Recent improvements and maximum covariance analysis of aerosol and cloud properties in the EC-Earth3-AerChem model

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Abstract. Given the importance of aerosols, clouds and their interactions in the climate system, it is imperative that the global Earth system models accurately represent processes associated with them. This is an important prerequisite if we were to narrow the uncertainties in future climate projections. In practice, this means that the continuous model evaluations and improvements grounded in observations are necessary. Numerous studies in the last few decades have shown both the usability and the

5   limitations of utilizing satellite-based observations in understanding and evaluating aerosol-cloud interactions, particularly under varying meteorological and satellite sensor sensitivity paradigms. Furthermore, the vast range of spatio-temporal scales at which aerosol and cloud processes occur adds another dimension to the challenges while evaluating climate models.

   In this context, the aim of this study is two-fold. 1) We evaluate the most recent, significant changes in the representation of aerosol and cloud processes implemented in the EC-Earth3-AerChem model in the framework of the EU project FORCeS

10   compared to its previous CMIP6 version. We focus particularly on evaluating cloud physical properties and radiative effects, wherever possible, using a satellite simulator. We report overall improvements in EC-Earth3-AerChem model. In particular, the strong warm bias chronically seen over the Southern Ocean is reduced significantly. 2) A statistical, maximum covariance analysis is carried out between aerosol optical depth (AOD) and cloud droplet (CD) effective radius based on the recent EC-Earth3-AerChem/FORCeS simulation to understand to what extent the Twomey effect can manifest itself in the larger

15   spatio-temporal scales. We focus on the three oceanic low-level cloud regimes that are important due to their strong net cooling effect and where pollution outflow from the nearby continent is simultaneously pervasive. We report that the statistical covariability between AOD and CD effective radius is indeed dominantly visible even at the climate scale when the aerosol amount and composition are favourably preconditioned for allowing aerosol-cloud interactions. Despite this strong covariability, our analysis shows a strong cooling/warming in shortwave cloud radiative effects at the top of the atmosphere in our

20 study regions associated with an increase/decrease in CD effective radius. And this cooling/warming can be attributed to the increase/decrease in low cloud fraction, in line with the previous observational studies.

1 Introduction

Aerosols can potentially act as cloud condensation nuclei (CCN) or ice nuclei, influencing cloud formation, structure, and properties. The type, concentration, and size distribution of aerosols impact cloud microphysics, altering droplet or ice crystal
25 size, number concentration, and cloud albedo (Twomey, 1974, 1977; Albrecht, 1989; Ramanathan et al., 2001). Conversely, clouds can also affect aerosols through wet deposition processes and also, indirectly via impact on meteorology and gas phase photochemistry, thereby, influencing their distribution and removal from the atmosphere. This interdependency of aerosol and clouds mediated by local meteorology plays a crucial role in climate regulation, radiative forcing and the distribution of precipitation. However, even after decades of research, a full grasp of the nature of aerosol-cloud interactions remains one
30 of the big challenges and constitutes one of the largest sources of uncertainty in our understanding of climate forcing and feedbacks (Solomon et al., 2007; Quaas et al., 2009; Carslaw et al., 2010; Bellouin et al., 2019).

The vast range of spatio-temporal scales at which aerosol-cloud interactions occur helps us to appreciate why it is so difficult to pin down their role in the climate system. For example, processes like condensation and sublimation occur at nano- to micrometer spatial scales, while the interplay between aerosols and clouds over the major pollution outflow regions occur at
35 much larger spatial scales. The temporal component of aerosol-cloud interactions also stretches from microseconds, to days and weeks of persistent pollution outflow, to the decades of emission policy changes. To account for these wide ranges of spatio-temporal scales, the harmonization and representation in a physically consistent and observationally constrained manner in global climate models is extremely challenging. At the same time, the evaluation of climate models must cover these different spatio-temporal scales. This can help us to assess, among others, at what spatio-temporal scales the aerosol-cloud interactions
40 can distinctly manifest themselves and could even dominate the local variability.

In this context, this study focuses on understanding variability at larger scales. We specifically aim to understand the co-variability between AOD and CD effective radius at the climate scale. We acknowledge that both AOD and CD effective radius can be independently influenced by a number of processes and that CCN number concentration, especially at the cloud base, are better suited to investigate aerosol impact on cloud albedo via changes in droplet radius, as discussed in a number of previous
45 studies (e.g. Quaas et al. (2020) and the references therein). The correspondence between AOD and CCN number concentration has been observed in a number of studies. Andreae (2009) identified a correlation between AOD and CCN concentration by comparing AERONET AOD with CCN measurements. Romakkaniemi et al. (2012) suggested the potential use of AOD as a proxy for CCN concentration. While uncertainties persist, Tang et al. (2014) noted the limitations of AOD as an indicator of CCN, acknowledging variations in CCN concentrations for a given AOD based on regional characteristics and meteorological
50 conditions, such as relative humidity. Despite these complexities, Andreae (2009) observed a high correlation coefficient of 0.98 between AOD and CCN concentration. Using the ground measurements of CCN at the Zeppelin Observatory (78.91°N, 11.89°E), Ahn et al. (2021) derived a CCN–AOD correlation with a coefficient of determination R^2 of 0.59.

We choose AOD and CD effective radius here because these variables are better grounded in observations and can be evaluated more directly using satellite-based observations. Furthermore, we assume that if aerosols indeed are one of the primary drivers of changes in cloud properties, especially under favourable meteorological conditions (Fanourgakis et al., 2019), their co-variability at a much larger, temporal and spatial scales shall visibly manifest itself in a relation between AOD and CD effective radius.

In this study, we first evaluate the recent improvements made in the EC-Earth3-AerChem model using a suite of satellite-based observations and we then employ maximum co-variance analysis on AOD and CD effective radius to explore the intricate interplay between aerosols and clouds. We primarily focus on the oceanic regions that sustain low-level liquid water clouds and where the pollution outflow also occurs at least to some degree. The net cooling effect of low-level liquid water clouds over open oceans is very important and their susceptibility to aerosols has big implications for the energy budget.

This work has been carried out as part of the European Union (EU) Horizon 2020 project FORCeS (Constrained aerosol forcing for improved climate projections (https://forces-project.eu/)). The project aims to identify the important aerosol and cloud processes that could reduce the uncertainties in the aerosol forcing estimates. In this framework, several improvements of the description of these processes that could potentially reduce the uncertainties were implemented in three global models that were previously used for CMIP6 ((https://pcmdi.llnl.gov/CMIP6/): NorESM2-LM (Seland et al., 2020), MPI-ESM1.2-HAM (Mauritsen et al., 2019; Neubauer et al., 2019) and EC-Earth3-AerChem (van Noije et al., 2021). In this paper we focus on EC-Earth3-AerChem model.

2 Model, observations and methods

2.1 EC-Earth3-AerChem model description

The EC-Earth3-AerChem model is based on the EC-Earth3 family of models (Döscher et al., 2022). The atmospheric component of the model is based on IFS cycle 36r4 which includes the land surface model H-TESSEL (Balsamo et al., 2009). The AerChem configuration features the aerosol and chemistry model, Tracer Model version 5 (TM5) (Krol et al., 2005; Huijnen et al., 2010). The resolution of the model is T255L91 with a grid spacing of approximately 80 km and 91 levels in the vertical that extends to 0.01 hPa. However, TM5 is run at a coarser resolution of 3^ox2^o with 34 vertical levels. The details of the CMIP6 version of the EC-Earth3-AerChem model are described in van Noije et al. (2021).

2.2 EC-Earth3-AerChem model updates within FORCeS

The FORCeS project aimed at the improvement of various crucial processes in the global climate models that would have the potential to influence aerosol radiative forcing and climate feedbacks, particularly concerning aerosols and clouds so as to improve our future climate projections. In this context, the different minerals present in dust aerosols are now explicitly traced and their interaction with specific climate processes has been implemented and the cloud activation scheme was updated. The following paragraphs give a more detailed, but, a brief overview of these updates.

2.2.1 Cloud activation scheme

85 An analysis of the different cloud droplet activation parameterization schemes (Partridge et al., in preparation) used in the global models showed systematic biases for parameters describing the aerosol size distribution and updraft velocity compared to the cloud parcel model that can lead to very low (high) maximum supersaturation for cases when high (low) aerosol loads combined with low (high) updraft velocity. Simpler schemes such as the Abdul-Razzak and Ghan (2000) and Shipway (2015) parameterizations typically used in GCMs showed larger biases compared to the more complex schemes such as the Barahona

90 et al. (2010) and Morales-Betancourt and Nenes (2014) schemes in clean marine regions, particularly over the Southern ocean. Hence, the cloud activation scheme of Abdul-Razzak and Ghan (2000) is replaced by the Morales-Betancourt and Nenes (2014) scheme in the FORCeS version of the EC-Earth3-AerChem model. This scheme makes use of the population splitting concept wherein the growing population of droplets is divided into three enabling a more accurate estimate for the number concentration of droplets formed at the time of maximum supersaturation.

2.2.2 Dust mineralogy

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The model developed in FORCeS explicitly incorporates the atmospheric cycle of dust minerals relevant for their climate impacts. Namely, iron oxides which control the absorption of shortwave radiation by dust aerosols (e.g., Sokolik and Toon (1999), Di Biagio et al. (2019)), quartz and feldspars that constitute efficient ice-nucleators (e.g., Atkinson et al. (2013), Harrison et al. (2019)), and calcite, which intervenes in atmospheric chemistry processes and affects the aerosol pH. The emission

100 of the different minerals is calculated relying on the soil mineralogical map of Claquin et al. (1999) with the modifications from Nickovic et al. (2012). To account for differences in the mineral's size distribution reported in the soil and those found in the aerosols, we apply an extension of the Brittle Fragmentation Theory for dust emission proposed by Kok (2011). This approach has shown a better agreement with observations (e.g., Perlwitz et al. (2015a, b)) than relying exclusively in the soil size fractions. In the soil map we consider, hematite as a proxy for iron oxides, therefore the optical properties for dust in the

105 SW are calculated online assuming an internal mixture of hematite and other host minerals with a Maxwell Garnett mixing rule. The refractive indices are taken from Scanza et al. (2015). The ice nucleation processes are sensitive to the K-feldspar fraction, which we assume to be a 35% of the total feldspar (Atkinson et al., 2013; Chatziparaschos et al., 2023). An extension of the soil mineralogy that accounts for the presence of feldspar in small particle sizes has also been included (Chatziparaschos et al., 2023).

2.2.3 MPOA source

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Marine organic aerosols have been suggested as a relevant source of ice-nucleating particles, particularly in remote marine environments (Wilson et al., 2015). The EC-Earth3-AerChem FORCeS model version includes a new source of Marine Primary Organic Aerosols (MPOA), that considers the partitioning between insoluble marine organics and sea salt. The MPOA emission is calculated as a fraction of the submicron sea salt aerosols dependent on the chloropyll-a (Chl-a) present in the

115 ocean surface layer (O'Dowd et al., 2008; Vignati et al., 2010). In addition, a coarse mode MPOA is included (Facchini et al.,

2008; Myriokefalitakis et al., 2010), which also depends on the online calculated sea salt emission (Gong, 2003). The model uses Chl-a concentration, a monthly averaged product derived from MODIS (Moderate Resolution Imaging Spectroradiometer) satellite observations.

2.2.4 Primary and secondary ice production

120 In regions such as the remote Arctic/Antarctica where the primary ice nucleating particles (INP) are sparse (Moore et al., 2024; Wex et al., 2019), a considerable source of ice crystals is via secondary ice production (SIP) processes in supercooled clouds (temperatures $\sim > -10^{o}$C) (Järvinen et al., 2022; Järvinen et al., 2023). This process by which ice crystals are generated outnumber those generated by primary INPs (Field et al., 2017). The mechanisms that contribute to the INP formation are not adequately represented in the models resulting in a significant underestimation of the observed ice crystal number concentrations

125 which in turn would impact the radiative forcing estimates (Vergara-Temprado et al., 2018; Zhao et al., 2023). In this project, an aerosol-and-temperature-sensitive parameterization (Costa-Surós et al., in preparation) has been developed to substitute the temperature-based parameterization by Meyers et al. (1992) for estimating the ice crystal number concentrations (ICNC) in mixed phase clouds. Specifically, it considers the primary ice crystal formation by immersion freezing dependent on K-feldspar and quartz (Harrison et al., 2019), deposition nucleation of soot and dust (Ullrich et al., 2017) and immersion freezing of marine

130 organic aerosols (Wilson et al., 2015). The ICNCs formed via SIP are quantified using the RaFSIP (Random Forest Secondary Ice Production) scheme version 1, as detailed in Georgakaki and Nenes (2023). RaFSIP is a data-driven parameterization developed from a two-year simulation with a 10-km horizontal grid spacing, covering the period from 2016 to 2017 over the pan-Arctic region using the Weather Research and Forecasting (WRF) model (https://www.mmm.ucar.edu/models/wrf) with explicit SIP microphysics (Sotiropoulou et al., 2021; Georgakaki et al., 2022). The effect of SIP through collisional break-up,

135 droplet-shattering and Hallett-Mossop rime splintering is described in the model through the introduction of an Ice Enhancement Factor (IEF) - a multiplication factor applied to primary ice production rates.

2.2.5 ISORROPIA-Lite for inorganic aerosols

A new thermodynamic module for inorganic aerosols, ISORROPIA-Lite (Kakavas et al., 2022), is also implemented in the updated EC-Earth3-AerChem model replacing EQSAM (Metzger et al., 2002). ISORROPIA-lite is based on the ISORROPIA-

140 II (Fountoukis and Nenes, 2007) code and it treats the thermodynamics of aerosol containing Ca^{2+}, K^+, Mg^{2+}, $SO_4{}^{2-}$, Na^+, $NH_4{}^+$, $NO_3{}^-$, Cl^-, H_2O and their equilibrium with gas-phase HNO_3, NH_3, HCl and H_2O. Furthermore, the bulk calculation of ammonium nitrate has been replaced by one which distributes these components over the accumulation and coarse soluble modes.

2.2.6 Model tuning

145 The model needed to be re-tuned after these updates and the parameters related to the autoconversion of cloud water to rain were slightly adjusted: the conversion efficiency, RPRCON was increased from $1.34*10^{-3}$ to $1.41*10^{-3}$ and the critical radius

was reduced from 8.75 to 7.5 μm. Also, the standard deviation of the updraft velocity distribution was reduced by 0.2 m/s (from 0.8 m/s to 0.6 m/s). Therefore, the cloud forcing was reduced by ~1 W/m^2.

In the following we will refer to the CMIP6 configuration of the EC-Earth3-AerChem model as ECE3-CMIP6 while the
150   configuration including FORCeS updates will be referred to as ECE3-FORCeS.

2.3   Observational datasets

To evaluate the impacts of the model updates detailed in Section 3, we use a suite of satellite-based datasets of cloud and aerosol properties. These are described below.

MODIS-Aqua: Retrievals of cloud properties from the MODIS onboard the Aqua satellite in the framework of NASA's
155   Earth Observing System (EOS) are used. We specifically use Collection 6 Level-2 and Level-3 (MYD06_L2 and MYD08_L3) information on cloud fraction, optical thickness, liquid water path and droplet effective radius. The cloud droplet number concentration is computed from CD effective radius and cloud optical depth products retrievals following methodologies suggested in Quaas et al. (2020) and the references therein. Eighteen years of data from 2003 through 2020 is used.

CERES-EBAF: The radiative flux components from the Clouds and the Earth's Radiant Energy System (CERES) Energy
160   Balanced and Filled (EBAF) dataset are used to evaluate the top of the atmosphere shortwave and longwave fluxes. The latest Edition 4.2 data at Level-3b is used for the analysis (Loeb et al., 2018; Kato et al., 2018). The time period from 2000-2016 is made use of in this study.

CPR-CloudSat: The latest Release 5 of CloudSat Level 2B-CWC-RVOD and 2B-CWC-RO (CloudSat Radar-Only Cloud Water Content) products are used to evaluate the cloud liquid (CLWP) and ice water paths (CIWP) respectively. These products
165   are derived for each radar profile as seen by CloudSat's Cloud Profiling Radar (CPR) for those profiles for which clouds are likely based on the radar profile analysis (https://www.cloudsat.cira.colostate.edu/cloudsat-static/info/dl/2b-cwc-ro/2B-CWC-RO_PDICD.P1_R05.rev0_.pdf). In 2B-CWC-RVOD product, the retrievals of CLWP are constrained using cloud optical depth information from MODIS (https://www.cloudsat.cira.colostate.edu/data-products/2b-cwc-rvod). Unlike the passive sensors, the CPR onboard CloudSat can sense the entire cloud column, day and night, thus providing better estimates of CLWP and
170   CIWP. We use the data from 2007 through 2011.

CALIPSO: We further use the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) Lidar Level-3 Global Energy and Water Cycle Experiment (GEWEX) Cloud, Standard Version 1-00 data product (CAL_LID_L3_GEWEX_Cl oud-Standard-V1-00) to obtain information on total cloud fraction and its subdivision into low-, medium- and high-cloud fraction (NASA/LARC/SD/ASDC, 2019). This latest Level-3 version is based on the version 4.20 Level-2 5-km merged layer
175   product for the period 2007-2016.

The climatologies of cloud and aerosol properties from these datasets averaged over their respective time periods are statistically compared with the model simulations in the section below.

2.4 Maximum Covariance analysis: description and methodology

Maximum Covariance Analysis (MCA) (Bretherton et al., 1992; Cherry, 1996) is a statistical technique that extracts coherent
180   patterns in two different data sets that explain the maximum fraction of the covariance between them. This means that the
analysis identifies the regions where the two fields co-vary to a maximum extent in the different spatial modes. Rotated MCA
is employed here to maximize the separation between the patterns and to improve their interpretability. This analysis provides
two maps: 1. homogeneous regression map and 2. heterogeneous regression map. The homogeneous patterns are the correlation
co-efficients between the input data of one field and scores (see the section below on how scores are calculated) of the same
185   field, whereas the heterogeneous patterns are the correlation co-efficients between the input data of one field and scores of the
second field and vice versa. The resulting heterogeneous maps are a typical characteristic of the MCA as they bring out the
covariability between the two data sets. The scores are similar to the principal components (PCs) that captures the amplitude
and temporal variation or in other words, the variability associated with each spatial pattern (or mode). The squared covariance
fraction (SCF) is an invariant quantity of this analysis. This fraction assesses the relative importance of each mode.

2.4.1   Mathematical background

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A brief overview of the MC analysis is given below.

Assume two data sets, X and Y with variables a and b respectively. These two data sets can be represented as matrices, X(a x
n) and Y(b x n), n is the number of observations. Both X and Y are first standardized and detrended.

The homogeneous patterns for X and Y fields are defined as,

195   $hom_X = Corr(X, A_x)$

$hom_Y = Corr(Y, A_y)$

Similarly, the heterogeneous patterns for X and Y fields are defined as,

$het_X = Corr(X, A_y)$

$het_Y = Corr(Y, A_x)$

200   where '$Corr$' refers to the correlation co-efficient between the two fields within the brackets, A_x and A_y are the scores of X
and Y. The scores are defined as projections of X and Y on to the singular vectors of the cross covariance matrix of X and Y.
The cross covariance matrix C_{xy} between X and Y are computed as in the equation below.

$$C_{xy} = (\frac{1}{n}) XY^T$$

where Y^T is the transpose matrix of Y.

205   A singular value decomposition is performed on the cross covariance matrix as,

$$C_{xy} = UDV^T$$

where U and V are correspondingly the eigen vectors for X and Y, and D is a diagonal matrix with singular values.

The scores are then calculated as,

$$A_x = U^T X$$

210   $$A_y = V^T Y$$

The squared covariance fraction quantifies the extent to which each mode i accounts for the explained proportion of the total squared covariance and is defined as,

$$SCF_i = \frac{\sigma_i^2}{\sum_{i=1}^{m} \sigma_i^2}$$

where m is the total number of modes and σ_i is the ith singular value of the covariance matrix.

3   Results-I: Evaluation of the recent improvements in ECE3-FORCeS model

Historical atmosphere-only (AMIP) simulations for the period 1980-2020 are used for the analysis. The AMIP simulations are forced by sea surface temperature (SST) and sea ice concentrations from the input4MIPs.CMIP6.CMIP.PCMDI.PCMDI-AMIP-1-1-8 database (https://www.wdc-climate.de/ui/cmip6?input=input4MIPs.CMIP6.CMIP.PCMDI.PCMDI-AMIP-1-1-8.ocean.mon.tosbcs.gn.v20220622). The forcing dataset is based on UK MetOffice HadISST and NCEP OI2 (Durack et al., 2022). The CMIP6 simulations cover a period from 1980-2018.

To facilitate the evaluation of the model simulated variables against satellite observations in a consistent manner, the COSP (Cloud Feedback Model Intercomparison Project (CFMIP) Observation Simulator Package) (Bodas-Salcedo et al., 2011) that is developed by the CFMIP community is available in EC-Earth3-AerChem. COSP provides tools and algorithms to simulate what a climate model would "see" from the viewpoint of the satellite that is used. This allows for a more direct comparison with satellite observations (Pincus et al., 2012).

In the following sections, we focus on the evaluation of the ECE3-FORCeS version and provide a comparison against both the ECE-CMIP6 version and observations.

3.1   Evaluation of COSP-simulated cloud fractions

In this section, simulated cloud and aerosol from the ECE3-FORCeS version of the model are evaluated against satellite observations and compared against the simulations of ECE3-CMIP6 version of the model. Here, the simulated cloud parameters derived from the COSP simulator of the model are evaluated.

The EC-Earth3-AerChem model outputs the following COSP parameters: CALIPSO-COSP total, low-, middle- and high-cloud fractions, MODIS-COSP total, water and ice cloud fractions. The CALIPSO-COSP simulated climatological mean total and the cloud fractions at three altitudes are presented in Fig. 1. The values in brackets correspond to the global mean. The ECE3-FORCeS model simulates the spatial distribution of the total cloud fraction reasonably well compared to the observations, with an overestimation in the polar latitudes. This overestimation stems from the biases primarily seen in the low-cloud
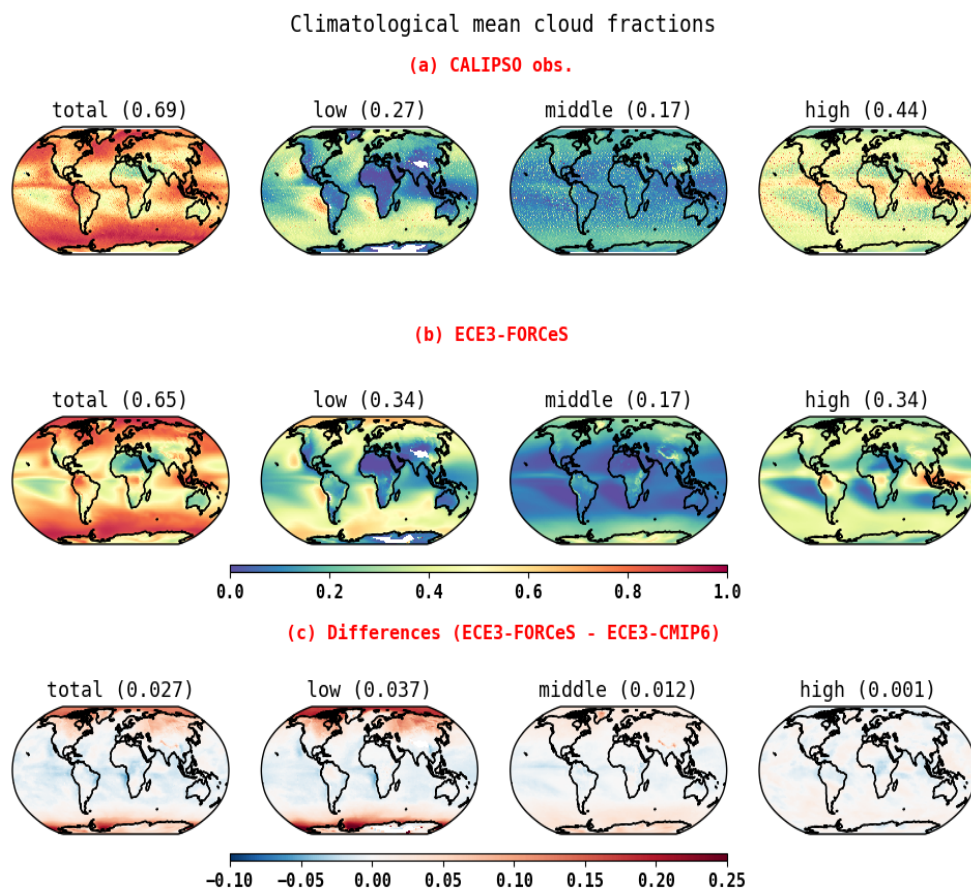
Climatological mean cloud fractions

Figure 1. Climatological mean total, low-, middle- and high-cloud fractions respectively in the four columns from (a) CALIPSO observations, (b) COSP-CALIPSO simulations from ECE3-FORCeS and (c) differences with respect to ECE3-CMIP6 simulations. The values in the brackets refer to the global mean of each category. The observations extend from 83^oS - 83^oN.

fraction. The high-cloud fraction, on the other hand, is considerably underestimated with respect to the observations. This explains the underestimation of the total cloud fraction over the oceanic regions west of South America, Africa and Australia. The simulated global mean total cloud fraction is 65%, whereas the observed mean fraction is 69%. The aerosol and cloud

240 processes have resulted in an increase in the total cloud fraction in the high latitudes in both hemispheres with respect to the corresponding ECE3-CMIP6 simulations and this increase can be attributed to the increase in low-cloud fraction and partly to an increase in middle-cloud fraction. No significant change is seen in the tropics and mid-latitudes with the model updates.

The ECE3-FORCeS and ECE3-CMIP6 zonal averaged climatological mean total, low-, middle- and high-cloud fractions are evaluated against CALIPSO observations (see Fig. 2 (a) - (d)). The black solid lines in each category of the cloud fraction

245 refer to the observed value. The total cloud fraction in the Southern Hemisphere (SH) high latitudes is much closer to the observations in the ECE3-FORCeS model. It can be clearly seen that the improvements in the ECE3-FORCeS version of the

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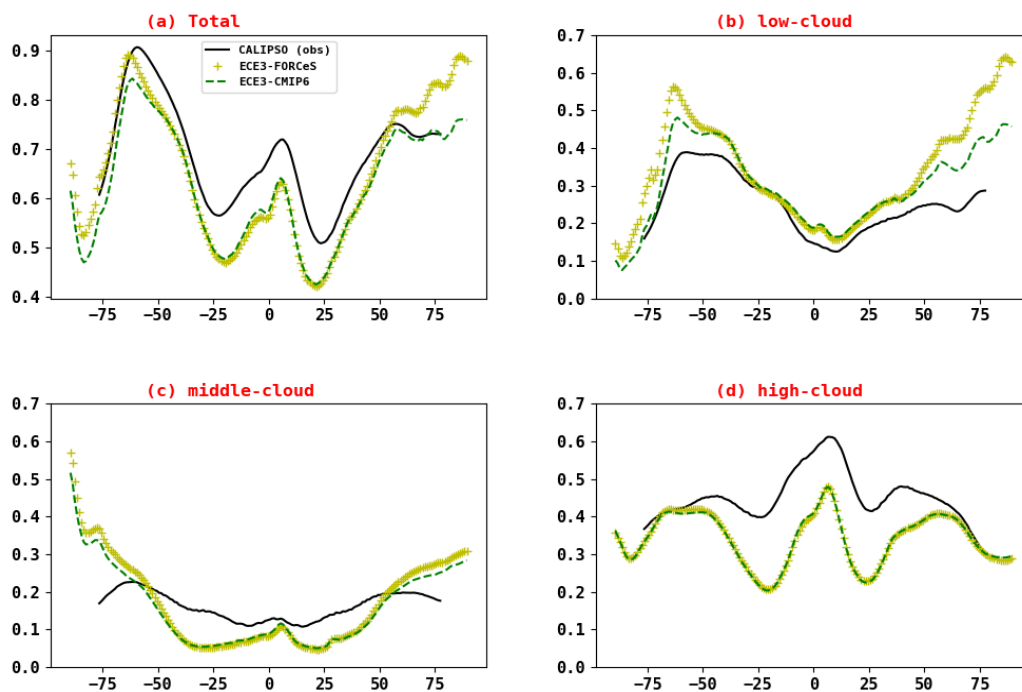Zonal mean cloud fraction: CALIPSO obs. vs CALIPSO-COSP simulations

Figure 2. Zonally averaged climatological mean CALIPSO-COSP simulated (a) total, (b) low-, (c) middle- and (d) high-cloud fractions. Solid black lines correspond to the CALIPSO observations. The green dashed lines are the ECE3-CMIP6 simulations and the dotted yellow lines correspond to the ECE3-FORCeS simulations.

model do not bring a significant change in the cloud fraction over the tropics and mid-latitudes compared to its ECE3-CMIP6 counterpart. However, the total and the low-cloud fractions in ECE3-FORCeS simulations in the Northern Hemisphere (NH) high latitudes tend to deviate even further from the observations compared to those in ECE3-CMIP6 runs.

250   A comparison of the simulated climatological mean total/water/ice cloud fractions with observations (in Fig. 3) shows that though the model simulates the spatial distribution of total cloud fraction well, it fails to reproduce the magnitude over the stratocumulus region west of the continents. The model captures the liquid water cloud fraction more realistically and, over the aforementioned regions, there is a slight overestimation compared to the observations. The global mean values of the water cloud fraction are close to 30% in both observations and the simulation. The ice cloud fraction, on the other hand, is

255   overestimated over the mid and high latitudes in both hemispheres in the model, thereby overestimating the global mean values of the ice cloud fraction by 8%. It can be noted that the simulated ice cloud fraction over the stratocumulus cloud decks is comparable to what is seen by MODIS. This would indicate that the mixed cloud fraction in the model is underestimated which could partly explain the underestimation seen in the simulated total cloud fraction across these regions.
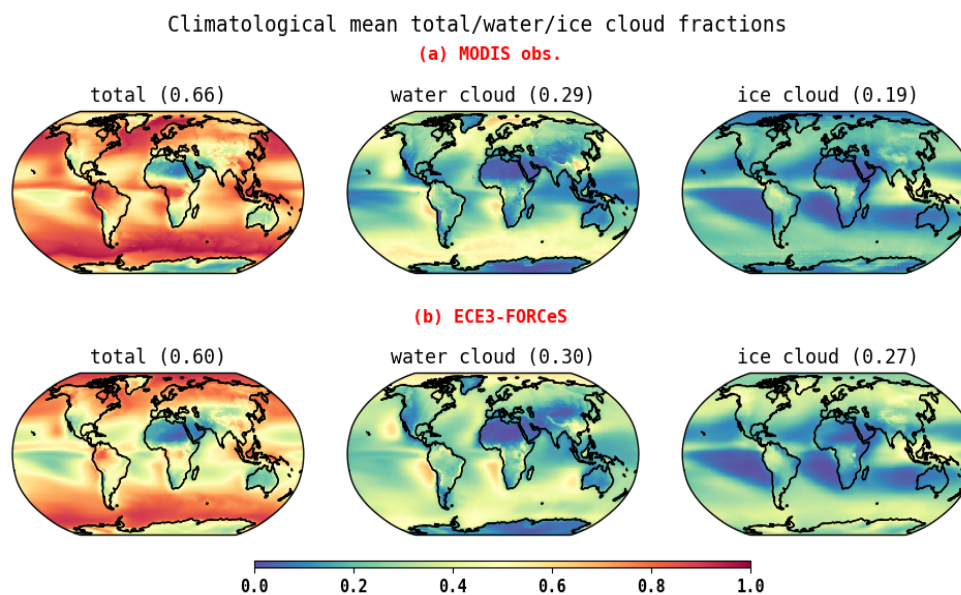
Figure 3. Climatological mean total (left), liquid water (middle) and ice (right) cloud fractions respectively in the three columns from (a) MODIS observations and (b) ECE3-FORCeS simulations.

3.2 Evaluation of cloud radiative effects

260   The cloud radiative effects (CRE) at the top of the atmosphere (TOA) is defined as the difference between the TOA all-sky and clear-sky fluxes. The spatial distribution and zonal means of the simulated climatological mean shortwave (SW) and longwave (LW) TOA CRE are evaluated against CERES-EBAF observations and the results are illustrated in Fig. 4 as differences in the simulated values from the observed values. Both model versions overestimate the SW cloud forcing over the Southern ocean and along the equator and over the pollutant outflow regions of Africa, North and South Americas and eastern China. However,

265   the positive biases are almost halved in ECE3-FORCeS compared to ECE3-CMIP6 simulations where the biases were more than 40 W/m^2 ((Fig. 4, column #1). On the other hand, the LW CREs are underestimated, particularly along the equatorial belt (Fig. 4, column #2). The slight positive biases in LW in ECE3-CMIP6 simulations in the stratocumulus regions over the western coast of the continents is reduced in ECE3-FORCeS. However, we overestimate the LW forcing in the high latitudes in the ECE3-FORCeS model. It can be seen more clearly in the zonal mean plots shown to the right in Fig. 4. There is a

270   considerable improvement in the SW CRE with the FORCeS model updates. The overestimation in the SW CRE at the TOA is notably reduced by more than 20 W/m^2 over the Southern ocean. In contrast, there is an overestimation in the LW CRE at latitudes > 50° in the FORCeS version, with biases in the range of 3-5 W/m^2, while in the mid-latitudes (between 50°S and 50°N), there is a slight improvement.
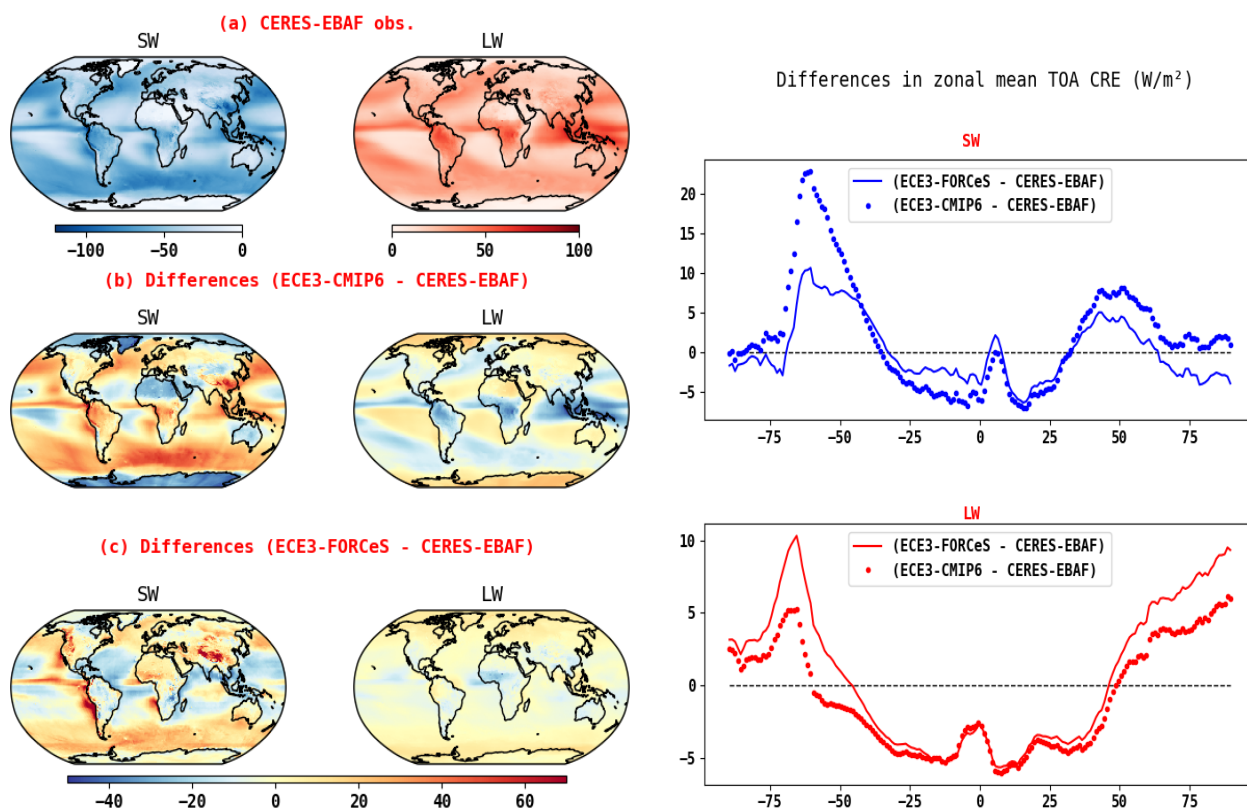
Figure 4. (left) Climatological mean CRE (in W/m^2) in SW (left) and LW (right) at the TOA from (a) CERES-EBAF observations, (b) differences of CMIP6 simulations and (c) ECE3-FORCeS simulations from observations. (right) Zonally averaged climatological mean CRE in SW and LW shown as differences from the observations.
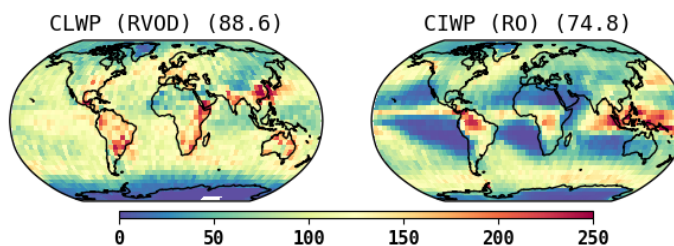
3.3 Evaluation of cloud microphysical properties

275 Measuring cloud water is crucial for investigating microphysical processes and the indirect impacts of aerosols. Cloud water is commonly represented using column-integrated metrics like cloud liquid water path and cloud ice water path. Since the COSP simulator for CloudSat products is not available, we use the standard model output for CLWP and CIWP for comparison with CloudSat retrievals. This comparison could still be reasonable as CloudSat can see through the whole cloudy column. Furthermore, using the MODIS visible optical depth constrained product for CLWP and Radar-Only product for CIWP provide

280 the best reference for cloud water free from the contamination by precipitation signal.

The spatial distribution of the ECE3-FORCeS simulated climatological mean fields against CloudSat observations and the differences from ECE3-CMIP6 simulations are presented in Fig. 5. These maps need to be interpreted with caution as the color bar range varies between model and observations. Despite the model's realistic simulation of spatial distribution, both CLWP
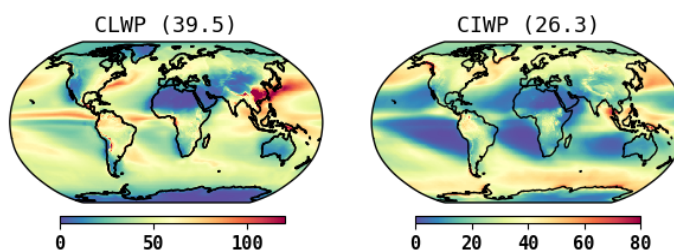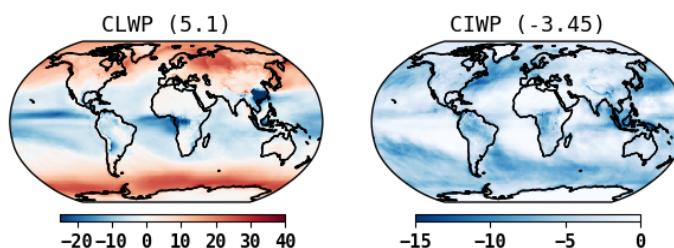
Figure 5. Climatological mean cloud liquid water (left) and ice (right) paths (g/m^2) from (a) CloudSat observations, (b) ECE3-FORCeS simulations and (c) their differences with respect to ECE3-CMIP6 simulations. The global mean values are given in the brackets.

and CIWP are markedly underestimated when compared to the actual observations. Following the FORCeS updates, CLWP
285 increased over high- and mid-latitudes in both hemispheres, while decreased over the tropics compared to the ECE3-CMIP6
version of the model. A significant global decrease of up to 3.5 g/m^2 in CIWP is simulated in the ECE3-FORCeS model with
regional decreases of up to 10-15 g/m^2, particularly over the oceans in the NH and SH.

The simulated climatological mean CD effective radius is evaluated against MODIS observations as in Fig. 6. The global
mean values are shown in the brackets for the same region (75°S- 75°N) as for the MODIS observations. The magnitude and
290 spatial pattern along the equatorial oceans are very close to the observations. However, the droplets sizes are comparatively
smaller in the simulations than in the observations. The observed global climatological mean is 14.7 μm compared to a value of
7 μm in the ECE3-FORCeS model simulation. The differences in the CD effective radius are in the range of uncertainty of the
observations wherein the biases in regional monthly mean MODIS derived values are at least 1 to 10 μm depending on cloud

horizontal heterogeneity and solar zenith angle (Fu et al., 2019). Notably, the latest model updates have led to an increase in
295 CD effective radius, especially over the tropical oceans, as observed in comparison to the ECE3-CMIP6 simulations.
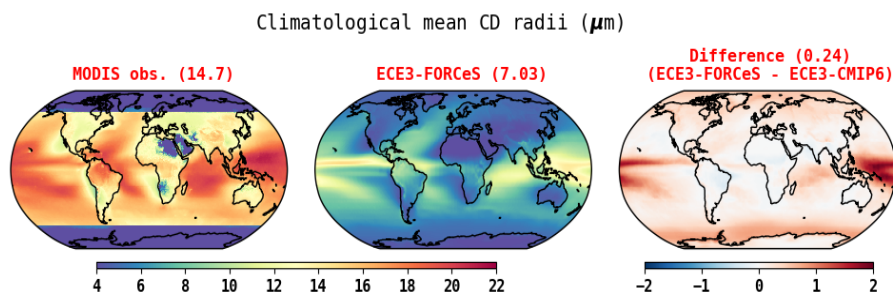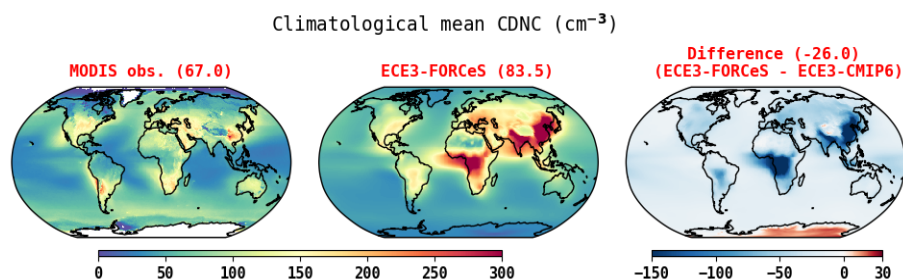
Climatological mean CD radii (**μ**m)

Figure 6. Climatological mean CD effective radius (μm) from MODIS observations (left), ECE3-FORCeS simulations (center) and their differences with respect to ECE3-CMIP6 simulations (right). The global mean values are given in brackets.

Climatological mean CDNC (cm^{-3})

Figure 7. Climatological mean in-cloud CDNC (cm^{-3}) from MODIS observations (left), ECE3-FORCeS simulations (center) and their differences with respect to ECE3-CMIP6 simulations (right). The global mean values are given in brackets.

Cloud droplet number concentration (CDNC) is a crucial quantity that influences the cloud properties, precipitation and radiative transfer. In-cloud mean CDNCs derived from MODIS observations and ECE3-FORCeS model simulation are shown in Fig. 7. The spatial distribution of the simulated CDNC resembles the MODIS observations relatively well. However, the ECE3-FORCeS model overestimates the CDNCs over sub-Saharan Africa and Asia, particularly over the highly polluted

300 regions of China and India. The discrepancies seen here are due to the fact that MODIS observations focus exclusively on the cloud top, thereby extracting the cloud properties, such as CDNC, only at the topmost layer of the cloud. On the other hand, the simulated CDNC corresponds to vertically averaged values up to around 680 hPa. This altitude level is chosen based on this cloud classification by ISCCP (International Satellite Cloud Climatology Project) where the this level is considered to be the low cloud top (https://isccp.giss.nasa.gov/cloudtypes.html). The differences between the two model versions show that with

305 the latest updates, the overestimations over Africa, India and China have been considerably reduced by more than 100 cm^{-3}.

4 Results-II: Maximum Covariance Analysis: Covariability between AOD and CD effective radius

4.1 Regions selected for the analysis

In order to investigate the maximum covariance between AOD and CD effective radius, we select the regions based on two prerequisites: a) the regions shall sustain low-level liquid water clouds and b) the regions experience at least some degree of pollution outflow from the continents. Among all the oceanic cloud regimes, these low-level clouds are most likely to be influenced by aerosols and stand the highest chance of testing the hypothesis that aerosol-cloud covariability can also be visible in the larger spatio-temporal scales. We selected three such regions based on our previous study, Devasthale and Thomas (2011), which investigated the frequency of occurrence of aerosol-cloud overlap globally in different seasons. They are shown in Fig. 8 by the blue boxes.
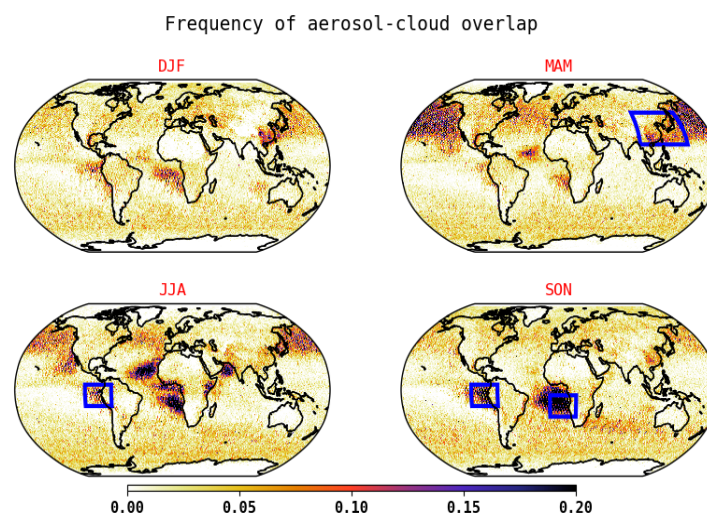

Figure 8. Frequency of aerosol-cloud overlap estimated based up on 4 years (May 2006 - June 2010) of CALIPSO satellite observations (Devasthale and Thomas, 2011). The regions selected are marked by the blue boxes.

The table below provides details about the selected regions, each distinguished by specific types of aerosols. For instance, the Eastern China region is marked by anthropogenic aerosols, persisting throughout the year but with a reduced intensity during the June-July-August season due to pollutant removal by wet deposition. The remaining two regions, designated as BB1 and BB2 respectively, for the west coast of Africa and the west coast of South America, are notably influenced by biomass burning aerosols in the free troposphere and marine aerosols in the boundary layer (Bourgeois et al., 2015, 2018). The prevalence of transported biomass burning aerosols is higher during September - November for Africa and June - November for South America during the dry season.

The climatological mean AOD and low-cloud fraction for the three regions selected for this study are shown in Fig. 9. It should be noted that the color bar varies according to the regions. Among these regions, Eastern China experiences the highest

Table 1. Regions selected for the study. 'BB' refers to biomass burning.

Regions	longitude; latitude	time periods
Eastern China	95 E - 150 E; 20 N - 50 N	All months
BB1: Africa	10 W - 20 E; 25 S - 5 S	Sept - Nov
BB2: South America	100 W - 70 W; 15 S - 5 N	Jun - Nov

AOD, reaching 0.7, in the area of maximum anthropogenic pollution. In the other two regions, AOD peaks over land with values around 0.25, corresponding to areas where biomass burning occurs. The Andes mountain range, situated to the west of the South American continent, acts as a barrier that partially hinders the transport of pollutants to the oceans. The low-cloud fraction ranges from 0.3 to 0.7 over these regions. It is also to be noted that the regions where the maximum pollution outflow over the oceans occurs and the regions where the maximum frequency of low-level liquid clouds is observed are not the same. The maximum aerosol-cloud overlap in fact occurs over the regions further remote from the individual maxima in AOD and cloud fraction.

4.2 MC analysis over Eastern China

As explained in Section 2.4, maximum covariance analysis is applied to assess the covariability between AOD and CD effective radius across Eastern China. Before applying the MCA, both these variables are deseasonalized and detrended. The heterogeneous patterns, the patterns that brings out the areas of covariability between the two fields, are presented. Only regions exhibiting statistical significance at the 95% confidence level are showcased. We present the first three modes derived from the analysis for Eastern China in Fig. 10 (a) - (c). These three modes collectively explain 66% of the total observed variability. A distinct pattern exhibiting opposite signs between AOD (left) and CD effective radius (right) emerges and each mode captures the different regions where this inverse covariability is dominant. The opposite signature between these two fields clearly suggests that the Twomey effect over those regions in the corresponding modes can influence the variability at the large spatio-temporal scales. The first two modes capture the covariability over the outflow regions where the aerosol advection over nearby oceanic low-level clouds is quite frequent (Devasthale and Thomas, 2011), whereas the third mode captures the covariability over the continental source regions. It is intriguing that the covariability between AOD and CD effective radius is not only noticeable in oceanic outflow regions but also extends to land regions. This is mainly due to the fact that different cloud regimes and thermodynamical conditions exist over the land and the oceanic regions in the selected study area. The covariability is especially stronger just off the Eastern Chinese coast in the first two modes, suggesting a noticeable influence of pollution advection on the low-level clouds, possibly dominating over other factors that can also cause covariability.
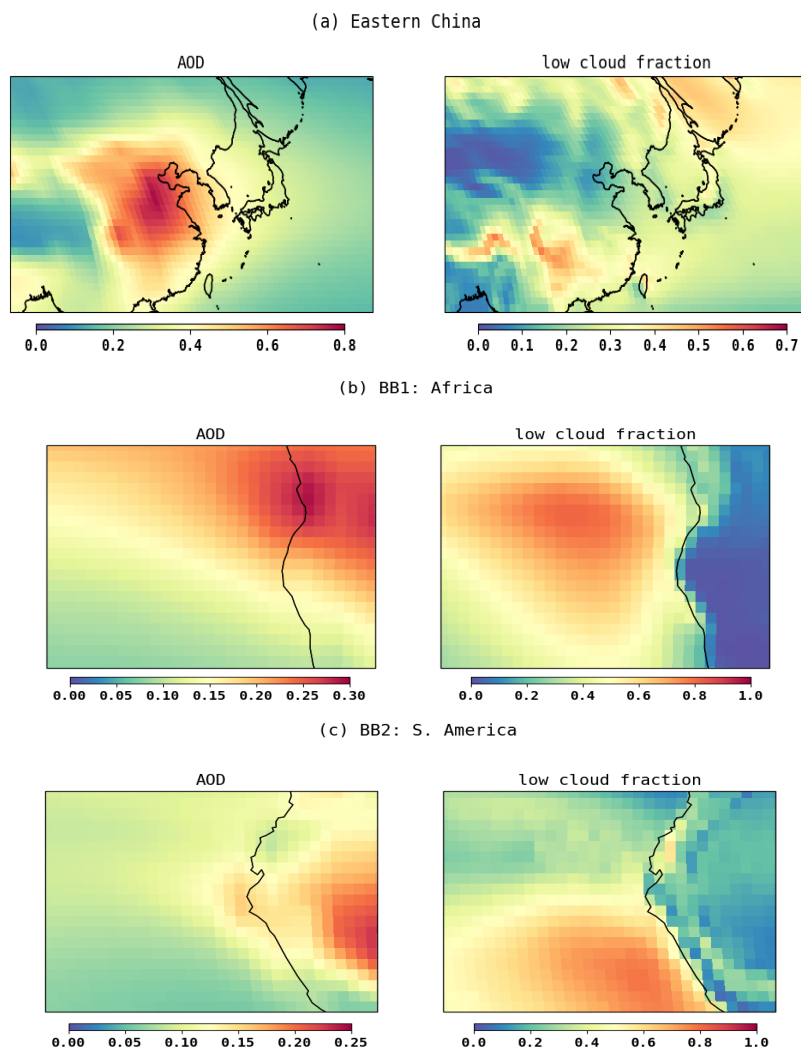
Figure 9. Climatological mean AOD (left) and low-cloud fraction (right) over the selected three regions: (a) Eastern China (b) BB1: Africa and (c) BB2: South America.

4.3 MC analysis over BB1: Africa

The maximum covariance analysis applied to AOD and CD effective radius over this region reveals that only the first mode is the most significant and dominant mode explaining a variability of up to 20%. The remaining modes do not exhibit statistically significant covariability and the SCFs in the subsequent modes are less than 10%. So, here we focus only on the first mode. The covariability seen in this only one mode is nonetheless very significant and noteworthy, as this mode exhibits the regions where the Twomey effect is likely dominant, particularly over the pollutant outflow regions. The heterogeneous pattern in these two fields for this mode is shown in Fig. 11.

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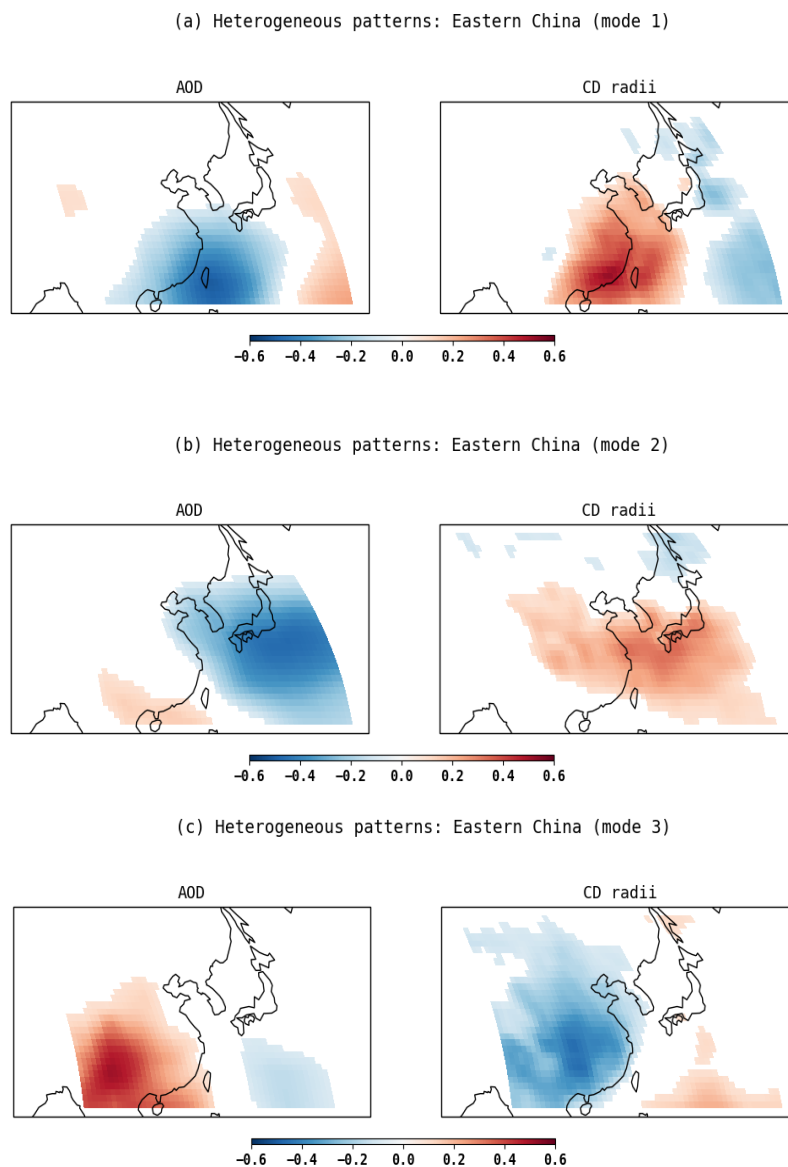
Figure 10. Statistically significant heterogeneous patterns over Eastern China in the three modes (a)-(c) derived from the maximum covariance analysis.

As mentioned earlier, this study region is dominated by the seasonal biomass burning in southern Africa that occurs during the months of June through November, transporting enormous amounts of absorbing aerosols across the southeast Atlantic over the extensive stratocumulus decks. This outflow region has therefore been the center of many measurement campaigns and studies focusing on characterizing aerosols and clouds and their interactions (see, for example, Redemann et al. (2021)).

To what extent these aerosols affect the underlying stratocumulus decks is still the topic of intense research. Although the freshly emitted biomass burning aerosols are not ideal condensation nuclei, numerous studies have previously shown that the morphology, composition and size distribution can change considerably during the transport, making them one of the dominant local sources of condensation nuclei, thereby increasing cloud droplet number concentration and changing droplet size (Lu et al., 2018; Royer et al., 2023; Che et al., 2022; Petters et al., 2009).

The significant covariability between AOD and CD effective radius here indeed suggests that as these biomass burning aerosols are advected over the oceanic stratocumulus regions in the model simulation, they undergo aging and mixing with other anthropogenic aerosols as well as marine aerosols, possibly becoming more efficient CCNs. In the ECE3-AerChem model, the open biomass burning emissions are distributed up to 2 kms following the vertical profiles for forest fires as given in Table A1 of van Noije et al. (2014). The accompanying advection of humidity however makes the aerosol-cloud interactions over these regions less predictable as the local meteorology together with the changing aerosol aging, coating and size distribution introduces high internal variability.
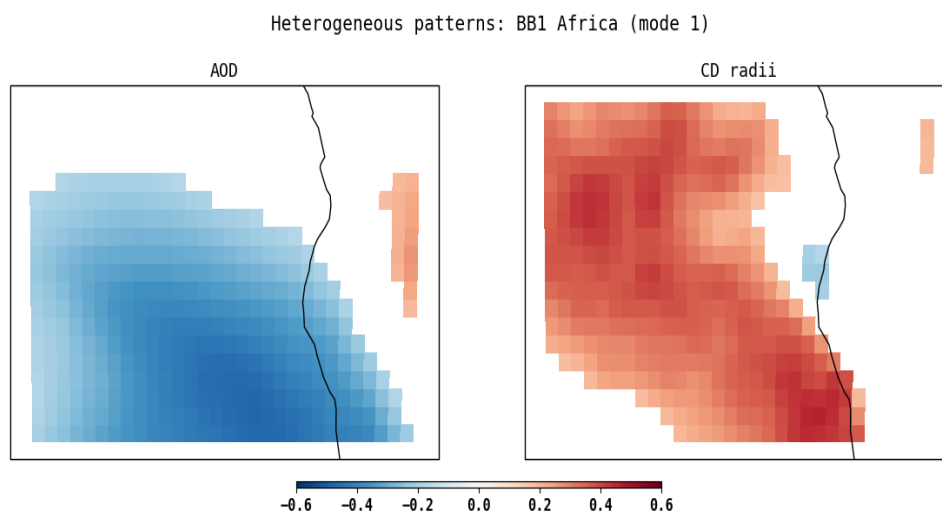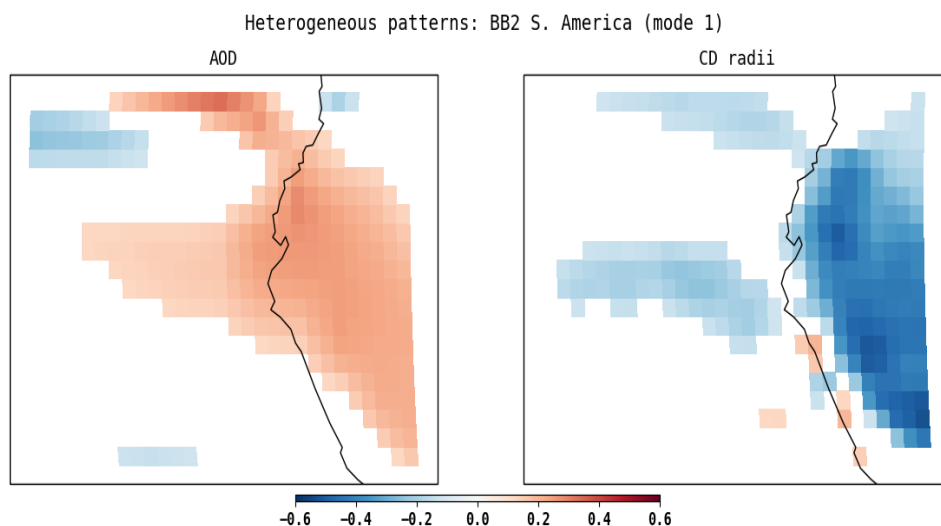

Figure 11. Statistically significant heterogeneous patterns in mode 1 derived from the MCA over biomass burning (BB1) outflow region of Africa.

4.4 MC analysis over BB2: S. America

Although the frequency of aerosol-cloud overlap is also high in this outflow region, the weakest covariability between AOD and CD effective radius is observed over the ocean compared to previous two study areas as shown in Figure 12. Although the first mode alone accounts for 59% of the total variability, the spatial extent of the statistically significant covariability and its magnitude is limited and weak over the ocean compared to the land. A number of factors that can affect the variability in AOD and clouds need to be considered here. The aerosols need to be convected and advected over much longer distances in

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the free troposphere crossing the Andes mountain range. The high residence time and aging during the transport likely lead to strong changes in aerosol properties and size distribution before they are available as condensation nuclei after the descent over the cooler upwelling waters off the western coast of south America. Furthermore, the large-scale dynamical variability associated with the El Niño Southern Oscillation (ENSO) strongly influences the variability of low-level cloudiness in this

380 region. ENSO also likely induces variability in the background marine aerosols through changes in sea surface temperatures and winds. Therefore, the correspondence between AOD and CD effective radius at larger temporal scales is much weaker.

Figure 12. Statistically significant heterogeneous patterns in mode 1 derived from the MCA over biomass burning (BB2) outflow region of South America.

4.5 Implications for cloud radiative effects

Additionally, we examined the implications of this covariability between AOD and CD effective radius for the cloud radiative effects. To investigate this, a composite analysis is carried out based on the PCs of AOD as shown in Fig. 13 as an example for

385 mode 1 over Eastern China.

Two composites were generated, one for which the PCs of AOD are greater than zero and another where the PCs of AOD are less than zero. In the composite analysis for PCs of AOD > 0, we investigated the anomalies of TOA SW CRE, AOD, CD effective radius, CLWP, CDNC, specific humidity, and low-cloud cover fraction. The composite analysis for the PCs of AOD < 0 is exactly the opposite of those obtained for PCs of AOD > 0. For brevity, we present the outcomes (Fig. 14 for the three

390 modes specifically over Eastern China for one composite analysis when PCs of AOD > 0.

In this composite, the anomalies in TOA SW CREs indicate pronounced cooling over the pollution source regions and the pollutant outflow region in modes 1 and 2, while the opposite is observed in mode 3 as can be seen in Fig. 14. The significant cooling observed in the first two modes can stem from two processes, either enhanced aerosol-cloud interactions
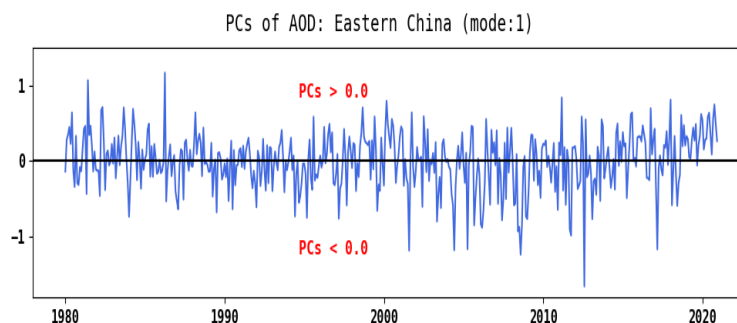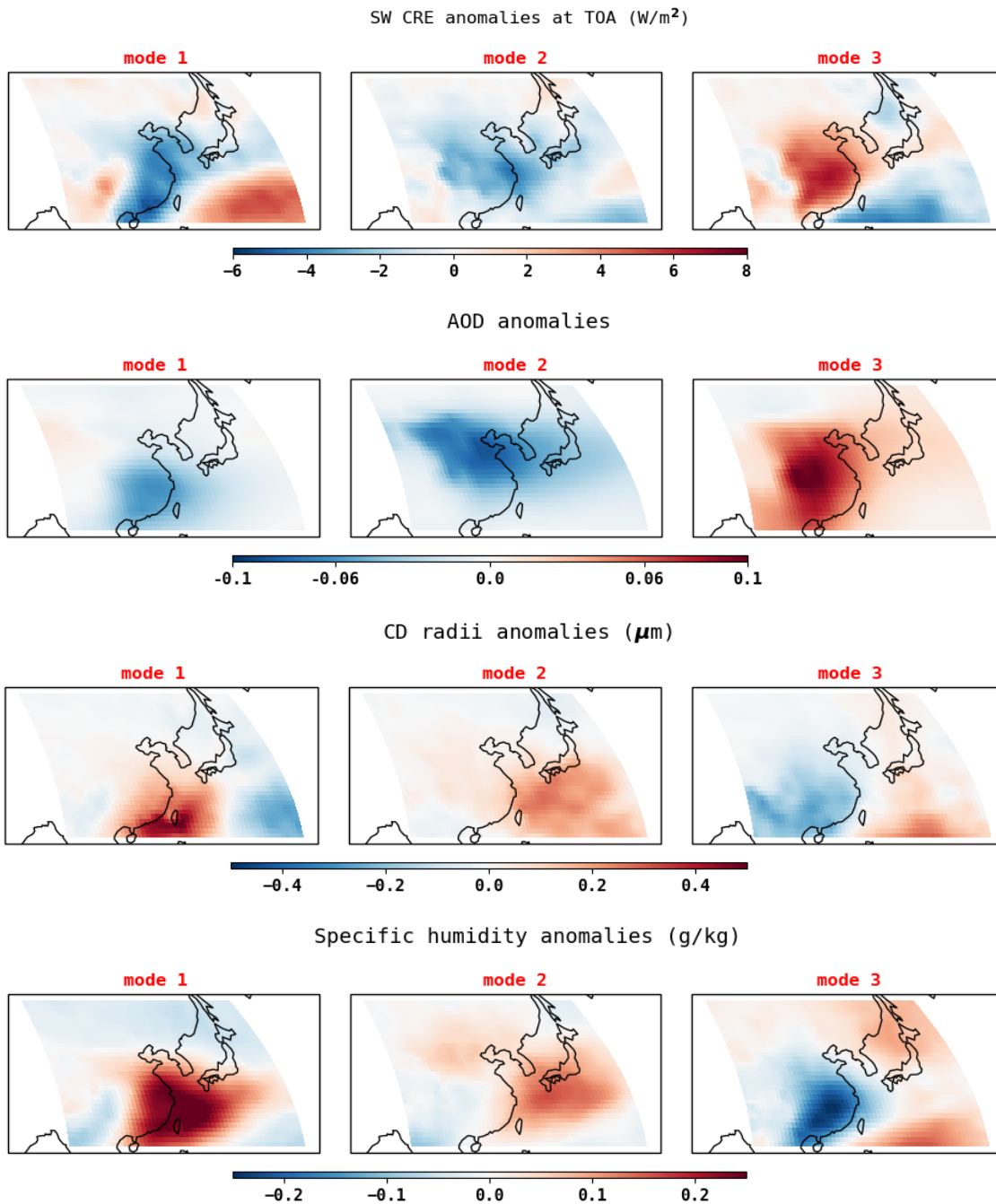
Figure 13. The monthly principal components of AOD corresponding to the mode 1 of the MCA.

(Twomey effect) and/or to an increase in low-cloud cover, leading to brighter clouds and increased reflection. Regarding the
395 first hypothesis, the AOD anomalies are in fact lower in this composite together with a corresponding positive anomaly in CD effective radius, indicative of larger droplets. The cloud droplet number concentration is also lower. This indicates that the Twomey effect cannot explain the cooling anomalies in the TOA SW CREs. However, a higher low-cloud fraction in tandem with higher CLWP and specific humidity is seen. This higher low-cloud fraction can adequately account for the marked cooling in TOA SW CREs. Consequently, the results show that, despite the existence of robust aerosol-cloud interactions at the larger
400 spatio-temporal scales, the TOA SW CREs are predominantly driven by the changes in low-level cloud fraction in these simulations. Previous studies (Gryspeerdt et al., 2019) have shown the inverse relationship between CDNC and CLWP over the heavily polluted regions based on satellite observations. The results of this composite analysis are in line with those previous studies, suggesting that CDNC-CLWP control can indeed have larger impact on cloud radiative effects than the Twomey effect.

Similar conclusions can be derived for the first modes over the biomass burning regions of Africa and S. America. The
405 results are shown in the Appendix Figs A1, A2 for Africa and A3, A4 for S. America.
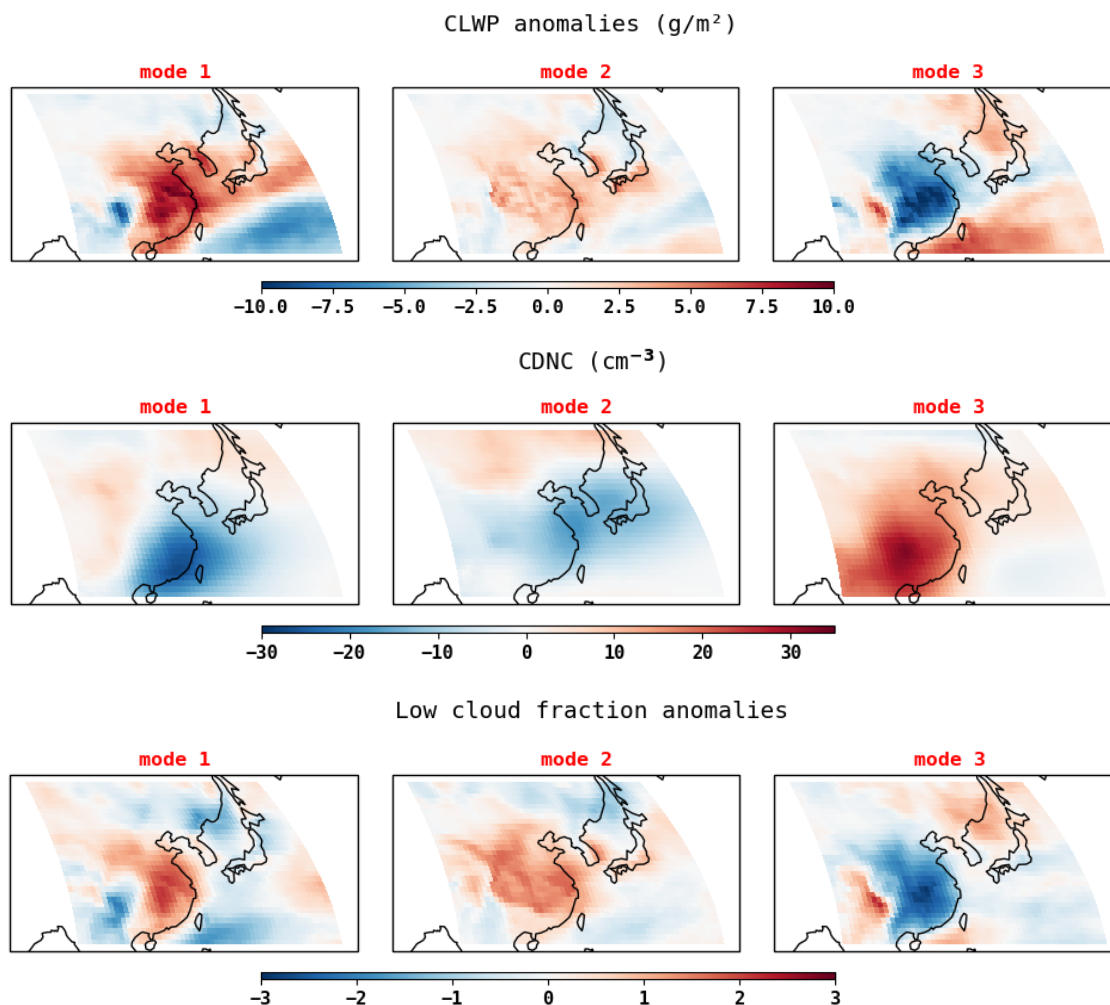
SW CRE anomalies at TOA (W/m²)

AOD anomalies

CD radii anomalies (μm)

Specific humidity anomalies (g/kg)

*

Figure 14. Each panel represents respectively the anomalies of TOA SW cloud radiative effects (W/m^2), AOD, CD effective radius (μm), specific humidity (g/kg), cloud liquid water path (g/m^2), CDNC (cm^{-3}) and low cloud fraction in the three modes over eastern China.

5 Discussions and conclusions

In the framework of the EU project FORCeS, a number of significant improvements in the representation of aerosol and cloud processes were implemented in the ECE3-AerChem model, ECE3-FORCeS. The ECE3-FORCeS includes now a representa-
410   tion of the atmospheric cycle of dust minerals that are relevant for their climate impacts, as well as a new source of marine organic aerosols. As a result, the absorption of radiation by dust in the SW is now dependent on the online calculated abundance of hematite. The model also incorporates an aerosol-sensitive scheme for the estimation of primary ice crystals, which depends on the abundance of quartz, k-feldspars and marine organics for the immersion freezing, and on dust and soot for

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deposition nucleation. This primary ice can lead to secondary ice production in supercooled conditions, which is quantified
415 via RaFSIP (Georgakaki and Nenes, 2023). A new warm cloud activation scheme by Morales-Betancourt and Nenes (2014) is
also implemented, enabling a more accurate estimate for the number concentration of droplets formed at the time of maximum
supersaturation. Finally, a new thermodynamic module for inorganic aerosols, ISORROPIA-LITE, has been also implemented
in the model.

These changes and improvements are bound to have an impact on the cloud properties and their radiative effects. Therefore,
420 we used 41 years of historical atmosphere-only (AMIP) simulations for the period 1980-2020 from this recent FORCeS model
version to confront them with the satellite-based observations. A suite of satellite sensor retrievals and simulators are used to
facilitate the comparison. Due to increased cloud fraction and cloud liquid water path compared to its previous CMIP6 version,
the strong warm biases often seen over the Southern ocean is reduced significantly in the FORCeS version of ECE3-AerChem.
The bias in the SW TOA CREs is reduced by nearly 50% over this region.

425 Using these simulations from the ECE3-FORCeS, we further carried out a maximum covariance analysis between AOD and
CD effective radius. The aim of this exercise was not to identify any new climate process, but rather to test if the covariability
between AOD and CD effective radius could also be seen at the larger spatial and temporal scales. Among the three oceanic
regions that were chosen based on the high frequency of aerosol and low-level cloud overlap, the strongest statistically sig-
nificant inverse covariability between AOD and CD effective radius was observed over the eastern Chinese outflow region.
430 Here, the aerosol loading is generally quite high and the aerosol sources are mostly anthropogenic. The conditions favourable
for allowing aerosol-cloud interactions, such as persistence of westerly winds, high humidity and aerosol load in the lower
troposphere, favourable aerosol composition and size distribution over the outflow region likely lead to strong inverse covari-
ability between AOD and CD effective radius seen even at larger spatio-temporal scales. This means that the Twomey effect
can manifest itself even at a climate scale by dominating the other possible drivers of local variability. Note that the model
435 simulations do not suffer from the limitations often discussed when using satellite data for similar analyses, such as seeing
only cloud top information or sampling issues related to simultaneous aerosol and cloud retrievals. Furthermore, the most of
the aerosol outflow in this region occurs in the lowermost troposphere and hence AOD could be indeed a good proxy for the
cloud condensation nuclei.

In the case of the other two regions in the southeast Atlantic (along the west coast of Africa: BB1) and eastern equatorial
440 Pacific (along the west coast of S. America: BB2), biomass burning is a significant source of aerosols during the months
selected for the study. The covariance between AOD and CD effective radius is statistically significant only in the first mode of
variability in these regions. However, it can be seen that the covariance is more pronounced over the pollutant outflow region
in BB1 and the source region in BB2. This is most likely due to fact that the aerosol vertical distribution and its variability is
more disengaged from the variability in the underlying cloud decks due to longer transport distances in BB2 due to the Andes
445 mountain range and the subsequent changes in aerosol composition and size distribution that may not favor the covariability.
In such case, the total AOD may indeed not be a proxy for cloud condensation nuclei, as argued by the previous studies (Quaas
et al., 2020). Our analysis also show a strong cooling/warming in the TOA SW CREs despite strong covariability (larger
droplets with low AOD and vice versa) and can be attributed to stronger/weaker control by the relationship between CDNC

and CLWP. Further investigations are underway to carry out similar study using satellite observations, but the lack of reliable,

450 multidecadal data of AOD, CCN and CDNC is currently making such an analysis challenging.

Code and data availability. The EC-Earth3 code is available from the EC-Earth development portal for members of the consortium. All code related to CMIP6 forcing is implemented in the component models. The model codes developed at ECMWF, including the atmosphere model IFS, are intellectual property of ECMWF and its member states. Permission to access the EC-Earth3 source code can be requested from the EC-Earth community via the EC-Earth website (http://www.ec-earth.org/about/contact/, EC-Earth-Consortium (2023)) and may be

455 granted if a corresponding software license agreement is signed with ECMWF. The repository tag for the version of EC-Earth that is used in this work is "projects/FORCeS". Currently, only European users can be granted access due to license limitations of the atmosphere model. However, the model code is made accessible to both the Editor and the reviewers throughout the review process. The component models NEMO, LPJ-GUESS, TM5, and PISM are not limited by their licenses.

All model output data are freely available from any ESGF data node as part of CMIP6 (https://esgf-data.dkrz.de/search/cmip6-dkrz/);

460 search for $< model\_id >$=EC-Earth3-AerChem, $< experiment\_id >$=amip, and $< variant\_label >$=r1i1p1f1 for ECE3-CMIP6 or $< variant\_label >$=r1i1p4f1 for ECE3-FORCeS. The python scripts used in generating the figures in the manuscript and the observational data for model evaluation are archived in Zenodo under https://doi.org/10.5281/zenodo.10781927
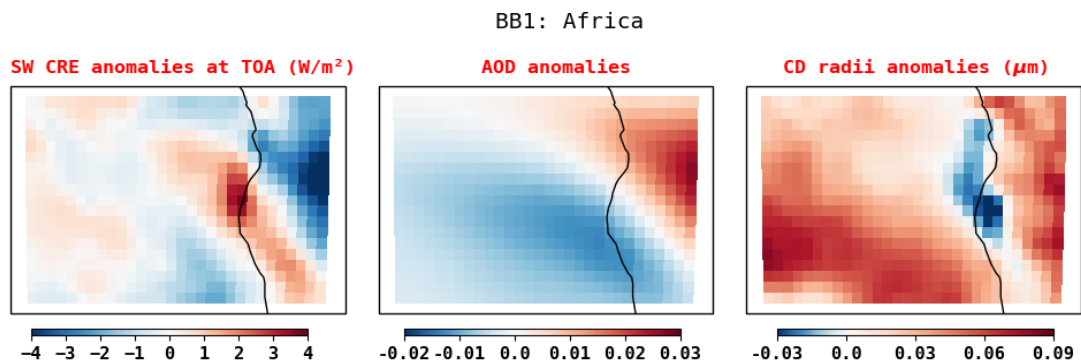
Figure A1. Anomalies of top of the atmosphere SW CRE (W/m^2), AOD and CD radii (μm) in the three modes over BB1: Africa
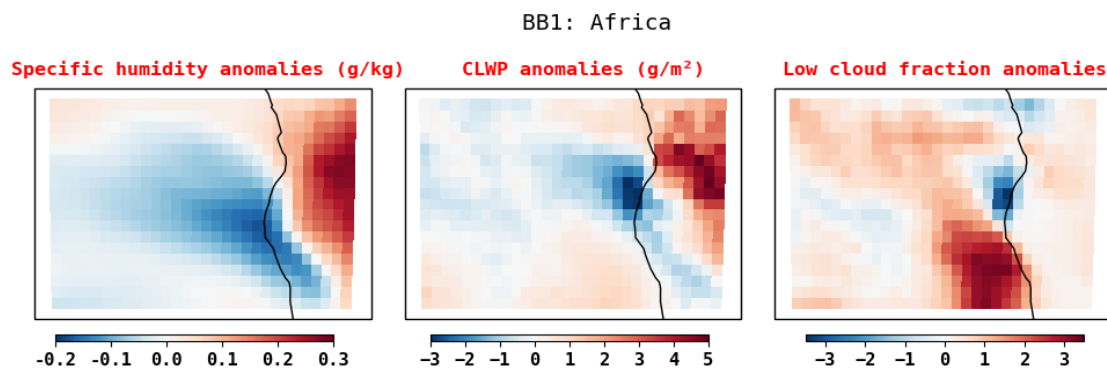

Figure A2. Anomalies of specific humidity (g/kg), CLWP (g/m^2) and low cloud fraction in the three modes over BB1: Africa
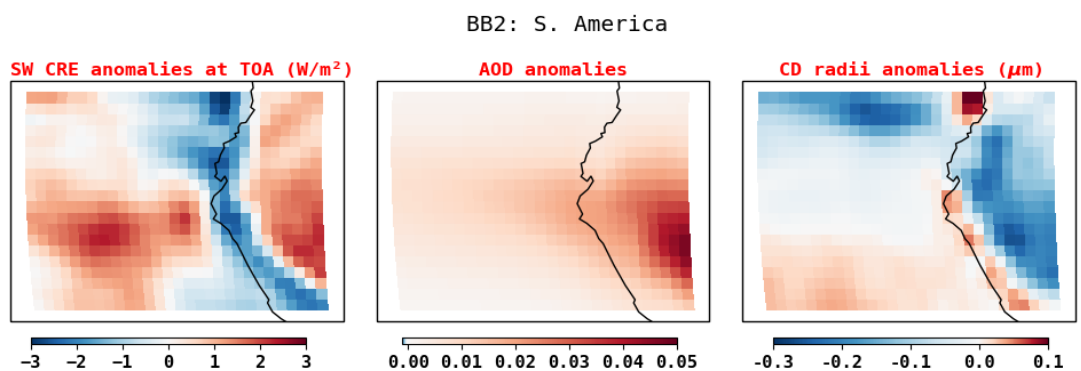

Figure A3. Anomalies of top of the atmosphere SW CRE (W/m^2), AOD and CD radii (μm) in the three modes over BB2: S. America
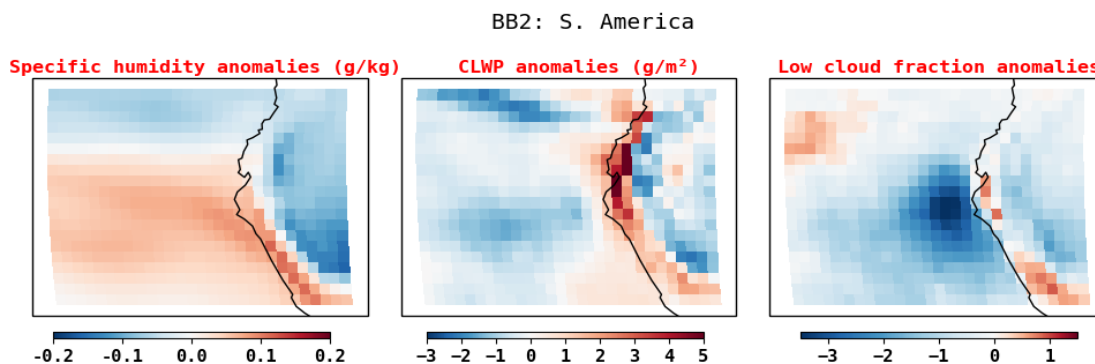
Figure A4. Anomalies of specific humidity (g/kg), CLWP (g/m^2) and low cloud fraction in the three modes over BB2: S. America

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