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# Limitations of passive satellite remote sensing to constrain global cloud condensation nuclei

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## Abstract

Aerosol–cloud interactions are considered a key uncertainty in our understanding of climate change (Boucher et al., 2013). Knowledge of the global abundance of aerosols suitable to act as cloud condensation nuclei (CCN) is fundamental to determine the strength of the anthropogenic climate perturbation. Direct measurements are limited and sample only a very small fraction of the globe so that remote sensing from satellites and ground based instruments is widely used as a proxy for cloud condensation nuclei (Nakajima et al., 2001; Andreae, 2009; Clarke and Kapustin, 2010; Boucher et al., 2013). However, the underlying assumptions cannot be robustly tested with the small number of measurements available so that no reliable global estimate of cloud condensation nuclei exists. This study overcomes this limitation using a fully self-consistent global model (ECHAM-HAM) of aerosol radiative properties and cloud condensation nuclei. An analysis of the correlation of simulated aerosol radiative properties and cloud condensation nuclei reveals that common assumptions about their relationships are violated for a significant fraction of the globe: 71 % of the area of the globe shows correlation coefficients between  $CCN_{0.2\%}$  at cloud base and aerosol optical depth (AOD) below 0.5, i.e. AOD variability explains only 25 % of the CCN variance. This has significant implications for satellite based studies of aerosol–cloud interactions. The findings also suggest that vertically resolved remote sensing techniques, such as satellite-based high spectral resolution lidars, have a large potential for global monitoring of cloud condensation nuclei.

## 1 Introduction

Aerosol–cloud interactions play an important role in the global climate system through modification of aerosol and cloud properties and abundance (Boucher et al., 2013; Twomey, 1974; Albrecht, 1989; Lohmann and Feichter, 2005). The activation of suitable aerosols (cloud condensation nuclei, CCN) to cloud droplets is the primary aerosol

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effect on warm clouds (and ice or mixed-phase clouds initiated from the liquid phase). Knowledge of the global abundance of aerosols suitable to act as cloud condensation nuclei is fundamental to determine the strength of the anthropogenic perturbation causing the radiative effect of aerosol–cloud interactions. Most estimates of the effect of aerosol–cloud interactions on the global radiation balance rely on global aerosol models. However, large uncertainties associated with the representation of clouds and aerosol effects on cloud microphysics and dynamics in current climate models (Boucher et al., 2013; Stevens and Feingold, 2009) demand for independent observational constraints. Unfortunately, direct observations of CCN are spatio-temporally sparse (Andreae, 2009; Spracklen et al., 2011) and provide insufficient constraints on their global distribution. Consequently, satellite retrieved aerosol radiative properties, such as aerosol optical depth (AOD), have been widely used as proxy for CCN in satellite based studies of aerosol–cloud interactions (Kaufman and Nakajima, 1993; Kaufman et al., 2005; Rosenfeld et al., 2008; Grandey and Stier, 2010; Boucher et al., 2013; Gryspeerdt et al., 2014).

Assuming identical size, shape, composition and humidity, CCN concentrations at fixed supersaturation are linearly related to aerosol light extinction, so that AOD, the column integrated aerosol extinction, could be expected to provide a first order proxy for CCN. However, for realistic aerosol distributions extinction and CCN concentrations are non-linearly related to size, complicating the retrieval of CCN based on extinction measurements (Ghan and Collins, 2004; Kapustin et al., 2006). It has been suggested from theory and an analysis of satellite retrievals (Nakajima et al., 2001) that aerosol index (Deuze et al., 2001)

$$AI = AOD \times \alpha \quad (1)$$

where the Ångström parameter

$$\alpha = - \frac{\ln(AOD_{\lambda_1} / AOD_{\lambda_2})}{\ln(\lambda_1 / \lambda_2)} \quad (2)$$













unchanged when using the non-parametric Spearman's rank correlation coefficient (Fig. 7e).

### 3 Results

The ECHAM-HAM simulated annual-mean surface CCN concentrations (Fig. 1) show distinct land–sea contrast, with maxima over the main aerosol source areas. CCN concentrations at the lower 0.2% supersaturation (activating only the larger particles of the CCN spectrum into cloud droplets) are lower than at the higher supersaturation of 1.5% (activating also smaller particles of the CCN spectrum).

The corresponding annual-mean AOD (Fig. 2a) shows similar maxima in the main aerosol source areas. However, it also shows high values over the sea-salt aerosol dominated storm track regions, dust source regions and generally higher levels downwind of the source areas (mainly because AOD is a column integrated quantity while (Fig. 1) shows surface CCN values). AI, giving lower weight to large particles, is in better spatial agreement with the annual mean CCN distribution than AOD (Fig. 2b).

These results confirm the common understanding that CCN are related to AOD and (better) AI for large spatial scales and long averaging periods. This is also confirmed in the fit of regional annual mean AOD and CCN pairs (Fig. 4a) for the main continents defined in Fig. 3.

The derived fit of the mean values ( $y = 0.0002x^{1.074}$  for  $\text{CCN}_{0.5\%}$ ) is statistically robust ( $r^2 = 0.70$ ) and compares to a fit ( $y = 0.0027x^{0.640}$  for  $\text{CCN}_{0.4\%}$ ,  $r^2 = 0.88$ ) of campaign mean co-located surface based CCN measurements and AERONET sun-photometer retrievals of AOD (Andreae, 2009). The inclusion of oceanic regions dominated by large sea salt particles with high extinction per particle, deteriorates the relationship of AOD and  $\text{CCN}_{0.2\%}$  (Fig. 4b) and  $r^2$  decreases from 0.65 to 0.47. The fit for land and ocean regions combined (Fig. 4c) improves using AI instead of AOD ( $r^2 = 0.84$ ).

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significantly stronger than the correlation of CCN at each layer with vertically integrated AOD (red). The extinction coefficient is temporally well correlated with relative humidity throughout the troposphere (pink). The correlation of vertically integrated AOD with CCN at cloud base is generally comparable to the correlation with surface CCN. For the Indian region, surface level CCN are even anti-correlated with vertically integrated AOD. Here, surface level extinction is strongly correlated with relative humidity (pink), while surface level CCN are efficiently removed by scavenging during high relative humidity events associated with strong precipitation. Over Europe and North America, correlations of column integrated AOD and surface CCN are generally intermediate to high, in particular north of the sub-tropical subsidence areas. Correlations of extinction coefficients with relative humidity are higher over Europe than over North American and particularly than over South America, where the predominant carbonaceous aerosols take up relatively little water. Interestingly, correlations of extinction coefficients with relative humidity are also low for the Southern Ocean region, despite the fact that the dominant (by extinction) sea salt aerosol is highly hygroscopic. This is likely due to the fact that not only aerosol water uptake but also aerosol removal via scavenging is positively correlated to relative humidity (via clouds and precipitation). This hypothesis is supported by the drop off of this correlation around and below cloud base (green line). However, correlations of column integrated AOD and surface CCN are consistently high for this region as well as for the northern high-latitude oceans.

These results suggest that vertically integrated aerosol radiative properties, as retrieved from satellite imagers, are of limited suitability as proxy for global surface or cloud base CCN: 71 % of the area of the globe shows correlation coefficients between  $CCN_{0.2\%}$  at cloud base and AOD below 0.5 (i.e. AOD variability explains only 25 % of the CCN variance). The fractional area of  $r < 0.5$  increases to 83 and 96 % for  $CCN_{0.5\%}$  and  $CCN_{1.5\%}$ , respectively. Corresponding areas for  $r < 0.5$  between CCN and aerosol index are somewhat lower (52, 66, 91 % for  $CCN_{0.2\%}$ ,  $CCN_{0.5\%}$ ,  $CCN_{1.5\%}$ , respectively, Fig. 8).

Could vertically resolved aerosol radiative properties, e.g. from space-born lidars, provide stronger constraints on CCN and ultimately the radiative effect of aerosol cloud interactions?

The correlation of surface CCN with surface aerosol extinction coefficients (AEC) (Fig. 9a) is significantly improved for most of the globe. Correlations further improve for surface extinction aerosol index  $AI_{AEC}$  (Fig. 9b) with  $r > 0.8$  for most of the globe.

Note that also correlations between surface layer CCN and  $AI_{AEC}$  deteriorate for higher supersaturations (sampling smaller particles of the aerosol size distribution), as expected from Mie theory, as the smaller particles contribute less to total extinction (Fig. 10). This is particularly evident over the continents with significant primary fine mode aerosol emissions.

## 4 Conclusions

Direct measurements of cloud condensation nuclei are limited and sample only a very small fraction of the globe so that remote sensing from satellites and ground based instruments is widely used as a proxy for cloud condensation nuclei. However, the underlying assumptions cannot be robustly tested with the small number of measurements available so that no reliable global estimate of cloud condensation nuclei exists.

This study overcomes this limitation using a fully self-consistent global model (ECHAM-HAM) of aerosol radiative properties and cloud condensation nuclei.

An analysis of the correlation of simulated aerosol radiative properties and cloud condensation nuclei confirms findings from earlier work that continental mean CCN are related to AOD ( $r^2 = 0.65$ ) for large spatial scales and long averaging periods but  $r^2$  drops to 0.47 when oceanic regions are included. Use of AI improves the goodness of fit, including oceanic regions, to  $r^2 = 0.84$ .

The mean goodness of fit for CCN and AI pairs over continental and oceanic regions deteriorates from  $r^2 = 0.57$  to  $r^2 = 0.46$  and  $r^2 = 0.41$  varying the averaging period from monthly via daily to 6 h instantaneous data.

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interactions possible. Such simulations should be increasingly used to test common assumptions in the assessment of aerosol cloud interactions from space (e.g. Gryspeerdt et al., 2015).

The findings in this work have important implications for satellite based studies of aerosol–cloud interactions. They suggest that vertically resolved remote sensing techniques, such as satellite-based high-spectral resolution lidars as ATLID on the ESA/JAXA EarthCare satellite, have a large potential for global monitoring of cloud condensation nuclei. The additional improvement in correlations using the dual-wavelength extinction measurements in AI, suggests that multi-wavelength high-spectral resolution lidars, such as the NASA airborne HSRL (McPherson et al., 2010), could further advance observational constraints on CCN from space.

While the sparse sampling of lidars from space (the CALIOP space-born lidar, Winker et al., 2009, samples the globe sparsely in 16 days, in comparison to e-folding aerosol lifetimes ranging from about 1/2 day for sea salt to 7 days for black carbon, Textor et al., 2006) may introduce sampling errors, these could be potentially mitigated through synergistic retrievals with co-located imaging radiometers. Ultimately, the assimilation into global aerosol models may provide the best observationally constrained dataset of global cloud condensation nuclei.

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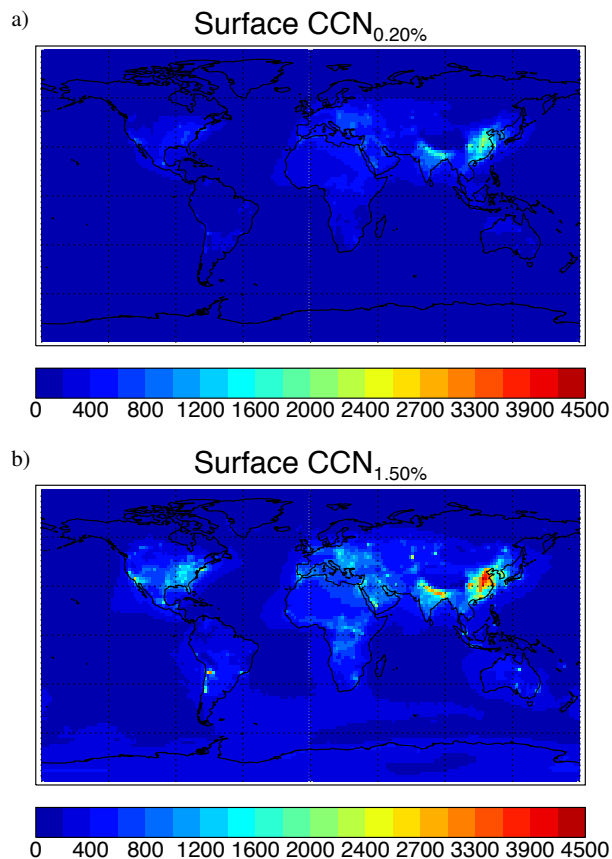
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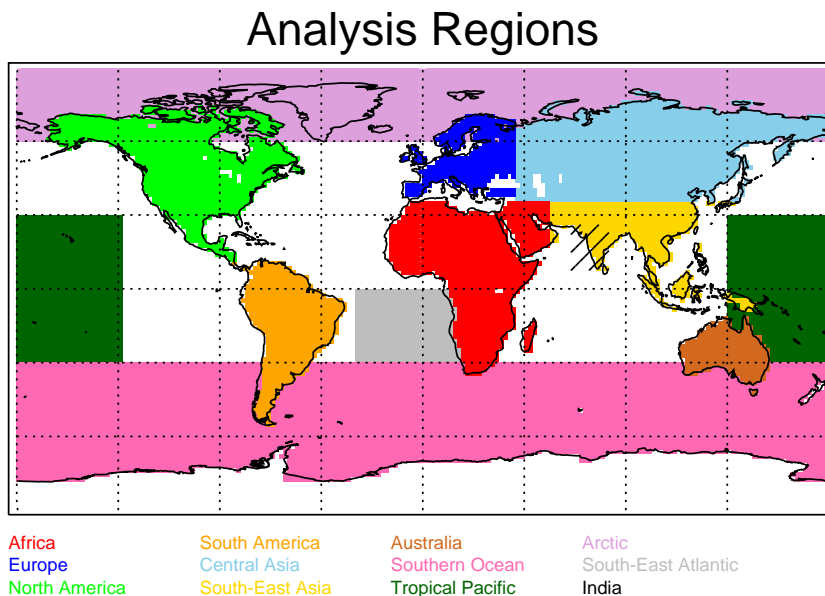


**Figure 1.** Annual-mean simulated surface cloud condensation nuclei concentrations [ $\text{cm}^{-3}$ ] at **(a)** 0.2 % and **(b)** 1.5 % supersaturation.



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**Figure 3.** Map of regions used in the analysis.

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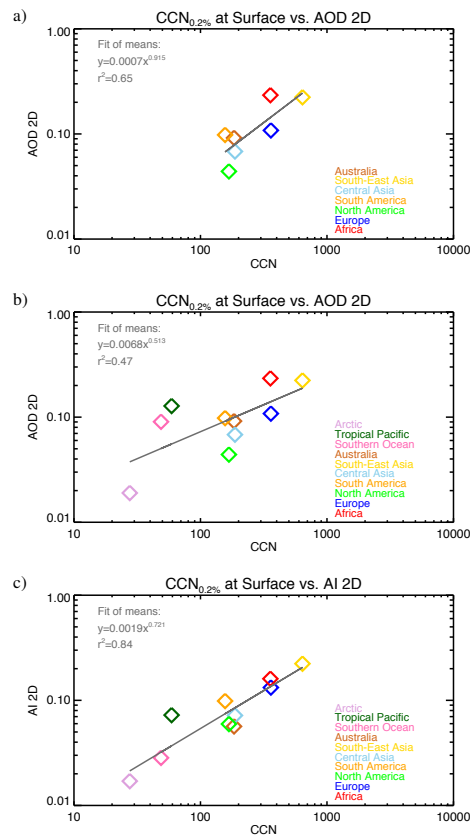
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**Figure 4.** (a) Annual continental mean AOD [1] as function of CCN<sub>0.2%</sub> [cm<sup>-3</sup>] and their fit derived from linear regression (gray), (b) as (a) but including three ocean regions, (c) annual continental mean simulated AI as function of CCN<sub>0.2%</sub> for continental and ocean regions as in (b); Regional colour coding as in Fig. 3.

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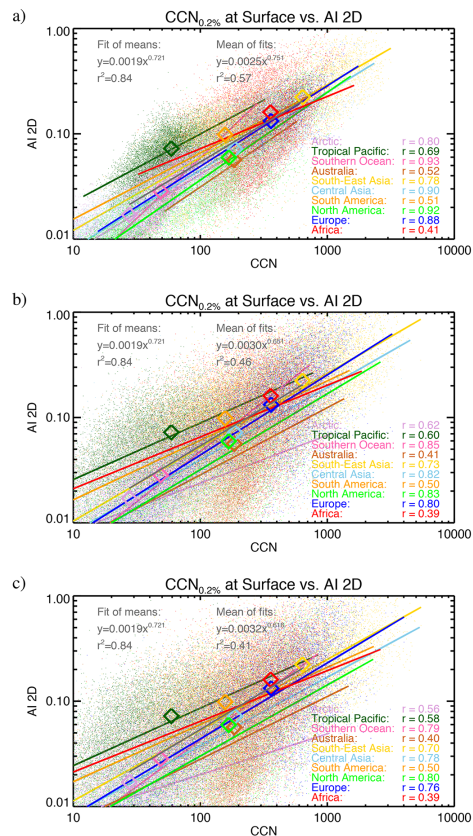
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**Figure 5.** Annual continental mean AI [1] as function of  $\text{CCN}_{0.2\%}$  [ $\text{cm}^{-3}$ ] (symbols) and their fit derived from linear regression (gray); overlay of (a) monthly mean, (b) daily mean and (c) instantaneous 6 hourly pairs of AI and  $\text{CCN}_{0.2\%}$  (scatter) and their fit derived from linear regression. For visualisation, data in scatterplot randomly sub-sampled to 10 000 pairs. Regional colour coding as in Fig. 3.

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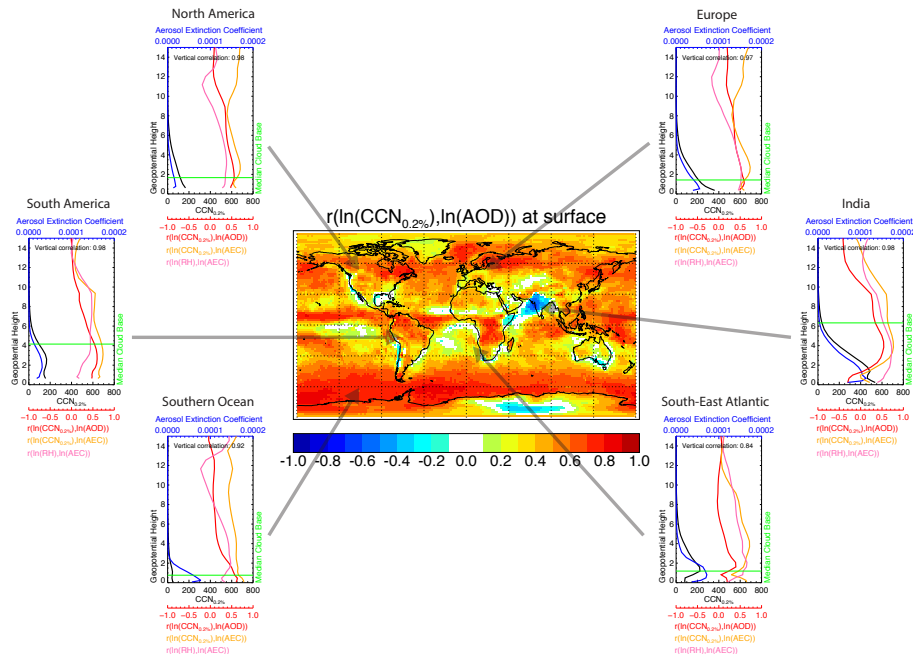
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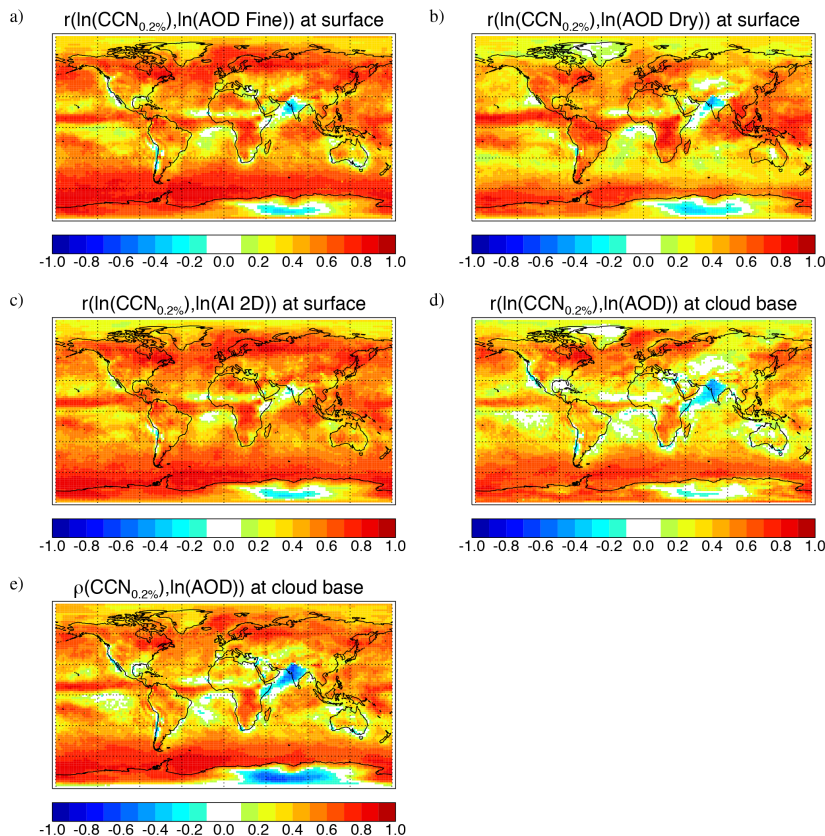
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**Figure 6.** Map of Pearson's correlation coefficient  $r$  of surface  $\ln(\text{CCN}_{0.2\%})$  with column-integrated aerosol optical depth  $\ln(\text{AOD})$  calculated for each model grid box from one year of 6 hourly pairs. Annual-mean vertical profiles of  $\text{CCN}_{0.2\%}$  [ $\text{cm}^{-3}$ ] (black), aerosol extinction coefficient (AEC) [ $\text{m}^{-1}$ ] (blue), profile of temporal correlation of  $\ln(\text{CCN}_{0.2\%})$  with column integrated  $\ln(\text{AOD})$  (red), profile of temporal correlation of  $\ln(\text{CCN}_{0.2\%})$  with vertically resolved extinction coefficient  $\ln(\text{AEC})$  (orange) and temporal correlation of  $\ln(\text{RH})$  with vertically resolved extinction coefficient  $\ln(\text{AEC})$  (pink). Also shown is the model median stratiform cloud base for each region (green) – note that this corresponds to the lowest detrainment level in regions dominated by convection, such as India. Regions defined as in Fig. 3.

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**Figure 7.** Map of Pearson's correlation coefficient of  $\text{CCN}_{0.2\%}$  with aerosol radiative properties for **(a)** surface  $\text{CCN}_{0.2\%}$  with vertically integrated fine mode aerosol optical depth, **(b)** surface  $\text{CCN}_{0.2\%}$  with vertically integrated dry aerosol optical depth, **(c)** surface  $\text{CCN}_{0.2\%}$  with vertically integrated AI, **(d)**  $\text{CCN}_{0.2\%}$  sampled at cloud base with vertically integrated AOD and **(e)** map of Spearman's rank correlation coefficient for  $\text{CCN}_{0.2\%}$  sampled at cloud base with vertically integrated AOD.

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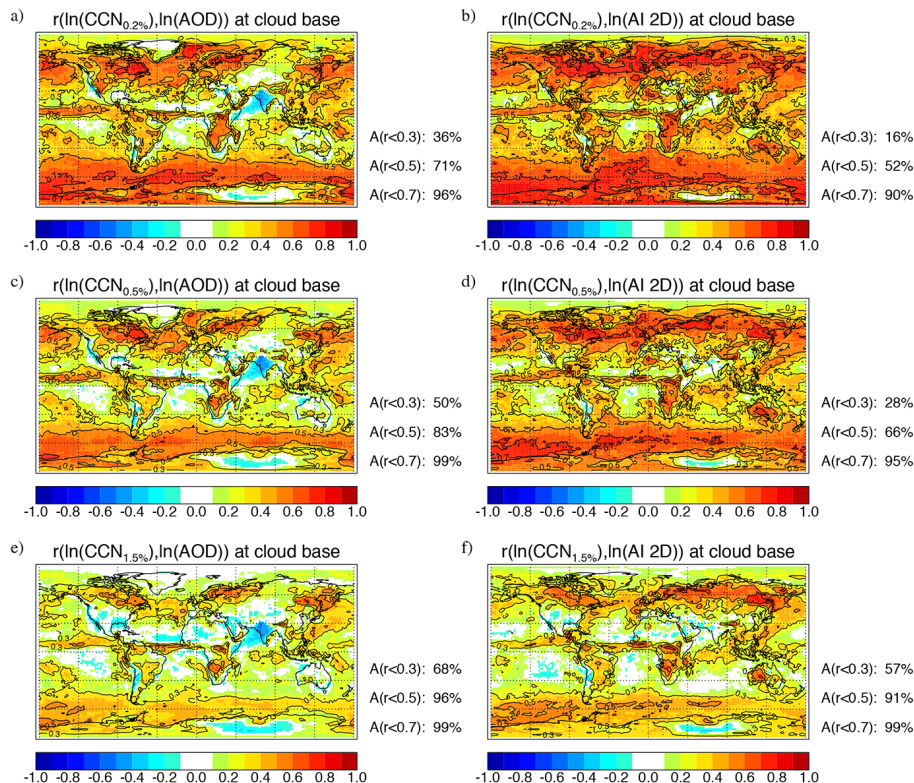
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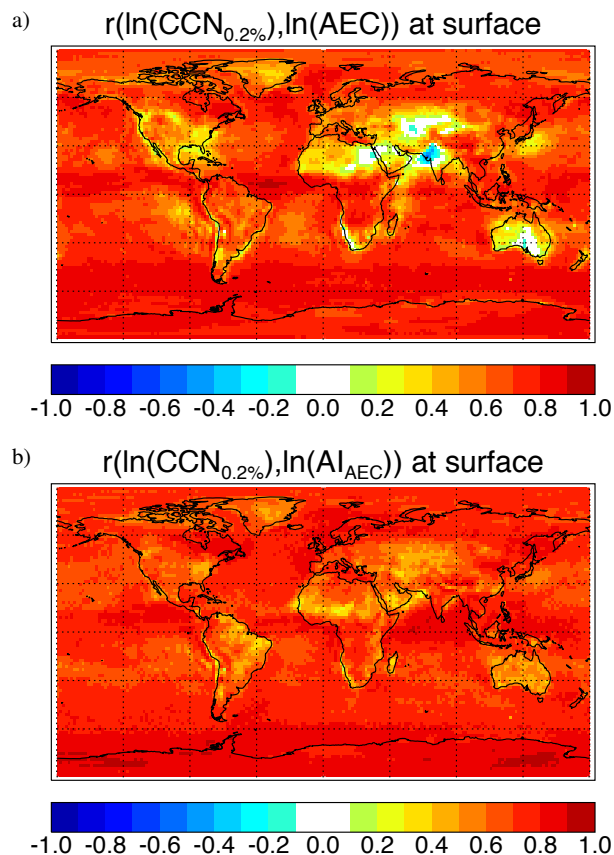
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**Figure 8.** Map of Pearson's correlation coefficient of cloud base CCN with aerosol radiative properties for **(a)**  $\text{CCN}_{0.2\%}$  with vertically integrated aerosol optical depth, **(b)**  $\text{CCN}_{0.2\%}$  with vertically integrated aerosol index, **(c)**  $\text{CCN}_{0.5\%}$  with vertically integrated aerosol optical depth, **(d)**  $\text{CCN}_{0.5\%}$  with vertically integrated aerosol index, **(e)**  $\text{CCN}_{1.5\%}$  with vertically integrated aerosol optical depth and **(f)**  $\text{CCN}_{1.5\%}$  with vertically integrated aerosol index. Fractional area (A) of the globe with  $r < 0.3, 0.5, 0.7$ .

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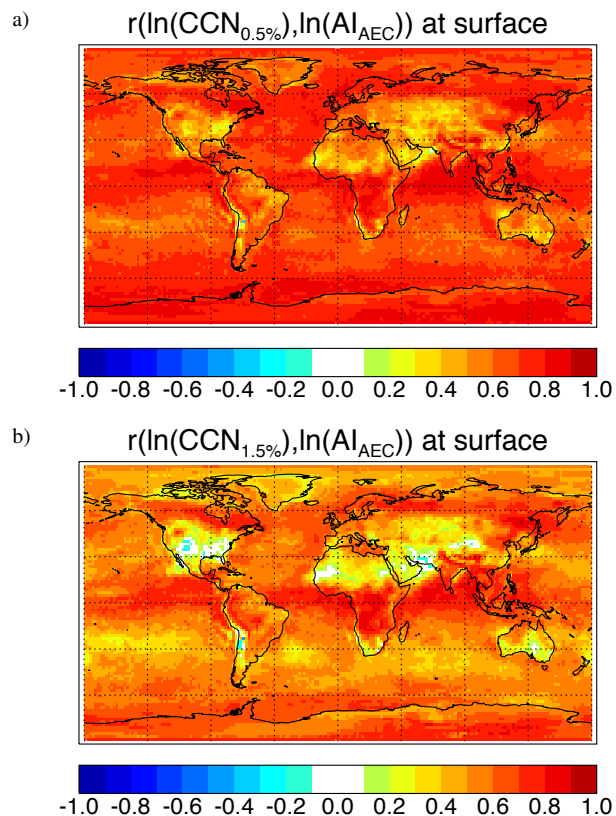
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**Figure 9.** Map of Pearson's correlation coefficient of CCN with vertically resolved aerosol radiative properties: **(a)** surface  $\ln(\text{CCN}_{0.2\%})$  with surface  $\ln(\text{AEC})$  and **(b)** surface  $\ln(\text{CCN}_{0.2\%})$  with surface  $\ln(\text{AEC}-\text{AI})$  calculated for each model grid box from one year of 6 hourly pairs.

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**Figure 10.** Map of Pearson's correlation coefficient of surface layer  $\ln(\text{AI}_{\text{AEC}})$  with  $\ln(\text{CCN})$  at higher supersaturations: **(a)**  $\text{CCN}_{0.5\%}$ , **(b)**  $\text{CCN}_{1.5\%}$ .