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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 $\mathrm{CCN}_{0.2\%}$ 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)

$$\mathrm{AI} = \mathrm{AOD} \times \alpha \tag{1}$$

where the Ångström parameter

$$\alpha = -\frac{\ln(\mathrm{AOD}_{\lambda_1}/\mathrm{AOD}_{\lambda_2})}{\ln(\lambda_1/\lambda_2)} \tag{2}$$

provides a superior proxy of CCN, as it gives lower weight to large (low α) aerosol and reduces the impact of large but low number-concentration sea salt and dust particles.

A significant body of prior work has evaluated the suitability of aerosol radiative properties as proxy for CCN based on local in-situ data and local remote sensing using sun-photometers or lidars. Ghan and Collins (2004) devised a method for estimating CCN at cloud base from lidar retrievals and surface CCN measurements, which they evaluated in (Ghan et al., 2006) using aircraft, surface in-situ, and surface remote sensing measurements. This study highlighted deteriorating retrieval quality for higher supersaturations and for scenes with vertical inhomogeneity. A continental-scale compilation of co-located observations of AERONET sun-photometer (Holben et al., 1998) retrieved AOD and ground-based CCN measurements revealed a statistically robust power–law relationship between AOD and CCN for continental scales and (month) long averaging periods (Andreae, 2009). Limited-scale in-situ observations (Kapustin et al., 2006) show that also the relationship of AI to aerosol number and CCN is strongly affected by relative humidity (increasing particle size and extinction but not aerosol number) and complex aerosol size distributions. An analysis of aircraft measurements for the ARCTAS measurement campaign over Canada shows reasonable temporal correlations between CCN and AOD ($r^2 = 0.59$) and a significant improvement in correlation when using in-situ dry extinction instead of vertically integrated ambient AOD (Shinozuka et al., 2015). An analysis of a large compilation of aircraft measurements over the Pacific reveals that regional campaign-average vertical profiles of extinction and CCN proxies show generally a strong correlation (Clarke and Kapustin, 2010) but it is unclear how representative this is for the temporal correlation of extinction and CCN at cloud base. Liu and Li (2014) investigate the correlation of CCN and aerosol radiative properties using data from five Atmospheric Radiation Measurement (ARM) Climate Research Facility sites. They find variable correlations between surface CCN and AERONET retrieved AOD for the different sites, with lower correlations for the Azores and Niger sites and attribute this to the dominance of large particles. They generally find improved correlations using AI as compared to AOD and best correlations between in-situ surface

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scattering/extinctions coefficients and in-situ measured scattering aerosol index. The importance of many of the above factors has also been realised in the context of deriving surface aerosol mass from AOD retrievals for air pollution applications over the continental United States (van Donkelaar et al., 2010), employing a chemical transport model to derive local linear conversion factors from AOD to surface mass.

In summary, previous work on the relationship between aerosol radiative properties and CCN has been based on in-situ CCN data in combination with aerosol radiative properties from in-situ measurements or remote sensing. Studies have found: a variable degree of correlations for different regions and aerosol regimes; generally improved correlations between AI and CCN as compared to AOD and CCN; a degradation of correlations in regions of high relative humidity; an impact of vertical layering on the correlation of surface CCN and aerosol radiative properties. However, the limited availability of direct measurements has made it impossible to provide a global, statistically robust, assessment of the suitability of aerosol radiative properties as constraint for CCN. Consequently, no reliable global observational dataset of cloud condensation nuclei exists and a large body of literature uses AOD/AI almost synonymously for CCN.

This work provides a global assessment of the link between aerosol radiative properties and CCN, overcoming the insufficient global coverage of direct observations through use of a fully self-consistent global model (ECHAM-HAM, Stier et al., 2005, 2007; Zhang et al., 2012) of aerosol radiative properties and CCN. It is clear that no perfect global model of aerosol radiative properties or CCN exists (e.g. Myhre et al., 2013; Mann et al., 2014) so *self-consistent* in this context refers to the fact that the calculation of the aerosol radiative properties (based on Mie theory) and CCN (based on Köhler theory) are fully consistent in terms of the size-distribution, composition and mixing state, unaffected by any independent assumptions or errors common to remote sensing retrievals. Therefore, use of this model allows to consistently assess the relationship between aerosol radiative properties and CCN as biases in the simulated fields are expected to be consistent. Nonetheless, it should be noted that the ability

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of models to mimic the spatial (in particular vertical) and temporal (co-)variability of aerosol and humidity fields introduces some uncertainty.

While the introduced methodology would lend itself to the derivation of CCN retrieval from satellite retrieved aerosol radiative properties, this is not the focus of this study. Likewise, it should be pointed out that this work does not investigate the link between aerosol radiative properties and the number of activated cloud droplets, which additionally requires the knowledge of (highly uncertain) updraft velocities at cloud base or the point of activation. Instead, this work aims to provide the first consistent global analysis of the suitability of aerosol radiative properties as observational constraint for CCN.

2 Methods

In this study we employ the aerosol-climate model ECHAM-HAM, version ECHAM-6.1_HAM-2.2, with a prognostic representation of the composition, size distribution, and mixing state of the major global aerosol components: sulfate, black carbon, particulate organic matter, sea salt, and mineral dust. More details and an extensive evaluation of this base model can be found in (Stier et al., 2005, 2007; Zhang et al., 2012; Schutgens and Stier, 2014) as well as part of the AeroCom intercomparison (Myhre et al., 2013; Stier et al., 2013; Mann et al., 2014).

2.1 The atmospheric general circulation model ECHAM6

The atmospheric general circulation model (GCM) ECHAM6 (Stevens et al., 2013) is the sixth-generation climate model developed at the Max Planck Institute for Meteorology. ECHAM6 solves prognostic equations for vorticity, divergence, surface pressure, and temperature, expressed in terms of spherical harmonics with a triangular truncation. Non linear processes and the physical parameterisations are solved on a corresponding Gaussian grid. Water vapour, cloud liquid water, cloud ice, and trace components are transported in grid-point space with a flux form semi-Lagrangian trans-

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port scheme (Lin and Rood, 1996). ECHAM6 contains a microphysical cloud scheme (Lohmann and Roeckner, 1996; Lohmann et al., 2007) with prognostic equations for cloud liquid water and ice. Cloud cover is represented using an assumed humidity distribution function (Sundqvist et al., 1989). Convective clouds and convective transport are based on the mass-flux scheme of Tiedtke (1989) with modifications by (Nordeng, 1994) and a modified triggering related to a prognostic treatment of the temperature variance in the planetary boundary layer (Stevens et al., 2013). Radiative transfer is represented using the rapid radiation transfer suite of models optimised for general circulation modeling (Iacono et al., 2008) with 16 and 14 bands in the longwave and shortwave parts of the spectrum, respectively.

2.2 The aerosol module HAM

The microphysical aerosol module HAM (Stier et al., 2005, 2007; Zhang et al., 2012) predicts the evolution of an ensemble of seven interacting internally- and externally-mixed log-normal aerosol modes. In the current setup, the components comprise: sulfate, black carbon, particulate organic matter, sea salt, and mineral dust. The microphysical core M7 (Vignati et al., 2004) calculates coagulation among the modes and the condensation of gas-phase sulfuric acid on the existing aerosol population. In the revised version HAM-2.0, the equilibrium water update is based on κ-Köhler theory (Petters and Kreidenweis, 2007) and a range of aerosol nucleation parameterisations have been introduced (Kazil et al., 2010) in addition to the original binary nucleation scheme. In this study we are employing a parameterisation of neutral and charged nucleation Kazil and Lovejoy (2007) as described in Kazil et al. (2010). Aerosol radiative properties, as well as the sink processes of dry deposition, sedimentation, and wet deposition, are parameterised in dependence on the prognostic aerosol size distribution, composition, and mixing state and coupled to the ECHAM meteorology. Emissions of mineral dust, sea salt and DMS from seawater are calculated online. For all other compounds, emission strength, distribution, and height are based on the AEROCOM aerosol model inter-comparison (http://aerocom.met.no) Phase II ACCMIP-MACCity

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emission inventory (Lamarque et al., 2010) for the year 2000. We implement an explicit Köhler theory based activation scheme with empirical estimation of maximum supersaturation in updrafts derived from explicit parcel model calculations (Abdul-Razzak and Ghan, 2000). The total number of activated particles is calculated as sum of the integrated log-normal aerosol number distribution from the radius of activation for each mode. Köhler theory requires detailed information about the aerosol composition. While the composition is relatively well defined for some aerosol components, such as sea salt, the detailed composition of other components, such as particulate organic matter is insufficiently understood. In many measurements of aerosol chemical composition, a non-negligible fraction of the aerosol mass cannot be identified and is often attributed to organics (Jimenez et al., 2009). Given the large uncertainties in the identification and simulation of organics, as well as low measured κ values (Petters and Kreidenweis, 2007), we ignore ionic contributions of organics to the solute and treat organics as well as dust in the hydrophilic modes as wet-able and in the hydrophobic modes as entirely hydrophobic.

2.2.1 Cloud Condensation Nuclei

In addition to the application of Köhler theory in the activation scheme of HAM, this scheme is also used for a consistent diagnostics of cloud condensation nuclei at fixed, prescribed supersaturations.

2.2.2 Aerosol radiative properties

Aerosol radiative properties are calculated in the framework of Mie theory. For each aerosol mode, effective refractive indices are calculated by volume-averaging the refractive indices of all components, including aerosol water, which is parameterised in terms of ambient relative humidity. The effective complex refractive indices and the Mie size-parameters for each mode serve as input to look-up tables for the aerosol radiative properties, providing extinction cross-section, single scattering albedo, and asymmetry

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parameter to the ECHAM radiation scheme. Long-wave (LW) radiative properties have been introduced and coupled to the ECHAM LW radiation scheme and black carbon refractive indices have been revised for version HAM-2.0 (Stier et al., 2007). We additionally introduce diagnostics of: Aerosol Index (AI) calculated online from the aerosol optical depth at wavelengths comparable to the MODIS AI product: $\mathrm{AI} = \mathrm{AOD} \times \alpha$, where the Ångström parameter $\alpha = -\frac{\ln(\mathrm{AOD}_{550\,\mathrm{nm}}/\mathrm{AOD}_{865\,\mathrm{nm}})}{\ln(\lambda_{550\,\mathrm{nm}}/\lambda_{865\,\mathrm{nm}})}$; fine mode AOD calculated as sum of Aitken and Accumulation mode AOD; dry AOD approximated from total AOD minus the AOD times the volume fraction of aerosol water.

We further investigate the role of the vertical aerosol distribution using the local (model layer) aerosol extinction coefficient (AEC) as well as the extinction aerosol index ($\mathrm{AI}_{\mathrm{AEC}}$), defined here as local aerosol extinction coefficient times the local Ångström parameter.

2.3 Simulation setup

All simulations were performed from October 1999 to December 2000 and constrain the large-scale meteorology to the year 2000 by nudging (Jeuken et al., 1996) the model to the ECMWF ERA40 reanalysis data (Simmons and Gibson, 2000). Only the year 2000 data are analysed. We employ a horizontal resolution of T63 in spectral space with a corresponding resolution of $1.8^\circ \times 1.8^\circ$ on a Gaussian grid. The vertical resolution is set to 31 levels, extending from the surface up to 10 hPa.

2.4 Statistical analysis

The statistical analysis is performed on 6 hourly instantaneous model output, unless longer averaging periods are described. Correlations are reported as linear Pearson correlation coefficient of log-transformed parameters, providing consistency with the majority of prior work. Fits are derived from linear regression of the log-transformed parameters to derive power-law expressions. Note that the results remain largely

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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 $\mathrm{CCN}_{0.5\%}$) is statistically robust ($r^2 = 0.70$) and compares to a fit ($y = 0.0027x^{0.640}$ for $\mathrm{CCN}_{0.4\%}$, $r^2 = 0.88$) of campaign mean co-located surface based CCN measurements and AERONET sunphotometer 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 $\mathrm{CCN}_{0.2\%}$ (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 ($r^2 = 0.84$).

Despite the good fit of the regional annual mean CCN and AI, the fit of individual monthly means within each region (colour coded scatter and fits in Fig. 4d) is variable, with r ranging from 0.41 to 0.93 for individual regions.

The mean goodness of fit deteriorates from $r^2 = 0.57$ to $r^2 = 0.46$ and $r^2 = 0.41$ varying the averaging periods of CCN and AI pairs from monthly via daily to 6 h instantaneous data (Fig. 5).

The global distribution of (temporal) Pearson's correlation coefficients between surface $\mathrm{CCN}_{0.2\%}$ and vertically integrated aerosol optical depth (map in Fig. 6) reveals variable suitability of vertically integrated AOD as proxy for surface CCN. While correlations are generally positive and exceed $r = 0.6$ for large parts of the high latitudes and the tropical oceans, significant areas of the continents and subtropical subsidence regions show low or even negative correlations.

Note that maps of global correlations for alternative aerosol radiative properties proposed as superior proxies of CCN (Fig. 7), specifically (a) fine mode aerosol optical depth, (b) dry aerosol optical depth and (c) aerosol index do not show significantly improved correlations. Sampling $\mathrm{CCN}_{0.2\%}$ at the model simulated lowest cloud base gives slightly reduced but spatially very similar correlations with AOD (d). Usage of the non-parametric Spearman's rank correlation coefficient (e) gives slightly enhanced but spatially very similar correlations.

The analysis of the vertical structure of aerosol extinction and CCN reveals the reasons for the large spatial variability in these correlations (Fig. 6). In agreement with observations (Clarke and Kapustin, 2010), the correlation between the annual mean vertical profiles of the aerosol extinction coefficient (AEC) and CCN is robust (r ranges from 0.84 to 0.98 for the selected regions), even in areas for which the temporal correlation of column integrated AOD and surface CCN is poor, such as the South-East Atlantic. This can be explained by vertical decoupling: while the temporal correlation of surface CCN with vertically integrated AOD is low ($r \approx 0.2$) in this region, the correlation of surface CCN with the surface extinction coefficient is robust ($r > 0.6$). Generally, the temporal correlation of extinction coefficients with CCN at the same layer (orange) is 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 $\text{CCN}_{0.2\%}$ 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 $\text{CCN}_{0.5\%}$ and $\text{CCN}_{1.5\%}$, respectively. Corresponding areas for $r < 0.5$ between CCN and aerosol index are somewhat lower (52, 66, 91 % for $\text{CCN}_{0.2\%}$, $\text{CCN}_{0.5\%}$, $\text{CCN}_{1.5\%}$, 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.

However, aerosol–cloud interactions occur locally (McComiskey and Feingold, 2012): an analysis of the temporal correlation on the local (grid) scale 71 % of the area of the globe shows correlation coefficients between $\mathrm{CCN}_{0.2\%}$ at cloud base and AOD below 0.5 (i.e. AOD variability explains only 25 % of the CCN variance). The areas with low correlations include the main marine stratocumulus decks, considered most susceptible to aerosol perturbations (Boucher et al., 2013). This suggests particularly limited constraints from passive satellite remote sensing in areas key for radiative forcing due to aerosol–cloud interactions. Note that correlations for alternative aerosol radiative properties proposed as superior proxies of CCN such as fine mode aerosol optical depth, dry aerosol optical depth and aerosol index do not show significant improvements.

A number of reasons contribute to the low correlations between aerosol radiative properties and CCN, in particular over sub-tropical subsidence areas: aerosol extinction is heavily affected by humidity, in particular at cloud base, and often the local correlation of relative humidity with aerosol extinction coefficients is larger than the correlation of local CCN with column integrated AOD. Satellite retrievals based on visible wavelengths are most sensitive to larger particles, corresponding to CCN at small supersaturations. Correlations between CCN and AI decrease with increasing supersaturations, in particular over the continents with significant primary fine mode aerosol emissions. Additionally, surface or cloud-base CCN and column AOD are often decoupled: and correlations of CCN with local aerosol extinction coefficients throughout the troposphere significantly exceed the correlations with column AOD. Consequently, correlations of surface CCN with surface AEC are significantly larger than with column AOD and are further improved for surface $\mathrm{AI}_{\mathrm{AEC}}$ for which $r > 0.8$ for most of the globe.

While the ability of this global model to mimic the spatial (in particular vertical) and temporal (co-)variability of aerosol and humidity fields introduces some uncertainty, it should be noted that this approach is free from retrieval errors, which would add additional uncertainty when using real satellite data. Advances in computational capabilities now make high-resolution, large-domain simulations of aerosols, clouds and their 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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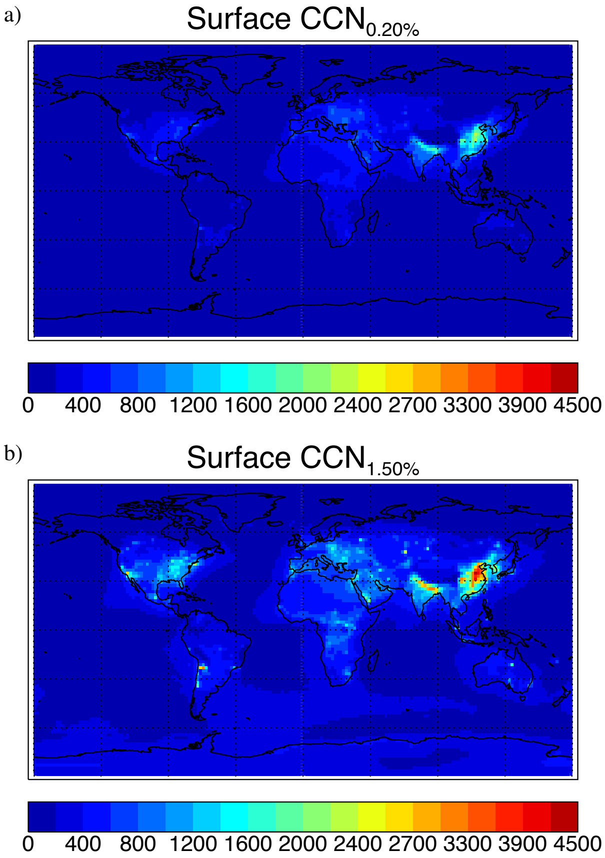
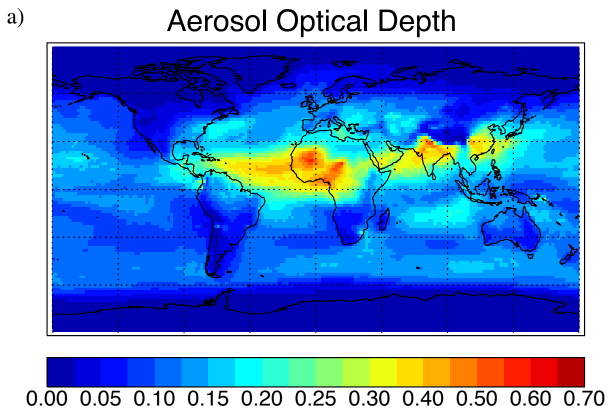


Figure 1. Annual-mean simulated surface cloud condensation nuclei concentrations [cm^{-3}] at **(a)** 0.2 % and **(b)** 1.5 % supersaturation.

a) Aerosol Optical Depth



b) Aerosol Index

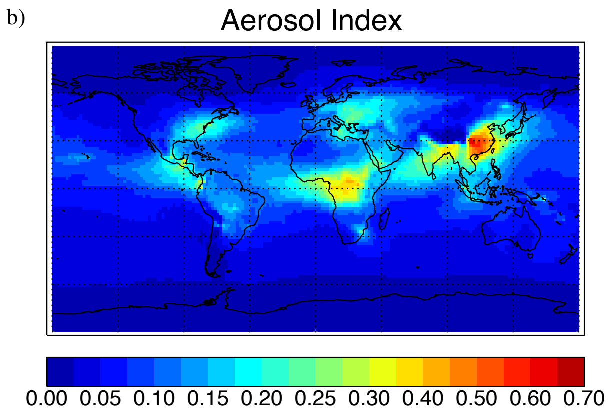


Figure 2. Annual-mean simulated **(a)** aerosol optical depth at 550nm and **(b)** aerosol index between wavelengths of 550 and 865nm. Note the non-linear scale, comparable to Fig. 1.

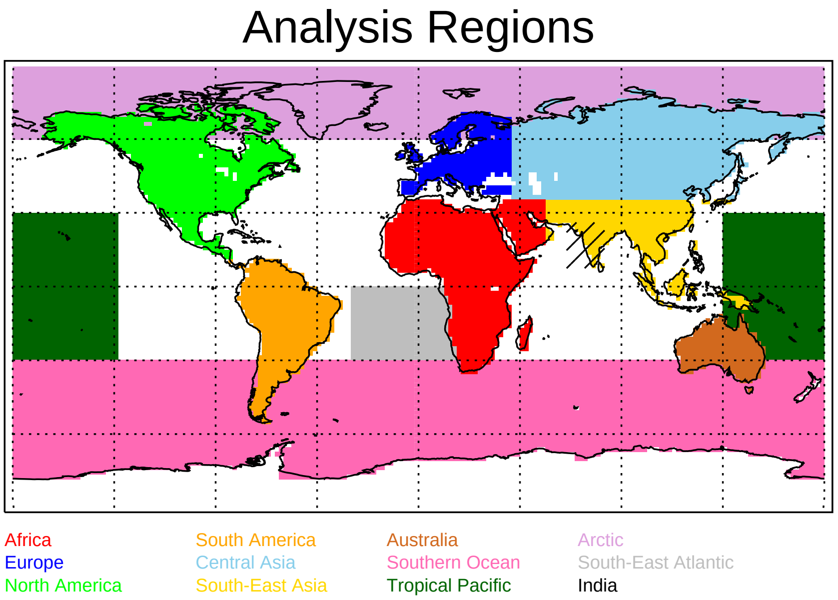


Figure 3. Map of regions used in the analysis.

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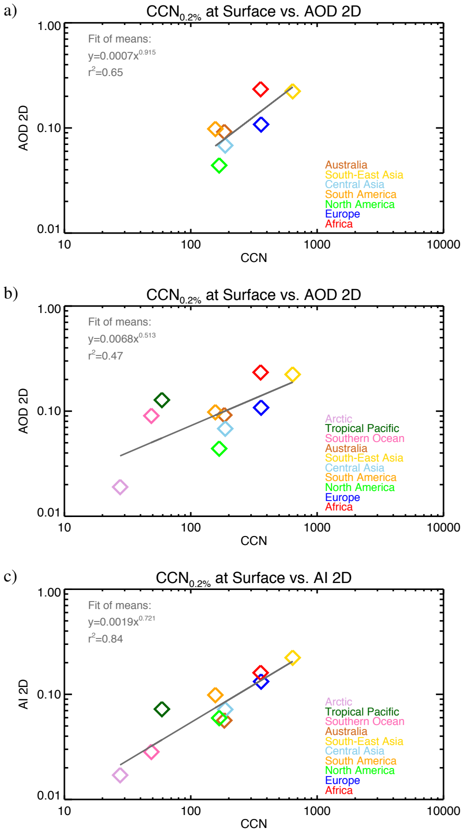

Figure 4. (a) Annual continental mean AOD [1] as function of CCN$_{0.2\%}$ [cm^{-3}] and their fit derived from linear regression (gray), **(b)** as **(a)** but including three ocean regions, **(c)** annual continental mean simulated AI as function of CCN$_{0.2\%}$ for continental and ocean regions as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

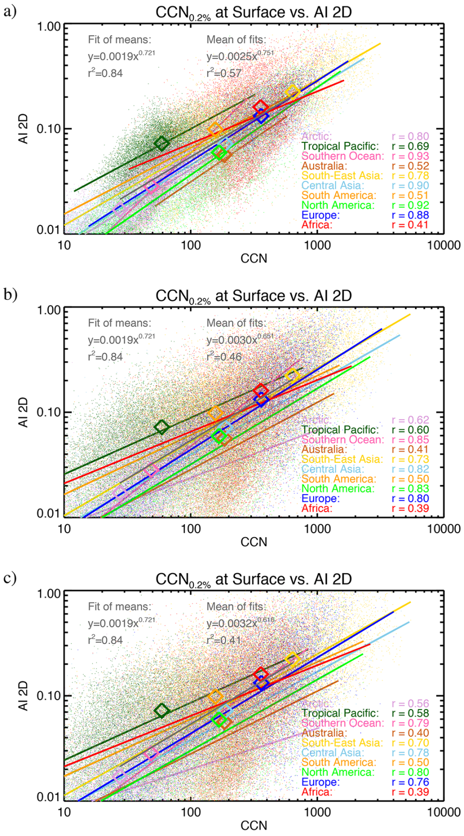

Figure 5. Annual continental mean AI [1] as function of CCN$_{0.2\%}$ [cm^{-3}] (symbols) and their fit derived from linear regression (gray); overlay of **(a)** montly mean, **(b)** daily mean and **(c)** instantaneous 6 hourly pairs of AI and CCN$_{0.2\%}$ (scatter) and their fit derived from linear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

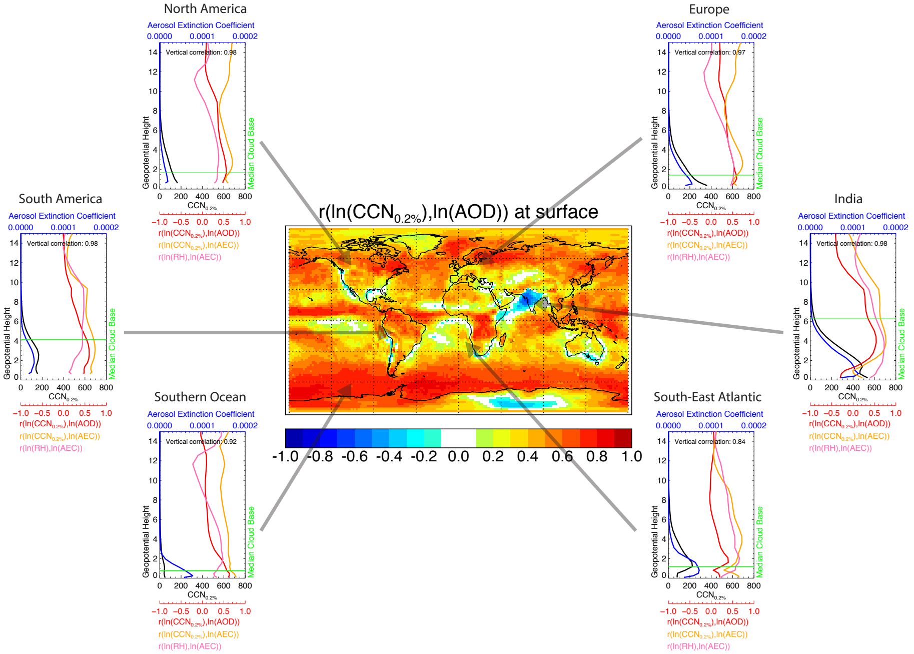

Figure 6. Map of Pearson's correlation coefficient *r* of surface ln(CCN$_{0.2\%}$) with column integrated aerosol optical depth ln(AOD) calculated for each model grid box from one year of 6 hourly pairs. Annual-mean vertical profiles of CCN$_{0.2\%}$ [cm^{-3}] (black), aerosol extinction coefficient (AEC) [m^{-1}] (blue), profile of temporal correlation of ln(CCN$_{0.2\%}$) with column integrated ln(AOD) (red), profile of temporal correlation ofln(CCN$_{0.2\%}$) with vertically resolved extinction coefficient ln(AEC) orange) and temporal correlation of ln(RH) with vertically resolved extinction coefficient ln(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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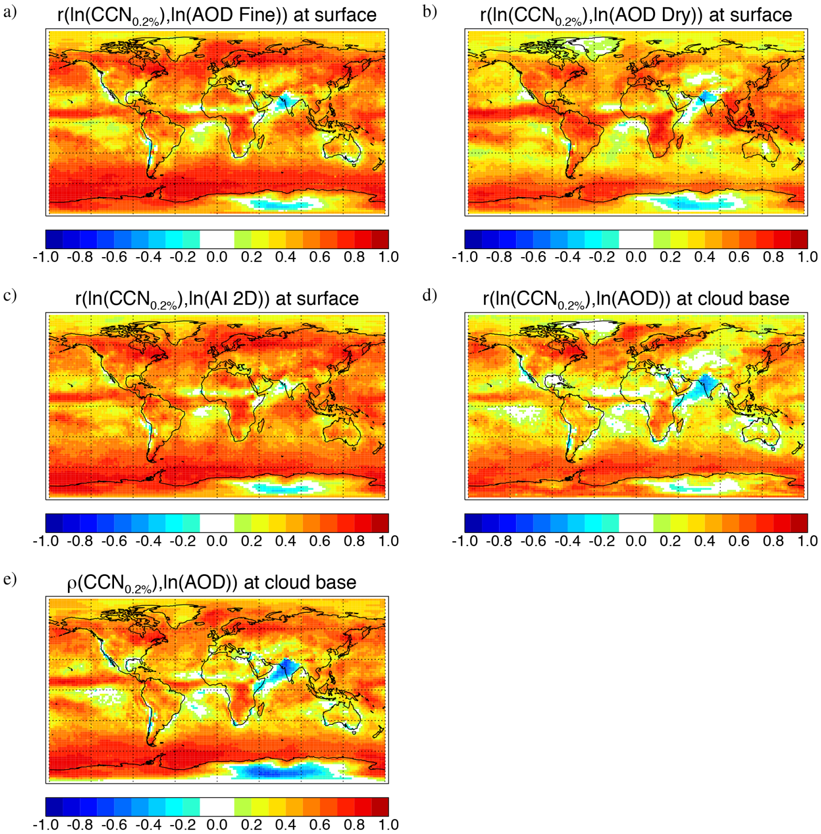


Figure 7. Map of Pearson's correlation coefficient of CCN$_{0.2\%}$ with aerosol radiative properties for **(a)** surface CCN$_{0.2\%}$ with vertically integrated fine mode aerosol optical depth, **(b)** surface CCN$_{0.2\%}$ with vertically integrated dry aerosol optical depth, **(c)** surface CCN$_{0.2\%}$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

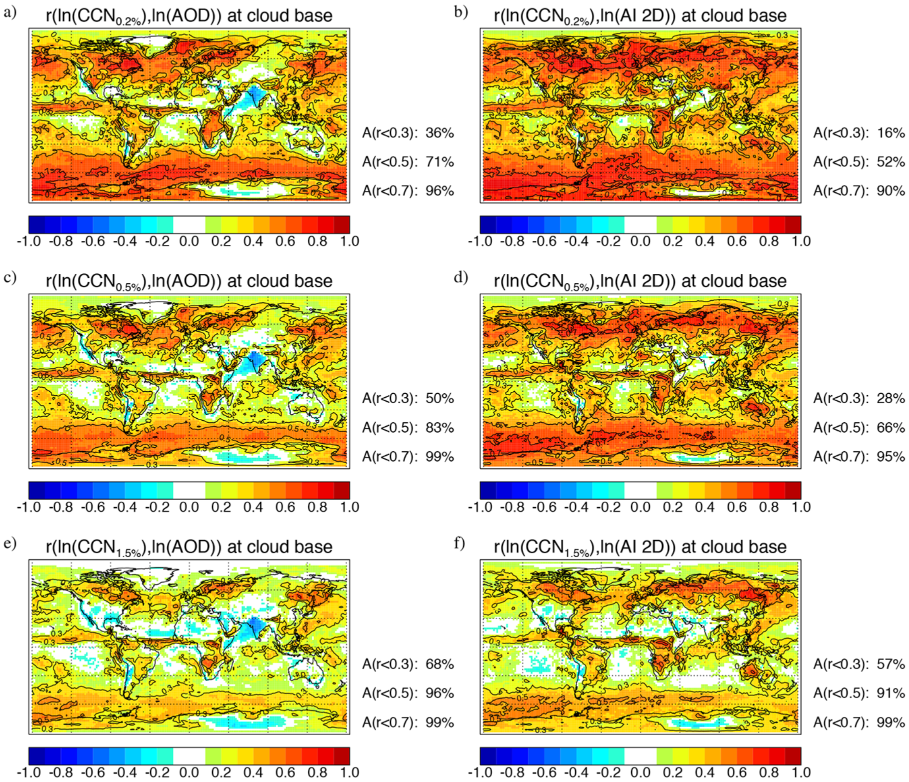
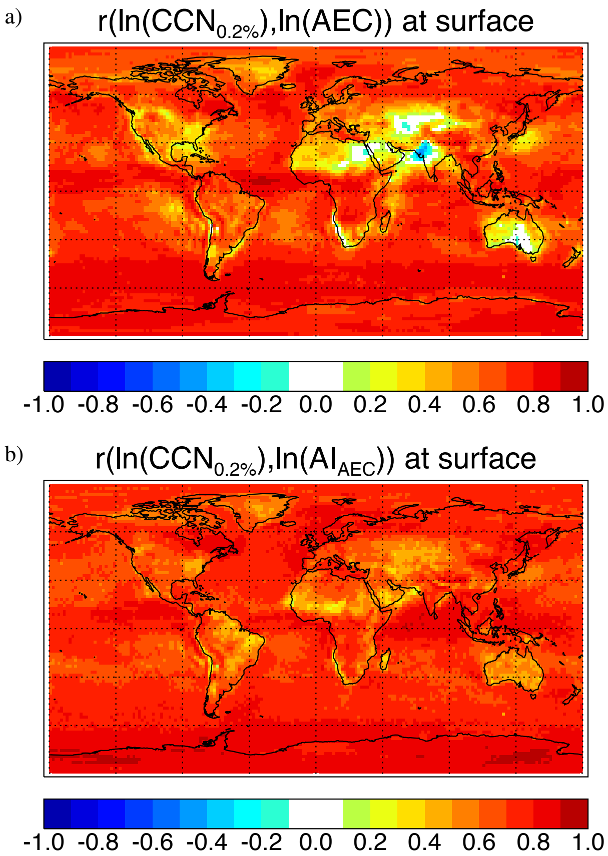


Figure 8. Map of Pearson's correlation coefficient of cloud base CCN with aerosol radiative properties for **(a)** $\mathrm{CCN}_{0.2\%}$ with vertically integrated aerosol optical depth, **(b)** $\mathrm{CCN}_{0.2\%}$ with vertically integrated aerosol index, **(c)** $\mathrm{CCN}_{0.5\%}$ with vertically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



a) $r(\ln(\mathrm{CCN}_{0.2\%}),\ln(\mathrm{AEC}))$ at surface

b) $r(\ln(\mathrm{CCN}_{0.2\%}),\ln(\mathrm{AI}_{\mathrm{AEC}}))$ at surface

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

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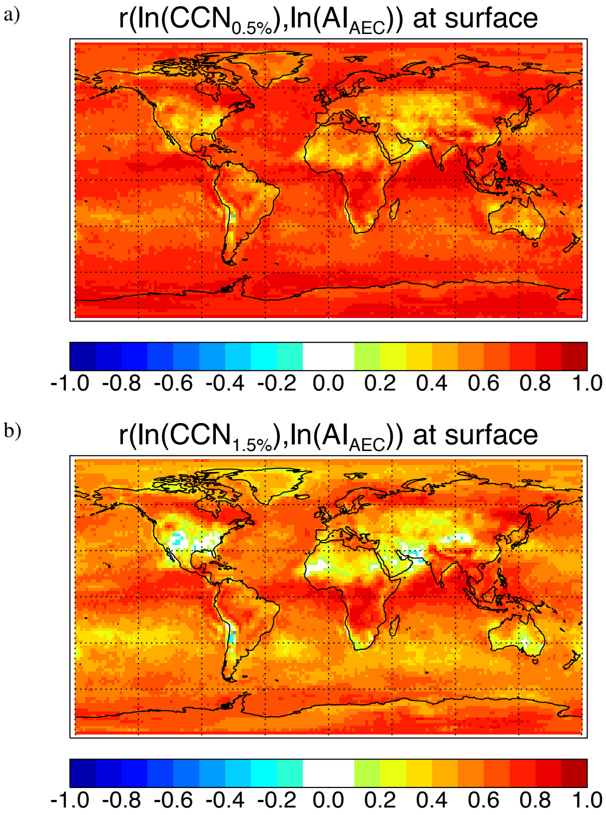

Figure 10. Map of Pearson's correlation coefficient of surface layer ln(AI$_{AEC}$) with ln(CCN) at higher supersaturations: **(a)** CCN$_{0.5\%}$, **(b)** CCN$_{1.5\%}$.