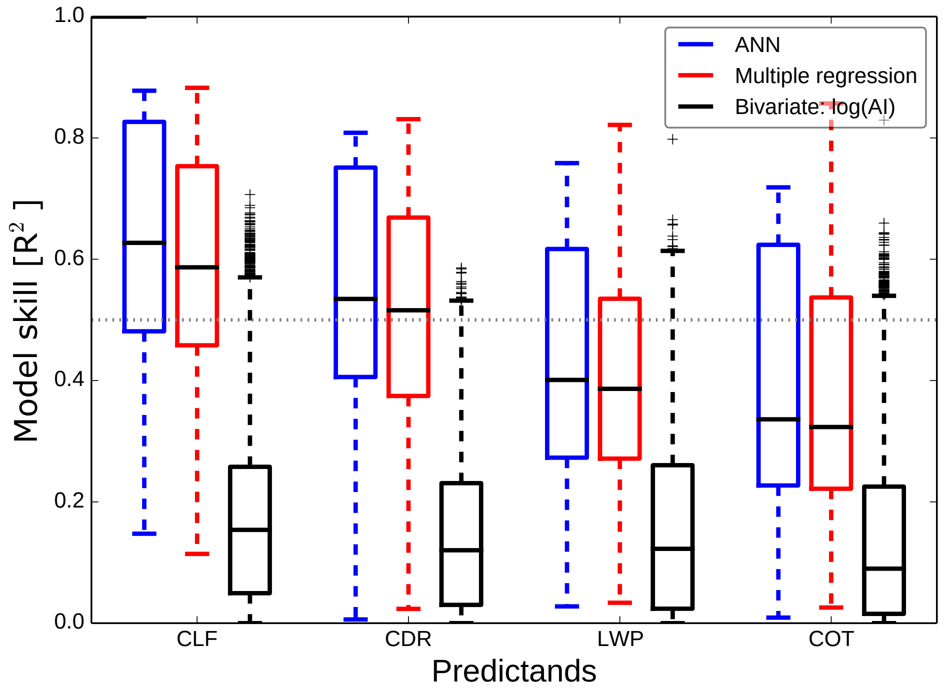

Figure 2. Predictand correlation with ANN test output, multiple linear regression (both multivariate) ~~test output~~ and ~~AOD~~ log(AI) (bivariate). The median is represented by the black horizontal line, framed by the interquartile range (boxes), whiskers expand the boxes by 1.5 interquartile ranges.

For all predictands there is a large spread in model skill, leading to distinct regional patterns as illustrated in Fig. 3. The skill of the ANNs is generally higher in the atmospherically stable regions off the western continental coastlines that are dominated by stratocumulus clouds. Less skilled ANNs can generally be found in the (sub-)tropic Pacific and the Indian Ocean.

The global spatial patterns of ANN skill are likely linked to the spatial patterns of the variability of the specific predictands (Fig. 4). A strong dependence on the variability can be noted for CLF and CDR (Fig. 4a and 4b), i.e. a higher variability enables the ANN to more skillfully represent the inherent relationships. This is sensible, as a higher predictand variability offers the ANN a stronger signal from which it can learn.

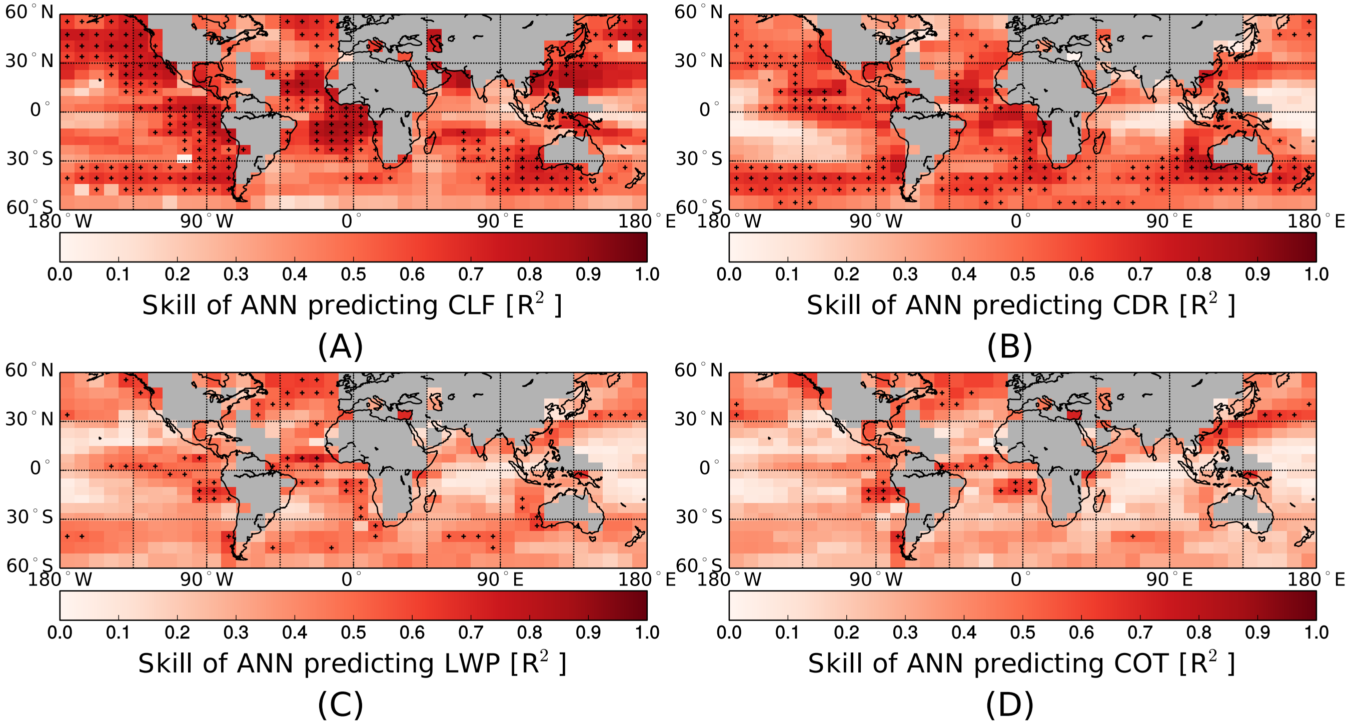

Figure 3. Global patterns of ANN skill [R^2] predicting a) cloud fraction, b) cloud droplet effective radius, c) cloud liquid water path and d) cloud optical thickness. As only ANNs with $R^2 > 0.5$ and rel. RMSE < its global average are used to compute the sensitivities, these are marked by a '+'.

3.2 Determinants of cloud occurrence and properties

Sensitivities are analyzed in all ANNs with a skill of $R^2 > 0.5$ and with a rel. RMSE that is smaller than its global average. Figure 5 shows globally summarized mean and standard deviation of all predictor sensitivities for CLF (Fig. 5a), CDR (Fig. 5b), LWP (Fig. 5c) and COT (Fig. 5d). Positive sensitivities point towards a positive response to an increase in the specific predictor while holding the other predictors constant at their regional average values. CLF shows the greatest sensitivity to LTS, where an increase in LTS leads to a strong increase in CLF, underlining the importance of LTS found in earlier studies (e.g. Klein and Hartmann, 1993; Matsui et al., 2004; Andersen and Cermak, 2015). CLF is also positively related to relative humidity at all assessed pressure levels, with the strongest sensitivity at 950 hPa, where stratocumulus clouds and transitional clouds between stratocumulus and shallow cumulus are located (Gryspeerdt and Stier, 2012; Andersen and Cermak, 2015). While boundary layer height and aerosol are also positively connected to CLF, W sensitivity varies in sign. Sensitivities associated with W can generally be interpreted as the change in the predictand when W changes from updrafts to downdrafts. The most relevant pressure level in terms of W seems to be 700 hPa, with strong positive sensitivities, illustrating that the downdrafts at 700 hPa associated with stable conditions in the lower troposphere correspond to an increase in CLF. In terms of CDR sensitivities,

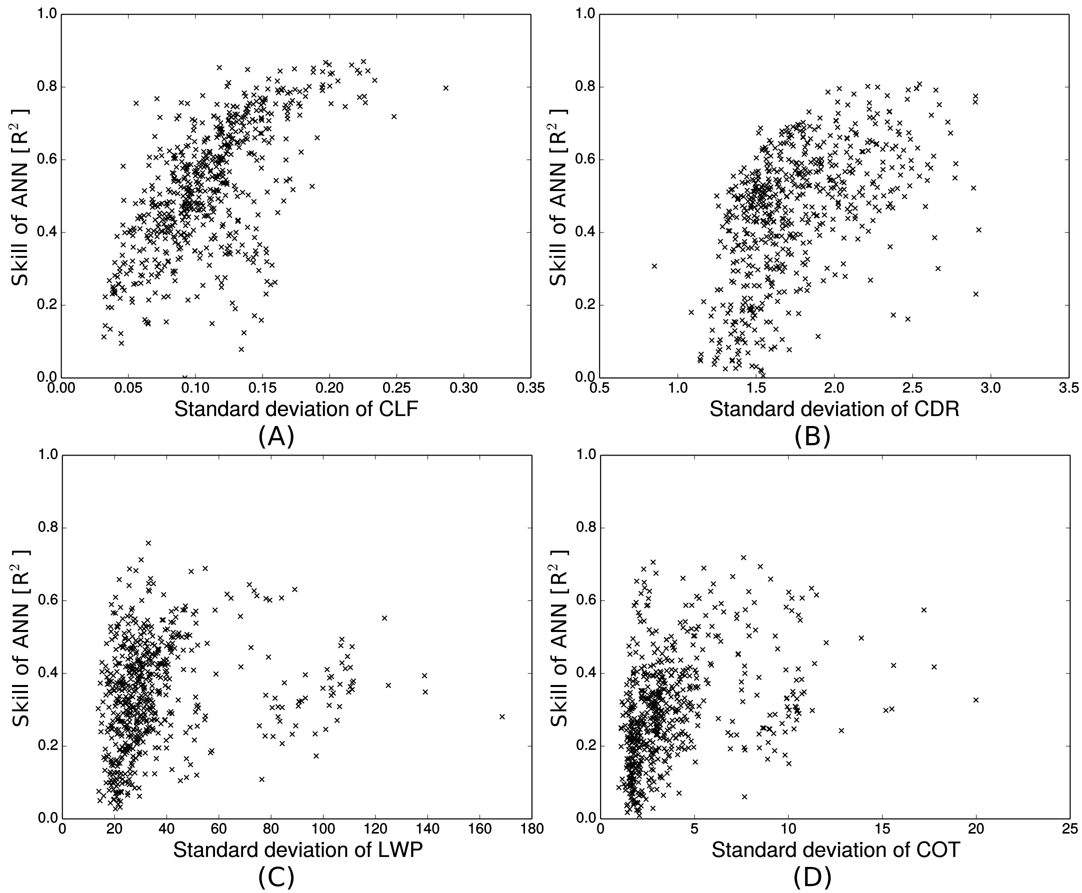

Figure 4. The skill of the ANN predicting a) CLF, b) CDR, c) COT and d) LWP as a function of the predictand variability (standard deviation). Each point illustrates the combination of skill and variability for a specific equal area region (pixel regions in figure 3).

LTS also displays the strongest effect, with an increase in LTS connected to a distinct reduction in droplet size. RH at 850 hPa exerts the strongest positive CDR sensitivity, with many of the cloud tops located at this pressure level. ~~AOD~~ AI has a notable sensitivity, showing a distinct negative association to CDR as previously assumed. Generally, updrafts favor larger CDR, with a stronger sensitivity at higher altitudes. Results of LWP and COT sensitivities are similar in terms of the signs and magnitudes of the individual sensitivities. Both are mainly determined by BLH and LTS, both positively associated with the respective cloud property. RH facilitates thicker clouds containing more liquid water, especially free tropospheric relative humidity at 700 hPa seems to have a positive impact on LWP and COT, as higher humidity levels at 700 hPa are likely to weaken drying effects of entraining air masses (Ackerman et al., 2004; Chen et al., 2012, 2014). While increases in aerosol lead to a negative LWP response, this does not lead to a similarly strong COT reduction. W is negatively related to both cloud properties, as situations with updrafts generally produce thicker clouds, the most relevant pressure level is at 850 hPa.

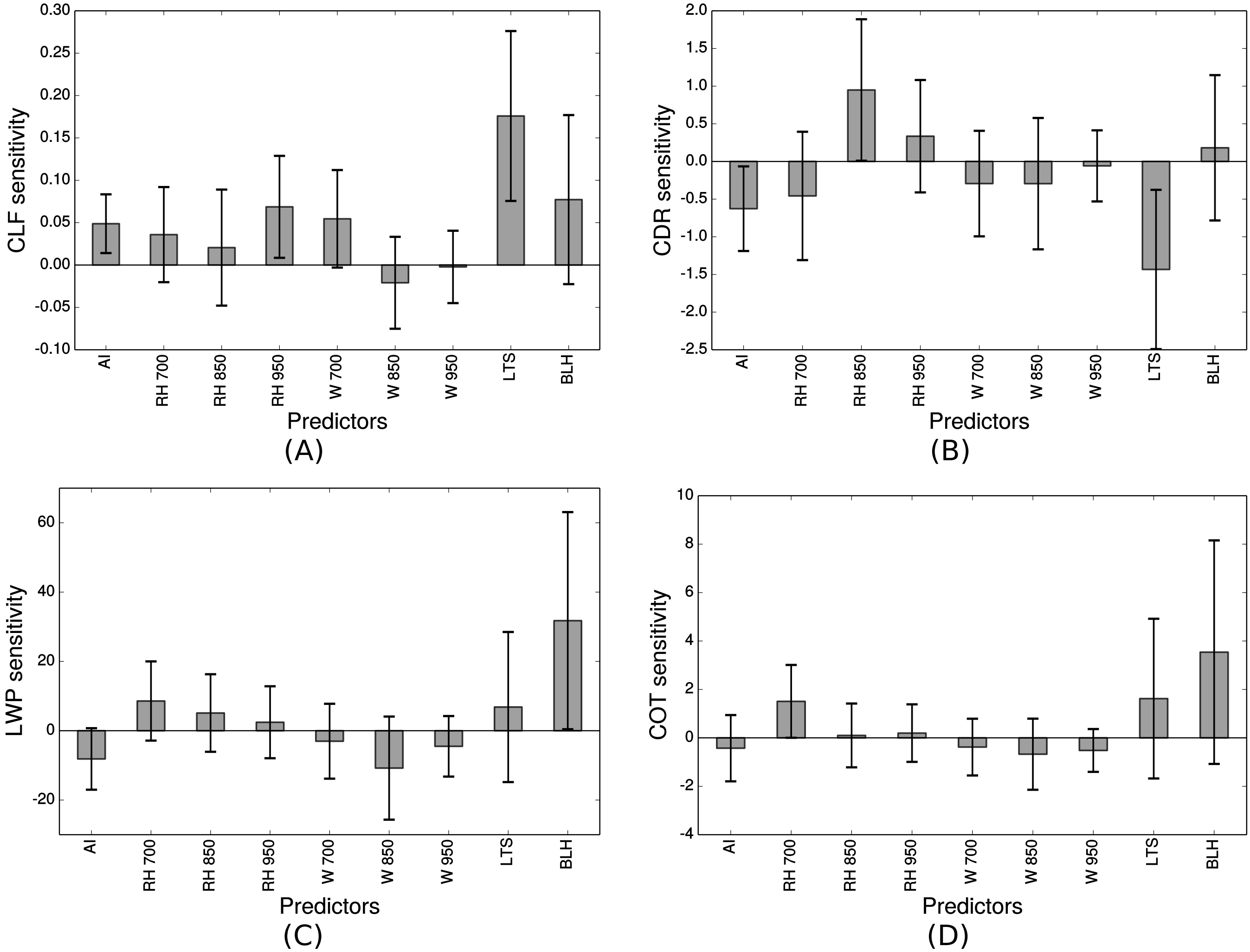

Figure 5. Global mean relative sensitivities as defined in section 2.3 of a) CLF, b) CDR, c) LWP and d) COT for all predictors of the ANNs (x axes). Error bars illustrate the regional variability of the sensitivities (global standard deviation).

The application of an individual ANN in every grid cell enables the analysis of regional patterns of the derived sensitivities. Panels of the left-hand column of Fig. 6 show regional patterns of CLF sensitivities, the panels of the right-hand column show regional patterns of CDR sensitivities. The range of the colorbars is identical within each column, so that both the overall magnitude as well as the spatial patterns of the sensitivities can be compared. LTS is the strongest determinant for CLF and is positively related to CLF everywhere on Earth, with especially strong sensitivities in atmospherically stable regions off the western coasts of the continents, where stratocumulus clouds are predominant (Klein and Hartmann, 1993; Russell et al., 2013). In these regions RH shows a strong positive CLF sensitivity at 950 hPa, pointing to the relevance of low-level humidity in these regions of low boundary layer clouds. Liquid water cloud fraction in the intertropical convergence zone is more sensitive to

RH at 850 hPa, reflecting the thicker boundary layer in this region. The most pronounced relation between the aerosol and CLF can be found at latitudes around 30°, especially over the Northwest Pacific.

CDR is markedly reduced by ~~AOD~~ AI in the Northwest Pacific and the Southwest Atlantic and negatively associated with ~~AOD~~ AI to a lesser degree in most other marine regions. The ~~regions over the Northeast Atlantic and~~ region close to the coastline of the Arabian Sea ~~are exceptions. In these regions~~ is an exception. Here, dust particles make up a significant portion of the aerosol species composition (Prospero, 1999; Kaufman et al., 2005), which may lead to larger droplet sizes when dust aerosols act as giant CCN (Levin et al., 2005; Barahona et al., 2010). The Southeast Atlantic features weakly positive sensitivities of CDR to changes in AI. While CDR is typically found to be negatively related to AI in the Southeast Atlantic (e.g. Costantino and Bréon, 2013), Andersen and Cermak (2015) found that AI and CDR can be positively associated in very stable atmospheric conditions. Issues of sampling (few aerosol retrievals in high CLF regions), scale (highly aggregated data) or their combination might affect the observed CDR sensitivity to AI in this region. One should note that sensitivity maps were also produced using AOD as a proxy for cloud condensation nuclei instead of AI. While the overall results were very similar, changes in sensitivity of CDR to the aerosol quantities were observed in the Northeast Altantic that is dominated by Saharan dust aerosols. Here, the difference between AOD and AI is substantial due to the abundance of coarse dust particles. LTS is negatively associated with CDR, especially south of 30° and in the subtropical Atlantic as found by Matsui et al. (2006), as high LTS environments are connected with weaker updrafts and a shallower boundary layer, limiting cloud droplet growth. This excludes the Southeast Atlantic, where stable conditions may trap the humidity in the boundary layer (Johnson et al., 2004; Painemal et al., 2014; Andersen and Cermak, 2015). Similar effects may occur in the Southeast Pacific as well. RH features the strongest positive CDR sensitivity at 850 hPa with distinctly strong sensitivities in the subtropic regions, where cloud tops are frequently located at this pressure level (Gryspeerdt and Stier, 2012). Compared to these factors, W at 700 hPa seems to be a relevant determinant in very selected, mostly tropical regions only.

4 Summary and conclusions

The central aim of this study was to identify and analyze the main determinants of marine liquid-water clouds and their sensitivities at the system scale. Artificial neural networks were shown capable of predicting cloud patterns on a global scale well, although ANN skill is dependent on the cloud property and its variability. Regions with a strong monthly variability such as the stratocumulus regions that feature a strong seasonal cycle are most skillfully represented.

Sensitivities were derived for all predictor-predictand combinations, revealing LTS to be the main determinant of monthly liquid-water cloud occurrence and properties. LTS is positively related to CLF on a global scale, with especially strong regional sensitivities in the subsidence regions and the mid-latitudes. In most of these regions, LTS features a strong negative sensitivity towards CDR. One exception to this negative CDR-LTS relationship is the Southeast Atlantic, where high LTS conditions may trap humidity in the boundary layer, causing larger CDR and hence a positve CDR-LTS relationship (Johnson et al., 2004; Painemal et al., 2014; Andersen and Cermak, 2015). The sensitivity of cloud properties to changes in relative humidity is dependent on both region and pressure level. CLF in regions that feature predominantly stratocumu-

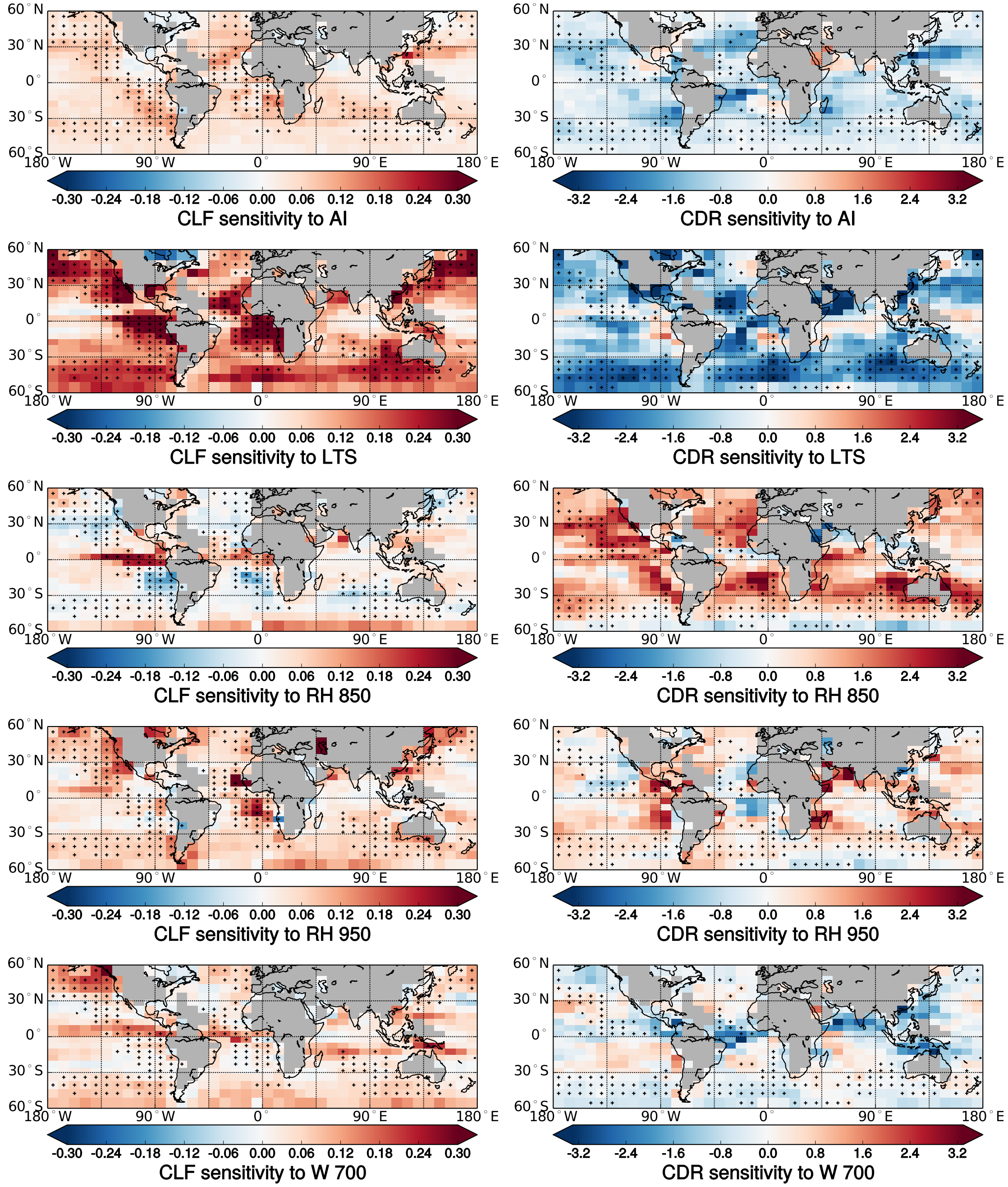

Figure 6. Global relative sensitivity patterns of selected CLF predictors in the left-hand column and CDR predictors in the right-hand column. Gray regions a) are over land or b) do not contain at least 2000 data points. Regions where the ANN test skill (R^2) is $>$ 0.5 and rel. RMSE $<$ its global average are marked with a '+'.

lus clouds or other low-level clouds is most sensitive to RH at 950 hPa, whereas tropical regions with thicker boundary layers are more sensitive to RH at higher altitudes. CDR sensitivity to RH is stronger at higher pressure levels, where the cloud-tops are likely located. In addition to this, BLH is found to be a main determinant of LWP and COT. One should note though, that not all of the observed predictor-predictand sensitivities are necessarily a result of a direct physical relationship between the predictor and the predictand, but may in part be ~~due to spurious covariations~~spurious due to cloud contamination of the satellite aerosol retrievals (Grandey et al., 2013), or due to the influence of confounding factors on both predictor and predictand (Gryspeerdt et al., 2016). For example, CLF and AI/AOD are both positively related to RH, potentially contributing to the observed positive ~~AOD-CLF sensitivity~~ CLF sensitivity to AI/AOD. Gryspeerdt et al. (2016) found that, by constraining potential aerosol-induced effects on CLF to situations where cloud droplet number concentration is simultaneously increased, the MODIS log(AOD)-CLF relationship is reduced by about 80 %. Issues of this kind are addressed here by including information on all relevant ~~parameters~~ confounding factors directly in the ANN, and - for comparison measure - when all other inputs are held constant at their grid-cell specific average, the log(AI)-CLF relationship is on average about 40 % weaker than the originally observed log(AI)-CLF relationship. While the decrease in the sensitivity is not quite as strong as in Gryspeerdt et al. (2016), the results correspond well in the sense that bivariately determined aerosol-cloud sensitivities as in Quaas et al. (2008) are likely to overestimate aerosol indirect effects significantly. While the influence of confounding factors is limited by the multivariate approach, effects concerning data quality (e.g. cloud contamination) are not accounted for and need to be considered, especially when interpreting the CLF sensitivity to AI.

The ramifications of the interactions between aerosols and cloud occurrence and properties seem to be represented well in the ANN, following the general understanding of ACI. Specific regions of interest arise, such as the Northwest Atlantic with strong sensitivities to ~~AOD~~ AI and regions that are affected by high dust loadings, with positive ~~AOD-CDR~~ AI-CDR relationships and an above average positive ~~AOD-CLF~~ AI-CLF sensitivity.

The results lead to the conclusion that on the system scale the aerosol may be viewed as a relevant determinant of marine liquid-water cloud fraction and microphysical properties, but only a secondary determinant for cloud optical thickness. On the scales considered here, lower tropospheric stability is the key controlling factor of cloud occurrence and droplet size, while boundary layer height controls the liquid water path and thus optical thickness of the cloud. The results confirm findings of previous studies that analyzed determinants of cloud properties in a more isolated manner (e.g. Klein and Hartmann, 1993; Johnson et al., 2 The results give confidence that the combination of observational and reanalysis data sets in a multivariate statistical approach is able to capture the natural variability of cloud occurrence and properties, and that meteorological and aerosol effects similar to those found in other studies can be identified in this system. In the future, a focussed, cloud-regime specific ANN approach similar to Gryspeerdt and Stier (2012) or Oreopoulos et al. (2016) could add to our system understanding. To address climate effects in a straight-forward manner, future research may also apply this study's approach to investigate the global determinants of cloud radiative effects.

5 Data availability

MODIS data used in this study were acquired as part of the NASA's Earth-Sun System Division and archived and distributed by the MODIS Adaptive Processing System (MODAPS). MODIS data were obtained from the Goddard Space Flight Center (http://ladsweb.nascom.nasa.gov/data/search.html). ECMWF ERA-Interim data used in this study were obtained from the ECMWF data server (http://apps.ecmwf.int/datasets/data/interim-full-moda/levtype=sfc/).

Author contributions. J. Cermak had the initial idea and performed a precursor study. H. Andersen fully developed the method and the software, obtained and analyzed the data sets, conducted the original research and wrote the manuscript. H. Andersen, J. Cermak, J. Fuchs, R. Knutti and U. Lohmann contributed to study design and interpretation of findings.

Competing interests. The authors declare that they have no conflict of interest.

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References

Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, Nature, 432, 1014–1017, doi:10.1038/nature03137.1., 2004.

Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227–1230, doi:10.1126/science.245.4923.1227, 1989.

Andersen, H. and Cermak, J.: How thermodynamic environments control stratocumulus microphysics and interactions with aerosols, Environmental Research Letters, 10, 24 004, doi:10.1088/1748-9326/10/2/024004, http://dx.doi.org/10.1088/1748-9326/10/2/024004, 2015.

Andersen, H., Cermak, J., Fuchs, J., and Schwarz, K.: Global observations of cloud-sensitive aerosol loadings in low-level marine clouds, Journal of Geophysical Research: Atmospheres, 121, 12 936–12 946, doi:10.1002/2016JD025614, http://doi.wiley.com/10.1002/2016JD025614, 2016.

Andreae, M. O.: Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions, Atmospheric Chemistry and Physics, 9, 543–556, doi:10.5194/acp-9-543-2009, 2009.

Barahona, D., West, R. E. L., Stier, P., Romakkaniemi, S., Kokkola, H., and Nenes, A.: Comprehensively accounting for the effect of giant CCN in cloud activation parameterizations, Atmospheric Chemistry and Physics, 10, 2467–2473, 2010.

Bennartz, R. and Rausch, J.: Global and regional estimates of warm cloud droplet number concentration based on 13 years of AQUA-MODIS observations, Atmospheric Chemistry and Physics Discussions, 2017, 1–32, doi:10.5194/acp-2016-1130, http://www.atmos-chem-phys-discuss.net/acp-2016-1130/, 2017.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S. K., Sherwood, S., Stevens, B., and Zhang, X.-Y.: Clouds and Aerosols, in: Climate Change 2013: The Physical Science Basis, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, [Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V. and Midgley, P.M. (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA., 2013.

Brenguier, J.-L., Pawlowska, H., Schüller, L., Preusker, R., Fischer, J., and Fouquart, Y.: Radiative Properties of Boundary Layer Clouds: Droplet Effective Radius versus Number Concentration, Journal of the Atmospheric Sciences, 57, 803–821, doi:10.1175/1520-0469(2000)057<0803:RPOBLC>2.0.CO;2, http://journals.ametsoc.org/doi/abs/10.1175/1520-0469(2000)057{%}3C0803:RPOBLC{%}3E2.0.CO;2, 2000.

Bréon, F.-M., Tanré, D., and Generoso, S.: Aerosol effect on cloud droplet size monitored from satellite, Science, 295, 834–838, doi:10.1126/science.1066434, http://www.ncbi.nlm.nih.gov/pubmed/11823636, 2002.

Cermak, J. and Knutti, R.: Beijing Olympics as an Aerosol Field Experiment, Geophysical Research Letters, 36, L10 806, doi:10.1029/2009GL038572, http://www.iac.ethz.ch/people/knuttir/papers/cermak09grl.pdf, 2009.

Chen, Y.-C., Christensen, M. W., Xue, L., Sorooshian, A., Stephens, G. L., Rasmussen, R. M., and Seinfeld, J. H.: Occurrence of lower cloud albedo in ship tracks, Atmospheric Chemistry and Physics, 12, 8223–8235, doi:10.5194/acp-12-8223-2012, http://www.atmos-chem-phys.net/12/8223/2012/, 2012.

Chen, Y.-C., Christensen, M. W., Stephens, G. L., and Seinfeld, J. H.: Satellite-based estimate of global aerosol-cloud radiative forcing by marine warm clouds, Nature Geoscience, 7, 643–646, doi:10.1038/ngeo2214, 2014.

Costantino, L. and Bréon, F.-M.: Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations, Atmospheric Chemistry and Physics, 13, 69–88, doi:10.5194/acp-13-69-2013, http://www.atmos-chem-phys.net/13/69/2013/, 2013.

Cybenko, G.: Approximation by superpositions of a sigmoidal function, Mathematics of Control, Signals, and Systems, 2, 303–314, doi:10.1007/BF02551274, http://link.springer.com/10.1007/BF02551274, 1989.

Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Quarterly Journal of the Royal Meteorological Society, 137, 553–597, doi:10.1002/qj.828, http://doi.wiley.com/10.1002/qj.828, 2011.

Di Noia, A., Sellitto, P., Frate, F. D., and De Laat, J.: Global tropospheric ozone column retrievals from OMI data by means of neural networks, Atmospheric Measurement Techniques, 6, 895–915, doi:10.5194/amt-6-895-2013, www.atmos-meas-tech.net/6/895/2013/, 2013.

Engström, A. and Ekman, A. M. L.: Impact of meteorological factors on the correlation between aerosol optical depth and cloud fraction, Geophysical Research Letters, 37, L18 814, doi:10.1029/2010GL044361, http://doi.wiley.com/10.1029/2010GL044361, 2010.

Funahashi, K.-I.: On the approximate realization of continuous mappings by neural networks, Neural Networks, 2, 183–192, doi:10.1016/0893-6080(89)90003-8, http://linkinghub.elsevier.com/retrieve/pii/0893608089900038, 1989.

Gardner, M. and Dorling, S.: Artificial neural networks (the multilayer perceptron) – a review of applications in the atmospheric sciences, Atmospheric Environment, 32, 2627–2636, doi:10.1016/S1352-2310(97)00447-0, 1998.

Glassmeier, F. and Lohmann, U.: Constraining precipitation susceptibility of warm, ice- and mixed-phase clouds with microphysical equations, Journal of the Atmospheric Sciences, 73, 5003–5023, doi:10.1175/JAS-D-16-0008.1, http://journals.ametsoc.org/doi/10.1175/JAS-D-16-0008.1, 2016.

Grandey, B. S., Stier, P., and Wagner, T. M.: Investigating relationships between aerosol optical depth and cloud fraction using satellite, aerosol reanalysis and general circulation model data, Atmospheric Chemistry and Physics, 13, 3177–3184, doi:10.5194/acp-13-3177-2013, 2013.

Gryspeerdt, E. and Stier, P.: Regime-based analysis of aerosol-cloud interactions, Geophysical Research Letters, 39, L21 802, doi:10.1029/2012GL053221, http://doi.wiley.com/10.1029/2012GL053221, 2012.

Gryspeerdt, E., Quaas, J., and Bellouin, N.: Constraining the aerosol influence on cloud fraction, Journal of Geophysical Research: Atmospheres, 121, 3566–3583, doi:10.1002/2015JD023744, 2016.

Hartmann, D. L., Ockert-Bell, M. E., and Michelsen, M. L.: The Effect of Cloud Type on Earth's Energy Balance: Global Analysis, Journal of Climate, 5, 1281–1304, doi:10.1175/1520-0442(1992)005<1281:TEOCTO>2.0.CO;2, 1992.

Hartmann, H., Becker, S., King, L., and Jiang, T.: Forecasting water levels at the Yangtze River with neural networks, Erdkunde, 62, 231–243, doi:10.3112/erdkunde.2008.03.04, 2008.

Hartmann, H., Snow, J. A., Su, B., and Jiang, T.: Seasonal predictions of precipitation in the Aksu-Tarim River basin for improved water resources management, Global and Planetary Change, 147, 86–96, doi:10.1016/j.gloplacha.2016.10.018, 2016.

Hornik, K., Stinchcombe, M., and White, H.: Multilayer feedforward networks are universal approximators, Neural Networks, 2, 359–366, doi:10.1016/0893-6080(89)90020-8, http://linkinghub.elsevier.com/retrieve/pii/0893608089900208, 1989.

Johnson, B., Shine, K., and Forster, P.: The semi-direct aerosol effect: Impact of absorbing aerosols on marine stratocumulus, Quarterly Journal of the Royal Meteorological Society, 130, 1407–1422, doi:10.1256/qj.03.61, http://doi.wiley.com/10.1256/qj.03.61, 2004.

Kaufman, Y. J., Koren, I., Remer, L., Rosenfeld, D., and Rudich, Y.: The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean, Proceedings of the National Academy of Sciences of the United States of America, 102, 11 207–11 212, doi:10.1073/pnas.0505191102, http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=1182178{&}tool=pmcentrez{&}rendertype=abstract, 2005.

Kecman, V. V. and Vojislav: Learning and soft computing: support vector machines, neural networks, and fuzzy logic models, MIT Press, http://dl.acm.org/citation.cfm?id=559026, 2001.

Klein, S. A. and Hartmann, D. L.: The Seasonal Cycle of Low Stratiform Clouds, Journal of Climate, 6, 1587–1606, doi:10.1175/1520-0442(1993)006<1587:TSCOLS>2.0.CO;2, 1993.

Koren, I., Feingold, G., and Remer, L. A.: The invigoration of deep convective clouds over the Atlantic: Aerosol effect, meteorology or retrieval artifact?, Atmospheric Chemistry and Physics, 10, 8855–8872, doi:10.5194/acp-10-8855-2010, 2010.

Koren, I., Altaratz, O., Remer, L. A., Feingold, G., Martins, J. V., and Heiblum, R. H.: Aerosol-induced intensification of rain from the tropics to the mid-latitudes, Nature Geoscience, 5, 118–122, doi:10.1038/ngeo1364, http://www.nature.com/doifinder/10.1038/ngeo1364, 2012.

Koren, I., Dagan, G., and Altaratz, O.: From aerosol-limited to invigoration of warm convective clouds, Science, 344, 1143–1146, doi:10.1126/science.1252595, http://www.sciencemag.org/content/344/6188/1143.full, 2014.

Latham, J., Rasch, P., Chen, C.-C., Kettles, L., Gadian, A., Gettelman, A., Morrison, H., Bower, K., and Choularton, T.: Global temperature stabilization via controlled albedo enhancement of low-level maritime clouds, Philosophical Transactions of the Royal Society of London A: Mathematical, Physical and Engineering Sciences, 366, 3969–3987, doi:10.1098/rsta.2008.0137, file:///Users/yangyang/YANGYANGXU/work/lynn{_}research/papersfromlynn/Lathametal.-2008-Philosophicaltransactions.SeriesA, Mathematical,physical,andengineeringsciences-Globaltemperaturestabilizationviacontrolledalbedoenhancementoflow, 2008.

Le, J., El-Askary, H., Allali, M., and Struppa, D.: Application of recurrent neural networks for drought projections in California, Atmospheric Research, 188, 100–106, doi:10.1016/j.atmosres.2017.01.002, http://linkinghub.elsevier.com/retrieve/pii/S0169809517300157, 2017.

Levin, Z., Teller, A., Ganor, E., and Yin, Y.: On the interactions of mineral dust, sea-salt particles, and clouds: A measurement and modeling study from the Mediterranean Israeli Dust Experiment campaign, Journal of Geophysical Research: Atmospheres, 110, 1–19, doi:10.1029/2005JD005810, 2005.

Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The Collection 6 MODIS aerosol products over land and ocean, Atmospheric Measurement Techniques, 6, 2989–3034, doi:10.5194/amt-6-2989-2013, http://www.atmos-meas-tech.net/6/2989/2013/amt-6-2989-2013.html, 2013.

Loeb, N. G. and Schuster, G. L.: An observational study of the relationship between cloud, aerosol and meteorology in broken low-level cloud conditions, Journal of Geophysical Research: Atmospheres, 113, D14 214, doi:10.1029/2007JD009763, http://doi.wiley.com/10.1029/2007JD009763, 2008.

Matsui, T., Masunaga, H., and Pielke Sr., R. A.: Impact of aerosols and atmospheric thermodynamics on cloud properties within the climate system, Geophysical Research Letters, 31, L06 109, doi:10.1029/2003GL019287, http://doi.wiley.com/10.1029/2003GL019287, 2004.

Matsui, T., Masunaga, H., Kreidenweis, S. M., Pielke, R. A., Tao, W.-K., Chin, M., and Kaufman, Y. J.: Satellite-based assessment of marine low cloud variability associated with aerosol, atmospheric stability, and the diurnal cycle, Journal of Geophysical Research: Atmospheres, 111, D17 204, doi:10.1029/2005JD006097, http://doi.wiley.com/10.1029/2005JD006097, 2006.

McComiskey, A. and Feingold, G.: The scale problem in quantifying aerosol indirect effects, Atmospheric Chemistry and Physics, 12, 1031–1049, doi:10.5194/acp-12-1031-2012, http://www.atmos-chem-phys.net/12/1031/2012/acp-12-1031-2012.html, 2012.

McComiskey, A., Feingold, G., Frisch, a. S., Turner, D. D., Miller, M. a., Chiu, J. C., Min, Q., and Ogren, J. a.: An assessment of aerosol-cloud interactions in marine stratus clouds based on surface remote sensing, Journal of Geophysical Research: Atmospheres, 114, D09 203, doi:10.1029/2008JD011006, http://doi.wiley.com/10.1029/2008JD011006, 2009.

Nakajima, T., Higurashi, A., Kawamoto, K., and Penner, J. E.: A possible correlation between satellite-derived cloud and aerosol microphysical parameters, Geophysical Research Letters, 28, 1171–1174, doi:10.1029/2000GL012186, 2001.

Olden, J. D. and Jackson, D. A.: Illuminating the "black box": A randomization approach for understanding variable contributions in artificial neural networks, Ecological Modelling, 154, 135–150, doi:10.1016/S0304-3800(02)00064-9, 2002.

Oreopoulos, L., Cho, N., Lee, D., and Kato, S.: Radiative effects of global MODIS cloud regimes, Journal of Geophysical Research: Atmospheres, 121, 2299–2317, doi:10.1002/2015JD024502, 2016.

Painemal, D., Kato, S., and Minnis, P.: Boundary layer regulation in the southeast Atlantic cloud microphysics during the biomass burning season as seen by the A-train satellite constellation, Journal of Geophysical Research: Atmospheres, 119, 11 288–11 302, doi:10.1002/2014JD022182, http://doi.wiley.com/10.1002/2014JD022182, 2014.

Peters, K., Stier, P., Quaas, J., and Graßl, H.: Aerosol indirect effects from shipping emissions: sensitivity studies with the global aerosol-climate model ECHAM-HAM, Atmospheric Chemistry and Physics, 12, 5985–6007, doi:10.5194/acp-12-5985-2012, http://www.atmos-chem-phys.net/12/5985/2012/, 2012.

Pincus, R. and Baker, M. B.: Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer, Nature, 372, 250–252, doi:10.1038/372250a0, 1994.

Prospero, J. M.: Long-range transport of mineral dust in the global atmosphere: impact of African dust on the environment of the southeastern United States, Proceedings of the National Academy of Sciences of the United States of America, 96, 3396–403, doi:10.1073/PNAS.96.7.3396, http://www.ncbi.nlm.nih.gov/pubmed/10097049http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=PMC34280, 1999.

Quaas, J., Boucher, O., Bellouin, N., and Kinne, S.: Satellite-based estimate of the direct and indirect aerosol climate forcing, Journal of Geophysical Research, 113, D05 204, doi:10.1029/2007JD008962, 2008.

Quaas, J., Ming, Y., Menon, S., Takemura, T., Wang, M., Penner, J. E., Gettelman, A., Lohmann, U., Bellouin, N., Boucher, O., Sayer, A. M., Thomas, G. E., McComiskey, A., Feingold, G., Hoose, C., Kristjánsson, J. E., Liu, X., Balkanski, Y., Donner, L. J., Ginoux, P. A., Stier, P., Feichter, J., Sednev, I., Bauer, S. E., Koch, D., Grainger, R. G., Kirkevåg, A., Iversen, T., Seland, Ø., Easter, R., Ghan, S. J., Rasch, P. J., Morrison, H., Lamarque, J., Iacono, M. J., Kinne, S., and Schulz, M.: Aerosol indirect effects - general circulation model intercomparison and evaluation with satellite data, Atmospheric Chemistry and Physics, 9, 8697–8717, doi:10.5194/acp-9-8697-2009, 2009.

Quaas, J., Stevens, B., Stier, P., and Lohmann, U.: Interpreting the cloud cover - aerosol optical depth relationship found in satellite data using a general circulation model, Atmospheric Chemistry and Physics, 10, 6129–6135, http://www.atmos-chem-phys.net/10/6129/2010/, 2010.

Russell, L. M., Sorooshian, A., Seinfeld, J. H., Albrecht, B. A., Nenes, A., Ahlm, L., Chen, Y.-C., Coggon, M., Craven, J. S., Flagan, R. C., Frossard, A. A., Jonsson, H., Jung, E., Lin, J. J., Metcalf, A. R., Modini, R., Mülmenstädt, J., Roberts, G., Shingler, T., Song, S., Wang, Z., and Wonaschütz, A.: Eastern Pacific Emitted Aerosol Cloud Experiment, Bulletin of the American Meteorological Society, 94, 709–729, doi:10.1175/BAMS-D-12-00015.1, http://journals.ametsoc.org/doi/abs/10.1175/BAMS-D-12-00015.1, 2013.

Shinozuka, Y., Clarke, A. D., Nenes, A., Jefferson, A., Wood, R., McNaughton, C. S., Ström, J., Tunved, P., Redemann, J., Thornhill, K. L., Moore, R. H., Lathem, T. L., Lin, J. J., and Yoon, Y. J.: The relationship between cloud condensation nuclei (CCN) concentration and light extinction of dried particles: indications of underlying aerosol processes and implications for satellite-based CCN estimates, Atmospheric Chemistry and Physics, 15, 7585–7604, doi:10.5194/acp-15-7585-2015, http://www.atmos-chem-phys.net/15/7585/2015/, 2015.

Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461, 607–613, doi:10.1038/nature08281, http://www.ncbi.nlm.nih.gov/pubmed/19794487, 2009.

Stier, P.: Limitations of passive remote sensing to constrain global cloud condensation nuclei, Atmospheric Chemistry and Physics, 16, 6595–6607, doi:10.5194/acp-16-6595-2016, http://www.atmos-chem-phys.net/16/6595/2016/, 2016.

Su, W., Loeb, N. G., Xu, K.-M., Schuster, G. L., and Eitzen, Z. A.: An estimate of aerosol indirect effect from satellite measurements with concurrent meteorological analysis, Journal of Geophysical Research: Atmospheres, 115, D18 219, doi:10.1029/2010JD013948, 2010.

Twomey, S.: The Influence of Pollution on the Shortwave Albedo of Clouds, Journal of the Atmospheric Sciences, 34, 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2, 1977.

Werbos, P.: Backpropagation through time: what it does and how to do it, Proceedings of the IEEE, 78, 1550–1560, doi:10.1109/5.58337, http://ieeexplore.ieee.org/document/58337/, 1990.

Wood, R.: Stratocumulus Clouds, Monthly Weather Review, 140, 2373–2423, doi:10.1175/MWR-D-11-00121.1, 2012.